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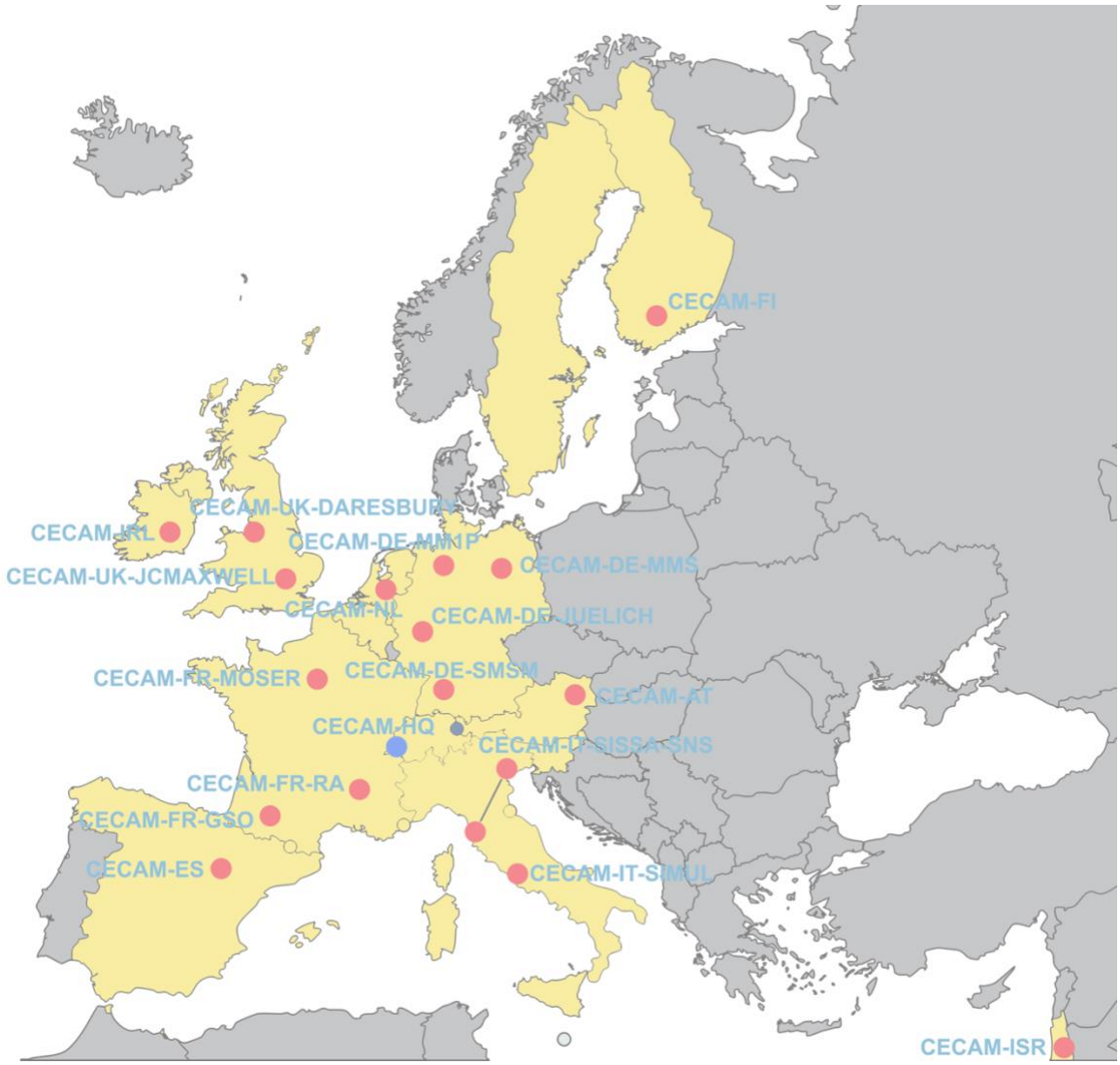
2021



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Centre Européen de Calcul Atomique et Moléculaire

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INTRODUCTION

CECAM activities in 2021 were strongly affected by the perduring COVID pandemic. As a result, most events in the Flagship program were held online with a gradual return to in presence workshops and schools towards the end of the year. Despite these difficulties, a rich program was deployed online or in person at the Headquarters in EPF-Lausanne and in the node network.

The Flagship program consisted of 25 events (20 online, 5 hybrid) at EPF-Lausanne, and 37 events (16 online, 6 hybrid, 15 onsite) in the node network. Activities were organised by 101 scientists from 19 countries at Headquarters, and 195 from 20 countries in the Nodes. Participants numbers were very high, reflecting in part the larger attendance for hybrid or online events. Headquarters events totaled 1838 attendees – 98 onsite – from 77 countries; with 1750 participants to node activities – 384 onsite – from 68 countries.

In addition to the workshops and schools of the Flagship program, the yearly program featured several other online or hybrid activities. Recordings of all events were posted on the CECAM website and YouTube channel, with an average of 250 views.

The series *Classics in molecular and materials modelling*, co-organised with the MARVEL NCCR, continued its successful run in webinar form. Three sessions were held in 2021, covering topics in condensed matter physics and statistical mechanics. We signal, in particular, the session on *Free energy of solids: origins and consequences of the 1983 CECAM workshop* with D. Frenkel and T. Ladd due to its close connection with the Center.

The 2021 CECAM MARVEL Mary-Ann Mansigh Conversation was organized in collaboration with the Fondazione Adriano Olivetti and focused on the history of the first desktop computer, PROGRAMMA 101. This was a hybrid event that gathered about 60 participants at EPF-Lausanne and was followed by ca 150 remote participants.

A new series of webinars was inaugurated to facilitate learning and networking for PhD students and early career researchers: *The Mixed-Gen series*. This series comprised 6 events in its first season and 8 (3 of which took place in 2021) in the second (split between 2021 and 2022). This innovative format was conceived at first to mitigate the negative impact of the pandemic on the opportunities for the younger members of the community to share their work and interact with leaders in the field. Each session consisted in an extended lecture by an experienced speaker, followed by contributions from early career researchers. A GatherTown poster and networking session concluded the event. The response of the community to this proposition was truly remarkable, with an average of 100 live participants and numerous views of the recordings posted on the CECAM YouTube channel, with a peak of 400+views.

In depth descriptions of all activities described above are gathered in this report.

Location: Online event/CECAM-UK-DARESBURY
Webpage: https://www.cecam.org/workshop-details/8
January 18-22, 2021

1 State of the art

The purpose of this Extended Software Development Workshop (ESDW) is to introduce students to mesoscale simulation via Dissipative Particle Dynamics and to parallel programming of hybrid CPU-GPU systems via OpenACC directives.
Mesoscale simulations have grown recently in importance due to their capacity of capturing molecular and atomistic effects without having to solve for a prohibitively large number of particles needed in Molecular Dynamic (MD) simulations. Different approaches, emerging from a coarse approximation to a group of atoms and molecules, allow reproducing both chemical and physical main properties as well as continuum behaviour such as the hydrodynamics of fluid flows.
One of the most common techniques is the Dissipative Particle Dynamics (DPD): an approximate, coarse-grain, mesoscale simulation method for investigating phenomena between the atomic and the continuum scale world, like flows through complex geometries, micro fluids, phase behaviours and polymer processing. It consists of an off-lattice, discrete particle method similar to MD but with replacement of a soft potential for the conservative force, a random force to simulate the Brownian motion of the particles and a drag force to balance the random force and conserve the total momentum of the system.
OpenACC directives allow to easily offload to GPU sections of the code and they are the ideal starting point to introduce GPU programming. We used the NVidia Course on OpenACC from the Deep Learning Institute (DLI) to present the basic concepts and then provided instructions on how to implement into DL_MESO code

2 Training provided

We presented the different topics in the following main points:
INTRODUCTION TO DPD AND DL_MESO: we covered basic background and theory on DPD, like the pairwise thermostat, the soft repulsive interaction and the applications of DPD. In the hand session we presented DL_MESO, how to use it and how to setup the parametrization for a proper simulation. We also presented the parallel version of DL_MESO based on MPI library and domain decomposition as well as its performance on different systems size and complexity.
INTRODUCTION TO OpenACC (NVidia DLI COURSE): we introduced what are GPUs, the hardware and the software abstraction concepts and why to use them. We then started the NVidia DLI course which focus on the profiling of applications, compute directives, data management and loop optimization. The use of Jupyter notebook has strongly simplified the interaction and presentation of the different concepts.
PORTING DL_MESO to OpenACC: we explained how to implement the OpenACC directives to DL_MESO starting from the performance analysis using NSight and gradually offloading the different compute loops to DL_MESO. We also presented the single and multi GPU versions of DL_MESO developed in CUDA language to give a comparison on the speedup and algorithms changes required to achieve high performance. The students had access to the Hartree Centre Supercomputer ScafellPike and be able to run different simulations on the latest Volta GPU cards as well as profiling their application and identify hotspots.
We had 19 people registered for the event from 4 different continents, but only 9 actually attended. The difference in time zone difference could have been the main reason for the

missing attendees. During the hand on sessions, we realized that the lack on face-to-face interaction slowed down the understanding. We used break-out rooms to improve the interactions and successfully allowed all participants to stay at the same level of exercises.

3 List of the software development projects

The following modules have been started during the ESDW: from 1-5 are related to DL_MESO master version. They are mainly updated of previous modules developed by Silvia Chiacchiara to make the software compatible with version 2.7. The modules from 6-9 are all around the porting of DL_MESO to OpenACC. Every student has started offloading the first Verlet Velocity step and gradually they will extend the porting to the rest of the code.

1) Updated moldip_af_dlmeso_dpd to use DL_MESO version 2.7 HISTORY files (by Michael Seaton)
2) Updated dipole_af_dlmeso_dpd to use DL_MESO version 2.7 HISTORY files (by Michael Seaton)
3) Updated dipole_dlmeso_dpd to use DL_MESO version 2.7 HISTORY files (by Michael Seaton)
4) Updated tetra_dlmeso_dpd to use DL_MESO version 2.7 HISTORY files (by Michael Seaton)
5) Updated format_dlmeso_dpd to use DL_MESO version 2.7 HISTORY files (by Michael Seaton)
6) DL_MESO OpenACC version (by Bruna Franciele Faria)
7) DL_MESO OpenACC version (by Portell-Canal Xavier)
8) DL_MESO OpenACC (by Sorush Khajepor)
9) DL MESO OpenACC (by Lianne Gahan)

4 Future plans

There is no current plan for a follow up, despite students were very well engaged and enthusiastic. However, we are keeping in contact via the ECAM GitLab repository to allow them to continue developing the OpenACC version and eventually merge all their contributes into the OpenACC branch of DL_MESO created on the STFC GitLab repository. We also had a following up meeting on the 3rd of February 2021 with professor S. Ramasamy from the Addis Abada Science and Technology University in Ethiopia for possible future collaboration with EU and UK.

5 Participant list

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Materials Design for Energy Storage and Conversion: Theory and Experiment

Location: Online event - hosted by CECAM-HQ
Webpage: https://www.cecam.org/workshop-details/25
March 2-5, 2021

1 State of the art

First-principles calculations as well as ab initio thermodynamics, kinetics, dynamics, and continuum-scale modelling have been applied to materials and interfaces in energy storages and conversion systems. [1,2] Continuum-scale modelling of interfaces and space charge have been developed. [3,4] However, most of the computational parameters in these models have been either provided by experimental data or were considered as free parameters. Treatment of chemical reactions at the active interfaces in energy conversion devices remains a great challenge for theory. The long-range effects of charge redistribution between electrodes, adsorbates, and electrolyte play an important role, and must be taken into account. [3,5,6] A remaining problem is to account for statistical effects at finite temperatures, in particular configurational entropy and interplay of chemical reactions. A pre-requisite to address these problems from first principles is to accelerate the energy evaluation per structure without losing the accuracy, e.g. by combining ab initio calculations with machine learning to find more transferrable interatomic potentials.[7-9] Data mining and machine learning can be also used to find descriptive parameters (descriptors) [10,11] that establish correlations between easily computable or measurable properties of materials (e.g., properties of involved atoms and interfaces) and their (electro)chemical and catalytic properties. Characterization of primary particles and microstructures as well as interfaces and grain boundaries are a major challenge for both experimentalists and theoreticians. Multiscale modelling of these complex systems is still in its infancy stage and needs to be further developed. It is also an enormous challenge for experimentalist to characterize energy materials in atomic scale, in particular at realistic operational conditions.

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2 Major outcomes

Multiscale modelling of solid-liquid interfaces was discussed. A method and software developed by Yoshitaka Tateyama caught interest of several attendees and corresponding contacts were established. Artem Abakumov described experiments on Li-rich layered oxides as battery cathodes. He emphasized the complexity of these materials, whose performance crucially depends on point defects and grain boundaries. Understanding and design of such materials requires concerted effort of theory and experiment. Axel Gross demonstrated how descriptors can help in designing more stable electrode materials. In particular he showed how ionic migration barriers that affect dendrite growth depend on ion features such as radii and electronegativity. Robert Muecke showed how detrimental stresses inside cathode materials can be reduced by designing appropriate microstructural textures. This was achieved by combining modelling with experimental measurements to reconstruct real microstructures. Keith Stevenson discussed prospects and challenges of beyond-Li batteries. He reported a discovery of high-voltage cathode materials containing Ti. For this, novel synthesis technique had to be designed. Also, formation of solid-electrolyte interphase was identified as the main reason of failure of hard-carbon based anodes. It was suggested to add organic components, which were demonstrated to partially remedy this problem. Michael Eikerling opened the discussion on multiscale modelling in electrocatalysis. He emphasized the importance of taking into account local reaction conditions. Solving for the potential distribution in the interfacial region was identified as the fundamental problem. It was addressed by combining DFT, molecular dynamics, and classical approaches for electrolyte. Piotr Kowalski focused on the problems in describing electronic structure of typical battery materials containing transition metals. He showed how DFT+U can help to solve these problems, but also pointed out that it should be applied carefully and more accurate and universal approaches have to be developed. The topic of multiscale modelling of electrochemical interfaces was continued by the group of Timo Jacob. Dynamics of electrode-electrolyte interfaces at realistic conditions were modelled by a combination of first-principle calculations and reactive force field. The importance of controlling synthesis conditions for a better theory/experiment connection was emphasized and demonstrated. A novel method for creating highly active electrocatalysts using plasma treatment was introduced. Minoru Otani presented an approach for modelling energy storage devices, combining DFT with classical liquid theory. He also showed applications of his method to MXene electrodes. The method induced great interest from the audience. Leonid Kahle presented a high-throughput computational infrastructure and its application to Li-ion battery design. The framework includes software tools for automation of calculations and fast prescreening of materials. The software and underlying methods induced raised great interest in the audience. Alexander Shapeev presented a powerful machine-learning approach to interatomic potentials, which was also of great interest to the audience, showing the demand for the approaches accelerating energy materials modelling. The accuracy of the approach for the complex materials used in batteries and electrochemistry was discussed. Sergey Levchenko presented

an approach to find descriptors and identify mechanisms of catalytic reactions based on theoretical or experimental data. At the end of the workshop, modelling of 2D nanomaterials with unique electronic and structural properties was extensively discussed. The methodological challenges of modelling such materials were pointed out, and application of presented methods to nano-structured energy materials was discussed.

3 Community needs

The workshop was very positively regarded by the audience. Key participants admitted the necessity for combined theoretical/experimental approaches in the field of energy conversion and storage. There is a need to develop a common language for an efficient communication between theory and experiment. The complexity of real energy materials requires development of multiscale theoretical and data-analysis approaches. On the other hand, awareness of experimentalists about theory challenges and efforts to perform well-controlled experiments were recognized as equally important. The interest from the diverse audience clearly showed the need for codes that implement multiscale approaches in a user-friendly way. The demonstrated need for combined efforts of theorists and experimentalists in studying energy materials, and the great interest in such materials in the modern times of growing energy demand and environmental awareness, shows a great potential for a series of CECAM events on this topic. Importantly, the events should not be too specialized, but should include discussions of advances in methodology in other fields that can be applied for designing energy materials.

4 Funding

The typical funding schemes included national funding such as DoE in US, DFG in Germany, and RSF in Russia. Projects in Japan received governmental support via ministry of education, and in Japan and Russia via educational organizations. Also, industrial funding was mentioned (IBM, BASF). Dual funding schemes such as German-Russian, and facility-based proposals (in particular with Oak Ridge National Laboratory) were discussed during the meeting. The possibility of participating in Horizon calls was also discussed.

5 Will these developments bring societal benefits?

The topic of the workshop has outstanding potential societal benefits. Energy is the key component of modern society, wellness and development of humanity depends on energy generation and conversion. Sustainability of energy generation and conversion is also the key factor. Climate change was recognized as an existential threat to the humanity. Functional materials are at the heart of modern energy generation and conversion devices. More stable and efficient materials for photovoltaics, batteries, catalysts, fuel cells, and other applications are urgently needed to prevent devastating climate changes. However, finding such materials turned out to be a great challenge. Despite tremendous efforts by both industry and academia, the progress in this area is slow. The complexity of energy materials and processes requires a concerted effort combining advanced theoretical and experimental approaches. Despite the fact that many groups in different countries work in the area of energy conversion and storage, the communication between them remains unsatisfactory. in particular communication between theory and experiment remains difficult due to the huge range of scales at which energy devices operate. While atomic scale is very difficult to study experimentally, especially at operational conditions, theory has difficulties connecting atomic and macroscale, with the latter being no less important. In this situation, workshop discussions have a potential to establish communications worldwide and to develop a common language for theorists and experimentalists, greatly advancing the development of sustainable energy solutions. Due to

its importance, the work in this area is and will continue to be supported by various funding sources, including government, industry, and research funding agencies.

6 Participant list

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Interdisciplinary Challenges in Nonequilibrium Physics

Location: CECAM-AT
Webpage: https://www.cecam.org/workshop-details/34
April 12-16, 2021

1 State of the art

Non-equilibrium processes play important and often vital roles at many length- and time-scales, ranging from those characteristics of the organisation of the cell nucleus to those relevant for the flocking of birds. Non-equilibrium statistical mechanics encompasses fundamental tools that are actively being developed and refined to understand such complex phenomena. A crucial factor that has hindered, so far, the formulation of a unified approach to nonequilibrium is the variety of different ways in which various systems can be out of equilibrium. Biological living complexes, glasses or active matter, represent such different systems and pose different challenges that are generally approached from different perspectives.
A key step to improve our understanding of universal non-equilibrium mechanisms at play across scales is to discuss and compare different ideas coming from all different research fields. The meeting will focus on finding a common denominator for theoretical, computational and experimental approaches, so to maximally promote collaborations and knowledge exchange among early career researchers.
The workshop included four sessions focusing on the following topics: 1) biophysics & polymers, 2) active matter, 3) non-equilibrium statistical mechanics, 4) glasses & disordered systems.
These topics have been chosen so as to cover a broad spectrum of different research directions in which non-equilibrium physics plays a major role and that display some degree of overlap. This is to ensure: (i) smooth transitions between the sessions, thus conveying the idea that all these topics are interconnected, (ii) stimulating cross-talks among invited speakers from all sessions and (iii) opportunities for the participants to discuss ideas and take them further in the form of new collaborations (independent of their respective PIs) or joint applications for research grants.

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2 Major outcomes

The Workshop, in its online format, has delivered three key outcomes:

1. **Gathered together an interdisciplinary community to discuss common challenges and new research direction.** The workshop managed to cross field-specific boundaries, bringing together researchers working in very disparate areas, from the mechanical properties of cells to the energy landscapes of glasses, from colloidal active particles to the rheology of ring polymers.
2. **Provided the opportunity for early-career researchers to showcase their work.** The workshop included several activities that allowed researchers and students with different levels of expertise to present their work and their interests. These activities included a Meet & Greet session (where all participants - regardless of their status - were able to introduce themselves and briefly illustrate their interests) the scheduled invited seminars (where selected early-career speakers provided impactful lectures) and the poster session (where discussion flourished for typically more than one hour and a half). Also, the poster sessions brought the spotlight on three specific contributions, selected by the participants, as particularly outstanding and consequently awarded by a prize (books).
3. **Produced high-quality and pedagogical material in the form of recorded seminars.** The Erwin Schroedinger Institute has adapted and uploaded and made available online video recordings of all of the presentations using the YouTube platform, at the following link https://youtube.com/playlist?list=PLjvY6sC_0lhIdmKmmUihEAegk6_fz5dFY
As the speakers made a substantial effort to reach out beyond their specialised research areas, all of the presentations are highly accessible and provide an accessible means to understand the most recent ideas in nonequilibrium physics. This is potentially extremely valuable for a variety of audiences: undergraduate and postgraduate students interested in the most recent advances in physics; lecturers presenting new topics in their courses; researchers approaching novel research lines.

