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# Ultrafast Dynamics of Photogenerated Electrons at $NO_3$/CdSe QD Interface Driven by Capture of Hot Electrons-*Ab Initio* Quantum Dynamics Simulation

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## Abstract

Harnessing hot carriers under visible light presents a promising strategy for selective and efficient nitrate ($NO_3^-$) reduction to lower-valence nitrogen species. Using ab initio quantum dynamics simulations, we unveil the ultrafast charge transfer processes at the CdSe quantum dot (QD)/$NO_3$ interface, revealing efficient hot electron (HE) extraction. Taking metal-rich CdSe QD as the surface, our time-dependent ab initio nonadiabatic molecular dynamics simulations depict rapid interfacial charge transfer, with 70% of the electron population captured by the lowest unoccupied molecular state (LUMO) of NO3 within a few hundred femtoseconds. The impulsive two-state model elucidates the role of molecular excitations in facilitating bond cleavage, highlighting the activation of two key vibrational modes: antisymmetric stretching and bending, facilitating the dissociation of $NO_3$ into $NO_2$. Similarly, $NO_2$ undergoes further reduction, leading to NO formation within 25–35 fs, followed by desorption from the QD surface. These findings provide an atomistic perspective on hot-carrier-driven $NO_3$ reduction and establish fundamental design principles for engineering QD-based photocatalysts with enhanced selectivity and efficiency.

D1-002

We explore the pressure-dependent structural, optoelectronic, and dynamic properties of the two-dimensional layered halide perovskite (2D LHP) (4F-PhDMA)PbBr$_4$, comprising inorganic layers of corner-sharing PbBr$_4$ octahedra interleaved with fluorinated organic spacer cations (4F-PhDMA) in the Dion-Jacobson (DJ) phase. Application of pressure induces pronounced distortions in the PbBr$_4$ octahedra, which modify the non-covalent van der Waals interactions between the organic and inorganic layers. These pressure-induced structural alterations significantly influence key optoelectronic parameters, such as the bandgap, effective masses of charge carriers, and electron-phonon coupling strength. The fluorinated spacer cations contribute to enhanced material performance by increasing the polarity of the organic layer, modulating dielectric confinement, and influencing exciton binding energy. Our findings underscore the critical role of pressure in optimizing the photophysical properties of 2D LHPs. Through a combination of static and dynamic structural and electronic analyses, we demonstrate that pressure-driven tunability provides a robust approach to improve the performance of 2D LHPs in photovoltaic and optoelectronic applications. This study establishes a framework for the design of next-generation materials with tailored optoelectronic properties via controlled pressure application and strategic chemical functionalization.

# Interfacial stability of Electrolytes at pure and oxidized Mg surfaces- Insights from Ab Initio Molecular Dynamics

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## Abstract

The majority of the current electric vehicles use lithium-ion batteries (LIBs) for energy storage. However, for long-range vehicles that require high energy densities (> 350 Wh/kg), current LIBs are not suitable. Among post-Li ion batteries, Magnesium (Mg) metal ion batteries present a promising alternative, offering high volumetric capacity, dendrite-free cycling and natural abundance. Yet, the practical realization of Mg-based batteries is fundamentally hindered by the formation of ion-blocking surface films resulting from electrolyte decomposition. A key contributor to this interfacial instability is the native magnesium oxide (MgO) layer that forms spontaneously upon air or electrolyte exposure. Here, we employ ab initio molecular dynamics (AIMD) simulations to investigate the interfacial stability of different salts and solvents on three Mg-based surfaces: pristine Mg, stoichiometric MgO, and undercoordinated oxidized MgO. While electrolyte components remain largely intact on metallic Mg and stoichiometric MgO, the oxidized MgO surface exhibits pronounced reactivity, triggering decomposition of both salts and solvents at ambient conditions. This instability arises from low-coordination surface sites and reactive oxygen species that promote electron transfer and bond cleavage. Our results elucidate the atomic-scale mechanisms of early-stage SEI formation and highlight the pivotal role of surface structure in governing interfacial stability, offering valuable guidance for the design of robust electrolytes for Mg-based batteries.

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# Effect of anionic and cationic doping on Fe-based superconducting series: interplay of magnetism and superconductivity

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The Fe-chalcogenide series has retained the relevant focus because of their multitudes of strongly correlated attributes like superconductivity or charge density wave coexisting with the underlying magnetic long-range order. Interplay of these correlated phases with multiple structural transitions leads to a complex phase diagram in this series of materials. In the present work, with the help of low temperature magnetization measurements and first-principles theory, we have investigated the role of both cationic and anionic substitution on the correlated behaviour of the series FeSe, FeTe, FeSeTe and CuFeTe [1]. First principles density functional calculations imply that all four systems are stabilized in tetragonal symmetric structure with A-type antiferromagnetic spin arrangements. Further, the dynamical electronic correlations in these systems are studied using the DFT+DMFT approach [2]. Within DFT+DMFT, the intersite exchange parameters of this series are extracted and used as an input for a spin-dynamical calculation. The solution of this spin dynamical equations results into a complex chiral ground state for all these systems. Low temperature magnetic measurements indicate the superconducting properties of FeSe are less suppressed with anionic Te substitution than the cationic Cu substitution. We have analyzed these results in the light of the substitution-induced modification of electronic properties.

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# First Principles Analysis of In-Plane Heterostructure of 2D Materials

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Transition metal dichalcogenides (TMDCs) ($MX_2$, M = transition metal, X = chalcogen) are layered materials with rich electronic and topological properties. Monolayers exhibit unique behaviors distinct from the bulk, with tunable properties. This versatility in their properties depends on monolayer structure and chemistry. These properties give them the potential for different application. Unlike vertical heterostructures, where different layers are stacked on top of each other, in-plane heterostructures are formed by seamlessly joining different materials or phases in the plane of the monolayer. Here we present results for $WTe_2$-based in-plane heterostructure.This work highlights the significance of phase engineering of $WTe_2$ and the potential of in-plane heterostructures as a versatile platform for future quantum and electronic applications.

# AAPALI PSI-K 2025

# Computational and Experimental Investigation of Morphology and Metal Influence of ZIFs in $CO_2$ Cycloaddition Reaction

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Zeolitic Imidazolate Framework (ZIF)s show noteworthy catalytic activity owing to the proximal presence of Lewis acidic metals and functional groups on the linkers.[1] They are potential catalysts for the cycloaddition of $CO_2$ with epoxides to form cyclic carbonates, which is an atom-efficient route for using $CO_2$.[2] Herein, we report a systematic study of ZIF catalysts for $CO_2$ cycloaddition with propylene oxide (PO) using periodic density functional theory (DFT) simulations, validated by experiments, to unravel the influence of ZIF morphology and metal centre on catalytic activity and stability. The (002), (011), and (222) surfaces of ZIF-67 (Co + 2-methyl imidazole), representing the dominant surface exposures of ZIF-67 nanocube (NC), rhombic dodecahedron (RD), and nanosphere (NS), respectively, were considered for the DFT simulations. Surface termination with 3-coordinated Co on ZIF-67(011) surface was the most stable with the lowest specific surface energy (0.36 $J/m^2$). Reaction mechanism and energy profile analyses on Lewis acidic Co indicated higher catalytic activity on ZIF-67(002) surface which has 2-coordinated Co, with the lowest activation barrier for ring opening (RO) of 0.38 eV. ZIF-67 (011) and ZIF-67(222) surfaces (CN=3) exhibited higher RO barriers of 0.70 eV or more and was unfavourable. Experiments with ZIF-67 in these three morphologies validated the predicted activity trends as ZIF-67 NC >ZIF-67 RD> ZIF-67 NS. The large energy penalty for removal of Co and $Co(mIm)_2$ units from ZIF-67(011) surface indicated stability of the ZIF-67 RD catalyst which was validated by reusability studies. In contrast, on ZIF-8 (Zn + mIm) with the ZIF-8(011) surface (stable RD), the PO ring opening and $CO_2$ insertion were kinetically more favourable ($E_a$ = 0.50 eV and 0.14 eV) than on ZIF-67(011) ($E_a$ = 0.70 eV and 0.48 eV) suggesting that ZIF-8 RD could be more active than ZIF-67 RD.

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AAPALI PSI-K 2025

# Tuning Hot Carrier Dynamics in Vacancy-Ordered Halide Perovskites through Lattice Compression: Insight from *ab initio* Quantum Dynamics and Machine Learning

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The efficient harvesting of hot carriers (HCs) from high-energy photons can significantly enhance optoelectronic performances. However, ultrafast HC cooling through intra-band transitions poses a significant challenge for extraction using traditional semiconductor absorbers. The stable and compressible vacancy-ordered halide perovskites with isolated octahedra exhibit discrete electronic states at the conduction band edge, indicating possible slow cooling of hot electrons (HE). Using state-of-the-art ab initio quantum dynamics simulations and unsupervised machine learning (ML), we investigate the effect of lattice stress on HE dynamics in vacancy-ordered $Cs_2SnBr_6$. The moderate stress enhances structural rigidity and weakens dynamic electron-phonon interactions at the conduction bands. Such modifications and the widened energy gap at the conduction band edge partially suppress intra-band non-adiabatic transitions, eventually elongating the HE lifetime. The pairwise mutual information extracts hard-to-find highly non-linear dynamic structure-excited state property correlations, offering the unique opportunity to strategically design efficient lead-free halide perovskites for HE-based optoelectronics.

# Thermoelectric characteristics of silver-based semiconductors using the non-empirical range-separated dielectric-dependent hybrid

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We explore the intricate domain of thermoelectric phenomena within silver (Ag) based chalcopyrites, focusing on compositions such as $AgXTe_2$ (where X=Ga, In) and the complex quaternary system $Ag_2ZnSn/GeY_2$ (with Y=S, Se) [1, 2]. The thermoelectric parameters are estimated from the Boltzmann transport theory in combination of the electronic structures that, in turn, are obtained using the non-empirical screened dielectric-dependent hybrid (DDH) functional approach within the density functional theory (DFT) [3]. This approach allows us to analyze critical thermoelectric properties, including electrical conductivity, Seebeck coefficient, and power factor. We investigate the often-overlooked phenomena of phonon scattering by leveraging both the elastic constant tensor and the deformation potential method [4, 5]. This enables a rigorous examination of electron relaxation time and lattice thermal conductivity, enhancing the robustness of our predictions [4]. We identify materials with a thermoelectric Figure of merit (ZT = $\sigma S^2 T/\kappa$) exceeding the critical threshold of unity. Our findings delineate a promising trajectory, laying the groundwork for the emergence of a new class of Ag-based chalcopyrites distinguished by their exceptional thermoelectric characteristics. .

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# Probing Magnetism in Iron Embedded $MoTe_2$: A First-Principles Study

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Two-dimensional (2D) materials have drawn significant interest due to the unique physical properties and potential applications in advanced technologies. Unlike graphene, transition metal dichalcogenides (TMDs) are notable for their ability to adopt multiple crystal structures, and structural metal-to-insulator phase transitions. Embedding metal atoms into the lattice voids further enhances these properties, offering new possibilities for quantum applications like ultrathin flexible electronics and valleytronics devices. In particular, modifying $MoTe_2$ monolayers by incorporating metal atoms provides a new pathway to engineer novel quantum phenomena. Using first-principles calculations, we investigate structural defects isomers such as: Interstitial (Int), Substitutional (Sub) and adatom configurations for post growth Fe atom deposition in the 2H and $1T'$ phases of $MoTe_2$. The formation energy calculation reveals the Fe atom preferentially occupies "Int" sites with a stronger affinity and enhanced magnetism of $2\mu_B$ under Te-poor conditions, in $1T'$ phase. For the first time, we present the possibility of enhancing the properties of $MoTe_2$ by embedding impurity metals within the zigzag chains of the $1T'$ phase emphasising its superiority for magnetic applications. This work opens new avenues for impurity engineering in 2D materials, enabling tailored functionalities for advanced applications leading in electrocatalytic activity, energy storage, spintronic devices, and emerging quantum technologies.

