Lennard-Jones Centre / CECAM Meeting 2025

From Electrons to Atoms to Molecules and Materials

Monday, 1st September to Friday, 5th September, 2025

Ray Dolby Centre, University of Cambridge

Full Programme

Monday, 1 September

8:30	—	9:15	**Registration**
9:15	—	9:30	**Opening remarks**
			AM Session Chair: Christoph Schran
9:30	—	10:15	**Marzari/Chiarotti**: Theory, algorithms, and applications of spectral functionals
10:15	—	10:30	**Li**: Density functional approximations guided by exact constraints
10:30	—	11:00	**Tea and Coffee Break**
11:00	—	11:45	**Burke/Daas/Crisostomo**: Fundamental Issues in DFT
11:45	—	12:30	**Mostofi/Maity**: Shining light on moiré materials: From atomic structure to optical excitation
12:30	—	13:30	**Lunch**
			PM Session Chair: Erin Johnson
13:30	—	14:15	**Tozer/Huynh**: Classical reaction barriers in DFT: An adiabatic-connection perspective
14:15	—	15:00	**Yang/Fan**: Development in functional approximations and in theory for excited states
15:00	—	15:30	**Tea and Coffee Break**
15:30	—	16:15	**Blum/Graf**: Pushing the boundaries of hybrid DFT based simulations for inorganic and hybrid semiconductors
16:15	—	16:30	**Lee**: CASTEP studies of NLO crystals: A 30 years journey
16:30	—	16:45	**Caussé**: Search for ternary superhydrides metastable at ambient pressure in the Y-Fe-H system: A new prototype structure
16:45	—	17:00	**Kumari**: In silico engineering of piezoelectric biomolecular assemblies
17:00	—	19:00	**Poster session and drinks reception**

Tuesday, 2 September

			AM Session Chair: Chuck Witt
9:00	—	9:45	**Perez/Vilhena/Trujilo**: Unraveling the potential of atomic force microscopy with DFT-based simulations and deep learning; Nanomanipulation dynamics of single molecules at surfaces; Accurate forces for CO tips on cobalt phthalocyanines on Ag(111)
9:45	—	10:30	**Car/Xie/Gao**: Ab initio bulk free energy surface of proper ferroelectrics; Modeling the solvated electron: Structure, reactivity, and new machine learning methods
10:30	—	11:00	**Tea and Coffee Break**
11:00	—	11:45	**Sprik/Zhang/Bui**: On the thermodynamics of crystals under hydrostatic pressure; Modelling of electrified solid-liquid interfaces with finite-field DFTMD; Liquid state DFT in the 21st century: Electrolytes, electromechanics and dielectrocapillarity
11:45	—	12:30	**Probert/Donaldson**: Finite temperature crystal structure prediction
12:30	—	13:30	**Lunch**
			PM Session Chair: Fabian Berger
13:30	—	14:15	**Cole/Moore**: DE-FF and MACE-OFF: Data-driven interatomic potentials for molecular simulations
14:15	—	15:00	**Tkatchenko/Kabylda**: Should I stay or should I go: From empirical potentials to machine-learned force fields in atomistic biomolecular dynamics
15:00	—	15:30	**Tea and Coffee Break**
15:30	—	16:15	**Reuter/Carbogno/Poths**: Next-level machine-learning interatomic potentials: Molecular dynamics with local electronic degrees of freedom; Relevance of catalyst restructuring under reaction conditions: An automated process explorer approach
16:15	—	16:30	**Bourne-Worster**: Tackling large embedded systems: environmental control in photosynthetic antennae
16:30	—	16:45	**Kavanagh**: Exploring defect energy surfaces with machine-learned foundation models
16:45	—	17:00	**Goodwin**: Exploring charge density waves in twisted bilayer $NbSe_2$ with machine learning
17:00	—	17:15	**Conference photo**
19:00	—	22:00	**Conference banquet at Christ's College**

Wednesday, 3 September

			AM Session Chair: Chris Pickard
9:00	—	9:45	**Norskov/Ghan**: First-principles study of heterogeneous catalysis: the spin promotion effect and revisiting electronic descriptors
9:45	—	10:30	**Marom/Carter-Fenk**: Computational discovery of new materials for singlet fission in the solid state; Single-Reference Linearized Coupled Cluster Theory for Strongly-Correlated Systems
10:30	—	11:00	**Tea and Coffee Break**
11:00	—	11:45	**Clark/Ravindran/Breeze**: Some new functionality in CASTEP, inverted densities and non-collinear DFT+U
11:45	—	12:30	**Haynes/Prentice/Xiao**: The ONETEP linear-scaling density functional theory program
12:30	—	13:30	**Lunch**
			PM Session Chair: Christoph Schran
13:30	—	14:00	**Alavi**: A brief history of the early days of Theoretical Chemistry in Cambridge
14:00	—	14:30	**Payne**: The rise of electronic structure calculations
14:30	—	18:00	**Excursions**

Thursday, 4 September

			AM Session Chair: Alex Thom
9:00	—	9:45	**Gross/Tu**: The concept of exact forces on the nuclei and other lessons from the exact factorization; Electronic decoherence along a single nuclear trajectory
9:45	—	10:30	**Hutter/Battaglia**: Quantum embedding of solid-state materials in the near-term quantum computing era
10:30	—	11:00	**Tea and Coffee Break**
11:00	—	11:45	**Galli/Somjit**: Electronic structure and coherent states of spin defects in solids
11:45	—	12:30	**Reining/Mohamed**: Excitonic effects beyond textbook models
12:30	—	13:30	**Lunch**
			PM Session Chair: Mike Payne
13:30	—	14:15	**Foulkes/Perez-Fadon**: Neural wave functions for quantum chemistry and materials physics
14:15	—	14:35	**Gori-Giorgi**: Accurate and scalable exchange-correlation with deep learning
14:35	—	14:55	**Lobato**: FEAT: Free energy Estimators with Adaptive Transport
14:55	—	15:15	**Muñoz Ramo**: Latest adventures on quantum phase estimation and quantum dynamics experiments on an ion trap quantum computer
15:15	—	15:45	**Tea and Coffee Break**
15:45	—	17:00	**Panel discussion**: Csányi (chair), Gori-Giorgi, Ivanov, Simperler, others TBA
17:00	—	19:00	**Networking event**

Friday, 5 September

			AM Session Chair: Angelos Michaelides
9:00	—	9:45	**Drummond/Johnson/Simula**: Quantum Monte Carlo methods and the CASINO program: Applications to positronic materials and atomic gases
9:45	—	10:00	**Agosta**: Hydrophobicity from molecular diffusion at the solid-liquid interface
10:00	—	10:15	**Park**: Deciphering the structural and electronic properties across the gold–water interface
10:15	—	10:30	**Govindarajan**: Multiscale modeling of electrochemical CO_2 conversion
10:30	—	11:00	**Tea and Coffee Break**
11:00	—	11:45	**Wentzcovitch**: Modeling Earth from Atomic to Global Scale
11:45	—	11:55	Poster talk 1
11:55	—	12:05	Poster talk 2
12:05	—	12:15	Poster talk 3
12:15	—	12:25	Poster talk 4
12:25	—	12:40	**Closing remarks**

Talk Abstracts

Theory, algorithms, and applications of spectral functionals

Tommaso Chiarotti[a] and Nicola Marzari[b,c,d,*]

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[b]Theory and Simulation of Materials, EPFL, Lausanne, Switzerland
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Electronic correlations play a central role in determining the ground state and response properties of many functional materials—from Mott-Hubbard insulators to high-Tc superconductors. While density-functional theory (DFT) has been the workhorse for first-principles calculations for more than three decades, these more complex classes of materials elude a DFT description, both for practical and conceptual reasons. Dynamical frameworks, where frequency-dependent self-energies appear, as in many-body perturbation theory or dynamical mean-field theory, provide instead a formally grounded approach to tackle excitations and electronic correlations.

Here we discuss an alternative approach, where a functional theory of the occupied spectral density is introduced,[1] generalizing density-functional theory to allow for the description of excitations and correlations in a very efficient and physically transparent formulation. The theory gives rise to dynamical functionals and dynamical potentials, for which a novel computational framework is also introduced—the algorithmic-inversion method on sum-over-poles (AIM-SOP)—tailored for these formulations and their implementation into computer codes.[2,3] Moreover, we approximate the unknown part of the spectral functional with a local and dynamical Hubbard functional, and we apply the framework to calculate both spectral and thermodynamic properties in the presence of correlations. We present fully self-consistent results for thermodynamic and spectral properties in the paradigmatic correlated solid $\mathrm{SrVO_3}$, showing close agreement with state-of-the art theory and experiments,[3] followed by the Mott-Hubbard/charge-transfer transition-metal monoxides, highlighting the accuracy of the present approach against state-of-the art calculations and experiments.[4] Last, we present the generalization of AIM-SOP to the case of strong correlations, as they emerge in defects and solids, and discuss its links to dynamical mean-field theory and to quantum embedding.

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[3] T. Chiarotti, A. Ferretti, and N. Marzari, *Energies and spectra of solids from the algorithmic inversion of dynamical Hubbard functionals*, Phys. Rev. Research **46**, L032023 (2024).

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Density Functional Approximations Guided by Exact Constraints

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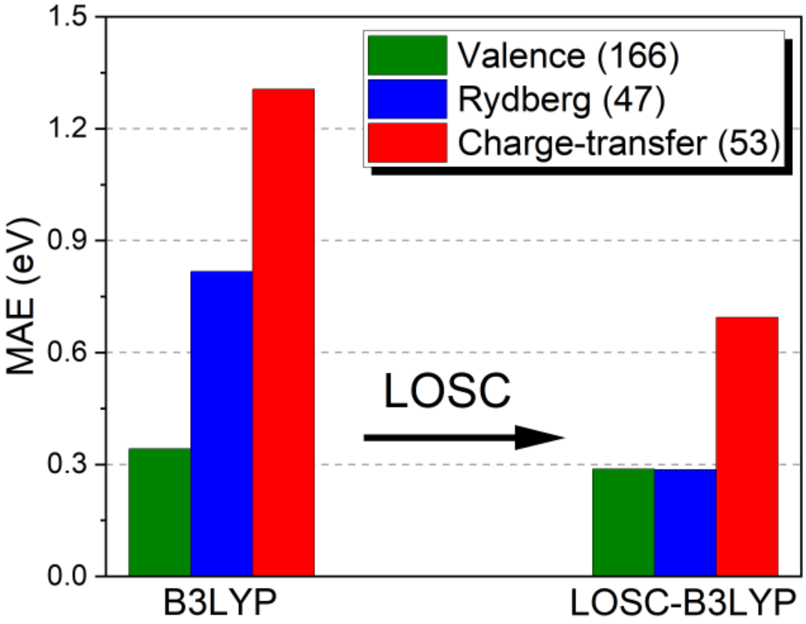
The present-day commonly used density functional approximations (DFAs) suffer from various systematic errors, and they can all be attributed to the violation of some exact constraints. I will focus on two important constraints: (i) the Perdew-Parr-Levy-Balduz (PPLB) linearity condition for fractionally charged systems; and (ii) proper energy behavior in the semi-classical limit $\hbar \to 0$. Guided by the first constraint, we have developed the localized orbital scaling correction (LOSC) functional that restores the PPLB condition through specially designed localized orbitals, called orbitalets. The LOSC functional can largely improve molecular dissociation problems, HOMO-LUMO gaps and photoemission spectra. Our most recent development of combining LOSC with linear-response time-dependent density functional theory (LR-TDDFT) has greatly improved excitation energies, particularly for Rydberg and charge-transfer excitations. The second constraint is highly relevant to strongly correlated systems. Through a model atomic calculation, we show that the exact total energy as $\hbar \to 0$ saturates to a finite value, whereas the mainstream DFAs have qualitatively wrong divergent behavior of $\hbar^{-1/6}$. By introducing an effective $\hbar$ for valence electrons, we estimate that the $\hbar_{\text{eff}}$ for such systems can be as small as 0.2. Thus, this exact constraint might inspire novel functional approximations for describing strong correlation.



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Fundamental Issues in DFT

Kimberly J. Daas[a], Steven Crisostomo[a] and Kieron Burke[a,b,*]

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[b]Department of Physics and Astronomy, University of California, Irvine, USA.

Density functional calculations drive modern materials science. While there are marvelous computational and algorithmic advances, there is still enormous room for improvement of the underlying theory. The focus of this session will be fundamentals of electronic structure theory, with an emphasis on unifying well-established but apparently very distinct approaches. Steven Crisostomo will discuss new relations between Kohn-Sham DFT and Green's functions approaches.[1] Kim Daas will unite two distinct DFT approaches to optical excitations, namely time-dependent DFT and ensemble DFT.[2] Kieron Burke will be the session leader.

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Shining Light on Moiré Materials: From Atomic Structure to Optical Excitations

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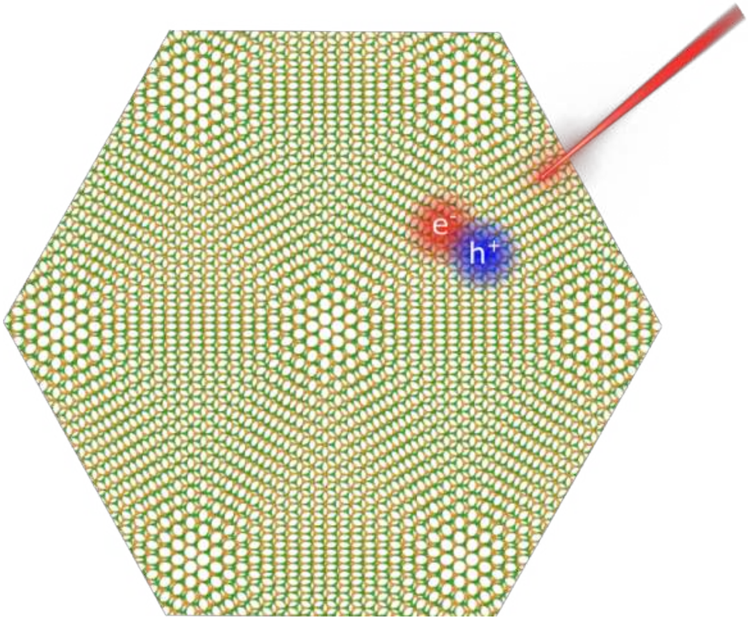
[c]Departments of Materials and Physics and the Thomas Young Centre for Theory and Simulation of Materials, Imperial College London, UK.

If one places a regularly-ruled transparent sheet on top of another identical sheet and then rotates the top sheet while holding the bottom one fixed, a beautiful moiré pattern emerges. Since 2018, similar moiré patterns have been created at the atomic scale, using two-dimensional materials such as graphene and transition-metal dichalcogenides. These novel moiré materials exhibit fascinating electronic, vibrational, and optical properties, including superconductivity and long-lived excitons, which are tunable through the twist angle, doping and device engineering.

A major bottleneck in accurate atomistic modelling of such materials is that the moiré unit cell can contain thousands of atoms at the small twist angles of interest. The direct application of well-established first-principles electronic structure approaches, therefore, is challenging due to the significant or prohibitive computational cost.

In this talk, we will discuss how we combine large-scale classical simulations and first-principles electronic structure calculations based on density-functional theory to study the electronic and vibrational properties of moiré materials. We reveal the emergence of chiral phonons,[1] and sliding phonon modes that generate surfing electrons.[2] Finally, we introduce a framework for efficiently computing optical excitations in moiré materials using many-body perturbation theory using a basis of localized Wannier functions.[3]



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Classical reaction barriers in DFT: An adiabatic-connection perspective

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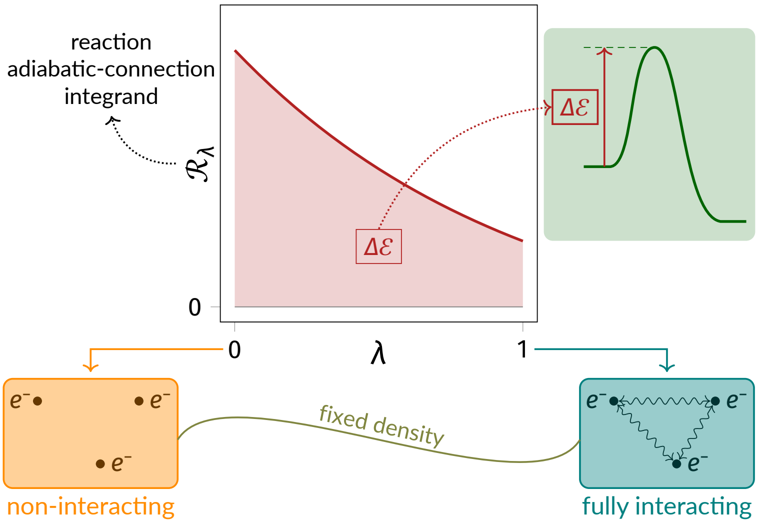
Classical reaction barriers in density-functional theory are considered from the perspective of the density-fixed adiabatic connection, which expresses the exchange-correlation energy as $E_{\text{xc}} = \int_0^1 \mathcal{W}_{\text{xc},\lambda} d\lambda$, where λ is the electron–electron interaction strength. In the first part of this talk, we describe how maximisation of the Lieb functional using a correlated electronic structure method can be used to determine an accurate reference $\mathcal{W}_{\text{xc},\lambda}$. We also show how $\mathcal{W}_{\text{xc},\lambda}$ can be determined for an arbitrary density functional approximation (DFA) by coordinate scaling. In the second part of this talk, we introduce a 'reaction adiabatic-connection integrand', $\mathcal{R}_\lambda$, involving the difference between the $\mathcal{W}_{\text{xc},\lambda}$ of the transition state and reactants, for which $\int_0^1 \mathcal{R}_\lambda d\lambda$ equals the barrier, meaning the barrier can be easily visualised as the area under a plot of $\mathcal{R}_\lambda$ vs. λ. We then compare reference $\mathcal{R}_\lambda$ curves, computed at the coupled-cluster level of theory, with approximate $\mathcal{R}_\lambda$, calculated from common DFAs for coupled-cluster densities. The comparison provides a simple way to visualise and understand functional-driven errors and trends in barriers from DFAs, whilst allowing a clean separation of the role of exchange and correlation contributions to the barrier. Specifically, the accuracy of $\mathcal{R}_0$ is determined entirely by the accuracy of the exchange functional, whilst the shape of $\mathcal{R}_\lambda$ is determined entirely by the correlation functional. The results clearly illustrate why the optimal amount of exact (orbital) exchange in hybrid functionals differs between reactions, including forward and reverse directions in the same reaction, and hence why simply introducing larger amounts of exact exchange may not be a reliable approach for improving barriers. Instead, the shape of $\mathcal{R}_\lambda$ must be captured more accurately through more accurate correlation functionals and the numerical data presented may be useful for this purpose. Finally, we consider density-driven errors from the perspective of $\mathcal{R}_\lambda$, highlighting the possible cancellation with functional-driven errors, noted in prior studies when Hartree–Fock densities are used.[1]



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Development in Functional Approximations and in Theory for Excited States

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The accuracy of DFT predictions hinges on the quality of density functional approximations (DFAs). Despite advancements, significant delocalization errors persist, leading to underestimating energy gaps in molecules and materials, incorrectly predicting the dissociation limits of chemical bonds, overdelocalizing charge distributions, and misaligning energy levels at interfaces. These errors stem from the violation of exact conditions on fractional charges, which are consequences of quantum mechanical degeneracy manifested in the classical variable of electron density. Our recent development of the localized orbital scaling correction (LOSC) addresses these errors and elevates DFT to a new level of robustness and accuracy. We will present results for molecules and bulk systems.

The orbital energies from our LOSC calculations reveals hidden excited state information from ground calculations, guiding us to the theory for excited states. Similarly in terms of total energies, since the 1970s, Kohn-Sham functionals have been employed for ΔSCF calculations of excited state energies, achieving accuracy comparable to that for ground state results, despite a lack of theoretical justification. Our recent research has established the theoretical foundation for ΔSCF calculations of excited states, showing that it is necessary to go beyond electron density and use the first-order density matrix of the noninteracting reference system to define the energy functional. The minimum of this functional corresponds to the ground state energy, consistent with ground state DFT, while the stationary solutions yield excited-state energies and electron densities, consistent with ΔSCF calculations. We also established the linear conditions for fractional charges in the excited-state theory and introduced the concepts of excited state chemical potentials. This in turn leads to the clear physical meaning of occupied and virtual orbitals energies as excited-state chemical potentials.

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Pushing the Boundaries of Hybrid DFT Based Simulations for Inorganic and Hybrid Semiconductors

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This talk targets electronic structure based simulations of crystalline multicomponent inorganic and organic-inorganic semiconductors, especially chalcogenides and hybrid organic-inorganic perovskites. Target properties include structure, phase stability, electronic, light emission and spin degrees of freedom. We summarize recent progress in the FHI-aims code,[1] especially a recent development of hybrid density functional theory that comfortably covers extreme system sizes, demonstrated for over 30,000 atoms.[2] Using this framework, numerically tabulated atom-centered basis sets enable precise all-electron simulations of energy levels and their alignments.

Energy level alignments between different components (e.g., organic and inorganic parts of layered perovskites) are captured in close agreement with experiments. Broken inversion symmetry and the deliberate inclusion of chiral molecules allow one to impart spin selectivity in electronic transport and excitons, including giant spin splitting in a series of specially designed organic-inorganic perovskites.[3] We also show how phonons impact the properties, including coherent phonon-mediated charge transport, chiral phonons, and involvement of phonons in the formation of a macroscopically coherent quantum state and superfluorescence at room temperature.[4]

We finally comment on next steps for physically complete simulations of multicomponent semiconductors, including a quasi-four-component relativistic treatment for accurate descriptions of energy levels (important in the presence of heavy elements such as Pb or Bi) that now covers system sizes of several hundred atoms, as well as more refined density functionals[5] to capture structural properties and electronic energy levels more rapidly and/or with higher accuracy.

This work would not be possible without the very large community of developers of the FHI-aims code around the world, as well as numerous experimental and computational collaborators. This work was partially supported by the Center for Hybrid Organic-Inorganic Semiconductors for Energy (CHOISE), an Energy Frontier Research Center funded by the Office of Basic Energy Sciences, Office of Science within the U.S. Department of Energy, and by NSF Award Number DMR-2323803.