3 Community needs

The present workshop has gathered together a broad community of early career researchers interested in non-equilibrium problems, spanning complex problems across different sub-fields.
During the workshop, it has become clear that the complexity of the problems addressed, for instance concerning biopolymers, collective behavior of active systems or slow glassy dynamics, requires extensive computer simulations. Several talks have presented results resulting from large scale simulation studies, many of them performed in HPC facilities. Some of the investigations carried on by the community require access to such equipment, which is accessible only through competitive calls like, for instance, HPC Europe. Also, running simulations in HPC environments requires the use of robust codes than run in parallel. A significant part of the community makes use of existing codes such as LAMMPS or GROMACS, but many young researchers are unfamiliar with them. It appears as a need for the community to get some **training on parallel computing and existing simulation packages**. Also, the workshop helped to disseminate the different schemes to get access to HPC facilities.
The participants have been particularly enthusiastic and active during all the sessions of the workshop. We have received quite some feedback acknowledging the usefulness of a workshop with such a format to establish a long-lasting network of scientist interested in non-equilibrium phenomena. Indeed, young participants felt more confident surrounded by other scientist at their same career stage. This had a clear impact on the nature of the interactions between participants: the main sessions were unusually lively and friendly, as well as the poster sessions in gather.town. We thus strongly believe there is a real need to **pursue workshops of this kind**, between early-stage researchers sharing doubts, raising questions and feeling as part of a lively community.

4 Funding

The main novel aspect of the workshop is its focus on the young career scientists. Although the funding was not explicitly discussed during the meeting, some discussions during the breaks touched upon the common need to increase the support of early career scientists. Some have just established their groups with the help of national or international grants (e.g. ERC StG), while others will be applying shortly. In light of that, grants specifically aimed at young scientist in the form of e.g. computational time (such as PRACE's calls or HPC Europa) would be very welcome. There was no dedicated session on joint research proposals, but a number of collaborations have been established among the young scientists, promising such outcomes in the near future. The participants explicitly discussed the plan to organize the next iteration of this meeting for its usefulness for the early-career researchers. Plans to submit a proposal for its funding are already being prepared

5 Will these developments bring societal benefits?

The interdisciplinarity nature of the workshop provided an opportunity for young researchers to expand their research lines as well as for approaching old problems from a new perspective, supplying critical novel insight. This constitutes the first benefit of an interdisciplinary meeting. Some contributions had immediate technological and industrial relevance. For example, T. O'Connor investigated the rheology of polymers in elongational flow, which is akin to the extrusion process ubiquitously employed in the industrial production of polymeric materials: a better understanding of the physics of these systems may bring a direct improvement of the production of fibrous materials. On the long term, many future developments, stemming from the contributions of the workshop, can be envisaged:

1. New materials, like a DNA-based material that can tune its mechanical and rheological properties by re-designing its internal network (D. Michieletto) and new ways to characterize (D. Truzzolillo), as well as manipulate these materials at the micro scale (S. Ramananarivo, J. De Graaf).
2. A better understanding of the cellular cortex (E. Fischer-Friedrich) will lead to a future generation of nano-vessels; the control over Brownian engines (P. Pietzonka, G. Verley) and of non-equilibrium chemical networks (G. Falasco, I. Neri) will transform them into autonomous units.
3. The ability to control bacteria (H. Massana-Cid) and understand their navigation system (A. Zoettl, B. Liebchen) will be exploited in the future for many different purposes, e.g. to avoid/promote biofilm formation, water purification or selective regulation of micro-environments.
All the possible developments of these research topics go towards improving societal wellness, producing sustainable, biocompatible materials, controlling our environment in a less disruptive fashion and providing novel ways to efficiently bolster our immune system against new threats.

6 Participant list

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Open Databases Integration for Materials Design

Location: Online meeting, hosted by CECAM-HQ
Webpage: https://www.cecam.org/workshop-details/52
June 7-11, 2021

1 State of the art

In the last few years, materials design has changed quite dramatically. Thanks to the increase of computing power, large sets of first-principles calculations can be performed automatically, adopting a high-throughput (HT) approach. Large databases (DBs) can then be created with the calculated properties for existing and hypothetical materials. In the same spirit, experimental databases have also developed, gathering different kinds of materials properties. All these DBs can be interrogated for discovering materials with desired properties. Furthermore, machine-learning models can be trained to predict the properties of other materials.
Many such DBs have appeared online (e.g., AFlow, COD, Materials Cloud, Materials Project, NOMAD, and OQMD, etc.). In most cases, a Representational State Transfer (REST) Application Program Interface (API) is available to interrogate the DB through scripts. However, till recently, it was only possible to query one DB at a time, and the APIs are very different from one DB to another.

Thanks to the OPTIMADE consortium (gathering all the key players involved in these different efforts), a common API was developed [https://arxiv.org/abs/2103.02068, submitted for publication in Sci. Data] 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, and from 2020-06-08 to 2020-06-12 (virtual event).

2 Major outcomes

Building on the results achieved in the previous OPTIMADE workshops and subsequent discussions on the OPTIMADE mailing list, the present workshop continued this effort. The objective was to make the various DBs interoperational on a broader range of properties through the development of the OPTIMADE API and its implementations. Furthermore, the workshop was meant to try to increase the extent of the OPTIMADE API to molecular dynamics.
The digital events comprised two plenary meetings per day (one in the morning and one in the morning) in order to gather as many people around the globe as possible (given the time difference), and small-group sessions in between and after the plenary meetings. The first plenary meeting 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. Most of the other plenary meetings consisted of longer presentations by members of other communities. Among others, talks were given by representatives of the new databases, who discussed how the OPTIMADE API could be useful for their databases (focusing on what is currently missing). Finally, some of the plenary meetings were dedicated to general discussions. One representative of the different small groups would present what had been achieved in their group and gather the opinion of the rest of the attendees. Then, depending on the results of the discussions, new groups would be formed in order either to continue to advance on the topic or to start a new topic. The small group sessions were dedicated to focused discussions, with the three most significant detailed below:

Tutorials

The tutorials focused on the OPTIMADE Python tools for which tutorials were first proposed to the newcomers, and then further improvements were made. All the talks and the tutorials were recorded and will be available through a dedicated OPTIMADE YouTube channel. The tutorials directly led to three databases asking for help to develop an OPTIMADE API interface, which will broaden the impact of the OPTIMADE API.

Molecular dynamics

The workshop was an ideal opportunity to bring together the postdoc, Johan Bergsma, tasked with developing the OPTIMADE API for application to molecular dynamics simulations, other OPTIMADE developers, and invited experts in molecular dynamics and bio-simulations. The outline specification developed will be a solid foundation for Johan Bergsma to define a full API over the months ahead.

Developments to the OPTIMADE API

The third session 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

The common OPTIMADE API will clearly benefit materials design by making it possible to interrogate all the databases using the same query. The OPTIMADE API will also contribute to making all these data FAIR (Findable, Accessible, Interoperable, and Reusable). The effort that has been started is therefore really important and could have a very high impact.
The OPTIMADE effort should definitely be continued, trying to involve as many important players as possible. For example, at the present CECAM workshop, we welcomed developers from the Computational 2D Materials Database (C2DB), the Open Reaction Database, the Quantum-chemical database for MOFs, the High Throughput Experimental Materials database, the Open Catalyst 2020 Dataset, Atomly.net, BioExcel, OpenKIM, ESPEI, and the Topological Material Database. 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

Strong links have also been established with the European Materials Modeling Council. They have a strong interest in the OPTIMADE API that is clearly in line with their objectives.
Various members of the OPTIMADE community participate in some of the boards of the European Materials Modeling Council. This opens some possible funding schemes at the European level. Indeed, the European Materials Modeling presently benefits from a Coordination and Support Action (CSA) from the EU, which started in November 2016. There are plans to submit a new proposal when an appropriate call is made. OPTIMADE will consider joining when this happens. More recently, OPTIMADE has also established different connections with the Materials Research Data Alliance (MaRDA) in the United States.

5 Will these developments bring societal benefits?

Materials design has clearly a great impact on society. Many new applications require specific materials with targeted properties. Furthermore, improving the efficiency of existing technologies (e.g., denser batteries, photovoltaics, faster computers, ...) also benefits from the design of materials with better properties.
The development of the OPTIMADE API contributes to making materials design easier. Indeed, the users will be able to interrogate many different databases with the same query. This will give them access to such more materials knowledge without the need to learn a different API for each database. Furthermore, machine learning methods can source information from multiple databases. 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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Excitonic and competing orders in low-dimensional materials

Location: Online meeting, hosted by CECAM-HQ
Webpage: https://www.cecam.org/workshop-details/21
June 21-23, 2021

1 State of the art

In the last thirty-six months, the field of the excitonic insulator has moved extremely fast.
This research has its origin in a heretic prediction formulated more than 50 years ago by a group of visionary physicists, including Leonid Keldysh and Walter Kohn [1-3]: If a narrow-gap semiconductor, or a semimetal with slightly overlapping conduction and valence bands, failed to fully screen the interaction among its intrinsic charge carriers, then excitons---electron-hole pairs bound together by Coulomb attraction---would spontaneously form. This would destabilize the ground state, leading to a reconstructed 'excitonic insulator'---a condensate of excitons at thermodynamic equilibrium. This chimeric phase shares fascinating similarities with the Bardeen-Cooper-Schrieffer superconductor: a distinctive broken symmetry, inherited by the exciton character, and collective modes of purely electronic origin. Its observation was deterred for many decades by the trade-off between competing effects in the semiconductor: as the size of the energy gap vanishes, favoring spontaneous exciton generation, the screening of the electron-hole interaction increases, decreasing in turn the exciton binding energy. Recently, mounting evidence [4-12] has been accumulating in low-dimensional materials, as they combine optimal band structures, poor screening behavior, truly long-ranged interactions, and giant excitonic effects (see also the list of recent literature maintained at www.nano.cnr.it/index.php?mod=men&id=196). This was the topic of our first Cecam Workshop in September 2018.
The last three years have witnessed mounting indications that the most promising excitonic insulator candidates are 2d materials. These candidates however exhibit also other kinds of order: a variety that includes topological insulators [4,13-19], ferroelectrics [19,20], unconventional superconductors [21-24], often depending on tiny variations of tunable parameters, such as doping, pressure, strain. This has introduced new urgent and far-reaching questions, concerning the role of excitonic correlations in a plethora of allegedly unrelated phenomena, whose interplay is just beginning to be explored. At the same time, the long-term challenge of establishing the excitonic insulator through the signatures of macroscopic quantum coherence is attracting renewed interest in this class of materials.
By collecting the key actors of theoretical and experimental research, who were spread among different communities, this workshop aimed at in-depth analysis of common themes and novel

challenges, both theoretical and computational, to progress our understanding of interacting systems in low dimensions.

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2 Major outcomes

The workshop put together the leading scientists in the field of the excitonic insulator (EI) by effectively mixing computational, theoretical and experimental talks, which could be broadly categorized into the following themes:
(i) new and old low-d materials as EI candidates (MacDonald, Varsano, Calandra, Codben, Georges, Da Como, Bechstedt);
(ii) C-based materials (Ilani, Behnia, Wirtz, Sorella);
(iii) light-driven excitonic coherence (Kaiser, Stefanucci, Littlewood, Latini);
(iv) competing/coexisting orders (Abbamonte, Kunes);
(v) fundamentals of excitons (Reining).
Such diversity reflected in lively discussions, with many questions and comments after each talk. A key outcome was the awareness that the field and the community of the EI is now established, with several candidate bulk materials presently under scrutiny as well as significant advances in the realization of novel 2d bilayers. A main (and basic) open question at this time remains the unambiguous identification of the EI phase in bulk systems, as the coupling between excitons and lattice is often so strong (as in Ta_2NiSe_5) to spoil the simple picture of exciton condensation. Therefore, in two very lively discussion sessions (chaired respectively by Peter Abbamonte and Allan MacDonald), participants struggled to agree on a rigorous definition of the EI phase as well as its clear-cut signatures.
It was tentatively agreed that the key concepts defining to the EI phase and its properties are as follows:

1. The exciton wave function, $< c^{+}v >_{EI}$, averaged over the EI ground state, breaks a crystal symmetry that is otherwise preserved in the “normal” phase (c and v are the annihilation operators of electrons filling in the conduction and valence band of an intrinsic semiconductor/semimetal, respectively). This definition may be an idealization for those real materials which only approximately exhibit the symmetry $< c^{+}v >_{normal} \sim 0$ in the normal phase (e.g., the interlayer coherence in bilayers is tunable in the normal phase). Furthermore, the symmetry breaking $< c^{+}v >$ may be due to non-excitonic channels, like e.g. electron-phonon interaction, spin-orbit coupling, other terms in the Hamiltonian, etc. In all these cases, the criterion reduces to a quantitative assessment of the excitonic contribution to the symmetry breaking as compared to other possible mechanisms.
2. Whereas the kind of symmetry breaking associated with the EI is obvious in bilayer systems (interlayer coherence), in bulk systems it may be either the translational or the point group symmetry of the crystal, or even time-reversal. In such a case, the EI picture applies only to a subset of possible situations, which fit the following scenario: the excitation energy of a bound state (exciton), present in the excitation spectrum below the electron-hole continuum, goes to zero as a function of an adiabatically tuned parameter (e.g. pressure, screening, temperature, etc.). Special considerations may apply to a BCS-like phase transition in which excitons do not exist above the critical temperature.
3. The existence of an acoustic-like collective excitation in the EI phase (excitonic sound), whose velocity is much higher than that of the ordinary sound---due to its purely electronic origin---may be regarded as a clear-cut signature of the EI.
4. More generally, experimental tests that can discriminate between the excitonic and phononic origin of the EI broken symmetry are highly desirable. This is especially urgent for excitons with finite center-of-mass momentum, since their condensation leads to a charge- or spin- density wave that may be confused with a Peierls-like distortion. The use of advanced ab initio tools to disclose the excitonic character of the phase transition was recognized as a breakthrough by the participants.

Finally, the possible manifestation of the macroscopic coherence of the EI phase, as well as the relation between the excitonic order and other types of order, e.g. those realized in topological insulators, magnetic materials, superconductors, remain totally open issues at this

time. The investigation of such fundamental questions, relevant to other fields such as high-Tc superconductivity, are still in their infancy.

3 Community needs

As the EI topic has started really exploding in the last three years, the workshop has been extremely successful in focusing theoretical, computational and experimental talks in order to outline the perspectives of simulations in this field. The participants have recognized this meeting, which is the second CECAM workshop covering exciton condensation, as an already established event that gathers the leading scientists working on the EI, as well as it outreaches other communities---including experimentalists. Given the urgency of improving networking within the new community, an obvious and immediate need is to consolidate the workshop series, by establishing CECAM as the home of a series of meetings on the EI topic. Furthermore, the participants have unanimously expressed the desire that the next event will take place in presence. This necessity is clearly demonstrated by the fact that the two online discussion sessions that have taken place during the workshop were actually insufficient to accommodate all expected contributions.
Concerning computational infrastructures, the workshop has clearly shown that advances in the understanding of real EI materials essentially depend on massive, first-principles state-of-the-art calculations, where screening and electron-hole interactions must be described with extreme accuracy to describe very delicate phenomena and energy balances. Code development has progressed rapidly, and it should be noted that most of the first-principle codes adopted in this workshop for DFT and beyond DFT calculations, as well as Quantum Monte Carlo (QMC), are developed and made available open source in Europe. In view of the rapid evolution of the HPC and the future exascale perspectives, a massive effort is needed to ensure that these codes remain at the forefront of computational technologies, and even contribute to co-designing future HPC systems. Some of the effort is supported in Europe through the H2020 CoEs, but this covers only a fraction of the actual needs, so it would be important to raise broader awareness in that direction. The same need of broader awareness exists for training and professional valorisation of the scientists who are investing not only in their research but also in the code evolution for the benefit of the whole community, and CECAM could certainly help in this direction. Finally, concerning HPC computer time, the amount that would be required is often much beyond the possibilities offered in Europe by PRACE and by the national HPC systems (this is e.g. the case for QMC, but also for some of the GW+BSE calculation campaigns). Computational work in this field will certainly provide eager users of the future EuroHPC architectures.

4 Funding

The research on the EI encompasses both fundamental and technological aspects. In this field the latter aspects are quite visionary, as they rely on the exploitation of macroscopic quantum coherence in real materials, e.g.: the occurrence of dissipationless current, the possibility of encoding information using electronic collective degrees of freedom, the ability to control superconductivity through excitons. Some of these long-term applications might fit in the European FET scheme, even the perspective of industrial interest at the moment is quite far. On the other hand, the core of the EI research has clearly a fundamental character, which may well fit curiosity-driven science programs such as the ERC scheme. It is worth stressing that the EI theme seems particularly fit to proposals combining theory and experiments together. The support of a workshop series on the EI topic at CECAM could certainly foster joint funding actions (given the online format of the workshop, funding actions were not discussed at this time).

5 Will these developments bring societal benefits?

The search for EI phases could lead to novel manifestations of macroscopic quantum coherence, which historically have always led to breakthrough applications. Indeed, exciton superfluidity would allow for the exploitation of quantum effects at temperatures much higher than those of the BCS state (since the exciton mass is much lighter than that of a Cooper pair). At variance with exciton-polaritons, where Bose-Einstein condensation occurs out of equilibrium, in the EI excitons would condense at equilibrium, which makes any achievement in this field extremely appealing. A deeper understanding of the proximity between the superconducting and the putative excitonic insulator phase, which is suspected to occur in 2d materials under present scrutiny (monolayer WTe_2, $TiSe_2$, MoS_2 under pressure) could enlighten the puzzle of high-T_c superconductivity.

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Memory effects in dynamical processes: theory and computational implementation

Location: Erwin Schroedinger International Institute for Mathematics and Physics (ESI)
Webpage: https://www.cecam.org/workshop-details/70
June 23-25, 2021

1 State of the art

If one develops a coarse-grained model of a complex system by systematically integrating out degrees of freedom, in general the resulting equations of motion are non-local in time. Hence Markovian models have shown their limitations when modeling complex many-body processes and there is now considerable research effort to go beyond the Markovian approximation. In this meeting speakers reviewed recent advancements in the study of coarse-grained models and they discussed the impact of memory effects on the system dynamics. New theoretical and numerical methods to analyze memory were presented and compared, such as recent advancements in time-dependent projection operator techniques and mode coupling theory. Also many practical examples from various fields of research were shown, e.g. biological reaction networks, phase transition kinetics, the physics of glasses and mechanical properties of soft materials. The topic was addressed by physicists, mathematicians and chemists and there was a lively interdisciplinary discourse.

2 Major outcomes

We had vivid discussions on the following topics:
Projection operator techniques: H. Meyer discussed the generalized Langevin equation in time-dependent contexts such as active matter and driven systems; B. Rothenberg, R. Vuilleumier and H. Vroylandt showed new approaches to reconstruct the noise history of a system and to propagate observables according to the orthogonal dynamics; F. Schmid showed recent work on Fluctuation-Dissipation relations far from equilibrium.
Numerical techniques: M. Hanke-Bourgeois introduced a new approach for computing a data-driven Markov model with extended variables. T. Franosch introduced a noise-cancelling algorithm that allows to study auto-correlation functions with high precision. V. Klippenstein presented a novel route for the parametrization of generalized Langevin thermostat models. K. Palacio-Rodriguez used transition path sampling trajectories to construct Langevin equations. F. Höfling discussed spectral approaches to memory.
Glasses: T. Voigtmann presented a recent theory to describe the dynamics of aging on the basis of microscopic approaches leading to self-consistent generalized Langevin equations for the non-stationary state.
Biological systems: P. Sollich analyzed networks in biological processes, like protein interaction and gene regulation networks, by means of a new net-work coarse-graining

technique. J. Yeomans discussed the dynamical changes from 2D layers to 3D structures in biofilm formation.
Materials: M. Müller presented a dynamic self-consistent field theory to predict the kinetics of structure formation in multicomponent polymer systems; S. Jabbari discussed mechanical memory as is seen in the Bauschinger effect in polymeric materials; S. Fielding discussed memory effects in ductile and brittle yielding in amorphous materials.
Other topics: A. Vulpiani discussed causation in relation to correlations and linear response theory; M. Schmidt gave an overview of an alternative route to non-equilibrium coarse-grained dynamics: Power Functional Theory; R. Wittkowski presented a field theory for soft-matter systems with two different time scales, which allows to obtain a phase field crystal model for polar (i.e., nonspherical or active) particles with translational and orientational memory; M. Dijkstra discussed how machine-learning can be used to find effective many-body potentials for colloidal systems; S. Bonella concluded the workshop with an analysis of generalized Langevin equations in the context of nuclear quantum effects.
There were new and exciting results in all contributions. As the field is young, there are many open questions, such as e.g. how to relate the equations of motion derived by projection operator techniques to those derived by Power Functional Theory, which in principle have to be equivalent, but have a different mathematical structure.

