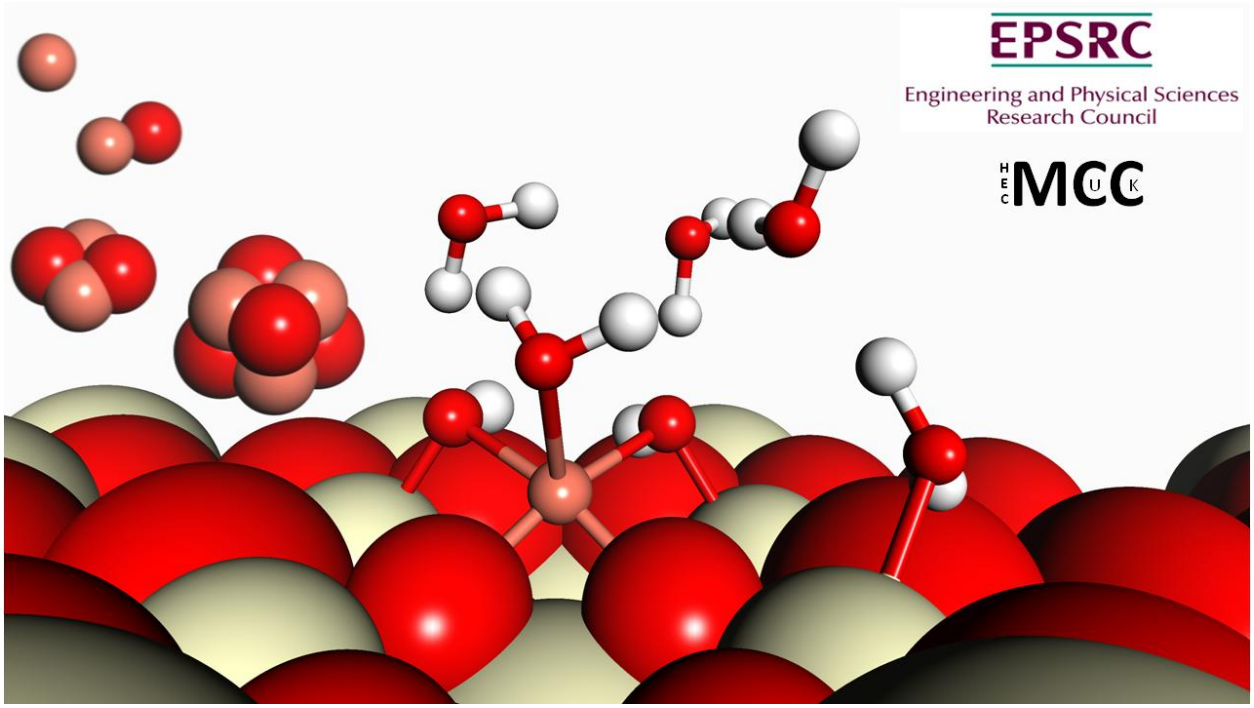
MCC 8th Conference



Daresbury

Conference: Wednesday 1st July 2026 – Friday 3rd July 2026
MCC meeting with group leads: Friday 3rd July 2026

Sponsors

The MCC is very grateful for funding from EPSRC (**EP/X035859**).

We are grateful that this event is also sponsored by:

STFC (Daresbury)

Department of Chemistry (UCL)

Organising Committee

Georgia Lomas, Alin-Marin Elena, Thomas Keal, Alexey Sokol, and Scott Woodley

We also would like to thank all members, our theme leaders, and our invited guests, who have supported this event by joining us in-person in Daresbury, albeit one, two, or all three days.

MCC 8th Conference - Programme

Wednesday 1st July			
11:30	**Registration Desk Open**		**Daresbury Laboratory**
12:00	**Lunch**		
Session 1:	Algorithms	**Chair** Tom Keal	
13:10	ML-PEG: machine learning performance and extrapolation guide		Elliott Kasoar STFC (COSEC)
13:30	Machine-learned potentials for silica polymorphism and MgO defect chemistry		Jamal Abdul Nasir UCL (smw)
13:50	**Invited:** Simulating molecules and materials on an ion trap quantum computer		David Munoz Ramo Quantinuum
14:30	AI for clusters: extracting structural data from images through deep learning		Cyril Xu UCL (smw)
14:50	MolSim++: GPU confined particle dynamics project		Joseph Thacker STFC (COSEC)
15:10	PATINA: A modular software approach towards composable & reproducible computational materials workflows		Gokay Avci UCL (smw)
15:30	**Tea**		
Session 2:	Discovery	**Chair** Natalia Martsinovich	
16:00	A multi-scale mixture of experts model for cross-size structural prediction of Cu nanoparticles		Yunyu Zhang UCL (cat)
16:20	Integrating ML and AI with classical simulations for automating materials discovery		Chris Collins Liverpool (dar)
16:40	FAIR-MOFs: Structure-centred synthesis inference from three-dimensional structures of metal-organic frameworks		Dinga Wonanke NTU (add)
17:00	DeepHydride: A machine-learned interatomic potential based framework for high-throughput discovery of metal hydrides		Lei Lei Nottingham (lin)
17:20	The morphology of sub-nanometer clusters on surfaces		Thomas Hill Cardiff (cat)
17:40	**Poster Session** Starting with 30s-lightning presentations (1 slide per poster) Session will include food and refreshments		
20:00	Session ends		

Thursday 2nd July			
Session 3:	Bulk	**Chair/Theme Leader** Keith McKenna	
9:00	**Invited:** Equations to Enterprise: Turning Computational Materials Research into Industrial Impact		David Gao Nanolayers
9:40	The solid-state chemistry of thorium-229 nuclear clocks		Harry Morgan Manchester (hwt)

10:00	Computational discovery of symmetry-protected and distortion-induced topological phonons in half-Heusler alloys	Sam Ginzburg Cambridge (sjj)
10:20	Bridging the Gap Between Theory and Experiment: Machine Learning Inter-atomic Potentials for Metal Organic Frameworks	Alin Marin Elena STFC
10:40	QM/MM investigations of defects in MgO and La_2CuO_4 and other related materials	Liam Morgan UCL (sok)
11:00	**Coffee**	

Session 4:	Surfaces and Interfaces	**Chair** Sanliang Ling	
11:20	**Invited:** CRYSTAL, an ongoing project for a chemical approach to solids		Silvia Maria Casassa University of Torino
12:00	Phonon and thermal transport effects of extended defects in reduced γ-WO_3 via machine learned interatomic potentials		Anas Siddiqui York (mck)
12:15	Towards modelling vibration-assisted hot-carrier processes in metallic nanoparticles		Damian Contant KCL (iva)
12:30	Oxygen scavenging at a SiO_2/Ta interface: a first-principles study		Margherita Buraschi UCL (shl)
12:45	Oxygen-driven evolution of novel Σ5 tilt boundaries in Anatase TiO_2 by in-situ eSTEM/TEM		Jack Fawcett-Hough-ton, York (mck)
13:00	**Lunch**		
Session 5:	Reactivity	**Chair/Theme Leader** Richard Catlow	
14:00	**Invited:** Advancing MOF simulations with Machine Learned Potentials		Guillaume Maurin Montpellier II
14:40	Effects of surface nanostructure of metal co-catalysts for photocatalytic conversion of methane: a theoretical study		Natalia Martsinovich Sheffield (nat)
14:55	Impact of the presence of Au on activity and selectivity of Cu/ZnO catalysts for methanol synthesis		Michael D. Higham Cardiff (cat)
15:10	Nitrogen evolution reaction study on Ru supported clusters and Ru(0001) surface		A. Amanzhol Cardiff (rol)
15:25	Investigating the formate pathway for CO_2 to MeOH over Pd/ZnO catalysts		E. McCarthy Cardiff (wil)
15:40	**Coffee**		
Session 6:	Power	**Chair/bulk Theme Leader** Alex Shluger	
16:10	**Invited:** Chemo-mechanics and Kinetics of Li_xNiO_2 - unravelling hidden complexities		Karsten Albe TU Darmstadt
16:50	Vibrational spectra of zirconolite		Joseph Flitcroft Manchester (bulk-ske)
17:05	Correlation of polarons and highly charged interstitials in metal oxides		Isaac Mackley UCL (bulk-shl)
17:20	Multiscale modelling of grain boundaries and extended defects in halide perovskites using machine-learning potentials and DFTB		Junke Jiang York (power-mck)

17:35	The Monte Carlo threshold algorithm in crystal structure prediction: introducing intramolecular degrees of freedom		Pedro Juan-Royo Southampton (discov-day)
17:50	session ends		
18:00 – 21:00	**Conference BBQ Dinner**		Daresbury Caterers

Friday 3rd July			
Session 7:	Reactivity	**Chair/Theme Leader** David Willock	
9:00	Rational design of sulphur-terminated MXenes for efficient CO_2 conversion into carbon neutral methanol production		Moses Abraham Leeds (lee)
9:20	The pressure-dependent surface state of cobalt during Fischer-Tropsch synthesis		Jack Swallow Manchester (swa)
9:40	Single- and dual-atom catalysts for the CO_2 reduction reaction		L. Rivera-Blair Cardiff (rol)
9:55	Updates to phonopy-spectroscopy: first-principles modelling of infrared and Raman spectra		J. Skelton, Manchester (bulk-ske)
10:15	Cage electron mediated hydride formation in Mayenite electride		Navaratnarajah Kuganathan Nottingham (lin)
10:30	**Coffee**		
Session 8:	Algorithms	**Chair** Andrew Logsdail	
11:00	**Invited:** From Lithium Batteries to Perovskite Solar Cells - Atomistic Insights into Energy Materials		Saiful Islam Oxford (isl)
11:40	Reliable and reproducible benchmarking of DFT codes on GPUs: A CASTEP case study		Oscar van Vuren Cardiff (log)
11:55	DL-FIND: Benchmarking methods for geometry optimisation		Thomas R. Durrant STFC (COSEC)
12:10	Implementing extendable workflows for QM-in-QM embedding		Gabriel Bramley Cardiff (log)
12:25	**Lunch**		
Session 9:	Enviro / Biosoft	**Chair** Umberto Terranova	
13:30	Field-free coexistence of skyrmions and anti-skyrmions induced by higher-order interactions and biaxial strain in the NiI_2 monolayer		Zebin Wu UCL (nano-sok)
13:45	Machine learned potentials for Li based reactive hydride composites		Oliver Morrison, Nottingham (surfin-lin)
14:05	Effects Of Isomerism on Porous framework materials: Window Edition – How Does Isomerism Affect A Guest Molecules Ability To Enter A Pore		Saad Choudry NTU (discov-add)
14:20	Probing nuclear extremes in lithium ceramics: multiscale simulations with machine-learnt interatomic potentials		Cillian Cockrell Bangor (enviro-cil)

14:40	Close		
15:00	**MCC Meeting of Group Leaders** The future of the MCC: open discussion		
16:45	Close		

Summary of Poster Presentations

1	A Density Functional Theory study on the Point defects in PuO_2	Paris-Rose Dawson Huddersfield (bulk-mol)
2	Atomistic Simulation of Liquid Electrolytes on Argyrodite Surfaces for Hybrid Solid-Liquid Lithium Batteries	Felix Scott Newcastle (surfin-jad)
3	Using DFT Simulations to Investigate Ammonia Conversion to Hydrogen on Transition-Metal Based Catalysts	Sebastian Teodorescu-Oros, Surrey (react-cai)
4	Computational Modelling of Photocatalytic Conversion of Organic Molecules into Biofuel	Luke Andrew Sheffield (react-nat)
5	Computational Study of Sodium-Ion Migration and Defect Chemistry in Na_8SnP_4-Based Solid Electrolytes	Patabandi Mudiyanselage Birmingham (power-dos)
6	Exploring the Defect Chemistry of Potassium Tantalate	Urvi A. Joshi Birmingham (bulk-dos)
7	A Computational and Experimental Study of the Methanol Synthesis catalyst	Matis Ferrini Lincoln (react-arc)
8	Amorphous-like Thermal Conductivity and High Thermoelectric Figure of Merit in "π" SnS and SnSe	Min. Zhang Manchester (power-ske)
9	Investigating Vacancy Defects in a Candidate Transparent Conducting Oxide, $LaSb_3O_9$	Jack N. Beattie Birmingham (bulk-dos)
10	Pressure Driven Evolution of Various Metal Hexaborides	Abubakar Imran Manchester (bulk-hwt)
11	First-Principles Investigation of Band Alignment and Defect-Induced Electronic States at 2D–3D Perovskite Interface	Shaojun Hao Newcastle (surfin-jad)
12	Computational Study of Thorium-Containing Host Materials for use in Thorium-229 Nuclear Clocks	Dewi Parry Manchester (bulk-hwt)
13	Benchmarking the Energy Ranking of Molecular Crystals at the Meta-GGA Level	Namrata Ramesh Leeds (bulk-arp)
14	Computational Investigation of CO_2 Methanation over Ni and Ni-Alloy Catalysts	M. Alotaibi UCL (react-cat)
15	Periodic DFT Investigation of CO_2 Hydrogenation on Clean and Fe-Doped Ni(111) Surfaces	Linqing Dong UCL (react-cat)
16	A Study of H_2O-Promoted CO_2 Coupling to Ethanol on the Cu(100) Surface	Zhenze Zhao, UCL (react-cat)
17	Modelling Charge Carriers in Cuprate High-Temperature Superconductors	Taochun Ma UCL (bulk-sok)
18	QM/MM Calculations for ZnO (10-10) and the CO_2 Adsorption	Shumin Li Cardiff (react-wil)
19	Modelling Excited States of MOFs NU-1000 and NU-901 Using Cluster and Periodic Approaches	Selin Kilic UCL (discov-rco)
20	Machine-Learned Structure Prediction of TiO_2 Clusters Using Fine-Tuned MACE Potentials	Anna-Lena Singer UCL (nano-smw)
21	Machine Learning Study of Charge Compensation Mechanisms in Zr-Doped ZnO at low doping Concentration	Yulin Song UCL (power-smw)
22	Spin-Polarised DFT Study of Antiperovskite Materials for Lithium-Ion Battery Applications	Ruojie Huang UCL (power-smw)
23	Sampling Defect Complexes in Beryllium Doped GaN Using a Machine Learning Interatomic Potential	Yuxuan Liao UCL (power-smw)
24	Model Collapse under Iterative Self-Training of Generative Models	Joley Yuhui Lin UCL (discov-but)
25	Atomistic Modelling of CoO_x/TiO_2 interfaces: Elucidating Metal-	Akash Hiregange

	Support Interactions in Fischer-Tropsch catalysts	Cardiff (surfin-log)
26	Time-Resolved Simulation of Shock-Driven Energy Redistribution in Crystalline Energetic Materials	Mateusz Mojsak Birmingham (bulk-adm)
27	Mechanochemically Promoted CO_2 Methanation over Co-CeO_2 Catalysts via Collision-Induced Charge Effects	Weiwei Xu Liverpool (react-xue)

Invited Talks

Simulating molecules and materials on an ion trap quantum computer

David Munoz Ramo

Quantinuum, UK

Quantum computing is receiving increasing attention for molecular and materials simulation tasks due to their capability to efficiently represent and evolve quantum systems. This presentation highlights recent advances in my team's research in quantum dynamics, electronic structure, and quantum error correction demonstrated on our ion trap quantum computers. We demonstrate quantum simulations of NMR dynamics and light-matter interactions, alongside studies of actinide chemistry models using the Quantum Phase Estimation algorithm . We also present the first end-to-end quantum chemistry workflow using a quantum error-correction code on trapped-ion hardware. These results showcase the progress toward fault-tolerant quantum chemistry and the key developments needed to achieve practical quantum advantage.

