Systematic DFT Study of Phthalocyanine Derivative Molecules on Rocksalt Substrate

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Phthalocyanine (Pc) derivative molecules, a representative class of planar π-conjugated systems, have attracted significant attention as a promising platform material for molecular electronics and surface-based quantum devices. However, one of the critical challenges hindering their integration in nanoscale architectures is the lack of awareness for their adsorption configuration and their changed MO level. Scanning tunneling microscopy (STM) investigations have identified multiple adsorption geometries and dynamic behavior[1], yet the energetics and diffusion mechanisms of Pc molecules remain poorly understood due to their complex potential energy surfaces and rotational degrees of freedom.

To address this issue, we have performed first-principles calculations to systematically investigate the adsorption energy and rotational barriers of Pc molecules on rock-salt substrates. In this presentation, we will introduce the analysis of energy profiles obtained from FHI-aims and VASP. Additionally, we will present a detailed molecule-substrate interaction, highlighting the interplay between adsorption site and MO gap. Our results offer deeper insights into the finding local minima from the unknown multi-dimensional system.

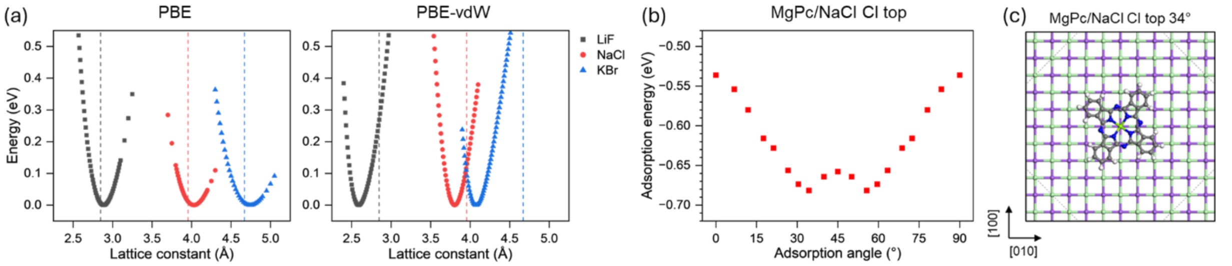

Figure 1: (a) Calculated lattice parameter of LiF, NaCl, and KBr substrate with PBE functional. (b) Adsorption energies of MgPc according to its alignment angle with NaCl, and (c) optimized structure of MgPc on NaCl with 34° alignment degree.

References

[1] Miwa, K., Imada, H., Kawahara, S., & Kim, Y. (2016). Effects of molecule-insulator interaction on geometric property of a single phthalocyanine molecule adsorbed on an ultrathin NaCl film. *Physical Review B*, *93*(16), 165419.

Topological confinement states in ABA trilayer graphene with antiparallel electric field

In AB-stacked bilayer graphene, the introduction of an out-of-plane electric field can break the inversion symmetry to open up a nontrivial bulk gap hosting the quantum valley Hall state. However, in ABA-stacked trilayer graphene, the out-of-plane electric field cannot open up a nontrivial valley band gap. By applying a pair of antiparallel electric fields, we theoretically propose three kinds of schemes to open up bulk gaps that harbor the quantum valley Hall effect in the ABA-stacked trilayer graphene. By further considering the small-angle twisted trilayer graphene, we can obtain the topological confinement states along the naturally formed domain walls between ABA- and BAB-stacked regions. It is noteworthy that the synergic effect between the antiparallel electric fields and the magnetic field can open up a nontrivial band gap possessing the quantum valley Hall effect and quantum Hall effect simultaneously. Our work not only theoretically proposes how to realize the quantum valley Hall effect in ABA-trilayer graphene, but also provides an ideal platform to explore the hybrid topological phases.

Poster Number 2, Presenter: Jiaqi An, University of Science and Technology of China

Hydrogen adsorption on goldene: A study based on Density Functional Theory

This theoretical study explores the adsorption behavior of hydrogen on goldene using computational methods based on density functional theory (DFT). The Fritz Haber Institute ab initio materials simulations package (FHI-aims) was employed, using the optB86b exchange-correlation functional to accurately account for van der Waals interactions. A K-point mesh was generated following the relation $a = 40 \times K$, and a vacuum spacing of 20 Å was applied between layers to avoid interlayer interactions.

Both the primitive cell and three supercells (2×2, 3×3, and 4×4) were considered to investigate different surface coverage ratios, analyzing hydrogen adsorption at various potential sites. The structural behavior of goldene in the presence of hydrogen atoms and molecules was examined. Additionally, the study includes the effect of surface doping with adatoms of K, Li, Mg, and Sn. The results are intended to be compared with the (111) surface of bulk gold, providing insights into the potential of goldene for hydrogen-related applications.

Poster Number 3, Presenter: Ivan Arellano-Ramirez, Universidad Tecnologica de Pereira

Accurate Adsorption Energies of C-Species on Copper Catalysts

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Abstract

Accurate prediction of adsorption energies for C-species on copper catalysts is critical for understanding catalytic behavior and catalyst design. However, conventional density functional theory often falls short, as exemplified by the "CO adsorption puzzle." Here, we demonstrate that the hybrid PBE-D3/M06 method overcomes these limitations, achieving near-chemical accuracy for C-species on Cu(100), Cu(110), and Cu(111) surfaces. This approach resolves the CO puzzle by correctly predicting adsorption sites and energies in excellent agreement with experimental data. The method yields remarkably low mean absolute errors (MAEs) of 0.06 eV for reaction intermediates (vs. RPA calculations) and 0.04 eV for molecules (vs. experimental values). To accelerate these high-fidelity calculations, we further developed a machine learning model that rapidly predicts accurate adsorption energies (MAE: 0.08 eV) from standard PBE values. When applied to electrochemical CO_2-to-CO conversion, our approach predicts onset and equilibrium potentials to within 0.04 V of experimental measurements across all low-index copper facets. This combined computational strategy provides an efficient and reliable framework for the rational design of copper-based catalysts.

Electron-phonon coupling in chiral 2D metal-halide perovskites

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Chiral two-dimensional metal-halide perovskites (2D-MHPs) exhibit intriguing spin-dependent transport phenomena linked to chirality-induced spin-selectivity (CISS). Their microscopic origins, especially the role of electron–phonon interactions, remain poorly understood.[1] Accurately capturing phonon coupling is essential to quantify spin dynamics in these materials.

Conventional first-principles techniques—finite-displacement methods (FDM) and density functional perturbation theory (DFPT)—offer high accuracy but incur prohibitive cost for large, complex unit cells, making routine studies impractical.

To address this, we validate the deformation potential approach first proposed by Bardeen and Shockley.[2] This theory approximates long-wavelength ($\mathbf{q} \to 0$) acoustic phonons by relating lattice deformations to perturbations in the electronic structure. While inherently less precise than FDM or DFPT, deformation potential theory has successfully predicted transport coefficients and carrier mobilities in achiral 3D hybrid perovskites.[3]

We further propose integrating machine learning models—such as DeepH and neural-network DFPT frameworks that predict DFT Hamiltonians and perturbative potentials via automatic differentiation.[4,5] These methods enable high-throughput, near-ab initio-level predictions of electron–phonon coupling with dramatically reduced computational expense. They can also serve to cross-validate the deformation potential approach.

Combining deformation potential theory with modern deep-learning DFPT techniques yields a scalable, efficient workflow for exploring spin dynamics in chiral 2D-MHPs, supporting both theoretical discovery and experimental validation.

Acknowledgments

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References

[1] B. P. Bloom et al., "Chiral Induced Spin Selectivity", Chemical Reviews **124**, 1950–1991 (2024).

[2] J. Bardeen et al., "Deformation Potentials and Mobilities in Non-Polar Crystals", Phys. Rev. **80**, 72–80 (1950).

[3] J. Xi et al., "Temperature-dependent structural fluctuation and its effect on the electronic structure and charge transport in hybrid perovskite CH3NH3PbI3", Journal of Computational Chemistry **42**, 2213–2220 (2021).

[4] H. Li et al., "Deep-learning density functional theory Hamiltonian for efficient ab initio electronic-structure calculation", Nature Computational Science **2**, 367–377 (2022).

[5] H. Li et al., "Deep-Learning Density Functional Perturbation Theory", Phys. Rev. Lett. **132**, 096401 (2024).

Vibrational Spectra of Solids, Liquids and Molecules from Partially-Adiabatic Elevated-Temperature Centroid Molecular Dynamics

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Abstract

Nuclear quantum effects (NQEs) significantly affect vibrational line shapes and temperature-dependent frequency shifts. Path integral molecular dynamics (PIMD) methods capture these effects by representing each nucleus as a ring polymer of P replicas connected by harmonic springs. Within this framework, centroid molecular dynamics (CMD) computes vibrational spectra by evolving the ring-polymer centroids at temperature T_{sys} under mean-field forces from the internal modes [1]. However, CMD suffers from spectral artifacts—especially in high-frequency regions—most pronounced at low temperatures [2], where NQEs are strongest and experimental data most abundant.

The elevated-temperature T_e-CMD ansatz [3] mitigates these issues by computing mean-field forces at an elevated temperature $T_e > T_{\text{sys}}$. Two implementations exist: the original, fully adiabatic path- integral coarse graining T_e-PIGS, which relies on machine-learned (ML) forces to describe the centroid potential of mean force (PMF), and a partially adiabatic alternative, PA-T_e-CMD [4], developed in this work. PA-T_e-CMD evaluates the PMF on-the-fly using a two-temperature Langevin thermostat implemented in i-PI [5], removing the need for prior ML training.

We applied both implementations to three representative systems. In gas-phase water, PA-T_e-CMD closely reproduces the results of T_e-PIGS and the exact vibrational density of states and infrared spectra. In methylammonium lead iodide (MAPI), accurate spectra for multiple phases were obtained using a single elevated temperature corresponding to a third phase, demonstrating the robustness of the ansatz across structural changes in solids. In contrast, simulations of a fluorinated carbonic acid complex (CAF) reveal a limitation of PA-T_e-CMD: strong anharmonicities introduce spurious coupling between physical modes and internal ring-polymer fluctuations, leading to spectral distortion. In such cases, the fully adiabatic T_e-PIGS proves more reliable.

Overall, the elevated-temperature ansatz systematically suppresses CMD artifacts. While T_e-PIGS is preferred when a reliable ML PMF is available —particularly in strongly anharmonic regimes— PA-T_e-CMD offers a flexible, user-friendly alternative when training such a model is impractical or data are scarce. Our study clarifies the trade-offs between the two implementations and supports the broader applicability of elevated-temperature path-integral techniques for quantum vibrational spectroscopy. Additionally, we propose a robust and transferable procedure for selecting T_e based on the target spectral region and vibrational frequency range.

As an outlook, we show that integrating flavors of T_e-CMD with a dynamical evaluation of tip-enhanced Raman spectra [6] further allows the investigation of spatially-resolved signatures of quantum nuclear motion.

References

[1] J. Cao and G. A. Voth, J. Chem. Phys. **101**, 6168–6183 (1994).
[2] A. Witt et al., J. Chem. Phys. **130**, 194510 (2009).
[3] F. Musil et al., J. Chem. Phys. **157**, 181102 (2022).
[4] J. Castro, V. Kapil and M. Rossi, *in preparation* (2025).
[5] Y. Litman et al., J. Chem. Phys. **161**, 062504 (2024).
[6] K. Březina and M. Rossi, *in preparation* (2025).