3 Community needs

Although the online format worked well, we realized once more that it is much more efficient to sit down and discuss science in presence (especially for explaining and rearranging equations or negotiating joint funding proposals). Thus, a follow-up workshop on the same topic would be very helpful. In this way, the ideas that were discussed during the past workshop would have some time to grow and could then be exchanged again.
To the best of our knowledge, none of the participants has issues with HPC resources. The codes to numerically study memory effects are usually written by the respective researchers (and their research group). What is appreciated by most of them is to share or get more data (simulation and experiment), in order to draw further conclusions by analysing larger/more data sets. Therefore, a joint meeting with experimentalists might also be an interesting future option.
Data storage and management is, of course, a general problem in the simulation community. Services for research data management would be appreciated.

4 Funding

A typical national funding scheme in Germany is the DFG "Normalverfahren", an open call where researchers apply for funding for a specific, smaller scale project. Most of the German participants of the meeting as well as two of the organizers have got funding from this source. Larger scale formats in Germany are the "research unit", the "priority program" and the "collaborative research center". T. Schilling is involved in a DFG research unit which works on a topic closely related to the workshop and which has just received funding.
Typical European funding could be acquired through a research training network. Maybe an ERC Synergy grant would be an option, too. However, these options were not discussed at the meeting. The online format is not suited to the type of negotiation that proceeds writing large scale collaborative proposals.

5 Will these developments bring societal benefits?

Studies of systems in their equilibrium state are the first steps to understand and control their behavior. However, in everyday life and in operation, systems are usually not in their equilibrium state. Responsive ("smart") materials and biological tissues are typical complex systems out of thermal equilibrium. If one develops models to predict the properties of these materials, one inevitably encounters memory effects. Typically, the effective equations of motion are stochastic integro-differential equations, which are challenging to handle. Thus, the workshop, which was predominantly theoretical/mathematical in nature (and hence has not generated direct societal benefits) has the potential to generate new computational and numerical approaches to aid the modeling and design of modern materials. As all contributors to the workshop also work in teaching, there is the additional benefit that the latest advances in method development will very soon enter the curricula of students.

6 Participant list

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Local structure meets machine learning in soft matter systems

Location: Online event - hosted by CECAM-HQ
Webpage: https://www.cecam.org/workshop-details/1028
June 28-July 1, 2021

1 State of the art

Many soft-matter systems, such as fluids and glasses, are clearly disordered materials, lacking the long-ranged translational order of crystals. However, hiding in the apparent structural disorder of fluids can be an impressive degree of local order which has profound effects on the behavior of the system, especially in the contexts of crystal nucleation and glassy dynamics. This workshop explored the use of machine learning (ML) techniques to these areas of soft matter.
In crystal nucleation research, system- and phase- specific algorithms that classify local structure have traditionally been used to separate particles that have crystallized from those that remain in the fluid phase [1, 2]. These algorithms, used to probe the structure and evolution of the crystal nucleus [3, 4, 5], were typically designed by trial and error, and are system and structure specific. Inspired by the successful application of ML to efficiently approximate molecular interaction potentials [6, 7, 8, 9, 10], ML has already proven highly successful in the development of new order parameters, allowing one to train algorithms to recognize specific crystalline environments [11, 12], and even to autonomously pick out groups of particles in different phases [13, 14].
In the study of the glass transition, the link between local structure and dynamics is a long-standing question [15, 16, 17]. While structure clearly plays a key role in glassy relaxation [18, 19, 20, 21, 22,23], pinpointing a structural explanation for e.g. the length scale of dynamical heterogeneities remains a challenge [24]. To help address this, many methods have been developed to detect different local motifs in glassy fluids which are linked to dynamics [17, 25, 26]. Here, ML has made a significant impact, leading to new algorithms that can be trained to identify soft spots in glasses [27, 28, 29, 30], or even spot structural variations in glasses without relying on dynamical information [31].

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2 Major outcomes

Broadly, the workshop was focused on the discussion of using machine learning techniques to capture, classify, and analyze local structure in a variety of many-body systems. The discussion can be roughly broken down into three topics:
1) Machine learning techniques for use in approximating force fields and interaction potentials. A core question when using ML for the description of many-body systems is how we should capture the local structure of particles in a set of numbers that can be processed by an ML algorithm. A natural source of inspiration is the field of molecular simulation, where accurate simulations require an accurate determination of the forces or energy of a system based on the particle configuration. Machine-learned force fields and potentials now lie at the forefront of this field, and tremendous work has been done to find efficient sets of descriptors of the structure of systems. The workshop included talks from several experts from this field, discussing new ways to best describe local structure, as well as the optimal use of ML to use with these descriptors. A tentative consensus from these discussions is that good descriptors should ideally be smooth and mathematically complete, while respecting the known symmetries (e.g. translation/rotation) of the underlying system. For the choice of ML algorithms, there is a strong preference for linear or kernel methods over more complex methods (e.g. deep learning), due to the fact that these ``simple'' methods tend to require less data for training and extrapolate much better to new configurations outside of the training set.
2) Prediction of glassy dynamics using machine learning. One area in soft matter where machine learning techniques have drawn particular attention is the exploration of the link

between structure and dynamics in supercooled fluids. The workshop included a number of speakers who are experts in this field, leading to interesting discussions of both the non-ML and ML techniques currently being used to understand how the dynamics of glassy fluids are impacted by their local structure. One clearly intriguing observation is the fact that both supervised and unsupervised ML techniques can now make good predictions about the future dynamics of a glassy configuration. However, an open question is how to extract physical insights from these predictions, especially when complex ML methods are used. Additionally, as most of these approaches focus on very local structural features, an interesting point raised during the workshop is that many theories of the glass transition suggest that this highly local structure may not be sufficient for a full understanding of glassy dynamics. Hence, it is suggested that ways should be found to include data spanning multiple length scales.

3) Machine learning for the study of self-assembly. When studying self-assembly processes, where particles in multiple distinct phases coexist, it is vital to be able to readily distinguish between particles in different phases. ML offers a number of possibilities here. Presentations at the workshop highlighted ML techniques for examining crystal nuclei during the nucleation process and for analyzing crystal structures. These methods can typically easily be trained to identify crystal structures, and often provide more accurate results than hand-crafted order parameters. However, these approaches rely on some set of local descriptors, which should be chosen to capture the core features of the phases in question. Hence, open challenges still exist for capturing the behavior of *e.g.* quasicrystals or chiral liquid crystals, where longer-ranged information may be required to correctly classify particles. Another highlight was the use of ML-based order parameters with advanced optimization algorithms in order to `inverse-design' particle interactions that lead to the self-assembly of a desired phase.

3 Community needs

The scientific community that touches on ML methods and soft matter is quite varied, and the computational needs of the individual researchers will vary depending on the scope of their research. Some of the projects discussed at the workshop require significant computing power including large HPC clusters. These are needed *e.g.* for the purpose of obtaining training data that is used for training ML algorithms (often from simulations of glassy systems, quantum DFT calculations, etc.). Additionally, many ML techniques benefit strongly from GPU-based computing for their training, and hence small GPU clusters are often beneficial. On the other hand, a number of presentations at the workshop highlighted the benefits of ML techniques with a relatively low computational cost, and in many cases the training time for the problems discussed was negligible.

The future organization of events that allow for networking between ML users in different fields or different areas of soft matter will certainly be beneficial: developments in this field are rapid, and there many possible techniques to compare and contrast. Additionally, it is extremely helpful to bring soft-matter researchers with experience in ML into contact with researchers who may have (experimental or simulation) data that could benefit from some form of ML analysis, possibly sparking new collaborations. Likely, it would be good to encourage the organization of multiple future workshops along these lines, perhaps each focusing on a different aspect of soft matter where ML can play a role.

Given the success of this online workshop which drew participants from all over the world, a series of small online workshops/updates on specialized aspects culminating in a final in person larger workshop might be worth considering, perhaps with an attached school to train young researchers in a domain that is still not present in all the curricula.

4 Funding

Typical funding channels for this area include the standard local, national, or European funding schemes for research grants. Additionally, the application of machine learning is a highly active field with significant interest from both national funding agencies as well as industry. As a result, a number of target proposal calls aimed at appropriate topics such as “Artificial Intelligence”, “Data Science”, or “Machine Learning” have appeared in different countries over recent years, and these are likely to continue to appear in the future. No joint research proposals were discussed during the meeting in the group discussions, although these may have been discussed in private conversations.

5 Will these developments bring societal benefits?

Machine learning is a rapidly expanding field with increasingly many applications being used on a daily basis. This workshop was aimed at sharing and spreading knowledge on the application of ML techniques in soft-matter systems. As these techniques seem to be highly useful in a variety of ways, spreading their use in the scientific community has a number of potential long-term benefits. First, it has the potential of enhancing the research done in the field of soft matter, which has its own applications and benefits depending on the topic studied (e.g. development of new sustainable materials, study of crystallization and vitrification which may impact the food or pharmaceutical industry, etc.). Second, usage of ML techniques in new academic fields may also lead to new insights into the design and optimization of ML algorithms, which may flow back into other fields of science and industry. Third, students using ML are trained in the use of new techniques that are highly sought-after by potential future employers, including industry.

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Multi-approach modeling of alloy nanoparticles: from non-equilibrium synthesis to structural and functional properties

Location: On-line event
Webpage: https://www.cecam.org/workshop-details/1068
July 7-9, 2021

1 State of the art

Strong advancements were made in modelling of alloy nanocrystals. Similarly, new techniques emerged for the realization of non-equilibrium alloys in a relatively easy and scalable fashion, such as the laser synthesis and processing of colloids, which permitted the investigation of new phenomena and advanced technological applications. [1,2,3,4]
Often, progresses in the field are allowed by the combination of theoretical outcomes with synthetic and experimental endeavours. [2,5,6,7,8]
Nonetheless, there is still a lot to understand about how to predict structural motifs arising in the metastable assembly of atomic constituents in real conditions. Accurate modelling and prediction of the physical properties and chemical characters enabled by the organization in non-equilibrium phases also remain challenging. Improving the theoretical description of nanocrystals formation during the laser-assisted synthesis is crucial as well for expanding the range of accessible nanoalloys.
This workshop has successfully put together experts from the communities active in computational modelling of nanocrystals formation, structure and properties as well as from

the laser synthesis and processing of non-equilibrium nanoalloys. The transversal competences collected in the workshop allowed to highlight the edge issues in modelling.
The workshop succeeded in bringing together a balanced group of participants from both the theoretical and experimental communities, which have been allowed to discuss about the key points of the experimental validation of theoretical predictions.
The emergence and exchange of ideas and constructive criticism have been stimulated by interactive platforms made available to participants throughout all the workshop (Gather platform) described below.
In addition to key references provided below, the book of abstracts offers a panorama of the recent significant developments of the field.

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2 Major outcomes

The workshop collected ca.45 participants with 25 talks and 12 poster presentations. 50% of participants were early-stage researchers and 50% senior. They were from 15 different countries and 33 different institutions (as first affiliation). 40% of participants were experimentalists and 60% theoreticians.
All the participants were gathered together with the "Gather Metropolis" on line platform, which provides the maximum possibilities for one-to-one discussion and a real-conference-like virtual experience. We received several enthusiastic feedbacks about workshop topics and organization from participants.
The main sessions were about Laser Synthesis and Formation of Nanoalloys (I, II); Nanoalloys Structure and Properties (I, II, III); Poster session and an on-line Demonstration Session with Live Laser Synthesis, Live Cavitation Bubble Imaging, Live Atomistic Modelling (DFT and Molecular Dynamics).
The titles and abstracts of the talk and poster contributions have been analysed to extract the main topics of the workshop, which are resumed as following:
nanoalloys; core shell nanoparticles; laser ablation in liquid and in air; molecular dynamics; chemical ordering in nanoalloys; density functional theory; non equilibrium nanoalloys; plasmonic properties; lithium metal; catalytic activity; montecarlo method; atomistic simulations; control over composition and structure; janus like structures; cohesive energy in alloys; laser irradiation in liquid; cooling rate; crystal structure; gas phase synthesis; platinum alloys, gold alloys, silver alloys.
The Workshop conclusions are:

- The worlds of nanoalloys (laser) synthesis and modelling have met for the first time in a dedicated "house"

- Laser synthesis in liquid is now recognized as a powerful technique for accessing nanoalloys, also from the outstanding and active community of theoreticians working on nanoalloys
- Senior and early-stage/young researchers exposed their work and discussed together on the above topics, ensuring a tans-generational scientific activity on the topic
- Research edges on the two fronts emerged as following:
    - Experimental side:
        - Control of nanoalloys characteristics with synthesis (composition, size, structure)
        - Probing ultrafast formation processes in real time to support modelling and understand key aspects still obscure
        - Accessing the chemistry in addition to the physics of the process with experimental techniques
        - Need for single particle experiments to avoid average over ensemble of nanoalloys with different features in terms of size and composition
    - Modelling/theoretical:
        - Convergence of different approaches and benchmarking of the best one is foreseen for the future, given the various different theoretical methodologies adopted for specific systems
        - Longer timescales required for molecular dynamics is key to understand how to control nanoalloy synthetic parameters
        - Effect of medium (liquid, gas, vacuum) should be implemented more to fill the gap with experiment reality
        - Addressing the chemical reactivity in simple and complex environments is the most challenging and unfilled effort at the moment.

A special issue has been connected to the workshop (in ChemPhysChem, Wiley) to follow up the presented research with scientific publications.

3 Community needs

The modeling activity on nanoalloys covers a wide range of size and time scales exploiting a variety of computational approaches: Density Functional Theory (DFT) and Time-Dependent (TD) DFT, Molecular Dynamics (MD), Monte Carlo (MC) and Kintetic (K) MC methods. DFT and TDDFT provide accurate results, but are typically restricted to nanoclusters or bulk alloys, whereas MD and KMC methods allow treating larger nanoparticles and longer timescales, but at a lower level of accuracy. A better integration of ab-initio and classical methods is desirable to treat realistic systems in a proper multiscale approach.
Computational outputs are produced using a number of commercial (VASP, MolCAS, ...) and open-source (Quantum-ESPRESSO, abinit, LAMMPS, ...) software, locally modified versions of them, homemade MC and KMC codes and analysis tools. The community is also active in developing new approaches to simulate inherently complex phenomena such as alloy nanoparticles formation under laser irradiation, time-evolution of non-equilibrium nanoalloys, optical responses and surface reactivity.
Calculations in this field demand a faily large number of resources to treat realistic systems and access to large national and European HPC infrastructures is of pivotal importance.
Networking between experimentalists and theoreticians is required also in the future to have a fruitful flux of information between the two fields, towards real world needs, especially in terms of current global challenges in catalysis, optics, information technologies, quantum technologies and medicine.
A series of CECAM workshops specifically addressing this topic is not required at the moment, because the size of the community is limited so ca. 100 scientists in Europe and ca. 200-300 in the world, that better meet annually or semiannually with standard CECAM workshops and

other initiatives (such as the French cluster on nanoalloys and the annual conference on nanoalloys).

4 Funding

Typical funding channels at the moment are national funding and international networking funding (COST actions, French research networking fundings). Joint applications to European projects have not been specifically discussed in the Workshop because of a lack of basic research support from the European Union. Access to specific funding is possible only when the theoretical and experimental scientists active in nanoalloys join on a topic with high chance of real application in catalysis optics quantum technologies or nanomedicine, which is indeed possible between some of the participants to the workshop in next months, taking advantage of calls about development of new catalysts or new nanomedicines.

5 Will these developments bring societal benefits?

Nanoalloys are at the center of several key technology problems of our contemporary society. The principal one is the development of energy efficient and sustainable processes for energy conversion, accumulation and storage, as well as for fine chemicals production. Nanoalloys will be even more embedded in any effort of this type in the future and are a key component of research projects and applications for all calls (national and European) about these topics. The sustainability and circular economy are key topics of Horizon Europe and the research groups participating to the workshop already are active in the field. The possibility of connecting even more the experimental and theoretical worlds offered by this workshop will clearly benefit the possibility to participate at European projects with high competitive and state-of-the-art proposals.
Nanoalloys are emerging also as biodegradable components for the development of novel anticancer drug design. EIC Open calls allows establishing networks on new nanomedicine developments, that are in the agenda of some of the groups participating to the workshop.

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CHARMM-GUI CECAM School

Location: CECAM-FR-GSO
Webpage: https://www.cecam.org/workshop-details/1072
September 5-10, 2021

1 State of the art

Molecular modeling and simulation play important roles in biomedical and material research as they provide molecular-level insight into the underlying mechanisms of molecular functions that are difficult to elucidate only with experiments. Current state-of-the-art molecular simulations and force fields (FFs) have been broadly tested and are sufficiently accurate to reproduce experimental results and guide new experiments with testable hypotheses.

However, as larger spatial scales, longer time scales, and higher levels of realism become possible and necessary, the generation of realistic molecular systems becomes a major obstacle, even for the experts. In addition to the reproducibility of simulation outcomes, the grand challenges are: how to effectively use simulation techniques for practical problems (for experts) and how to lower the entry barrier for their use (for non-experts). CHARMM-GUI (http://www.charmm-gui.org) has, in the past 15 years, been trying to address these challenges by providing a web-based graphical user interface (GUI) to interactively build complex biomolecular systems and prepare input files for state-of-the-art molecular simulations using CHARMM, NAMD, GROMACS, AMBER, GENESIS, TINKER, LAMMPS, Desmond, and OpenMM.

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2 Major outcomes

The school combined a detailed overview of the capabilities of CHARMM-GUI with practical exercises. In addition to the standard modules, ***PDB Reader***, ***Solution Builder*** and ***Membrane Builder***, new functionality was presented and an outlook to almost ready tools given.
Recently integrated modules into CHARMM-GUI include ***Glycolipid Modeler*** for the generation of various glycolipid structures; ***LPS Modeler*** to generate lipopolysaccharide (LPS) structures from various Gram-negative bacteria for bacterial outer membrane (OM) system building; ***Nanomaterial Modeler*** and ***Polymer Builder*** for modeling and simulation of various nanomaterial and polymer systems. In addition, for wide applications of free energy simulations (FES) to drug discovery in academia, we just released ***Free Energy Calculator*** with four submodules: *Absolute Ligand Binder* (for absolute protein-ligand binding FES), *Relative Ligand Binder* (for relative protein-ligand binding FES), *Absolute Ligand Solvator* (for absolute ligand solvation FES), and *Relative Ligand Solvator* (for relative ligand solvation FES).
These modules were presented and demonstrated both in lectures, as well as the practicals. Additional lectures gave a detailed introduction to force fields (J.B.Klauda) and the theoretical foundations of minimization and molecular dynamics (E.Paci). The video tutorials from the CHARMM-GUI website were the starting point for the practical exercises, which were held in two sessions: A morning block for participants from Europe and Asia, and an evening block for students from the Americas. Through Zoom breakout rooms and the feedback form on the CHARMM-GUI website, students could get in touch with individual CHARMM-GUI developers and obtain help and feedback.
Participants in the school acquired a thorough refresher course in the theoretical underpinnings of biomolecular MD. They acquainted themselves with the full range of functionality available in CHARMM-GUI and learned how to make efficient use of the tools offered.
Last but not least, the slide decks and recordings for all lectures held in the course of the school are available to the public through the CECAM website of the school. These constitute an important resource, complementing the documentation available at the CHARMM-GUI website.

3 Community needs

Since this was a school, not a workshop, training early-stage researchers in the efficient use of CHARMM-GUI, the questions in this section are not directly applicable. However, the need to carry out the school in a hybrid fashion demonstrated the challenges and needs arising in such a format.
Given the uncertainty of the Covid-19 situation, we had to restrict onsite participation to a small number; in the end 20 students participated in Toulouse. For online attendance we accepted over 200 applicants. The main tool used, Zoom, made available through EPFL Lausanne, worked flawlessly, but it cannot fully replace personal interaction. CHARMM-GUI has a contact form to report problems; this proved extremely helpful to address difficulties students had during the practical exercises. Ideally, a room for workshops and schools held in a hybrid fashion should have the technical means that presentations held by speakers *onsite* can be given "traditionally", with the slides and the stream of a camera on the podium being transmitted to online participants. Similarly, questions and lectures from *online* participants/speakers should be presented via a projector and a sound system to *onsite* participants in the workshop room, *without* causing feedback loops.