Equations to Enterprise: Turning Computational Materials Research into Industrial Impact

David Gao

Nanolayers, UK

Nanolayers Research Computing was founded to bridge the gap between advances in computational materials science and real-world industrial applications. In this talk, I will provide an overview of Nanolayers, how we operate at the interface of research and commercialisation, and our experience building spin-out companies from scientific innovation. I will then introduce our latest venture, focused on machine-learning models for electronic structure applications, and discuss how these emerging approaches are enabling faster and more scalable materials modelling. Together, these examples illustrate a pathway from fundamental science to industrial impact in the materials sector.

CRYSTAL, An Ongoing Project for a Chemical Approach to Solids

Silvia Maria Casassa

University of Torino, Italy

TBA

Advancing MOF simulations with Machine Learned Potentials

Guillaume Maurin

University of Monpellier II, France

TBA

Chemomechanics and Kinetics of Li_xNiO_2: Unraveling the hidden complexities by combining electronic structure calculations, cluster expansion and machine-learning potentials

Karsten Albe

Technische Universitat Darmstadt, Germany

Layered transition metal oxides with ultra-high nickel content are currently experiencing renewed interest as cathode active materials for next-generation lithium-ion batteries. Because these materials are closely related to the model system $LixNiO_2$, understanding its structure, thermodynamics, chemo-mechanics, and defect physics on the atomic scale is essential for the development of more advanced nickel-rich cathodes. In this contribution I will show that understanding the chemo-mechanical effects of 0- to 2-dim. defects is key for describing the real material behavior.

I will start with discussing the dynamic Jahn–Teller distortions in $LixNiO_2$, explaining the coexistence of local monoclinic distortions with an average rhombohedral structure [1] . Then I will show that cluster expansion techniques allow to simulate experimentally observed phase diagram, if the presence of native defects, particularly Ni ions occupying Li sites (NiLi), is taken into account, which modify the thermodynamics by promoting solid-solution behavior and smoothing voltage profiles [2,4].

In a next step, the role of dislocations in LiNiO is discussed, which can act as sinks for Li and O vacancies, thereby influencing transport processes and serving as precursors for degradation phenomena [3]. Finally, to enable the simulation of defect-mediated chemomechanical processes on larger length and time scales, a machine-learning interatomic potential for $LixNiO_2$ and its delithiated phases is presented. This Atomic Cluster Expansion (ACE) potential was trained on an actively expanded r²SCAN-rVV10 density-functional-theory database and validated against bulk, defect, and interface configurations. The resulting model accurately reproduces thermodynamic and elastic properties and captures key structural transformations, including the dissociation of screw dislocations into Shockley partials enclosing O1 stacking faults. Using the ML-MD simulations, I will show how strains induced by coherent interfaces affect the stability of $LixNiO_2$.

[1] Chem. Mater. 2020, 32, 23, 10096–10103; [2] J. Mater. Chem. A, 2021,9, 14928-14940; [3] Chem. Mater. 2023, 35, 2, 584–594; [4] Chem. Mat, 2024 36 (1), 492-500

From Lithium Batteries to Perovskite Solar Cells - Atomistic Insights into Energy Materials

Saiful Islam

Department of Materials, University of Oxford

Further breakthroughs in materials for low carbon energy applications require advances in new compositions and underpinning materials science. Indeed, a greater fundamental understanding of new materials for lithium-ion batteries and solar cells require atomic-scale characterization of their structural, electronic and transport behaviour. In this context, advanced modelling methods (including ab initio, MD and ML potentials) are powerful in investigating these properties on the atomic scale. This presentation will describe such materials modelling studies in two topical areas: (i) structural and redox chemistry of lithium-rich cathodes for lithium-ion batteries; and (ii) ion migration and molecular passivation effects in metal-halide perovskites for solar cells.

Contributed Talks

ML-PEG: Machine Learning Performance and Extrapolation Guide

Elliott Kasoar*, Joseph Hart, Ilyes Batatia, Domantas Kuryla, Alin M. Elena, Gábor Csányi

Scientific Computing, STFC UKRI / Engineering Laboratory, algor-cosec** University of Cambridge*

Recent advances in machine learnt interatomic potentials (MLIPs) are revolutionising atomistic simulations, enabling results with accuracy comparable to ab initio calculations at significantly greater time and length scales. In addition to improved model architectures, access to large, high-quality datasets has enabled the creation of foundational models, trained on sufficiently diverse data to act as a general-purpose model, or starting point, for applications ranging from molecules to proteins. However, as model architectures continue to develop, and new training data becomes available, it has become clear that we are limited in our current methods for rigorously benchmarking foundational MLIPs. Many frequently cited benchmarks are becoming saturated in their scores, focus on a narrow range of systems, such as bulk inorganic crystals, and favour low energy and force errors, rather than more complex physical properties. These limitations inhibit meaningful comparisons of state-of-the-art models, as well as our ability to explain quite diverse performances of models when studying different properties or systems. To address this, we introduce the Machine Learning Performance and Extrapolation Guide (ML-PEG), a hierarchical, customisable, interactive interface, through which the results of benchmarks spanning a wide range of material/molecular applications and physical properties can be examined, ranging from a single numerical score to visualisation of individual atomic structures from a dataset.

Machine-Learned Potentials for Silica Polymorphism and MgO Defect Chemistry

Jamal Abdul Nasir*, Alin Marin Elena, Alexey A. Sokol, Bruno Camino, Aurora Gherson Beckwith, Keith Butler, C. Richard A. Catlow, Scott Woodley*

*Department of Chemistry, University College London algor-**smw***

Silica-based materials and ionic oxides remain important model systems for understanding structure, stability and reactivity in materials chemistry[1][2]. In this work, we use machine-learned interatomic potentials to study two related problems: the stability of silica frameworks and polymorphs, and defect formation and migration in MgO.

For silica, we apply the MACE-MP-0 potential to siliceous zeolites and dense silica phases. The model describes structures with different Si coordination environments without the need for re-parameterisation, which is a major advantage over many classical potentials. Our calculations reproduce the metastability of microporous zeolite frameworks relative to α-quartz and describe the compression behaviour of quartz, coesite and stishovite. The predicted transition pressures, around 3.5 GPa for quartz→coesite and 9 GPa for coesite→stishovite, are close to experimental values, showing that this approach can give a reliable description of silica polymorphism.

We also benchmark machine-learned interatomic potentials for MgO, where Schottky defect energies depend strongly on electrostatics, lattice polarisation and supercell size. Paired neutral Schottky defects and isolated charged vacancies therefore define different reference scales and should not be mixed. A general foundation MACE model does not capture the expected size dependence. We therefore use two MgO-specific models fine-tuned on our PBE0 MgO dataset: MACE-POLAR, which includes explicit electrostatic and polarisation terms, and MACE-mh-1-v1.1, a short-range multi-head MACE model fine-tuned from the MATPES-r2SCAN head that is not polarizable. Both models better reproduce the PBE0 paired-defect energy scale; notably, MACE-mh-1-v1.1 also gives an improved Mg-vacancy migration barrier of ~2.04 eV, consistent with previous DFT and experiment.

References

1. Abdul Nasir, J., Guan, J. Jee, W., Woodley, S. M., Sokol, A. A., Catlow, C. R. A., & Elena, A. M. (2024). Modelling Silica using MACE-MP-0 Machine Learnt Interatomic Potentials. arXiv preprint arXiv:2411.00436.

2. Catlow, C.; Faux, I.; Norgett, M. Shell and breathing shell model calculations for defect formation energies and volumes in magnesium oxide. Journal of Physics C: Solid State Physics 1976, 9 (3), 419–430.

AI for Clusters: Extracting Structural Data from Images through Deep Learning

Cyril Xu*, Alexey A. Sokol, Scott M. Woodley

*Department of Chemistry, University College London algor-**smw***

The growth in computational power and data availability now enables alternative approaches to materials modelling. While GNNs (Graph Neural Networks) have advanced the field by explicitly modelling atomic connectivity, they often abstract away the continuous 3D geometric context preserved in raw imaging data — information that is valuable for property prediction. Traditional cheminformatics approaches have similarly relied on finite descriptor sets (e.g., molecular weight and dipole moments), which, while efficient for regression modelling, fail to capture underlying structural topology.

By leveraging Computer Vision (CV) techniques, complex geometric features can be recovered efficiently by feeding raw images to Deep Neural Networks. The distinct advantage of this approach is that it bypasses feature engineering, allowing the network to learn latent physical properties such as electron density for high-fidelity predictions. While recent literature has begun to employ this methodology in predicting molecular properties such as HOMO-LUMO gaps, it has not yet been widely adopted in modelling clustered materials, leaving a significant opportunity for methodological development.

Atomic clusters exist on complex energy landscapes characterised by multiple competing low-energy minima, routinely represented in the literature as static 2D images. Extracting high-fidelity structural data, such as 3D atomic coordinates, from these 2D representations is an ill-posed problem. Standard methodologies struggle to resolve depth ambiguity caused by atomic occlusion, where atoms are obscured in the projected view. We introduce a preliminary deep-learning architecture utilising state-of-the-art CV techniques to resolve depth ambiguity. The extracted structural data can then be used for downstream applications, including populating the HIVE database.

MolSim++: GPU confined particle dynamics project

Joseph Thacker, Benjamine Speake, Michael Seaton, Ilian Todorov

STFC Scientific Computing *algor-**cosec***

We will present a new particle dynamics driver (to address Dissipative Particle Dynamics and Molecule Dynamics simulation) aimed at performing on GPU architectures while extensible to future hardware advances. Embodying best software practices, the project was conceived in C++ and adopts the SYCL standard to leverage GPU parallelism while also being vendor and compiler agnostic. We will present the project evolution, the core algorithms underpinning our GPU confined performance and compare our performance to that of other leading particle dynamics engines' GPU ports (e.g, LAMMPS-kokkos and GROMACS-CUDA). We will discuss various aspects of our strategy to balance performance gains and compromises on a GPU, the performance-portability of SYCL kernels, and how our project strives to maximise code reuse and sustainability by adopting core software design principles. We will also outline the benefits and consequences of starting a software project from scratch (as opposed to building upon our previous software experiences DL_POLY, DL_MESO and their CUDA ports).

PATINA: A Modular Software approach towards Composable and Reproducible Computational Materials Workflows

Gokay Avci*, Alexey A. Sokol, Scott M. Woodley

*Department of Chemistry, University College London algor-**smw**

Computational materials discovery increasingly depends on the ability to connect atomistic simulation, workflow automation, provenance tracking, structural analysis, and reproducible data generation across heterogeneous software environments. [1] Existing materials-modelling workflows often combine multiple simulation engines, scripting layers, file formats, databases, and job schedulers. Although this flexibility is powerful, it can also produce fragmentation: atomic structures, bonding information, electrostatic descriptors, simulation metadata, and optimisation histories are often represented differently across workflow stages. This reduces maintainability, weakens scientific verification, and limits the reuse of computational protocols across materials classes. [2] Conversely, monolithic workflows are usually easier to understand and deploy, but they often struggle to support extensible, multi-paradigm materials discovery workflows that combine global optimisation, machine learning, high-throughput simulation, and human-guided scientific reasoning.

Here we introduce PATINA (Platform for Atomistic Topology, Inference and Networked Assembly), a single binary computing platform designed to support reproducible and extensible atomistic simulation workflows, written in Rust language, inspired from previous KLMC implementations. [3] PATINA is built around native data structures for atomic configurations, bonding graphs, topological relationships, simulation state, and workflow metadata. The platform is designed around three core ideas: 1) A consistent internal representation of atomistic structures, enabling structural comparison, validation, mutation, and analysis to be performed independently of the original simulation code. 2) A modular architecture which supports global structure search algorithms across multiple materials classes, including clusters, molecular materials, oxides, porous frameworks, and non-stoichiometric systems. 3) Choice of Rust as the main language provides memory safety, strong typing, and safe concurrency, making the platform suitable for distributed simulation submission, active learning loops, and long running stable computational campaigns.

As a demonstration, we discuss an evolutionary structure search workflow for non-binary oxide clusters. Candidate structures are generated, mutated, filtered, submitted to external simulation engines, and verified using PATINA's internal structural and workflow abstractions. This enables the optimisation history, simulation provenance, and structural evolution of the population to be tracked in a reproducible form.[4, 5] The same abstraction can be extended towards machine learning assisted workflows, where surrogate models, interatomic potentials, or active learning agents select new structures for evaluation.

PATINA is intended to complement the wider computational materials ecosystem, including workflow platforms, simulation engines, databases, and community infrastructures. By providing a strongly typed, single binary orchestration layer with native structural verification and concurrent execution protocols, PATINA offers a route towards more maintainable, reproducible, and agent ready materials simulation workflows. This contribution will discuss the design principles of PATINA, its current capabilities, and possible routes for integration with the MCC community.

[1] Scientific Data 7, 300 (2020); [2] Scientific Data 7, 299 (2020); [3] PCCP 16, 21119–21134 (2014); [4] Nanoscale 9, 3850–3864 (2017); [5] J Chem Phys 124, 244704 (2006).

A Multi-Scale Mixture of Experts Model for Cross-Size Structural Prediction of Cu Nanoparticles

Yunyu Zhang*, Keith T. Butler, C. Richard A. Catlow

*Department of Chemistry, University College London discov-**cat**

Predicting structures and energetics of metallic nanoparticles across wide size ranges remains challenging because the balance of interaction scales changes rapidly with system size. We introduce a multi-scale Mixture-of-Experts (MoE) architecture for Cu clusters and nanoparticles that explicitly separates short-, medium-, and long-range interactions using three specialised machine-learning interatomic potential experts combined through a learnable size-conditioned gating network. The resulting MoE aggregates per-atom energies into a single conservative potential, ensuring forces are obtained as energy gradients and enabling stable molecular dynamics. Across mixed cluster-nanoparticle test sets, the MoE improves accuracy relative to both an off-the-shelf foundation potential and a finetuned single-expert baseline. Stress tests show that force errors remain comparatively stable across cluster sizes and that the model retains robust energetics under morphology out-of-distribution shifts quantified using a structural outlier score based on similarity measures. The learned gating weights further provide an interpretable, size-dependent decomposition of interaction scales. Finally, validation against linear-scaling density functional theory using the ONETEP code, together with finite-temperature molecular dynamics tests, demonstrates consistent energetics, stable force behaviour, and well-behaved dynamical trajectories, supporting the use of the model for efficient structural optimisation and configurational sampling across nanoparticle sizes.

Integrating Machine Learning and Artificial Intelligence with Classical Simulations for Automating Materials Discovery

Collins, C. M., J. Clymo, K. Atkinson, M. S Dyer., M. W. Gaultois., V. Gusev, Darling, G. R., Claridge, J. B., & Rosseinsky, M.

*Department of Chemistry, University of Liverpool discov-**dar***

In the grand challenge of discovering new inorganic solids numerous tools continue to emerge: for the prediction of compositions, chemical structures, physical properties, and high throughput synthesis. In this talk I will discuss our recent work in prediction for materials discovery, supported by compute time from the materials Chemistry consortium. I discuss our recent work, where we are developing workflows to guide the exploration of new materials, including the integrated use of machine learning (ML), artificial intelligence (AI) and crystal structure prediction (CSP).

The integration of generative machine learning with heuristic crystal structure prediction[1]. I present the most recent implementation of my crystal structure prediction code FUSE, where the method is adapted to use generative AI, reinforcement learning & machine-learnt inter atomic potentials. And illustrate it with some recent predictions and resulting experimental outcomes.

