CCP9 Conference
&
Community Meeting

Mercure Chester Abbots Well Hotel
Chester
19-22 March 2024

Programme & Abstracts

CCP9 Community Meeting and Conference 19-22 March 2024, Chester

	Tuesday 19	Wednesday 20
09:00		Peter Elliott (STFC)
09:15		
09:30		Urmimala Dey
09:45		Paul Dirk
10:00		Kapildeb Dolui
10:15		Manuel dos Santos Dias
10:30		Break
10:45		
11:00		Livia Bartok-Partay (Warwick)
11:15		
11:30		Pico talks
11:45		
12:00	Arrival and registration	
12:15		
12:30	Lunch	Lunch
12:45		
13:00		
13:15		
13:30		
13:45		
14:00	Welcome from CCP9 Chairperson	Theodoros Papadopoulos (Chester)
14:15	Carla Molteni (KCL)	
14:30		Neil Drummond
14:45		Juliana Morbec
15:00	Arihant Bhandari	Matthew Foulkes
15:15	David Bowler	Abyay Ghosh
15:30	Break	Break
15:45		
16:00	Dermot Green (QUB)	Stephen Dugdale (Bristol)
16:15		
16:30	Christopher Coveney	James Hague
16:45	Dan Criveanu	Kamil Iwanowski
17:00	Chris Pickard (Cambridge)	Clio Johnson
17:15		Venkat Kapil
17:30		Poster session
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18:00		
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19:00		
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	Thursday 21	Friday 22
09:00	Johannes Lischner (IC)	
09:15		Dylan Storan (IOP)
09:30	Sun-Woo Kim	ONETEP - Samuel Magorrian
09:45	James Middleton	CASTEP - TBC
10:00	Bo Peng	QUESTAAL - Jerome Jackson
10:15	Visagan Ravindran	CRYSTAL - Barry G. Searle
10:30	Break	
10:45		
11:00	Marina Filip (Oxford)	CCP9 Chairperson's Report
11:15		CCP9 Community meeting
11:30	Christoph Schran	
11:45	Pavel Stishenko	
12:00	Shirui Wang	
12:15	Nick Hine	
12:30	Lunch	
12:45		
13:00		
13:15		
13:30		
13:45		
14:00	Myrta Grüning (QUB)	(meeting ends)
14:15		
14:30	Christopher Woodgate	
14:45	Xue Yong	
15:00	Erin Johnson	
15:15	Dylan Storan (IOP)	
15:30	Break	
15:45		
16:00	Andrew Turner (EPCC)	
16:15		
16:30	Stephen Longshaw (STFC, CoSeC)	
16:45		
17:00	Round table discussion	
17:15		
17:30		
17:45		
18:00		
18:15		
18:30		key:
18:45		**INVITED AND PLENARY TALKS**
19:00		**CONTRIBUTED TALKS**
19:15		**COMMUNITY CODE UPDATES**
19:30	Conference dinner	**CCP9/COMMUNITY NETWORKING**

Decoding the complexity of matter through electronic structure calculations

Carla Molteni

King's College London

Atoms can assemble and disassemble in a huge variety of ways to create complex materials and biomolecules, guided by the laws of quantum mechanics. In principle, we can write down the mathematical equations that precisely describe atoms' behaviour, but in practice we cannot solve them except in simple cases. However, the introduction of density functional theory 60 year ago and the many subsequent related advancements have provided practical tools for accurately calculating the electronic structures of increasingly complex systems, aided by the availability of powerful computers and the development of sophisticated algorithms. I will present a personal perspective of the success and challenges of electronic structure calculations to unravel the behaviour of matter, drawing from my own experience and research on nanomaterials and bio-systems. I will argue that electronic structure methods need to be part of an extended computational toolbox including complementary techniques, to harness their strengths and address their limitations.

Ab initio electrochemistry via grand canonical DFT

A. Bhandari, C. Peng, J. Dziedzic, L. Anton, J. R. Owen, D. Kramer,
C.-K. Skylaris

University of Southampton

Electrified electrochemical interfaces occur frequently in devices for energy conversion and storage. Transfer of charge carriers across electrochemical interfaces is a key process which can be controlled by an electric potential difference. A description of such interfaces in terms of the electronic structure and electrolyte ions requires a formulation in the grand canonical ensemble. To describe this phenomenon at the atomic scale, we have developed a method for performing density functional theory (DFT) calculations under potential control [J. Chem. Phys., 2021, 155, 024114]. The charge developed on the electrode is neutralized by a build-up of counter charge in the electrolyte, which is represented via Poisson-Boltzmann theory in a grand canonical ensemble [J. Chem. Phys., 2020, 153, 124101]. This model has been implemented in the ONETEP linear-scaling DFT program [J. Chem. Phys., 2020, 152, 174111] and works in both fully periodic as well as open boundary conditions [J. Phys. Chem. C, 2020, 124, 7860]. The model parameters are calibrated vs. standard electrochemical reference electrodes and electrolytes. The model has been applied to study differential capacitance of graphene-based electrodes in supercapacitor applications, and lithium nucleation on graphite anode in Li-ion batteries [J. Mater. Chem. A, 2022, 10, 11426]. The predictions from the model agree well with experiments.

Large scale and linear scaling DFT modelling of polarisation textures in perovskites

David Bowler

University College London

I will discuss the recent application of CONQUEST[1] to complex ferroelectric systems with up to 5,000 atoms[2,3]. We studied the local polarisation textures[2] of $PbTiO_3$ thin films on $SrTiO_3$, finding that the film thickness is key: in relatively thick films (more than 6 formula units thick) polar vortices can be clearly seen to form, while thinner films cannot support these, instead showing a polar wave with chiral bubbles forming at the surface. We have extended these studies using linear scaling to investigate the interaction of domain walls with surface trenches[3]. I will end with a perspective on the opportunities available for large scale DFT simulations of dielectric materials.

References: [1] A.Nakata et al., J. Chem. Phys. 2020 **152**, 164112. [2] J. S. Baker and D. R. Bowler, Adv. Theory Simul. 2020 **3**, 2000154. [3] J. S. Baker and D. R. Bowler, Phys. Rev. Lett. 2021 **127**, 247601.

Many-body theory of positron-molecule interactions

Dermot G. Green*

Queen's University Belfast

Low-energy positron interactions with atoms and molecules are characterized by strong correlations including polarization of the molecular electron cloud, screening of the electron-positron Coulomb interaction, and virtual-Ps formation see e.g., [1]. They substantially modify scattering, enhance annihilation rates by orders of magnitudes, and can be essential for positron binding. They also make the theoretical description of the positron-atom/molecule system a challenging many-body problem.

I will review our recent progress in developing the diagrammatic many-body theory of positron-molecule interactions and its computational implementation in our Gaussian-basis code EXCITON+ [1] (which is adapted from EXCITON, see e.g., [2]). Accounting for the dominant correlations, we have used it to calculate binding energies for a range of polar and non-polar molecules achieving unprecedented agreement with experiment [1,3,4], but also making predictions (e.g. of positron binding to DNA nucleobases [1], the effect of halogenation in hydrocarbons [3], and the competing role of dipoles and pi bonds in heterocyclic molecules [4]. Delineating the effects of the correlations, we showed, in particular, that virtual-positronium formation significantly enhances binding in polar molecules, and moreover, that it can be essential to support binding in non-polar molecules. We have also extended the method to the calculation of positron scattering [5] and to describe positronic-bonded molecules [6].

Beyond providing fundamental understanding required to supporting ongoing experiments and advancing antimatter technologies (traps, accumulators, beams and PET), our results provides benchmarks for the development of general methods to tackle the quantum many-body problem, for which the positron-molecule problem provides an (somewhat unforgiving!) testbed

* In collaboration with C. H. Patterson (Trinity College Dublin), and my QUB group members J. Hofierka, B. Cunningham, C. M. Rawlins, A. R. Swann, J. P. Cassidy and S. K. Gregg. This work is supported by the ERC, StG 804383 'ANTI-ATOM'.

References: [1] J. Hofierka,...,DGG, Nature 606, 688 (2022). [2] C. H. Patterson, Phys. Rev. Mater. 3, 043804 (2019). [3] J. Cassidy, ..., DGG, arXiv:2303.05359, Phys. Rev. A Letter (in press, 2024). [4] E. Arthur-Baidoo, ... , DGG, arXiv:2312.02779 (under review). [5] C. M. Rawlins, ..., DGG, Phys. Rev. Lett. 130, 263001 (2023). [6] J. Cassidy,..., DGG, J. Chem. Phys. 160, 084304, Emerg. Inv. Spec. Collec. (2024).

Rearrangement collision theory of phonon-driven exciton dissociation

Christopher J. N. Coveney

University of Oxford

Understanding the processes governing exciton dissociation is of central importance for optoelectronic devices such as solar cells. Recently, we have introduced a new ab initio approach to compute the phonon-driven exciton dissociation rate in optoelectronic materials [1, 2]. In this talk, I will present a rigorous theoretical framework that unifies the approaches of many-body perturbation theory (MBPT) and Fermi's Golden Rule (FGR), allowing us to derive expressions for phonon-driven exciton scattering rates. I will show how the formalism of rearrangement collision theory [3] can be applied to extend the usual FGR approach to describe phonon-driven exciton dissociation, as well as the FGR exciton to exciton scattering and uncorrelated charge carrier scattering rates [4, 5]. I will present applications of these expressions based on the Wannier-Mott exciton and Fröhlich electron-phonon coupling approximations [6] to demonstrate the importance of accounting for exciton dissociation as well as exciton-exciton scattering in polar materials.

References: [1] Coveney, Haber, Alvertis, Neaton & Filip, In Review (2023). [2] Alvertis, Haber, Li, Coveney, Louie, Filip, Neaton, Submitted (2023). [3] Sunakawa, Progr. Theo. Phys. **24**, (1960). [4] Antonius & Louie, PRB **105**, 085111 (2022). [5] Marini, PRL **101**, 106405 (2008). [6] Filip, Haber & Neaton, PRL **127**, 067401 (2021).

We acknowledge funding from the UK-EPSRC and computation resources from TACC (Frontera) at UT Austin.