4 Funding

Not directly applicable since this was a school, and not a workshop. However, the skills and competences taught in the course of the school will be helpful for scientists in the early stages of their career to formulate concisely how simulation techniques can be applied to the study of biomolecular system. This should prove useful for writing grant applications, either to funding agencies in their respective home countries, or to, e.g., EU funding sources (Horizon 2020, etc.).
Among the speakers present onsite, joint research and, hence, the possibility of joint research proposals was discussed.

5 Will these developments bring societal benefits?

The spread of the SARS-CoV-2 virus has caused an urgent need for the rapid development of vaccines, drugs and a deeper understanding of the infection mechanism to counter the adverse effects of this pandemic on societies worldwide. The role of molecular dynamics simulations and methods derived from it is still rapidly increasing. CHARMM-GUI is an essential tool for the correct setup of such calculations; its application to the study of SARS-CoV-2 proteins is amply demonstrated (http://www.charmm-gui.org/docs/archive/covid19). Used correctly, CHARMM-GUI frees the user from having to worry about routine aspects and tedious details, and makes it possible to focus on the physico-chemical and biological aspects relevant to the system to be studied. This school helped train well over a hundred scientists in the early stages of their career, and we hope that it will prepare them to contribute to research against health threats, such as Covid-19, now and in the future. We expect them to carry the knowledge and expertise gained back to their home countries and institutions, serving as multiplicators.

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Nanopore Translocation and Nanochannel Confined Biopolymers: bridging theory and experiments

Location: CECAM-IT-SISSA-SNS
Webpage: https://www.cecam.org/workshop-details/41
September 8-10, 2021

1 State of the art

The goal of the workshop, as stated in the original proposal, was to bring together researchers with diverse backgrounds, from physics and chemistry to molecular biology, and engineering, to discuss topics central to the behavior of biopolymers driven through nanopores or confined in nanochannels.
Since its discovery about three decades ago nanopore sensing has remained an active field of research and various research avenues have witnessed spectacular growth in the last 5 years, as reflected in the order of magnitude increase in several publications, largely due to the advancement of single-molecule manipulation techniques that have provided unprecedented quantitative insight into the ubiquity of various forms of pore translocation or channel-like confinement in biological contexts, such as, during RNA transcription, mRNA translation and degradation of ubiquitinated proteins by the proteasome. The wealth of available experimental data has, in turn, spurred much activity on the theoretical and modeling side aimed at understanding the general physical mechanisms that underpin the observed phenomenology, as well as understanding their biological implications.
Taking into account these premises, we planned the workshop with a balance between theorists involved in modeling and simulations of the aforementioned systems and experimentalists.
The covered pivotal topics were two: (i) translocation through a multi-nanopore system of different designs and (ii) confinement of biopolymers in channels whose dimensions are much smaller compared to the length of the biopolymer. These focus areas were selected for their general relevance and broader implications. For the former, we recall that by capturing and pulling DNAs and proteins through nanopores and straightening them in nanochannels, it is now possible to sequence a single human genome without amplification. It is believed that nano-pore and nano-channel-based technologies will have a major impact on human health and disease, such as cancer, with accurate, albeit cheaper, and faster routine desktop analyses of the human genome. For instance, the emerging field of protein sequencing by nanopore opens the possibility to profile gene expression at a single molecule level.
For the second, instead, we note the ongoing debate in the community of polymer theorists regarding the existence of previously unforeseen metric scaling regimes that bridge between the two classic ones named after de Gennes and Odijk. The richer than expected theoretical background poses more challenges for interpreting experimental data and begs to address by simulations large segments of channel-confined genomic DNA for further numerical expediency and a new outlook on coarse-graining. Recent developments in self-consistent field theories of confined polymers may provide clues for bridging experimental and simulation length and time scales. Deviations from ideal conditions in experiments, such as the presence of fluorescent dyes, charges at the channel surfaces, etc. need to be incorporated in theory and modeling.

2 Major outcomes

The workshop was held at SISSA from Sept. 8 to Sept. 10, 2021 in hybrid mode. The participants were 75 in total (35 from Europe) and 32 of them attended the activity on site. The workshop was attended with 95% of the invited speakers as in the original plan.

During the three-day activity there were 29 invited talks (half on site and half remote), which were broadcasted over Zoom. An online poster session on the gather.town platform was also kept open during the activity and also afterwards.
The talks spanned a broad range of experimental, modeling and simulation, and theoretical topics. While some of these were directed towards the fundamental understanding, some of them address technical challenges to increase the resolution and the speed of the reads. The breadth of the topics and the depth and subtleties involved in various subtopics much exceeded the original expectation of the organizers. Both solid-state nanopores and different biological nanopores were discussed.
Theoretical ideas for the tension propagation theory, topological friction, dynamics of confined biopolymers as a function of the equilibrium phase diagram were presented. These theoretical ideas were used to explain experimental findings. Various types of numerical work from all-atom to coarse-grained description were presented. Translocation in presence of active rods and the idea of stochastic resonance was explored indicating their relevance in biological processes. The inclusion of co-ions and counter-ions were discussed in mesoscopic modeling using lattice-Boltzmann fluid. These theoretical ideas and modeling work were very much appreciated by the experimentalists and opened the door for joint collaborative work. On every occasion, there was quite a lot of discussion during the break which included remote participants also.
A broad spectrum of experiments was presented where nanopores were used to analyze and develop a physical understanding of how biological entities, such as how an enzyme works, use nanopores in mapping the energy landscape of the important protein – kinase and decoding digital information in biological macromolecules are to mention a few.
There were several talks on the extension of a solitary solid-state nanopore to multi nanopore systems – active control logic to scan a dsDNA multiple times, engineering of an adjustable two-pore system for ion transport, etc.
A good number of talks also covered the application of engineered biological nanopores, such as, to use of nanopores for peptide sequencing and size selectivity of large macromolecules, such as PEG, and small molecules, such as antibiotics and amino acids.
Overall, the organizers felt that there was quite a lot of synergy among these talks and there was always a constant exchange of ideas and discussions during the meeting, during the breaks, lunch, and dinner. The remote participants were also active in asking questions during the sessions.

3 Community needs

The fundamental tenet of nanopore sequencing is to extract the physico-chemical characteristics of the translocated molecule from the current blockade data only. Despite much success with nanopore technology, this is still a formidable task with challenges to overcome. Experimentalists have used machine learning algorithms to interpret the data. However, a large fraction of the simulation and modeling is done with coarse-grained models with implicit solvent for higher speed and longer chains. Extracting current blockade data directly from such simulation by design is not possible. On the numerical side, a much-debated aspect was the treatment of electrostatic effects in simulations of translocating polyelectrolytes. From the several discussions it became apparent that there is still a chasm between the atomistic simulations with explicit counterions, which yield accurate results albeit for relatively short timescales, and coarse-grained models, which can cover much longer timescales at the expense of using mean-field or other simplified treatments of electrostatic screening and condensation effects.
It will be worthwhile to address both issues in future workshops. If possible, one can foster collaborative numerical work through one or more CECAM nodes. A next CECAM workshop, presumably in two years in Lyon, can focus on this important topic "Bridging length scales from all-atom to coarse-grained models in nanopore simulation and theory" to decipher current

blockade data efficiently and improve the numerical treatment of electrostatic effects. Close collaboration with experimentalists will be much needed for the success of this topic.

4 Funding

This workshop was partly funded by a private enterprise, Elements sarl. This is a concrete indicator that more industries and manufacturers of lab equipment for nanopore sensing devices could be involved as external funders of activities on this topic.
The workshop itself, with the numerous formal and informal discussions – especially those that happened on site – opened new possibilities for collaborative research and thus of prospective funding. The possibilities of joint research proposals were discussed in small groups. One possibility for Global and USA collaboration is through NIH who funds proposals with Co-I all around the world. This will be an avenue worth exploring for nanopore-related work due to its direct connection to human health and disease. Some of the participants share such grants and more NIH grants are likely to be written from the ideas discussed in this workshop. Another funding avenue that naturally emerged was with the Human Frontiers Science Program, that is particularly geared towards projects involving teams across different continents and aimed at problems of relevance for biological systems.

5 Will these developments bring societal benefits?

Because of the extremely high sensitivity of nanopores, able to detect tiny molecules as well small changes in volumes, the science of nanopores is growing rapidly, embracing the technology. The workshop provided an excellent platform for future initiatives in many ways as follows:

- During discussions on various occasions (break, lunch, dinner) small groups discussed collaboration among various participants in the form of short-term mutual visits.
- Nanopore sensing is a hot topic for venture capitalists, regular private patrons, industrial and federal funding opportunities due to its immense prospect for transforming the existing today's medical diagnosis platform. Low cost, more accurate, and rapid diagnosis are the vision of the next decade which will bring transcendental changes across the globe. Early and accurate prognosis of the tests will reduce the number of deaths, mitigate suffering, and provide low-cost affordable treatment plans. Some of the talks in the workshop were directly targeted towards this vision.
- These initiatives will be sustained for a few decades as fundamental research will bring in new information to make these platforms better.
- In addition to the philanthropic sponsors in each country, agencies such a Human Frontiers Science Program, National Human Genome Research Institute (NHGRI), National Cancer Institute at the NIH will honor proposals with global partners.
- Currently the focus is more on sequencing and genome maps, however with time nanopore work will provide a transcendental change for molecular biology, such as piecewise physical understanding of post-translational modification of proteins, molecular motors, stochastic resonance in molecular biology. The application of this fundamental research in drug design will open doors for more industrial partnerships.

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Second discussion Meeting on Quantum Crystallography: expectations and reality

Location: ONLINE
Webpage: https://www.cecam.org/workshop-details/1051
September 9-12, 2021

1 State of the art

Quantum Crystallography is an emerging research area that aim at investigating a large spectrum of quantum phenomena occurring in crystals. They range from the interaction of crystalline materials with the electromagnetic radiation to quantum mechanics-based properties of materials. This field is witnessing a constant expansion (a consequent broadening of its scope) as also testified by the organization of the first Erice School of Crystallography dedicated to this subject (2018), by the previous edition of the CECAM discussion meeting (2017) and by the increasing number of symposia and presentations at international crystallographic conferences.
Originally the focus of quantum crystallography was mainly on electron density and derived electrostatic properties. However, a major goal has always been also the calculation of wavefunctions using experimental observables, in particular X-ray diffraction intensities. In fact, this would allow the exhaustive determination of all quantum mechanical properties for a crystal under investigation. This has become possible in the last 20 years thanks to the technique devised by Jayatilaka and coworkers. Many similar methods were developed and are continuously improved together with alternative approaches to reach the same goal. Often the interest is on the constituents of a crystal (such as molecules in crystals), thus making quantum crystallography a natural partner of computational quantum chemistry. Methods based on the direct determination of the electron density also evolved significantly even

allowing to distinguish tiny polarizations of core electrons. Moreover, by coupling charge and spin models and using an appropriate mixing of experimental information, mappings of spin-resolved electron densities were also possible. Finally, the investigation of the quantum behavior of materials at extreme conditions or under particular stimuli is of increasing interest within the scope of Quantum Crystallography.

2 Major outcomes

Three were the main goals of the meeting: 1) presenting the most recent results and developments within the quantum crystallography community; 2) establishing and reinforcing the collaboration with scientific communities working in close research fields; 3) discussing hot-topics and problems of the modern quantum crystallography. These objectives were inter-related and fully achieved during the four days of the workshop, characterized by a very active and vivid participation during all the sessions (especially during those explicitly dedicated to discussion).

Concerning the first goal, almost all the different traditional topics of quantum crystallography were quite fairly represented during the 27 short (20-minute) talks that were given by the invited speakers: recent advances in quantum chemical topology, modeling of crystal field effects and polarizabilities, new developments and applications of the traditional multipole model techniques, modeling of thermal motion in charge density studies, improvements in software for periodic solid-state calculations, experimental and theoretical modeling of Compton scattering, strategies for refining wavefunctions and density matrices from experimental scattering/diffraction data, recent advancements in Hirshfeld atom refinement, experimental determination of transient (time-dependent) charge densities, treatment of disorder and polymorphism, extension of quantum crystallography techniques to the modeling of electron diffraction data, application of quantum crystallography strategies to the refinement of protein crystal structures. These topics were also covered by most of the 17 posters that were presented during the workshop (split in two poster sessions).

The second objective was reached thanks to the invitation of four top-scientists working in research fields very close to quantum crystallography, but till now only sporadically in contact with the community. They gave longer (40-minute) talks on the following topics: Van-der-Waals quantum effects in molecular crystals (Alexandre Tkatchenko), charge density investigation by electron diffraction (Lukas Palatinus), development of new quantum methodologies for the study of complex system (Jason Goodpaster), recent advances and accomplishments through the X-ray free electron laser (X-FEL) technique (Peter Weber). They very clearly illustrated the possible connection between their respective areas of interest and quantum crystallography. The success of these presentations was also evident from the long and vivid discussions that followed each of the four presentations.

Finally, to achieve the third goal, two round-table discussions were organized (each of them subdivided into an evening and a morning session). As the discussions that followed each talk, they were highly participated and different themes and problems were tackled. Among them we can mention: i) the real meaning and usefulness of wavefunctions/orbitals resulting from X-ray restrained wavefunction calculations; ii) the possible combination of experimental information resulting from spectroscopy with the one obtained from diffraction experiments; iii) state of the art and future of the traditional multipolar model techniques; iv) soundness of quantum crystallographic methods with respect to recent advancements in electron crystallography, X-FEL, time-resolved experiments; v) usefulness of quantum crystallography in the development of technological applications.

3 Community needs

The previously mentioned interplay of the community with other scientific fields reflects the needs to “establish quantum crystallography”. This means finding the ideal locus at the interface between quantum physics/chemistry and crystallography. This also implies a clear definition of the research objects in quantum crystallography, of the existing and the potentially available methods and techniques, and of the neighboring fields with which to interact. Having this in mind, the short lectures of scientists active in the quantum crystallography community presented the state of the art of their research activities, while the invited talks given by top-scientists from outside the community helped drawing a better definition of the surrounding scientific areas and enabled creating new scientific connections. In the future, further tightening these scientific bonds through continuous interactions/contaminations will be essential, particularly with the quantum chemistry community.
During the meeting we also discussed the possibility of creating a common platform to interface existing and future (multi-purpose) software of quantum crystallography. This option might become reality soon thanks to a UK funding opportunity (see the next point for other details).
Moreover, given the success of this 2nd discussion meeting on Quantum Crystallography, the possibility to start a series of these CECAM workshops was enthusiastically considered by all the participants. Some scientists already exhibited their willingness in organizing the next meeting. The possible main topic of the next workshop was also discussed, with a focus on software development being an option.
Finally, a need of standardization in quantum crystallography was envisaged. This implies standardization of the language (like definitions, symbols, etc.), of the quantities to share for interfacing different software, and of the expected results from each technique and methodology.

4 Funding

Although this meeting was organized at zero costs and therefore without the need to raise funding, new and efficient ways to obtain grants for the discussed research activity are necessary. As anticipated above, a UK national funding scheme will be explored to finance a project aiming at the creation of a common workbench for current and future quantum crystallography software. This possibility was discussed during the meeting and, although the project (if funded) will be overseen by only two scientists based in UK (Simon Coles and Horst Puschmann), the whole quantum crystallography community will be involved.
The possible future interactions between the quantum crystallography community and other neighbor research areas (e.g., quantum chemistry, X-FEL, electron crystallography) will also naturally lead to the submission of joint proposals in upcoming calls (e.g., ERC synergy grants, COST actions, bilateral programs between national research agencies).

5 Will these developments bring societal benefits?

Although some of the goals targeted by quantum crystallography are mainly of academic interest, the existing quantum crystallographic techniques and the related software are already mature enough to answer challenging research questions in different research fields, ranging from materials science to drug design.
Just to give an enlightening example, the traditional multipolar model technique for the reconstruction of experimental electron densities from high-resolution X-ray diffraction data was successfully used in experiential investigations on molecular magnets. Furthermore, although this aspect has not been sufficiently explored yet, the possibility of obtaining wavefunctions compatible with experimental (X-ray diffraction) measurements might open unprecedented perspectives in the search of new materials with pre-designed properties.

Finally, the availability of quantum crystallographic libraries (of multipoles, extremely localized molecular orbitals or polarizabilities) to reconstruct electron distributions or polarizabilities of large systems (e.g., proteins), at a reduced computational cost, could also potentially open new routes in pharmaceutical research, with the perspective of performing electron density-based *in silico* high-throughput screenings or docking calculations for the search/design of new possible drugs.

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Location : CECAM-FR-GSO, Grand Hôtel d'Orléans, Toulouse, France
Webpage: https://www.cecam.org/workshop-details/35
September 27-30, 2021

1 State of the art

Energy transfer plays a prominent role in many dynamical processes at surfaces. The internal degrees of freedom (DOFs) of adsorbates typically couple to phonons and electron-hole pairs (EHP) of the surface. These couplings can be revealed by investigating fundamental steps of more complex reactions involved, e.g., in heterogenous catalysis. Improvement in state--to--state molecule-surface scattering experiments, together with the advent of novel atomic sources and high-power free-electron laser light sources, will provide mechanistic information about collisional inelasticity with unprecedented detail. The challenge for theory is now to understand the dynamical implications of the coupling of substrate EHP and phonons to the DOFs of atoms and molecules in the vicinity of surfaces.
Density Functional Theory (DFT) has become a standard approach to investigate molecule-surface interactions and full-dimensional, DFT-based on-the-fly calculations (e.g. AIMD) can be performed almost routinely for the ground electronic state, but the computation of excited states and of electronic processes remains a formidable challenge. Yet, schemes such as Independent Electron Surface Hopping and AIMD with electronic friction have been developed. Also, excited states can now be treated via Time-Dependent DFT (TD-DFT), but on-the-fly calculations are too time-consuming for simulations beyond a few typical trajectories, preventing meaningful comparison to experiment. Approximations to DFT such as Density Functional Tight Binding (DFTB) or TD-DFTB, constitute another promising route.

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2 Major outcomes

Density functional theory (DFT) is the established choice for characterizing the electronic structure of molecules interacting with surfaces. In this CECAM workshop, this was identified as a key challenge for the future. For example, it was shown that sticking probabilities depend strongly on the quality of the reference quantum chemistry data used for constructing potential energy surfaces (PES). Quantum Monte Carlo was advocated as a promising method to benchmark the important topological points of the PES, helping refine the reaction barriers and adsorption energies. The need to develop alternative methods, such as wave function-based cluster embedding theories and novel hybrid density functionals for DFT, was also emphasized.
Another major issue to treat fundamental surface reactions is the representation of the electronic structure in the form of a PES. A large part of the CECAM meeting was devoted to the recent developments in PES representations, which is evolving from traditional force fields and interpolation schemes towards machine learning (ML) models. The success of ML models to fit PES in both electronic ground and excited states should be seen as the major highlight of the workshop, as ML is establishing itself as both the state-of-the-art and the way of the future for surface science. Important open questions were discussed, such as how to quantify and improve the transferability of ML force fields, how to choose the appropriate descriptor for a given system, and how to create a reliable database for the ML model.
ML models were also demonstrated to provide an accurate and efficient alternative to variational methods for high-dimensional vibrational spectroscopy at surfaces, and to have sufficient flexibility to provide faithful representations of nonadiabatic coupling elements – in particular the friction tensor – over the whole configurational space relevant for the dynamics. Here, highly accurate perturbative treatment of EHP coupling up to second order was shown to be necessary and sufficient to provide a quality database that could be used for future ML models of nonadiabatic coupling.
Another element of novelty was the discussion of dynamical methods allowing to treat open quantum systems using stochastic wave packet methods. These could allow investigating surface dynamics beyond the current paradigm of ab initio molecular dynamics with electronic friction, and also beyond the Markovian approximation. For the latter, the potential of novel effective time-dependent mean-field Hamiltonians to include phonons and EHP coupling in simulations of molecule-surface scattering was also demonstrated.

3 Community needs

As for computing most of us use several levels of resources, including local institute clusters, regional meso-centres and national HPC infrastructures. These distinct levels of resources are complementary and provide a flexible, adequate framework for development and computational production tasks. Obviously, high-DOF scattering, *ab initio* molecular dynamics and non-adiabatic dynamics require a large amount of HPC resources and will necessitate further adaptation to HPC and parallel computing.