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# Engineering two dimensional electron and hole gases at Si/Si$_{1-x}$Ge$_x$ heterostructure interfaces: Insights from DFT calculations

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Si/Si$_{1-x}$Ge$_x$ heterostructures are key materials for the development of semiconductor-based spin qubits [1], offering promising prospects for scalable, low-noise quantum computing architectures. Beyond their potential in quantum computing, these materials are also widely used in the fabrication of electronic devices, such as MOSFETs [2]. A two-dimensional electron or hole gas (2DEG or 2DHG) can be formed at the interface of modulation-doped Si/Si$_{1-x}$Ge$_x$ heterostructures [3], [4], making them highly relevant and significant for such applications. Despite extensive experimental studies, a comprehensive and accurate first-principles investigation of these systems still seems to be missing. To understand the factors responsible for formation of a two-dimensional electron and/or hole gas at Si/Si$_{1-x}$Ge$_x$ interfaces, we have performed accurate Density Functional Theory (DFT) calculations using HSE06 hybrid functional [5]. Our calculations have been aimed to design the heterostructures for making electron or hole based quantum dot qubits.

In semiconductor based heterostructures, band alignment between the two materials forming the heterojunction is an essential property to know if a 2DEG or 2DHG forms, the direction of charge transfer at the interface, and the position of 2DEG or 2DHG. For Si and SiGe, the type of band alignment and band offsets depend on the amount of Ge concentration in SiGe alloy, and the strain condition (due to lattice mismatch between Si and SiGe) in the heterostructure layers. We have studied these factors by calculating band alignment between bulk Si (strained) and bulk SiGe for several concentrations of Ge ($x$) in the alloy. Our calculations have shown that the type of alignment changes with $x$. We have found optimum concentrations of Ge to obtain a 2DEG or 2DHG based on the type of alignment and offsets. We have found that for some low values of $x$, the type of alignment does not fall into the usual classification of band alignment into types 1, 2 or 3. Our results have successfully explained the experimental reports on formation of 2DEG and 2DHG in these heterostructures for different values of $x$. I will present and discuss the results from DFT calculations, along with the physical implications of the band alignment type and dopant choice, which influence the formation of a 2DEG or a 2DHG.

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# AAPALI PSI-K 2025

# Chiral Split Magnon in Altermagnet β-$MnO_2$: A First-principles Study

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Altermagnetism represents a newly identified class of magnetism characterized by alternating spin polarizations in both real and reciprocal space [1]. Unlike conventional ferromagnets and antiferromagnets, altermagnets exhibit spin splitting in electronic bands despite preserving time-reversal symmetry. Recent theoretical predictions suggest that this alternating spin splitting extends to magnon bands, giving rise to nontrivial chiral magnons [2]. These chiral magnons are particularly attractive as they enable robust spin transport without requiring strong spin–orbit coupling, making altermagnetic materials promising candidates for next-generation spintronic applications. In this study, we investigate the emergence of chiral magnons in tetragonal β-$MnO_2$, an altermagnet, using first-principles calculations based on density functional theory. We reveal the presence of non-degenerate chiral magnons, confirming the altermagnetic nature of β-$MnO_2$. The observed magnon splitting is attributed to a symmetric-exchange interaction rather than relativistic effects, distinguishing it from conventional Dzyaloshinskii–Moriya-driven chiral magnons. Furthermore, we demonstrate that external perturbations, such as lattice strain, effectively modulate the exchange parameters, leading to tunable chiral magnon transport. Our analysis shows that these modifications can significantly influence key spin transport phenomena, including the spin Seebeck effect and transverse spin conductivity, with the potential to reverse their transport direction [3]. These findings provide new insights into the fundamental properties of altermagnets, establishing β-$MnO_2$ as a model system for exploring chiral magnons. The ability to manipulate magnon transport in altermagnets through external control parameters opens new avenues for designing energy-efficient spintronic devices. This work highlights the technological potential of altermagnetic materials for spin-based information processing and magnons applications.

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# Plant-derived Medicinal Biomolecules in an Aqueous Solution: A Quantum Mechanical Prediction for Efficacy Parameters

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**Abstract:** The present study explores the profound legacy of medicinal autotrophs, recognizing their historical significance in human healthcare and emphasizing the enduring collaboration between nature and humanity in the pursuit of therapeutic remedies. The research centers on biomolecules derived from natural autotrophs, namely Lupeol, Hederagenin, Quinovic acid, and Morolic acid are investigating their properties through a comprehensive computational analysis. Commencing with an emphasis on the historical context of medicinal plant usage, exemplified by Castanopsis indica and Anthocephalus cadamba in our country, the current study highlights the diverse medicinal applications of plant-derived biomolecules. The computational analysis utilizing density functional theory reveals a fundamental understanding of Lupeol, Hederagenin, Quinovic acid, and Morolic acid. Guided by the broader framework of Conceptual Density Functional Theory, the study explores energies, global reactivity parameters, and solvation energies, providing crucial insights into the biomolecules' potential as natural drug candidates.Molecular Electrostatic Potential (MESP) surfaces unveil potential sites for electrophilic and nucleophilic reactions, critical for drug-receptor interactions.The values of Global recativity paramerters for them provide valuable

# Exploring Multifunctional Prospects of Metal-free Group-IV Carbide Quantum Dots and Their Single Layer Heterojunctions

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The recently successful large-area bottom-up synthesis of a two-dimensional honeycomb silicon carbide (SiC) monolayer (Phys. Rev. Lett. 130, 076203 (2023)) has undeniably opened up new avenues for exploring the fascinating physics of graphene-like monolayers across various functional domains. In this study, we have meticulously engineered group-IV carbide monolayers, including SiC, GeC, and their single-layer heterojunction (J. Appl. Phys. 132, 184301 (2022)), to design six stable quantum dots (QDs) and comprehensively investigate their electronic properties, magnetic edge states, and electronic transitions. The QDs are deliberately designed such that the in-plane junction line between two carbide monolayers goes through the middle of the structures. These QDs have been further characterized through circular dichroism and their exceptional sensitivity to environmentally hazardous weakly interacting target gases using Density Functional Theory (DFT) calculations. Notably, QDs with zigzag edges exhibit spin-split electronic states alongside fluorescence signatures extending into the short-wave infrared (SWIR) region. Interestingly, polar SiC-GeC QDs showcases chiral characteristics due to their asymmetric point group symmetry ($C_1$), as revealed by vibrational and electronic circular dichroism spectroscopy. Moreover, the presence of heterojunctions between distinct carbide components significantly enhances gas sensitivity, reaching approximately upto 70% using DFT-D3 correction, a substantial improvement over their individual counterparts- paving the way for advanced environmental friendly and multifunctional applications.

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# Phonon-assisted control of magnonic and electronic band splittings

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Spin-split antiferromagnets, also known as altermagnets, have emerged as a new class of magnetic materials exhibiting properties distinct from conventional antiferromagnets and ferromagnets [1]. Altermagnets break time-reversal symmetry, similar to ferromagnets, while maintaining the characteristic anti-parallel spin arrangement of antiferromagnets. In our study, we theoretically demonstrate the tunability of non-relativistic magnonic and electronic spin splitting by manipulating phonon modes. Using $\mathrm{MnF_2}$ as a representative altermagnet with non-relativistic spin splitting in its electronic bands [3], we identify an analogous $d$-wave splitting in magnon modes of specific handedness. Our results [4] reveal a direct correlation between magnonic and electronic splittings, showing that the energy splitting in both bands can be adjusted by jointly modulating the $\mathrm{A}_{2u}$ and $\mathrm{A}_{1g}$ phonon modes, with frequencies of 8.52 and 9.74 THz, respectively. The experimental detection of magnonic and electronic band splitting remains an open challenge. Our analytical investigation identifies a set of $k$-points where polarized neutron scattering may facilitate its observation in altermagnets. This work highlights the intricate interplay between charge, spin, and lattice degrees of freedom, opening new possibilities for magnonic applications.

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# Exploring Energy Materials from First Principles Study

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The thermoelectric effect describes a material's ability to create electric power when subjected to a thermal gradient without moving components [1]. Thermoelectric generators (TEGs) offer a promising way to boost the efficiency of numerous processes, with applications in power production, household products, transportation, wearable gadgets driven by human body heat and solid-state refrigeration[2]. A thermoelectric material's efficiency is characterized by the dimensionless thermoelectric figure of merit(ZT), given by $ZT = \frac{S^2\sigma}{(k_l+k_e)}T$ where 'S' is thermopower, '$\sigma$' is electrical conductivity, 'T' is temperature and '$\kappa_l$' and '$\kappa_e$' are the lattice and electronic contributions to the thermal conductivity, respectively. A good thermoelectric material should possess a large thermopower as well as high electrical conductivity and low thermal conductivity. The power factor PF = $\sigma S^2$ is an important parameter for any thermoelectric material. Materials with high power factor along with high ZT has been sought after for thermoelectric device applications. Unfortunately, improving the power factor and ZT is particularly challenging due to the conflicting needs of material properties to increase S and $\sigma$ simultaneously. However, due to continuous effort from scientist across the world, a high figure of merit greater than one has been achieved in state of the art materials like $Bi_2Te_3$ , PbTe and SnSe operating at temperatures ranging from room temperature to mid-high.

In this work, we studied the thermal and electronic transport properties of layered oxychalcogenides LaAgXO (X = Se, Te) using density functional theory. Different scattering mechanisms such as acoustic deformation scattering (ADP), ionized impurity scattering (IMP) and polar optical scattering (POP) are included to calculate the scattering rates at various doping concentrations and temperatures. The calculated scattering rates are used in the Boltzmann transport equation to get the absolute values of thermoelectric coefficients. The Seebeck coefficient of both the compounds is nearly 400 $\mu V/K$ for optimal p-type doping. The lattice thermal conductivity of both LaAgXO is ultra-low with values around 0.20 W/mK along the 'c' axis at 300 K due to low lifetime and low group velocity. This is lower than other well-known thermoelectric materials like PbTe and SnSe. Rattling motion observed in the Ag-Te tetrahedral layer might be the reason for the significant suppression of the $\kappa_l$. We predict huge ZT values of 1.63 for p-type and 2.8 for n-type LaAgTeO at 900 K, which are higher than that of promising thermoelectric materials such as BiCuSeO (1.4) and LaCuSeO (2.71). There is a crossing in phonon band dispersion which forms a nodal line in the 001 plane that may lead to topological behaviour. This study highlights LaAgXO as potential thermoelectric materials for future device applications.

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# A novel phase of germagraphene — Quasi-direct bandgap and anisotropic carrier mobility with potential optoelectronic response

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Experimental feasibility of implanting germanium into a graphene system (Tripathi et al., ACS Nano 12 (2018) 4641–4647) motivates us to investigate the prediction of a new phase of monolayer germa-graphene structure using first-principles calculations. In this work we verified the stability and explored the mechanical response and electronic/optical properties of monolayer rectangular-germagraphene. The structure displays auxetic behavior and exhibits a brittle nature. Tiny and anisotropic carrier effective masses demonstrate rapid carrier transport properties and increase the efficiency of photogenerated electron-hole separation. Based on GGA-PBE results, the rectangular-germagraphene monolayer exhibits a semiconducting electronic nature with a quasi-direct band-gap of 0.4 eV, whereas the HSE06 estimation indicates that the monolayer exhibits a band-gap of 0.5 eV, which is highly encouraging for use in nanoelectronics. According to optical results, rectangular-germagraphene has the ability to absorb visible, infrared, and near-infrared light. All these exotic properties could be a good option for nanoelectronics and nanooptics devices.

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# FIRST PRINCIPLES STUDY OF MULTIGAP SUPERCONDUCTIVITY IN $V_3$Si USING MIGDAL – ELIASHBERG THEORY

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In the last few decades, A15 intermetallic compounds with the formula $A_3B$ (such as $V_3Si$ and $Nb_3Sn$) have been extensively studied as many of them display superconductivity at relatively high temperatures. They exhibit the best combination of critical current density, superconducting temperature and critical magnetic field strength[1]. $V_3Si$ is an important member of A15 superconductors. It has the highest known superconducting transition temperature ($T_c$) of 17K among silicides and hence is being considered as a potential candidate for quantum circuit applications.