Computational treatment of lanthanide dopants in oxides by DFT with self-consistent Hubbard corrections

Dan Criveanu and Katherine Inzani

University of Nottingham

Lanthanide-doped, wide band gap oxides are technologically important materials that have applications in lighting, lasing, scintillation and more recently have shown promise for use in quantum technologies. These applications are possible due to the localised f-electron density of the lanthanide dopant and the wide band gap of the host, resulting in well-defined optical transitions. To predict if a lanthanide-doped oxide is a good candidate for optical-based applications, a good description of its quantum chemical properties is required. To this end, density functional theory (DFT) is a computationally efficient choice for computing the quantum chemical properties of material systems. However, DFT suffers from self-interaction error which leads to erroneous delocalisation of electron density and incorrect predictions of energy gaps, especially for systems that contain lanthanide elements and transition metals. One way to address this is with additive corrections to the DFT energy such as DFT+U, in which a Hubbard U parameter is introduced to enforce localisation of a given subshell and improve energy gaps.

DFT+U has commonly been used to correct the self-interaction error in transition metal compounds, but has rarely been applied to lanthanide-containing materials. More recently there have been DFT+U methods adapted to target lanthanide bulk systems. These methods should also be applicable to lanthanide-doped systems, which we verify using a test system – cerium-doped yttrium aluminium garnet (YAG:Ce). Here we show that the self-consistent application of simplified, rotationally-invariant DFT+U using the linear response approach is able to qualitatively capture the structure of the electronic ground state of YAG:Ce. The electronic structure description is further improved with methods that attempt to address the metastability issues that arise from the use of DFT+U, namely occupation matrix control and U-ramping. We see these results as a promising start for the self-consistent application of DFT+U to lanthanide-doped systems.

A computational hunt for conventional high temperature superconductivity

Chris J. Pickard

University of Cambridge

First principles methods for the prediction of the structure of materials have delivered a powerful tool for generating candidate structures for comparison with experimental analytical methods. Early studies focused on the exotic properties and structures of relatively simple systems, typically the elements and binary compounds. The promise of discovering materials with extreme properties relies on the ability of screen a wide variety of compounds.[1] I will reflect on why ab initio random structure searching (AIRSS) is particularly suited to these challenges, focussing on the dramatic acceleration that ephemeral data derived potentials (EDDPs) afford,[2] and their role in the uncovering of Mg_2IrH_6 as a feasible ambient pressure high temperature superconductor.[3]

References: [1] Shipley, Alice M., Michael J. Hutcheon, Richard J. Needs, and Chris J. Pickard, Phys. Rev. B **104**, 054501 (2021). [2] Chris J. Pickard, Phys. Rev. B **106**, 014102 (2022). [3] Dolui, Kapildeb, Lewis J. Conway, Christoph Heil, Timothy A. Strobel, Rohit Prasankumar, and Chris J. Pickard. arXiv preprint arXiv:2310.07562 (2023)

Simulating real-time dynamics with TDDFT

Peter Elliott

STFC

In recent years, we have applied real-time time-dependent density functional theory (TDDFT) to study ultrafast charge and spin dynamics, including ultrafast demagnetization and optically induced spin transfer (OISTR). In this work, we:

a) show how coupling to the nuclear degree of freedom leads to femto-phono-magnetism, a powerful route to control magnetic order at ultrafast times. We identify strong non-adiabatic spin-phonon coupled modes that dominate early time spin dynamics.

b) study the spin Hall effect which plays a crucial role in spintronics, as both a generator (and detector) of spin current, which may be subsequently be used in spin-orbit-torque devices. We generalize the spin Hall angle to laser pulses of finite frequencies in the linear response regime and predict a giant optical spin Hall effect. Namely, for certain transition metal elements, at particular frequencies, the spin current can be a significant fraction of the charge current, and even exceed it for XUV frequencies.

c) show that a hybrid laser pulses combining linear (IR) and circularly polarized (visible) light allow for the excitation of free carriers of chosen spin at arbitrary k-vector with the Brillouin zone of WSe_2.

Prediction of Room Temperature Electric Field Reversal of Magnetization in the Family of $A_4B_3O_9$ Layered Oxides

Urmimala Dey and Nicholas C. Bristowe

Durham University

Magnetoelectric (ME) multiferroic (MF) materials with intrinsic cross-coupling between electrical and magnetic order parameters are promising for next generation memory devices where external electric fields can switch the direction of magnetization leading to enhanced speed and reduced power consumption. Despite extensive research in the past few decades, only a handful of bulk ME-MF materials that allow for reversal of magnetization with an external electric field have been discovered. Furthermore, all the observed bulk systems have below room-temperature (RT) magnetism which hinders their practical applications. Using first-principles calculations guided by group-theoretical analysis, we predict a hitherto unknown polar phase of bulk $A_4B_3O_9$ layered oxides where applied electric field can switch the magnetization between 180° symmetry equivalent states. Our phonon calculations reveal that the high-symmetry paraelectric phase of these layered oxides is unstable against a zone-centre polar distortion. Furthermore, a weak ferromagnetic (wFM) mode arises spontaneously in the resulting polar phase via a canting of the ground state antiferromagnetic spin ordering of the magnetic B-site cations. The polar mode couples to the wFM mode and the AFM order in an ‘improper’ manner giving rise to a non-linear ME effect where magnetization can be reversed by an electric field via the reversal of the polar mode. Previous experimental studies on these layered oxides demonstrated long-range magnetic ordering of the spins above RT, indicating the possibility of RT electric field switching of magnetization in $A_4B_3O_9$ layered oxides which is further supported by our calculations of the magnetic exchange interaction parameters.

Band gap tuning in Brownmillerites for applications as photoferroic materials

Paul Dirk, Nicholas C Bristowe, Emma E McCabe

Durham University

Photoferroics are photoactive ferroelectric materials that exhibit a bulk photovoltaic effect enabled by the lack of inversion symmetry and are, in contrast to conventional semiconductor photovoltaic devices, capable of achieving above band gap photovoltages. Some ferroelectric oxide perovskites have been studied for this application, but their often large band gaps make applicability for new solar cell technologies challenging.

Brownmillerites are anion-deficient perovskite-related structures with ordered vacancies that form alternating layers of corner-linked BO_6 octahedra and layers of BO_4 tetrahedra. The existence of three distinct cation coordination sites allows for flexibility in accommodating a wide range of p-block and transition metal cations, which makes them suitable for designing materials with a range of properties. Cooperative rotations of the BO_4 tetrahedra can break the inversion symmetry of the ideal high-symmetry structure (of Imma symmetry) making them interesting candidates to design new photoferroics.

A first-principles investigation into some of the effects that composition could have on the electronic structure of Brownmillerites revealed some candidates with potentially suitable band gaps for photoferroic applications. Our experimental studies support the findings from our computational work regarding crystal structures and optical band gaps. Some of the materials investigated show properties that could make them potential candidates for designing new solar energy harvesting devices.

Feasible route to high-temperature ambient-pressure hydride superconductivity

Kapildeb Dolui

University of Cambridge

A key challenge in materials discovery is to find high-temperature superconductors. Hydrogen and hydride materials have long been considered promising materials displaying conventional phonon-mediated superconductivity. However, the high pressures required to stabilize these materials have restricted their application. Here, we present results from high-throughput computation, considering a wide range of high-symmetry ternary hydrides from across the periodic table at ambient pressure. This large composition space is then reduced by considering thermodynamic, dynamic, and magnetic stability, before direct estimations of the superconducting critical temperature. This approach has revealed a metastable ambient-pressure hydride superconductor, Mg_2IrH_6, with a predicted critical temperature of 160 K, comparable to the highest temperature superconducting cuprates. We propose a synthesis route via a structurally related insulator, Mg_2IrH_7, which is thermodynamically stable above 15 GPa and discuss the potential challenges in doing so.

Topological magnons driven by the Dzyaloshinskii-Moriya interaction in the centrosymmetric ferromagnet Mn_5Ge_3

Manuel dos Santos Dias

STFC Daresbury Laboratory

The phase of the quantum-mechanical wave function can encode a topological structure with wide-ranging physical consequences, such as anomalous transport effects and the existence of edge states robust against perturbations. While this has been exhaustively demonstrated for electrons, properties associated with the elementary quasiparticles in magnetic materials are still underexplored. Here, we show theoretically and via inelastic neutron scattering experiments that the bulk ferromagnet Mn_5Ge_3 hosts gapped topological Dirac magnons [1]. Although inversion symmetry prohibits a net Dzyaloshinskii-Moriya interaction in the unit cell, it is locally allowed and is responsible for the gap opening in the magnon spectrum. This gap is predicted and experimentally verified to close by rotating the magnetization away from the c-axis with an applied magnetic field. Hence, Mn_5Ge_3 realizes a gapped Dirac magnon material in three dimensions. Its tunability by chemical doping or by thin film nanostructuring defines an exciting new platform to explore and design topological magnons. More generally, our experimental route to verify and control the topological character of the magnons is applicable to bulk centrosymmetric hexagonal materials, which calls for systematic investigation.

References: [1] Nat Commun **14**, 7321 (2023).

Nested sampling: Exploring the configuration space of carbon with machine learned interatomic potentials

George Marchant, Miguel Caro, Bora Karasulu, Livia Bartók-Pártay

University of Warwick

Nested sampling facilitates unbiased top-down exploration of atomic configuration space, automatically generating all relevant atomic configurations without hindrance from high barriers. One of its most appealing advantages is the ease of calculating the global partition function, giving access to thermodynamic properties such as heat capacity and compressibility.

The advantages of the nested sampling method have been demonstrated in sampling the potential energy landscape of various model systems, enabling the calculation of their complete pressure-temperature phase diagrams. These calculations have frequently uncovered previously unknown thermodynamically stable solid phases, highlighting that the macroscopic properties of interatomic potential models can be very different from what is expected or intended.

Recently, we have showcased how nested sampling can assess the reliability of a machine-learned interatomic potential for carbon, GAP-20. This application included the evaluation of its behaviour at a wide pressure range, and identified an erroneous high-density graphite phase. Our identification of necessary configurations has extended the model's training set, thereby enhancing its performance. We conducted a performance comparison between the GAP-20 model, several empirical models, and an ACE potential.

Nanoscale surface and interface materials modelling for emerging photovoltaic technologies

Theodoros Papadopoulos

University of Chester

Organic photovoltaic technologies require organic semiconductors to have a long enough exciton diffusion length in order to reach the n-type/p-type semiconductor interface; excitons would then separate into electrons and holes, and would be collected at the device electrodes, i.e. the cathode and anode respectively. The metal electrode needs to have a work function that is sufficiently low to facilitate efficient collection of electrons at the cathode, while the transparent metal-oxide electrode needs to have a valence band maximum that is sufficiently high for efficient collection of holes to the anode.