First-principles study of electronic transport properties in thermoelectric materials using NEGF-DFT

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Accurate characterization of electronic transport properties is vital for optimizing the performance of thermoelectric materials. In this study, we employed non-equilibrium Green's function combined with density functional theory (NEGF-DFT) to calculate electronic transport behaviors of classical thermoelectric materials. The preliminary results exhibit notable differences compared with traditional computational methods, motivating us to further investigate the underlying physical mechanisms responsible for these discrepancies. Additionally, we plan to integrate machine learning techniques into our research to explore and identify novel thermoelectric materials with high power factors.

Poster Number 7, Presenter: Sizhen Chen, Shanghai University

The Multiface of Boron Species in Oxidative Dehydrogenation of Propane

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Oxidative dehydrogenation (ODH) of propane provides an energy-saving strategy alternative to the direct propane dehydrogenation in its conversion to olefins, and boron-based materials revealed remarkable catalytic ability. According to our studies, boron shows the multiface character under ODH conditions, and can adopt sp- (-B=O), sp^2- (>BOH, >BOOH), and sp^3- (≡BOH) hybridization modes. The electronic nature of boron in these hybridization modes can critically influence its reactivity in ODHP. The sp^2- (>BOH, >BOOH) and sp^3-hybridized (≡BOH) boron species could selectively activate 2°-C-H bond of propane over its 1°-C-H, which promotes the generation of propylene. In contrast, sp-hybridized boron species (-B=O) is inert to activate C-H bond, but exhibits excellent ability to eliminate alkoxyl radicals, which can prevent the further oxidation of oxygen-containing radical intermediates to deep oxidation products. These insights revealing the multifaceted nature of boron species improve our understanding of the reaction mechanism and are expected to guide the rational design of ODH catalysts.

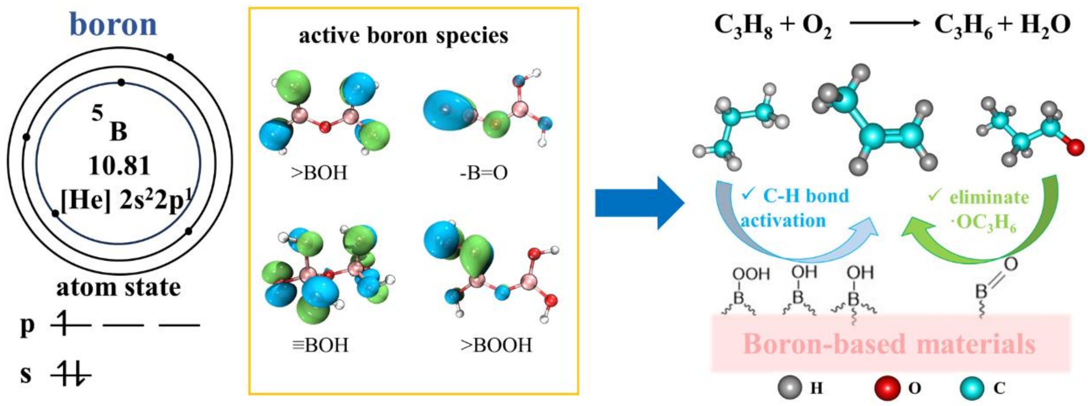

Fig 1. The electronic nature and schematic elucidation of the role of active boron sites in ODHP.

Key words: electronic nature, boron, ODH, reaction mechanism, DFT

Reference

[1] Chen, W.-X.; Liu, Y.-C.; Liu, Z.-Y., et al, *J. Phys. Chem. Lett.* **2024**, *15* (35), 8984-8989.
[2] Liu, Y.-C.; Liu, Z.-Y.; Lu, W.-D., et al, *J. Phys. Chem. Lett.* **2022**, 13(50), 11729-11735.
[3] Liu, Z.-Y.; Xu, D.-T.; Xia, M.-R., et al, *J. Phys. Chem. Lett.* **2021,** *12* (36), 8770-8776.
[4] Liu, Z.-Y.; Lu, W.-D.; Wang, D.-Q.,et al, *J. Phys. Chem. C.* **2021,** *125* (45), 24930-24944.
[5] Liu, Y.-C.; Liu, Z.-Y.; Wang, D.-Q.,et al, *J. Phys. Chem. C.* **2022,** *126* (50), 21263-21271.
[6] Zhou, Q.; Liu, Z.-Y.; Zhu, L.-H.; Lu, W.-D., et al, *J. Phys. Chem. C.* **2023,** *127* (27), 12942-12952.

Dissecting Light-Atom Transport Mechanisms During Titanium Nitride Oxidation via First-Principles Calculations

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Titanium nitride is widely used in harsh environments due to its excellent mechanical and chemical stability. However, its susceptibility to oxidation poses a major challenge to its long-term stability and practical use. Although numerous theoretical studies have been conducted to explore its oxidation mechanism, direct experimental validation remains limited, largely due to the complex, multiscale nature of the oxidation process. In this work, we employed DFT calculations, guided by time-resolved atomic-resolution High-Angle Annular Dark Field imaging, to investigate the oxidation of titanium nitride. Two distinct oxidation pathways were identified experimentally, each exhibiting branching behavior under varying local conditions. By comparing first-principles energies, we analyzed various oxidation modes and atomic rearrangement sequences. Fitting the calculated energy curves revealed a composite mechanism involving simultaneous oxygen interstitial insertion and nitrogen substitution, an ABBA-type rearrangement of Ti atoms, and an oxygen migration pathway aligned with Ti in a Ti–O configuration. This work advances the fundamental understanding of oxidation mechanisms in titanium nitride by revealing the atomic migration behavior of light elements.

Explicitly Correlated Driven Similarity Renormalized Group

Strongly correlated systems requires methods that extend beyond mean-field theory due to their inherently multiconfigurational nature. Dynamic correlation plays a critical role in refining predictions. Recent advancements of Driven Similarity Renormalization Group (DSRG) method, offer intruder-state-free frameworks with promising applications in challenging systems like the Cr2 dimer and organic molecule singlet-triplet gap calculations. Basis Set Incompleteness Error (BSIE) remains a key limitation in wavefunction-based ab initio electronic structure theory. Explicitly correlated F12 theory has emerged as a robust solution, systematically accelerating basis set convergence by incorporating explicit electron correlation terms. In this work, we demonstrate that the Canonical Transformed F12 (CT-F12) method provides a universal, accurate, and computationally efficient strategy to eliminate BSIE in multireference systems. This approach enables high-precision electronic structure calculations while maintaining simplicity in implementation.

Poster Number 10, Presenter: Sy Dong, Beijing Normal University

Poster Abstract

Hands-on Workshop on Electronic-Structure Theory and Artificial Intelligence for Materials Science, Shanghai 2025

Title: *Advancing Relativistic DFT: A Roadmap for Quasi-Four-Component Implementation in FHI-aims*

The accurate modeling of heavy-element systems and spin-dependent phenomena necessitates the incorporation of relativistic effects within electronic-structure calculations. Recognizing this, our recent efforts have been directed towards the development of a quasi-four-component (Q4C) framework within the FHI-aims code, with the goal of achieving a fully relativistic description that maintains computational efficiency.

During my internship under Prof. Volker Blum, we laid the groundwork for this Q4C approach, focusing on a self-consistent treatment of relativistic effects while integrating seamlessly into the existing FHI-aims infrastructure. This initiative aligns closely with the broader objectives detailed in Chapter 2.3 of the FHI-aims roadmap, which emphasizes the expansion of relativistic capabilities in all-electron simulations.

FHI-aims already includes both scalar-relativistic corrections and spin-orbit coupling, which enable the study of many relativistic effects relevant to materials with heavy atoms. However, the Q4C formalism offers a pathway to go beyond these approximations, enabling fully relativistic four-component treatments while retaining variational stability and numerical control. By projecting out negative-energy states using the no-pair approximation and enforcing exact atomic kinetic balance, this method avoids variational collapse and allows consistent inclusion of complex spinor solutions and spin currents. These features are critical for accurately describing chiral molecules, topological materials, and systems with intricate magnetic textures.

This poster outlines the current status of the Q4C implementation and its theoretical underpinnings. We invite discussion and feedback from the community to discuss this exciting development.

Poster abstract

Liu Xinqi

May 30, 2025

Although ab initio calculations based on density functional theory (DFT) have achieved great success in the field of electronic structure calculations, they are still un- suitable for large-scale materials. Twist vdW materials have attracted significant atten- tion from condensed matter physics researchers due to their novel properties. However, performing ab initio calculations on twisted systems incurs enormous computational costs, while semi-analytical models often lack precision in predicting the properties of most twisted systems. In recent years, the rapid development of deep neural networks has given rise to a new approach—deep learning ab initio calculations which holds promise for addressing the accuracy and efficiency challenges faced by traditional ab initio calculations when dealing with large-scale systems such as Twist vdW materi- als. This work focuses on using deep learning methods to calculate and study the band structure of bilayer twisted tellurene and twisted bismuthene, thereby analyzing the ef- fectiveness of this approach. Specifically, by comparing the band structures obtained from deep learning with those from ab initio calculations, we analyze multiple aspects including band matching degree, computational time, and memory usage, ultimately demonstrating the accuracy and efficiency of deep learning methods

Title: Enhancing Thermoelectric Performance of Black Phosphorus via Interlayer Twisting and Organic Molecule Intercalation: A First-Principles Study

Layered materials have garnered significant attention in recent years for thermoelectric due to their quasi-two-dimensional crystal structures. These materials exhibit anisotropic electron and phonon transport, along with low-dimensional electronic characteristics, providing an ideal platform for decoupling thermal and electrical transport. Among them, black phosphorus (BP) is a promising thermoelectric candidate due to its intrinsically high carrier mobility and layer-dependent bandgap. However, its practical application is hindered by low conversion efficiency and high lattice thermal conductivity.

In this work, we employ first-principles calculations combined with Boltzmann transport theory to systematically investigate the impact of interlayer twisting and organic molecule intercalation on the thermoelectric performance of BP. Our results show that both strategies significantly enhance anharmonic phonon scattering, leading to a notable reduction in lattice thermal conductivity. Simultaneously, modulation of the band structure introduces multi-valley features and enables band convergence, resulting in improved Seebeck coefficient and power factor. These effects raise the room-temperature zT value of BP to approximately 0.5.

This study provides new insights into improving the thermoelectric properties of layered materials and opens new directions for the design of non-toxic and flexible high-performance thermoelectric systems.

Poster Number 13, Presenter: Shuai Duan, Qufu Normal University

Electronic Properties via DFTB and Machine Learning

Density functional tight binding (DFTB) parameters, especially parameters in atomic basis functions, require improvement. Machine learning (ML)-based, chemical environment-dependent DFTB parametrization may offer a promising solution for accurate calculations across diverse chemical environments at low computational cost. We introduce a DFTB-ML workflow that allows for the optimization of electronic properties by generating two-center integrals, either by training basis function parameters directly or by training the diatomic integrals, which are then used to build the Hamiltonian and the overlap matrices for the following diagonalization. Using this DFTB-ML workflow, we have managed to obtain improved electronic properties. While both machine learning approaches enabled us to improve on the electronic properties of the molecules as compared with existing DFTB parametrizations, only by training on the basis function parameters we were able to obtain consistent Hamiltonians and overlap matrices in the physically reasonable ranges or to improve on multiple electronic properties simultaneously. Finally, we extend the DFTB-ML workflow to systems with periodic boundary conditions, enabling band structure calculations of large systems at the accuracy level of expensive hybrid functionals. The approach is particularly accurate for predicting the valence band maximum (VBM) and conduction band minimum (CBM). The DFTB-ML model demonstrates robust performance across diverse chemical environments, including molecules, bulk materials, slab models, and defect systems.