I will demonstrate how FUSE is coupled with a new explainable AI tool COMGEN[2], which allows a user to specify sets of chemical constraints and then explore the consequence of them providing explainable results. I integrate COMGEN into an automated workflow able to perform large scale CSP screening on compositions suggested by COMGEN to identify candidate compounds for investigation.

References:

1 Collins, C. M., Sayeed, H. M., Darling, G. R., Claridge, J. B., Sparks, T. D. & Rosseinsky, M. J. Integration of generative machine learning with the heuristic crystal structure prediction code FUSE. *Faraday Discussions*, (2024), doi:10.1039/D4FD00094C.

2 Clymo, J., Collins, C. M., Atkinson, K., Dyer, M. S., Gaultois. M. W., Gusev, V., Rosseinsky, M. J., Schewe, S., *Angew. Chem. Int. Ed.* 2024, doi: 10.1002/anie.202417657

FAIR-MOFs: Structure-Centred Synthesis Inference from Three-Dimensional Structures of Metal-Organic Frameworks

A.D. Dinga Wonanke*, Antonio Longa, Asha Pankajakshan, Joseph O. Ogar, Lauri Himanen, Alvin N. Ladines, José A. Márquez, Matthew A. Addicoat, Deborah L. Crittenden, Markus Scheidgen, Pietro Lio, Stefanie Dehnen, Christof Wöll, Thomas Heine

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Material discovery often begins with the computational design of new three-dimensional (3D) structures. For metal-organic frameworks (MOFs), this has led to the prediction of millions of high-performing hypothetical materials. However, when one consults the Cambridge Structural Database (CSD), only approximately 120,000 MOFs have been synthesised. Furthermore, despite the promise of MOFs for a wide range of applications, their translation to industrial-scale deployment remains limited. This clearly indicates that the barrier from computational prediction to experimental synthesis and scaling is still very high.

To lower this barrier, we implemented FAIR-MOFs, a structure-centred framework that enables synthesis inference directly from 3D MOF structures. FAIR-MOFs is built on over 47,000 curated experimentally synthesised MOFs, in which 3D structures are systematically deconstructed into chemically meaningful building units and mapped to literature-derived synthesis records. Building on this, we trained a graph neural network that infers the most probable metal-salt precursors directly from a target structure, while organic ligands are identified through fragment recognition and cheminformatics. Solvents and complementary reagents are recommended using a retrosynthetic recommender trained on reagent co-occurrence patterns and deployed as an interactive web platform, cheminteraction.com**.**

We demonstrate the practical utility of this approach by successfully synthesising three MOFs originating from hypothetical databases using only model-proposed precursor sets, obtaining phase-pure products confirmed by PXRD and FTIR spectroscopy. FAIR-MOFs therefore establishes a direct bridge between computational structure prediction and experimental synthesis by providing a pathway towards targeted, reproducible and experimentally accessible MOFs.

DeepHydride: A Machine-Learned Interatomic Potential Based Framework for High-Throughput Discovery of Metal Hydrides

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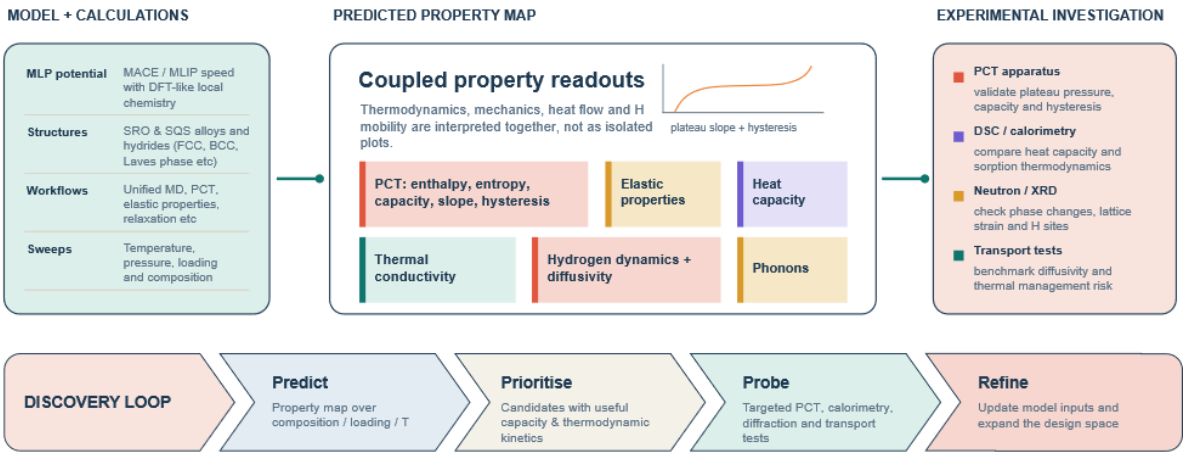
Discovering optimal metal hydrides requires co-optimising thermodynamic, mechanical, and transport properties across vast compositional spaces—a task for which Density Functional Theory (DFT) is prohibitively expensive. To bridge this gap, we present DeepHydride, an open-source Python framework centred on a MACE[1] machine-learned interatomic potential (MLIP) offering near-DFT accuracy at molecular dynamics (MD) throughput. Trained on the Materials Project trajectories supplemented with ~2 million structures spanning high-entropy alloys, AB_2 Laves phases, and hydrides, the model achieves energy and force MAEs of 5.2 meV/atom and 6.5 meV/Å on 346,000 unseen frames. This improves upon the MACE-MP-0 baseline by 20× and 18×, alongside a 9-fold stress error reduction. PlatonicRep[2] analysis confirms the model encodes fundamentally new local chemistry for hydrogen and titanium rather than merely re-weighting existing features. DeepHydride automates workflows for: (i) pressure–composition–temperature (PCT) isotherm prediction; (ii) MD-based hydrogen diffusivity and Green-Kubo thermal conductivity; (iii) elastic constant calculation; and (iv) complex structure generation. MD simulations on $Ti_{0.1}Zr_{0.9}Mn_{0.9}V_{0.1}Fe_{0.5}Ni_{0.5}$ yield experimental-level hydrogen diffusivities (10^{-6}–10^{-5} cm^2/s). Thermal conductivity calculations reveal that the hydrogen diffusion contribution to heat transport is negligible (< 10 m W m^{-1} K^{-1}) at all conditions, while the lattice contribution remains glass-like (0.3–0.6 W m^{-1} K^{-1}). Embedded within a targeted predict–prioritise–probe–refine discovery loop, DeepHydride substantially compresses the experimental search space for next-generation metal hydrides.



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The Morphology of Sub-Nanometer Clusters on Surfaces

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*discov-**cat***

Metal–support interactions strongly influence the structure and catalytic activity of supported metal clusters.[1] Computational studies often involve methodologies that use gas-phase clusters placed on surfaces, which can bias optimisation toward local minima, which do not truly replicate the interaction between cluster and surface.

The work presented aims to accelerate cluster discovery with machine learning (ML) techniques. Using the ReMatch + SOS algorithm in combination with pre and post screening techniques promises to reduce the computational expense of Monte Carlo /Global Optimisation methods.[2]

Au/CeO2 and Cu/TiO2 systems were investigated where 12 and 13 atom clusters were generated on the stoichiometric and defective surfaces ((111) CeO2, (110) rutile TiO2, (101) anatase TiO2). Comparison with optimisation of equivalent gas-phase clusters showed that the methodology could identify lower-energy supported structures, including the global minimum for the target cluster size.

Strong interactions of the metal species with surfaces showed donation of electrons towards the metal oxide to form cations. On defective surfaces dispersion of the metal clusters was observed, a direct result of the formation of anionic metal species, causing repulsion within the clusters.

The developed method has shown how dependent morphology of clusters is based on the characteristics of the surface. With this method, we can demonstrate how import morphology of small metal clusters is to catalytic activity.

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The Solid-State Chemistry of Thorium-229 Nuclear Clocks

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The thorium-229 isotope has a nuclear excited state at 8.4 eV above the ground state that can be excited with a tabletop laser, as demonstrated for the first time in 2024 after a 50-year search.[1,2] The frequency of this transition is extremely well defined and, being a purely nuclear process, insensitive to environmental perturbations. It therefore holds great potential as a next-generation clock, outperforming atomic clocks by orders of magnitude. A major goal is development of a solid-state nuclear clock, where ^{229}Th is present in a host material rather than an optical trap.[3,4] A solid-state clock would have a high concentration of ^{229}Th nuclei, allowing us to reach high accuracy in rapid measurements, and it would be chemically and physically durable, making it suitable for applications from millimetre-accurate GPS to earthquake prediction.

The properties of the device will be determined, in part, by the host material. In this talk I will present the story of ^{229}Th from a chemist's perspective, emphasising structure, electronics, and materials discovery. I will also describe developments in other directions, from thin films to nonlinear optics, and highlight the role that materials chemistry can play in this exciting new field.

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Computational Discovery of Symmetry-Protected and Distortion-Induced Topological Phonons in Half-Heusler Alloys

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Topological phonons offer a pathway to robust vibrational behaviour in crystalline solids, with promising implications for thermal transport, spin-phonon coupling, and symmetry-guided materials engineering. Yet predicting such states remains challenging because the presence and character of phonon band crossings are highly sensitive to crystal symmetry, structural distortions, and the underlying lattice-dynamical description.

Here, we present a first-principles study of topological phonons in half-Heusler compounds, focusing on the interplay between symmetry, strain, and vibrational band topology. To enable systematic exploration, we developed TopoloPy, an automated workflow designed to identify and classify both trivial and non-trivial band crossings throughout the Brillouin zone.

For a family of structurally isomorphic face-centred-cubic half-Heuslers - NiMnSb, CoMnSb, CoVSb - we demonstrate that exchanging atoms between the transition-metal sublattices, while preserving the FCC framework, reshapes the phonon dispersions and gives rise to symmetry-inequivalent Weyl families on different band pairs, alongside extended nodal features. By progressively lowering the symmetry, we then examine how strain reorganises the phonon-crossing landscape. In the uniaxially distorted tetragonal phase, non-trivial, axis-localised nodal precursors emerge and evolve into Weyl crossings, while further lowering to an orthorhombic geometry opens a distinct route in which Weyl charge is interchanged between separate reciprocal-space channels. Together, these results provide first-principles evidence for a non-local, strain-driven mechanism of topological reorganisation, in which Weyl charge can be “teleported” through nodal-line-like manifolds, thereby extending mechanisms previously proposed in minimal-model Hamiltonians to realistic half-Heusler materials.

Cage Electron Mediated Hydride Formation in Mayenite Electride

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The storage of hydrogen as atomic hydride ions (H^-) is important for developing advanced materials for catalysis, electron emission, energy conversion, and hydrogen activation because hydride species are highly reactive and difficult to stabilize under ambient conditions. Electrides are a unique class of materials in which electrons act as loosely bound anions localized within cavities or channels of the crystal lattice, making them promising for applications such as catalysis, electron emitters, batteries, and sensors [1-3]. Using density functional theory (DFT) simulations, we study the formation of H^- ions from gaseous H_2 in $12CaO•7Al_2O_3$ (C12A7), also known as mayenite electride in its electron-rich form. Both stoichiometric and electride phases strongly encapsulate hydride species, with charge analysis confirming H^- formation and showing enhanced encapsulation in the electride phase due to cage-confined electrons. Furthermore, increasing the electron concentration through Si doping at the Al site promotes additional hydride formation, providing further insight into the hydrogen storage capability of C12A7-based materials

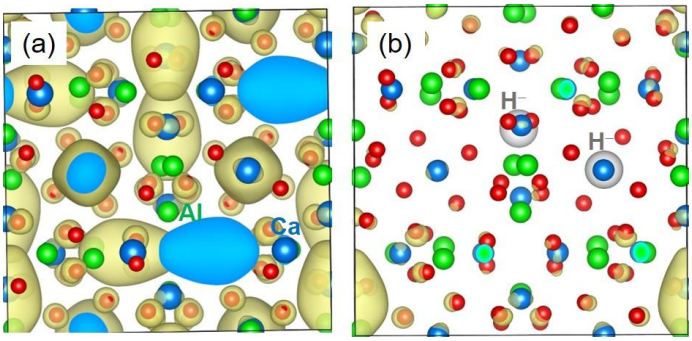

Fig. 1. Electron density distribution of C12A7 electride (a) before and (b) after the formation of H^- ions

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QM/MM Investigations of Defects in MgO and La_2CuO_4 and Other Related Materials

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The defect chemistry of magnesium oxide has been extensively studied for many years, with a myriad of applications in catalysis, optoelectronics and many other fields. Both vacancies and substitutionals have been shown to be effective in providing MgO with additional useful properties. Our work employs modern hybrid quantum mechanical – molecular mechanical (QM/MM) techniques, using the ChemShell code[1], to both reassess previously investigated defects and probe others that have been less researched. The use of QM/MM methodologies, and specifically with ChemShell, has previously been shown to be valuable for investigating defects in energy materials, for example in the cases of SrO[2], CeO_2[3] and GaN[4].

We present investigations of Cu-doped MgO, in particular the behaviour of hole states in the material. This work has been completed alongside complementary experimental EPR data on the material. The classical F-centres in MgO and the formation Schottky pair defects are revisited with our methods, as well as unconventional oxygen configurations around cation vacancies. Preliminary modelling of superconducting La_2CuO_4 has also been completed, in conjunction with development of an interatomic potential for open-shell transition metal ions, that will allow for complete replication of the materials structure within the QM/MM framework.

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Rational Design of Sulfur-Terminated MXenes for efficient CO_2 Conversion into Carbon Neutral Methanol Production

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The global rise in atmospheric CO_2 concentrations has become one of the most pressing environmental challenges, demanding urgent strategies for carbon mitigation and sustainable energy production. Electrocatalytic CO_2 reduction into value-added fuels offers a promising pathway toward carbon-neutral energy technologies, yet achieving selective and energetically favorable multi-electron conversion remains a major challenge. In this context, MXenes have emerged as promising catalytic platforms because of their tunable surface terminations, chemically active transition metal sites and strong capability to optimize adsorbate-surface interactions. Here, we present first-principles calculations of the catalytic conversion of CO_2 into CH_3OH over sulfur-terminated MXenes (M_2CS_2; M = Sc, Ti, V, Cr, Y, Mo, Mn, Zr, Nb, and W) to identify efficient catalysts for selective methanol production. The reaction pathways reveal that the catalytic activity strongly depends on the stabilization of reaction intermediates, thereby governing the thermodynamic feasibility of the multistep hydrogenation process toward CH_3OH formation. Notably, Ti_2CS_2 and V_2CS_2 emerge as promising catalysts owing to their reduced limiting potentials and favorable reaction energies, whereas W_2CS_2 shows overly strong binding of oxygenated intermediates that limits catalytic turnover. The thermodynamic analysis further identifies the hydrogenation of *COOH to *HCOOH and the subsequent conversion of *CHO to *CHOH/*CH_2O as key elementary steps that strongly influence the overall reaction pathway. Charge density difference analysis of the key *CHO intermediate reveals pronounced charge redistribution between the adsorbate and the MXene surface, promoting efficient electron transfer. Competing hydrogen adsorption calculations further confirm suppressed HER activity, highlighting their strong selectivity toward CO_2 reduction over hydrogen evolution. Overall, sulfur-functionalized MXenes emerge as efficient electrocatalytic platforms for selective CO_2 to methanol conversion, offering fundamental insights for the rational development of next generation two-dimensional catalysts for sustainable fuel generation.