In this talk, I will firstly focus on how to predict electronic structure properties of materials surfaces and interfaces, that are crucial for improving the efficiency of photovoltaic devices, such as surface work-function modification and materials interface energy level alignment. Via density functional theory (DFT) calculations, I will show how to modify the surface properties of cathode electrodes in terms of charge collection across the interface as well as work function modification. The results to be shown are in very good agreement with ultra-violet photoemission spectroscopy (UPS) measurements [1-4].

Secondly, a combination of DFT and experimental measurements via ultraviolet and X-ray photoelectron spectroscopies (UPS and XPS) is used to explore the nature of the interface between anodes and organic hole-transport layers. Attention is paid to the appearance of gap states, and energy barriers between the anode's valence band maximum and the organic semiconductor's highest occupied molecular orbital, which relates to a facile hole collection process, vital for the fabrication of efficient photovoltaic devices [5, 6].

References: [1] Y. Zhou et al, Science 336, **327** (2012). [2] T. A. Papadopoulos et al., Isr. J. Chem. **54**, 779 (2014). [3] M.-K. Lin et al., Phys. Rev. B **95**, 085425 (2017). [4] K. Arumugam et al., Sci. Rep. **11**, 12645 (2021). [5] T. A. Papadopoulos et al., Adv. Funct. Mater. **23**, 6091 (2013). [6] E. Polydorou et al., J. Mater. Chem. A **4**, 11844 (2016).

Adhesion of Graphene to Hexagonal Boron Nitride

Marcin Szyniszewski, Elaheh Mostaani, Angelika Knothe, Vladimir Enaldiev, Andrea Ferrari, Vladimir Fal'ko and Neil Drummond

Lancaster University

We investigate interlayer binding and search for metastable structures in van der Waals heterobilayers of two-dimensional materials with near-aligned hexagonal lattice vectors but incommensurate lattice constants, presenting numerical data for bilayers of hexagonal boron nitride (hBN) and graphene. Diffusion quantum Monte Carlo methods, which provide an accurate treatment of van der Waals interactions, are used to parametrise an adhesion potential as a function of local lattice offset between monolayers of hBN and graphene. Using a continuum model of the elastic energy and the adhesion potential, we find a unique solution for the displacement field at each small misalignment angle. Our diffusion quantum Monte Carlo interlayer binding energies provide benchmark data for the development of dispersion-corrected exchange-correlation functionals, and we have used them to parameterise carbon-nitrogen and carbon-boron interatomic pair potentials for use in atomistic modelling of van der Waals heterostructures.

Interaction between pentacene molecules and monolayer transition metal dichalcogenides

Edward Black and Juliana Morbec

Keele University

Combining two-dimensional (2D) materials with organic materials can be very attractive for applications that require flexibility and where size and weight are important parameters to be considered, such as in wearable, portable and mobile applications. Organic materials usually exhibit excellent optical absorption efficiency and photo- and temperature-induced conformational changes, while 2D materials often show relatively high carrier mobility, superior mechanical flexibility, and tunable electronic and optical properties. Combining both systems can stabilize the organic materials and lead to heterostructures with both high carrier mobility and high optical absorption efficiency, which is promising for solar energy conversion. In this work we investigate, by means of density-functional-theory calculations, heterostructures composed of organic molecules (for example, pentacene and azulene) and transition metal dichalcogenides (TMD) for application in photovoltaic devices. We examine the interaction between the molecules and monolayer TMDs as well as the band alignment of the heterostructures, considering effects of the molecular coverage, rotation and dielectric screening. [1]

References: [1] E. Black, P. Kratzer and J. M. Morbec, Phys. Chem. Chem. Phys. **25**, 29444 (2023), arxiv.org/abs/2304.08619.

Approximating Many-Electron Wave Functions using Neural Networks

W.M.C. Foulkes, Gino Cassella, Wan Tong Lou, Andres Perez Fadon,
Halvard Sutterud, David Pfau, James Spencer

Imperial College London

The use of neural networks for approximating many-electron wave functions is gaining popularity. Neural wave functions are universal approximators in principle and remarkably flexible and accurate approximators in practice. Furthermore, by making use of the variational formulation of the Schrödinger equation, they can be optimized without the use of externally generated data. Their use gives results of consistent quality across highly diverse systems, including molecules, nuclei, and periodic solids. In some cases, the variational optimization is capable of discovering quantum phase transitions unaided.

Spin and orbital degrees of freedom in FeSe: ab-initio perspective

Abyay Ghosh, Piotr Chudzinski, Myrta Gruening

Queen's University Belfast

FeSe is the structurally simplest quasi-two-dimensional iron chalcogenide superconductor. In spite of the structural simplicity, its phase diagram presents exotic phases (superconductivity, spin density wave, nematicity) which are the object of intense study by the condensed matter community. At fundamental level, the description of the orbital and spin degrees of freedom and of their interaction is key to understand the properties of FeSe in its different phases.

We present the calculations of structural, electronic, and magnetic properties of FeSe in the tetragonal phase within density-functional theory at the generalised gradient approximation level. First, we studied how the bandwidth of the d-bands at the Fermi energy are changing by adding simple corrections: Hubbard U, Hund's J and by introducing long-range magnetic orders. We found that introducing either a striped or a staggered dimer antiferromagnetic order brings the bandwidths—which are starkly overestimated at the generalized gradient approximation level—closer to those experimentally observed. Second, for the ferromagnetic, the striped, checkerboard and the staggered dimer antiferromagnetic order, we investigate the change in magnetic formation energy with local magnetic moment of Fe at a pressure up to 6 GPa. The bilinear and biquadratic exchange energies are derived from the Heisenberg model and noncollinear first principles calculations, respectively. We found a non-trivial behaviour of the spin-exchange parameters on the magnetization and we put forward a field-theory model that rationalizes these results in terms of strongly coupled, two-dimensional spin-orbital fluctuations. The implications for the experimentally observed phase diagram are then discussed.

Exploring the impact of correlations on the electronic structure and Fermi surfaces of metals

Stephen Dugdale

University of Bristol

For several decades now it has been possible to measure the Fermi surface of metals via the electron momentum distribution by Compton scattering and positron annihilation [1]. The momentum density measurements resolve real momentum, but the dependence on crystal momentum can be straightforwardly inferred, meaning that the occupation number n(k) can be obtained. For much of this time, the predictions about Fermi surface topologies from density-functional theory (DFT) calculations using the LDA or GGA were sufficient to please both experimentalists and theoreticians regarding their level of agreement with experiment. More recently, we've been able to start to look more carefully at the discrepancies.

In this talk I will showcase the power of the experimental technique for resolving n(k) through a Compton scattering experiment on a so-called high-entropy alloy, NiFeCoCr, where DFT is able to reproduce the experimental Fermi surface, together with its extraordinary disorder-induced smearing, to a remarkable degree.

I will then move on to look at at some materials where we have been able to move the theoretical description beyond DFT, separately looking at the impact of electron correlation within dynamical mean-field theory (DMFT) [3—5] and the GW approximation [6]. I will highlight the impact that this has on the level of agreement with the experimental electronic structure at the Fermi energy.

References: [1] S.B. Dugdale, Fermi surface measurements, Editor(s): Tapash Chakraborty, Encyclopedia of Condensed Matter Physics (Second Edition) **1**, 815, (Academic Press, 2024). [2] H.C. Robarts et al., Phys. Rev. Lett. **124**, 046402 (2020). [3] A.D.N. James, E.I. Harris-Lee, A. Hampel, M. Aichhorn, and S.B. Dugdale, Phys. Rev. B **103**, 035105 (2021). [4] A.D.N. James, M.Sekania, S.B. Dugdale, and L. Chioncel, Phys. Rev. B **103**, 115144 (2021). [5] W. Chen, A.D.N. James, and S.B. Dugdale, Electronic Structure **4**, 045002 (2022). [6] A.D.N. James, E.I. Harris-Lee, and S.B. Dugdale, arXiv:2402.00799 [cond-mat.str-el].

Quantum simulators for unconventional interactions from arrays of optical tweezers

Jim Hague*

The Open University

We calculate Hamiltonian parameters for dressed Rydberg atoms (DRAs) in arrays of optical tweezers and discuss how such arrays can be used to make quantum simulators for electron-phonon interaction and multi band effects in the presence of strong correlation. Arrays of optical tweezers are highly flexible: both depth and shape of individual lattice sites can be controlled dynamically for the quantum simulation of varied problems. Our goals are to understand how (1) Hubbard parameters differ between arrays of optical tweezers and standard optical lattices (2) quantum simulators can be used to emulate quantum materials in the weak- and intermediate-coupling regimes where multiple bands interact, and the basis is not localised and (3) quantum simulators built from arrays of optical tweezers can be used for other key interactions such as electron-phonon coupling.

We determine expressions for the hopping and Hubbard U of arrays of optical tweezers finding that strongly correlated systems with arbitrary structures can be simulated, including those with multiple-site basis and impurities [1]. We consider experimental parameters for quantum simulation of Hubbard models with an arbitrary basis. Finally, we discuss the onset of charge transfer insulating states in ionic Hubbard models. We then describe a quantum simulator for the Hubbard-Holstein model (HHM), comprising two DRA species held in a monolayer by independent arrays of optical tweezers, predicting that boson-mediated preformed pairing and Berezinskii-Kosterlitz-Thouless (BKT) transition temperatures are experimentally accessible [2,3]. We discuss phonon-mediated pairing and condensation.

Finally, as proof of concept, density-functional calculations are made for the bandstructure for a ‘graphene’ quantum simulator made from arrays of optical tweezers [4]. We discuss our ambitions for simulation of quantum simulators using density functional approaches.

* with Calum MacCormick, Pavel Kornilovitch and Leon Petit

References: [1] J.P.Hague, L. Petit and C. MacCormick, Phys. Rev. A. **104**, 053321 (2021). [2] J. P. Hague P. E. Kornilovitch and C. MacCormick, Phys. Rev. A, **102**, 033333 (2020). [3] J. P. Hague and C. MacCormick, Phys. Rev. Lett. **109**, 223001 (2012). [4] J.P. Hague and C. MacCormick, Phys. Rev. A, 95, 033636 (2017).