The workshop aimed to networking theoreticians from different communities in order to bring the richest and most diverse panel of methodological expertise that can be useful to advance the community core applications. Thus, suggestions of collaborative work were prolific.
Overall, the after-talk (30 + 10 minutes of discussion) and the (three) daily (50 minutes) discussions were truly intense. There were so many questions, comments and debates, that none of the discussion periods came to a sudden end and that the round tables had to be interrupted on or beyond the schedule, leaving for informal sessions the opportunity to discuss further about work that was insufficiently addressed. Clearly, this second CECAM workshop (initially scheduled in 2020) was highly useful and fruitful in learning how to solve a number of problems, which in turn called for a number of new problems to be considered and solved. Indeed, there remain open issues that have just begun to be explored and that may impact on diverse fields, such as *e.g.* the interplay between electronic friction and nuclear quantum effects, the effect of a non-vanishing memory in the electronic friction kernel in gas-surface dynamics and catalysis, the need of internal consistency checks for theory that can overcome the unavoidable uncertainties of experimental results. Hence, it would be most profitable to continue this "series" of CECAM workshops started in Albi, France, in 2017.

4 Funding

Funding was not discussed in the official sessions. We nevertheless expect that joint research proposals will come out of the lively and fruitful discussions, of the many informal exchanges and of the overall excellent assiduousness throughout the workshop. Yet, the presentations frequently acknowledged national funding schemes as well as several existing collaborations. Right after the workshop some joint research proposals between participants have been submitted to the Horizon Europe program (e.g. in response to the ERC-Syg-2022 call) and others are being considered, in the form of either research or training networks.

5 Will these developments bring societal benefits?

The topics addressed by the workshop aim at pure fundamental research and the benefits are at the moment exclusively scientific. It is rather hard to think about a problem beyond the academic world (economical, technical, societal or cultural) that could take advantage from any development discussed in the workshop. Yet, the kind of challenging problems that were addressed is exactly the one that every once in a while triggers developments in wider research areas, and impact on society.
While the work is fundamental in nature, in conjunction with UHV experiments it allows us to explore and understand how reactions take place on surfaces. This impacts our understanding of things like catalysis, semiconductor processing and other surface or nano technologies. What we learn about the dynamics of these fundamental processes guides those who design better catalysts, leading to industrial processes that are both more efficient and more environmentally friendly. Thus, the impact on society is significant.
More generally, the workshop addressed the theme of energy conversion that can be of relevance to scientific problems related to the production, consumption and storage of energy in all its forms.

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Location: Online
Webpage: https://www.cecam.org/workshop-details/1069
October 11-15, 2021

1 State of the art

Methods for simulating complex phenomena are increasingly becoming an accepted mean of pursuing scientific discovery. Especially in molecular sciences, simulation methods have a key role. Modern computing units are inherently parallel machines where multiple multi-core CPUs often paired with one or many accelerators such as GPUs or, more recently, FPGA devices. In this context, High performance computing (HPC) is the computer science field that specifically addresses the task of optimizing the performance of software through code refactoring, single/multi thread/process optimization. Despite several excellent codes already exist, the requirement of properly accelerating simulative codes is still compelling with several engines not fully leveraging the actual capabilities of current architectures. An example of such codes is NanoShaper [1]; NanoShaper is a software for computing the molecular surface both for the aim of visualization and for solving the Poisson-Boltzmann equation. NanoShaper is already a multi-threaded software, its core algorithm is prone to parallelization, however it does not leverage yet GPUs nor distributed memory architectures. As NanoShaper presents these features is the ideal platform for learning, practicing and testing parallelization paradigms including the already existing parallelization scheme and further developments.

Key References
*[1] S. Decherchi, W. Rocchia, PLoS ONE, **8**, e59744 (2013)*

2 Training provided

In this E-CAM Extended Software Development Workshop (ESDW) we introduced participants to High Performance computing using as reference platform the NanoShaper algorithm and code base. This was achieved through frontal lessons on NanoShaper algorithm/code, computer architectures, applications and via hands-on sessions where participants had the possibility to learn parallelization techniques (e.g. Boost threading and OpenACC) directly on the NanoShaper code base. This ESDW was hence focused on the technological aspects in HPC optimization/parallelization procedures and techniques. The primary educational goal of the workshop was to show to the participants which are the main challenges in code optimization and parallelization and the correct balance between code readability, long term maintenance and performance. This included technical lessons in which parallelization paradigms were explained in detail. A second goal was to allow participants to find computational bottlenecks within software (profiling) and to setup and imagine an optimization/parallelization strategy including re-implementing the existing NanoShaper parallel code and extending the parallelization scheme to GPUs and distributed memory architectures.
The frontal lessons covered:

- An introduction to single thread code optimization techniques including:
    - o CPU inner workings: instruction level parallelism via the pipeline, memory hierarchies (e.g. cache), Single Instruction Multiple Data paradigm
    - o Optimization techniques: dead-code clean up, constant folding, loop unrolling, loop fusion, SIMD etc.
- Introduction to NanoShaper algorithm and key routines.

- Code profiling (via tau)
- Shared memory architectures parallelization (OpenMP, Boost) and common parallelization patterns (e.g. reductions)
- Distributed memory multi-process parallelization (MPI): send/received, asynchronous paradigms, the load balancing problem.
- GPU oriented parallelization (CUDA, OpenACC): a first general introduction to CUDA followed by an in-depth introduction to OpenACC.

The workshop also had talks from world-wide HPC experts which shown their own applications and HPC developments ranging from industrial complex cloud architectures to the bit level optimization of GPU codes. This allowed people to understand the highly multi-faceted scenario of HPC from code optimization to big distributed, cloud infrastructures.

3 List of the software development projects

During the workshop and the hands-on sessions we concentrated on the NanoShaper code base. We covered two types of developments. In a first phase people was asked to re-implement already existing parallelization and or analysis of NanoShaper. These included:

- Single thread CPU code profiling via tau profiler. Production of profiling signatures of NanoShaper under the SkinSurface workload.
- Parallelization of NanoShaper ray casting phase via the Boost libraries both trying a cache friendly and not cache friendly strategy.

In a second phase instead, we focused on the development of genuinely new unreleased software modules. This includes chiefly an initial OpenACC (hence GPU) parallelization of the ray casting phase of the NanoShaper algorithm. While this process is obviously long, during the workshop time window we provided the participants an initially optimized OpenACC code base and asked participants to "fill some gaps" on the code to complete a first running proof-of-concept implementation. This initial version of the OpenACC code included the switching from the Array of Structures to the Structure of Arrays format for the patches of the SkinSurface, memory transfers between GPUs and CPU and the usage of OpenACC in an object oriented environment as is the one provided by NanoShaper.

4 Future plans

The IIT HPC group is developing now a fully parallelized version of NanoShaper of which the one provided to participants OpenACC version was a initial draft. This new NanoShaper will support GPUs as minimal goal and possibly fully support distributed memory architectures. This will enable to deal faster with the current molecular targets in terms of size and also to triangulate much bigger molecules particularly in the case the distributed memory version is realized. Additionally, during the workshop, we started a collaboration with Politecnico of Milano colleague Prof. Carlo De Falco, to link NanoShaper to the BIM solver with the aim of building a new adaptive Poisson-Boltzmann (PB) solver that takes fully advantage of NanoShaper features. Hence overall we expect to have in perspective two new software modules: an enhanced NanoShaper in terms of performance and its interfacing to the BIM code to support PB equation solution.

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Extended Software Development Workshop: Improving bundle libraries

Location: CECAM-HQ-EPFL, Lausanne, Switzerland
Webpage: https://www.cecam.org/workshop-details/23
October 11-22, 2021

1 State of the art

The landscape of Electronic Structure Calculations is evolving rapidly. On one hand, the adoption of common libraries greatly accelerates the availability of new theoretical developments and can have a significant impact on multiple scientific communities at once. On the other hand, electronic-structure codes are increasingly used as "force drivers" within broader calculations, a use case for which they have initially not been designed. All these trends further push the development of electronic-structure software more and more towards the provision of standards, libraries, APIs, and flexible software components. At a social level, they are also bringing different communities together and reinforce existing collaborations within the communities themselves. Ongoing efforts include an increasing part of coordination

of the developments, enhanced integration of libraries into main codes, and consistent distribution of the software modules
Since 2014, the Electronic Structure Library [esl, els-gitlab] has been paving the way towards broader collaborations within the Electronic Structure community, with a web site, data-exchange standards, refactoring code of global interest into integrated modules, and regularly organising workshops. An essential component of the ESL is the ESL Bundle, a consistent set of libraries broadly used within the Electronic Structure community. This bundle solves various installation issues for end users and enables a smoother integration of the shipped libraries into external codes.
With the above points in mind, the goals of this ESDW were: (i) improving the ESL libraries by letting developers react to feedback from third-party codes, (ii) extending functionality of libraries and (iii) adding new libraries to the ESL Bundle.
[esl] http://esl.cecam.org/
[esl-gitlab] http://gitlab.e-cam2020.eu/esl

2 Training provided

The initial discussions provided an ample view into specific code developers contributions and thoughts on the modularisation that the ESL and others are supporting. There was a broad range of presentations from community codes and libraries and how they were dealing with the modular software development framework. These included Quantum Espresso, Libxc, Abinit, DFTB+, Siesta, libOMM and MatrixSwitch, i-PI, ELSI, FHI-aims, Wannier90, INQ, AiiDA, SIRIUS, Exciting. In addition, there was also a presentation of new libraries involving kinetic Monte Carlo simulations that are currently under development. Other related communities that have similar prospects and are important collaborators, such as the MolSSI [molssi] and EESSI [eessi] projects, were also given the chance to present themselves and their work.
A discussion point at the workshop was how to unify an interoperability mechanism to share common objects and data-structures between simultaneously running codes. In the long term this would enable top-level DFT codes to exchange data between other DFT codes thus leveraging mechanisms in each of these. This is something that is currently being investigated in various other implementations openPMD with interfaces for ADIOS2 etc.
Another discussion point was error-handling in libraries and codes. It was acknowledged by the participants that this is still a considerable issue in Electronic Structure software and some strategies and solutions were debated. In particular, an *ad hoc* talk was organized to present and discuss error-handling in Object-Oriented Fortran.
During the second part of the workshop, which consisted of coding sessions, the participants organised themselves into small groups. Whenever possible, less experienced participants were teamed with more senior developers, so that they could benefit from their experience and knowledge of the different ESL projects. The format of the coding sessions also allowed for ample time and opportunity for the participants to interact, discuss, and share experiences.
[molssi] https://molssi.org/
[eessi] https://eessi.github.io/

3 List of the software development projects

During the coding sessions participants worked on the following projects:

- Upgrading the supported package versions included in the ESL Bundle and releasing a new version of it. This comprised ensuring that the build procedure and interoperability of the different packages were up to date.
- In the context of maintainability and providing developer access to latest versions of libraries included in the ESL Bundle, we have created a more versatile build-scheme enabling developers to install different versions of libraries without manually editing files. This will help

developers test their codes against the ESL Bundle shipped libraries and the bundles build mechanisms.

• Coordinate various developments on Libpspio (updated CI, code cleanups, preparation for ABINIT 8 file format inclusion, removal of interpolations…).

• Present and discuss a simple way to support an error stack in Fortran 2003.

• Inclusion of the ESL Bundle in the official release of EasyBuild [easybuild].

• Work on a new library for Green-Function based methods suitable for exascale architectures.

4 Future plans

It is part of our long-term strategy for the ESL Bundle to release a new version of it every 6-12 months. These new versions include updated versions of current libraries, as well as new ones. Already planned is the inclusion of the DFTB+ library and of LibPAW, which is part of the ABINIT project.

Spurred by the discussions about Object-Oriented Fortran, we are organizing a series of monthly on-line seminars on the subject.
A follow-up to this workshop is planned. The dates are still to be determined, but the goal would be to have 5-10 participants on-site for a week in Spring 2022.
With the generous financial support of CECAM, another ESL workshop is currently being organized. This workshop will cover the topic of best practices in Electronic Structure software development, as this was identified by many participants of previous workshops as a very important issue.

5 Participant list

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Recent developments in quantum Monte Carlo

Location: Rome, Italy
Webpage: https://www.cecam.org/workshop-details/1050
October 21-22, 2021

1 State of the art

With advances in algorithms and growing computing power, quantum Monte Carlo (QMC) methods have become a powerful tool for the description of a variety of quantum fluids and a viable alternative for high-accuracy calculations of the electronic structure of many atoms, molecules and solids. They are, however, at a less mature stage than today's ready-for-use quantum-chemistry or density-functional packages: while the production of results on a variety of quantum many-body systems has been going on for at least three decades, a significant effort is still devoted to research and development of methods and algorithms, including real-time dynamics, super-accurate optimization strategies for trial wavefunctions and molecular structures, inputs for the density functional theory of van der Waals forces, and eventually neural networks which at least for small molecular systems seem to improve the accuracy of variational quantum Monte Carlo to the point where it outperforms other ab-initio quantum chemistry methods.
Organized by his mentors and former students currently active in the field of computational physics, the workshop is also meant as a celebration of the 60th birthday of Saverio Moroni, a sound condensed-matter theorist who has given key contributions to quantum Monte Carlo methods, their application to a wealth of quantum fluids and chemical systems, and their dissemination in Europe.

2 Major outcomes

This event gathered, despite limitations still on for the COVID19 pandemic, scientists from thirteen countries all over the world to discuss the status and recent developments in the field of Quantum Montecarlo. The event was organized in order to permit the attendance also by remote, joining the in-person session via Zoom. The logistics was kindly provided by the CREF (Centro Ricerche Enrico Fermi) who made available their premises and the technology to permit remote attendance.
The workshop spanned two days, organised in three topical sessions: New methods and algorithms (chaired by David Ceperley, University of Illinois at Urbana Champaign), Quantum fluids (chaired by Gaetano Senatore, University of Trieste) and Solid State Physics (chaired Lilia Boeri from the Sapienza University of Rome). Each of the session consisted of a set of 30-minute, each including some time for questions and discussion. All the talks received a good and lively feedback from the audience, including those given from the (few: 5 out of 19) remote invited speakers.
New methods and algorithms (chair: Ceperley)

- Carleo: Neural-network quantum states in continuous space
- Foulkes: Approximating many-electron wave functions using deep neural networks
- Sorella: The phase diagram of the Hubbard model by Variational Auxiliary-Field QMC
- Baroni: Stochastic perturbation theory: a prequel to Reptation quantum Monte Carlo
- Guidoni: Quantum Computing for Correlated Systems
- Filippi: Variational principles and excited states in quantum Monte Carlo
- Mitaš: Quantum Monte Carlo and spins: systems with spin-orbit interactions
- Umrigar: Green's functions for cross-node and fixed-node diffusion Monte Carlo

Quantum fluids (chair: Senatore)

- Boninsegni: The (never ending?) search for exotic phases of helium on graphite
- Holzmann: Quantum fluids: from helium to electrons
- Fantoni: A quantum Monte Carlo method for nuclear matter
- Gori-Giorgi: Noncovalent interactions from models for the Møller−Plesset adiabatic connection
- Vitali: Ground and excited states of Fermi superfluids from QMC calculations
- Pierleoni: Energy gap closure and metal-insulator transition in solid and fluid hydrogen with pressure
- Casula: Phase diagram of high-pressure hydrogen including nuclear quantum effects

Solid state physics (chair: Boeri)

- Grosso: The young Saverio: the years in Pisa
- Attaccalite: Optical excitations in layered materials: the case of hexagonal boron nitride
- De Palo: Long-range order in electron-hole bilayer with valley degeneracy
- Varsano: Evidence of ideal excitonic insulator in MoS2 under pressure

In addition, at the end of the first day, we had a session with six posters contributed by in-person participants.
A relevant, noteworthy consideration is that the talks were spanning a large landscape, from methodological aspects to applications, and that all contributions fueled active discussions within the audience. The public of the workshop was very heterogenous, extending from people with a long, acknowledged expertise in QMC to much younger students and postdocs. Such a mixture, which the social event further enhanced, certainly played an important role in the unquestionable success of the workshop.
Nonetheless it is important to mention how this workshop was meant to be a tribute to the role that Saverio Moroni had in the development of Quantum Montecarlo methods. During the two days of workshop, it was easy to touch the fact that many generations of scientist have grown up and still are able to contribute to this field of research thanks to the Moroni's work.

3 Community needs

This workshop was a good opportunity to gather a community which, for several decades, has been committed to the invention of new statistical and computational approaches to the solution of condensed-matter and physical-chemistry challenges, and to the development of existing ones. In all of their many different flavors, Quantum Monte Carlo methods take enormous benefit from the exploitation of their intrinsically parallel nature. The technological evolution and the availability of large-scale infrastructures has boosted the opportunity tof inventing, testing and improving algorithms based on stochastic methods for the resolution of a large variety of problems. In the context of HPC exploitation, the TREX Center of Excellence was presented during the poster session. It turned out that many of the participants are already involved in it, which clearly witnesses a structured connection between the HPC and the people working on QMC methods. A promising aspect, touched upon during the discussion, was the possibility of monitoring how beneficial, in the medium term, Quantum Computing might be to the topics discussed during this workshop.
Although the core business of this workshop was a wide variety of continuum and lattice Quantum Monte Carlo methods, some detours on Density Functionals, Materials Science and as said Quantum Computing also took place. Such a large view on diverse approaches amounted to a very valuable networking opportunity (much more so being one of the very first on-person international meetings after the COVID19 "isolation") both for young researchers and more experienced people, to discuss and confront problems and solutions.

4 Funding

In this workshop there was not a specific discussion about the funding opportunities.
All presentations of the workshop were funded by national funding agencies of the country of affiliation of the speakers.
As of computing time, many results presented in the workshop were funded by PRACE.
In the last year the TREX Center of Excellence has started in the European call for Horizon2020. This project involves many different existing QMC codes (some of them discussed within the workshop), with the ambition of improving their efficiency within the next-generation HPC systems in EU.

5 Will these developments bring societal benefits?

Understanding the properties of materials, at the atomistic and electronic structure level is not of mere academic interest but play a key role in chemical processes governing the possibility of efficient light-energy conversion, energy storage, and electronic transport phenomena, which are at the heart of novel green technologies. The energy scales at play in these types of (photo)chemical reactions are small, significantly smaller than the systematic error usually plaguing standard approaches in their nominal range of applicability. To date, Quantum Montecarlo is among the few - if not the only -possible methods able to offer the needed predictive power through realistic simulations of these systems. Having new methodologies in the field of Quantum Montecarlo and the possibility of integrate these methods in efficient computer codes is a turnkey to implement societal benefits. For this reason, it is of extreme importance to foster the discussion and the confrontation in events such as this workshop.

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Benchmarking of electronic structure methods for non-covalent interactions

Location: Bat. Bienvenue (former IFSTTAR building), U. Gustave Eiffel 14-20 Bd Newton, 77420 Champs-sur-Marne, France
Webpage: https://www.cecam.org/workshop-details/1040
November 8-10, 2021

1 State of the art

The accurate description of molecular non-covalent interactions is a fundamental piece of the puzzle for the development of predictive computational models in chemistry. The *in silico* design of new catalysts, screening of drug molecules, materials simulation, all depend on the quality of the underlying model, the balance between strong and weak chemical forces. Even in an age of rapid progression in machine learning tools, our development in these areas will be ultimately limited by the quality of the data available. Furthermore, one needs to consider the level of resolution needed to achieve chemical predictions.
This workshop was created to consolidate joint experimental-theoretical benchmarking. It built on the success of previous initiatives, [1,2] providing a forum:

- for an open exchange on the blind challenges performed so far, including discussions on:
    - electronic structure methods for structure optimisation and configurational minima and search of gas phase molecular clusters
        - approximations in the simulation of vibrational/rotational spectra
        - procedures in error analysis for both experiment and theory

        - shortcomings and/or strengths of the workflows adopted
- to set the stage for future blind challenges or alternative benchmarking events, featuring
    - challenges for electronic structure theory in the description of non-covalent interactions
    - limits of experimental resolution in the study of non-covalent molecular clusters
    - discussion of standards for data exchange and benchmarking practice
    - discussion of negatives in experimental investigations and their potential publication

Key References

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*[2] H. Gottschalk, A. Poblotzki, M. Fatima, D. Obenchain, C. Pérez, J. Antony, A. Auer, L. Baptista, D. Benoit, G. Bistoni, F. Bohle, R. Dahmani, D. Firaha, S. Grimme, A. Hansen, M. Harding, M. Hochlaf, C. Holzer, G. Jansen, W. Klopper, W. Kopp, M. Krasowska, L. Kröger, K. Leonhard, M. Mogren Al-Mogren, H. Mouhib, F. Neese, M. Pereira, M. Prakash, I. Ulusoy, R. Mata, M. Suhm, M. Schnell, J. Chem. Phys., **152**, 164303 (2020)*

2 Major outcomes

The major goal of the workshop was to build connections between theoretical groups and experimental research units in areas particularly affine to quantum chemical benchmarking. This has been successfully achieved by hosting a significant number of participants from gas phase spectroscopy. The discussion was geared towards identifying weaknesses in current standard quantum chemical protocols, with several challenges being pointed out:

- the accuracy of dispersion corrections for large molecular systems was questioned in several talks. Particularly in supramolecular chemistry, current approaches to the description of dispersion interactions seem to overshoot. Experimentally predicted structures are not correctly ranked, with the most compact conformers being signaled as most stable.
- the problem of conformational sampling. Although several advances have been made in computational protocols for the search of local structural minima (e.g., CREST from the Grimme group), some outstanding issues remain. When shifting from lower to higher levels of theory, the number of minima is not necessarily kept and the energetic ordering can often times be faulty. This is a factor to consider when evaluating quantum chemical methods, and which should be featured/discussed in benchmarks. How well can lower-level methods cover the conformational space away from the global minimum?
- diversity in datasets was also actively discussed. In the last day of the workshop, one had the opportunity to discuss developments in open-shell datasets. Open-shell systems are often not considered in benchmarking and present unique challenges, not only to theory but also to experiment.
- discussion of errors in benchmarks should be more careful. From the talks on error analysis, a discussion took place on the generalised use of mean average errors, and how other quantities should be included in these assessments.
- the general problems faced in designing experiments well suited for benchmarking. Particularly for kinetics experiments, this can be a challenging task. An active discussion took place around the choice of models for interpreting gas phase kinetics.

