

The background of the poster is a collage of scientific imagery. The top half features a dense field of small, multi-colored spheres (purple, blue, yellow, orange) that resemble a molecular simulation or a data visualization. The bottom left shows a cluster of red, white, and black spheres, likely representing water molecules. The bottom right features a 3D ball-and-stick model of a purple molecular structure. A large white diamond shape is centered on the page, containing the event's title and date.

Lennard Jones Centre

Showcase Day

9th October 2023
The Maxwell Centre



Lennard Jones Centre Showcase Day

The Lennard Jones Centre (LJC) brings together researchers from different departments across the University of Cambridge with a common interest in materials and molecular modelling. Our core aim is to foster an inclusive environment from which new ideas and collaborations can emerge. We take ‘materials and molecular modelling’ to be loosely defined, but some of our interests include quantum mechanical systems, exploration of high-dimensional landscapes, and the interfaces between materials.

This *Showcase Day*, –now in its second iteration – gives early career researchers (particularly postgraduate students) within the remit of the LJC an opportunity to present their work to their peers as well as leading academics within Cambridge and beyond. Building on the success of last year, we have expanded the program to include a talk from industry, as well as a panel discussion with some experts in the field who we are sure will give great insight into their careers and the progress of the field.

Finally, we are very grateful to our sponsors IBM, Quantinuum and UKCP for supporting the event.

Showcase Day Organising Committee

BENJAMIN XU SHI

Yusuf Hamied Department of Chemistry

DR. CHRISTOPH SCHRAN

Cavendish Laboratory, Department of Physics

FLAVIANO DELLA PIA

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LILA CADI TAZI

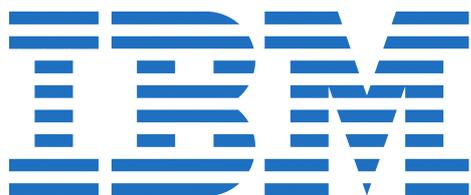
Yusuf Hamied Department of Chemistry

NIAMH O’NEILL

Yusuf Hamied Department of Chemistry

XAVIER R. ADVINCULA

Yusuf Hamied Department of Chemistry



Schedule

09:45 Arrival and Welcome (*coffee and tea provided*)

Session 1

Chair: Rokas Elijošius

Session 1

10:00 **Ilyes Batatia**, *Department of Engineering*

MACE: Higher Order Equivariant Message Passing Neural Networks for Fast and Accurate Force Fields

10:15 **Lars L. Schaaf**, *Department of Engineering*

Accurate Energy Barriers for Catalytic Reaction Pathways

10:30 **Elena Gelžinytė**, *Department of Engineering*

Machine Learning Interatomic Potentials to Predict Bond Dissociation Energies

10:45 Break (*coffee, tea, juice, and pastries provided*)

Panel Discussion

Chair: Dr. Steve Cox

11:00 **Prof. Sally Price, Prof. Emilio Artacho, Prof. Chris Pickard**

Attendees are encouraged to ask questions about the evolution of the field of materials modelling and panel member's careers

12:00 **Lunch**

12:30 **Poster Session**

Company Talk

Chair: Dr. Christoph Schran

13:30 **Dr. Fabian Thiemann, IBM**

Session 2

Chair: Dr Sun-Woo Kim

14:00 **Esmee Woods**, *Department of Chemistry; Department of Physics*

Analysis of First Passage Time Distributions on Large, Ill-Conditioned, Multifunnel Energy Landscapes

14:15 **Maryna Bakumenko**, *Department of Applied Mathematics, Aston University*

Multiscale Hybrid Model of Liquids: Modified Equations of Motion

14:30 **Dingeman (Danny) van der Haven**, *Department of Materials Science and Metallurgy*

The Importance of Particle Shape in Granular Matter; An Efficient Method for Simulating Arbitrarily Shaped Particles

14:45 Break (*coffee, tea, juice, and pastries provided*)

Session 3

Chair: Dr Lewis Conway

15:15 **Benjamin Xu Shi**, *Department of Chemistry*

It's Not a S(KZ)CAM! High Accuracy Surface Chemistry at Low Cost

15:30 **Paula Teeuwen**, *Department of Chemistry*

Accelerated Discovery of Metal-Organic Cages Self-Assembled from Trapezate Building Blocks

15:45 **Honghao Yu**, *Department of Chemistry*

Is Laning a Phase Transition?

16:00 Conclusion and Awards (*drinks and snacks provided*)

19:00 Formal Dinner at Churchill College

Panel Member Bios

Prof. Sally Price

Department of Chemistry, University College London

BIOGRAPHY

Sally, officially Sarah, Price gained a PhD in theoretical chemistry at the University of Cambridge under the supervision of Anthony Stone, specialising in deriving models of the forces between molecules from their wavefunctions. She worked at the Universities of Chicago and Cambridge, before becoming a lecturer at UCL (University College London), where she is now a Professor specialising in Computational Chemistry. In developing the theory and computer codes to model the organic solid state, she has collaborated widely with experimental solid-state chemists, pharmaceutical scientists, theoretical physicists and computational scientists, including leading the Basic Technology Project “Control and Prediction of the Organic Solid State” (www.cposs.org.uk). She was awarded the RSC Interdisciplinary Prize in 2015 and elected to the Fellowship of the Royal Society in 2017 in recognition of the value of this collaborative work that has revealed and continues to reveal the complexities of organic crystallisation.

