Activities & Scientific Report

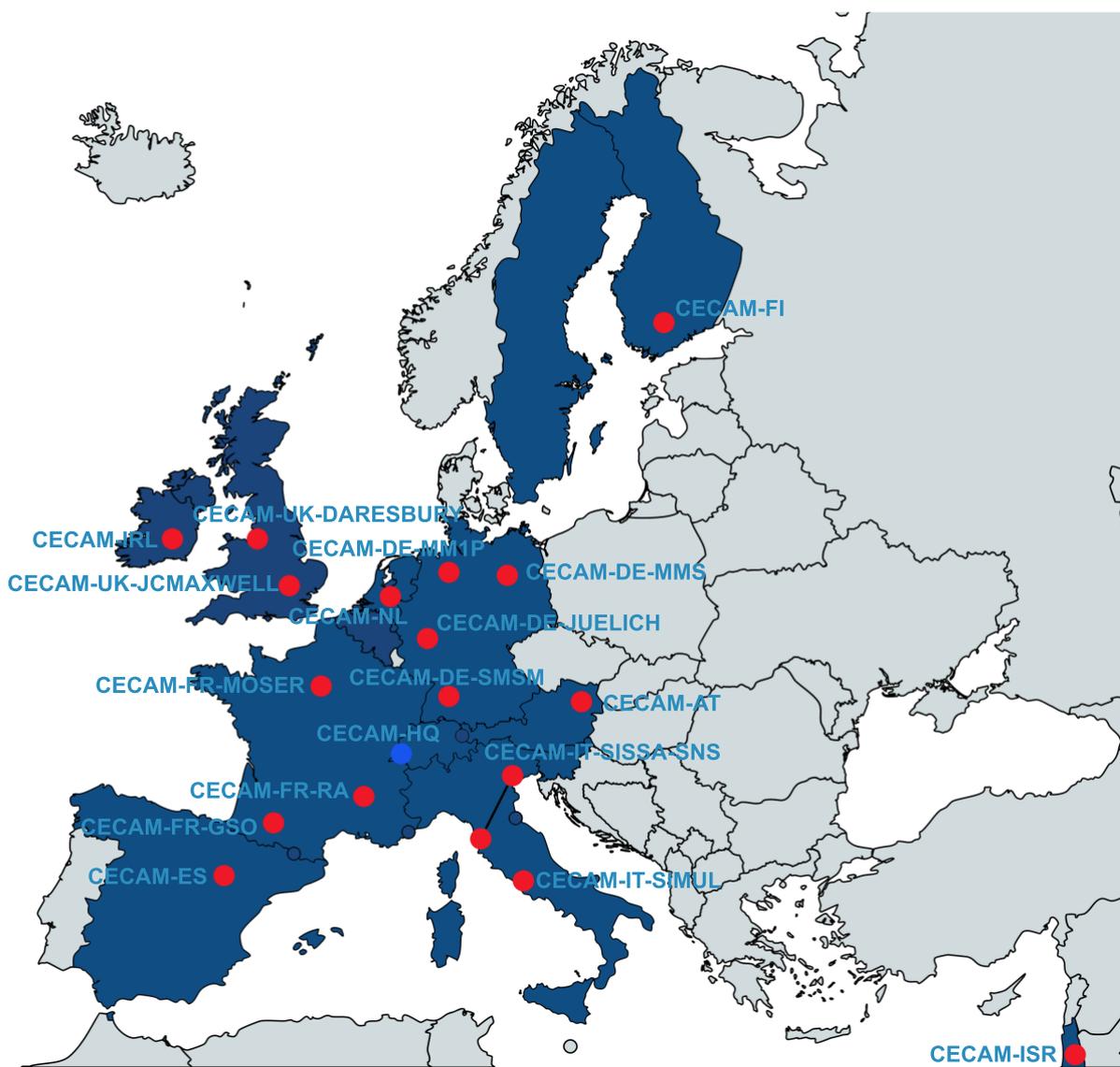# 2022

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INTRODUCTION

2022 marked the return to a normal deployment of the CECAM Flagship after the CoVid-19 pandemics. The year was characterised by an enthusiastic response of the community to the resuming of in-person activities, enabling more effective scientific exchange and networking opportunities, in particular for early career researchers. The technological infrastructure for online events, perfected in the previous two years, however, enabled a broader offer, including a small number (5 in total) of fully online events and exploitation of hybrid formats (17 events in total) to enable broader participation, specifically, in schools and a reduction of the carbon footprint of some events.

Activities at the Headquarters at EPF-Lausanne were back to pre-pandemic counters, with a total of 22 workshops and 3 schools held in the year. Of these, only 4 were online, with 22 workshop and 3 schools delivered in fully presential or hybrid format. 104 organizers from 16 different countries shaped this program, with a total number of 1602 participants (662 onsite, 940 online) from 60 countries.

The node network also assured a rich program of interesting events, delivering 33 workshop and 17 schools. 245 organizers from 19 countries were funded and 2103 participants (1570 onsite, 533 online) hosted in these events.

The scientific topics discussed in the 2022 Flagship program covered traditional and emerging areas in our field. Notably, the number of events dedicated to Machine Learning approaches (both in first principles and statistical mechanics) was quite high, 7, indicating the continued growth of the community's interest and contributions in this domain. Interestingly, CECAM also hosted a young researcher's workshop on Machine Learning for materials, indicating that the Center is identified as an incubator of new ideas across different generations. Among the leading-edge topics investigated, we signal a workshop on Theory and Simulation of small quantum systems and quantum thermodynamics, held at the Headquarters.

Following very positive feedback on the Mixed-Gen series, originally devised to enable PhD students to benefit from insight on their work from experts during the pandemic, it was decided to continue this successful webinar format with 7 more sessions.

The online format was maintained also for the Mary Ann Mansigh CECAM MARVEL conversation series, dedicated to the role of materials modeling in industry, and for 3 CECAM MARVEL Classics, covering topics in electronic structure and spin glass dynamics. Notably, this last lecture was delivered by the Nobel Prize winner Giorgio Parisi together with Marc Mezard.

Recent advances on the glass problem
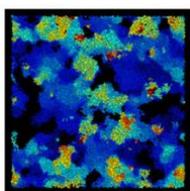
Location: on-line, hosted by CECAM-HQ
Webpage : https://www.cecam.org/workshop-details/48
Dates: Jan 5, 2022 - Jan 7, 2022

1 State of the art

Glass inhabits a complex and ramified energy landscape. Because a supercooled liquid that solidifies into a glass result from the system's failure to fully explore it, statics and dynamics are inextricably linked. The *transition* into a glass is therefore an entirely different phenomenon from the transition into a crystal. Cooling a liquid to low temperatures (the glass transition) and compressing a zero-temperature disordered collection of particles (the jamming transition) are two ways of obtaining amorphous solids. Jamming is athermal and static, and is in some ways simpler than glass formation, where thermal and dynamical effects also play crucial roles, yet also presents a remarkably rich phenomenology.

Recent advances in the glass field have discovered and characterized the jamming transition for dimension $d \geq 2$ and obtained a solution for the statics and the dynamics of liquids in the mean-field, $d \to \infty$ limit. The recent development of a field-theoretic framework to describe glassy dynamics away from $d = \infty$ as well as a harmonic theory for jammed solids near zero temperature further provide two broad insights, which make this workshop particularly timely: (i) the crucial role played by dimension as a control parameter, and (ii) the realization that real-space and phase-space approaches converge to the same critical exponents. A number of these advances would have been impossible without the constant support, confrontation, and validation from computational sciences. This research program has indeed benefited from and stimulated a vast program of molecular simulations, in order to both test the theoretical ideas and to provide a bridge between the mean-field results and physical systems in $d = 2,3$. New computational concepts and algorithms are pushing forward our numerical capabilities far beyond what was once considered conceivable, and are currently playing a central role in solving the glass problem.

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2 Major outcomes

The main themes of the workshop were articulated in four sessions (overlapping over the three days of the online workshop):

- **Activated and aging dynamics in the supercooled liquid state:** Activated dynamics in supercooled liquids is often pictured as driven by the hopping of barriers separating different energy basins. We discussed that even though barrier hopping does occur, it is not always the dominant activation mechanism. In fact, in some regimes the slow dynamics should be attributed not to the height of barriers, which are low or absent, but rather to the scarcity of convenient directions. We discussed two out-of-equilibrium regimes, where the dynamics is activated. These regimes differ qualitatively in the way

the landscape is explored: activation is driven by the height of energy barriers at lower temperatures, and by the availability of convenient pathways at higher temperatures.

- **Anomalous elasticity in amorphous solids:** (1) It was shown that mechanical strains in amorphous solids are screened via the formation of plastic events that are typically quadrupolar in nature. At low densities the screening effect is reminiscent of the role of dipoles in dielectrics, while the effect at high density has no immediate electrostatic analog, and is expected to change qualitatively the mechanical response, as seen for example in the displacement field. High-density screening results in an undulating displacement field that strictly deviates from elasticity theory, leading to a Kosterlitz-Thouless (or Hexatic like) phase transition from quasi-elastic to anomalous mechanical response. (2) Simple mean field spin glasses were discussed as models of low temperature excitations in amorphous solids. We discussed how marginal stability in the sense of the Gardner phase is realized in the spin glass phase both from the linear and non-linear excitations point of view.

- **Using machine learning to build a theory of glassy dynamics:** Disordered solids flow at high enough applied stress and melt at high enough temperature via localized particle rearrangements whose probability depends on the local structural environment. We discussed several machine learning methods to obtain simple variables that are highly correlated with the propensity of the particle to rearrange. The probability that a particle will rearrange yields a measure of the local free energy barrier to rearrangement. From these and other observations, we discussed how we can use these methods to construct coarse-grained theories of glassy dynamics.

- **Glassy phases in active matter:** We discussed how standard glassy physics can describe well the ergodic regime of active matter, provided the replacement of the ambient temperature by an effective 'temperature' associated to activity: higher temperature implies faster relaxation. However, beyond the glass transition, the relaxation of the nonergodic system abruptly slows down at low but nonzero activity. As we increase further activity, the relaxation speeds up until it exceeds the passive situation. This nonmonotonic behavior cannot be described by a simple increase in temperature. To explain this phenomenon, we discussed particle displacement orientation: relaxation in sufficiently active nonergodic phase follows collective motion mechanisms, while cooperative motion dominates at zero and low activities. Directed motion makes cage exploration less efficient and thus slows down cooperative relaxation with respect to a passive glass.

3 Community needs

Most of the theoretical advances we discussed at the workshop would have been impossible without the constant support, confrontation, and validation from computational sciences. This research program has indeed benefited from and stimulated a vast program of molecular simulations, in order to both test the theoretical ideas and to provide a bridge between the mean-field results and physical systems in d=2,3. New computational concepts and algorithms are pushing forward our numerical capabilities far beyond what was once considered conceivable, and are currently playing a central role in solving the glass problem. Our community is thus very heavily using computational methods, ranging from classical molecular dynamics to smart Monte Carlo algorithms, or to transition path finding algorithms such as the NEB, and to state-of-the-art machine learning methods using both simple and deep architectures. Our workshop included experimentalists such as Jeppe Dyre and Matthieu Leocmach.

4 Funding

Typical funding channels for our community are local grants (e.g. ANR grants from the French government), European grants (such as Horizon 2020 grants, i.e. ERCs and networks). A very important source of funding has also been the Simons collaboration "Cracking the glass problem", funded by the Simons Foundation, which was supposed to provide important funding to the workshop prior to its cancellation, for reasons related to the Covid pandemics. For the same reason, no informal discussions were possible during the workshop, so no joint research proposal were discussed.

5 Will these developments bring societal benefits?

Our workshop covered mostly fundamental science, so no immediate societal benefits of the science discussed can be expected in the short term. Long-term scientific impact can be expected, but it is hard to predict which way it will happen.

However, our workshop has important societal impacts in terms of workforce development. Both the organizing team and the speaker list contained a very diverse array of people, thus helping advance the teaching and research career of an academic community that is more representative of the population it serves. In addition, the astonishingly high number of registered participants from all over the world will result in a broad educational opportunity for the scientific community and help distribute frontier knowledge in a more democratic and discrimination-free fashion. We therefore expect our workshop to have improved the pipeline to science and technology careers for a large cross-section of its stakeholders. We should thus expect more CECAM proposals on this topic to emerge in the coming years.

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Theory and Simulation Methodologies for Small Quantum System Dynamics and Quantum Thermodynamics (QUANTDYNA)
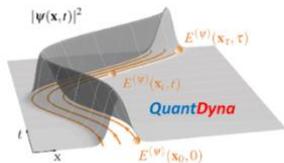
Location: On-line, hosted by CECAM-HQ
Webpage : https://www.cecam.org/workshop-details/40
Dates: Jan 17, 2022 - Jan 19, 2022

1 State of the art

The field of dynamics of small quantum systems is undergoing a rapid phase of development (the Second Quantum Revolution [1]). An outstanding scientific and technical challenge in this field is to understand and control the influence of an external environment to the dynamical state and operation of quantum devices coupled to it [2].

To this end it is crucial to model: (i) the thermodynamic behavior of small quantum systems, both isolated and open to the external environment (natural or engineered); (ii) the fluctuation relations that connect thermodynamic quantities such as work and free energy of the device; (iii) effects of the environment beyond weak coupling; (iv) the effect of the environment on strongly-correlated many-body physics and the emergence of new types of physics in open many-body quantum systems. The aim of this workshop was to bring together leading researchers from different backgrounds to cross-fertilize theoretical and numerical methodologies, and to present new results and methodologies to attack and discuss open outstanding problems. In particular, the unique feature here was to bring together experts on methods including Master Equation (Lindblad) and Quantum Langevin Equation approaches [3], Tensor Network Methods [4], Transfer Tensor Methods [5], Dynamical Mean Field Theory [6], and Machine Learning [7] just to mention a few. The invited and contributed talks were originally planned to cover many theoretical and computational aspects related to open quantum systems and their application to quantum computing, quantum heat engines, and optimal control of quantum systems. However, due to the rapidly escalating pandemic (Dec. 2021) the workshop was conducted fully online, and the program accommodated accordingly. The online version of the workshop comprised two invited talks each day, followed by a panel discussion where the audience actively participated.

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2 Major outcomes

The topic of the first day was "Computational development of open quantum systems: Beyond Markovian master equations (MME)". The first talk was by prof. Klaus Molmer. He gave an in-depth review of the history and various applications of MMEs, especially in the field of quantum optics. He explained why the MMEs are useful even in many modern device applications of. The numerical methods to solve MMEs were also briefly discussed. The second talk was by Juan M.R. Parrondo, whose topic was wave-particle duality in quantum thermodynamics (QT). He discussed the benefits and disadvantages of using either particle of wave picture in QT. Following the talks there was an active panel discussion led by Ala-Nissila, who gave a summary of recent works on the range of validity of MMEs for open qubits, and the correlation picture of open quantum dynamics beyond MMEs.In addition to the other panel members (Molmer, Aurell, Chenu, Vinjanampathy) there was a long and interesting discussion with several audience members on MMEs. The main research direction in this field was identified as going beyond MMEs and developing efficient numerical tools that could be used to interpret experimental data and design new quantum devices.

The theme of Day 2 was "Quantum Thermodynamics (QT) -- what is it good for?". The first talk was by Eric Lutz (Stuttgart) on controlling the stability of thermal machines. The background to the topic is the trade-off which exists also classically between efficiency of a thermal machine (limited by Carnot formula) and power. Lutz reviewed recent developments and walked through both numerical examples and experimental results. The second talk was given by Geraldine Haack (Geneva) on the possibility of using nanoscale thermal machines to intrinsically accomplish quantum tasks: "Can we achieve Quantum Teleportation with Quantum Thermal Machines?", to be understood as if it is possible to create quantum entanglement as the work realized by quantum device driven by heat. Prof. Haack gave a pedagogic overview of quantum teleportation as a means of moving a quantum state from one location A to another location B, if the agents in A and B (Alice and Bob) share an entangled quantum state. The panel discussion included Ronnie Kozloff and Joachim Ankerhold. Prof Kozloff gave a mini overview of quantum thermodynamics as whole, supplementing the views presented by the speakers. Active partcipants included the panelists, the speakers, several of the workshop Organisers, Michele Campisi, Sai Vinjaynampathy, Berislav Buca, Viktor Holubec, Klaus Moelmer, Juan Parrondo, and others. It was concluded that QT is part of a new research frontier where new phenomena wait to be discovered. Geraldine Haack argued that quantum thermal machines can be used to produce purely quantum resources which do not exist classically. Finally, several participants pointed to the important technical problem of qubit reset for quantum computation.

The topic of Day 3 was "New directions" The focus in this session was mainly on quantum computing (QC) and related issues. The first talk was by Jacob Biamonte (Skolkova) entitled "Results on variational quantum circuits to minimize effective Hamiltonians". His group is constructing specific quantum circuits (to be mapped to quantum algorithms) to find ground states for a large class of Hamiltonians using quantum computers. This is an important development in QC related to specific physical systems. The second talk was by Michele Campisi (Pisa) on QT on quantum computers. He presented ways of connecting QC with related thermodynamic quantities in the quantum setting, and presented his recent results obtained by using state-of-the art quantum computers.

The panel discussion of Day 3 included A. Imparato, B. Kappen, B. Buca. In particular, B. Kappen and B. Buca gave each a short presentation on their work on and put their results in perspective of Day 3's topics. A lively discussion ensued.

3 Community needs

The open quantum system community has traditionally been very diverse for historical reasons. The field first emerged in the context of low-T experiments on quantum gases in the field of quantum optics, and also rather independently in solid-state physics where the focus was on elecronic transport. Since the 1990s there has been a huge expansion in the field and now most of low-T physics deals with (open) quantum systems and/or devices. There are well-developed codes for electronic structure and transport, but no systematic code development for open quantum dynamics based on master equations or other systematic approaches. The original idea of this workshop was to bring together many of the different communities working on similar problems, but this ambitious plan had to be toned down due to the pandemic. The two main issues in this field include cross-fertilization between the different fields where open systems are studied and exchange of methodologies and systematic development of numerical tools and codes to solve for quantum dynamics, e.g. based on various master equation approaches. There are several Python-based open-source codes for solving the (Markovian) Lindblad equation, but no systematic code development beyond this. One reason is that the community does not agree what is the most efficient and accurate way to model open quantum systems, and how to go beyond the Markovian limit. It is thus imperative that further workshops be organized in this field and the present ones are planning to submit a new application to realize this in the near future within the CECAM Flagship series.

4 Funding

The funding for open quantum systems and especially devices based on the is growing hugely, both on national and international levels. Due to the rapid development of quantum computing (QC) most EU countries have national initiatives to build up QC computing infrastructure (e.g. FiQCI in Finland which aims to couple quantum-classical computing within the European HPC network). The most recent drafts of the EU Horizon 2020 schemes have multiple programs related to both building up QC infrastructure and to facilitate large-scale QC for academic and commercial use. CECAM can play an important supportive role in the basic sciences related to QC and quantum devices by targeted workshops and events.

5 Will these developments bring societal benefits?

The second quantum revolution is expected to impact many aspects of our society, such as healthcare, the military, finance, cybersecurity, weather modelling, and much more. Harnessing quantum technology could, for instance, help us build computer systems that are unhackable. We could also rapidly solve complex science questions in high-energy or nuclear physics, create incredibly accurate quantum GPS, and solve complex optimisation challenges – such as in traffic congestion, logistics, and fraud detection. Harnessing quantum computing could also help us advance even further in artificial intelligence. Quantum technologies will likely transform the life sciences sector as well, to help us explore novel medical treatments and improve our health. Emerging quantum technologies could also transform the energy sector by optimising power grids and predicting the environmental effects of certain types of energy production. To realize all this, we need a thorough understanding how quantum mechanics works in action in real-life systems -- this is the essence of open quantum systems that are inevitably coupled to other quantum or classical systems (environments) that are needed for controlling them.

6 Participant list

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Combining multi-scale simulation and scattering for structural analysis of complex systems
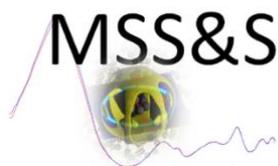
Location: on-line, hosted by CECAM-HQ
Webpage : https://www.cecam.org/workshop-details/144
Dates: Jan 26, 2022 - Jan 27, 2022

1 State of the art

Atomistic simulation is increasingly being used to interpret small angle and total scattering data in disordered and soft matter systems. The method allows introduction of known constraints (e.g. molecular geometry and interaction energies) into the structure determination and refinement and give as output an atomistically-detailed structural ensemble consistent with the data. Two key examples include Empirical Potential Structure Refinement (EPSR) [1] and its application to the interrogation of liquid and amorphous systems utilising total neutron and X-ray scattering data, and SASSIE for calculations of SAXS/SANS from biological molecules in solution [2]. These tools, and other similar methods [3] have yielded a large volume of very high-quality science in a wide number of fields. In particular, EPSR has transformed structure determination in molecular liquids and allowed the study of increasingly complex and varied systems [4,5,6].

The recent developments of wide Q-range neutron scattering instruments (e.g. NIMROD at ISIS (UK) and NOVA and TAIKAN at J-PARC(Japan)) and the enhanced availability of combined SAXS/WAXS at both synchrotrons and as lab sources, increasingly mean that both atomistic and mesoscale structures can be investigated simultaneously. Furthermore, there is a push to study even more complex and applied systems e.g. large surfactant micelles, polymer conjugates and heterogeneous systems. These trends mean that computational and methodological limits are frequently being met, particularly when both atomistic and mesoscale structure need to be understood together.

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2 Major outcomes

For large biological molecules in solution, simulation techniques have become established tools to generate a conformational ensemble to be compared to data, allowing a subset of structures to be selected as the most likely structures (e.g. SASSIE – Perkins and Kruger). Advances are being made in these techniques for more complex systems, such as Bayesian Inference, to most appropriately select and weight different conformers/molecules within a polydisperse solution. Many talks highlighted the considerable benefit on combining multiple sources of data with simulation (e.g. inclusion of NMR, cryo-EM). However, as highlighted by D. Scott, general tools for combining these simultaneously with simulation are not sufficiently advanced – key research problems include how to appropriately weight different data sets, account for experimental errors and difference in sampling between the experiment and the.

Where simulation does not agree with data, J. Hub and N. Rhys discussed the use of refinement methods on simulations, to push an atomistic simulation towards SAS and neutron diffraction data respectively, this also highlighted the great potential for scattering techniques to inform simulation, by helping to provide better force-fields.

For soft-matter systems, these types of combined computation/scattering approaches are already being used e.g. for polymers (T. Gkourmpis) and asphaltenes (M. Hoepfner). One "bottom-up" approach is to use the tools developed for understanding liquid structure through a combined use of neutron diffraction with isotopic substitution and EPSR. As shown by K. Edler, this has revealed impressive detail in micelle structure, particularly in counterion binding – however the system was "fitted to the experiment" using C10TAB, rather than larger more common surfactants due to computational constraints. Developments to increase the size of system that can be studied are underway e.g. H.B. Kolli and P. Carbone showed the use of coarse-grained simulation – improving the Q-range of data that can be simulated and sampled. Talks from A. Paul and A. Seddon demonstrated the need for these combined simulation and computational techniques to be further developed for important soft matter problems such as drug delivery and gel formation.

Porous materials are another area that are increasingly using scattering experiments to understand their structure. K. Stefanopoulos demonstrated the ability of this data to form important conclusions about confined CO_2 in idealised systems and applied (reservoir rock) systems. Computational approaches to assist in analysing the data from confined fluids (e.g. EPSR) are possible but to date have only been used on relatively simple systems. N. Skipper demonstrated this approach to understand interlayer structure in a vermiculite clay but noted difficulties in the approach particularly with regard appropriate simulation of components weakly weighted in the system (e.g. cations).

The meeting also highlighted other potential tools and methods of interest to the community: B. Shanks and M. Hoepfner discussed novel methods for potential refinement from scattering data - fitting to simple potentials, such as the Mie potentials, therefore allowing thermodynamic data to be predicted from scattering data for simple noble gas fluids. B. Capone demonstrated the use of scaling theories in predicting designs for macromolecular systems for absorption – linking theory, computation and experiment. V Kapil explored bridging the scales down to quantum behaviour, combining computational methods and deep inelastic neutron scattering to understand the deviation of the proton momentum distribution in water from classical Maxwell-Boltzmann behaviour.

3 Community needs

It is clear from the workshop contributions that access to neutron and X-ray facilities are vital to support the community through SAXS/SANS and diffraction and other spectroscopy experiments. Other national facilities for applied measurements, such as NMR were also highlighted – and as mentioned above, are vital for a multi-technique approach.

Existing codes that exploit the combined power of scattering and atomistic simulation are now mature and yielding important scientific outputs (e.g. SASSIE, EPSR). Development of new codes to interrogate more complex systems, and/or sensibly combine large datasets is underway but in need of further support.

It should be noted that many users of these codes are not experts in computational modelling and so resource is required to ensure the codes are made user friendly, appropriately documented and that suitable starting structures can be easily made. For the later this can be a big barrier to entry for soft matter systems.

Following this meeting, it is clear that there is a significant opportunity for cross-fertilisation of ideas and approaches across SANS/NS boundary and scattering/computation/other methods. This was presented both in terms of simulation helping understand scattering (e.g. through structure refinement) and in helping simulation through development of new force-fields.

The meeting was held virtually over 3 afternoons, and while there was a good level of discussion, the format could not offer the same level of in-depth discussion in small groups that would have occurred at a face-to-face meeting. It was a starting point but further opportunities for discussion would be of great benefit to this broad community.

4 Funding

In the UK funding for combined computational projects (CCP's) has been vital in this area. The jointly funded UK-US (NSF-EPSRC) CCP-SAS has been a great success in developing user-friendly codes for calculation of scattering for biological-systems – options to extend this to soft matter systems are being investigated. New calls, such as the EPSRC's "Software for Research Communities" also provide similar funding, but however are very oversubscribed.
Short term funding has been available through the facilities (notably the Ada Lovelace Centre at STFC) to continue to advance some of the progress made through CCP-SAS in the area of soft-matter.
Perhaps due to the virtual nature event not allowing prolonged, informal discussions the possibility of joint proposals through wider funding opportunities was not discussed in the meeting.

5 Will these developments bring societal benefits?

The combined use of scattering and simulation is being used in a wide range of different science areas; from understanding virus capsids to understanding the porous structure of interstellar ice, resulting in a diverse range of potential societal, industrial, economic and health benefits.
As outlined above the combined use of small-angle scattering and simulation to understand the structure of biomolecules in solution is well established and being utilised to understand structure and therefore function across a number of different systems. Further developments are in the combined use of additional experimental probes for more complex systems and the use of wide-angle scattering to understand structure and hydration at a molecular level.
The methods discussed in the meeting are also starting to yield important results in the field of soft-matter. Talks demonstrated the application of these methods to understanding personal product formulation (e.g. shampoos) and polymer structure. The application of methods to porous materials and confined fluids was shown to be of relevance to the understanding conventional energy reservoirs, CO_2 sequestration and also to storage of energy carriers (e.g. H_2) in confined media.

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From disordered biomolecular complexes to biological coacervates
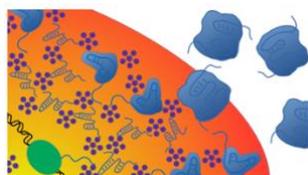
Location: CECAM-ETHZ, HIT building, Hönggerberg campus, Zurich, Switzerland & on-line (hybrid)
Webpage : https://www.cecam.org/workshop-details/1033
Dates: Mar 8, 2022 - Mar 11, 2022

1 State of the art

Recent work has increasingly demonstrated the importance of disorder in biology, exemplified by intrinsically disordered proteins (IDPs). Some IDPs may fold into ordered structures under certain conditions, but this workshop focused on the large class of IDPs or intrinsically disordered regions (IDRs) which perform their functions while remaining largely disordered, as well as their interactions with other biomolecules, in particular nucleic acids. Such proteins include intrinsically disordered linker sequences in large, multidomain proteins, disordered molecular chaperones, disordered proteins lining the nuclear pore complex, disordered protein complexes and those which form biological coacervates by liquid-liquid phase separation.

Understanding the fascinating properties of disordered proteins is a challenging problem, precisely because of their disordered state, which limits the applicable experimental techniques. In addition, for most experiments, the observables are averages over a very broad distribution of configurations, making their interpretation in terms of underlying molecular structure and dynamics a challenging inverse problem. Polymer theory is clearly an important part of the solution, but cannot easily account for the coexistence of both ordered and disordered regions in the protein. Molecular simulation is therefore a key tool for investigating such disordered proteins. Because of the large size of many of the individual proteins and especially for large assemblies and coacervates, the development of coarse- grained models is critical – and likely the most appropriate approach given that the forces driving these interactions are transient and non-specific in nature. Progress will require models with different resolutions, ranging from atomistic with implicit solvent to hyper coarse-grained approaches in which a protein is represented by a single bead, and possibly a multi-scale combination of these.

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2 Major outcomes

The final program including the titles of all presentations and posters illustrates the impressive list of world-class experts on the topic of disordered proteins and their interactions that presented and discussed their latest results. Posters enabled especially junior scientists to present their work to the participants, discuss them in depth and obtain valuable feedback on their work. After two years where hardly any in-person events could take place owing to the pandemic, this was for many of the on-site participants the first workshop or conference that again enabled the informal, open, and flexible discussions that are such an important driver of scientific discourse and exchange. In the intense and lively plenary discussions, the key outstanding questions and possible approaches were discussed both on the first and the last day of the workshop.

Among the topics that attracted particular attention were:

- measures of how to close the gap between biology and physics in our understanding of phase separation.
- measures of how to close the gap between molecular properties and mesoscopic behavior both in experiment and simulations.
- the use of multiscale simulation approaches to cover the vast range of length scales (atomic level to mesoscopic structures such as droplets or chromatin) and timescales (nanosecond chain dynamics to ageing processes on timescales of hours or days)
- the specific role of 'aging' from liquid to solid phase-separated droplets by gel and aggregate formation
- the possibility to obtain systematic experimental data under well-defined conditions for testing and parameterizing simulation force fields.
- the challenges of understanding processes at interfaces between dense and dilute phase.
- the role of posttranslational modifications and their representation in molecular or coarse-grained simulations
- the development of new experimental methods required for investigating the molecular properties of phase-separated systems that might be particularly informative for simulations.
- the role of evolution to rationalize the biological function of biomolecular condensate formation.

3 Community needs

From the very active participation in these and all other discussions, and from the extremely positive feedback we received, it was clear that such exchange was very helpful for focusing future work in the field and for enabling exchange between the different disciplines in this highly multidisciplinary field involving scientists from biology, chemistry, physics, and material science, using approaches that range from experiment to simulation and theory. We expect

that these interactions will help to bridge the gaps between disciplines, trigger new activities, and lead to new collaborations, especially between experimental and simulation/theory groups, to advance the field.

One thing that became clear during the meeting was that there had been a proliferation of new computational methods and theories—including coarse-grained methods at various resolutions—and that there was a need to finding ways of benchmarking and comparing these. This will only be possible with more standardized ways of comparing experiments and simulations and there were extensive discussions on this point. Future work requires the collection of more homogenous biophysics' datasets—expanding to other molecules than proteins—and measurements that go beyond simple single conformer properties. Benchmarking simulations against data on condensates and phase properties will require improvements in algorithms and simulation software and sustained access to HPC. By bringing together experts in simulation, theory, and experiments, the CECAM meeting is an excellent venue for discussing such efforts.

More broadly, in view of the enthusiastic response of the participants, it is very clear that another workshop on this topic should be planned. The organisers of the current workshop envisage future CECAM workshops in years alternating with the Gordon Research Conference on Intrinsically Disordered Proteins. We are thus likely to apply for a workshop to take place in 2025.

4 Funding

Funding for this topic at the national level is from the dominant funding agencies for basic and biomedical research, such as the NIH and the NSF in the US, the SNF in Switzerland, the DFG in Germany, the ANR in France, etc. Topics related to disease (e.g. ALS, FTD, cancer etc.) are also funded by disease-specific foundations. At the international level, an important source are grants within the Horizon 2020 framework (many of the speakers have been funded by ERC grants; other examples include COST initiatives and MSCA Doctoral Networks). Since much of the pioneering activity in the field takes place in the US, more international collaborative grants, such as the International Programs of the Swiss National Science Foundation, would be desirable.

5 Will these developments bring societal benefits?

Several aspects of this research field are likely to yield societal benefits. Of central importance is that an increasing number of disease-related processes are being discovered to involve intrinsically disordered proteins and/or liquid-liquid phase separation (LLPS), including neurodegenerative diseases, such as Alzheimer's disease, amyotrophic lateral sclerosis (ALS), frontotemporal dementia (FTD), and various forms of cancer. Being able to dissect the molecular mechanisms underlying these diseases has obvious biomedical implications and is likely to result in previously unrecognized drug targets. Especially for the latter aspect, the availability of simulation models and force fields that are suitable for describing the relevant interactions between different biomolecules, and the interactions between biomolecules and potential drugs will be essential for further progress of both fundamental understanding and biomedical applications.

At a more fundamental level, the stringent link and tight connection between theory, simulation and experiment that has been characteristic of the field of disordered proteins has over the years provided a key example on how interdisciplinary science can be done. Indeed, we are witnessing a "spill-over" of these ideas and methods to other fields including—for example—the study of biomolecular condensates more broadly.

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Recent Advances in Machine Learning Accelerated Molecular Dynamics
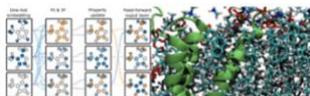
Location: CECAM-IT-SISSA-SNS: Scuola Normale Superiore, Italy
Webpage : https://www.cecam.org/workshop-details/1063
Dates: Mar 16, 2022 - Mar 18, 2022

1 State of the art

Computer simulation with molecular dynamics (MD) acts as a bridge between microscopic models and macroscopic phenomena. Machine learning (ML) - an emerging data-driven approach in this context - can provide new impetus and accelerate MD simulations to tackle new challenges in both method developments and applications.

Traditionally, force fields behind MD simulations in biophysics and materials chemistry applications were constructed using simple functional forms, which do not always properly describe complex chemical environments such as, for example, metal sites in enzymes or (electro)chemical interfaces. In this context, ML-based reactive force fields (or potentials) are now emerging as a promising alternative approach, with their ability to give quantum mechanical accuracy without explicitly including the electronic degrees of freedom. This allows simulations of a range of relevant systems using time and length scales that are unfeasible today, particularly when combined with enhanced sampling methods such as metadynamics and Markov state models.

The success of ML relies on the availability of data and knowledge of how to design a dataset that is representative yet small enough. Thus, the synergy between sampling methods, such as the Markov state model, metadynamics, and ML techniques, such as active learning, may stimulate new ideas. ML methods that can also provide property predictions in both compositional and conformational spaces are urgently needed. In turn, such methods should also allow posterior data analytics for exploring correlations and discovering new descriptors in real-world applications.

Thus, this workshop aims to bring scientists from MD simulation and ML backgrounds within areas of biophysics and material science under the same roof to spark discussions and new ideas.

2 Major outcomes

The scientific presentations and discussions during our CECAM workshop focused on the different computational approaches, i.e. the theoretical and computational developments, as well as the applications in both materials' science and biophysics. The applied computational methodologies were discussed in depth during the workshop and ranged from quantum mechanics (QM) and enhanced molecular dynamics (MD) sampling techniques to coarse-grained (CG) molecular dynamics accelerated by machine learning approaches. In particular, we had 19 talks on machine learning potential, 10 talks on enhanced sampling, and 7 talks on high-throughput screening, drug and electrolyte designs as well as on analyzing MD trajectories. In terms of the application areas, the number of talks was equally divided between biophysics (18 talks) and materials science (18 talks)

Regarding the development and the future of machine learning potential, Gabor Csanyi provided a perspective at the end of his talk, which stimulated a number of interesting follow-up questions. This includes how to describe radicals which might need more electronic information, how to generate a generic organic force field that outperforms the existing

empirical force fields used in the biophysics community, how to come up with new strategies to have uniform prediction accuracy for both short-range and long-range interactions, and how to identify proper local descriptors to address the charge self-consistency challenge encountered in materials modelling. This resonates with the views on the 5th generation machine learning potential from Joerg Behler and Michelle Ceriotti, in which both believe that the next generation machine learning potential will not only predict potential energy surface but also electronic properties. When it comes to enhanced sampling, this workshop also brought up two complementary perspectives. Michele Parrinello highlighted the importance of machine learning in identifying better collective variables that can be used in conjugation with free energy calculations, while Frank Noe showed how the Boltzmann generator can be used to obtain the probability distribution directly without explicit MD simulations that reveal the power of generative ML models.

Overall, the mix of biophysicists and materials scientists in this workshop turned out to work quite well because the topics are interrelated and method-oriented. A cross-disciplinary and forward-looking perspective on ML accelerated MD simulations can be considered as the main outcome.

3 Community needs

The workshop clearly showed very promising and intriguing possibilities for the field in the future. The rapid development in graphics processing units and new opportunities from the field of big data with the emerging power of machine learning tools allow for unprecedented possibilities that, however, will also yield new challenges and needs for our computational community. One of the main needs will remain the shareability and accessibility of data throughout the community. One ultimate goal is to optimize the reuse of data to ensure efficient collaboration and transfer of knowledge throughout the scientific communities. In the future, we should aim to organize more interdisciplinary workshops like this one to bring both biophysicists and material scientists under the same roof and to invite other communities such as computer science and engineering sciences to join. Additional needs of the community identified during the meeting show the shared view about the necessity to receive more direct support from High-Performance Computing centers at both the National and EU levels (e.g. the HPC Europa projects), especially for helping researchers in computational development activities towards more efficient methods and models.

4 Funding

The meeting nurtured very active discussions between researchers from different EU countries as well as the US on state-of-the-art materials and molecular modelling techniques. This naturally invites future exchanges and collaborations between the participants. During the exchange and networking (guaranteed through the organized lunches, coffee breaks, and social events), discussions on applying for EU funding have been also considered. We believe that follow-up workshops, seminars, and scientific collaborations, as well as grant applications and student exchanges, are going to be facilitated through the newly established and enforced cooperation network that originated from the meeting.

5 Will these developments bring societal benefits?

Materials and molecular modelling can certainly bring societal benefits by identifying novel targets in material (e.g. for a new material design in all-solid-state batteries) and drug design (e.g. in a responsive mode as was shown to be necessary for the Covid-19 pandemic).

This is also in alignment with the sustainable development goals (SDGs) set by the United Nations. For instance, materials modeling of metal oxide-based nanoparticles can accelerate the development and design of new energy storage materials and water purifiers, which support fully SDG 7 and 6 on "affordable and clean energy" and "clean water and sanitation" respectively. Similarly, molecular modeling of membrane protein and drug-like molecules for curing malaria will help to meet SDG 3 on "good health and well-being". Apart from the impacts led by research activities themselves on materials and drug discovery, our workshop on materials and molecular modelling also made positive contributions to achieving SDG 4 "quality education" and 5 "Gender equality". In particular, our workshop included a significant number of participants who are young PhD students and postdocs as shown by 6 contributed talks from a pool of enthusiastic candidates. On a similar note, our workshop featured 14 talks from female scientists, which is about 40% of the total number of talks. By promoting female participation in our research area, we are also working towards better gender equality and diversity and setting a better example for the next generation.

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Multiscale modelling of irradiation-driven processes for emerging technologies
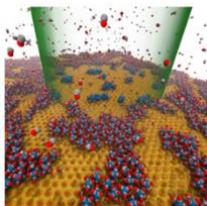
Location: CECAM-HQ-EPFL, Lausanne, Switzerland & on-line (hybrid format)
Webpage : https://www.cecam.org/workshop-details/31
Dates: Mar 16, 2022 - Mar 18, 2022

1 State of the art

Many modern and emerging technologies exploit irradiation of molecular systems which results in alteration in both the systems' structure and morphology and hence changes in their electronic, mechanical & catalytic properties on both the nano- and the microscale [1-4]. The interaction of radiation with molecular systems initiates quantum processes (e.g. electronic excitation, ionization, radiation-induced fragmentation), electronic and thermal relaxation of the deposited energy as well as production of secondary species, their subsequent transport and the irradiation-driven chemistry (IDC). These processes are followed by further transformations resulting in the chemical equilibrium in the system. The complex cascade of IDC processes takes place on different time & spatial scales and its understanding must be based on a multiscale approach that treats the entire multistage scenario in a consistent manner. Such an approach must combine discrete theoretical and experimental methods into a coherent framework whilst going beyond the limitations of the particular methods [5,6]. This can be achieved by means of novel computational methods such as Irradiation Driven Molecular Dynamics [7], which provides a link between Molecular Dynamics and Monte Carlo methods for the efficient simulations of IDC of complex molecular systems exposed to radiation [7,8]. The resolution of the aforementioned challenge in the field requires strong synergies of the experts in computational modeling with different background, experimentalists and technologists, capable of validating the outcomes of computational modeling and building closer ties to the concrete technological applications. The Flagship Workshop aimed to establish such synergies for three technology-related case studies suitable for the exploitation of a multiscale approach, namely (i) surface deposition techniques, nanofabrication & nanolithography [4], (ii) 3D nanoprinting [9,10], and (iii) novel light sources [11,12].

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2 Major outcomes

The scientific discussions held during the Workshop highlighted the importance of developing consistent multiscale modeling methodologies and novel computational methods which enable us to go beyond the limitations of the particular existing methods (such as TDDFT, classical MD, or track-structure Monte Carlo methods). Illustrative examples highlighted in the Workshop are Stochastic Dynamics (SD) [13], Reactive and Irradiation-Driven Molecular Dynamics (RMD & IDMD) [7,14] and relativistic MD [15,16] implemented in the advanced software package MBN Explorer [17]. These approaches enable efficient simulations of the structure and dynamics of various condensed matter systems (ranging from atomic clusters and nanoparticles to nanostructures to macroscopically large crystals, etc.) exposed to different radiation modalities (such as X-rays, electrons and ion beams) in connection to the selected technological applications discussed in the Workshop [4,9-12]. These computational developments represent an important example of continuity of research efforts in the community, which has become possible through the continuous series of funding through different European and national projects.

The second major outcome concerns a strong need for close links between modeling and experiment [18-25]. Such interlinks, on one hand, provide relevant input parameters for atomistic simulations and, on the other hand, enable the full validation of the methodologies on a macroscopic, technologically relevant scale. Particular examples highlighted in the Workshop are recent breakthroughs in the atomistic modeling of the Focused Electron Beam Induced Deposition (FEBID) [4] process by means of IDMD [7]. A recently developed generic model for the atomistic simulation of FEBID [7] enables the interpretation of many different experimental results and their predictability [8,26].

The third major scientific outcome concerns the important role of computational multiscale modeling in connection to the development of future crystal-based light sources with photon energies of MeV and above [11,12], based on the concept of charged particle channeling [15,16,24,25]. State-of-the-art multiscale modeling approaches permit the complete atomistic-level characterization of materials exposed to such beams and the description of the underlying physical phenomena [16].

A common suggestion was the need for a comprehensive set of databases [27-30] containing reviewed and recommended data (e.g. collisional cross sections, rate constants, force field parameters).

Finally, it was emphasized that the presented computational advances are not limited to the particular technological areas presented in the Workshop, but can be applied to other challenges, e.g. the investigation of irradiation-driven processes in space [31,32], effects of radiation in biological systems [1-3] or radiation-induced nanocatalysis [33].

3 Community needs

The meeting confirmed the ongoing development of an active community engaged in multiscale modeling of irradiation-driven processes and demonstrated increasing links between academics and different communities that apply Multiscale Modeling (MM) to a wide range of technological and scientific applications. This community necessarily includes the experimental community which provides validation of the multiscale models.

In building a community to both develop and exploit MM it is necessary to engage researchers exploring the interaction of different types of radiation with atoms and molecules in different phases. These interactions lead to the production of secondary products that subsequently transform the local environment by physical changes (e.g. DNA rupture in cells) or chemical changes by local reactions (e.g. on a surface). Computational models of collisional processes must be integrated with models of the transport of the secondary species produced in the irradiation process. Such transformations engage experts drawn from the condensed matter community and exploit existing tools but complement these with models describing Reactive and Irradiation-Driven Molecular Dynamics and Stochastic Dynamics. Only a complete MM approach, within which all of these processes and models are combined, will provide the detailed simulations necessary for accurate replication of the processes discussed in the meeting. Therefore, the need to build such an interdisciplinary and intersectorial community to advance MM of irradiation-driven processes was apparent throughout the meeting. The award of COST Action CA20129 "MultIChem" provides an opportunity for staff exchanges to create networks that address the challenges and needs of the MM community. During the meeting delegates agreed to prepare a 'Roadmap' that will provide an immediate *modus operandi*, creating a deeper sense of community whilst simultaneously defining the future direction of the research.

4 Funding

Funding is a composite of National and International funding. Much of the research is conducted in universities with notable exception of MBN Research Center, a SME company developing the MBN Explorer software package for the advanced MM simulations which is purchased by many user groups. National funding supports temporary researchers (PhDs & Postdoctoral researchers) and most experimental work used to validate and test MM. European programs provide funding for collaborative projects, networks and conferences. Successful awards in MM include COST action CA20129 - Multiscale Irradiation and Chemistry Driven Processes and Related Technologies (Oct. 2021) and 2 Horizon 2020 projects RADON and N-Light support academic-industry interchange and direct applications of MM. TECHNO-CLS a 5-year Horizon Europe project developing new light sources using MM begins in June 2022. Thus, core of funding is in place and the community is proactive in applying to diverse range of funding schemes.

5 Will these developments bring societal benefits?

The workshop entitled 'Multiscale modelling of irradiation-driven processes for emerging technologies' discussed the application of MM to a range of technical and scientific applications. Three areas of direct social benefit were highlighted: (i) The design of nanoparticle radiosensitizers for exploitation in radiotherapy; (ii) The optimization and operational control of FEBID to develop a new methodology for fabrication of nanostructures, and (iii) The design of new high intensity and high energy crystal-based light sources using leptonic transport through oriented crystals.

In each case MM is essential for the design of novel instrumentation as well as providing detailed knowledge of the underpinning science. In each case major industrial exploitation of the knowledge will lead to wealth creation and employment whilst simultaneously providing new products for societal benefit. The most immediate societal benefit probably lies in the design of nanoparticles to act as radiosensitizers and the use of MM in design of treatment planning protocols for next generation radiotherapy (e.g. ion beam cancer therapy).

The meeting identified several new thematic areas where MM can advance technologies beneficial to Society including irradiation processing of spaceborne materials, environmental remediation and plasma technologies. The desire to exploit space as a resource for human development in the next decade is predicted to be a multibillion industry but requires a fuller understanding of the radiation environment in space for which the tools and expertise presented at this meeting are very appropriate. Similarly, the MM of plasma systems is a natural extension of present studies with a wide range of applications for society from manufacture of products to human health and environmental control (e.g. waste water treatment).

Finally, the commercial exploitation of MM software has already been demonstrated e.g. by MBN Research Center GmbH.