Entropy in the bond topology controls thermal conductivity of disordered polymorphs

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The structural and thermal properties of disordered solids play a pivotal role in many diverse energy technologies. For example, amorphous alumina is used as a high-dielectric-constant material and its thermal conductivity determines miniaturisation and efficiency in electronics; defective graphite is employed as a moderator in nuclear reactors, where neutron irradiation causes structural changes and material's aging. Although experiments in these materials indicate a strong dependence of thermal conductivity on structural disorder, the fundamental relationship between atomistic structure and macroscopic conductivity remains unclear. Here we address this challenge, using the recently developed Wigner formulation of thermal transport [1] and quantum-accurate machine-learning potentials to shed light on how disorder in the atomic bond network affects thermal conductivity [2]. We introduce a descriptor – topological entropy – which quantitatively captures the variability of local coordination environments, and we show that it correlates with thermal conductivity at a given density [3]. Our research establishes disorder in the atomic bond topology as a fundamental degree of freedom to control and engineer thermal conductivity, calling for studies on how to practically control the local bond topology. Finally, our findings also suggest the possibility to probe atomistic structural properties using thermal-conductivity measurements.

References: [1] M. Simoncelli, N. Marzari, and F. Mauri, Physical Review X **12**, 041011 (2022). [2] A. F. Harper, K. Iwanowski, C. W. Witt, M. C. Payne, and M. Simoncelli, accepted in Phys. Rev. Materials (2024), arXiv:2303.08637. [3] Iwanowski, Csányi, and Simoncelli, Topological entropy controls thermal conductivity in disordered carbon polymorphs. Talk accepted for the 2024 March Meeting of the American Physical Society.

Three-body Backflow Displacement in Quantum Monte Carlo Simulations

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In the field of quantum Monte Carlo simulations (QMC), Variational Monte Carlo (VMC), and Diffusion Monte Carlo (DMC) are often used in tandem. VMC iteratively updates a configuration of electron positions and optimises other free parameters in a trial wavefunction to produce optimised wavefunctions and variational ground state energies. DMC propagates a set of walkers stochastically through imaginary-time, their density is used to recover an approximation to the ground state wavefunction, and produce expectation values. Since their density is positive, walkers are constrained from moving across the nodal surfaces of the wavefunction, leaving the nodes fixed and in approximate positions. This motivates research into techniques for first optimising the nodal surface of a wavefunction using VMC before using DMC to produce results. One well used technique for doing this is backflow, first developed for studying Helium, backflow involves displacing particle positions within a Slater determinant by an amount dependent on the entire configuration of particles. Typically, in VMC, the nodal surface is solely determined by a single Slater determinant, in a Slater-Jastrow wavefunction. Results obtained by Taddei et al. in 2015 suggest that recursively performing two-body backflow displacements can improve ground state energies in QMC. However, this recursive technique proves computationally expensive. This may be mitigated by implementing higher order backflow terms to achieve the same effect. This project is implementing a three-body backflow displacement for use in studying 2D Fermi gases, alongside existing two-body and particle-Ion displacements. In particular, the critical density for Wigner crystallisation will be determined and compared with existing literature.

Machine Learning for full quantum first-principles simulations

Venkat Kapil

University College London

Computational chemistry and material science hinges on the precision and efficiency of first-principles simulations. Ideally, these simulations should incorporate the quantum nature of all electrons and nuclei, achieving predictive accuracy across areas from protein folding, drug design, and catalysis to nanoscale thermodynamics and quantum materials. Traditional "full quantum" simulations are, however, computationally prohibitive. This presentation introduces a modern first principles framework that significantly reduces these costs while maintaining high fidelity. Our method leverages physics-based machine learning to estimate the system's Born-Oppenheimer potential energy surface and other essential electronic quantum effects, such as polarization and polarisability. Demonstrating its efficacy, we predict hitherto unfeasible first-principles phase diagrams of nanoscale systems [1] and relative stabilities of molecular crystal polymorphs [2].

Further, we address the challenge of modelling nuclear motion by mapping quantum dynamics to an effective classical correction akin to effective potentials by Feynman and Hibbs [3]. Our work translates quantum nuclear motion to simple classical molecular dynamics. To showcase our method's capability, we predict vibrational spectra of bulk and interfacial aqueous phases, achieving quantitative agreement with experiments for the first time [4]. Our model offers a path for comprehensive quantum simulations, combining accuracy with the ease of prevalent classical methods.

References: [1] V. Kapil, C. Schran, A. Zen, J. Chen, C. Pickard, and A. Michaelides. Nature 2022, **609**, 7927. [2] V. Kapil, and E. Engel. Proc. Nat. Acad. Sci. 2022, **119**, 6. [3] F. Musil, I. Zaporozhets, F. Noé, C. Clementi, and V. Kapil, J. Chem. Phys. 2022, **157**, 18. [4] V. Kapil, D. Kovács, G. Csányi, and A. Michaelides, Faraday Discuss., 2023.

Modelling the optical properties of large systems

Johannes Lischner

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Modelling the optical properties of large systems, such as plasmonic nanoparticles, quantum defects or twisted bilayer materials, is highly challenging for current ab initio techniques. To address this challenge, we have developed efficient techniques that are based on localized basis functions (such as Wannier functions or atomic orbitals). This allows us to study the generation of ”hot” electrons and holes in metallic nanoparticles with more than one million atoms or the properties of inter- and intralayer excitons of twisted bilayers of transition metal dichalcenides.

Two room-temperature superconductivity claims in 2023: Separating fact from fiction

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University of Cambridge

In 2023, two experimental claims of room-temperature superconductivity shocked the world. The first claimed to show near-ambient superconductivity in lutetium-hydride compounds coinciding with drastic colour changes of the samples from blue to pink to red [1]. We present a full microscopic theory of colour in lutetium hydride, revealing that hydrogen-deficient LuH_2 is the only phase which exhibits colour changes under pressure consistent with experimental reports, with a sequence blue-violet-pink-red-orange [2]. In addition, we find no phonon-mediated superconductivity near room temperature in the claimed pink phase. The second claim concerns "LK99" [3], a copper-doped lead apatite compound which has motivated numerous contradictory theoretical reports. Puzzlingly, previous theoretical works claimed that experimentally reported structures of both the parent and copper-doped lead apatite are dynamically unstable. By pioneering the inclusion of anharmonic phonon-phonon interactions, we show that both compounds are dynamically stable at room temperature, consistent with the experimental reports [4]. We resolve all existing inconsistencies about the dynamical stability of the LK99 compound by clarifying the role of volume and electronic correlation strength. Furthermore, we demonstrate it is not a superconductor but instead a strongly correlated Mott insulator whose phenomenology differs from that of d^9 cuprate superconductors by performing comprehensive DFT+DMFT calculations in Hubbard U-J parameter spaces [5].

References: [1] N. Dasenbrock-Gammon et al., Nature **615**, 244 (2023). [2] S.-W. Kim et al., Nature Communications **14**, 7360 (2023). [3] S. Lee et al., arXiv:2307.12037 (2023). [4] S.-W. Kim et al., npj Computational Materials **10**, 16 (2024). [5] S.-W. Kim et al., in preparation.

Investigating the triboelectric charging properties of the functional groups of common pharmaceutical materials using density functional theory calculations

James R Middleton, Andrew Scott and Mojtaba Ghadiri

University of Leeds

Triboelectrification is a ubiquitous and poorly understood phenomenon in pharmaceutical powder processing. Charged particles can adhere to vessel walls causing sheeting, they can cause agglomeration threatening the stability of powder formulations, and in extreme cases cause electrostatic discharges which present a serious fire and explosion hazard. Triboelectrification is highly sensitive to environmental and material conditions which makes it very difficult to compare experimental results from different publications. In recent years density functional theory (DFT) has been leveraged to better understand the mechanisms of charging at the nanoscale, allowing for precise control of the environmental and material properties of the system being modelled. In this work the charging properties of individual pharmaceutically relevant molecules are investigated by studying the structure and electronic properties at the molecule/substrate interface. Using this Molecule-substrate approach the charging contributions of individual functional groups are explored by examining the Hirschfeld charges, the charge density difference between the molecule and substrate, and the location of the frontier orbitals (HOMO and LUMO) of a given molecules. This information gives us insight into the expected magnitude and polarity of charge transfer and which functional groups play the largest role in triboelectric charging.

Photo-induced electronic and spin topological phase transitions in monolayer single-element ferroelectrics

Bo Peng, Gunnar F. Lange, Daniel Bennett, Kang Wang, Robert-Jan Slager, Bartomeu Monserrat

University of Cambridge

Monolayer group-V materials exhibit rich physics such as single-element ferroelectricity, nontrivial topology, varied spin-orbit coupling, and light-induced structural dynamics. We show with ab initio calculations that light can induce hidden transient phases in both the ferroelectric and paraelectric monolayers. We illustrate the nontrivial topological character of these system by using the recently introduced idea of spin bands and spin-resolved Wilson loops, and show that the topology changes via the closing of the respective band gaps in the presence or absence of photo-induced structural phase transitions. The evolution of topological edge states is also discussed. Our study provides multiple strategies to tailor electronic and spin topology via ultrafast control of photo-excited carrier and structural dynamics.

Local Exchange-Correlation Potentials by Density Inversion in Solids

Visagan Ravindran, Stewart Clark, Nikitas Gidopoulos

Durham University

The vast majority of DFT calculations are performed within the Kohn-Sham (KS) scheme where the exchange-correlation (XC) potential needs to be approximated. Although many approximations have been proposed to date, the quality of the potentials and the electronic ground state densities they yield are difficult to assess, especially since the exact KS potential is unknown. Using the density inversion method of Hollins et al.[1], we invert target densities of various materials obtained by non-local potentials from Hartree-Fock, DFT+U and generalised KS schemes (GKS) to find the local, multiplicative KS potential that adopts these densities as an electronic ground state. We further invert accurate (almost) “exact” densities from fixed-node diffusion Monte Carlo[2,3] (DMC), thereby allowing us to gain insight into the “exact” KS potential in these materials.

Using the resulting local XC potentials from the inversion of these densities, we calculate the band structure of these materials and compare them with the original GKS and DFT+U band structures to assess the quality of these densities and potentials to the reference “exact” KS potential. Furthermore, comparison of the “exact” KS band gap and experimental band gap allows one to gauge the size of exchange-correlation discontinuities and the strength of correlations.