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CASTEP Studies of NLO Crystals: A 30 Years Journey

Ming-Hsien Lee

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This paper review the scientific and people stories of utilizing CASTEP code on searching for new NLO crystals for the past three decades (since 1995). The technical evolution of the methodology start from an inspiration of brave adaptation of the belief that KS orbital is chemically meaningful.[1] An important mission of CASTEP study of those NLO crystals was to clarify a long debate in the research literature, namely ionic group contribution is more important or cations? By zeroing the Kohn-Sham wavefunction within a chosen space, usually a sphere as a unit, I switch off optical properties calculated for that crystal. Tests was made to demonstrate the additivity of cut parts summing back to roughly original value, if reasonable partition is defined, such as cations and anion functional groups , in this sense, the mechanism of NLO and birefringence of beta-BaB2O4 was explained.[2]

Although atom-cutting method was direct, it require a human choice of partitioning to divide the whole crystals into subsystem. By taking the advantage of triple summation loop of KS bands, SHG can be divided into two types of process, namely Virtual Electron (v-c-c transition) Virtual Hole (v-v-c transition). For example, fixed one nv and sum the rest of band index loops, one obtains the effective band contribution to the total VE or VH which is strictly additive, and therefore a perfect decomposition of SHG into band contribution, one can use this to identify the "SHG-relevant KS orbitals" in molecules or crystals. Unexpectedly, for molecules and AgGaS/Se2. only a very small subset of the entire valence and conduction bands manifold contribute to SHG, this allow the mechanism of SHG been discussed, energy levels and optical matrix elements in an un-entangled way.[3]

In spite of detail information band-resolved analysis provided, when applied such analysis on important crystalline oxides such as BBO, there is still way to many lines to handle and to make sense. I therefore proposed constructing orbital density based on individual SHG weight. This SHG-weighted density gives the hot-spot of SHG in crystal because any probability density of SHG-unimportant orbitals simply disappear from the visualization. One can see very chemical / local visualization of physical property related Bloch states without the use of Wannier functions type techniques. Being able to see this hot-spot bring the design of NLO crystals to the next level, one know which part to keep and which part to replace for improved performance for other mechanical chemical properties of a given family of crystals.[4]

After DFPT has been implemented into CASTEP, to our delight, most of the "anomaly" (or at least "puzzling") materials giving poor birefringence through using SOS method[5] can be largely improved with DFPT. There is no surprise that DFPT give better result, after all, SOS is based on zero-th order wavefunction, but DFPT is up to 1st-order. However, we perform an extra computer experiment by fixing to original crystalline symmetry when performing DFPT E-field perturbation. quite unexpectedly roughly half of those SOS puzzled then DFPT improved crystals fall back to similar values of SOS under this "wrong way of doing" PDFT. This mean for some puzzled crystals, the failure of SOS is due to it limitation of no symmetry breaking, whereas full DFPT take that into account.[8]

B. Zhang has performed high-throughput screening on borate[6] and germannate[7] from ICSD. Some new candidates of NLO crystals has been found. Later with Tomoyuki Tamura, all structures without inversion center symmetry in ICSD is used to tarin a machine learning model based on ALiGNN, then search on Materials Projects data results in the prediction of 10 new promising NLO crystals.[9]

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Search for ternary superhydrides metastable at ambient pressure in the Y-Fe-H system: a new prototype structure

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Computational studies have successfully predicted the dramatic uptake of hydrogen by metals under pressure to the formation of superhydrides. These materials demonstrate exceptional properties, such as high-temperature superconductivity, hydrogen storage and superionicity, like in LaH_{10}.[1–3] Up to now, most of the studies have been performed on binary hydrides in the 100 GPa range and none of the discovered superhydrides could be recovered at ambient pressure. Ternary hydrides offer possibilities to stabilize superhydrides at ambient pressure.[4] We have thus undertaken the search of ternary superhydrides in the system Y-Fe-H by coupling calculations and experiments. Remarkably, we synthesized a novel superhydride above 80 GPa that could be recovered metastable at ambient pressure at least for few hours. This is validating a promising avenue for future-experimental investigation in high-temperature superconductivity within hydrides at ambient pressure.

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In silico Engineering of Piezoelectric Biomolecular Assemblies

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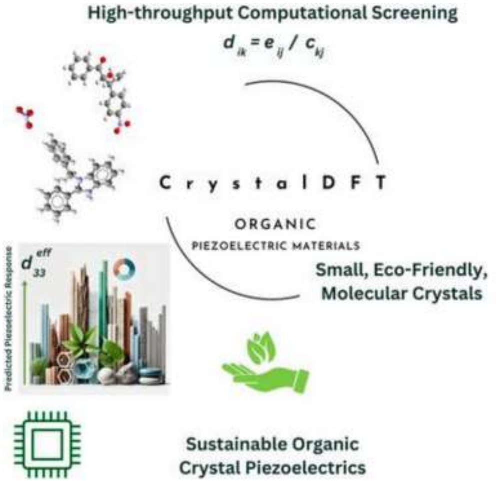
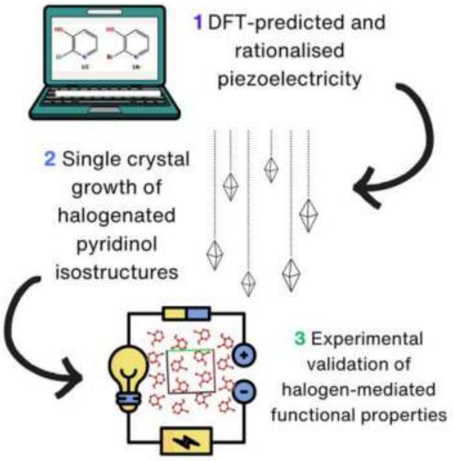
The ability of certain materials to generate electric charge in response to mechanical stress is derived from their non-centrosymmetric crystal structures, which produces a wide array of applications in sensing, actuation, and energy harvesting.[1] Organic molecular crystals, characterized by their structural tunability, sustainability, and biocompatibility, present a promising platform for the next generation of piezoelectric materials.[2] Utilizing crystal engineering and Density Functional Theory (DFT), we have developed CrystalDFT a structured database of crystals accompanied by predicted electromechanical properties.[3] This dataset reveals a diverse range of piezoelectric responses, featuring many materials exhibiting natural (unpoled) longitudinal piezoelectricity. A focused study on hydrated organic crystals, involving high-throughput DFT screening, identified thirty hydrates with longitudinal piezoelectric coefficients exceeding 10?pC/N, including one reaching 386?pC/N. These results underscore the role of hydrogen bonding in enhancing dipole alignment and facilitating polar phases. Further investigation of structure property relationships led to the design and synthesis of halogenated pyridin-3-ol derivatives (2-X-pyridin-3-ol; X = Cl, Br, I). Stabilized by both hydrogen and halogen bonding,[4] these crystals exhibit DFT-predicted shear responses up to 99.19?pC/N. Experimental shear responses of 54 74?pC/N were accompanied by longitudinal responses of 5 10?pC/N.

Overall, these studies showcase the effectiveness of computational screening and molecular design in discovering high-performance organic piezoelectrics, bridging theory and practical application in sustainable electromechanical materials.





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Unraveling the potential of Atomic Force Microscopy with DFT-based simulations and Deep Learning

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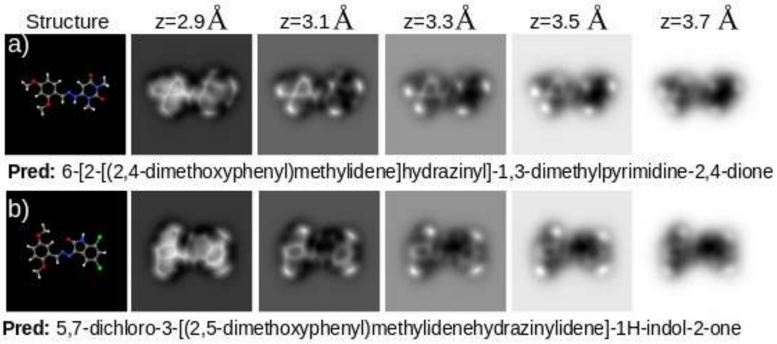
Atomic Force Microscopy (AFM) operated in the frequency modulation (FM) mode achieved true atomic-scale imaging and manipulation on both conducting and insulating materials.[1,2] AFM with metal tips functionalized with a CO molecule at the apex (HR-AFM) revealed the internal structure of adsorbed organic molecules with unprecedented resolution, resolving intermolecular features and determining bond orders.[3] Density Functional Theory (DFT) has played a key role in understanding the AFM image contrast and extending its capabilities. This talk and the two following ones explore this fruitful collaboration.

DFT-based AFM simulation methods[4,5] applied to the interpretation of the contrast observed in porphycenes[6] and on self-assembled molecular layers driven by either halogen[7] or hydrogen bonds,[8] show clear connections between fundamental chemical properties of the molecules and key features imprinted in AFM images. Here, we show how Deep Learning (DL) models[9] –trained with QUAM-AFM,[10] a very large data set of theoretically simulated HR-AFM images–, can achieve a complete identification (structure and chemical composition) of an arbitrarily complex molecule from a stack of HR-AFM images taken at different heights. Multimodal recurrent networks (M-RNNs),[11] that combine convolutional neural networks (CNNs) for image analysis and recurrent NN (RNNs) for natural language processing can determine the chemical groups and their connection within the molecule to predict the IUPAC name from HR-AFM images (see Figure) with an accuracy better than the best language translation models. Some of the limitations of M-RNNs can be overcome using Conditional Generative Adversarial Networks (CGANs)[12] or exploiting other molecular as the molecular fingerprints.[13] This approach has the potential to become the next breakthrough in molecular identification,[14] overcoming the limitations of traditional spectroscopic methods like nuclear magnetic resonance (NMR).



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Nanomanipulation & Dynamics of Single-Molecules at Surfaces

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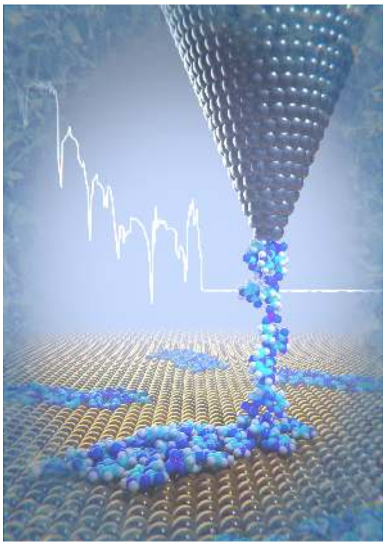
Understanding the motion/dynamics of a single molecule over a surface is a problem of a paramount importance in designing advanced molecular nanostructures/assemblies capable of meeting specific needs.[1–5] Almost 20 years passed after the first scanning probe microscopy[6] atomic-manipulation until it became possible to record the time variations of mechanical forces during manipulation.[7] As this technique[7–12] – cryo-force-spectroscopy – relies on a dynamic AFM operated at 4.8K in Ultra-High-Vacuum it allow us to perform successive manipulations a in a clean and controlled environment but also enable us to characterize both the surface and the molecule with atomic resolution prior and after the manipulation. This breakthrough opened new and exciting possibilities to explore mechanical properties at a single molecule level with an unrepresented resolution and control over the system. In this talk I shall discuss some of the major achievement and the versatility of this technique including: the discovery of a state of almost vanishing friction of graphene nanoribbons when sliding over a gold surface;[8,9] measuring the stiffness of a single DNA nucleotide and the detection[10] of a C-C bond twist at a single bond level,[11] among others.[12,13] Altogether this provides a comprehensive understanding of complex on-surface dynamics of the molecules over surfaces and how dynamic balancing of intra-molecular mechanics and surface interaction play out in a broad range of conditions.



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Accurate forces for CO tips on cobalt phthalocyanines on Ag(111)

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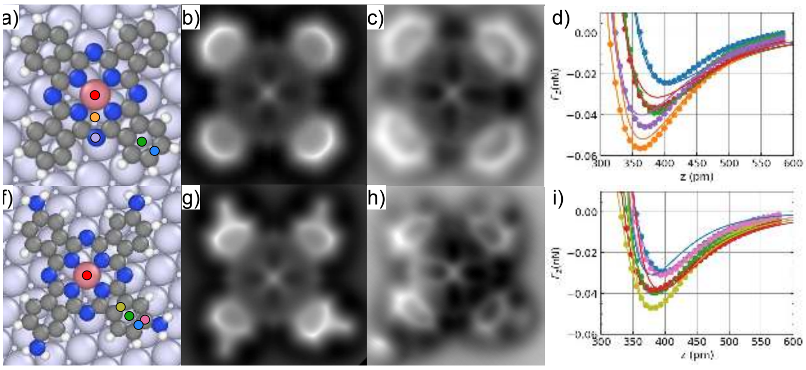
Recent studies have revealed that cobalt phthalocyanines (CoPcs) immobilized on carbon nanotubes, a hybrid catalyst, enhance methanol production from CO_2.[1] In particular, the interaction strength between the intermediate CO and the central Co atom is a critical factor influencing catalytic performance. To explore the subtle characteristics of this interaction, Wang et al.[2] have used a novel 3D scanning methodology with a high-resolution atomic force microscope (HRAFM) with a CO-functionalized tip to study CoPc molecules on Ag(111) at the single-molecule level. Here, we validate this extensive experimental study with density functional theory (DFT) and the full-density-based model (FDBM)[3,4] for HRAFM. Our DFT simulations reveal that the most stable adsorption configuration for a CoPc molecule on Ag(111) is the cobalt metal atom on a bridge site with one of the wings aligned with the silver's close-packed direction, as seen in Fig. 1 a). Then, using the FDBM, we confirmed Wang et al.'s method for isolating CO tip-sample interactions from substrate and metallic tip effects. The differences between the experimental and theoretical force curves indicated the molecule was being bent by effect of the CO tip. Taking into account the stiffness of the molecule, the curves show a remarkable agreement, Fig 1. d).[5] We extended this study to $CoPc(NH_2)_4$ (Fig. 1 second row) and compared the electronic structure of both molecules, analyzing the effect of the amino groups. Finally, we have looked for possible explanations to the unusual spot that appears quite frequently associated to $CoPc(NH_2)_4$ (lower left corner of the experimental image in Fig. 1 h).



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Ab Initio bulk free energy surface of proper ferroelectrics

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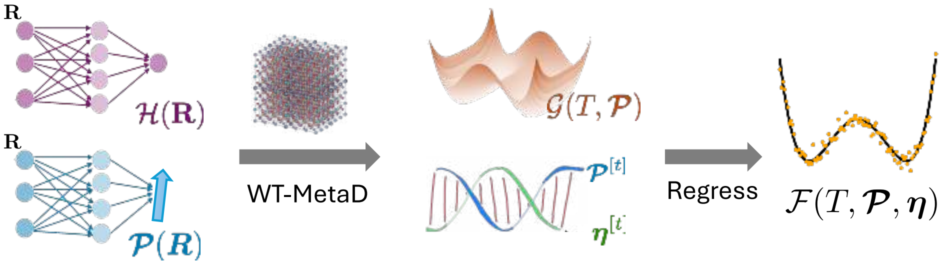
We report a systematic and accurate approach for deriving the bulk free energy surface (FES) $\mathcal{F}(T, \boldsymbol{\mathcal{P}}, \boldsymbol{\eta})$, a function of temperature T, polarization $\boldsymbol{\mathcal{P}}$, and strain $\boldsymbol{\eta}$, from the first-principles density functional theory (DFT) of proper ferroelectrics. The core of our approach is the well-tempered metadynamics (WT-MetaD) algorithm that extracts the polarization dependence of the FES from all-atom molecular dynamics simulations without an *a priori* ansatz. The rest of the FES is derived from the WT-MetaD trajectories that span the relevant phase space. We demonstrate our approach in the case of lead titanate. The errors across the phase transition, due to DFT numerics, all-atom molecular dynamics, and free energy evaluation by enhanced sampling, can be systematically controlled and are of the order of 1meV/atom. The accuracy of the resulting *ab initio* FES is only limited by the adopted functional approximation of DFT.



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Modeling the Solvated Electron: Structure, Reactivity, and New Machine Learning Methods.

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We study the solvated electron in bulk water with our newly developed neural network methods. Our simulation corroborates the cavity picture, revealing a robustly localized electronic state. We characterize various structural and dynamical properties of the solvated electron. We further explore the reactivity of the solvated electron with a hydronium ion, and map a proton-transfer–mediated recombination pathway that yields a hydrogen radical. It turns out that accurately modeling this seemingly simple system places strong demands on machine-learning methods; to meet them we have developed DP-MP (Deep Potential with Message Passing), an accurate yet lightweight neural-network force field, as well as an iterative Wannier-center prediction method that tracks the electron's position on-the-fly. The code for these methods is available at `https://github.com/SparkyTruck/deepmd-jax`.

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On the thermodynamics of crystals under hydrostatic pressure

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Viewed on the length scale of macroscopic thermodynamics, what distinguishes a uniform solid under hydrostatic pressure from a liquid? The answer, agreed on in textbooks on continuum mechanics, is the harmonic elastic response to small changes of volume relative to a reference volume. This is a manifestation of solid rigidity. No such reference is needed for liquids. The pressure is determined by the actual density only. On the other hand, liquids can gain mass by inserting particles. This is much more difficult for solids, one would have to search for the rare vacancy. The chemical potential of a crystal diverges approaching full lattice site occupation. As a result, the conventional Gibbs-Duhem relation for liquids breaks down even under hydrostatic pressure. This is recognized, to varying degree, in the material science literature but generally not accepted by physical chemists. Using a modification of a mean field compressible lattice gas model common in materials science[1] we first show that the Gibbs-Duhem relation is indeed violated in this model crystal and then propose a way to restore it by including the number of lattice sites in the set of extensive state variables.[2] With M fixed, the open system is stable under isothermal isobaric conditions (μ, p, T). This also introduces a new thermodynamics field, the force conjugate to M, which is useful for the evaluation of open system isobaric isothermal response coefficients. As an example we will work out an expression for the generation of vacancies induced by the application of pressure.

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Modelling of Electrified Solid-Liquid Interfaces with Finite-Field DFTMD

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The development of novel materials for electrochemical energy storage and conversion is central to the electrification and sustainable advancement of our world. Achieving atomistic-level understanding, control, and design of these materials is therefore crucial. In this context, molecular modeling grounded in quantum and statistical mechanics—and increasingly empowered by machine learning—offers both deep physical insight and predictive capability.

In this short talk, I will present our recent efforts on modelling electrified solid–liquid interfaces in aqueous environments. Metal oxides, for example, can become strongly charged due to acid–base surface chemistry, especially when the operating pH deviates from the point of zero charge, leading to the formation of a protonic double layer (pDL). In contrast, the classical electric double layer (EDL) arises at metal–electrolyte interfaces, where charge accumulation is induced by external bias relative to the potential of zero charge.

Despite their distinct origins, both types of double layers can be treated on an equal footing using finite-field density functional theory-based molecular dynamics (DFTMD). This unified framework enables detailed investigations of capacitive charging and electric field effects at electrochemical interfaces, while also generating high-quality reference data for the development of machine learning-accelerated models.

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Liquid state DFT in the 21^{st} century: Electrolytes, electromechanics and dielectrocapillarity

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Understanding the structural and thermodynamic behavior of complex fluids—especially electrolytes—requires theoretical approaches that bridge multiple length scales. Density functional theory (DFT) for classical fluids offers a first-principles route to this challenge, but practical applications remain difficult due to the greater diversity and complexity of intermolecular interactions in molecular fluids compared to quantum DFT. In this work, we will explore how combining classical density functional theory (cDFT) with deep learning offers a powerful and flexible framework for uncovering emergent phenomena in electrolyte solutions and dielectric liquids. This hybrid methodology not only enables accurate and efficient predictions of fluid structure and thermodynamics far beyond the reach of traditional molecular simulations,[1] but also opens the door to discovering new physics. One such example is "dielectrocapillarity"—a novel phenomenon describing how electric field gradients can control phase behavior and criticality in polar fluids.[2] We will discuss recent developments that provide a first-principles understanding of such electromechanical effects[3] and point toward new strategies for manipulating complex fluids, both in bulk and under confinement.

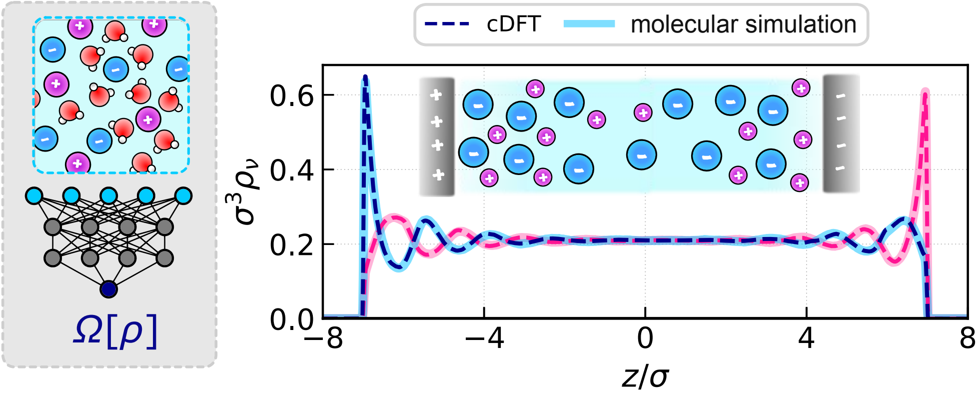

Figure 1: The structure of the electric double layer predicted by cDFT, computed at a fraction of the cost of a molecular simulation.