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Understanding plastic deformation via Artificial Intelligence

Location: on-line, hosted by CECAM-HQ
Webpage : https://www.cecam.org/workshop-details/56
Dates: Mar 28, 2022 - Mar 30, 2022

1 State of the art

Plastic deformation of solids is a paradigmatic problem for multiscale materials modeling. Relevant processes range from the atomistic scale where the detailed atomic arrangement of a material is of crucial importance for its deformation properties, up to macroscopic scales where deformation instabilities manifest themselves in the form of various macroscopic plastic localisation and oscillation phenomena. However, the crucial question of how the properties of defects and microstructure of the material link to the macroscopic constitutive equations of continuum mechanics is still open. Homogenisation, where the transition from discrete defects (dislocations in the case of crystal plasticity) to an appropriate "smooth" continuum description is assumed to be reached by simple averaging of the dynamics of individual defects, has not been accomplished.

The use of AI methods has recently exploded in two relevant fields for the proposed workshop: materials science and statistical physics. Computational materials science allows – compared to most experiments – to pose well-defined problems for AI applications in the presence of large enough datasets for the typical AI paradigm of teaching algorithms with sufficient data. Much of this effort is on the atomistic level, both for short-cutting heavy calculations via learning faster potentials and for the sake of classifying and predicting novel compounds. Likewise, the classification problem is close to one of the key questions of both equilibrium and non-equilibrium statistical physics: can we work from the data and establish from that the whole phase diagram of a system. This can be tried in various contexts from models (classical and quantum) and in and out of equilibrium systems like for instance for active matter. The progress in this direction itself would seem to encourage us to try seriously to understand plasticity by the toolbox of AI.

2 Major outcomes

As suggested by the title of the workshop, the main discussions focused on the overarching problem of applying artificial intelligence methods such as machine learning to make progress in understanding plastic deformation. In terms of the studied systems or materials undergoing plastic deformation, the scope was quite broad, covering not only the "usual" problems of plasticity of crystalline and amorphous solids, but also for instance metamaterials and chains of bi-stable springs.

In terms of the AI/machine learning methods discussed in the context of the above-mentioned plastically deforming systems, the scope was similarly broad, including but not limited to ordinary and convolutional neural networks (deep learning), reinforcement learning, etc. In addition, there were interesting discussions related to, e.g., interpretability of the predictions of machine learning algorithms in the context of amorphous plasticity, as well as to the importance of definition of the task when applying machine learning in plasticity and beyond. In addition to discussing studies where AI/machine learning was applied to study plasticity, the program also contained talks pointing out possibilities to do so in the future.

Due to uncertainties related to COVID-19, the workshop was, after initially postponing it, organized online on Zoom. While this was not ideal from the point of view of facilitating interactions and discussions among the participants, we still feel it was important to organize the event anyway, even if the online format certainly has its drawbacks. This is because the topic of the workshop is very timely and fast developing, and at least the Organisers thought the online workshop provided a valuable venue for getting an update on the current developments in the field. The online format also made it possible to have an audience which likely exceeded in size what we would have had at an on-site workshop – at best there were around 50 simultaneous participants in the Zoom meeting, and for most of the time the number of people in the meeting was more than 30. Having said that, the online workshop also highlighted the need to organize an on-site, physical workshop on a similar topic in the near future.

3 Community needs

Application of artificial intelligence and machine learning to understand plastic deformation is a new and quickly evolving field. Its emergence and growth are obviously linked to the recent and current overall trend in almost every sector of our society experiencing explosive growth in application of artificial intelligence and machine learning, fueled by the combination of increasing availability of large data sets and rapid development of hardware suitable for, e.g., training increasingly complex machine learning models. Hence, there is clearly a need for a forum where the physics/materials science community would be able to stay up to date regarding new developments in this rapidly evolving field. In the broader field of materials informatics operating at the interface of materials science and informatics methods such as AI and machine learning, examples include, e.g., the 1st NOMATEN International Conference on Materials Informatics. On the other hand, for the community of people working at the interface between AI and plasticity highly specialized and smaller workshops (such as a CECAM Flagship Workshop) with up to 30-40 participants might be something that is well appreciated by the community also in the near future.

4 Funding

As is the case for any field of research, further advancing the study of AI and plastic deformation relies on the availability of sufficient funding. In addition to the various and quite scattered national funding schemes, instruments under Horizon Europe may offer interesting possibilities. In addition to individual grants such as those offered by European Research Council (ERC), Horizon Europe encompasses instruments for Europe-wide collaboration, something that would likely be highly beneficial for advancing the topic of the workshop. Out of these, for instance applying for a Marie Curie Innovative Training Network (ITN) with a topic related to application of artificial intelligence/machine learning to understand plastic deformation could be a viable option in the near future.

5 Will these developments bring societal benefits?

The talks and related discussion during the workshop not only provided nice examples of artificial intelligence and machine learning applied to the problem of plastic deformation of materials but also emphasized the technological importance of the main aims of the workshop, that is, predicting plastic deformation and studying optimization problems with the aim of discovering novel materials with desired properties. A key aim of the more applied branch of research on the topic of the workshop is to improve the accuracy and interpretability of, e.g., the machine learning models applied, something that could in the longer term allow to more precisely design various engineering applications. Such advances should contribute to more reliable (and thus safer) components and structures in engineering applications, which may be, thanks to the improved accuracy, also designed to be lighter and thus more energy efficient. Hence one can conclude that research work discussed during the workshop has great potential for medium- and long-term societal benefits.

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Machine Learning for Quantum Many-Body Physics
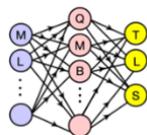
Location: CECAM-FR-GSO, Toulouse
Webpage : https://www.cecam.org/workshop-details/1045
Dates: Apr 4, 2022 - Apr 8, 2022

1 State of the art

The flagship school organized in Toulouse was dedicated to machine learning approaches for quantum many-body physics and chemistry. The quantum many-body problem is crucial to predict and understand quantum material properties and it is known to be a very hard problem to solve numerically due to the exponentially large number of configurations. Recently, artificial intelligence tools and algorithms have been applied in all fields of science and beyond. Quite naturally but not straightforwardly, some of these ideas were applied to the quantum many-body field. Let us mention for instance successes in (i) using image recognition to sort quantum phases of matter or detect quantum phase transition; (ii) performing a quantum state reconstruction (tomography) using measurements outcomes of experiments; (iii) performing a variational optimization for the ground-state of a quantum hamiltonian using neural networks inspired quantum wavefunctions.
After an introduction to machine learning within the quantum physics context, all these topics (and more) were introduced in the school.

2 Major outcomes

Since this was a school targeting young PhD or postdoc researchers, there were mostly lectures and tutorials. The lectures were quite exhaustive and did not require any prior knowledge. The speakers were able to present major ideas in artificial intelligence (learning, supervised/unsupervised learning, kernel methods etc.) as well as many state-of-the-art applications in quantum physics and chemistry.
More precisely, the lectures were:
- 2 lectures by. Florian Marquardt on Introduction to Machine Learning, Neural Networks, and Reinforcement Learning (with a focus on quantum systems for the later)
- 2 Lectures by Evert van Nieuwenburg on Supervised / Unsupervised Learning and Learning phase transitions by confusion
- 2 lectures by Matthias Rupp on Kernel methods, and their applications in atomistic models
- 2 lectures by Giuseppe Carleo on neural quantum states, and their application to learn ground-states and quantum dynamics
- 1 lecture by Stefanie Czischek on quantum tomography with neural networks
- 1 lecture / tutorial by Mohamed Hibat-Allah on Recurrent Neural Networks and application to learn ground-states of quantum systems
We also had a few specialized seminars on spiking and non-spiking neuromorphic hardware, open quantum systems with neural quantum states, generative models for Rydberg arrays, quantum spin liquids and neural quantum states, quantum error correction with machine learning.
Moreover, there were also tutorials were very much appreciated since they allowed to make real computations quickly using standard libraries in the field: introduction to Jax and automatic differentiation, Varitional Monte Carlo to learn ground-states (Netket library), quantum dynamics with Netket.
Finally, we had a lively poster session as well as a session with short talks by participants.

We have used a Slack workspace to allow people to ask questions and download the required instructions, which was quite efficient.

In the last session, we summarized the key ideas in the field and the open problems, and also checked what did the students learn in the form of a fun quiz.

3 Community needs

This was a very broad community ranging from solid state physics to quantum chemistry, including also more fundamental quantum information researchers or statistical mechanics student. But precisely, the artificial intelligence represents a novel framework with lots of potential applications in all fields. In the future, it is clear that more libraries will be written (using developments from more fundamental computing science) and the best performance will be obtained using HPC resources. Already now, the major libraries take advantage of the GPU acceleration. Our school was mostly aiming at theoreticians but it seems to us that applications could also be envisioned for experimentalists (data sorting, protocol optimization etc.) which could be done in the future. This was a school intended for beginners in the field, which gathered a large number of students. We feel that, in part thanks to this school and others, there is a new community on the topic of machine learning in quantum physics that is emerging with a large momentum. It is clear that this community will gather around specialized workshops, as recently organized for instance by the Netket developers (located in Lausanne), and we believe CECAM should have many opportunities to support these meetings and structure this new community.

4 Funding

Given the current interest in quantum technologies, it should be possible to get funding from different sources (French national "Plan quantique", regional "Institut Quantique Occitan" etc.). Indeed, the near-term Noisy Intermediate-Scale Quantum devices (NISQ) require quantum simulations to predict outcomes and some novel algorithms could be the ones inspired from machine learning.

While we have not directly pushed towards a discussion of research proposals during the meeting (this was a school after all), it is certain that the many discussions between junior and more senior participants will lead in the long term to fruitful collaborations.

5 Will these developments bring societal benefits?

Quantum materials are the cornerstones of many near-future applications including new computing chips, energy or quantum computing platforms. The success of these future applications depends heavily on a detailed understanding of the microscopic quantum phenomena at play. Unfortunately, the quantum advantage ultimately emanating from the entanglement between microscopic degrees of freedom comes with an exponential complexity when one needs to perform accurate modelling of quantum materials.

Hence it is crucial to develop and improve computational techniques for such complex systems and unveil the microscopic mechanisms in quantum devices or materials.

The use of machine learning techniques is a new apparatus in the toolbox to perform better simulations of such complex quantum many-body systems and fulfil this goal.

Let us also note that machine learning marries well with quantum computing, for instance to optimize the quantum gates or to post-treat data from quantum computers in a feedback loop.

Our school allowed many young researchers to have a direct and practical introduction to the field. Several of these students will for sure be involved in quantum technologies within academic or industrial settings, increasing the potential to make disruptive changes for economy and society.

Ion adsorption and electrokinetic transport at interfaces
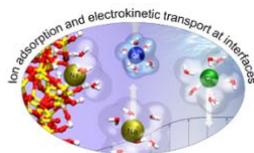
Location: CECAM-FR-GSO, Marcoule
Webpage: https://www.cecam.org/workshop-details/1141
Dates: May 4, 2022 - May 6, 2022

1 State of the art

The field of charged solid-liquid interfaces is of particular interest in physical chemistry and material science because it concerns many scientific fields and applications: electrochemistry, biology, geology, pollution control, formulation, etc. Two important phenomena are essential to any description of these systems: the adsorption of ions on the surface, which drives in particular the electrical properties of the double layer, and the fluid transport that results from the external and internal forces of the medium. Electrokinetic phenomena, such as electroosmosis or electrophoresis, are particularly intriguing examples of a macroscopic motion originating from the interfacial charge distribution and fluid dynamics, allowing the design of controlled nanofluidic devices. They are also very much used by the engineering sciences because they allow the measurement of global characteristic quantities like the zeta potential, which can be directly related to experiments. Despite their practical interest, these quantities are interpreted with effective models, which idealize the molecular reality.
Since the original works of Gouy, Chapman, and Smoluchowski, the common description of these systems relies on continuous solvent theories that are not well justified microscopically. In this context, in the last few years, many molecular studies have tried to justify and improve these descriptions. In parallel to these dynamic studies, many works on equilibrium were interested in the specific ion effects in particular at the interfaces. The goal of the workshop was to define, in the light of these recent works, a program to establish a truly modern theory of ion adsorption and fluidic transport at interfaces, bringing together theoreticians that develop models at different scales and different methods.

2 Major outcomes

A central theme throughout the meeting was the ambiguous definition of the zeta potential. We have discussed multiple ways to calculate this property from simulations and to determine it from electrokinetic experiments, often with contradicting results. Some of the methods that we have discussed relied on the assumption of adjacent stagnant and fully mobile fluid layers whereas others considered a local viscosity and yet others used the local mobility of individual components. Although this issue is far from being solved, we have agreed that there is no clear separation between a stagnant and a mobile layer, the mobilities of all components change continuously, nor is viscosity truly a local property.

Molecular simulations, alongside experiments, prove to be instrumental in finding the relation between ion adsorption and electrokinetic transport both from the theoretical point of view as well as applications to particular systems.

A comparison of several presentations also revealed using the term 'surface potential' in different contexts. While the electrochemical community rather interprets this term as the difference of electrochemical potential between the solid electrode and bulk liquid, some presentations rather used this term to describe the properties of a solid surface including the strongly adsorbed immobile layer of ions and solvent. Therefore, care must be paid to avoid misinterpretation of the published data.

The meeting has also highlighted that the currently available force fields are not optimal for accurately predicting ion adsorption and transport. Indeed, while truly impressive force fields have been developed in the recent years to describe aqueous electrolytes in bulk, based on ionic charge rescaling (the so-called electronic continuum correction), as exemplified by the Madrid force field, describing liquid-solid interfaces remains a challenge. While we agreed that the mixing rules approach to determine liquid-solid interaction parameters can at best provide a fair approximation, the cost of determining interaction parameters for each specific liquid-solid interface is prohibitive. Accurate force fields could nevertheless be developed for a few selected interfaces of particular interest. A radical alternative would be to switch to ab initio simulations, or to ab initio-based machine learning potentials. However, although in principle ab initio simulations describe matter at a more fundamental level, they also include some degree of empiricism, e.g. the choice of the exchange-correlation functional in density functional theory. We concluded that more work is urgently needed to address this matter.

Another aspect that was identified in relation to differences between simulation studies and experiments is the fact that many simulations are based on infinite pores with an atomic roughness, whereas transport through real pores might be strongly influenced by entrance effects and macroscopic roughness. Moving forward, this is something to keep in mind when interpreting experiments and comparing them to simulations, and also when setting up a simulation system. Simulations of finite pore lengths also exist, but these might be in fact so short that no fully developed flow forms, making it difficult to decouple the entrance/exit effects from other phenomena.

Future work will possibly try to distinguish the relevant concept to interpret molecular dynamics simulations and experimental measurements on electrokinetic phenomena, including those presented in the workshop.

3 Community needs

The available resources vary strongly between countries and even universities and departments. Overall, the meeting has not specifically indicated any limitation in computational resources. In terms of codes, a large part of the research presented during the workshop was based on molecular dynamics simulations, for which existing codes (e.g. LAMMPS, Gromacs, CP2K, VASP) were mostly used. One of the themes throughout the meeting was that coupling between simulations and experiments is key to advancing this field. At the same time, we acknowledged that the computational and experimental community often used different nomenclature and had limited knowledge of the possibilities and limitations that the other community was dealing with. This indicates a need for more events like the one that we organized and more networking between computational and experimental research in general. Some of the participants have strongly indicated that they would like our meeting to be a recurring event, which is a possibility that we have also considered among the Organisers.

4 Funding

The possibility of joint research proposals was not explicitly discussed during the meeting, although multiple opportunities for collaborations were identified on an individual basis. One funding instrument that would be suitable is the ERC Synergy grant. Many other funding instruments, especially those within Horizon Europe, are more focused on specific applications and the development, integration, and upscaling of corresponding devices than on more fundamental research. As such, various funding instruments could be suitable, depending on which application we would focus on.

5 Will these developments bring societal benefits?

The topic of the meeting has indirect far-reaching economic and social benefits since the insight that we provide is important for the design of optimal nanofluidic (i.e., lab-on-a-chip) devices, membranes, porous electrodes, electrochemical processes, and colloidal suspensions (for example in the foods industry, paints, and cement paste). Some of the applications that were specifically discussed during the meeting included the removal and long-term storage of radioactive materials in relation to the aging of materials as well as the electrochemical removal of ions from water (water purification/desalination). An exciting middle-term benefit discussed during the workshop concerns new methods for sustainable energy harvesting based on aqueous electrolytes at interfaces and in confinement. A prototypical example is the harvesting of the so-called blue energy (i.e. the energy that arises from the osmotic force between seawater and fresh water) with nanoporous membranes; the underlying phenomena (diffusio-osmosis, reverse electro-dialysis, etc.) share many similarities with the standard electrokinetic effects discussed during the workshop, and their description faces the same challenges.

6 Participant list

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Young Researcher's Workshop on Machine Learning for Materials

Location: CECAM-IT-SISSA, Miramare Campus, Italy
Webpage: https://www.cecam.org/workshop-details/1150
Dates: May 9, 2022 - May 13, 2022

1 State of the art

Data-driven methods have emerged as a novel paradigm to advance materials discovery over the past decade. Machine learning potentials (MLPs) enable the sampling of trajectories with the same accuracy of high-level electronic structure methods but at a fraction of their cost. MLPs have been established as a means to rationalize puzzles previously unapproachable by atomistic simulations. Elsewhere, the chemical and physical properties of large chemical spaces are now screened in a high-throughput fashion by leveraging artificial intelligence methods, materials simulations, and automation protocols. The screening is not only viable for the case of known structures, but generative models can now autonomously generate previously-unseen, and tailored, molecules and crystals structures with a target property. Machine learning (ML) methods therefore serve as formidable surrogates to accelerate expensive computational screening, but also to guide experimental screening and extract

knowledge from data gathered via high-throughput or from literature. Furthermore, the advances in the theoretical understanding of how machine learning algorithms work is demystifying and surpassing the vision of data-driven approaches as magic black-boxes.

This event built upon the state-of-the-art in the field of machine learning for materials in two ways. Firstly, it helped instruct the next generation of young researchers on the latest advancements in methods and applications of AI for material discovery through didactic lectures and hands-on tutorials. Secondly, the workshop promoted a discussion on the implications of the latest advancements in data-driven methods on the different sub-areas of Materials discovery, bringing together experts of different fields in the world of machine learning for materials and promoting cross-contamination of ideas and techniques.

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2 Major outcomes

The introductory part of the workshop paved the way to an overview of supervised and unsupervised methods which well represented the in-machine learning for materials. A number of tutorials on publicly available open source and documented codes were offered to participants.

Many discussions during the workshop focused on the design of atomic descriptors for supervised tasks in materials science. Two techniques were highlighted by various speakers as top performers: atom-density (e.g., atomic cluster expansion (ACE)) representation, and equivariant learnt representations via message-passing networks (MPE(3)N). Both methods efficiently encode information about local atomic environments and allow for very accurate learning of atomic or structural properties (e.g., forces, energies, etc.). A discussion of a unified theory to reconcile the dichotomy between ACE and MPE(3)N was a recurring trend across invited and contributed speakers. State-of-the-art Pareto fronts of prediction speed-accuracy, and an analysis on the memory and time requirements for training were also often reported.

A second set of common themes and techniques related to the use of generative models. Their application ranged across disciplines: SMILES-based short-term memory recurrent neural networks were used to design drugs; cartesian coordinates and autoencoders were utilized to unbiasedly obtain equivariant representation for quantitative structure-activity relationships; classical descriptors and variational autoencoders were adopted to map states during dynamics and/or glass phenotypes.

A third set of common themes related to the use of machine learning methods to accelerate the first principles screening of material stability. The application of this method ranges from energy materials (e.g., perovskites) to molecular crystals (e.g., drugs) and leveraged

uncertainty-driven methods to iteratively and accurately chart convex-hulls and establish thermodynamically stable phases.

A final class of major scientific points of discussion encompasses a broader spectrum of topics which enables to bridge complexity gaps between data models and experiments, as so to establish rational design paradigms from structure-property relationships. A heterogeneous list of techniques debated includes (but is not limited to): machine learning potentials for fast-and-accurate simulation of complex dynamics; transfer learning to derive universal predictors which work well across different chemistries; experimental characterization and manipulation of materials via data-driven optimization.

Overall, the need for cross-contamination of expertise emerged as a strong and resonant topic throughout the conference. During panel discussions, presentations, and face-to-face interactions, participants expressed the need to escape scientific bubbles and gather information about techniques, applications, and developments in fields adjacent to their own research. In this regard the presence of leaders in atomistic modeling, computer science, machine learning, experimentalists, and industry representatives enabled an interdisciplinary exchange of perspectives and experiences.

Our workshop was indeed specifically designed to address the needs for multi-disciplinary cross-contamination, and we received resounding feedback about how such an effort was successful. All (to our knowledge, and according to a currently ongoing survey) participants to the workshop, be it an invited speaker, an online attendee, a poster presenter, or a young researcher that attended their first conference on the topic, was largely positive about the structure, topics, and organization of the event.

Finally, all talks, tutorials, and panel discussions that took place during the workshop have been recorded and uploaded on YouTube, the ICTP website, and the conference's website, thus making high-quality scientific content available to anyone.

3 Community needs

The workshop allowed researchers in adjacent fields to meet each other, learn about the most recent advances of their colleagues, and network in a scientifically fertile environment. Moreover, the presence of an introductory school in the workshop allowed for young researchers that are new to the field to learn about potential applications of machine learning technologies to their area of interest, and to better appreciate the advancements presented by invited leading researchers during the "workshop" part of the conference. From the networking perspective, the event north star was indeed to enable the cross-fertilization of research networks, promoting the encounter and collaboration between domain experts.

From the computational perspective, tutorials used Google Colabs seamlessly. All the codes discussed were open-source, and all tutorials presented during the workshop are available on the conference's website for anyone to follow.

The computational expense associated with the machine learning for materials codes described is mostly related to data-generation. In this case data from the literature were utilized. A discussion on how to develop accessible and efficient machine learning codes which do not necessarily necessitate expensive computational architectures (e.g., GPU highly parallel facilities) has been put forward. Similarly, a reflection on the need to push open-science (open code, open data, etc.) to ensure the democratization of the field has been discussed. While there exist open repositories and robust generation routine for computational data (e.g., Materials Cloud, IoChemBD, NOMAD, Materials Project, AFLOW), a discussion on how to promote the creation of FAIR compliant routines and databases for experiment-related data and codes was initiated

4 Funding

While the possibility of joint research proposals has not been formalized yet during the meeting, the in-person nature of the event led to naturally-forming discussion groups that can lead to collaborations between experts and principal investigators in the field that would not have worked together otherwise.

5 Will these developments bring societal benefits?

The discovery of novel materials for catalytic applications, energy storage, diagnostics, and therapeutics is one of the key ways in which the goal of a sustainable and equitable development can be reached (see also UN Sustainable Development Goals (SDGs))

The recent years have seen a surge in the development and application of machine learning technologies in materials science. The initial results are extremely promising, with applications ranging from the design of novel catalysts for CO_2 reduction to the exploration of the chemical space of energy materials for novel Lithium-free batteries. While giant steps have been made within the design of algorithms and the understanding of the theoretical backbone of machine learning in materials and chemical science, widespread and large-scale applications are just starting to bloom and to have a real-world impact, allowing, e.g., for the discovery of novel stable materials or the design of never-seen-before catalysts or drugs.

This workshop pushed forward research in these critical fields by providing both a way to spread research advancements to young researchers, and a way to initiate collaboration between widely renowned scientists of different fields. This workshop further equipped the next generation of scientists (in academia and industry alike) with skills in tackling the complex problems related to high-performance materials design.

The presence of industry representatives (Roche, Bayer, Microsoft Research, AIndo) on one hand offered an overview of possible career pathways to participants. From an alternative perspective, the state-of-the-art methods and achievements obtained by our community were promoted to these R&D teams.

6 Participant list

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Machine Learning Augmented Sampling for the Molecular Sciences
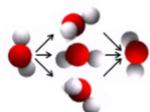
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1124
Dates: May 11, 2022 - May 13, 2022

1 State of the art

Machine learning is rapidly becoming an indispensable tool for the computational sciences. As neural networks, generative modeling, and other machine learning techniques enter computational workflows, many important questions about efficiency, accuracy, and performance remain. Efforts to incorporate machine learning techniques to enhance and augment dynamics and sampling are among the most promising avenues for applications of machine learning in the computational molecular sciences [1], nevertheless, these techniques are still in their infancy. To date, research on this topic has followed two distinct paths: one route relies on generative neural networks to produce samples from a given target distribution [2] and a distinct strategy aims to enhance sampling or dynamics by biasing dynamics [3]. What is more, the literature on this topic is currently somewhat siloed---similar computational problems arise in applications in molecular dynamics [4] and nonequilibrium statistical mechanics [5], quantum mechanics [6], high-energy physics [7], and Bayesian estimation [8], but there is not yet a robust dialogue among the participants in these different literatures.

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2 Major outcomes

The workshop facilitated interactions and dissemination across fields where machine learning strategies can assist sampling the high-dimensional and multimodal distributions that arise throughout the molecular sciences. The workshop presentations recapped recent advances across communities and fueled the reflection on the next steps to tackle several critical challenges in the field.

- *How do we learn non-local sampling procedures to target multiple modes?* An important strategy identified is to combine deep generative modelling with the original workhorse for complex landscape sampling that is annealing. However, questions remain onto the limitation of the proposed approaches: both in the guarantees coming with the investigated approaches as a well as with the limit in complexity of which systems are amenable to these hybrid strategies.

- *Which strategies can be used to leverage learning where no data is available a priori?* In many molecular problems, we do not have a large initial training set with which to train a model, so seeking adaptive or active methodologies is a priority. Here again, it seems that driving learning with annealed targets is a possible solution.
- *What are good measures of success for sampling strategies involving some learning and can we obtain performance guarantees?* The objective functions used for training often do not suffice to evaluate the quality of sampling because they lack global information. This point remains an important challenge in the field of MCMC in itself as highlighted by the introductory tutorial by Werner Krauth. However, it was also pointed out that the differentiability of the machine learning models can also offer extra validation strategies, for instead by validating both the fit of a potential and the corresponding forces. Another important line of research aims at estimating uncertainties in learned models which allows to evaluate a posteriori the level of trust in a given problem scientists can have in fitted models.
- *How do we ensure these methods scale to very high-dimensional, physically relevant, systems?* Most systems of biological or chemical relevance have many degrees of freedom, as such, the scaling of machine learning strategies to high-dimensional spaces appears as critical as much as it is challenging. While systematic methods to scale algorithms acting directly on the entire state-space are still lacking, designing dimensionality reduction scheme is probably key.

3 Community needs

The workshop gathered researchers at the intersection of multiple communities: chemists working on rare events, statistical physicists' experts in sampling methods, physicists interested in many-body problems and machine learners interested in generative models.

- We need events fostering discussions between members of these different fields to foster the emergence of new ideas and to put to the test of applications the emerging methodologies. This CECAM event was a great opportunity given the size of the workshop and the very welcoming environment at Batochime. The repetition of such events every couple of years would be very valuable: the field of probabilistic machine learning is extremely active and moves at an impressive speed. Meetings between communities should enable to assess the opportunities created by newly developed machine learning methods.
- In terms of computational infrastructure, the field is still young. There are no common numerical tools used by the community, each group building its own test bed for the moments. While this has not been discussed at the workshop, it would be interesting to assess the potential of developing common packages. This is all the more relevant for molecular dynamics (MD) applications as these typically combine well-established MD CPU-codes with custom-made python machine learning packages, sometimes allowing for GPU acceleration.

4 Funding

These lines of research can benefit from traditional funding sources, to provide a few examples:
- National funding schemes (such as the National Science Foundation in the USA, Agence Nationale de la Recherche in France).
- European projects (such as European Research Council grants).
- Private foundations particularly keen to fund machine learning research (for instance Hi!Paris in Paris collected donations from the French industry).
There was no discussion of joint proposal during the meeting.

5 Will these developments bring societal benefits?

The research topic of this workshop aims at accelerating computations of statistical properties of physical systems using machine learning tools. While still young, these ideas have a substantial potential for impacts across fields as they tackle a fundamental question. The free energy governs the macroscopic states of physical systems and is well defined for any system associated with a Hamiltonian. However, computing free energies exactly is most of the time out of reach because of its exponential scaling with dimension in terms of computational burden. Relying on learnable surrogate models has the potential to drastically accelerate sampling algorithms, saving computational resources where lengthy computations will be avoided. It will also allow to study physical phenomenon at scales previously impossible to reach. Areas of interest are the fundamental study of phase transitions, fundamental properties of matter in lattice quantum field theories or behaviors of bio-molecules potentially for new drug designs.

6 Participant list

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Wannier 2022

Location: International Centre for Theoretical Physics (ICTP) - Trieste, Italy
Webpage : https://www.cecam.org/workshop-details/1148
Dates: May 16, 2022 - May 27, 2022

1 State of the art

The last two decades have witnessed a tremendous growth in the use of Wannier functions (WFs) for first-principles electronic structure calculations. Beyond providing fundamental insights on several aspects of the electronic structure, from chemical bonding to electrical polarisation, topological invariants, Berry curvature and more, WFs have found applications in a plethora of different domains. The software package WANNIER90 has become a reference for calculating maximally-localised Wannier functions (MLWFs) [1,2] and related properties [2,3,4]. As Wannier functions are independent from the basis sets used to represent the electronic structure in the underlying first-principles calculations, WANNIER90 can be interfaced to virtually any electronic-structure code. Indeed, most of the major electronic-structure packages have already an interface to WANNIER90, including Quantum ESPRESSO, ABINIT, VASP, Siesta, Wien2k, Fleur and Octopus. The availability of a robust MLWF code that is connected to several ab-initio engines has acted as a fertiliser for the birth of independent computational efforts aimed at calculating complex materials properties by leveraging WFs. Several independent packages exploiting MLWFs and WANNIER90 exist nowadays targeting a number of properties, from electron-phonon coupling [5] (EPW) to topological properties [6,7] (Z2Pack, WannierTools), Berry-phase related properties [8] (Wannier Berri), tight-binding models (PythTB, TBModels), high-throughput calculations [9] (AiiDA-Wannier90), and more. Wannier 2022 has been an event that put together the community behind these symbiotic packages that form a research and software ecosystem built upon the concept of MLWFs. The workshop has served the two-fold objective of teaching several techniques enabled by Wannier functions to young researchers and fostering an integration between all the packages composing the Wannier ecosystem, contrasting fragmentation and duplication of efforts.

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2 Major outcomes

The workshop comprised two parts, a summer school (first week) and a developer meeting (second week). The summer school consisted of lectures and hands-on sessions on a wide range of electronic-structure methods based on Wannier functions (WF). The school included highlight talks that provided a historical and broad perspective on WFs in electronic structure, dedicated lectures to the theory and methods of WFs, as well as hands-on tutorials at the basic and advanced level that covered: Maximally-localised Wannier functions (Wannier90); Hybrid Wannier functions, orbital magnetisation, electronic polarization and quantum geometry of the insulating state; Advanced Wannier functions methods: symmetry-adapted, SCDM, transport (Wannier90); Partly occupied Wannier functions (ASE); Tight-binding models (PythTB); Topological properties (Z2pack, IrRep & WannierTools); Berry-phase properties (WannierBerri); Automated wannierisation (AiiDA); Electron-phonon coupling (EPW); Dynamical mean-field theory (TRIQS).

The full program can be found at https://indico.ictp.it/event/9789/, including the recordings of all lectures. The participants of the school had also the chance to present their research activity thanks to two poster sessions (in-presence and online on gather.town). Lecturers and directors voted for the best poster presentation; a monetary award was given to Chakraborty Atasi for her presentation on "Berry Curvature dipole senses topological phase transition in a moiré superlattice". The developers meeting has gathered the community that sustains various software packages built around the concept of maximally-localised Wannier functions (MLWF), strengthening interactions between the developers and promoting a synergetic research and software ecosystem. The full program can be found at https://indico.ictp.it/event/9851/, including the recordings of all talks. We had a round of invited and contributed technical talks (15 in total) to update all participants on current efforts in the Wannier ecosystem, while a large part of the schedule was devoted to group discussions between participants. The talks covered recent developments in WFs - such as Wannier function perturbation theory, Wannier90 library developments and symmetrisation - as well as related subjects such as Koopmans functionals and quantum computing. Every day we had one or two sessions of 3-min "flash talks" that helped to form several groups of 3 to 10 participants focusing on specific areas. Participants had the opportunity to cooperate on integrating and interconnecting different packages into an organic software ecosystem, share capabilities and exchange ideas, while contributing to drawing a roadmap for future developments of the software related to Wannier functions. We include here the list of the working groups formed during the workshop: symmetry, library. electron-phonon, bottlenecks, coulomb, position operator, tetrahedron, hybrid wannier functions, python i/o, high-throughput, alloys and defects, initial projections. Thanks to the workshop, substantial progress has been made in several of the areas mentioned above. We discovered that several participants wanted to solve the same problems, and that often multiple software implementations—that would solve those—had been developed and needed to be merged and shared with the community. In addition, the workshop emphasised how already-existing solutions given problem (e.g. hybrid Wannier functions) could easily be adapted to serve a very different application (in this example, device simulations of 2D materials). Among the many advancements, we mention some concrete steps towards an interface between Wannier90 and Qiskit for quantum computing simulations. Finally, the workshop has been invaluable to share the latest updates and most recent developments in the broad Wannier functions community, which has triggered a number of collaborations that started thanks to the workshop.

3 Community needs

The electronic-structure community crucially depends on robust open-source first-principles codes and continuous access to HPC resources. These workshops are vital to support the maintenance and development of codes, especially in the Wannier functions ecosystem, from small software packages (e.g. WannierBerri) to intermediate-size codes (e.g. Wannier90), to large ab-initio codes (e.g. Quantum ESPRESSO).

The Wannier functions community would benefit from 5-day annual developer meetings, based on the model of our Wannier 2022 Developers Meeting. In addition, advanced schools like the Wannier 2022 Summer School-which covered the broad Wannier ecosystem beyond Wannier90 (from DMFT, to electron-phonon, to topological properties and more)-should probably be scheduled every two years. At the workshop, we discussed the pros and cons of having remote-only summer schools, while there was a strong consensus that developers meeting should be run in presence.

The community would definitely benefit from more specific funding for the maintenance and development of open-source simulation codes, for instance in the form of permanent positions dedicated to code development (as opposed to research-focused) at national labs and research institutions.

4 Funding

This workshop has been funded by ICTP (15'000 EUR plus extra accommodation and staff), Psi-k (5'000 EUR) on CoE MaX funds, NCCR MARVEL (5'000 EUR) and CECAM (2'500 EUR). The computational infrastructure to run the hands-on tutorials was provided by ICTP through their ICTP Cloud.

We have discussed the possibility of joint research projects that would require proposal for funding both postdocs/PhDs and computing time on HPC facilities.

The Italian newborn national center on HPC might be an interesting opportunity to monitor, together with several parallel efforts all over Europe funded through the NextGeneratioEU plan.

5 Will these developments bring societal benefits?

Wannier functions enable to calculate from first principles an enormous number of important materials properties, ranging from electronic polarization, to transport coefficients, to electron-phonon coupling (e.g. superconducting critical temperatures), to topological properties and more. In addition, Wannier Hamiltonians are routinely employed in multi-scale device simulations, to bridge atomistic first-principal calculations with transport simulations—that is particularly relevant to understand and design transistors (and other electronic components). As discussed during the workshop, Wannier functions might also play an important role in connecting quantum computing algorithms with first-principles codes.

Hence, the advancements in the Wannier functions ecosystem—that have been supported by the workshop—definitely contribute to improve our capabilities to compute relevant physical properties of complex materials at low computational cost (especially thanks to Wannier interpolation). These techniques are fundamental to computationally screen or design novel materials with improved performance and/or higher sustainability.

6 Participant list

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Hybrid Quantum Mechanics / Molecular Mechanics (QM/MM) Approaches to Biochemistry and Beyond

Location: CECAM-HQ EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1030
Dates: May 16, 2022 - May 20, 2022

1 State of the art

This School, following former editions on the same theme, was intended as an answer to the growing need for multiscale approaches accounting for different levels of accuracy in the description of realistic systems. Advances in methods, computer codes and high-performance computing (HPC) architectures are constantly expanding, becoming increasingly multidisciplinary, whereas standard University courses and school have difficulties in coping with these developments.

To meet requests and expectations from students and young researchers approaching this rapidly evolving field, we extended, complemented and updated our School with respect to all former editions, following precious indications from previous participants and colleagues worldwide. We did our best to present in a comprehensive way scope, implementation and domain of applications of hybrid quantum mechanics/molecular mechanics (QM/MM) approaches. To this aim advanced computational tools and free energy sampling techniques used nowadays were presented and discussed. Analogously, the issue of excited states and quantum treatment of the nuclear degrees of freedom was added to the lectures for a comprehensive overview of the methodologies available nowadays. With respect to the state of the art, since a formation of this type is rarely available in University Master and Ph.D. courses, we designed specific QM/MM exercises to be done by participants with the scope of making them understand the physical and chemical meaning of data and parameters supplied as input files in the computer codes. This was accompanied by a critical analysis and post-processing of the numerical outcome.

Morning lectures on the underlying theory were followed by afternoon sessions of training with two major codes, CPMD (http://www.cpmd.org/) and CP2k (http://www.cp2k.org/). New exercises were designed with respect to previous editions.

Key References

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2 Major outcomes

Lectures, exercises and the constant interaction with the participants, allowed us to provide them a precise knowledge about know how to set up a QM/MM simulation starting from pristine crystallographic data, available directly from experimentalists or from the protein data bank (PDB), and following the various steps to complete the system, including the addition of missing hydrogen atoms and solvent molecules. Lectures were generally animated by questions and specific requests from the participants, honored with additional files provided by us directly on the CECAM web server granting easy access to everybody. The participants could acquire a discrete knowledge about the basic protocol and fundamental algorithms used to perform QM/MM dynamical simulations. Particular attention was given to the interface between the QM and the MM subsystems and their mutual interaction. The students learned about the calculation of the different energy terms and forces, including different levels of approximation employed in the most popular QM/MM codes available.

In the hands-on sessions, the participants could experience directly the handling of a large system in term partitioning between an embedded QM subsystem and the larger MM one surrounding the quantum part. Different QM size and QM-border terminations were experienced by the participants in the afternoon sessions, allowing then to handle the subtle effects on the dynamics of the electronic charge distribution and the electrostatic potential of the composed QM/MM system. Due to time limitations and computer accessibility, the examples were based on simple systems (small QM region, small plane wave cutoff energy, etc.). Nonetheless, the protocols employed are valid for more realistic and complex QM/MM systems. Advanced techniques, such as Blue Moon, metadynamics and string methods, for the sampling of rare events and activated processes were also presented. These were complemented by dedicated lectures on different levels of complexity, including numerical grids and the treatment of long-range interactions (CP2k), the treatment of nuclear quantum effects, and both time-dependent density functional (TD-DFT) and surface hopping methods in QM/MM simulations for the treatment of excited electronic states.

3 Community needs

We received an unprecedented number of applications (86) for this specific edition of our School. While we were mainly expecting to accept participants formerly excluded for technical and budget reasons in past editions, we had to cope with an increased interest in this School from nearly all European and several non-European countries. Budget constraints, limitations in the number of seats in the terminals room and still persisting consequences of the pandemic covid-19 crisis did not allow us to accept onsite all candidates. Only 32 onsite participants, among which 6 unsupported, could be accepted. Nonetheless, in an attempt at satisfying a larger number of students, we included, on the basis of our former experiences, 23 online participants who could follow in real time lectures, ask questions, and have access to the slides of the presentations. Recordings of our lectures are still available at the CECAM web site, so that one can download the PDF versions of all the lectures and listen to the explanation

simultaneously. A repetition of this tutorial in a couple of years would be highly welcomed by the large number of applicants who could not be accepted. We remark that keeping the number of participants at 20-30 is an optimal compromise: On one hand, the number of terminals in the computer room at the CECAM HQ and the available computing nodes are limited. On the other hand, a larger number of attendants would further make the execution of practical sessions more difficult, in particular responding promptly to the many requests we experienced from the students during the hands-on sessions, often resulting to specific, time-intensive explanations. Students expressed their warm gratitude for having given them this opportunity.

4 Funding

This time, the regular CECAM allocated funding (20000 CHF) was barely sufficient to cover the accommodation expenses, social dinner, coffee breaks and allowed for a small per diem (20 CHF/day) to the supported participants and the tutors. Especially the per diem, keeping into account the cost of eating in Switzerland, even at the canteens and facilities at EPFL, for the student was judged insufficient. Actually, a slightly larger budget (~additional 1000 CHF) would have been welcome in view of the increased life cost worldwide in the last two years.

5 Will these developments bring societal benefits?

The active participation of all the students, including the ones following on-line, and strict interactions also during the breaks and at the hotel, where both tutors and students were lodged, made the tutor-student interaction one of the main successes of this tutorial. Indeed, students of this edition of our School demonstrated a high interest in the subjects and started autonomously their own WhatsApp virtual community to continue the discussion and their mutual interactions even after the official time of the school. On the basis of the comments, feedback and discussions with the participants, we feel that the formation we provided is likely to have an impact in their research career and, hence, on the society in which this future generation of researchers will work and to which they will contribute. Afternoon sessions, after an intensive learning day for the participants, were also an important moment of exchange between tutors and students and among the participants. The poster sessions also remained very lively all along the week (including flash presentations from the participants), a clear sign of motivation from all the students. The poster prizes, which were increased to 6 this year, as opposed to 4 in former editions, testify the high quality of the participants and poster prizes (a certificate plus a chocolate box) were highly appreciated. With many of them, communication (via e-mail) with requests for further details, checks and help in understanding the hands-on exercises has continued also after the end of the tutorial, providing us with highly useful feedback about the practical utility of this event. Indeed, in this edition, one of the speakers, Xi Chen, was in fact one of our former students in this same School. A clear sign of the importance and significance of this event. The school that we have organized will facilitate future developments in these fields, i.e. making our planet great again.

6 Participant list

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Les Houches - TSRC Workshop on Protein Dynamics

Location : Centre Paul Langevin, Aussois, France
Webpage : https://www.cecam.org/workshop-details/54
Dates: May 22, 2022 - May 27, 2022

1 State of the art

The **study of complex biomolecular dynamics is a considerable challenge**, as it requires not only to determine a set of atomic coordinates in 3D space, but also how they evolve in a fourth dimension, time. **No single experimental technique is able to achieve this task by itself**. Remarkable developments over the last decade, enabled by steady progress and ground-breaking new technologies both in computational algorithms and experimental approaches have, however, provided much insight into the dynamics of proteins, often by merging different experimental approaches and computational methods. Increasingly, the spatial and temporal domains accessible to state-of-the-art computational models and experiments are commensurate, providing **opportunities for previously unprecedented critical comparisons of the predictions of computational models as well as the molecular-level interpretation of experimental data**. Arguably, connecting experimental with computational data has turned out to be game-changing in order to understand the function of increasingly complex biomolecular systems at ever-growing details.

Important challenges remain to be overcome both for experimental and computational approaches. For example, a particularly important and active research field in computational biophysics aims at simulating molecular dynamics over sufficient time scales (micro- to milliseconds) to observe the biologically relevant conformational changes, which coincide with many biomolecular processes (e.g. enzymatic turnover). Computational investigation of millisecond events in large biological objects is computationally very expensive, calling for e.g. enhanced sampling and accelerated simulation methods, new hardware and algorithms. Another active research area is related to understanding the solvation of biomacromolecules through computational and experimental approaches.

2 Major outcomes

During the workshop researchers from all over the world, specialists in the characterization of biomolecular dynamics, exchanged on the latest advances in the field, covering a wide spectrum of disciplines between biology, physics, (bio-)computer science and chemistry. This multi-disciplinarity was the richness of this workshop, and also a challenge, as it required a very pedagogical approach from the speakers in order to exchange. The 35 speakers were asked to spend 15-20 minutes on a pedagogical presentation of their research technique and methods, followed by a presentation of their latest research. The discussion after each presentation was an important part of the workshop. In addition to the speakers, 29 participants (PhD students, post-doctoral fellows, researchers) took an active role in the discussions, and each participant presented a poster and a 3-minute flash presentation of their research. Several young researchers were selected, based on the submitted abstract, to give an oral presentation. A prize from the French Biophysical Society was awarded to the best poster.

There were several highlights to be mentioned here.

One was the integration of experimental data with numerical/simulation approaches. Kresten Lindorff-Larsen (Copenhagen) presented several of their latest developments that use reweighting algorithms to simulation data to match experimental data. This talk sparked great interest and lively discussions from both the experimental and computational participants. Among such theory/simulation/experimental integrations were also studies of intrinsically disordered proteins, addressed by several speakers (Robert Best, Ben Schuler, Steve Pressé).

Another focus was the discussion of machine-learning based approaches in the prediction of protein properties and analysis of data. Along these lines, Basily Wicky (Washington University; Baker group) presented latest developments in protein design, which take into account the conformational landscape. Partyush Tiwari, with a provocative title "A match made in heaven? Integrated theory, simulations and AI for discovering protein dynamics", explained multi-disciplinary AI-assisted studies.

there was intensive discussion about single-molecule experiments, from experiments and theory (following talks by Dima Makarov, Steve Pressé, Rob Best, and others).

We explicitly encouraged discussions, at least 15 minutes after each talk, and provided also time for discussions during evenings and a hike (one afternoon).

We are aware of several collaborations that emerged from this meeting.

3 Community needs

As the participants were very multi-disciplinary, covering many experimental techniques in addition to computational approaches. Given the heterogeneity of the crowd, discussing the community needs was not the focus (as this was so diverse). However, it became clear that the integration of experimental and computational approaches is absolutely required. It was agreed that it is important that these different communities meet each other, in meetings such as this one.

We launched a survey after the meeting about this meeting and which was extremely positive, with a strong call for organising this meeting again (planned for 2024).

Access to high-performance computational infrastructure is important for some (but not all) the computational people. It turns out that for the experimentalists - even though they often also deal with large data sets - it is not so important to obtain external HPC resources, because usually this is provided by their home institutions.

4 Funding

Sponsoring of the conference: In addition to CECAM funding which we gratefully acknowledge, we have reached out to several other funding bodies, and have been successful to some extent. We have obtained significant funding from the "Deutsch-Französische Hochschule"/"Université Franco-Allemande" (12 k€). Moreover, the institute of one of the organisers (Institute of Science and Technology Austria) supported the workshop (2 k€), as well as the institution of a French organiser (GRAL, GrenobleAlliance). We unsuccessfully applied for support from EBSA.

We have not explicitly discussed channels for funding the research. Available funding for the research work includes national funding schemes as well as European (ERC) funding.

5 Will these developments bring societal benefits?

Contemporary molecular medicine and drug discovery heavily rely on the output of structural biology. The determination of molecular structures (static ones!) has been revolutionized lately by machine-learning based approaches, such as AlphaFold. Dynamics, however, would presumably be very hard to predict, and it requires still the combined efforts of experiments, theory and simulation. Moreover, many proteins that are central to human diseases do not even have a stable 3D structure, but are dynamically disordered. The mechanisms by which proteins interconvert to form, for example, insoluble protein aggregates involved in Alzheimer's or Parkinson's diseases, still are far from being resolved. The development of drugs that target intrinsically disordered proteins is a very challenging field, and it is clear that it has strong potential.

The multi-disciplinary study of protein dynamics is of societal interest, because it has the potential to help tackle diseases for which the contempary methods are insufficient.

The study of protein structure and dynamics also involves the developments of cutting-edge instrumentation, such as free-electron lasers and electron microscopes. These developments regularly entail solving technical problems, which then turn out to be useful for other fields.