The nature of superconducting gap in $V_3Si$ has been a subject of great interest among the research community. While experimental studies such as scanning tunnelling microscopy (STM) and photoemission spectroscopy (PES) indicate the presence of single gap in $V_3Si$ [2,3], other studies such as microwave responses point towards the two-gap superconductivity [4]. In this work, we investigate the multigap nature of superconductivity in $V_3Si$ using ab initio Migdal-Eliashberg theory.

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# A Computational Framework for Accurate Estimation of Theoretical Specific Capacity: A Case Study of α−Graphyne

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**Abstract:** Sodium-ion batteries are considered as potential alternatives to conventional lithium-ion batteries. To realize their large-scale practical applications, it is essential to identify suitable anode candidates exhibiting promising electrochemical properties such as high specific capacity, low diffusion energy barrier, and excellent cyclic stability. In this work[1], using density functional theory (DFT) calculations and ab initio molecular dynamics simulations, we examine α-graphyne – a carbon-based 2D material – as a potential anode candidate. Our results show that AGY exhibits an ultra-low diffusion barrier of 0.23 eV along both the horizontal and vertical directions and a low average anodic voltage of 0.36 V. Our AIMD studies at 300 K show excellent thermodynamical stability with the loading of four sodium atoms, resulting in a theoretical specific capacity (TSC) of 279 mA h $g^{-1}$. Doping studies show that varying the nature of acetylenic links of AGY with electron-rich (nitrogen) or electron-deficient (boron) elements, the adsorption strength and diffusion barriers for Na atoms on AGY can be tuned. Furthermore, treating AGY as a case study, we find that conventional DFT studies are expected to overestimate the TSC by a huge margin. Specific to AGY, this overestimation can be up to ~300%. We identify that ignoring the probable formation of temperature-driven metal clusters is the main reason behind such overestimations. Furthermore, we develop a scheme to calculate TSC with higher accuracy. The scheme, which can be easily generalized to the universal class of electrodes, is evolved by concurrently employing AIMD simulations, DFT calculations, and cluster analysis.

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## AAPALI PSI-K 2025

# High-temperature magnetism and strong spin-lattice coupling in two-dimensional $CrTe_2$

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**Abstract:** Recent experimental studies on ultrathin films of $CrTe_2$ have shown potential for achieving room-temperature ferromagnetism. However, the results in single-layer $CrTe_2$ are widely debated in the literature, with conflicting findings ranging from intrinsic ferromagnetism[1,2] to a ground state characterized by zigzag antiferromagnetism[3]. We correlate these contrasting results to a very strong spin-lattice coupling, which is atypical for two-dimensional van der Waals materials. We examine the underlying phase diagram to understand these phenomena better and explore exchange interactions and magnetic anisotropies. Moreover, we employ long-range anisotropic Heisenberg Monte Carlo simulations to calculate ordering temperatures and investigate methods for manipulating the magnetic properties, focusing on compatibility with potential device applications.

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# Statistical Properties of Confined System

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The properties of a confined system is different from the open system due to the interaction with their environment. The confinement changes the system's energy levels and dynamics. In this work, we study the statistical properties (i.e. average energy, entropy, and specific heat) of a confined system. We use the Finite Element Method (FEM) to solve the Schrodinger equation numerically. In FEM, we define the interpolation functions locally to approximate the unknown wave function rather than define the trial function globally. This study shows that FEM can capture the statistical behavior of confined systems very effectively.

# Altermagnetism in Weyl semimetal GdAlSi

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Spintronics has emerged at the forefront of technological applications in current times and is set to play a major role alongside conventional electronics. In the past decade, magnetic topological semimetals have generated considerable interest as they exhibit variety of quantum phenomenon coupled with emergent Dirac or Weyl Fermions which has helped gain a deeper understanding of fundamental Physics as well as enabled exciting spintronic applications. “Altermagnetism”, a fundamentally new type of collinear staggered magnetic order similar to a collinear antiferromagnet but showing non-relativistic spin-splitting, was proposed theoretically and subsequently verified by experiments recently. This new order has been shown to be promising for antiferromagnetic spintronics with high frequency switching and immunity to stray fields. In this work, we show the coexistence of non-trivial topology and altermagnetism in a single crystalline material GdAlSi. Our *ab-initio* calculations show GdAlSi to have a collinear antiferromagnetic ground state with momentum-dependent non-relativistic spin-splitting which is a consequence of crystalline symmetries. Further investigations reveal GdAlSi to be a topologically non-trivial Weyl semimetal hosting exotic and robust spin-polarized surface states known as Fermi arcs. Second-harmonic generation and magnetization measurements confirm the non-centrosymmetric crystal structure and antiferromagnetic nature of GdAlSi respectively whereas angle-resolved photoemission spectroscopy reveals the presence of Fermi arc-like features. Such coexistence of altermagnetic order and Weyl Physics in a single material is unprecedented and could pave the way for novel spintronic devices.

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# Design of ferroelectric double perovskite oxides as photovoltaic materials

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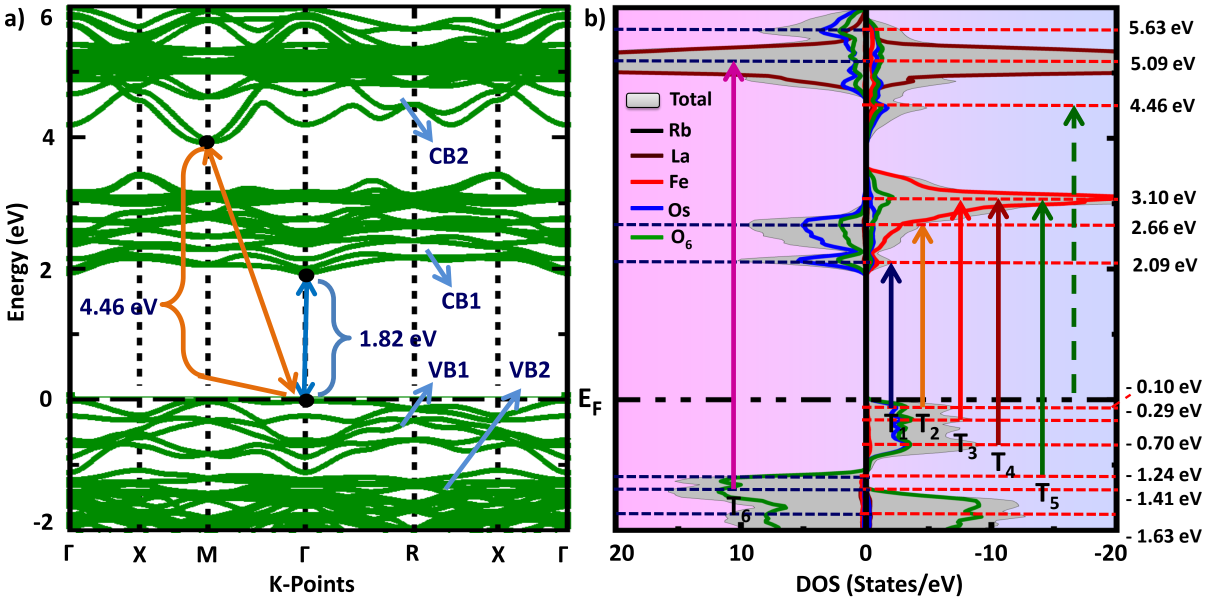
Ferroelectric photovoltaic materials are of great interest due to their coupling between spontaneous polarization, electronic, and optical properties, making them promising candidates for solar energy applications. This study explores hybrid improper ferroelectricity in $AA'BB'O_6$ (A = Na, K, Rb; A′ = La, Y) with $3d$-$5d$ transition metal MnW and FeOs at the BB′ sites. Density functional theory calculations reveal that these DPO materials stabilize in a polar $P2_1$ phase with an $a^-a^-c^+$ tilt pattern. The polarization originates from the noncancellation of layered polarization between AO and A′O layers, which can be tuned via relative displacement. In MnW-based systems, $Mn^{2+}$ ($d^5$) ions couple antiferromagnetically, while $W^{6+}$ ($d^0$) remains non-magnetic, leading to a net zero spin moment (1). In contrast, FeOs-based systems exhibit ferrimagnetic ordering via $Fe^{3+}$ (S = 5/2) and $Os^{5+}$ (S = 3/2) interactions mediated by O-2$p$ hybridization, yielding a net spin moment of 4.33 $\mu_B$ for $Fe^{3+}$ and -1.84 $\mu_B$ for $Os^{5+}$ (2). These materials are semiconducting with $E_g < 3$ eV and strong absorption in the visible and ultraviolet regions. Ab initio molecular dynamics simulations confirm robust ferroelectric polarization switching above 300K, making them suitable for room-temperature applications. These findings highlight the potential of these DPOs for advanced spintronic and optoelectronic technologies.



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# Electronic and magnetic properties of $Sr_2TiFeO_6$ implementing *ab initio* density functional theory

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Double perovskite oxides with the formula $A_2BB'O_6$, featuring 3d transition metals at the B and B′ sites, offer a versatile platform for investigating diverse electronic and magnetic properties. Electron correlation (U) and Hund's coupling ($J_H$) play a crucial role in determining those properties. $Sr_2TiFeO_6$ is a notable compound in this category, featuring a mixed valence state of Fe/Ti occupying the B/B' sites. The transport and thermoelectric properties of $Sr_2TiFeO_6$ have been investigated in some experimental investigations [1-3].

In this study, we have performed a theoretical investigation of the ground state structure, and electronic, and magnetic properties of $Sr_2TiFeO_6$ using Density Functional Theory (DFT). The Hubbard U parameter was included to accurately represent the correlation effects within the Fe d orbitals. Our electronic density of states (DOS) calculations reveal that the FM state exhibits metallic behavior, with distinct contributions from the two Fe atoms indicating mixed valence states of $Fe^{3+}$ and $Fe^{4+}$. This underscores the crucial role that Fe d-states play in determining the electronic and magnetic characteristics of $Sr_2TiFeO_6$. Furthermore, the calculation of exchange interactions suggests a predominant antiferromagnetic interaction between Fe atoms, complemented by a weaker ferromagnetic component. This complicated interplay of magnetic interactions demonstrates the complexity of magnetic behavior and the frustration of $Sr_2TiFeO_6$.

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# Comparative Investigation of PdFeGa alloy using DFT and DFT+U for Multifunctional Applications

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The study of half-Heusler alloys has gained significance in materials science due to their exceptional electronic, mechanical, magnetic, and dynamical properties. In this research, a detailed theoretical comparative investigation of the PdFeGa alloy is conducted using Density Functional Theory (DFT) and DFT+U methods. We analyzed its structural stability, electronic band structure, elastic properties, and magnetic behavior to assess its potential as a half-metallic ferromagnet. Half-Heusler alloys, as ternary compounds, are widely utilized in spintronic, thermoelectric, and optoelectronic applications. This study explores stable half-metallic ferromagnetic materials with Curie temperature exceeding room temperature, making them suitable for practical implementation. The results indicate that PdFeGa exhibits half-metallic ferromagnetism, with a significant magnetic moment localized on the Fe atom. The inclusion of on-site Coulomb interaction (U) in DFT+U calculations enhances the description of electron correlation effects, leading to more accurate predictions of its electronic and magnetic properties. The Generalized Gradient Approximation (GGA) was employed for the exchange-correlation functional, with a dense k-point mesh and a high-energy cutoff to ensure computational accuracy. The findings confirm that PdFeGa is a half-metallic material with a high Curie temperature and strong spin polarization. This study provides new theoretical insights into PdFeGa, highlighting its potential for future applications in spintronic and electronic devices.

