



Lennard Jones Centre

Showcase Day

28th October 2024
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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 third 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 two talks from our industry sponsors. We are also very excited to invite Prof. Volker Heine for a talk and extended discussion, where he will offer a unique historical perspective and share his vast experiences in the field. Finally we are very happy to welcome Prof. Rosana Colleparado, who will lead a thought-provoking discussion on gender equality in research, organized by members of the LJC Gender Equality Network.

Finally, we are very grateful to our sponsors Radical AI, Valence Labs and Quantinuum for their generous support of the event.

Showcase Day Organising Committee

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Schedule

09:30 Arrival and Welcome (*coffee and tea provided*)

Session 1

Chair: Pascal Salzbrenner

10:00 **William Baldwin**, *Department of Engineering*

Paths to liquid water at coupled cluster accuracy

10:15 **Niamh O'Neill**, *Yusuf Hamied Department of Chemistry / Department of Physics*

To Pair or not to Pair? Ascending Jacob's Ladder for ions in solution

10:30 **Anna Bui**, *Yusuf Hamied Department of Chemistry*

Learning classical density functionals for ionic fluids

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

Gold Sponsor Talk

Chair: Dr Fabian Berger

11:00 **Radical AI**, Janosh Riebesell

Invited Guest Talk 1

Chair: Dr Christoph Schran

11:30 **Prof. Volker Heine**, *Theory of Condensed Matter, Cavendish Laboratory*

Understanding metal bonding

Followed by extended discussion and historical perspective.

12:15

Lunch

12:30

Poster Session

Invited Guest Talk 2

Chairs: Lila Cadi Tazi, Chiara Leadbeater, Yuthika Pillai and Paula Teeuwen

13:30 **Prof. Rosana Colleparodo**, *Yusuf Hamied Department of Chemistry*

LJC Gender Equality Network : Discussing gender issues in research careers

Silver Sponsor Talk

Chair: Dr Fabian Berger

14:15 **Valence Labs**, Hatem Helal

14:30 Break (*coffee, tea, and juice provided*)

Session 2

Chair: Benjamin Shi

15:00 **Kamil Iwanowski**, *Department of Physics*

Bond-network entropy controls thermal conductivity of coordination-disordered solids

15:15 **Ingvars Vitenburgs**, *Department of Physics, Imperial College London*

Extended Coupled Cluster approach to Twisted Graphene Layers

15:30 **Fanzhi Su**, *Department of Materials Science*

Deep Learning-Enhanced Multivariate Analysis of EDX Signals: Pushing the limits of Elemental Analysis

Session 3

Chair: Dr Maria-Andreea Filip

15:45 **Patryk Adam Wesolowski**, *Yusuf Hamied Department of Chemistry*

Multilevel Frameworks for Studying Protein Energy Landscapes

16:00 **Pin Yu Chew**, *Yusuf Hamied Department of Chemistry*

Physical determinants of multiphase organisation in protein/RNA condensates

16:15 **Chiara Leadbeater**, *Yusuf Hamied Department of Chemistry*

Non-unitary Trotter circuits for imaginary time evolution

16:30 Conclusion and Awards (*Followed by wine and cheese*)

Guest Speaker Bios

Prof. Volker Heine

Theory of Condensed Matter, Cavendish Laboratory, University of Cambridge

BIOGRAPHY

Born in Germany in 1930, I was lucky to get out to New Zealand with the help of Quakers and grew up there with a local family. After a M.Sc. degree in both maths and physics, I came to Cambridge on a Shell Scholarship in 1954, and have got stuck here! It was the same year as the arrival of Prof. N.F. Mott as Cavendish Professor and the start of the TCM group (then called Solid State Theory). My thesis with him was on calculating the electronic band structure of aluminium and relating it to experimental work in the Low Temperature Group. Since then my main work has been developing the concept of pseudopotentials, some surface physics, incommensurately modulated structures, application of ‘computer experiments’ to problems in mineralogy, and promoting sharing of computer codes across Europe. A recent paper (with S. Chen) is on ‘Understanding Metal Bonding’ [1].

[1] Heine, V., & Chen, S. (2024). Understanding metal bonding. *Journal of Physics: Condensed Matter*, 36(35), 353002

Prof. Rosana Colleparado

*Professor of Computational and Molecular Biophysics, Yusuf Hamied Department of Chemistry and Department of Genetics
Winton Advanced Research Fellow, Department of Physics*

BIOGRAPHY

Rosana Colleparado is an Interdisciplinary Professor of Computational and Molecular Biophysics at the Departments of Genetics and Chemistry, University of Cambridge. Her research focuses on developing multi-scale models to explore the mechanisms governing DNA packaging within cells, including the formation of biomolecular condensates.

A native of Mexico City, Rosana earned her undergraduate degree in Chemistry and a specialization in High-Performance Scientific Computing from the National Autonomous University of Mexico. In 2009, she completed her DPhil in Physical and Theoretical Chemistry at the University of Oxford.