Poster Number 14, Presenter: Guozheng Fan, Xiaogan Sichuang Information Technology Co., LTD.

Study on Interfacial Layer of SnTe Device with Cu Electrode

PbTe is one of the best thermoelectric materials in the medium - temperature range (500 - 900 K), but its application is limited by Pb's toxicity. As Sn and Pb are in the same group, SnTe has sparked great interest as a PbTe substitute.

However, most current SnTe research focuses on boosting material performance. The lack of module studies severely restricts SnTe's practical application, a dilemma faced by many advanced thermoelectric materials. Consequently, although SnTe's ZT value has been enhanced, its device - fabrication performance remains unsatisfactory.

The actual energy conversion efficiency and service life of thermoelectric devices largely depend on the assembly process and contact interfaces. Connecting electrodes with thermoelectric materials is central to assembling thermoelectric modules.

To boost SnTe's thermoelectric conversion efficiency, finding a suitable interfacial layer between SnTe and electrode materials has become a key task and a focal point of our research.

Poster Number 15, Presenter: Xiaoyu Fei, Qufu Normal University

Dynamic Simulation of Copper Catalysts for CO_2RR by Machine Learning Potential

ABSTRACT: Catalytic processes are inherently dynamic phenomena at elevated temperatures, and understanding the dynamic reconstruction of catalyst surfaces is crucial for catalyst design. Traditional molecular dynamics methods, whether based on ab initio (AIMD) or empirical potentials, face trade-offs between spatiotemporal scales and computational accuracy. Starting from high-accuracy DFT data, we employ machine learning potential to achieve large-scale, long-duration, high-precision dynamic simulations of catalyst surfaces, capturing the dynamic reconstruction process. We investigate the thermally induced dynamic reconstruction of copper catalyst surfaces and its effects on CO_2 adsorption behavior, aiming to advance the understanding and design of copper catalysts.

KEYWORDS: copper catalysts, heterogeneous catalysis, machine learning potential, CO_2RR

Poster Number 16, Presenter: Borui Feng, Changchun Institute of Applied Chemistry

Low-Scaling GW Approach Based on Pseudopotential and Numerical Atomic Orbitals

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Abstract

The GW method, a cornerstone of many-body perturbation theory in condensed matter physics, offers superior accuracy in quasiparticle band structure calculations compared to conventional density functional theory (DFT). However, its notorious $\mathcal{O}(N^4)$ computational scaling has severely restricted practical applications to small-scale systems. We report a novel low-scaling GW implementation based on pseudopotentials and numerical atomic orbitals (NAOs) basis sets, developed within the open-source DFT software, ABACUS, integrated with the LibRPA library for post-DFT GW calculations framework. This implementation achieves unprecedented $\mathcal{O}(N^2)$ computational complexity through space-time formalism, strict locality of NAOs basis functions, and localized resolution of identity (LRI) approximations for efficient product basis construction. Validations against the established FHI-aims code demonstrate exceptional agreement in semiconductor band structure predictions. Systematic analysis of pseudopotentials and basis sets reveals that their choice critically influences both the convergence behavior and computational efficiency of GW calculations, despite comparable accuracy in predicted bandgaps. Our results emphasize the critical role of strategic pseudopotential-basis set pair optimization in achieving an optimal balance between computational accuracy and resource efficiency for realistic many-body simulations. The ABACUS+LibRPA framework thus enables robust, high-throughput GW calculations for large-scale material systems, bridging the gap between accuracy and efficiency in electronic structure theory.

Keywords: GW method, FHI-aims benchmark, pseudopotentials, numerical atomic orbitals, low-scaling algorithms, ABACUS+LibRPA

Implementation of full BSE and TDA BSE with NAO and pseudo potential

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Abstract

We have developed an implementation of Bethe-Salpeter equation (BSE) in ABACUS, which is an ab-initio electron structure package utilizing numerical atomic orbital (NAO) and pseudo potential, while Screened Coulomb potential and GW energy are obtained by LibRPA. Both Tamm-Dancoff approximation version and full version BSE Hamiltonian are constructed based on Resolution of Identity technique, and are solved efficiently with ELPA. The result of exciton energy and absorption spectrum are compatible with FHI-aims.

Given that full BSE Hamiltonian is a non-hermitian matrix, we carefully consider the bi-orthogonal property of its right eigenvector and left eigenvector. This property was ignored by many packages to the best of our knowledge.

Keywords: Bethe-Salpeter equation, non-hermitian, beyond Tamm-Dancoff approximation

Magnetoelectric Coupling Mechanisms in Altermagnetic Multiferroic Moiré Superlattices

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Abstract: Altermagnetism, as an emerging magnetic order characterized by compensated collinear moments and momentum-dependent spin splitting, has recently attracted significant attention for its potential in next-generation ultrafast spintronics [1-3]. The investigation of interaction between altermagnetism and ferroelectricity is particularly intriguing—such magnetoelectric coupling effects could enable novel memory and logic devices. This raises a fundamental scientific question regarding altermagnetic superlattices: Can moiré twisting induce intrinsic magnetoelectric coupling in altermagnetic multiferroics, and what governs its evolution? Meanwhile, the scarcity of intrinsic 2D altermagnetic materials has substantially hindered progress in this field.

In this work, we present the first explicit physical mechanism for altermagnetic-order-induced ferroelectric polarization through combined symmetry analysis and microscopic electric dipole theory. By establishing the symmetry relationship between polarization vectors and Néel order parameters in altermagnets, we reveal their strong interdependence and classify 2D altermagnets into eight distinct magnetoelectric coupling types based on layer group symmetry. First-principles calculations on prototype monolayer $MgFe_2N_2$ validate our theoretical framework. We further propose utilizing magneto-optical Faraday rotation to detect Néel vector orientation and associated polarization in altermagnetic multiferroics.

Moreover, through high-throughput screening of a 100,000+ 2D material database with symmetry criteria for altermagnetism, we've identified over 1,000 intrinsic 2D altermagnetic multiferroics. Notably, we anticipate employing machine learning on our established altermagnetic material database to discover more intrinsic altermagnets and investigate their magnetoelectric coupling mechanisms in moiré superlattices. Our work bridges the strong magnetoelectric coupling of Type-II multiferroics with the unique advantages of altermagnets, while incorporating machine learning tools to pioneer new pathways for developing multifunctional altermagnetic spintronics.

Keywords: altermagnetic multiferroics, moiré superlattices, machine learning, first-principles calculations, high-throughput screening

Reference

[1] L. Šmejkal, J. Sinova, T. Jungwirth, Emerging Research Landscape of Altermagnetism, *Phys. Rev. X* 12, 040501 (2022).

[2] L. Šmejkal, J. Sinova, T. Jungwirth, Beyond Conventional Ferromagnetism and Antiferromagnetism: A Phase with Nonrelativistic Spin and Crystal Rotation Symmetry, *Phys. Rev. X* 12, 031042 (2022).

[3] J. Krempaský, L. Šmejkal, S. W. D’Souza, *et al.*, Altermagnetic lifting of Kramers spin degeneracy, *Nature* 626, 517 (2024).

[4] Wen-Ti Guo, Junqi Xu, Yurong Yang, Huaiqiang Wang, Haijun Zhang, Altermagnetic type-II Multiferroics with N´eel-order-locked Electric Polarization, *Phys. Rev. Lett.* under review (2025).

[5] Wen-Ti Guo, Junqi Xu, Xinqi Liu, Rongxiang Zhu, Huaiqiang Wang, Haijun Zhang, High-throughput screening and classification of intrinsic altermagnetic monolayers, *Nat. Commun.* to be submitted (2025).

Title:

Optimization Scheme for Reduced Density Matrix Functional Theory

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Abstract:

Reduced density matrix functional theory (RDMFT) is a theory with great potential. It uses the single-particle reduced density matrix (1-RDM) as the basic variable, expresses the kinetic energy of the particle as an explicit functional of the density matrix, and allows orbitals to be occupied in fractional form. It can better describe strongly correlated quantum systems and breaks through the limitations of traditional density functional theory. A key challenge of RDMFT is how to reliably and efficiently optimize 1-RDM to obtain ground state properties. Currently, we are trying to use iterative diagonalization methods and direct minimization-based methods to solve the optimization problem of RDMFT, especially its convergence behavior and computational efficiency, and hope to extend it to solid systems.

Keywords:

RDMFT, ground-state optimization, strongly correlated systems

Multi-Level Mapping of Lithium Transport Pathways in Semi-Crystalline PEO Electrolytes

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Solid-state lithium batteries promise safer, higher-energy storage than current liquid-electrolyte systems, yet their progress is throttled by the modest ionic conductivity of the benchmark solid electrolyte, polyethylene oxide (PEO). The origin of this limitation lies in PEO semi-crystalline morphology: amorphous regions enable Li+ motion, whereas crystalline ones act as virtually insulating towards Li+ conductivity. A quantitative, atomistically grounded picture of Li^+ diffusion is therefore essential, but difficult to obtain because parameterising long-chain polymers for faithful large-scale simulations remains an open challenge. To overcome this gap, we construct an integrated, three-tier computational framework that sequentially combines (i) density-functional theory (DFT) for undertanding bonding chemistry and T=0 K limit, (ii) classical molecular dynamics (MD) for mesoscale conformational sampling, and (iii) a tailored machine-learning potential (MLP) MD that fuses the accuracy of DFT with the efficiency of MD. Here, by DFT calculations, we identify the range of Li^+ local configurations in PEO, revealing distinct low- and high-energy sites that set the fundamental diffusion limits, while by using conventional force-field MD, we reveal why crystallinity affects Li diffusion. However, we find that comparison with DFT shows force-field diffusion barriers can deviate, leading to an order-of-magnitude spread in predicted Li^+ diffusivity. Training a PEO-specific MLP on the DFT data (initially prototyped with the MACE framework) reduces those errors while also reducing the computational cost to that comparable to classical MD. The resulting potential permits ns-scale trajectories for polymer cells, a regime inaccessible to brute-force DFT. By linking together DFT, MD, and MLP, we establish a coherent storyline from quantum-level interactions to identifying the diffusion-limiting step to rational design of next-generation PEO. Beyond clarifying the Li^+–PEO chemistry, we develop a workflow that is a template for other high-molecular-weight polymer electrolytes, accelerating the rational design of solid-state batteries that couple high energy density with uncompromised safety.

AI-enabled computation of high-temperature Raman spectroscopy

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Accurate simulation of Raman spectroscopy is important to understand corresponding experimental measurement and revolve materials' atomistic structure. In this work, We combined Latin Hypercube Sampling (LHS) and the anharmonic special displacement method (A-SDM) to perform thermodynamic calculations for elemental zirconium, Which not only reduces the computational cost of phonon renormalization modeling, but also dramatically decreases the number of samples required to describe the parameter space of anisotropic thermal expansion

Using this approach, we computed thermodynamic quantities, including Gibbs free energy and phase diagrams, for three phases of pure zirconium (α, ω, and β phases). The predicted phase boundaries and lattice expansion show qualitative agreement with experimental characterizations, with accuracy surpassing that of traditional quasi-harmonic approximation models.