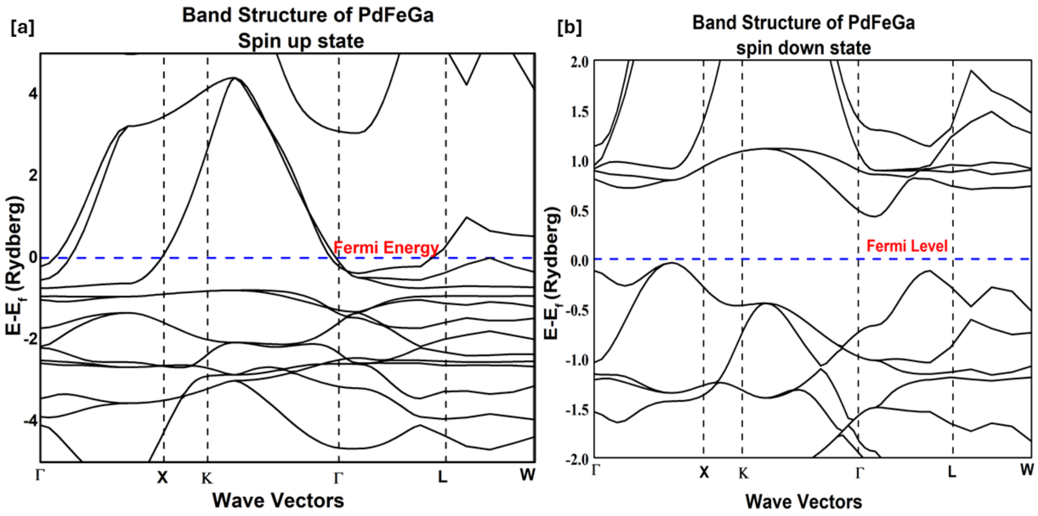

**Figure 1** Band Structure of PdFeGa alloy in (a) spin up and (b) spin down state.

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# The enhanced efficiency of two-dimensional polyaramid as a glucose sensor

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## ABSTRACT

The development of highly efficient glucose sensors is of paramount importance in medical diagnostics, particularly for diabetes management. Among various two-dimensional (2D) materials, polyaramide (2DPA) has emerged as a promising candidate due to its large surface area, exceptional chemical stability, and tunable functional properties. In this study, we investigate the adsorption behavior of glucose molecules on 2DPA surfaces using density functional theory (DFT) simulations. The adsorption energy ($E_{ad}$) was calculated to be -0.25 eV, indicating a favorable interaction between glucose and the 2DPA surface. To gain deeper insights into the adsorption mechanism, a comprehensive analysis of electronic properties was performed, including the density of states (DOS), partial density of states (PDOS), electronic band structures, and charge density distributions. These analyses revealed a strong binding affinity and potential electronic interactions conducive to effective glucose detection. The proposed sensing material demonstrated a rapid recovery time of 23.15 ns, underscoring its suitability for real-world healthcare diagnostic applications. Furthermore, to enhance the adsorption capacity of glucose on 2DPA, the surface was functionalized with transition metal (TM) atoms. The results show that TM decoration significantly increases the binding affinity of glucose molecules, suggesting a synergistic effect that enhances sensor performance. Overall, this study highlights the potential of 2DPA as a versatile material for next-generation glucose sensors, offering high performance, stability, and biocompatibility for practical medical applications.

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# Self-assembly of a Saddle Shaped Molecule on Metal Surface: Insights from First Principles

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Self-assembly of organic molecules on metal substrate is a fundamental process in nanomaterials design, having possibilities of wide range of applications especially in the field of organic electronics. In our combined experimental and theoretical study, we investigate molecular self-assembly and adsorption of saddle-shaped molecule on metal surface. We explore how molecule's orientation with respect to the underlying substrate affects the adsorption geometry and charge transfer. Our DFT simulations reveal the lowest energy configuration, in agreement with experimental findings. The adsorption process leads to the flattening of molecular curvature due to charge transfer, which prevents the molecular stacking along the direction perpendicular to the substrate. We also corroborated our electronic structure calculations with experimental data and the excellent agreement found between the two re-confirms the experimental findings.

# Chemical tuning of metavalent bonding at the interfaces in chalcogenide superlattices: A first-principles study

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Group IV chalcogenides with metavalent bonding (MVB), a form of bonding intermediate between metallic and covalent interactions, exhibit unusual combination of delocalization and gapped electronic structure that enables exceptional properties. In this study, we consider superlattices consisting of alternating layer of MVB and ionic chalcogenides, and analyze the evolution of MVB across an interface using first-principles theory. We use Born effective charge ($Z^*$) as the key local descriptor of MVB and determine its spatial variations within PbTe-SrTe superlattices. We establish that MVB saturates precisely within 2.5 unit cells of the interface thus involving 5 bonds along (-X-M-)$_n$ chains, in contrast to the three-center four-electron (3c-4e) bonding. When substitution of Sr atom for Pb disrupts the (-Pb-Te-)$_n$ chain, local electric field altered through redistribution of charges result in suppression of MVB. Such tunable MVB uncovered here offers routes to design of superlattices with tailored thermoelectric, IR and electronic properties.

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AAPALI PSI-K 2025

# Interaction of Li/Na/K with Holey Graphyne-Nickel Sulfide Hetero System for Metal-Ion Battery Anode

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As global energy demands continue to rise, metal-ion batteries have emerged as a promising solution for sustainable energy storage applications. However, conventional battery electrode materials often suffer from low electrical conductivity and high diffusion barriers, leading to performance degradation over repeated charge/discharge cycles. In this study, we employ density functional theory (DFT) calculations to investigate the electrochemical potential of holey graphyne-nickel sulfide (HGY-$Ni_2S_2$) hetero system as a prototype anode material for metal-ion batteries. Our computational analysis confirms the thermodynamic stability of the hetero system, exhibiting metallic characteristics that ensure excellent electrical conductivity and enhanced ion mobility. The strong interaction of alkali metal atoms (Li, Na, and K) with the hetero system is demonstrated by intercalation studies, yielding binding energies of -3.48 eV per Li atom, -3.25 eV per Na atom, and -3.64 eV per K atom. Notably, the binding energy remains negative for up to four adatoms while maintaining a positive open-circuit voltage, indicating the system's ability to accommodate additional metal atoms and, consequently, high storage capacity. Furthermore, the calculated low diffusion barriers (0.32 eV for Li, 0.24 eV for Na, and 0.11 eV for K) suggest facile ion transport within the HGY-$Ni_2S_2$ hetero system, which is highly desirable for fast-charging battery applications. These findings provide valuable insights into the potential of HGY-$Ni_2S_2$ as a next-generation anode material and pave the way for further experimental validation toward the development of high-performance alkali metal-ion battery technologies.

# Exciton-phonon coupling-mediated anomalous emission dynamics in a newly emerged twisted quasi-2D Piezoelectric Perovskite

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In the past few years, quasi-2D Ruddlesden Popper (RP) perovskites have been a fascinating topic of interest due to anomalous ability of their surface edge states to dissociate excitons into long-lived free carriers. As a consequence of which, several researchers have investigated the fundamental origin of edge states in quasi-2D RP perovskites.[1] However, it is still debatable whether these are intrinsic electronic states due to crystal lattice distortion at the surface edge or self-form 3D perovskites on the edge of quasi-2D RP perovskites.[2]

To the best of our knowledge, our work is the first fundamental, experimental, and computational study on a newly emerged twisted layered 2D RP perovskite single crystal, and one of the first Iodide-based quasi-2D perovskites with RT ferroelectric/piezoelectric behaviour. We investigated this intriguing and favourable system to demonstrate the effect of Piezoelectricity and exciton-phonon interactions on emissive properties and further investigate whether this can be augmented for phonon-modulated hot-carrier relaxation process. Herein, we report its piezoelectric and ferroelectric properties for the first time using quantitative nanoscale imaging and identify both, in-plane and out-of-plane piezoelectricity, with a significantly high $d_{33}$ piezoelectric co-efficient value as compared to other 2D perovskites. To elucidate the origin of dual PL emission, we investigated the photoluminescence (PL) properties of single crystals, exfoliated single crystals, and atomically thin layers. Subsequently, we correlate the emergence of edge states with synergetic effect of charge-transfer Moiré excitons, piezoelectric-Paraelectric phase transition, and strong electron-phonon coupling. Interestingly we observe that the piezoelectric phase at RT evolves to a stable Quantum Paraelectric phase at lower temperature due to quantum fluctuations, manifested via a well-defined polaronic emissions. Using Ab-initio simulation technique, we investigate the underpinning fundamental origin of the anomalous behaviour in the twisted layers of a newly emerged quasi-2D perovskite.

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# MXene/Blue Phosphorene for Photocatalytic Overall Water Splitting Application.

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MXenes, represented by the general formula $M_{n+1}X_nT_x$, are a class of two-dimensional (2D) materials composed of transition metals (M), carbon or nitrogen (X), and surface functional groups (T) such as hydroxyl or oxygen. Their exceptional electrical conductivity, high surface area, good mechanical properties, chemical stability, and low toxicity make them promising for a wide range of applications, including energy storage, electronics, catalysis, and biomedicine. To further enhance their properties, MXenes can be combined with other 2D materials to form van der Waals (vdW) heterostructures. Our work focuses on the MXene/blue phosphorene (BlueP) heterostructure, where strain engineering is employed to tune their electronic and mechanical properties. By systematically studying the effects of biaxial and uniaxial strain, we demonstrate how strain modulates key properties such as bandgap, electronic states, and material anisotropy. Specifically, the $Sc_2CO_2$/BlueP heterostructure exhibits type-II band alignment, which enhances photocatalytic performance by facilitating efficient charge separation. Electrons migrate to the conduction band minimum (CBM) of one layer, while holes remain confined to the valence band maximum (VBM) of the other, aided by a built-in electric field that reduces charge recombination. Using both empirical and density functional theory (DFT) methods, we evaluated its potential for photocatalytic water splitting, supported by absorption spectra analysis. Notably, our findings reveal that applying compressive strain further enhances photocatalytic efficiency, underscoring the heterostructure's suitability for water-splitting applications. This work highlights the significance of strain engineering in optimizing 2D materials for sustainable energy applications.

**Keywords**: First principle calculation, Mxene, Blue phosphorous, heterostructure, External tuning parameter, Photocatalysis

**Reference**: Sarga, P. K.; Karthik, H. J.; Bhattacharyya, S. Electronic and Interfacial Properties of 2D MXene/Blue Phosphorene Heterostructures: Impact of External Strain for Thermoelectric Applications. *ACS Appl. Electron. Mater.* **2024**, *6* (11), 8039–8058. https://doi.org/10.1021/acsaelm.4c01386.

# Electronic Structure of the Mixed Compounds, $Ca_{1-x}La_xFe_{1-x}Mn_xO_3$

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The unusually high valence state of $Fe^{4+}$ ions in $CaFeO_3$, despite their orbital degeneracy ($t^3_{2g} – e^1_g$), remain free from the Jahn-Teller effect, unlike isoelectronic $Mn^{3+}$ in $LaMnO_3$ [1,2]. In the case of $CaFeO_3$, this orbital degeneracy is overcome by Breathing Mode distortion, resulting in charge disproportionation [3]. To investigate the interplay of these structural and electronic properties, the first-principles density functional theory (DFT) calculations were carried out in the plane-wave basis with projector augmented-wave potentials [4], as implemented in the Vienna Ab initio Simulation Package (VASP) [5]. We explored the mixed compounds $Ca_{1-x}La_xFe_{1-x}Mn_xO_3$ (x = 0.25, 0.50, 0.75) [6, 7, 8, 9], where intersite charge transfer from Mn to Fe results in non-degenerate electronic states for both Mn ions ($Mn^{4+}$: $t^3_{2g}$) and Fe ions ($Fe^{3+}$ : $t^3_{2g}$ $e^2_g$). Our findings reveal a transition from an insulating parent phase to a half-metallic state at x = 0.25 and 0.75 [10], while it remains insulating at x = 0.50. Additionally, a local structural study uncovers atomic-scale phase separation in La, Mn- and Ca, Fe-rich regions driven by strong Fe-O covalency [11]. Magnetic calculations indicate ferromagnetic ordering in the half-metallic state, whereas an A-type antiferromagnetic ordering emerges in the insulating phase[12, 13, 14]. The origin of these magnetic states is attributed to a kinetic energy-driven mechanism, offering insights into the complex electronic and magnetic behavior of these materials.

