



Thomas Young Centre Student Day 2022

Tuesday 10th May 2022

10.00 to 17.00

10:00 - 10:05	Introduction
Session One chaired by Kemal Atalar	
10:05 - 10:20	Structural origins of the low-temperature orthorhombic-to-tetragonal phase transition in high-T_c cuprates Christopher Keegan, imperial College London
10:20 - 10:35	Ultralong organic phosphorescence and thermally activated delayed fluorescence in phenone-carbazole derivatives Amir Sidat, Queen Mary University of London
10:35 - 10:50	An insight into Sr₂Sb₂O₇ as a potential n-type thermoelectric material Luisa Herring Rodriguez, University College London
10:50 - 11:05	High-temperature superconductivity in the lanthanide hydrides at extreme pressures Yao Wei, Kings College London
11:05 - 11:20	Refreshments
Session Two chaired by William Morton	
11:20 - 11:35	Charge transport in bulk hematite and at the hematite/water interface Christian Ahart, University College London
11:35 - 11:50	Multiscale screening of plasmon enhanced photocatalysts Robert Michael Jones, Kings College London
11:50 - 12:20	Sam Tippetts, UKAEA
12:20 - 14:00	Lunchbreak
Session Three chaired by Chengcheng Xiao	
14:00 - 14:15	Optimum reaction conditions in RAFT copolymerisations based on kinetic Monte Carlo Fiona Sander, Imperial College London
14:15 - 14:30	Multi-scale Modelling of Conjugated Polymers to Understand the Role of Side Chain Chemistry in Mixed Transport Conduction Nicholas Siemons, imperial College London
14:30 - 14:45	Shaping the Ripples: Exploring Routes to Nanoengineer the Morphology of Graphene Fabian Thiemann, University College London
15:00 - 15:15	An atom centred representation of the RPA density-density response function Mario Zauchner, Imperial College London
15:15 - 15:30	Refreshments
Session Four chaired by Robert Jones	
15:30 - 16:00	Lara Roman Castellanos - DeepMind
16:00 - 16:15	(quantum) Gaussian Process States - A Bayesian learning perspective for representing many-body wavefunctions Yannic Rath, Kings College London
16:15 - 16:30	Monte Carlo Simulations for Fitting Neutron Diffraction Data Camilla Di Mino, University College London
16:30 - 16:35	Closing remarks ,
16:35 - 17:00	Posters
17:00 - 17:05	Winning poster / talk announcement
17:05 - 18:00	Drinks reception

Plenary Presentations

Sam Tippetts, UKAEA

Fusion energy is entering a golden age, with more public and private facilities being built than ever before, and the United Kingdom Atomic Energy Authority (UKAEA) is at the forefront of international research and development. We operate two world leading experiments, JET and MAST Upgrade, that are paving the way for future fusion power plants, and are both undergoing the most exciting phases of their operational histories. Fusion research and development is heavily dependent on Engineering Analysis, High-Performance Computing (HPC) and Materials Research, and we will be showcasing the work of our specialists in these areas, as well as discussing the benefits of working at UKAEA. There will be opportunity to ask questions of our panellists and to get into contact for careers information.

Lara Roman Castellanos, DeepMind

After finishing my PhD in nanoplasmonics, I joined DeepMind as a research scientist. I will talk about my career path to DeepMind, how is it to do research in a non-academic environment and I will give an overview of our recent publication: a neural network functional for Density Functional Theory, DM21, which solves the fractional electron problem and has state of the art performance on main chemistry benchmarks.

Poster Presentations

Anharmonic lattice dynamics of superionic lithium nitride

Gabriel Krenzer

Ionic transport in conventional (non-superionic) solids is usually considered as being effected by a sequence of discrete “hops”, where individual mobile ions undergo stochastic moves between available crystallographic sites. While this model is usually appropriate for ionic solids with low-to-medium ionic conductivities, fast ionic conduction is often associated with highly-concerted ionic motion, in which groups of ions undergo cooperative near-simultaneous motion. This observational correlation between fast-ion conduction and cooperative ion-transport mechanisms suggests that ionic motion in superionic conductors might be better described in terms of appropriate collective degrees of freedom, rather than independent single-atom coordinates. One appealing choice of basis for describing collective dynamics in a crystal is the set of normal modes of vibration, which correspond to phonon eigenvectors. It has also been proposed that superionic transitions are accompanied by the breakdown of specific phonon modes linked to characteristic diffusion processes. Any changes in vibrational properties across the superionic transition may therefore also provide insights into the underlying physics of this phenomenon. Here, we apply a combination of lattice dynamics, ab initio and classical molecular dynamics to probe the vibrational properties of the archetypal superionic conductor Li₃N. We assess harmonic, quasiharmonic, and anharmonic descriptions of the phonons. The harmonic and quasi-harmonic models show no change in features across the superionic transition. The anharmonic model, however, exhibits a breakdown for all modes. The implications for developing lattice-dynamics-based descriptors for superionic conductors are discussed.

Efficient electron open boundaries for electrochemical applications

Margherita Buraschi

A major challenge in modelling interfacial processes in electrochemical (EC) devices is the need to perform simulations at constant potential. This means an open-boundary description of the electrons is needed so that electrons can enter and leave the computational cell. To enable realistic modelling of EC processes under potential control we have introduced the hairy probe method in the weak coupling limit [1]. This is an efficient open-boundary formalism appropriate for electrochemical problems and is compatible with Density Functional Theory. The implementation we have developed will be employed initially to model electrochemical nanojunctions: these are three terminal functional devices of great interest because of their switching functionality. However, the formalism can be applied to all electrified interfaces. Enabling efficient potential control in a simulation, our work paves the way to make substantial steps forward in our understanding of EC phenomena.