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Finite Temperature Crystal Structure Prediction

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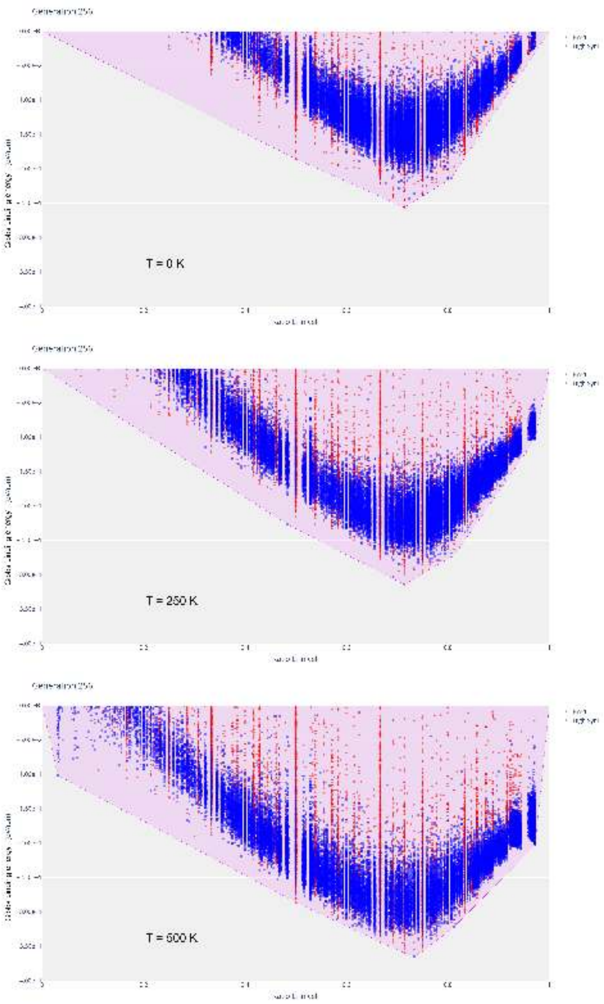
Crystal structure prediction is a key starting point for materials discovery. Convex hulls are a powerful tool for the prediction of thermodynamically stable compounds of 2 or more elements. In order to construct a convex hull, the complete set of stable structures at a given temperature and pressure must be known, but this is not achievable for all but the most simple cases. Furthermore, a search for minimum energy structures can be computationally expensive. Many algorithms have been developed for crystal structure prediction, including the CASTEP Genetic Algorithm[1–3] which has recently been extended to convex hulls.[4] However, until recently, almost all convex hulls have been calculated at T=0. A few (e.g. ref. [5]) have attempted to go further by including the vibrational free energy, but until now, there has been no inclusion of configurational entropy. We have recently developed a simple expression for the configurational entropy of a crystal.[6] In this work, we combine this with the vibrational free energy to generate a complete description of the Gibbs binding energy at modest temperatures. At higher temperatures, it is expected that anharmonic effects will become important, and these have not been included yet. The figure shows an example 2D convex hull of the Li-Si system, at 3 different temperatures, calculated using MatterSim.[7] It can be seen that temperature changes the shape of the hull and new structures become stable.



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DE-FF and MACE-OFF: Data-driven interatomic potentials for molecular simulations

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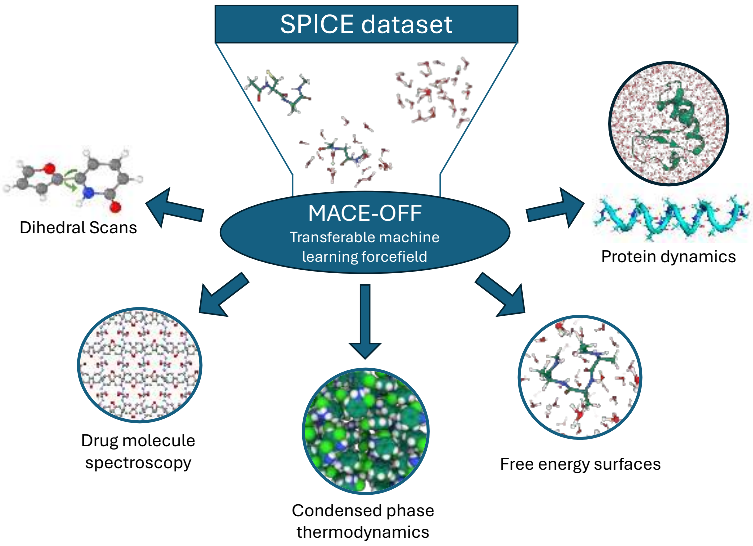
Drawing on computational methods that are based around training to extensive condensed phase physical property and quantum mechanical datasets, we will describe some of our efforts to design accurate and transferable inter- and intra-molecular potentials, with a view to applications in condensed phase atomistic modelling and computer- aided drug design.

We will explain how recent collaborations with the Open Force Field Initiative enable the development of a fast, accurate alternative to the Lennard-Jones non-bonded potential.[1] With OpenFF, we developed Smirnoff-plugins as a flexible framework to extend the software stack to include custom force field functional forms. We deployed the infrastructure that OpenFF has provided for optimising parameters against condensed phase data, to train a transferable, small molecule force field based on a double exponential functional form (DE-FF). The automated framework allowed us to train and test a full small molecule force field in just a matter of weeks (as opposed to many years for traditional force fields), with promising accuracy in the condensed phase.

Finally, we will describe the MACE-OFF series of transferable force field for organic molecules created using state-of-the-art machine learning technology and first principles reference data.[2] MACE-OFF demonstrates the remarkable capabilities of short-range models by accurately predicting a wide variety of gas and condensed phase properties of molecular systems, including dihedral scans, descriptions of molecular crystals and liquids, and even properties of a solvated small protein.



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Should I Stay or Should I Go: From Empirical Potentials to Machine-Learned Force Fields in Atomistic Biomolecular Dynamics

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Biomolecular simulations at atomic resolution provide a powerful "computational microscope" on the dynamical behavior of living systems. Machine-learned force fields (MLFFs) have accelerated *ab initio* MD simulations by 6-7 orders of magnitude over the last decade.[1,2] Although MLFFs are expected to revolutionize biomolecular simulations,[3] empirical molecular-mechanics potentials are widely trusted to provide actionable insights. Here we present a comprehensive investigation of the similarities and differences in simulation results and phenomena obtained with a general-purpose SO3LR MLFF[4] and a range of widely used empirical potentials. Both approaches are consistent in the prediction of infrared and NMR spectra of solvated and gas-phase (bio)molecules. However, a comprehensive analysis of the trajectories shows qualitative differences in the energetic, enthalpic, entropic, and vibrational mode distributions, such that even the dynamics yielding similar spectra may have altered character. These differences stem from the anharmonicity and extended range of interatomic interactions captured by the MLFF trained on quantum forces, in contrast to the dominant harmonic and short-range physics underlying empirical potentials. We conclude that biomolecules are strongly collective and dynamical beyond conventions based on empirical potentials and static crystal structures.

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Next-Level Machine-Learning Interatomic Potentials: Molecular Dynamics with Local Electronic Degrees of Freedom

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Over recent years, machine-learned interatomic potentials (MLIPs) have quickly become an invaluable tool in physical chemistry and material science. By modeling potential-energy surfaces and associated forces with comparable accuracy, but at fraction of the cost of *ab initio* approaches, they allow for thermodynamic sampling at previously unreachable time and length scales. In turn, this provides the opportunity to simulate several pivotal mechanisms for energy conversion and storage, including the *in operando* evolution of electrochemical interfaces, including those of (semi-)amorphous systems, as well as charge intercalation and stabilization processes. However, these applications typically require to explicitly account for *local* electronic charges, charge redistribution, and electric fields. While several important developments have recently focused on incorporating long-range electrostatic interactions, the aforementioned local effects have, so far, mostly been neglected.

In this work, we discuss strategies on how to overcome these shortcomings by explicitly reintroducing local electronic degrees of freedom on top of the MLIP. In the first example, we present a response-augmented approach that leverages local descriptors including Born effective charges to predict local electric fields. This gives access to work-function changes and hence allows to model electrified metal surfaces at variable potentials, even in explicitly solvated, complex interfacial environments. We demonstrate the power of the proposed approach by investigating the pH-dependent, non-Nernstian adsorption-site switching of OH on Cu(100) surfaces.[1]

Second, we discuss how localized, but percolating electronic charges such as small polarons can be modeled within MLIP-frameworks as semi-classical degrees of freedom that follow the nuclei adiabatically. We demonstrate the approach for lithium titanium oxide (LTO), a prototypical anode material for which polaron hopping is known to play a decisive role.[2] Our large-scale molecular-dynamics simulations reveal that polarons do not only diffuse orders of magnitude faster than ions, but that they are also able to enhance lithium diffusivity. Thereby, polarons do not merely serve as spectators, but lower the effective Li-diffusion barrier by thermodynamically adapting to the ionic motion. This corroborates the occurrence of a correlated polaron-ion dynamics with profound implications for the design of energy materials.

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Relevance of Catalyst Restructuring Under Reaction Conditions: An Automated Process Explorer Approach

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Catalysts are known to restructuring under operando conditions; the formation of the $\sqrt{5}$ surface oxide on the Pd(100) surface is a prototypical example of this. Despite extensive investigation, details of its formation remain unknown. By combining a Machine-Learned Interatomic Potential (MLIP) with a newly-developed Automatic Process Explorer (APE),[1] I will present an example of O-induced restructuring at the step edge of the Pd(410) surface, as the early stages of surface oxide formation. Our MLIP-APE methodology allows us to identify 3,000 unique elementary processes for surface restructuring. Applying a fuzzy classification approach[2] to changes in local environment during these processes, we take first steps towards a comprehensive surface restructuring mechanism. Many identified restructuring processes involve unintuitive complex collective motion, which can explain experimentally-observed processes e.g. step-bunching or microfaceting. Critically, the timescale of the majority of identified restructuring pathways is comparable to that of catalysis, implying that surface restructuring can take place simultaneously to catalysis.[3]

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Tackling large embedded systems: environmental control in photosynthetic antennae

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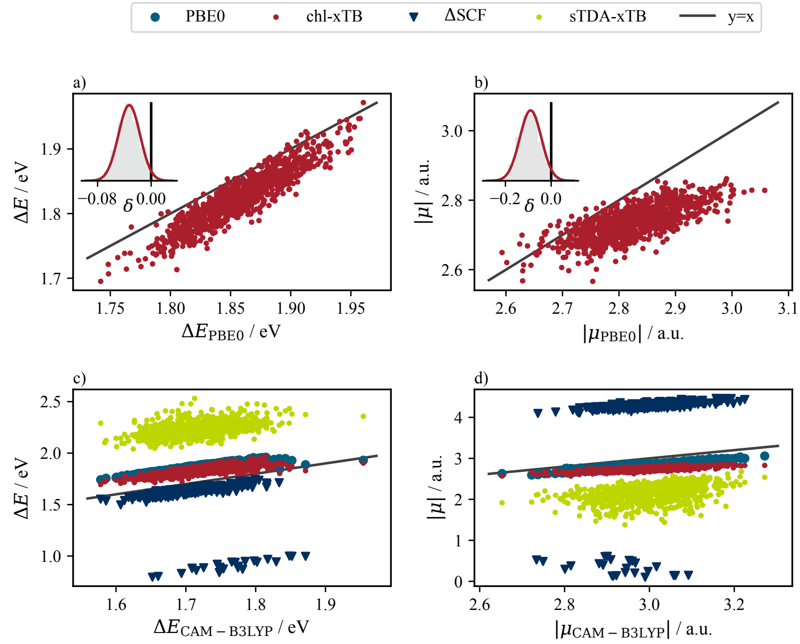
Natural photosynthesis provides copious inspiration for a circular solar economy where components of the light-harvesting machinery are constantly broken down and reused in an entirely self-sustaining way. A strikingly interesting feature of photosynthetic systems is that the same fundamental components (chlorophyll pigments) are used to perform radically different functions in different parts of the photosystem: exciton transfer across the antenna system and charge transfer in the reaction centre. The implication is that nature has not selected the most optimal pigment for each job but rather made use of available chromophores and altered their behaviour to create the desired functionality.

Understanding how the features of the protein environment drive these different functionalities is a challenging task due to the size and complexity of the system. I will discuss ways in which recent advances in electronic structure theory, including highly efficient, targeted quantum chemical calculations[1,2] and machine-learned forcefields,[3] can be brought together to start exploring the crucial role of the environment in controlling chromophore photochemistry.



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Exploring Defect Energy Surfaces with Machine-Learned Foundation Models

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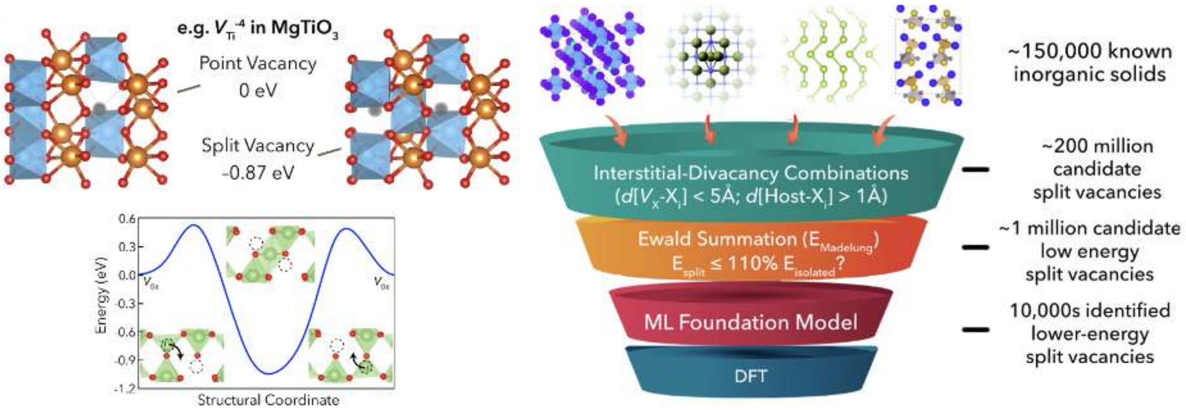
Recently, the defect modelling community has made steps toward adopting machine learning approaches, to avoid the limits imposed by costly DFT supercell relaxations.[1,2] However, challenges remain before these ML methods can be widely-adopted for defects.

In this work, I focus on the formation of split-vacancy defect geometries; a motif which can dramatically lower vacancy formation energies and alter behaviour, yet are difficult to predict with conventional approaches due to enormous configuration spaces and 'non-local' reconstruction. I show how recently-developed machine-learned foundation models can be readily applied to this task, but with important caveats.[3] Combined with geometric and electrostatic analysis to produce an efficient tiered screening approach, these methods permit the screening of all solid-state compounds in the Materials Project and ICSD databases to identify thousands of low energy split vacancy configurations, hitherto unknown. Recent developments of foundation models in the NequIP and Allegro model family will also be discussed.[4–6]



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Exploring charge density waves in twisted bilayer NbSe2 with machine learning

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Niobium diselenide has garnered significant attention over the past few decades because of the coexistence of superconductivity and charge density waves (CDWs), observable down to the monolayer limit. Introducing relative twist angles between monolayers, in the field of twistronics, offers a new variable to tune these systems, yet a fundamental question remains: do CDWs persist in moiré structures, and how are they altered compared to the pristine monolayer/bilayer? Traditional first-principles methods face limitations due to the computational resources required for long-wavelength moiré patterns; for instance, a 1-degree twist angle necessitates modeling over 10,000 atoms, making simulations impractical. This study employs first-principles data to develop machine learning interatomic potentials with the Allegro architecture, enabling scalable and accurate simulations. We investigate the formation and evolution of CDW order in monolayers and twisted bilayers, validating our results against density functional theory calculations. Beyond niobium diselenide, our goal is to establish a protocol for studying CDWs in two-dimensional systems. We outline strategies for producing training data and perform a detailed hyperparameter scan to identify key aspects for studying these systems.[1]

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First-principles study of heterogeneous catalysis: the spin promotion effect and revisiting electronic descriptors

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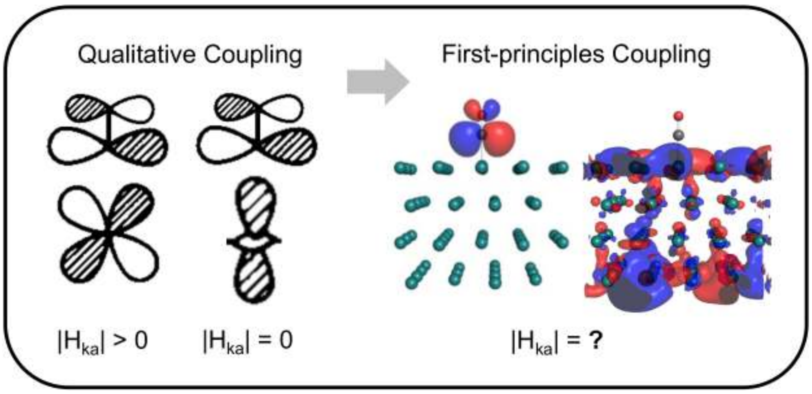
In the first part of our contribution, we discuss recent work on a spin-promotion effect for heterogeneous catalysis, with examples including thermal synthesis and decomposition of ammonia.[1–3]

In the second part, we discuss the coupling and overlap matrix elements between the orbitals of adsorbates and surfaces. While these parameters are ubiquitous in the literature, they are usually approximated or invoked qualitatively. Using a recently developed diabatization procedure,[4] we calculate these parameters directly from Density Functional Theory. We find the resulting matrix elements suitable for the description of a broad range of phenomena on surfaces, including ultrafast electron transfer, scanning-tunneling spectroscopy and the catalytic activity of transition metals. The approach yields quantitative agreement with experiment and enables interpretation of phenomena in the familiar language of orbital phase.



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Computational Discovery of New Materials for Singlet Fission in the Solid State

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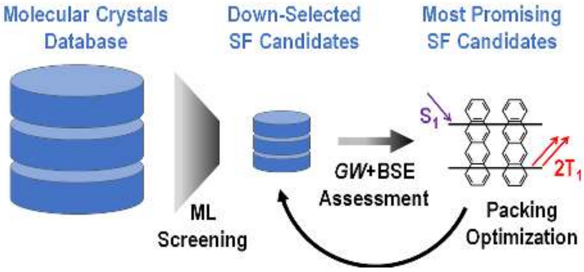
Intermolecular singlet fission (SF) is the conversion of a photogenerated singlet exciton into two triplet excitons residing on different molecules. SF has the potential to enhance the conversion efficiency of solar cells by harvesting two charge carriers from one high-energy photon, whose surplus energy would otherwise be lost to heat. The development of commercial SF-augmented modules is hindered by the limited selection of molecular crystals that exhibit intermolecular SF in the solid state. Computational exploration may accelerate the discovery of new SF materials. The GW approximation and Bethe–Salpeter equation (GW+BSE) within the framework of many-body perturbation theory is the current state-of-the-art method for calculating the excited-state properties of molecular crystals with periodic boundary conditions. However, its high computational cost is prohibitive for large-scale materials screening. Therefore, we develop materials discovery workflows that combine GW+BSE with low-cost physical or machine learned (ML) models.[1] For example, we implemented a genetic algorithm fitness function based on a dimer model for the SF rate combined with structural stability and used it to discover a putative polymorph of tetracene predicted to have improved SF performance.[2] To explore the chemical space in search of new types of SF materials, we trained symbolic regression ML models to predict the GW+BSE values of the thermodynamic driving force for SF based on properties calculated at the density functional theory (DFT) level.[3] To this end, we generated the PAH101 dataset of GW+BSE calculations for 101 polycyclic aromatic hydrocarbons (PAHs).[4] More recently, we reused the PAH101 set to train ML models to predict additional device-relevant excited-state properties of molecular crystals, including the optical gap (corresponding to the first singlet exciton energy), the triplet exciton energy, the singlet-triplet gap, and the singlet exciton binding energy.[5]



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Single-Reference Linearized Coupled Cluster Theory for Strongly-Correlated Systems

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Linearized coupled cluster theory with double substitutions (LinCCD) and its singles-inclusive counterpart, LinCCSD, approximate the exponential form of the coupled-cluster wave function to linear order. While these linearized approaches offer simpler gradients, straightforward Hermitian formulations, and improvements to parallelizability, LinCCD produces near-singular results in strongly correlated systems. Furthermore, the LinCCD approach is akin to configuration interaction with double substitutions (CID) and is not size-consistent. We have recently shown that size-consistency and regularity can be restored in LinCCD by removing terms associated with ring and crossed-ring diagrams,[1] leaving only the ladder and driver contributions. The resultant linearized ladder coupled-cluster doubles (LinLCCD) method can smoothly break covalent bonds. Through a further approximation wherein the particle-particle ladder is removed, a regular size-consistent theory can be obtained that scales as $\mathcal{O}(N^4)$ with density fitting. While LinLCCD does not diverge for single bond dissociations, the predicted equilibrium energies and dissociation limits are generally much too high. Furthermore, LinLCCD does diverge in the spatially symmetric dissociation of N2. Recently, we have found that a simple linear perturbative correction to the LinLCCD wave function can restore the important contributions from the ring/crossed-ring diagrams to the energy. Our xLinCCD(2) and xLinCCSD(2) methods retain size-consistency, recover full CCD-quality bond dissociation curves for single bonds, and can smoothly dissociate N2 without the spurious barrier encountered in CCD. Moreover, we can solve the xLinCCD(2) equations in two cycles and without amplitude storage for substantial cost savings over CCD approaches while recovering similar results in strongly correlated systems. Our xLinCCD(2) method provides some of the most accurate results reported for a theory that is entirely linear in the wave function amplitudes.

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Some new functionality in Castep, inverted densities and non-collinear DFT+U

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Stewart, as Chair of CCP9, the UK's UKRI funded network on electronic structure in solids will give a brief overview. He will then introduce some new functionality his group has been doing adding new science to Castep. Two of his PhD students will then give more details and results of their work.

Theo will discuss his work on the addition of the Hubbard U parameter to the CASTEP plane wave density functional theory (DFT) code for calculations involving non-collinear magnetism. The DFT+U scheme entails the addition of a Coulomb interaction potential (U), to improve the accuracy of a calculation's description of a material's electronic structure. CASTEP has for a long time supported the addition of a Hubbard U potential to non-spin polarised calculations and to calculations that contain collinear spins constrained to a single quantisation axis. We have recently implemented the Hubbard U approach for non-collinear magnetic calculations. I shall present results with a focus on $Dy_2T_2O_7$, a frustrated pyrochlore magnet, where using the source-corrected LDA exchange correlation functional, we are able to reproduce[1] the spin ice ground state.