Prof. Emilio Artacho

*Theory of Condensed Matter, Cavendish Laboratory, University of Cambridge
CIC nanoGUNE BRTA and DIPC, Tolosa Hiribidea 76, 20018 San Sebastián, Spain
Ikerbasque, Basque Foundation for Science, 48011 Bilbao, Spain*

BIOGRAPHY

Emilio Artacho arrived at Cambridge in 2001, starting in the Department of Earth Sciences, where he was granted a Professorship in 2006. In 2011 he transferred to the Cavendish. Before Cambridge, he was an associate professor in condensed matter physics in Madrid since 1995. He was an Invited Professor at the École Normale Supérieure de Lyon (1999) and a Visiting Miller Professor at the University of California at Berkeley (2007). He is also an Ikerbasque Research Professor at nanoGUNE (San Sebastián, Spain). Artacho’s research is within the field of computational and theoretical condensed matter physics, especially (but not exclusively) developing and using first-principles simulation methods. Recent work includes the study of strongly out-of-equilibrium quantum processes in forced electronic systems, mostly as affected by nuclei shooting through matter, as happens in radiation-damage processes. For this purpose, real-time time-dependent DFT is used. Water nano-films are also studied within a longer line of work in the study of liquid water, based on first-principles molecular-dynamics simulations. Other topics of interest respond to collaborations with other groups, such as the study of complex and functional oxides and their heterostructures, two-dimensional materials, etc. The SIESTA linear-scaling DFT method is the main software tool used in most computational work, which is developed and maintained in the group within a wider collaboration.

Prof. Chris Pickard

Department of Materials Science and Metallurgy, University of Cambridge

BIOGRAPHY

Chris Pickard's research helps modern scientists "see" and discover the universe at the atomic scale through Quantum Mechanics – from the centres of giant exoplanets to pharmaceutical compounds, new battery materials and high-temperature superconductors. He is the inaugural Sir Alan Cottrell Professor of Materials Science in the Department of Materials Science and Metallurgy, University of Cambridge. Previously he was a Professor of Physics, University College London, and Reader in Physics, University of St Andrews. He has held EPSRC Advanced and Leadership Research Fellowships, and a Royal Society Wolfson Research Merit Award. He is a lead developer of the widely used CASTEP computer code and introduced both the GIPAW approach to the prediction of magnetic resonance parameters and *Ab Initio* Random Structure Searching (AIRSS). CASTEP has provided a significant source of licencing income for Cambridge Enterprise for over 20 years, while his AIRSS software is freely available through an open-source license. In 2015 he won the Rayleigh Medal and Prize of the Institute of Physics, awarded for distinguished research in theoretical, mathematical or computational physics.

Abstracts of Student Talks

Ilyes Batatia

Department of Engineering

MACE: HIGHER ORDER EQUIVARIANT MESSAGE PASSING NEURAL NETWORKS FOR FAST AND ACCURATE FORCE FIELDS

Creating fast and accurate force fields is a long-standing challenge in computational chemistry and materials science. Recently, several equivariant message passing neural networks (MPNNs) have been shown to outperform models built using other approaches in terms of accuracy. However, most MPNNs suffer from high computational cost and poor scalability. We propose that these limitations arise because MPNNs only pass two-body messages leading to a direct relationship between the number of layers and the expressivity of the network. In this talk, we will introduce MACE, an equivariant MPNN model that uses higher body order messages. We show that using four-body messages reduces the required number of message passing iterations to just two, resulting in a fast and highly parallelizable model. We show that MACE generally outperforms alternatives for a wide range of systems from amorphous carbon, universal materials modelling, and general small molecule organic chemistry to large molecules and liquid water. We demonstrate the capabilities of the model on tasks ranging from constrained geometry optimisation to molecular dynamics simulations and find excellent performance across all tested domains.

Lars L. Schaaf

Department of Engineering

ACCURATE ENERGY BARRIERS FOR CATALYTIC REACTION PATHWAYS

This talk highlights the indispensable role of machine learning force fields (MLFFs) in computational catalysis. We present a robust protocol that automates training and enables efficient exploration of reaction pathways. Notably, the protocol accurately replicates DFT reaction barriers and identifies improved reaction paths. Validated on carbon dioxide hydrogenation to methanol over indium oxide, the protocol finds a 40% reduction in the rate-limiting step compared to existing literature. Additionally, we emphasize the significance of accounting for finite temperature effects and compute reaction free energy barriers. The ability of MLFFs to enhance our understanding of such extensively studied catalysts underscores the need for fast and accurate alternatives to direct *ab initio* simulations. Automatic training procedures play a central role in making MLFFs more accessible for routine academic and industrial use.

Elena Gelžinytė

Department of Engineering

MACHINE LEARNING INTERATOMIC POTENTIALS TO PREDICT BOND DISSOCIATION ENERGIES

Empirical force fields are valuable tools in computational chemistry, however, they suffer from limitations in terms of accuracy, transferability and their lack of applicability to open-shell structures. Recently, Machine Learning Interatomic Potentials (MLIPs) have emerged as versatile surrogate models capable of accurately reproducing *ab initio* potential energy surfaces. However, most of their applications have been targeted at near-equilibrium closed-shell structures. The presented work aims to address this limitation by developing highly accurate and transferable MACE MLIP that can be applied to both closed- and open-shell molecules. An accurate description of radical species extends the scope of possible applications to Bond Dissociation Energy (BDE) prediction, for example, with relevance to cytochrome P450 metabolism modelling. We show that the MACE interatomic potential that is applicable for open- and closed-shell drug-like molecules containing C, H, O chemical elements. We explore MACE transferability to the COMP6 dataset and show that it reaches accuracy on par with state-of-the-art transferable ANI2x potential for closed shell molecules. MACE potential reaches similar accuracy on two CYP substrate datasets, with open- and closed-shell structures relevant to CYP metabolism. We apply MACE to aliphatic C-H bond dissociation energy prediction where it reaches RMSE below 1.6 kcal/mol and a better rank prediction than currently used AM1 semi-empirical method. The MACE model extrapolates well over bond dissociation potential energy surface scans, which shows promise for extension to predict not only reaction energies but also reaction activation energies.