Following her doctorate, Rosana held a Schlumberger Faculty for the Future Fellowship at New York University (2009–2011) and a Marie Curie Research Fellowship at the Institute for Biomedical Research in Barcelona (2011–2013). In October 2013, she joined the University of Cambridge for postdoctoral work with Daan Frenkel and David Wales. Soon after, Rosana took her first career break for maternity leave in July 2014 (six months), followed by six months of part-time work. In March 2016, she launched her independent research group at Cambridge under the Winton Advanced Research Fellowship. Later that year, in September, she took a second career break for maternity leave (six months), again followed by six months of part-time work. Rosana became an Assistant Professor in 2020 and was promoted to Professor in 2022. She currently serves as the Director of Postgraduate Education in Chemistry and as Fellow and Overall Director of Studies in Natural Sciences (Physical) at Clare College.

Sponsors Talks

Radical AI

Janosh Riebesell

Radical AI is a start-up focused on combining machine learning, atomistic simulations and laboratory automation to develop new materials. During the talk, we will dive into the science Radical AI have been working on across data generation and Machine Learning Interatomic Potentials.

Valence Labs

Hatem Helal

Electronic structure simulation (ESS) has been used for decades to provide quantitative scientific insights on an atomistic scale, enabling advances in chemistry, biology, and materials science, among other disciplines. Following standard practice in scientific computing, the software packages driving these studies have been implemented in compiled languages such as FORTRAN and C. However, the recent introduction of machine learning (ML) into these domains has meant that ML models must be coded in these languages, or that complex software bridges have to be built between ML models in Python and these large compiled software systems. This is in contrast with recent progress in modern ML frameworks which aim to optimise both ease of use and high performance by harnessing hardware acceleration of tensor programs defined in Python. We introduce MESS: a modern electronic structure simulation package implemented in JAX; porting the ESS code to the ML world. We outline the costs and benefits of following the software development practices used in ML for this important scientific workload. MESS shows significant speedups on widely available hardware accelerators and simultaneously opens a clear pathway towards combining ESS with ML.

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Abstracts of Student Talks

William Baldwin

Department of Engineering

PATHS TO LIQUID WATER AT COUPLED CLUSTER ACCURACY

There is much interest in making a truly first principles model of liquid water. The MB-pol model, parameterised to calculations of 4 water molecule clusters, predicts many properties well, but a limiting factor is that it cannot handle proton transfer. Similarly, Marx and coworkers recently presented a new machine learning potential for liquid water trained to coupled cluster which, while not limited to whole molecules in principle, was not trained or demonstrated on proton transfer. To address this, we present a modification to the MACE MLIP architecture which is capable of extrapolating from clusters to condensed phase and which has sufficient physics to model proton transfer. By training only on clusters, our model can correctly recover bulk water properties including the diffusivities of excess protons. So far, we have demonstrated this by training on density functionals where an AIMD reference is available. Our models contain a coarse grained description of the charge and current density, allowing one to access properties like the infrared spectra, or apply external fields.

Niamh O’Neill

Department of Chemistry, Department of Physics

TO PAIR OR NOT TO PAIR? ASCENDING JACOB’S LADDER FOR IONS IN SOLUTION

The extent of ion pairing in solution is an important phenomenon to rationalize transport and thermodynamic properties of electrolytes. A fundamental measure of this pairing is the potential of mean force (PMF) between solvated ions. The relative stabilities of the paired and solvent shared states in the PMF and the barrier between them are highly sensitive to the underlying potential energy surface. However, direct application of accurate electronic structure methods is challenging, since long simulations are required. We develop wave function based machine learning potentials with the random phase approximation (RPA) and second order Møller–Plesset (MP2) perturbation theory for the prototypical system of Na and Cl ions in water. We show both methods in agreement, predicting the paired and solvent shared states to have similar energies (within 0.2 kcal/mol). We also provide the same benchmarks for different DFT functionals as well as insight into the PMF based on simple analyses of the interactions in the system. We finish with an outlook on using these models to target transport properties of electrolytes, where both an accurate electronic structure description yet long simulation times are crucial.

Anna Bui

Department of Chemistry

LEARNING CLASSICAL DENSITY FUNCTIONALS FOR IONIC FLUIDS

Owing to their importance across both science and technology, developing accurate yet manageable theoretical descriptions of electrolyte solutions and ionic liquids has challenged scientists for over a century. The difficulties arise from the fierce competition between the strong, long-ranged Coulombic interactions and steric repulsions at shorter range, which is completely missing in traditional Debye-Hückel and Poisson-Boltzmann theories. While classical density functional theory (cDFT) in principle offers an exact statistical mechanical framework for this problem, the approximations inherent to existing functionals have limited its applicability, pushing researchers to rely on costly molecular simulations.