Poster Number 22, Presenter: Jinchang Hu, Shanghai University

p-type Dopability in Half-Heusler Thermoelectric Semiconductors

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Keywords: p-type dopability, half-Heusler, defect formation energy, complex

Half-Heusler (HH) semiconductors with high valence band degeneracy are promising p-type thermoelectric materials. However, effective p-type dopability in HH semiconductors remains a significant challenge, hindering the further development of high-performance p-type HH thermoelectrics. In this work, we investigate p-type dopability in NbFeSb, NbCoSn, and ZrNiSn through comprehensive first-principles defect calculations. We identify that interstitials (B_{i}) and vacancies (V_A) primarily limit dopability by setting the maximum achievable doping level. Using dopability limits and pinned Fermi levels as indicators, we systematically evaluate the doping feasibility of p-type dopants at three different Wyckoff positions. Moreover, by taking NbCoSn as an example, p-type dopability was experimentally scrutinized, validating the calculation results. This work offers valuable insights into the challenges associated with p-type doping in HH compounds and provides practical guidance for the experimental selection of suitable host materials and dopants, which can help facilitate the realization of effective p-type doping.

Molecule Activation Mechanism on Cobalt Facets towards Rational Catalysts Design

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Keywords: Cobalt, F-T Synthesis, CO_2 Activation, Reaction Kinetics

Visualizing surface reactions through the combination of density functional theory calculations and high-resolution scanning tunneling microscopy is critically important for advancing the fundamental understanding of catalysis. This approach provides unique and intuitive evidence for numerous significant reaction mechanisms, enabling insights into the elementary steps, transition states, and key intermediates of surface and interface catalytic reactions at the atomic level. We have conducted in-depth investigations into the elementary processes of important small molecules, such as CH_3OH、H_2、CO、H_2O、O_2 and CO_2, on the single-crystal surfaces of cobalt-based catalysts. Our work focuses on the fundamental mechanisms of adsorption, diffusion, dissociation, activation, and interaction for these molecules.

References

[1]. **Huang C.*** et al., *J. Phys. Chem. Lett.* **2022**, 13, 5131.

[2]. **Huang C.*** et al., *J. Phys. Chem. Lett.* **2023**, 14, 9385.

[3]. **Huang C.*** et al., *J. Phys. Chem. C.* **2024**, 128, 20811.

[4]. **Huang C.*** et al., *J. Phys. Chem. Lett.* **2025**, 16, 2529.

[5]. **Huang C.*** et al., *J. Am. Chem. Soc.* **2025**, 147, 19, 16164.

The photocarrier lifetime of monolayer MoS_2 modulated by surface-adsorbed oxygen molecule

Abstract

Two-dimensional molybdenum disulfide (MoS_2) is a promising material for advanced photonic and optoelectronic devices due to its unique properties and strong light-matter interactions. The performance of these devices is largely determined by the lifetime and behavior of photoexcited carriers. During preparation or transfer, monolayer MoS_2 is often exposed to ambient gaseous environments, where oxygen molecules can significantly influence carrier dynamics. Using ab initio nonadiabatic molecular dynamics within surface-hopping framework, we investigate how single oxygen molecule adsorption modulates photocarrier lifetimes in monolayer MoS_2. Our simulations reveal that both physisorbed and chemisorbed O_2 molecules accelerate electron-hole recombination but through distinct pathways and physical mechanism. Physisorbed triplet O_2 reduces carrier lifetimes from nanoseconds in pristine MoS_2 to hundreds of picoseconds via high-frequency phonon excitation and enhanced electron-phonon coupling, challenging predictions of the Shockley-Read-Hall model. In contrast, chemisorbed O_2 at sulfur vacancies slightly shortens lifetimes through defect-assisted recombination. These findings provide a simple yet effective strategy to tune photocarrier lifetimes in monolayer MoS_2, expanding its potential applications in optoelectronics.

Poster Number 25, Presenter: Longjuan Kong, National University of Defense Technology

Impact of material descriptors on the prediction of thermoelectric property in a machine learning-based approaches

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Abstract:

The performance of thermoelectric devices, which can harness waste heat into useful energy, has been mainly hinged on their underlying material physics. Machine learning has enabled the search for effective thermoelectric materials within the extensive chemical combinatorial space to be more practical. Material descriptors, which encapsulate material information, play a vital role in the development of effective machine learning models for predictive and inverse-design applications. We can utilize descriptors derived from physical characteristics or chemical composition to generate feature vectors for ML algorithms. Identifying suitable descriptors that capture essential aspects of material chemistry and physics can greatly improve the thermoelectric property prediction of ML models, aiding in the inverse design of materials for thermoelectric applications. Our study aims to compare various structure-based physical descriptors, including Sine and Ewald-sum matrices, with composition-based chemical descriptors, such as combination-based feature vectors and atomic element property composition vectors for ML applications of thermoelectric property prediction. We investigated the impact of these descriptors on the predictive capability of ML algorithms such as XG-Boost (XGB), Random Forest (RF), and Deep Neural Network (DNN) for prediction of power factor, a crucial indicator of thermoelectric performance, and provided insights into their performance. The R^2 scores of models utilizing chemical descriptors consistently outperform those using physical descriptors across various investigated ML models, with the highest R^2 score of 0.92 being achieved by the RF model with an average mean absolute percentage error (MAE) of 16%. Our findings indicate that chemical descriptors effectively capture the underlying material physics for thermoelectric property prediction tasks, surpassing structure-based physical descriptors.

AI-driven intelligent Raman spectrometer: Optimizing optical design and component selection to achieve high performance

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The design of traditional Raman spectrometers relies on expert experience and repeated trial and error, making it difficult to achieve the optimum under multiple objective constraints such as cost, size, sensitivity and spectral range. This study proposes an innovative artificial intelligence (AI) -assisted design framework for the development of the next-generation Raman spectrometer. We have proved the effectiveness of this method through simulation and experimental verification, and successfully designed and built an optimized prototype of a compact and high-performance Raman spectrometer. Compared with traditional design, the AI-driven approach significantly shortens the design cycle and achieves an improvement in signal-to-noise ratio under the same cost constraints. This research provides a new paradigm for the development of more intelligent and more adaptable Raman instruments to diverse needs.

Poster Number 27, Presenter: Sida Li, Shanghai University

First-Principles Study of Magnons in Spin-Frustrated Two-Dimensional VI_2

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Spin-frustrated magnets, due to the presence of a large number of energy-equivalent ground states in their systems, exhibit some novel quantum effects and have thus attracted widespread attention. Here, we systematically study magnons, the fundamental collective excitations in magnetic materials, in spin-frustrated two-dimensional VI_2. The magnon band structures, density profiles and wavefunctions are computed, using a fully first-principles approach for spin dynamics based on density functional perturbation theory. The influence of the non-collinear magnetic structure and the effect of spin-orbit coupling are revealed. Our study promotes the understanding of collective spin excitations in spin-frustrated magnets and provides critical guidance for designing magnetic spintronic devices.

Rational Design of Perovskite Electrocatalysts for Oxygen Evolution Reaction via Center-Environment Deep Learning

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This study develops an integrated computational-experimental framework combining deep learning, high-throughput screening, and first-principles calculations to accelerate the discovery of efficient perovskite-based oxygen evolution reaction (OER) catalysts. We established a comprehensive dataset of OER overpotentials and adsorption free energies for key intermediates (*OH, *O, *OOH) across 171 perovskite oxides. A novel Center-Environment based artificial neural network (ANN) model achieved exceptional predictive accuracy (R^2=0.99). Systematic exploration of 73 potential cation substitutions at A/B sites generated 5,329 new perovskite configurations, with doping strategies expanding the virtual library to 101,251 structures. Experimental validation identified CrFeO3 as the most promising candidate, exhibiting a remarkably low overpotential of 319 mV. DFT calculations elucidated atomic-level OER mechanisms, establishing structure-activity relationships to guide rational electrocatalyst design.

Poster Number 29, Presenter: Yihang Li, Shanghai University

AI-enabled computation of Molecular Dynamics and Electronic Structure

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Accurate simulation of large-scale material systems is crucial for understanding the corresponding experimental measurements and atomic structures of device materials. In this work, we used a deep learning method based on equivariant graph neural network, DeepH, to simulate the electronic structure of graphene/hexagonal boron nitride system at different twist angles. In the future, we will also combine large-scale molecular dynamics simulations to simulate the structure of large-scale two-dimensional structures and perform more accurate property calculations.

Poster Number 30, Presenter: Yilin Li, Shanghai University

Recently, the quantum anomalous Hall effect (QAHE) has been theoretically proposed in compensated antiferromagnetic systems by using the magnetic topological insulator model [Phys. Rev. Lett. 134, 116603 (2025)]. However, the related and systematic study based on a realistic material system is still limited. As the only experimentally realized antiferromagnetic topological insulator, $MnBi_2Te_4$ becomes a vital platform for exploring various topological states. In this work, by using comprehensive first-principles calculations, we illustrate that the QAHE can also be realized in compensated antiferromagnetic even-septuple-layer $MnBi_2Te_4$ without combined parity-time (PT) symmetry. Using a magnetic topological insulator model, the layer-resolved Chern number is calculated to further understand the presence of different Chern numbers. The application of external hydrostatic pressure will strengthen the Te-Te quasicovalent bond due to the dramatic compression of the van der Waals gap. Thus, the topological nontrivial gap exceeds the room-temperature energy scale in a wide range of pressures. Additionally, we find that constructing $MnBi_2Te_4/CrI_3$ heterostructure can realize the compensated antiferromagnetic configurations with QAHE. Our work demonstrates the realization of QAHE in compensated antiferromagnetic even-septuple-layer $MnBi_2Te_4$ and provides a reliable strategy to obtain the corresponding magnetic configurations.

Poster Number 31, Presenter: Zeyu Li, University of Science and Technology of China

Abstract

In recent years, rapid progress has been made in solid-state lithium batteries. Among various technologies, coating the surface of electrodes or electrolytes has proven to be an effective method to enhance interfacial stability and improve battery cycling performance. Recent experimental studies showed that gas-solid reactions offer a convenient approach to form modified coating layers on the solid electrolyte. Here, we performed computational simulations to investigate this surface reaction process. Specifically, we simulated the gas-solid reactions of Li_6PS_5Cl(LPSC) solid-state electrolytes in pure CO_2 and in mixed CO_2/O_2 atmospheres using ab-initio molecular dynamics (AIMD) and machine-learning force fields (MLFF)-accelerated molecular dynamics (MD) approaches. In the former case, LPSC surfaces primarily form Li_2CO_2Sbecause it is difficult to dissociate another oxygen atom from the second CO_2 molecule. While in CO_2/O_2 mixed atmosphere, O_2molecules preferentially adsorb onto LPSC, which supplies oxygen sites for subsequent CO_2 adsorption to form carbonate -CO_3 units. This reaction pathway ultimately generates an interfacial product dominated by Li_2CO_3. These coatings exhibit distinct electronic and ionic conductivity characteristics, allowing the possibility to control coating compositions and configurations by adjusting the gas-solid reactions. Key criteria for applying this strategy are extracted from the current research.

Poster Number 32, Presenter: Zicun Li, Institute of Physics, Chinese Academy of Sciences

Relativistic configuration-interaction and coupled-cluster calculations of Ir^{17+} transition energies and properties for optical clock applications

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The transition energies and properties of the Ir^{17+} ion are calculated using the Kramers-restricted configuration-interaction (KRCI) and Fock-space coupled-cluster (FSCC) methods within the Dirac-Coulomb-Gaunt Hamiltonian framework. These calculations show several forbidden optical transitions between the $4f^{13}5s$ ground state and the $4f^{14}$ and $4f^{12}5s^2$ excited states, underscoring their potential as candidates for optical clock applications. Additionally, key properties of the ground and low-lying excited states are reported, including Landé g_J factors, lifetimes, electric dipole polarizabilities, electric quadrupole moments, hyperfine structure constants, relativistic sensitivities, Lorentz-invariance coefficient tensor, and isotope shifts. The excellent agreement between the results from the KRCI and FSCC methods demonstrates the robustness of the calculations and confirms the reliability of the proposed clock transitions.