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# A Physically Inspired Machine-Learnable Representation of Electronic Structure

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The development of machine learning-based surrogate models for expensive electronic structure calculations have resulted in the development of both predefined and learnable descriptors. However, existing descriptors often involve trade-offs between accuracy, interpretability, and computational cost. We propose a physically inspired representation of the electronic structure of atoms - the electronic structure vector (ESV), based on atomic orbital occupancies and quantum numbers. The ESV representation, along with atomic numbers Z, (Z-ESV) are used as the initial embeddings for a Crystal Graph Convolutional Neural Network (CGCNN) based model, to predict atomization energies on the QM7 dataset [1] within hybrid DFT-level accuracy (test MAE = 0.075 eV). This approach differs from most state-of-the-art models, wherein atomic identities are fit to data. The versatility of the ESV with atomic numbers is demonstrated on tabular data, via the prediction of formation energies of dual-atom catalysts on graphene [2] (test MAE = 0.09 eV) and adsorption energies of *COCOH on various configurations of Cu-based binary alloy catalysts [3] within DFT accuracy (test RMSE = 0.12 eV). We also show that catalysts organize themselves in a physically meaningful manner in the space of Z-ESV. Properties related to the band structure, such as the d-band centre of binary alloy catalysts [4] and band gaps [5], have also been predicted with good accuracy. Additionally, the ESV enables the classification of inorganic crystals into zero- and finite-gap materials with good accuracy (test ROC-AUC = 0.8988). These results demonstrate that versatile, physically inspired representations of chemical identity can be used to develop novel surrogate models for electronic structure calculations.

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# Three dimensional higher-order van-Hove singularity in approximant quasicrystals

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Van Hove singularities (VHS) are critical points in the electronic band structure where the density of states (DOS) diverges, often enhancing electronic correlations and driving emergent quantum phases. However, such singularities are rarely observed in three-dimensional systems. Using first-principles calculation and symmetry analysis, we demonstrate the presence three-dimensional higher-order saddle-point VHS in approximant quasicrystals, with substantial flat energy dispersion near the Fermi level. This results in a power-law divergence in the DOS, significantly amplifying correlation effects that can drive instabilities toward charge density wave (CDW) formation, superconductivity, and nematic ordering. Using $\mathrm{Os}_4\mathrm{Al}_{13}$ as an exemplary system, we show that the large DOS near the Fermi energy enhances superconductivity, with an experimentally measured high superconducting transition temperature of 5.47 K. A detailed electronic structure and symmetry analysis further reveals a nontrivial electronic state characterized by a $\mathbb{Z}_2$ topological invariant ($\mathbb{Z}_2 = 1$) and spin-polarized conducting surface states [1]. The interplay between superconductivity, topological effects, and higher-order VHS offers new insights into the possible realization of topological superconductivity in quasicrystals and their approximants.

1. Pavan Kumar Meena*, Rahul Verma*, Arushi, Sonika Jangid, Roshan Kumar Kushwaha, Rhea Stewart, Adrian D. Hillier, Bahadur Singh†, and Ravi Prakash Singh† *Observation of superconductivity in a nontrivial* $\mathbb{Z}_2$ *approximant quasicrystal,* arXiv:2501.00554

# Accelerating diffusion analysis on halide solid electrolyte via on-the-fly machine learning force fields

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Due to the lengthy time frames and enormous system sizes, realistic finite-temperature simulations of complex materials create a considerable barrier for first-principles approaches, frequently requiring years of computational work. In this study, we use an on-the-fly machine learning force field (MLFF) technique that creates interatomic potentials automatically while simulating molecular dynamics. This method enables large-scale simulations while preserving the chemical accuracy of first-principles techniques and minimizing the need for human intervention. We apply this framework to study the solid-state electrolyte α-$Li_3YCl_6$ (space group P-3m1) for lithium-ion battery applications. Using a Bayesian inference-based on-the-fly learning technique, we generate an accurate interatomic potential and perform temperature-induced diffusion analysis on a 500 K-equilibrated structure of $Li_3YCl_6$. Our MLFF training spans a wide temperature range (100 K – 1200 K) and allows us to get a well-equilibrated 500K structure. During MLFF training, only 5,979 first-principles calculations were performed out of 250,000 molecular dynamics steps, effectively bypassing 97.6% of the computationally expensive ab initio evaluations. From these reference calculations, 1,602 local configurations were selected as basis sets to construct the regression model and thereby it predicts energies, forces, and stress tensors with near first-principles accuracy of 0.94 meV/atom, 0.029 eV/Å, and 0.27 kbar, respectively. We further extend our study for the production run by applying the trained MLFF to a larger 480-atom system for long-timescale (say100 ps) simulations. Our calculated lithium-ion conductivity 4.87 mS/cm closely matches the value 5.01 mS/cm obtained via direct *ab* initio molecular dynamics on that smaller 500 K-equilibrated system (60 atoms, 40 ps simulation) at RT. This study highlights the transferability of inter-atomic potential of on-the-fly MLFF in modeling complex battery materials at finite temperatures. By enabling highly efficient phase-space exploration with minimal human effort, this approach provides a powerful and accurate framework for predicting ionic conductivity in energy storage materials.

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# Structural and Dynamic Changes in the High Pressure Phases of $MAPbBr_3$ Unveiled by Machine Learning Force Field Simulations

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Hybrid perovskites, such as $MAPbBr_3$, exhibit intriguing structural and dynamic changes under varying pressure conditions, which are critical for understanding their phase transitions and functional properties, particularly for optoelectronic and energy applications [1]. Here, we develop a machine learning force field (MLFF) capable of representing the pressure-dependent potential energy surface of $MAPbBr_3$, up to $\sim 2$ GPa, trained on data from prior ab initio molecular dynamics (AIMD) simulations [2]. This MLFF enables molecular dynamics simulations over extended timescales and larger length scales, which we use to characterize the dynamic behavior of the pressure-induced phases. We observe the emergence of two distinct MA configurations in the intermediate pressure cubic phase in accordance with previous experiments. Order parameters tailored to capture guest-host dynamics are used to describe them and to explain their origin. Additionally, the free energy landscape of the cubic-tetragonal phase transition is investigated by quantifying entropy contributions from various types of molecular motion. In the high-pressure tetragonal phase, previously found to host polar and anti-polar domains [2], we detail their dynamics and associated time scales using specialized order parameters. These findings provide insights into the interplay between local structure and dynamics in the pressure-induced phases of $MAPbBr_3$.

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# Quadruply Fused Porphyrinoid Nanotapes on Metal Substrate: Insight from DFT

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Coordination polymers draw great attention from the research community due to their various applications, ranging from gas adsorption and packing to catalysis and molecular magnets. Here, we report a combined experimental & theoretical study of self-assembled one-dimensional coordination polymeric chains formed by semiconducting building blocks and Fe atoms co-deposited on metal substrates. Our Density Functional Theory (DFT) calculations confirm the different structures seen by Scanning Tunneling Microscopy experiment at different annealing temperatures. DFT calculations also shed light onto the different magnetic orderings and the corresponding electronic structure of the novel macrocycles formed on the surface. Also, we explored the aromaticity of those novel macrocycles formed and commented about the stability of those systems from our calculations. These systems present an ideal platform for studying the interplay between magnetic ordering, molecular structure and substrate effects. Further, since the magnetic orderings of these 1D chains can, in principle, be tuned by external perturbations, it can be used for a variety of technological applications.

# Topological Phase Transitions in Kagome Ferromagnets: The Role of Intrinsic Rashba Spin-Orbit Coupling

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The theoretically predicted Chern insulators have highlighted the potential of easy-axis kagome ferromagnets to host the quantum anomalous Hall effect. This phenomenon can also emerge from in-plane ferromagnetism in kagome systems via the breaking of both out-of-plane and in-plane mirror symmetries [1, 2]. We demonstrate that the interplay between magnetism and mirror symmetries makes ferromagnetic kagome systems a versatile platform for realizing non-trivial topological phases, with the orientation of magnetic moments $\hat{m}(\theta, \phi)$ at lattice sites serving as a key tuning parameter. We show that the Rashba spin-orbit coupling (SOC) induced by broken out-of-plane mirror symmetry together with the intrinsic SOC incorporated in a tight-binding model captures the rich topological phase diagram of kagome systems as a function of $\hat{m}(\theta, \phi)$. In particular, the restoration of in-plane mirror symmetry for specific values of $\phi$ promotes topological phase transition upon variation of in-plane orientation of the moments $\hat{m}(\theta = 90°, \phi)$ [3]. However the topological phase transition for the variation of $\hat{m}(\theta, \phi$=constant) is dictated by a competition between Rashba SOC and intrinsic SOC. Density functional theory calculations for the ferromagnetic kagome monolayer $\mathrm{Co_3Pb_3S_2}$, a representative compound belonging to the family $\mathrm{Co_3X_3Y_2}$ (X=Sn, Pb; Y=S, Se) further corroborate our predictions based on the proposed tight-binding model.

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# Symmetries in zero and finite center of mass momenta excitons

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Exciton calculations often face computational bottlenecks due to the need for diagonalization of large-scale Bethe-Salpeter Equation (BSE) Hamiltonians. This problem becomes more severe for fine exciton center-of-mass (COM) momentum sampling, where we need to diagonalise Hamiltonians for many exciton COM momenta. We use a crystal symmetry-based approach to reduce this computational cost. We implement the rotation of finite COM exciton wavefunctions based on crystal symmetries from a calculation of exciton wavefunctions on the irreducible Brillouin zone. In addition, we introduce a symmetry-adapted basis to block-diagonalize the BSE Hamiltonian at zero exciton COM momentum, further reducing the computational costs. This method enables efficient calculation of the quantities like exciton-phonon couplings and exciton spectra.

# Berry Curvature-Induced Hall Effect in $Mn_2PtSn$ Heusler Alloy: A First-Principles Study

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The transport properties of spintronic materials are strongly influenced by the Hall effect, which originates from spin–orbit coupling and the underlying electronic structure. This effect becomes particularly significant in surface-related studies, as spintronic materials are often employed in thin-film form. Heusler alloys, known for their exceptional magnetic and transport characteristics, are promising candidates for such applications. Despite their potential, no prior reports exist on the Berry curvature-induced Hall effect in the $Mn_2PtSn$ Heusler alloy and its surfaces. In this study, we used combined approach of of density functional theory and maximally localized Wannier functions to investigate the Berry curvature contributions to the Hall effect. Our analysis reveals prominent Berry curvature peaks along high-symmetry k-points, which play a crucial role in determining the anomalous Hall conductivity of this system. These findings provide valuable insights into the transport properties of $Mn_2$ PtSn, highlighting its potential for spintronic applications.

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# Identification of $C_3N$/Silicene heterostructure as anode materials for Li-ion battery and its origin

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Silicene, the silicon analogue of graphene, has been identified as a high energy density anode material for lithium-ion batteries. However, due to its low mechanical stability and high volume expansion, developing a suitable combination of silicene as a 2D anode material remains challenging. In this work, using first-principles study, we have demonstrated that the ratio of two-dimensional polyaniline ($C_3N$) and silicene in the multilayer heterostructures plays a major role to define the Li storage properties and to provide metallicity for easy conduction of electrons and increase the stability and specific capacity with moderate open circuit voltage. The volume expansion for fully lithiated heterostructures is within 22 %. The proposed 2D heterostructures could be a future material for anode in LIB’s and the description of the synergetic effect on Li storage properties will help for further development of 2D heterostructure materials. The low interlayer diffusion energy of Li-ion makes all the heterostructure as a potential anode material. The proposed study will help to understand the Li storage properties of the $C_3N$/silicene based composite to develop the anode materials with a certain approach.

Keywords: DFT, Li-ion Battery, C3N, silicene, Anode, synergetic effect, Heterostructure.