[1] Zauchner M, Horsfield AP, Todorov T (2018) Efficient electron open boundaries for simulating electrochemical cells, Physical Review B, 97 045116 (<https://doi.org/10.1103/PhysRevB.97.045116>)

Fantastic electriles and where to find them

Chengcheng Xiao

Electriles are materials with electrons localized on interstitial sites in the crystal lattice and exhibit an array of interesting properties that show promise for applications such as catalysts, electron emitters and superconductors. Electriles can be found among many different classes of materials, including elemental metals at high pressure, organic crystals, intermetallic compounds and ceramic materials.

In this work, we present a general theory that provides a unified understanding of the origin of interstitial electrons in all currently known types of electriles. Using this theoretical understanding, we have devised an automated high-throughput algorithm based on first-principles electronic structure calculations for identifying systems that are potentially electriles. Applying our workflow to the ~55,000 materials in the Materials Project crystal structure database, we identified ~10,000 potential candidates. These candidate systems demonstrate a smooth transition from the typical (inter)metallic systems to prototypical electrile systems and the results show that the chemical bonding of electrile materials can be categorized multicentered bonding.

Degradation And Dielectric Breakdown In Modern HfON Based Devices

Teo Cobos

The performance of day-to-day devices are governed by the reliability of the metal-oxide-semiconductor field-effect transistors (MOSFETs) and ever so often they are affected by random telegraph noise and bias temperature instability. These are caused by the trapping and release of holes and electrons found in the devices' architecture, but the atomistic nature of these processes remains unclear. As the dimensions of the devices get smaller as advances in MOSFETs continues, we are reaching a limit where a single charging and discharging event is the predominant factor of device degradation. This research focuses on the molecular modelling of the effect of Negative Bias Temperature Instability (NBTI) in c-Si/a-SiO₂/a-HfO₂ devices using Density Function Theory (DFT) methods. This work investigates a wide range of defects that trap holes and possible candidates for NBTI in bulk a-SiO₂ and in the Si/SiO₂ and Si/SiO₂/HfO₂ interface and aims to evaluate the accuracy of the Reaction Diffusion Drift (RDD) model which invokes the dissociation of hydrogen passivated defect precursors and can explain the directly measured trap generation kinetics across many types of devices. We have found two possible candidates for the initial trap assisted hydrogen dissociation mechanism and have formulated a complex mechanism to which further defect generation can occur.

The Mixing Behavior of MgSiO₃ and CaSiO₃ Melts

Isaac Taschimowitz

During Earth's early formation, the solid rocky mantle seen today was, in fact, molten. We call this the magma ocean. The exact details of Earth's transition from liquid to solid are largely unknown, however the effects of this transition can possibly be observed today such as ultra-low velocity zones (ULVZs) at the base of the mantle. It is crucially important to understand the processes governing this transition to fully understand Earth today and even to model the future evolution of our planet. One of the key processes involved is the transport of the crystallizing minerals, where the controlling factor is the density relationship between the solid and liquid phases. For example, if minerals crystallize and are neutrally buoyant in the melt, they may form a layer which splits the magma ocean in two, forming an upper and basal ocean which evolve separately. However, constraining the density of the melt phases has been challenging due to its evolving composition. Therefore, we aim to develop a model to give the volume of the melt for a given composition, pressure, and temperature. As volume does not always vary linearly between endmembers, the mixing behaviour of the melts must be investigated. Modelling these mixing volumes also allows understandings into the change of Gibbs free energy across composition, which could affect other properties such as melting temperatures. We present results from DFT calculations, giving the mixing behaviour between the endmembers of MgSiO₃ and CaSiO₃. A third-order Birch-Murnaghan equation of state was fitted to the results of various compositions (between and including the endmembers), which allow volumes to be obtained across composition for a fixed pressure and temperature. By plotting these volumes against composition, the mixing behaviour for various pressures and temperatures were found. We find that the mixing behaviour between the endmembers MgSiO₃ and CaSiO₃ can be considered ideal.

Free Energy Surfaces from sets of asynchronous enhanced molecular dynamics simulations subject to multiple biases

Antoniu Bjola

Molecular dynamics (MD) simulations have become a powerful tool to study and predict the thermodynamics of molecular systems due to great advances in algorithm development, as well as an explosive growth of computational power. These simulations allow scientists to develop unique insight into the behavior of complex systems with atomistic detail, and provide a route to obtain average properties of interest, allowing to bridge the gap between microscopic mechanisms and experimental observables. Often, however, molecular systems contain multiple metastable states separated by high free energy barriers, rendering the simulation of some systems unfeasible.

To address this issue, numerous methods have been proposed. Two widely used methods are umbrella sampling (US) and metadynamics (MetaD), which introduce external bias potentials that perturb the Hamiltonian of the system to improve the sampling of the molecular system. While US makes use of time-independent, attractive bias potentials that enable parallel simulations, MetaD introduces a history dependent repulsive bias potential that favors the exploration of configuration space.

With this work we further develop Mean Force Integration (MFI) to combine the parallelization of US, with the autonomous exploration of configuration space given by MetaD. Using MFI, we compute Free Energy Surfaces (FES) from sets of multiple, asynchronous simulations subject to time-dependent and/or time-independent biases. Furthermore, we provide an on-the-fly estimate of the convergence of the local mean force and demonstrate the calculation of the position-dependent error of FES by bootstrapping.