It should be highlighted that this was the only in-person event to enable contact between several of the groups who had already participated in blind challenges from the

Suhm/Schnell/Obenchain groups [10.1063/5.0004465]. These were seminal double-blind studies and included work from over 12 research groups. It was important to have talks from both the theory and experimental side reviewing the outcomes and major conclusions. Following the discussion of the latter challenge, the setup and design of the next blind study (HyDRA, Setting up HyDRA) was presented, with key contributions from the blind challenge designers, and included detailed summary from the PhD student who is managing the international challenge. This provided the perfect setting to clear some of the participant's questions on the scope of the challenge. Also new participants in the challenge have been enrolled.

3 Community needs

One of the primary topics of discussions centered on what we benchmark against. A good majority of the presentations focused on using the benchmarking theory against better theory, namely the gold standard (CCSD(T)) or its explicitly correlated version (CCSD(T)-F12). In addition, however, there were calls for more benchmarking against experimental data.
An aspect of needing more experimental data is choosing the correct data. The workshop identified that what experimentalists are looking for and the problems they are looking to solve are not congruent with the studies of theory groups. An example of this was the talk given by Georg Jansen, where it was identified the current data set available to compare DFT-SAPT energies, but that, despite a few key exceptions, the know experimental benchmarks are limited. After the talks, it was identified in discussions that the experimental community doesn't need to provide more data but needs to consider more carefully the targeted systems.
In the end, we could agree that collaboration is critical. However, there are still some lingering thoughts on how we move forward. Indeed, we recognized that the communication between groups had been limited due to the decreased availability of conferences and workshops following the pandemic. The valuable discussions that happen in a conference room or dinner table are lost in the digital meetings. Recognizing that there will still be a slow return to regular conference presentations, we must look to the communication and collaboration methods that have thrived in the lockdown. The recent and current blind challenges stood out at the workshop as a promising and refreshing path to future collaborations between theory and experiment.
A series of CECAM workshops should be considered as a forum for the discussion of blind challenges within the community. In a yearly basis, one would have the opportunity to gather participants from recurring or newer challenges.

4 Funding

The workshop benefited from the fundings allocated by CECAM-FR-MOSER. Moreover, the workshop was partially funded by the Research Training Group BENCh (RTG2455) and the NFDI4Chem, both German initiatives, the first with a focus on benchmarking and the second on research data management. The possibility of joint research proposals was considered for the second funding phase of the RTG2455, to be submitted by mid 2022. The interaction with NFDI4Chem is equally important, as it established contact to repositories and database archives, essential partners for upcoming proposals in the field.
The full amount of funding was used to cover the local expenses (housing, meals and conference diner) for most of the attendees (Seniors and Post Docs and PhD students). We also used part of the money to cover the travel expenses of some participants.
Smaller collaborations were also discussed which could later result in joint proposals, with focus on Franco-German interactions.

5 Will these developments bring societal benefits?

Benchmarking is a fundamental activity to assess the predictive power of quantum chemistry towards the development of new methodologies for large molecular systems (e.g. enzymes, drugs encapsulators, ...). By building experiments of varying degree of complexity whereby the methods can be tested and evaluated, one is building a blueprint for automated procedures to come in future years. Although we did not include any speaker from the industry, some of the participants have ties to the chemical industry and their work is a stepping stone for a stronger integration of molecular modelling and industrial/pharmaceutical research.
The other point which should be made is that the curation of experimental data will become an increasingly important activity, as the variety of computational tools increase and one requires a clear reading of the limitations in each approach. In the era of big data, it is important to care for the quality of the latter. Regression, classification methods can only be as good as the data from which they are based. This is an important step for a landscape of open science in this field, which can only increase the confidence of society in our work and methods.
By the way, more than 1/3 of the participants (in-person and online) were young researchers. The workshop was an excellent occasion for their training on benchmarking activities, both theory and experiment. The cross-fertilisation discussions were beneficial, in particular after 2 years of pandemic. Several PhD students were attending such event (conference/workshop) for the first time.

6 Participant list

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Paramagnetic NMR in f elements

Location: CECAM-FR-GSO
Webpage: https://www.cecam.org/workshop-details/1043
November 21-24, 2021

1 State of the art

NMR is among the most versatile tools to probe chemical bonding. Paramagnetic NMR (pNMR) refers to the NMR shifts in paramagnetic systems such as open-shell metal complexes, which are very different from their diamagnetic counterparts and poorly understood—despite decades of research [2,3]. The pNMR shift may be split (using pseudo-relativistic terminology) into a contact, a spin-dipolar (or 'pseudo-contact'), and a paramagnetic spin—orbital (PSO) term.

The modeling of pNMR relies typically on approximate expressions for the chemical shift in terms of electron paramagnetic resonance (EPR) pseudo-spin Hamiltonian parameters (g and hyperfine coupling tensors) or the magnetic susceptibility. Soncini et al. recently derived the NMR shielding tensor as a temperature-dependent bilinear derivative of the Helmholtz free

energy, providing a very general expression [9]. Multi-reference wavefunction-based pNMR calculations based on Soncini's expression, without recourse to a spin Hamiltonian mapping, have been reported in the meantime. However, these calculations are far from routine, and presently suffer from a poor description of the spin-polarization (which is very sensitive to the dynamic correlation) [5,10].

DFT based approaches are suitable to describe the spin polarization-delocalization in a paramagnetic system, but they are limited to states that can be described reasonably well with a single determinant, and the magnetic coupling between states requires an elaborate treatment in a response theory setup. Wave Function Theory (WFT) based approaches describe correctly the magnetic properties of a paramagnetic metal center, but with the currently available approaches it is difficult to treat the spin polarization-delocalization. One may foresee range-separation approaches which combine the advantages of DFT and WFT [11] or DMRG methods [12] which permit to handle large active spaces for a proper description of pNMR shifts.

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2 Major outcomes

The workshop gathered different communities, experimental with concerns with the underlying theory, computational chemists and quantum chemical tools developers.
There were 10 participants attending in person, and 25 persons online. Most of the participants attending in person had never met before, and the scientific exchange all along the workshop were really fruitful since all of them have the same interests, the same culture, but different points of view.
Informal discussions about some theoretical concepts such spin Hamiltonians, time relaxation, the limits of Soncini's equation were of prime importance.
The attendance online was very constant, showing the interest of the participants. Every presentation (limited in time to 20 or 30 mn) was followed by long discussions, unlimited in time, this up to 30 mn. Thanks to the good technical equipment of the conference room, the discussions were going forth and back between the room and the screen, without inconvenience.
There are two major issues for the calculations of paramagnetic chemical shifts, i) which formalism should one use, and ii) with which quantum chemical method. The applicability of Soncini's equation is now clear: one needs fast electronic relaxation, otherwise it is not applicable. The mapping onto the spin Hamiltonian formalism has been a subject of debate, since the description of the many low-lying states of the f elements by Spin Hamiltonian is no straightforward. The old question of whether the susceptibility tensor can be used for the long-range effects is now clear, and the old approach reaffirmed. Going beyond the dipolar terms may be needed if the magnetization distribution is extended (5f) rather than strongly localized (4f).
The calculation from first principles of the hyperfine coupling was addressed by two participants and this opens the door to the calculation of the contact contribution to the

chemical shifts. Numerous practical difficulties remain in the quantum chemistry part of all this, because of the underlying electronic structure approximations.
The necessity to have good experimental results was addressed I order to benchmark the new methods.
DMRG methods should permit the description of very large active spaces, and consequently, the description of the contact term, at the least for the atoms close to the paramagnetic center.

3 Community needs

Discuss the needs of the community in terms of computational infrastructure (e.g. existing codes, use of HPC resources), networking (e.g. outreach to other communities including experimentalists), event organization (e.g. should a series of CECAM workshops on this topic be considered and if so, why?)
The modelization of paramagnetic chemical shifts in f elements is performed using different codes of quantum chemistry, including relativistic effects.
Some of them are commercial, other are open source. Often, the use of development modules is necessary. The developers for all those codes did attend the workshop.
One can mention
the code MOLCAS for the wave function-based calculations where Jochen Autschbach recently implemented the calculation of hyperfine coupling and Stefan Knecht implemented DMRG with spin-orbit coupling. Interdependently to Jochen Autschbach, Nicholas Chilton developed a code in python for the calculation of hyperfine coupling, based on the same scheme.
The code ReSpect, for the 2-components relativistic DFT calculations. This code is devoted to the calculation of the magnetic spin parameters and is developed by Stanislav Komorovsky.
The code Dirac, for the 4-component relativistic calculations. This code is developed by Trond Saue and Hans-Jorgen Jensen.
Those codes are usually run on local clusters or national computing facilities.

4 Funding

The benefit of this workshop in order creates a community concerned by the same interests have been highly appreciated by the participants. Specially that it was a joined experimentalists - theoreticians workshop.
Nicholas Chilton and Stefan Knecht have started a collaboration.
Martin Kaupp and Radek Marek plan to apply against a bilateral German-Czech funding program.
Peter Kaden, Michael Patzschke , Claude Berthon and Hélène Bolvin plan to apply for a bilateral grant against ANR-DFG funding agencies.
Jochen Autschbach and Herman Cho have a common project funded by the US Department of Energy.
As a main issue, the team of the Helmholtz-Zentrum Dresden-Rossendorf center proposed this workshop to happen every second year. As a start, they send an invitation to all particpants of this workshop for the first “Actinides revisited” conference which will be organized in Dresden in September 2022.

5 Will these developments bring societal benefits?

The NMR of paramagnetic substances has become an important tool in many areas of research, in particular in the development of new magnetic materials in studies of biological systems or nuclear waste treatment.

Among the most interesting areas of research where paramagnetic systems containing heavy elements play an important role are
A. Targeted drug delivery and prodrug formulation
B. Understanding of paramagnetic NMR spectra (e.g. assignment of pNMR signals)
C. Nuclear waste treatment (e.g. actinide extraction to lower radio toxicity and heat load)
D. Extracting information about structure of metalloproteins
E. Advanced magnetic materials for data storage
NMR of paramagnetic species (pNMR) has the potential to gain deep insight into the electronic and molecular structures of molecules with unpaired electrons. This includes radicals used in biomedical research, lanthanide based molecular magnets, and actinide compounds found in nuclear waste. Therefore, the development of pNMR toward routine applications in these fields has the potential to transform them, with obvious benefits to society and the economy.

6 Participant list

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Location: On-line, hosted by CECAM-HQ-EPFL
Webpage: https://www.cecam.org/workshop-details/59
December 13-16, 2021

1 State of the art

Optical excitations dominate emerging applications, including energy conversion and storage and quantum information science. The involved processes typically include energetically-excited states, and in particular, energy carriers called *excitons*, composed of electrons and holes bound together through Coulomb interaction. The exciton scattering and relaxation dynamics, as well as their ballistic and diffusive transport, are key ingredients in device functionality and are closely related to the material composition. Understanding the underlying excited-state processes can introduce design pathways to optimize material functionality.
In recent years, extensive experimental research is dedicated to studying time-resolved excited-state phenomena in solid-state functional materials, such as transition metal dichalcogenides, organic molecular crystals, organic-inorganic hybrids, quantum dots, and metal-organic complexes. Such systems often host strongly-bound excitons as energy carriers. Advanced time-resolved spectroscopy and microscopy experiments suggest that those excitons can exhibit a rich variety of dynamics due to complex phenomena. These interaction processes are shown to have strong coupling to phonons and structural inhomogeneities, such as atomic defects, local strain, or non-uniform screening.
This workshop brings together researchers from different scientific communities, who study time-resolved exciton phenomena in functional materials using a broad variety of approaches. While the main focus is computational developments, an important aspect of it is a state-of-the-art experimental perspective. The main aim of this joint cross-community computational-experimental meeting is thus to encourage exchange of ideas and identify emerging questions for future research directions and collaborations, as well as to share and advance current theoretical methods to exciton dynamics and transport in functional materials.

2 Major outcomes

The scientific outcome of the workshop can be divided into three main topics:
Exciton decay selection rules in materials of reduced dimensionality: Exciton momentum and spin selection rules are coupled to the underlying material structure and symmetry. It is of increasing interest to better understand the possibility for structural design and tunability of the associated quantum selection rules. Of particular interest are the mechanisms dominating exciton dynamics in 2D transition metal dichalcogenides (TMDCs)- based materials, and specifically the relation between exciton momentum and spin properties to increased lifetimes associated with intervalley dephasing, dark state occupation, and multiexciton formation. In layered perovskites, structural design of the dielectric environment and lattice fluctuations dominate exciton decay and diffusion, and advanced computational approaches are required to realize the associated optical and dynamical interaction mechanisms. In particular, the role of light-field and exchange-induced interactions in the many-body description and its relation to exciton dispersion was discussed, as it is studied in recent years from analytical and many-body perturbation theory perspectives, both were represented in the workshop. In addition, several talks discussed direct observations of momentum-resolved excitations and associated non-equilibrium dynamics. The development of both the experimental and theoretical tools involved is new and ongoing, making this topic fundamental in emerging exciton dynamics research.

Advances in computational approaches to exciton-phonon interactions: In recent years, a significant development was achieved in experimental, theoretical, and computational capabilities to explore exciton-phonon coupling, essential for reliable predictions of excited-state dynamics in realistic systems. During the workshop, several talks discussed this progress in detail. Advances in experimental methods allow for a careful investigation of thermal effects in exciton migration. Related approaches and observations were discussed, presenting for example the case of phonon drift in TMDCs and vibrational coupling due to exciton delocalization in organic crystals. From the theoretical perspective, newly-developed approaches based on Green's function methods and time-dependent density functional theory, as well as density-matrix and Hartree-Fock-based approaches were presented; several aspects of this recent development were mentioned throughout the workshop, among them its application to polaronic coupling in organic molecular crystals, and a related investigation of the Rashba effect in perovskites. Other theoretical directions include the merging of phenomenological methods with ab initio understanding when accounting for exciton-phonon coupling. Associated studies discussed related developments and applications, such as ab initio computations of beyond-harmonicity effects, and phonon-assisted dark state exciton emission.
Exciton dynamics and structural complexities – hybrid 2D materials, heterostructures, defects: The relation between structural complexity and excited-state properties and interaction dynamics was a major topic of discussion in the workshop. From the experimental side, related talks discussed the effect of heterostructure composition, twist angle, and encapsulation; exciton self-trapping in hybrid layered inorganic materials; exciton effective mass and fine structure in perovskites of varying structures; defect-induced Zeeman splitting across layered interfaces; and multiexciton coupling in quantum dots. Related theoretical studies include structural effects on exciton-exciton and exciton-light coupling, spatially varying dielectric environments in layered hybrid systems, and the complex exciton nature and dynamics in TMDC-based moiré lattices and with atomic defects.

3 Community needs

The theoretical approaches and method development presented in this workshop represent new computational horizons of non-equilibrium and time-resolved dynamical excited-state phenomena, including explicit time propagation methods as well as perturbative approaches. The vast majority of these new theoretical directions is based on ongoing code development within existing packages, most of them are publicly available. It is of growing interest to connect such different codes for optimal application, and related efforts are forming these days through mutual grant applications and cross-methodological collaborations. The networking aspect within this community is, however, not trivial. Most of the related scientific meetings typically consider either specific methods - e.g. electronic structure methods, wavefunction-based approaches, or phenomenological models- or specific materials and applications, limiting the scope of the discussed underlying phenomena.
The main goal of this workshop is to bring together researchers and students from different backgrounds and identify common interests primarily associated with the underlying phenomena. Our aim is to generate an open and diverse environment for all participants to familiarize themselves with a variety of relevant theoretical and computational methods from one hand, and a collection of materials and applications from the other. The large number of 50-60 participants throughout all four days of the workshop, despite the challenging online form, as well as the fruitful and enriching scientific discussions which emerged during and after the workshop, is evidence of such common interests and will hopefully lead to future interactions in the field. It is hence our aim to establish this workshop in the form of a series, for instance once every two years.

4 Funding

The workshop was funded by both CECAM and psi-K. Possible funding sources for related collaborations include personal Europe- and US-based funding grants of individual participants (ERC starting and consolidator, NSF), as well as collaborative grants, such as an Horizon-DN networking recently-submitted grant applications including a number of participants, as well as ongoing collaborative PRACE proposals for associated computing allocations. Joint research is already performed by many of the participants. One of the main goals of the workshop is to establish pathways for new collaborations, as well as new connections between theoretical and computing approaches that do not trivially merge. Despite the online format, the workshop included stimulating discussions which can potentially lead to such new interconnections.

5 Will these developments bring societal benefits?

Current advances in science and technology rely on an ever-expanding base of nanostructured materials promising new device functionalities, sustainable operation, and improved performance. The materials systems involved typically combine high structural complexity with strong interactions between fundamental electronic excitations, lattice vibrations, and light. Successful technological application thus ultimately requires developments that go far beyond a phenomenological understanding of the associated phenomena. It demands system-specific predictions and a high level of theoretical modeling combined with reliable experimental input. This state of affairs motivates a broad variety of approaches from a diverse scientific community to address these challenging goals.
Bringing together researchers from different parts of the field within the scope of this workshop, opening up discussions for a broad variety of methods as well as proposing alternative solutions to key challenges represent a crucial step in this direction. Already today, computational approaches play a key role in state-of-the-art technology and guide future developments. It includes critical areas as information processing and storage, communication, renewable energy, sensing, and lighting. The platform provided by the workshop to advance these fields through exchange of ideas, resulting interactions, and possible connections between fundamental physics and applications is thus significant from the economic perspective and in view of the future impact. The resulting avenues to advance sustainable energy sources, develop quantum computation, as well as propose novel strategies to manipulate matter are promising and will serve as a platform for future developments. Combined with the education aspect of bringing young scientists together with established researchers, enabling exchange and advancing future collaborations renders this workshop a highly successful endeavor.

6 Participant list

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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.

David Vanderbilt, Rutgers University
Raffaele Resta, IOM-CNR Democritos
March 23 2021

Conceptual Aspects of the Theory of Electric Polarization and Orbital Magnetization
David Vanderbilt, Rutgers University

In this talk I will take a conceptual approach, rather than a historical or mathematical one, to the "modern theory" of electric polarization and orbital magnetization. In particular, I will present a series of physical arguments that severely constrain the form of any such theory. For example, I will explain why we expect electric polarization to be well defined only up to a quantum, and discuss the theorem relating bulk polarization to surface charge in general terms. As for orbital magnetization, I will explain why this quantity is not subject to the same kind of quantum of indeterminacy that was found for the electric polarization. Only then will I introduce the mathematical concepts of Berry phase and Berry curvature, and briefly point the way to their utilization as the basis for the by-now well-established theories of electric polarization and orbital magnetization.