TABLE I. Energy levels (in cm^{-1}) of the Ir^{17+} ion. The KRCI (e32-CISD) calculation and FSCC (e60-CCSD) calculations consider the influence of basis set size, truncation of virtual orbitals, and triple excitation. Analysis of uncertainties (given in parentheses) is provided in the Appendixes.

			CI					CC	
Config.	Term	g factor	KRCI	Ref.[1]	Ref.[2]	Ref.[3]	Ref.[4]	FSCC	Ref.[5]
$4f^{13}5s$	$^3F_4^o$	1.2477	0	0	0	0	0	0	0
	$^3F_3^o$	1.0490	4769(45)	4838	4236	4777	4647	4655 (74)	4662
	$^3F_2^o$	0.6642	26194(26)	26272	26174	25186	25198	25476(129)	25156
	$^1F_3^o$	1.0296	31381(109)	31492	30606	30395	30167	30579(232)	30197
$4f^{14}$	$^1S_0$	0.0000	12006(1392)	5055	5091	12382	7424	10203(3563)	13599
$4f^{12}5s^2$	$^3H_6$	1.1612	28848(2117)	35285	33856	30283	29695	27445(3007)	24221
	$^3F_4$	1.1359	38221(2019)	45214	42199	39564	39563	36062(2774)	33545
	$^3H_5$	1.0309	53162(2115)	59727	58261	53798	53668	51059(3120)	47683
	$^3F_2$	0.8426	60530(1509)	68538	63696	61429	62140	56931(2523)	55007
	$^1G_4$	0.9927	61953(2058)	68885		62261	62380	59318(2977)	56217
	$^3F_3$	1.0809	64577(1795)	71917	68886	65180	65438	61492(2886)	58806
	$^3H_4$	0.9141	85218(2150)	92224	66296	84524	84662	82019(3066)	78534
	$^1D_2$	1.1313	89257(1064)	98067	117322	89273	91341	84415(2351)	82422
	$^1J_6$	1.0005	100121(688)	110065		101136	103487	96331(2120)	93867

References

[1] J. C. Berengut, V. A. Dzuba, V. V. Flambaum, and A. Ong, Phys. Rev. Lett. 106, 210802 (2011).
[2] U. I. Safronova, V. V. Flambaum, and M. S. Safronova, Phys. Rev. A 92, 022501 (2015).
[3] C. Cheung, M. S. Safronova, S. G. Porsev, M. G. Kozlov, I. I. Tupitsyn, and A. I. Bondarev, Phys. Rev. Lett. 124, 163001 (2020).
[4] V. A. Dzuba and V. V. Flambaum, Phys. Rev. A 108, 053111 (2023).
[5] A. Windberger, J. R. Crespo López-Urrutia, H. Bekker, N. S. Oreshkina, J. C. Berengut, V. Bock, A. Borschevsky, V. A. Dzuba, E. Eliav, Z. Harman, U. Kaldor, S. Kaul, U. I. Safronova, V. V. Flambaum, C. H. Keitel, P. O. Schmidt, J. Ullrich, and O. O. Versolato, Phys. Rev. Lett. 114, 150801 (2015).
[6] Y. M. Yu and B. K. Sahoo, Phys. Rev. A 109, 023106 (2024).
[7] Y. M. Yu, P. Duo, S. L. Chen, B. Arora, H. Guan, K. Gao, and J. Chen, Atoms 10, 123 (2022)

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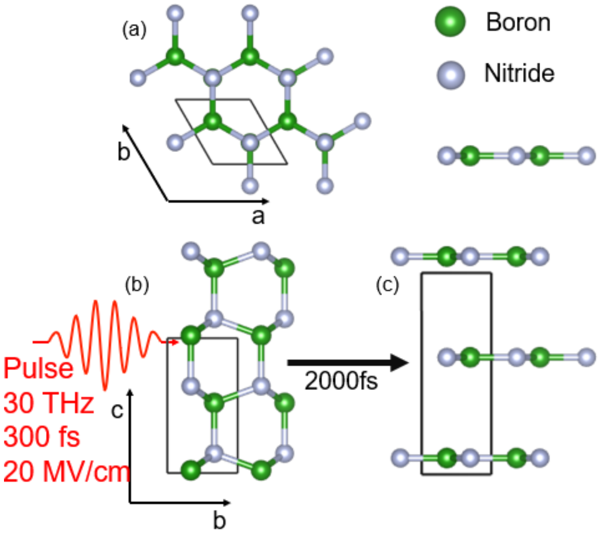
DFT study: Laser Driven Bulk-to-Layered Phase Transition

Light-induced phase transitions are pivotal in the study of light–matter interactions. While previous research has focused on symmetry changes in bulk materials or sliding in vdW structures, transitions bridging these two phases are rarely explored.

In this project, we construct a new computational method, phonon selective dynamics (PSD), for studying light-induced phase transition. We found that with a 30 THz, 300 fs duration time, 20 MV/cm laser pulse, wurzite Boron Nitride (wBN, bulk structure) can be transformed to hexagonal Boron Nitride (hBN). This transition is associated with the excitation of polar and anti-polar phonon modes, which involve strain along the crystallographic c-axis. Our computational results also suggest the optimal conditions for the experiment.

Our results filled the gap of phase transition induced by light from bulk to vdW structure, provide insights into controlling phase transitions via targeted phonon excitations, offering potential pathways for material property modulation.



Poster Number 34, Presenter: Shuang Liu, Guangdong Technion-Israel Institute of Technology

The main influencing factors of strong correlation system DFT+U scheme

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Supervisor : Xinguo Ren

The DFT+U method is a widely used scheme of density functional theory (DFT) for describing strongly correlated systems, particularly those involving localized d and f electrons. However, it has often been treated as a black-box approach, with limited understanding of how specific parameters affect the results. In this study, we test the impact of several key factors in DFT+U. The most critical are the Hubbard U and Hund's J parameters, which represent on-site Coulomb repulsion and exchange interactions. U and J, are obtainable through empirical fitting or first-principles methods such as linear response theory, and have a direct influence on the system's electronic and magnetic properties. For example, without introducing U and J, DFT cannot correctly open the bandgap of Mott insulator. Another essential element is the double counting correction, which compensates for correlation effects already included in the DFT functional. The Fully Localized Limit (FLL) is typically used for systems with strong electron localization, while the Around Mean Field (AMF) scheme may be more appropriate for weakly correlated or itinerant systems. Equally important is the projection framework used to define the correlated subspace. On-site, dual (Mulliken), and full representations differ in how they localize orbitals and affect occupancy. Among them, dual and full projections yield similar results, while on-site projection shows deviations but offers lower computational cost. Overall, the choice of U and J values is the most influential factor, followed by the projection scheme and double counting correction. A proper choice of these parameters is essential to ensure the accuracy and reliability of DFT+U results for correlated electron materials.

AI-Driven Innovation in Semiconductor Materials Development

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This study presents AI4Semiconductor, a groundbreaking AI-powered multiscale materials R&D platform that revolutionizes semiconductor development through the synergistic integration of adaptive optimization algorithms and cross-scale dynamics simulations. The platform's transformative capabilities stem from two core innovations: an Intelligent Material Design System and a Dynamic Evolution Simulation Engine. The Intelligent Material Design System uses machine learning-enhanced high-throughput computational methods to establish predictive models for composition-structure-property relationships, while simultaneously constructing a semiconductor material genome database with over 100,000 material property parameters. Complementing this, the Dynamic Evolution Simulation Engine employs advanced equivariant geometric neural networks to model interatomic interaction potentials with unprecedented precision and integrates metadynamics-enhanced sampling techniques to achieve atomic-scale visualization of nanosecond-level epitaxial growth dynamics. Empirical validation demonstrates that the platform reduces the R&D cycle of semiconductor materials by over 60% while enhancing computational accuracy in multiscale simulations by two orders of magnitude. These advancements establish a scalable framework for redefining design paradigms in next-generation semiconductor technologies, particularly in emerging fields such as perovskite semiconductors and two-dimensional electronic devices. By bridging computational materials science with industrial innovation, AI4Semiconductor addresses critical challenges in accelerating the discovery-to-production pipeline.

Theoretical Insights into Atomically Exposed Pd@BN Catalysts for Selective Acetylene Hydrogenation

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Strong metal–support interactions (SMSI) traditionally rely on reducible oxide supports to stabilize metal species. In this work, a new approach to extend SMSI to non-reducible supports via h-BN encapsulation was explored, enabling the stabilization of atomically dispersed Pd species and offering unique opportunities for reaction pathway control. Density functional theory (DFT) calculations were employed to explore the structural and electronic properties of Pd atoms confined at the h-BN interface. The results reveal that electron transfer from Pd to the h-BN support creates an electron-deficient Pd center, which plays a pivotal role in modulating adsorption energetics. In particular, the weakened binding of ethylene suppresses over-hydrogenation, favoring its desorption and thus enhancing selectivity. DFT-based reaction pathway analysis indicates that the potential energy surface for acetylene hydrogenation is significantly altered by the atomic-scale Pd environment. The activation barrier for the first hydrogenation step remains low, while further hydrogenation of ethylene becomes kinetically less favorable due to the modified electronic structure of Pd. These theoretical insights are supported by experimental characterizations (XPS, CO-DRIFTS), which confirm the presence of atomically isolated Pd and strong SMSI effects. This study highlights the power of DFT to guide the design of stable, selective hydrogenation catalysts through electronic structure engineering at the metal–support interface.

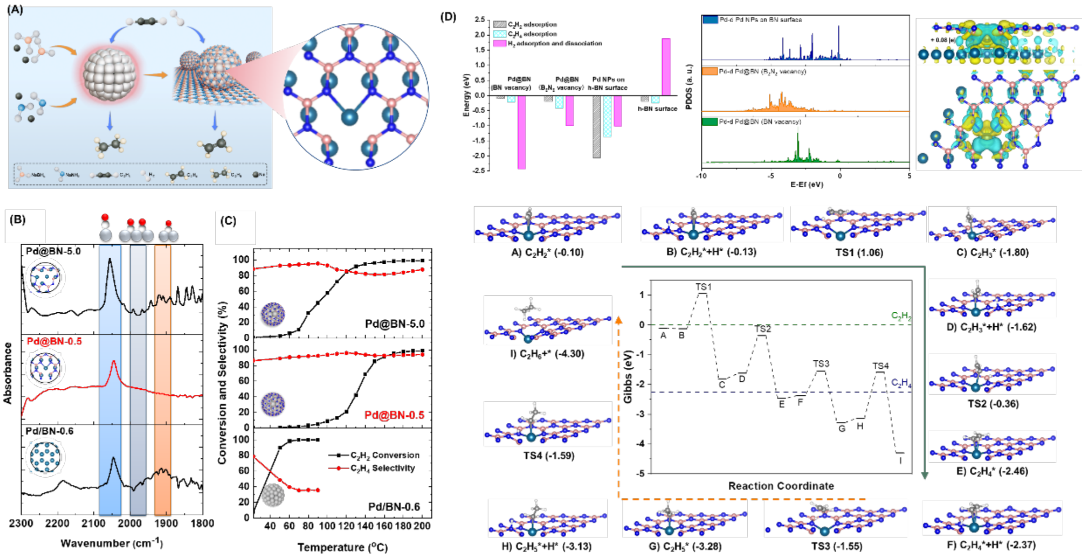

Figure (A) Schematic illustration of the growth strategy for achieving atomically dispersed Pd species via h-BN encapsulation of Pd nanoparticles; (B) CO-DRIFTS spectra of various BN-supported catalysts; (C) Acetylene conversion and ethylene selectivity over different catalysts in selective hydrogenation reactions; (D) Proposed reaction pathway and underlying mechanism for acetylene selective hydrogenation catalyzed by atomically dispersed Pd sites.