Visagan will present work on density inversion within DFT. Assessing the quality of Kohn-Sham (KS) potentials $v_s(\mathbf{r})$, particularly the exchange-correlation (XC) potential $v_{xc}(\mathbf{r})$ remains a challenging problem, not least due to the fact that the exact $v_{xc}(\mathbf{r})$ is unknown and not a physical observable, thereby making direct comparison with experiment impossible. Density inversion allows one to "reverse-engineer" $v_s(\mathbf{r})$ from a given target ground-state density $\rho_0(\vec{r})$. Using the inversion method of Refs.,[2,3] we investigate and quantify the degree of non-locality of various generalised Kohn-Sham schemes in insulating solids, including meta-GGAs, hybrids and DFT+U by comparing differences between the GKS and KS band structures.[4] We also compute the accurate ground-state densities for a selection of these solids using variational and diffusion quantum Monte Carlo,[5] allowing us to gain some insight into the behaviour of the exact $v_s(\mathbf{r})$ and to benchmark $v_{xc}(\vec{r})$ and $\rho_0(\vec{r})$ of some common XC approximations.

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The ONETEP linear-scaling density functional theory program

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The ONETEP code[1,2] was originally designed to enable large-scale electronic structure calculations using an implementation of density functional theory capable of achieving linear scaling with system size. Through the use of a basis set equivalent to plane waves and well-controlled approximations, comparable accuracy to the plane-wave pseudopotential formulation can be achieved, while the real-space approach is well-placed to exploit low-dimensional systems and embedding approaches.

ONETEP was originally designed for self-consistent calculations that only sample the Γ-point of the Brillouin zone (BZ). However, many systems of interest are periodic at short scales in one or two dimensions (e.g. nanowires and surfaces), where significant computational efficiency can be achieved via using smaller supercells and sampling the BZ. Here, we introduce two types of BZ sampling methods in ONETEP. The first is targeted at small periodic systems and exploits the fact that the localisation constraints of the non-orthogonal Wannier functions (NGWFs) can be lifted. The second targets intermediate-sized systems and keeps the NGWFs localised, with k-dependence introduced via phase factors in the Hamiltonian and other matrix elements. These developements extend the operability of ONETEP to short- and intermediate-scale periodic systems, including 2D materials, nanowires, nanotubes, surfaces, and interfaces, and enable ONETEP to be benchmarked against other DFT codes by the Delta-codes DFT test.[3] The resulting delta value of < 1 meV/atom suggests that our method is not only reliable but also highly accurate.

ONETEP allows us to model not only the ground state electronic structure of large-scale systems, but also their excited state behaviour through linear response time-dependent DFT. However, higher level functionals (e.g. hybrids) are required for quantitatively accurate results, which significantly increases computational cost if applied to large systems. This can be remedied by using quantum embedding, where different levels of theory are applied to different regions self-consistently – something that ONETEP's local basis is ideally suited for – significantly reducing the number of atoms treated at the higher level of theory, and therefore computational cost. Here, we introduce the combination of embedded mean field theory (EMFT)[4] and linear-scaling DFT within ONETEP, including compatibility with TDDFT and implicit solvation.[5,6] We exemplify this on molecules (in solvent), carbon nanotube systems, and molecular crystals. These methods allow ONETEP to perform high level DFT calculations to be performed on previously inaccessibly large systems, and point the way towards including even more accurate methods within the ONETEP framework.

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A brief history of the early days of Theoretical Chemistry in Cambridge

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With the advent of quantum mechanics in the late 1920's, it soon became apparent that the subject of chemistry would be for ever changed by this (still-enigmatic) physical/mathematical theory. So a Theory group, lead by John Lennard-Jones, was set in the early 1930s in the chemistry department. Lennard-Jones contributed profoundly to the development of the new field, both through his own work, and through his doctoral students, who included Charles Coulson, Frank Boys and John Pople, to name but a few, and their PhD students... I will try to give a sense of the scientific background and work of these early pioneers, and the lasting impact they had in shaping the subject, both in the UK and internationally.

The Rise of Electronic Structure Calculations

Mike Payne

Cavendish Laboratory, University of Cambridge, Cambridge, UK.

Dirac famously stated 'The fundamental laws necessary for the mathematical treatment of a large part of physics and the whole of chemistry are thus completely known, and the difficulty lies only in the fact that application of these laws leads to equations that are too complex to be solved.' In this talk I shall present a personal, and somewhat Cambridge-centric, overview of the development of electronic structure calculations.

The concept of exact forces on the nuclei and other lessons from the exact factorization

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The exact factorization[1] writes the full molecular wave function as a single product of a nuclear wave packet and a many-electron wave function which parametrically depends on the nuclear coordinates and which has the meaning of a conditional probability amplitude. The variational principle then leads to formally exact equations of motion for these two wave functions. The nuclear equation is a standard time-dependent Schrödinger equation which contains a single time-dependent scalar potential energy surface and a vector potential which has the structure of a Berry connection. We will deduce Ehrenfest equations for the nuclear subsystem[2] which allow the identification of unique and formally exact forces on the nuclei. In the classical limit, one obtains Lorentz-type forces where the Berry curvature associated with the vector potential plays the role of a magnetic-like field, and one obtains an electric-like field given by the gradient of the scalar potential energy surface plus the time-derivative of the (Berry-connection) vector potential. Consequences of these unusual forces will be explored. In particular, for neutral atoms exposed to a constant magnetic field, the exact Berry connection cancels the external vector potential completely. Moreover, the exact Berry connection leads to an exact molecular geometric phase whose calculation does not invoke the adiabatic approximation. The value of this exact Berry phase can differ significantly from the standard (adiabatic) Berry phase. Cases will be presented where the adiabatic Berry phase is π while the true geometric phase is zero, implying that in such cases the standard Berry phase is an artifact.[3] For other systems, the value of the exact geometric phase turns out to be close to the adiabatic Berry phase.[4] Possibilities to measure the exact geometric phase will be discussed.[5]

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Electronic decoherence along a single nuclear trajectory

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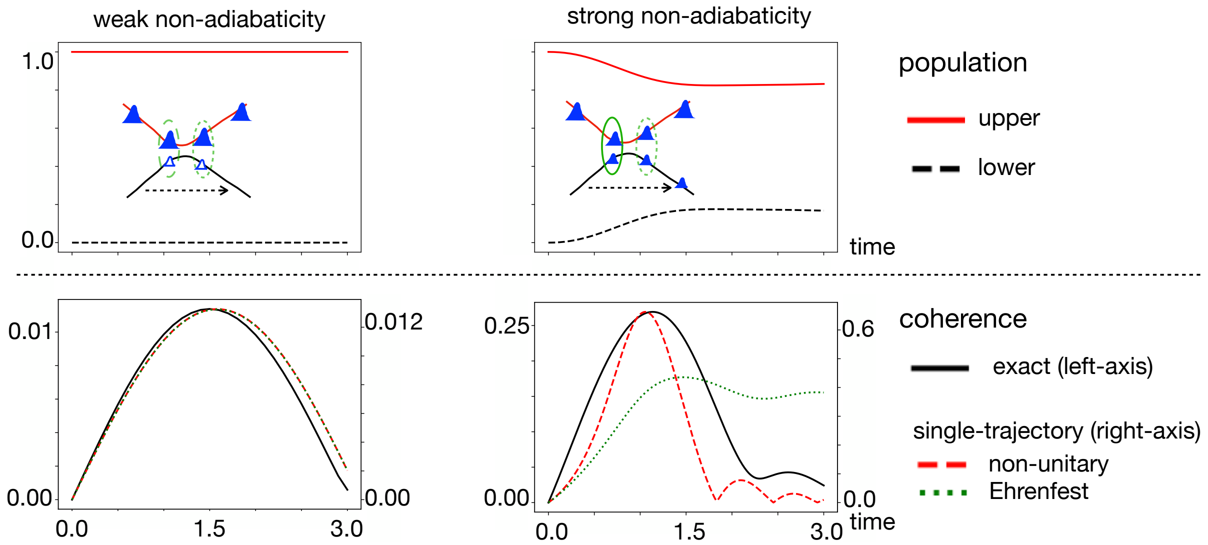
We describe a novel approach to subsystem decoherence without the usual tracing-out of the environment. The subsystem of focus is described entirely by a pure state evolving non-unitarily along a single classical trajectory of its environment. The approach is deduced from the exact factorisation framework for arbitrary systems of electrons and nuclei.[1] According to ”common wisdom”, consideration of electronic decoherence necessitates the finite spatial extension of the nuclear wave packets. Uniquely through the looking glass of the exact factorisation, without explicit inclusion of the finite spatial extension of the nuclear wave packets, one can closely investigate factors native to a single classical trajectory of the nuclei that are possibly responsible for electronic decoherence. Using this approach, first, we analytically show that the non-adiabatic correlations between electrons and nuclei dominate the non-unitarity of electronic subsystem's pure-state dynamics.[2] Second, using the prototypical example of a nuclear trajectory passing through an avoided crossing, where non-adiabatic electron-nuclei correlation play essential roles, we contrast our non-unitary approach with the full-unitary Ehrenfest dynamics, both work under a single nuclear trajectory. It's well known that the electronic coherence described by Ehrenfest dynamics as the nuclear trajectory passes through the avoided crossing fails to decay, namely, it is unable to describe decoherence. By seeing the consistency of this latter fact with the orthogonalisation-maintaining feature of the Ehrenfest dynamics' unitary aspect, we propose that the de-orthogonalisation process behind the non-unitarity of the pure-state electronic dynamics is responsible for decoherence. This thus establishes a single-nuclear-trajectory factor behind electronic decoherence, distinct from those factors that explicitly evoke the finite spatial extension of the nuclear wave packets.



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Quantum embedding of solid-state materials in the near-term quantum computing era

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Quantum computers hold the transformative potential to enable efficient quantum mechanical simulations of materials and their properties beyond what is feasible by their classical counterpart. However, current near-term quantum devices permit fully ab initio calculations on systems containing a few atoms only. To overcome the current limitations of quantum computers, and leverage their power, we have developed a hybrid quantum-classical periodic DFT embedding scheme by interfacing CP2K to IBM quantum devices through the Qiskit API.[1]

In this approach, the quantum computation is restricted to a localized region of a solid-state material selected in orbital space, while the remaining part of the system — the environment — is handled on a classical computer. This is particularly relevant when the localized region entails strongly correlated electrons and requires a description beyond the mean-field. This results in a hybrid algorithm, whereby the quantum device is used for obtaining the ground state of the embedded Hamiltonian, and the corresponding electron density is used to iteratively update the embedding potential on a classical computer until charge self-consistency.

First, we will demonstrate the accuracy of this new approach by showing the results obtained with noiseless simulations of optically addressable defects in MgO based on the variational quantum eigensolver. We have computed the absorption and emission band maxima of the neutral oxygen vacancy and obtained results on par with state-of-the-art approaches and experimental data.[1] Secondly, we will show how we have extended the embedding method to use the sample-based quantum diagonalization algorithm,[2] which allows us to run the embedding scheme on larger embedded fragments and noisy quantum devices. To demonstrate its applicability, we will present results obtained for the onset of magnetism in defective graphene.[3]

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Electronic structure and coherent states of spin defects in solids

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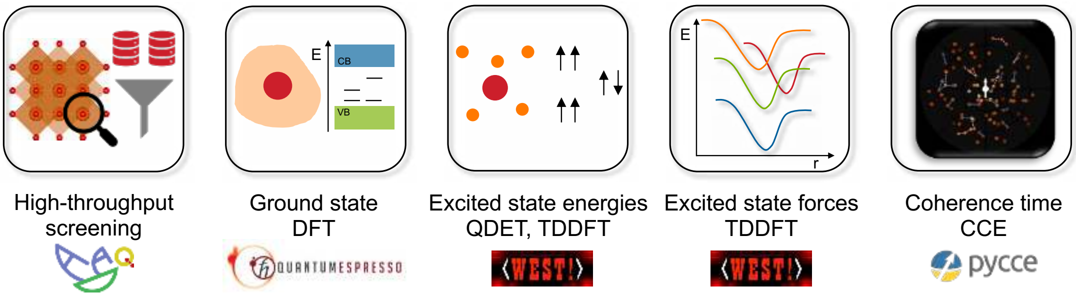
We present electronic structure calculations using time dependent DFT,[1] many-body perturbation theory, and quantum defect embedding theory[2] to predict the properties of spin defects for quantum sensing and quantum communication technologies. We also discuss simulations of the spin dynamics of spin defects, using closed and open quantum system formulations.[3] We then focus on oxide hosts, predicted to have long coherence times and thus offering the exciting prospect of designing defects and hosts that are compatible with current telecommunication and manufacturing infrastructure. In particular, we present our recent work on discovering and characterizing spin defects in calcium oxide (CaO),[4] magnesium oxide (MgO),[5] and cerium oxide (CeO_2) following the framework shown in the figure below. We identified an NV- center (similar to the NV- center in diamond) to be a promising spin defect in MgO, with several favorable ground and excited state electronic and optical properties, including spin triplet ground and excited states, large zero-field splitting, and singlet shelving excited states. We discuss the vibronic coupling in this system and its impact on optical properties. In addition, we discuss the progress made in computing the electronic structure and optical properties of Er^{3+}-doped CeO_2, which has been demonstrated to be a promising system for quantum communications applications.[6,7]



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Excitonic effects beyond textbook models

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Excitation spectra of valence electrons are often dominated by interaction effects, even in the absence of strong correlation. In this talk, we start by reminding how taking into account a screened electron-hole interaction in ab initio calculations allows one to understand excitonic effects in semiconductors and insulators that go beyond textbook exciton models.[1] We will then see examples where the electron-hole interactions can lead to surprising findings, both in the case of high and of low density materials.

In the first example, we uncover a striking interplay of the direct and exchange electron-hole interactions at large momentum transfer in solid helium.[2] This interplay leads to an unexpected and very strong angular dependence of the spectra that cannot be explained by single particle matrix elements. Most importantly, a prominent excitonic peak is entirely suppressed for specific directions of momentum transfer. We show that this effect manifests dramatically near Bragg points, where even infinitesimal changes in the direction of the wave vector can switch the excitonic peak on or off. Our analysis reveals that these angular effects stem from a strong anisotropy in crystal local field effects, which modulate the electron-hole interaction by orders of magnitude. Our findings shine new light on existing experimental data and allow us to predict when such an effect will occur in other materials.

In the second example, we will discuss exotic collective excitations that may be found in low-density metals and that are of excitonic origin, in spite of perfect macroscopic screening.[3] We show that ghost modes in the irreducible polarizability and low energy modes in the dynamic structure factor cannot be captured by the standard approximations to the electron-hole Bethe Salpeter equation. Instead, one has to take into account the screening that is appropriate for fermions, and not the one that shields classical charges. Approximate vertex corrections based on time dependent density functional theory allow us to overcome this limitation and to discuss the nature of the collective modes. In particular, we highlight the importance of the short-range region of the electron-hole interaction to explain these phenomena.

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Neural Wave Functions for Quantum Chemistry and Materials Physics

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Neural networks were first successfully used[1–3] to approximate many-electron wave functions of molecules in 2020 and have since produced impressive results for molecules, quantum dots, the electron gas, periodic solids, and nuclei.[4] Most electronic structure methods rely on uncontrolled approximations such as the choice of exchange-correlation functional in density functional theory or the form of the parameterized trial wavefunction in conventional quantum Monte Carlo simulations. Neural wave functions, by contrast, are built from multilayer perceptrons and/or transformers, which are universal function approximators. The weights and biases that define a neural wave function may be optimized efficiently without the use of externally generated data by combining variational Monte Carlo methods with automatic gradients calculated using back propagation. This talk will start with a general introduction to the field, moving on to discuss some very recent applications.

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Accurate and scalable exchange-correlation with deep learning

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Density Functional Theory (DFT) is the most widely used electronic structure method for predicting the properties of molecules and materials. Although DFT is, in principle, an exact reformulation of the Schrödinger equation, practical applications rely on approximations to the unknown exchange-correlation (XC) functional. Most existing XC functionals are constructed using a limited set of increasingly complex, hand-crafted features that improve accuracy at the expense of computational efficiency. Yet, no current approximation achieves the accuracy and generality for predictive modelling of laboratory experiments at chemical accuracy — typically defined as errors below 1 kcal/mol. In this work, we present Skala, a modern deep learning-based XC functional that bypasses expensive hand-designed features by learning representations directly from data. Skala achieves chemical accuracy for atomization energies of small molecules while retaining the computational efficiency typical of semi-local DFT. This performance is enabled by training on an unprecedented volume of high-accuracy reference data generated using computationally intensive wavefunction-based methods. Notably, Skala systematically improves with additional training data covering diverse chemistry. By incorporating a modest amount of additional high-accuracy data tailored to chemistry beyond atomization energies, Skala achieves accuracy competitive with the best-performing hybrid functionals across general main group chemistry, at the cost of semi-local DFT. As the training dataset continues to expand, Skala is poised to further enhance the predictive power of first-principles simulations

FEAT: Free energy Estimators with Adaptive Transport

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[b]Angstrom AI.

We present Free energy Estimators with Adaptive Transport (FEAT), a novel framework for free energy estimation – a critical challenge across scientific domains. FEAT leverages learned transports implemented via stochastic interpolants and provides consistent, minimum-variance estimators based on escorted Jarzynski equality and controlled Crooks theorem, alongside variational upper and lower bounds on free energy differences. Unifying equilibrium and non-equilibrium methods under a single theoretical framework, FEAT establishes a principled foundation for neural free energy calculations. Experimental validation on toy examples, molecular simulations, and quantum field theory demonstrates improvements over existing learning-based methods.

Latest adventures on quantum phase estimation and quantum dynamics experiments on an ion trap quantum computer

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Quantum computers are emerging as a promising platform for performing accurate electronic structure calculations that are currently unfeasible using standard computing systems. They are also expected to enable the scalable simulation of quantum dynamics, one of the most resource-intensive tasks in quantum chemistry. In this talk, I will lay out some basic concepts of the application of quantum computers to the simulation of molecules and materials at ab initio level. Then, I will show some results obtained in my team about electronic structure calculations for molecules and materials using the Quantum Phase Estimation algorithm, as well as quantum dynamics simulations using the Linear Combination of Unitaries method. I will finish with an overview of future developments in this field.

Quantum Monte Carlo methods and the CASINO program: applications to positronic materials and atomic gases

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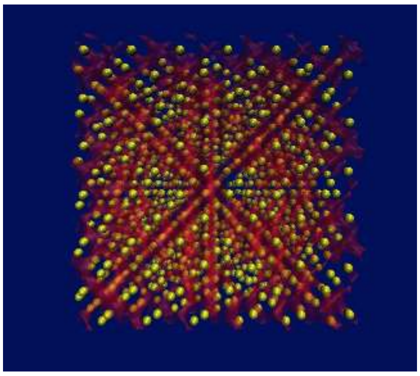
The CASINO quantum Monte Carlo (QMC) software[1] originated with the work of Richard Needs and Guna Rajagopal in the Cavendish Laboratory's Theory of Condensed Matter group in the early 1990s, and has gone on to be used in many hundreds of publications. In this presentation we give an overview of some recent developments and applications of CASINO, including work to support offloading to accelerators, the introduction of new ways of describing correlation in many-body wave functions, and better support for the simulation of particles other than electrons. We will focus on two topics of current interest: the use of QMC methods to study positrons in bulk materials, and the simulation of atomic gases using QMC.

Positrons provide a sensitive experimental probe of a material's electronic structure because the characteristics of the annihilation radiation directly encode the positron state, which is dependent on the local electronic environment. First-principles computational methods therefore play a fundamental role in the analysis and interpretation of positron annihilation spectroscopy data. Two-component density functional theory (DFT) is the most widely used method for modelling positrons in materials, so it is crucial to investigate and improve the accuracy of this technique. We have used QMC to develop electron–positron correlation functionals by simulating positrons immersed in homogeneous electron fluids,[2] and more recently we have developed the ability to perform *ab initio* QMC simulations of positrons in crystalline solids.[3,4] Parameter-free QMC delivers more accurate positron lifetimes and Doppler-broadening profiles of annihilation radiation than even the most refined semi-empirical two-component DFT approaches.



Positronic charge density occupying interstitial regions in bulk Si.

Ultracold systems of fermionic atoms in optical traps are currently of significant interest as quantum simulators of electronic systems, but there is a need to develop theoretical and computational approaches to support experimental work. We describe QMC simulations of homogeneous two-dimensional fluids of fermionic cold atoms interacting via dipolar potentials. These simulations provide energy functionals that will enable the use of DFT to model inhomogeneous cold atomic systems with dipolar interactions.

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Hydrophobicity from molecular diffusion at the solid-liquid interface

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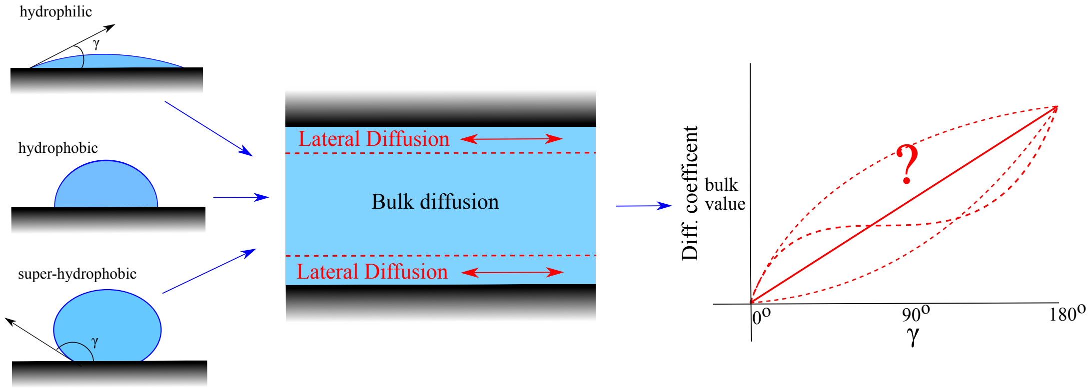
A quantitative assessment of wetting properties is essential for applications in material science and biology. However, this measurement often depends experimentally on the evaluation of the contact angle between a water droplet and the underlying surface. Several factors affect the water contact angle (WCA),[1] making it quite unreliable for quantitative usage, especially in nanotechnology applications, where nano- and sub-nano features strongly modify interfacial interactions, or where the assessment is completely hampered due to the nano-resolution geometry of the environment. This problem also persists in molecular dynamics simulations, where evaluating a realistic droplet is limited by system size (a few nanometers for droplet diameter), droplet relaxation time (tens of nanoseconds to milliseconds), and surface-water interaction availability (ad hoc parametrization from ab initio data is usually necessary).[2]

I present a new paradigm that overcomes these problems by quantitatively estimating wettability based on molecular diffusion near the surface. I show that the parallel component of molecular diffusion with respect to the surface (calculated from the mean square displacement of molecular dynamics trajectories) uniquely relates to the water contact angle.[3] This is possible by assuming that the interfacial change in water's free energy when in contact with a surface has a pure entropic contribution.[4] I will prove that this assumption is valid.