# Exploring Yb-based triangular lattice a potential candidate for quantum Spin liquids: $HYbO_2$

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Early ideas of quantum spin liquids (QSLs) were first established from the magnetic frustration encountered in 2D triangular antiferromagnets [1]. More recently, the family of compounds with a chemical formula $AYbX_2$ [A = Na, K and X = O, S, Se], which feature 2D isotropic traingular lattices, has been shown to exhibit QSL ground states [2,3]. Interestingly, the compound $HYbO_2$ shares a similar chemical formula with this family but contains spatially anisotropic 2D triangular layers. The compound is synthesized by a group at IIT Tirupathi for investigating its probable candidacy for QSLs. Motivated by this, we have investigated the basic electronic structure of $HYbO_2$ using *first-principles* based calculations. From our calculations, we found that spin-orbit coupling (SOC) plays a crucial role in driving antiferromagnetism in $HYbO_2$. Notably, we have also observed that the orbital magnetic moment is higher than the spin moment, suggesting that the system exhibits $J_{eff}$ = 1/2 magnetic moments and significant magnetic anisotropy in the system, both due to the presence of substantial SOC.

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# Atomically Injected Ni-Atom over Graphene and Analogous Substrates for Hydrogen Evolution Reaction: A DFT Study

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A rational catalyst for electrocatalytic hydrogen evolution reaction (HER) is a long-standing challenge that researchers are confronted with[1]. In view of this, tiny particles of transition metals (TMs) spread over a substrate acting as an active site for the reaction, scientifically known as single-atom catalysts is seen as an efficacious way for designing an efficient catalyst[2]. Herein, we comprehensively investigated catalytic activity of Ni-atoms spread over various kinds of two-dimensional (2D) substrates like graphene, AlC, AlN, h-BN, BeO, and MgO (Ni@2D) towards HER using density functional theory calculations. All the considered 2D substrates have various inequivalent anchoring sites like top, hollow, bridge, and vacancy sites for Ni-atoms. So, there are total 34 anchoring sites, and we computed binding energy ($E_b$) of Ni-atom over all the sites. Having large number of configurations, we first applied a screener on stability of Ni@2D and only considered those configurations for which the $E_b$ value is $\leq$ 3.00 eV for further calculations. Out of 34, 17 configurations were falling in this range. Further, we computed the differential Gibbs free energy of H-adsorption ($\Delta G_H$) and generated volcano plot between $\Delta G_H$ and exchange current density ($i_0$) as a prime indicator of HER activity. Then, we screened these configurations based on $\Delta G_H$ values that $\Delta G_H \leq |0.5|$ eV, and out of 17, 10 systems were falling in this region. At last, we examined complete reaction profile of HER via Volmer-Heyrovsky (VH) and Volmer-Tafel (VT) mechanisms over the remaining 10 configurations, and the lowest activation energy for HER are 0.12 eV and 0.21 eV for Ni@AlN and 0.28 eV and 0.36 eV for Ni@h-BN via VT and VH mechanism, respectively. Our findings show Ni@AlN and Ni@h-BN could be a non-noble TM candidate for eco-operational HER catalyst.

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# Compression-Induced Comproportionation in Palladium Trifluoride

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Transition metal compounds offer a versatile platform for studying crucial chemical and physical phenomena, such as electron transfer, metal-to-insulator transition, charge ordering, spin-spin interactions, to just name a few. On the other hand, oxides and fluorides of palladium shows immense technological importance, both in current (optoelectronics, photovoltaics) and future (superconductivity, quantum computing) applications. Palladium trifluoride ($PdF_3$) is a mixed-valent system better formulated as $Pd^{II}Pd^{IV}F_6$. In an attempt to explore the high pressure polymorphs (p >1 GPa) of this compound we perform a joint theoretical and experimental study on its properties at large compression. Hybrid density functional calculations predict the thermodynamically stable ambient-pressure $LiSbF_6$-type polymorph of $R\bar{3}$ symmetry. The ambient-pressure $LiSbF_6$-type structure remains the most stable polymorph of palladium trifluoride up to 27 GPa, in contrast to previous PBE-calculations which predicted a transition into a tetragonal ($P4_12_12$) structure above 19 GPa.[1] In fact, our calculations indicate that this tetragonal structure is dynamically unstable, i.e. exhibits imaginary modes in phonon dispersion curves. Applying the distortion of one of these modes and re-optimizing the structure yields a orthorhombic structure of $P2_12_12$ symmetry which turns out to be the most stable polymorph up to around 50 GPa. The predicted phase sequence at room temperature and below 70 GPa is: $R\bar{3} \xrightarrow{27\ GPa} P2_12_12 \xrightarrow{53\ GPa} P\bar{1} \xrightarrow{55\ GPa} C2/c$. The triclinic $P\bar{1}$ and monoclinic C2/c structures have been predicted earlier, but with very different stability fields.[1]

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# Coexistence of spin valley coupled Dirac semimetal and robust quantum spin Hall state in halogenated BiAs film

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Two-dimensional (2D) honeycomb layered structure (HLS) has emerged as a significant pathway for exploring the intercorrelation between two distinct degree of freedom in Dirac semimetal. The Dirac spin-valley-based characteristics of electrons in HLS offer an unparalleled opportunity to investigate various physical properties [1]. Though, the availability of suitable spin-valley-coupled Dirac semimetals (svc-DSMs) exhibiting robust Dirac spin-valley coupling, is quite bounded. Here, we utilize *first-principles* calculations to explore multiple topological phases in functionalized bismuth arsenic (BiAs) monolayer. More intriguing, we demonstrate that it harbors an exceptionally uncommon svc-DSMs state under modest tensile strain, which can be attributed to the orbital filtering phenomenon and giant spin splitting (helpful to cut down spin-flip scattering in spintronics) induced by strong spin-orbit coupling (SOC). The Dirac fermions residing in K/-K valleys exhibit opposite spin moment and Berry curvature, indicating that the two valleys are inequivalent. We found that strong SOC and broken space inversion symmetry leads to the Rashba spin splitting in band structure with a noteworthy Rashba parameter of 1.03 eV $\mathring{A}$ around the M point of the Brillouin zone. On further increment of strain, the topological phase undergoes a transition from svc-DSMs to a quantum-spin Hall insulator. Through alterations in stacking order with strain, a novel bilayered halogenated BiAs is showcased quantum spin Hall state with a large gap ($E_g(k) = 0.26$ eV) appearing at the K point with the inclusion of SOC. Moreover, our finding with the hBN substrate exemplifies that the interaction between substrate and monolayer does not alter the svc-DSMs state of the system. Our findings greatly enhance our understanding of svc-DSMs and offer valuable insights for experimental detection and possible applications at high temperatures [2].

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# AAPALI PSI-K 2025

# Machine Learning Force Field-Accelerated Prediction and Modulation of Lattice Thermal Conductivity in 2D TMDs

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Thermal transport is a fundamental property of condensed matter, in which heat is transmitted through microscopic collisions of particles such as phonons and electrons. The successful synthesis of single- to few-layer Transition Metal Dichalcogenides (TMDs) has revolutionized the field of nanoelectronics. Characterizing their thermal transport properties is crucial for efficient device integration and overheating management. While Density Functional Theory (DFT) effectively simulates thermal properties, it incurs high computational costs. Our study proposes a convenient strategy to evaluate the phonon dispersion and $K_L$ of $WS_2$ monolayer as one of the TMDs, including strained variants, by integrating on-the-fly machine learning force field (MLFF) with the phonon Boltzmann transport equation [1]. We found MLFF predicted phonon dispersion and the room temperature $K_L$ of $WS_2$ (99.4 $Wm^{-1}K^{-1}$) monolayer which agree well with DFT calculated value (100.6 $Wm^{-1}K^{-1}$). Further, the MLFF predicted $K_L$ falls perfectly with temperature as DFT-based values [2]. MLFF found to be much efficient over DFT in terms of time and recourses consumption to calculate these thermal properties. This highlights the high accuracy and efficiency of MLFF in predicting thermal properties.

Furthermore, biaxial strain can modulate the thermal transport properties of TMDs monolayer. Using the above strategy, we extended our predictions to room-temperature $K_L$ of biaxially strained $WS_2$ monolayers (2% to 20%), finding instability at 21% strain due to imaginary phonon frequencies. The $K_L$ decreases non-monotonically with increasing strain in $WS_2$, from 99.4 $Wm^{-1}K^{-1}$ at 0% to 44.5 $Wm^{-1}K^{-1}$ at 20%. This trend is similar to other 2D TMDs [3]. The minimum $K_L$ of 11.1 $Wm^{-1}K^{-1}$ was found at 16% strain. We analyzed the harmonic and anharmonic properties of the strained $WS_2$ monolayer and observed an overall impact of decreasing phonon group velocity and phonon lifetime, while the increasing trend of specific heat with strain, induces the resultant reduction trend of the $K_L$ in the strained $WS_2$ monolayer. Considering remarkable advancements in 2D TMDs materials, the proposed methodology offers an efficient approach for investigating and modulating thermal properties in these materials.

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# AAPALI PSI-K 2025

# $RbGeBr_3$ Perovskite Solar Cells: Explored via DFT, Machine Learning and SCAPS-1D studies

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This study is aimed to predict the performance of $RbGeBr_3$ perovskite based solar cell from Density Functional Theory (DFT) study and Machine Learning (ML) models and validated through the Solar Cell Capacitance Simulator (SCAPS-1D). We first investigated a single-junction $RbGeBr_3$ perovskite solar cell, achieving a power conversion efficiency (PCE) of 18.25% from DFT calculations. For ML the dataset, sourced from the open-access database by T. Jesper Jacobsson et al. and extracted from the MaterialsZone repository [1], comprises 43,252 perovskite solar cell devices with 100+ parameters per device, generated from DFT calculations. We refined the dataset to 11,706 entries by filtering columns, removing duplicates, validating input descriptors and by systematic exploration to identify patterns, duplicates, and missing values, followed by the removal of abnormal data, validation of input descriptors, study conducted with 445 solar cell devices. Using ML techniques, we predicted PCE of solar cell device based on material layers, properties, and device configurations. Among Decision Tree (DT), K-Nearest Neighbor (KNN), Support Vector Regression (SVR), Random Forest (RF), and Gradient Boosting Regression (GBR), the RF model performed best, predicting a PCE of 25.34% with a test RMSE of 0.604% and a 97% test correlation coefficient. GridSearchCV and fivefold cross-validation optimized the hyperparameters. SCAPS-1D validation for $Al/FTO/SnS_2/RbGeBr_3/P_3HT/Ni{:}Au$, with and without a Hole Transport Layer (HTL), closely matched ML predictions, achieving a PCE of 25.79% with a 0.45% error. These findings highlight the potential of $RbGeBr_3$ for high-performance perovskite solar cells, warranting further experimental validation.

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# DFT Perspective on Reactive Glass-Metal Interactions Enables Surface Modification of Gold Nano-Islands

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Strong metal-support interactions (SMSI) at the interface of metal nanoparticles and support material is known to give rise to interesting interfacial properties [1]. On similar lines, an intriguing development is the emergence of reactive metal-support interactions (RMSI), wherein chemical exchange takes place at the metal-support interface facilitating the formation of multielement nanoparticles (MENPs), which are otherwise unattainable through conventional methods.[2] These unique multi/bimetallic nanoparticles show novel electronic and geometric configurations such that the modified surface composition and atomic arrangement significantly enhance molecular adsorption-desorption which is crucial for catalytic and sensing applications. [2, 3]

In this work we have experimentally demonstrated that as a result of RGMI at the interface of sodium aluminophosphosilicate (NAPS) glass and gold nano-islands (GNIs) at 550 °C under ambient conditions, produces thermally robust and tailored Au surfaces. Extensive characterization studies reveal that Na and P from the glass support are incorporated at the GNIs surface following which lattice distortions in the Au(111) planes are observed. To understand the properties and behavior of the modified GNIs we have performed first-principles density functional theory calculations using Quantum ESPRESSO software package on Au(111) slab doped with Na and P. Our results indicate adsorption of both Na and P on Au(111) surfaces are responsible for lattice distortions. We find that interaction of Na with the host atoms is electrostatic in nature while that of P is chemical which results in formation of hybridized states near the Fermi level of Au(111).