6 Participant list

Organisers
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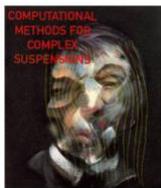**Schanda, Paul**
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Computational methods and tools for complex suspensions

Location: Ensanche Building in Bilbao, Spain
Webpage : https://www.cecam.org/workshop-details/1112
Dates: May 23, 2022 - May 27, 2022

1 State of the art

The scientific goal of this new CECAM workshop is to move beyond so-called "simple suspensions" to **"complex suspensions"** where complexity can come from different (possibly concomitant) sources that include but are not restricted to:

(1) **Non-Newtonian properties of suspending phases**
How can existing computational models be generalized to handle viscoelastic, plastic, or thinning/thickening liquid phases? How can far-field hydrodynamics interactions between suspended particles be modelled accurately and efficiently? Can new short-range non-Newtonian lubrication force models be developed? How can we model partial/full slippage effects on the particle surface, and translate them into effective hydrodynamic interactions? How can non-Newtonian models be generalized to the fluctuating/Brownian regime?

(2) **Complexity of the dispersed phase, particle anisotropy and deformability**
Suspensions are often composed of deformable and anisotropic particles. Topics of specific interest include novel models and numerical schemes for suspensions of irregularly-shaped particles (including roughness), deformable particles, including Brownian motion, as well as applications in biophysics or technology/engineering.

(3) **Complex inter-particle interactions and Multiphysics**
The workshop will examine novel schemes and/or applications focusing on the effect of complex non-hydrodynamic interactions in suspensions, and in particular many-body interactions such as magnetic nanoparticles or dielectric nanoparticles under optical fields.

(4) **Particle activity**
Active suspensions are a topic of great interest, and often require resolving complex hydrodynamic phenomena. Of special interest to this workshop are new approaches to reduced-order (coarse-grained) modeling of active suspensions.

2 Major outcomes

In regards to topic (1) **non-Newtonian properties of suspending phases**
Presentations focused on: (1) computational methods for direct numerical simulation of suspensions with a non-Newtonian solvent. (2) Comparison to experimental measurements of elongational an extensional viscosity in dense suspensions. (3) Models that combine frictional and lubrication forces between nearly touching particles in dense suspensions. The round-table discussion focused on the contrast between modeling lubrication in dense suspensions using a hydrodynamic model based on rough particles (e.g., Brady, Swan), and modeling them with dissipative frictional contact forces (e.g., Ellero, Morris). A major open question we identified for future work is whether experimental measurements can distinguish and thus select between these two models. Another topic discussed was how to model the Brownian motion of particles suspended in a non-Newtonian fluid. Some gaps were identified in the existing literature, notably going beyond linear models such as Oldroyd-B, and the need

for new computational methods and comparisons among different simulation techniques (e.g., SDPD, LB, FHD).

In regards to topic (2) Complexity **of the dispersed phase, particle anisotropy and deformability**

Presentations focused on deformable red blood cells and vescicles in confined flows, the (hydro)dynamics of semi-flexible polymer chains in complex flows, efficient modeling of suspensions of sphero-cylinders, the dynamics of flexible vescicles filled with active particles, and systematic coarse-graining to Langevin equations. In the roundtable discussion, the focus was on continuum modeling of Brownian fluctuations of flexible objects in a solvent, such as polymer chains or membranes, and the avoidance of collisions between rigid particles in Brownian Dynamics simulations. We identified as a direction of future work more systematic study of the sensitivity of models of Brownain flexible objects to the details of the discretization, and the need to develop more efficient yet accurate methods for many-body hydrodynamics that mediate between expensive boundary integral methods and cheaper but not very accurate methods like rigid multiblobs.

In regards to topic (3) **Complex inter-particle interactions and Multiphysics**

Presentation of experimental and numerical works on magnetic and optical forces on nanoparticles. J. de Vicente showed the rich spectra of pattern formation of magnetic nanoparticles (MNP) and their rheology under flow. Jonhanat Leliaert introduced state-of-art numerical modelling for the evolution of the magnetic moment of MNP dominated by either Neel or Brownian relaxation; and applications to hyperthermia and Pablo Palacios presented simulations including both magnetic interactions and hydrodynamic coupling of supraparamagnetic NPs. M. Marqués presented several examples of light driving plasmonic nanoparticles owing to the primary and secondary optical forces induced by laser beams. Bioinspired problems involving either physical virology with AFM (de Pablo) or cell motility.

In regards to topic (4) **Particle activity**

Presentations focused on accurate computational modeling of propulsion mechanisms and the importance of hydrodynamic interactions in active suspensions of phoretic colloids, driven colloidal micro-rollers coated in reactive agents, the importance of accounting for particle shape for bacterial suspensions, efficient computational modeling of propulsion using hinged bodies and reduced models, and collective dynamics in drops of active nematics. The round-table discussion identified the need for a basic experimental system of active particles where propulsion can be understood and controlled and modeled, and the need to identify common mathematical and physical questions that go beyond the wow factor common in the field of active matter.

3 Community needs

One of the interesting conclusions from this workshop was to realize that we were forming a new "community". Many of the participants did not previously know each other, and come from different "standard fields" such as magnetism, optics, hydrodynamics, rheology, physical virology, experimental softmatter, applied mathematics and either experimental, theory or computation. However round discussions clearly showed the need of combining these dispare communities to bypass broader challenges in applied and fundamental problems. In fact, the workshop showed that fundamental and applied science are much closer than they seem. Several collaborations arised over the workshop between hard theoretists on coarse-graining (P. Español, T. Schilling) and experimental groups in biology, bridged by groups on mesoscopic modeling (Pagonabarraga, Ellero, ...) using high performance computing (Delgado, Pelaez...) and solid applied math (Donev, Brenan, Schkafe, Delmotte). New interdisciplinary communities need time and space to meet and networking: i) organizing workshops in the line of this one and ii) organizing courses of different type, to mix knowledge on theoretical/experimental/numerical basis of soft matter and molecular biology. The scope and domain of this interdisciplinary community is quite large and a series of events would be

most welcome. As an example, concerning numerical techniques, over the final discussion session, it was proposed to foster further know-how exchange between the magnetic, optic and hydrodynamic communities studying nanoparticle transport, as they all deal with many-body green functions. This could be the subject of a more specialized workshop or course. Interdisciplinarity has been a "hot concept" for more than one decade, however it is still in its infancy and faces strong difficulties due to the inertia of the comfort zones. Organizations, such as the CECAM, need to take care of the promising seeds of interdisciplinary communities.

4 Funding

As a collective, we did not discuss the possibility of joint research proposals during the meeting. However, several new collaborations were started over this meeting and some of them might lead to EU funding in the near future. Fostering these initiatives and collaborations between groups were part of the objective of the workshop, and we believe we were really successful. As a new collective, we discussed the need for further workshops of this kind, to keep the momentum going, and also the need to share knowledge between disciplines in the form of short courses, probably in the summer. In fact, to ease scientific communication, we need to start sharing *basic knowledge* on experimental techniques, theory and numerical schemes between students of different experimental and theoretical groups. We discussed options for funding via national and local programs. A COST action would be also attractive, but difficult to set, precisely due to the interdisciplinary character of the group.

5 Will these developments bring societal benefits?

One of the keynote talks focused on the study of toxicity of nanocolloidal titanium suspensions commonly used in cosmetics, medical imagining, and other situations of great societal prevalence and importance. We discussed medical applications of suspensions of nanocolloidal magnetic particles in tumor treatment via local heating. Understanding the shear thinning and thickening rheology of dense suspensions is also of great importance in practical applications, and requires closing the gap between first-principles theoretical understanding and empirical modeling currently used in engineering. Another medical application discussed in the meeting was the effect of blood cell diseases on the flow and fractionation of red blood cells in capillaries. To bring these applications to fruition requires combined experimental, theoretical, and computational work to bridge the gap between the microscopic structure of the suspensions and their macroscopic behavior. The possibility of using neural networks to bridge the gap between data and practical clinical and other applications was identified as a future direction of research.

6 Participant list

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New Horizons in Photoelectrochemical Water-Splitting and Heterogeneous Catalysis: Bridging Theory, Simulation and Experiment
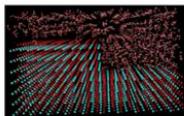
Location: CECAM-FR-RA, Lyon France and CECAM-IRL, Dublin, Ireland
Webpage : https://www.cecam.org/workshop-details/1171
Dates: May 23, 2022 - May 31, 2022

1 State of the art

This timely workshop aims to bring together both leading experimentalists and theorists/simulators to share their perspectives, knowledge and experiences on state-of-the-art progress in PEC [1-4]. The high-level workshop goals will center around tackling the pressing problem of removing obstacles towards efficient and sustainable PEC water-splitting - using a broad suite of state-of-the-art molecular-simulation methods to elucidate mechanistic insights for leading, careful experiments to investigate. In so doing, we advocate a pragmatic philosophy of simulation-led electronic/atomistic design as a cost-effective prototyping tool. The workshop will elucidate the role of doping and surface defects in promoting the efficiency of cost-effective metal oxides, such as TiO_2, Ta_2NO_5 and Fe_2O_3, etc.

Further workshop objectives include discussions on progress in:
1. static electronic structure and excited-state Density Functional Theory-based calculations (e.g., non-adiabatic MD for electron-injection kinetics), in particular with emphasis on non-equilibrium charge-carrier dynamics;
2. molecular dynamics of explicitly-solvated surfaces by reactive force-fields, tight-binding MD approaches and linear-scaling electronic-structure methods;
3. training of highly sensitive machine-learning potentials to allow for higher-fidelity and longer-time deterministic or free-energy-biased dynamics simulations (*e.g.*, to gauge charge-carrier dynamics in a more rigorous, statistical manner);
4. experimental spectroscopy determinations of charge-carrier lifetimes, and on how this can drive forward progress in state-of-the-art molecular simulation.

Key References
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2 Major outcomes

Some very important talks were given on experimental methods (T. Cuk, P. Farras, V Sofianous), device physics (V. Andrei, V. Artero), multiscale modelling (S. Haussner), explicit solvation (A. Selloni, D. MacKernan, N. English) and DFT and simulations (C. di Valentin) – and, intriguingly, on polarons (R. Long, A. Walsh and P. Deak).
Some key themes which emerged from open and targeted/general discussions were the need to develop pathways and solar-conversion devices that are effective. In this sense, the topic of preservation of charge-carrier lifetimes, to avoid recombination, was seen as being

particularly important. To this end, making defects selective to trapping holes and electrons is important. Of central interest is whether electric fields can preserve charge separation for long enough? In this respect, the cultivation and development of an insulator layer is to be assessed carefully. It was also discussed as to what limits heterogeneous catalysis and PEC itself, as compared to charge-carrier motion and lifetimes.

Beyond these more high-level considerations, there was also a good deal of targeted discussion on the perennial practical and configurational matters of how electrodes should face each other. This includes matters of light management (and how to deal with diurnal fluctuations therein, as plants accomplish) – e.g., the consideration of going through two devices in a tandem set-up – and what the techno-economic benefits of this may be. Technical questions, such as what the limiting flux of charge is, are also important.

On the broad and important matter of CO_2 conversion, one problem with this area is that it is too early to discuss performance in detail, given that a lot of further underlying principles need to be clarified. In this respect, the talk by Motiar Rahman was very important in highlighting this. Certainly, on CO_2 conversion, making the liquid product directly, such as formic acid, methanol, etc., is a challenge: product separation is vitally important here, although a lot of energy is needed for this. In another sense, catalytic degradation is another problem to be understood in the broader context of what, and which, activation steps can make a catalyst more robust, and confer longer device life, bearing in mind that solar-light concentration does not work so well for diffuse light?

On the type of surfaces for optimal heterogeneous catalysis, very different catalysis has taken place on different materials, especially ones with large surface areas for efficient catalysis. Here the on/off shuttering of light – or pulse electrolysis – was discussed (e.g., Beatrice Roldan Cuenya, FHI), which featured a greater stability of CO_2 reduction on copper catalyst. In this context, the work of Virgil Andrei was discussed in the workshop, who does 50 min on/10 min off – which is needed to be considered in the context of pico- and micro-seconds. In electrolysis itself, reversing polarity to stability-test is another approach for an inversion of polarity - which has big ramifications of fuel-cell operation. 'Annealing' of the sample is also important: if the charges cause it to degrade, they make the electronic states go back to 'heal'. It was discussed that altering the bias voltage, such as on/off, and even trying to reverse polarity of the bias potential, are very effective.

The electric double layer (EDL) and capacitance were also discussed: this is slow to rearrange. In salts, we can think of Coulombic charges and timescale, and the strength of electric field. This suggests that some underlying periods might be effective as a whole, rather than on identifying individual sub-processes (by trial and error). With big electrodes, it takes milliseconds for these processes to occur, but these may need to go to shorter micro-second scales.

3 Community needs

Turning to theory/simulation, and how this can make contributions to experiments, theory and simulation, theory needs to be better on kinetics. Even for basic thermodynamic bases (e.g., Noskov theory), this would benefit from better treatment of kinetics for a more holistic understanding. The role of explicit solvent is also important, in that we need better solvent models, and this was discussed by Selloni, MacKernan and English. The role of rare-event methods was also highlighted (as well as the calculation of order parameters): femtoseconds for hydrogen atoms ranging to milliseconds for macro processes, and on how to handle bond-breaking and creation (particularly in the context of work by C. di Valentin and N. English on tight-binding DFT and reactive forcefields).

For wider theory, the talks by MacKernan on the path-integral propagation of the system are important. For coarse graining, we do not need to know all of details, but can rather focus on the core of the system. This is also the case for the rigorous Henderson theorem, which is like the classical analogue to the Kohn one: for every pair distribution function, there is a pair potential to reproduce it. By getting the PDF of effective variables for a large number of atoms,

we can define an average object - extracting PDFs of those beads, validated from structure-factor information.

Combining machine-learning (ML) with experiments and DFT/electronic-structure simulation is also important, in that doping of the NP systems, and how to modify OH and co-catalysts, needs to be clarified – in an effort to engineer photo-anodes, so as to minimise optical and energy losses. For actual construction of AI-based potential models trained on such high-quality data (particularly accurate electronic-structure simulations), the work of A. Selloni and her colleagues in Princeton is particularly important here, with recent efforts in the group of N. English being of note.

4 Funding

Reflecting upon this double-workshop, scientific and funding priorities for future work in this area are:

1. Higher-fidelity, time-resolved experimental measurements
2. More accurate potentials for longer-time molecular simulation (e.g., machine learning)
3. Accurate determination of excited hole-/electron- diffusion lengths and timescales
4. Pulse methods for photo-driven or heterogeneous catalysis
5. Advances in multiscale modelling (over time and space), for double-layer phenomena

These grant-submission priorities, for both individual groups (national schemes) and wider consortia (e.g., Horizon Europe), reflect key outstanding challenges in this general field. Taking this into account is important in seeding potential collaborations in the future for joint efforts – grants, workshops, conferences, community events, community codes, etc.

5 Will these developments bring societal benefits?

At present, sustainable conversion efficiencies of incident solar radiation to liquid fuels are only a few percent. Harvesting ~0.25% of the ubiquitous and vast incident solar radiation on the earth (some 120,000 TW) and conversion at ~8.5-10% efficiency (*i.e.*, ~25-30 TW) would meet the projected doubling of domestic, transport and industrial needs of the world's population by mid-century (from ~17 TW today to ~30-35 TW). Although solar energy is diffuse and subject to diurnal and seasonal fluctuation, it does, in large part, overcome fundamental intermittency/volatility problems associated with other renewable-energy forms, *e.g.*, wind and wave. However, although no system including a single photo-absorber has shown *truly efficient, sustainable and economic* water splitting to date, the gap is certainly closing with respect to non-renewables for conversion of solar energy into hydrogen or electricity from photoelectrochemical (PEC) and photovoltaic (PV) approaches, or, equally, tandem-cell approaches. In particular, PEC has seen some major developments in recent years, to the point of potentially being competitive with PV methods for real-world deployment in the coming years.

The "paradigm shift" in boosting PEC's longer carrier dynamics, as well as reducing charge recombination and canny band-gap manipulation, will serve to facilitate the development of sensible atomistic/electronic "design rules" for experiment and theory/simulation working hand in glove in a symbiotic relationship – a genuine overhaul of intellectual infrastructure and thinking - to design better solar and carbon-conversion catalysts.

6 Participant list

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Open Databases Integration for Materials Design

Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1120
Dates: May 30, 2022 - Jun 3, 2022

1 State of the art

During the last decade, materials design has undergone a revolution. The increase of computing power and the development of simulation codes have led to the development of the so-called high-throughput (HT) computational approach through which large sets of calculations can be performed automatically. As a result, large databases (DBs) have been created with the calculated properties for existing and hypothetical materials. The empirical approach has obviously not been left out and experimental databases have also developed, gathering different kinds of materials properties. By interrogating all these DBs, it is now possible to screen for materials with targeted properties. Moreover, all the available data can be used to train machine-learning models for predicting the properties of other materials at a much lower cost.

Many materials DBs have become available online (e.g., AFLOW, COD, Materials Cloud, Materials Project, NOMAD, and OQMD, etc.). In most of the cases, a Representational State Transfer (REST) Application Program Interface (API) is available to interrogate the DB through scripts. Nonetheless, up to a few years ago, queries could only be performed on one DB at a time, and they would have to be adjusted to the very different APIs of each DB.

Thanks to the OPTIMADE consortium (gathering all the key players involved in these different efforts), a common API was developed [1] through a series of meetings held at the Lorentz Center in Leiden, Netherlands from 2016-10-24 to 2016-10-28, and at the CECAM in Lausanne, Switzerland from 2018-06-11 to 2018-06-15, from 2019-06-11 to 2019-06-14, from 2020-06-08 to 2020-06-12 (virtual event), and from 2021-06-07 to 2021-06-11 (virtual event).

Key References

[1] C. Andersen, R. Armiento, E. Blokhin, G. Conduit, S. Dwaraknath, M. Evans, Á. Fekete, A. Gopakumar, S. Gražulis, A. Merkys, F. Mohamed, C. Oses, G. Pizzi, G. Rignanese, M. Scheidgen, L. Talirz, C. Toher, D. Winston, R. Aversa, K. Choudhary, P. Colinet, S. Curtarolo, D. Di Stefano, C. Draxl, S. Er, M. Esters, M. Fornari, M. Giantomassi, M. Govoni, G. Hautier, V. Hegde, M. Horton, P. Huck, G. Huhs, J. Hummelshøj, A. Kariryaa, B. Kozinsky, S. Kumbhar, M. Liu, N. Marzari, A. Morris, A. Mostofi, K. Persson, G. Petretto, T. Purcell, F. Ricci, F. Rose, M. Scheffler, D. Speckhard, M. Uhrin, A. Vaitkus, P. Villars, D. Waroquiers, C. Wolverton, M. Wu, X. Yang, Sci. Data., **8**, 217 (2021)*
*[2] M. Evans, C. Andersen, S. Dwaraknath, M. Scheidgen, Á. Fekete, D. Winston, JOSS., **6**, 3458 (2021)*

2 Major outcomes

The present workshop started with a tutorial with a series of lectures and of two hands-on sessions, as well as a colloquium with 4 talks and a round table. The talks showcased possible use of OPTIMADE either already implemented, or in development, or even suggested extensions. The second afternoon consisted of a series of brief presentations (elevator pitches of 5 minutes) during which the participants to the previous OPTIMADE meetings discussed the state of their implementations. From the third day on, the workshop alternated parallel sessions (one focusing on the improvement of the API, one focusing on molecular dynamics trajectories, one focusing on GitHub issues and implementation) and plenary meetings in which the progress achieved in the parallel sessions. The workshop took place in hybrid mode with about 20 persons onsite and 30 people online (especially for the tutorial).

Tutorial

After a brief general introduction, different ways to use the OPTIMADE API were first presented together with some examples. One of the presentations also focused on the OPTIMADE Python tools [2]. During the hands-on sessions, the participants went through the different exercises available online. They could ask questions to the developers. During the second session, they could even either set up their own OPTIMADE server or come with their own possible usage of OPTIMADE.

Molecular dynamics

This parallel session was mainly attended by molecular dynamics and biomolecules experts and various OPTIMADE developers to help coordinating the discussion. After a presentation of the recently developed prototype, the participants first installed it on their own machine and then were able to upload a few trajectories in their database. Then, various discussions took place about the specification of the trajectories reaching an agreement which will be translated into a pull request in the coming weeks and will then be polished online. Finally, the rest of the discussions were dedicated to the extension of the structure specification for biomolecules (including residues, chain, …). Here, a few more discussions will be needed to achieve a final agreement.

Developments to the OPTIMADE API

This parallel session started about a discussion regarding the possible use of GNU units within the OPTIMADE specification. Then, it mainly focused on the extension of the OPTIMADE API to cover a broader range of properties. Many very fruitful discussions took place (both during the plenary and small-group sessions). The range of improvements can be measured by the >100 pull requests in the OPTIMADE GitHub repository during and in the days immediately following the workshop.

The OPTIMADE workshop can definitely be considered a success since very important improvements were achieved for the common API and its actual implementation, for its extension both in terms of new properties and MD trajectories. More importantly, very tight connections have been established between the different projects, and new projects have been incorporated. The participants really appreciated the workshop and indicated that the chosen online tools had been key to its success.

3 Community needs

Materials design (including materials informatics) will clearly benefit from the common OPTIMADE API which makes it possible to interrogate all the DBs using the same query. The OPTIMADE API will also contribute to the FAIRification (Findable, Accessible, Interoperable, and Reusable) effort that is currently taking place in the materials community.

The OPTIMADE initiative should definitely be continued, trying to involve as many important players as possible. For example, at the present CECAM workshop, we welcomed developers from ColabFit and MPDD. Some of them have already started implementing the OPTIMADE API to query their database.

It is important to maintain this community with a common target (the OPTIMADE API) that will be beneficial for the community. Furthermore, the links that have been created among the different persons can also serve a different purpose. Indeed, there has already been discussion about the possibility of using the same community to define some standards and an ontology for materials (actually, a workshop will soon be organized involving many people of the OPTIMADE consortium). Moreover, since the meeting gathers an important number of players in the field, this number can be used as leverage to push towards the adoption of a standard.

For all the reasons above, we believe that the OPTIMADE API effort should be continued. We hope that the CECAM will continue to support the organization of this series of workshops.

4 Funding

The OPTIMADE consortium has established links with various key players world-wide. At the EU level, there is a connection with the European Materials Modeling Council (EMMC). The OPTIMADE API is in line with their objective and interactions have already taken place. Various members of the OPTIMADE community participate in some of the boards of the EMMC. There have been discussions to apply together to some EU calls. For instance, the EMMC presently benefits from a Coordination and Support Action. When a call is issued, some European members of the OPTIMADE consortium will be involved in the new proposal. In the US, OPTIMADE has been in touch with the NIST Research Data Framework and the Materials Research Data Alliance (MaRDA).

5 Will these developments bring societal benefits?

Given the challenges that it is facing, our society will greatly benefit from all the improvements that materials design can bring. Indeed, many applications related to the energetic transition require specific materials with targeted properties (e.g., denser batteries, photovoltaics, thermoelectric, catalyst, ...). The OPTIMADE API is an important advance in this field. Thanks to it, the users are now able to interrogate many different databases with the same query. Thus, they have access to much more materials knowledge without the need to learn a different API for each database. Furthermore, information from multiple databases can now be exploited for machine learning methods. The OPTIMADE API will also avoid replicating calculations by different databases allowing them to spend resources broadening the bounds of materials knowledge.

The social benefit is thus evident, though it is indirect in the sense that the OPTIMADE API is a tool that will contribute to making it easier to design materials that provide benefits to society.

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Integrative approach to the theoretical study of chromatin

Location: CECAM-ES
Webpage: https://www.cecam.org/workshop-details/43
Dates: May 31, 2022 - Jun 2, 2022

1 State of the art

This workshop was intended to introduce new interactive molecular simulation tools for the multiscale study of DNA (1) analysis tools, software in virtual research environments (http://www.multiscalegenomics.eu/MuGVRE) and workflows that unifies from the 1D to the 3D/4D genomic data (2-3) (http://mmb.irbbarcelona.org/NucleosomeDynamics; http://mmb.irbbarcelona.org/MCDNA), predictions and experimental data. The aim of the workshop is to teach to Ph.D. students and postdoc to work not only with a large variety of multiple resolution simulation methods, but also to use new interfaces and software platforms to fuel their research projects. Emphasis will be made to integrate experimental data. During this workshop, the participants will learn about the biological background, the experimental and the computational techniques to study the DNA, from free DNA to the chromatin complex, and will interactively apply the knowledge acquired using new graphical interfaces and databases (4). In detail DNA is studied from the atomic resolution, using molecular dynamic simulation and analysing them at the base pair level; passing through the nucleosome level (computationally and experimentally resolving MNase maps), to the chromatin level, experimentally by Hi-C, TAD techniques to coarse grained model to rebuild the chromatin structure.

Key References

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*[3] A. Hospital, P. Andrio, C. Cugnasco, L. Codo, Y. Becerra, P. Dans, F. Battistini, J. Torres, R. Goñi, M. Orozco, J. Gelpí, Nucleic. Acids. Res., **44**, D272-D278 (2015)*

2 Major outcomes

During the workshop the different methodologies to study DNA and the chromatin assembly have been discussed. In particular, how to go from the DNA base resolution to the chromatin structure, in a multi-scale, multi-resolution approach. Thanks to the combination of experimental and theoretical expertise from the speakers, different types of experimental data analysis and its integration to build computational models keeping the base pair resolution and containing at the same time all the chromatin folding information were presented. In more detail, the workshop was divided in three days, focused on different levels of the multi-scale, multi-resolution scheme: 1) Atomistic representation of DNA/RNA, with a focal point on the extraction of dynamic and flexibility properties, applied then to protein-DNA recognition (e.g. transcription factors) and also to the development of new parameters for the Coarse-grain algorithms used in higher multi-resolution levels, presented in the following days of the workshop; participants were mainly interested in how to generate and analyse their data (Molecular Dynamics simulations and helical parameter analysis) for modified nucleotides and comparing different simulations or different sequences; 2) Experimental techniques, from the nucleosome positioning to the chromosome structure determination. During the workshop, the

main discussion involved how to integrate all this data, with different resolutions, to build a chromatin model and how the cell conditions could affect the results and the chromatin compaction. 3) Chromatin organization using mesoscopic- coarse grained methods. the different approaches to include the experimental data to build a chromatin model and how the binding of intrinsically disordered proteins (IDPs) could change the nucleosome arrangements and the overall compaction.

On the bioinformatics side, we discussed the different platforms available to integrate experimental and computational tools (Virtual Research Environments - VRE) and the advantages of using GitHub repositories.

3 Community needs

In our field when we study the molecules at atomic level using technique such as molecular dynamic we need HPC resources. Often people that need these resources to study biological systems, and need to write proposals (PRACE, RES) contact us as experts, to be able to take advantage of the computational power they ask for.

The development of platform such as VRE can integrate the experimental results and make them available to the community, not only to experimentalists, integrating analysis tool to be able to use the results for computational aims.

A series of workshops, going from the atomic to the mesoscopic level integrating all the developed platforms and tools at the experimental and computational level will be very useful. These workshops will go from the bioinformatic to the biology and could be of interest to a wide spectrum of scientists. Having the bioinformatic tools integrated in VRE, Githubs and built in workflows allows the experimentalist to approach the computational studies in an easier way. The workshop was a summary of all this multiscale analysis but the field would benefit from a series of workshops where everything would be analyzed with more time and details.

4 Funding

Joint research proposals were not discussed during this workshop, although the workshop was designed as a follow-up of a previous workshop assembled by a collaboration between two H2020 European funded grants: Multiscale Complex Genomics (MuG - 676556) and BioExcel Centre of Excellence (BioExcel - 675728). However, as stated above, we think the content of the workshop could be transformed into a series of events introducing the multiscale analyses of the genome with the most updated bioinformatic methodologies and platforms.

5 Will these developments bring societal benefits?

The topics discussed during the workshop are already playing a key role in the health benefit for the community. The importance of the nucleic acid's structures (in particular the dynamics and flexibility), the impact of the epigenetic modifications on the chromatin structure and the implication of mutations in the chromatin and associated diseases are currently main research topics in the pharmaceutical field. Clear examples are the well-known development of the COVID-19 vaccine as an RNA structure (RNA Messenger -mRNA- vaccine), or the DNA methylation diseases studies (Parkinson, Alzheimer). Developments presented in the workshop have already been part of funding opportunities (H2020 Multiscale Complex Genomics, H2020 BioExcel Centre of Excellence, Ris3Cat Emergent Cluster of the Human Brain), and are expected to be included in future funded projects.

As stated in the previous section, this workshop was in fact organized as a follow-up of a previous workshop assembled by a collaboration between two of these H2020 European funded grants: (MuG - 676556) and (BioExcel - 675728).

6 Participant list

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Quantum Chemistry Methods for Materials Science
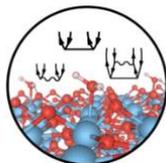
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1116
Dates: Jun 8, 2022 - Jun 10, 2022

1 State of the art

Density-functional theory (DFT) has been the method of choice for electronic-structure calculations in materials science over many decades. However, certain well-documented failures such as unsatisfactory prediction of atomization energies and underestimation of weak interactions and reaction barriers, limit the predictive power of current density functional approximations, including (semi-)local and hybrid functionals in materials science.
The desire for general-purpose electronic-structure methods with high accuracy is pressing, especially in materials science. Together with the rapid growth of computational capacity, this has drawn attention to the sophisticated quantum-chemistry methodologies rooted in wavefunction theory. The improvable accuracy together with potentially richer electronic-structure information and the ability to study electronically excited states via the equation of motion formalism make them very promising in materials science. In this context, the central goal of the workshop was to discuss the state of the art and challenges of using quantum chemistry methods in materials science, to share the recent progresses in quantum chemistry, and to deepen the coalescence of two communities: molecular quantum-chemistry and solid-state physics.

In addition to the theoretical topics described above, this workshop did focus on the computer implementation of massive parallel algorithms to perform quantum chemical calculations on modern supercomputers. The evolution of computer architecture towards larger multicore machines partly equipped with GPUs makes it necessary to adapt existing simulation software and employ libraries tailored to run ab initio calculations efficiently on modern hardware. This workshop will bring together some of the world's leading experts in the development of massively parallel algorithms for quantum chemistry calculations to foster cooperation and catalyze scientific software innovation.

2 Major outcomes

The topics discussed at this workshop can be roughly categorized as follows:
1. Scope of highly accurate quantum chemical wave function theories in materials science
2. Different approaches to simulate materials
3. Technical challenges of massive parallel computer implementation of quantum chemical wave function theories

Regarding the scope of highly accurate quantum chemical wave function theories in materials science, this workshop had several presentations that showed the substantial progress of these methods in the last few years. Both the number of applications and the accessible system sizes have also increased significantly. Furthermore, these methods give access to a growing number of properties in materials. While most applications were limited to ground state properties 5 years ago, nowadays there exist many implementations and applications that seek to predict excited state properties. In particular, excitation energies for optical spectra and band structures become accessible. However, it was clear from the discussions at the meeting that their remain many challenges to be addressed by the community, e.g. the convergence of excited state properties with respect to computational parameters.

Since this field of research is situated at the crossroad of chemistry and physics, the community employs different approaches when applying wave function theories to materials. This is reflected by the different basis sets, boundary conditions and computer codes employed. However, each of these approaches has their advantages in certain situations, e.g. molecular crystals or metals are challenging cases for plane-wave or atom-centered basis set type approaches, respectively. At the same time, it is important for the entire community to be able to cross-validate and test their approaches by studying well-understood benchmark systems. Many of these and related points were discussed by the attendees of this workshop. Another important topic of the present workshop concerned the implementation and massive parallelization of quantum chemical wave function theory calculations. The change of modern hardware and parallel programming paradigms due to the growing importance of accelerators such as GPUs possess a challenge to computational chemists and computational physicists. This workshop brought together computational scientists from both academia and industry to foster the discussion and exchange of ideas.

3 Community needs

The needs of the community at this workshop (computational quantum chemists and quantum physicists with a focus on materials science) in terms of computational infrastructure is well aligned with most communities represented by CECAM.

Overall the continuous development of novel and improved theoretical methods, growing challenges possed by more complex materials and increasingly available computational resources demonstrates that this research community is in need of more networking and workshop activities. These activities help to concert the joint efforts in a more efficient manner and address challenges in a more collaborative manner. On the one hand it is recommendable

to embed these activities in the wider research communities. However, on the other hand, it is equally important to further strengthen joint intra-community activities. For these activities future CECAM workshop would serve as an ideal setting and such plans were already discussed at the present workshop.

4 Funding

Potential joint research proposals and collaborations were discussed amongst different workshop participants. These included Horizon 2020 projects and European centers of excellence for HPC projects. Furthermore, EURO HPC JU compute time proposals were also discussed.
It should be noted that the discussion of joint funding opportunities is an activity that could be emphasized more strongly in a future version of this workshop. The focus of most discussion was mostly about the scientific aspects of the research topic.

5 Will these developments bring societal benefits?

Computational materials science is expected to contribute substantially to the development of novel or better materials that fulfill key tasks in chemical processes or devices. Due to the growing need for energy and resources, our society benefits significantly from improved theoretical and computational methods that can be used to study and predict materials properties using computer simulations. Although computational materials science was already incredibly successful in the last decades, it remains a challenge to predict materials properties with controllable accuracy. Addressing this challenge is the goal of the methods, which were at the center of this workshop. We have experienced that these methods make significant progress and are expected to fill this gap in the current toolbox of practicing computational materials scientists in the foreseeable future. As such the topics and developments discussed at the present workshop will bring societal benefits even if they should still be considered as fundamental research.

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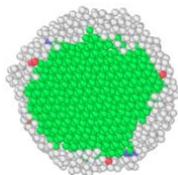**Goswami, Rohit** - University of Iceland, Iceland
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Current Challenges in Materials for Thermal Energy Storage

Location: CECAM-ES
Webpage: https://www.cecam.org/workshop-details/1159
Dates: Jun 8, 2022 - Jun 10, 2022

1 State of the art

Human activities produce vast amounts of energy. Being able to store energy more efficiently would have far-reaching economic and societal impacts. These objective drives efforts to design Thermal Energy-Storage Materials (TESMs). TESMS store and release energy via ´sensible heat' or the latent heat of a phase transition. The latter ensures that the uptake/release of energy happens at well-defined temperatures. TTESMs design relies primarily on empirical knowledge, targeting a few observables – melting enthalpy & temperature or the Thermal Conductivity (TC), albeit with limited physical insight. The TC influences heat-transfer dynamics substantially, and interfacial heat transport imposes additional dynamic constraints.

Computer simulations (CS), classical and ab initio add much value to laboratory studies. CS provide insight into barrierless growth kinetics, challenging classical nucleation theory. CS of phase-change materials revealed new kinetic scales, not accounted for by classical heat-transfer theory, and provide a toolbox to study thermal transport mechanisms or phase coexistence. However, most simulations do not focus on TESMs, making it difficult to benchmark CS against experiments.

There are yet-to-be-tapped synergies in TESMs by bringing together CS and state-of-the-art radiation-scattering techniques (neutrons and X-rays), which have evolved significantly

(European Spallation Source in Sweden; XFELs in Germany, Italy or Switzerland). These techniques have not been used widely to study TESMs and they go well beyond thermophysical properties or time-averaged observables. The dynamic structure factor can be interrogated with neutrons and X-rays, providing insight into thermal-transport mechanisms in the search for new materials: molecular media, barocalorics, or carbon-based nanofluids. Simulations and experiments can work in tandem, to identify key experimental observables and assist with the interpretation of experimental results.

2 Major outcomes

The workshop provided an excellent and timely overview of the advances and limitations of computational approaches to investigate TESMs. While machine-learning methods are becoming extremely popular by providing high-quality, accurate data for complex calculations (particularly for costly ab-initio methods), there is still room for improvements in this area by selecting more representative training sets, whilst not losing the physical insight provided by traditional computational approaches. This can become frustrating, not only in this area but in general, as future developments would need to be placed on firm ground based on well-defined physical concepts.

The experimental exploration of materials has advanced considerably, and it is possible to measure properties at ultra-high heating rates in bulk materials. However, there are still challenges in the study of the thermophysical properties of interfaces. Experimental work can profit from computer simulations, as the latter are particularly suitable for investigating interfaces of nanoscopic dimensions. Untapped synergies between experimental and simulation were identified, e.g. obtaining information from neutron scattering on anharmonic effects and spectroscopic measurements to benchmark new computational models. The experiments also revealed important information on the dynamics of fluids in nanoporous materials and nanomechanics. Regarding experimental development, we would like to mention also emerging opportunities with last-generation spallation neutron sources and XFELs – e.g., extended coverage of length and timescales, high temporal resolution, coherence. There are good opportunities in these areas for computational studies to assist, interpret and advance the use of these powerful experimental techniques.

During the workshop, we also discussed the community's capabilities to manufacture and characterize new materials. The community has acquired a great deal of expertise in making carbon-based media, and experimentalists can control the composition of phase-change nanocomposites and, in general, the properties of nanostructured materials with increasing precision. As a result, new opportunities in the area of phase transitions of carbon-based materials could be explored, as well as in the area of multifunctional materials. Similarly, plastic crystals and (more generally) dynamically disorded condensed matter offer exciting avenues, such as barocalorics, which were also discussed at the workshop.

Rigorous ab-initio methods can provide unprecedented details on the behaviour of complex materials, such as semiconductors, which are of particular interest in the area of thermoelectric devices. The theoretical techniques offer insight into bonding models and metavalency, which should help develop and improve new materials. Classical models also provide insight on fundamental aspects giving further inspiration to understand the basic interactions that control the behaviour of solutions under equilibrium and non-equilibrium conditions, as well as methods to study complex nucleation processes to rationalize observed nucleation rates. The interaction between ab-initio, classical computations and practical approaches provides a unique toolset to explain experimental data and help discover new TESMs.

3 Community needs

The investigation of TESMs requires significant computational effort due to the needs associated with simulating sufficiently large systems that are representative of the microstructure of the materials in question. It is a genuine multiscale problem where the concurrent use of ab-initio, classical and continuum models is needed to characterize the thermophysical properties of the material and its associated phase behaviour. However, it also involves specific aspects connected to phase transitions and the need to consider interfaces explicitly, which further motivate the use of atomistic-based methods. At present, the community benefits hugely from highly parallelized codes such as LAMMPS, DIPOLY or Gromacs, all of which have contributed to the investigation of increasingly complex systems using more sophisticated models. There is a need, however, to increase the level of communication between experimentalists and simulators to profit mutually from the information that can be obtained using the different approaches, as well as to focus together on well-defined challenges and classes of materials. The workshop provided an excellent example of how this interaction can work by simulators showing the capabilities of computational methods to experimentalists. Reciprocally, experimentalists presented current and emerging opportunities to use state-of-the-art radiation-scattering techniques at large facilities to study TESMs, as well as how these efforts are linked to the design and development of new materials in the laboratory. There is a need to make these interactions less "random" and sporadic. We felt that this latter point is particularly important in the area discussed at the workshop, where such integration is not as strong as it should be.

4 Funding

Traditional funding channels include European funding and funding from national research agencies. Currently, the situation of some countries, specifically the UK, is unclear regarding EU funding, and mechanisms in this direction need to be investigated to facilitate trans-national collaborations. Additional funding opportunities might come from research projects involving experimental studies at large facilities, providing simulators unique opportunities to engage face-to-face with experimentalists and adapt their computational approaches to help interpret the experimental data and guide future efforts. Regular calls at, e.g. ILL, ESRF, ISIS, or DIAMOND, or the use of the EU-funded EUSMI network could be exploited for future collaborations in TESMs. During the workshop, there were informal discussions on potential research projects. Hopefully, some of these will materialize into concrete submissions to the relevant funding agencies in the near future.

5 Will these developments bring societal benefits?

As discussed in the original proposal to CECAM leading to this workshop, human activities produce and require vast amounts of energy, and being able to store energy would bring significant benefits to society, potentially reducing costs. This is particularly important to achieve carbon-neutral technologies, and energy recovery has an essential role in this context, as there is a need to enhance efficiency and reduce the economic cost associated with developing the relevant materials.

The discussions in the workshop revolved around methods for materials exploration. These methods could bring additional benefits by reducing the costs associated with materials discovery and result in new strategies that experimentalists can use to design novel approaches to synthsize new materials in the laboratory. In addition, there are essential synergies between simulation and experiment that we can exploit to design more efficient and sustainable materials to maximize energy storage in a cost-effective manner.

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Defects in solids for quantum technologies

Location: Stockholm University, Stockholm, Sweden
Webpage: https://www.cecam.org/workshop-details/1125
Dates: Jun 12, 2022 - Jun 17, 2022

1 State of the art

Quantum devices of the next generation are expected to actively create, manipulate and read out quantum states of matter. This may revolutionize information and communication technology, and also has great potential for life-science and medical applications. Considerable effort has been spent to develop a basic unit of quantum information processing (or qubit) from different individual quantum systems, such as single atoms or ions trapped in a crystal lattice, single Josephson superconducting devices, single photons emitted from quantum dots or single photons/spins associated with point defects in semiconductors. However, most of these systems can only operate at cryogenic temperatures or in ultra-high vacuum. Quantum states due to point defect in wide band gap semiconductors may realize single photon sources and quantum bits that can be harnessed in quantum information processing and nanoscale sensor applications at room temperature.

The leading contender is the nitrogen-vacancy center in diamond that may be considered as a robust quantum bit. However, the possibility to realize bright single-photon emitters and single spin sources (single defects with spin) in SiC, Si, and hBN have been demonstrated. Researchers face many materials science challenges in fabricating point defect quantum states with favorable intrinsic properties that can be perturbed by other defects either in bulk or at the surface of the devices. First principles theoretical simulations have been demonstrated as an essential tool in understanding the underlying physics of these atomic scale systems as well as in identification of potential new quantum bits and single photon emitters in wide band gap semiconductors. Therefore, tight collaboration of experimental research and atomistic simulations is essential for a rapid progress in the field.

2 Major outcomes

Before discussion the scientific outcome, let us briefly summarize the statistics of DSQT22 Flagship workshop. Including the Organisers, 119 participants from 21 countries registered for the workshop. Vast majority of participant showed up in person at the workshop site and attended the scientific talks. Selected invited speakers, e.g. from countries with remaining hard travel restrictions, were able to make their presentations on-line. The final scientific program of the workshop lasted for five full days, included 34 invited talks (10 female presenters), 48 contributing talks (8 female presenters), and a poster session with 30 posters (5 female presenters).

DSQT22 workshop succeeded to bring together world-leading theoreticians and experimentalists active in the field of quantum devices based on solid state qubits. Our discussions improve interdisciplinary cooperation overcoming traditional boundaries between scientific disciplines. Invited speakers at DSQT22 workshop represented electronic structure theory community, theory at phenomenological and atomic scale level, materials growth, defect engineering, surface chemistry, quantum optics and spin physics. Recognized leaders in the field, like Giulia Galli (University of Chicago), Chris Van de Walle (University of California), David Awschalom (University of Chicago), Igor Aharonovich (University of Technology Sydney) and early career researchers, like Marina Radulaski (University of

California), Lukas Razinkovas (Center for Physical Sciences and Technology, Lithuania), Daniil Lukin (Stanford University) and Joel Davidsson (Linköping University) highlighted recent progress and discuss challenges and opportunities in their invited talks. Invited talks, contributed talks and posters stimulated the exchange of methodological expertise and new developments between scientists working on different aspects of the field, illuminate opportunities for optimizing the materials properties and device design aided by theory. Discussions during the breaks provided opportunity to form new interdisciplinary collaborations on solid state quantum bits for the mutual benefit of theoretical, experimental and applied researchers, especially following talks demonstrating the potential of this field in various interdisciplinary areas, such as biosensing presented by Romana Schirhagl (Groningen University) and potential improvements in nano-NMR covered by Daniel Cohen (Hebrew University of Jerusalem).

The workshop program included scientific sessions on NMR with spin defects, Growth and defect fabrication in SiC, Advances of the field of 2D and 3D materials, Deterministic defect fabrication for photonics and implantation defects, Photoionization and recombination of NV centers, Photo-physics and electrical readout of defects, Theoretical and experimental spectroscopy of point defects, Automated defect studies and databases, Quantum control of defect spins, Spin physics and spin-environment couplings, Quantum defects in silicon, Decoherence and spin relaxation, New approaches in ESR, First-principles theory of point defects in semiconductors for quantum applications, Sensing with point defects, Defects in wide-band-gap semiconductors for quantum technology and Defect based photonic applications.

In summary, based on numerous feedbacks from the participants, we conclude that the DSQT22 workshop goals are achieved. Fulfilment of the meeting objectives ensured the benefits of the research community and promoted numerous international research collaboration. The interdisciplinary character of the workshop helped finding solutions for challenging scientific questions in the field of defects in wide-band gap semiconductors for the next generation quantum technologies.

3 Community needs

The study of excited properties of point defects is a notoriously difficult task due to sizable supercell models used for modelling individual defects as well as the limitations of DFT based approaches for predicting excited state properties. At the forefront of the field, various embedding theories have been developed that combine hybrid-DFT, cRPA, and CI. In these methods hybrid-DFT is used to describe the delocalized states of the host wide band gap semiconductor, cRPA is used to describe the screening effects of the host material on the Coulomb interaction of the localized defects states, and CI is used to describe the spectrum of the localized defect states. Different flavors of this approach have been tested on the NV center in diamond, which serves as a test bed for method development nowadays. In order to evaluate the true potential of these methods, comprehensive benchmarks are needed that requires the establishment a benchmark set of experimentally well characterized color centers in various wide band gap semiconductors.

High throughput computational and experimental studies were unanimously identified as a promising direction of future development. Handling and analyzing the data pose challenges that requires a versatility and unified solution from the community. Especially, integration of experiential data together with computational results needs more attention. Experimental colleagues seem not to be aware of the solutions developed by the computational physics community, therefore the crosstalk between these communities needs to the further strengthened.

4 Funding

In addition to CECAM and the Psi-k Network, Stockholm University and Linköping University also significantly contributed to financing our conference. Furthermore, funding from the Knut and Alice Wallenberg Foundation was essential for the organization of the conference. It is important to underline that the use of these sources was possible due to having the conference in Stockholm.

Within Horizon Europe not only European Research Council but also various programs of the European Innovation Council (EIP) could be considered for applications for funding for future conferences. Current studies in the field often possess great innovative potential and generate industrial interest that may help acquiring funding from EIP.

5 Will these developments bring societal benefits?

Semiconductor technology lies at the heart of modern industry, science and communication networks. Its vast importance is given by the versatility of semiconductors as power electronics, information processors, sensors and mechanical devices. Keeping the current pace of increasing the information processing power or the number of components in a microprocessor, the bit – the basic unit of information – will have to be stored in atomic- and subatomic-scale systems, such as individual atoms, ions, electrons or photons, whose behavior is governed by the laws of quantum mechanics. Sensors on the atomic scale are also required for monitoring single molecules. The transition from conventional technologies to quantum technologies is therefore unavoidable. To develop a basic unit of quantum information processing based on point defects in semiconductors has huge potential for applications. Spins associated with defects in semiconductors, such as the complex of a vacancy and a nearby N atom (NV center) in diamond, have shown excellent optical and spin properties suitable for room-temperature qubits. Recently, coherently controlled single electron spins associated with the divacancy and the Si vacancy in silicon carbide (SiC) have also been engineered, demonstrating that SiC can combine its well-developed semiconductor technology and the best of diamond, allowing for a single optically addressable spin, with long coherence time even at room temperature, embedded in a high-performance electronic material. Scientific and industrial developments in this direction may ultimately lead to the realization of quantum applications operating at elevated temperatures, which may eventually enable the widespread of quantum devices and influence our society in many respects.