Furthermore, I will demonstrate how this assumption can be used to derive an analytical expression based on physical principles relating the WCA to interfacial diffusion. Finally, I will validate the accuracy of this new law through simulations of water interacting with surfaces of increasing hydrophilicity.



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Deciphering the Structural and Electronic Properties Across the Gold–Water Interface

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Metal-water interfaces play a crucial role in electrochemical reaction mechanisms relevant to energy storage and conversion devices. However, since these interfaces are buried between solid and liquid phases, it remains challenging to experimentally resolve the intrinsic structural and electronic properties of such electrified interfaces, in particular those of the interfacial water molecules. As a result, their fundamental nature is still far from being fully understood up to being controversial.

In this work, we quantify the structural and electronic interactions at the gold-water interface. Our findings reveal that the dipole moments and electronic properties of water molecules near the metal surface can vary significantly depending on their orientation and hydrogen-bonding states.

By employing a novel efficient implemention for localizing molecular orbitals in hybrid systems such as water interacting with a metallic surface via partially occupied Wannier functions—which allows us to compute the contributions of individual water molecules to the total interfacial electronic dipole—we uncover that water molecules in close proximity to the gold surface exhibit significantly increased dipole moments. We disclose that this is due to distinct polarization of those electron lone pairs which point toward the metal surface, thus belonging to water molecules with their O-H bonds pointing toward the water layer.

Additionally, we present evidence of electronic polarization and charge transfer effects in vibrational spectra through peak broadening and frequency shifts of dangling O-H bonds near the metal surface, as observed in both sum-frequency generation (SFG) spectroscopy and fully atomistic ab initio molecular dynamics simulations.

Overall, the strong electronic interactions between metal and water that we reveal give rise to orientation-dependent electronic heterogeneity. Our findings provide new insights into the fundamental properties of metal-aqueous interfaces with broad implications for electrochemistry, catalysis, and energy-related applications.

Multiscale modeling of electrochemical CO_2 conversion

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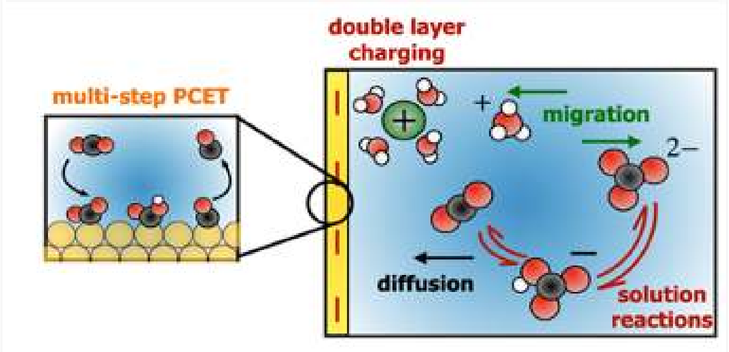
Electrochemical conversion of renewable inputs such as CO_2 has the potential to decarbonize the production of fuels and high-value chemicals. Electrocatalysts are the cornerstone to such conversion processes as they enable efficient and selective transformations. As a result, it is essential to develop electrocatalytic interfaces with high intrinsic activity, selectivity and stability to enable widespread implementation of electrochemical CO_2 conversion technologies.[1] In this talk, I will discuss our work on using multiscale simulations involving atomistic, microkinetic and mass transport modeling to understand electrochemical CO_2 conversion on Cu and Au surfaces. I will highlight the importance of active site identity,[2] surface coverage,[3] electrochemical kinetics and transport,[4,5] and electrolyte effects [6] in understanding electrode stability, reaction mechanisms and product selectivity, and the need for joint theory-experiment efforts to fully uncover this complex multi-step process.



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Modeling Earth from Atomic to Global Scale

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Geophysics relies on a synergistic interplay between seismology, geodynamics, and mineral physics, advancing through close collaboration between these computationally intensive fields. Mineral physics provides essential materials properties for interpreting seismic tomography and refining geodynamic simulations. Materials simulations have played a pivotal role in integrating these disciplines, complementing experiments by extending the pressure-temperature range of accessible data and revealing atomic-scale phenomena that revise experimental and seismological interpretations.

I will highlight recent mineral physics contributions to geophysics using advanced materials simulation methods. This interdisciplinary dialogue has fostered a modeling approach that bridges atomic to global scales. A striking example is the spin state change in iron oxide alloys under pressure, a quantum phenomenon that leaves a visible imprint on global seismic tomographic models.

Poster Abstracts

Proton Behaviour and Water Self-Dissociation: From Graphene–Water Interfaces to Nanodroplets

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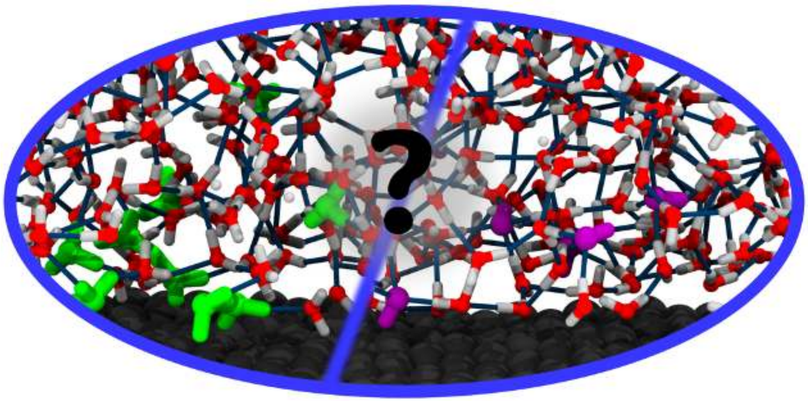
Water's ability to autoionize into hydroxide and hydronium ions profoundly shapes interfacial chemistry, rendering surfaces effectively acidic or basic. While proton affinity at the air–water interface is well known, far less is understood at solid–liquid interfaces. Here, I will present machine learning-based molecular dynamics simulations with first-principles accuracy that reveal self-dissociation behavior at the graphene–water interface. We find that protons preferentially accumulate at the interface, while hydroxide exhibits a bimodal distribution between surface and bulk.[1] Extending this to confined nanodroplets, we observe that confinement enhances dissociation, with the extent and mechanism strongly dependent on the confining material. In graphene droplets, dissociation occurs without chemical alteration of the substrate, whereas in hBN, hydroxide can show chemisorption, directly coupling water chemistry to the material. These findings highlight how nanoscale confinement and surface chemistry together regulate proton behavior, with implications for aqueous reactivity, ion transport, and functional interface design.



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DFT+U Study of Magnetic Configurations and 5f Electron Behaviour in UAu_2

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UAu_2 is a uranium-based compound where a frustrated stacked triangular lattice of uranium atoms leads to rich and unusual magnetic behaviour, including an incommensurately modulated antiferromagnetic state that exhibits non-Fermi liquid characteristics,[1,2] a field-induced transition to a ferrimagnetic state,[2] as well as the emergence of multicomponent odd-parity superconductivity at high pressure.[3] In this project, the DFT+U method was used to explore how different magnetic configurations (non-magnetic, ferromagnetic and ferrimagnetic) affect the structure and electronic properties of UAu_2. Structural optimizations and total energy comparisons across a range of Hubbard U values were carried out to assess the relative stability of these configurations, indicating that the ferrimagnetic state is likely the most stable. We also examined the band structure and density of states, where the uranium 5f electrons show strong sensitivity to U, becoming more localized and spin-polarized with increasing values. These changes significantly impact hybridization with the gold 5d states. In addition, the Fermi surface of the ferrimagnetic state is compared to quantum oscillations observed in recent measurements. While this work focuses on UAu_2, it also lays the foundation for exploring other strongly correlated systems. By combining theoretical predictions with experimental observations, this study aims to deepen our understanding of the complex behaviour found in uranium-based heavy fermion materials.

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Modelling of CO_2 electroreduction catalysed by small copper clusters: the effect of cluster size on product distribution

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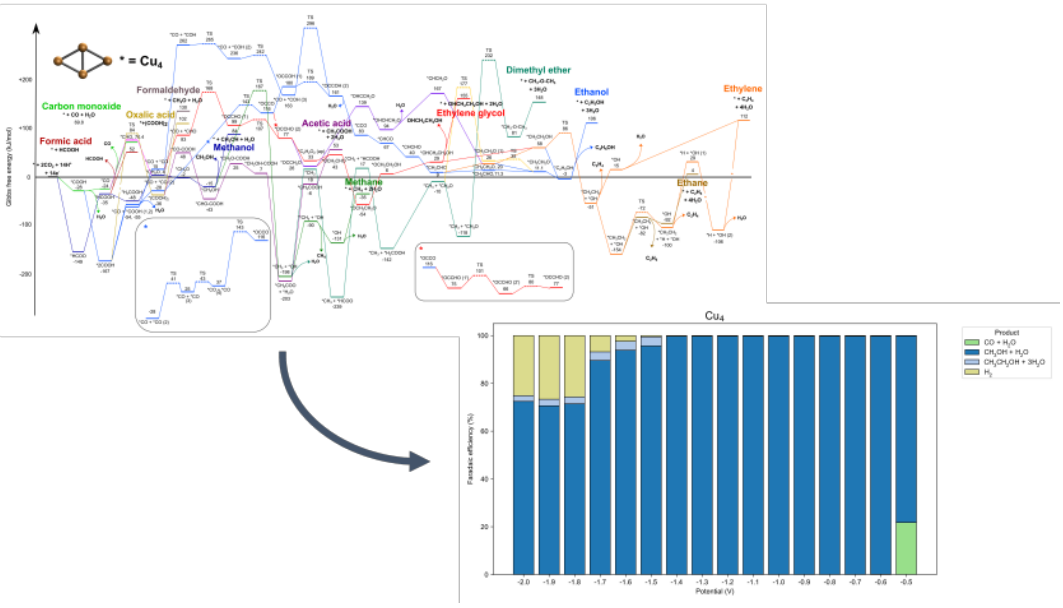
The rising atmospheric CO_2 concentration is a global challenge, and its mitigation through CO_2 capture and conversion into valuable chemicals (e.g., methanol, ethanol) represents a promising strategy. However, the development of efficient catalysts remains a key research focus. Small copper clusters have recently attracted interest due to their catalytic activity in CO_2 reduction.[1–3] Advancing catalyst design requires a deeper understanding of the reaction mechanism.

In our DFT-based study, we investigated the CO_2 electroreduction mechanism toward C_1 and C_2 products, focusing on the size dependence of Cu_n clusters (n = 3–6, 12). A microkinetic model was subsequently developed to interpret the complex mechanisms identified, and the effect of catalyst support was investigated using a graphene-Cu_4 model system.

Our results indicate that the ground-state spin multiplicity of the clusters plays a key role in C_1 selectivity: methanol is the main C_1 product on Cu_4, Cu_6, and Cu_{12}, while Cu_3 and Cu_5 may also yield methane. Among C_2 products, ethanol formation appears most likely across all cluster sizes, though the required cell voltage varies considerably. On Cu_4 and Cu_{12}, product formation begins at relatively low potentials, favouring C_1 products on Cu_4 and C_2 products on Cu_{12}.



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When Are Dopant d-States Free-Atom-Like? Periodic Trends and Confinement Effects in Single-Atom Alloys

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The narrow, free-atom-like d-states of dopants in single-atom alloys (SAAs) have been widely regarded as a defining feature responsible for their remarkable catalytic selectivity.[1,2] However, whether this property is universal across all transition metal dopants remains an open question. Here, we use density functional theory (DFT) calculations to systematically investigate the d-band characteristics of 4d transition metals embedded in various metallic hosts. We find that only late transition metal dopants exhibit truly narrow, free-atom-like d-states. In contrast, early dopants exhibit significantly broader d-bands, not because of stronger hybridization with the host, which remains minimal, but due to their more spatially extended orbitals and greater confinement within the host lattice.

We identify key physical parameters that govern d-band width, including dopant confinement, spatial extent of d-orbitals, and the filling of d-states. These factors also impact dopant spin states, with reduced confinement stabilizing high-spin configurations. Building on these insights, we establish design principles for tuning dopant electronic structure in SAAs and predict d-band widths with high accuracy using only four simple structural and electronic descriptors.

Our findings clarify ambiguities in the literature, demonstrate that the free-atom-like behavior is not universal, and show how d-band shape modulation directly influences catalytic selectivity. These insights provide additional levers in rational SAA design by showing that catalyst performance can be tuned not only through the choice of dopant element but also by engineering the local environment of the active site to modulate the dopant's electronic structure.

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Accelerating Ab-Initio calculations of Superconducting Transition Temperatures

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First-principles computational predictions of the electron-phonon coupling strength λ and superconducting critical temperature T_c have underpinned the discovery of several high T_c hydride superconductors, including H_3S ($\approx$ 200 K)[1] and LaH_{10} ($\approx$ 260 K),[2] and have recently theorised Mg_2IrH_6 as a promising candidate.[3,4] These calculations, which involve self-consistently solving the Eliashberg equations, require Brillouin zone integration of the electron-phonon matrix elements on dense electron grids, rendering them computationally intensive and presenting a bottleneck for high-throughput materials screening. In this work, we present a novel method to accelerate the convergence of T_c and λ with respect to the electronic grid, tested on a range of conventional superconducting systems. Even on coarse grids, the method is shown to reproduce converged results for λ to within 20% for most systems, resulting in up to two orders of magnitude speed-up in ideal cases. The improvement is particularly pronounced for high T_c superconductors with features that necessitate fine Brillouin zone sampling, which would typically be missed in a standard high-throughput search. Our approach is easy to integrate into existing materials screening workflows, to enable rapid identification of promising candidates that might otherwise be overlooked.

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Tunable π-magnetism in carbon-based nanostructures

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Harnessing the quantum properties of π-magnetic nanostructures holds great promise for the development of next-generation molecular spintronic and quantum devices.[1] Among these, triangulene, a non-Kekulé polycyclic hydrocarbon, is particularly notable for its intrinsic open-shell character and its ability to host stable unpaired spins.[2] However, achieving strong and tunable magnetic interactions between molecular units remains a key challenge.[3]

In this work, we explore a chemical tuning strategy by introducing topological defects and heteroatom doping into triangulene dimers. Using broken-symmetry DFT and high-level wavefunction methods, we calculate magnetic coupling constants (J) and spin polarization energies (ΔE_{spin}) and their spin density distributions. Our results show that both the connectivity and the chemical composition of the bridging units critically affect the ground-state spin and the strength of coupling. We identify stable high-spin ground states with ferromagnetic exchange coupling up to 260 meV, highlighting the potential of rational chemical design in tailoring π-magnetism.

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Bayesian Optimization-Guided Discovery of Optimal OH Adsorption on High-Entropy Alloy Catalysts

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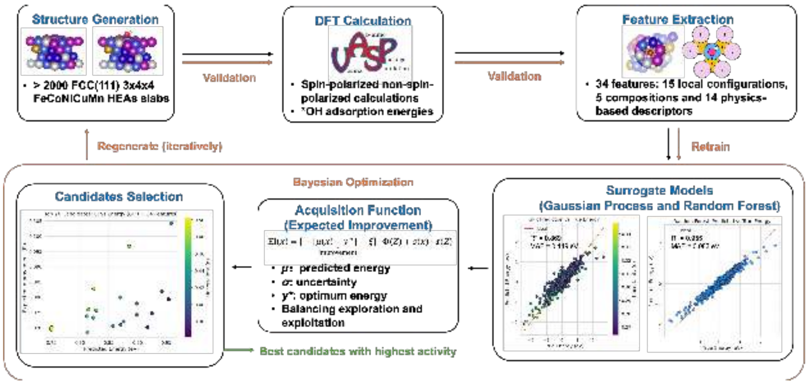
To identify high-performance high-entropy alloy (HEA) catalysts for oxygen reduction reaction (ORR), we screen for optimal compositions and atomic configurations for the adsorption energy of *OH on quinary FeCoNiCuMn, combining density functional theory (DFT) and machine learning (cf. figure below). Both spin-polarized and non-spin-polarized DFT calculations are performed, revealing that spin-polarization consistently leads to more negative adsorption energies, highlighting the critical role of magnetism in surface reactivity. Using 34 structural (local configurations and compositions) and physics-based descriptors (e.g., Bader charge), we construct a surrogate model and applied Bayesian optimization (BO) to iteratively refine the candidate compositions via acquisition functions balancing exploration and exploitation strategies. In particular, 10 nearest neighbors around the adsorption sites are fixed whereas the other 38 atoms in the supercells used in DFT calculations are randomized. Based on more than 2000 calculations, it is observed that there exist significant variations in the *OH adsorption energies across these configurations, confirming substantial influence of long-range structural and magnetic interactions. Our BO+DFT approach can efficiently sample both the atomic configurations and chemical compositions for the optimal ORR activity. This framework provides a robust and data-efficient strategy for rational HEA catalyst discovery.



Coexisting charge density waves in twisted bilayer $NbSe_2$

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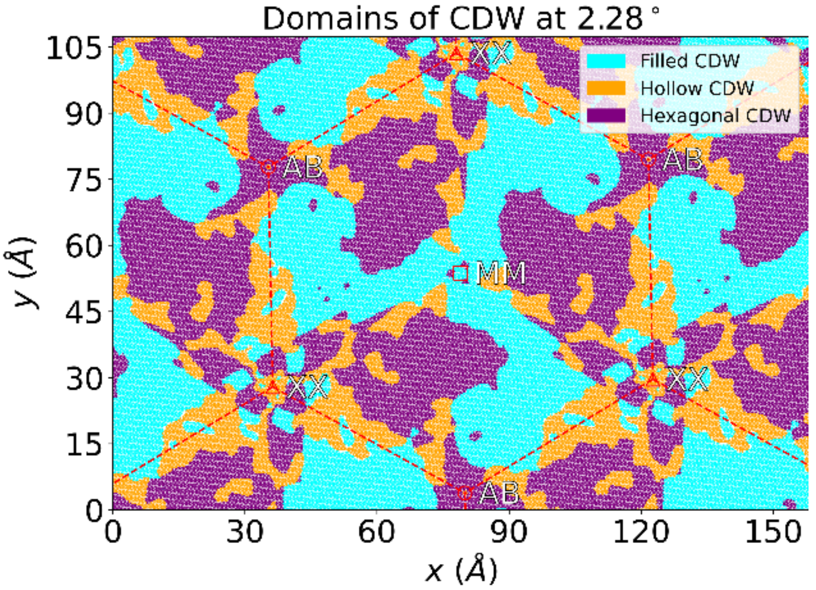
Twisted moiré bilayers are formed by stacking and rotating two monolayer materials. Whilst moiré bilayers of semiconducting transition metal dichalcogenides (TMDs) such as $MoSe_2$ have been widely studied, there has been less attention on those composed of metallic TMDs such as $NbSe_2$.

In its monolayer form, $NbSe_2$ hosts superconductivity and different types of charge density waves (CDWs), which are atomic reconstructions that increase the unit cell size. In this work we address the question of whether and how these CDWs coexist with the long-range relaxations inherent to moiré systems.

We have carried out large-scale first-principles relaxations based on density functional theory to study charge density waves in twisted bilayer $NbSe_2$ at twist angles down to 2.28 degrees. We have also developed new methods to disentangle the smaller and more local CDW displacements from the larger and more long-ranged moiré relaxations, and for locally identifying the type of CDW formed.

We find that domains of different types of CDW coexist within the large moiré unit cell. The formation of the domains is due to the interaction with the strain induced by moiré relaxations. Our work provides insight into the interaction between moiré and CDW relaxations.[1]



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Integrating machine learning and artificial intelligence with classical simulations for automating materials discovery.

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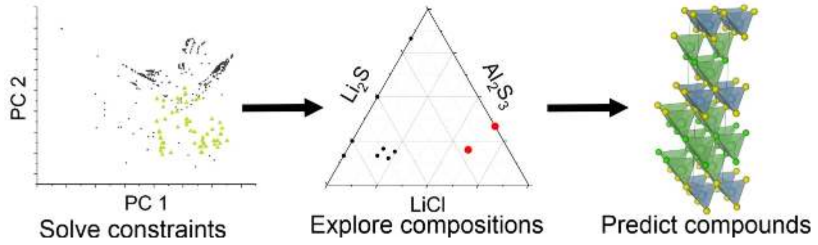
In the grand challenge of discovering new inorganic solids numerous tools continue to emerge for the prediction of compositions, chemical structures and physical properties, many of which are using recent advances in machine learning (ML) and artificial intelligence (AI), while still being underpinned by density functional theory calculations (DFT). In this poster I will discuss our work from two recent publications, where we are developing our own ML and AI driven tools and deploying them in workflows to guide the exploration of new materials.

1. The integration of generative machine learning with heuristic crystal structure prediction.[1] We present a new implementation of our crystal structure prediction method FUSE, where the method is adapted to use both generative machine learning models to generate trial crystal structures & machine learnt inter atomic potentials. The combination of the introduction of the two ML methods results in a speed up in calculations of up to 8 times, with no loss in the accuracy of energy calculation compared to pure DFT, while also obtaining lower energy structures.

2. The exploration of chemical with automatable workflows. We present a new automated reasoning tool COMGEN,[2] which allows a user to specify sets of chemical constraints and then explore the valid compositions which are a consequence of them, thus providing explainable results. Numerous constraints are available in COMGEN, including (but not limited to); minimum distance from a given set of known chemical compositions, limits on chemical composition, number of atoms within the formula unit, and restricting the search to compositions which are accessible from a given set of starting materials. We then integrate this into a workflow, where we perform probe structure prediction with FUSE[2] and DFT on compositions suggested by COMGEN and use composition only ML properties prediction to identify several candidate compounds for investigation as electrolytes in all solid-state Li batteries.