Esmee Woods

Department of Physics, Department of Chemistry

ANALYSIS OF FIRST PASSAGE TIME DISTRIBUTIONS ON LARGE, ILL-CONDITIONED, MULTIFUNNEL ENERGY LANDSCAPES

Local minima on an energy landscape and the transitions connecting them can be represented by a kinetic transition network. Multifunnel energy landscapes exhibit a range of transition timescales between states, which correspond to competing pathways. We focus on computing and understanding the first passage time distributions between energy minima. Analysis of large networks is challenging due to the limit of computation time and space. Furthermore, classical methods which analyse networks that exhibit a large separation of time scales, termed ill-conditioned networks, break down due to finite precision.

We introduce partial graph transformation, which iteratively eliminates and renormalises well-chosen states to produce a lower rank model, whilst approximately conserving the full first passage time distribution. We then combine methods which accurately compute the mean first passage time, with those that compute the short time first passage time distribution, to produce the full first passage time distribution, in the ill-conditioned regime. This enables us to investigate and understand the kinetics of larger networks and at lower temperatures, than previously possible.

Maryna Bakumenko

Department of Applied Mathematics, Aston University

MULTISCALE HYBRID MODEL OF LIQUIDS: MODIFIED EQUATIONS OF MOTION

Classical Molecular Dynamics (MD) methods provide highly accurate simulations by solving Newtonian equations of motion and calculating inter-particle forces. These methods offer detailed atomistic-scale information, enabling the representation of macroscopic thermodynamic properties as well as microscopic characteristics such as radial distribution functions and autocorrelation functions. While modern computers can simulate liquid molecular systems of considerable size (tens of nanometers across) using MD, modelling biologically relevant processes within feasible timescales (microseconds to milliseconds) remains a challenge. MD-continuum coupling is an approach that allows for larger-scale modelling by replacing part of the MD region with a continuum representation (HD). However, coupling MD and HD models presents significant difficulties. The proposed Multiscale hybrid model of liquids is based on Smith's approach [1], which applies the Gaussian principle of least action to derive equations of motion for particles in extreme cases, ensuring momentum correspondence between MD and HD. The liquid system is described as a two-phased mixture, with a Lagrangian representation (MD) and an Eulerian representation (HD) of the same substance. The concentrations of the MD and HD phases, determined by parameter s ($0 \leq s \leq 1$), control the atomistic information in specific regions. The Control Volume formulation establishes a strict link between discrete and continuous representations. To couple MD and HD, the momentum in the hybrid region has been adjusted to match MD by employing a constraint force.

[1] Smith, E. R., Zaki, T. A., et al. (2015). A localized momentum constraint for non-equilibrium molecular dynamics simulations. *The Journal of Chemical Physics*, 142(7), 074110

Dingeman (Danny) van der Haven

Department of Materials Science and Metallurgy

THE IMPORTANCE OF PARTICLE SHAPE IN GRANULAR MATTER; AN EFFICIENT METHOD FOR SIMULATING ARBITRARILY SHAPED PARTICLES

Granular materials, such as coal, corn, powders, and sand, are ubiquitous yet poorly understood. The predictiveness of analytical theories for granular matter lags behind compared to theories for gases, liquids, and pure solids. This is largely because of the complex dependence of the bulk behaviour on the properties, such as size, shape, and stiffness, of the constituent particles. Numerical methods such as the discrete element method (DEM) have shown more success and are a trending topic. However, although DEM has been able to provide new insights, particle shape remains a difficult property to take into account.

We present the volume-interacting level-set discrete element method (VLS-DEM), a novel method for simulating granular materials with particles of any shape. Individual particles are represented by a discrete level set (also called a signed-distance function). The use of a discrete level set allows the representation of any particle geometry, be it convex, concave, simple, or complex. Furthermore, by integrating the overlap volume between two particles instead of using only the penetration distance, VLS-DEM is physically constituent even for concave geometries. For non-trivial particle shapes, VLS-DEM outperforms other methods used to present more complex particle shapes. Finally, VLS-DEM was used to perform a case study on the Platonic solids and their hyperbolised variants. A continuous hyperbolisation function was used to create new concave shapes. Each of these shapes have a different sphericity, angularity, and concavity that affect the properties of the bulk materials. Comparing the angle of repose and porosity of these materials shows that the angularity and concavity both strongly affect the angle of repose and porosity to an extent that even convex polyhedra with rolling friction cannot capture. These observations show that the full complexity of particle geometry need to be considered, underling the need of new methods such as VLS-DEM.