In this work, we generalize a recently proposed machine learning (ML) approach for representing the exact free energy functional of “simple liquids” to ionic fluids. Our approach solves the critical problem of incorporating non-local effects within the ML framework by leveraging the connection between cDFT and local molecular field theory. The resulting ML-based cDFT functional to be applied to much larger systems than those in the training set, quantitatively captures simulation data and ensures thermodynamic consistency. Our work opens a new avenue for studying ionic fluids and electrolyte solutions, not just by existing practitioners of cDFT, but also by those relying on molecular simulations. Furthermore, this work represents a critical step in evolving cDFT from a tool typically used in soft matter—where interaction potentials are often simpler—into a robust framework for chemical physics, where more complex potentials, such as those for ions and water, are common.

Kamil Iwanowski

Department of Physics

BOND-NETWORK ENTROPY CONTROLS THERMAL CONDUCTIVITY OF COORDINATION-DISORDERED SOLIDS

Understanding how the vibrational and thermal properties of solids are influenced by atomistic structural disorder is of fundamental scientific interest, and paramount to designing materials for next-generation energy technologies. While several studies indicate a strong dependence of thermal conductivity on structural disorder, the fundamental physics governing the disorder-conductivity relation remains elusive.

Here we show that order-of-magnitude, disorder-induced variations of conductivity in network solids can be predicted from a ‘bond-network’ entropy, an atomistic structural descriptor that quantifies heterogeneity in the topology of the atomic-bond network. We employ the Wigner formulation of thermal transport to demonstrate the existence of a relation between the bond-network entropy, and observables such as smoothness of the vibrational density of states (VDOS) and macroscopic conductivity. We also show that the smoothing of the VDOS encodes information about the thermal resistance induced by disorder, and can be rigorously related to phenomenological models for phonon-disorder scattering based on the semiclassical Peierls-Boltzmann equation. Our findings rationalize the conductivity variations of disordered carbon polymorphs ranging from nanoporous electrodes to defective graphite used as a moderator in nuclear reactors.

Ingvars Vitenburgs

*Department of Physics and Astronomy, University of Manchester & Molecular Sciences
Research Hub, Imperial College London*

EXTENDED COUPLED CLUSTER APPROACH TO TWISTED GRAPHENE LAYERS

A study of correlation effects in twisted bilayer graphene is presented using the extended coupled cluster method at consistent mean-field and beyond mean-field truncations. In order to describe the phase transitions in this strongly correlated system, equations and a suitable implementation is developed for the method. Combining modern tensor contraction techniques with singular value decomposition, the correlation effects are successfully described in a qualitative manner, with the majority of both short-range and long-range Coulomb interactions included. Superconductivity is derived, analytically, to be possible only when beyond-mean-field correlations are included at non-zero fillings. Furthermore, the superconducting gap is found to peak at an angle $\theta_c = 1.00$ with a BCS value for the critical temperature of $T_c^{\text{BCS}} = 0.8$ K, matching closely with experimental data. Additionally, while a homogeneous (s-wave) gap is obtained, processes for the insulating phases are found to be not described by these effects, a fact observed in previous experimental studies. The simulations are somewhat limited by the truncations used, but nevertheless, a strong candidate for the mechanism behind superconductive phases in twisted bilayer graphene is presented.

Fanzhi Su

Department of Materials Science and Metallurgy

DEEP LEARNING-ENHANCED MULTIVARIATE ANALYSIS OF EDX SIGNALS: PUSHING THE LIMITS OF ELEMENTAL ANALYSIS

Nanoscale materials characterization often uses highly energetic probes which can rapidly damage beam-sensitive materials, such as hybrid organic–inorganic compounds. Reducing the probe dose minimizes the damage, but often at the cost of lower signal-to-noise ratio (SNR) in the acquired data. This work reports the optimization and validation of principal component analysis (PCA) and nonnegative matrix factorization for the postprocessing of low-dose nanoscale characterization data. PCA is found to be the best approach for data denoising. However, the popular scree plot-based method for separation of principal and noise components results in inaccurate or excessively noisy models of the heterogeneous original data, even after Poissonian noise weighting. Manual separation of principal and noise components produces a denoised model which more accurately reproduces physical features present in the raw data while improving SNR by an order of magnitude. However, manual selection is time-consuming and potentially subjective. To suppress these disadvantages, a deep learning-based component classification method is proposed. The neural network model can examine PCA components and automatically classify them with an accuracy of >99% and a rate of ~2 component/s. Together, multivariate analysis and deep learning enable a deeper analysis of nanoscale materials' characterization, allowing as much information as possible to be extracted.

Patryk Adam Wesolowski

Department of Chemistry

MULTILEVEL FRAMEWORKS FOR STUDYING PROTEIN ENERGY LANDSCAPES

Proteins, which make up over 50% of dry cell mass, are vital to nearly all biochemical processes, with their functionality depending on proper folding into native structures. However, simulating the complex structural dynamics of proteins over relevant timescales is beyond the reach of accurate quantum mechanical methods. Therefore, mechanical force fields and coarse-grained models have been developed to simplify protein behaviour by reducing interaction sites. Despite advances like AlphaFold improving our ability to predict native structures, understanding the thermodynamic stabilities and pathways driving these processes remains challenging, particularly for larger proteins.