We demonstrate the effectiveness of our approach by computing FES for a range of systems including analytical Langevin models, single-molecule systems, and large collective processes such as the nucleation of a liquid phase from a supersaturated vapour. Finally, we show that based on MFI it is possible to radically improve the scaling of performances for biased sampling methods on large computational platforms.

Exploring Ligand Binding in a Glycine-Gated Ion Channel with Molecular Dynamics and Enhanced Sampling Methods

Jacob Clark

Ion channels are fundamental units of the nervous system responsible for mediating intercellular signals and propagating action potentials: this makes them critical in brain function, disease and pharmacology. Pentameric Ligand Gated Ion Channels, like the glycine receptor, are responsible for modulating the process fast synaptic communication.

Understanding the activation of these channels requires detailed structural and functional investigation, which is limited by current experimental techniques. Atomistic simulations can overcome these limitations by providing atomic resolution dynamics. Here we investigate an atomistic model of the glycine receptor using molecular dynamics, with the goal to elucidate the interaction network of the neurotransmitter within the binding pocket. The model was developed from a glycine bound cryo-EM structure.

The timescales of binding and unbinding are beyond the capabilities of standard molecular dynamics. Therefore, to investigate these rare events, we use the enhanced sampling methods metadynamics to overcome molecular dynamics limitations in the exploration of glycine binding and unbinding processes.

Data-Driven Design of Nature-Inspired Photocatalytic Materials

Gowan Whalley

Inspiration is taken from nature in the design of materials for more efficient photocatalytic materials for CO₂ reduction. The goal is the efficient production of two carbon products, such as ethanol. This will revolve around two areas: product selectivity and sensitization efficiency. Cucurbiturils (CB[n]) will be utilized to achieve these aims, these are barrel-shaped molecules with a cavity large enough to fit multiple guests with high binding affinities. These properties allow the formation of structures analogous to the active sites of enzymes and energy-transfer complexes in photosynthesis. Both applications are backed up by encouraging evidence with prior observation of cucurbiturils influencing both product distribution in electrocatalytic CO₂ reduction due to its high affinity for CO₂ molecules and energy transfer efficiency between donor-acceptor pairs. The use of CB to form these Energy Transfer complexes will have a protective effect on the dye molecules, separating them from the catalyst surface to prevent degradation, a common problem in dye-sensitized systems. The active site design approach will revolve around screening potential molecular candidates for their binding affinity to the CB[7] / CB[8] cavity using a machine learning model trained on a dataset of computational calculations. These candidates will then be selected based on their ability to stabilize the intermediates of CO₂ photoreduction and encourage the formation of two carbon products such as ethanol. The photocatalyst material that will be focused on will be titanium dioxide, a common semiconductor used in photocatalysis which whilst having benefits such as low cost and toxicity has disadvantages including the absorption of light exclusively in the UV region of the electromagnetic spectrum and a tendency to form single carbon reduction products. This project aims to tackle these challenges using a combination of Data-Science and Computational Chemistry techniques.

Oxygen vacancies defect of crystalline (α , β , and ϵ)-Ga₂O₃

Chaiyawat Kaewmeechai

Ga₂O₃ is a transparent semiconductor possessing ultrawide bandgap (5 eV), a high breakdown electric field and a large Baliga's figure of merit (BFOM), which is a potential candidate for the power electronics. The three most stable phases of Ga₂O₃ are β , ϵ , and α respectively, which contain different portions in octahedron and tetrahedron. In this work, we investigate geometry optimizations and charge transition levels (CTL) of oxygen vacancy defects in these Ga₂O₃ phases using the density functional theory (DFT) with optimized PBE0-TC-LRC nonlocal functional satisfying the Koopmans' theorem. We found that the CTL changes from 0 to +2 in Ga₂O₃. Then, tetrahedron Ga moves toward the vacancy with a displacement ~ 0.3 Å for the 0 charge state but it moves opposite the vacancy with around ~ 0.9 Å for +2 charge state relative to the perfect atomic position. While the α -phase that contains only octahedrons, two Ga atoms with the shorter bonds move toward and opposite the vacancy with the displacement ~ 3 Å for 0 and +2 charge states respectively. We discuss that the very large displacement is observed in tetrahedron Ga especially +2 charge state due to weaker bonds and lower density in tetrahedron-contained Ga₂O₃ (β and ϵ). By possessing tetrahedron in Ga₂O₃ also decreases density and electronic band gap significantly.

Temperature dependence of the charge mobility in rubrene from fragment orbital-based surface hopping

Jan Elsner

We present the temperature dependence of the charge mobility in single-crystal rubrene, calculated using our fragment orbital-based surface hopping (FOBSH) method. This is a non-adiabatic dynamics method developed by the Blumberger group which makes use of Tully's fewest switches surface hopping and allows us to simulate charge transport on the nanometer scale. This method is ideally suited to study organic semiconductors, where charge transport operates in a regime somewhere in between the traditional limits of band theory and thermally activated hopping. In our application of this method to rubrene, we have carefully parameterized a force field to reproduce the distributions and timescales of electronic coupling matrix elements in the system by comparison to expensive DFT(optPBE-vdW)-MD results. We find excellent qualitative agreement with experiment, as well as near quantitative agreement with transient localization theory, another theoretical framework for studying charge transport in organic semiconductors. This work provides validation of the FOBSH method, as well as providing insight into what drives the temperature dependence in rubrene, namely how the density of states is affected by temperature. This also marks the first step in an exciting new project; the application of the FOBSH method to study thermoelectric physics in organic semiconductors, for example calculation of the Seebeck coefficient.