Aitomia: Your Intelligent Assistant for AI-Driven Atomistic and Quantum Chemical Simulations

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The automation of quantum chemical and atomistic simulation workflows continues to draw considerable attention, motivating the development of efficient, scalable, and user-friendly approaches—particularly those that enable interaction through natural language. In present era one of best scalable solution is Large Language Models (LLMs) and Artificial Agents with their excellent capability in natural language processing and logical workflows gained attraction to make the workflow semi or fully automated in different domain of science. Here we develop Artificial Intelligent (AI) based assistant named Aitomia for quantum chemical and machine learning enhanced atomistic simulations. Aitomia leverages the power of AI agents and logical workflows, to provide users-friendly and easy to handle, monitor and analyze these simulations. Aitomia utilizes MLatom software package for running all these quantum chemical and machine learning enhanced simulations. Aitomia will be made accessible integrated with Aitomistic Hub. Now demo version can be accessible at https://www.aitomistic.xyz/ . It is expected to accelerate and make easier exploration of chemical space for basic research, material and drugs design. More description about how to use and obtain this platform can be obtained from http://mlatom.com/aitomia [1].

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[1] Hu, J.; Nawaz, H.; Rui, Y.; Chi, L.; Ullah, A.; Dral, P. O. *arXiv preprint arXiv:2505.08195* **2025**.

AI-enabled computation of high-temperature Raman spectroscopy

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Accurate simulation of Raman spectroscopy is important to understand corresponding experimental measurement and revolve materials' atomistic structure. In this work, we developed an accurate method for Raman spectroscopy simulation at high temperature based on molecular dynamics powered by a force field trained with the equivariant graph neural network method MACE. We show that our method successfully reproduced experimental Raman peaks on the defected $LaFeO_3$ system; and revealed the abnormal peak shift of V_2O_5 at certain temperature. Understanding and detailed analysis of the results is in progress.

Poster Number 39, Presenter: Hengkang Pan, Shanghai University

RPA100 : All-electron RPA atomization energies for 100 molecules without single-particle basis set errors.

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Supervisor : Xinguo Ren

Among various electronic structure methods, the random phase approximation (RPA) stands out as a cornerstone that bridges quantum chemistry methods and density functional theory. Practical applications show that RPA-based methods are suitable for describing delicate ground-state energy differences for both molecular and extended systems. However, a major hurdle in the RPA calculations is its very slow convergence with respect to one-electron basis set, rendering the numerically fully converged results different to obtain. Calculating numerically converged RPA atomization energies via extrapolation is highly non-trivial. To obtain reliable results free from basis set errors, we numerically solve the Sternheimer equation in the finite element space, representing the first-order wavefunctions on a dense finite element grid. This approach avoids expressing the first-order wavefunctions in terms of unoccupied states, which are dependent on the single-particle basis set. As a result, we obtain a density response function and RPA correlation energy that are free from single-particle basis set errors. This enables us to compute reliable molecule atomization energies, providing benchmark data for future basis set development and offering guidance for solving basis set errors in other correlated methods.

First-Principles Insights into Multi-Component Alloying Effects on the Stability and Mechanical Properties of Molybdenum-Based Alloys

LinQin Qin, Yuchao Tang, Yiheng Sheng, Jiong Yang, and Yi Liu

Abstract

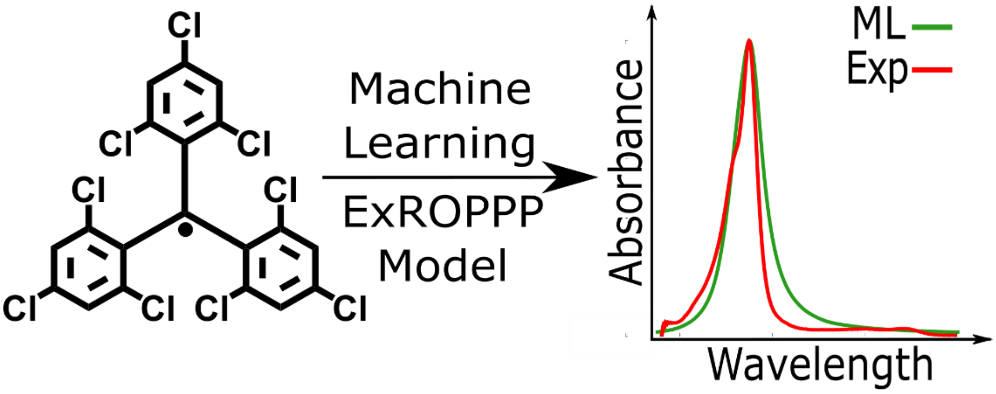
This study systematically investigates the single- and double-site nonequivalent substitution effects of 3d (Ti, V, Cr, Mn, Fe), 4d (Y, Zr, Nb), and 5d (Ta, W, Re) transition metal elements in body-centered cubic (BCC) and face-centered cubic (FCC) molybdenum (Mo) matrices using first-principles density functional theory (DFT). Through the construction of 418 alloy models (209 for BCC-Mo and 209 for FCC-Mo), the regulatory mechanisms of multi-component alloying on the stability, plasticity, and mechanical properties of Mo-based alloys are revealed by combining substitution energy calculations, new materials index, elastic constants analysis, and solid solution strengthening models. Substitution energy calculations demonstrate that in single-site substitution, Ti, V, Zr, Nb, Ta, and W enhance the stability of BCC-Mo, with Ti exhibiting the most pronounced effect. For FCC-Mo, Ti, V, and Fe significantly improve stability. Due to the significantly larger atomic radius of Y compared to Mo, its substitution energy shows high positive values in both BCC- and FCC-Mo systems, causing severe lattice mismatch. In double-site substitution, combinations of Ti, Re, W, Nb, Ta, and Zr within the BCC-Mo system enhance its stability. For FCC-Mo, the absolute substitution energy values are notably smaller when alloy element pairs occupy second nearest neighbor (2NN) sites than first nearest neighbor (1NN) or third nearest neighbor (3NN) sites. The analysis of correlation effects between alloying elements in the BCC-Mo system reveals that these effects exhibit a pronounced distance dependency, with distinct alloying element pairs demonstrating diverse evolutionary patterns in their correlation behavior. Specifically, most alloying element pairs show strong correlation effects at 1NN sites. And these effects progressively weaken or even undergo qualitative transformations as the inter-atomic distance increases. Results from the new materials index indicate that in double-site substitution systems, when the double-site substitution elements occupy the 2NN sites, it is more favorable for enhancing the plasticity of the alloy compared to their placement in the 1NN or 3NN sites. A comprehensive evaluation of stability and mechanical properties suggests that minor compositional adjustments can significantly influence the strength-plasticity balance, with double-site substitution offering broader tunability than single-site substitution. Finally, a "thermodynamic stability-strength-plasticity regulation" strategy for multi-component alloying is established, providing a theoretical framework for designing and optimizing Mo-based alloys.

Key Words: First-Principle, Substitution Energy, New Materials index, Correlation Effects, Mechanical Property Evaluation.

Poster Number 41, Presenter: Linqin Qin, Shanghai University

Learning Radical Excited States from Sparse Data

Emissive organic radicals are currently of great interest for their potential use in the next generation of highly efficient organic light emitting diode (OLED) devices and as molecular qubits. However, simulating their optoelectronic properties is challenging, largely due to spin-contamination and the multiconfigurational character of their excited states. Here we present a data-driven approach where, for the first time, the excited electronic states of organic radicals are learned directly from experimental excited state data, using a much smaller amount of data than typically required by Machine Learning. We adopt ExROPPP, a fast and spin-pure semiempirical method for the calculation of the excited states of radicals, as a surrogate physical model for which we learn the optimal set of parameters. To achieve this we compile the largest known database of organic radical geometries and their UV-vis data, which we use to train our model. Our trained model gives root mean square and mean absolute errors for excited state energies of 0.24 and 0.16 eV respectively, improving hugely over ExROPPP with literature parameters. Four new organic radicals are synthesised and we test the model on their spectra, finding even lower errors and similar correlation as for the training set. This paves the way for the high throughput discovery of next generation radical-based optoelectronics.



Poster Number 42, Presenter: Jingkun Shen, University College London

First Principles Study of the Structure and Dynamics of Silicate Glass-Formers

Tiannan Shen

Silicates are widely present in nature and industry and understanding the structure-property relations in these materials has attracted much attention in the past decades. In this work, we are motivated to understand the interplay between atomic-level structure, dynamics and electronic properties in prototypical silicate glass-formers. To this end, we use first-principles method within the framework of density functional theory to investigate alkali silicates in both the molten and glassy states. We find that the alkali ions mainly act as network modifiers that interrupt the SiO network. The structure becomes more ordered with changing the alkali species from Li to Na to K (increasing size). The coordination number of the alkali ions increases with increasing the size of the alkali species, whereas the SiO coordination is basically independent on the type of alkali species. Furthermore, it is found that the dynamics of the alkali ions is significantly faster than oxygen and silicon but decrease with increasing alkali size. Moreover, our results show that the structural differences as inherented from the liquid state have profound effect on the vibrational and electronic properties of the glasses. Our findings provide fresh microscopic insights into the structure-property relations in silicate glass-formers.

References:

1) Liu, Y., Madanchi, A., Anker, A.S. et al. The amorphous state as a frontier in computational materials design. Nat Rev Mater 10, 228–241 (2025)

2) L. Pedesseau, S. Ispas, and W. Kob. First-principles study of a sodium borosilicate glass-former. Phys. Rev. B 91, 134202 (2015)

Optimizing Batch Parameters and Angular Momentum Cutoffs for Machine Learning Prediction of Self-Energies in Molecular Systems: Balancing Efficiency and Accurac

Author Information : Wudong Shen, IOPCAS

Supervisor : Xinguo Ren

This study investigates the trade-offs between batch parameters and angular momentum truncation (l_max) in predicting molecular self-energies using equivariant neural networks. For a dataset of 200 H_2O molecular configurations, we demonstrate that training with batch_size=10 and l_max=2 achieves optimal computational efficiency but introduces a ~15% increase in relative error. In contrast, the highest accuracy (H-H block self-energy real-part error <0.05 eV) is attained with batch_size=1 and l_max=4, albeit at the cost of significantly prolonged training time due to the intrinsic computational complexity of equivariant architectures. Validation via quasiparticle energy level reconstruction and spectral function $A(\omega)$ calculations reveals that the model captures key experimental features (peak position errors <0.2 eV), yet systematic deviations persist in peak intensities within strongly correlated regimes. Visualized error analysis further highlights the advantage of small-batch training in resolving sharp spectral features. Our work establishes a scalable optimization framework for machine learning applications in quantum chemistry, providing quantitative guidelines for parameter selection that balances computational resources and prediction fidelity. These findings advance the practical implementation of neural networks in ab initio electronic structure simulations.