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Materials Discovery with Ephemeral Data Derived Potentials

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The development of Machine Learned Interatomic Potentials (MLIPs) has revolutionised atomistic simulations over recent years. MLIPs enable simulations with accuracy comparable to quantum mechanical approaches at up to 10 million times smaller computational cost. The ephemeral data derived potentials (EDDPs[1,2]) are a flexible class of MLIP that can utilise comparatively small neural networks for fast and accurate simulations. Materials discovery techniques, such as random structure searching, can exploit this acceleration in a number of ways. Larger system sizes and chemical spaces can be explored, or more computationally intensive structure optimisers, utilising molecular dynamics, can incorporated[3]. Through a range of applications, we demonstrate that more complex optimisers can improve search performance for large systems. Through example simulations in LAMMPS[4], we further show that the EDDPs can be employed beyond structure searching, to explore complex dynamical behaviour across a range of conditions and chemical systems.

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Energy Landscapes for Membrane Proteins using NMR based Hybrid Restraint Potentials

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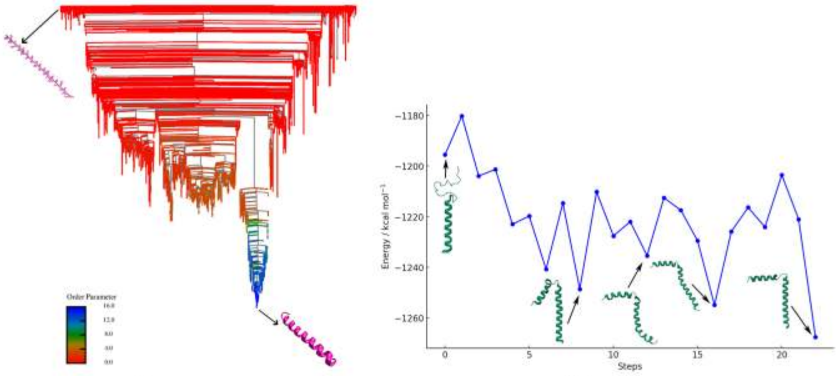
Understanding the relationship between a protein's three-dimensional structure and its function is a prime cornerstone of structural biology. Nuclear Magnetic Resonance (NMR) spectroscopy is a key experimental method used to determine protein structures. As of June 2025, it has been employed in resolving 12,645 out of the 204,506 protein structures archived in the Research Collaboratory for Structural Bioinformatics Protein Data Bank (RCSB PDB), accounting for approximately 6.2% of the total entries.[1] The use of acquired NMR data as constraints to infer the structural and orientational characteristics of the protein's native state lies at the heart of the entire procedure.

Our research focuses on the simultaneous utilization of multiple protein NMR measurements, specifically chemical shift, dipolar coupling and inter-atomic distances, in conjunction with appropriately weighted force field potentials to enhance structure prediction and energy landscape analysis using basin-hopping and discrete path sampling. We employ Napshift,[2] an in house developed artificial neural network, and bond-length based physical models to respectively compute chemical shift and dipolar coupling values from the sequence and structure of the biomolecule at each sampling step of the simulation. Hybridization of these constraints as penalty energies with empirical force fields facilitates the biasing of sampling towards native states and the exclusion of incompatible configuration space. Our optimized hybrid restraint potentials thus contribute to the enhancement of biomolecular simulations and other computational methods reliant on the accuracy of foundational force fields.

The systems under study encompass integral membrane proteins, including sarcolipin, and phospholamban. NMR restraint hybrid potentials provide a reliable way to account for their environments thereby preventing the need to use explicit or implicit solvation and lipid membrane molecules. The figure shows some results obtained for (a) the folding pathway for sarcolipin in the absence of NMR constraints, colored according to the number of hydrogen bonds stabilizing the secondary structure, and (b) the conversion pathway of phospholamban between its two stable states. Our findings underscore the efficacy of hybridizing multiple constraints with empirical biomolecular force fields to enhance the landscape analysis approaches for membrane proteins. Moving forward, our research will center on harnessing additional NMR observables, such as residual dipolar couplings and heteronuclear nuclear Overhauser effect (hetNOE), to derive additional restraints and further refine our methodology.



(a) (b)

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First-principles phase behaviour of nanoconfined water at extreme conditions

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Understanding the behaviour of water under nanoscale confinement is crucial in various industrial and scientific contexts, ranging from catalysis to planetary science and nanofluidics. Experiments have revealed a variety of unusual properties in confined water, such as ferroelectric ordering and an unusually low dielectric constant, highlighting its distinct nature compared to bulk water.

Even more exotic behaviour is expected when water is subjected to extreme pressures at the nanoscale, where reduced interatomic distances are likely to induce fundamental changes in its structural and electronic properties. Yet, this regime remains largely unexplored—both experimentally and theoretically. In particular, a deeper understanding of the electronic structure in such conditions could offer valuable insights into charge transport, defect formation, and a potential transition to a metallic state.

This work addresses this knowledge gap by combining state-of-the-art computational tools, ranging from ab-initio electronic structure methods, machine learning potentials to molecular dynamics, to explore confined water with first-principles accuracy over realistic time and length scales. We focus on a bilayer phase of water within a graphene-like channel, investigating its behaviour under increasing pressure, from 10 GPa up to the high-pressure regime of 250 GPa.

Our simulations reveal the emergence of novel phases at high pressures, including a non-molecular, superionic, hexagonal bilayer phase. Notably, this phase exhibits signs of potential metallisation, with a predicted band gap closure at GGA level. This suggests that, unlike bulk water, nanoconfined water under pressure may transition into a metallic phase.

Machine Learnt Interatomic Potentials for High-Throughput Phonon Calculations of Metal-Organic Frameworks

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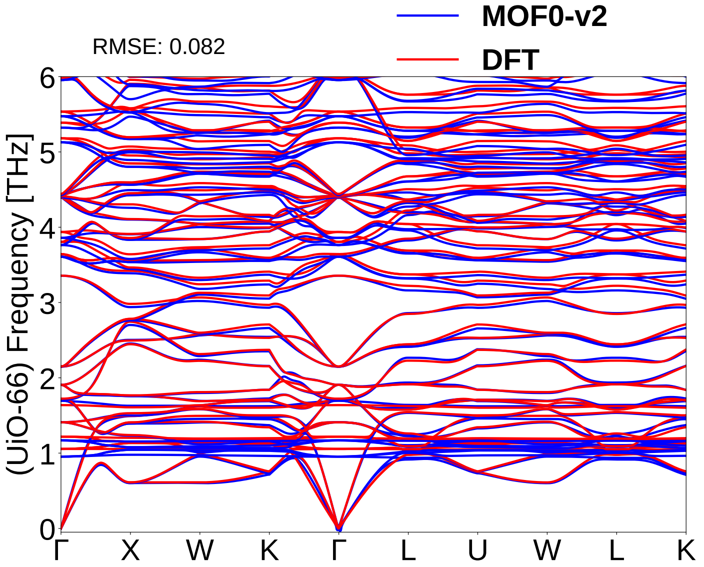
Metal–organic frameworks (MOFs) are highly porous and versatile materials studied extensively for applications such as carbon capture and water harvesting. However, computing phonon-mediated properties in MOFs, like thermal expansion and mechanical stability, remains challenging due to the large number of atoms per unit cell, making traditional Density Functional Theory (DFT) methods impractical for high-throughput screening. Recent advances in machine learning potentials have led to foundation atomistic models, such as MACE-MP-0, that accurately predict equilibrium structures but struggle with phonon properties of MOFs. In this work, we developed a workflow for computing phonons in MOFs within the quasi-harmonic approximation with a fine-tuned MACE model, MACE-MP-MOF0. The model was trained on a curated dataset of 127 representative and diverse MOFs. The fine-tuned MACE-MP-MOF0 improves the accuracy of phonon density of states and corrects the imaginary phonon modes of MACE-MP-0, enabling high-throughput phonon calculations with state-of-the-art precision. The model successfully predicts thermal expansion and bulk moduli in agreement with DFT and experimental data for several well-known MOFs. These results highlight the potential of MACE-MP-MOF0 in guiding MOF design for applications in energy storage and thermoelectrics.[1]



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Electrode potential dependent differential capacitance in electrocatalysis: a novel, ab initio computational approach

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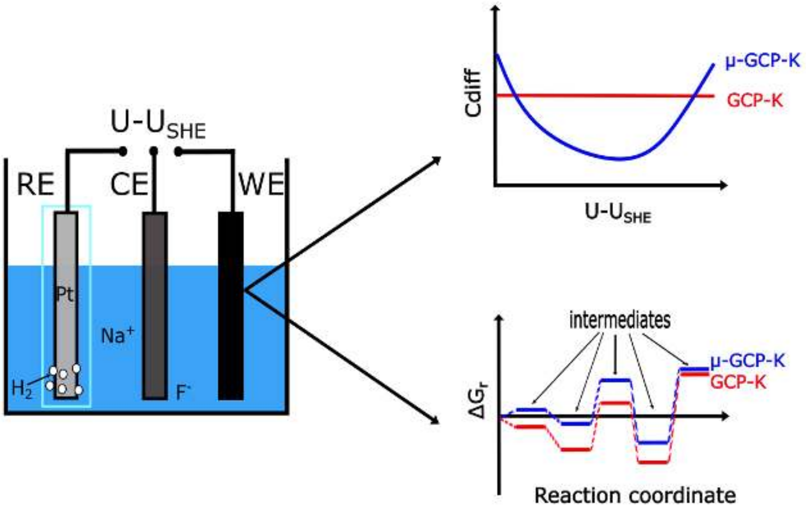
With the growing interest in nanomaterials, the role of ab-initio simulations has become increasingly important in the design of electrochemical catalysts. As electrochemical reactions are driven by electrode potential, leading to variations in electron number, the utility of the grand-canonical representation becomes evident, particularly with its integration into the formalism of Density Functional Theory, referred to as GC-DFT.[1] The purpose of the Grand Canonical Potential - Kinetics (GCP-K) method[2] is to determine the grand canonical potential based on constant charge computations and quadratic charge-thermodynamic free energy relation. This implies a linear relationship between electrochemical potential and charge, which might not always be a valid approximation depending on the electronic structure of the electrode.[3] We developed a new approach, which we call μ-GCP-K, that is based entirely on the electronic structure, without any restriction on the charge-free energy relationship, making it more broadly applicable than the GCP-K. Rather than assuming a quadratic relationship for free energy in charge as in GCP-K, we applied interpolation to accurately capture the charge dependence of the chemical potential and free energy, while also determining the correct form of the grand canonical potential. We demonstrate the efficiency of our method by computing the differential capacitance and highlighting the significance of accurately calculating the grand canonical potential. In summary, we present a more general and precise approach that incurs no increase in computational cost, enabling the design of novel electrocatalysts and enhancing comparability with experimental results.



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Multiscale Simulation of Hydroxide Ion Transfer in Anion-exchange membranes: Comparing Classical and Machine Learning Approaches

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We present two complementary multiscale strategies to model hydroxide-ion transport over the length and time scales relevant to practical anion-exchange membrane (AEM) applications. Our first method harnesses machine-learned interatomic potentials, fine-tuned on high-level quantum-chemical data, to reproduce hydroxide dynamics with near-*ab initio* accuracy at time scales unattainable by direct AIMD simulations.[1] Starting from the MACE-MP-0 foundation model, we develop a systematic protocol that yields excellent agreement with AIMD reference data and even captures complex phenomena such as hydroxide–hydroxide hydrogen bonding at elevated concentrations. Our second framework couples classical molecular dynamics with lattice Monte Carlo, extending simulation times from picoseconds to milliseconds while retaining the essential quantum-chemical features of the Grotthuss mechanism.[2] This hybrid scheme not only matches the predictive accuracy of machine-learned potentials but also affords clear mechanistic insight: by decomposing the overall conductivity into proton-transfer kinetics (between hydroxide and water) and oxygen-framework rearrangements, we directly quantify how O–O separation governs hydroxide mobility. Both methods accurately reproduce experimental hydroxide conductivities in aqueous solutions across a range of concentrations and temperatures, with computational efficiency that permits application to complex membrane environments. We further demonstrate their utility on an anion-exchange membrane: benchmarking our fine-tuned MACE model against AIMD, we exploit extended length- and time-scale sampling to converge local transfer rates and map hydroxide migration pathways as a function of hydration level. By capturing mechanistic details over all relevant scales, our multiscale frameworks provide powerful, predictive tools for the design of next-generation hydroxide-conducting membranes.

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Benchmarking Short- and Long-ranged Machine Learning Potentials for Aqueous Interfaces

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Machine Learning Potentials (MLPs) enable large-scale molecular dynamics simulations with near ab initio accuracy. However, accurate modelling of interfacial systems remains challenging as their behaviour is often governed by long-range electrostatic interactions - features that are hard to capture with the inherently short-ranged architecture of MLPs. Using aqueous NaCl as a model system, we analyse the performance of long- and short-range MLPs trained on density functional theory data in bulk water, at the air–water interface, and at the graphene–water interface. To probe slowly converging properties such as diffusion coefficients and ion/water density profiles in slabs, we also train models on classical force fields, benchmarking against long-timescale simulations. We highlight the capabilities of a locally charge-conserving MACE model towards accurately reproducing both static and dynamic properties and implicitly learning water dipoles.

Quantum Simulations of Chemistry in First Quantization with any Basis Set

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Quantum computation of electronic energy is one of the most promising applications of quantum computers. Previous work has mainly focused on quantum algorithms where the Hamiltonian is represented in second quantization with compact basis sets while existing methods in first quantization are limited to plane waves. In this talk, we present a new method[1] to solve a generic ground-state chemistry problem in first quantization using any basis set. We achieve asymptotic speedup in Toffoli count for molecular basis sets, and orders of magnitude improvement using real space grids as compared to the second quantization. In some instances, our approach requires similar or even lower resources compared to previous first quantization plane wave algorithms. The developed methodology can be applied to variety of applications, where matrix elements of a first quantized Hamiltonian lack simple circuit representation.

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Digital infrastructure for machine learnt interatomic potentials

Elliott Kasoar[a,b], Patrick Austen[a], Harvey Devereux[c], Kieran Harris[d], Joe Hart[b], David Mason[e], Jacob Wilkins[a], Federica Zanca[a*], Alin M. Elena[a], Gábor Csányi[b]

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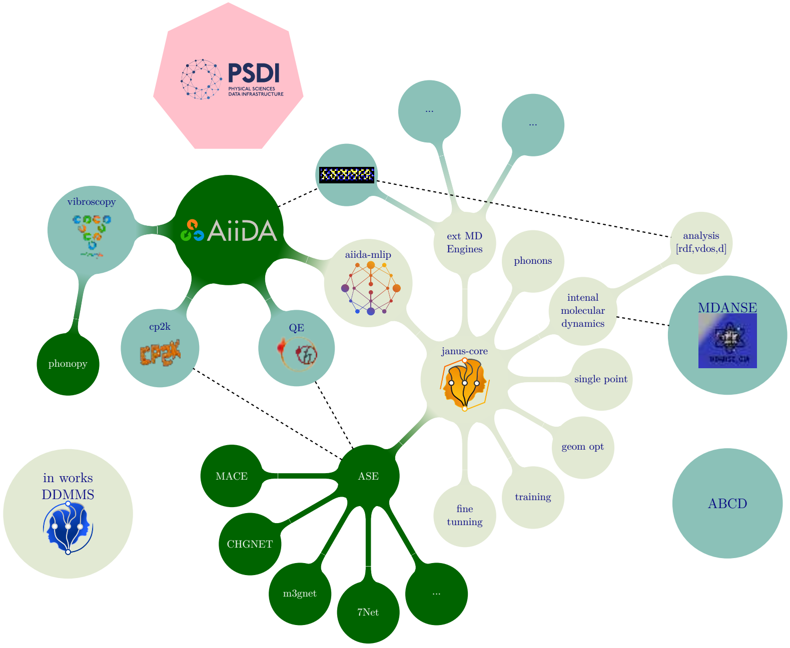
Recent advances in machine learnt interatomic potentials (MLIPs) are revolutionising atomistic simulations, enabling results with accuracy comparable to ab initio calculations at significantly greater time and length scales. However, in order for researchers to be able to take full advantage of these advances, a software framework is needed to facilitate scientific benchmarking, training and fine-tuning of MLIPs, as well as to enable their integration into simulation workflows to study properties of interest. To address this need, we introduce (a) janus-core,[1] a Python package that provides tools to train, fine-tune, and perform a range of calculations with MLIPs; (b) aiida-mlip,[2] an AiiDA plugin to provide full provenance tracking when using janus-core; (c) an OpenSearch implementation of the abcd[3] database, to provide storage, summary, and discovery of atomistic data required for training MLIPs; and (d) mlip-testing,[4] a benchmarking framework to develop, run, and visualise an automated, modular, hierarchical test suite for MLIPs.



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What is the best density functional for adsorption?

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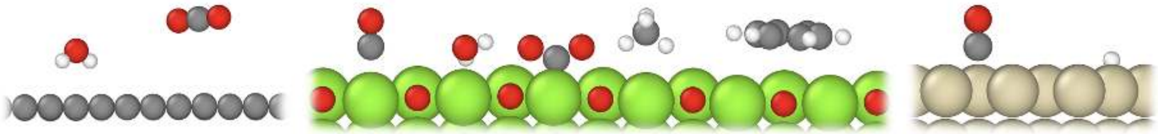
Despite the widespread use that density functional theory has found in the field of surface science, there is a distinct lack of applicable benchmarks. We report a diverse set of benchmark data examining the accuracy of modern density functional approximations (DFAs) for adsorption of molecules on surfaces. This includes covalent, ionic and metallic surfaces with diverse adsorbates, and a relevant selection of DFAs including the popular ωB97X-V and ωB97M-V by Head-Gordon et al. In line with previous indications, we find that the current hierarchy of DFAs seen for molecular main-group and transition-metal chemistry does not directly translate to surface chemistry. Taking statistics over the considered test set, we thus present a data-informed answer to the question of which is the best general-purpose DFA for adsorption on surfaces.



Towards Practical State Preparation for Hamiltonian Simulation in Chemistry

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Quantum computers have originally been proposed as tools for effective simulation of quantum systems, such as electrons in molecules. While ground states of many molecular systems can be efficiently computed with traditional methods of computational chemistry, electronic structure of large strongly correlated systems is classically difficult to model due to exponential time and space complexity. A quantum algorithm, Quantum Phase Estimation, is known to be capable of finding the ground state wavefunction and the ground state energy of electronic systems in polynomial time, given an initial state with sufficient overlap with the ground state. However, preparation of such initial states is exponentially expensive in the worst case, and existing methods typically require multi-controlled rotation gates (resulting in deep circuits) and/or rely on variational optimization of circuit parameters. In this contribution, we will present a method for construction of state preparation circuits, for use as initial states in phase estimation protocols, for an important class of electronic structure problems. Our approach does not require variational optimization, yields high overlaps with the ground state and results in orders-of-magnitude lower circuit depths than best-in-class general state preparation methods. We will present a theoretical explanation of the effectiveness of the Ansatz, as well as numerical benchmarks. We believe our state preparation method will broaden the toolset available to researchers in the field, and may become the tool of choice for a certain subclass of electronic states.

How Accurate Are DFT Forces? Unexpectedly Large Uncertainties in Molecular Datasets

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Training of general-purpose machine learning interatomic potentials (MLIPs) relies on large datasets with properties usually computed with density functional theory (DFT). A pre-requisite for accurate MLIPs is that the DFT data are well-converged to minimize numerical errors. A possible symptom of errors in DFT force components is nonzero net force. Here, we consider net forces in molecular datasets including SPICE, Transition1x, ANI-1x, ANI-1xbb, AIMNet2, QCML, and OMol25. Several of these datasets suffer from significant nonzero DFT net forces. We also quantify individual force component errors by comparison to recomputed forces using more reliable DFT settings at the same level of theory, and we find significant discrepancies in force components averaging from 1.7 meV/Å in the SPICE dataset to 33.2 meV/Å in the ANI-1x dataset. These findings underscore the importance of well converged DFT data as increasingly accurate MLIP architectures become available.

The $H_2O - NH_3$ binary system at planetary interior conditions

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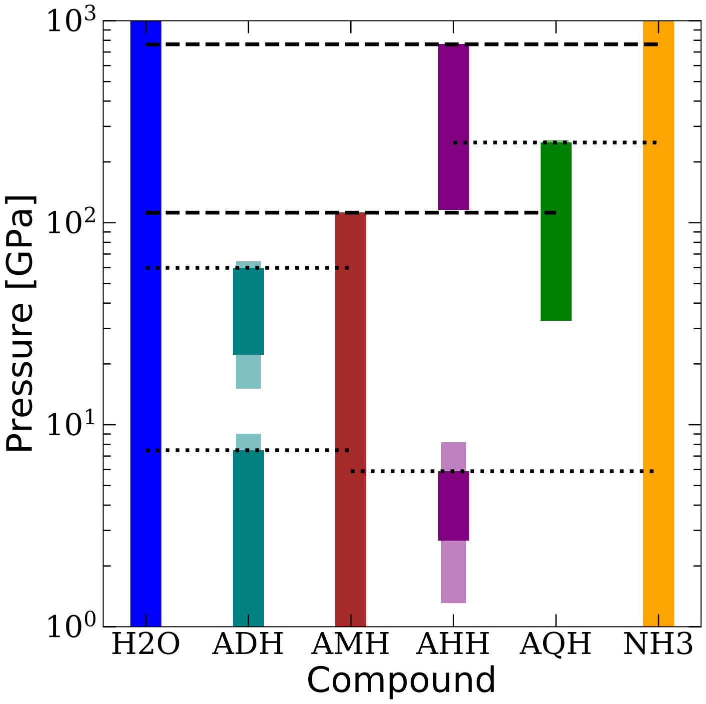
The interiors of Uranus and Neptune, the "Ice Giants", are not directly accessible to observations so their composition and structure, which determine their magnetic fields, are difficult to constrain. However, these planets are predicted to house vast reservoirs of planetary ices, such as water (H_2O) and ammonia (NH_3), in their interiors at extreme thermodynamic conditions: high pressure and high temperature.[1,2] To study these planetary ices at extreme conditions, we build on previous work[3,4] and apply density functional theory (DFT) combined with molecular dynamics to the $H_2O - NH_3$ binary system, including the ammonia hydrates $(H_2O)_x(NH_3)_y$. From these simulations we characterise the range of phases, including fluid and superionic states, which arise in the $H_2O - NH_3$ system and are crucial for our understanding of ice giant interiors.