Ab Initio study of the onset of Al corrosion

Rashid Al-Heidous

Aluminum (Al) is an essential element in several industries such as aeronautics, wastewater treatment, and batteries. All the mentioned industries have limited their use of Al and its alloys, due to pitting corrosion. In this work we investigate the onset of Al corrosion and surface re-oxidation. Using an ab initio, realistic, dynamical representation of the Al(111)-water interface, we have compared the morphology of clean and oxidized Al(111) substrates, and revealed a wealth of interfacial features, including interfacial solvation structures, potential drops, mass, angle and charge distributions. In addition, we have quantified equilibrium coverages and adsorption strength for O and OH species on the metal surface, as a function of the electrode potential; Our findings represent the base for a deeper fundamental understanding of Al corrosion, which will ultimately enable a molecular based engineering of targeted solutions to counteract it.

p-type Impurities and Doping in 4H-SiC

Niamh Smith

To create a p-type 4H-SiC MOSFET device, 4H-SiC is generally bombarded with high energy ions of either Al⁺ or B⁺ during ion implantation. Out of these two dopant species, Al is most preferred as B typically experiences significant in- and out-diffusion. However, several recent experimental studies have discovered that a drop in device channel mobility occurs as, what appears to be, a direct consequence of the implantation of high aluminium implantation dosages into 4H-SiC. The most practical way forward for industry at present would be to use an alternative p-dopant species in the creation of p-type 4H-SiC MOSFET devices. However, no alternative dopants, which possess similar dopant levels to that of aluminium (experimentally found to be ~0.2eV), are currently known. We have conducted investigations, via the use of density functional theory (DFT) calculations, into the incorporation and dopant levels of point defects produced as a result of insertion of fluorine, platinum, and several trivalent atomic impurities (namely B, Ga, In, Tl, and Sc), alongside aluminium, to compare p-dopant potential in the aim to find a suitable p-dopant alternative. Given the shallowness of Al's dopant level, single electron Kohn-Sham energy defect levels, thermal ionisation energies, and total energy differences between charge transferred states of the system have been assessed in the pursuit of obtaining and verifying accurate doping levels for the impurities studied. Impurity point defect electronic structures have also been evaluated. We find that when a point defect's electron density is partially localised on the impurity atom alongside one or two of the impurity atom's neighbouring carbons that Kohn-Sham defect levels tend to be higher than ionisation energies. When electron density is localised on neighbouring carbons only, Kohn-Sham defect levels are found to be lower or approximately equal to ionisation energies. B, Ga, and In are found to all have doping levels below 0.5eV.

Machine Learning Guided Discovery of Catalysts for CO₂ Conversion

Marco Anselmi

The rising levels of CO₂ in Earth's atmosphere is one of the most worrying problems that society must confront. One of the most attractive solutions is the chemical conversion of CO₂ into added-value chemicals and fuels. The main challenge in this solution is in the inertness of CO₂. Thus, making catalyst to reduce the energy required for the conversion is CO₂ is a necessity. Quantum chemistry methods have been widely used to guide the discovery of better and cheaper catalysts through the calculation of the properties of the material and the determination of the elementary steps controlling the kinetic and thermodynamic of the conversion of the reactant. Despite significant advances in quantum chemistry methods and codes, the computational cost associated with screening large numbers of candidate catalysts is still significant. Deep Learning, a subset of Machine Learning, is becoming increasingly popular as the methodology to use to accelerate the design of new materials. One of the most appealing Deep Learning techniques that can be used in computational chemistry is the Geometric Deep Learning technique. This project will thus focus on the development of Geometric Deep Learning techniques as a substitute to conventional computational chemistry methods for the prediction of materials properties.

The activation mechanism of GPR68, a computational study

Christos Matsingos

GPR68 is an orphan GPCR that is part of a family of proton sensing GPCRs. The members of this family are unique in that they are activated not by a ligand directly but rather by changes in extracellular pH. GPR68 has been implicated in various physiological processes and has been found in a variety of tissues. Interestingly there is recent evidence being gathered which implicates low and high levels of GPR68 with the progression of neoplastic pathologies. The receptor, however, remains dark pharmacologically and little is known about its activation mechanism. Our research focuses on employing an in silico approach to study the activation mechanism of the receptor. We have generated homology models of the receptor and have run molecular dynamics simulations on different protonation states of the receptor. The evolution of the pKa values of key residues involved in pH sensing during the simulations have been monitored to understand which amino acids are affected by changes in pH and how these changes lead to receptor activation. We have determined a possible sequence of protonation events on the extracellular and transmembrane side of the receptor which are associated with changes in microswitches indicative of activation.

Analysis of structures and properties for design of OLED materials

Anant

Phosphorescent organic light-emitting diodes (PhOLEDs) have attracted attention because of their prospective use in display monitors and lighting applications and their higher energy efficiencies. However, designing blue PhOLEDs with longer operational lifetimes remains to be a challenge. In this work, we use a database of PhOLEDs [1] to analyse the structures and theoretical properties of 191 hosts and 11 guests to gather property-structure and property-property relationships. Donor (D), acceptor (A) and linker (L) moieties of the hosts were identified, with carbazole (Cz), pyridine (Py) and benzene (Bz) as the most commonly occurring D, A and L, respectively. We also analysed the alignment of the energy levels of the hosts and guests to find combinations of dopant-host with reduced charge trapping. We believe these results contribute to a better understanding of how we can design PhOLED devices with the most promising host and dopant combinations.