The Pressure-Dependent Surface State of Cobalt during Fischer-Tropsch Synthesis

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Fischer-Tropsch synthesis (FTS) plays an important role in the global transition to sustainable fuels, particularly in hard-to-decarbonise sectors such as aviation and shipping. These industries account for over 5% of global CO_2 emissions and require high-energy-density liquid fuels. By converting syngas (a mix of H_2 and CO) derived from renewable feedstocks into long-chain hydrocarbons, FTS offers a scalable route to sustainable synthetic fuels. Commercially, cobalt nanoparticles (Co NPs) are the catalyst of choice in FTS due to their high selectivity for liquid paraffins under industrial conditions (200-250°C, 20-40 bar). Despite its importance, the nature of the catalytically active species and the role of surface structure remain unresolved. Metallic cobalt is commonly regarded as the active phase, although oxidic Co phases and Co carbide have also been reported to display catalytic activity too. Understanding how the surface of this catalyst evolves under reaction conditions is therefore of extreme importance. Most surface-sensitive techniques operate under vacuum or near-ambient-pressure conditions, far removed from the multi-bar pressures of many industrial reactions. This longstanding “pressure gap” obscures key surface intermediates and chemical states that are stabilised under reaction conditions, impeding efforts to resolve active species and establish reliable structure-activity relationships.

In this talk I will report on our recent efforts to bridge this critical pressure and materials gap, through the implementation of a high pressure *operando* soft X-ray platform that unlocks surface-sensitive studies of NP catalysts at near-industrially relevant pressures. Using a graphene-sealed microreactor compatible with both electron yield X-ray absorption spectroscopy and X-ray photoemission spectroscopy, we resolve the surface chemical state of Co NPs under the FTS reaction at pressures up to 4 bar. This enables the identification of a previously unobserved Co-carbonyl intermediate, stabilised under elevated pressures. We use density functional theory to calculate the energetics of terminated Co surfaces with different degrees of surface roughness and carbonyl coverage to further elucidate on adsorption-induced restructuring. Through this we demonstrate that higher pressures lead to adatom migration, surface restructuring, and link Co-carbonyl formation to pressure-driven restructuring of the NP surface.

Updates to Phonopy-Spectroscopy: First-Principles Modelling of infrared and Raman spectra

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P.-R. Dawson, M. Molinari and A. R. Pallipurath
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Infrared (IR) and Raman spectroscopy are routine materials-characterisation tools, but the spectra of complex materials can be difficult to interpret. We have developed a comprehensive workflow for predicting infrared (IR) and Raman spectra using density-functional theory (DFT), which we have implemented in our open-source Phonopy-Spectroscopy code.

Phonon frequencies and eigenvectors are obtained within the harmonic approximation. IR spectra are modelled by computing the mode dipole oscillator strengths and infrared dielectric function, providing access to absorption, reflectivity and transmission spectra, as well as related properties such as the static dielectric constant. Raman spectra are obtained by computing the Raman activity tensors from one of several methods, including the possibility to use e.g. time-dependent DFT (TD-DFT) to capture resonance effects. The code can also perform "on the fly" non-analytical corrections and can optionally include calculated per-mode linewidths.

The code can perform IR and Raman simulations on single crystals and powders, accounting for the orientation of the crystal and the polarisation of the incident and detected radiation. This allows for the simulation of a wide variety of measurements, spanning routine laboratory characterisation to the more sophisticated experiments possible with high-end or bespoke instrumentation. The code has also been designed to “decouple” the preparatory DFT calculations and spectrum modelling, to better facilitate collaboration between theory end experiments.

We have benchmarked the code for a wide range of applications including inorganic photovoltaic absorbers, pharmaceutical molecular solids, organometallic coordination polymers, materials for nuclear waste disposal, alloys, and systems with defects.

Recent developments include a new and cheaper approach to modelling Raman spectra, an updated approach for simulating IR spectra, including "off-axis" measurements, and tools for characterising phonon modes and for working with defective systems.

Field-Free Coexistence of Skyrmions and Anti-Skyrmions Induced by Higher-Order Interactions and Biaxial Strain in the NiI_2 Monolayer

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Department of Chemistry, University College London *nano-**sok***

The spin spiral (SS) state in monolayer (ML) NiI_2 presents a promising avenue for the exploration of two-dimensional multiferroics, but the mechanisms underlying this SS state and its effective modulation remain insufficiently understood. In this study, we employ first-principles calculations to reveal the major features and physical mechanisms governing the magnetism of ML-NiI_2 under in-plane strain, explicitly considering the effects of higher-order interactions (HOIs), which have been overlooked in prior research. Our findings identify the magnetic ground state of ML-NiI_2 as an SS state characterized by a propagation vector q = 0.23 and demonstrate that in-plane biaxial strain serves as an effective method for tuning the period and stability of the SS state, the frustrated ratio, magnetic anisotropy energy and the strength of HOIs. Notably, the strength of HOIs significantly increases under higher strain, particularly for the four-site-four-spin exchange interaction, which is substantial and crucial to the properties of ML-NiI_2. Additionally, through atomistic spin dynamics simulations, the coexistence of skyrmions and anti-skyrmions with diameters of at least 2 nm is achieved in ML-NiI_2 via in-plane biaxial strain, without the necessity for an external magnetic field, which is more conducive to spintronic applications. The presented results establish ML-NiI_2 as a prospective candidate for next-generation spintronic devices and contribute to a deeper understanding of HOIs in two-dimensional magnets.

Machine Learned Potentials for Li Based Reactive Hydride Composites

Oliver Morrison*, David Grant, Martin Dornheim, Sanliang Ling

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This work investigates lithium-based reactive hydride composites (RHCs) using machine-learned interatomic potentials (MLPs). RHCs are promising solid-state hydrogen storage materials because reactions between hydrides can reduce hydrogen desorption temperatures while improving reversibility and kinetics [1]. Since hydrogen desorption in RHCs is diffusion-controlled, understanding lithium transport is important for improving hydrogen storage performance [1,2].

High-accuracy MLPs were developed using the MACE (Message Passing Atomic Cluster Expansion) framework [3] to study lithium transport in Li–Mg–B–N–H systems. The training dataset contains tens of thousands of density functional theory (DFT) reference configurations from ab initio molecular dynamics across multiple stoichiometries, phases, temperatures (400–1000 K), and defect structures relevant to lithium migration. The resulting MACE potential achieves near-DFT accuracy, with force predictions reaching a root mean squared error of 41 meV/Å on unseen test data. This enables nanosecond-scale molecular dynamics simulations of Li-based RHCs at a fraction of the cost of direct DFT simulations. Using these machine-learned interatomic potentials, we perform numerous long-timescale molecular dynamics simulations, uncovering lithium migration pathways and diffusivities across a range of compositions.

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First-Principles Investigation of Band Alignment and Defect-Induced Electronic States at 2D–3D Perovskite Interface

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2D/3D hybrid perovskite architectures have attracted increasing interest due to their potential to combine high environmental stability with excellent optoelectronic performance. However, several critical band-structure issues—such as band misalignment, quantum-well-induced band bending, deep-level defect formation, and weakened electronic coupling—remain insufficiently understood at the interface and limit further efficiency improvement. In this work, we employ density functional theory (DFT) to investigate the electronic structure of the $(CH_3(CH_2)_3NH_3)PbBr_4$ (PEA-$PbBr_4$) / $CsPbBr_3$ perovskite interface. Particular attention is given to Br vacancies, which introduce deep trap states and are known to directly suppress open-circuit voltage in bromide perovskite devices

Our results show that the wide bandgap and strong quantum-confinement characteristics of PEA-$PbBr_4$ cause pronounced conduction- and valence-band offsets relative to $CsPbBr_3$, creating potential barriers for charge extraction. The insulating nature and low dielectric constant of the PEA organic spacer weaken wave-function overlap across the interface, leading to localized band flattening and reduced carrier mobility. Defect calculations further reveal that Br vacancies at the interface introduce deep trap states with energies on the order of a few hundred meV, promoting non-radiative recombination and contributing to open-circuit-voltage losses.promote non-radiative recombination and contribute to open-circuit-voltage losses. We also explore how variations in interfacial atomic arrangement and organic-chain orientation could be leveraged to mitigate band discontinuities, suppress defect states, and establish more favorable charge-transfer pathways.

Probing Nuclear Extremes in Lithium Ceramics: Multiscale Simulations with Machine-Learnt Interatomic Potentials

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*enviro-**cil***

Fusion fuel systems, particularly breeder blankets, demand advanced materials capable of withstanding high temperatures, intense neutron flux, and evolving chemical environments. Lithium-bearing ceramics are leading material candidates for breeder blankets, but, as is typical for solid state nuclear materials, their response to irradiation and solute transport properties are difficult to study. This is compounded by the challenging experimental conditions of neutron irradiation and tritium generation.

Here, in collaboration with the UKAEA on their flagship LiBRTI project, we present the deployment and refinement of machine-learned interatomic potentials (MLIPs) for tritium-bearing lithium metatitanate (LTO). We curate a high-fidelity DFT database of pristine, defective, and tritiated structures, also including amorphous and off-stoichiometric configurations to ensure model fidelity of MLIPs at nuclear extremes. Using these MLIPs, we perform equilibrium and non-equilibrium molecular dynamics (MD) simulations to estimate the evolution of LTO under irradiation and the transport of tritium therein. These simulations inform parameters for our finite-element tritium-transport models (TTMs). We investigate how the crystal lattice and granular microstructure respond to radiation damage and tritiation, thereby predicting effects on macroscopic operating performance and informing future research directions in the LiBRTI programme.

Our MLIP MD simulations reveal qualitative differences between MLIPs and classical empirical potentials while accessing regimes beyond the reach of ab-initio methods, contextualised by our mesoscale TTMs. By integrating these predictions into the broader multiphysics pipeline, we demonstrate the importance of model fidelity and multiscale reach for the prediction of material performance under reactor conditions.

Molecular Architecture and Oligomerisation as Determinants of Micelle Structure

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Covalent oligomerisation of surfactant monomers fundamentally reshapes micellar self-assembly, yet the structural consequences of linking repeat units into a polymer backbone remain poorly understood relative to their monomeric counterparts. Using molecular dynamics simulations of eight surfactant systems, which span monomeric, dimeric, mixed, linear, glucoside-based, and polymerised architectures, we show that oligomerisation drives a pronounced shift toward aspherical aggregate morphologies, as seen in the contrast between Tyloxapol and its monomer analogue Triton X-100. In designed binary mixtures, intermediate structural behaviour emerges when tail-unit parity is preserved, revealing that local packing geometry rather than molecular stoichiometry governs micelle shape. Headgroup chemistry exerts a strong secondary influence: the glucoside amphiphile C18GA exhibits an anomalous odd-even alternation in tail order absent in the linear analogue C12E6, indicating tight coupling between headgroup stereochemistry and chain conformation. These findings establish molecular architecture as the primary lever controlling soft-matter self-assembly, with direct implications for the rational design of surfactant systems in pharmaceutical and environmental contexts.

Contributed Mini Talks

Phonon and Thermal Transport Effects of Extended Defects in Reduced γ-WO_3 via Machine Learned Interatomic Potentials

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Tungsten oxides are receiving increasing interest as functional materials for a range of applications including catalysts for water splitting, Li-ion batteries, thermoelectric, optoelectronic and electrochromic devices. The stable stoichiometric monoclinic phase, γ-WO_3, upon reduction shows a range of complex defects including line defects. These defects can influence phononic behaviour and thermal transport, which are critical in determining the performance, efficiency and thermal stability of WO_3 based electronic and energy devices. However, investigating the influence of these extended defects on vibrational and thermal transport properties using accurate ab initio methods such as density functional theory (DFT) remains prohibitive due to the large spatial and temporal scales required. To combat this, we leverage Machine learned interatomic potentials (MLIPs) which can learn atomic environments at near DFT accuracy while enabling simulations at a fraction of the computational cost.

In this work, we trained a MACE MLIP for WO_3 systems containing line defects, with configurations generated from the MACE-MATPES foundational model and sampled across a range of temperatures and applied strains, and labelled using DFT. We demonstrate the capability of this potential to predict vibrational spectra in both the harmonic limit via finite difference methods and the anharmonic regime via molecular dynamics simulations (MD) for different types of line defects. We are further able to compute their thermal conductivities using the Green Kubo formalism, obtaining trends and values in good agreement with experimental measurements. Finally, we show that this potential can be extended to grain boundaries and their reduced structures, where the structural units are similar to those observed in line defect systems, enabling successful screening and relaxation of candidate structures and accurate prediction of experimentally observed configurations.

Towards modelling vibration-assisted hot-carrier processes in metallic nanoparticles

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Metallic nanoparticles (NPs) stand as ideal photocatalysts for inducing chemical reactions in adsorbates [1]. Under irradiation, plasmon modes can become excited on the NP surface. Their decay through Landau damping leads to the formation of hot carriers (electrons/holes) which, when transferred to the adsorbates present on the NP surface, lead to a change in the activation barriers [2,3]. In this context, the interaction between hot carriers and the NP vibrational modes is usually perceived as a hindrance to the catalysis. Nevertheless, because they can scatter electrons and holes between different bands, vibrations may actively modify the generation and energy distribution of hot carriers, thus opening different reaction routes. In this regard, the evaluation of electron-vibration interactions is essential. In this work, we perform a tight-binding parametrisation of the electronic band structure of Ag and Au featuring an atomic arrangement that is slightly distorted from equilibrium [4]. The tight-binding model used is constructed by fitting Slater-Koster parameters to density functional theory calculations carried out on non-ideal bulk structures such as strained crystals [5]. We then check whether such a model can describe the electron-vibration coupling accurately by direct comparison with ab-initio calculated electron-phonon matrix elements and the Eliashberg spectral functions.

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Oxygen scavenging at a SiO_2/Ta interface: a *first-principles* study

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Resistive random-access memory (ReRAM) devices have emerged as promising candidates for advanced memory technologies. These devices offer high scalability, high endurance and fast programming speed. Typically, ReRAM consist of a metal oxide sandwiched between two metallic electrodes. Application of a small voltage induces the formation and destruction of a conductive oxygen vacancy filaments in the oxide, enabling switching between high (HRS) and low resistive states (LRS)[1]. As oxygen vacancies are central to the working principle of ReRAM devices, a complete understanding of the formation of such defects at the metal/oxide interface is essential. In this project, we investigate from *first-principles* the interface between silicon dioxide (SiO_2) and tantalum (Ta). Tantalum has shown excellent ability to promote the formation of oxygen vacancies at the metal/oxide interface by extracting oxygen from the oxide in a process called scavenging[2]. To better understand this exchange, we examine two types of models: **full-interface models**, consisting of a Ta(110) slab on a SiO_2 surface, and **cluster models**, consisting of small Ta_n clusters adsorbed on a SiO_2 substrate. Our goal is to evaluate the transfer of oxygen from SiO_2 to Ta at different stages of film growth. Here, the full-interface models represent a fully formed film, while the cluster models correspond to the initial stages of film growth. For each model, we explore the formation of oxygen vacancies in the SiO_2 substrate and the incorporation of the displaced oxygen into the Ta, either slab or cluster. We investigate this oxygen-transfer process considering its energetic and kinetic aspects. Our findings show that the process is energetically favourable only for the cluster models, suggesting that spontaneous scavenging is likely to initiate during the early stages of film growth. At the same time, however, each model indicates the existence of an energy barrier associated with the breaking of Si-O bonds. With this computational analysis, we aim to shed light on the atomistic processes during a possible scavenging event at the interface between the metal oxide and the metallic electrodes.