References: [1] T W Hollins, S. J. Clark, K. Refson, N. I. Gidopoulos, J. Phys.: Condens. Matter, **29**, 04LT01 (2017). [2] R. J. Needs, M. D. Towler, N. D. Drummond, P. López Ríos, J. Phys.: Condens. Matter, **22**, 023201 (2010). [3] R. J. Needs, M. D. Towler, N. D. Drummond, P. López Ríos, J. R. Trail, J. Chem. Phys. **152**, 154106 (2020).

Phonon screening of excitons in semiconductors and insulators from first principles computational modeling

Marina R. Filip

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Excitons are composite quasiparticles which consist of correlated photoexcited electron-hole pairs. Information on exciton formation, delocalization and dissociatiation is of key importance to the functionality of semiconductors and determines a wide range of applications, including photovoltaics, lighting and lasing. Development of new computational modeling techniques based on density functional theory (DFT) and many body perturbation theory which are capable to describe interactions between excitons and other quasiparticles constitutes a frontier first principles computational modeling of materials. The GW+Bethe-Salpeter Equation (BSE) approach [1,2] is the state-of-the-art approach to compute optical excitation energies in semiconductors and insulators and provides the foundation of new methods aimed at describing complex excited state phenomena.

In this talk, I will present a new approach we have developed, which allows us to understand the impact of ionic vibrations have on the dielectric screening of excitons from first principles [3,4]. I will first present the main theoretical approach, and show how we can capture the effect of phonon screening by approximating the exciton-phonon self-energy at the level of phonon exchange. This allows us to compute temperature dependent exciton binding energies, and the rate of dissociation of excitons upon scattering with phonons from first principles [4,5]. I will secondly show how this approach can be applied to several well understood semiconductors and insulators and how our results follow very similar trends with data available in the literature.

References: [1] Hybertsen & Louie, Phys. Rev. B **34**, 5390 (1986). [2] Rohlfing & Louie, Phys. Rev. Lett. **81**, 2312 (1998). [3] Filip, Haber & Neaton, Phys. Rev. Lett. **127**, 67401 (2021). [4] Alvertis, Haber, Li, Coveney, Louie, Filip & Neaton, submitted (2023), arXiv:2312.03841i. [5] Coveney, Haber, Alvertis, Neaton & Filip, submitted (2023).

To pair or not to pair? Machine-learned explicitly-correlated electronic structure for NaCl in water

Christoph Schran

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The extent of ion pairing in solution is an important phenomenon to rationalise transport and thermodynamic 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. Leveraging developments in machine learning potentials and electronic structure methods, we obtain wavefunction based models with RPA and MP2 for the prototypical system of Na and Cl ions in water. We show that even among these methods, discrepancies in the PMF still remain, and also highlight shortcomings of density functional theory and classical force-field predictions. Towards the end of this talk, we show how these methods can be applied to understand ion pairing under confinement.

Improvements of an implicit solvent model in FHI-aims DFT code

Pavel Stishenko, Jacob Filser, Gabriel Bramley, Reinhard Maurer, Volker Blum, Harald Oberhofer, Andrew Logsdail

Cardiff University

FHI-aims is a density functional theory (DFT) code utilizing numerical atomic-centered basis functions. It supports simulations of both periodic and non-periodic systems and employs basis set locality to achieve linear scaling with the size of the simulated system. These features make it efficient for simulation of large heterogeneous systems with large empty volume, such as surfaces, zeolites, metal-organic frameworks. Many important processes occurring in these systems also take place in solvents. Therefore, there is a high demand for efficient methods to simulate solvents in FHI-aims. Recently the multipolar expansion (MPE) implicit solvent model has been implemented in FHI-aims. The MPE showed high accuracy compared with experimental data, but its performance was limited by need to solve a large system of linear equations (SLE). We have exploited the locality of multipolar functions to sparsify the SLE matrix thereby increasing the performance of the MPE model. Moreover we have enabled the MPE model to support periodic systems, that is crucial for adsorption and catalysis applications.

Computational insights into emerging chalcogenide perovskite photovoltaics

Shirui Wang

Imperial College London

Chalcogenide perovskites are of interest due to similarities in their electronic structure with the lead halide perovskites. Both have intense optical absorption and electronic structures characteristic of facile charge transport. Furthermore, the chalcogenides are earth-abundant, highly stable, and relatively non-toxic. Hence the toxicity concerns of lead in the lead halide perovskites are eliminated. Here, we systematically assess the phase space of chalcogenide perovskites (including perovskite polytypes) for the $BaZrCh_3$ series where Ch = O, S, Se using hybrid density functional theory. We calculate the maximum theoretical photovoltaic device performance based on optical properties. We also calculate their fundamental band alignment to suggest potential electron and hole contact materials. Our work identifies the most promising chalcogenide perovskites and gives directions to experimentalists working on building next-generation devices. Discovering a cost-effective solar photovoltaic will have enormous industrial and economic impacts and minimise the effects of climate change.

Exploring Twisted 2D materials with Linear-Scaling DFT and Machine Learned Interatomic Potentials

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University of Warwick

The electronic and vibrational properties of 2D materials whose minimal models are large, such as disordered alloys, twisted bilayers and misaligned heterostructures, are challenging for conventional approaches to Density Functional Theory (DFT) due to their unfavourable scaling with system size. I will give an overview of the ONETEP code [1] and the simulation and modelling work enabled by it in recent years on 2D materials, focussing on twisted bilayers, heterostructures and alloys, particularly theoretical spectroscopy including simulated ARPES via spectral function unfolding. I will also briefly demonstrate our work accessing theoretical spectroscopy of even larger systems through construction of Machine-Learned Interatomic Potentials, which has produced MLIP surrogate models for 2D systems whose accuracy closely matches that of DFT, constructed using equivariant neural networks such as MACE [2]. We show predicted Raman spectra of ternary and quaternary alloys of form $\mathrm{Mo}_{1-x}\mathrm{W}_x\mathrm{S}_{2-2y}\mathrm{Se}_{2y}$, and of twisted bilayer TMDs.

References: [1] www.onetep.org and J.C.A. Prentice et al, J. Chem. Phys. **152**, 174111 (2020). [2] I. Batatia et al., Advances in Neural Information Processing Systems **35**, 11423 (2022).

Nonlinear optics from first principles

Myrta Grüning

Queen's University Belfast

In the past decades, many-body approaches based on the GW approximation and the Bethe-Salpeter equation have become state-of-the-art for calculating optical absorption in solids and nanostructures. In this talk, I'll present a real-time approach derived from the non-equilibrium Green's function, that allows extending the GW+BSE approach beyond the linear regime.[1,2] Using this approach, I'll address the importance of many-body effects and in particular of excitonic effects for nonlinear optical properties.[3] For example, I'll look at the case of single-layer monochalcogenide whose strong Second Harmonic Generation cannot be reproduced within the independent-particle approximation.[4] In the second part of the talk, I'll address the computational cost of these approaches and so discuss the possibility of a real-time approach based on time-dependent density-functional theory, that can describe excitonic effects [5] or of alternatives to the real-time approach. [6]

References: [1] Attaccalite, C., Grüning, M. & Marini, A., (2011) Physical Review B **84**, 24, 245110. [2] Attaccalite, C. & Grüning, M., (2013) Physical Review B **8**, 235113. [3] Grüning, M. & Attaccalite, C., (2014) Physical Review B **89**,081102. [4] Claudio Attaccalite, Maurizia Palummo, Elena Cannuccia, Myrta Grüning, (2019) Phys Rev Mat **3**, 074003. [5] Grüning, M., Sangalli, D. & Attaccalite, C., (2016) Physical Review B **94**, 035149. [6] Alliati, I.M, Grüning M. (2023), Electron. Struct. **5** 017001.

Connections Between Magnetism and Preferred Atomic Arrangements in Multicomponent Alloys: Insights From An All-Electron Linear Response Theory

Christopher D. Woodgate, Julie B. Staunton

University of Warwick

Metallic alloys remain a fundamental and fascinating class of materials to study, as systems which are both of fundamental physical interest and also of use in an enormous range of technologically relevant applications. An important question to address when modelling these systems is: can the phase stability of a given system be successfully predicted without reliance on empirical data? In this talk, we will present results from an ab initio, all-electron, Landau-type theory [1,2] suited for modelling the phase behaviour of a wide class of alloy systems, which is based on density functional theory calculations of materials' electronic structure. In particular, we will demonstrate the importance of the correct treatment of magnetism when modelling these systems, showing how both the nature and predicted temperature of atomic ordering are dramatically affected depending on the simulated magnetic state. We will consider example systems including the pathological, 3-component, CrCoNi alloy [3], as well as the technologically relevant binary FeNi system [4], the ordered state of which is under consideration as a candidate rare-earth-free permanent magnet.

References: [1] C. D. Woodgate, J. B. Staunton, Phys. Rev. B **105**, 115124 (2022). [2] C. D. Woodgate, J. B. Staunton, Phys. Rev. Mater. **7**, 013801 (2023). [3] C. D. Woodgate, D. Hedlund, L. H. Lewis, J. B. Staunton, Phys. Rev. Mater. **7**, 053801 (2023). [4] C. D. Woodgate, L. H. Lewis, J. B. Staunton, arXiv:2401.02809 (2024).