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Ab-initio Probing of Hydrogen Sorption on Metal-Overlayered Pd(111) Surfaces

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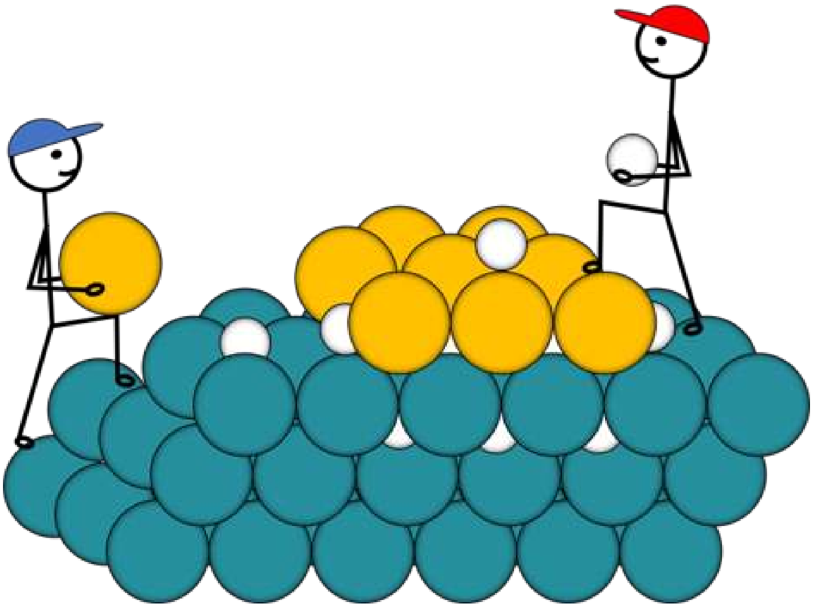
Understanding and tuning the surface reactivity of palladium (Pd) is key to enhancing hydrogen sorption and catalytic performance. While many studies focus on low hydrogen coverages, exploring surface behavior under higher coverages is essential for practical applications. Motivated by recent advances in overlayer and metallene-based nanomaterials, we use density functional theory to investigate hydrogen sorption on Pd(111) surfaces modified with a single metal overlayer—denoted $\mathrm{M_{ML}Pd(111)}$, where M includes Ag, Au, Co, Cu, Ir, Ni, Pt, and Rh. Using ab initio-based phase and surface Pourbaix diagrams, we determine the thermodynamically preferred hydrogen coverage on these systems under varying conditions of temperature, hydrogen pressure, pH, and electrochemical potential. Notably, Cu and Pt overlayers enhance surface stability compared to pristine Pd(111), while Ag and Au are also stable but bind hydrogen weakly. Among the studied systems, $\mathrm{Cu_{ML}Pd(111)}$ stands out with favorable hydrogen adsorption energies and low diffusion barriers from surface to subsurface sites, making it a promising candidate for selective hydrogenation and reduction reactions. Overall, this work highlights the critical role of hydrogen coverage in tuning surface and subsurface reactivity and provides design principles for next-generation Pd-based hydrogenation catalysts.



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Localized Orbital Scaling Correction to Linear-response Time-dependent Density Functional Approximations

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The localized orbital scaling correction (LOSC) method, which was developed for eliminating the delocalization error in density functional approximations (DFAs), is extended to the linear-response regime for calculating excitation energies with time-dependent density functional theory (TDDFT). Extensive numerical tests on various datasets show that LOSC-DFAs are able to maintain the good performance of parent DFAs for valence excitations while systematically improve the excitation energies for Rydberg and charge-transfer excitations by reducing the delocalization error. For charge-transfer excitations, LOSC can produce correct asymptotic behaviors with the donor-acceptor separation R as well as the excitation energy at the infinite separation limit. Moreover, through the example of *trans*-polyacetylene oligomers, we demonstrate that the performance of LOSC does not deteriorate with increasing system size, holding promise for the application in bulk systems.

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From Atomic Interactions to Electronic Tunability : An In Silico Analysis of MoS_2/Perylene Hybrid Interfaces

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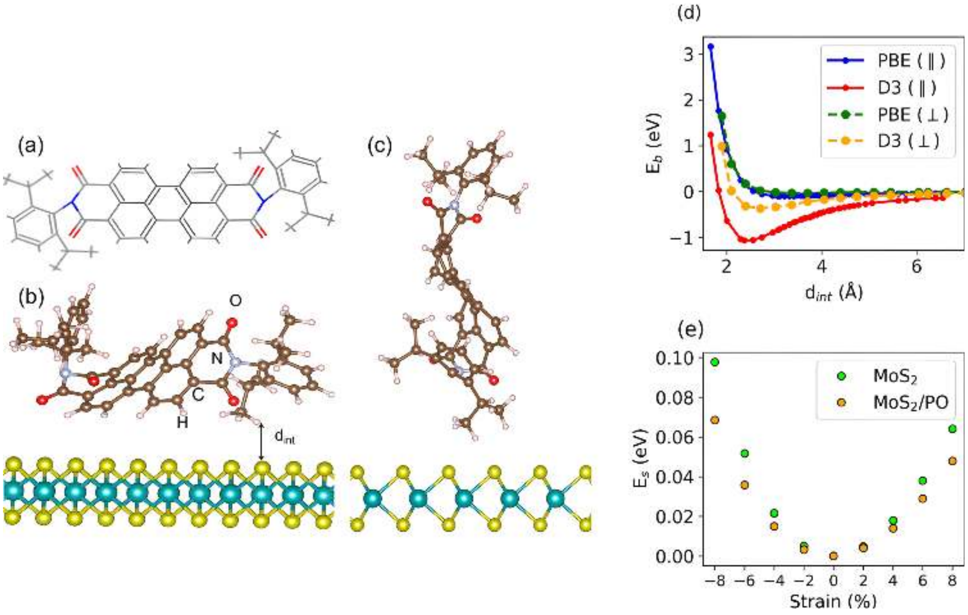
Combining organic molecules with two-dimensional (2D) materials offers a transformative approach to engineering van der Waals (vdW) heterostructures, bringing together the exceptional properties of 2D inorganic layers and the chemical versatility of organic compounds. Organic molecules, characterised by strong light absorption but limited charge mobility, complement inorganic 2D materials that provide excellent charge transport yet lower absorption. This synergy enables novel functionalities by harnessing the strengths of both material classes.[1, 2]

Molybdenum disulfide (MoS_2), a prototypical transition-metal dichalcogenide, is a promising 2D semiconductor with applications in electronics and photonics. Perylene-based dyes, notable for their tunable optical properties, offer opportunities to tailor hybrid interfaces with MoS_2. Using density functional theory (DFT), we systematically explore the structural, electronic, and optical properties of MoS_2/perylene hybrids, including derivatives such as perylene diimide (PD) and perylene orange (PO). Our study highlights how interfacial strain and molecular design enable precise control over hybrid properties, revealing synergistic effects that enhance performance. These insights advance the understanding of 2D–organic combinations and guide the design of materials with tailored functionalities for next-generation optoelectronic devices.[3]



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Revealing the role of surface disorder in H_2 desorption from metal surfaces via machine learning enhanced simulations

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Understanding interactions between hydrogen and metal surfaces as well as the desorption of hydrogen is essential for hydrogen storage and heterogenous catalysis. Conventional computational methods such as the nudged elastic band together with the harmonic approximation are limited in capturing the dynamical behaviour of surface atoms, especially at high coverages. With the assistance of machine learning interatomic potentials, we investigate the thermodynamic and kinetic aspects of associative desorption of H_2 from Cu(111) and Pt(111) at different coverages. Through the application of enhanced sampling techniques, including umbrella sampling and metadynamics, our results reveal a significant reduction in the desorption barrier on both surfaces. Of importance, a significant entropic-driven decrease in desorption barrier is observed on Pt(111) at high coverage. Further analysis suggests that the concerted motions of H atoms on Pt may attribute to configuration disorder, accelerating the desorption.

From accurate quantum mechanics to converged thermodynamics in solution with machine learning potentials

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Obtaining accurate predictions of thermodynamic properties, especially free energies that define the state of a system, is a key goal in atomistic simulations. This demands pushing the frontiers of two paradigmatic problems: (1) achieving high electronic structure accuracy and (2) reaching converged sampling of the nuclear motion. Coupling advances across electronic structure theory, machine learning potentials and enhanced sampling, we demonstrate a data-efficient approach to perform finite temperature simulations at the 'gold-standard' CCSD(T) level. This approach has been applied to constant pressure simulations of liquid water, combined with nuclear quantum effects to enable agreement with experiments for structural and transport properties. We further extend this framework to ions in solution, achieving excellent experimental agreement for the ion pair association free energy of CaCO3 in water - a challenging property for both classical force fields and density functional theory. This framework sets the stage for routine CCSD(T)-quality predictions for complex aqueous systems.

Dielectric Breakdown at Solid-Liquid Interfaces in Short Range MLIPs, and How to Fix It.

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Machine-Learnt Interatomic Potentials (MLIPs) are enabling studies of solid-liquid interfaces with *ab initio* accuracy at affordable computational cost. The most modern and accurate architectures—many-body equivariant GNNs—typically (but not exclusively) capture only short-range interactions. In this work, we show that the lack of treatment of long-range electrostatics can lead to an unphysical net alignment of water dipoles above a surface, generating a polarizing field that causes artificial dielectric breakdown of the water. We find that training on models that treat long-range electrostatics alleviates the issue. We propose a strategy of training on datasets that include thin water layers above surfaces and slabs of water, and show it to be effective for long-range models. This work lays the groundwork for creating reliable MLIPs for studying processes at solid-liquid interfaces, such as heterogeneous catalysis.

Understanding SCF landscapes and their applications in materials, machine learning and beyond

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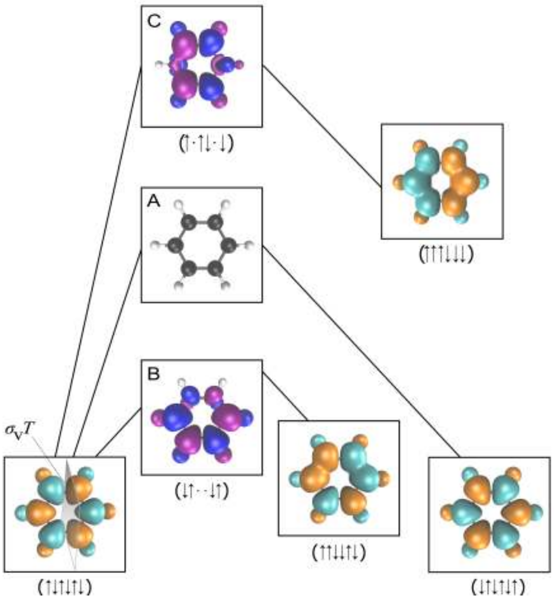
There has been substantial effort to understand the potential energy landscapes of self-consistent field (SCF) methods, primarily aimed at locating excited states relevant to phenomena such as charge transfer and core electron excitations. However, the topology of the SCF landscapes remains comparatively underexplored.[1,2] In our work, we use coefficients to iteratively explore the electronic structure landscape, drawing parallels to the molecular Potential Energy Surface (PES) framework. These landscapes contain stationary points: minima and saddle points, whose interconnections reveal non-trivial pathways, particularly those involving spin conversion from global minima to higher-order saddles. Using model systems such as benzene and cyclobutadiene, we analyse these pathways and investigate the role of symmetry constraints that are either preserved or broken as the system transitions between stationary points. Notably, we identified a simple combinatorial model that predicts the number of minima on these landscapes.[3] The implications of this work are significant: deficiencies in widely-used single-determinant methods (e.g., Hartree-Fock or DFT) lead to multiple solutions producing more complicated landscapes than expected. For example, in related projects on battery degradation pathways, we observed unexplained ground-state instabilities, where the SCF solution spontaneously collapsed into different electronic configurations. Similarly, in machine learning applications for chemistry, datasets are often trained on energies and geometries derived from single-determinant approximations—introducing fundamental inaccuracies into the resulting potentials. Through my poster, I aim to bridge the gap between fundamental understanding of SCF theory and the more applied, often opaque, issues of “convergence” in electronic structure calculations.



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Computational Study of Surface-Catalyzed Ullmann Coupling Reaction of Bromobenzene on Coinage Metals

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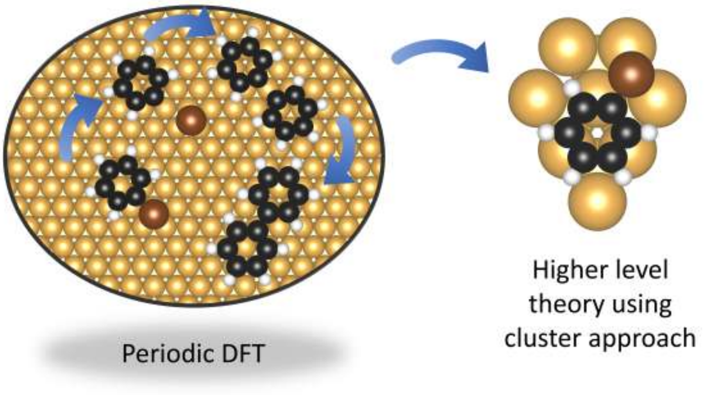
Experimental studies of the on-surface Ullmann coupling reaction on the coinage metal surfaces have gained increasing interest in recent years, as it provides a promising path to *e.g.* 2D graphene-like structures, while allowing for a high flexibility with concern to the product.[1] To rationalise experimental findings, density functional theory (DFT) methods are mainly used. However, a variety of combinations of different exchange-correlation (XC) functionals and dispersion corrections often make the results hard to compare.[2–5]

The different steps of the Ullmann coupling reaction were investigated using different XC-functionals and dispersion corrections on the Cu(111), Ag(111), and Au(111) metal surfaces with periodic boundary conditions.

The computed adsorption energies and the adsorbate–surface distances were analysed in detail. To predict the structure and energy of the adsorbed chemical systems, various DFT approximations have started to be benchmarked against the correlated *ab initio* methods using a cluster approach.[2]

Further, in cooperation with experimentalists, a comparison of the D3(BJ) and D4 dispersion corrections was performed for large organic compounds on a Au(111) surface by means of adsorption energy and structure.[6]



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Adsorption of CO_2 on Defective Graphene in the Presence of Water

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Defects in materials are as ubiquitous as water in real-world systems, yet a molecular-level understanding of their effects remains limited [1]. We investigate how the presence of water influences CO_2 adsorption on defective graphene – a system with important environmental and industrial relevance. Using the foundation machine learning model MACE-MP-0 and density functional theory (DFT), we study the thermodynamics of CO_2 and H_2O interactions at single vacancy (SV), double vacancy (DV), and Stone–Wales (SW) defects in graphene. In contrast to pristine graphene, both H_2O and CO_2 can partially or fully dissociate upon adsorption at SV and DV sites. Regardless of defect type, a second CO_2 or H_2O molecule adsorbs much more weakly and remains intact, as the initial dissociative adsorption quenches the defect reactivity. These findings provide molecular-level insights into water–CO_2 co-adsorption on defective graphene and can inform the design of carbon-based adsorbents for wet carbon capture applications.

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Leveraging generative AI to reconstruct unknown positions of hydrogen sites

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Generative AI methods are rapidly evolving to speed up and improve materials discovery. Diffusion based models can not only be adopted to generate new materials with desired properties but also to reconstruct crystal structures for which structural information is only partially available.[1] In this work, we use Microsoft's MatterGen,[2] a diffusion based model originally designed to generate new stable crystal structures, and extend and apply it to reconstruct missing hydrogen sites in crystal structures reported in experimental databases. This is particularly useful as the experimental measurement of hydrogen sites with standard XRD is typically challenging due to weak scattering of hydrogen. We show how to leverage approaches known from image inpainting in the field of computer vision, combined with universal machine learning interatomic potentials, to improve the success rate of correctly identifying the missing sites while significantly lowering the computational cost with respect to a direct DFT approach. Moreover, the adopted approaches exhibit superior performance compared to existing inpainting approaches in the context of conditional crystal structure generation.

Thanks to the generality of the method, future applications range from reconstructing experimental structures as in the case of missing hydrogen positions to analyzing intercalation by predicting the most probable ion positions in DFT optimized crystal structures of cathode materials.

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C2x: a code-agnostic DFT pre- and post-processor

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The c2x code[1] has been developed to assist in a wide variety of pre- and post-processing for DFT codes, including interfacing with common visualisation software. It supports Abinit, Castep, Onetep, Quantum Espresso, Siesta and Vasp, and can assist in converting input and output files between those codes. It supports Gnuplot, Jmol, Vesta, VMD, and XCrysDen for visualisation.

It is a command-line program, written in C, and continues to be developed and extended. It uses the same internal data representation regardless of the input code, so all features are available for all codes (with the exception of wavefunction processing for non planewave codes). Its only build dependency is SPGlib,[2] without which it will build but will be unable to perform symmetry analysis.

Its list of features, when used in conjunction with suitable visualisation software, includes:

- supercell and primitive cell generation
- surface creation and nanotube rolling
- charge, spin and spinor visualisation
- the same for individual bands
- charge and spin density differencing between two systems
- Fermi surface visualisation
- DoS and LDoS (slabs and spheres) plots
- band structures (crossings not resolved)
- band inversion symmetry reporting
- interpolation (trilinear and Fourier)
- averaging and integration (planar, spherical and cylindrical)
- electrostatic potential calculation
- dipole moment calculation, and *post hoc* corrections
- calculation of Brillouin zones

Perhaps almost as important as the software is its website, `www.c2x.org.uk`, which contains example calculations for many electronic structure codes.

C2x has been developed for over a decade, and has gained much functionality since its paper was published seven years ago. It has been cited by over 70 publications, and ideas for new features are always welcome.

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Machine Learning Potential Study for the Unknown Reaction of the Calcium Polyhydride Synthesis

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Room-temperature superconductivity is one of the most significant goals in physics. Polyhydrides, with their remarkable critical temperatures (260 K), have emerged as promising candidates. In this field, theoretical structure search methods based on density functional theory (DFT) calculations have proposed novel structures as an initial step. Subsequently, proposed polyhydride are realized by high-pressure synthesis using a diamond anvil cell (DAC). In this synthesis in DAC, not only the high pressure but also high-temperature heating by laser irradiation are usually required to promote reactions. This implies that polyhydride synthesis involves kinetically slow processes or reactions with high activation energies. Therefore, physical insights into the reactions are important to obtain efficient synthesis routes.

Under such circumstances, we have revealed the polyhydride synthesis mechanism using machine learning potential molecular dynamics simulations.[1] In this MLP-MD simulations, the CaH_2 surface melts at high pressure and temperatures like 40 GPa and 1500 K upon contact with high pressure H_2. During the reaction, the melting (disordered) structure poses hydrogen network similar to CaH_4. Therefore, the hydrogenation reaction occurs via liquid CaH_4 phases. Based on the MLP-MD simulations, we propose the thermodynamic model for the hydrogenation under high pressure, the surface melting via liquid product hydrides,

$$\mathrm{CaH_2(s) + H_2 \longleftrightarrow CaH_4(l) \longleftrightarrow CaH_4(s).}$$

as follows,

$$\Delta \mathrm{G} = \Delta \mathrm{G}_{\mathrm{fus}} + \Delta \mathrm{G}_{\mathrm{hyd}} + \Delta \gamma_{\mathrm{interface}}.$$

Here, ΔG_{fus} and ΔG_{hyd} represent the free energy of CaH_4 fusion ($CaH_4(s) \leftrightarrow CaH_4(l)$) and CaH_2 hydrogenation ($CaH_2(s) + H_2 \leftrightarrow CaH_4(s)$), respectively. $\Delta\gamma_{interface}$ is the interfacial energy difference before and after the reaction. Since ΔG_{hyd} is negative under high pressure, the hydrogenation enthalpy lowers the activation energy to obtain liquid CaH_4 phase as an intermediate state. This thermodynamics shows why we need to apply "overpressure", the higher pressure compared to the equilibrium one. Details of the reaction and thermodynamic analysis will be presented at the conference.