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Oxygen-Driven Evolution of Novel Σ5 Tilt Boundaries in Anatase TiO2 by in-situ eSTEM/TEM

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Hydrogen production via photocatalytic water splitting is vital for achieving net-zero targets, but the efficiency of promising TiO_2 catalysts is limited by large bandgaps and rapid electron-hole recombination. Understanding defect structures at the atomic level is prudent to design effective catalysts. Using in-situ environmental scanning transmission electron microscopy (eSTEM/TEM), we induced electron beam reduction and recrystallisation of anatase TiO_2 nanoparticles (NPs) under an O_2 background, complemented by hybrid density functional theory (DFT) simulations. STEM imaging at 550°C captured the dynamics of newly formed reduced TiO_2 phases and grain boundaries. The reduced phase was identified as a having Ti_2O_3 stoichiometry, forming reproducibly along anatase {011} planes due to electron beam and thermal effects. This reduction is fully reversible upon oxygen exposure, yielding grain boundaries dominated by Σ5[100](012) tilt boundaries (TBs). These TBs have not been previously reported, and this study provides the first insight into their atomic and electronic structure. Hybrid DFT calculations, employing 10.5% Fock-exchange to maintain Koopman's compliance in TiO_2 [1, 2], were performed to assess the electronic structure of the Σ5 TB. Calculations were also performed on the TiO_2-Ti_2O_3 boundary to assess its stability. In the Σ5 TB, we demonstrate that hole-polaronic trapping occurs readily on interfacial oxygen sites, which also yield the most stable structures upon oxygen removal. These insights lay the foundation for future studies of controlled extended defects in TiO_2 NPs while demonstrating the power of in-situ eSTEM for creating and analysing grain boundaries at the atomic scale within a single NP.

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Effects of surface nanostructure of metal co-catalysts for photocatalytic conversion of methane: a theoretical study

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Photocatalysis offers an efficient way to drive thermodynamically and kinetically challenging chemical reactions, such as water splitting, CO_2 reduction, and conversion of organic molecules. To achieve high efficiencies, photocatalysts are typically combined with co-catalysts, such as noble metals, to promote separation of photogenerated electrons and holes and provide active sites for reactions. Co-catalysts are usually few-nanometres-sized nanoparticles. Theoretical modelling typically represents co-catalysts either as two-dimensional flat surfaces, or as very small clusters containing few atoms. In this work, we used a more realistic theoretical representation of a metal co-catalyst as a nanostructured surface containing defects, and explored the effect of these defects on the model reaction of methane dissociation as the first step of photocatalytic conversion of methane.

We modelled palladium, one of the most commonly used noble metal co-catalysts, using density functional theory calculations. The co-catalyst was represented as a Pd (111) surface containing defects, such as various arrangements of Pd adatoms and vacancies. We found that clusters of adatoms or vacancies are preferred to single defects. Further, we modelled the mechanism and energetics of methane adsorption and dissociation on the flat and defect-containing surfaces. We found that adsorption of the products (dissociated methyl and hydrogen) was slightly more energetically favourable than adsorption of methane, and that single adatoms particularly favoured adsorbed methyl and hydrogen compared to methane. Furthermore, single adatoms and vacancies and pairs of adatoms or vacancies reduced the activation barrier for dissociation of adsorbed methane by up to 0.6 eV. Thus, this study shows that presence of adatoms and vacancies on noble metal co-catalyst surfaces is beneficial for the first step of catalytic conversion of methane, both kinetically (by reducing the energy barrier) and thermodynamically (by stabilising the dissociated product).

Impact of the Presence of Au on Activity and Selectivity of Cu/ZnO Catalysts for Methanol Synthesis

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Cu-based catalysts for the synthesis of methanol from syngas ($CO/CO_2/H_2$) are firmly established in industry, in particular the alumina-supported Cu/ZnO system (CZA), which has been used successfully for over fifty years. Despite this success, however, the precise nature of the active catalytic phase and the origin of catalytic activity remain poorly understood. In addition, recent work has demonstrated the potential promotional impact of the presence of Au on ZnO supported metal nanoparticle catalysts. It was found that the supported monometallic Au/ZnO catalyst was found to exhibit high selectivity to methanol, but low CO_2 conversion, whilst the Cu/ZnO system was found to exhibit higher CO_2 conversion but poorer methanol selectivity. The AuCu/ZnO system, meanwhile, exhibited both superior CO_2 conversion rates and methanol selectivity. This suggests a bifunctional mechanism with the Cu component enhancing CO_2 conversion rates, and the Au component directing methanol selectivity. As such, it is of interest to investigate the mechanism of methanol formation on representative Cu/ZnO surface models, as well as the impact of dilute Au on key elementary reaction processes, in addition to developing highly representative models for supported stoichiometric AuCu alloy clusters.

In the present work, plane-wave DFT techniques are applied to investigate the impact of the dilute presence of Au on the mechanism of CO_2 hydrogenation to methanol on a highly representative model for a small Cu cluster supported on a reconstructed polar ZnO surface. The calculations showed that substitution of a single Cu atom for Au did not significantly affect CO_2 adsorption and activation, however higher activation barriers were reported for subsequent hydrogenation processes such as for formate formation compared to the Cu/ZnO model. Conversely, methoxy hydrogenation was found to be less energy-demanding when Au is included. Additionally, unbiased Monte Carlo global optimization techniques were applied to identify low-energy structures for 1:1 stoichiometric AuCu clusters supported on a ZnO slab, with partial segregation of the Au species to the lower coordinate sites.

Nitrogen Evolution Reaction Study on Ru Supported Clusters and Ru(0001) surface

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Ammonia decomposition is a promising route for carbon-free hydrogen production, addressing key challenges in hydrogen storage and transport. The nitrogen evolution step is widely recognised as the rate-limiting process, particularly at low temperatures. In this work, we employ density functional theory (DFT) calculations to compare the behaviour of Ru clusters supported on carbon with that of an extended Ru(0001) surface. We focus on how structural and electronic properties influence nitrogen adsorption, reaction pathways, and transition states associated with N–N recombination.

Our analysis shows that low-coordinated, sub-nanometre Ru clusters generate a distinct adsorption environment that facilitates nitrogen exchange between the metal cluster and the carbon support, thereby modifying nitrogen binding and activation relative to nitrogen adsorption on terrace sites of extended Ru surfaces. The Ru15/C cluster exhibits stronger binding to atomic nitrogen than the extended surface does, significantly reshaping the reaction energy profile for N_2 evolution. In particular, the energetic barriers for N–N recombination and N_2 desorption differ markedly between the cluster and the extended surface.

These results demonstrate that reduced coordination and local electronic structure in supported Ru clusters alter the activation barrier for nitrogen recombination, highlighting the carbon support's active role in the catalytic process. Overall, the adsorption energetics, reaction pathways, and transition state analysis provide a mechanistic understanding of how supported Ru clusters tune the kinetics of the nitrogen evolution step in ammonia decomposition and suggest that nitrogen incorporation into the carbon support may be promoted at low temperatures.

Investigating the Formate Pathway for CO_2 to MeOH over Pd/ZnO Catalysts

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As the effects of climate change become increasingly evident, strategies to utilize rather than just reduce greenhouse gases are becoming more compelling than ever. The method under investigation here is the use of palladium/zinc oxide catalysts to hydrogenate the greenhouse gas carbon dioxide into methanol,[1] which is in demand for its applications as a fuel, solvent or platform chemical.

Selectivity towards methanol is greatly influenced by the interaction between the palladium metal and zinc oxide support. Lawes *et al.* demonstrated a markedly improved MeOH selectivity ratio at higher ratios of Zn:Pd than the 1:1 ratio required to form a PdZn alloy.[2] It is possible that use of ZnO as a support provides a Zn reservoir for in-situ generation of the active catalytic phase,[3] as well as the metal oxide aiding anchoring oxygen-containing adsorbates at the catalyst/zinc oxide interface for direct hydrogenation.[4]

The current research uses the all-electron FHI-aims code, using a 'light' basis set and a k-grid of $3 \times 3 \times 1$, studying Pd_{10} adsorbed on a 5×5 $ZnO(000\bar{1})$ slab. It aims to investigate the formation of a local Pd/Zn alloy, using hydrogen spillover to reduce surface Zn species. (Figure 1) This allows the comparison of the impact of this site change on the energetics of key reactant intermediates in the hydrogenation of CO_2 to methanol.

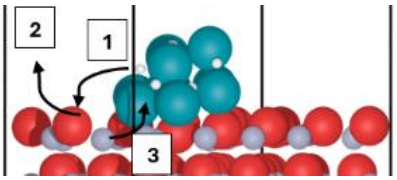

Figure 1: Surface reduction of ZnO(000^{-}1)

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Vibrational Spectra of Zirconolite

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The UK government has committed to disposing of the 140 tonnes of legacy Pu currently held at Sellafield in a geological disposal facility (GDF), and this is likely to remain the preferred disposal route for future waste materials. Zirconolite is one of two main candidate materials for development as a disposal waste form for encapsulating Pu.

One on the major concerns of geological disposal is tracking wasteform corrosion over the extended lifetime of the active materials, to ensure environmentally-toxic actinide species remain contained. Non-destructive remote monitoring using vibrational spectroscopy is a potentially attractive option for *in situ* monitoring. However, for this to be effective requires a comprehensive understanding of the vibrational spectra of the "host" material, and the spectral signatures associated with the doped forms and potential corrosion products.

In this work, we take the first steps toward this by using a bespoke modelling workflow developed in the group, based on density-functional theory (DFT), to perform a complete characterisation of the infrared (IR) and Raman spectra of Zirconolite. We aim to build on this to create a library of reference spectra to support the characterisation of unknown samples, which could ultimately enable tracking the corrosion of wasteforms *in situ* and targeted early interventions to prevent the release of Pu into the environment.

Correlation of Polarons and Highly Charged Interstitials in Metal Oxides

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Defects play a central role in determining the electronic and functional properties of materials, particularly in applications such as microelectronics, solar cells, and batteries. The characterisation and analysis of defect charge states is key to understanding charge trapping, charge transfer and defect-related reactions, which are critical factors in performance and reliability.

In this work, we use density functional theory (DFT) to investigate nitrogen and carbon interstitial defects in hafnium oxide (HfO_2), an important dielectric material, and compare their behaviour across a set of technologically relevant oxides: silicon dioxide (SiO_2), gallium oxide (Ga_2O_3), and zinc oxide (ZnO). These materials span a range of electronic structures and applications, providing a broad basis for identifying trends in defect behaviour.

Many oxides undergo nitridation, which can passivate intrinsic defects and modify electronic behaviour. We investigate the configurations of nitrogen interstitials, which form covalent N-O bonds up to the singly negative charge state. However, this bond can be broken, with nitrogen forming stable configurations up to the triply negative state. To understand the origin and stability of these highly charged states, we further examine how additional electrons are accommodated within the lattice across all four oxides.

Mechanisms enabling localisation of high charge are identified, including lattice distortions that accommodate additional electrons within the interstitial region. These distortions resemble those associated with self-trapped polarons present in stoichiometric HfO_2, suggesting a correlation between polarons and ion interstitials. Crucially, this correlation is shown to hold across the oxide set: oxides capable of supporting electron polarons - such as HfO_2 and SiO2 - permit nitrogen to reach higher charge states, while those that cannot, such as SiO_2 and ZnO, do not exhibit this behaviour.

These findings highlight the interplay between polaronic effects and interstitial defects, and illustrate how an oxide's ability to localise polarons acts as a key indicator of its capacity to stabilise highly charged interstitial defects. This work offers a unifying framework for understanding nitrogen passivation and charge trapping across oxide dielectrics, with broader implications for the design and engineering of defect-tolerant materials.

Multiscale Modelling of Grain Boundaries and Extended Defects in Halide Perovskites Using Machine-Learning Potentials and DFTB

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Metal halide perovskites (MHPs) are promising materials for photovoltaic and optoelectronic applications, but their performance and stability are strongly influenced by microstructural imperfections, including grain boundaries, defects, stoichiometric imbalance, and impurity phases. Therefore, establishing structure-property relationships at the atomic scale is essential.

In this study, I will present a computational workflow for investigating perovskite grain boundaries and extended defects by combining density functional theory (DFT), density functional tight binding (DFTB), and machine-learning interatomic potentials (MLIPs). This work builds on the development of efficient DFTB parameters for iodide and bromide perovskites, including 3D and 2D materials, mixed-halide systems, and perovskite heterostructures. These parameters were designed to enable reliable structural relaxation and electronic-property prediction at a lower computational cost than conventional DFT, while maintaining good agreement with reference data for lattice parameters, band gaps, and effective masses.

Here, I further extend this approach to more complex defective systems. DFT calculations are used to generate reference data for training MLIPs, enabling efficient exploration of grain-boundary structures, defect configurations, and local reconstruction mechanisms. After benchmarking against DFT for representative grain-boundary systems, DFTB is then applied to evaluate the corresponding electronic properties, including changes in band-edge states and possible trap formation. By linking accurate first-principles calculations, scalable atomistic modelling, and efficient electronic-structure analysis, this work aims to provide a practical multiscale framework for understanding how grain boundaries and defects control the stability and optoelectronic response of MHPs.

The Monte Carlo Threshold Algorithm in Crystal Structure Prediction: Introducing Intramolecular Degrees of Freedom

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Traditional crystal structure prediction (CSP) workflows are a global optimization problem, where we aim to find the lowest energy packing of a molecule in a crystal. CSP workflows generate a large number of minima in the potential energy surface (PES), and the general assumption is that the lowest energy structure in that set is the most likely thermodynamically stable packing for the chosen molecule. In practice, this is not true, due to many reasons: crystallisation conditions, impurities, and effects of finite temperature and pressure to the relative energy ranking of different packings. Better predictions of what crystal packing a molecule will take can be obtained by considering more information than just the local minima in the PES, such as the energy barriers separating them.

Our group proposed the use of the Monte Carlo threshold (MCT) algorithm [1] to obtain more information from the crystal packing PES [2, 3]. The MCT algorithm can obtain an estimation of the energy barriers between local minima in the PES. We have previously shown how the data obtained through the MCT algorithm can be used to rationalise the appearance of different polymorphs of three small polycyclic aromatic hydrocarbons [4]. In that study, we treated the molecular conformations as rigid, but for many systems of interest this assumption might not hold. Extending the method to include flexible molecules would allow the exploration of molecules relevant for industrial applications (eg. pharmaceuticals). We have decided to investigate the effectiveness of the MCT algorithm when intramolecular degrees of freedom can't be ignored, starting by applying the method to the ROY molecule. This molecule has 15 known polymorphic forms [5, 6], therefore investigating whether the MCT algorithm can provide insights on the reason of this high degree of polymorphism will be very valuable.

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Single- and Dual-Atom Catalysts for the CO_2 Reduction Reaction

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In the last 50 years alone, CO_2 emissions have more than doubled, and the impact on our environment has been considerable. Global temperatures have risen considerably, affecting our ecosystems, wildlife and food stocks, among countless other facets of our planet. With current technology, it is not possible to completely eradicate the use of fossil fuels. There are, however, existing methods under investigation to mitigate CO_2 emissions, known as carbon capture, use, and storage (CCUS).