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Atomistic simulations of interfacial processes in energy materials
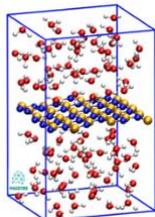
Location : CECAM-FR-MOSER, Sorbonne University Pierre et Marie Campus Paris France
Webpage: https://www.cecam.org/workshop-details/1136
Dates: Jun 13, 2022 - Jun 15, 2022

1 State of the art

Over the past decades, the increase of the efficiency of atomistic simulation methods enabled the very efficient characterization of materials for energy applications, both for bulk properties and for interfacial processes. In particular, density functional theory and molecular dynamics have become essential tools for the interpretation of experimental results, and they are now involved at the initial stages for prediction purposes. In the case of energy applications, simulations have for example played a key role in:
-characterizing the electronic structure of Li-ion battery electrodes [1,2] and suggesting new cathode materials [3] or solid-state electrolytes [4]
-providing useful descriptors for ranking materials for catalyzing the reactions involved in the electrochemical production of fuels [5] and exploring the impact of the liquid structure on the energetics of water reduction [6]
-understanding the capacitive energy storage mechanisms in nanoporous carbon electrodes [7] and metal-organic-frameworks [8]
-identifying key chemical species involved in the transport of CO_2 inside molten carbonate fuel cells [9]
The conventional methods are now supplemented by machine-learning tools, that can increase further the accuracy of the predictions and the number of studied systems in future years [10].

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2 Major outcomes

The first day of the workshop opened with three of the most prominent experts of ab initio simulations, M. Tuckerman, B. Kirchner and G. Galli, presenting recent methodological developments in the study of water under confinement and at interfaces of interest for energy matierials, by means of DFT-based molecular dynamics. Among the key points addressed we

mention hydronium and hydroxide diffusion in exchange membranes for fuel cells, and ion clustering in ionic liquid electrolytes. The first day closed with flash presentations and a poster session.

The second day started with the presentations of young researchers, C. Zhang and L. Scalfi, who discussed new approaches to model the polarization or metallicity of electrode surfaces and the formation of a protonic double-layer at the electrode-electrolyte interface. Structural aspects as well as free-energy calculations were addressed. The session ended with a presentation of M. Sulpizi about numerical techniques to compute sum frequency generation spectra at water interfaces, including equilibrium and non-equilibrium effects.

The following session focused on a long-standing challenge in computational science: the prediction of crystal nucleation mechanisms and rates. J. Rogal and P. Bolhuis made important contributions to this field in recent years, and discussed the delicate interplay between enhanced sampling techniques and the definition of reaction coordinates, an open problem. The role of precursors and temperature was investigated with respect to polymorphism.

In the afternoon, M. Iannuzzi discussed ab initio molecuar dynamics investigations of osmotic transport at interfaces between hBN or graphene and water. A combination of enhanced sampling methods and modeling of equilibrium simulations were employed. C. Dellago introduced the important of how machine-learning potentials trained on DFT calculations can contribute to simulate larger systems for longer times: results on bulk and interfacial water described via neural network potentials and Behler-Parrinello descriptors are quite impressive. The day was closed by S. Cox, who tried to resolve controversies in the determination of the dielectric response of water films, and by S. Sgroi who presented an ab initio protocol for the study of battery materials in a manufacturing company.

During the last day, several aspects introduced in previous sessions were further discussed, such as the properties of nanoconfined water, in particular its phase diagram from first principles (V. Kapil, B. Cheng), electrode/electrolyte interfaces with new developments for classical molecular simulations (S. Bonella) and reactivity (L. Giordano), or nucleation (B. Cheng, J. Lam). Some of these contributions highlighted the increasingly important role of machine-learning tools. T. Schilling and R. Semino discussed the challenges for the prediction of thermodynamic and transport properties in composite materials. Finally, B. Cheng and P. Robin presented new simulation strategies and new applications of molecular simulations to understand transport properties, with illustrations on heat transport in bulk fluids or ion transport in nanochannels.

3 Community needs

This workshop has brought together researchers from quite different communities of atomistic simulations, and has thus shed light on the different approaches adopted to undertake the complex study of interfacial problems, typically ranging through different time- and length scales. Those communities generally use different methods and codes, ranging from electronic-structure/DFT ones to advanced sampling/molecular dynamics-based ones, also including new computational tools based on artificial intelligence/machine learning approaches.

If the mutual interest and intellectual flexibility of researchers from the different communities allows them to constructively interact with one another, this workshop has shed light on the fact that a significant added value of broad interest would (or will) be achieved by bringing more systematically those communities together, at first, and then by helping to interface, in a professional manner, those codes. Creating specific workflows capable, for a given system or problem, to perform different kinds of calculations at different length/timescales and possibly, if interesting, keep track of the generated calculations data for potential machine learning training seems to be one of the main needs of the atomistic community interested in interfacial systems.

4 Funding

There are now many sources of funding on this topic, both at the national and international scale. This can be seen from the development of consortiums such as the Materials Genome Initiative (https://www.mgi.gov/) in the USA or MARVEL (https://nccr-marvel.ch) in Switzerland. The organisers of this workshop have recently created the MAESTRO (Materials for Energy through STochastic sampling and high-peRformance cOmputing) consortium, at the Institut des Sciences du Calcul et des Données (ISCD) of Sorbonne University in Paris, combining mathematicians, physicists, chemists, and computer scientists, precisely to provide multidisciplinary and transdisciplinary insights to these challenges. At the European level, specific calls such as the ones opened within Batteries 2030 offer many opportunities.

5 Will these developments bring societal benefits?

The social impact of energy materials is huge: The energy transition aims at replacing fossil fuels by greenhouse-gas-free resources. The future energy mix will certainly include larger shares of renewable sources, mainly wind and sun. All these energy productions involve the use of advanced materials: Wind farms extensively use magnetic materials and the next generation of solar cells will probably involve perovskites. In addition, renewable energy sources suffer from strong intermittency. Even on a country scale, the fluctuations of wind and solar energy cannot phase well with the population needs, which induces the need to use large shares of coal or gas power plants to complement them. To solve this problem, the main solution would be to enable electricity storage using batteries or conversion towards hydrogen fuel through the use of fuel cells. In such electrochemical devices, the materials play a central role as well. For decades, the development of these materials was based on experimental approaches only, but now atomistic simulations are one of the most important tools to characterize the systems and optimize their performance. However, the case of interfaces still requires important developments, following which it will be possible to propose new strategies for enhanced batteries, fuel cells or solar cells based on computer simulation initial results.

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Challenges of molecular spectroscopy: Theory meets experiment
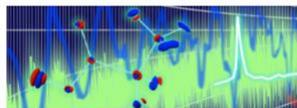
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1034
Dates: Jun 13, 2022 - Jun 17, 2022

1 State of the art

Advanced spectroscopic techniques have given us detailed pictures of molecular structure and properties, and served as the driving force for the development of new theories and approximations for quantum dynamics. Yet, at the moment, there is no theoretical method capable of exactly reproducing spectroscopic experimental results fully from first principles; each method makes a more or less severe approximation to the Schrödinger equation [1]. It is of fundamental importance to understand if and how approximated theory can still meet experiment.

The fields of infrared and rotational spectroscopies are expanding quickly, and these traditional methods are being implemented into various new techniques [2, 3]. Special chemical physical conditions can be set up for such experiments [4, 5], which can be, in principle, reproduced in theoretical simulations [6-12]. However, limitations exist due to the errors in the potential energy surface and quantum dynamics; only an experimental validation can provide the necessary assessment. Electronic spectra pose other challenges [13], including the necessity to describe excited electronic states and their potential energy surfaces. Going beyond the traditional harmonic method [14], many research groups nowadays account for the anharmonicity with vibrational perturbation theory [15, 16] or with semiclassical trajectory-based approaches combined with on-the-fly ab initio evaluation of the potential energy surface [17-22]. Other participants of the workshop made important contributions to the treatment of solvent effects on condensed-phase spectra [23-25]. As time-resolved spectroscopies are becoming almost routine, more recently, 2D spectroscopies interrogating both nuclear and electronic motion have come to the lime-light [26-30]. Vibrational and electronic coupling can be monitored, whereas linear spectroscopies do not easily reveal microscopic details of the underlying processes in the condensed phase.

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2 Major outcomes

The workshop served as a meeting point for theorists working in the field of computational spectroscopy and experimentalists developing state-of-the-art laser set-ups. Here, we list several topics discussed during the workshop and conclude with some of the main points common to all topics.

On the theoretical side, speakers presented developments of new computational methods for simulating steady-state and time-resolved spectra in the terahertz, mid-infrared, visible, ultraviolet, and X-ray regimes, involving both vibrational and electronic transitions in molecular systems. Presented methods included, for example, semiclassical wave packet-based techniques, phase-space mapping methods, thermofield dynamics with Gaussian wave packets or tensor trains, or variational and perturbation theory-based methods for vibrational wavefunctions. Alongside these, known methods, like the nuclear ensemble approach or classical molecular dynamics were tested in new contexts.

Experimental work presented in the workshop involved time-resolved electron imaging techniques, attosecond pump-probe experiments, ultrafast X-ray spectroscopy, and terahertz spectra of liquids and hydrogen-bonded clusters. A need for theoretical modeling was emphasized both during talks and in formal or informal discussions. Whereas the theoretical description of these spectroscopies exists, first-principles modeling is still a challenge, as noted by many participants.

To circumvent the high computational cost of accurate electronic structure and exact quantum dynamical calculations, most participants proposed semiclassical or classical-like, trajectory-based methods. These allow for an on-the-fly implementation, in which the electronic structure problem is solved only where needed along the classical trajectory and a full potential energy surface construction can be avoided. Also, as emphasized by some of the participants, apart from being practical, these methods often reveal a more intuitive picture compared to full quantum solutions. Thanks to the vibrant ambient and common interests in these topics, the participants mutually questioned the proposed computational methods, subjecting them to scrutiny. This led to questioning best/worse-case scenarios for different methods, their utility and failure modes, as well as their comparison to other existing approaches. Questions and answers exchanged between experimental and theoretical researchers have been very common at the end of each talk. This helped the audience to understand better the development of experimental interpretations and the limits of each computational approach.

3 Community needs

In the workshop, we brought together experimentalists as well as theorists that are leading experts in the fields of rovibrational, infrared, visible-ultraviolet, and other spectroscopies (including both linear and ultrafast subfields). As reported above, spectroscopic techniques are nowadays most often articulated as a set of different methods which go beyond the traditional optical setup. It is, therefore, essential to stimulate the interactions and collaborations between the theoretical and experimental communities in order to facilitate the advance towards the common objective—a better understanding of light-matter interactions in general. In our view, the theorists participating in this type of workshops get inspired by the newest experimental techniques and, vice versa, the experimentalists become aware of the recent developments in quantum dynamics and related theories used for spectra calculations, including the limitations and accuracy of these methods.

Discussions of these challenging and sometimes controversial questions were enabled by the specific format of the CECAM workshops, which encouraged an open discussion of "what *does not* work", in contrast to the more standard conference-type contributions composed of successful examples only. Workshop's size and structure, including a high fraction of time dedicated to discussion, social dinner, and excursion, generated many informal and in-person interactions, which were particularly appreciated by the numerous students and post-doctoral fellows attending the workshop. Therefore, we believe a similar workshop to ours should be held from time to time, as both theoretical and experimental techniques are updated.

4 Funding

Fundamental research, which was the focus of this workshop, is eligible for various funding schemes, including the Horizon Europe and national grants. Indeed, many of the workshop participants were or still are holders of such grants, including the prestigious European Research Council funding. Joint research proposals were not discussed in the workshop. Nevertheless, the open discussion inspired new ideas and will certainly result in further collaborations or joint projects. This aspect has been also important for the numerous students attending the workshop, since it helped them to understand that research is made not only of interesting new results but also of exploiting suitable funding schemes.

5 Will these developments bring societal benefits?

By addressing some of the most fundamental questions in chemical physics, such as the correspondence between the quantum and classical worlds or the nature of interatomic and intermolecular interactions, this workshop improved our understanding of state-of-the-art methodologies related to molecular spectra. By verifying computational models with both theory and experiments, researchers learn about intrinsic properties of molecular systems. These, in turn, find applications in the design of new drugs, functional materials, or sustainable energy sources and storage. Furthermore, it is known that molecular spectra, especially recent ultrafast techniques, provide insight into key quantum-mechanical phenomena for the development of quantum computers, namely, the processes of environment-induced decoherence and energy dissipation. Therefore, it is not surprising that some of the biggest industrial leaders in this field, including Microsoft, IBM, and Google, are actively developing their own research departments for computational and theoretical chemistry. These could also serve as potential funding sources for fundamental chemistry and chemical physics research. More generally, the direct interaction between experimental and theoretical researchers allows them to better shape the future developments of their research mutually and, in this way, increases the positive impact of spectroscopy on people's everyday life.

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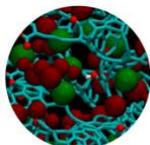**Tommasini, Matteo** - Politecnico di Milano, Italy
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Modeling adsorption in microporous carbons: Bridging methods and crossing boundaries between applications

Location: Meriadeck library, Bordeaux, France
Webpage: https://www.cecam.org/workshop-details/1142
Dates: Jun 14, 2022 - Jun 17, 2022

1 State of the art

Microporous carbons are an important class of materials, present as natural or artificial compounds in many industrial and technological applications including hydrocarbon recovery, gas storage, water purification and energy storage. In all these applications knowing the structure of the carbon, both in the pristine state and under operation, usually involving adsorption/desorption of species, is key to understanding and predicting the performance of the process. The accurate characterisation of microporous carbons and their evolution upon adsorption/desorption presents a number of challenges associated with the disordered nature of these materials and the Multiphysics, and often multi-scale, character of the processes at play.

In a context where experiments become more and more complex, with in particular the rise of in situ techniques, and where understanding and predicting materials behaviour and performance in given applications is crucial, this workshop focuses on modeling adsorption in these microporous carbons. The modelling process involves i) the generation of carbon structure models, ii) the simulation of adsorption and iii) the determination of relevant properties; all of these steps raising specific challenges. In many of the applications mentioned above, several phenomena occur concomitantly. While couplings between chemistry, mechanics, adsorption and transport in microporous carbons are present, they still remain often neglected in simulations.

One of the main motivations for this workshop is to bring together people from different communities focusing on diverse applications in order to get a broad view of the current simulation methods used in distinct fields, discuss similarities and disparities between applications and identify ways of improving techniques.

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2 Major outcomes

Questions this workshop aimed to address were both technical and fundamental:

- How to best model adsorption in porous carbons (interactions, statistical mechanics, algorithms ...)?
- Can we develop unified interaction potentials adequate for both generation of carbon structures and simulation of fluid adsorption?
- What are the differences and similarities between gas and ion adsorption/desorption?
- How does the carbon flexibility affect charge storage mechanisms and performance in supercapacitors and other electrochemical energy storage systems?
- Is there a way to model the interplay between electronic structure and mechanical properties of the porous carbon?
- What can models bring to the understanding of electrochemical CO_2 capture and geological CO_2 sequestration?

A number of talks and discussions focused on experimental to probe the structures of microporous carbons, the adsorption of fluids in these materials and the transport properties of the adsorbed molecules. Some of the techniques presented provide very precise information: the surface force balance brings insights into the adsorbed fluid structure under strong confinement through the identification of characteristic lengths, and nuclear magnetic resonance experiments and small angle X-ray scattering allows one to quantify the amount of fluid adsorbed in a porous carbon with different experimental conditions and characterise its dynamics. Nevertheless, all techniques have limitations, and the deep interpretation of the results requires the use of models and simulations. The determination of pore size distributions from adsorption isotherms for example, although applied for many years, still poses challenges: seemingly inconsistent adsorption isotherms obtained for different probe molecules can constitute an opportunity to better understand the structure of microporous carbons.

One interesting point made during the workshop regarding models is the fact that different approaches can be followed: conceptual models and simulation models. Conceptual models aim at predicting or generalising properties for a range of systems, examples of these included the use of free volume theory to estimate diffusion coefficients and the related idea of

calculating diffusion coefficients in hierarchical porous carbons knowing the densities of the micro- and macro-porous parts and the corresponding diffusing coefficients. In some cases, simulation models are needed. The workshop was an opportunity to discuss the progress of machine-learning based force fields to generate atomistic structures. While it is currently not possible to conduct simulations including adsorbed species with these models, research is active to explore this possibility. Research results obtained using more classically developed force fields confirmed, if it was needed, the power of atomistic simulations to explore molecular phenomena at carbon / fluid interfaces. In that regard, the recent generation of very large (1 million atoms) structures opens new doors for simulations and link with experiments. Regarding the simulation of electrochemical systems, there were interesting discussions on various ways to include the semi-metallicity of the electrode materials in the models. In addition to conceptual and atomistic models, other models and methods such as a "platelet" based model and classical DFT were presented. Recent developments in classical DFT including i) the idea of a mixed continuous, for the fluid, and atomistic, for the solid, view of the interface and ii) time-dependent DFT open new perspectives for the fields of research of interest in the workshop.

Overall, the workshop was a great opportunity to discuss recent developments in methods and models, but also and occasion to realise that simulating and understanding multi-scale and multi-physics systems is still a real challenge.

3 Community needs

This workshop was an opportunity to discuss the advantages of using atomistics simulations of microporous carbons and devices to understand some of the molecular phenomena at play and interpret experimental results. These atomistics simulations require the use of HPC resources, thus a necessity in this field of research. A number of codes and force fields are developed and available to the community: force fields to represent the carbon flexibility during simulations and algorithms to include the semi-metallic character of the carbon materials are examples of tools that need to be widely available. Efforts in the future could be made to increase the facility to use the state-of-the-art models on the one hand and to conduct multi-scale simulations (at the atomistic and carbon particle scales for example) on the other hand to get a more and more accurate picture of the systems and devices. All participants enjoyed the workshop and agreed that such an event with both experimentalists and modelers focusing on the carbon and microscopic phenomena but not dedicated to an application was missing in the landscape of scientific events. A series of similar workshops could be envisioned to structure this community and facilitate exchanges of codes and resources.

4 Funding

The research in this area is typically funded through national calls (French ANR, German DFG, etc...), through research in companies or through collaborations with the industry (petroleum, energy storage, …), through transnational funding, etc... No session was dedicated to joint research proposals but many discussions gave ideas for possible scientific interactions in the future. The discussions have in particular highlighted some complementarity between some of the research groups represented.

5 Will these developments bring societal benefits?

The applications for adsorption in microporous carbons are numerous and related to important societal questions: hydrocarbon recovery, gas storage, water purification and energy storage. Modelling is conducted to understand and predict the properties and performance of microporous carbons with respect to these applications. Following the idea of understanding and predicting, there is always the possible benefit of suggesting and preparing materials with optimised properties, or made in a more sustainable fashion, compared to current solutions to the technological challenges at hand. Developing faster and/or more accurate models is an important step in that respect. It is worth noting that it is potentially beneficial not only for the scientific and technological outcomes. Indeed, more computationally efficient models can be considered more sustainable as employing them would not require large computational resources. Overall, the research topic of this workshop can lead to economic and environmental developments.

6 Participant list

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Challenges in Alzheimer's, Parkinson and Amyotrophic Lateral Sclerosis diseases from experimental (in vitro and in vivo) to theoretical/ computational perspectives
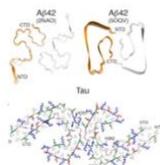
Location: CECAM-FR-MOSER
Webpage: https://www.cecam.org/workshop-details/1131
Dates: Jun 20, 2022 - Jun 23, 2022

1 State of the art

This workshop aims to be a comprehensive, authoritative, critical state-of-the art on protein misfolding and aggregation which is the fundamental cause of more than 20 amyloidogenic diseases affecting either the central nervous system or a variety of peripheral tissues, with disorders ranging from Alzheimer's (Abeta and tau), Parkinson (alpha-synuclein) to amyotrophic lateral sclerosis or ALS (superoxide dismutase, SOD) and type II diabetes (IAPP). Experimental determination of oligomers is challenging because of their transient natures. Simulations of aggregation at different time and length scales can complement experiment, but requires the development of accurate models ranging from all-atom, coarse-grained to mesoscopic or super-mesoscopic representations). Understanding how amyloid aggregates actually become toxic is the challenge in developing a treatment for such diseases. There is evidence from in vitro and in vivo studies indicating crosstalk between different amyloid proteins and interactions between Abeta, tau and alpha-synuclein promote protein aggregation and accelerate cognitive dysfunction.
The central aim of this workshop is to bring together scientists from different disciplines working on different amyloid proteins so as to discuss common and different features between

the diseases and their possible links with a critical and an in-depth overview of our current knowledge on Abeta genesis, amyloid oligomers and formation (kinetics and thermodynamics) and toxicity using a very broad range of biophysical and biochemical methods, computer simulations, liquid-liquid phase separation of disordered proteins, in vivo studies and systems biology.

2 Major outcomes

We have discussed the following aspects:
- Structures of Amyloid-beta assemblies at the beginning and the end, structures of TDP-43 filaments from human brain and modeling and design of oligomeric amyloid aggregates and nanofibrils.
- Probing the principles governing amyloid protein aggregation and fibril formation (a) in the bulk solution using atomistic, coarse grained or minimal models' simulations, and in particular the probability of the amyloid-prone aggregate state or intermediate states, the liquid-liquid phase and the primary and secondary nucleation mechanisms, (b) on the surface of membrane models using all-atom or very simplified protein models, and (c) moving to in vivo conditions.
- From the experimental structure and toxicity side, many interesting results were reported, including the cross-seeding between different amyloid proteins, the search for biologically relevant tau aggregates and elusive alpha-synuclein oligomers, the influence of zinc on modulating aggregation, the conformational ensemble of the prion protein and of superoxide dismutase 1 in the early phases of aggregation, the interaction of amyloid-beta protein with ApoE4, and how cholesterol levels modulate phospholipids' critical micelle concentration driving amyloidogenic proteins' membrane damage and notably the detergent-like model.
- Simulations to understand the stability of amyloid-beta protein fibrils or the disassembly of amyloid fibrils by using laser and ultrasound excitations, and the interaction of insulin-degrading enzyme with small amyloid oligomers. Modulations of gamma-secretase interactions were also discussed.
- An important aspect of the workshop was also aimed at understanding the molecular and cellular systems interactions in neurodegenerative diseases, the design of new drugs and notably structure-based discovery of small molecules that disaggregate tau fibrils from Alzheimer's disease, the challenges in drug development, and the very promising result of of a new secondary structure of amyloid-beta oligomers to diagnostics for Alzheimer's disease.

3 Community needs

The intrinsically disordered proteins (IDPs) involved in neurodegenerative diseases have a few aggregation-prone regions and overall, all IDPs have a low mean hydrophobicity and a high mean net charge. IDPs are structurally flexible and lack stable secondary structures in aqueous solution. When isolated, they behave as polymers in a good solvent and their radii of gyration are well described by the Flory scaling law. The insolubility and high self-assembly propensity of IDPs implicated in degenerative diseases have prevented high-resolution structural determination by solution nuclear magnetic resolution (NMR) and X-ray diffraction experiments. Local information at all aggregation steps can be obtained by numerous experiments, and long-range tertiary contacts or low-resolution 3D information can be deduced from other experiments However, the information obtained from most experimental observables represents an average over the free energy landscape and gives time- and space-averaged properties.

Computer simulations at different time and length scales can in principle provide the dominant microstates of IDPs using multiple sampling techniques and various representations ranging from all-atom and coarse-grained (CG) to mesoscopic models. However, they are limited by

the accuracy of the force field and the size of the energy landscape to be explored. An important issue is to model a system in mimicking in vivo conditions, and very importantly conditions which recapitulate aging conditions.

In the field of amyloid aggregations, people use existing codes such as NAMD, CHARMM, AMBER, GROMACS or develop their own codes. Overall, the community needs more and more CPU and GPU times to sample relevant disease-causing microstates for more realistic systems.

4 Funding

All speakers of the workshop are funded by national research agencies from USA, Mexico, India, China, Italy, France (ANR), UK, Germany and Austria. Some speakers are also funded by ERC grants and contracts with pharmaceutical companies. This workshop will establish new collaborations(a) between computational and experimental scientists, (b) between computational scientists working at different protein model representations and therefore promoting multi-scale simulations, and (c) between experimental scientists to share a bank of oligomers for Abeta, hIAPP, alpha-synuclein, tau, prion, superoxide dismutase 1, and TDP-43 proteins.

5 Will these developments bring societal benefits?

There is still a long road for a treatment of each disease as (1) there is growing evidence from *in vivo* and *in vitro* studies of co-occurring pathologies across common neurogenerative diseases, indicating cross-talk between the amyloid proteins and interactions and cross-seeding between the Aβ, tau and α-synuclein proteins which promote aggregation, generate different strains and accelerate cognitive dysfunction, (2) we still lack efficient biomarker of disease progression, and all diseases have long prodomal phases.

The workshop has identified the main challenges and has ignited new ideas allowing ultimately an efficient treatment and diagnostic of neurodegenerative diseases. New drugs have been discussed but they have not entered into clinical phase trials yet. Very promising results have been presented at the experimental and computational levels to better understand the role of amyloid oligomers in the development of the diseases and for a better diagnostic of normal patients and Alzheimer's disease patients. Health benefits may come out of this research.

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Error control in first-principles modelling

Location: CECAM-HQ-EPFL, Lausanne, Switzerland & online (hybrid format)
Webpage: https://www.cecam.org/workshop-details/1115
Dates: Jun 20, 2022 - Jun 24, 2022

1 State of the art

The determination of errors and their annotation in the form of error bars, is widely established in the experimental branches of chemistry, physics, and material science. In contrast, a rigorous error analysis for first-principles materials simulations is lacking, and hence the results of such simulations are generally quoted without simulation-specific errors. We

consider this a severe obstacle for innovation, given the important role that multiscale numerical simulations now play in chemical and physical research.

The aim of this workshop was to bring researchers from different communities, from quantum chemistry, materials sciences, scientific computing to mathematics, and establish links between these disciplines with the common goal of quantifying errors simulation scales. The themes discussed included: how to go beyond existing benchmarking procedures or explicit convergence studies via an understanding of errors, reliability and efficiency of numerical procedures, as well as the determination of errors in observables, which are the result of long molecular dynamics simulations. We explored interactions with the field of Uncertainty Quantification (UQ) to link (i) numerical and modeling errors in electronic structure calculations, to (ii) the statistical inference of interatomic potentials and to (iii) the error in global/observable quantities.

2 Major outcomes

The workshop provided an overview of the current state of error control and error analysis in first-principle simulations. This included contributions from the aforementioned communities, targeting applications in chemistry, materials science and physics, such as the atomistic simulation of molecular as well as extended systems. Error estimation approaches based on numerical analysis, statistical inference, benchmarking as well as uncertainty propagation approaches were presented. Particular focus areas of the presentations were machine-learned interatomic potentials, the numerical analysis of density-functional theory methods as well as benchmarking techniques under uncertainty.

During the talks it became apparent that error estimation techniques are not developed on an equal level amongst communities. Moreover, their use is not equally prominent. This offered the opportunity not only to learn from the approaches in different communities, but also to exchange on the expected "community standard" of error control. To provide one example: Following up from the Delta project (https://molmod.ugent.be/deltacodesdft), the necessity to systematically compare numerical implementations across codes is now established as a very important tool to validate implementation choices in plane-wave density-functional theory. For the molecular community such a systematic comparison is still missing to the best of our knowledge. On the other hand, benchmarking of (e.g. DFT) models against higher-order reference simulations has been established for many years in the molecular community, while such procedures are seen less frequently in the solid-state context.

Originating from these differences we also noted a differing mindset when it comes to error estimation, in particular between mathematicians and application scientists. As we expected already during planning keeping the workshop accessible across the different communities thus proved to be a challenge. Obstacles we observed were, e.g., (a) differences in the meaning associated to terminology or (b) the different research "speed" across communities originating from their deviating motivation when tackling a problem (e.g., solid mathematical analysis versus empirical understanding of errors). This results in progress to occur at different paces rendering research results in one community apparently inaccessible to the other.

Our approach in this workshop to overcome this common issue in interdisciplinary research has been to propose discussion session centered around broad multi-disciplinary topics to stimulate cross-fertilization. These sessions focused on (a) active learning, (b) connecting numerical and statistical approaches for error estimation and (c) developing a common language and cross-community guidelines with error estimation in atomistic modelling. We successfully stimulated initial advances on overcoming these barriers as exemplified by a GitHub repository (https://github.com/mfherbst/error-atomistic-simulations), which openly publishes our outcome from the discussion on topic (c). In the future we hope this repository to develop into a list of examples of error control strategies taken by researchers from various contexts and we wish to use this as a basis to develop both a common language as well as community guidelines like a minimal standard for error control. Moreover, we discussed ideas how error control more prominent and visible in the communities. Multiple people expressed

interest to continue this work beyond the context of the workshop and it was proposed to structure these results into an opinion paper to foster error analysis as a tool towards reproducible, validated simulation workflows.

3 Community needs

We do believe further events on this topic are needed. Firstly, a single workshop has by no means been sufficient to overcome aforementioned interdisciplinary barriers. In particular we hope for a better integration of the molecular and chemistry community in future events, which have been underrepresented this time. Secondly, our workshop clearly showed the remarkable progress that has happened in the past years when it comes to statistical and numerical error control. We believe a follow-up in 2023/24 thus highly suited to follow up on developments as well as the discussions on cross-community error control guidelines, which got kick-started at his event.

On the more practical side, we believe that the hybrid mode should be made the default setting for all CECAM workshops. This model makes events more open to researchers who cannot travel, either for funding, environmental or personal reasons. We experimented successfully with a conference-specific Slack workspace featuring one channel per talk. This simplified interactions between remote and local participants/speakers exemplified by frequent discussions on Slack after the talks. Regarding the discussion sessions we got the feedback that it was sometimes difficult in particular for juniors to participate in the group discussion. As improvements we suggest to e.g. (i) set and communicate the discussion subject in advance to allow interested people to get prepared, (ii) motivate PostDocs to coordinate the discussion, which are able to curate material and formulate stimulating research questions and (iii) get these PostDocs to start the session with an introductory presentation on open problems.

The boat trip, which we scheduled at the end of the workshop made it more family friendly, allowing participants to choose to take part in the social event or to return home early without missing parts of the program. For the future events we would like to encourage CECAM to offer child-care support.

4 Funding

There are no specific funding channels related to error control we are aware of. In fact, one aspect discussed during the workshop were possibilities to increase the visibility regarding the need of error estimation for reproducible science. In this regard we expect stronger demands from funding agencies and proposal reviewers to be an effective measure towards promoting error quantification as a standard in atomistic simulations (similar to demanding an open source and open data policy). Moreover, we argued there *should* be specific funding calls to enhance reproducible science and error control in simulations.

Opportunities for novel research proposals have not been discussed at a workshop level, but many participants have ongoing collaborations, which were continued in private conversations during the workshop.

5 Will these developments bring societal benefits?

Computational approaches to understand and simulate the behaviour of matter are widely established and play an integral role in materials research. Since the advent of cheap computational power, the volume of supercomputing power devoted to first-principle materials simulations is on a steep rise. Outcomes have been, for example, the development of novel materials fully in silico, which already successfully contribute to overcoming materials-related

challenges of the 21st century (e.g. new materials for batteries, green catalysis, quantum computing, ...). Such developments routinely require millions of first-principle simulations.

In this regime error control of simulations provides significant advances:

1. Estimating simulation error allows to compare results on an equal footing with experiment, where error analysis is well-established.

2. Error control allows to balance errors in a simulation workflow, allowing to save computational efforts in part of the workflow, where less accuracy is required.

3. Error control procedures contribute to making simulation results more robust and reproducible. Moreover, they contribute to improving heuristics for selecting computational parameters (as error estimation allows to understand which parameter limits accuracy) and thus reduce the fraction of failing calculations in workflows.

Being thus central improving efficiency and reliability of simulations, thus their computational cost, improved error control contributes significantly to finding the materials of tomorrow. Given the overall impact of first-principle simulations we find the effort devoted to error qualification in this field is still insufficient.

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Interdisciplinary workshop on the theoretical study of periodic systems

Location: CECAM-FR-RA
Webpage: https://www.cecam.org/workshop-details/1137
Dates: Jun 20, 2022 - Jun 23, 2022

1 State of the art

Periodic systems, i.e., systems that are translationally invariant at least in one dimension, have been of keen interest not only in physics and chemistry, but also mathematics. Scientists in these disciplines have been trying to solve important problems related to a large variety of periodic systems, with important theoretical and technological impact. The aim of this workshop is to bring together some of the best experts in the various disciplines, in order to tackle a given problem from different points of view.
Physics:
1) The description of properties of crystalline solids [4,5], in particular using density-functional theory or many-body perturbation theory [6]. These methods are very powerful but are limited to weakly correlated materials since the electron-electron interaction is hidden in an effective potential.
2) The study of the uniform electron gas and its various phases (Fermi liquid, Wigner crystal, etc.) [7,8,9]. The uniform electron gas is a model to study electron correlation in many-electron systems. Although many of its properties are known, its features at low density where it is predicted to form a Wigner crystal is still largely unknown.
Chemistry:
1) the ab initio treatment of crystals [10,11,12]. Ab initio chemistry approaches such as configuration interaction and coupled cluster theory are known for their great accuracy but their explicit treatment of the Coulomb interaction between electrons makes their application to periodic solids non-trivial.
2) the study of polymers and nanotubes [13]. Since in this case the system is periodic in only one direction, one often extrapolates results of finite systems to estimate the infinite-size limit. However, results may depend on the choice of the extrapolation [14]

2 Major outcomes

The goal of the workshop was to gather experts coming from different disciplines (Chemistry, Physics, Mathematics), in order to discuss the problems that arise in the theoretical treatment of periodic systems. The major outcomes of the workshop mainly consist of new contacts that have been established among the participants, and that will very probably lead soon to new scientific collaborations. In particular, we mention:
1) Giovanni Vignale discussed the orbital Hall effect for graphene-like materials, and strongly interacted with Ivo Souza and Raffaele Resta.
2) Iann Gerber, Maria Peressi and Tobias Schäfer presented different aspects of plane wave methodology to study periodic systems.
3) Marie-Bernadette Lepetit, Emanuel Gull, and George Booth planned to interact on the subject of quasi-degenerate perturbation theory.
4) The mathematical aspects were covered by Richard Schwartz, and partly by Emanuel Gull. In particular, the talk of Richard Schwartz about the solution of the Thomson problem for five electrons on a 2-sphere, gave some hints on how similar problems could be tackled on 2- and

3-tori for a given number of electrons. However, the difficulties for a formal proof of these general cases are indeed formidable.

5) Several contacts have been established by the Toulouse group (Miguel Escobar Azor, Véronique Brumas, Stefano Evangelisti, Arjan Berger):

a) with Lorenzo Maschio, for the possible implementation of the Clifford formalism in the CRYSTAL code. Moreover, Maschio presented a very comprehensive view of advantages and drawbacks of the use of local basis sets (Gaussians) for periodic systems, compared to the plane-wave technique.

b with Neil Drummond, who will test the Clifford boundary conditions in his QMC code for the study of the Wigner-crystal phase in a low-density electron gas.

c with Ivo Souza, who presented a formalism that is both gauge and translationally invariant for electric polarization and magnetization. We plan to translate this formalism to the case of Clifford boundary conditions.

3 Community needs

Many of the concepts here reported will benefit from an increase support of fundamental research. This is today particularly important in a moment in which public research is funded via contracts and projects. In many countries, fundamental research suffers with respect to applied or industrial research. Very often national calls give high priority to joint research+industry applications. To this we can add the perverse ramification of calls (european, national, regional, local) that far from multiply the offer, rather it creates a frustrating scattered and highly inefficient funding procedure. For what concerns specifically software and hardware, we have both good and bad news. Many groups have already partnerships with local software engineers, few others decided to ask support to the PRACE facilities. This is a crucial step: it is indeed true that the computer power available today (both in national and European calls) is good news to tackle bigger and more complicate systems. However, without a proper writing (and often, re-writing) of our computer codes (and this is the bad news), to take full advantage of the modern paradigms (blue-gene low memory nodes, GPUs accelerators, mics, and all hybrid architectures), there will not be any possibility to tackle such complex materials and behaviours. The scalability required to take full advantage of this new kind of resources is in fact such (1-1000000 cores) that only a professional approach can succeed. This professional procedure comes not only in term of parallelization of the codes, but also in their everyday maintenance and in all aspects of collaborative development (revision control, tickets, pull requests, forums, etc.), as previously mentioned. Finally, in order to make these new codes and capabilities widely available, training strategies have to be devised, at both hard (summer dedicated schools) and soft (e-learning platforms, MOOCS) levels.

4 Funding

It is crucial for the development of the fundamental research involved in this workshop, that funding agencies continue or even increase their funding of fundamental research. The workshop brought together many established experts in the theoretical treatment of periodic systems. Several channels have been analyzed for the funding of fundamental research:
- ERC grants at the European level (starting, consolidator and advanced, together with synergy) are the most important calls in which fundamental theory can be funded.
- Marie-Curie calls (Individual fellowship, CoFUND, and ITN) are still active and available also for fundamental research
- National funding agencies are uneven (good for Germany, bad for Italy, worsening in France, etc)
- local/regional calls are sometimes good sources of funding but are increasingly more dedicated to applied research

5 Will these developments bring societal benefits?

Progress in the theoretical description of periodic systems is of great importance to fundamental research but also to applied research since many technological advances that could have societal benefits involve solid-state materials, such as photovoltaics for a sustainably energy supply and solid-state batteries for electric vehicles. Although much progress can be made from experiment, the theoretical description of such devices can complement the experimental data by providing analysis and also by predicting which materials are potentially of interest and which are not. This is of course only possible if the solid-state theory is sufficiently accurate and if the computer code in which the theory is implemented is sufficiently optimized. Therefore, the accurate and efficient treatment of periodic systems in numerical calculations is crucial to predict and analyse materials that are of potential technological interest. However, this applies to the field globally. Concerning the more restricted focus of the workshop in particular, it is about fundamental research preparing the long-term future on a scale of, say, 10 years which unfortunately is not always of direct interest for a private company.

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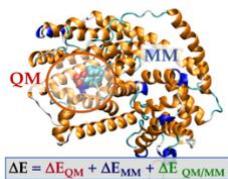**Resta, Raffaele** - CNR, Italy
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Present and Future of Hybrid Quantum Chemical and Molecular Mechanical Simulations

Location: CECAM-IT-SIMUL, Politecnico di Milano, Polo Territoriale di Lecco, Lecco
Webpage : https://www.cecam.org/workshop-details/1152
Dates: Jun 20, 2022 - Jun 23, 2022

1 State of the art

Hybrid QM/MM methods were originally developed over 30 years ago to allow the simulation of chemical reactions in complex environments. Nowadays, they are probably the most widely used multi-scale method in molecular simulation, employed for studying processes in all types of systems, including biomolecules, solutions, materials and surfaces [1]. Recent examples include electron transfer [8,9], metals and metalloproteins [10,11], mechanochemistry [12], photochemistry [13,14], and tunneling [15]. There are also several QM/MM implementations available, with their own strengths and weaknesses, for example in the codes CP2K [2], CPMD [3], NAMD [4], ORCA [5] and pDynamo [6,7].

Research in the QM/MM area is still very active, with developments in algorithms often in tandem with advances in other areas and in applications. Important examples are: the use of different QC methods, from generic semi-empirical methods such as XTB [16], to various DFT and higher-level ab initio schemes, and machine-learning corrections [17,18]; advances in coupling of the QM region to the MM environment by employing electrostatics treated by multipolar expansions or by considering MM polarization [19]; alternative MM representations, such as reactive force fields [20]; and the best way to employ QM/MM potentials with different sampling algorithms, such as those designed to study complex transitions and reactions [21]. Finally, there are also substantial questions with so-called adaptive QM/MM algorithms in which the partitioning of atoms between QM and MM regions is allowed to vary [22,23], and with how to integrate QM/MM approaches with other multi-scale representations, such as coarse-grained and continuum schemes [24,25]. The aims of the workshop were to take stock of the state-of-the-art in the QM/MM area by bringing together as diverse a range of QM/MM practitioners as possible and to identify directions for future development and application.

2 Major outcomes

The workshop addressed the current and future challenges and development of QM/MM hybrid methods. Several young researchers including post-docs and junior PI exposed their new research lines, discussing work still not published. More senior speakers gave nice overviews of the past and present state of the method.

The limitation of insufficient sampling is a common theme in all dynamical methods, and addressing it in QM/MM is hard because it is computationally expensive. Some improvement in path sampling methods were discussed, but the number of enhanced sampling methods used is still less than in traditional molecular dynamics (MD). The combination with deep learning algorithms seems promising when studying related compounds, but it is still far from easy for new users. In fact, because of their multi-scale nature, QM/MM simulations are more complex to setup and visualize than traditional MD. Efforts were shown to facilitate this process with new graphical user interfaces, which will be especially useful for expanding the QM/MM practitioners and to smoothen the learning curve for newcomers.

As a multi-scale method, improvements due to new methods and faster hardware can be introduced into QM/MM approaches. As such, polarizable force fields could represent an important step for the MM part. The use of large basis sets and the associated technical challenges were also addressed. The field is still far from having standardized file-types and setup protocols, and that hampers the use of different codes and the ability to reproduce results from other groups.

Alternative methods, such as the cluster model, reactive force fields or EVB represent an alternative to QM/MM, that were shown to give similar results when used properly.

Most current applications of QM/MM are in the biological sciences, and in enzyme catalysis in particular, probably because this is the discipline that created this tool. As Organisers, we looked for speakers that could expand this view, showing applications in more distant fields, such photochemistry, nanoparticles, electrochemistry or polymer physics. The reasons that may hinder QM/MM applications in other fields, such as the lack of transferable force fields, were also discussed.

Finally, an important outcome of this workshop was the decision to write a review describing all the previously mentioned points and other issues discussed in this workshop. On the one hand, we had a very positive response from the participants when this was addressed in the discussion session, with many volunteering to contribute. We are starting to organize how these contributions will be written and collected. On the other hand, we already have an invitation from Francesca Novara, editor of *Chemistry-Methods* journal (from Wiley), that would welcome such a review.

3 Community needs

During a final round-table session, the definition of standards for QM/MM simulations emerged as an important need for our community. We should be able to define:

1. Protocols which are "more appropriate" for QM/MM simulations in a particular case or for a particular type of simulation. This is connected to the "validation" of a method as a meaningful way of answering a given research question (see Konrad Hinsen's "Computation in Science", ISBN 168174029X). We pointed that a set of well-studied test cases (say, the chorismate mutase enzyme) could be employed to benchmark and compare different QM/MM approaches. Validations would also profit from direct comparisons to other experimental observables than reaction rates, commonly used in enzymology;

2. Standards for reporting the method and its detailed setup, so that a complete reproduction of the simulations could be performed and the computed results could be "verified" (see DOI:10.1021/acs.jctc.2c00286, for a recent example). More dialog between developers of QM/MM software should be stimulated, so that calculations could be reproduced with different packages;

3. Standards for formatting output and data sharing of QM/MM computations. Although databases for pure QC and MM methods already exist, it is unclear if these are sufficient for hybrid QM/MM simulations.

Extensions of graphical interfaces (such as PyMOL) for dealing with QM/MM simulations would be important particularly for new users, but also for analysis and setup of simulations. For hardware, we noted that impressive speed-ups seen for pure MM simulations with GPUs are not often observed for QC methods - which are usually the bottleneck in a QM/MM simulation.

Regular workshops and online material such as software documentation, videos and websites are relevant for disseminating information on QM/MM simulations and should be pursued in the near future.

4 Funding

The workshop did not include a dedicated session on fundings. As Organisers, we did our best to stimulate personal interactions between individual participants, that hopefully will lead to joint research projects.

The Horizon Europe program (2021-2027) offers several opportunities for research in computational molecular sciences, from the fundamentals (e.g. by the ERC program) up to specific applications (e.g., within the "missions" of fighting cancer, climate change, water pollution).

A EU network of scientists active in QM/MM simulations could possibly be funded by an MSCA (Marie Sklodowska-Curie Action) grant, to train a dozen PhD students. The exchange of PhD students between different groups could be an important tool to address some of the "community needs" described above, especially documentation, cross-validation and reproducibility of QM/MM simulations.

5 Will these developments bring societal benefits?

The workshop offered several examples of problems and applications bringing potential benefits to society.

For example, one talk (by R. Suadiaz) described QM/MM simulations of the mechanisms behind antibiotic resistance in microbes (one of the greatest threats to human health according to the World-Economic-Forum-Global-Risks report, in 2018), in particular against the last-resort antibiotic Colistin. Another talk (by M. De Vivo) discussed computational drug discovery, starting from QM/MM studies of enzymatic catalysis of pharmaceutically relevant targets, such as metalloproteins that process DNA and RNA. As a final example, a talk by B. Zappone discussed the mechanisms behind (bio)polymer adsorption, adhesion, and cohesion on wet surfaces. These are important for biofouling (on the negative side), but also for surgery and tissue engineering (on the positive side).

The workshop had a truly international attendance. Several speakers came from outside Europe, especially north and south America. Collaboration between scientists there and in Europe could be funded by travel grants. There are several such schemes, funded both by the EU and through bilateral agreements. Also, private funds, such as the Bill and Melinda Gates Foundation, finance ambitious interdisciplinary and international projects in "Discovery & Translational Sciences", especially on human health and the environment.