Benjamin Xu Shi

Department of Chemistry

IT'S NOT A S(KZ)CAM! HIGH ACCURACY SURFACE CHEMISTRY AT LOW COST

Achieving the level of chemical accuracy needed for reliable predictions of surface properties is a major challenge for first-principles calculations. Notoriously, it has not been possible to reach a consensus on the adsorption energy of CO bound to MgO despite intense scrutiny. In this talk, I will present the SKZCAM (pronounced “scam”) protocol, which offers a low cost and accurate framework for applying ‘gold-standard’ techniques such as CCSD(T) to surfaces. Our approach has allowed for the most precise determination of the CO on MgO adsorption energy to date, bringing theory and experiments into consensus for the first time. Comparisons to other reference methods demonstrate that it is between one and two orders cheaper, and comparable in cost to DFT. This affordability enables us to address more challenging surface problems, involving molecules that could not have been tackled before.

Honghao Yu

Department of Chemistry

IS LANING A PHASE TRANSITION?

Non-equilibrium systems exhibit a wide variety of pattern formations. One example is the laning in binary driven systems. We consider mixtures of oppositely driven particles, showing that their non-equilibrium steady states form lanes parallel to the drive, which coexist with transient jammed clusters where particles are temporarily immobilised. We analyse the interplay between these two types of non-equilibrium pattern formation, including their implications for macroscopic demixing perpendicular to the drive. Finite-size scaling analysis indicates that there is no critical driving force associated with demixing, which appears as a crossover in finite systems. We attribute this effect to the disruption of the long ranged order by transient jammed clusters.

Paula Teeuwen

Department of Chemistry

ACCELERATED DISCOVERY OF METAL-ORGANIC CAGES SELF-ASSEMBLED FROM TRAPEZATE BUILDING BLOCKS

In this work, an interdisciplinary approach aimed towards accelerating the discovery new of metal organic cages (MOCs) is proposed. These materials have potential applications in catalysis, separations, sensing, drug delivery and biotechnology. Rational design principles, high-throughput computational methods and experimental methods will be combined to select new building blocks with high potential of self-assembling into new MOCs. A focus is placed on MOCs with anisotropic cavities capable of binding lower symmetry guests and MOCs with large enclosed cavities capable of interacting with biomolecules (artificial virus capsids). This work proposes a new series of MOCs with progressively larger cavity sizes, called the 'Propello MOCs'. It is hypothesised that these MOCs could be self-assembled from trapezate building blocks, where different topologies in this series could be accessed through tuning their structural variables. The in-silico construction and optimization of potential MOC-geometries from these building blocks, including cubic and Propello MOCs, will be automated using the Supramolecular Toolkit (stk), as developed by the Jelfs group [1]. Automated computational methods, with increasing levels of theory, will be employed to virtually screen the most favourable MOC-geometry for each building block. The screening method will include an evaluation of different diastereomers possible for each topology, where a total of 22104 unique diastereomers were identified for (pseudo-)cubic MOCs from trapezate building blocks. This work is expected to provide more insight into the fundamental design principles of MOCs and lead to the discovery of new materials with new functionality.

[1] Turcani, L., Jelfs, K. E., et al. (2021). stk: An extendable Python framework for automated molecular and supramolecular structure assembly and discovery. *The Journal of Chemical Physics*, 154(21), 214102

Abstracts of Posters

Lila Cadi Tazi

Yusuf Hamied Department of Chemistry

QUANTUM ERROR MITIGATION - APPLICATION TO QUANTUM CHEMISTRY

The promising processing power of quantum computers remains largely elusive, primarily due to the presence of noise in both the processing units (qubits) and hardware control systems. Many algorithmic methods have been developed to alleviate the impact of noise in quantum hardware; these are known as quantum error correction (QEC) or quantum error mitigation (QEM). While QEC bears similarities to classical error correction, demanding additional quantum resources (more qubits, more gates), QEM represents a near-term strategy aimed at mitigating noise using limited resources, often involving additional measurements of quantum circuits.

In this work, we evaluate the potential of specific error mitigation techniques, such as Zero Noise Extrapolation (ZNE), for reducing noise in realistically simulated quantum hardware within the Variational Quantum Eigensolver (VQE) algorithm applied to quantum chemical problems. Additionally, we introduce a novel noise characterization approach called FCI projection, providing a more chemical interpretation of quantum noise effects in molecular simulations on small systems.

Niamh O'Neill

Yusuf Hamied Department of Chemistry

TO PAIR OR NOT TO PAIR

The extent of ion pairing in solution is an important phenomenon to rationalise transport properties of electrolytes. A fundamental measure of this pairing is the potential of mean force (PMF) between the solvated ions. The relative stabilities of the paired and solvent separated states in the PMF are highly sensitive to the underlying potential energy surface. However direct application of accurate electronic structure methods to resolve this property is challenging, since long simulations are required to obtain statistical convergence and reach the dilute limit. Leveraging developments in machine learning potentials (MLPs) and electronic structure methods, we obtain wavefunction based models with the random phase approximation (RPA) and second-order Møller-Plessett perturbation theory (MP2) for the prototypical system of Na and Cl ions in water. The RPA model is in excellent agreement with experiment for structural properties, which we then use to compute the PMF, revealing a more stable contact ion pair over the solvent separated ion pair state, while also highlighting shortcomings of density functional theory and classical force-field predictions. These models are primed for application to computationally intensive electrolyte properties including transport coefficients and even confined systems, all of which are highly sensitive to their chosen reference electronic structure method.