Here, developed multilevel frameworks that integrate various levels of theory to analyse protein energy landscapes are presented. A key outcome of this work is the standalone multilevel, multicentre lwONIOM library, which is freely available online. This tool enables accurate and efficient analysis of complex proteins across relevant timescales. Applied to the bovine pancreatic trypsin inhibitor (BPTI), it effectively balances computational cost and precision, providing valuable insights into structural stability and dynamics. Additionally, we also integrated the UNited RESidue (UNRES) coarse-grained potential into the Cambridge energy landscapes software, facilitating efficient calculations for large biomolecular structures. This approach maintains accuracy while significantly reducing computational time with results aligning well with experimental data and all-atom force fields.

Furthermore, by combining methodologies such as machine learning potentials, coarse-grained energy landscape searches, and structural analysis, we investigated amyloid monomers related to Alzheimer's disease. This work identified key structural features linked to protein misfolding, offering insights that could guide the development of therapeutic strategies for neurodegenerative diseases. These multilevel frameworks, therefore, provide powerful tools for studying complex biomolecular systems and have significant potential for enhancing drug design and other health-related research.

Pin Yu Chew

Department of Chemistry

PHYSICAL DETERMINANTS OF MULTIPHASE ORGANISATION IN PROTEIN/RNA CONDENSATES

Biomolecular condensates are fundamental players in cellular (dys)function, and understanding the phase behaviour and underlying driving forces of their formation is important to both determine how functionality arises in cellular structures, and also to create new functions or alter dysfunctions. Internal subcompartmentalisation of biomolecules to form multiphase architectures has been observed in several biomolecular condensates, where immiscible liquid-like phases with different compositions can coexist within the same compartment. Computer simulations are a useful tool to help us probe the phase behaviour of biomolecular mixtures, as well as the properties of the resulting condensates at a molecular level. Here, we couple a genetic algorithm to molecular dynamics simulations using a residue-resolution coarse-grained model to evolve protein/RNA mixtures towards demixing into compositionally distinct liquid-like phases. We use this method to design multiphasic protein condensates by predicting a partner protein to a given target protein that supports multiphasic compartmentalisation, as well as further investigate the underlying thermodynamics of multiphasic condensate formation, and the role of protein-protein and protein-RNA interactions in stabilising multiphasic architectures in protein/RNA condensates.

Chiara Leadbeater

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, using mid-circuit measurements on a single ancilla qubit. Imaginary time evolution (ITE) is widely used for obtaining the ground state (GS) of a system on classical hardware, computing thermal averages, and as a component of quantum algorithms that perform non-unitary evolution. Near-term implementations on quantum hardware rely on heuristics, compromising their accuracy. 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 ITE (PITE) algorithms, 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. Compared with previous PITE proposals, the suggested block encoding results in shorter circuits and is simpler to implement, requiring only a slight modification of the Pauli gadget primitive. This scheme was tested on the fermionic Hubbard model and is demonstrated to converge to the GS of the system [1].

[1] Leadbeater, C., Fitzpatrick, N., Ramo, D. M., & Thom, A. J. W. (2024). Non-unitary Trotter circuits for imaginary time evolution. *Quantum Science and Technology*, 9(4), 045007

Abstracts of Posters

Javiera Quiroz-Fernandez

Department of Physics

BAROCALORIC STUDY OF AgI USING MACHINE LEARNING POTENTIALS

Barocalorics offers a sustainable path to more efficient cooling devices using solid-state materials. To date, experimental research has dominated the field — largely due to the complexity of simulating the phase transitions observed during the thermodynamic cycle, and the diverse family of materials with promising caloric properties. In this poster, I will introduce different protocols that use Machine Learning potentials to obtain barocaloric effects, using the solid electrolyte AgI as case study.

Anastasia Kryachkova

Molecular Sciences Research Hub, Imperial College London

INVESTIGATING NUCLEAR QUANTUM EFFECTS ON WATER ENCAPSULATION IN FUNCTIONALIZED FULLEROID CAGES USING MACE FORCEFIELDS

This ongoing study investigates the encapsulation and release dynamics of water molecules within oxygen-functionalized fulleroid cages, with a focus on the influence of nuclear quantum effects. By employing MACE-MP-0 forcefield in molecular dynamics (MD) and path integral molecular dynamics (PIMD) simulations, we explore how quantum effects alter water stabilization and release behavior. Initial findings suggest that water molecules are temporarily stabilized near oxygen groups, as supported by potential energy surface (PES) and nudged elastic band (NEB) analysis. Differences between the MACE-MP-0 and MACE-OFF23 forcefields were also observed, with MACE-MP-0 providing a significantly more accurate representation of potential energy. Additionally, metadynamics simulations highlight the importance of quantum nuclear effects on the energy barriers involved in water encapsulation. While the research remains a work in progress, these early results suggest that nuclear quantum effects, captured through MACE-MP-0, are critical for accurately modeling fulleroid cage behavior and optimizing them for molecular encapsulation applications.