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Local vs Collective Interactions in Polaritonic Chemistry
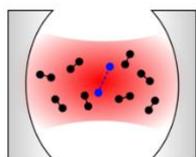
Location: CECAM-FR-GSO
Webpage: https://www.cecam.org/workshop-details/1140
Dates: Jun 22, 2022 - Jun 24, 2022

1 State of the art

Polaritonic chemistry is an emerging field of research that opens new ways to control the properties and dynamics of molecules and materials by engineering their surrounding electromagnetic vacuum by micro- and nano-cavities. A striking experimental fact is that the electromagnetic cavity modes can couple to a molecular transition (electronic or vibrational) and strongly influence the matter subsystem. These changes are accompanied by the formation of hybrid light-matter excitations called *polaritons*. In this regime of strong light-matter coupling, materials get dressed by the cavity modes and thus can partially inherit properties of the confined light field. This phenomenon has an impact on both optical and chemical properties of materials and molecules and ***paves the way to design a new generation of materials in confined electromagnetic environments.*** Pioneering experiments reported a wealth of different properties that are altered significantly in optical cavities. In parallel to these flourishing experimental activities, a wealth of theoretical works has appeared that try to describe these cavity-induced modifications. However, some recent disagreements and controversial issues were reported between theories and experiments addressing, for instance, the role of vibrational strong coupling in the modification of ground-state chemical reactivity observed in various experiments. It thus appears that there is, on the experimental side, a strong need to perform further experiments (e.g. issue of reproducibility), and on the theoretical side, to develop a general picture and qualitative understanding of the physical mechanism responsible for cavity-modified chemistry. ***This workshop aimed at clarifying and scrutinizing the different experimental scenarios and theoretical methods for investigating the light-matter strong-coupling mechanism in polaritonic chemistry.***

2 Major outcomes

We structured the workshop along the following questions: **1.) Day one: « collectivity vs locality »** How to reconcile the description of polaritons as collective and spatially extended light-matter excitation, with the locality implied by chemical reactions? **2.) Day two: « quantum vs classical »** What is the *quantum nature* of the collective *polaritonic* state in a cavity, and its corresponding role on chemistry? 3.) **Day three: « symmetries and new directions »** What are the consequences of symmetries in polaritonic chemistry? Those questions were extensively discussed but no conclusive answers could be given. There was a broad consensus that there is still a discrepancy between the most promising experimental results (modifications of chemical reactions via vibrational strong coupling) and the available theoretical methods. ***For that reason, it was proposed to start a concerted effort of experimentalists and theoreticians to identify a "fruitfly" chemical reaction that is still feasible with different theoretical techniques yet shows at least quantitative changes in rates when reaching strong coupling.*** A further suggestion was that experimentalists should try to provide further observables than only the change in rates and the Rabi splitting and report also systems where no effect could be observed. **With respect to point 1.)** No

collective approach could so far recover the results of the chemical-reaction experiments. Local approaches, assuming that in the ensemble we have strong local fields or non-equilibrium effects, were able to attain qualitatively similar results as in the experiments. There is now a general interest to understand under which conditions such local-field or non-equilibrium situations could arise and **how to compute microscopically the coupling strength**. The development of a mesoscopic approach to cavity QED is a complementary path that sounds promising to quantify **the cooperative coupling between single-molecules** and to probe the reaction thermodynamics of large molecular ensembles. **With respect to point 2.)** A distinction was proposed between single-particle and many-body quantum effects. It was argued for that the observed changes in chemical reactions should be qualitatively captured by classical considerations and if quantum effects are decisive, they should be single-particle quantum effects. Robust many-body quantum effects need a high level of control of all detrimental processes. **With respect to point 3.)** chiral cavity modes and geometrical phases (light-induced conical intersections) were seen as promising routes to engineer even stronger cavity-induced effects. Also, a stronger integration of various ab-initio QED methods and phenomenological models was requested to tackle the theoretical complexity of polaritonic chemistry.

3 Community needs

During the Workshop, it appeared that there is a strong need to propose a catalogue of simpler (intramolecular) chemical reactions or reactions that are reasonably controlled and of which the reaction mechanism is sufficiently known. This could enable to deal with the issue of reproducibility and to test experimentally the modification of chemical reactivity upon entering either the electronic or vibrational strong coupling regime. ***For this purpose, it appeared relevant for the future to set up a database shared by the community about those reactions***, and collect all positive or negative results of experiments. From the point of view of theory, there is a strong need to use large computational resources. Indeed, the development of new *ab initio* approaches that enable to describe on the same footing electronic and vibrational structure of molecules, solvent effects and the cavity-electromagnetic field, were clearly identified as being crucial for understanding the experiments. ***The demanded computational resources/ open-source codes could also be shared by making an European node on this field of research.*** The knowledge acquired by various communities in quantum chemistry, quantum optics, plasmonic and effective models could be shared and ***the organisation of interdisciplinary summer schools*** on polaritonic chemistry is relevant for this purpose. This Workshop was the first CECAM Workshop organized on polaritonic chemistry. There was a clear need of having a regular series of Workshops in this field (which is still not existing), which is an emergent one with fast developments. ***A series of recurrent CECAM Workshop could really be relevant for the future following this Workshop.***

4 Funding

5 Will these developments bring societal benefits?

At the present stage it is premature to project if there will be (and what kind of) social/economic benefits and/or industrial applications of polaritonic chemistry. But it is clear that the potential is there due to the possibility of engineering chemical reactivity of ensembles of molecules simply by coupling them to the vacuum state of the electromagnetic modes of a cavity or to finely designed nanophotonic structures. Of particular interest for the future are the possibility (still under debate) of *modifying enantioselectivity of molecules* or *stereochemical reactions, modulating material properties* (superconductivity, electronic and energy transport, energy harvesting) by electromagnetic confinement. For this to become a more applied field of research, *this workshop really pinpointed the necessity to collectively address in the community the issue of reproducibility, data sharing and synergies between theory and experiments*.

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Topological phases in condensed matter and cold atom systems
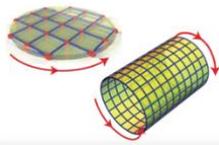
Location : Institut d'Etude Scientifique de Cargese, France
Webpage : https://www.cecam.org/workshop-details/1139
Dates: Jun 27, 2022 - Jul 9, 2022

1 State of the art

The 2016 Nobel Prize in Physics was awarded to pioneering work opening the field of topological phases of matter. This field has matured later on in the study of the fractional quantum Hall effect, which continues to deliver exciting physics, in the form of non-abelian excitations and the observation of neutral edge modes. Inspired by the quantum Hall effect, the study of non-abelian particles has branched into different topics, such as the study of topological phases emerging in (spin) lattice models and recently topological insulators and superconductors. During recent years, the field of topological phases has been boosted by the possible application to quantum computing. Implementing topological quantum computation in realistic experimental systems is one of the holy grails of the community. Most notable are the newly discovered topological insulators (or superconductors), which combine physics from the quantum Hall effect and graphene. Currently, most of the interesting physics in topological insulators emerges from combining non-interacting band theory with the notion of topology, which has led to some spectacular results.

2 Major outcomes

However, the fact that most of the developments in the field of topological insulators have focused on the effects of the topological properties alone means that consideration of the consequences of adding electron interactions are largely missing. While the latter give rise to very interesting physics in their own right, combining them with topological structures will most certainly lead to many interesting discoveries. The fractional quantum Hall effect is a prime example of where this interplay indeed has led to very exciting new physics.

Classifying topological phases in the presence of interactions is a daunting task, so that making even a little progress will greatly enhance our understanding of topological phases. This is one of the main questions that has been addressed during the workshop.

Invited speakers were asked to give two 1h lectures, mostly on the blackboard (outside). The initial goals of the event were fully accomplished with following lectures:

• Adolfo Grushin: Models and experiments in amorphous topological matter
• Yasir Iqbal: Symmetry classification of quantum spin liquids
• Sid Parameswaran: Symmetry, Topology, and Correlated Insulators in Moiré Graphene
• Vedika Khemani: Topological Floquet phases and time-crystals
• Chris Laumann: Spin ice / quantum spin ice

- Kirill Stenghel: Order by disorder in chiral antiferromagnets
- Rachel Queiroz: A local view on topology
- Ashley Cook: Topological skyrmion phases of matter I & II
- Maia Vergniori: Crystalline symmetries and topological materials
- Ronny Thomale: Topological phases in electric circuits
- Adam Smith: Non-abelian anyons: the basics of topological quantum field theory
- Frank Pollmann: Intro on MPS and TPS / Using quantum computers to simulate topological phases
- Cécile Répellin: Chern insulators: from single-particle to many-body properties

3 Community needs

It is worth emphasizing the dynamism that the field has received through the development of NISQ processors. It is just now becoming feasible to emulate/simulate a wide variety of many-body phases -- both static and dynamic properties -- using these near-term quantum computing platforms, with new non-equilibrium topological properties -- such as various types of Floquet insulators -- being of particular interest.

The enthousiasm and dynamism of the junior (PhD, posdocs, etc...) participants testifies to the need to learn the new developments in this rapidly developping field. The format of the workshop was ideal for such an exchange of knowledge. Most talks were given outside on the blackboard in a rather informal manner; the IESC offers a unique framework for junior participants and more experienced lecturers to interact.

Many interactions/discussions between lecturers and participants have been initiated at the end of these lectures and have continued for the rest of the event. Posters have been shown for the whole time of the School/Workshop and most junior participants had a chance to publicize their work and discuss about it with others.

4 Funding

The economic model was to wave registration fees for invited speakers and lecturers while, for other participants, an affordable fee was asked to cover lodging, breakfast and lunches for the 2 weeks of the workshop. Travels expenses were not provided to anyone, except the transportations from/to Ajaccio airport.

The GSO-CECAM node has provided most of the local cost charged by the IESC. We also benefited from a limited funded from the Max Planck Institute (R. Moessner's prize).

5 Will these developments bring societal benefits?

The research topics of the workshop are clearly of fundamental research nature and at the forehead of scientific knowledge. However, many topics discussed during this event may have potential applications to future technologies associated to (topological) quantum computing or quantum simulator platforms. It is part of a European effort to develop quantum technologies. This goal could only be achieved when fundamental concepts are fully understood and when theoretical and numerical tools will be completely developed to attack highly non-trivial complex systems to be encountered in this field.

It is worth emphasizing the dynamism that the field has received through the development of NISQ processors. It is just now becoming feasible to emulate/simulate a wide variety of many-body phases -- both static and dynamic properties -- using these near-term quantum computing platforms, with new non-equilibrium topological properties -- such as various types of Floquet insulators -- being of particular interest.

6 Participant list

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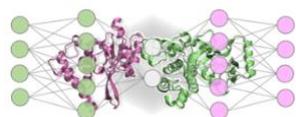Ecole Normale Supérieure de Lyon, France
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Chasing CVs using Machine Learning: from methods development to biophysical applications

Location: Inria Paris,
Webpage : https://www.cecam.org/workshop-details/1134
Dates: Jun 28, 2022 - Jun 30, 2022

1 State of the art

The dynamical behaviour of molecular systems in chemistry, biophysics and materials science, is typically governed by a small number of collective modes, alternatively collective variables (CVs) or reaction coordinates. Until a few years ago, CVs were identified using empirical approaches and chemical intuition. Intense efforts have been invested in automating the definition of CVs from molecular simulation data, using either supervised or unsupervised Machine Learning (ML) techniques [1-11]. These CVs are then used to obtain new understanding on the system at hand, perform molecular design, and guide enhanced sampling [12,13]. They also provide a way to perform dimensionality reduction.

Although the details and specifics differ, most CV discovery techniques fall into two categories: those seeking high-variance CVs and those seeking slow CVs [12]. The best known high-variance CV estimation technique is principal component analysis (PCA), but nonlinear analogs were developed [12,13], possibly in conjunction with an iterative procedure to combine CV discovery and accelerated sampling [14-19].

Most of the approaches proposed for the identification of slow CVs rely on the variational approach to conformational dynamics or in the (extended) dynamical mode decomposition [13]. The inputs in these approaches can be the (Cartesian or internal) coordinates of the system itself, or average values of indicator functions as in Markov State Models. Nonlinearity can be brought into the model through the use of neural networks [20].

The choice of the proper dimensionality of the CVs to be found is a delicate question, which still requires further research efforts. For linear models such as PCA and its variations, there are clear mathematical guidelines for selecting an appropriate number of dimensions based on variance explained by the model. A similar framework for nonlinear models, based on neural networks for instance, is currently missing.

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[19] G. Bussi, A. Laio, Nat. Rev. Phys., **2**, 200-212 (2020)
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2 Major outcomes

Besides the many discussions and exchanges following the talks, there were also two more formal discussion times, where the audience brainstormed on the current challenges in the field of finding reaction coordinates with machine learning techniques. The topics discussed fall into two general categories.

1) Technical aspects of CV discovery:

- the choice of the loss/objective function should be more carefully thought of, as this implies limitations in the use and interpretation of output results. One also needs to carefully think about mandatory associated regularizations strategies in order to avoid overfitting (by introducing additional penalty terms in the loss such as ridge or lasso; by changing the architecture via dropout; by modifying the learning framework with the addition of extra noise to the input as in denoising autoencoders; by turning to alternative training strategies via the use of stochastic gradient descent instead of Adam, or by resorting to early stopping, etc.)

- the transferability of CVs is a very interesting issue. It may well be that the CVs obtained for subclasses of problems (e.g. family of kinases) are sufficiently similar so that the learning performed on one system can benefit others. One would need to this end to create a repository or dictionary of learned CVs, and then to quantify the similarity between these functions; it is likely that there may be a trade-off between physically meaningful CVs that are based on user-defined feature maps, with good transferability and possibly sparse, machine-learned CVs with less transferability.

- there is some general consensus about the transferability of data, often generated with one method (e.g. simulating Langevin dynamics, possibly with enhanced sampling) while used on the premises of another framework (e.g. relying on effective Brownian dynamics as in diffusion maps).

- another technical issue is to perform unbiasing in the context of slow CV discovery and enhanced sampling, as the biased trajectories need to be reweighted using some Girsanov weight in addition to the usual importance sampling weight arising from the free energy bias. There are some preliminary works in this direction, suggesting that reweighting is not necessary for sufficiently short trajectories (see Wang/Tiwary, J. Chem. Phys. 152, 144102, 2020) but more work is definitively needed here.

2) General ML procedures and good practices. Besides the technical issues spontaneously encountered in the current machine learning based approaches to CV discovery, there were also discussions on more general aspects of machine learning, which have proven useful and sometimes decisive in other scientific fields, and may also have an impact here

- relevance of data whitening, especially for slow CV discovery

- resorting to staging procedures with a progressive featurization of the initial high dimensional input

- using reinforcement learning to directly learn the bias to be applied on the system, without the need to parametrize it as the gradient of a free energy. Of course, one may suffer there from the curse of dimensionality, as the bias to be learned is a priori a vector field of the same dimension as the system. Preliminary works in this direction include Das/Rose/Garrahan/Limmer, J. Chem. Phys. 155, 134105, 2021; Yan/Touchette/Rotskoff, Phys. Rev. E 105, 024115, 2022; Lelièvre/Robin/Sekkat/Stoltz/Victorino Cardoso, arXiv preprint 2205.02818.

- many fields benefited a lot from well-established benchmarks (as for image classification in the field of machine learning, or machine learned potentials). The field would greatly benefit from the consensus around clear common objectives and the availability of reference data.

- the quality of force fields was discussed, as this can have a strong influence on the quality of the final results. The participants from materials science were surprised that the many efforts aimed at developing machine learned potentials are not being picked up in the biophysics community. One reason is that the quality of the underlying models (e.g. type of membranes protein, structure and composition of proteins obtained by homology modeling, etc) is lower than in systems from materials science, so that there would be little need of quantum quality reference energies/forces given the larger modeling error.

3 Community needs

Most of the researchers in this conference are using HPC infrastructures, including PRACE projects, but also computing offered by private companies such as Amazon, Microsoft and Google. There seems to be no need to further organize this. In terms of software, many of them are also developing python packages or libraries to be used in conjunction with existing reference molecular dynamics codes. This is a definite and clear trend in the community towards dissociating the molecular dynamics engine, seen as a tool to produce data, and the analysis/machine learning tool, natively written in python most of the time since most of the software for machine learning methods is in this language (in particular PyTorch for neural networks, and scikit-learn for other techniques).

There is, on the other hand, a clear need for places to exchange ideas and good practices. Many researchers saw this workshop as the natural continuation of the 2019 CECAM flagship workshop "Learning the collective variables of biomolecular processes", also held at Inria Paris (July 10-12th, https://www.cecam.org/workshop-details/92); and even more expressed an explicit interest in gathering again in a couple of years at most on a similar format. Some other researchers, closer to materials science, are thinking of organizing a similar event in their field of application.

In addition, it would make sense to organize a hands-on version of such a workshop, where good practices in terms of using machine learning methods would be discussed in detail -- with questions such as determining the topology of neural networks, setting hyperparameters for training, choosing regularization strategies, etc. Given the craftsmanship currently needed to make the best use of machine learning techniques, a similar training by specialists in machine learning (e.g. researchers working on image classification, etc.) would be extremely beneficial in setting up a sub-community in molecular dynamics more fluent in these modern techniques.

4 Funding

There were two main sources of funding for the workshop: (1) the CECAM funds of the local nodes, namely CECAM-FR-MOSER (8000 euros), which covered coffee breaks, snacks and drinks for the poster session on Tuesday, the conference dinner, and the accommodation of invited speakers from institutions outside of Germany; and CECAM-DE-MMS (2000 euros) which covered travel expenses of speakers and some participants from Germany; (2) the ERC

5 Will these developments bring societal benefits?

There was a clear and strong relationship between the topics of this workshop and private companies in the pharmaceutical industry at large. This was demonstrated through one of the talks given by Paul Maragakis, from D.E. Shaw Research, a private research company, who showed that the analysis of MD trajectories using machine learning for mechanistic insight is indeed used in the pharmaceutical industry. This was also demonstrated by the high level of sponsorship from such companies (Sanofi, Aqemia, Discngine, Qubit), and the lively discussions following the presentations of these companies in the "industrial session" of the workshop. All these companies nowadays seamlessly integrate machine learning techniques and molecular simulations for quantitative estimation of protein-ligand binding thermodynamics or kinetics or of protein conformational dynamics. The industry session also showed to the younger members of the audience that careers in the private sector on their research topics are possible.

Moreover, as therapeutic targets become less and less attainable through traditional drug discovery techniques, the use of ML to enhance their conformational sampling is gaining attention. Techniques like the ones presented and discussed during the meeting could potentially revolutionise Computer-Aided Drug Design in the context of targeting allosteric mechanisms or yet unknown protein or RNA conformations.

6 Participant list

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Quantum Transport Methods and Algorithms: From Particles to Waves Approaches
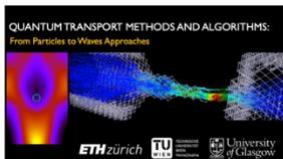
Location: ETHZ, Zurich, Switzerland
Webpage: https://www.cecam.org/workshop-details/49
Dates: Jul 6, 2022 - Jul 8, 2022

1 State of the art

With the rapid decrease of the semiconductor device dimensions, technology computer aided design (TCAD) has entered a new era where one-dimensional models and (semi-) classical approximations such as the drift-diffusion or Boltzmann Transport equations are reaching their limit to simulate the properties of nanoscale components. These approaches should be complemented by more advanced, but computationally more intensive models based on quantum mechanics: energy quantization, geometrical confinement, and tunneling currents are all important effects that should be properly taken into account. Moreover, when possible, accurate device simulations should go beyond the ballistic limit of transport and include relevant scattering mechanisms such as electron-phonon, alloy disorder, interface roughness, or electron-electron interactions.

Different techniques have been proposed to solve the resulting quantum transport problems. They can be divided into two main categories: one relying on a wave formulation of transport and typically expressed with so-called Non-equilibrium Green's Functions (NEGF) [1] and one considering electrons as particles and dealing with a quantum phase-space [2]. Two research communities have formed around these methods that might better collaborate with each other and could benefit from enhanced exchanges of ideas, physical models, simulation methods, or numerical algorithms.

Key References

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2 Major outcomes

The goal of this CECAM workshop was to bridge the wave- and particle-based quantum transport communities in order to solve the *multiscale* challenges that the *nonequilibrium* modelling of contemporary and future nano-devices, e.g., ultra-scaled transistors, photo-diodes, thermoelectric generators, or memory cells, is facing, especially when going beyond electron transport phenomena.

22 invited speakers from both academia and industry participated to the workshop and shared their view on the challenges and future developments of physics-based nano-device modeling. The major outcomes of the five sessions that took place are the following:

- Quantum Transport Basics and Theory: The fundamental principles and assumptions of the wave- (NEGF) and the phase space-based (Wigner) formalisms were critically introduced by the doyen of our quantum transport community, Dave Ferry. This practically opened the discussion of the scope of their applicability and the current challenges. The following talks presented the achievements and future development of the NEGF approaches, a basic for the description of entangled states model of electron-electron interactions and a method for transport modeling based on Bohmian trajectories. The latter is an alternative phase space (particle) approach. The establishment of a link to Wigner particle approaches was identified

as a critical future research step. During the discussion the participants agreed that wave- and particle-based methods are rather complementary: A conjunction between them offers a synergistic approach to the multiscale challenges of the modeling of contemporary and future nano-devices.

- From Classical to Quantum: In this session the five talks covered topics such as quantum-corrected Monte Carlo methods, the embedding of a NEGF window into a classical simulation domain, and Finite-Difference Time-Domain techniques. During the discussion ideas were shared on how to compare these different approaches and assess which one is the most appropriate for a given application. It was agreed that one common template device structure will be prepared and all available tools/methods will simulate it. The results will be made available to the community through a shared publication or via the nanoHUB.

- Numerical Algorithms: The two first presentations highlighted how professional software development can help domain scientists overcome programming difficulties and focus on their core activity, the development of novel physical models. Two researchers then provided an overview of the implementation of their own TCAD tool, one relying on the Wigner functions and the other one on the NEGF formalism. Although the underlying methods are different, the challenges are similar and concern the parallelization of the workload, the need for efficient numerical solvers, and memory management.

- Electronic Transport: This session was mainly dedicated to the usage of the NEGF method to solve different type of devices. Examples that were discussed include 2D materials, Si nanowires, and carbon nanotubes. During the discussion the participants talked about how NEGF methods can be unified and how to speed up these calculations.

- Beyond Electronic Transport: The last session concentrated on thermal transport and light-matter interactions. The talks were given by researchers who are at the forefront of the modeling of such systems, some of them having pioneered the field. Two presentations were dedicated to particle-based simulation approaches, two others reported recent advances in NEGF applied to large-scale thermoelectric systems and optoelectronic devices, while the remaining one successfully reconciled the wave and the particle nature of phonons.

3 Community needs

Nano-device simulations with particle- or wave-based approaches are typically computationally very intensive. A lot of different tools (e.g., ATOMOS, BITTLES, NanoTCAD ViDES, NEMO5, NESS, OMEN, QuantumATK...) and useful libraries (DaCe, FEAST, libNEGF, SPIKE...) have been developed and are maintained by the groups who attended the workshop, some of them are open-source packages, others have become commercial products, most of them can take advantage of or even require large-scale HPC resources, while few can run on a single desktop.

Several researchers present in Zurich are already intensively collaborating with experimental groups and with industry. The goal of device modeling is typically two-fold: shedding light on the behavior of existing components and providing accurate design guidelines for next-generation nano-systems. This can only be realized with a direct connection to labs actually fabricating such compounds. While the link to experimentalists can always be strengthened, the one to the materials science and physics communities should definitively be enhanced. With the advance of *ab initio* methods, more and more data and inputs are takes from tools that were developed in these communities. If instead of exchanging parameters between different codes, new packages that integrate both types of functionalities would be developed, results of higher physical accuracy could potentially be obtained at lower computational cost. Possible future directions of device modeling and technology computer aided design have been discussed during this CECAM workshop. Benchmarks to compare different tools have also been proposed. Hence, another such event in 2-3 years could be really useful to monitor the progresses that have been made since the first meeting.

4 Funding

As participants from Europe and from the USA were present, no funding scheme available to all of them exists. Nevertheless, there have been bi- and multilateral discussions between several groups to launch new projects together. For example, through the EuroHPC initiative, there might be opportunities to support the upgrade of existing modeling tools towards pre- and exascale supercomputers and to run them on these machines. For researchers based in Switzerland, the Swiss National Science Foundation offer different "lead agency" schemes with e.g., Germany, Austria, France, and now also the USA to initiate collaborations between two or more principal investigators. Horizon Europe projects are more difficult to set up, mainly for two reasons: (i) consortium mainly consisting of tool developers are possible only in very specific calls and (ii) the majority of the workshop attendees cannot participate to EU projects (USA) or currently only with difficulty (Switzerland).

5 Will these developments bring societal benefits?

Technology computer aided design (TCAD) tools have been used in the semiconductor industry since the end of the 1970's, when the first 2-D simulations of complementary metal-oxide-semiconductor field-effect transistors (MOSFETs) became feasible. Over the last 40 years, MOSFETs have undergone tremendous evolutions, their dimensions being reduced by two orders of magnitude. At the same time technology boosters have been gradually introduced, e.g. strain, high-κ dielectrics, or 3-D FinFET structures to keep improving their switching characteristics. While the driving force behind these innovations has always been the intuition of clever researchers, TCAD tools have been used to validate their ideas, to optimize the underlying design parameters, and to understand their influence on the transistor performance. Hence, TCAD has critically contributed to the success of the semiconductor industry.

Now that the dimensions of transistors have reached the nanometer scale, it is no more possible to use TCAD tools relying solely on the classical laws of physics and to expect reliable performance predictions. It is our hope that the CECAM workshop we organized in Zurich paved the way for the emergence of a new class of device simulators that take advantage either of particle- or wave-based methods and that will optimally support the design of next-generation transistors and other nano-devices.

6 Participant list

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Light-matter interaction and ultrafast nonequilibrium dynamics in plasmonic materials
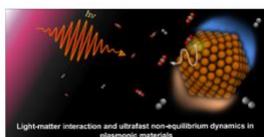
Location: University of Warwick, UK
Webpage : https://www.cecam.org/workshop-details/1164
Dates: Jul 18, 2022 - Jul 21, 2022

1 State of the art

Plasmonic materials (e.g. Au, Ag, Cu, Mg) can often absorb a huge amount of energy when the incoming light is in tune with the localized surface plasmon resonance frequency or Surface Plasmon Polaritons. In the last few decades, ultrafast phenomena that occur upon plasmonic excitation have garnered the interest of a large part of the scientific community due to their potential relevance in applications ranging from optical sensors, catalysis to biomedical applications.

A full theoretical description of light-matter interaction and plasmon-induced ultrafast non-equilibrium dynamics is a formidable challenge that demands an intrinsically multidisciplinary and multiscale approach. A variety of different approaches based on time-dependent Density Functional Theory, many-body perturbation theory, molecular dynamics, Mie theory, continuum electrodynamics, and combinations thereof have emerged to address many of the open questions in plasmonic. Further improvements in theoretical descriptions are crucial to optimize SPP generation and amplification in materials, to tailor losses and plasmonic

lifetimes, as well as to integrate plasmonic effects into semiconductor technology to create new quantum materials. Due to the diverse aspects of this problem, a coherent research community around theoretical plasmonics is only slowly emerging. The aim of this workshop was to assess the state of computational methods in this field, to identify major challenges, as well as to provide engagement between disparate communities to create space for cross-community collaboration.

The meeting was held from 18th to 21st of July 2022 at the University of Warwick. It featured 28 talks, 4 discussion sessions, and 10 posters. It was attended by 42 in-person attendees from 12 different countries and broadcast as a webinar with between 3 and 17 virtual attendees at any time.

2 Major outcomes

The workshop brought attention to the diversity of materials, physical phenomena, and theoretical and experimental methods that are relevant for plasmonic and for its application to the field of photocatalysis. Several presenters emphasised the opportunities to discover new ultrafast physics when strong plasmonic resonances at interfaces are triggered such as optical nonlinearity and plasmon-induced photoemission. An emerging research stream is the use of polarised light and chiral structures to engineer unique materials properties. Exciting new experimental techniques are emerging and their capability to harness strong near-field effects was among the discussion topics of this workshop. These include tip-enhanced Raman spectroscopy, EELS-STEM, as well as photon-induced near-field electron microscopy. Theoretical investigations need to be able to predict the non-linear response that these measurements observe.

The common understanding of plasmon-assisted chemical reactions typically relies on the energy exchange among hot-electrons (photo or plasmonically excited on plasmonic nanoparticles) and molecular adsorbates. An important issue that still remains to be addressed in this field consists in revealing the influence of more complex many-body excitations and quasiparticles for chemical reactions in plasmonic compounds. The question of whether plasmons directly couple to phonons or only act a source of hot electrons still remains to be addressed.

A clear need was identified for electronic structure techniques that are scalable and able to address strong correlation effects in materials as well as strong light-matter interaction. Currently few viable methods exist to study electronic excited states at molecule-metal interfaces, which also directly limits the use of nonadiabatic molecular dynamics techniques and real-time time-dependent DFT to study how electronic excitation drives dynamics at the interface. Promising methods were identified as exchange-correlation functionals that correctly predict the level alignment at the interface and many body perturbation theory methods that capture frequency and lifetime of 1-particle and 2-particle excitations. The special properties of plasmonic and other collective resonances in unusual plasmonic materials such as Magnesium or two-dimensional materials was also discussed. In the latter case, it was raised that this likely requires electronic structure methods that can address strong correlation effects as they occur in the presence of flat bands (e.g. the constrained RPA method).

The coupling of the mesoscopic optical response of plasmonic nanostructures and the local atomic-scale dynamics remains a key challenge in this field. Several speakers introduced techniques to couple quantum and continuum methods to describe nonlinear optical response at interfaces. These include classical electrodynamics approaches with some level of quantum embedding. Such methods unfortunately often break down when quantum effects become dominant as, e.g., in the presence of molecule-metal charge transfer. Other techniques that were presented are real-time TD-DFT with explicit account of the quantized nature of the electromagnetic field. These methods emerged as suitable for describing strong light-matter interaction in plasmonic materials and the formation of polaritons in optical cavities.

It was discussed that observed dynamical mechanisms likely differ between pulsed lasers and continuous white light illumination. It remains unclear if dynamics at the interface will be dominated by thermalised electrons at elevated temperatures or truly non-equilibrium electron distributions created by light excitation. Collective plasmonic excitations mainly decay into electronic excitations at the Fermi level. The origin of this effect is currently being investigated, but a full theoretical understanding is currently lacking. However, real-time TD-DFT simulations of plasmon decay also show evidence of this effect. This is an example where two different communities found corroborating evidence of the same effect, which we established as part of this conference.

3 Community needs

Coupling of different classical and quantum mechanical methods that operate on different scales (ultrafast electron and nuclear dynamics at the few femtoseconds and nanometer scale to electrodynamic field enhancement effects at the millimeter scale) is crucial to study ultrafast plasmonic dynamics. Therefore, modern software will need to offer sustainability, interoperability, modularity to combine classical and quantum approaches and approaches that tackle different scales. Most first principles codes are not modular and relatively monolithic in nature. The attendees identified a great need in future software to tackle ultrafast condensed phase dynamics in strong light-matter interaction.

Many attendees positively commented on the interdisciplinary nature of this workshop, and on the diversity in the background of the attendees. Many attendees were exposed to new topics, methods, and approaches for tackling problems pertaining to the field of ultrafast plasmonics. A clear outcome of the workshop was the need to enable more space for such cross-community gatherings in a field that is defined by an unusually broad spread of classical and quantum methods that originate from condensed matter physics, chemistry, and optics. During the final discussion session, participants were very vocal about wanting to establish a series format of this workshop running every 2-3 years that reflects the fast progress of different communities on ultrafast plasmonics. We believe that a series of CECAM workshops would be the best way to serve the need for different communities to exchange ideas on a regular basis.

4 Funding

All attendees agreed that meetings with space for discussions between participants from different theoretical communities, in particular from plasmonics, electronic structure theory, quantum chemistry, and surface science together should continue. As this field is intrinsically multidisciplinary and multi-method focused, regular exchanges across communities are crucial.

During one of the discussions, several possible avenues to fund future meetings and platforms were discussed. These included the option of continuing the meeting as a CECAM flagship workshop via a new proposal in 2-3 years, but also other funding opportunities such as an EU cost action. This is a particularly appealing option as so many European countries were represented at this workshop (participants from 9 European countries and China, USA, Israel). Many attendees left the meeting with new research ideas, having met new potential collaborators.

5 Will these developments bring societal benefits?

Novel optoelectronic and spintronic devices based on plasmonic field enhancement effects offer great benefits in terms of improved sensitivity, size integration and cost effectiveness. One of the most interesting processes associated with plasmonic excitation is the efficient production of hot electrons as part of a non-radiative plasmon decay channel. These highly energetic electrons have been shown to increase the reaction rate of different surface chemical reactions such as CO_2 reduction, water splitting, and H_2 dissociation reactions. The transformation of the chemical industry towards renewable feed stocks and more selective and cost-effective catalytic processes is vital to meet sustainability targets set by the UN, the EU, and many national governments. Plasmonic and hot-electron based energy conversion offers a way to selectively provide free energy to chemical reactions. Mechanistic understanding of all relevant processes, including the plasmonic excitation, ultrafast electron dynamics and the electron-driven molecular dynamics is crucial to design optimal plasmonic devices and catalysts. Theoretical plasmonics and dynamics simulations that utilize a broad spectrum of classical and quantum mechanical methods are vital to achieve such an understanding.

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SIGN 2022

Location: CECAM-ISR
Webpage : https://www.cecam.org/workshop-details/1048
Dates: Sep 4, 2022 - Sep 9, 2022

1 State of the art

The workshop was dedicated to the sign problem. The sign problem is a computational challenge for simulations of many physical problems. It usually occurs in systems that include fermions, as well as when chemical potential is introduced and in systems with frustration. In particular, the problem arises in the study of QCD at high density and as such is relevant to our attempts to understand dense nuclear matter. Related difficulties arise in models of superconductivity, in the simulation of the time-development of quantum field theories, and in simulating the behavior of future quantum computers.

The origin of the sign problem can be traced to large cancellations among terms that lead to a final result that is much smaller in absolute value. While this might seem like a simple nuisance, it has been argued that in particular cases the sign problem is an NP-hard problem. Hence, it is not expected that a complete solution will be found. Nonetheless, various approaches in particular cases have led to significant improvement in our computational abilities. Among these approaches one can mention the Complex Langevin Method, (generalized) Lefschetz Thimbles, and other methods based on contour deformations and analytic continuation. Some approaches are based on machine learning. Algorithms for quantum computers have also been developed.

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[2] P. de Forcrand, Simulating QCD at finite density, 2010
[3] L. Scorzato, The Lefschetz thimble and the sign problem, 2016

2 Major outcomes

Several different approaches towards the sign problem in various statistical systems were presented in the workshop. The approaches divide naturally into several types.

One major family of methods uses the complex plane for handling the sign problem. The complex Langevin method doubles the degrees of freedom of the system and integrates over the whole complexified space of field configurations. On the other hand, path deformation methods, e.g., Lefschetz-thimble methods, do not double the degrees of freedom. Hence, it can be seen as if the latter define a delta-supported distribution in the complexified space. Yet another possibility is to double the degrees of freedom but to integrate only over some parts of the doubled space. This can be thought of as averaging different contours, but since the physical results do not depend on the contour this is similar to averaging over different gauge choices, in a way that is computationally beneficial. There is still no single clear choice for moving into the complex plane and different strategies can be beneficial in different cases.

The choice of an optimal contour deformation is a wide-open question. One does not have to choose a single contour, but in many cases, this is the most natural choice. Naturally, machine learning is a leading tool for performing this task. Other approaches were also presented and discussed.

Another general direction that was discussed is the use of canonical instead of grand canonical formulations, since cancellation between different canonical sectors can reintroduce a signal to noise problem quite similar to the sign problem. Also, there can be sectors with zero contribution due to conservation laws, for example when there is a Z_n symmetry, that would effectively contribute noise in a grand canonical formulation.

One more important point that was raised in several talks in the context of the Langevin method was the reliability of this method. One figure of merit is the contribution of large field values to statistical averages. A number of ways to evaluate this, and to minimize it, were proposed.

Some methods may be useful for a wide variety of physical problems, while others are designed for only one problem at a time. The theoretical and physical applications discussed in the sessions included random matrix models, the doped Hubbard model of superconductivity, sigma models of interacting qubits, quantum spin ladders, emergent Yang-Mills field theory, and QCD for nuclear matter at high temperatures and densities.

3 Community needs

For direct calculations with a strong sign problem, no machine will be efficient enough to give reliable results. When, however, the sign problem is ameliorated, the computational needs in many cases turn out to be quite modest. This is especially true for low-dimensional problems. As a result, many of the simulations performed by researchers in the field run on personal workstations, especially simulations that are intended to be a proof of concept for some technique.

Nonetheless, when more involved physical systems are simulated, and in particular when one attempts to improve our understanding of the QCD phase diagram, the computational cost can be quite significant and large dedicated clusters are used. Beyond this, access to HPC is often needed even to provide proof of feasibility for new approaches and algorithms.

The current workshop was the latest in a series of SIGN workshops, which began in 2009. To the best of our knowledge this was the first time that CECAM supported such an event. The interdisciplinary importance of the sign problem, its frequent appearance in condensed matter and statistical physics, and the scale of computing needs makes the area a natural fit for CECAM. We suggest that CECAM adopt the conference series as its own.

4 Funding

Most researchers in the field are funded by various standard national channels, along the lines of the US National Science Foundation, the Israel Science Foundation, DFG, and ERC.

There are also some specific networks, for example the Collaborative Research Center CRC-TR211, funded by the DFG. Several researchers from this network participated in the workshop.

Possibilities of joint research proposals were not discussed during the meeting sessions. However, members of the SIGN community interact with each other frequently and already conduct long-distance collaborations.

5 Will these developments bring societal benefits?

The focus of the workshop was the sign problem in QCD and, more generally, in physical theories of both high energy and condensed-matter physics. The sign problem and similar signal to noise problems occur in a wide variety of contexts, from QCD to condensed matter systems to molecular structure and dynamics. Hene, the methods discussed in the meeting can benefit society in a more general context than that of fundamental high energy research. As researchers in the field are active also in other communities, we foresee that ideas presented in the workshop will find wide application.

Apart from solution of sign problems themselves, some novel ways to implement machine learning and new algorithms for quantum computing, which were discussed in the workshop, will be useful also for many other problems.

This specific workshop, the eighth in the series, was postponed three times due to the changing constraints of the Covid-19 pandemic. It is thus important to note that the convening of scientists for a meeting in person, after more than two years of Zoom meetings, was important for the community and gave fresh impetus to international collaboration.

6 Participant list

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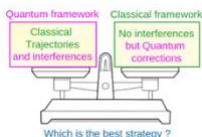Holon Institute of Technology, Israel
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Theories of Molecular Processes and Spectra based on the Quantum-Classical Synergy

Location: Bordeaux Centre Meriadeck Library
Webpage: https://www.cecam.org/workshop-details/1044
Dates: Sep 5, 2022 - Sep 7, 2022

1 State of the art

Nowadays, to ensure the crucial synergy between experiment and theory, theoretical modeling and computations need to increase their precision in order to be of comparable accuracy as the experiments. For few atom systems, exact quantum calculations of the dynamics and kinetics are feasible. However, the exponential scaling with dimensionality makes exact quantum calculations unfeasible for systems involving many atoms. Classical trajectory simulations can be applied to any system, whatever the dimensionality. There are mainly two ways to add quantum mechanical effects to classical dynamics: one is by approximating the quantum propagator, the other is by using ad hoc corrections. Each one presents different strengths and weaknesses. The first approach, pioneered by Miller and Heller, is the more rigorous one. Starting from the exact and intuitive formulation of the quantum propagator given by the Feynman path integral, classical trajectories are some of the paths and the semiclassical limit of the propagator is given by the sum over all possible classical path. [1, 2, 3] Consequently, the resulting methods replace the difficult wave propagation step by a simple run of classical trajectories while accounting for the superposition principle and, hence, quantum interferences. The second avenue of approach, especially for chemical dynamics, amounts to introduce ad hoc quantum corrections in classical trajectory simulations to deal with major quantum effects such as tunneling,[4] non adiabatic transitions [5, 6] or the quantization of initial and final internal motions. This is performed on the basis of intuitive grounds (ZPE corrections, Gaussian binning) and from pre-existing semiclassical models (WKB, Landau-Zener). These methods are extremely popular as they can be applied to high-dimensional systems. In this workshop, representatives from both approaches have presented their progresses.

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[1] W. Miller, Classical-Limit Quantum Mechanics and the Theory of Molecular Collisions, 2007
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*[4] W. Miller, The Journal of Chemical Physics, **51**, 3631-3638 (1969)*
*[5] W. Miller, S. Cotton, Faraday Discuss., **195**, 9-30 (2016)*
*[6] M. Richter, P. Marquetand, J. González-Vázquez, I. Sola, L. González, J. Chem. Theory Comput., **7**, 1253-1258 (2011)*

2 Major outcomes

The major scientific points discussed in the meeting are all related to the development of new quantum dynamics approximated methods. These can be divided into three groups:
1) methods for ground electronic state dynamics;
2) methods for non-adiabatic transition and electronic transition dynamics;
3) method for reactive dynamics and tunneling effects.
As far as 1) is concerned, the methods spam from path integral approaches to semiclassical and quasi-classical ones. Each method has its own advantages or disadvantages, which

includes the possibility to be implemented for on-the-fly ab initio approaches, the amount of accuracy, the system dimensionality limits. The variety of the speakers allowed the audience to appreciate the full range of methodological options and provided a full spectrum of possible ground electronic state quantum dynamics options.

As far as 2) is concerned, the field is less mature than the ground state one, and present a huge number of options. Some methods are more accurate than others but, at the same time, more limited in applications to real systems. All speakers has stressed a certain amount of liability in this field of theoretical chemistry. Most of the approaches are based on classical trajectory procedures with an *ad hoc* quantum expression, ranging from the Landau-Zener to the Surface hopping and the Meyer-Miller Hamiltonian formulation.

Finally, the methods of point 3) ranged from one dimensional analytical formula to many dimensional semiclassical corrections of the rate calculations or even classical condensed phase simulations. This field is still offering room for improvements at the very basic level of theoretical formulation and it is surely the more challenging in terms of accuracy, when comparing with the experimental results.

One issue surely concerns all the methods presented in this workshop. And this issue is the accuracy of a potential energy surface. It should be either provided on-the-fly or with a pre-computed potential energy surface. In the opinion of the great majority of the speakers the accuracy and the calculation computational cost are the main concern for the applicability of quantum dynamics methods to real systems.

3 Community needs

The theoretical quantum dynamics community has been organizing the CECAM workshops without loss of continuity since many years now. Since this community is mainly focused on methodological developments, no needs for new computational infrastructures has emerged from the workshop. In addition, since most of the methods presented are still in their infancy and benchmarking stage, also the need to collect all the methods into one software has not emerged. Nevertheless, some methods are more advances and tested, and some groups are developing some user-friendly software. However, the workshop showed that these groups do not share a common will to have even these more developed methods into one software because they would prefer first to test the software on its own and then, eventually, associate them to a bigger software project, as for example E-CAM.

All participants appreciated very much the format of the workshops, that allowed them for plenty of time for questions, discussion sessions and round tables, and they all agree that this is a peculiarity of the CECAM meeting that should be preserved as much as possible. More often, discussions were occurring during lunch and coffee break times and for this reason hybrid meetings, including zoom sections, should be avoided as much as possible. We received feedbacks from participants that they were going back home with new ideas and implementation for their work thanks to the format of the workshop.

4 Funding

There was no joint proposals discussion programmed during the meeting. We prefer to let the single participant to pick up the chance of the workshop to create groups for proposal after new ideas has been generated after the talks. The workshop had a strong international component, including a large representative from overseas, and there are no many available calls for proposal that can represent the different workshop components at the same time. Europeans' participants, excluding British and Swiss participants, may join together in some sort of Marie-Curie Training Network. This was not taken into consideration during the workshop. However, the workshop offers the chance for the people to meet and to update each other of their research activities and this is the first step for setting up a possible

European network. Thus, we think that if there are the conditions that a quantum dynamics network may flourish after this workshop, it will surely do.

5 Will these developments bring societal benefits?

The topic of this workshop is about fundamental methodological developments. For the methods and the applications presented, social benefits are probable in the very long terms. Nevertheless, there can be new benefits for the society and the community only if this kind of research is performed. In fact, the topic of this workshop is just the first step of a series of steps toward a possible societal impact. We are part of the first gear that bring benefits to the society. Indeed, private companies are appreciating very much our work and more and more often theoretical chemists are employed in private companies, such as Microsoft, IBM, Google, or other international companies more related to chemistry and new material developments. We believe that the possibility in the future to have a quantum dynamics software could greatly enhanced the simulation compartment of private corporations and greatly facilitate the design of new products by replacing the present trial-and-error approaches that most often experimental chemists are forced to adopt. Simulations can be a valid ally in the new product developments strategy and quantum mechanical accuracy of the simulations is very important in order to provide and design a strategy for new discoveries.

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20 years of Metadynamics
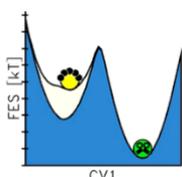
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage : https://www.cecam.org/workshop-details/1117
Dates: Sep 5, 2022 - Sep 8, 2022

1 State of the art

Molecular dynamics (MD) simulations allow complex phenomena to be studied at a unique spatial and temporal resolution. While conventional simulations provide insights into processes that occur on short time scales, the study of rare events such as protein folding, chemical reactions and phase transitions was considered, 20 years ago, prohibitive in many cases of interest. To extend the time scale that is accessible in MD, several enhanced-sampling methods have been developed over the years. Metadynamics was introduced in 2002 as a method to accelerate sampling in MD simulations and at the same time reconstruct free-energy profiles as a function of a small number of descriptors of the system, often known as collective variables (CVs) [1]. Since its introduction, metadynamics has been used to tackle a large variety of problems in computational biology, chemistry, physics, and material science [2,3]. These include problems close not only to basic research in Academia but also to industry, such as applications in the drug discovery field. Over the past 20 years, there has been active methodological development to extend the capabilities of metadynamics beyond its original formulation [4]. Strong efforts have also been dedicated to overcoming the challenge of using a small number of CVs to describe complex processes.
The objectives of this workshop were to:
- provide an overview of recent methodological advances in the metadynamics field;
- illustrate applications of metadynamics in the fields of computational biology, drug discovery, chemistry, and material science;
- discuss open issues and challenges in the field;
- give the opportunity to students and early-career researchers to discuss their projects in a poster session and contributed talks;
- promote networking between students, early-career and more experienced researchers, and developers of the metadynamics approach.

Key References
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2 Major outcomes

The workshop was lifeful and attracted a very significant interest, as demonstrated by the very large number of applications. We decided to allow participation on site only to the speakers, and to accept as on-line participants all the other applicants. The on-line audiance was often above 100. The program was dense and rich. Several recent developments in the areas of algorithms and applications in computational biology, chemistry, physics, and material science emerged. In particular:

- machine learning approaches for the identification of collective variables to bias in metadynamics simulations were presented in different talks, highlighting the important role that these techniques are currently having (and will continue to have) in the field;
- several approaches were presented to characterize kinetics properties of molecular systems;
- on the computational biology side, applications were presented in the areas of membrane proteins, RNA, protein aggregation, liquid-liquid phase separation and viral proteins, in particular SARS-CoV-2;
- several talks illustrated the impact of metadynamics in the drug-discovery area, with contributions from both the academic and the private sector;
- in the chemistry and material science areas, several talks highlighted the impact of metadynamics in the study of nucleation and crystallization processes.

In summary, the workshop clearly demonstrated how method development in the metadynamics field is still extremely active after 20 years and how applications cover a wide range of scientific disciplines.

During the discussion panel, several open questions and challenges in the field were identified:

- need for rigorous approaches to monitor the convergence of metadymics simulations: block analysis on long trajectory versus the use of replicate simulations was discussed, the advantage of using a static bias was mentioned, the need to assess convergence using the raw trajectory rather than the evolution of the bias potential was stressed;
- the computational cost of metadynamics was discussed. This is an issue when "global" collective variables are used. Several tricks (neighbor lists, multiple-time step approaches, GPU implementation of CVs) were mentioned.
- the importance of having human-interpretable rather than machine-learning CVs was discussed. The fact that it is often possible to trace back machine learning variables to simple variables and to add "physics" into ML CVs was mentioned.
- the problem of dealing with sub-optimal CVs was discussed. It was stressed that sub-optimal CVs are very instructive and there are methods, like DeepTICA, that can be used to improve them. In general, it is advisable to use methods that can exploit previous simulations with sub-optimal CVs to improve them. However, in many cases results obtained with sub-optimal CVs might be extremely useful and inspiring to make novel predictions, despite results are not extremely accurate.