Xavier R. Advincula

Yusuf Hamied Department of Chemistry

UNDERSTANDING PROTON TRANSFER UNDER CONFINEMENT

Nanoconfined water is ubiquitous and of broad and growing technological and environmental importance. The fundamental question –perhaps the most fundamental– to ask of nanoconfined water is “Is it H₂O?”. Or, to be more precise, how does the tendency for nanoconfined water to dissociate differ from that of bulk liquid water and how does the solvation of protons and hydroxide differ in the bulk and under confinement? Using a combination of *ab initio* and machine learning techniques, we unravel the properties of the excess proton and the proton hole under confinement to obtain a more fundamental understanding of proton transfer (PT) near interfaces. In particular, we observe a propensity of the excess proton towards the interface and, in a contrasting manner, an aversion of the proton hole towards the interface. To rationalise this, we provide insights into the structural diffusion of such protonated defective species, the microscopic mechanisms involved in PT and the implications of these observations on the dynamics of the system.

Dr. Fabian Berger

Yusuf Hamied Department of Chemistry

BRINGING MOLECULES TOGETHER: SYNERGISTIC CO-ADSORPTION AT DOPANT SITES OF SINGLE ATOM ALLOYS

Development of improved catalysts for industrially relevant processes is an ongoing task for chemists that has lost none of its importance in the last century. To design new generations of catalysts, a reliable and accurate understanding of the fundamental reaction mechanisms and important steps such as barriers of rate-determining steps is required. A major challenge for catalysis is the high stability and thus inertness of common feedstocks. Activation of rather unreactive molecules requires highly active catalysts which typically comes at the cost of reduced selectivity. Still in its infancy, the novel material class of single atom alloy (SAA) catalysts offers a promising solution to avoid this activity-selectivity tradeoff. A crucial feature of effective catalysts, apart from lowering reaction barriers, is the ability to bring reactants together at active sites. We provide evidence that co-adsorption at dopant sites on SAAs is favored over separate adsorption at dopant sites and on the inert host metal surface. By investigating three adsorbates (CO, NO, H), 13 transition metal dopants (3d: Ni, Cu; 4d: Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ag; 5d: Ir, Pt), and two host metal surfaces (Cu(111), Ag(111)), a general preference for co-adsorption at dopant sites is unveiled. This phenomenon is in contrast to pure metal surfaces, as repulsive lateral interactions between adsorbates can be overcompensated by strong interactions with a dopant atom. The effect does not utilise steric constraints like other common catalysts such as zeolites (i.e. pore space) or enzymes (i.e. active pockets).

Kripa Panchagnula

Yusuf Hamied Department of Chemistry

EXPLORING THE PARAMETER SPACE OF AN ENDOHEDRAL ATOM IN A CYLINDRICAL CAVITY

Endohedral fullerenes, or endofullerenes, are chemical systems of fullerene cages encapsulating single atoms or small molecules. These species provide an interesting challenge of Potential Energy Surface (PES) determination as examples of non-covalently bonded, bound systems. While the majority of studies focus on C_{60} as the encapsulating cage, introducing some anisotropy by using a different fullerene, e.g., C_{70} can unveil a double well potential along the unique axis. By approximating the potential as a pairwise Lennard-Jones (LJ) summation over the fixed C cage atoms, the parameter space of the Hamiltonian includes three tunable variables: (M, ϵ, σ) representing the mass of the trapped species, the LJ energy, and length scales respectively. Fixing the mass and allowing the others to vary can imitate the potentials of endohedral species trapped in more elongated fullerenes. We choose to explore the LJ parameter space of an endohedral atom in C_{70} with $\epsilon \in [20\text{cm}^{-1}, 150\text{cm}^{-1}]$, and $\sigma \in [2.85\text{\AA}, 3.05\text{\AA}]$. As the barrier height and positions of these wells vary between $[1\text{cm}^{-1}, 264\text{cm}^{-1}]$ and $[0.35\text{\AA}, 0.85\text{\AA}]$ respectively, using a 3D direct product basis of 1D harmonic oscillator (HO) wavefunctions centred at the origin where there is a local maximum is unphysical. Instead we propose the use of a non-orthogonal basis set, using 1D HO wavefunctions centred in each minimum and compare this to other choices. The ground state energy of the $X@C_{70}$ is tracked across the LJ parameter space, along with its corresponding nuclear translational wavefunctions. A classification of the wavefunction characteristics, namely the prolateness and “peanut-likeness” based on its statistical moments is also proposed.

Chiara Leadbeater

Yusuf Hamied Department of Chemistry

NON-UNITARY TROTTER CIRCUITS FOR IMAGINARY TIME EVOLUTION

We propose an imaginary time equivalent of the well-established Pauli gadget primitive for Trotter-decomposed real time evolution on quantum circuits [1]. Imaginary time evolution (ITE) is widely used for obtaining the ground state of a system on classical hardware. Near-term implementations on quantum hardware rely on heuristics, compromising their accuracy [2,3]. As a result, there is growing interest in the development of more natively quantum algorithms. Since it is not possible to implement a non-unitary gate deterministically, we resort to the implementation of probabilistic imaginary time evolution (PITE) algorithms [4-6], which rely on a unitary quantum circuit to simulate a block encoding of the ITE operator – that is, they rely on successful ancillary measurements to evolve the system non-unitarily. This scheme was tested on the transverse Ising model and the fermionic Hubbard model and is demonstrated to converge to the ground state of the system.