John Hayton

Department of Chemistry

CHALLENGES OF CRYSTAL MORPHOLOGY PREDICTION: HYDRATION EFFECTS AT THE GROWING HALITE SURFACE

Modelling growing ionic surfaces is challenging, due to the high computational cost of measuring attachment rates, and the countless number of unique growth sites on the crystal. However, any attempt to simplify or parameterize models of crystal growth encounters the challenge of strong hydration effects at the sodium chloride surface. Here, we explore these strong solvent binding effects on the sodium chloride 100 surface. We look at both the relative free energy of different sites, and the detachment rates into solution. We find a qualitative match between the two measurements, demonstrating the free energies can act as a useful starting point for identifying the most important growth units on the crystal.

Jad Jaafar

Department of Engineering

MODELLING DEFECT DYNAMICS AND REACTIONS IN TMDs

Transition metal dichalcogenides (TMDs) are a class of 2D materials that hold potential to transform nanoelectronics. These materials often feature defects that significantly affect their properties. Such defects are pivotal in material oxidation processes, with substitutional oxygen being a notably prevalent defect in TMDs. As exemplified by WS_2 , oxidation can either cause unwanted corrosion or be utilized for precise etching and interface creation. Recent studies using operando scanning electron microscopy have revealed that the oxidation of monolayer WS_2 exhibits complex behavior due to various competing mechanisms. This complexity challenges existing kinetic models and calls for a deeper atomistic understanding, prompting the use of machine learning interatomic potential (MLIP) models trained on density functional theory (DFT) data. The introduction of the message passing neural network, MACE, has significantly improved the accuracy of DFT parameterization. Our research involves training a MACE potential for WS_2 to investigate defect dynamics and oxidation processes.

Kun Yang

Department of Physics

MECHANICAL AND ELECTRICAL PROPERTIES OF NOVEL ULTRA-THIN LAYERED FERROELECTRIC BISMUTH OXIDE STRUCTURES FROM FIRST PRINCIPAL CALCULATIONS

Ferroelectric materials at the nanometre scale play a crucial role in the miniaturization of essential electronic components, such as memory devices, capacitors, piezoelectric sensors, and thin-film transistors. Recent research has proposed a new Bismuth Oxide structure that retains ferroelectricity down to the 1-nanometer scale. In our study, we applied first-principles calculations to both the bulk and thin-layered forms of this novel structure. Using density functional theory (DFT), we successfully confirmed the bulk paraelectric, bulk ferroelectric, and layered paraelectric phases. Phonon calculations allowed us to identify distortion patterns, and our band structure analysis revealed a band gap of 1.4 eV, further demonstrating the material's ferroelectric properties. This work offers additional insights into the potential applications of this material and similar ones in future research and industrial developments.

Bogdan Rajkov

Department of Physics

COUPLED ELECTRON-PHONON TRANSPORT AND VISCOUS THERMOELECTRIC EQUATIONS

Non-diffusive, hydrodynamic transport of charge or heat has been observed in several materials, and recent, pioneering experiments have suggested the possible emergence of electron-phonon bifluids. Here we introduce the first-principles computational framework to describe these phenomena, showing that the viscosity of electron-phonon bifluids is microscopically determined by composite ‘relaxon’ excitations. We show how the coupled Boltzmann transport equation can be coarse-grained into a set of mesoscopic Viscous Thermo-electric Equations (VTE).

Kieran Bozier

Department of Materials Science and Metallurgy

ELIASHBERG THEORY IN SUPERCONDUCTING ELEMENTS

First principles calculations of superconducting properties, notably the critical temperature (T_c), have been widely used to search for superconductivity at ambient conditions. Theoretical methods have successfully predicted high T_c hydride superconductors such as H_3S [1], and recently suggested that Mg_2IrH_6 [2][3] is an excellent candidate for high-temperature conventional superconductivity. We investigated the accuracy of the predicted transition temperature for metallic elements using Eliashberg theory, a commonly used many-body green’s function method, with electron-phonon coupling parameters derived from density functional perturbation theory. Our results show that the standard literature values for the coulomb pseudopotential parameter μ^* cause T_c to be overestimated compared to experimental results, particularly for transition metals. This suggests that μ^* may be structure-dependent or that other factors, such as spin interactions, influence the electron-phonon coupling strength.

[1] Duan, D., Liu, Y., Tian, F., Li, D., Huang, X., Zhao, Z., Yu, H., Liu, B., Tian, W., & Cui, T. (2014). Pressure-induced metallization of dense $(H_2S)_2H_2$ with high- T_c superconductivity. *Scientific Reports*, 4(1), 6968

[2] Dolui, K., Conway, L. J., Heil, C., Strobel, T. A., Prasankumar, R. P., & Pickard, C. J. (2024). Feasible Route to High-Temperature Ambient-Pressure Hydride Superconductivity. *Phys. Rev. Lett.*, 132, 166001

[3] Sanna, A., Cerqueira, T. F. T., Fang, Y.-W., Errea, I., Ludwig, A., & Marques, M. A. L. (2024). Prediction of ambient pressure conventional superconductivity above 80 K in hydride compounds. *npj Computational Materials*, 10(1), 44

Eleanor Davison

Department of Materials Science and Metallurgy

EXCITONIC VAN HOVE SINGULARITIES: A NEW APPROACH TO TUNING EXCITON TRANSPORT

Excitons – a coulomb-bound electron and hole – are the particles that govern light-matter interactions in many semiconductor materials, particularly in 2D materials and their heterostructures. Optimising the transport properties of excitons is a crucial step in improving these systems for advanced optoelectronic applications, such as in solar cells, LEDs, and light sensors. Since excitonic properties are determined by the exciton bandstructure, most successful methods of improvement focus on precise tuning of the excitonic dispersion to enhance materials performance.