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# Abstract template for AAPALI PSI-K 2025

## Enhanced thermoelectric properties of Zinc-Indium co-doped $Sn_{1.03}Te$

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### ABSTRACT

The development of high-performance thermoelectric materials is pivotal for energy conversion technologies. This study presents the synthesis and characterization of Zinc Indium doped $Sn_{1.03}Te$, which exhibits excellent thermoelectric properties. The doping process involved the incorporation of varying concentrations of Zinc (Zn) and Indium (In) into the $Sn_{1.03-x-y}Zn_xIn_yTe$ (x = 0.0, 0.01; y =0.0, 0.0025, 0.01) matrix using a solid-state reaction method, followed by hot pressing to enhance the density and homogeneity of the samples. Detailed structural analyses through X-ray diffraction (XRD) and scanning electron microscopy (SEM) revealed that Zn and In atoms were successfully incorporated into the $Sn_{1.03}Te$ lattice without forming secondary phases, leading to a refined grain structure and reduced lattice thermal conductivity. The electrical transport properties were systematically investigated from 300 K to 830 K. In $Sn_{1.03-x-y}Zn_xIn_yTe$ (x = 0.01, y = 0.01), the Seebeck coefficient, electrical conductivity, and thermal conductivity measurements indicated a significant enhancement in the power factor (PF) throughout the temperature range (i.e. 10 $uW/cmK^2$ at RT to 19 $uW/cmK^2$ at 830K) and a huge reduction in the total thermal conductivity (8.0 W/mK) compared to undoped $Sn_{1.03}Te$ (3.8 W/mK) at RT due to heavy phonon scattering. Consequently, the dimensionless figure of merit (ZT) achieved an enhanced value of 0.8 at 830 K compared to only Indium (1%) doped (ZT = 0.7). We are looking to tune the zinc-indium concentration in $Sn_{1.03}Te$ further and hope for further improvement in its thermoelectric properties.

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# DAY 1 : 19 MAY 2025

D2-001

# Theoretical Investigation of Half-Metallic Ferromagnetism, Thermoelectric, and Optical Properties in Novel Half-Heusler RbXSb (X = V, Nb, and Ta) Alloys

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The structural, electronic, magnetic, thermoelectric, and optical properties of RbXSb (X = V, Nb, Ta) Half-Heusler alloys using the first-principles approach as implemented in the WIEN2k Code [1]. Generalized gradient approximation (GGA) [2] (Perdew *et al.*1997) [3] and Tran-Blaha modified Becke-Johnson (TB-mBJ) (Tran & Blaha 2009) [4] were used. By rearranging the atoms in the half Heusler XYZ structure, three structures (Type I, Type II and Type III) can be formed. Total energy calculations have been performed for all three structures. It has been found that all these alloys are stable in the Type I structure. Hence, further calculations are focused on the Type I structure. The nonmagnetic (NM) and ferromagnetic (FM) states are investigated, and all RbXSb alloys are found to be stable in the FM state. According to spin-polarized calculations, RbXSb (X = V, Nb, Ta) alloys are ferromagnetic and also have a perfect half-metallicity (HM) these alloys exhibiting 100% spin-polarization at the Fermi level obey the Slater-Pauling rule with an integer magnetic moment of 3.00 $\mu_B$ per formula unit at equilibrium volume. The magnetic moment mainly originates from the strong spin-polarization of n*d* electrons of X (V, Nb, Ta) atoms and partial involvement of n*p* electrons of Sb atom. Our findings provide insight into the temperature-dependent transport properties of RbXSb (X = V, Nb, Ta) alloys. A significant figure of merit of 0.40 is observed for RbVSb at 1000 K. Optical absorption and conductivity were found to be maximum in the UV region of the spectrum. Half Heusler RbXSb (X = V, Nb, Ta) alloys, could be potential materials for spintronic, thermoelectric and optoelectronic devices.

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**AAPALI PSI-K 2025**

# Development of machine learned interatomic potential for thermal transport

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Interatomic potential plays a crucial role in characterizing the atomistic behaviour of a system and capable to retain the Density Functional Theory (DFT) [1-2] level accuracy that minimizes the high computational cost. Over the past years, several types of interatomic potentials have continuously been developed to investigate atomistic properties of large systems. Recently synthesised gamma-graphyne [3] sparked our interest to develop a machine learned interatomic potential (MLIPs) [4] to study its thermal conductivity, that allows us to gain insights into thermal transport mechanism [5]. An efficient training approach have been implemented in our study that focuses to study thermal behaviour of the system by varying length scale. Structural properties [6] are also analysed that provides validation of our machine learned interatomic potential and exhibit strong agreement with DFT results. Furthermore, our developed machine learned potential can be leveraged for the designing of novel devices.

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# A comparative study of a-$TiO_2$ bulk and its Ni-modified (101) surface for photocatalytic applications

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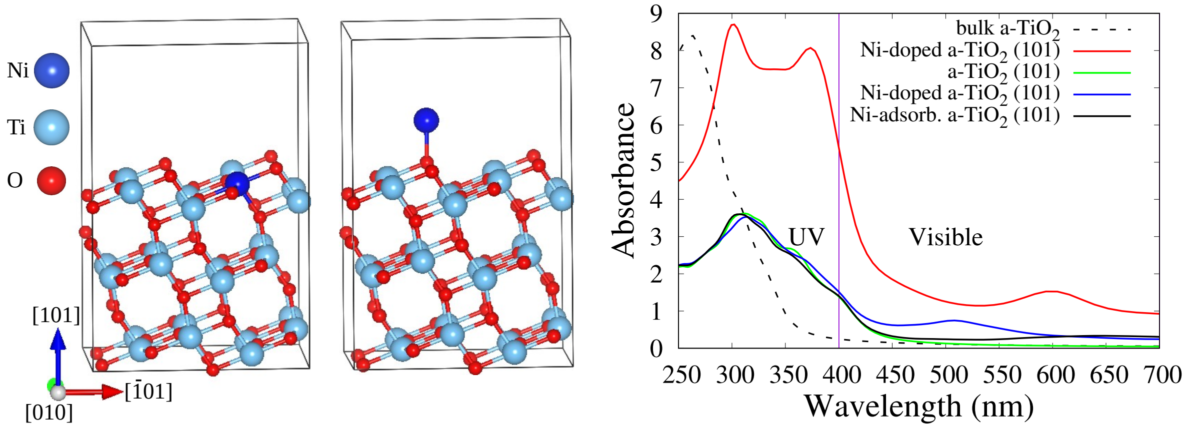
Titanium dioxide (a-$TiO_2$) is the most commonly used semiconductor for photocatalysis applications like green hydrogen production, environmental pollutant degradation, and solar energy conversion. Its practical efficiency however is constrained by a wide bandgap (~3.2 eV) and charge carrier recombination rates. The wide gap limits its absorption to the ultraviolet (UV) region. To overcome this limitation and increase its photocatalytic activity, transition metal doping and adsorption on a-$TiO_2$ (101) surface has been explored as a way to enhance UV and visible-light absorption (Fig.1(c)).

In this study, we conduct density functional theory (DFT) based investigation of the structural, electronic, and photocatalytic properties of bulk a-$TiO_2$, a-$TiO_2$ (101) surface, Ni-doped a-$TiO_2$ (101) surface, and Ni-adsorbed a-$TiO_2$ (101) surface. Our results reveal that Ni doping and adsorption introduces impurity states within the bandgap narrowing it compared to bulk a-$TiO_2$ and extending the material's light absorption into the visible spectrum.

A detailed comparison of adsorption energies, charge density differences, and density of states analysis provides fundamental insights into the stability and photocatalytic performance of Ni-modified a-$TiO_2$. The study highlights that both Ni doping and Ni adsorption are effective for enhancing the photocatalytic efficiency of a-$TiO_2$. These findings provide valuable guidelines for the rational design of next-generation a-$TiO_2$-based photocatalysts for sustainable energy and environmental remediation applications.



(a) (b) (c)

Simulation structure of (a) Ni-doped $TiO_2$ (101) surface, and (b) Ni-adsorbed $TiO_2$ (101) surface. (c) absorption spectra of a-$TiO_2$ bulk, Ni-doped a-$TiO_2$ (101), a-$TiO_2$ (101) surface, Ni-doped a-$TiO_2$ (101) surface and Ni-adsorbed a-$TiO_2$ (101) surface in the x direction.

# Half-Heuslers to Double Half-Heusler: A potential route towards improved thermoelectric performance

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During the past few decades, half-Heusler (hH) materials have garnered significant research attention for thermoelectric applications due to their cost-effectiveness, high thermal stability, mechanical strength, high power factor ($PF$), non-toxicity and moderate efficiency, $ZT$. Our previous study regarding Bi-based hH alloys reveals their potential performances towards thermoelectric device applications. These hH compounds exhibit semiconducting bandgaps in the range 0.62-1.25 eV. Bi-based hH alloys particularly ZrRhBi, ZrIrBi and HfRhBi possess significantly higher degrees of electronic transport compared to common thermoelectric materials like $Bi_2Te_3$. However, their moderate lattice thermal conductivity (5-10 $Wm^{-1}K^{-1}$) limits their thermoelectric performance up to a specific limit. To suppress phonon transport as well as enhance electronic transport further, double half-Heuslers $XX'YY'Bi_2(X, X'$ : Zr, Hf; $Y, Y'$ :Rh, Ir$)$ are formed based on Bi-based potential hH alloys. Initial screening regarding the stability of these double hH alloys is satisfied by means of energetic, dynamical and thermal stability. $Zr_2Rh\,IrBi_2$ and $ZrHfRh_2Bi_2$ exhibit semiconducting bandgaps of 0.66 and 0.59 eV respectively. Other materials are semi-metallic, thus excluded from thermoelectric computations. In case of double hH alloys, electrical conductivity plots showcase increasing behaviour with carrier concentration whereas Seebeck shows a decreasing trend which is in accordance with Pisarenko's relation. In terms of electronic transport, performance of double hH alloys do not exceed hH alloys rather lies in between hH range. However, lattice thermal conductivity of double hH materials is a bit lower compared to their hH counterparts. Depending upon moderate electronic transport and reduced phonon transport, these double hH materials can achieve significant $ZT$ values up to 2.03 around 1000 K. Considering the cost of Ru for experimental fabrications, this work also includes the possibility of making Ru-free double hH alloys.

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# Investigation of the active Cu sites for $CO_2$ reduction on 2D $CuSiO_3$

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Copper metasilicate ($CuSiO_3$) derived from the mineral Dioptase, is an anisotropic two-dimensional (2D) compound with single chains of silicate tetrahedra running along [001]. The $Cu^{2+}$ atoms occupy the edge-sharing $CuO_{4(+2)}$ octahedral chains connected to two distant neighbouring O-atoms. [1] Herein, we study the electronic structure of $CuSiO_3$ employing Density Functional Theory (DFT) and examine the electrocatalytic $CO_2$ reduction (ECR) on [100] facets of $CuSiO_3$. Unlike metallic Cu, which was explored for ECR initially, Cu-based materials have drawn more attention lately as they (i) exhibit selective formation of products due to the presence of $Cu^{\delta+}$ ($0 < \delta < 1$) sites (ii) facilitate easy C-C coupling and are (iii) conducting. Few recent studies have claimed to utilize $CuSiO_3$ either in form of $Cu/CuSiO_3$ or $CuO/CuSiO_3$ interfaces for its improved ECR potential through interface engineering. [2, 3] In the present study, we investigate the synergistic effects of multivalent Cu states and extra stability imparted by $SiO_4$ chains in $CuSiO_3$ and probe the [100] facets for efficient ECR. The spin-polarized studies show [100] planes of $CuSiO_3$ are half-metallic (**Figure 1.a-d**) which is promising for ECR. Comparative study shows $CO_2$ adsorption is stronger on [100] (-1.31 eV) than [020] (-0.26 eV) facets of $CuSiO_3$ and are therefore more active. The detailed mechanistic pathways of ECR and the calculated Gibb's free energies show formation of *CHO is the potential determining step with an energy barrier of 0.31 eV. For effective conversion to products, we studied how the Cu-O-Si interaction controls the formation of various intermediates. Due to its higher conductivity, stability, and abundant active sites on the surface, $CuSiO_3$ [100] exhibits a reduced activation barrier and a greater $CO_2$ conversion ratio for ECR.