3 Community needs

This workshop highlighted the importance of bringing together experts in the field with younger scientists who recently approached this area of research. We selected the on-site participants exactly with the criterion of including young scientists and newcomers in the community. In this regard, two types of events would be extremely useful in the future:

- Workshops open to both computational and experimental communities working in the field. The current workshop was attended by computational researchers only. As most of our studies would benefit from experimental collaborations, in the future we will seek to organize events that favor collaborations between the two communities.

- Practical schools to learn how to use metadynamics and other enhanced-sampling techniques using problems of sufficient complexity. Previous schools in the field teach these methods using oversimplified systems that do not fully reflect the complexity of typical problems in the field. In the future, we will organize practical schools with the concept of "bring your own problem", in order to learn methods on more advanced problems relevant to the participants. Furthermore, these schools should focus on teach students the techniques to address the challenges discussed in this workshop, in particular: convergence of metadynamics simulations, improve computational cost, identifying sub-optimal CVs.

4 Funding

Joint research proposals were not discussed during this meeting, but could be envisioned in the future. This workshop was significantly cofinanced by other funding sources. Other possible founding sources for future activities could be, for example,
- ERC Synergy grants
- International Grants (PRCI) of the French Agence nationale de la recherche (ANR)
- Human Frontier Science Program
- EMBO Advanced Collaboration grants
- SNSF
- EPSRC
- COST Actions

5 Will these developments bring societal benefits?

The meeting also highlighted how metadynamics can have an important role in the drug discovery pipeline. This was shown by speakers coming both from the academic and the private sector.

For instance, in drug discovery, metadynamics is increasingly used both in hit-to-lead and lead optimization phases, with many talks showing its usefulness to design more effective and less toxic drugs. A talk from an industrial speaker (H Kettenberger from Roche) has also shown how Metadynamics-based methods can be effectively used to help design therapeutic antibodies, which are increasingly used in anticancer therapy and other complex diseases. Thus, the topic of the workshop has potential societal benefits also in terms of healthcare.

Another research highlight that has been reported can benefit society by addressing the global climate emergency. A talk from M Parrinello has shown how metadynamics, OPES and similar methods are being used to understand the Haber Bosch catalytic process and eventually decrease the energy necessary for ammonia production. Since ammonia is one of the most important raw materials (for fertiliser production), it is clear that the use of the methods discussed in the workshop can be tremendously impactful.

Finally, a number of talks and posters focussed on the question of degradation of "forever" chemicals and polymers such as PET. Also, in this respect it is clear that metadynamics and similar enhanced sampling methods can help better understand how enzymes that can degrade such polymers work and provide insights on how to make them more efficient.

Thus, the topics covered by the workshop have shown a number of potential societal benefits, ranging from economic to healthcare to addressing the many global emergencies facing us all.

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Understanding function of G-Protein Coupled Receptors by atomistic and multiscale simulations

Location : Aula polivalente, Campus Est USI-SUPSI, Lugano, Switzerland
Webpage : https://www.cecam.org/workshop-details/39
Dates: Sep 12, 2022 - Sep 14, 2022

1 State of the art

The G-protein coupled receptors are the largest family of proteins in the human genome, containing approximately 800 members. They transduce chemical signals across cell membranes in every imaginable physiological context, from immune signaling, to regulating excitable tissue like heart and brain, to the sense of taste and smell. Many members of the family are the target of pharmaceuticals, both already brought to market and in various stages of development. Understanding GPCR function is therefore of broad significance.

Simulations of GPCRs have been an integral part of these efforts since the very first GPCR structure was published in 2000. Since then, computers have become significantly faster and simulation algorithms more sophisticated, while at the same time more and more structure/function experimental data has accumulated. Simulations continue to make major contributions to our understanding of the functional dynamics of GPCRs, to the discovery of new pharmaceutical tools, and to the determination of lipid-dependent function. Increases in computer power have made routine the simulation of GPCR/G-protein complexes, which is leading to a better understanding of hoe GPCRs interact with their signaling partners.

Recent advancements focus on the characterization of activation mechanisms and the capabilities of certain ligands to induce interaction with specific effectors upon binding, a phenomenon known as "biased signaling". Another emerging topic is the elucidation of the

homo- or hetero-oligomerization processes involving two or more of GPCRs, acting as another allosteric mechanism regulating the receptor signaling cascade.

2 Major outcomes

Several topics emerged as focal points:
- **Mechanisms of interaction between GPCRs and their signaling partners.**
  Driven especially by recent advances in cryo-electron microscopy, a great deal of data has accumulated showing GPCRs interacting with a variety of G-proteins, arrestins, and other signaling partners. This has in turn has driven a wave of simulations to understand how different ligands activate distinct signaling pathways in a biased way.
- **Improvement of experimental settings for better investigation of GPCRs.**
  On the experimental side, the development of membrane mimetics (lipid nanodiscs) has opened new possibilities for control of lipid composition, structural biology (especially cryo-EM), and functional characterization, including NMR.
- **Rationalization of the environment influence on GPCRs.**
  It has long been recognized that membrane proteins, including GPCRs, are influenced by their membrane environment. These interactions are transient, and often very difficult to resolve by experimental methods. Simulations have helped fill this knowledge gap for many years, but a modern understanding of mammalian membranes recognizes the complexity of the lipidome both in terms of its composition and asymmetry with respect to the leaflets.

The following open questions were discussed:
- **Full characterization of the biased signaling mechanism.**
  Different ligands can elicit specific interactions between GPCRs and their signaling partners. While some advances have been made, a comprehensive elucidation of the driven forces behind this phenomenon has yet to be reached.
- **Definition of a minimum membrane model for mimicking physiological conditions.**
  It remains an open question to what extent the membrane mimetics employed in experimental/computational settings are faithful to the native lipid environment. Modern simulations on state-of-the-art machines are poised to help resolve this question.
- **Investigation of new classes of GPCRs.**
  GPCRs belonging to class A have always attracted more attention than the others, mainly due to their involvement in many pharmacologically relevant processes. Recent advances with experimental techniques (NMR, CryoEm, etc.) have unlocked the possibility of characterizing previously difficult-to-obtain structures of Taste and Class D GPCRs. Preliminary evidence showed that these receptors have a slightly different activation mechanism with respect to A class GPCRs. Further investigations are needed to better elucidate them.

Main outcomes of the discussion:
- **New tools available for rationalization of GPCR activation mechanism.**
  New approaches and methods have been developed to address the mechanistic aspects of the GPCRs conformational equilibrium. In time, they will provide invaluable insights that will accelerate pharmaceutical development.
- **Advances in the elucidation of the biased signaling mechanism.**
  While an overall rationalization of the aspects regulating such process is still missing, significant discoveries were made. They are expected to have a substantial impact on the pharmaceutical field.
- **Clarification of the importance of the lipidomic complexity for GPCRs function.**
  Membrane composition is an aspect that is attracting considerable interest due to its influence on GPCRs. Simulations are just beginning to address this point, but a breakthrough is expected on the short term.

3 Community needs

The participants reported that the mix of simulation and experimental talks was extremely effective at driving substantive conversations and developing new collaborations. A few specific examples of new collaborations founded at the workshop: (i) A computational drug discovery group with a biophysics/cell biology experimental group, (ii) A bioinformatics group with a molecular simulation group, (iii) NMR spectroscopists/structural biologists with several different simulation groups, (iv) A structural biology group with a simulation group focused on lipid-protein interactions.

Although this field of biophysics (membrane protein function) has long been characterized by impactful collaborations between simulation and experimental groups, it was noted by many participants that these kinds of meetings (bringing together simulation and experimental groups) have disappeared of late, especially regarding the GPCR field. There was thus a real need which was filled by the meeting, and there appears to remain such a need going forward. Based on extensive positive feedback provided by the participants, we do feel that a series of such workshops is warranted, in order to continue the long and successful history of collaboration between structural biologists and simulation groups focused on GPCRs.

4 Funding

The workshop promoted several discussions about the importance of collaborations between experimental and computational groups. It was noted by many that meetings fuelling this kind of interaction have disappeared as of late, which has negatively impacted on the research activity. As such, discussion about joint research projects has been lively and several funding channels, either European or Swiss, are currently being tested for promotion of these collaborations. However, the field would definitely benefit from ad-hoc grants designed to bring together structural and computational groups, which are presently limited in number.

5 Will these developments bring societal benefits?

As mentioned in the introduction, GPCRs are the single most important drug target in the human proteome. 35% of the marketed drugs target a GPCR, and the global GPCR market size is estimated at USD 3.0 billion. While many drugs targeting GPCRs have already been brought to market, many more targets await. An emerging theme focuses on more selective GPCR activation — not targeting at the protein level, but at the biochemical signaling pathway. Many distinct signaling pathways run through a single GPCR, and different native ligands and drugs couple to different pathways. Rationally designing this kind of functional modulation is a major current challenge in the GPCR field. Progress would have an enormous impact. For example, it is now well established that opioid tolerance and abuse is intimately linked with activation of the arrestin pathway over the "canonical" G-protein pathway. Efforts underway leverage massively scaled molecular simulations to learn how exactly distinct drugs preferentially activate different pathways. Success would open a new and promising therapeutic strategy for safely treating pain.

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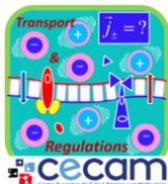**Tan, Zihan** - Forschungszentrum Jülich, Germany
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Charged Species in Bulk and Interfaces: Transport and Regulation

Location: CECAM-AT
Webpage: https://www.cecam.org/workshop-details/1130
Dates: Sep 13, 2022 - Sep 16, 2022

1 State of the art

We have discussed transport of charged species in bulk and at interfaces, focusing on the role played by electrostatics in the dynamics and structure in biologically relevant systems, thus spanning a wide range of length and time scales. This includes the important role of water in the mass transport of ions through confining geometries. The interfacial friction between planar and nanoscopic layers lubricated by water, have been shown to have a strong effect on ion correlations and electro-osmotic flow [1, 2]. Moreover, confinement-induced structure of water affects membrane-desalination and osmotic power harvesting [3,4], and elastic sheets show unusual structures under stress [5,6]. There is a qualitative difference between the degree of ionic hydration ions near the air-water interface as compared to bulk [7,8], while the deswelling behaviour and accompanied transport properties of microgel particles depend on the release/capture of ions confined inside the microgel [9].

Charge regulation of macromolecules have been discussed, both in equilibrium and induced by external fields, which can affect their interactions to an extent that it plays an important role in their phase behaviour. The pH and salt concentration of bacteriophages regulate the chemical dissociation equilibrium of the amino acids, and thereby their electrostatic interactions [6]. The ionic-strength dependent osmotic pressure plays an important role in biological systems [10,11], for which a fluorescence-based experimental technique has been used to visualize the small osmotic differences at hand [12]. Disregard of the variation/regulation of charges and the effect of complex charge distributions, shape and size clearly matter in cell assemblies (organoids), which area often affected by mechanical forces [13], and for active particles, including both single-cell motility [14] and the dynamics and structure in complex and crowded environments [15,16].

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2 Major outcomes

The following subtopics in charged species were extensively discussed, leading to a general improvement of our fundamental understanding:

T1: Transport of charged species in bulk and confinement
T2: Charged active particles near charged surfaces
T3: Collective behavior of charges species in external electric fields
T4: The role of charges in nanopore DNA/RNA sensing
T5: Membrane deformation, permeation, budding, and encapsulation

Concerning T1 and T2, the discussions included the electric field generated at a water-air interface, where simulations were performed on 352 water molecules, over a time span of 200 psec. These effects play a role in the theoretical microscopic investigations of the electrophoretic mobility of charged colloids at high concentrations and electric conductivity. An alternative theoretical approach based on Mode Coupling Theory (MCT), has been discussed the swelling/deswelling behaviour of microgels, governed by electrostatics and elasticity, as well as the resulting structural and dynamics. Here, electrostatic interactions and charge regulation play an important role, which has also been addressed for in homogeneously charged spherical particles. Macromolecules with an even more complex structure, that of ring polymers, have a distinct different phase behaviour and glass formation as compared to linear polymers. Subtopic T3 considerably overlaps with T1 and T2, but is more focused on external-field induced effects, like transport of charged particles in non-polar media and reversible micelle formation under an electric field, where it has been shown that micelles can exchange charges. Further quantification of the effects of external electric and magnetic fields with varying frequency in terms of generalized Green-Kubo response functions is in simulations of electrolyte solutions. Quadrupolar nuclear magnetic resonance (NMR) relaxation experiments concerning the response to frequency-dependent fields of electrolytes in aqueous solutions, and frequency-dependent impedance measurements were presented.

As T4 and T5, theoretical ideas concerning the origin of attractive electrostatic interactions between like-charged surfaces with mobile charges are considered interactions of non-uniformly charged colloids, with the dielectric polarizability of polyelectrolytes in electrolyte solutions. Subsequently, the role of charges in nanopore DNA/RNA sensing (in T4) was presented, on the basis of a MARTINI program. Also, subtopic T5 has been focused on the electro-permeability of hydroperoxide lipid membranes. In addition, some of the results of a detailed experimental investigation in an angle-resolved XPS (ARXPS) of supported ionic liquid phases concerning their microstructural order and anisotropic diffusive transport.

Finally, the physics of active particles (either synthetic particles or bacteria, sperm) is a quite active field of research, between active Brownian particles in analogy to electrostatic interactions. In a comparison to experiments on peroxide-driven Janus colloids, which still unclear how interactions are affected by concentration gradients of the degradation products of peroxide. Also, a single Brownian active particle embedded in a viscoelastic fluid is discussed with small inertial forces in a micro-fluidic channel exhibits quite complex behaviour.

3 Community needs

Action to follow up our series of CeCam/DACam 2022 Flagship Workshops
By closing the 3-consecutive series of our discussion workshops on "Charged Species in Bulk and Interfaces", we have decided to share their outcome to the scientific community, by publishing a special issue in a peer-reviewed journal (tentative "EPJE"). We will invite all the speakers since the 1st CeCam/DACam workshop in 2016, 2nd in 2018, and 3rd 2022, to contribute to the special issue. The topics that have been defined in the three CECAM events will be presented in this special issue in such a way that theory, simulations, and experiments will form a coherent set of publications, on a coherent set of topics: structure, transport, charged colloids, external fields, and molecular aspects.

This special issue will also address the possible future directions of research in the scientific directions addressed in the workshops, the scientific questions that remain open, with an emphasis on what simulations can contribute.

4 Funding

Most of the expenses have been covered by CECAM, while most of the invited speakers paid part of their travelling and housing costs through their home institution. The FZJ offered to contribute in this respect with 5000 euro to account for part of the travelling costs for long-distance oversees from US, Brazil, and Asia. Due to the few participants from such remote countries, this budget was only used this time for the three participants from Jülich (Kang, Dhont, and Nägele paid for their own travel and housing).

5 Will these developments bring societal benefits?

The workshop contained total 12 sessions (with invited speakers) and 3 discussion panels for 3 full days, covering topics over a broad suit of aspects concerning charged particles, from pure theory to sophisticated computer simulations, and experiments, including a number of applications where charge-charge interactions play an important role. We have discussed charged species in bulk and at interfaces, which are significant in many practical applications. These discussions ranged from molecular to the mesoscopic and macroscopic length and time scales, and including a broad scope of systems. Most practical applications are concerned with water-based systems, to which special attention has been paid, including microscopic simulations on the single-water-molecular level. We therefore think that the special issue discussed above will serve the application community towards a rational design of various kinds of products where electrostatics plays an important role.

In this workshop, we have also discussed future applications, like artificial neuromorphic circuits through the design of 3d ionic structures with cylindrica/conic channels (as discussed by Rene van Roij (RvR)), the control of microheterogeneities in bio-ink 3d printings with cell viability (by Claude Oelschlaeger (CO)), and the improvement of screen-imaging performance (Kristiaan Neyts (KN)). This application aspect of the CECAM meetings will most probably serve the community in the development of future applications.

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Building pandemic models on the fly: How to develop data-based models when time is at a premium and good data are scarce
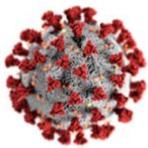
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1114
Dates: Sep 21, 2022 - Sep 23, 2022

1 State of the art

In 2020 the world was faced with a global pandemic: COVID-19, which has claimed millions of lives. The response to this pandemic has involved medics, scientists and engineers. Parts of this response, for example the rapid development of vaccines, have been triumphs of modern science, but not all has been so successful. Epidemiological models such as the Susceptible-Exposed-Infected-Recovered (SEIR) model, can predict the short-time evolution of the number of infected individuals, if one is able to estimate the parameters entering in the model. However, SEIR is in effect a mean-field model for transmission when it is now clear than transmission is extremely heterogeneous (e.g., super spreaders).

SEIR models population data, a complementary approach is to study the biology, chemistry and physics of transmission. The transmission of a virus is in part a physical transport process, out of one body, across air and into another. The fluid mechanics part of this is increasingly well understood, however, the biology often has little predictive power, and the physical chemistry of the disease carrying droplets is complex and poorly understood. These droplets are composed of a virus in mucus (a complex soft matter mixture of large natural polymers, plus salts and surfactants). We also have almost no information on how these droplets are formed, inside the respiratory tract of an infected person.

Despite the interdisciplinary nature of the models needed here, little interdisciplinary research is being done. This leaves us no better prepared for the next pandemic than we were in 2019. What we need, but don't have, is transferable (from one disease to another) models, which need to be based in part on the physical processes (eg droplet formation, aerosols) that are

common to airborne disease transmission. We also need to bridge the large gap between those working on these physical models, and epidemiologists and others working at the level of populations.

2 Major outcomes

The talks, which covered a range of different topics, triggered a lot of discussion and many new ideas emerged for possible topics of future CECAM workshops, or for CECAM mediated collaborations. We analysed carefully our level of understanding of the mechanism of viral transmission.

A first key ingredient is the correct modelling of aerosol transport which, we agreed, is reasonably well described by fluid mechanics. A second key ingredient is modelling correctly the chemistry in the aerosol droplets: indeed, evaporation and diffusion of CO_2 through the air changes the osmotic pressure and the pH, which, in turn, affect the survival of the virus. Understanding these changes by targeted experiments seems difficult, pointing to a possible role for simulations across a range of length scales. A third key ingredient is the size distribution, variability, viral load and impurity concentration of aerosols in the respiratory tract of infected persons. These data, crucial to model transmission, are almost entirely unknown, calling for more experimental and modelling work. For example, it has been suggested to investigate aerosol formation for a simple model of lung that is expanding/contracting due to air flow.

The role of multivalency in targeting of cells by viruses, but also the targeting of viruses by pharmaceutical compounds emerged as an important topic. Again, this topic is well suited for a numerical (but not exclusively numerical) approach. We also discussed our level of understanding of the physics of mask filtration, concluding that on this topic a quantitative analysis is already available. At this stage, there seems no obvious need for a CECAM initiative on this topic.

Many contributions of the workshop were also devoted to epidemiological models, and the state-of-the-art in that field was discussed in depth. We discussed what data we need to make epidemiological models robust, in particular to make predictions reliable at least a week or two in advance. From the talks it became clear that for political/legal reasons, some key information has not been made available to the scientific community during the epidemic, in particular the more sensitive data related to contact tracing. As a consequence, crowd-sourcing of pandemic modelling is difficult, and the role of the broader community is necessarily limited.

Another topic that emerged is the importance of using the appropriate level of complexity in epidemic models: possibly some data are available, but are not necessarily useful, so could be dropped to simplify our current models. It would be appropriate to exploit tools from information theory and advanced Bayesian inference to address this issue quantitatively.

We also noticed that heterogeneity - of many forms - is likely to be very important, but often is neglected, as it cannot be accounted for in the most widely used models, such as, for example SIR models. It is well known that some infected people are much more infectious than others, some have far more contacts, and/or are far more mobile. For example, one can imagine a situation in which in geographic location R0 > 1 while in another, perhaps nearby, R0 < 1, but the mobility between the two is large. It would be interesting to study and expand the models taking explicitly into account these large deviations. We finally noticed that current models do not provide insight into some of the striking qualitative features of the Covid epidemic curves, such as the typical duration of the wave (~ 2 months) and the presence of long periods of time in which the epidemic looks pretty stationary.

3 Community needs

Aspects of COVID-19 transmission span many decades in space and time, i.e., are multiscale phenomena. Transmission seems dominated by micrometer-sized aerosol droplets containing viruses with a diameter of around 100 nm, nanoscale mucin proteins, and Angstrom-size salt ions. Both the formation and evaporation of these droplets takes a fraction of a second, i.e., a timescale orders of magnitude larger than molecular timescales. Due to the difficulty of obtaining direct experimental data on both the formation and evaporation of these droplets, multiscale simulations can play a role here. Currently, these are not being done, so CECAM could catalyse such work, for example by bringing simulators with relevant expertise together with experimentalists, and those interested in understanding the transmission of airborne diseases.

Such work would not require new codes, LAMMPS and similar should be sufficient, just awareness amongst simulators, and links between this community and others who understand the key problems.

4 Funding

A H2020 ITN could bring scientists together into an interdisciplinary team. The work needed to take this field forward is interdisciplinary, and cohorts of researchers with complementary skills would be far more effective than isolated individuals. These schemes allow funding for cohorts of PhD students, although the interdisciplinary nature of the required research may make obtaining funding challenging. Since the topics treated in this workshop are of possible interest also for healthcare, one can consider co-financing future activities involving hospitals and civil protection.

5 Will these developments bring societal benefits?

Thus far, the COVID-19 pandemic has killed approximately ten million people. Many more people have had COVID symptoms. Moreover, long COVID is now affecting many. The flu pandemic a hundred years ago probably killed more than fifty million people. So, the benefits of better understanding airborne transmission are obvious. Airborne transmission is a complex process, both in the physical mechanism (multiscale phenomena) and the epidemiology (highly heterogeneous transmission).

In the workshop we heard of examples where the pandemic had brought together the interdisciplinary teams together to understand these complex phenomena. If future work can build on this interdisciplinarity to deliver further research, we should be better prepared for the next pandemic. Better understanding of transmission in various environments will inform more rational Non-Pharmaceutical Interventions (NPIs) such as mask wearing and room ventilation, while better epidemiological models will inform restrictions on movement, working from home, and similar measures, which better balance reduced transmission with societal and economic costs.

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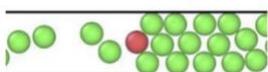**Morand, Jules** - University of Trento, Italy
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Large deviations, extremes and anomalous transport in non-equilibrium systems

Location: CECAM-AT, Erwin Schrödinger Institute, University of Vienne, Austria
Webpage : https://www.cecam.org/workshop-details/984
Dates: Sep 26, 2022 - Sep 30, 2022

1 State of the art

Characterizing the steady-state and the dynamical properties of systems driven out of thermal equilibrium is currently a subject of intensive theoretical and experimental studies. Considerable theoretical progress in understanding various aspects of such non-equilibrium systems has been made recently and the aim of this workshop was to bring together leading researchers and young scientists around three main themes:
(i) Theory of large deviations: large deviation functions have been considered as possible candidates for an effective free energy in out-of-equilibrium situations. In particular, the idea of detecting a phase transition by studying the singularities of the large deviation function has been used in various areas ranging from anomalous transport and stochastic thermodynamics to active matter and disordered systems.
(ii) Rare events, extremes and first-passage properties in stochastic processes: Extreme value statistics plays an important role in statistical physics, e.g., in the context of disordered and non-equilibrium complex systems. One of the major current issues is to understand the role of correlations between different degrees of freedom in characterizing the

universality classes of extreme value distributions. There has been some recent progress in calculating extreme value distributions exactly in strongly correlated systems such as in random matrix theory.

(iii) Anomalous transport in low dimensional systems: Very recently, important progress has been made in this highly active area of research whereby a general framework has been proposed for understanding anomalous transport in a broad class of systems. This framework, referred to as non-linear fluctuating hydrodynamics, provides a successful approach, which has generated considerable activity aiming at testing the general validity of this theory.

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2 Major outcomes

At the workshop, which was co-funded by the Erwin Schrödinger Institute (ESI) in Vienna, there were four to seven talks per day, with a total of twenty-seven speakers -- and more than fifty participants-- from all over the world. Most of the talks have been recorded and can be viewed at https://www.esi.ac.at/events/e431/. Between the talks, there were many slots for discussions and the participants could enjoy the stimulating atmosphere provided by the ESI to share ideas and initiate collaborations. For many participants, this workshop was the first opportunity since the pandemic to meet colleagues in person, which was particularly appreciated. The themes of the Workshop were discussed along the following main lines:

1. Active matter
Active particles form a class of non-equilibrium systems which generate dissipative directed motion through self-propulsion and consuming energy from their environment. The study of active particles is relevant in a wide variety of biological and soft matter systems ranging from bacterial motion, formation of fish schools and bird flocks, as well as granular matter and colloidal surfers. Recent years have seen a tremendous surge of research on active matter. This workshop gathered many leading experts of this lively subject who presented recent developments on the subjects. These include theoretical as well experimental progresses. On the theoretical side, on can mention the recent development of analytical tools to describe stochastic processes which are relevant for active systems as well as the characterization of the effects of quenched disorder. On the experimental side, recent progresses were reported on the detailed study of the motion of a single active particle in viscoelastic fluid.

2. Large deviations and their applications
Large deviations were at the core of several talks and discussions during this workshop. Indeed, large deviations have recently found a lot of applications and several new results were presented during the workshop. This includes in particular macroscopic fluctuations in interacting particle systems (e.g., exclusion processes), applications in chemical physics or in the context of microbial growth or the development of new theoretical tools (e.g., tensor network techniques).

3. Stochastic thermodynamics
Stochastic thermodynamics provides a universal framework for analyzing nano- and micro-sized non-equilibrium systems. Prominent examples are single molecules, molecular machines, colloidal particles in laser traps and biochemical networks. Thermodynamic notions like work, heat and entropy can then be identified on the level of individual fluctuating trajectories. They obey universal relations like fluctuation theorems, which have generated a lot of activities during the last two decades. This was also an important theme of this workshop during which several theoretical advances were presented, including the concept of thermodynamic inference which uses consistency constraints derived from stochastic

thermodynamics to infer otherwise hidden properties as well as the quantum extension of the classical fluctuation theorems.

4. Resetting dynamics

During the last ten years, resetting of stochastic processes has become a major theme of study in non-equilibrium statistical physics. An archetypal example is a diffusing particle that is reset to the origin after random waiting times with exponential distribution. It has been shown that (i) the system reaches a non-equilibrium stationary state at large time and (ii) the mean first-passage time to some target is rendered finite, rather than infinite as in the absence of resetting. During the workshop, several new results on resetting were presented, in particular one of the very first experimental realization of this resetting protocol using optical tweezers.

3 Community needs

This workshop, which was part of a longer thematic program at the ESI in Vienna, brought together researchers working in various subfields of statistical physics and included theorists, computationally oriented scientists as well as experimentalists. As such, the workshop was a very useful forum to exchange ideas and tools that are central and established in one field, but perhaps not yet fully developed in another. For instance, the large deviation formalisms, widely applied to transport in interacting particles systems, is also very promising for the understanding of motility induced phase transitions in active matter. The community would certainly benefit from such cross-disciplinary meetings in the future.

Another important issue is that the non-equilibrium nature of driven systems complicates also the numerical simulation of rare events occurring in these systems. The reason is that, in contrast to equilibrium systems, the underlying stationary distribution is usually unknown. As a consequence, importance sampling methods developed for equilibrium systems cannot be applied straightforwardly to non-equilibrium systems. New numerical approaches for large deviation simulations would be very valuable for the community.

4 Funding

Funding channels for the community were not really the focus of the workshop. However, funding opportunities for networking but also for future meetings were discussed in small groups. On the European level, doctoral networks within the Marie Sklodowska Curie scheme of the European Commission offer interesting possibilities to bring different institutions together to train PhD candidates. Various possibilities for research networks involving a number of scientists exist also at the national level in different countries, as discussed in the workshop. Future efforts to bring the community together might rely on applications to the Lorentz Center in Leiden or the Kavli Institute of Theoretical Physics at UC Santa Barbara.

5 Will these developments bring societal benefits?

As the discussions of the workshop centered mainly on theoretical concepts and tools, it is difficult to tell, which societal benefits they will bring except to say very generally that they will advance our understanding of non-equilibrium systems. Nevertheless, one can argue that many systems in biology but also in materials are out of equilibrium and hence methods to describe such states on a basic level can eventually be beneficial for society. For instance, extreme events occurring in weather/climate systems can obviously be very consequential and the ability to quantify the probability of such events based on numerical models is very important. Rare events may also play a role in the design of new materials self-assembling under non-equilibrium conditions driven by active particles. Finally, the large deviation formalism, and simulation techniques developed to evaluate large deviations functions, might help us to better understand and manipulate biological systems on the microscopic level with implications for health applications.

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Co-Design for HPC in Computational Materials and Molecular Science

Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1113
Dates: Oct 3, 2022 - Oct 5, 2022

1 State of the art

Co-design is a cooperative and concurrent process of designing both hardware (HW) and software (SW) components. In high-performance computing (HPC), co-design helps to identify specific code features that are relevant to isolate, classify, and optimize key compute-intensive sections of the codes (kernels), and eventually guide the development of future exascale systems.[1]

Researchers in atomistic computational materials and molecular science have developed several leading software used worldwide on massively parallel computers. While researchers in computational fluid dynamics, seismology or weather have routinely used their codes to apply the co-design, the engagement and effort put so far from the side of the atomistic simulation community to push and influence the development of future architectures for exascale systems have been more limited. [2-5] Co-design for exascale applications is a hot topic in the computational science community and is pushed by the European Commission and the EuroHPC Joint Undertaking. The European HPC Centres of Excellence (CoEs) are particularly engaged in co-design for various scientific domains and with the involvement of E-CAM, two CoE workshops on co-design have been organized by the FocusCoE CSA in Spring 2021. These workshops had a rather broad scope involving all domains covered by CoEs. Because of this, the full potential and possible practical strategies of co-design in materials science and related disciplines are still partially unexplored.

Additionally, in the UE context, the European Processor Initiative (EPI) is an open opportunity to impact the development of future architectures. Ensuring that, during the path to exascale, ab initio and atomistic modelling applications can achieve optimal performance on EPI hardware in terms of execution times and power efficiency is of crucial importance, making co-design in materials and molecular sciences particularly timely and relevant.[6]

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2 Major outcomes

This workshop gathered the contributions of leading academic scientists, industry researchers, coordinators of HPC centres, as well as software and hardware engineers. It covered a quite broad range of timely topics, taking atomistic materials and molecular science codes as reference HPC applications and possible co-design vehicles. Given the relatively

small size of the meeting, and that all participants attended in person, one of the main outcomes of the workshop was its strong potential to foster close contact and exchange of ideas among scientific and technical communities, which are otherwise not usually in touch with each other.

The main topics were covered in individual sessions, which contained invited presentations and round-table discussions. The four selected topics were the following:

• HPC centre co-design, where members of major European supercomputing centres shared their views on co-design and how the layout of new centres is influenced.

• Application development and co-design, where leading members of the scientific software development community discussed the challenges of adopting state-of-the-art technologies while targeting the porting of their codes on emerging computer architectures, shared existing examples of HPC co-design, and open questions about code adaptations and co-design approaches.

• Library development and co-design, where scientific library developers broadly used in materials science applications, presented leading-edge technologies for supporting new HPC architectures. In this session, the development of general-purpose and domain-specific libraries, kernels, and mini-apps which can be potentially used in co-design was discussed.

• Hardware vendor and integrator perspectives, where state-of-the-art HPC hardware architecture manufacturers, designers and integrators discussed how they are influenced by the scientific software community.

In particular, the workshop focused on and drew key conclusions about the relationship between algorithms and computer architectures in materials science, the connection of parallel programming technologies and runtime systems, and the interplay of the above layers for a wide spectrum of computer architectures. In terms of output and goals, there was a long discussion on how scientific software developers in materials science can influence hardware manufacturers as well as middleware and system-level software developers (and vice versa). There were also some inspiring examples of how the academic research community can influence the decisions of hardware manufacturers. On the other hand, hardware HPC vendors and integrators exposed their visions on co-design, exposed their openness to hearing the requests from researchers and code developers, and highlighted their willingness to support the community.

3 Community needs

Simulations of materials and molecules are constantly enabled by the increasing computing power and the upcoming availability of HPC exascale computing. The evolution of these research branches requires larger and more powerful computing resources, which will allow the users to face challenges that were not achievable before due to the large computational cost of the most accurate simulation methods, with the subsequent limitations in systems sizes and structural complexity. Moreover, the heterogeneity of computer architectures is also expected to increase in complexity with more specialized and efficient devices. In this context, co-design between computational scientists and the hardware industry has to be seen as a bidirectional effort, where the application developers and hardware designers provide feedback to each other. This workshop showed how co-design is currently employed in the development of the new HPC processors and of the related software stack, ranging from compilers to optimized libraries, to schedulers and IO, to container technologies. These developments were discussed from the perspective of current research in materials and molecular science.

4 Funding

This workshop was co-organized by the NOMAD, MaX, BioExcel and TREX Centres of Excellence (CoE). The collaboration between these CoEs is quite active, and it will be enhanced after this workshop. Most of the participants of the workshop were based in Europe, which will certainly feature in new grant proposals and collaborations in the context of the Horizon Europe programme. The event also had a very clear industry-oriented component. Some of the industry participants are already involved in the above-mentioned CoEs, for example, Intel and NVIDIA are part of the NOMAD's industrial second shell, while SiPearl, ATOS and ARM have taken part of different MaX projects. After this event, new synergies between hardware designers, European HPC centres and academic researchers were privately discussed, and we expected these to be set in the near future.

5 Will these developments bring societal benefits?

Research of novel materials and molecular design has a clear impact on the economy, environment and public health. Predicting and understanding novel materials and molecules with target properties is a constant requirement in fundamental science, engineering and industry. While computational modelling techniques have been widely used for the simulation of all types of materials and molecules, the accuracy and complexity of the models are often limited by the availability of computational resources. Therefore, the constant evolution and improvement of these models will be further enhanced by the incoming exascale HPC resources, which are also expected to come along with the increasing heterogeneity of the computer architectures. On this ground, co-design is a key vehicle to obtain the best performance on each architecture and therefore allowing the fastest development of new materials and drugs, at the lowest computational (and therefore environmental) cost. The research resulting from these activities will certainly have an immense impact on the IT, energy, environment or medical sectors.

6 Participant list

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International Workshop on "Reduced Density-Matrix Functional Theory: Improving its foundation and extending its scope"

Location: ECT, Trento, Italy
Webpage: https://www.cecam.org/workshop-details/24
Dates: Oct 3, 2022 - Oct 14, 2022

1 State of the art

It is a fact that the energies and other electronic properties of atomic and molecular ground states can be computed directly from the two-electron reduced density matrix. Yet, since each subfield of the quantum sciences typically restricts to systems all characterized by a fixed pair interaction, the ground-state problem should *de facto* involve the one-particle reduced density matrix (1RDM), only. The corresponding exact theory is known as Reduced Density Matrix Functional Theory (RDMFT) which is based on the existence of an *exact* energy functional of the 1RDM [1]. Compared to the widely used Density Functional Theory (DFT), RDMFT has some significant conceptual advantages: the kinetic energy is described exactly and strong electronic correlations can be described efficiently. Benchmark calculations have revealed indeed that the common functionals in RDMFT can yield correlation energies that are a precision comparable to Møller-Plesset second-order perturbation theory. RDMFT has also succeeded in predicting more accurate gaps of conventional semiconductors than semi-local DFT. Recently, we have witnessed remarkable progress on RDMFT along several lines:
1. The first RDMFT software code has been made public [2].
2. RDMFT has been extended to excited states by generalizing the work on ensemble DFT [3].
3. Novel ideas were proposed to improve the numerical minimization of functionals [4].
4. RDMFT has been extended to bosonic quantum systems, including ultra-cold gases [5].
5. The cusp condition due to the Coulomb interaction has finally been translated into a property of the 1RDM [6].
6. Modern machine learning techniques were put forward for improving functional approximations [7].
Most of these novel ideas are rather challenging, fundamental, and of potentially high impact. A crucial challenge is to combine them to improve the foundation of RDMFT, overcome its recent limitations, and extend its scope.

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2 Major outcomes

The aim of this international workshop was to discuss and explore new aspects and challenges in Reduced Density Matrix Functional Theory (RDMFT). For this, 13 experts in RDMFT were present at the ECT* Center in Trento (Italy) for an intensive and informal meeting during 3-14 October. This in-person workshop was complemented by five mini-symposia (hybrid format), open to a broad international audience: 85 scientists (ranging from Ph.D. students and postdocs to professors and leading experts) were present during the discussions. Each symposium consisted of 6 talks of 22 min + 8 min for questions and further discussions. The schedule was the following:

Symposium 1 - October 3, 14:30-18:00 CEST

Exact results in RDMFT: properties of universal functionals, the role of N-representability.

Talks: E.K.U. Gross (Jerusalem), David Mazziotti (Chicago), Sarina Sutter (Amsterdam), Federico Castillo (Santiago de Chile), Christian Schilling (Munich), and Tomasz Maciazek (Bristol).

Main topics: the challenge of strong correlations, RDMFT for the canonical ensemble, and recent results in convex N-representability.

Symposium 2 - October 5, 9:30-13:00 CEST

RDMFT for quantum chemistry: Computational and theoretical state-of-the-art and open challenges

Talks: Jerzy Cioslowski (Szczecin), Mario Piris (San Sebastián), Neil Qiang Su (Nankai), Emmanuel Fromager (Strasburg), Jian Wang (Zhejiang), Lexin Ding (Munich).

In this symposium, some common misconceptions concerning the construction and properties of approximate functionals and novel ways to combine RDMFT with embedding quantum theories were discussed.

Symposium 3 - October 7, 9:30-13:00 CEST

Extending the scope of RDMFT (Bosons, ultracold gases, and superconductors)

Talks: Alessio Recati (Trento), Christian Schilling (Munich), Julia Liebert (Munich), E.K.U. Gross (Jerusalem), Carlos Benavides-Riveros (Dresden), and Tomasz Maciazek (Bristol).

This symposium focused on novel opportunities for RDMFT like bosonic mixtures, quantum droplets, and superconductors.

Symposium 4 - October 10, 9:30-13:00 CEST

Conceptual aspects of RDMFT

Talks: Thierry Deutsch (Trento), Paul Johnson (Laval), Sofia Bousiadi (Athens), Derk Kooi (Amsterdam), Julia Liebert (Munich), and Mauricio Rodríguez-Mayorga (Amsterdam).

Main topics: Efficient algorithms for non-interacting ensembles, analytical properties of exact functionals, and relativistic RDMFT.

Symposium 5 - October 12, 9:30-13:00 CEST

Excited states, time evolution, machine learning, and more

Talks: Julia Liebert (Munich), Pina Romaniello (Toulouse), Carlos Benavides-Riveros (Dresden), Christian Schilling (Munich), and Giuseppina Orlandini (Trento).

Main topics: machine learning for RDMFT, ensemble RDMFT for excited states, and the mathematical definition of exchange forces in the context of RDMFT.

Several other aspects like the extension of RDMFT for magnetic systems and the relation between correlation energy and entropic measures of the natural occupation numbers were

also discussed in the informal meetings that the workshop promoted among the participants. The quite flexible structure of the workshop was pivotal to promoting discussions not originally planned by the Organisers.

3 Community needs

During the workshop, it was clear that the community requires better coordination of efforts such that the results in RDMFT can be quickly acknowledged and improved by other groups. The need to have better codes that are accessible to the community at large was also highlighted. Machine learning is an opportunity for that purpose.

It is important to differentiate the term RDMFT from other acronyms used in different fields (for instance, DMFT sometimes is also used for density matrix functional theory which is somehow confusing). The participants agreed that a more convenient acronym is 1RDMFT.

It is also quite important to organize a similar workshop in the future. Between the workshop organized in Lausanne in 2017 and this one in Trento in 2022, 5 years elapsed, which may be too much time to structure a better development of the field. One possibility is to establish a workshop on a regular basis every two years with the support of CECAM and other funding agencies.

Finally, the communication of new ideas could be done outside of papers and conferences. A step in that direction is the site on YouTube where the talks of our workshop can be found (shorturl.at/FLO79).

4 Funding

CECAM and Psi-k are quite useful funding channels for the organization of workshops on RDMFT. Since several members of the community have received funding from European agencies (Marie Skłodowska-Curie Actions or the German Emmy Noether program), the possibility of joining different research projects was discussed. In the near future, we foresee an increase in the collaboration efforts of the members of the community. The recent extension of 1RDMFT to ultracold gases and bosonic mixtures opens a new opportunity to receive funding outside the community of electronic-structure theory. For this 2022 workshop, we acknowledge the support of the Austrian CECAM node.

5 Will these developments bring societal benefits?

As Organisers, we wanted that our workshop reached people who could not be physically in Trento. For that reason, we organized a sequence of 5 hybrid symposia. The talks can be found on YouTube. Furthermore, the slides of the presentations and other materials can be found on the webpage of the workshop: https://indico.ectstar.eu/event/153 . As a result, any person in the world can follow the main outcomes of our work in Trento.

Yet, the main benefit of developing RDMFT is the idea of capturing strong correlations in an effective but exact manner. This will result in the exploration of new strong-correlated quantum regimes that can lead to the understanding of new chemical reactions or find new stable chemical compounds that cannot be found using DFT.

A more ambitious goal is the fact that RDMFT for superconductors can potentially lead to a novel route for understanding non-conventional superconductivity, an outstanding yet unsolved problem in electronic-structure theory.

6 Participant list

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Immobilizing peptides and proteins: Interplay between theoretical and experimental approaches
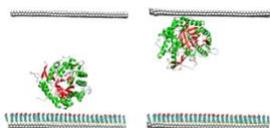
Location : CECAM-FR-MOSER, Institut de Biologie Physico-Chimique, Paris
Webpage : https://www.cecam.org/workshop-details/1132
Dates: Oct 4, 2022 - Oct 7, 2022

1 State of the art

The immobilization of peptides and proteins on membranes and solid supports has attracted growing interest in the biomaterials field over the last 40 years, as this issue plays a central part in numerous applications [1-3]. When investigating how a biomolecule and a surface interact, one must consider both the stability of the interface, and the biomolecule orientation on it. For example, in bio electrocatalysis devices, ensuring correct protein orientation will enable direct electron transfer between the adsorbed redox enzyme and the electrode. One must also make sure that the enzyme active site remains accessible after immobilization. The conservation of the adsorbed protein's structure and dynamics is important too, as perturbations in the protein conformation or internal mobility are likely to result in a decrease of the catalytic activity. These questions are central for devices such as biosensors or biofuel

cells [3], which use rely on the conservation of biomolecular function out of the cellular environment

Over time, computational models have played an increasingly important part in this field, as they can bring greater hindsight on the processes taking place at the bio nanointerface. The last decade has seen a wealth of methodological developments, with empirical force fields parametrization for a wide range of materials, and multiscale approaches combining all-atom and coarse-grained representations. As a consequence, molecular simulations techniques are now a powerful tool in the biomaterials field [4-7]. Simulations can provide us with information regarding the orientation of the adsorbed molecules, its strength and its impact on their structure. This is of particular interest, since an important issue addressed when dealing with immobilized systems is the conservation of their reactivity. When setting up an immobilization strategy, one must achieve a delicate balance between the enzyme stability and its catalytic activity [8].

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2 Major outcomes

The workshop gathered roughly 35 participants, including students, with a balanced program and sessions dedicated to both theoretical and experimental approaches. The interest from the experimentalist and theoretician and computer chemists was very high as reflected the exciting discussions.

The program was organized in different sections dealing with experimental and computational studies as well as combination of theoretical/experimental approaches to address protein immobilization of surfaces. The interest from the experimentalist and theoretician and computer chemists was very high as reflected the exciting discussions during the session as well as during the breaks. Various computational approaches were discussed throughout the meeting. Among them, the use of coarse-grained representation of protein-surface systems were presented (V. Tozzini) and their combination with atomistic molecular mechanic description (G. Brancolini S. Corni, V. Lobaski and J. Subbotina). Sampling of the protein/peptide conformational states on complex surfaces was addressed by R. Wade using Brownian Dynamics or by using Free energy calculations (N. Reuter) and enhanced sampling approaches such as molecular dynamics simulations (J. Dragelj and S. Muralikumar). Although force field parameters for proteins (and lipids) are well established, accurate force field for surfaces are rare and, in most cases, parametrization is performed for specific purposes. There was a general consensus on the need for developing and sharing force field parameters for, for example, gold surface. FF parametrization for interfacial processes can profit from machine learning techniques as stated by S. Sarupria.

Among the experimental techniques, the coupling of electrochemistry with various spectroscopical methods is being currently used for investigating, in particular, the adsorption of redox proteins on electrodes. A mayor issue in these experiments is to assess and control the optimal orientation of the macromolecules on surfaces by assuring a good electronic communication with the electrode and without losing activity. Various examples were presented by A. Poulpiquet, A. Fischer, J. Kaar and the C. Karafoulidi-Restou. Other experimental approaches were introduced, such as SEIRAS spectroscopy were nicely

explained by K. Ataka, K. Hauser, A. Waffo and J. Kozuch. Promising novel nanostructured materials for protein immobilization such as metal organic frameworks, a promising platform as they can enhance the enzyme resistance against solvents and high temperatures, or polypeptide surface coatings were presented by P. Falcaro and P. Lavalle. A common problem in these approaches is the accurate characterization of the surface-biomolecule interface. None of the experimental techniques presented in this meeting can provide an atomistic description of the complex interface. Most of the information is obtained indirectly through systematic analysis of spectroscopical patters, transmembrane currents, catalytic activity or fluorescent quantum yield among other. In basically all cases computational models are used for the interpretation of the complex set of observables.

Thus, the following key issues for the development of modeling approaches were addressed the final round table: 1) development of new surfaces for protein immobilization and proper surface treatment enabling its functionalization so that one can tune surface properties to interact specifically with a target biomolecular system; 3) accurate modeling of surface reactivity since the adsorbed biomolecule may form covalent interaction with it, 4) sampling as the investigation of phenomena like the surface coverage and the interaction between the adsorbed biomolecules requires large-scale simulation setups with a high computational cost and 5) validation of in silico structural models with adequate experimental data.

3 Community needs

- The discussions highlighted the importance of communication between experimental and modeling groups. The need for common databases sampling structural information on solid interfaces was also discussed. The final discussion highlighted the need for simple experimental setups for validating and testing computational models.
- Simulations including biomolecules and solid surfaces are still non-standard in the molecular modeling field and the scientific community needs the development of new force fields to describe the energy between the interacting partners. In addition, we still lack online resources (such as tutorials) for researchers in the field.
- During the final discussion, there was a common agreement that this topic is not sufficiently addressed in most scientific events dealing with biomolecular modeling and would probably deserve the organization of specific sessions. Some participants already asked for the next CECAM event on this topic.
- The possibility of organizing a GRC was also discussed, in particular with the American participants of the workshop.