[1] Chan et al, Organic Electronics, 63, 257-266, (2018)

Exploring Enhanced Sampling Methods to Calculate Kinetics Processes in Ligand-Gated Ion Channels

Guangpeng Xue

Pentameric ligand gated ion channels (pLGIC) play an important role in the nervous system: studying the mechanism of their conformational changes is of great importance in regards to the treatment of neurological diseases. In this work, we aim to explore the performance of methods to calculate kinetics effects, by evaluate the isomerization rates of proline dipeptide with a view to then estimating it for Pro281 in the 5-HT₃ receptor, a pLGIC. The dipeptide models of proline in vacuo and in water environments were investigated by means of enhanced sampling methods. A reliable protocol to setup the metadynamics (MetaD) simulations for obtaining kinetics information was provided in this work, avoiding extra deposition in transition regions and at the same time saving computational resources. The Poisson distributions of this rare event were characterized. The timescales of proline dipeptide isomerization in two directions (Trans-Cis and Cis-Trans) were obtained in our work. The presence of water molecules increased the isomerization energy barrier and the timescales of isomerization in water systems were roughly of ten times larger than in vacuo systems. The influence of thermostats, initial position and water models were also investigated, and the number of simulations in obtaining reliable and converged kinetics values was also studied. Due to the complexity of the receptor system, a new method Gaussian Mixture Based Enhanced Sampling (GAMBES) was tested in the vacuum system. The GAMBES method is promising in the kinetics calculation of the receptor system. The advantages and disadvantages of both methods MetaD and GAMBES were discussed in this project.

Unravelling the impact of disorder on the electronic properties of mixed-metal chalcogenides

Adair Nicolson

Mixed-metal mixed-anion systems have seen a significant rise in interest as ‘perovskite-inspired materials’ as are expected to combine the excellent stability seen in metal chalcogenide solar cells with the well-known performance of hybrid halide perovskite solar cells.[1]

$\text{Sn}_2\text{SbS}_2\text{I}_3$ is a promising solution-processed photovoltaic absorber having achieved efficiency above 4% in initial devices.[2] Theoretical work predicts that the material family of $\text{A}_2\text{BCh}_2\text{X}_3$ mixed-metal chalcogenides could also be ferroelectric, with $\text{Sn}_2\text{SbS}_2\text{I}_3$ having strong lattice polarization and large dielectric constants.[3]

However, this family has not been rigorously explored resulting in confusion in the literature regarding the structure of these materials, with some works observing disorder in room temperature crystals.[4] Without a proper description of the structure, prediction of the electronic properties cannot be accurately performed. Understanding the extent of the disorder in these systems is of key importance due to its tendency to quench favourable properties such as macroscopic polarisation.

Using Density Functional Theory, Cluster expansion and Monte Carlo techniques, we have systematically examined the cation disorder in $\text{Sn}_2\text{SbS}_2\text{I}_3$ for the first time and will discuss its likely impact on the potential for this material family to produce ferroelectric and photovoltaic devices.

[1] Nie, R. et al., Lead-Free Perovskite Solar Cells Enabled by Hetero-Valent Substitutes. *Energy Environ. Sci.* 2020, 13 (8), 2363–2385. <https://doi.org/10.1039/D0EE01153C>.

[2] Nie, R. et al., Heteroleptic Tin-Antimony Sulfoiodide for Stable and Lead-Free Solar Cells. *Matter* 2020, 3 (5), 1701–1713.

[3] Kavanagh, S. R. et al., Hidden Spontaneous Polarisation in the Chalcogenide Photovoltaic $\text{Sn}_2\text{SbS}_2\text{I}_3$. *Materials Horizons* 2021

[4] Doussier, C. et al., Crystal Structure of $\text{Pb}_2\text{SbS}_2\text{I}_3$, and Re-Examination of the Crystal Chemistry within the Group of (Pb/Sn/Sb) Chalcogeno-Iodides. *Solid State Sciences* 2007, 9 (9), 792–803.

INVESTIGATING THE QUASI-LIQUID LAYER ON ICE SURFACES: A COMPARISON OF ORDER PARAMETERS

Jihong Shi

Ice surfaces are characterized by pre-melted quasi-liquid layers (QLLs) which mediate both crystal growth processes and interactions with external agents. Understanding QLLs at the molecular level is necessary to unravel the mechanisms of ice crystal formation. Computational studies of the QLLs heavily rely on the accuracy of the methods employed for identifying the local molecular environment and arrangements, discriminating solid-like and liquid-like water molecules. Here we compare the results obtained using different order parameters to characterize the QLLs on hexagonal ice (Ih) and cubic ice (Ic) model surfaces investigated with molecular dynamics (MD) simulations in a range of temperatures. For the classification task, in addition to the traditional Steinhardt order parameters in different flavours, we select an entropy fingerprint and a deep learning neural networks approach (Deeplce), which are conceptually different methodologies. We find that all the analysis methods give qualitatively similar trends for the behaviours of the QLLs on ice surfaces with temperature, with some subtle differences in the classification sensitivity limited to the solid-liquid interface. The thickness of QLLs on the ice surface increases gradually as the temperature increases. The trends of the QLLs size and of the values of the order parameters as a function of temperature for the different facets may be linked to surface growth rates which, in turn, affect crystal morphologies at lower vapour pressure. The choice of the order parameter can be therefore informed by computational convenience except in cases where a very accurate determination of the liquid-solid interface is important.