First principle studies on single atom catalyst for electrochemical synthesis of ammonia

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Designing efficient single-atom catalysts (SACs) with high selectivity for the electrocatalytic reduction of nitrate to ammonia formation is both crucial and challenging. This challenge arises due to the intricate and competitive electronic interactions among intermediates, metal active centers, and coordination environments. In this work, we present a comprehensive investigation detailing how to enhance the activity and selectivity of the electrocatalytic nitrate reduction reaction (NO_3RR) by transitioning from single-layer SACs to bilayer SACs (BSACs). This enhancement is achieved through axial d–d orbital hybridization, as elucidated by a systematic study of 27 SACs and BSACs utilizing density functional theory (DFT) calculations. Considering potential pathways involving O-terminal, N-terminal, NO-terminal, and NO-dimer configurations, our calculations reveal that among monolayer SAC candidates, Ti-Pc and V-Pc exhibit low limiting potentials (UL) of −0.24 and −0.48 V, respectively. Furthermore, analyses of formation energy, dissolution potential, and ab initio molecular dynamics results demonstrate the robust stability of these catalysts under reaction conditions. In these single-layer transition metal (TM)-Pc complexes, the d-band energy levels and occupation numbers are influenced by d_{xz}/d_{yz} and p_z orbital hybridizations. Notably, the presence of axial d_{z^2} orbitals introduces a novel avenue for fine-tuning d-band characteristics and reactivity through d_{z^2}–d_{z^2} interactions. Building on these insights, the formation of BSACs using Ti-Pc and V-Pc as substrates, facilitated by axial d–d orbital hybridization, offers a distinctive approach to modulating the catalytic performance of NO3RR. Significantly, we establish a two-dimensional volcano correlation encompassing the d-band center ($_d$), $d_{xz} + d_{yz}$ orbital occupation numbers, and UL to describe NO_3RR catalytic efficacy. Optimal BSACs should possess concurrent appropriate $_d$ and $d_{xz} + d_{yz}$ occupation numbers. Remarkably, Ti-Mo and Ti-Ta BSACs emerge as exceptional NO_3RR catalyst candidates, both displaying a remarkably low UL of −0.13 V. The hybridization between d_{z^2}–d_{z^2} orbitals heightens charge transfer and structural stability within double-layer metals. The scarcity of contiguous metal sites introduces a substantial energy barrier hindering NO_2, NO, and N_2 formation, effectively suppressing NO_3RR by-products. In summation, this investigation imparts valuable insights into effectively enhancing nitrate reduction on SACs and BSACs, offering valuable guidance for advancing electrocatalyst development.

London Dispersion in Density-Functional Theory and the Exchange-Hole Dipole Moment Model

Erin R. Johnson

Dalhousie University and University of Cambridge

Inclusion of London dispersion in density-functional calculations is now standard practice in computational chemistry and materials science. In this talk, we review how the dispersion energy can be written as an asymptotic series expansion from perturbation theory, which can be added to the self-consistent energy. We will then focus on the exchange-hole dipole moment (XDM) model, in which the dispersion coefficients are non-empirical and depend directly on the electron density and related properties. XDM offers simultaneous high accuracy for a diverse range of chemical systems, including molecular crystals, layered materials, and metal surfaces. Applications to the chemistry of molecular crystals, and crystal structure prediction, will be highlighted.

Community Meeting Presentations

Electronic Structure: a journal by IOP Publishing that supports the electronic structure community

Dylan Storan

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Change and sustainability in UK national HPC services

Andy Turner

EPCC, The University of Edinburgh

The Computational Science Centre for Research Communities: The Next Steps

Stephen Longshaw

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ONETEP

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University Of Warwick

CASTEP

Keith Refson

Durham University

QUESTAAL

Jerome Jackson

STFC Daresbury Laboratory

CRYSTAL

Barry G. Searle

STFC Daresbury Laboratory

Unravelling the Role of Ruthenium on the Overpotential and Cyclability of Litihum-Oxygen Batteries

Zhaohan Shen[1], Wei Yu[1], Alex Aziz[2], Takeharu Yoshii[1], Eiichi Kobayash[2,3], Hirotomo Nishihara[1]

[1] *Tohoku University, Japan,* [2] *Manchester Metropolitan University,* [3] *Kyushu Synchrotron Light Research Center, Japan*

Lithium-oxygen (Li-O_2) have extremely high theoretical energy densities but suffer from high overpotentials and thus poor cycle stability. The addition of a ruthenium catalyst has long been investigated to reduce the overpotential and increase cyclability, but its impact remains uncertain. This work employed a combination of experimental techniques and density functional theory (DFT) simulations to elucidate the role of Ru catalysts on the overpotential and cyclability of Li-O_2 batteries. Both Ru-hcp and Ru-fcc nanoparticles were synthesised and their catalytic behaviour investigated. DFT was used to gain an insight into the overpotentials of the different phases and comprehensive characterizations, including in situ differential electrochemical mass spectroscopy using isotopic labelling ^{13}C-based cathodes, successfully decoupled the side reactions taking place at the carbon cathode and the electrolyte. Our results indicate that Ru-fcc has greater catalytic activity than Ru-hcp. This mitigates the degradation of the carbon cathode due to the lower overpotentials required but leads to a more pronounced decomposition of the electrolyte compared to Ru-hcp.

Searching for High Thermoelectric Performance Molecular Junctions

Asma Alajmi, Bashayr Alanazi, and Ali Ismael

Lancaster University

If high-thermoelectric-performance molecules could be identified, then they could be used as building blocks for organic thin-film materials able to convert waste heat into electricity. A key parameter governing thermoelectric performance is the Seebeck coefficient S, which determines the voltage generated when a temperature difference is applied to a single-molecule junction. In the literature, ensemble-averaged values of S are typically reported. Here we demonstrate that the search for high-performance molecules can be accelerated by focusing attention on fluctuations in S. We study the electrical conductance and Seebeck coefficient of three endohedral metallofullerenes (EMFs) for $Sc_3C_2@C_{80}$, $Sc_3N@C_{80}$, and $Er_3N@C_{80}$, which based on their structures, are selected to exhibit different degrees of charge inhomogeneity and geometrical disorder within a junction.

Hydrogen defects in perovskite ($CaTiO_3$)

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Perovskite ($CaTiO_3$) is of great technological interest because of its ferroelectric, optoelectrical, and catalytic properties. Hydrogen is ubiquitous in most materials and is likely to be present in perovskite as hydroxyl group defects (-OH). The presence of hydrogen in perovskite is poorly constrained [1], in terms of OH solubility and H incorporation mechanisms, despite the significant impact hydrogen defects can have on the physical and chemical properties of a material [2,3]. Here we present for the first time a comprehensive study combining X-ray diffraction, electron microprobe analysis, Fourier transform infrared (FTIR) and Raman spectroscopy on a suite of natural and high-pressure/temperature (up to 13 GPa/1200°C) synthesised perovskite crystals. FTIR measurements indicate a complex array of sharp and broad OH absorption bands between 3560 and 3300 cm^{-1} with OH contents ranging from 106 to 714 ppm H_2O. To identify the local structures responsible for the observed OH IR bands, ab initio calculations were performed to model theoretical IR spectra of various hydrogen incorporation mechanisms in perovskite. These calculations indicate that each observed IR signature is related to OH defects located at isolated and coupled Ca and Ti vacancies, a Ti vacancy combined with a Ti exchanged with the Ca site, and interstitial OH defects in perovskite.

References: [1] A. Beran, E. Libowitzky, T. Armbruster, Can. Mineral. **34** (1996) 803–809. [2] M. Wöhlecke, L. Kovács, Crit. Rev. Solid State Mater. Sci. **26** (2001) 1–86. [3] S. Chuangchote, K. Roongraung, Mater. Hydrog. Prod. Conversion, Storage, John Wiley & Sons, Ltd, 2023: pp. 539–573.

Atomistic Modelling of Electrified Lithium Anode-Electrolyte Interfaces under Potentiostatic control

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[1] *University of Southampton,* [2] *STFC-UKRI*

Utilising ONETEP's grand canonical DFT implementation [1] in conjunction with the soft-sphere continuum solvation model [2] and the Neutralization by Electrolyte Concentration Shift (NECS) schemes [3], we can introduce bias to systematically investigate the evolution of relative surface energies among the [100], [110], and [111] crystal facets with respect to voltage variations; with a specific focus to possible surface reconstructions under bias.

References: [1] Bhandari, A.; Peng, C.; Dziedzic, J.; Anton, L.; Owen, J. R.; Kramer, D.; Skylaris, C. K., Journal of Chemical Physics 2021, **155**. [2] Fisicaro, G.; Genovese, L.; Andreussi, O.; Mandal, S.; Nair, N. N.; Marzari, N.; Goedecker, Journal of Chemical Theory and Computation 2017, **13**, 3829–3845. [3] Dziedzic, J.; Bhandari, A.; Anton, L.; Peng, C.; Womack, J. C.; Famili, M.; Kramer, D.; Skylaris, C. K., Journal of Physical Chemistry C2020, **124**, 7860–7872.

A General Purpose Framework for QM/MM Embedding

Gabriel Bramley, Pavel Stishenko, Andrew Logsdail

Cardiff University

The QM/MM embedding methodology enables unprecedented accuracy for simulations within a region of interest, while maintaining low computational costs for the wider environment. Such embedding schemes have been implemented in a variety packages, including wrappers and internal implementations for given electronic structure packages. However, general purpose implementations of embedding schemes are limited by the workflows ability to transfer data structures (eg., electrostatic potentials and density matrices) from one SCF task to another. By leveraging the import/export capabilities of the recently developed ASI (Atomic Simulation Interface) API, we demonstrate a generalisable python-based interface for implementing projector-based QM/MM embedding simulations for the FHI-aims software package. We will furthermore show the capability of the embedding framework for other strategies such as DMET (Density Matrix Embedding Theory).

Novel ab-initio Machine Learning Simulation Suite

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University of Lincoln

We introduce NaiMLeSS (Novel ab-initio Machine Learning Simulation Suite): an open-source Python package designed to streamline the training of machine learning potentials (MLPs) using data from ab initio electronic structure calculations and subsequent machine learning molecular dynamics (ML-MD) simulations. NaiMLeSS seamlessly interfaces CP2K[1] for ab initio calculations, NequIP[2] for MLP training and LAMMPS[3] for ML-MD simulations. Built as a python module, NaiMLeSS can be integrated into larger workflows, as well as used as a stand-alone solution, and it is developed with a strong emphasis on modularity, ensuring that community contributions (such as support for additional ab initio, ML or MD tools) can be incorporated without any modifications to the core architecture. The utility and flexibility of NaiMLeSS are demonstrated through a comprehensive workflow example that showcases its application in conducting committee ML-MD simulations on a toy system.

References: [1] Kühne, T.D. et al, J. Chem. Phys. (2020) **152**, 194103. [2] Batzner, S. et al, Nature Communications (2022) **13**, 2453. [3] Thompson A.P. et al, Comp. Phys. Comm. (2022) **271**, 108171.

PyDFT and PyRTP: accurate and accessible DFT/RT-TDDFT calculations for all

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Theoretical and computational methods, in the form of two Python programs entitled ’PyDFT’ and ’PyRTP’, are presented. These methods attempt to emulate the density functional theory and real time time-dependent density functional theory methods implemented in the open source electronic structure and molecular dynamics software package CP2K. The creation of these programs allows for effective teaching of DFT and RT-TDDFT in an accessible programming language for undergraduate and graduate-level users, without sacrificing the accuracy of these calculations.