View video

Electric Polarization, Orbital Magnetization, and Other Geometrical Observables
Raffaele Resta, IOM-CNR Democritos

Electric polarization and orbital magnetization have a very similar definition at the textbook level; yet within quantum mechanics they are intensive geometrical observables of a very different nature. At the fundamental level polarization is essentially a one-dimensional quantity and orbital magnetization a two-dimensional one: the difference between them can be traced back to basic features of algebraic geometry in odd vs. even dimensions. One- and three-dimensional observables make sense in insulators only and are well defined only up to a quantum: the former case is polarization; the latter case occurs in magnetoelectric coupling. When a bounded sample is addressed, the quantum indeterminacy is fixed by the chosen termination. Two-dimensional observables exist in metals as well and are exempt from the quantum indeterminacy; their value does not depend on sample termination. Besides magnetization, I will discuss anomalous Hall conductivity in insulators and metals.

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Free-energy of solids: origins and consequences of the 1983 CECAM workshop

Tony Ladd, University of Florida
Daan Frenkel, Cambridge University
May 20 2021

Thermodynamics of hard spheres: 1950's to 1980's

Tony Ladd, University of Florida

View video

Follow-up of the 1983 workshop

Daan Frenkel, Cambridge University

Liquid-state physics was the first subject to benefit from the advent of computer simulations, because there is no reference state for a liquid and so in the early 1950's there was no "Theory of simple liquids". However, after perturbation theories were developed by Barker-Henderson and Weeks-Chandler-Andersen, the hard-sphere fluid has played a key role as a reference state for all theories of simple liquids. This has been the basis for huge theoretical advances, using simulations to determine the properties of the hard-sphere reference system.
On the other hand, solid-state physics had the harmonic solid as a reference with an exact statistical mechanics, and an analytical basis for phonon perturbation theories. In that community the need for computer simulations was only perceived by a few visionaries such as J.P. Hansen and M.L Klein. But for a crystal of hard spheres, there is no harmonic approximation, and that is why the first numerical calculations of the free-energy of solids by Hoover and Ree used hard spheres. In the years that followed, very few free-energy calculations of solids were reported; again J.P. Hansen, along with L. Verlet, was one of the few exceptions. In the 1980s' hard sphere crystals moved from being a theoretical oddity to something that could be studied experimentally in colloidal suspensions, provoking additional interest in these systems that led to an experiment on the space shuttle mission STS-93.
At a 3-week workshop held at CECAM (then in Orsay) in 1983, the present speakers were intrigued by the problem of finding a robust and accurate way to compute the free-energy of solids. It was completely off-topic: the focus of the workshop was on novel equations of motion for molecular dynamics (constant T, p, and NEMD). But the wonderful thing about these workshops was: there were almost no talks, there was no structure (except coffee and our daily metro ride from and to the center of Paris), and you just worked on whatever interested you.
There was an unanswered question for hard sphere systems that interested us, namely which crystal phase was the most stable: FCC or HCP. Earlier attempts by Hoover and Ree had failed to reach the required accuracy, because the difference is so small. And this stimulated us to try to do better. In the end, we also failed, but we could put much more precise bounds on the free-energy difference. In fact, it took another 15 years before the problem was resolved: FCC is more stable, in agreement with the contemporaneous results from the Columbia mission, but at considerably less expense! The result, as such, is not so important, because the difference is so small (about 10^{-4} k_BT) but the work set standards for the kind of accuracy that free-energy calculations should achieve, and that development continues to this day.
In our talk we will discuss the workshop, the history of the interest in hard-sphere models, and some of the follow-up studies. In the discussion, we may even tell some personal stories - if prompted.

View video

Athanassios Panagiotopoulos, Princeton University
Dominic Tildesley, University of Southampton

December 9 2021

Simulation of Fluid-Phase Equilibria
Athanassios Panagiotopoulos, Princeton University

The phase behavior of pure components and mixtures plays an important role in science and technology, so there has been interest in molecular-based calculations of phase equilibria since the early days of computer simulation. In this presentation, I plan to review the origins of the Gibbs ensemble methodology [1,2] and its development in the intervening thirty years, which have enabled the routine calculation of phase diagrams for fluids composed of complex molecules. An alternative to the Gibbs ensemble is provided by histogram reweighting grand canonical Monte Carlo, which have greater accuracy near critical points. A number of other efficient Monte Carlo sampling algorithms have also been proposed over the years. In addition, open-source fast and scalable molecular dynamics codes have been developed that can be used in direct interfacial simulations of phase equilibria. The main remaining challenges in this area are related to the inadequacies of the current generation of force fields and the development of machine-learning methods to allow *ab initio*-based predictions of the phase behavior.

1. A.Z. Panagiotopoulos, "Direct determination of phase coexistence properties of fluids by Monte Carlo simulation in a new ensemble," *Mol. Phys.*, **61**, 813-826 (1987).
2. A. Z. Panagiotopoulos, N. Quirke, M. Stapleton and D. J. Tildesley, "Phase equilibria by simulation in the Gibbs ensemble: alternative derivation, generalization and application to mixture and membrane equilibria," *Mol. Phys.*, **63**, 527-545 (1988).

Athanassios Panagiotopoulos received an undergraduate degree from the National Technical University of Athens in 1982 and a PhD from MIT in 1986, both in Chemical Engineering. He was a postdoctoral fellow in Physical Chemistry at the University of Oxford (1986-87) and held faculty positions at Cornell (1987-97) and the University of Maryland (1997-2000). He is currently the Susan Dod Brown Professor and Department Chair of Chemical and Biological Engineering at Princeton University. He has received numerous awards and honors, including the AIChE Colburn Award, and the Prausnitz Award in applied chemical thermodynamics. He was elected to the U.S. National Academy of Engineering in 2004 and to the American Academy of Arts and Sciences in 2012. He is the author of more than 300 technical papers and of the undergraduate textbook "Essential Thermodynamics" (2011).

View video

Molecular Simulation of Fluid-Phase Equilibria: multicomponent mixtures and pores
Dominic Tildesley, Department of Chemistry, the University of Southampton

I want to begin by recalling the day that the original Gibbs ensemble Monte Carlo paper dropped onto my editorial desk in Southampton and the range of opportunities that immediately opened up with this breakthrough. As an example, we recall that the Gibbs ensemble Monte Carlo method (GEMC) is readily extended to multicomponent mixtures by increasing the number of independent boxes in the simulation. In this way, all six classes of phase behaviour for binary liquid mixtures can be reproduced with the simplest potential models. The technique can also be extended to three component mixtures exhibiting up to

four coexisting phases (LLLV). It is not easy to study the effects of confinement on fluid-phase equilibria in irregular geometries by experiment or classical liquid state theory. This is where simulation comes into its own and GEMC can provide a testbed for analytical theories where the pore is modelled as a static external field of hard spheres. One of the beauties of the method is the ease with which it can be extended to fluids of small rigid molecular fluids, fluids of flexible molecules and even to polymers. We will examine a more recent application of GEMC to a quantum fluid, ^{4}He, using a path integral approach. Before the development of GEMC, fluid phase equilibria were often simulated using a gas-liquid interface in a periodic slab geometry and changing the temperature to move along the LV coexistence curve. We want to illustrate some of the hurdles in the study of phase equilibria using this direct approach and how they have been gradually overcome with particular reference to liquid water and liquid argon.

Dominic Tildesley received his BSc in Chemistry from Southampton University in 1973 and his DPhil from Oxford University in 1977. After postdoctoral positions at Pennsylvania State University, Cornell and Oxford, he joined the Chemistry Department at Southampton University in 1981. He was awarded the Chair of Theoretical Chemistry at Southampton in 1990. In 1998 Dominic joined Unilever Research Port Sunlight as the Head of the Physical Sciences Group. In 2003 he became Chief Scientist for Unilever's Home and Personal Care Division. He retired from Unilever in June 2012. He moved to Switzerland in January 2013, to become Director of CECAM (Centre Européen de Calcul Atomique et Moléculaire) based in Lausanne in Switzerland. He was awarded the title of Professor Titulaire at the École Polytechnique Fédérale de Lausanne. He retired in December 2017 and is currently a visiting Professor at the University of Southampton, where he holds an honorary doctorate. He is the author of The UK e-Infrastructure Strategy for Science and Business: a roadmap for the development and use of advanced computing, data and networks, and he co-chaired the UK E-infrastructure Leadership team. He has been President of the Royal Society of Chemistry and received a CBE for services to science, technology and business in 2014.

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MARY ANN MANSIGH CONVERSATION 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.

Beniamino de' Liguori Carino, Adriano Olivetti Foundation
Pierpaolo Perotto, Finsa-Technology for people
November 9 2021

Coding a legacy: Adriano Olivetti and the factories of good(s)
Beniamino de' Liguori Carino, Adriano Olivetti Foundation

Entrepreneur, industrialist, publisher, intellectual, politician, social innovator and pioneer of urban planning, Adriano Olivetti (born in Ivrea in 1901 – died in Aigle in 1960) is one of the most singular and extraordinary figures of the 20th century. His community-based social reform project, which centered on the interplay between physical progress, technical efficiency and the ethics of responsibility, is today recognised as one of the most up-to-date and advanced models of sustainability. The presentation frames the Olivetti legacy providing a brief overview of Adriano Olivetti's philosophy as well as placing specific focus on links which bound together the Olivetti entrepreneurship experience with some of the most intriguing challenges we are nowadays facing as society.

Beniamino de' Liguori Carino is General Secretary and member of the Board of Trustee of the Adriano Olivetti Foundation. Since 2009 he has been a member of the Adriano Olivetti Foundation's Scientific Committee, of which he became Secretary in 2016. He has been a member of the Advisory Committee of the Olivetti Historical Archive Association since 2016 and has been appointed Vice President. Since 2012 he is Publisher and editorial director of Edizioni di Comunità. Beniamino graduated from La Sapienza University of Rome with a degree in Modern and Contemporary History.

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The Programma 101, a small big idea
Pierpaolo Perotto, Finsa-Technology for people

In the October of 1965 Olivetti presented the Programma 101 at the New York World Fair in a small room of its stand. That was the beginning of a revolution that strongly effected the future of Olivetti, the future of the information technology market and the future of many of us. The presentation frames the historical, cultural, and technological contest that allowed the project team of the Programma 101, led by Pier Giorgio Perotto, to make real what, at that time, was considered a dream so impossible to be difficult even to dream and that impacted so significantly the future events inside and outside Olivetti.

Pierpaolo Perotto is the CEO and partner of Finsa s.p.a. founded in Genova in 1996 by his father Pier Giorgio when he left Olivetti group after almost 40 years. Finsa is a consulting firm focused on Advisory, Design and Technology, which also founded App2check, a state-of-the-art CX analytics platform that collects and analyzes customer feedbacks through a proprietary Artificial Intelligence engine. Pierpaolo started his career in 1986 in Silicon Valley where he worked for two years at the Olivetti Artificial Intelligence Center. In 1988 moved back to Italy working at Olivetti in Ivrea for other 10 years and later at Ernst & Young Consultants until 2002. In 2003 he took the lead of Finsa. Pierpaolo Perotto took a master's degree in Electronic Engineer at the University of Genoa in 1986 and he was a guest professor at the University of Economic Science of Turin from 2003 to 2005.

View video

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.

Christoph Dellago, University of Vienna
Luigi Bonati, ETHZ
Yanyan Liang, Ruhr-Universität Bochum
January 28, 2021

Bridging time scales in molecular simulations
Christoph Dellago, University of Vienna

The microscopic dynamics of many condensed matter systems occurring in nature and technology is dominated by rare but important barrier crossing events. The resulting wide ranges of time scales are a challenge for molecular simulation. For instance, a supercooled liquid can exist in a metastable state without freezing for very long times before it eventually crystallizes. In this case, typical waiting times may vastly exceed the time scales accessible to molecular dynamics simulations. Similarly, the time scales of chemical reactions or of biomolecular reorganizations can be many orders of magnitudes longer than those of basic molecular motions. This rare event problem is also related to the problem of finding reaction coordinates, i.e., of identifying important degrees of freedom that capture the mechanism of the process under study. In this talk, I will give an overview of several approaches developed to address these computational problems and illustrate them with various examples including the crystallization of supercooled liquids and cavitation of water under tension. I will conclude with a discussion of some open problems and an outlook to possible future directions.

View video

Training collective variables for enhanced sampling via neural network-based discriminant analysis
Luigi Bonati, ETHZ

The design of an appropriate set of collective variables is crucial to the success of several enhanced sampling methods. Here we focus on deriving such variables from information limited to metastable states. We characterize these states with a large set of descriptors and use neural networks to compress this information into a lower-dimensional space, using Fisher's linear discriminant as an objective function [1]. This approach results in an efficient sampling of a variety of rare events, while also identifying the physical descriptors that undergo the most significant changes in the process. We illustrate these features through two applications. The first is the study of an intermolecular aldol reaction characterized by a concerted mechanism, while the second one is the calculation of ligand binding free energies for a set of host-guest systems [2].

References
[1] L. Bonati, V. Rizzi, M. Parrinello, J. Phys. Chem. Lett., ***11****, 2998-3004 (2020)*
[2] V. Rizzi, L. Bonati, N. Ansari, M. Parrinello, Nat Commun, ***12****, 93 (2021)*

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Identification of a multi-dimensional reaction coordinate for crystal nucleation in Ni3Al
Yanyan Liang, Ruhr-Universität Bochum

Crystal nucleation during solidification in multi-component alloys is a complex process that comprises competition between various crystalline phases as well as chemical composition and ordering. In this work, we combine transition interface sampling method with extensive committor analysis to investigate the atomistic mechanism during the initial stages of crystallization in bimetallic Ni3Al. The analysis of the path ensemble indicates the existence of multiple reaction channels leading to the formation of face-centred cubic (fcc) or body centered cubic (bcc) crystallites. The committor analysis shows that the formation and growth of crystalline clusters and the polymorphs selected in Ni3Al depend explicitly on the interplay between cluster size, crystallinity, and chemical short-range order. In particular, the chemical short-range order exhibits a key role in the stability of the small cluster triggering either continuous growth or shrinkage. In contrast to classical nucleation theory, it is essential to include all three features in a multi-dimensional reaction coordinate to correctly describe the nucleation mechanism in Ni3Al. The identification of a multi-dimensional reaction coordinate is expected to be of key importance for the atomistic characterization of crystal nucleation in complex, multi-component systems.

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Mixed-Gen Season 1 Session 2: Active matter

Julia Yeomans, *University of Oxford*
Bruno Ventejou, *CEA Saclay*
Supriya Bajpai, *IITB-Monash Research Academy*
March 4, 2021

Phase field models for mechanobiology
Julia Yeomans, University of Oxford

A lot is known about the ways in which single cells move over a surface, but there are still many questions about the motion of confluent layers where cells coupled through intercellular junctions show a range of behaviours. In vivo cells move through the extracellular matrix, a viscoelastic polymer network. Here even single cell motility is not well understood and the mechanobiology of the collections of cells that form tissues and tumors is little explored.
Several ways to model cell motility have been described in the literature. One of the most successful has been active nematic (a.k.a. active gel) continuum models. However, these do not resolve individual cells and it is hard to distinguish the roles of intercellular and intracellular forces. Models which do resolve individual cells and the interactions between them include Potts, vertex and phase field models.
This talk will concentrate on phase field models where each cell is represented by a field variable. I shall discuss possible choices of forces acting on the cells, in particular polar driving forces and intercellular activity, show how these affect the movement of confluent cell layers and compare the model results to experiments. There are many open questions. Can coarse-grained physical models give useful insight into complex biological processes? What is the most physical way to model the forces acting between cells? What exactly is contact inhibition of locomotion and what initiates the flocking of a layer of cells?

View video

Chirality disorder in the Vicsek model
Bruno Ventejou, CEA Saclay

We study how chirality disorder affects the collective motion ordered phases of the Vicsek model. We present results on two cases, a Gaussian and a bimodal distribution of chirality. In both cases, we observe that the long-range ordered Toner-Tu liquid phase does not resist chirality disorder, whereas the coexistence phase made of traveling bands persists under finite disorder. Two new micro-phase separated regimes emerge, where spontaneous sorting of negative and positive chirality leads to clockwise and counter clockwise vortices or rotating polar packets.

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Cell motility and polarity controls notch signaling pathway patterns in active tissues
Supriya Bajpai, IITB-Monash Research Academy

Cell motility and polarity dynamics in active tissues plays a very important role during the development of multicellular organisms. Intercellular signaling interactions regulate collective cell motion by allowing cells to communicate with each other by transmitting signals. A key question is how collective cell movement itself influences the spatiotemporal intercellular signaling patterns in tissues. On the one hand, the motility of cells coupled with their polarity can lead to collective motion patterns. On the other hand, intercellular signaling is responsible for generating spatial patterns of the signaling molecules. Although modeling efforts have, thus far, these two processes separately, experiments in recent years suggest that these processes influence each other. Hence, we present a model to study how the dynamics of cell motility coupled with cell polarity and intercellular signaling influence the spatiotemporal patterning of signaling molecules.
We observe a rich variety of spatiotemporal patterns of signaling molecules that is influenced by the cell motility polarity and signaling dynamics of the cells. We also observe that the collective motion of signaling patterns is due to the combined effect of the individual cell motion and spatiotemporal shift in signaling molecules that leads to an emergent time-scale of spatial rearrangement of the patterns.

View video

Mixed-Gen Season 1 Session 3: Quantum Monte Carlo

David Ceperley, University of Illinois Urbana-Champaign
Juha Tiihonen, Oak Ridge National Laboratory
Michele Ruggeri, CNRS
March 25, 2021

Simulating quantum systems
David Ceperley, University of Illinois Urbana-Champaign

In this talk I will present an overview of quantum Monte Carlo methods which have been used to simulate many-body continuum systems, with emphasis on the Diffusion Monte Carlo and the Path Integral Monte Carlo methods and their applications to the Wigner Crystal, superfluid helium and dense hydrogen.

View video

Prospects of Forces and Geometry Optimization with Quantum Monte Carlo
Juha Tiihonen, Oak Ridge National Laboratory

Quantum Monte Carlo methods (QMC) describe the potential energy surface (PES) of strongly correlated structures with high accuracy and efficiency. However, the QMC forces face challenges due to systematic biases and statistical cost, which can be controlled but not fully mitigated with state-of-the-art estimator techniques. We show novel scaling relations of QMC force variances with the effective valence charge Z_{eff}, along with an outlook of how large and heavy systems might be within reach for the modern applications of the QMC forces [1].
Additionally, we present a novel energy minimization technique [2] to relax multiple structural parameters using sparse sampling of the DMC PES. It uses density functional theory as a surrogate to obtain the optimal, the most rapidly converging line-search directions in the reduced parameter space. Resampling techniques are used on the surrogate model to predict and further minimize the computational cost of the DMC PES search. The approach can be generalized to any method preferring accurate and possibly noisy PES rather than forces. We present examples of the method as applied to flake-like 2D molecules and 2D monolayers.

References
[1] J. Tiihonen, R. C. Clay III, and J. T. Krogel, Towards Quantum Monte Carlo Forces on Heavier Ions: Scaling Properties*, in preparation (2021)*
[2] J. Tiihonen, and J. T. Krogel, Surrogate Hessian Accelerated Structural Optimization*, in preparation (2021)*
**This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division, as part of the Computational Materials Sciences Program.*

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Quantum Monte Carlo determination of the principal Hugoniot of deuterium
Michele Ruggeri, CNRS,

We show Coupled Electron Ion Monte Carlo results for the determination of the principal Hugoniot of deuterium. Particular care is given to the determination of the reference initial state, as well as to the evaluation of the influence of finite temperature and nuclear quantum effects on the computed Hugoniot curve. Our QMC results are in agreement with experimental data in low T, P conditions, while at higher T, P the computed Hugoniot is still more compressible than experimentally observed.

Reference
*[1] M. Ruggeri, M. Holzmann, D. Ceperley, C. Pierleoni, Phys. Rev. B, **102**, 144108 (2020)*

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Claudia Draxl, Humboldt University Berlin
Adam Hospital, IRB Barcelona
Pierre-Paul De Breuck, Université catholique de Louvain
April 8, 2021

From data to knowledge
Claudia Draxl, Humboldt University Berlin

Research data paired with Artificial Intelligence (AI) enable a new quality of science. The ultimate goal in our research domain is to predict novel candidate materials for a given application, possibly even in regions of the materials space that no-one would think of. For a real breakthrough, key prerequisites have to be brought together: Data – not only Big but most relevant and reliable – and novel AI tools with predictive power. In this session, we will review where we are on this road. A special focus will be on recently-developed data-analysis tools.