An off-lattice hybrid kinetic Monte Carlo (kMC) modelling methodology: Bridging the time and length scales between the atomic level and the continuum

Stavros Ntioudis

The numerical investigation of thin-film formation and evolution is considered a major research challenge. On one hand, models of the continuum (e.g., Finite Element Analysis) lack the necessary resolution to accurately describe the underlying growth mechanisms (i.e., adsorption, desorption, diffusion, reactions). On the other hand, quantum (e.g., Density Functional Theory) and molecular approaches (e.g., Molecular Dynamics) can effectively track the significant atomistic phenomena by simulating the detailed trajectory of atoms. Nonetheless, in the direction of capturing such trajectories, it becomes mandatory to deploy femtosecond or sub-femtosecond time increments and estimate numerically expensive integrals at each timestep. This limits the system size and the timescale of lower-level theories to tenths of thousands of atoms and a few nanoseconds per GPU day, respectively. Thin films involved in modern engineering applications (e.g., lubricant additive tribofilms or Solid Electrolyte Interphases) typically require seconds, minutes, or even hours to grow and their thickness can reach up to several hundreds of nanometres. Consequently, traditional approaches of molecular-level precision are unsuitable when it comes to describing the realistic evolution of thin films. In this poster, we present a novel physics-based multiscale computational methodology capable of bridging the time and length scales between the atomic level and the continuum. The proposed numerical framework uses a bottom-up modelling approach with data from DFT and MD calculations being supplied to a kinetic Monte Carlo (kMC) scheme, which in turn deploys an efficient stochastic algorithm to accelerate the state-to-state system evolution. The kMC method is coupled with a computationally inexpensive equilibration step in a hybrid (kMC/MD) implementation, which not only captures the significant atomic-scale processes but also accounts for the residual elastic strains between atoms. The thermal decomposition of lubricant additive TCP molecules on a pristine Fe (110) substrate is utilized as a proof of concept of the introduced modelling methodology.

Polarisation of water under thermal fields: the effect of the molecular dipole and quadrupole moments

Aidan Chapman

Thermal gradients induce a wide range of powerful non-equilibrium coupling effects. These effects have important applications in biomolecule sensing and nanoscale heat collectors. A more recently discovered coupling effect is thermopolarisation (TP) [1]. The application of thermal gradients gives rise to molecular orientation and electric fields in polar fluids. Both water [1–3] and acetonitrile [4] have both exhibit this effect in simulations. The importance of thermopolarisation has been discussed in applications such as optothermoelectrics, mechanisms for sonoluminescence, in the study of bioelectric effects, and the microwave drying of materials.

In this work [5], we have used non-equilibrium molecular dynamics (NEMD) simulations to investigate thermopolarisation for four popular rigid water models (OPC, TIP3P, TIP4P/2005 and SPC/E). The effect is quantified by the thermopolarisation coefficient, S_{TP} , which is the ratio between the induced electric field and the applied thermal gradient. Below an inversion temperature the coefficient is positive [3,5], but above which the coefficient changes sign and the field changes direction. We have precisely determined the temperatures of inversion for each model by applying thermal gradients that covers the temperatures of inversion. Clear lines of inversion can be seen on the density-temperature phase diagrams of all four water models investigated. We have derived an expression for the thermopolarisation coefficient in terms of the molecular multipole moments, the average molecular orientation, and the thermal expansion coefficient.

References

- [1] F. Bresme, A. Lervik, D. Bedeaux, and S. Kjelstrup, *Physical Review Letters* 101, 020602 (2008).
- [2] I. Iriarte-Carretero, M. A. Gonzalez, J. Armstrong, F. Fernandez-Alonso, and F. Bresme, *Physical Chemistry Chemical Physics* 18, 19894 (2016).
- [3] J. Armstrong and F. Bresme, *Physical Review E* 92, 060103 (2015).
- [4] O. R. Gittus, P. Albella, and F. Bresme, *The Journal of Chemical Physics* 153, 204503 (2020).
- [5] A. Chapman and F. Bresme, Under Submission (2022).

Lone pair driven anisotropy in antimony chalcogenide semiconductors

Xinwei Wang

Antimony sulfide (Sb_2S_3) and selenide (Sb_2Se_3) have emerged as promising earth-abundant alternatives among thin-film photovoltaic compounds. A distinguishing feature of these materials is their anisotropic crystal structures, which are composed of quasi-one-dimensional (1D) $[Sb_4X_6]_n$ ribbons. The interaction between ribbons has been reported to be van der Waals (vdW) in nature and Sb_2X_3 are thus commonly classified in the literature as 1D semiconductors. However, based on first-principles calculations, here we show that inter-ribbon interactions are present in Sb_2X_3 beyond the vdW regime. The origin of the anisotropic structures is related to the stereochemical activity of the Sb 5s lone pair according to electronic structure analysis. The impacts of structural anisotropy on the electronic, dielectric and optical properties relevant to solar cells are further examined, including the presence of higher dimensional Fermi surfaces for charge carrier transport. Our study provides guidelines for optimising the performance of Sb_2X_3 -based photovoltaics via device structuring based on the underlying crystal anisotropy.

Studying the thermoelectric performance of CuInTe₂ using hybrid-DFT and anharmonic lattice dynamics

Sabrina Hachmioune

Fossil fuels provide 80% of the world's energy and of that energy, an estimated 80% is wasted in the form of heat. This has motivated world leaders to invest in alternative forms of energy generation. One such alternative to be explored is the use of thermoelectric materials (TE).

Thermoelectric materials can convert heat into electricity through the Seebeck effect allowing them to increase the overall efficiency of existing processes, as well as serve as a clean source of electricity. Materials in use today are composed of toxic elements like lead or are plagued by low efficiencies. To measure thermoelectric efficiency, we use the figure of merit, ZT. For comparison, the ZT of a well-known TE that has been used in many NASA space missions, PbTe, has an average ZT of 1.4.