One unexplored strategy is to introduce a van Hove singularity (VHS) into the excitonic dispersion. When present in electronic dispersions, VHSs are known to cause remarkable electronic phenomena such as magnetic phases and unconventional superconductivity, and thus mirroring the concept in exciton bands could lead to similarly useful enhancements in excitonic properties. Here, we present a novel excitonic bandstructure that incorporates a VHS. Using a fully microscopic many-particle theory, we investigate how the introduction of an excitonic VHS changes exciton dynamics, phonon-mediated scattering behaviour and diffusion coefficients, revealing its significant effects on the physical properties of excitonic materials. We find that highly anisotropic diffusion behaviour can be induced, with a strong dependence on the VHS shape and the relative energy of optical phonons within the material.

This work introduces an exciting method for tailoring exciton behaviour and highlights the importance of exploring more exotic exciton dispersions further.

Connie Fairchild

Department of Chemistry

ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY VIA MOLECULAR SIMULATION

An impedance spectrum contains a wealth of important information regarding the electrochemical performance of a device, however it is often unclear how to disentangle the molecular origins of the different frequency dependencies observed. This is commonly addressed using equivalent circuit fitting, however the lack of physical meaning behind this procedure can cause issues that this work aims to circumvent. We tackle these ambiguities by looking to understand the molecular-level mechanisms of charge transport in a bulk electrolyte that will consequently give rise to resistive, capacitive and inductive behaviours in an electrochemical cell. We show that a low frequency expansion of the expression for the bulk impedance produces a physically meaningful equivalent circuit representation for a bulk electrolyte. While this is often approximated in the literature by a parallel resistor-capacitor arrangement, molecular dynamics simulations suggest a three-component unit may extract more physically meaningful information.

Joshua Wu

Department of Physics

TOWARDS ONE-STEP HYDROGEN ECONOMY: PHOTOCATALYTIC PRODUCTION AND TUNABLE STORAGE IN ENDOHEDRAL-DOPED FULLERENE MONOLAYERS

Hydrogen presents both prospects in renewable energy and challenges in material design. Two crucial steps in the hydrogen economy are clean production and reversible storage. Recent synthesis of 2D covalent fullerene monolayers provides a promising platform for both applications, combining the large surface area of fullerenes and the extended structure of graphene. Previous work has shown that these structures are effective photocatalysts for overall water splitting, but poor storage materials owing to weak binding with much higher formation energy than the goal of -0.8 to -0.2 eV. We investigate the possibility of overcoming this challenge using doped monolayers, starting with Mg dopants at different configurations that were experimentally synthesised. Interstitial doping exhibits novel molecular adsorption through orbital interactions, while endohedral doping shows strong binding energies within the ideal energy range. Further high-throughput screening of endohedral dopants demonstrates a wide range of adsorption energies from -4 to $+0.5$ eV, showing prospects for tunable properties. The effect of endohedral doping on photocatalytic performance is evaluated through band structure calculations. Isolated Mg states that lie in the band gap participate weakly in the photoexcitation of electrons, hence preserving the catalytic activity. Our work sheds light on achieving generation and storage of hydrogen through a single material.

Balazs Pota

Department of Physics

THERMAL CONDUCTIVITY PREDICTIONS WITH FOUNDATION ATOMISTIC MODELS

Advances in machine learning have led to the development of foundation models for atomistic materials chemistry, enabling quantum-accurate descriptions of interatomic forces across diverse compounds at reduced computational cost. Hitherto, these models have been benchmarked relying on descriptors based on atoms' interaction energies or harmonic vibrations; their accuracy and efficiency in predicting observable and technologically relevant heat-conduction properties remains unknown. Here, we introduce a framework that leverages foundation models and the Wigner formulation of heat transport to overcome the major bottlenecks of current methods for designing heat-management materials: high cost, limited transferability, or lack of physics awareness. We present the standards needed to achieve first-principles accuracy in conductivity predictions through model's fine-tuning, discussing benchmark metrics and precision/cost trade-offs. We apply our framework to a database of solids with diverse compositions and structures, demonstrating its potential to discover materials for next-gen technologies ranging from thermal insulation to neuromorphic computing.