4 Funding

- The UniSysCat cluster of Excellen from the TU Berlin sponsor the oversee travel costs of Prof.Sarupria (2000€)
- The workshop also received funding (500€) from the French GGMM (Groupe de Graphisme et Modélisation Moléculaire).
- Other potential funders for later meeting are CNRS Gdr (Groupe de Recherche), such as BIM (Bioinformatique Moléculaire, https://www.gdr-bim.cnrs.fr/) for the modeling aspects and B2I (Bioingénierie des interfaces, https://events.femto-st.fr/GdR_B2i/fr) for the experimental side of the topic.

5 Will these developments bring societal benefits?

- The immobilization of enzymes already represents an important topic for issues related to biotchnologies (like energy production in biocells) and the development of medical devices (biosensors, implants, drug-carriers...)
Understanding the interaction between solid surfaces and biomolecules is a key issue from an environmental perspective. For example, participants of the workshop (J. Subbotina) are taking part to the SmartNanotox project (http://www.smartnanotox.eu/) whose goal is to determine the toxicity of nanomaterials. Modeling tools are also important for determining how nanoparticles can degrade in the environment. Finally, biotechnological developments (such as biosensors, or bioanalytical devices) will often involve biomolecules coupled to solid surfaces. A. de Poulpiquet talked about redox enzymes grafted on electrodes and how they can be used to develop biofuel cells, and P. Falcaro presented nanostructured materials for protein immobilization such as metal organic frameworks, which are a promising platform as they can enhance the enzyme resistance against solvents and high temperatures.

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Electronic Structure Software Development: Best Practices and Tools

Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1121
Dates: Oct 10, 2022 - Oct 16, 2022

1 State of the art

There are now a sizable number of mature electronic-structure codes under very active development, becoming increasingly more complex as new methods emerge and existing methods and workflows evolve. The relentless advance in the development of novel computer architectures is putting pressure on the developers to provide performance portability in addition to new functionalities. New, open paradigms for collaborative development have emerged. This shift has contributed to diluting the former silo mentality, both at a scientific level (one research group, one code) as well as at the business model level (libre software vs. open-source vs. proprietary), allowing collaborations between communities and making new public-private partnerships possible.

The Electronic Structure Library initiative promotes the development of standardized libraries and their release in a bundled fashion for ease of use. This bundle solves various installation issues for end users and enables a smoother integration of the shipped libraries into external codes. In order to maintain the compatibility of the bundle with the main electronic-structure codes on the long run, its development has been accompanied by the creation of the ESL Steering Committee, which includes representatives of both the individual ESL component libraries and of the codes using them. Therefore, the visibility of the ESL expands and the developers are exposed to an increasing amount of feedback, as well as requests from third-party applications.

The ESL initiative has been focused on content and usability of the libraries. However, with the growth in the number of contributed libraries and users, it has become clear that a minimum set of requirements is needed to sustain the quality and usability of the components. At the same time has come the realization that in the electronic structure community there is still a lot of effort required to sustain paradigms for developing software using *best practices*, the focus of the current workshop.

2 Major outcomes

The present workshop brought together speakers and participants who shared experiences on best practices in community development, including:

- Ensuring that the library and codes are sufficiently tested and correct by providing tests that check functionality and correctness.
- Automating infrastructure for testing, building and addressing compiler compatibility issues.
- Allowing researchers with non-software backgrounds to employ *best-practice* methods when developing code, such as for performance critical aspects as well as for maintainability and interface issues.
- Providing sufficient documentation, both for developers interfacing libraries in a hosting code, and for users who require knowledge of the functionality provided.

The first two days consistent of talks sharing and highlighting best practices around key Electronic Structure projects, including abinit, cp2k, bigdft, siesta, questaal and quantum espresso, but also from more focused projects like libxc, libmbd or workflows, aiida, asi. Importantly, the workshop included perspectives and a panel discussion from numpy developers, a major effort that is distinct from the electronic structure community (although widely used). One major outcome of the first two days is the intent to create a joint paper that focuses on technical best practices in scientific software development, as well as on best practices in software community building and community coordination. This paper would also highlight CECAM's central role in fostering this community over many years.

The perspectives presented by the participants will serve as key components of this paper, augmented by contributions from other leading community organizations in the field (notably the U.S. Molecular Sciences Software Institute, MolSSI).

A CECAM software seminar was suggested as a way to present ESL initiative to other communities. Steering committee will have to liaise with CECAM about the best time.

In the remaining days of the workshop, small working working groups met to discuss specific topics:

1. IO: Damien Caliste, Martin Lueders, Dimitar Pashov and Alin Elena, discussed about the importance of having a unified hdf5 interface for fortran codes, and how this should shape the development of the electronic structure common data format (ESCDF), and the corresponding library. https://gitlab.com/dmt/hfh
2. Unit testing: Developers were reporting on their current practices, and available tools were investigated, e.g. the fortuno framework, developed by Balint Aradi. Alin Elena tested fortuno for a small proof of concept fortran code and Pietro Delugas implemented it in one of the quantum espresso modules (fft) replacing the in-house code. https://github.com/aradi/fortuno
3. Libxc version 6 was released during the workshop and members tested it prior to adoption by the major electronic structure codes.
4. Easybuild and spack packages are de facto standards for HPC deployment. We discussed adopting spack in addition to easyconfigs which we already support. Additional resources will need to be invested by the community to support these. Critical knowledge of spack was identified in the community, with packages like siesta already providing them.
5. We have discussed the importance of having unified and standardised build systems for our packages. A de facto standard was identified as cmake. CMake already has features to help with continuous integration and testing. A need for recipes to help the community to adopt them was identified and will be addressed via the whitepaper. Integration with github and gitlab ci was identified as a community need. Enforcing modern software engineering techniques like library versioning was demonstrated.

3 Community needs

The ESL initiative is, in fact, centered around a key community need in the experience of the Organisers, i.e., a venue to communicate and (where possible) create bridges between the diverse software projects and developments surrounding electronic structure theory.

Ongoing needs of the community, in the Organisers view, include continued open communication channels to high-performance computing centers as well as general developers of high-performance computing environments, which are essential to the mission and success of the core electronic structure community. Our software developments have relied on hardware and software stacks as well as usability choices that are separate from our community for decades - cases in point are (for example) compilers and message passing interface libraries, but also separate communication libraries that underlie these obvious libraries, drivers accessing network interfaces etc. None of these software stacks are inherent to the electronic structure community, yet they create an increasingly complex ecosystem and points of failure for our users. Interfacing with this ecosystem was partly addressed by the workshop, but broader and continuous interactions between the various developer communities are needed.

Another need of the community (as always) is, simply, continuous training and updating of scientists' knowledge and practice in modern software development techniques, as well as recognition of such efforts, e.g. vast majority of tools and techniques used nowadays for code development did not exist five to ten years ago. Recognition of excellent scientific software developments is one important motivation to become involved in the first place, especially at the outset of a scientific career. Efforts that promote and recognize important, well executed and sustainable software developments, which ultimately enable better science, should be a continuous priority of our community.

Finally - perhaps obviously, but not always a strong point of a community focused on scientific computer programming: It is essential to create an environment that is welcoming and open to all. We need to foster a broad and diverse range of individuals, including at the beginning of their careers, in order to keep our community vibrant, productive and realize the myriad scientific opportunities which are now possible thanks to the computational equipment at our fingertips.

4 Funding

Funding of scientific software developments is historically difficult since it is often viewed as distinct from the scientific outcomes enabled by that software. This problem has been recognized by funding agencies in Europe and in the U.S., with specific calls and initiatives targeting software development. In Europe, large centers (MaX, NOMAD) now exist; the U.K. has had a continuous program focused on software development (e.g., ARCHER2 and its Exascale Initiative), the U.S. Department of Energy has large initiatives and ESL-related activities have also been funded through the US National Science Foundation. All of these funding mechanisms remain intensely competitive, however, creating new divisions through a perceived need to protect funding resources (e.g., competing teams may not be in a position to adjust goals and streamline efforts). A key role played by the ESL is that it bridges these different groups, national initiatives and distinct projects, creating open communication channels towards future proposals and funded projects.

5 Will these developments bring societal benefits?

The potential societal benefits enabled by electronic structure techniques are enormous. Chemistry, pharmacology, materials science and products that build on them ultimately rely on being able to quickly understand and harness benefits from as-yet unrealized functionalities of matter. Electronic structure codes are widely used in academia and industry for materials research, and in general for development and innovation, from drug design to materials for green energy. Having robust, well tested and maintainable software will not only increase the confidence in the results obtained from those codes, but also help faster development of new features. The latter will especially be facilitated by modular codes, which make use of libraries to encapsulate the low-level numerical tasks from the high-level physics or chemistry question, the codes aim to address.

The ongoing and planned activities of the ESL initiative will help in the point, outlined above, and furthermore, will prepare students and young career researchers better for software-development related positions both in academia as well as in industry. ESL has a long tradition of engaging with small and medium size businesses, QuantumATK and Simune, to name just a few, and we will continue this trend.

6 Participant list

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Machine Learning Meets Statistical Mechanics: Success and Future Challenges in Biosimulations
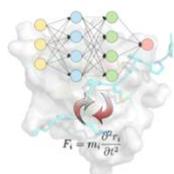
Location: CECAM-IT-SIMUL, Grand Hotel Vesuvio, Sorrento
Webpage : https://www.cecam.org/workshop-details/1153
Dates: Oct 12, 2022 - Oct 14, 2022

1 State of the art

Since the first applications in the 1970s, molecular dynamics (MD) has progressively become a valuable tool for the investigation of complex biological phenomena such as protein folding and conformational changes, ligand association and dissociation, and many other biologically relevant events. Nevertheless, standard MD calculations yet hardly reach the timescale of an entire biological process (millisecond-to-second range). Such a limitation can be overcome by employing coarse-grained modelling and enhanced sampling methods, or a combination of both. However, the success of enhanced sampling methods like umbrella sampling and metadynamics, depends on the choice of the system's reaction coordinates, namely collective variables (CVs), that are used to accelerate the sampling and that should depict the slowest degrees of freedom, finally providing an accurate description of the thermodynamic and kinetic properties of the system. Unfortunately, selecting the right CVs remains a challenging task, which generally requires both expertise of the investigator and time-consuming trial-and-error procedures. In this scenario, the rapid growth of molecular simulations data has highlighted the need of having a simplified representation of the available data manifold, leading to an increased interest in algorithms capable of organizing and analysing such data. To this end, a number of machine learning (ML) methods have been developed to manage simulations data with different scopes: i.e. define CVs, solve dimensionality reduction problems, deploy advanced clustering schemes, build thermodynamic and kinetic models.

2 Major outcomes

The present workshop successfully established a brainstorming on the main implementation of ML algorithms in the field of biosimulations, with a critical evaluation of their benefits, current limitations and future perspectives.
Fruitful discussions were held on the following main topics:

- **Switching from atomistic data to ML-readable representations**

Many different strategies have been presented to convert an atomic configuration A (coming from biosimulations) into a representation B, suitable to ML algorithms. This preliminary step

is fundamental in any different application of ML. Indeed, the reliability and the consistency of the ML predictions is strictly correlated to the capability of the input representation *B* of retaining the most important physico-chemical properties of the investigated system.

- **ML-trained interatomic potentials**

The application of ML methodologies to accelerate or improve the performances of classical MD simulations is emerging as a possible solution to the timescale problem. Indeed, different kinds of neural network are being trained by simulation data to compute interatomic potentials more efficiently than the molecular mechanics-based force fields or quantum-mechanical treatments.

- **Dimensionality Reduction and Collective Variables Identification**

As previously mentioned, a limiting step in the applications of enhanced sampling methods like metadynamics or umbrella sampling is the identification of the proper reaction coordinates (collective variables). The solving of this dimensionality reduction problem trough ML is thus drawing the attention of a large group of scientists. Particularly, various kinds of deep neural networks have been developed to analyze the huge amount of data produced by vanguard MD simulations to identify the most suitable physical descriptors of the phenomenon under study. Then, combinations (usually non-linear) of these variables can be used as reaction coordinates in new, more-accurate, enhanced sampling runs.

3 Community needs

Community needs were thoroughly examined in the discussion sessions. All the participants highlighted the necessity of a more active and efficient crosstalk between the scientists belonging to the Machine Learning and Biosimulations communities. A fruitful combination of these two disciplines requires indeed a continuous knowledge exchange among researchers with different backgrounds, spanning from mathematics, informatics, physics, biology, and theoretical chemistry. In addition, the need of a repository of simulation trajectories and input files was highlighted. Such a repository could be useful to provide training data for the development of new algorithms and methodologies. Moreover, to improve the large-scale applications of the newly developed algorithms, it was also suggested to:

1. Make public both the codes and the input data used to obtain the published results
2. Provide detailed documentation relative to the method and all the tools needed to perform the research

Many participants mentioned their wish for events similar to this CECAM meeting in the future. It was apparent that the mixture of scientists with different theoretical backgrounds and of various career stages was very appreciated.

4 Funding

This event promoted several discussions about the importance of collaborations between different computational and experimental groups. It was noted by many that meetings like this one represents a great chance to establish networks and new collaborations. As such, the discussion readily moved about the identification of proper funding channels. The field would benefit from ad-hoc grants designed to bring together more computational groups with different skills, which are presently limited in number.

5 Will these developments bring societal benefits?

During the workshop the social benefits that can arise from the development of Machine Learning applied to Biosimulations were highlighted. The latters already represent a valuable tool for the investigation of complex biological phenomena such as protein folding and conformational changes, ligand association and dissociation mechanisms. The social benefits span from a better understanding of biomolecules behavior to the knowledge of the main mechanisms ruling binding-unbinding of ligands, which contribute to the design and development of new and more efficient drugs. The biosimulations coupling with ML can importantly enhance the gained social impacts since in this way the current limits in terms of reached timescales and system's dimensionality boundaries can be overcome. Moreover, the development of new ML algorithms devoted to a specific research project could be adapted for different research fields, as often happens in the scientific research.

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9th Workshop on Time-Dependent Density-Functional Theory: Prospects and Applications

Location: Benasque Center for Science, Spain
Webpage: https://www.cecam.org/workshop-details/1157
Dates: Oct 25, 2022 - Oct 27, 2022

1 State of the art

Time-dependent density-functional theory (TDDFT) is one of the many methods that can be used to understand the excited electronic structure of atoms, molecules, or materials. It can be viewed as an extension of density functional theory (DFT) to time-dependent problems. The basic rationale behind DFT is the reformulation of the many-electron problem (roughly speaking, an equation with 3N variables, where N is the number of electrons) as a problem whose basic variable is the one-particle density, an object depending on the three spatial variables. Alternatively, we can view DFT as a manner to tackle with the interacting many-electron problem by studying a much easier non-interacting one. TDDFT is based on the same reduction of complexity. Usually this is obtained using an auxiliary system of non-interacting electrons that feel an effective time dependent potential, the time-dependent Kohn-Sham potential. Its exact form is, however, unknown, and has to be approximated.

TDDFT is by no means the only approach to the excitations of many-electron systems. In fact, more accurate (yet more expensive) techniques (based on many-body perturbation theory, for example) exist, and therefore these alternatives have also been covered at the workshop. However, TDDFT achieves a good balance between accuracy and computational cost. In consequence, its use is increasing. Around 300 publications every year contain results computed with TDDFT, and this number is increasing. This interest has been motivated by the recent developments of TDDFT and include the description of photo-absorption cross section of molecules and nanostructures, electron-ion dynamics in the excited state triggered by laser fields, van der Waals interactions, development of new functionals coping with memory and non-locality effects, applications to biological systems (chromophores), transport phenomena, optical spectra of solids and low- dimensional structures (as nanotubes, polymers, surfaces...).

Key References

[1] Time-Dependent Density Functional Theory, (Springer Berlin Heidelberg, Berlin, Heidelberg, 2006)
[2] Fundamentals of Time-Dependent Density Functional Theory, (Springer Berlin Heidelberg, Berlin, Heidelberg, 2012)
[3] C. Ullrich, Time-Dependent Density-Functional Theory, 2011
[4] N. Maitra, The Journal of Chemical Physics, **144**, 220901 (2016)

2 Major outcomes

This has been the ninth edition of a combined event (School + Workshop) that regularly reviews the fundamentals, progress, and applications of time-dependent density-functional theory, and related electronic structure techniques. The event has included a workshop, but also a previous school, at which early-stage researchers are exposed to the fundamentals of the theory, and practice hand-on exercises.

The school presented (1) an introduction to density-functional methods for absolute beginners; (2) an introduction to many-body theories in general and many-body perturbation theory in particular; (3) advanced topics in TDDFT (such as the problem of non-adiabaticity, etc.), (4) a discussion on the applications of TDDFT, the validity of the various approximations to the exchange and correlation kernel, etc.; (5) other topics such as quantum control, the microscopic-macroscopic connection, etc. In addition, the afternoon sessions were dedicated to hands-on exercises. The octopus and BerkeleyGW codes were chosen as the tools to show the students the capabilities of the theory. The calculation of spectra of molecules and solids were the main exercises, both with TDDFT and with GW and the Bethe-Salpeter equation. Almost 40 students participated in these sessions. Furthermore, they could present a poster displaying their research at two poster sessions: one during the school, and one during the workshop. As we usually do in this event, we asked all the school instructors to vote on the student posters, and elected the four best, who were offered the opportunity to present their research during the workshop with a short talk.

The workshop was made up of 27 talks (counting the 4 student presentations), and one poster session. The presentation slots were 40 minutes long, with ample time for discussion. There was a big variety regarding the topics: new theoretical developments (e.g. fundaments of the theory on lattices, the merger of TDDFT and electrodynamics, conditional probabilty density-functional theory…), applications in the high-field regime (the description of attosecond experiments, extreme optics in solids with the use of a combined Maxwell-TDDT setup…), development of functionals (e.g. range separated hybrids, Koopmans functionals…), and a number of description of applications to biology, 2D materials, transport, etc.

The program will always be available at:

https://www.benasque.org/2022tddft/cgi-bin/talks/allprint.pl

There, the school presentations are also available for download.

3 Community needs

Many codes have implemented TDDFT, as we could learn from the presentations of the workshop, in which we could see results obtained with many different programs. The number of researchers that work on methodological software advances for TDDFT seems to be growing. Some of the results displayed during the presentations highlighted the good scalability of these programs. TDDFT is praised for its applicability to larger systems, but even in this case there is clear need to access high performance computing resources. This is not always the case for many researchers.

This event is organized regularly, and has always been funded by the CECAM-ES node. We believe that it is necessary to keep doing so, as there are not so many TDDFT-focused events. Fortunately, there are events about electronic structure and electronic excitations in general (and all the related spectroscopies). Although ours was a theoretical / computational - only

event, there have been many successful stories of collaboration with experimentalists based on TDDFT, and the networking in that respect has worked well in these last decades.

4 Funding

Unfortunately, we must repeat almost verbatim a main concern that was also a consensual comment of many researchers after the 2018 workshop: the difficulty of accessing funding through many of the usual channels, due to the basic-research nature of the discipline, and to the orientation of many of those channels to more applied science. Basic research in DFT or TDDFT is performed under the umbrella of large projects with a more technologically oriented topic, in which the basic science aspect is regarded as a necessary but marginal sub-project. The situation has not improved since 2018, and therefore we must repeat the same concern here.

No particular funding proposal was discussed during the workshop.

5 Will these developments bring societal benefits?

TDDFT has demonstrated its predictive power in a certain applicability domain. Software solutions have been developed over the years, and can be found both at the academic and industrial level. Therefore, its use benefits the industrial research on materials science and photo-chemistry, for example. Modelling based on TDDFT and related techniques may help industry, by easing the interpretation of the observations made in their research & development processes. There are numerous technological processes and areas in which the excited-state electronic-structure of atoms, molecules, and materials (the object of study of TDDFT) is relevant. For example, during this workshop, we could witness reports on the application of TDDFT to model the proton stopping power of materials – an area of interest for the study of radiation damage, and for the construction of containers of nuclear reactors. Also, the application of TDDFT to the excited state chemistry of molecules of biological relevance was discussed both during the workshop and during the previous school.

6 Participant list

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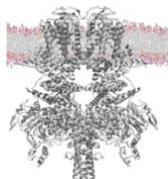Martin-Luther University Halle Wittenberg, Germany
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Ions, membrane and channels: Multiscale simulations from quantum to coarse-grain. A symposium in honor of Mike Klein.

Location: Rome, Italy
Webpage: https://www.cecam.org/workshop-details/159
Dates: Oct 27, 2022 - Oct 29, 2022

1 State of the art

Membrane proteins such as ion channels and molecular transporters play a crucial role in the biological function of cells, such as molecular signaling, detection, and transport. They are the main focus of many drug target and therapeutic approaches, as understanding their function is pivotal to the development and design of new technologies that rely on mimicking biological processes. This is particularly interesting in nanobiotechnology, and suitable to move towards more sustainable technologies in future. During the workshop, collective phenomena in membranes, such as cooperative ion channels, mechanisms of metal transporter, the quantification of energy landscapes of bio-membrane curvatures, as well as as specific proteins and systems of interest were singled out and discussed about experts in the field of bio-molecular simulations. With remarkable progress in protein structure determination of the past decade and the increasing speed and availability of supercomputers and new hardware technologies in combination to algorithmic developments, more and more detailed insight can be gained on the functions and mechanisms of membrane protein and ion channels.

2 Major outcomes

The scientific discussions during our CECAM workshop focused on the different computational approaches, i.e. the theoretical and computational developments, as well as the applications of this challenging research area that was strongly influenced and shaped by Mike Klein throughout his scientific career. The applied computational methodologies were discussed in depth during the workshop and ranged from quantum mechanics, enhanced molecular dynamics sampling techniques and coarse-grained molecular dynamics to machine learning approaches. While quantum chemical techniques allow to obtain insights into the electronic structure and properties of ions, and is fundamental to the development of polarizable force fields and metal parametrization, classical molecular dynamics techniques using enhanced sampling or coarse-grained models are necessary to study the dynamical behavior of membrane proteins at different time scales. To understand the impact and potential of membrane proteins for technological applications, different global and local aspects need to be addressed from the experimental as well as theoretical side. Therefore, the invited speakers presented different approaches to elucidate the biological functions of membrane proteins. Selected examples comprise the study of the hERG potassium channel and the KDEL receptor, the modulation of a mitochondrial proton carrier and the CLC transporter. Phenomena such as the hydrophobic effect, permeation, selectivity and gating of ion channels, and the formation of pores in membranes and lipid assemblies were discussed. Hereby, a critical review of the computational techniques employed for these challenging tasks was undertaken throughout the Q&A and discussion sessions. while perspective ones by borrowing ideas from other fields. The highlights during the discussion were also underlined

by the experimental counterparts from experimental collaborations which developed equally rapidly in recent years. At the same time, current limitations in the field concern the limitations of National and International research funding directed towards basic research and computational development programs, in place of highly specific and application-targeted funding programs.

3 Community needs

The workshop clearly showed very promising and intriguing possibilities for the field in future. The rapid development in protein structure determination and new opportunities from the field of big data with the emerging power of machine learning tools allows for unprecedented possibilities that, however, will also yield to new challenges and needs for our computational community. One of the main needs will remain the shareability and accessibility of data throughout the community. One ultimate goal is to optimize the reuse of data to ensure efficient collaboration and transfer of knowledge throughout the scientific communities. In the future, we should aim to host and organize more interdisciplinary workshops that join life-sciences with other communities such as computer science and engineering sciences. Additional needs of the community identified during the meeting show the shared view about the necessity to receive more direct support from High Performance Computing centers at National and EU level, especially for helping researchers in computational development activities towards more effective methods and models.

4 Funding

The meeting nurtured very active discussions between researchers from 11 different countries including the US, inviting future collaborations between the participants. During the exchange and networking (guaranteed through the organized lunches, coffee breaks and the social event), several opportunities to apply for EU funding have been also considered, among which some calls within the Human Brain Initiative and H2020 Era-net programs. We believe that follow-up workshops, seminars and scientific collaborations, as well as grant applications and student exchanges, are going to be facilitated through the newly established and enforced cooperation network originated from the meeting.

5 Will these developments bring societal benefits?

The workshop presented and stimulated collaboration in ion channel pharmacology, of great economic and societal benefit (ion channels are very important pharmaceutical targets). Opportunities for funding this very important research field were also discussed. Generally speaking, the biological targets discussed during the workshop are found in all living organisms and play critical roles in a wide range of physiological processes, including muscle contraction, neural signaling, and hormone secretion. Therefore, their may lead to the development of new therapies and treatments for a range of diseases and disorders. The organization of the workshop allowed to facilitate collaborations and cross-talking among scientists from different fields and institutions, which will ultimately lead to more efficient and effective research, accelerating scientific discoveries and advancements in the field. This may further have broader implications for human health and well-being as an increased understanding of ion channels and membranes can also contribute to the development of innovative technologies such as biosensors, e.g. to detect changes in ion channel activity and aid in the diagnosis and treatment of diseases. Overall, the workshop can have far-reaching

societal benefits, advancing both scientific knowledge and practical applications that can improve human health and quality of life.

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Data Modeling and Computation: Capturing Biomolecular Processes

Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1122
Dates: Oct 31, 2022 - Nov 4, 2022

1 State of the art

Understanding the molecular basis of life requires synergy of molecular simulations, experiments, data analysis, and theory. There is however a significant disconnect among these disciplines, with, e. g., oversimplified phenomenological theories commonly employed to analyze both experimental and simulation data. This workshop is motivated by recent theoretical, computational, and experimental advances that offer novel opportunities for connecting these disciplines. These advances include achieving overlap of experimental and simulation timescales, development of data driven computational approaches to single molecule signals, advances in machine learning and its applications to molecular data, and breakthroughs in fundamental theory of molecular kinetics and stochastic dynamics. Tools from Data Science including Neural Nets, Bayesian and likelihood-based methods, and even Information Theory collectively offer new approaches to tackling longstanding challenges present in the world of biomolecular dynamics. This workshop promoted synergy between those who simulate the dynamics of molecules and those who infer their behaviors from raw data, thereby facilitating interaction between computational scientists and experimentalists who can capture dynamics at state-of-the-art spatiotemporal scales. While temporal scales available to molecular simulations remain severely limited from above and both spatial and temporal resolution of single-molecule data remains severely limited from below, this workshop was specifically intended to bridge this gap and leverage novel tools to unravel life at the length scale (single molecule) and across time scales at which it occurs.

2 Major outcomes

The specific open questions/focus themes explored by the workshop were:
- Can thermodynamic and kinetic properties of a molecular system be reliably estimated from molecular trajectories (simulated or experimental) given sparse sampling inherent to computational limitations and experimental constraints (e.g., because a molecular motor detaches from its track or a dye photobleached)? Moreover, can accurate low-dimensional models capturing the stochastic evolution of single-molecule signals be inferred from such sparse signals? Such models will likely need to go beyond currently used phenomenological models (such as, e.g., one-dimensional diffusion along a reaction coordinate) to capture the complex assembly, say, of many biomolecular actors.
- What information regarding underlying high-dimensional molecular processes is encoded in the observed low-dimensional signals (e.g., often just single photon arrivals)?
- What are optimal ways of combining data from molecular simulations and experiments?
- How can we combine the relative strengths and weaknesses of neural net approaches and, more traditional, computational statistics approaches to unravel biomolecular events down to microsecond timescales?

To explore these themes, this 5-day workshop was structured into focused 0.5-1-day sessions, each exploring one or more of the following overlapping topics: Optics, Photonics and Imaging with application to single-molecule and cellular phenomena, Optimal methods for combining molecular simulations with experimental data and advanced computational techniques, Polymer dynamics and networks, Proteins and allostery, Biochemical dynamics and kinetics, and Single-molecule spectroscopy. Each of the 34 workshop participants gave a talk, followed by a discussion, in one of the sessions.

The talks given by the workshop participants reflected impressive advances achieved recently in addressing the questions raised above. Specifically, we have learned about (1) new machine learning methods for single-cell Raman imaging and for free energy reconstruction from force spectroscopies, (2) Bayesian inference methods for ultrafast X-ray scattering, cryo-electron spectroscopy, and optical imaging/photonics, (3) new methods for estimating path probabilities and conditional first passage time distributions from force-spectroscopy, (4) detection of hidden states and time-directionality from single-particle/molecule signals and (5) coarse-grained models and first-principle prediction of liquid-liquid phase separation in proteins. Workshop participants reported that these ideas and techniques have resulted in important biological insights on allostery, protein folding, kinetics of biochemical reactions, protein-DNA interactions, synaptic transmission, and function of antimicrobial peptides.

During the many discussions that took place during the workshop, a number of open issues have been identified. In particular,

- What are the fundamental and practical limitations of data inference in application to single-molecule data, particularly single-photon data? Given low-dimensional nature of single-molecule signals, when is it possible to differentiate between kinetic models of different dimensionality or between time-reversible and irreversible phenomena?
- Given many competing methods of single-molecule data analysis (various hidden Markov-type models, Baeysian nonparametrics, traditional apprtoaches relying on data-fitting etc.), how to evaluate their relative performance?
- What can machine learning talk about underlying energy landscapes and about dynamics?

3 Community needs

While some the more general data analysis tools reported in the workshop have mostly been tested in application to synthetic data, many experimental groups use in-house analysis methods. Often similar experiments (e.g. force spectroscopy of protein folding) are being analyzed using different methods, leading to different conclusions. There is a need for a broader survey of existing data analysis methods and their critical evaluation in application to experiments. Experimentalists participanting in the workshop will make large quantities of data (e.g. single-photon arrival times or bead trajectories in optical tweezers experiments) available with the purpose of comparing various methods. At the same time, in order to explore the fundamental and practical limitations of data inference in application to single-molecule data, workshop participants volunteered to generate synthetic data where the ground truth (i.e. the underlying dynamics) is known and where the challenge is to uncover the underlying dynamics. Such "challenges" are becoming increasingly popular and often lead to joint publications involving multiple groups.

We anticipate that evaluation and discussion of the outcomes of pursuing these challenges to the community will warrant a future CECAM workshop on this topic.

4 Funding

Data analysis tools discussed in this workshop often require significant computation. As such they will benefit from several existing and future funding opportunities for high performance computing resources. Examples include the new petascale computing system, Frontera, which is supported by the US National Science Foundation (NSF), and which is available through the NSF Petascale Computing Resource Allocation program [Dan Stanzione, John West, R. Todd Evans, Tommy Minyard, Omar Ghattas, and Dhabaleswar K. Panda. 2020. Frontera: The Evolution of Leadership Computing at the National Science Foundation. In Practice and Experience in Advanced Research Computing (PEARC '20), July 26–30, 2020, Portland, OR, USA. ACM, New York, NY, USA]

5 Will these developments bring societal benefits?

The societal benefits of this workshop include the following:

1. Understanding of the biological function of molecular machines and of the molecules of life are crucial to developing better drugs and therapies. Some of the workshop participants are pursuing applied research of immediate biomedical significance, such cell imaging, interaction between different drugs, protein aggregation, and viral replication mechanisms. Others focus on more fundamental aspects of allostery, protein folding, biochemical kinetics, and chemical dynamics, which, likewise, may ultimately enable better healthcare tools.
2. Data science is essential to all aspects of everyday life, and statistical data analysis tools developed specifically for biochemical phenomena may have significance outside the immediate field.
3. Given its highly interdisciplinary nature, this workshop contributes to development of educational resources, as most of the workshop participants are either educators (e.g. University faculty) or postdocs/graduate students. Likewise, given the highly international composition of the workshop, it is bringing the European and American molecular science circles closer together.

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Development of coarse-grained models
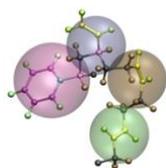
Location : CECAM-FR-RA École Normale Supérieure de Lyon, France
Webpage : https://www.cecam.org/workshop-details/1041
Dates: Nov 14, 2022 - Nov 16, 2022

1 State of the art

The purpose of the workshop was to bring together active researchers in the field of the design of coarse-grained (CG) models for applications ranging from biology to materials science and engineering. The development of realistic CG models, designed to account for the chemical nature of the grain can be achieved through top-down and bottom-up approaches but also by machine learning-based approaches. All these approaches have been discussed and presented through different applications. Fundamental aspects have been addressed: what are the strengths and weaknesses of these different approaches? How can we improve the CG models to account for deformable grain shapes, to consider the transferability of these models over a larger range of temperatures and pressures and to use a larger degree of coarse-gaining. In terms of applications, CG models have been presented in the framework of polymeric materials (mechanical properties, entanglements, long-time process) and biological systems (proteins, membranes, long-time dynamics).

2 Major outcomes

Different approaches were presented for building coarse grain models and their merits were discussed. The top-down approach allows for good reproduction of particular experimental data, by tuning the model definition and parameters. The bottom-up approach require more modeling assumptions and approximations. It allows to make prediction for new systems, but the predictions are not always accurate. Several bottom-up strategies were discussed and

compared (Iterative Boltzmann Inversion, Inverse Monte Carlo, Statistical Trajectory Matching, Machine Learning, Relative Entropy, Force Matching...).

Machine learning can be used, which require some control to guide the learning toward a physically acceptable model. The future of machine learning was debated in the discussions. It is already quite powerful. It will probably be even more and more accurate and faster, but the user would usually gain no physical insight, because it works as a black box.

Hierarchical coarse-gaining was also discussed, as well as reverse mapping to fine grains, which may rise technical difficulties.

The type of models was also discussed. In particular, the occasional necessity to build anisotropic models, and ways to do so. The grains can be modeled as ellipsoids with fixed shape or the shape may evolve in time. Specific equations for the evolution of coarse degrees of freedom, derived from first principles, were presented with simplifying assumptions to make them more usable. Many-body interactions can be taken into account by cluster expansion (3-body, 4-body...) or by local density potentials. The development of models where one grain represents several small molecules was introduced and discussed. The very definition of the grain in this case was discussed. The electrostatic interactions can be taken into account at the coarse-grain level. The appropriateness of taking into account or not time correlations in the random and dissipative interactions was discussed, in relation with the separation of time scales between coarse and fine degrees of freedom. So discussions focused as well on structure as on dynamics. How can current strategies be improved to account for dynamics in addition to structure?

Specific applications of the coarse-grain models were presented, and the corresponding coarse grain model design was motivated. In particular, applications were presented in polymers, biological molecules (DNA, proteins), or simple liquids (water, ions in solution). In each case, the coarse grain model has to retain system-dependent physically relevant detail (entanglements, rigidity, charges, shape...).

3 Community needs

The use of coarse-grained models currently requires many theoretical developments on all approaches (bottom-up, top-down, machine learning). Few codes are available on these aspects and the number of competing methods is important, so it is difficult to standardize the approach. The teams develop by themselves the codes which are still confined to the laboratories because of the difficulty to be operational and transferable to other systems. It is however useful to consider structuring the community around the "coarse-grained model design". This type of research activity is well suited to CECAM workshops due to the size of the community, which is not so important around the world. We can indeed think of extending the initiative with the programming of other workshops in the future to see the evolution of theoretical practices and the diversity of applications on which they were tested. Indeed, there are many progresses in the field of CG models and the challenges raised by the industry and academic community is strong in this field. It would then be interesting to plan another workshop within two years.

4 Funding

The possibility of connecting different teams in response to national and EU calls for proposals was not specifically discussed. Nevertheless, the organisers, through the SimatLab Joint Research Laboratory, are planning to submit a European project on polymer materials and could call on teams who participated in the workshop. Collaborations could take place between groups in the field of artificial intelligence or machine learning to develop coarse grain models with a specific chemistry, in the domain of theoretical physics to design models with a high level of coarse-graining to access large time and length scales.

5 Will these developments bring societal benefits?

It is clear that the contribution of molecular simulation is undeniable for the industry. Industry will be able to reduce its research costs by reducing the number of experiments, but it will also be able to innovate the design of materials by imagining the materials of tomorrow from molecular interactions. The difficulty lies in the ability of molecular simulation methods to model a representative sample of the experiment. The multi-scale approach and the connection between atomistic and mesoscopic scales is essential in this objective. The applications obviously concern the medical domain where expectations are numerous (membrane, cells, DNA) but also the field of material to design sustainable materials, for example. This is why we have focused our workshop on both biological and material applications. However, considerable theoretical efforts must be made in relation to more efficient means of calculation (GPU, exascale) to achieve the objectives of industrial interest systems and the support for organizing such workshops between specialists in the field is a significant help.

The question of the environmental impact of research on coarse-graining was discussed. On the one hand, developing coarse grain models requires heavy computations and storage. But on the other hand, having available accurate coarse-grain models allows to run much faster simulations and to save computational resources.

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Green's function methods: The next generation 5

Location: CECAM-FR-GSO
Webpage: https://www.cecam.org/workshop-details/1138
Dates: Nov 15, 2022 - Nov 18, 2022

1 State of the art

Green's functions have always played a prominent role in many-body physics. In particular, the one-body Green's function (GF) delivers a wealth of information about a physical system, such as ground-state energy, excitation energies, densities and other measurable quantities. Therefore, the development of approximate methods to calculate the one-body GF has been an active research topic in many-body physics since the 60's, and many routes have been explored in order to find increasingly accurate GFs. A prevalent class of methods is based on the iterative solution of an integral equation for the GF containing an effective potential, the so-called self-energy, which needs to be approximated. The well-known GW approximation belongs to this class; this approximation is the method of choice for calculating band structures, but it also shows several shortcomings, such as the wrong description of satellites in photo-emission spectra, particularly in so-called strongly-correlated materials. Therefore, more refined levels of approximations are needed to keep pace with the advances made in experiments. Recently much progress has been made in this direction both by going beyond standard methods and also exploring completely novel routes to calculate GF. A new wave of original ideas, understanding, and solutions, has pervaded the field and therefore we think it was timely to gather these new concepts in a workshop.

2 Major outcomes

It is clear that state-of-the-art approximations are not sufficient anymore to describe new phenomena and emerging new physics that the advances in experimental techniques allow us to explore. The so-called strongly correlated materials, for example, exhibit remarkable electronic and magnetic properties, such as metal-insulator transitions, half-metallicity, or unconventional superconductivity, which make them among the most attractive and versatile materials. Describing these systems requires going beyond today's state-of-the-art approaches and represents one of the greatest challenges for condensed-matter theory. New strategies start to emerge which revisit fundamental equations and reformulate them in such a way as to make clearer the physics they describe. New physical constraints are searched in order to obtain physical approximations. It is also clear that collaboration with other fields is essential since similar problems are common to various domains and solutions might already exist. According to this, the workshop has shown three big axes of development, promising to tackle the new challenges in material science:

1) Many novel approaches have been proposed to describe physical observables from Green's functions such as correlation satellites in optical spectra, exciton band structures, inelastic x-ray scattering, and photoemission from the three-body Green's function

2) Many efforts have been demonstrated to combine many-body perturbation theory with other theories such as dynamical mean-field theory and parquet theory and to include new physics such as electron-phonon coupling and non-collinearity. The "ab initio community" can learn many things from the "DMFT community" and vice versa.

3) The development of vertex corrections both in the self-energy and in the screening and determining for which observables and/or materials vertex corrections are important. Also, the impact of self-consistency on these calculations has been addressed.

3 Community needs

Many of the concepts here reported will benefit from an increased support of fundamental research. This is today particularly important in a moment in which public research is funded via contracts and projects. In many countries, fundamental research suffers with respect to applied or industrial research. Very often national calls give high priority to joint research+industry applications. To this, we can add the perverse ramification of calls (European, national, regional, local) that, far from multiplying the offer, rather creates a frustrating scattered and highly inefficient funding procedure. For what concerns specifically software and hardware, we have both good and bad news. Many groups have already partnerships with local software engineers, and few others have decided to ask for support from the PRACE facilities. This is a crucial step: it is indeed true that the computer power available today (both in national and European calls) is good news to tackle bigger and more complicated systems. However, without proper writing (and often, re-writing) of our computer codes (and this is the bad news), to take full advantage of the modern paradigms (blue-gene low memory nodes, GPUs accelerators, mics, and all hybrid architectures), there will not be any possibility to tackle such complex materials and behaviours. The scalability required to take full advantage of this new kind of resource is in fact such (1-1000000 cores) that only a professional approach can succeed. This professional procedure comes not only in terms of parallelization of the codes, but also in their everyday maintenance and in all aspects of collaborative development (revision control, tickets, pull requests, forums, etc.), as previously mentioned.

Finally, to make these new codes and capabilities widely available, training strategies must be devised at both hard (summer dedicated schools) and soft (e-learning platforms, MOOCS) levels.

4 Funding

It is crucial for the development of the fundamental research involved in this workshop, that funding agencies continue or even increase their funding of fundamental research. The workshop brought together many established experts in the field of Green's function methods as well as young promising talented researchers. Today, several channels have been analyzed for the funding of fundamental research:

-ERC grants at the European level (starting, consolidator, advanced, and synergy) are the most important call in which fundamental theory can be funded.

- Marie-Curie calls (Individual fellowship, CoFUND, and ITN) are still active and available also for fundamental research (even if some calls, useful in the past, like the career integration, are now closed)

- National funding agencies are uneven (good for Germany, bad for Italy, worsening in France, etc.)

- local/regional calls are sometimes good sources of funding, but only for punctual needs.

5 Will these developments bring societal benefits?

Progress in Green's function methods is of great importance to fundamental research. However, it has already shown its crucial usefulness also for European Industry. Many-body Green's functions contain a wealth of information that can be used to develop novel materials for various innovative technological applications. For example, the single-particle Green's function contains information on the electron addition and removal energies, which allows for an accurate description of band gaps, band alignments, etc. This is critical information in the development of photovoltaic devices. The single-particle Green's function also contains information on the current density, which can be used to describe accurately all kinds of phenomena related to electron transport. From the two-particle Green's function, we can obtain information about the interaction between two particles such as the interaction of an excited electron with the hole it leaves behind. This is crucial information for developing new photo-voltaic and photo-catalytic devices, for which its success largely depends on the ability of the material to split the electron and the hole after their formation. However, this applies to the field globally. Concerning the more restricted focus of the workshop in particular, it is about fundamental research preparing the long-term future on a scale of, say, 10 years which unfortunately is not always of direct interest to a private company.

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Challenges and Advances in Solving Eigenproblems for Electronic-Structure Theory
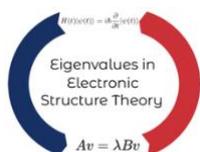
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/1128
Dates: Nov 15, 2022 - Nov 18, 2022

1 State of the art

Numerically solving (generalized) eigenvalue problems is one of the fundamental tasks in computational science with broad applications in quantum mechanics [1] and machine learning [2]. In particular, eigenvalue solvers play a fundamental role in electronic-structure theory at the density-functional theory (DFT) level, in which this step constitutes the computational bottleneck [3]. Besides critically limiting the time-to-solution, the cubic scaling of traditional eigenvalue solvers limits which physical systems can be addressed from first-principles codes with reasonable computational effort. To date, many electronic-structure software packages use highly optimized eigensolver libraries for the solution of eigenproblems, e.g., *EigenExa* [4] and *ELPA* [5,6]. These open-source libraries were developed over the last decade with the specific goal to accelerate and enable novel first-

principle calculations by leveraging synergetic collaborations between mathematicians, high-performance computing (HPC) experts, and electronic-structure code developers. With that, these libraries often serve as lighthouse examples of how software engineering and science should work hand-in-hand to enable more efficient numerical simulations.

Currently, new challenges have appeared on the horizon with the upcoming deployment of (pre-)exascale HPC systems featuring heterogeneous architectures with hundreds of cores per nodes as well as multi-purpose accelerators such as GPUs. Harnessing the computational power available on these resources requires significant efforts to port existing algorithms to novel architectures and to design new algorithms tailored to these architectures [7-9]. Tackling these challenges necessitates an even tighter collaboration between mathematicians, HPC experts, first-principles code developers and even hardware and software vendors.

Key References

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2 Major outcomes

Several important conclusions can be drawn from the presentations as well as from the discussion sessions. They are all related to the fact that the first-principles community needs **accurate**, **efficient**, and **scalable eigenvalue solver libraries**.

1) Upcoming HPC architectures offer enormous compute power via their GPUs and thus promise investigations of larger systems with higher accuracy than possible today. This requires the eigenvalue-solver libraries to efficiently exploit such multi-GPU configurations. All developers are already working on GPU-accelerated implementations and, for most eigensolver packages presented during the workshop, multi-node, multi-GPU benchmarks have already been shown. Still, this aspect involves quite challenging developments, also due to the heterogeneity of the GPU architectures.

2) So-called **mixed-precision approaches** have been identified as a potential route to save computational resources. Since DFT typically requires several self-consistency iterations (SCF) until convergence is achieved, the eigensolver can run with lower precision, for example single precision, during the first iterations and then switch to target precision, typically double precision, at a later stage. Several libraries are already developing and testing such a functionality as well as iterative refinement approaches for the eigenvalues and eigenvectors.

3) Standardized benchmarks for eigensolver libraries are needed: On the one hand, such benchmarks would allow electronic structure theory experts to better judge which eigenvalue solver is the best option under which circumstances. On the other hand, also the developers of eigenvalue solvers can profit from such **standardized benchmarks**, which would allow for an **objective** and **reproducible** comparison between different eigenvalue solvers.

Several participants of the workshop have agreed to collaborate for building such a standardized benchmark. In a first step, various library developers (from *ChASE*, *ELPA*, and *EigenExa*) will agree on a few representative setups that define matrix type (such as random or Toeplitz), data type (float, double, complex, or double complex), required fraction of the

eigenspectrum, and solution accuracy. Here, the problems will be chosen to give a fair overview over the abilities of the different types of eigenvalue solvers. In a second step, electronic structure theory experts will also propose benchmark setups from practical applications, which are representative for certain classes of physical systems treated in DFT simulations. Details of the benchmarks, such as detailed timings, the library version, compilation flags for the build, details of the computational architecture, and all other information necessary for reproducibility will be published in a common repository.

3 Community needs

The organized workshop and in particular the discussion sessions revealed that a tighter collaboration and better communication is needed and desired in this community. The challenges this field faces requires networking and close interaction between the DFT community, the eigensolver developers, and hardware vendors. For instance, the developers of different eigenvalue-solver packages are often facing similar challenges, e.g., when implementing support for different GPU architectures or when testing mixed-precision approaches. In turn, support from the hardware manufacturers can be extremely helpful for these tasks. Similarly, also the users of different eigenvalue-solver libraries are often not fully aware of all existing advancements in all libraries for all architectures, especially with respect to novel developments. To this end, the CECAM workshop was an important first step to bring the different communities together and to provide a platform for fruitful exchange between them. This is further substantiated by the fact that several developers and beneficiaries of eigenvalue-solver libraries agreed to work together to build a standardized benchmark set. Accordingly, we feel that a continuation of these workshops, for instance on a two- or three-year schedule, would be very beneficial both for the eigenvalue solver and the electronic-structure theory community.

4 Funding

The development of eigenvalue solver libraries is typically funded via the following ways:
- internal funding of research institutes, HPC centers, or companies (internal or in kind to open source)
- cross-institutional project funding via national research funding campaigns
- cross-institutional project funding via international research funding campaigns, such as the European Horizon 2020 initiative.

All eigenvalue solver packages represented at the workshop have been or are still funded via at least one of these approaches.

During the meeting a discussion has been started between the developers of the *ChASE* and *ELPA* library for a future joint proposal.