In this work, we explore a chalcopyrite semiconductor, CuInTe₂ (CIT) with an experimental ZT of 1.18 at 850 K which has been shown to increase up to 1.72 as a p-type material (10^{18} carriers/cm³). We combine density functional theory (DFT) with lattice dynamics to determine the transport and phonon properties of CIT. The hybrid-DFT functional (HSE06) with spin-orbit coupling (SOC) is used to accurately predict the band structure and the AMSET code is used to calculate its transport properties beyond the constant relaxation time approximation. We use the supercell approach employed by Phono3py to calculate third order phonon-phonon interactions and determine the lattice thermal conductivity.

These results allow us to calculate the ZT of CIT from first principles where previous studies have relied on an experimental or semi-empirical approach. A study of the intrinsic and extrinsic defects is also underway to provide a crucial understanding of the material for effective doping and to unlock the thermoelectric potential of CuInTe₂.

The Wadsley-Roth materials family as functional energy materials

Adrien Richard

The Wadsley-Roth (W-R) family of materials is made up of transition metal oxides mainly composed of Tungsten, Titanium and Niobium and are thought to be highly disordered. Members of the W-R family are defined by their structure based on ReO₃ – type blocks, which stack to form blocks of corner-sharing octahedra of size (m x n) delimited by crystallographic shear planes (CS) and are connected to each other via edge-sharing. Block units connect infinitely in directions perpendicular to the (m x n) plane. Tetrahedral sites can also be present in the same plane as the blocks to fill voids. An index was created to be able to easily classify these materials depending on their type of block structure.

Over recent years, these materials have gained much interest from the scientific community. However, publications almost systematically study these materials individually (i.e. interest in a particular composition) rather than as a family of materials. This has led to the properties of the W-R family remaining unknown, for the most part, especially in terms of how the block structure can influence electronic and insertion properties, for example. Typically, one would dope such materials in an aliovalent fashion to bolster their electronic properties such as electronic conduction and charge carrier migration, for example. Nevertheless, with the difference in Oxygen to Metal (O/M) ratio between structures of the W-R family being quite small, it proves to be an arduous task to identify which compound is synthesised. Furthermore, it is not clear whether doping through traditional methods for these materials (annealing to include oxygen vacancies etc.) would be effective here as a phase transition from a doped compound to a defect-free block structure is possible thanks to the proximity in O/M ratio values between different W-R members.

With no current or past studies on the relationship between structure and electronic properties in W-R compounds, it is impossible to predict which methods will be more effective than others to better these promising materials and thus leaves researchers to the time-consuming method of trial and error. For this reason, I have endeavoured throughout my PhD research to be able to present the first comprehensive computational prediction study on the W-R materials as energy materials.

A computational re-evaluation of selenium as a solar absorber

Alp Eren Samli

Crystalline hexagonal selenium (c-Se) was the first material to exhibit the photovoltaic effect. It was used to make the first solar cell in 1883 but the low efficiency of the cell (<1%) made it impractical. Starting with silicon in 1954, many other technologies reached much higher efficiencies and effectively made c-Se obsolete. However, c-Se shows potential due to its direct band gap, high earth abundance, high thermodynamic efficiency limit, and low temperature processing. In fact, selenium solar cells have improved considerably in the last 30 years with a current record of 6.5%. In this study, the Vienna Ab initio Simulation Package (VASP) was used to perform hybrid density functional theory (DFT) calculations on crystalline selenium's 5 known polymorphs (hexagonal, rhombohedral, β -monoclinic, γ -monoclinic, δ -monoclinic) to determine the ideal phase for solar cell operation. The DFT-D3 dispersion correction was found to greatly improve agreement with experimental values. Band structure calculations revealed that all polymorphs except γ -monoclinic have an indirect fundamental band gap with a slightly higher direct band gap. Hexagonal Se was found to have the lowest direct band gap (1.904 eV with spin-orbit coupling) and highest charge carrier mobilities, allowing it to reach the highest efficiencies. Analysis of the material's optical response showed that it can reach a maximum power conversion efficiency of 23% with a thickness of 2 μm , making the first photovoltaic material a promising cheap alternative to silicon, perovskites and other high efficiency solar absorbers. Its high band gap even allows it to be used in tandem with low band gap absorbers like silicon.

DFT study of phonon properties in barocaloric ammonium sulfate

Shurong Yuan

Ammonium sulfate is a giant inverse barocaloric material that is cheaply and commercially available. Exploiting its potential for cooling applications requires an understanding of the mechanism driving the entropy change. Here we report an investigation by density functional theory of the phonons under working conditions of temperature and pressure. We find excellent agreement between the experimental and calculated results. The ammonium librational modes that are crucial to the entropy change are identifiable by their negative Gruneisen parameter. Our results connect the differences in structure across the phase transition to those in the atomic dynamics, suggesting a route towards designing new caloric materials.

The cardioprotective effect of cocoa-flavanols: a computational study

Cambrin Kemble-Diaz

Cocoa-flavanols are a subset of a group of phytochemicals called polyphenols. Polyphenols are derived from foods commonly eaten in southern Europe including fruits, vegetables, nuts, olive oil and red wine. As such they have been proposed as a possible explanation for the positive effects of the Mediterranean diet. Studies have suggested their protective effect against ageing-associated diseases. Cocoa-flavanols themselves come from cocoa and have been associated with resistance to cardiovascular events, in particular with the recent landmark COSMOS trial. The molecule epicatechin and its derivatives have been identified via in-vitro studies as being the possible source of the effect. Although there is good evidence for this in in-vitro/in-vivo, and now even in clinical studies, there is currently a dearth of investigations into the method of action of these molecules. Via a combination of computational simulations and further in-vitro studies, we hope to elucidate it.