[1] Chiara Leadbeater, A. J. W. T., et al. (2023). Non-unitary Trotter circuits for imaginary time evolution

[2] Jones, T., Benjamin, S. C., et al. (2019). Variational quantum algorithms for discovering hamiltonian spectra. *Phys. Rev. A*, 99, 062304

[3] Motta, M., Chan, G. K.-L., et al. (2019). Determining eigenstates and thermal states on a quantum computer using quantum imaginary time evolution. *Nature Physics*, 16(2), 205–210

[4] Liu, T., Liu, J.-G., & Fan, H. (2021). Probabilistic nonunitary gate in imaginary time evolution. *Quantum Information Processing*, 20(6), 204

[5] Zhang, G.-L., Liu, D., & Yung, M.-H. (2021). Observation of exceptional point in a PT broken non-Hermitian system simulated using a quantum circuit. *Scientific Reports*, 11(1), 13795

[6] Kosugi, T., Matsushita, Y.-i., et al. (2022). Imaginary-time evolution using forward and backward real-time evolution with a single ancilla: First-quantized eigensolver algorithm for quantum chemistry. *Phys. Rev. Res.*, 4, 033121

Pin Yu Chew

Yusuf Hamied Department of Chemistry

DESIGNING MULTIPHASE BIOMOLECULAR CONDENSATES

Biomolecular condensates are highly multi-component systems in which complex phase behaviour can ensue, including the formation of architectures comprising multiple immiscible condensed phases. Relying solely on physical intuition to manipulate them is difficult because of the complexity of their composition. We address this challenge by developing a computational approach which couples a genetic algorithm to a residue-resolution coarse-grained protein model to design pairs of protein sequences that result in well-separated multilayered condensates. We find that the balance of homo- and heterotypic interaction energies between the two components is important for controlling multiphase organisation, and our approach for designing multilayered condensates achieves this balance of interaction energies by optimising the composition and patterning of the protein sequences.

Flaviano Della Pia

Yusuf Hamied Department of Chemistry

DMC-ICE13: AMBIENT AND HIGH PRESSURE POLYMORPHS OF ICE FROM DIFFUSION MONTE CARLO AND DENSITY FUNCTIONAL THEORY

Ice is one of the most important and interesting molecular crystals, exhibiting a rich and evolving phase diagram. Recent discoveries mean that there are now 20 distinct polymorphs; a structural diversity that arises from a delicate interplay of hydrogen bonding and van der Waals dispersion forces. This wealth of structures provides a stern test of electronic structure theories, with Density Functional Theory (DFT) often not able to accurately characterize the relative energies of the various ice polymorphs. Thanks to recent advances that enable the accurate and efficient treatment of molecular crystals with Diffusion Monte Carlo (DMC), we present here the DMC-ICE13 dataset; a dataset of lattice energies of 13 ice polymorphs. This dataset encompasses the full structural complexity found in the ambient and high-pressure molecular ice polymorphs, and when experimental reference energies are available, our DMC results deliver sub-chemical accuracy. Using this dataset, we then perform an extensive benchmark of a broad range of DFT functionals. Our results suggest that a single functional achieving reliable performance for all phases is still missing, and that care is needed in the selection of the most appropriate functional for the desired application. The insights obtained here may also be relevant to liquid water and other hydrogen-bonded and dispersion-bonded molecular crystals.

John Hayton

Yusuf Hamied Department of Chemistry

PREDICTING CRYSTAL MORPHOLOGY FROM SIMULATION - IS KMC ENOUGH?

A crystal's morphology plays a vital role in determining properties such as catalytic activity. The growth processes that determine morphology, however, are only poorly understood at the molecular level. In this work, we use a recently developed kinetic Monte Carlo (KMC) approach to explore the growth of NaCl crystals at different supersaturations. We demonstrate an alchemical method for obtaining the free energies of sodium chloride lattice sites relative to a kink site, which we then use to parameterize the KMC. The resulting crystals capture some of the known morphological behaviour of NaCl, but fail to capture the shift to octahedral morphologies at higher supersaturations. Finally, we discuss possible explanations for this shortcoming, and suggest possible improvements to the model to remedy this issue.

Josiah-Shem Davis

Aston University

CONSTRUCTING LAGRANGIANS FOR DISSIPATIVE SYSTEMS: APPLICATION TO HYDRODYNAMICS

It had been believed for almost a century that “The equations of motion of a dissipative linear dynamical system with constant coefficients are not given by a variational principle.” [1]; however only integer-order time derivatives were considered. This poster offers an introduction to an approach pioneered by Fred Riewe (1996) and later developed upon by Matheus J. Lazo and Cesar E. Krumreich (2014) by which the use of fractional differentiation allows for the derivation of dissipative dynamics from within the Lagrangian formulation of mechanics. In this poster a construction of a Lagrangian approach for dissipative systems is introduced in the context of application to hydrodynamics via the Smoothed Dissipative Particle Dynamics (SDPD) model [2]; a coarse-grained formulation of the non-isothermal Navier-Stokes’ equations for hydrodynamics.

[1] Bauer, P. S. (1931). Dissipative dynamical systems. i. *Proceedings of the National Academy of Sciences of the United States of America*, 17(5), 311–314

[2] Español, P., & Revenga, M. (2003). Smoothed dissipative particle dynamics. *Phys. Rev. E*, 67, 026705

Harry Moore

Department of Engineering

TRANSFERRABLE MACHINE LEARNING POTENTIALS FOR ORGANIC CHEMISTRY

Molecular dynamics is a workhorse in computational biophysics, enabling insight into the dynamics of large systems over long timescales. Typically, empirical forcefields are employed, which trade off accuracy for performance. Recently, machine learning potentials have been developed which are capable of describing potential energy surfaces at essentially quantum chemical accuracy, however these have largely been system-dependent and non-transferrable. In this work, we employ the MACE architecture to develop a general machine learning potential capable of accurately describing a range of molecular and biomolecular systems, including organic liquids, proteins, and drug-like molecules. We benchmark its predictive performance on free energy, secondary structure and condensed phase property calculations.