The focus of this work is on the CO_2 reduction reaction (CO_2RR), in which CO_2 is converted into other useful chemicals, such as syngas (CO and H_2). Some catalysts that have been considered in other works include metal oxides, organometallic compounds, and metal-organic frameworks. This study, however, focuses on single-atom and dual-atom catalysts (SACs and DACs, respectively), in which isolated or paired atoms are embedded in a support. More specifically, we are considering a graphene support with transition metal (TM) adatoms.

While SACs are limited by linear scaling relationships and lower metal loading, DACs are emerging as promising candidates. DACs may have synergistic effects, distinct electronic structures, and chemical flexibility, potentially allowing them to outperform SACs. Before investigating DACs, we studied SACs to understand how a single TM affects CO_2RR performance. Investigating the electronic and mechanistic properties helps narrow down the database of TMs considered for DACs, which, together with analysis of stability, catalytic performance, and possible reaction pathways, enhances understanding of these new catalysts

Reliable and Reproducible Benchmarking of DFT Codes on GPUs: A CASTEP Case Study

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Modern HPC architectures are increasingly heterogeneous, relying more heavily on accelerators, or GPUs, to push performance into the exascale. DFT codes are often optimised for CPU-only performance, though many codes have taken active steps to explore the potential for accelerating DFT through offloading computations that are more suited to GPU parallelisation, a particularly effective technique for large scale materials simulations.

Despite GPU offloading boasting potential speedups of orders of magnitudes, such developments often not readily embraced users. To bridge this gap, we are collaborating with the SHAREing project[1] to demonstrate the effectiveness of periodic planewave DFT performed on GPUs by designing a benchmarking suite comprised of realistic simulations designed to show the types of simulations that will benefit most from GPU acceleration. These benchmarks will be used to deliver a worked example of how to reproducibly prepare a DFT code for evaluation by the SHAREing benchmark procedure. High quality, independent benchmarking data from SHAREing can then be exploited by the DFT community to ensure that they are getting optimal performance from any HPC cluster.

We are working with the planewave DFT code CASTEP.[2,3] Using CASTEP as a testbed, we will develop benchmark runs and compilation instructions to ensure rigorous and fair testing of the code by SHAREing. Benchmarking inputs will be made available to the DFT and condensed matter physics communities, giving developers a worked example of how to ready their code for benchmarking. These benchmarks will be critical in directing the architecture of future HPC resources.

In this presentation, we will discuss the reliable and reproducible preparation of benchmarks for DFT codes, using CASTEP as a worked example, and the requirements such preliminary benchmarks and transferable build procedures must satisfy for the SHAREing procedure. The discussion and dissemination of optimal compiler and library combinations for given hardware will be of particular use to the MCC community to ensure that optimal performance on every HPC facility is achieved.

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DL-FIND: Benchmarking Methods for Geometry Optimisation

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Geometry optimisation tasks contribute a significant part of the computational effort for many chemical investigations. DL_FIND is a geometry optimisation library, used by ChemShell and available as a standalone library. We report on recent developments.

Firstly, we benchmark an optimiser based on using Gaussian Process Regression (GPR) [1] to build up a surrogate energy surface. This method demonstrates competitive performance with the standard L-BFGS method but requires significantly more memory. The use of a multi-level representation of the GPR, to reduce the memory requirements, is investigated.

Secondly, a modification to the L-BFGS method [2], where the memory of past steps is reset when an unphysically large step is suggested by the optimiser, is tested and found to produce a modest performance improvement.

Thirdly, we investigate reaction barrier calculations using the Nudged Elastic Band (NEB) method. Improved initial guesses of the reaction path using the Image Dependent Pair Potential (IDPP) [3] and Geodesic Interpolation [4] are compared. We also benchmark the energy-weighted (EW) extension to the Nudged Elastic Band method (EW-NEB) [5]. Taken together, these features improve the stability and performance of NEB calculations.

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Implementing Extendable Workflows for QM-in-QM Embedding

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Post-Hartree-Fock (HF) methods accurately describe the total energies of chemical systems at computational costs that scale poorly with system size. Hierarchical quantum mechanics in quantum mechanics embedding (QM-in-QM) allows one to balance between computational cost and accuracy. This compromise is achieved by describing a small region of chemical interest with a high accuracy electronic structure method, while the wider environment is represented as an embedding potential evaluated with a more computationally tractable method such as DFT.

At present, implementing embedding workflows into electronic structure software is complicated by the monolithic design of most packages. As many of the algorithms supporting QM-in-QM embedding are code agnostic, much of the duplicated development effort can be avoided by abstracting embedding specific functionality to an external wrapper.

Our talk presents a modular and open-source approach that extends QM-in-QM embedding to electronic structure codes. The Pythonic wrapper, EmbASI, controls the embedding workflow at the high-level and performs tasks such as subsystem partitioning, molecular orbital localisation, and embedding potential construction. The development effort for QM driver is limited to the integration of the Atomic Simulation Interface (ASI) Application Programming Interface (API) into the host software package, which controls the import and export of key data quantities, such as density and Hamiltonian matrices.

The efficacy of the workflow is demonstrated by implementing the projection-based embedding theory (PBET) scheme into the FHI-aims electronic structure software package. We show the significant compute time reductions enabled by QM-in-QM embedding for highly accurate post-Hartree-Fock methods with negligible reduction in energetic accuracy.

Effects Of Isomerism on Porous framework materials: Window Edition – How Does Isomerism Affect A Guest Molecules Ability To Enter A Pore

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Molecular Framework Materials are porous crystalline materials. Combined with high tunability these materials have a wide range of applications from gas storage to separation. One of the techniques used to investigate these materials is High Throughput Computational Screening (HTCS), HTCS allows researchers to screen a large library of structures for a particular application.

However, HTCS faces several notable limitations. Many implementations rely on low level of computational theory typically force field. Furthermore, some studies use already synthesized structures, essentially repurposing structures rather than new discoveries. An Additional constraint is that for sake of computational resources cage structures are often constrained to be rigid, which in turn fails to consider both the flexibility and isomerism that may be present.

We present our results showing how linker functionalization and isomerism affect the ability of guest molecules to enter a pore.

Posters

A Density Functional Theory study on the Point defects in PuO_2

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bulk-**mol**

Vibrational spectroscopy, particularly Raman, is an important technique when applied to plutonium dioxide (PuO_2), as it can provide valuable insights into a given sample ageing history as radiation defects develop over time. To underpin this spectroscopic feature of PuO_2, we are simulating vibrational spectra of PuO_2 containing defects.

Factor group analysis of the space group of PuO_2 ($Fm\bar{3}m$, face-centred cubic; O_h^5 point group) predicts the observation of one Raman active phonon (T_{2g}), experimentally observed at ~480 cm^{-1}, however alpha decay and formation of defects can cause the T_{2g} band to appear at lower wavenumbers, broaden and decrease in intensity. Radiation damage in PuO_2 generates new peaks in the Raman spectrum, such as those seen in the experimental literature in the 530-660 cm^{-1} region, attributed to the formation of various defects. These include various point defects, anionic Frenkel pairs, cationic Frenkel pairs and alpha helium. The Raman-inactive mode (1LO2) is thought to become Raman active through symmetry lowering and lattice disorder and appears in the region ~580 cm^{-1}, possibly following the formation of Frenkel oxygen defects generated through alpha decay.

Computational modelling of this subject provides a more fundamental understanding of the self-irradiated PuO_2 Raman spectra. This study employs Density Functional Theory including spin orbit interaction to investigate the vibrational fingerprint of pure and defective PuO_2, focusing on the IR and Raman signatures. The defects investigated in this study are various point defects, due to their relevance in the experimental literature.

Atomistic Simulation of Liquid Electrolytes on Argyrodite Surfaces for Hybrid Solid-Liquid Lithium Batteries

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Hybrid solid-state batteries have the potential to reduce two major shortfalls of solid-state batteries, namely, the surface contact between electrodes and electrolyte and the susceptibility for stress fractures within the electrolyte, without diluting the improved safety and resistance to dendrites that they possess over current liquid electrolyte-based lithium-ion batteries. The proposed design includes a small amount of liquid electrolyte around the electrodes to increase wettability of the electrolyte-electrode contact and give space for expansion without compromising the solid electrolyte. However, this presents a new interaction not present in other structural designs, the solid-liquid electrolyte interface (SLEI), which may dominate the bulk ionic transport and in essence the viability of any design.

Seeing where and how different liquid electrolytes interact in the formation of an SLEI on an atomic scale can grant us valuable insight into the SLEI and how to select appropriate compatible electrolytes to maximize functionality. As such we have been using DFT and ab Initio molecular dynamics to simulate likely primary interaction sites and start to observe the formation of the SLEI through time, as well as make predictions about the stability of the overall interface and ionic conductivity with multiple differing electrolytes. The relative unlikeliness of these interactions in other designs outside of hybrid systems means that relatively little analysis and simulations have been done and each simulation is populating a sparce yet potential vital database that will be critical for subsequent researchers to be able to predict interactions in any choice of electrolytes.

We are working with the Spencer-Jolly group at Birmingham who are undertaking experimental analysis on different hybrid systems and the resultant SLEI formations, which will allow us to gauge the accuracy of any simulations we make while providing them predictions of the interactions on a scale that is impossible to observe. Our simulations primarily focus on Li6PS5Cl argyrodite as the solid electrolyte, due to its excellent Li-ion conductivity, with liquid electrolytes simulated against this being EMIM/TFSI:LiTFSI and DOL/DME:LiTFSI to provide a selection of different organic and inorganic liquid electrolytes from which to gather any comparisons between.

Using DFT Simulations to Investigate Ammonia Conversion to Hydrogen on Transition-Metal Based Catalysts

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The transition to low-carbon energy systems depends critically on efficient energy conversion and storage technologies. Hydrogen is often highlighted as a promising energy carrier because of its high gravimetric energy density and environmental friendliness, but its practical storage and distribution remains challenging [1]. At ambient pressure, hydrogen has a very low volumetric energy density of about 0.01 MJ L^{-1}, and even when compressed to 700 bar this only increases to roughly 5.8 MJ L^{-1} [2], these limitations are unfavourable in terms of financial cost and safety for storage and transportation. Ammonia is emerging as a promising hydrogen carrier, because it can be readily liquefied under a moderate pressure of 10 bar at room temperature to achieve high gravimetric and volumetric energy densities of 22.5 MJ kg^{-1} and 13.9 MJ L^{-1} [3]. Ammonia could be easily transported using the existing transportation structure to applications sites and convert to hydrogen via thermochemical or electrochemical processes. Ammonia could also be directly used in combustion to generate heat and energy. Catalysts play an important role in all these conversion processes. Transition metal-based catalysts are in the spotlight because they are more abundant and cost effective than platinum group metals (PGMs), and have shown comparable performance to PGMs.

In this work, DFT simulations are used to investigate reaction pathways and thermodynamics of the electrocatalytic ammonia oxidation reaction (EAOR) on different Cu-based surfaces. Our collaborators at Cranfield University have experimentally introduced oxygen vacancies into Cu_2O and have provided the electrocatalytic performance results to corroborate this work. Oxygen vacancy free (OV-free), oxygen vacancy rich (OV-rich) Cu_2O (111) and Cu (111) surfaces are used to calculate adsorption energies of relevant NH_x* (x=0-3) intermediates and free energies of elementary dehydrogenation steps leading to conversion of ammonia to nitrogen and hydrogen. OV-rich Cu_2O stabilises all intermediates more favourably than OV-free Cu_2O however, both surfaces have a similar ΔG_{max} involving the final elementary step forming N* (rate-determining step). From experimental characterisation (XPS) of OV-rich Cu_2O, a high enough ratio of Cu^0 was observed to suggest some of the reaction may be happening on local clusters of Cu metal and it is the case that ΔG_{max} is reduced significantly on Cu.

In parallel, thermochemical ammonia cracking is simulated using DFT on Cu (111) and Ni (111) surfaces, which are used to elucidate adsorption energies, activation energies and rate coefficients of elementary steps. These values will be used to guide a larger workflow involving a collaboration with University of Durham that includes CFD, ammonia combustion and catalyst synthesis.

Acknowledgements: MMM Hub for computational resources, which is partially funded by EPSRC (EP/T022213, EP/W032260, EP/P020194) and UK-HyRES project (EP/X038963).

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Computational Modelling of Photocatalytic Conversion of Organic Molecules into Biofuel

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Biofuels are promising alternatives to fossil fuels for achieving global net zero targets. However, converting sustainable precursors such as long chain organic acids found in waste cooking oils requires highly selective pathways to avoid harsh, energy intensive reaction conditions. Photocatalysis offers a solution, using sunlight to drive the decarboxylation of organic acids into alkanes. Our experimental collaborators at the University of York have developed a $BiVO_4$-based photocatalyst, which is able to drive this decarboxylation process.[1]

This work describes our density functional theory modelling of the reaction mechanism and electronic behaviour of the decarboxylation process on the most stable $BiVO_4$ (001) surface. We use acetic acid as a model alkanoic acid, and co-adsorbed water as a solvent. We find that acetate binds strongly to the catalyst surface through dissociative adsorption, whereas water interacts weakly. Following decarboxylation, the resulting methyl radical exhibits a strong affinity for surface oxygen atoms, while the CO_2 binds very weakly, facilitating product release. Crucially, by simulating photoexcitation via a triplet-state, we observed a dramatic decrease in both the activation barrier and reaction energy (ΔG) compared to the electronic ground state. These initial findings will allow us next to investigate the dimerisation of alkyl radicals (methyl in this case) to form longer-chain alkanes.

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Computational Study of Sodium-Ion Migration and Defect Chemistry in Na_8SnP_4-Based Solid Electrolytes

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All-solid-state sodium batteries are attractive alternatives to lithium-ion technologies because of the natural abundance and low cost of sodium. A key challenge is identifying solid electrolytes that combine high Na^+ conductivity with sufficiently low electronic conductivity. Recently, pristine Na_8SnP_4 was reported as a promising sodium phosphide ion conductor, with its disordered high-temperature polymorph showing significant Na^+ mobility.[1] However, the relatively narrow calculated electronic band gap suggests that electronic leakage should also be considered when assessing its suitability as a solid electrolyte.

Here, we use first-principles calculations to examine the relationship between Na-ion transport and defect-controlled electronic leakage in Na_8SnP_4. Phonon calculations confirmed the dynamical stability of the ordered reference structure, while *ab initio* molecular dynamics and site analysis of the 500 K trajectory reveal repeated Na exchange between tetrahedral and octahedral sites, supporting a connected Na-ion hopping network. Representative nudged elastic band calculations were used to examine local migration pathways.

To assess electronic leakage, charged Na vacancy and Na interstitial defects were calculated in a 104-atom supercell. The resulting defect thermodynamics show that Na Frenkel-type defects can pin the Fermi level away from the band edges, giving low room-temperature electronic carrier concentrations under intrinsic Na-defect-dominated conditions. However, carrier concentrations increase strongly at elevated temperature, indicating that Na_8SnP_4 lies close to the boundary between ionic and mixed ionic–electronic conduction.

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Exploring the Defect Chemistry of Potassium Tantalate

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Potassium tantalate (KTO) is a wide bandgap, transparent material which demonstrates suitability for a wide range of applications, with recent years garnering a particular interest in its use for quantum technologies (i.e. spintronics).[1,2] Other notable recent work has achieved significantly high mobilities for *n*-type conductivity, reaching values of 100 V s cm^{-2}, indicating applicability for fields such as transparent thermoelectrics.[3] Few first principles studies have explored the nature of the intrinsic defects present in the system, or the fundamental properties which might contribute to the high mobility seen in experimental works; hence the origin of these characteristics should be further evaluated.