Ab-initio tight-binding models for twisted bilayer TMDs

Kemal Atalar

The discovery of superconducting and correlated insulating states in magic-angle twisted bilayer graphene has led to intense interest in other moiré materials. Twisted bilayer transition metal dichalcogenides (TMDs), for example, have been shown to exhibit correlated insulating states at different fillings, possible superconducting behaviour [1], and exotic optical properties [2].

The large size of the moiré unit cells of small-angle twisted TMD bilayers renders electronic structure calculations based on density functional theory (DFT) unsuitable for the routine study and exploration of their large chemical and structural phase space. Therefore, atomistic tight-binding models are often used. Whilst tight-binding models with position-dependent interlayer hoppings have been developed, a model that captures position dependence within a layer doesn't exist. This is essential to describe the effect of intralayer atomic relaxations on the emergence and nature of flat bands in the electronic structure [3], as well as to calculate electron-phonon coupling.

In this work, we develop a position-dependent intralayer TMD tight-binding Hamiltonian. Our model is based on Slater-Koster relations that are parametrized to hoppings obtained from first-principles DFT Hamiltonians of strained monolayer TMDs in a basis of maximally localized Wannier functions.

[1] Wang L. et al., Nat. Mat. 19, pages 861–866 (2020)

[2] Tran, K., et al., Nature 567, 71–75 (2019)

[3] Li, H., et al., Nat. Mater. 20, 945–950 (2021)

Shape-dependence of optical properties of coinage-metal nanoparticles

Wei Zhao

Metallic nanoparticles showing enhanced optical and plasmonic properties are at the heart of the development of new technologies for harvesting and converting sunlight into chemical and electrical energy. The size and shape-dependence of optical properties of coinage nanoparticles is a feature of high importance and drawing considerable scientific interest. In this talk, we will discuss our work on the electronic and photo-absorption properties of the “breathing” effect in the bare copper, silver and gold with a highly-symmetric fullerene shape [1], from which we can report that a red-shift of the optical spectrum is expected as the fullerenes inflate and these shift in the optical spectra are not directly linked to the changes in the HOMO-LUMO gap. We will also report our work on the absorption spectrum of bare Cu, Ag, Au nanoparticles with the same size and different geometrical shapes [2], from which we have shown that highly symmetric caged-like morphology display enhanced peaks in the absorption spectrum, mainly in the visible region due to symmetrical electronic configuration of cage-like structures. These effects can not be fully explained by the corresponding change in HOMO-LUMO gaps which will be enhanced by cage structures. Finally, we will introduce a new fast screening method using information from ground-state DFT calculation only as a robust way to predict morphology which is active in the visible range.

[1] W Zhao, R M Jones, R D'Agosta, and F Baletto, J. Phys.: Condens. Matter., 2022, 34 224005

[2] F Baletto and R Ferrando, Phys. Chem. Chem. Phys., 2015,17, 28256-28261

Quantum Embedding

Basil Ibrahim

Embedding methods are an approach to the electronic structure problem which make use of locality to reduce the complexity of the problem. To obtain a correlated solution to a problem, a number of independent calculations are performed on a fragments of a system obtained from a mean-field reference. These results must then be combined into a description of the whole system from which the expectation values of arbitrary observables can be obtained.

This poster will focus on the application of the highly accurate but expensive coupled cluster method to both solid and molecular systems, overcoming the huge computational expense via the use of an modified density matrix embedding theory with an improvable description of the environment of each fragment. Thus, in principle, convergence to the full coupled cluster solution may achieved. This method will be introduced, with a focus on obtaining expectation values, and ground state energies and equations of state for standard test systems will be presented with comparison to other methods.

In silico design of cardiac myotropes for the treatment of cardiovascular diseases

Fariha Akter

Heart disease is one of the leading causes of mortality worldwide, despite medical advancements. Recent research has indicated that new promising treatments can be developed by designing drugs that can directly bind the key cardiac motor protein myosin II and modulate its contractile activity in diseases such as hypertrophic and dilated cardiomyopathy [1]. Myosin can adopt different conformations during its functional cycle. Compounds that can selectively stabilise these conformations are expected to have different effects on myocardial contractility [2]. This research aims to rationally design new conformation-selective compounds of human β -cardiac myosin.

Various conformation-selective ligands were identified using virtual screening, molecular docking, and molecular dynamics (MD) simulations. Hydrogen bond and contact map analyses showed that selectivity towards a specific conformation was related to a higher number of protein-ligand interactions. MD simulations confirmed a stabler binding to the expected state for each ligand. Moreover, new possible target conformations were identified with enhanced- sampling MD simulations (steered MD and umbrella sampling) of the recovery stroke transition. Pocket tracking analyses revealed that the binding site is very dynamic with significant changes in physicochemical properties such as size and polarity. These findings will be used to guide the design of drugs that can selectively stabilise intermediate states in the myosin recovery stroke. The best candidates will be tested in vitro on single cardiac myosin molecules to measure their effect on myosin force, velocity, and power and determine the type of activity (activation or inhibition) of each ligand [3].

1 S. J. Lehman, C. Crocini and L. A. Leinwand, *Nat Rev Cardiol*, 2022, <https://doi.org/10.1038/s41569-022-00682-0>.

2 S. Hashem, W. G. Davies and A. Fornili, *J. Chem. Inf. Model.*, 2020, 60, 6438–6446.

3 J. Ochala, M. Li, M. Ohlsson, A. Oldfors and L. Larsson, *The Journal of Physiology*, 2008, 586, 2993–3004.