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BioExcel-CV19: a database of COVID-19 related Molecular Dynamics trajectories
Adam Hospital, IRB Barcelona

BioExcel-CV19 is a platform designed to provide web-access to atomistic-MD trajectories for macromolecules involved in the COVID-19 disease. BioExcel-CV19 main objective is to generate a tool for scientists interested in the COVID-19 research to interactively and graphically check key structural and flexibility features stemming from MDs. As these features vary depending on the structure analyzed, specific analyses were performed, uploaded to the database, and represented in the web portal. These analyses and key features were collected by direct interaction with the simulation's authors. As an example, trajectories corresponding to the virus Receptor Binding Domain (RBD) attached to the human Angiotensin Converting enzyme 2 (hACE2), the RBD-hACE2 complex, include interface observables (e.g., residue distances, hydrogen bonds), allowing an easy analysis of their behaviour along the simulation. The project is part of the open access initiatives promoted by the world-wide scientific community to share information about COVID-19 research.
All data produced by the project is available to download from an associated programmatic access API. Access: https://bioexcel-cv19.bsc.es/#/

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MODNet: accurate and interpretable property predictions for limited materials datasets by feature selection and joint-learning
Pierre-Paul De Breuck, Université catholique de Louvain

In order to make accurate predictions of material properties, current machine-learning approaches generally require large amounts of data, which are often not available in practice. In this work, a novel all-round framework is presented, named MODNet, which relies on a feedforward neural network, the selection of physically-meaningful features and, when applicable, joint-learning. Next to being faster in terms of training time, this approach is shown to outperform current graph-network models on small datasets. In particular, the vibrational entropy at 305K of crystals is predicted with a mean absolute test error of 0.009 meV/K/atom (four times lower than previous studies). Furthermore, joint-learning reduces the test error

compared to single-target learning and enables the prediction of multiple properties at once, such as temperature functions. Finally, the selection algorithm highlights the most important features and thus helps understanding the underlying physics.

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Mixed-Gen Season 1 Session 5: Machine Learning

Michele Ceriotti, EPFL
Dávid Péter Kovács, University of Cambridge
Yasemin Bozkurt Varolgünes, Max Planck Institute for Polymer Research
May 6, 2021

The thin line between physics and data
Michele Ceriotti, EPFL, Lausanne

As it has done with many other fields of science, machine learning has taken molecular and materials modeling by storm. There is virtually no simulation task to which machine-learning techniques have not been applied, usually very successfully.
In this talk I will take one step back and look at the relationship between this inductive, data-driven modeling paradigm, and the traditional physics-based approaches. I will take on both a historical and a conceptual perspective. First, I will put the latest wave of machine-learning potentials and models in the context of well-established atomistic simulation techniques. Then, I will discuss the interplay between general-purpose statistical learning ideas and the domain-specific insights that can, and should, be applied to obtain accurate and transferable predictions.
From the description of long-range interactions between atoms and molecules, to the estimation of structural and functional properties of materials in realistic conditions, the future of atomic-scale simulations straddles the line between data and physics-driven modeling.

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Atomic Cluster Expansion force fields for organic molecules: evaluation beyond RMSE
Dávid Péter Kovács, University of Cambridge

The efficient simulation of molecules and materials from first principles is a long-standing challenge in the physical sciences. Machine learned force fields promise to speed up quantum mechanical simulations by several orders of magnitudes, whilst maintaining the accuracy of high-level quantum mechanics. In the past 3 years several new approaches were proposed to fulfill this promise built on Gaussian Process Regression and Neural Networks. In this poster we demonstrate that highly accurate molecular force fields can be built using the Atomic Cluster Expansion (ACE) framework and linear least squares regression. Our model is built from body ordered symmetric polynomials, which are a natural extension of the traditional molecular mechanics force fields. We show that these relatively simple models are able to achieve state of the art accuracy on the MD17 benchmark dataset of small organic molecules. Furthermore, we also train several other machine learning models like sGDML, ANI and GAP, as well as a classical force field and compare them on tasks such as normal mode prediction and extrapolation to high temperature data. Finally, we fit the potential energy surface of a large flexible organic molecule. We show that the ACE model shows excellent transferability

across temperatures, and we compare how well the different models reproduce the complex dihedral torsional energy landscape of the molecule form as little as 500 reference calculation.
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Automated identification of collective variables and metastable states from molecular dynamics data
Yasemin Bozkurt Varolgünes, Max Planck Institute for Polymer Research

Extracting insight from the enormous quantity of data generated from molecular simulations requires the identification of a small number of collective variables whose corresponding low-dimensional free-energy landscape retains the essential features of the underlying system. Data-driven techniques provide a systematic route to constructing this landscape, without the need for extensive *a priori* intuition into the relevant driving forces. In particular, autoencoders are powerful tools for dimensionality reduction, as they naturally force an information bottleneck and, thereby, a low-dimensional embedding of the essential features. While variational autoencoders ensure continuity of the embedding by assuming a unimodal Gaussian prior, this is at odds with the multi-basin free-energy landscapes that typically arise from the identification of meaningful collective variables. In this work, we incorporate this physical intuition into the prior by employing a Gaussian mixture variational autoencoder (GMVAE), which encourages the separation of metastable states within the embedding. The GMVAE performs dimensionality reduction and clustering within a single unified framework, and is capable of identifying the inherent dimensionality of the input data, in terms of the number of Gaussians required to categorize the data. We illustrate our approach on two toy models, alanine dipeptide, and a challenging disordered peptide ensemble, demonstrating the enhanced clustering effect of the GMVAE prior compared to standard VAEs. The resulting embeddings appear to be promising representations for constructing Markov state models, highlighting the transferability of the dimensionality reduction from static equilibrium properties to dynamics.

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Mixed-Gen Season 1 Session 6: Activated Events

Alessandro Laio, SISSA, Trieste
Matej Badin, SISSA; Comenius University
Eugen Hruska, Emory University
Isabell Louise Grothaus, University Bremen
June 3, 2021

Estimating the free energy: from enhanced sampling to manifold learning
Alessandro Laio**,** SISSA, Trieste

Multidimensional free energy landscapes are able to provide a synthetic description of complex molecular systems, revealing their salient features. In the last decades we have witnessed the development of several powerful enhanced sampling methods, and estimating the free energy as a simultaneous function of 2-3 variables is nowadays routine. However, in many relevant systems one cannot project the free energy to a low-dimensional space without artificially merging distinct free energy minima, therefore losing relevant information. But how can one find the minimal number of variables which are really necessary to describe a system? How can one choose these variables among a set of many candidates? Can one compute the free energy as a simultaneous function of, say, 10 variables? And how can one visualize

and interpret a 10-dimensional landscape? We will illustrate how these questions can at least partially be addressed exploiting tools offered by unsupervised manifold learning and dimensional reduction.

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Nucleating different coordination in crystal under pressure: Study of B1-B2 transition in NaCl by metadynamics

Matej Badin, SISSA; Comenius University

Prediction of crystal structures has reached a high level of reliability, but much less is known about the mechanisms of structural transitions and pertinent barriers. The barriers related to nucleation of crystal structure inside another one are critically important for kinetics and eventually decide what structure will be created in experiment.
We show here an NPT metadynamics simulation scheme [1] employing coordination number and volume as collective variables and illustrate its application on a well-known example of reconstructive structural transformation B1/B2 in NaCl. Studying systems with size up to 64000 atoms we reach beyond the collective mechanism and observe the nucleation regime. We reveal the structure of the critical nucleus and calculate the free energy barrier of nucleation and also uncover details of the atomistic transition mechanism and show that it is size-dependent.
Our approach is likely to be applicable to a broader class of structural phase transitions induced by compression/decompression and could find phases unreachable by standard crystal structure prediction methods as well as reveal complex nucleation and growth effects of martensitic transitions.

Reference

[1] Matej Badin and Roman Martoňák, arXiv:2105.02036

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Benchmarking the accuracy of free energy landscapes generated by adaptive sampling strategies

Eugen Hruska, Emory University

Verifying the accuracy of free energy landscapes generated by sampling methods is limited due to the limited number of test systems. We investigate the accuracy of adaptive sampling methods on a test set of proteins with sizes ranging up to 70 amino acids, establishing a baseline of accuracy demonstration. Besides the free energy landscape; the convergence of kinetic results from sampling methods is crucial. The kinetic convergence, upper limits for sampling performance, and scaling of these sampling methods are considered.

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N-glycan conformers explored by enhanced sampling and machine learning
Isabell Louise Grothaus, University Bremen

Glycosylation is one of the bulkiest post-translational modification of proteins but has long been overlooked in molecular dynamics simulations, despite its omnipresence in the cell. However, the structure, function and interaction of many biochemical systems is governed by N-glycans covalently linked to asparagine residues in specific protein sequences. Due to the flexibility of their glycosidic linkages and their sugar units, N-glycans assume many different conformations, unlike the more rigid protein structure to which they are attached. A complete description of their conformational phase space requires thus the consideration of a large number of internal degrees of freedom. We show that an enhanced-sampling molecular dynamics scheme based on enhancing transitions of all relevant barriers with concurrent one-dimensional energy potentials in the framework of metadynamics can in fact capture effectively global conformers of branched glycans, importantly also including the monomer puckering states. Interestingly, our approach revealed altered N-glycan conformer populations depending on the puckering state of individual monosaccharides. These puckering-dependent conformer distributions, so far mostly ignored in glycoprotein simulations, might be crucial in explaining biological phenomena involving N-glycans.

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Mixed-Gen Season 2 Session 1: Multiscale simulations of complex materials

Friederike Schmid, Johannes Gutenberg University
Nicodemo Di Pasquale, University of Manchester
Javier Diaz, EPFL
Constantinos J. Revelas, National Technical University of Athens
October 27, 2021

Polymer simulations across multiple scales
Friederike Schmid**,** Johannes Gutenberg University

Polymers, i.e., macromolecules, are omnipresent in nature and technology, yet their simulation still represents a challenge. This is because the behavior of polymeric systems is governed by processes on a multitude of length scales ranging from Angstrom (chemical structure) to micrometers or more (morphologies of polymeric materials). By now, many decades of research efforts have been dedicated to developing efficient simulation methods for polymeric systems, yet new ideas and methods still keep emerging. In the lecture I will give a brief introduction into some basic concepts of polymer theory and then discuss polymer models on different scales, with some bias towards polymer field theory.

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Mori-Zwanzig projection technique in coarse-grained modelling
Nicodemo Di Pasquale, University of Manchester

Despite their successes, Fine-Grained (FG) or atomistic models suffer severe limitations in terms of the time- and length-scale accessible to investigation. Therefore, many real- world applications, especially in the class of soft matter materials, remain inaccessible to fully resolved simulations, due to their unsustainable computational costs, and must rely on semiempirical Coarse-Grained (CG) models. Significant efforts have been devoted in the last

decade towards improving the predictivity of these CG models and providing a rigorous justification of their use, through a combination of theoretical studies and data- driven approaches.
In this talk, we will explore one of the most promising frameworks to obtain such rigorous justification, the projection operator technique, developed by Mori and Zwanzig (MZ) in the context of non-equilibrium statistical mechanics. The Mori-Zwanzig (MZ) projection is a generic, yet systematic, theoretical tool for deriving coarse-grained models.
We will show that the dynamics of the beads is described by a Generalized Langevin Equation (GLE) obtained from the full FG description, in which a precise physical meaning can be given to each of its terms. In particular, we derive the fact that some of the well-known bottom-up methodologies used to obtain the CG interactions (i.e., IBI models [1], force matching [2], relative entropy models [3]) are all included within such MZ description [4]. We will then explore the problem of the memory in CG models, and propose a rational basis for a data-driven approach to an approximation of the memory and fluctuating terms in such GLE, which can be considered included in the class of the Markovian models [5].

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*[4] N. Di Pasquale, T. Hudson, M. Icardi, Phys. Rev. E, **99**, 013303 (2019)*
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Co-assembly of block copolymers and anisotropic nanoparticles
Javier Diaz, EPFL

Block copolymer (BCP) melts can self-assemble into ordered periodic morphologies (lamellar, BCC spheres, hexagonal cylinders, …), which has been exploited to control the localization of colloidal nanoparticles (NPs). Additionally, BCP domains can template the alignment of NPs with orientational degrees of freedom such as nanorods. In this work we computationally study the role of shape anisotropy with a generalized model that simulates colloids with a variety of shapes (spheroids, rhomboids, nanorods). Simulation results are compared with experimental setups both in 2D and in 3D, finding good agreement and novel co-assembled structures, thanks to the rich phase behavior of BCP melts and the various NP length scales.
Highly elongated ellipsoids are shown to induce a sphere-to-cylinder phase transition in the BCP, due to the symmetry-breaking effect in the vicinity of the nanoellipsoid.

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Multiscale Modeling of Polymer-Nanoparticle Systems
Constantinos J. Revelas, National Technical University of Athens

We present a multiscale simulation strategy for the prediction of the structure and thermodynamics of polymeric interphases (i.e., polymer-vacuum, polymer-polymer and polymer-solid) and polymer nanocomposites. The hierarchical multiscale strategy comprises three levels of description: i) atomistic Monte Carlo and Molecular Dynamics simulations, ii) mesoscopic self-consistent field calculations, and iii) macroscopic hybrid particle-field simulations. The intermediate level of the multiscale strategy is based on self-consistent field

calculations, which enable derivation of the potential of mean force that describes the repulsion and agglomeration tendencies of bare or polymer-grafted nanoparticles. Using these potentials one could undertake hybrid particle-field simulations to describe the phase behavior at macroscopic time and length scales.

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Mixed-Gen Season 2 Session 2: Machine learning in simulations

Gabor Csanyi, Cambridge University
Thorben Fröhlking, **Giovanni Bussi**, SISSA
Silvia Bonfanti, Università degli Studi di Milano
November 25, 2021

First principles force fields: status and challenges

Gabor Csanyi**,** Cambridge University

Over the past decade a revolution has taken place in how we do large scale molecular dynamics. While previously first principles accuracy was solely the purview of explicit electronic structure methods such as density functional theory, the new approaches have allowed the extension of highly accurate, first principles simulations to the atomic scale, where electrons are not treated explicitly anymore, and therefore hundreds of thousands of atoms can be simulated. These quantum mechanically accurate force fields and interatomic potentials are fitted to electronic structure data and at first used techniques inspired by those used in machine learning and artificial intelligence research: neural networks, kernel regression, etc. It is a quickly moving field, and - having learned key lessons about representation, symmetry and regularisation - there appears to be some semblance of convergence between the diverse methods, which now also include polynomial expansions carried to high dimension.

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Using machine learning to improve RNA force fields

Thorben Fröhlking, Giovanni Bussi**,** SISSA

The capability of current force fields to reproduce structural dynamics in agreement with experiment is limited. In the past years, several methods have been developed to take advantage of experimental data in order to enforce agreement with experiments. Since Maximum-entropy methods are limited with respect to the choice of the corrections functions, the approach introduced in Cesari et al., J. Chem. Theory Comput. 2019, 15, 6 provided a more flexible method. This approach allows arbitrary force-field terms to be corrected, and arbitrary ways to quantify agreement with experiment, together with a robust regularization protocol to avoid overfitting. We here extend this framework introducing and comparing a number of different regularization strategies (L1, L2, Kish Size, Relative Kish Size, Relative Entropy). The protocol is applied to difficult RNA systems, namely GACC, ccGAGAgg and ccUUCGgg, where specific intramolecular hydrogen bonds in the AMBER RNA force field proposed by Kührová, et al., J. Chem. Theory Comput. 2019, 15, 5 are corrected with automatically determined parameters. The identified parameters are in agreement with recently suggested corrections Mráziková et al., J. Chem. Theory Comput. 2020, 16, 12 as well as the experimental NMR data. Simulations involving systems present in the training set, but also unknown systems, using identified parameters display improvements regarding native population of Tetraloops as well as good agreement with NMR-experiments for Tetramers.

Furthermore, we test the possibility to introduce non-linear corrections in the form of two-layers artificial neural networks (ANN) applied on pairs of consecutive dihedral angles. By testing on an extended dataset, we find that the linear corrections on dihedral angles introduced in Cesari et al., J. Chem. Theory Comput. 2019, 15, 6 as well as introduced ANN potentials significantly reduce existing discrepancies between experiments and simulations regarding NMR observables.

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Machine Learning of Metamaterials

Silvia Bonfanti, Università degli Studi di Milano

Mechanical metamaterials are a new class of materials with exceptional properties derived from their designed internal geometry composed of multiple sub-elements or cells, rather than their composition. Through prudent choice of cells arrangement, mechanical metamaterials find a straightforward application as actuators that achieve predetermined input-output operations and can be 3D printed in a single block, with the advantage of removing the assembly phase since there is no longer need for all structural components. Despite the rapid progress in the field, there is still a need for efficient strategies to optimize metamaterial design for a variety of functions.
Here, we first present a computational method for the automatic design of mechanical metamaterial actuators that combines a reinforced Monte Carlo algorithm with discrete element simulations. The method allows us to get machine-generated structures that can reach high efficiency in terms of functionality, exceeding human-designed structures.
Next, we show that it is possible to design novel efficient actuators by training a deep neural network which is then able to predict the efficiency from the image of a structure, and to identify its functional regions which are those mechanically relevant.
The use of machine learning to assist the automatic design of mechanical metamaterial actuators opens intriguing possibilities in terms of algorithmic speed, as it could potentially allow to design larger structures that cannot be efficiently simulated by conventional methods, increasing the complexity for countless engineering applications.

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Emanuela Zaccarelli, La Sapienza University
Asaf Szulc, Ben-Gurion University
Fergus Moore, University of Bristol
Luca Tonti, University of Manchester
December 16, 2021

Anomalous slow dynamics in soft matter
Emanuela Zaccarelli**,** La Sapienza University

Colloidal suspensions form a wide variety of arrested states, from gels to glasses of various kinds. After a short introduction, I will focus on examples where slow dynamics becomes "anomalous", i.e., not characterized by a classical two-step behavior, but by a logarithmic dependence of the density auto-correlation functions and by a power-law (subdiffusive) behavior of the mean-squared displacement. I will describe a few case studies, from short-ranged attractive colloids to binary mixtures, where this behavior has been reported, highlighting the important role of computer simulations.

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Cooperative effects driving the multi-periodic response of cyclically sheared amorphous solids
Asaf Szulc, Ben-Gurion University

Plasticity in amorphous materials, such as glasses, colloids, or granular materials, is mediated by local rearrangements called "soft spots." Experiments and simulations have shown that soft spots are two-state entities interacting via quadrupolar displacement fields generated when they switch states. When the system is subjected to cyclic strain driving, the soft spots can return to their original state after one or more driving cycles. The response is multi-periodic when the system repeats after more than one driving cycle. To better understand the physical mechanisms behind multi-periodicity, we use a model of interacting hysterons. Each hysteron is a simplified two-state element representing hysteretic soft-spot dynamics. We show how interactions generate cooperative effects that allow multi-periodic cycles to emerge. The nature of the mechanisms that we uncovered has significant implications for the understanding, the modeling, and the uses of plasticity in amorphous solids.

Reference
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Crystallisation and polymorph selection in active Brownian particles
Fergus Moore, University of Bristol

We explore crystallisation and polymorph selection in active Brownian particles with numerical simulation. In agreement with previous work (Wysocki et al. in Europhys Lett 105:48004, 2014), we find that crystallisation is suppressed by activity and occurs at higher densities with increasing Péclet number (*Pe*). While the nucleation rate decreases with increasing activity, the crystal growth rate increases due to the accelerated dynamics in the melt. As a result of this competition, we observe the transition from a nucleation and growth regime at high *Pe* to

“spinodal nucleation” at low *Pe*. Unlike the case of passive hard spheres, where preference for FCC over HCP polymorphs is weak, activity causes the annealing of HCP stacking faults, thus strongly favouring the FCC symmetry at high *Pe*. When freezing occurs more slowly, in the nucleation and growth regime, this tendency is much reduced and we see a trend towards the passive case of little preference for either polymorph.

Reference
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Diffusion of globular macromolecules in liquid crystals of colloidal cuboids
Luca Tonti, University of Manchester

It is already known that macromolecular motion in intracellular environment is strongly hindered by the crowded environment in which they display, and this effect is known as macromolecular crowding. However, less effort has been invested in how the structural organization of environment influences the motion of guest particles. To this end, we investigated the dynamics of a model system of colloidal small hard spheres (HSs) immersed in isotropic and nematic suspensions of prolate and oblate hard board-like particles (HBPs), i.e., cuboids, performing Dynamic Monte Carlo simulations [1]. We found similar features in the dynamics of both HBPs and HSs: from slower diffusion in denser systems, to anisotropic diffusion in anisotropic phases. Anomalous diffusion has been observed at intermediate and longtime scales for both HSs and HBPs, with deviation from expected Gaussian particle displacement distributions for some cases, also in isotropic phases. We also found nematic-like clutters of HBPs in isotropic phases, whose local preferential orientation we suppose can explain observed non-Gaussian behaviour [2].

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