Kamil Iwanowski

Department of Physics

QUANTIFICATION OF DISORDER WITH TOPOLOGICAL ENTROPY IN DISORDERED CARBON STRUCTURES

Disordered forms of carbon find use in various industrial applications. Tetrahedral amorphous carbon is utilized in electronic devices and wear-resistant coatings; nanoporous carbon is used in batteries, supercapacitors and devices designed for energy conversion and water desalination. The relationship between the structural, vibrational and thermal properties of these materials is unclear, owing to a lack of effective descriptors that correlate with thermal properties in disordered systems. This study presents calculations of thermal conductivity for 20 models of amorphous and nanoporous carbon, using the Wigner formulation of thermal transport and quantum-accurate GAP potential for carbon. To quantify disorder in different carbon allotropes, we introduced a new quantitative measure of disorder called topological entropy, based on the distribution of local atomic environments, classified using the H_1 barcode. Topological entropy was extensive with respect to the number of atoms in local environments and the quotient between topological entropy and the number of atoms ($S(n)/n$) was used as an intrinsic measure of topological disorder. We found a negative correlation between room-temperature thermal conductivity divided by density and $S(n)/n$. By using $S(n)/n$ as a classifier of disorder in the coordination topology of a solid, amorphous carbon was the most disordered structure, while graphitic-like nanoporous carbon was the most ordered structure among the materials considered. The topological entropy used in this study provides fundamental insights on relationships between material's atomistic structure and thermal conductivity. These findings could potentially have technological applications, as they may allow to use the thermal conductivity - a quantity easy to measure in experiments - as a descriptor for the degree of topological disorder in solids.

Domantas Kuryla

Yusuf Hamied Department of Chemistry

AUTOMATED DISCOVERY OF REACTION PATHWAYS IN GAS PHASE USING MACHINE LEARNING POTENTIALS

Computer simulation is an important tool in understanding mechanisms and predicting reaction outcomes in organic chemistry. The large computational cost of the quantum chemical calculations limits the length- and time-scales available to the simulation. Machine learning potentials (MLPs) accelerate the simulation of chemical reactions by approximating the potential energy surface obtained using quantum chemistry. To train an MLP, a dataset representative of the system needs to be built. Using an active learning approach, we can explore gas phase reaction pathways when only the reactants and products are known at the start, with demonstration on S_N2 reactions.

Dr. Hrishit Banerjee

Yusuf Hamied Department of Chemistry

THE IMPORTANCE OF ELECTRONIC CORRELATIONS IN EXPLORING THE EXOTIC PHASE DIAGRAM OF LI ION BATTERY CATHODES

Using *ab initio* dynamical mean-field theory (DMFT) we explore the electronic and magnetic states of layered Li_xMnO_2 as a function of x , the state of charge. Constructing real-space Wannier projections of Kohn-Sham orbitals based on the low-energy subspace of Mn $3d$ states and solving a multi-impurity problem, our approach focuses on local correlations at Mn sites. The antiferromagnetic insulating state in LiMnO_2 has a moderate Néel temperature of $T_N = 296\text{ K}$ in agreement with experimental studies. Upon delithiation, the system has multiple inequivalent sites and we develop a computationally tractable method for DMFT calculations for multiple sites with low statistical noise. We observe that the system proceeds through a number of states on charging: ferrimagnetic correlated metals at $x = 0.92, 0.83$; multiple charge disproportionated ferromagnetic correlated metals with large quasiparticle peaks at $x = 0.67, 0.50, 0.33$; ferromagnetic metals with small quasiparticle peaks at $x = 0.17, 0.08$ and an antiferromagnetic insulator for the fully delithiated state, $x = 0.0$. At moderate states of charge, $x = 0.67 - 0.33$, a mix of $+3/ +4$ formal oxidation states of Mn is observed, while the overall nominal oxidation of Mn state changes from $+3$ in LiMnO_2 to $+4$ in MnO_2 . In all these cases the high-spin state emerges as the most likely state in our calculations considering the full d manifold of Mn based on the proximity of e_g levels in energy to t_{2g} . We observe a crossover from coherent to incoherent scattering behaviour on delithiation as function of state-of-charge. We show that the charge disproportionated mixed valence states arise in cases with different quasiparticle peaks at different sites, and postulate this ordering to be present for a range of x . The pathways leading to structural transformations from layered to spinel structure in LiMnO_2 may be traced back to such charge disproportionated state, and this helps us in understanding degradation in cathodes.