Oxygen adsorption effects on geometries and electronics of thermoelectric PbTe surfaces

Surface oxidation on PbTe during the synthesis and operation inevitably degrades its thermoelectric performance. We theoretically investigate oxygen adsorption on six stable PbTe surfaces. The oxygen adsorption on PbTe surfaces leads to a significantly strong adsorption energy (-3.06 eV/O) at the (111) surface, and ~30% expansion of the first interlayer spacing at the stepped surfaces. These facilitate the oxygen penetration and initiates the surface oxidation. From electronic structure analysis, oxygen adsorptions induce PbTe thermoelectric performance degradation due to density of states reduction at the Fermi level, defect levels within the bandgap, and carrier-type inversion. Thus, we propose the PbTe anti-oxidation strategies including eliminating surface defects, passivating active oxygen adsorption sites, and applying anti-oxidation coatings.

Poster Number 45, Presenter: Yaoling Shen, University of Science and Technology China

Deep learning potential model for High-*k* $Hf_xZr_{1-x}O_2$ solid solutions

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ABSTRACT

$Hf_xZr_{1-x}O_2$ (HZO) solid solutions exhibit exceptional high dielectric constant (high-*k*) properties near the morphotropic phase boundary (MPB), yet the microscopic origin of their high dielectric response remains unclear. In this work, we develop a neural network-based deep potential (DP) model for the HZO system using a training dataset derived from first-principles calculations. The DP model accurately captures atomic interactions in the HZO solid solution. Based on DP-assisted large-scale atomic simulations, we elucidate the critical role of composition in phase stability and successfully predict the high-*k* behavior of HZO. Our findings provide a promising pathway for designing next-generation high-density, low-power memory devices.

Keywords: $Hf_xZr_{1-x}O_2$, Dielectric Property, Deep Learning Potential

Theoretical Prediction of Solid Electrolyte Interphase of XPS: From First Principles to Machine Learning

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The solid electrolyte interphase (SEI) is critical for battery performance; therefore, understanding its composition is essential for electrolyte design and high-performance battery development. X-ray photoelectron spectroscopy (XPS) serves as a key experimental tool for SEI characterization, while atomic-level simulations provide theoretical insights into SEI formation. Accurate XPS simulation bridges theory and experiment, playing a crucial role in SEI analysis.Here, We developed two complementary approaches for XPS prediction: first-principles calculations and data-driven machine learning (ML) modeling. The first-principles calculations are based on density functional theory (DFT), implemented in VASP at the PBE-D3 level[1]. Three methodological approximations were evaluated: the Initial State (IS) approximation, Final State (FS) approximation, and Janak-Slater Transition State (JS) approximation[2]. Considering the balance between computational efficiency and accuracy, we selected the IS approximation for binding energy calculations to match experimental XPS data.To address the high computational cost of DFT, we developed a ML-based alternative. The selection of descriptors is critical for ML performance; here, we employed the Local Many-Body Tensor Representation (LMBTR), which encodes atomic structures into tensors through Gaussian-smeared two-body (g2) and three-body (g3) terms, weighted by interatomic distances (see reference[3] for mathematical details).The ML model was trained on a DFT-derived dataset, achieving a mean absolute error (MAE) of 0.04 eV and an R^2 score of 99.87% on the test set. Predicted XPS spectra show excellent agreement with both experimental and DFT-calculated results[4]. We further validated our approach on two additional battery systems (1M LiFSI in FDMB and 1M LiFSI in FDMH electrolytes), with predictions closely matching experimental observations[5]. In summary, leveraging DFT-generated training data, we developed a high-accuracy ML model for rapid XPS prediction. This approach integrates computational modeling with experimental characterization, providing a robust framework for battery material analysis and simulation validation.

References

[1] Lukas Köhler and Georg Kresse. Density functional study of co on rh(111). *Phys. Rev. B*, 70:165405, 2004.

[2] Noèlia Pueyo Bellafont, Francesc Viñes, Wolfgang Hieringer, and Francesc Illas. Predicting core level binding energies shifts: Suitability of the projector augmented wave approach as implemented in VASP. *J. Comput. Chem.*, 38(8):518–522, 2017.

[3] Haoyan Huo and Matthias Rupp. Unified representation of molecules and crystals for machine learning. *Mach. Learn.: Sci. Technol.*, 3(4):045017, 2022.

[4] Qintao Sun, Yan Xiang, Yue Liu, Liang Xu, Tianle Leng, Yifan Ye, Alessandro Fortunelli, William A Goddard, and Tao Cheng. Machine learning predicts the x-ray photoelectron spectroscopy of the solid electrolyte interface of lithium metal battery. 13(34):8047–8054, 2022.

[5] Peiping Yu, Qintao Sun, Yue Liu, Bingyun Ma, Hao Yang, Miao Xie, and Tao Cheng. Multiscale simulation of solid electrolyte interface formation in fluorinated diluted electrolytes with lithium anodes. 14(6):7972–7979, 2022.

Efficient Computational Approaches to Random Phase Approximation (RPA) for Molecular and Materials Systems

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13th May 2025

Abstract

The Random Phase Approximation (RPA) is a widely-used many-body theory that provides insights into electron correlation effects in molecular and material systems. However, the computational cost associated with RPA can be prohibitive for large-scale systems. In this work, we introduce novel techniques to enhance the efficiency and applicability of RPA across various systems. Our contributions include:

- The development of a **Localized Resolution of Identity (LRI)** approach to compute analytical gradients of the RPA ground-state energy, significantly reducing computational costs while maintaining accuracy.
- A novel algorithm for **structural relaxation** based on RPA, specifically tailored for water clusters, which accounts for electron correlation effects neglected by traditional methods.

Through benchmarking on a variety of systems, we demonstrate the effectiveness of these approaches in advancing both fundamental research and applied studies. By reducing computational costs and improving accuracy, these methods enable the study of large systems and complex molecular interactions, such as hydrogen bonding in water clusters and collective excitations in extended material systems. We believe that these advancements will be of significant interest to the computational chemistry and materials science communities.

Keywords

Random Phase Approximation (RPA), Resolution of Identity, Analytical Gradients, Water Clusters, Structural Relaxation, Computational Chemistry, Electron Correlation, Many-Body Theory

Abstract

Random Green's function Method for Large-Scale Electronic Structure Calculation

An accurate and linear-scaling electronic structure method is essential for large-scale applications. Here, we present a new linear-scaling approach for large-scale electronic structure calculations—the random Green's function (rGF) method—which combines the random state technique with Green's function formalism. In this method, the rGF is defined on a set of random states and is efficiently calculated by projecting onto Krylov subspace. In equilibrium calculations, the rGF method enables direct evaluation of the Fermi–Dirac operator, thereby avoiding the need for polynomial expansions of the Fermi–Dirac function. To demonstrate its applicability, we implement the rGF method within the density-functional tight-binding framework. It is shown that the Krylov subspace can be maintained at a small size for materials with different band gaps at zero temperature, including H_2O and Si clusters. Moreover, by applying a simple deflation technique, the rGF method achieves an accuracy of ~1 meV per H_2O molecule in total energy compared to deterministic calculations. Due to the wide applicability of GF, the rGF method can be readily combined with other electronic structure methods to accelerate large-scale materials property calculations. We also extend rGF to nonequilibrium Green's function (NEGF) formalism and obtain excellent results. The rGF method provides an efficient and robust stochastic framework for large-scale electronic structure simulations.

Poster Number 49, Presenter: Mingfa Tang, Xiaogan Sichuang Information Technology Co., LTD

Poster Abstract

Title: Twist-Angle Engineering of Moiré Excitons in WSe_2/WS_2 Heterostructures: GW-BSE Insights into Linear/Nonlinear Optical Phenomena

Introduction

Moiré potentials in twisted van der Waals heterostructures create tunable excitonic landscapes, enabling unprecedented control over correlated quantum states. While exciton physics in monolayer transition metal dichalcogenides (TMDs) is established, the role of twist-angle-dependent many-body interactions in moiré excitons remains unresolved.

Methodology

We combine ab initio **GW approximation** and **Bethe-Salpeter equation (BSE)** calculations within a moiré continuum model to decode excitonic properties in twisted WSe_2/WS_2.

Key Findings

Valley Polarization Control
Nonlinear Optical Response
Emergent Quantum Phenomena

Can ZnO/Cu catalyst provide promising activity for glycerol direct dehydrogenation? A combined density functional theory and coverage-dependent microkinetics study

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Keywords: glycerol dehydrogenation, coverage effect, microkinetic modeling, CSTR simulation, DFT, Cu-based catalyst

Glycerol, a byproduct of biodiesel production, can be utilized to synthesize high-value-added chemicals such as 1,3-dihydroxyacetone (DHA). DHA has garnered significant attention due to its high value and is currently primarily produced through the aerobic oxidative dehydrogenation of glycerol. However, the use of oxidants can lead to over-oxidation, reducing selectivity and increasing process complexity. Additionally, the high cost of noble metal catalysts limits their industrial application. In contrast, the non-oxidative dehydrogenation of glycerol is safer and more economical, as it does not involve oxygen or water, and the byproduct hydrogen has added value. Since the first report by Church and Joshi in 1951 on the use of Cu-based catalysts for the non-oxidative dehydrogenation of alcohols[1], Cu-Zr, Cu-Cr, and Cu-Zn catalysts have been successively applied in the dehydrogenation reactions of ethanol and cyclohexanol. Research indicates that Cu-based catalysts have potential in the non-oxidative dehydrogenation of glycerol to produce DHA, but related studies are still very limited, primarily due to a lack of theoretical guidance on reaction mechanisms and catalyst structure-activity relationships.

In this work, the glycerol NODH reaction on Cu (111) and ZnO/Cu(111) surface is systematically studied utilizing DFT calculations and microkinetic modeling (MKM). Firstly, based on the coverage-independent results, H, glycerol and two oxygen-containing intermediates (SO*, PO*) are chosen as the environmental species to build the coverage-dependent microkinetics model, where both self and cross adsorbate-adsorbate interactions and their effect on free energy of intermediate and transition state are thoroughly studied. The coverage-dependent microkinetic analysis and electronic structure analysis are employed to demonstrate the origin of enhanced catalytic performance of glycerol NODH on ZnO/Cu (111) surface, compared with Cu (111). The superiority of the ZnO/Cu (111) surface in glycerol conversion and DHA selectivity is further confirmed by the continuous stirred tank reactor (CSTR) model

Reference

[1] Church, J. M.; Joshi, H. K. Industrial & Engineering Chemistry 1951, 43, 1804-1811.

TransOpt 3.0: a package for electrical transport with multiple scattering mechanisms and versatile program interfaces

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Understanding and computing the electrical transport properties is crucial for fundamental and practical physics, governing key characteristics like thermoelectric efficiency and semiconductor performance. In this work, we developed an open-source code, TransOpt 3.0, based on the Boltzmann transport theory under the relaxation time approach for charge carriers in a full-band electronic structure. In the current 3.0 version, multiple carrier scattering mechanisms are implemented, including intrinsic scattering with acoustic phonons as well as polar optical phonons, and extrinsic interactions with ionized impurities. The code can interface with multiple DFT packages, such as Vienna *ab initio* Simulation Package (VASP), Quantum Espresso (QE) combined with Electron-phonon Wannier (EPW), and Atomic-orbital Based Ab-initio Computation at USTC (ABACUS). Notably, TransOpt 3.0 is an easily deployable Fortran code compilable with OneAPI or NVIDIA hpc_sdk toolchains. It supports both multi-core CPU and GPU architectures, with GPU version particularly improving efficiency for the calculations of relaxation times on the full k mesh. The large language model DeepSeek is adopted in the process of the development of TransOpt 3.0 and corresponding post-processing workflows. A series of case studies, including widely studied perovskites and typical thermoelectric materials, are presented to comprehensively demonstrate the major features and calculation efficiency of TransOpt 3.0.