Darius Kayley

Department of Physics

PROPERTIES OF VARIOUS PHASES OF BUCKMINSTERFULLERENE VIA SIMULATION

The electronic, optical, and acoustic properties of four phases of buckminsterfullerene, with space groups 71, 166, 202, and 205, were investigated using computational simulations on a high-performance computer using both GGA-PBE and PBE0 functionals. Results demonstrate that symmetry plays a crucial role in the properties of a material. The different space groups have different band structures, exciton contributions to energy eigenstates, and optical absorption. The 71 and 166 phases were found to have indirect band gaps of 2.0745 eV and 2.0271 eV, respectively. The band gap for the 202-Fm-3 phase was calculated to be 2.2929 eV using PBE0, and 1.1160 eV with GGA-PBE, which overestimate and underestimate, respectively, the experimental value of 1.86 eV. Optical absorption of the four phases was also studied, and it was found that excitons affected the absorption coefficient under different electromagnetic field polarisations anisotropically. Acoustic properties were studied by plotting phonon bands; Space groups 71 and 166 show typical optical and acoustic modes; space groups 202 and 205 space groups have imaginary modes, signifying the presence of dynamical instabilities in these structures.

Jenny Pan Pan

Department of Physics

INVESTIGATION OF MATERIALS PROPERTIES IN FULLERENE NETWORKS

In this work we constructed the 2D crystal structures of hexagonal, hexagonal closed-pack (hcp) and kagome lattices with the fullerene units as building blocks. We manually chose the fullerene superatoms of either C₃ or D₆ symmetry to ensure an overall symmetrical 2D structure. We performed geometry relaxation, self-consistent calculation and plotted their individual band structures to investigate their topological properties.

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A MACHINE-LEARNED POTENTIAL INVESTIGATION OF HEAVY METALS UNDER PRESSURE

Density functional theory, has been remarkably successful at describing the electronic structure and predicting the physical properties of a wide range of materials. This success is, at least in part, based on making sensible approximations which preserve much of the accuracy while reducing the computational expense. For instance, the relativistic spin-orbit coupling (SOC) is usually neglected, resulting in a speed-up of 1-2 orders of magnitude. Since SOC increases with increasing nuclear charge, this is usually justified for light elements. This has enabled applications which rely on DFT simulations of a large number of atoms, such as ab initio molecular dynamics (AIMD), as well as those relying on a large number of simulations of more moderately-sized systems. A prominent example of the latter is high-throughput first principles crystal structure prediction.

However, for heavier elements, SOC plays an important role in the electronic structure and hence in the energy landscape. As a result, accurate first-principles quality structure searches or AIMD have been practically out of reach for these materials. As a result, the pressure-temperature behaviour of the heavy metals is theoretically much more poorly understood than that of lighter elements. For polonium, for example, no ab initio phase diagram exists whatsoever.

Here, we rely on the ephemeral data-derived potentials (EDDPs), a flavour of neural network interatomic potential, to learn the energy landscape including SOC from small-cell DFT calculations. These potentials are then used to carry out extensive structure searches and explicit melting MD calculations. Using this technique, we present the first ab initio phase diagram for lead including SOC, and the first phase diagram for polonium altogether. In both cases, we demonstrate that taking SOC into account is essential for improving agreement with experimental results.

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ELECTRONIC PROPERTIES OF $\text{Re}_6\text{Se}_8\text{Cl}_2$: EXPLORING THE POTENTIAL OF A SUPERATOMIC MATERIAL

In this study, we investigated the electronic properties of $\text{Re}_6\text{Se}_8\text{Cl}_2$, a van der Waals superatomic semiconductor recognised for its unique excitonic behaviour, which could minimise phonon scattering and improve energy transfer efficiency. Our study leverages advanced computational techniques to reveal the material's electronic structure.

We start with structural relaxation, then do comprehensive band structure analysis of both bulk and monolayer form, comparing results with and without Spin-Orbit Coupling (SOC). The band gap values obtained in both bulk and monolayer $\text{Re}_6\text{Se}_8\text{Cl}_2$ are indirect, with a modest reduction with SOC. Initial findings show that SOC has a considerable influence on the band gap, emphasising the importance of adding relativistic effects in precise modelling.

Advanced computations utilising Wannier functions produce high-quality findings that are compatible with the reported band structure. The research moves on to include hybrid functionals, switching from HSEsol to PBEsol0 and create a band gap that is more closely aligned with experimental data.

Our study lays the framework for additional explorations into excitonic effects and phonon computations, hoping to discover the potential of $\text{Re}_6\text{Se}_8\text{Cl}_2$ for future semiconductor applications. Future research will concentrate on applying these more complex approaches to improve our knowledge of the material's electronic properties and dynamic stability.