5 Will these developments bring societal benefits?

Electronic-structure theory is a vital ingredient in modern material science that guides and steers the research for novel materials with optimal properties. Accordingly, this line of research has a huge societal impact, since it affects and drives several important fields. This has become evident from several examples discussed during the workshop. For instance, a transition to a carbon-emission-free society requires more efficient storage of electric energy in re-chargeable batteries and more efficient hydrogen catalysis. To be successful, this kind of research needs to address realistic materials with system sizes (i.e., the number of

considered atoms) much larger than possible today. This will only be possible with exascale HPC resources. Since the solution of eigenvalue problems is the computationally dominant step in DFT calculations, the availability of efficient and scalable exascale eigenvalue solvers is of fundamental importance for the research topics mentioned above. Indeed, this has been recognized in several European and international projects, e.g., MaX and NOMAD Centers of Excellence as well as the Exascale Computing Project in the US.

6 Participant list

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Quantum dissipation by swift nuclei in condensed matter

Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage : https://www.cecam.org/workshop-details/1126
Dates: Dec 7, 2022 - Dec 9, 2022

1 State of the art

Swift nuclei moving through condensed matter and slowing down by electronic friction give rise to a paradigmatic problem of quantum dissipation at the nanoscale that is both strongly out-of-equilibrium and non-adiabatic. Apart from fundamental importance, the relevance of the problem to medicine and to the nuclear and aerospace industries gave rise to a century of intense research [1]. Understanding and modelling were, however, limited to Lindhard's linear response [1], valid for weak perturbations, and the homogeneous electron gas, valid for idealized metals [1,2]. Since 2007, progress accelerated via the first-principles simulation of such processes based on real-time time-dependent density-functional theory (RT-TDDFT) [3-6]. However, the mean-field approximation embodied in TDDFT imposes serious limitations to describe dissipation and equilibration. Going beyond requires either functionals with memory [7], many-body perturbation approaches [8], or electronic excitations [9]. Simultaneously, there has been progress in the theoretical underpinnings, from general results at low velocities [10] to a Floquet formalism for periodic solids [11]. Ehrenfest coupled dynamics also has limitations such as lack of thermalization. Recent developments in the coupled quantum dynamics of electrons and nuclei proved successful in other contexts [12-17], and can also allow for progress here. The added challenge is the electronic continuum, as opposed to the discrete potential energy surfaces normally used in non-adiabatic dynamics [18].

Finally, the great difficulty in carrying out experiments in this field has also progressed in recent times, e.g. in the measurement of energy loss [19,20], of optical response [21], and the interaction of ions with metallic surfaces [22]. This workshop aimed at identifying and exploiting new synergies theory-simulation-experiment, including the identification of most suitable target systems to confront jointly to advance the field.

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2 Major outcomes

The workshop brought together key people in the fields of electronic stopping of nuclei in matter, non-adiabatic quantum dynamics, and density-functional theory and many-electron dynamics, to face the problem of quantum dissipation of swift nuclei in matter, from quantum friction effects of ions/molecules on surfaces and nanoconfined flow, to strong dissipation under irradiation. Invited speakers were prompted to talk about their recent work and ideas in their own topics which they think could connect to the other subfields. The general ambition was cross-fertilisation, and exploring how connections of advances in one field might contribute to the others. In the spirit of traditional CECAM workshops, ample opportunity for discussion and lateral collective thinking was provided.

The format consisted of three full days, including seven talks and a discussion session per day, after the afternoon coffee break. Slots of 40 min were allocated per speaker, aiming at 20-25 min of lecture and 15-20 min of discussion. Most of the talks were delivered in-person, although three of them were delivered remotely due to the speakers being unable to travel to Lausanne. Two talks had to be cancelled due to illness or unforeseen complications. One of them, the last talk of the workshop, was not replaced. The other one, on the first day, was replaced with a talk by one of the organisers. The workshop was structured in three interconnected themes, one per day, starting with nuclei as projectiles (experiment, theory, simulation), followed by levels of theory for the dynamics of the electronic subsystem, to conclude with quantum coupled dynamics of electrons and nuclei, including connection to other non-adiabatic contexts. Each day had an associated discussion session led by one of the participants who identified important open questions to be addressed in the future, as arising from the presentations.

Key needs identified and actions proposed to address them can be summarised as follows:
1. Promoting further interactions between modelers and experimentalists and ensuring that experimentalists' input on relevant questions and coherence between models and experimental set up is clearly disseminated. To foster this goal, the organization of a follow-up workshop driven by experimentalists was proposed and will be pursued for 2024.
2. Clarifying and disseminating state-of-the-art and open questions via a shared publication in the form of a roadmap paper. In particular, this work should include a more important participation by the cognate community of non-adiabatic dynamics applied to chemical processes, which has developed a number of quite advanced tools, especially in the field of photochemistry.
3. Identify challenges and benchmark systems for currently existing techniques. In particular, an adequate description of electron thermalisation after a strong energy pulse was considered a timely and suitable challenge for the dynamical simulation techniques being used (such as TDDFT).

3 Community needs

The needs identified for this community were mostly at the networking and event organization level, not so much computational resources. The possibility of proposing a COST Network to foster collaboration via short-term scientific missions and thematic workshops was discussed and positively met by the participants. Work to define the structure and participants to the

project will be pursued in the near future. Furthermore, increased communication and exchange with other scientific communities technically related to this one but traditionally separate in particular, simulators of non-adiabatic phenomena coming from physical chemistry or theoretical chemistry needs to be fostered. This will be pursued via participation to relevant workshops and promoting new activities with organisers coming from different communities. The creation of a bespoke communication channel, or participation to similar tools being developed in related communities, is also a potential action to be pursued in the future.

4 Funding

Typical funding channels have so far been mainly confined to individual research work. A need to look into collaborative projects to foster further exchange has been identified. The main source of funding identified for these follow-up activities is the COST Action mentioned above. The possibility to explore alternative funding sources - in particular to enable series of workshops - will also be explored. More organic involvement in on-going projects in the EU and USA and working towards prompt sharing of funding information would be beneficial.
At this stage, consideration of specific funding channels, beyond the COST action indicated above, is premature. However, we expect that the greater involvement of experimentalists in future meetings will facilitate identifying opportunities beyond those related to purely academic collaborations.

5 Will these developments bring societal benefits?

Although the main focus of the workshop was on methods and experiments, the topic has potential for applications of societal interest in areas such as medicine, nuclear research, and aereospace industry. There are research communities in the three mentioned contexts, all of them using quite coarse phenomenological descriptions in some of the several time and length scales relevant to them. Pushing the fundamental knowledge and first-principles simulation techniques should put those phenomenological methods on much firmer and quantitative footing.

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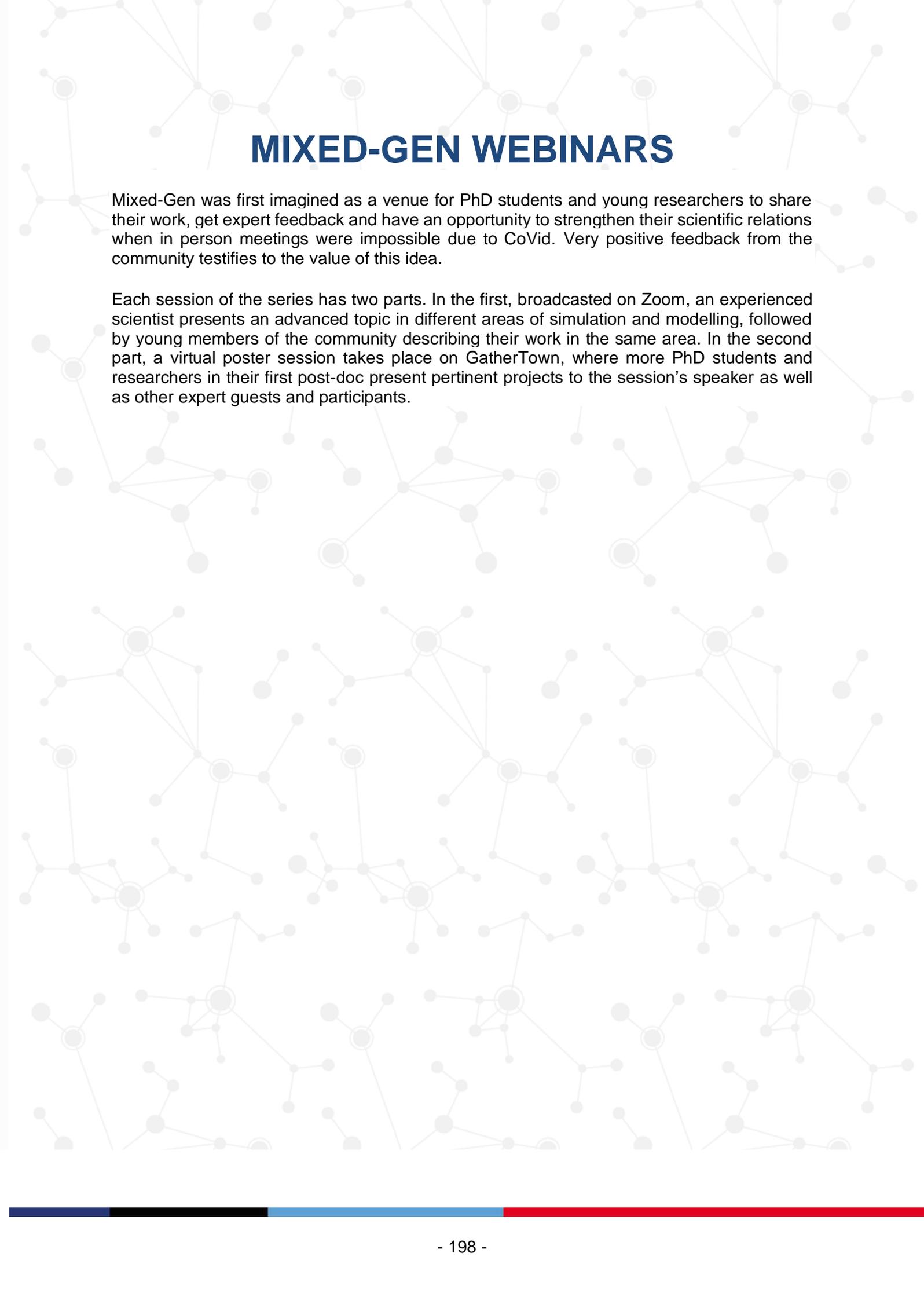
MIXED-GEN WEBINARS

Mixed-Gen was first imagined as a venue for PhD students and young researchers to share their work, get expert feedback and have an opportunity to strengthen their scientific relations when in person meetings were impossible due to CoVid. Very positive feedback from the community testifies to the value of this idea.

Each session of the series has two parts. In the first, broadcasted on Zoom, an experienced scientist presents an advanced topic in different areas of simulation and modelling, followed by young members of the community describing their work in the same area. In the second part, a virtual poster session takes place on GatherTown, where more PhD students and researchers in their first post-doc present pertinent projects to the session's speaker as well as other expert guests and participants.

Mixed-gen Season 2 – Session 5: Simulating glasses

February 24, 2022, 15:00 – 19:00 CET

Perspectives for the next generation of glass transition studies
Ludovic Berthier, University of Montpellier

Computer simulations give unique insights into the microscopic behavior of amorphous materials. It became recently possible to generate equilibrium configurations of deeply supercooled liquids using a simple swap Monte Carlo algorithm for a broad variety of model glass-formers. This novel computational tool, which provides an equilibration speedup of more than 10 orders of magnitude, has allowed the exploration of thermodynamic properties and putative equilibrium phase transitions underlying the formation of glasses with unprecedented accuracy. I will summaries these recent findings and provide a broad picture about the thermodynamic phase diagram describing the physics of liquids undergoing a glass transition.

From hard spheres to hard-core spins
Grace Sommers, Princeton University

A system of hard spheres exhibits physics that is controlled only by their density. This comes about because the interaction energy is either infinite or zero, so all allowed configurations have exactly the same energy. The low-density phase is liquid, while the high-density phase is crystalline, an example of "order by disorder" as it is driven purely by entropic considerations. Here we study a family of hard spin models, which we call hardcore spin models, where we replace the translational degrees of freedom of hard spheres with the orientational degrees of freedom of lattice spins. Their hardcore interaction serves analogously to divide configurations of the many spins system into allowed and disallowed sectors. We present detailed results on the square lattice in $d=2$ for a set of models with Z_N symmetry, which generalize Potts models, and their U (1) limits, for ferromagnetic and antiferromagnetic senses of the interaction, which we refer to as exclusion and inclusion models. As the exclusion/inclusion angles are varied, we find a Kosterlitz-Thouless phase transition between a disordered phase and an ordered phase with quasi-long-ranged order, which is the form order by disorder takes in these systems. These results follow from a set of height representations, an ergodic cluster algorithm, and transfer matrix calculations.

Many Body Correlations Are Non negligible in Simple Glassformers
Chengjie Luo, Eindhoven University of Technology

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Mixed-gen Season 2 – Session 6 Simulating biological systems

March 31, 2022, 15:00 – 19:00 CET

Using Computer Simulations to Advance our Understanding of Biological Systems at the Atomic Level

Benoit Roux, The University of Chicago

Classical molecular dynamics simulations based on atomic models play an increasingly important role in a wide range of applications in physics, biology and chemistry. The approach consists of constructing detailed atomic models of the macromolecular system and, having described the microscopic forces with a potential function, using Newton's classical equation, F=MA, to literally "simulate" the dynamical motions of all the atoms as a function of time. The calculated trajectory, though an approximation to the real world, provides detailed information about the time course of the atomic motions, which is impossible to access experimentally. Specialized free energy simulations are also an important route to establish a strong connection to experiments. The development of efficient methods for simulating slow conformational transitions is another subject of great interest in computational studies of biomolecular system. A powerful paradigm for mapping the conformational landscape of biomolecular systems is to combine free energy methods, transition pathway techniques and stochastic Markov State Model based massively distributed simulations. These concepts will be illustrated with a few recent computational studies of Src tyrosine kinases, K+ channels, and the P-type ion pumps.

Asparagine tautomerization in glycosyltransferase catalysis. The molecular mechanism of protein O-fucosyltransferase 1

Beatriz Piniello Castillo, University of Barcelona

Enzyme O-fucosyltransferase 1 (POFUT1) catalyzes the transfer of one moiety of L-fucose to a threonine or serine residue in the surface of epidermic growth factor-like domains (EGF-LD) [1], small peptides involved in the Notch signaling cascade. Unlike most inverting glycosyltransferases, POFUT1 lacks a basic residue in the active site that could act as a catalytic base to deprotonate the Thr/Ser residue of the EGF-LD acceptor during the chemical reaction. Using quantum mechanics/molecular mechanics (QM/MM) metadynamics methods [2][3], we have uncovered the enzyme's catalytic mechanism, which involves proton shuttling through a conserved active site asparagine that undergoes tautomerization [4]. Experimental kinetic analysis of *Caenorhabditis elegans* POFUT1 Asn43 mutants support this result, as the enzymatic activity was ablated even when mutated to aspartate. These findings will aid inhibitor development for Notch-associated *O*-glycosylation disorders.

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Entropy of water in protein condensates

Saumyak Mukherjee, Ruhr University Bochum

Liquid phase separation (LLPS) of proteins is a spontaneous and reversible process that generates membraneless organelle-like protein condensates. Although extensive research has been dedicated to understand this process, the underlying thermodynamic contributions remain elusive. In this project, we study the entropy of water in such protein condensates to

understand its contribution towards condensate formation. For our studies, we use all atom molecular dynamics simulations on two proteins, namely human gammaD-crystallin (globular protein) and the low complexity domain of fused in sarcoma RNA binding protein (FUS-LCD) (intrinsically disordered protein (IDP)).

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Mixed-Gen Season 2 – Session 7: Simulating non-equilibrium phenomena and rare events
April 28, 2022, 15:00 – 19:00 CET

How to build a theory for a coarse-grained system out of equilibrium
Tanja Schilling, University of Freiburg

Active matter, responsive ("smart") materials and materials under time-dependent load are systems out of thermal equilibrium. To construct coarse-grained models for such systems, one needs to integrate out a distribution of microstates that evolves in time. This is a challenging task. In this lecture, I will recall equilibrium coarse-graining methods, in particular projection operator formalisms. Then I will review recent developments in theoretical approaches to the non-equilibrium coarse-graining problem, in particular, time-dependent projection operator formalisms and numerical schemes to construct explicitly time-dependent memory kernels.

Unbiased sampling of HMC schemes for non separable Hamiltonian systems
Régis Santet, Ecole des Ponts

Hamiltonian Monte Carlo (or Hybrid Monte Carlo) [1] is a Markov Chain Monte Carlo method that allows to sample high dimensional probability measures. Girolami *et al.* [2] have used it to include a position-dependent diffusion coefficient, coming from the overdamped Langevin dynamics, that improves the convergence of the numerical method. It however requires simulating Hamiltonian dynamics with a non-separable Hamiltonian, which is done in practice with implicit methods in order to ensure the preservation of key properties of the Hamiltonian dynamics (symplecticity and time-reversibility in particular) [3]. Unfortunately, actual implicit numerical schemes cannot be reversible, as already noted in the context of constrained stochastic differential equations [4,5]. We show here how to enforce the numerical reversibility of the method to guaranteed that the sampling is unbiased. Our numerical results demonstrate that this correction is indeed relevant in practice.

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Passive particle in an active bath: can we tell it is out of equilibrium?
Jeanine Shea, Johannes Gutenberg University Mainz

We study a passive probe particle immersed in a fluid of active particles. In spite of the non-equilibrium nature of the system, the trajectory of the immersed particle does not exhibit obvious non-equilibrium signatures: its velocity distribution remains Gaussian, the second

fluctuation dissipation theorem is not fundamentally violated, and the motion does not show apparent signs of time reversal symmetry breaking. To tell that the particle is out of equilibrium requires the examination of the behavior of the passive particle *in tandem* with the behavior of the active fluid: the kinetic temperature of the passive particle does not equilibrate to that of the surrounding active particles. Instead, it is higher, even though already the kinetic temperature of the active fluid is enhanced compared to the underlying thermal bath. As a strategy to diagnose non-equilibrium from analyzing probe particle trajectories only, we propose to examine their response to a small perturbation which reveals a non-equilibrium signature through a violation of the first fluctuation dissipation theorem.

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Mixed-Gen Season 2 – Session 8: Electronic energies beyond Density Functional Theory
June 9, 2022, 15:00 – 19:00 CET

Strategies to deal with the many-body problem in materials: a unified view on functionals
Lucia Reining, École Polytechnique, Palaisea

Materials are many-body systems made of electrons and nuclei. Their properties can be expressed in terms of expectation values using many-body wavefunctions. In practice, unfortunately, these expressions cannot be straightforwardly evaluated, because one cannot calculate or store the wavefunctions, even if one decouples electron and nuclei. Alternatively, to using many-body wavefunctions, in principle one can also describe properties in terms of simpler quantities, such as the density or a one-body Green's function. However, the exact expressions, i. e., the exact functionals of the density, or of the Green's function, are most often unknown, and it is not easy to find good approximations.

In this talk, we will focus on general strategies that are common to different approaches relying on functionals. We will discuss ideas and major general concepts. We will investigate the difficulties of finding good approximations, and some strategies for improvement. In particular, we will consider Density Functional Theory, Many-Body Perturbation Theory, and Dynamical Mean Field Theory on the same footing, in order to highlight common ideas and different choices.

On the background of these considerations, selected examples of our recent work will illustrate the state-of-the art of applications in spectroscopy [1,2] and of the search for better approximations [3].

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Theoretical investigation of the molecular structure, vibrational spectra, thermodynamics and non linear optical properties of 4,5-dibromo-2,7-dinitro-Fluorescein
Jean Baptiste Fankam Fankam, University of Yaounde I

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Mixed-Gen Season 2 – Session 4: Simulating quantum materials

July 7, 2022, 15:00 – 19:00 CET

Electronic structure of quantum materials with strong correlations: a dynamical mean-field theory perspective

Antoine Georges, Collège de France, Paris and Flatiron Institute, New York

From copper-oxide superconductors to twisted bilayer graphene and transition-metal dichalcogenides, materials with strong electronic correlations have focused enormous attention over several decades. Solid-state chemistry, new elaboration techniques and improved experimental probes are constantly providing us with examples of novel materials with surprising electronic properties.
In this lecture, I will review some salient physical aspects of strong electronic correlations. I will emphasize that the classic paradigm of solid-state physics, in which electrons form a gas of wave-like quasiparticles, must be seriously revised for strongly correlated materials. Instead, a description accounting for both atomic-like excitations in real-space and quasiparticle excitations in momentum space is required. I will review how Dynamical Mean-Field Theory (DMFT) fulfills this goal. By providing both qualitative insights and quantitative predictions, this approach has significantly advanced our understanding of materials with strong electronic correlations.

Ab initio description of strongly correlated materials: combining density functional theory and dynamical mean-field theory

Sophie Beck, Flatiron Institute, New York

The use of density functional theory (DFT) and dynamical mean field theory (DMFT) has proven successful as a combined approach for electronic structure calculations of strongly correlated materials. In this talk, I will give an overview of how the computational approach works in practice, focusing also on our recent implementation of full charge self-consistency using the community codes Quantum Espresso, Wannier90, and TRIQS. I will discuss recent advances in realistic materials modeling and present our implementation of the DFT+DMFT method in the solid_dmft software package.

Dynamical Mean Field Theory on a Quantum Computer

Jannis Ehrlich, Fraunhofer IWM

Quantum computers promise to overcome the limitations in size which prevent the simulation of large correlated systems with conventional computers. As quantum computers are still in their early stage, we chose the two-site DMFT in order to investigate their potential and current limitations. Calculations based on simulators show, that quantum computing has the potential for these applications, but a specific treatment of statistical errors is required, to ensure that singularities lift each other even when the accuracy is limited.

Anharmonic lattice dynamics from vibrational dynamical mean-field theory

Petra Shih, Timothy Berkelbach, Columbia University

We introduce vibrational dynamical mean-field theory (VDMFT) as a non-perturbative and systematically improvable method for the simulation of anharmonic lattice dynamics [1]. Inspired by DMFT, VDMFT maps the dynamics of a periodic anharmonic lattice of atoms onto those of a self-consistently defined impurity problem with local anharmonicity and coupling to a bath of harmonic oscillators. We test VDMFT on models of anharmonic optical and acoustic

phonons with classical and quantum impurity solvers, and describe nonlocal anharmonicity via its cellular extension and the combination with self-consistent phonon (SCPH) theory (SCPH+VDMFT). With much fewer degrees of freedom in the impurity model than in the full supercell, we show the convergence to accurate results at affordable computational costs.

Reference
[1] Papers: P. Shih and T. Berkelbach, arXiv:2109.00028 (2021)

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Mixed-Gen Season 3 – Session 1: Simulating complex oxides

November 24, 2022, 15:00 – 19:00 CET

Correlated Transition Metal Oxides -- New Twists to Old Problems
Silke Biermann , Ecole Polytechnique, Palaiseau

Influence of Ge substitution on the structural and electronic stability of the competing VO2 phases
Peter Mlkvik, Claude Ederer, Nicola Spaldin, ETH Zurich

We present a density-functional theory (DFT) study of the structural, electronic, and chemical bonding behavior in germanium (Ge)-doped vanadium dioxide (VO_2). Our motivation is to explain the reported increase of the metal-insulator transition temperature under Ge doping and to understand how much of the fundamental physics and chemistry behind it can be captured at the conventional DFT level. We model doping using a supercell approach, with various concentrations and different spatial distributions of Ge atoms in VO_2. Our results suggest that the addition of Ge atoms strongly perturbs the high-symmetry metallic rutile phase and induces structural distortions that partially resemble the dimerization of the experimental insulating structure. Our work, therefore, hints at a possible explanation of the observed increase in transition temperature under Ge doping, motivating further studies into understanding the interplay of structural and electronic transitions in VO_2. Our plan is to continue studying VO_2 under doping and develop suitable methods to model the correlated behavior in this system.

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Mixed-Gen Season 3 – Session 2: Theory and numerical simulation of transport processes in condensed matter

December 15, 2022, 15:00 – 19:00 CET

Gauge invariance of transport coefficients: fathoming heat transport from the struggle to simulate it

Stefano Baroni, SISSA, Trieste

The simulation of adiabatic (*i.e.* non-electronic) heat transport from the Green-Kubo theory of linear-response has long been hampered by two misconceptions. First, the linear-response theory of heat transport was deemed incompatible with modern simulation techniques based on electronic-structure theory, because the quantum mechanical expression of the heat flux that enters the Green-Kubo formula is inherently ill defined. Second, it is commonly thought that the numerical application of this theory would require very long molecular-dynamics simulations, much longer in fact than the typical heat-flux auto-correlation times one is required to evaluate. In this talk I will describe the efforts done at SISSA over the past several years to overcome this state of affairs, which resulted in a methodology allowing one to compute heat transport coefficients from equilibrium molecular dynamics (both *ab initio* and based on deep neural-network potentials), no less than in a deeper understanding of the theory of hydrodynamic fluctuations and their numerical analysis through the statistical theory of stationary time series.

Quantum thermal transport in solids: coherences, disorder, and viscosities

Michele Simoncelli, University of Cambridge

Starting from the Wigner formulation of quantum mechanics, we derive a microscopic equation that describes thermal transport in very general terms and that covers on the same footing crystalline, disordered, and glassy materials. This Wigner transport equation [1] generalizes the Peierls-Boltzmann equation introduced in 1929, and naturally adds a tunnelling term to the drift and scattering of phonon wavepackets. In fact, we show that tunnelling is essential to describe correctly transport in materials that are engineered to be poor thermal conductors, such as thermoelectrics [1], thermal barrier coatings [2], and glasses [3]. We show the predictive accuracy of this formulation by calculating from first-principles thermal transport in various complex crystals and glasses, highlighting also the agreement with experiments at increasing temperatures, where the tunnelling term dominates and offsets the incorrect $1/T$ decay of Peierls-Boltzmann. Last, we coarse grain the atomistic equations of motion to obtain a set of mesoscopic viscous heat equations [4], which generalize Fourier's law accounting for the thermal viscosity, in addition to the thermal conductivity. These mesoscopic equationsexplain the experimental observation of hydrodynamic heat propagation in graphite around 100 K [Huberman et al., Science 364 (2019)], and we exploit them to propose a strategy to amplify signatures of hydrodynamic thermal transport [5].

References
[1] M. Simoncelli, N. Marzari, F. Mauri, Nat. Phys., **15**, *809-813 (2019)*
[2] M. Simoncelli, N. Marzari, F. Mauri, Phys. Rev. X, **12**, *041011 (2022)*
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[4] M. Simoncelli, N. Marzari, A. Cepellotti, Phys. Rev. X, **10**, *011019 (2020)*
[5] J. Dragašević and M. Simoncelli, Viscous heat backflow and temperature resonances in graphite, in preparation.

Sticky coupling as a control variate for the computation of transport coefficients

Shiva Darshan, Ecole des Ponts, ParisTech

A standard method to compute transport coefficients is to simulate Langevin dynamics perturbed by a small non-equilibrium forcing up to a time T and time-average over the trajectory a desired observable divided by the magnitude of the forcing, η. Unfortunately, this method suffers from large finite-time sampling bias and variance in the limit of small forcing—on the order of $(T\eta)^{-1}$ and $(T\eta^2)^{-1}$ respectively.

For overdamped Langevin dynamics, we propose a method to reduce the bias and variance of this computation using a version of the reference (unperturbed) dynamics sticky coupled [1] to the perturbed dynamics as a control variate. We will show that when the potential of the dynamics is strongly convex at infinity, this sticky coupling based estimator's reduces the bias and variance by a factor of η^{-1} compared to the standard method. The case of strongly convex at infinity potentials includes commonly used systems such as Lennard-Jones particles confined to box by a quadratic potential.

Reference
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Topology, oxidation states, and charge transport in ionic conductors

Paolo Pegolo, SISSA, Trieste

Recent theoretical advances, based on a combination of concepts from Thouless' topological theory of adiabatic charge transport [1] and a newly introduced gauge-invariance principle for transport coefficients [2], have permitted to connect (and reconcile) Faraday's picture of ionic transport---whereby each atom carries a well-defined integer charge---with a rigorous quantum description of the electronic charge-density distribution, which hardly suggests its partition into well-defined atomic contributions [3,5]. By relaxing some general topological conditions, one interestingly finds that charge may be transported in ionic conductors without net ionic displacements [4,5]. This allows a new connection between our topological picture and the well-known Marcus-Hush theory of electron transfer, which can be linked with the topology of adiabatic paths drawn by atomic trajectories [5]. As a significant byproduct, this permits the classification of different regimes of ionic transport according to the topological properties of the electronic structure of the conducting material [5].

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CECAM MARVEL CLASSICS IN MOLECULAR AND MATERIALS MODELLING

In this lecture series, we take a different look at fundamental developments of simulation and modelling. Milestone conceptual steps, methods and algorithms are presented by their originators. These technical lectures are followed by an interview in which the speakers recall for us the period, problems, people and circumstances that accompanied these developments, providing important and unusual insight in the birth and growth of tools that we now take for granted.

This lecture series is co-organized by CECAM and MARVEL (http://nccr-marvel.ch/) at EPFL and broadcasted in webinar format.

Ab initio studies of electronic excitations using many-body perturbation theory

Tuesday January 18 2022, 16:00 CET

Underlying concepts and practical implementations

Rex Godby, University of York, UK

Many-body perturbation theory (MBPT) is a set of theories of the quantum states and time-evolution of a system of interacting electrons. MBPT is based on the iterative solution of an exact set of coupled equations for the electron Green's function (which plays the central role in MBPT, rather like the Kohn-Sham wavefunctions in density-functional theory). The Green's function has a physical meaning: the probability amplitude for an electron, injected into the system at a given position, to have arrived at another position after a given time interval. The similarity between the MBPT and DFT formalisms gives a useful perspective on both theories. Exchange and correlation are described in MBPT by the self-energy, Σ, a non-local, energy-dependent potential that is the counterpart of the exchange-correlation potential in DFT. After one iteration of the coupled equations, Hedin obtained his 1965 "GW" approximation for Σ which has been the dominant approach in ab initio applications of MBPT over 35 years. I shall review the status of different implementations of GW and higher-order theories, addressing their theoretical and computational merits and also the variety of experimental characteristics of a system that can be predicted.

Rex Godby is emeritus professor of theoretical physics in the Department of Physics, University of York. His research focuses on the quantum dynamics of systems of interacting electrons and other complex systems, including studies of the exact Kohn-Sham potential of time-dependent density-functional theory (TDDFT), together with the development of improved approximate TDDFT functionals for dynamical problems. After completing his BA and PhD degrees at the University of Cambridge, Godby was a postdoctoral researcher at Bell Laboratories, New Jersey, from 1984 to 1986. He then returned to Cambridge as a research fellow, moving to York in 1995. He is one of the founding members of the European research network on the ab initio description of electronic excitations in matter, the ETSF (European Theoretical Spectroscopy Facility), which evolved from earlier European collaborations starting in the 1980s, via the "Nanophase" and "Nanoquanta" research networks.

Electronic excitations in a many-body world, from quasiparticles to spectra

Lucia Reining, Ecole Polytechnique, Palaiseau, France

Many-body effects influence observables quantitatively, and often also qualitatively. In particular, while a spectrum of independent electrons consists of single lines that correspond to "the" energy of an electron, real spectra can show broad peaks and a rich structure. These observations can be related to an effective hamiltonian that is frequency-dependent, even when the full many-body hamiltonian is static. In this part we will discuss the origin and physics of the frequency-dependence, as well as its consequences. Building on Rex Godby's introduction, we will examine the GW approximation with these questions in mind, highlighting the strong points and limitations of GW from a conceptual point of view, as well as in comparison with experiment. We will then see how this insight can guide us in the search for approximations beyond GW, and we will extend the discussion to the electron-hole problem. Finally, we will make a few points about the gap between theory and experiment.

Lucia Reining is an exceptional grade senior researcher of CNRS, working at the Ecole Polytechnique in Palaiseau, France. After her master's degree at the University RWTH Aachen in Germany with a pen-and-paper thesis, she moved to "Tor Vergata" University in Rome to work with Rodolfo Del Sole, who convinced her that computers are a useful tool to get numbers out of a tight binding model. She remained for a PhD about many-body perturbation theory in tight binding. Since people tended not to

believe semi-empirical approaches, she decided to learn more about first principles calculations. After her PhD she moved to CECAM, which was located at Orsay at that time. With a Marie-Curie fellowship, she discovered the excitement of speeding up ab initio calculations, more specifically working on GW. Towards the end of her stay at CECAM she got involved in European projects which laid the foundations for long-lasting collaborations and friendships, and which culminated in the European Theoretical Spectroscopy Facility (ETSF, www.etsf.eu). At that time, she and her coworkers started their activity on the first principles solution of the Bethe-Salpeter equation. She then entered the CNRS. Today, her work combines analytical and numerical approaches including the development of density functionals and Green's functions functionals, approximations and applications, often in collaboration with experimentalists. Besides journal publications, a joint book with Richard Martin and David Ceperley about "Interacting Electrons" reflects her view on many-electrons physics, and her work has been recognized by a silver medal of the CNRS in 2003, election as APS fellow in 2007, an ERC advanced grant in 2012 and the Gentner-Kastler prize of the French and German physical societies in 2020.

Understanding excited states from first principles: Single- and multi-particle excitations and time-dependent phenomena

Steven Louie, University of California at Berkeley
Lawrence Berkeley, National Lab, Berkeley, California, USA

Many fascinating phenomena in nature owe their emergence from interactions of a large number of particles. In particular, many-electron interactions are often dominant in excited-state phenomena of real materials. We present in this talk some of our understanding of and capabilities for ab initio computation of excited-state phenomena within the interacting many-particle Green's function approach using many-body perturbation theory. Different physical measurements and phenomena are connected to the different kinds of interacting Green's functions. Photoemission spectroscopy may be understood from the perspective of an interacting 1-particle Green's function (e.g., within the GW approximation for the electron self-energy) as discussed in the talks by Rex Godby and Lucia Reining. However, correlated multi-particle excitations such as excitons, trions, and bi-excitons require knowledge of the 2-, 3- and 4-particle interacting Green's functions, respectively. Moreover, nonequilibrium and time-dependent phenomena, such as field-driven phenomena and pump-probe experiments, require yet very different treatments from those of the equilibrium case. We shall discuss the concepts and methods behind the ab initio studies of these phenomena, and give some illustrative examples including single- and multi-particle excitations, linear and nonlinear optical responses, as well as field-driven time-resolved and angle-resolved photoemission spectroscopy (tr-ARPES).

Steven G. Louie received his Ph.D. in physics from the University of California at Berkeley in 1976. After having worked at the IBM Watson Research Center, Bell Labs, and U of Penn, he joined the UC Berkeley faculty in 1980, where he is professor of physics and a senior faculty scientist at the Lawrence Berkeley National Lab. He is an elected member of the National Academy of Sciences, the American Academy of Arts & Sciences, the Academia Sinica (Taiwan), and a foreign member of the Chinese Academy of Sciences, as well as a fellow of the American Physical Society (APS), the American Association for the Advancement of Science, and the Materials Research Society (MRS). He is a recipient of the APS Aneesur Rahman Prize, the APS Davisson-Germer Prize, the MRS Materials Theory Award, the Foresight Institute Richard Feynman Prize, the DoE Award for Sustained Outstanding Research in Solid State Physics, as well as named Jubilee Professor of the Chalmers University of Technology, Ørsted Lecturer of the Technical University of Denmark, and Benjamin Lee Professor Award of the Asia Pacific Center for Theoretical Physics, among others. Louie's research spans a broad spectrum of topics in condensed matter physics and nanoscience. He is known for his pioneering development of the ab initio GW method and for his studies of novel bulk and reduced-dimensional systems, with over 670 publications and a current Google Scholar h-index of 159.

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Simulation methods for spin glasses with applications in optimization

Thursday November 3 2022, 15:00 CET

3d simulations of spin glasses on dedicated architectures

Giorgio Parisi, Università di Roma La Sapienza

In this talk I will present very large-scale simulations that have been done on spin glasses in 3 and 4 dimensions. After an introduction on the theoretical framework based on replica symmetry breaking, I will present simulations, both at equilibrium at non-equilibrium, focusing on the large-scale behaviour.

Giorgio Parisi is professor emeritus of Theoretical Physics at the University of Rome La Sapienza and associate researcher at the INFN National Institute of Nuclear Physics. From 2018 to 2021 he was President of the Accademia Nazionale dei Lincei, and currently acts as President of the Class of Physical Sciences, Mathematics and Natural and Vice President of the Academy. Born in Rome in 1948, Parisi completed his studies at the Sapienza University of Rome where he graduated in physics in 1970. Throughout his scientific career, Giorgio Parisi has made many decisive and widely recognized contributions in different areas of physics: in particle physics, statistical mechanics, fluid dynamics, condensed matter, supercomputer. He has also written articles on neural networks, immune systems and movement of groups of animals.
Among other recognitions, Parisi was awarded, in 1992, the Boltzmann Medal for his contributions to the theory of disordered systems, the Dirac Medal in Theoretical Physics in 1999, the Max Planck Medal in 2011, the Lars Onsager Prize of the American Physical Society 2016, the Nature Award for Mentoring in Science 2013, and the Wolf Prize in Physics in 2021.
In 2021 Giorgio Parisi was awarded the Nobel Prize in Physics.

Spin glass concepts and algorithms in hard constrained satisfaction problems

Marc Mezard, Università Bocconi Milano

Spin glass theory has had a large impact on many fields. Among them, a new field of research is rapidly expanding at the crossroad between statistical physics, information theory and combinatorial optimization. It deals with problems which are very important in each of these fields, like spin glasses, error correction, or satisfiability. This talk will review how the cavity method, initially developed to understand spin glass theory in a framework more transparent than the replica method, can be transformed into message passing algorithms that turn out to be quite efficient for several large-scale problems of constraint satisfaction and statistical inference.

Marc Mezard is a Professor of Theoretical Physics. He studied physics at Ecole normale supérieure in Paris and obtained his PhD in 1984. Hired at CNRS in Paris, he was Research Director in Université Paris Sud. From 2012 to 2022 he was Director of Ecole normale supérieure, and then joined Bocconi University as a professor, in the newly created department of computational sciences. Prof. Mezard's work focuses on statistical physics of disordered systems, with applications in various fields like information theory, computer science, machine learning, biophysics. In recent years his research has focused on information processing in neural networks, machine learning and deep networks, with specific interest in the theoretical impact of data structure on learning strategies and generalization performance.

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Dynamical mean-field theory

Thursday December 8 2022, 15:00 CET

The Dynamical Mean-Field Theory perspective on strong electronic correlations

Antoine Georges, College de France & Flatiron Institute

From copper-oxide superconductors to twisted two-dimensional materials, strong electronic correlations have focused enormous attention over several decades. The classic paradigm of solid-state physics in which electrons form a gas of wave-like quasiparticles must be seriously revised for strongly correlated materials. Instead, a description accounting for both atomic-like excitations in real-space and quasiparticle excitations in momentum space is required. I will review how Dynamical Mean-Field Theory (DMFT) fulfills this goal and provides an original physical perspective on strongly correlated electron materials as well as an efficient computational framework to understand and predict their properties. Thanks to the efforts of a whole community over almost three decades, the theory has been developed to such a point that it can successfully be applied to real materials taking into account their structure and chemical composition. I will also outline how the theory is being extended and generalized in many fruitful directions.

*After initial steps in the statistical mechanics of disordered systems, **Antoine Georges** has focused his research on the physics of interacting quantum systems such as materials with strong electronic correlations, quantum impurity models and cold atomic gases in optical lattices. He is one of the co-inventors of Dynamical Mean-Field Theory, for which he shared the 2006 Europhysics Condensed Matter Prize, the 2020 Aneesur Rahman Prize and the 2022 Feenberg Medal. This theory has deeply transformed our understanding of materials with strong correlations and our ability to explain, calculate and predict their physical properties.*
Antoine Georges is a Professor at Collège de France, Paris and the Director of the Center for Computational Quantum Physics (CCQ) at the Flatiron Institute, Simons Foundation, New York. He also has an affiliation to the University of Geneva and is a member of the French Academy of Sciences.

Understanding and computing the properties of strongly correlated electron materials: the Dynamical Mean Field Theory perspective and challenges ahead

Gabriel Kotliar, Rutgers University and Brookhaven National Laboratory

Strongly correlated electron materials pose great conceptual and computational challenges. Quantum embedding methods such as Dynamical Mean Field Theory (DMFT) have enabled a community of scientists to achieve great progress in this area. After reviewing some basic ideas underlying this approach, such as the reduction of the full many body problem into a quantum impurity model satisfying a self-consistency condition. We will then provide examples of some progress using materials containing elements from different regions of the periodic table, outlining different classes of strongly correlated behavior: heavy fermions, Mott systems and Hund metals. We will conclude with the great challenges ahead.

***Gabriel Kotliar** got BSc and MSc at the Hebrew University inJerusalem and his PhD at Princeton University in 1983 under the supervision of PW Anderson, working on the physics of disordered systems. He was a postdoc at the ITP in Santa Barbara (1984-1986), an assistant professor at MIT (1986-1988) and has been at Rutgers University where he holds a Board of Governors Chair ever since. He is currently the director of the center of theoretical spectroscopy and materials design at Brookhaven National Laboratories and the director of the Center for Materials Theory at Rutgers.*
Among his research achievements are the prediction of d wave superconductivity and the pseudogap in the copper-based superconductors, early studies of the interplay of superconductivity and disorder, and advancing many techniques for treating strongly correlated electron systems. He has been the

recipient of numerous awards including the Alfred P. Sloan Research Fellow, the Presidential Young Investigator Award, the Lady Davies Fellowship, the Guggenheim fellowship, the Blaise Pascal Chair. He is a fellow of the American Physical Society, a member of the National Academy of Sciences and a member of the American Academy of Art and Sciences. He was one of the recipients of the (2006) Agilent Technologies Europhysics Prize, the 2021 Aneesur Rhaman prize for computational physics, the 2022 Feenbergmedal for his contributions to the development of Dynamical Mean Field Theory.

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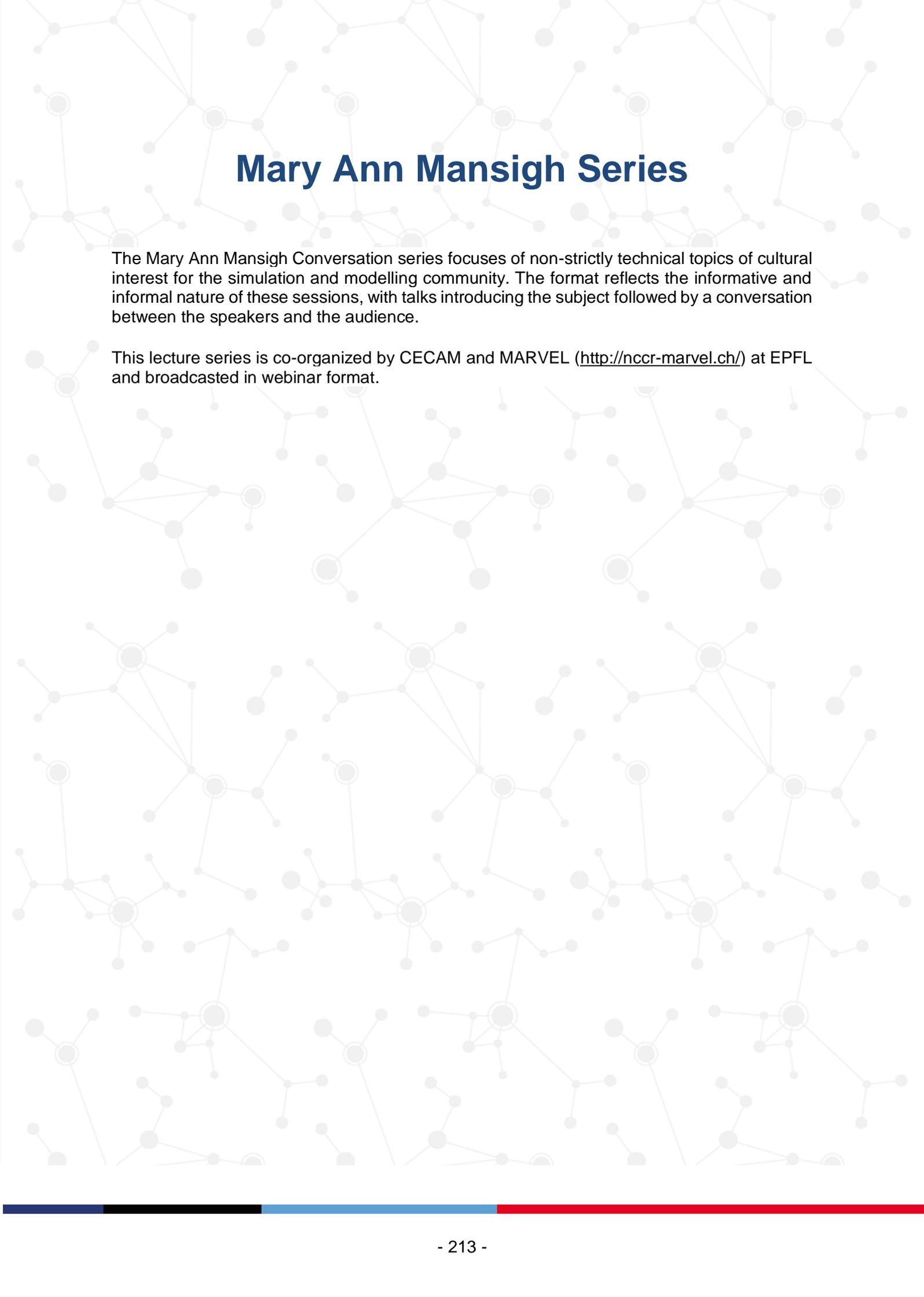
Mary Ann Mansigh Series

The Mary Ann Mansigh Conversation series focuses of non-strictly technical topics of cultural interest for the simulation and modelling community. The format reflects the informative and informal nature of these sessions, with talks introducing the subject followed by a conversation between the speakers and the audience.

This lecture series is co-organized by CECAM and MARVEL (http://nccr-marvel.ch/) at EPFL and broadcasted in webinar format.

Fascination and industrial value of materials modeling

Thursday May 5 2022, 15:00 CEST

Fascination and industrial value of materials modeling

Erich Wimmer, Materials Design

Understanding the marvelous complexity of nature is a deeply human desire, and to this end modeling is a uniquely powerful tool. Starting from this view, this lecture will revolve around an aspect of high actuality, namely energy. Moving from a society burning fossil fuels and thereby destroying our vulnerable environment to solutions that are sustainable and realistic, is perhaps one of the biggest technological and societal challenges of the 21st century. By discussing specific examples including metal alloys and polymers, batteries, electronic devices, and CO_2 capture, this lecture will show how modeling and simulations are applied in a wide range of length and time scales, supporting the path to better and sustainable technologies. With deeply founded theoretical concepts, highly efficient algorithms in sophisticated computer programs, and unprecedented compute power, never before had humans such fascinating tools in their hands. Thus, materials modeling creates industrial value and it satisfies our human desire for understanding the processes that form and transform materials around us.

Erich Wimmer is co-founder, Chief Scientific Officer, and Chairman of the Board of Materials Design. He has initiated major software development programs, organized industrial consortia, and led research efforts to create industrial value through computational chemistry and materials science. Dr. Wimmer is author and co-author of over 130 scientific publications and has given numerous lectures worldwide.

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