This study aims to evaluate the electronic structure of KTO using first principles techniques, as well as investigating the intrinsic defects present, to provide a foundational framework for future investigations. As defect states are defined with respect to the valence and conduction band edges, it is important to accurately model the electronic structure of the pristine material, to proceed with the defect calculations.[4] The evaluation is conducted using hybrid level functionals (for accuracy) and relativistic effects (i.e. spin-orbit coupling) are incorporated.

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A Computational and Experimental Study of the Methanol Synthesis catalyst

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Methanol is produced globally at an estimated 98 million tons per year.[1] The reaction uses syngas ($H_2/CO/CO_2$) as the starting materials and operates over a $Cu/ZnO/Al_2O_3$ catalyst at mild reaction conditions.[2] While the reaction pathways are well characterised, there are uncertainties regarding the complex states and interactions of hydrogen with the Cu component. In this work, we performed Density Functional Theory (DFT) based quantum chemical calculations using the Vienna Ab-initio Simulation Package (VASP) to understand the interactions of H atoms on the Cu(111) surface. We used the non-local vdW-DF2 by Lee et al.[3] exchange-correlation functional, along with the projector augmented wave (PAW) method. The surface was modelled using a 4 x 4 supercell with 5 atomic layers and a theoretical lattice constant 3.746 Å. A 3 x 3 x 1 k-point grid was used, and the plane-wave cutoff energy was set to 475 eV.

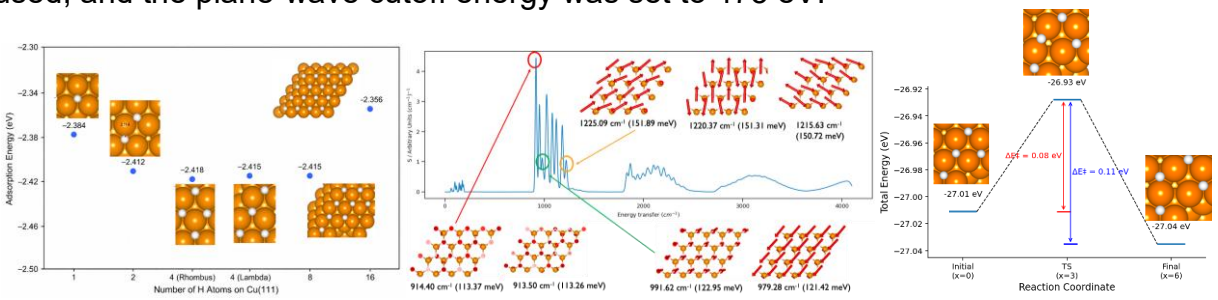

Figure 1: (Left) Adsorption energy per H atom change as coverage of Cu(111) surface increases (Middle) Phonon simulated INS spectra of the full monolayer coverage system. (Right) Diffusion energy barrier of single hydrogen atom in the presence of three nearest-neighbor adsorbed hydrogen atoms.

Results in fig.1 (left panel) of the low surface coverage of the fcc sites, on the Cu(111) surface, show the adsorption energy becoming more exothermic, suggesting increasing stability potentially due to cooperative stabilization effects. At increased coverages, however, stability decreases due to H-H lateral repulsions. (Middle) shows the simulated Inelastic Neutron Spectroscopy (INS) spectra of the full monolayer coverage system. The fig. shows the highlighted H atoms vibrational modes and combinational regions within the 850 cm^{-1} and 1300 cm^{-1} region. (Right) Depicts the diffusion energy barrier of a single hydrogen atom diffusing from an fcc site to an hcp, in the presence of three fixed fcc adsorbed hydrogen atoms. The result shows that the bridge site is a true transition state rather than an adsorption site, at low coverage. Additionally, the hydrogen atom tends to diffuse relatively quick, with a low energy barrier of 0.08 eV in the forward path. The aim of the project is to combine theoretical results with experimental INS data to deduce the interaction and state of hydrogen on Cu during the methanol synthesis process.

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Amorphous-like Thermal Conductivity and High Thermoelectric Figure of Merit in "π" SnS and SnSe

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The tin chalcogenides SnS and SnSe are an important family of optoelectronic materials with applications in, among other technologies, photovoltaics (PV), thermoelectric (TE) generators and photodetectors. In particular, SnS has been widely studied as a potential PV material, and orthorhombic *Pnma* SnSe is one of the current flagship high-performance TEs. The recently-discovered π-cubic phases of SnS and SnSe have similar structure coordinations to the *Pnma* phases and have been shown to be both dynamically stable and close to the convex hull. However, the TE performance of these structures at various temperature and carrier concentration conditions is largely unexplored.

We performed first-principles simulations to calculate the TE properties and figure of merit *ZT* of p- and n-type doping of π-cubic phases of tin monochalcogenides. The n-type doping of π-cubic SnS and SnSe was predicted to have a high *ZT* of 2.19 (860 K) and 2.83 (740 K) due to their low thermal conductivity κ and comparatively high electrical conductivity σ, which have better TE performance than their corresponding *Pnma* phases. Although the π-cubic phases with p-type doping have inadequate σ, their ZT_{max} were able to exceed 1, which makes them plausible matches for the industrially operated TE material e.g. Bi_2Te_3. Our results indicate that the n-doped π-cubic SnS and SnSe could be good substitutes for their *Pnma* phases in the thermoelectric technologies.

Investigating Vacancy Defects in a Candidate Transparent Conducting Oxide, $LaSb_3O_9$

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Transparent conducting oxides (TCOs) are materials which are both optically transparent and have metal-like conductivity. This unique combination makes them very useful, with a wide range of applications such as solar cells, smart windows and touch screen devices. [1] Despite this broad range of applications, industry currently relies on a small number of materials. Sn-doped In_2O_3 (ITO) is the most commonly employed, offering an excellent combination of low resistivity and high conductivity.[2] However, there are concerns over the long-term sustainability of this practice due to high costs and a scarce supply of indium. Therefore, finding new TCOs consisting of more earth-abundant elements is essential. Recently, Sb(V)-oxides have emerged as promising n-type TCO candidates due to their wide band gaps, favorable band alignments and high conductivity under appropriate doping.[3–6] $LaSb_3O_9$ was selected to extend this class of Sb(V)-oxide TCOs, due to its connected 3D network of SbO_6 octahedra, which is known to promote highly mobile electrons. Electronic structure and transport properties were calculated using hybrid density-functional theory, revealing a wide band gap of 4.3 eV and high electron mobility. However, intrinsic vacancy calculations show that compensating La vacancies remove the doping window, preventing sufficient carrier concentrations for n-type conductivity. This further highlights defect compensation as a key limitation in wide band gap oxides and provides guidance for the future design of Sb(V)-based TCO materials.

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Pressure Driven Evolution of Various Metal Hexaborides

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Metal hexaborides, MB_6, are a group of ionic crystals with large covalent bonding networks possessing a unique set of thermal, electrical, magnetic and physical properties. Thus, they have the potential for real world technological applications including thermionic emitters due to their low work functions [1], and as ceramics due to their hardness [2]. To date there are three known pressure-induced phase transitions for the hexaborides, one of which has been experimentally verified.

As such, it is important to understand effects underpinning the bonding and stability of the boron-framework. Building on previous work, which showed cation size is directly related to the transition pressure [3], I provide a comprehensive outline on the effects of changing the metal cation (M = Ca, Ba, Y, La, Th) on the phase transitions. It is shown that, alongside cation radius, valence electron count and d/f-orbital M-M and M-B hybridization contribute to the stability and drive towards certain phases. The use of the hybrid functional HSE06 over PBE is also shown to be important in accurately predicting phase transitions, the removal of "ghost" phases (ThB_6) and correctly predicting the electronic properties (semiconducting vs semimetallic behaviour) of the hexaborides.

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First-Principles Investigation of Band Alignment and Defect-Induced Electronic States at 2D–3D Perovskite Interface

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2D/3D hybrid perovskite architectures have attracted increasing interest due to their potential to combine high environmental stability with excellent optoelectronic performance. However, several critical band-structure issues—such as band misalignment, quantum-well-induced band bending, deep-level defect formation, and weakened electronic coupling—remain insufficiently understood at the interface and limit further efficiency improvement. In this work, we employ density functional theory (DFT) to investigate the electronic structure of the $(CH_3(CH_2)_3NH_3)PbBr_4$ (PEA-$PbBr_4$) / $CsPbBr_3$ perovskite interface. Particular attention is given to Br vacancies, which introduce deep trap states and are known to directly suppress open-circuit voltage in bromide perovskite devices

Our results show that the wide bandgap and strong quantum-confinement characteristics of PEA-$PbBr_4$ cause pronounced conduction- and valence-band offsets relative to $CsPbBr_3$, creating potential barriers for charge extraction. The insulating nature and low dielectric constant of the PEA organic spacer weaken wave-function overlap across the interface, leading to localized band flattening and reduced carrier mobility. Defect calculations further reveal that Br vacancies at the interface introduce deep trap states with energies on the order of a few hundred meV, promoting non-radiative recombination and contributing to open-circuit-voltage losses.promote non-radiative recombination and contribute to open-circuit-voltage losses. We also explore how variations in interfacial atomic arrangement and organic-chain orientation could be leveraged to mitigate band discontinuities, suppress defect states, and establish more favorable charge-transfer pathways.

Computational Study of Thorium-Containing Host Materials for use in Thorium-229 Nuclear Clocks

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A nuclear clock has the potential for time-keeping orders of magnitude more accurate than current atomic clocks. Thorium is an excellent candidate as a nuclear clock material because the thorium-229 nucleus has a unique low-energy excited state of 8.4 eV. A suitable solid-state nuclear clock material that contains thorium-229 must have a band gap larger than 8.4 eV to avoid competing electronic transitions [1,2]. Accurately predicting the band gap of thorium-containing host materials is a major obstacle that still needs to be overcome for thorium-229 nuclear clocks to become a reality, and calculating these band gaps computationally remains a significant challenge.

In this project I investigate the use of the modified Becke–Johnson exchange potential combined with DFT+U (MBJ+U) as a computationally efficient method for calculating the electronic structure of thorium-containing materials. Currently very expensive GW methods are considered the benchmark for these types of calculations, but GW calculations are not feasible for many of the most promising nuclear clock candidate materials such as those with large supercells. I show that a specific configuration of MBJ+U provides accurate band gap prediction for a range of simple thorium-containing materials in agreement with GW calculations. I then use this same MBJ+U configuration on more realistic thorium-229 nuclear clock candidate materials for which GW calculations are not possible.

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Benchmarking the Energy Ranking of Molecular Crystals at the Meta-GGA Level

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Accurate prediction of relative polymorph stability remains a central challenge for modelling molecular solids. Dispersion-corrected density functional theory (DFT-D) methods, particularly those using the exchange-hole dipole moment (XDM) dispersion, have established a reliable benchmark for crystal energy rankings. However, the computational cost of these approaches, especially when using hybrid functionals, motivates the search for computationally efficient alternatives that retain comparable accuracy.

In this poster, we assess the performance of the modern meta-GGA functional r^{2}SCAN with the rVV10 dispersion correction for polymorph energy ranking across a diverse set of molecular crystals. All candidate structures are taken from prior literature reports and have already been fully optimised at the B86bPBE-XDM level of theory, enabling a controlled comparison of energy models independent of structural relaxation effects.

The energy ranking obtained from single-point periodic DFT calculations at the r^{2}SCAN+rVV10 level are compared against that obtained using B86bPBE-XDM and B86bPBE0-XDM. Particular attention is given to systematic trends across different classes of materials, including rigid molecules and flexible molecules, and multi-component crystals. The results will help to determine whether r^{2}SCAN-based approaches can offer a computationally efficient, broadly applicable alternative to established hybrid DFT-D methods for polymorph prediction of molecular crystals.

Computational Investigation of CO_2 Methanation over Ni and Ni-Alloy Catalysts

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Carbon dioxide (CO_2) methanation is a promising route for carbon utilisation and renewable fuel production in power-to-gas (PtG) applications. Nickel-based catalysts are widely used owing to their high activity and low cost compared with noble-metal catalysts. However, a detailed understanding of the reaction mechanisms governing CO_2 hydrogenation and the influence of catalyst modification remains essential for the rational design of improved catalytic systems.

Density functional theory (DFT) calculations were performed using the Vienna Ab initio Simulation Package (VASP) with the BEEF-vdW and PBE-D3 functionals to investigate CO_2 methanation on low-index nickel surfaces. Surface energies of Ni(111), Ni(110), and Ni(100) were calculated, confirming Ni(111) as the most stable facet. The adsorption energies of key species, including CO_2, H_2, CO, and O, were subsequently evaluated to examine surface reactivity and adsorption behaviour. Following surface screening, detailed mechanistic studies were carried out on both pure Ni(111) and single Mn-doped Ni(111) surfaces. The complete reaction network for CO_2 hydrogenation was studied through reaction energy calculations, including the redox, formate, and carboxyl pathways associated with the reverse water-gas shift reaction (RWGS), as well as the C hydrogenation, HCO hydrogenation, and COH hydrogenation pathways that lead to methane formation. Activation barriers for the initial CO_2 and H_2 dissociation steps were also calculated to provide insight into the early stages of the reaction mechanism.

The comparison between pure and Mn-doped Ni(111) surfaces enables assessment of how a single Mn dopant influences adsorption energetics, reaction thermodynamics, and potential reaction pathways. The results contribute to a molecular-level understanding of CO_2 methanation on Ni-based catalysts and provide a foundation for ongoing transition-state calculations and subsequent kinetic modelling studies to identify improved catalyst formulations for efficient CO_2 conversion.

Periodic DFT Investigation of CO_2 Hydrogenation on Clean and Fe-Doped Ni(111) Surfaces

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CO_2 hydrogenation on Ni-based catalysts is of interest for carbon utilisation and the formation of value-added products, but the surface-level role of metal composition in CO_2 activation and early hydrogenation steps remains important to clarify. In this work, periodic spin-polarised density functional theory calculations are used to investigate CO_2 hydrogenation on clean and Fe-doped Ni(111) surfaces. The clean Ni(111) surface is first established as a reference model after comparing the relative stability of low-index Ni surfaces. Adsorption of key species, including CO_2, H_2, CO, O and H, is then examined at different surface sites using the PBE-D3 method, allowing both adsorption energies and structural changes of the adsorbed species to be analysed.

The clean Ni(111) calculations provide a baseline for understanding how molecular adsorption, C–O bond elongation, CO_2 bending and H–H bond cleavage depend on surface site. Dissociation-energy calculations are used to assess the energetic feasibility of forming surface CO, O and H species, while activation barriers for selected elementary steps are calculated to distinguish between thermodynamic favourability and kinetic accessibility. Fe-doped Ni(111) models are then introduced to examine how the dopant changes local adsorption geometry, adsorption energy and reaction energetics compared with the clean surface.

By combining adsorption, dissociation and activation-barrier calculations, this study compares the early CO_2 hydrogenation pathway on clean and Fe-doped Ni(111) surfaces. The results provide insight into whether Fe incorporation can modify the interaction between CO_2-derived intermediates and the Ni surface, and whether it can promote key elementary steps involved in CO_2 activation and hydrogenation.

A Study of H_2O-Promoted CO_2 Coupling to Ethanol on the Cu(100) Surface

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Carbon dioxide (CO_2) emissions are a major contributor to climate change, making efficient CO_2 utilisation an important environmental and industrial goal. Among the possible products from CO_2 hydrogenation, ethanol is particularly attractive as both a liquid fuel and a valuable chemical feedstock. However, under conventional thermocatalytic conditions, CO_2 hydrogenation mainly produces C_1 products such as methanol, CO, or methane, while selective ethanol formation remains challenging.