Understanding Electronic Structure in Ga_2O_3 Alloys: First Principles Investigation of In and Bi Alloying

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Ga_2O_3, a wide-bandgap semiconductor, has undergone extensive investigation for various potential applications, including high-power electronics and solar-blind photodetectors. While unintentional n-type doping is common, achieving p-type doping in Ga_2O_3 remains challenging. This presents a practical obstacle to fabricating p-n based devices despite extensive research in the field. Alloying, for example, with Al or In, represents a promising strategy for tuning optoelectronic properties and reducing mismatches in terms of structure or thermal expansion. Additionally, incorporating Bi or Ni through alloying can offer an alternative route to achieving p-type doping.

In this contribution, we investigate the electronic structure of Ga_2O_3 alloy. After the Ga site's occupation preference investigation Special quasi-random structures are employed for disorder modeling. The effective band structure of InGaO alloys shows that as the In content increases, there is a systematic reduction observed in the band gap and electron effective mass. The effect of Ni, as well as Bi alloying Ga_2O_3, is explored to engineer the band gap, pushing up the valence-band maximum. This strategy has recently been proposed to facilitate the achievement of p-type doping.

Simulating Materials at Extreme Conditions with Ephemeral Data Derived Potentials

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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.

First-principles Wigner formulation of coupled radiative and conductive heat transfer

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At ordinary temperatures, heat transfer in dielectric solids is mainly mediated by atomic vibrations, as accurately described by established quantum transport formulations for heat conduction. At extremely high temperatures, experiments in polar dielectrics show a very strong enhancement in their heat-transfer capability, which departs from predictions obtained using state-of-the-art conduction theories. Such behavior has been speculated to originate from the emergence of additional radiative effects, but no theoretical framework has been able to rationalize it from first principles.

Here, we employ the Wigner phase-space formulation of quantum mechanics to derive a set of transport equations for coupled conductive and radiative heat transfer. Their first-principles solutions rationalize experiments in materials with various degree of disorder and compositions, as we show with applications to thermal-barrier-coating crystals and oxide glasses. The approach developed sheds lights on the fundamental physics underlying heat transfer in solids, and paves the way for the control and theory-driven optimization of thermal insulators employed e.g. in industrial furnaces, jet engines, and heat shields.

Beating the Egg-box Effect: Making Plane Wave DFT Calculations More Accurate

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The ”egg-box effect” is a known challenge in Density Functional Theory (DFT) calculations which arises from the discretization of continuous quantities, e.g. the electron density. This effect is observed as an unphysical change in the system’s total energy, reminiscent of an egg-box (see figure), as the system is moved relative to the discrete grid. This can cause unphysical results such as geometry relaxations finding incorrect crystal symmetries or negative phonon modes in vibrational spectra calculations. The egg-box effect can be mitigated by using finer grids to represent all continuous quantities however this greatly increases computational cost.

The origin of the effect in plane-wave DFT is in the evaluation of the Exchange and Correlation (XC) energy. We present a novel technique for estimating the egg-box effect on the total energy, providing a degree of uncertainty quantification. This is done by Fourier interpolating the ground state density to a set of shifted grid positions and re-evaluating the change in XC energy at these grid offsets. This is found to be in near exact agreement with the change in total energy from the egg-box effect. Our results also show that for more advanced XC approximations, such as GGAs and meta-GGAs, the egg-box effect can be order of magnitudes worse than less advanced functionals. Performing this analysis at an early stage of a workflow can inform on the expected accuracy of subsequent calculations.

Further, our results demonstrate that by selectively computing the XC energy and its corresponding potential on a finer grid, via Fourier interpolation, the egg-box effect can be reduced and even eliminated from calculations altogether. Coupled with the uncertainty quantification method, egg-box related inaccuracies can be avoided more conveniently than just increasing all the grids until inaccuracies appear to have been converged out. This work offers a promising pathway towards mitigating the egg-box effect in a diverse range of materials modelling applications.

The Stability and Synthesisability of Quaternary Oxysulfides

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Oxysulfides are a family of mixed-anion materials containing both oxide and sulfide anions. When compared to the widely explored oxide materials, the incorporation of another (hetero)anion provides an ability to tune both structural and electronic properties, [1] leading to a range of candidates for non-linear optics, [2] superconductivity, and photocatalytic applications. [3] However, the discovery of these mixed-anion materials is hindered by the additional synthetic challenges faced (e.g. using more reactive/harmful reagents requires working under an inert atmosphere, and including a less electronegative anion species requires a sealed system to prevent oxidation).

While many computational studies focus on the discovery of stable structures, we also strive to understand whether these compounds can be synthesised. Using a synergistic approach of experiment and density functional theory (DFT), we look to identify stable products and likely reaction pathways for a given set of reagents by calculating reaction enthalpies for both solid state reactions and lower temperature intercalation routes. [4] DFT calculations have also been used to investigate the electronic structure of synthetic targets. We present here work on quaternary oxysulfides, specifically $La_2O_2MS_2$-related materials. [5]

References: [1] H. Kageyama, K. Hayashi, K. Maeda, J. P. Attfield, Z. Hiroi, J. M. Rondinelli and K. R. Poeppelmeier, Nat. Commun., 2018, **9**, 772. [2] Y.-Y. Li, W.-J. Wang, H. Wang, H. Lin and L.-M. Wu, Cryst. Growth Des., 2019, **19**, 4172-4192. [3] K. Maeda, F. Takeiri, G. Kobayashi, S. Matsuishi, H. Ogino, S. Ida, T. Mori, Y. Uchimoto, S. Tanabe, T. Hasegawa, N. Imanaka and H. Kageyama, Bull. Chem. Soc. Jpn., 2021, **95**, 26-37. [4] S. Sasaki, D. Driss, M. T. Caldes, E. Gautron, S. Cadars, E. Janod, B. Corraze, C. Guillot-Deudon, I. Braems, S. Jobic and L. Cario, Solid State Sci., 2024, **147**, 107383. [5] M. Orr, G. R. Hebberd, E. E. McCabe and R. T. Macaluso, ACS Omega, 2022, **7**, 8209-8218.

The effect of dihedral angle on the conductance of asymmetric molecules

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To employ a single molecule as an efficient electronic component, it is essential to understand the relationship between its conformation and charge transport properties. This study examines how the torsion angle between adjacent aromatic rings influences the conductance of symmetric and asymmetric molecules. Taking a chain of four benzene rings as an example, rotating about the middle bond preserves the symmetry, while rotating about the right-hand bond breaks the symmetry. We demonstrate that the switch performance could be enhanced by introducing asymmetry, thus providing a new ingredient to the design of a single-molecule switch.

Chemical interface damping of surface plasmon polaritons

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Au nanostructures exhibit fascinating optical properties enabled by surface plasmon polaritons (SPPs), collective surface oscillations of the conduction electrons. Functionalisation of the Au nanostructure by adsorbing molecules to its surface modifies the properties of the SPP. For example, the SPP propagation distance is reduced. This is known as the chemical interface damping (CID) of the SPP. However, the microscopic mechanism of this process remains ill-understood.

To make progress, we carry out first-principles density-functional theory calculations for a paradigmatic system: an Au 111 surface with adsorbed 4-ATP molecules. We present results for the relaxed atomic structure of the adsorbed molecules and also analyse the electronic properties of this system. We discuss how the resulting insights affect the interpretation of the CID.

Superconductivity in the elements

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First Principles calculations of superconducting properties, notably the critical temperature T_c, have been widely used to search for novel superconductors. These theoretical methods have had successes in the prediction of high T_c hydride superconductors such as H_3S [1], and more recently have suggested Mg_2IrH_6 [2][3] to be an excellent candidate for high-temperature conventional superconductivity. However, one limitation of the commonly used Eliashberg methods is the inclusion of the Coulomb pseudopotential $\mu*$, which acts as a semiempirical parameter. To investigate the limits of a fully-first principles approach, we calculated the T_c for a range of elemental systems using the EPW package [4] in Quantum Espresso for fixed $\mu*$. We find a general tendency to overestimate T_c in elemental systems, with particularly large discrepancies between the theory and the experimental values in the 3d and 4d systems. This suggests that additional mechanisms that have not been fully accounted for, such as paramagnetic spin fluctuations, may be required to fully reproduce the experimental results for elemental transition metals.

References: [1] Drozdov, A. P., Eremets, M. I., Troyan, I. A., Ksenofontov, V. & Shylin, S. I., Nature **525**, 73–76 (2015). [2] Dolui, K.,Conway, L.J., Heil, C., Strobel, T.A., Prasankumar,R., Pickard, C.J., arXiv:2310.07562. [3] Sanna, A., Cerqueira, T.F.T., Fang, YW. et al., npj Comput Mater **10**, 44 (2024). [4] H. Lee, S. Poncé, K. Bushick, S. Hajinazar, J. Lafuente-Bartolome, J. Leveillee, C. Lian, J. Lihm, F. Macheda, H. Mori, H. Paudyal, W. Sio, S. Tiwari, M. Zacharias, X. Zhang, N. Bonini, E. Kioupakis, E.R. Margine, and F. Giustino, npj Comput Mater **9**, 156 (2023).

An ab initio study to enhance electrical steel for transformer and motor applications

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Theoretical simulations have been carried out to predict the electrical and magnetic losses of pure Iron. This is done to establish a protocol for characterizing the effect of doped Iron and using this to model the losses in the operation of a transformer.

The CASTEP[1] code, based on the density functional theory (DFT), is used to simulate the spin-polarized density of states of BCC Iron. The density of states is employed within the BoltzTrap2 program[2] to calculate the electrical conductivity of Fe. The electrical conductivity of pure Fe, calculated in this study, showed good agreement with various experimental and theoretical calculations, once a fitted value of the relaxation time from previous works was included. This demonstrated that the electrical conductivity varies across temperature ranges and that the relaxation time is temperature-dependent in high-temperature ranges.

On the other hand, for investigating the magnetic losses of pure Fe, the exchange interaction constants and the magnetic anisotropy energy are deduced from vector spin-polarized DFT calculations. These values are then used in the VAMPIRE[3] program, which does atomistic simulation of magnetic materials, to calculate the Curie temperature and a magnetic hysteresis loop. The M-H loop of pure iron is too small to calculate within VAMPIRE for reasonable calculation effort. Consequently, the magnetic losses are difficult to quantify with an accurate DFT-based anisotropy energy of 10-26 J/atom. We find that with VAMPIRE, the anisotropy energy of 10-24 J/atom is considered to be an upper bound on the correct answer to produce a reliable hysteresis loop area.