Combining the hybrid functional method with dynamical mean field theory in the numerical atomic orbital basis set framework

Author Information : Zirui Wu, IOPCAS
Supervisor : Xinguo Ren

Combining the hybrid functional (HYF) method with dynamical mean-field theory (DMFT) within a numerical atomic orbital (NAO) basis provides a powerful first-principles scheme for correlated materials. We build on a recently developed NAO-based DFT+DMFT framework and introduce HYF+DMFT within this basis. In our scheme, the HYF (mixing a fraction α of exact Hartree–Fock exchange) offers an improved static mean-field description of the weakly correlated bands, while DMFT captures the dynamical local correlations of strongly localized electrons. Importantly, we show that the HYF's fraction α can be related to the DMFT Coulomb U parameter, linking the two parts of the method. We benchmark HYF+DMFT on several transition-metal oxides (e.g. NiO, CoO, FeO, MnO) and find that it yields band gaps much closer to experiment than conventional DFT or DFT+DMFT methods. In particular, previous studies have shown that hybrid functionals reproduce TMO gaps well, and our results demonstrate that the combined HYF+DMFT approach further improves gap predictions by treating weak and strong correlations on equal footing. This new methodology thus extends NAO-based DFT+DMFT to include hybrid exchange, enabling accurate modeling of materials with both band-insulating and Mott-insulating characters.

Spin-informed universal graph neural networks for simulating magnetic ordering

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The screening and discovery of magnetic materials are hindered by the computational cost of first-principles density-functional theory (DFT) calculations required to find the ground state magnetic ordering. Although universal machine-learning interatomic potentials (uMLIPs), also known as atomistic foundation models, offer high-fidelity models of many atomistic systems with significant speedup, they currently lack the inputs required for predicting magnetic ordering. In this work, we present a data-efficient, spin-informed graph neural network framework that incorporates spin degrees of freedom as inputs and preserves physical symmetries, extending the functionality of uMLIPs to simulate magnetic orderings. This framework speeds up DFT calculations through better initial guesses for magnetic moments, determines the ground-state ordering of bulk materials and even generalizes to magnetic ordering in surfaces. Furthermore, we implement a closed-loop anomaly detection approach that effectively addresses the classic "chicken-and-egg" problem of creating a high-quality dataset while developing a uMLIP, unearthing anomalies in large benchmark datasets and boosting model accuracy. Finally, the anomalies identified in the Materials Project dataset account for about 1% of the total data, and adsorbate-induced nonlocal spin effects are discovered in catalysis.

[1] Xu, W., Sanspeur, Y.*, Kolluru, A., Deng, B., Harrington, P., Farrell, S., Reuter, K., Kitchin, J. R.*, *Proc. Natl. Acad. Sci.* **2025**, 122, e2422973122.

[2] Deng, B., Zhong, P.*, Jun, K., Riebesell, J., Han, K., Bartel, C.J. and Ceder, G.*, *Nat.Mach. Intell.,* **2023**, 5, 1031.

HH130: a Database of Machine Learning Interatomic Potentials for Half-Heusler Thermoelectrics

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High-throughput screening of thermoelectric materials from databases requires efficient and accurate computational methods. Machine-learning interatomic potentials (MLIPs) provide a promising avenue, facilitating the development of database-driven thermal transport applications through high-throughput simulations. However, the present challenge is the lack of standardized databases and openly available models for precise large-scale simulations. In this work, we introduce HH130, a standardized database for 130 half-Heusler (HH) compounds in MatHub-3d (http://www.mathub3d.net), containing both MLIP models and datasets for the thermal transport of HH thermoelectrics. HH130 contains 31,891 total configurations (~ 245 configurations per HH) and 390 MLIP models (three models per HH), generated using the dual adaptive sampling method to cover a wide range of thermodynamic conditions, and has been made publicly available on MatHub-3d (Download: http://www.mathub3d.net/static/database/HH130.zip). Comprehensive validation against first-principles calculations demonstrates that the MLIP models accurately predict energies, forces, and interatomic force constants (IFCs). Based on the MLIP models in HH130, we efficiently performed four-phonon interactions for 80 HHs with phonon frequencies closely matching ab initio results. It is found that HHs with 8 valence electron count (VEC) per unit cell generally exhibit lower lattice thermal conductivities (κ_Ls) compared to those with 18 VEC, due to a combination of low 2nd IFCs and large scattering phase spaces in the former group. Additionally, we identified several HHs that demonstrate significant reductions in κ_L due to four-phonon interactions. HH130 provides a robust platform for high-throughput computation of κ_L and aids in the discovery of next-generation thermoelectrics through machine learning.

Keywords: Machine-learning Interatomic Potentials, HH130, Thermal Transport

The Impact of Zirconium and Silicon Doping on the Structure and Dielectric Properties of HfO_2

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Tetravalent dopant elements (zirconium and silicon) of hafnium dioxide (HfO_2) have significant effect on the relative thermodynamic stability and dielectric properties of different crystal phases (monoclinic, cubic, tetragonal, and orthorhombic). Our first-principles investigations indicate that an increase in the doping concentration of the Zr atoms leads to a linear trend in the energy and dielectric constant of the different phases of HfO_2. On the contrary, we also reveal that Si dopant in HfO_2 induces local SiO_2 quartz-like tetrahedral ordering when Si-Si distances reach 5 Å, which should in turn stabilize the tetragonal phase $Hf_xSi_{1-x}O_2$ with a dielectric constant of 32. This phenomenon can be explained by structural changes and charge transfer mechanisms, demonstrating dopant spatial distribution as a critical design parameter for high-k dielectrics. This study provides a theoretical basis and guidance for the development of a new generation high-performance dielectric materials.

First-principles and machine learning study on the coupled effects of defects and alloying elements on the mechanical properties of molybdenum alloys

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Mo-based alloys are prime candidates for high-temperature structural components due to their high melting point, low density, good high-temperature strength and creep resistance, and high thermal/electrical conductivity. However, similar to many high-temperature intermetallics, Mo alloys and common constituent phases exhibit severe brittleness at room temperature, severely hindering their application and development. Alloying with multiple elements is a common strategy to enhance their overall properties. However, research focused on understanding and improving their brittle fracture behavior remains limited. Experimental trial-and-error approaches are costly and slow, and the complex interplay of multiple alloying elements, point defects, and line defects on their stability and mechanical properties is still poorly understood.

Therefore, the project employs first-principles calculations based on Density Functional Theory, combined with machine learning, to investigate the correlated effects of various alloying elements and defects on the mechanical properties of Mo alloys. So far, we have calculated the vacancy formation energies and solute-vacancy interaction energies in molybdenum alloys containing various solute atoms using first-principles methods. This work aims to investigate the influence of vacancies on solute segregation in alloys to improve their mechanical properties. The detailed analysis of these results is currently in progress.

Abstract

The project involves theoretical calculations of a molecule called TET, to determine its magnetic properties, such as spin density, magnetic ground state, and other properties related to magnetism. Finally, through the molecular orbital simulation of this molecule and in conjunction with the experiments, the final results were obtained.

The research plan is to first model this molecule in the MS, then conduct some theoretical calculations using AIMS, and finally compare the experimental results.

AI and ML for Selecting Pd-based single-atom Electrocatalysts

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Rational design of efficient electrocatalysts is crucial for clean energy conversion technologies. This study develops a computational approach integrating artificial intelligence and machine learning to screen high-performance palladium-based single-atom electrocatalysts. Our current work involves constructing a multidimensional database containing electronic structure descriptors and active site characteristics to train a graph neural network-based predictive model. Subsequent validation of its accuracy through first-principles calculations is required to identify a research system that exhibits exceptional catalytic activity for the hydrogen evolution reaction.

Metal-N_4 single-atom catalysts (SACs) are regarded as ideal platforms for electrocatalytic ammonia synthesis thanks to their maximal atomic utilization. However, their activity and selectivity are highly sensitive to the local structure of the surrounding carbon matrix. Here we adopt a synergistic strategy that integrates defect engineering with heteroatom doping to systematically tune the electronic environment of the M-N_4 active center, with a particular focus on the spatial distance between the defect/dopant sites and the metal atom. We first build a model library-containing diverse vacancies, surface functional groups, and B, P, and S dopants via high-throughput density-functional-theory calculations. A graph neural network is then trained to rapidly predict key adsorption energies, after which the most promising candidates are examined by constant-potential constrained ab-initio molecular dynamics and detailed charge-orbital analyses to elucidate how distance effects modulate electronic coupling and reaction pathways. The resulting insights provide theoretical guidance for the rational design of efficient and sustainable single-atom catalysts for green ammonia synthesis.

Poster Number 60, Presenter: Yiyan Zhu, Changchun Institute of Applied Chemistry

Quantum Anomalous Hall Effect with Tunable Chern Numbers in High-Temperature 1T-PrN_2 Monolayer

Quantum anomalous Hall (QAH) states offer tremendous potential for realizing the next-generation electronic devices. However, most of the available candidates have fixed Chern number and low observable temperature. It is intriguing to explore the novel materials with tunable Chern number and high working temperature.

In this study, we theoretically predict two-dimensional ferromagnetic monolayer PrN_2 with abundant magnetically correlated topological states and high magnetic transition temperature above 500 K by DFT calculations. When spin-orbital coupling (SOC) is ignored, PrN_2 monolayer is a 2D Dirac Half semimetal. When SOC is considered and the magnetization lies in the xy plane, PrN_2 monolayer will transform into 2D Weyl half-semimetal in the case that the magnetization maintains twofold rotational symmetry. If magnetization breaks all rotational symmetries, the Weyl-like points are gapped and QAH states with lower Chern number($C = \pm 1$) are found. Significantly, with the rotation of magnetization direction, alterations in symmetry give rise to periodic variations in both band gap and topological states ($C = \pm 1$). More interestingly, when the magnetization are turning to out-of-plane, the PrN_2 monolayer realizes an QAH phase with high Chern number of ± 3.

The present work proposed an intrinsic QAH insulator with tunable Chern number and high working temperature, which provides a feasible platform for investigating exotic topological physics.

Study on Segregation and Mechanical Properties of Al-Mg Polycrystalline Alloy Based on Machine Learning Potential

Calculating the mechanical properties of Al-Mg alloys is of great significance in the fields of materials science and engineering. Studying the impact of solute segregation on grain boundaries helps to gain a deeper understanding of fundamental principles in materials science, such as the strengthening mechanisms, grain boundary stability, and overall performance of alloys. However, using existing interatomic potentials is either difficult to accurately describe or extremely costly, especially for large and complex systems. Our goal is to combine density functional theory with machine learning to:

1. Develop accurate machine-learned interatomic potentials that retain quantum-level fidelity while enabling large-scale simulations.
2. Reveal the impact of solute segregation on the mechanical properties of Al-Mg alloys.
3. Use the trained potentials to study the segregation behavior and its influence on the mechanical performance of polycrystalline Al-Mg alloys.

Through these workflows, we expect to develop deep learning interatomic potential models that are both highly accurate and computationally efficient. The results will provide theoretical insights into the design of Al-Mg alloys with enhanced mechanical properties.