Recent studies have shown that the introduction of water vapour can enable CO_2 conversion to ethanol over Cu-based catalysts with high selectivity. This suggests that water may influence the reaction pathway by modifying surface structure, stabilising key intermediates, or changing the activation barriers of elementary steps.

In this project, density functional theory calculations using VASP will be employed to investigate CO_2 and syngas hydrogenation toward ethanol formation on Cu-based surfaces. The study will focus on key C_1 intermediates, CHO decomposition, methanol-related pathways, and C–C coupling steps. Transition states will be searched using NEB and dimer methods to obtain activation barriers and compare different reaction pathways. The role of water and reconstructed Cu-based surfaces will also be explored to understand their influence on ethanol selectivity and to provide theoretical guidance for improved catalyst design.

Modelling Charge Carriers in Cuprate High-Temperature Superconductors

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Cuprate high-temperature superconductors are known for their high critical superconducting temperatures (T_c) which is of great interest to many researchers. Whilst the materials have been extensively researched experimentally, the superconducting mechanism still has not been explained theoretically. Doping is a key step in introducing charge carriers into the materials that pair and become superconducting. A series of doping methods has been established and the present project the present simulations using shell model interatomic potentials in GULP and the Mott–Littleton method to model defect and hole states in cuprate superconductors. Recent work reproduces defect formation energies in La_2CuO_4 and YCBO, as well as modelling polaronic structures in La2CuO4.

QM/MM Calculations for ZnO ($10\bar{1}0$) and the CO_2 Adsorption

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Au/ZnO is a typical photocatalytic CO_2 reduction catalyst. To further understand the reaction mechanism, CO_2 reduction process simulated with low coverage is needed, which could be close to the real situation. ZnO tends to be defective with oxygen vacancies (Vo) on the surface, which could be an effective CO_2 activation site, but the property is sensitive to the concentration, should be simulated in a diluted concentration to reflect the real condition. Besides, the system should be able to access the excitation state.

Herein, quantum mechanics/molecular mechanics (QM/MM) method is applied to ZnO, with a high accuracy QM region embedded in a forcefield representation of the extended surface. ZnO ($10\bar{1}0$), a non-polar surface chosen as the substrate, oxygen vacancy energy (E(Vo)) of the surface will be calculated to confirm the precision of electronic structure of the surface. On ZnO surface, CO_2 adsorptions with different configurations are calculated. To obtain high exposed QM region with good convergence property, a QM partition mode is proposed, which is non-stoichiometric, rectangular, and with at least 2 neighbor ions in QM region to largely remove the dangling bonds.

Modelling Excited States of MOFs NU-1000 and NU-901 Using Cluster and Periodic Approaches

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Excited state properties of metal-organic frameworks (MOFs) are of increasing interest due to their applications as photocatalysts and fluorescence sensors. Many modelling techniques have been employed to understand these processes; however, challenges arise due to their large unit cells and heavy transition metals. When coupled with the high computational cost of accurate excited state calculations, significant bottlenecks can arise. This work compares cluster and periodic-based approaches for two MOF systems, NU-1000 and NU-901, to investigate the effects of model choice and topology on absorption spectra. Simulated spectra for both methods were found to largely match the absorption bands found in the organic ligand of NU-1000 and NU-901, H_4TBAPy. However, neither model replicates the broadness found in the low energy region of the experimental spectra. This indicates that some broadening mechanisms are not well represented by the calculations. In future work, embedded QM:QM' cluster models could combine high accuracy calculations on a chosen region with the long-range interactions found in the extended network, giving a better description of the solid-state environment.

Machine-Learned Structure Prediction of TiO_2 Clusters Using Fine-Tuned MACE Potentials

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Titanium dioxide (TiO_2) is an important, semiconducting material with various applications in catalysis, solar cells, disinfection and coatings.[1] In particular, TiO_2 nanoclusters have attracted significant interest due to their increased catalytic activity compared to the bulk phase.[2] The properties of TiO_2 nanoclusters strongly depend on their atomic structure, which significantly deviates from the bulk structure and varies with the size of the cluster.[3] Unlike the bulk structure, the TiO_2 cluster structures remain a subject of ongoing research. Experimental determination of small cluster structures is extremely difficult, as single cluster sizes are hard to isolate, and conventional diffraction methods are limited by the absence of periodicity in the structure. Previous computational studies have shown that empirical interatomic potentials are unable to capture subtle energetic preferences between different coordination environments,[4] whereas DFT calculations become computationally expensive as the number of atoms increases. To address these limitations in structure prediction of TiO_2 clusters of different sizes, we employ machine-learned interatomic potentials.

Specifically, we build upon the pre-trained, multi-head MACE-MH-1 foundation model,[5] which was originally trained on a diverse set of inorganic materials. By fine-tuning this foundation model, we improve the prediction accuracy for TiO_2 clusters. The fine-tuning dataset comprises approximately 200 hybrid DFT (PBE0) calculations of diverse TiO_2 cluster structures, covering cluster sizes of up to 12 formula units (36 atoms). In particular, the training targets during the fine-tuning process are the energies and forces of the corresponding clusters. The resulting model aims to provide a computationally efficient yet accurate description of the energetics and structures of TiO_2 clusters, enabling more extensive investigations of their structure – property relationship.

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Machine Learning Study of Charge Compensation Mechanisms in Zr-Doped ZnO at low doping Concentration

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Zinc oxide (ZnO) is a wide bandgap semiconductor whose n-type conductivity can be improved by doping. Zirconium (Zr) is outstanding among tetravalent dopant candidates; compared to trivalent dopants, it provides one additional electron while maintaining an ionic radius similar to that of Zn^{2+}. However, experimental observations show inconsistent trends in electrical and optical properties with varying doping concentration, as well as conflicting bandgap behaviour. As a result, the charge compensation mechanism remains unexplained at the atomic level for low doping concentration (below 10%).

This study aims to analyse the mechanism, whether dominated by atomic defects (zinc vacancies or O interstitials) or by electronic redistribution, by combining hybrid density functional theory (DFT) with machine learning interatomic potentials. Defect configurations (Zr substitutes Zn site, with either a V_{Zn} or O_i) are generated by using a random sampling approach followed by symmetry degeneracy check with the spglib Python package to remove equivalent structures in 2x2x2 supercells. The pre-trained model MACE-MH-1 is used for rapid pre-optimisation and energy ranking. Candidates are then validated by geometry relaxation and single-point calculations using PBE and PBE0 in the CRYSTAL code.

Initial results show that MACE-predicted energy rankings agree with PBE and PBE0 for Zr_{Zn} + V_{Zn} complexes and Zr_{Zn} + O_i complexes. Currently, O_i calculations (O in octahedral sites, tetrahedral sites, random-generated sites, and split interstitials) are in progress. PBE0 convergence remains challenging. PBE-relaxed structures are considered for PBE0 single-point calculations. The next steps will explore 3x3x3 supercells and employ MACE-driven Monte Carlo and global optimization. This report paves the way for future investigations aimed at refining and further comparing the performances of different methods to identify the dominant charge compensation mechanism in the Zr-doped ZnO system and thereby facilitate the development of efficient transparent conducting oxides.

Spin-Polarised DFT Study of Antiperovskite Materials for Lithium-Ion Battery Applications

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Antiperovskites have shown promise for use in lithium ion batteries as they use earth-abundant Fe, rather than the more expensive currently used cobalt, and they have potentially a greater charge density than the materials currently used. The aim of my project is to try and understand the structural and electronic features of the antiperovskites Li_2FeSO, Li_2FeSeO and Li_2FeTeO that may be important in their potential operations as cathode materials. I will do this using computational chemistry, specifically by carrying out spin-polarised DFT calculations on the three species mainly using the CRYSTAL 23 software package. The initial aims will be structural optimization of all three species, the determination of the Fe spin state (high-, low-, or intermediate-spin), how spins on adjacent Fe ions interact (ferromagnetic or antiferromagnetic), the structural and electronic effects of substituting S with Se and Te, and how Li deficiency affects the stability, magnetism, electronic properties, and the lithium ion migration pathways of the materials. It is also of interest to carry out calculations on $Li_2FeS_xSe_{(1-x)}O$ species and to compare their properties relative to pure Li_2FeSO and Li_2FeSeO.The preliminary results obtained so far for all Li_2FeSO, Li_2FeSeO and Li_2FeTeO show clearly that the high-spin situation is energetically favoured, and the relatively large calculated band gap suggests this is an insulating state. The antiferromagnetic arrangement of spins is of lower energy than the ferromagnetic arrangement and appears to be anisotropic. Calculations involving looking at the structural and electronic effects of removing Li ions to create vacancies, and investigating the mixed $Li_2FeS_xSe_{(1-x)}O$ species are still in progress.

Sampling Defect Complexes in Beryllium Doped GaN Using a Machine Learning Interatomic Potential

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P-type doping in GaN is limited by compensation, passivation, and defect complex formation, which reduce the effective activation of acceptor dopants. Beryllium has been proposed as a promising alternative to magnesium because it may act as a shallower acceptor when occupying the gallium site. However, its behaviour in GaN can be complicated by interstitial beryllium, native point defects, and dopant clustering. This work investigates the configurational stability and compensation mechanisms of GaN doped with beryllium through an accelerated computational workflow combining density functional theory with machine learning interatomic potentials.

Defect complexes are generated in GaN supercells, including substitutional beryllium, interstitial beryllium, nitrogen vacancies, gallium interstitials, and nitrogen interstitials. Symmetry equivalent structures are removed, and the remaining configurations are screened using the MACE-MH-1 potential before validation with PBE and selected PBE0 calculations. This approach enables efficient identification of energetically favourable defect motifs and provides initial evidence for local defect association and beryllium clustering.

Building on these results, DFT calculations targeted to beryllium defect environments are being used to fine-tune the machine learning potential. The fine-tuned model provides a basis for Monte Carlo configurational sampling and global optimisation, allowing the most stable compensating complexes and clustering arrangements to be searched more efficiently than by first principles calculations alone. This framework aims to provide a systematic understanding of beryllium compensation in GaN and to guide future strategies for improving p-type doping efficiency.

Model Collapse under Iterative Self-Training of Generative Models

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Generative AI is transforming materials discovery, with projects like DeepMind's GNoME reporting the generation of millions of new crystal structures. These models are not only trained on existing experimental and computational databases but also keep expanding them with newly generated data. This rapid recursive training raises a critical question: Does the quality and diversity of the generated structures degrade when the models are continually trained on data of their own creation? In this work, preliminary experiments that form the first step towards addressing this question will be presented. Using the MP20 dataset and a transformer-based model, CrystaLLM, multiple iterations of a "generate–evaluate–retrain" scheme are implemented to examine how the diversity and novelty of generated structures evolve over successive generations. These findings provide an early insight into whether generative materials models remain sustainable when recursively trained on their own data.

Atomistic modelling of CoO_x/TiO_2 interfaces: Elucidating metal-support interactions in Fischer-Tropsch catalysts

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The use of transition metal catalysts in Fischer-Tropsch (FT) reaction, involving the conversion of syngas (CO and H2 mixture) to liquid hydrocarbons, is crucial in producing sustainable aviation fuels (SAFs).1 Cobalt-based metal oxides, dispersed on TiO_2 support, have proven to be highly effective precursors of metallic catalysts in FT synthesis, with Mn promoters enhancing the selectivity.1 The addition of Mn reduces nanoparticle size, enhances cobalt oxide dispersion across the support, and induces a phase transition from spinel Co_3O_4 to rocksalt CoO. Furthermore, at low Mn loadings, a preferential rocksalt CoO interfacial layer is observed between the TiO_2 support and spinel Co_3O_4 phase, indicating metal-support interactions between CoO_x (Co_3O_4 and CoO) and TiO_2. However, the nature of such interactions in the cobalt oxide catalytic precursor remains unknown.

In this study, we have used the universal machine-learned force field MACE-MP2 and density functional theory (DFT) to investigate the stability CoO_x/TiO_2 interfaces to have an atomic level understanding of metal-support interactions in cobalt-based FT catalysts. Periodic DFT calculations on the CoO_x/TiO_2 interface models were performed using the rSCAN exchange-correlation (XC) functional and 'light' basis set as implemented in the Fritz Haber Institute ab initio materials simulation (FHI-aims) package.3 Exhaustive interface model generation was performed using the Zur and McGill algorithm,4 which were then screened using MACE-MP to identify the most appropriate models. DFT calculations on the pre-screened models show that interfacial stability is higher for CoO/TiO_2 interface compared to Co_3O_4/TiO_2 interface. However, the presence of oxygen vacancy defects on TiO_2 surface enhances the interfacial stability of the Co_3O_4/TiO_2 interface. The oxygen vacancies on TiO_2 surface are stabilized by the movement of oxygen atoms from the Co_3O_4 phase across the interface. Furthermore, our calculations show that interfacial stability increases with increase in oxygen vacancy concentration on the TiO_2 surface. These insights lay the groundwork for future investigations into the effect of Mn-doping on interfacial stability. The atomistic understanding gained from this study provides valuable knowledge for rational catalyst design, enabling targeted modifications to enhance activity, stability, and selectivity in industrial FT processes.

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Time-Resolved Simulation of Shock-Driven Energy Redistribution in Crystalline Energetic Materials

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Shock-driven chemical reactions underpin the initiation, sensitivity, and performance of energetic materials.1 Despite their importance, the atomic-scale mechanisms that govern how a mechanical shock redistributes energy and leads to chemical activation remain poorly understood.2 This limits our ability to predict shock sensitivity, assess safety margins, and rationally design materials with tailored energetic behaviour.3

In this work, we develop a theoretical framework to simulate the vibrational response of crystalline materials to mechanical shock. Our framework focuses on the dynamics of kinetic energy redistribution among the vibrational degrees of freedom in a crystal via anharmonic phonon scattering. Using a time-resolved phonon population model informed by third-order force constants, we track how energy introduced impulsively into low-frequency vibrational modes propagates throughout the lattice. Specifically, we identify the vibrational modes into which kinetic energy is preferentially channelled. By exploring post-impact atomic dynamics under selective excitations informed by our model, we identify structural distortions that may act as microscopic precursors to bond rupture and shock initiation in crystalline energetic materials.

Our methods establish a new mechanistic link between mechanical excitation and chemical activation at the atomic scale. By resolving the earliest stages of shock-driven energy localisation, this work provides a foundation for predictive modelling of shock sensitivity, initiation mechanisms, and safer energetic material design.

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Mechanochemically Promoted CO_2 Methanation over Co-CeO_2 Catalysts via Collision-Induced Charge Effects

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CO_2 methanation is an important pathway for CO_2 utilisation and renewable-energy storage, but its low-temperature performance is restricted by the kinetic stability of CO_2 and the complex multi-step hydrogenation process. Mechanochemical activation has emerged as an effective strategy to enhance catalytic reactions, although its promotional mechanism is usually attributed to structural reconstruction and defect formation. Herein, density functional theory calculations were used to investigate the mechanism of mechanochemically promoted CO_2 methanation over Co-CeO_2. The Co-CeO_2 interface facilitates CO_2 activation and intermediate hydrogenation, while carbonate hydrogenation is identified as the rate-determining step. Extra-electron calculations further reveal that collision-induced charge transfer promotes interfacial electron redistribution and accelerates the key hydrogenation step, demonstrating a direct electronic contribution of mechanical energy to catalytic reaction kinetics.

Notes

Delegates @2026 MCC Conference: Daresbury Laboratory