Future work is ongoing to extend this study to include the effects of doping in Iron and how that impacts the material properties. This study may open the way to identify new improvements for further developing electrical steel and pushing it to obtain better performance.

References: [1] M. C. Segall, M. D., Lindan, P. J., Probert, M. A., Pickard, C. J., Hasnip, P. J., Clark, S. J., & Payne, J. Phys. Condens. matter, **14**, no. 11, p. 2717, 2002. [2] M. J. Madsen, G. K., Carrete, J., & Verstraete, Comput. Phys. Commun., **231**, pp. 140–145, 2018. [3] https://vampire.york.ac.uk/.

Unraveling the Oxidation of Realistic Platinum Nanoparticles using Linear Scaling DFT and Modern Forcefields

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Platinum Nanoparticles (Pt NPs) are pivotal catalysts in numerous industrial applications, encompassing fuel cells, catalytic converters, and chemical synthesis. The performance of Pt NPs tends to degrade over time under reactive conditions, leading to a decrease in catalytic activity. Understanding the mechanisms that drive the degradation of Pt NPs due to oxygen interactions is therefore essential for the creation of more resilient and effective catalysts. In this study, we delve into the oxidation of Pt NPs by employing a combination of linear scaling Density Functional Theory (DFT) in conjunction with ReaxFF and the new MACE-MP-0 [1] forcefields. Our simulations are done on a realistic nanoparticle which was reconstructed via Scanning Transmission Electron Microscopy images [2]. Oxidation is carried out using Grand-Canonical Monte Carlo simulations with the ReaxFF forcefield [3]. Subsequently, selected structures are optimized using the new MACE-MP-0 model. Finally, the electronic properties of the optimized structures are then calculated using our linear scaling DFT code ONETEP [4].

References: [1] Ilyes Batatia et al., doi:10.48550/arXiv.2401.00096. [2] Jolyon Aarons et al., Nano Letters **17** (2017),pp. 4003–4012. [3] Lili Gai et al., The Journal of Physical Chemistry C **120** (2016), pp. 9780–9793. [4] Joseph C. A. Prentice et al., The Journal of Chemical Physics **152** (2020), p. 174111.

Truncated Quasiparticle Mass Divergence in NiS_2

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NiS_2 is a well-known Mott insulator which has been successfully described using density functional theory with dynamical mean field theory (DFT+DMFT). The previous studies done looking at the Mott insulator transition (MIT) in this material have been with respect to adding charge to the system and through substitutionally doping with Se. Recent high-pressure quantum oscillation experiments have found that the pressure-induced MIT contains a truncation in the mass divergence. This form of mass divergence and more specifically truncation has not been able to be replicated through DFT. This work aims to replicate this phenomenon through DFT+DMFT.

Luminescent europium complexes for chasing biomolecules

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The detection of biomolecules is important for biomedical and other biological applications. Molecules containing phosphate groups, such as ATP and ADP are particularly interesting because they participate in significantly important biological reactions [1].

In particular, the complexes of europium and terbium which exhibit emission in the red and green regions of spectra have unique photophysical properties that are useful for anion sensing in biological media [2,3]. The usefulness of these europium and terbium complexes can be attributed to their long luminescence lifetimes and sharp line-like emission spectra. However, it is challenging to design complexes with high selectivity for the target anion [4].

Computations can play an important role for improving the design process by providing molecular-level insight and accurate energetics of the relevant processes. Our work involves illuminating the structure and binding properties of luminescent europium complexes. In a first step, we need to devise a robust computational strategy that is amenable to charged ions in solution, taking account of thermostatistical effects and overcoming basis set superposition error. In addition, a potentially vast conformational space has to be searched. Within this contribution, we present our early results in the development of such a strategy discussing encouraging results but also further challenges. We present DFT optimised structures of host-guest complexes to provide insight into the binding modes of the guest species.

References: [1] R. V. Velazquez-Castillo, M. K. Salomon-Flores, A. O. Viviano-Posadas, I. J. Bazany-Rodriguez, C. Bustos-Brito, J. M. Bautista-Renedo, N. Gonzalez-Rivas, L. D. Rosales-Vazquez, A. Dorazco-Gonzalez, Dyes and Pigments, 2021, **196**, 109827. [2] S. J. Butler, D. Parker, Chemical Society reviews, 2013, **42**, 1652-1666. [3] S. E. Bodman, C. Breen, S. Kirkland, S. Wheeler, E. Robertson, F. Plasser, S. J. Butler, Chemical Science 2022, **13**, 3386-3394. [4] S. E. Bodman, S. J. Butler, Chemical Science, 2021, **12**, 2716-2734.

Quantum Chemical Characterization of Rotamerism in Biological Thio-Michael Additions

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Myotonic dystrophy type I (DM1) is the most common form of adult muscular dystrophy and is a severe condition that affects a range of systems, with no treatment currently available. Recently, small molecules have been developed that have some selectivity for and covalently inhibit the protein cyclin-dependent kinase 12 (CDK12). CDK12 is involved in the transcription of elongated RNA sections that results in the DM1 condition. The covalent bond is achieved after nucleophilic addition to a warhead. Here, we use high-level quantum chemistry calculations to characterise the gas-phase nucleophilic addition of an archetypal nucleophile, methanethiolate, to various nitrogen-containing molecules which are representative of the small-molecule CDK12 inhibitors. By investigating the structural, energetic, and electronic properties of the enolate intermediates, we show that synclinal additions are energetically favoured over antiperiplanar additions due to the greater number of attractive non-covalent interactions permitted by the stereochemistry. The mechanistic insights from this study will inform the design of new derivatives with faster reaction rates and higher yields of the adducts required for CDK12 inhibition and treatment of DM1.

Simulating grain boundaries in anatase TiO2 photocatalysts

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Carbon-free hydrogen production is an important technology that will help the UK to meet its legally binding Net Zero targets, by 2050, supplanting the use of fossil fuels. A catalytic means to decompose water into hydrogen would be ideal; TiO2 is a promising candidate for a photocatalyst, owing to its high abundance and low toxicity. However, its efficacy is limited by a large bandgap (3 – 3.2 eV) and rapid electron-hole recombination. This poster outlines the use of density functional theory (DFT) to investigate grain boundaries in anatase TiO2, notably the $\Sigma 3$ and $\Sigma 5$, and how they affect its electronic structure. This will provide a foundation for our future studies of surface terminated defects for TiO2-based photocatalysis.

Electronic Structure of Endohedral Metallofullerenes

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The electronic structure of 3 endohedral metallofullerenes (EMFs), including $Sc_3N@C_{80}$, $Sc_3C_2@C_{80}$, and $Er_3N@C_{80}$, in addition to an empty cage such as C_{60} are intensively explored. Investigations start with wave function plots of the studied molecules. Three different methods including Mulliken, Hirshfeld and Voronoi population analyses, were employed to track down the charge transfer between metallic moieties and the cage. To determine the most probable orientation for metallic moieties such as Sc_3N, Er_3N and Sc_3C_2 inside the cage, the counterpoise method was employed for their different rotation angles θ, Φ, and β in the gas phase. A similar approach was used on a Au-substrate in which case, rotations about four different rotation axes θ, Φ, α, and β are explored. It also explores how θ, Φ, α, and β axes pass through the metallic moieties and how the total energy varies with the angle of rotation, both in the presence and absence of a gold substrate.

INTONE: A package for the post-processing of ab-initio magnetic susceptibilities

Kiran Jonathan

STFC

Inelastic neutron scattering is a popular form of spectroscopy in the condensed matter community, and is often used in the study of the magnetic properties of materials. With the availability of new modules in codes such as Quantum ESPRESSO and Questaal for the simulation of magnetic susceptibilities, there is an increasing need to validate these methodologies against existing and future experimental results. INTONE is an open-source Python-based package which aims to unify the post-processing of magnetic susceptibility data from different ab-initio sources, with hopes of facilitating these kinds of experimental comparisons in the future.

Relativistic electronic structure and photovoltaic performance of Ki_2CsSb

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Antimony-based materials have emerged as promising non-toxic and earth-abundant photovoltaic candidates with similar electronic properties to the lead hybrid perovskites. The family of Heusler alloys X_2YZ and XYZ have attracted considerable attention due to their highly symmetric structure, compositional flexibility, low thermal conductivity and remarkable transport properties. Among them, the full-Heusler K_2CsSb has shown its potential application as a photo-cathode and thermoelectric, while its possibility to serve as a photovoltaic remains unexplored. In this project, using hybrid DFT calculations with spin-orbit coupling, K_2CsSb is found to exhibit an ideal direct bandgap, dispersive conduction band, strong optical absorption, and a competitive theoretical efficiency of over 28% at a thickness of 200 nm. The point defect study will be included in this project to further understand the trap-limited efficiency of this promising photovoltaic candidate.

Catalysis with ferroelectric nanomaterials

Chiara Gattinoni

King's College London

Heterogenous catalysis based on ferroelectric materials has great potential to provide solutions for some of humanity's most pressing issues, such as renewable energy generation or carbon sequestration. The main advantage of ferroelectric materials as catalysts is that the interaction of the ferrocatalyst with its surrounding environment can be manipulated by the in-built polarization. In this work, I investigate the ferrocatalytic process in two prototypical ferroelectrics, barium titanate ($BaTiO_3$) and bismuth ferrite ($BiFeO_3$). I show that the interplay between the charge state of the ions in the materials and their ferroelectric polarization lead to radically different ferrocatalytic processes for the hydrogen evolution reaction and a click cycloaddition.

ARTEMIS and RAFFLE

Francis Davies

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The prediction of the atomic structure of interfaces is a considerable challenge. ARTEMIS[1] and RAFFLE[2] represent two complementary tools developed to aid in the exploration of interface structures with the goal of eliminating human bias and reducing computational cost; together, these methods explore the configuration space to find the lowest energy structures. The interface problem involves several factors including balancing of mismatch strain, intermixing and reconstruction. Our approach shows that different interface morphologies can be formed, between two bulk crystals, and offers a pathway to finding new materials.

References: [1] N. Taylor, F. Davies, I. Rudkin, C. Price, E. Chan, S. Hepplestone (2020) Comp. Phys., 2020, **257**, 107515. [2] J. Pitfield, N. Taylor, S. Hepplestone, Physical Review Letters, 2024, **132**, 066201.

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