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FINDING REACTION PATHWAYS EFFICIENTLY USING MACHINE LEARNING POTENTIALS

In chemical reactivity, the information about reaction pathways is important for determining reaction mechanisms and kinetics. Finding reaction pathways using quantum mechanics methods is computationally expensive. This cost can be circumvented using reactive machine learning potentials (MLPs), which are trained using two main approaches: by training on reaction datasets and by active learning. Both approaches have so far relied on precomputed reaction pathway information. The reaction dataset approach has been limited to training on full pathways, while other diverse datasets exist that only contain reactant, transition state, and product geometries. Active learning approaches have so far required initialization with reaction pathway or transition state data, introducing a large cost at the very beginning. We aim to make reactive MLP training more efficient by leveraging the stability of the MACE architecture. We show that MACE models can be trained using datasets containing only the reactant, transition state, and product configurations, to predict pathways and transition states of unseen reactions. We also use a nudged elastic band - active learning approach that only requires reactant and product information to find the reaction pathway and obtain a reactive MLP. These approaches provide a computationally cheap way to find reaction pathways and generate reaction training data without expensive quantum mechanics driven pathway search.

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THE GRAPHENE-WATER INTERFACE IS ACIDIC

Water's ability to autoionize into hydroxide and hydronium ions profoundly influences surface properties, rendering interfaces either basic or acidic. While it is well-established that the water-air interface is acidic, a critical knowledge gap exists in technologically relevant surfaces like the graphene-water interface. Here we use machine learning-based simulations with first-principles accuracy to unravel the behavior of the hydroxide and hydronium ions at the graphene-water interface. Our findings reveal that the graphene-water interface is acidic, with the hydronium ion predominantly residing in the first contact layer of water. In contrast, the hydroxide ion exhibits a bimodal distribution, found both near the surface and towards the interior layers. Analysis of the underlying electronic structure reveals strong polarization effects, resulting in counterintuitive charge rearrangement. Proton propensity to the graphene-water interface challenges the interpretation of surface experiments and is expected to have far-reaching consequences for ion conductivity, interfacial reactivity, and proton-mediated processes.

Lila Cadi Tazi

Department of Chemistry

QUACE : SYMMETRIZED MOLECULAR DESCRIPTORS ON A QUANTUM CIRCUIT

There is an evident need to develop fast and accurate methods for materials and chemical simulations, as ab initio quantum chemistry methods are not scalable. On the one hand, the machine learning (ML) literature offers an appealing approach in which ML force fields trained on a small number of ab initio computations can accurately predict properties of new structures with a reduced computational cost. On the other hand, quantum computing algorithms are being developed with the potential to reduce the scaling of classical computing methods for theoretical chemistry. In this work, we bring together quantum computing and the MACE ML framework to introduce the QUACE hybrid algorithm.

In the classical MACE architecture, the central operation is a tensor contraction that requires the manipulation of highly dimensional data and is the bottleneck step of the method. This tensor operation is well suited to be performed on a quantum circuit.

We implement a quantum algorithm that performs tensor contraction, thus reducing the load of classical processing and enabling improved scaling of the overall algorithm with system size. Although present NISQ quantum devices are not powerful enough to outperform classical performances, this work aims to demonstrate the potential of quantum computing for tensor contractions and its application to molecular simulations. QUACE could be a practical method for running molecular dynamics on near-term noisy quantum devices, where quantum noise would be harnessed as a source of stochasticity in the dynamics.

Leo Tan

Department of Physics

SUPERATOMIC MAGNETISM IN X@C60

Lattices of C60 have been shown to behave similarly to lattices of single atoms, where each C60 unit behaves collectively as a superatom [1]. Recently, triangular C60 monolayers have also been experimentally realised [2], providing confidence in the stability of similar structures with entrapped endohedral atoms. In this work, we investigate C60 lattices with spin-3/2 endohedral atoms (X@C60), aiming to study a class of similar systems with tunable magnetic properties. This is done via Density Functional Theory (DFT) calculations, using the Perdew–Burke–Ernzerhof functional corrected for solids (PBEsol0). Using the wavefunction obtained via DFT, a tight-binding model is built and used to infer the magnetic coupling constants. Preliminary results suggest that these structures closely realise a nearest-neighbour Heisenberg model. This opens the door to as further studies of complex magnetic order in similar structures, as well as applications in quantum simulation and scalable qubit systems.

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SUPRAMOLECULES AND SUPERCOMPUTERS

In the Nitschke group, we develop molecular capsules known as metal-organic cages (MOCs). These materials are created through subcomponent self-assembly, where both organic and inorganic building blocks are combined to form polyhedron-like frameworks. By tuning these building blocks, we control the shape, size and chemical environment of the cavity and influence its capacity to interact with ‘guest’ molecules. We aim to prepare MOCs with new topologies, particularly those with larger cavities capable of interacting with biomolecules (artificial virus capsids). To achieve this, we are employing automated computational methods at various levels of theory to virtually screen for the most favourable MOC-geometry for different building blocks. Here, we introduce an accessible (no-code) method for the in-silico construction of MOCs based on the chosen polyhedron-name and building blocks. This approach places the building blocks on topology graphs, which are sourced from the internet. Ongoing collaborative efforts are aimed at further automating the geometry optimization process for large sets of structures. One strategy involves the development of a machine learning interatomic potential (MLIP) tailored for MOCs using the mace framework. Ultimately, these methods aim to reduce cost and effort typically required in traditional trial-and-error approaches, allowing for the evaluation of MOC structures before going to the lab.