[1] Banerjee, H., Grey, C. P., & Morris, A. J. (2023). The importance of electronic correlations in exploring the exotic phase diagram of layered Li_xMnO_2 . *Phys. Rev. B*

Dr. Peter Cooke

Department of Materials Science and Metallurgy

SIMULATING MATERIALS AT HIGH-PRESSURE WITH EPHEMERAL DATA DERIVED POTENTIALS

Dynamic compression techniques have allowed new and interesting materials to be probed at extreme pressures. However, the transient nature of the pulse limits the information that can be extracted from a given experiment. Simulations can provide a useful tool to aid interpretation of this experimental data, but capturing the full complexity of the experiment requires large system sizes and time-scales of nano-seconds, all at a quantum mechanical level of accuracy. Classical simulations using data derived potentials have been shown to maintain this ‘quantum accuracy’ at a fraction of the computational cost, allowing much larger length and time-scales to be explored. We present results from the recently developed ephemeral data derived potential (EDDP) approach that has been integrated into the LAMMPS simulation package. Using a range of example systems, we demonstrate that the EDDPs are a versatile tool for fast and accurate simulations at high pressure.

Rokas Elijošius

Department of Engineering

ZERO-SHOT MOLECULE GENERATION USING SIMILARITY KERNELS

Diffusion, or score-based, generative models have achieved state-of-the-art results in molecular generation and docking. While the relationship between the score function and force has already been established, the associated energy remains unexplored. We analyse the energy surface of a trained diffusion model and propose a similarity kernel based on MACE descriptors that can generate molecules without any additional training. Furthermore, using a similarity kernel we can tailor the shape of the generated molecules by changing the distribution the generation starts from. Since the kernel optimizes local similarity, it is easily scalable and extensible compared to trained models.

Eszter Varga-Umbrich

Department of Engineering

LOW-BARRIER ESCAPE MECHANISM FOR H FROM Fe

Experimental findings regarding the ingress and egress of hydrogen from iron contradict theoretical perspectives on these processes. To gain deeper insights, we employed a machine learning interatomic potential based on CASTEP single-point energy calculations, enabling extensive pathway searches, large system sizes, and long time scale simulations. Our iteratively trained potential has successfully simulated escape mechanisms through the (110) and (100) surfaces, exhibiting energy convergence to the ground truth values. Hydrogen embrittlement, which is closely associated with the rapid diffusion of lattice H atoms in crystalline structures, potentially occurs through quantum mechanical tunnelling, even at room temperature. By employing zero-point energy (ZPE) correction and phonon calculations using the potential, we could predict the probability of this effect. The escape barrier per hydrogen molecule through the (110) surface decreased by 64% when aided by lattice H, indicating a significant reduction that counters the influence of low diffusing H concentration and likely is the dominant mechanism. Furthermore, we have found that the effects of temperature and pressure play a more critical role in the coverage and pathways through the (100) surface, which are currently under further investigation.

Bogdan Rajkov

Department of Physics

PHASE TRANSITIONS UNDER PHOTOEXCITATION IN GRAPHITE AND HALIDE PEROVSKITES

Recently, advances have been made in theoretical descriptions of materials under monochromatic illumination. By introducing separate chemical potentials for photoexcited populations of electrons and holes, we can explain a broad range of phenomena in which photoexcitation induces changes in crystal structure. In this work, we have examined graphite and an inorganic halide perovskite, CsPbBr₃, as examples of crystals where properties under photoexcitation are of great interest. In CsPbBr₃, we identified a possible orthorhombic-to-cubic phase transition, which could prove important to this material's performance as a photovoltaic. In graphite, the presence of a characteristic nodal line was examined under photoexcitation. Disappearance of this degeneracy in energy levels is an example of a topological phase transition, and it could signify a phase transition to a diamond-like structure, thus explaining the mechanism behind experimental observations.

Yuthika Pillai

Yusuf Hamied Department of Chemistry

SCF LANDSCAPES FOR SYMMETRY BREAKING IN BENZENE

Hartree-Fock theory is known to predict symmetry-broken ground state solutions for molecules like C_{60} , C_{36} and C_{20} . In this study, we investigate the spin symmetry breaking in benzene that was previously predicted by Tóth using SCF landscapes for electron structures. The novel method for electronic structures described by Burton and Wales was able to give much deeper insight into the and models by providing a connected database of the solutions obtained, giving qualitative insight into the problem. Using the Hartree-Fock level of theory, benzene was found to break S^2 symmetry. We investigate this symmetry breaking by constructing SCF landscapes using different basis sets and functionals. The size of the basis set was found to have a negligible effect on the symmetry breaking and hence the basis set 6-31G(D) was used for the remainder of the studies. To study the impact of the Hartree-Fock exchange on the symmetry breaking, the results show that by varying the exact Hartree-Fock exchange contribution for PBE functional, the system changed from a symmetry unbroken state to a symmetry broken state.

Se Hun Joo

Department of Materials Science and Metallurgy

EXPEDITING CATHODE MATERIAL DISCOVERIES THROUGH AB INITIO RANDOM STRUCTURE SEARCHES AND MACHINE-LEARNED INTERATOMIC POTENTIALS

Developing advanced lithium-ion battery materials is essential for electric vehicles, portable electronics, and renewable energy storage systems. However, traditional experimental methods are time-consuming and costly. As a promising computational approach for materials discovery without any prior knowledge, *ab initio* random structure searching (AIRSS) has gained considerable attention. For large and complex systems, however, it remains computationally demanding. To address this, we present here an approach that combines AIRSS with ephemeral data-derived potentials (EDDP), a machine-learned interatomic potential. It enables rapid and accurate computation of energy landscapes for extensive structure explorations. We discuss our iterative training scheme for EDDP using small unit cells over various stoichiometries. In testing different Li-containing systems, we were able to easily rediscover the known polymorphs. Additionally, EDDP-based structure searches can significantly reduce computation costs. We anticipate that this approach can be used effectively to accelerate the discovery of advanced battery materials.