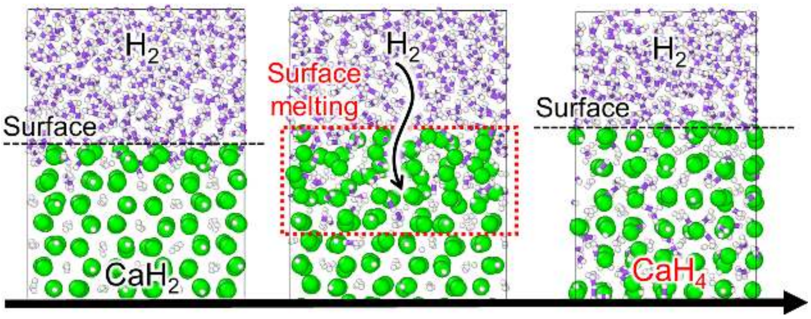

Figure Snapshots of $CaH_2(100)/H_2$ interface during MLP-MD simulations at 40 GPa and 1500 K.[1]

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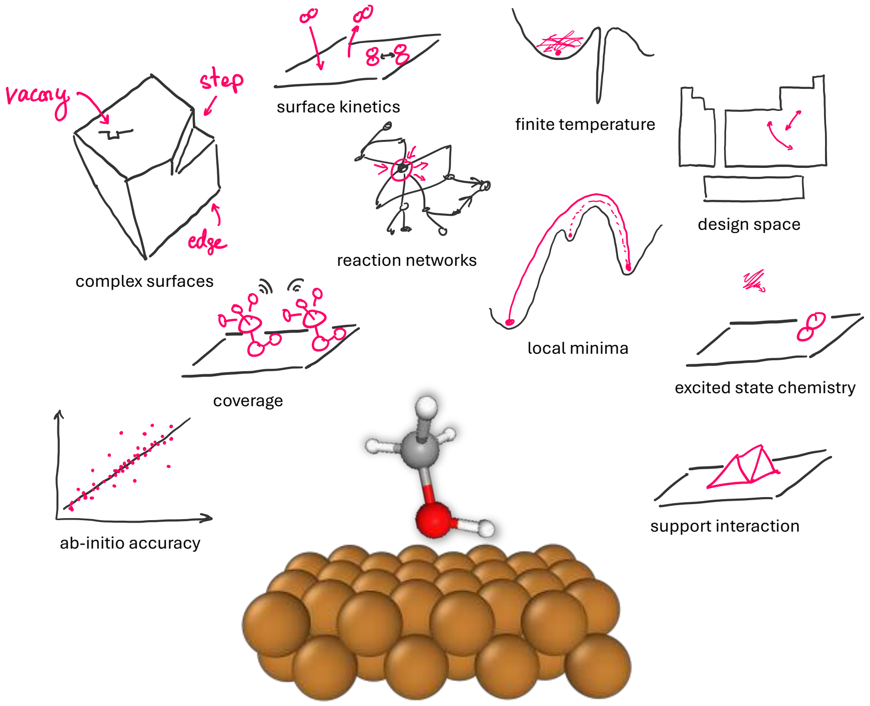
The Future of Machine Learning Interatomic Potentials for Heterogenous Catalysis

Lars L Schaaf, Christoph Schran, and Gábor Csányi

Machine learning interatomic potentials (MLIPs) are emerging as powerful tools for simulating catalytic systems with near ab-initio accuracy at a fraction of the computational cost. In this talk, we outline the unique challenges that catalysis presents to ab-initio modelling, ranging from complex surface reconstructions and temperature-dependant selectivity to reaction network complexity and coverage-dependent effects. We highlight the utility of MLIPs for these challenges and recent progress in addressing these issues. Furthermore, we outline current limitations and future developments in the domain of large foundation models, electrostatic architectures for modelling charge transfer and the effects of external fields. Finally, we argue that MLIPs do more than accelerate routine workflows; they enable predictive simulation of realistic, dynamic, and structurally complex catalytic surfaces, reducing the gap between computational modelling and experimental observations.



CuNi Bimetallic Catalysts for Hydrogen Production via Methanol Steam Reforming: A Theoretical and Experimental Investigation

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The methanol steam reforming (MSR) reaction is a key route for hydrogen production, featuring a favorable H_2/CO ratio of 3:1 and mild endothermicity. Bimetallic catalysts based on 3d transition metals offer enhanced performance due to synergistic geometric and electronic effects. Among various Cu-based alloys, CuNi has emerged as a particularly promising system. In this study, density functional theory (DFT) calculations were performed to investigate the MSR mechanism on CuNi(111) surfaces, revealing low activation barriers for key reaction steps. The presence of Ni significantly promotes the initial C–H and O–H bond activation, while maintaining the overall stability of the alloy surface. Experimental validation further confirms the superior catalytic activity of CuNi, consistent with computational predictions. This combined theoretical and experimental approach provides valuable insights into rational catalyst design for efficient hydrogen production.

It's Not a S(KZ)CAM! Accurate Surface Modeling at Low Cost

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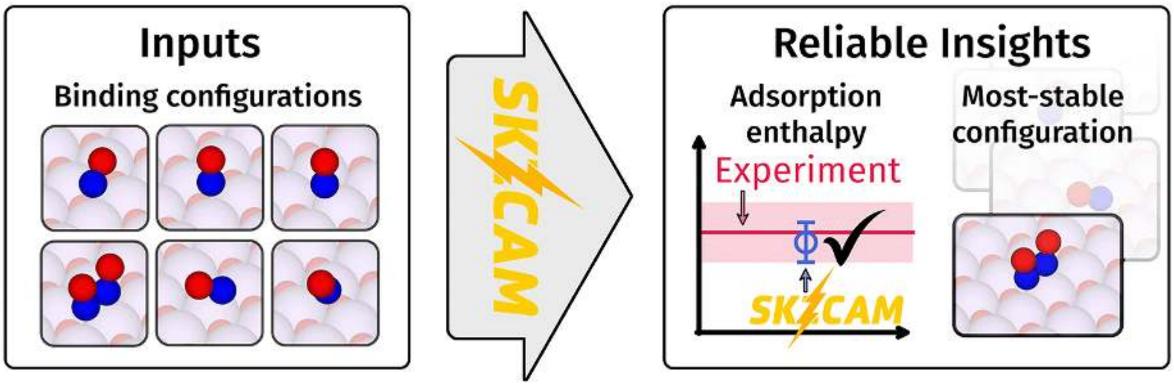
Reactions supported on solid surfaces form the cornerstone of both the chemical and energy industries. With computational modeling, it is possible to attain an atomistic-level understanding of these processes, paving the way towards designing improved materials. However, consistently reproducing experiments remains challenging: density functional theory (DFT) is efficient but can give inconsistent predictions, and while methods from quantum chemistry, notably the "gold-standard" coupled cluster theory [CCSD(T)], can readily reproduce experiments, their application to surfaces has been limited by their prohibitively high cost. I will present the SKZCAM (pronounced "scam") protocol,[2] which overcomes this cost-accuracy tradeoff to bring CCSD(T) to the surfaces of ionic materials at a cost approaching DFT. It successfully resolves longstanding discrepancies, notably the adsorption energy of CO on MgO(001)[2] – often dubbed the "hydrogen molecule of surface science" that has historically challenged both experiments and theory. Furthermore, its computational efficiency has allowed for a diverse set of molecules on technologically relevant metal-oxide surfaces to be studied,[3,4] revealing new insights into adsorbed gold nanoclusters and methanol clusters, important to applications ranging from waste gas removal to fuel production. Finally, the SKZCAM protocol is now available as an open-source package,[5] serving as a new tool to facilitate atomistic understanding of complex surface phenomena for other research groups.



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Structural Instabilities and Acidity at Conical Regions in Trivalent Metal Ion Nano-clusters

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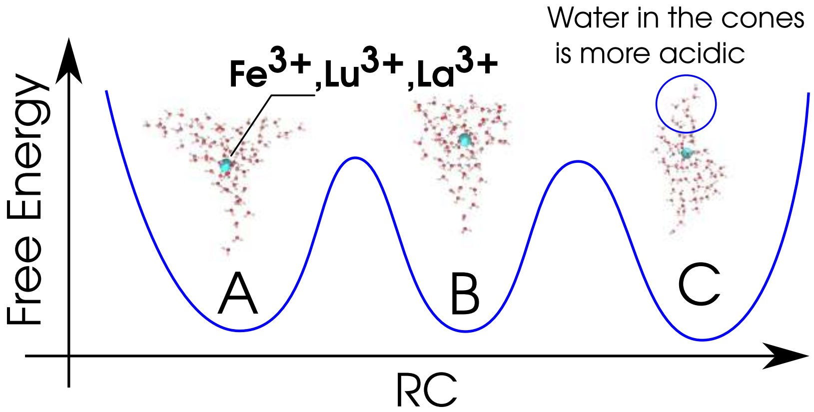
Sub-nanometer aqueous clusters containing a single trivalent metal cation can exhibit charge-induced structural instabilities. Here, we present computational evidence that clusters containing a single Fe^{3+}, Lu^{3+}, or La^{3+} ion undergo continuous geometric transformations as a consequence of this instability. These clusters dynamically evolve across their potential energy landscape, adopting triangular, elongated two-point, single-point, and more spherical configurations often with distinct conical surface protrusions. The manifestation of this instability differs from that observed in mesoscopic and microscopic droplets containing macroions, where stable "star-like" structures form, characterized by a specific number of conical protrusions that varies with the droplet size. In the present study, we find that the orientation of the H_2O molecules surrounding the metal ion is influenced not only by the electric field of the trivalent ion but also by the local conical protrusions. To further investigate the local acidity in the conical protrusions, we employ a proxy model system consisting of an aqueous nano-cluster containing three H_3O^+ ions, simulated using ab initio molecular dynamics. Within the conical regions of the cluster, protons exhibit diffusion across several water molecules, in contrast to the more localized proton delocalization observed in the compact body of the cluster. These findings suggest that local geometry can significantly modulate acidity in highly charged nano-clusters, with potential implications for understanding charge-transfer and ionization mechanisms in techniques such as electrospray ionization mass spectrometry. Additionally, the structural motifs and solvent organization reported here provide a molecular-level framework that can complement interpretations from infrared spectroscopic data.



Ring Polymer Neural Fields for spin-phonon systems

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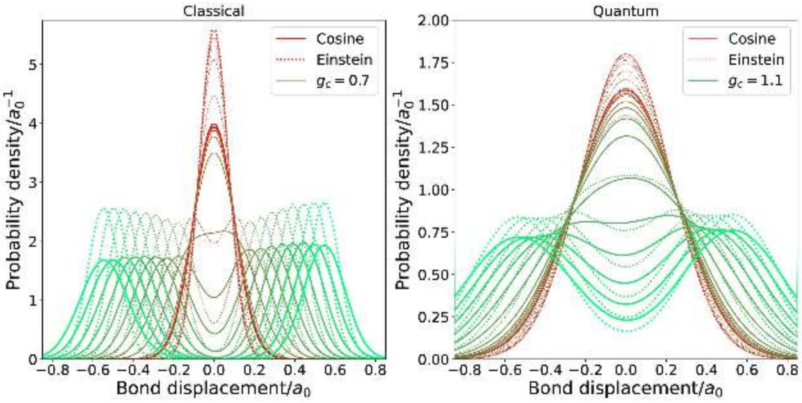
The coupling between the real space configurations of atoms and the strength of the spin-spin interactions gives rise to interesting spin-phonon effects such as spin-Peierls dimerisation, but it also makes the treatment of these systems incredibly tricky since one has to solve both the spin problem as well as the vibrational problem together. Previously, we built on existing Monte Carlo schemes used to tackle this by employing Molecular Dynamics, and with a suitable approximation to the Exact Diagonalisation scheme, we were able to sample the spatial configurations of the systems in order to calculate the real space observables, such as bond length distribution functions at different values of spin-phonon coupling (Figure below) and spin properties, such as spin-spin correlation functions with distance. While this demonstrated the principle behind the method, there was still space for improvement, mainly in the accuracy in the cases when long-range interactions were present, computational time and scaling up to systems beyond one dimension. All of these problems were addressed in the newest addition to the method that we wish to present, using neural networks for calculating forces coming from the spin-spin interactions, here, via Heisenberg model. In addition, the path integral formalism was used to account for nuclear quantum effects and Stochastic Series Expansion was used for spin calculations, including the training forces, and the combined method is called Ring Polymer Neural Field.



Analytical Structure of Excited-state Wave Functions in Many-body Harmonic System

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Peking University, Beijing 100871, China

The structure of the excited-state wave functions in many-body systems is very important for developing excited-state quantum chemistry methods. This study investigates the analytical structure of excited-state wave functions of the many-body system with both harmonic external potential and interaction (harmonic system), from one to higher dimensions. Using group-theoretic methods, we reveal the distribution of fermionic states in one-dimensional systems and establish a one-to-one correspondence between fermionic and bosonic states. We find that the excited-state wave functions of the harmonic system generally exhibit structures of multi-determinants multiplied by Gaussian correlation factors. This product structure can be efficiently approximated by a linear combination of few Slater determinants under in the weakly correlation region, whereas the linear combination approach requires significantly more terms to achieve comparable accuracy in the strong correlation region. Furthermore, we observe that the number of determinants in the product structure rapidly increases with both the number of particles and the excitation level. Based on these findings, this study proposes that the structure resulting from the exchange antisymmetric operator acting directly on the function of the particle coordinates represents the optimal structure for the harmonic system's wave function.

Kinetics: The Missing Link Between Theory and IRMPD in Cluster Chemistry

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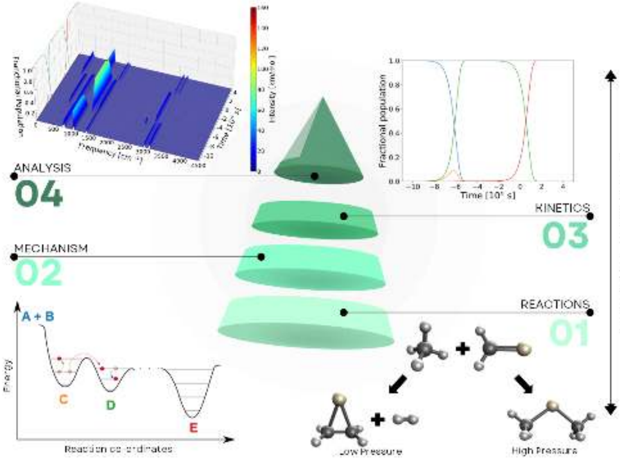
One of the most intensively studied areas of nanocatalysis is nanoscale clusters, as their size-dependent structural and electronic properties can lead to unique catalytic activity, distinct from bulk materials. To study adsorption and bond activation on clusters, Infrared Multiphoton Dissociation (IRMPD) spectroscopy offers a powerful tool due to its sensitivity to structural motifs and ability to distinguish isomers in the gas phase — a regime often inaccessible to traditional condensed-phase techniques. In this method, mass-selected cluster–bath gas adducts are irradiated with a tuneable IR laser, leading to vibrational excitation and fragmentation, from which infrared spectra can be reconstructed. Combining this with quantum chemical computations, providing information about precise structures, electronic configurations, and binding energies allows the investigation of complex, branched, multi-step reaction mechanisms. However, even combined experimental and computational approaches cannot unambiguously determine the progress of the reaction, making kinetic simulations — alongside experimental comparison — essential for understanding the feasibility of condition-dependent steps and guiding the design of high-performance catalysts. We show three different reactions where our master-equation based microkinetic simulation[1] has different purposes, from the unnecessary ($Cu_3Sc + CO_2$),[2] through the interpretative and supportive ($Pt^+ + CH_4$),[1,3] to the predictive ($Cu_4^+ + H_2$)[1,4] — highlighting how the role of kinetics can range from negligible to essential in uncovering mechanistic details and enabling reliable catalyst design.



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First-principles study of the crossover in the electronic stopping power

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We are using first principles real-time time-dependent density-functional theory to reproduce the cross over in electronic stopping power when increasing projectile velocity for oxygen and magnesium projectiles in bulk aluminium. These two projectiles are the most indicative cases of Z1 oscillations. We aim to correlate this electronic stopping power behaviour as function of velocity with the charge state of projectile. The charge state is difficult to define, and therefore we are following deformation density maps as well as charging effects as given by both the Voronoi and Hirshfeld population definitions. Both population methods (of very different character) give remarkably consistent behaviours. The results will be presented and discussed.

Simulations of the RuO_2-LiCl(*aq*) interface using finite field molecular dynamics

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The developments of various chemical systems and processes are needed to secure our future energy supply, storage, and usage. One piece of solutions to this challenge is electrochemical supercapacitors, providing a power output surpassing that of batteries and an energy storage capacity higher than traditional capacitors.[1] One of the quintessential supercapacitor systems is the aqueous interface of RuO_2, whose impressively high specific capacitance arises due to interplay between various structural and electrochemical factors.[1,2] To begin with, a polarised conducting oxides such as RuO_2 in contact with an electrolyte stabilizes itself by attracting electrolyte ions from the bulk electrolyte. However, the capacitive contribution of this electrochemical double layer does not tell the whole story. For one, the electron spillover at the conductor boundary give rise to additional capacitive effects. Furthermore, for several materials including RuO_2, there is additional charge storage possibilities from proton-coupled Faradic reactions at the interface. In certain cases these can provide a capacitor-like response, which is then known as pseudocapacitance.[1,2] While knowledge about the interaction and relative effect of these contributions would be useful for developing the next generation supercapacitors, this information is not easily extracted from experiments. On the other hand, the atomic and electronic resolution provided by computational chemistry can give fascinating insight into the fundamental processes of supercapacitors. Recently, so-called finite field molecular dynamics has proven successful in simulating the capacitive response of TiO_2[3] - and gold-aqueous interfaces.[4] Finite field simulations allow simulated systems to react to macroscopic-like fields, while still allowing for continuous forces also under periodic boundary conditions. We now apply finite field molecular dynamics to the arguably even more intricate case of the conducting oxides where the boundary between proton charge and electronic charge gets blurred. Control of the applied fields, the surface protonation, and the degree of hydroxylation allows us to disentangle the origin of the capacitive response at the RuO_2-LiCl(*aq*) interface.

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Altermagnetic fullerene monolayers

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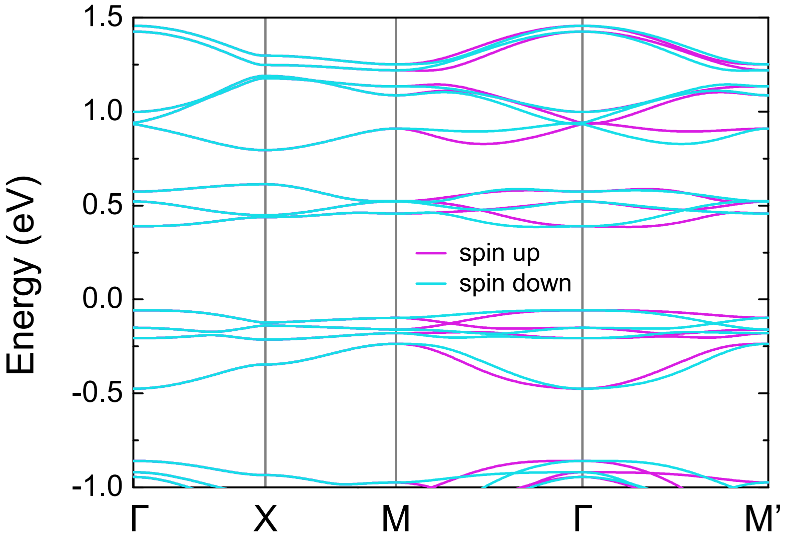
Fullerene monolayers have been predicted to be highly tunable through fullerene size[1] and varied dimensionality[2] since their successful synthesis.[3] However, most of these pure-carbon systems are non-magnetic semiconductors. Recently, we introduce magnetism to fullerene monolayers through molecular symmetry and valence-bond resonance. Here, we show one example of altermagnetic fullerene monolayers exhibiting spin-splitting and zero net magnetisation. Curiously, the effective local moments lie on the famous Shastry-Sutherland lattice with exchange constants close to the altermagnet-spin liquid phase transition, raising the intriguing possibility of realising a quantum spin liquid in a pure-carbon system.



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Machine learning interatomic potentials on Cu_water_ions interfaces

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Machine learning interatomic potentials (MLIPs) have shown a promise to simulate large-scale systems for a long period at the DFT-accuracy level. In this work, we refined MACE-based model[1] to construct MLIPs of Cu_water_ion systems based on dataset collected from ab initio molecular dynamics (MD). We evaluated performances of MLIPs on the simulation of water molecules on three facets of Cu (Cu(111), Cu(100) and Cu(110) and water coordination numbers of cations, which are in agreement with previous experiments and DFT calcuations.[2,3] Furthermore, we applied the refined potentials to compare the effect of different ions on interfaces of water and Cu(100) to interpret the gap of waters near the surface in experiments.

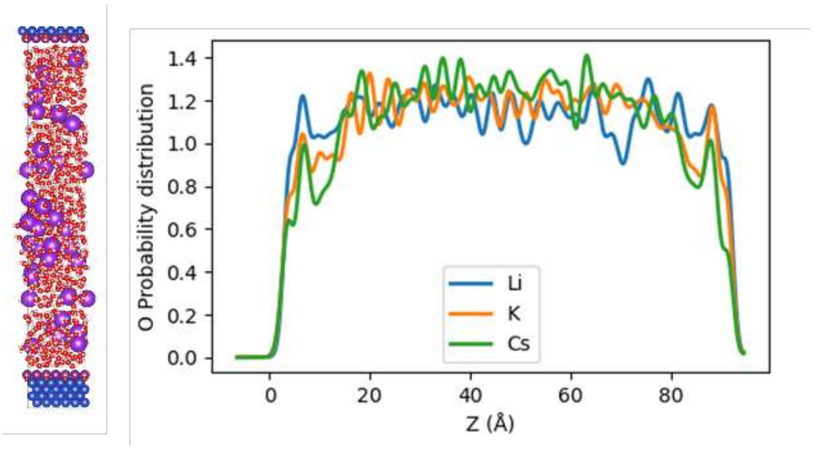

Figure.1 O distribution along Z axis in systems with Li^+, K^+ and Cs^+ as cations.

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Long-range polarization models for reactive molecular systems.

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The combination of the molecule-based many-body expansion (MBE) with machine learning interatomic potentials (MLIP) has proven highly potent in generating surrogate potential energy surfaces for fast computational sampling of condensed phases.[1] Key to accurate MBE-MLIP potentials is an accurate description of long-range electrostatics which requires the definition of element-specific fixed atomic parameters such as atomic charges and polarizabilities.[2,3] In the case of reactive systems, such a treatment falls short due to significant charge transfer generating atomic environment-dependent atomic charges and polarizabilities. In this work, we systematically investigate this problem at the example of protonated water clusters and discuss possible solution strategies from Thole-type polarization models up to MBE-corrected models.

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Ab Initio Investigations on the Quantum Dynamics of Excited Carriers in Condensed Matter Systems

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The ultrafast dynamics of charge carriers in condensed matter systems plays a crucial role in charge transport, optoelectronic performance, and solar energy conversion. Our research aims to develop an *ab initio* simulation framework for a state-of-the-art understanding of multi-dimensional carrier dynamics in solid-state materials. To this end, we have developed the Hefei-NAMD code, which enables the investigation of (i) single-particle dynamics, (ii) spin dynamics, (iii) exciton dynamics via GW combined with real-time Bethe–Salpeter equation (GW+rtBSE), and (iv) momentum-space dynamics.[1–5] In this talk, I will first give a brief overview of Hefei-NAMD and introduce the theoretical framework of momentum-resolved nonadiabatic molecular dynamics (NAMD-k). Using NAMD-k, we have studied the hot electron relaxation processes in a variety of materials and revealed a universal two-stage mechanism: (i) an initial momentum relaxation process, during which hot carriers rapidly scatter across the entire Brillouin zone within tens of femtoseconds, forming a hot carrier ensemble; (ii) a subsequent energy relaxation process, typically occurring over several picoseconds or longer. This provides a clear microscopic picture of nonequilibrium hot carrier dynamics in materials.

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