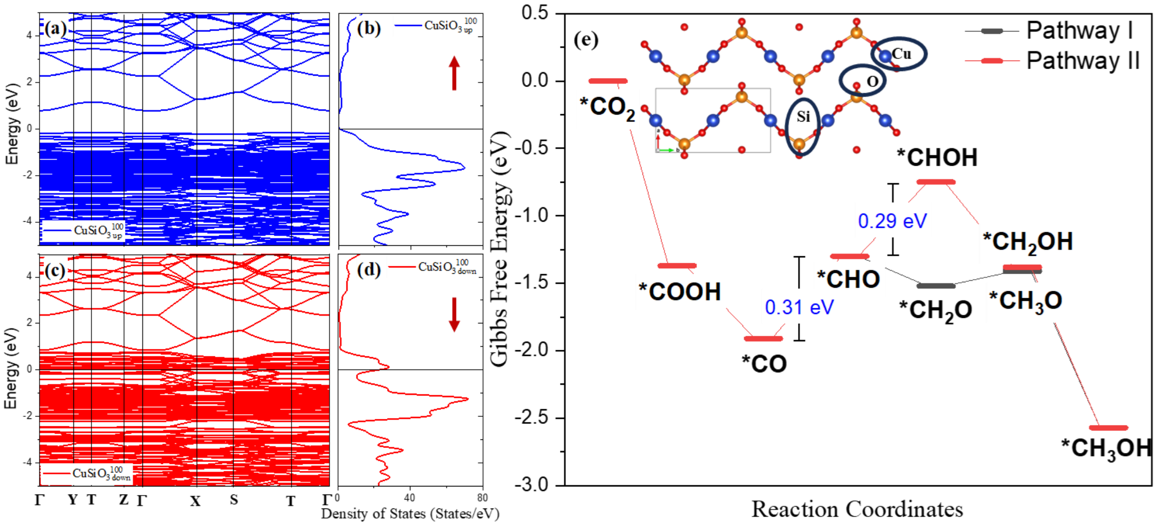

*Figure 1. (a-d) Spin-polarized DOS, and band structure of [100] facet of $CuSiO_3$ and (e) calculated reaction profile for ECR to $CH_3OH$*

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# Controllable Switching between Axion and Quantum Anomalous Hall States in $CrI_3/MnBi_2Te_4/CrI_3$ Heterostructures

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In recent years, intrinsic magnetic topological insulators have become a focal point of research due to their intriguing quantum phenomena and distinctive material topology. The inherent magnetism in these materials disrupts time-reversal symmetry, forming a magnetic gap in non-trivial topological surface states[1]. Notably, $MnBi_2Te_4$ stands out as a prominent intrinsic magnetic topological insulator[2], which, in the 2D limit, exhibits axion physics[3] in an antiferromagnetic (AFM) ground state and quantum anomalous Hall (QAH) state when tuned to ferromagnetic (FM) state. While the QAH state in the FM phase of $MnBi_2Te_4$ holds promise for advanced technologies, the requirement of an external magnetic field exceeding 6T for tuning poses practical challenges.

Using ab initio density functional theory, we showcase the feasibility of switching between AFM and FM phases within a heterostructure composed of two layers of $MnBi_2Te_4$ separated by monolayers of $CrI_3$ via electrostatic gating. Also, by utilizing a simplified four-band model[4] for the surface states, we demonstrate this topology switching. This result signifies the development of a controllable switch capable of toggling between axion and quantum anomalous Hall (QAH) states, thereby addressing magnetic field constraints and expanding the material's potential for applications in spintronics and quantum technology.

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D2-007

# $CsFeX_3$ (X= F, Cl, Br, I) Promising Materials for Spintronics and Infrared Optoelectronics

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The search for new materials that have the potential to revolutionize electronic and optical technologies is ongoing. In this study, we have explored various physical properties of $CsFeX_3$ ( X = F, Cl, Br, I) using first principles electronic structure calculations and highlight their promising applications. Our findings show that these materials exhibit 100% spin polarization, making them ideal candidates for spintronic devices. The spin-up states behave like semiconductors, while the spin-down states reveal metallic characteristics, demonstrating half-metallicity that remains consistent with various values of the Coulomb interaction parameter (U) in the DFT+U calculations. Additionally, $CsFeX_3$ compounds show excellent infrared (IR) absorption and a high refractive index, making them promising options for optoelectronic applications. In particular, $CsFeF_3$ shows the highest IR absorption, whereas $CsFeI_3$ displays the lowest. When it comes to mechanical stability, only $CsFeCl_3$ and $CsFeBr_3$ fulfill the required stability criteria. Further analysis indicates that $CsFeBr_3$ has greater ductility, while $CsFeCl_3$ maintains a more uniform structure, exhibiting very little anisotropy in its mechanical characteristics. Among all the studied materials $CsFeCl_3$ shows a unique combination of half-metallicity, strong infrared absorption, and remarkable mechanical durability, which make it a promising candidate for spintronic and IR-centric optoelectronic devices. This research highlights the immense promise that $CsFeX_3$ compounds hold for shaping the next generation of electronic and optical technologies.

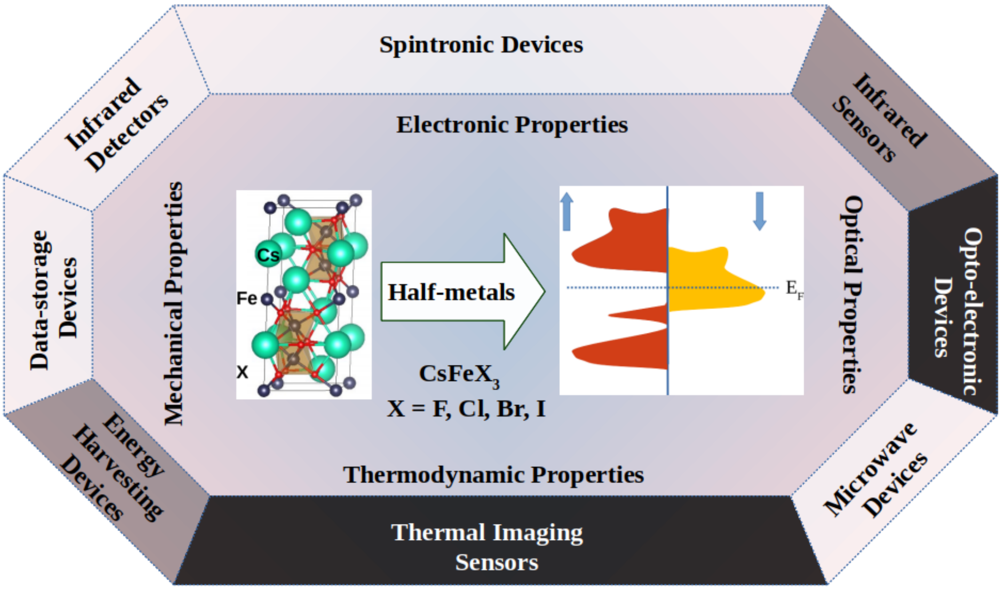

Figure 1: Schematic representing the properties of $CsFeX_3$ studied in this work and the list of their potential applications

# Are ultra thin transition metal dichalcogenides based excitonic solar cells exceptional ?

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## Abstract

We have studied two dimensional (2D) van der Waals bilayered heterostructures (vdWHs) made up of transition metal dichalcogenides $MX_2$ (M = Zr, Hf and X = S, Se) to explore their potential as materials for excitonic solar cells. We have theoretically predicted the power conversion efficiency (PCE) for six possible vdWHs formed from four $MX_2$ monolayers as suggested by Bernardi *et. al* [1]. Calculations carried out confirmed their structural stability and dynamical stability. Systematic investigations of the electronic properties like layer projected band structure, site projected density of states, partial charge density plots of conduction and valence states at their corresponding extremum points in the first Brillouin zone should provide an impetus for experimental realization of some of these vdWHs.

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# Melting of harder-than-diamond lonsdaleite using machine learned potential

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Lonsdaleite, also known as hexagonal diamond, is a rare poly-morph of carbon distinguished by its unique hexagonal structure and exceptional mechanical properties, which may exceed those of cubic diamond. Despite being identified in meteorite samples and generated under specific high-pressure conditions, questions about its electronic properties and stability at high temperatures remain unanswered. This study addresses these gaps by investigating the structural stability, electronic band structure, and thermodynamic properties of lonsdaleite using first-principles and molecular dynamics simulations. Phonon dispersion analysis confirms its structural stability, while band structure calculations reveal an indirect band gap of 3.06 eV. Our calculations yield a bulk modulus of B=450.946 GPa. We further employed non-equilibrium molecular dynamics simulations to evaluate its Gibbs free energy, indicating that lonsdaleite maintains stability up to approximately 2000 K; beyond this, a rapid decrease in free energy suggests a melting point close to 2100 K. These results provide new insights into the potential applications of lonsdaleite under extreme conditions.

# AAPALI PSI-K 2025

## Water dissociation on α-Titanium(0001): Insights into role of water vapour on early stages of high temperature oxidation of Titanium

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The exceptional high strength, light weight, and corrosion resistance of α-Ti make it a perfect choice for jet engine applications.[1] However, a major drawback of titanium and its alloys is their low oxidation resistance during prolonged exposure to oxygen-containing environments at elevated temperatures. Therefore, significant efforts have been made to develop Ti-based oxidation-resistant alloys that maintain titanium's mechanical properties and increase the maximum application temperature. [3,4,5] However, for rational designing of such alloys it is important to develop microscopic understanding of the oxidation process. In a recent study by some of the authors of this work, the early oxidation of Ti in presence of different alloying elements was studied using density functional theory (DFT) based calculations. [6] Since moisture is present under operating conditions, it is also essential to understand how water vapour impacts the oxidation process. Experimental studies indicate that the presence of water vapour accelerates the oxidation rate of Titanium compounds. [2,7-9] However, there is a lack of mechanistic understanding on how water vapour enhances the oxidation rate.

As a first step towards developing such an understanding, in this work, using DFT based calculations, we have studied dissociation of a water molecule on Ti(0001) surface and diffusion of the dissociated oxygen atoms from the surface to the subsurface in the presence of hydrogen atoms. Our findings indicate that water preferentially adsorbs on the top of Ti. We find that the OH bonds of water dissociate sequentially with activation energies of 0.30 eV and 0.58 eV for the first and second O-H bond dissociation resulting in two H atoms and an O atom bound to the surface. Further, we find that the dissociated H atoms are more likely to diffuse from the surface to the subsurface rather than desorb by forming $H_2$ gas. Additionally, we observed that, irrespective of the O sorption site, the diffusion barrier for oxygen is reduced in the presence of hydrogen atoms, compared to when hydrogens atoms are absent. In order to examine the impact of high temperature on the oxidation process, we have evaluated the change in free energy for the full reaction profile at different temperatures (400 K, 800 K, and 1200 K). These free energy calculations indicate that water would prefer to adsorb dissociatively on the surface. In contrast, the O diffusion process remains unaffected, albeit slight changes in the diffusion barriers.

**References:**

# Orientational Dependence of Spin Crossover Properties

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Spin crossover molecules exhibit a remarkable ability to switch between different spin states in response to external stimuli, making them promising candidates for applications in spintronics. To effectively implement these materials in nanotechnology, understanding the behaviour of thin films of spin crossover molecular crystals and the influence of substrates is crucial. This project combines experimental and theoretical approaches to study monolayers of spin crossover molecular crystals on metal substrates. Our Density Functional Theory (DFT) calculations identify the preferred orientations of the molecular overlayers and analyze the spin splitting energy associated with various orientations on metal substrates. This research enables the growth of thin films with tailored orientations and specific spin splitting energies, paving the way for technological applications in advanced electronic devices.

# Machine Learning - Driven Design and Synthesis of Materials for Solid-State Hydrogen Storage

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Solid-state hydrogen storage in metal alloys holds the potential to outperform conventional (liquid/compressed) methods on account of volumetric capacity, safety, and durability. Despite significant performance improvements in metal hydrides over the past two decades, their practical application is hindered by the unattainable thermodynamics and kinetics. Alloying is one of the most effective ways of tailoring the properties of these compositions. However, the traditional trial-and-error approach restricts the exhaustive screening of chemical space, necessitating more efficient strategies. To address this issue, we are integrating ML with experimental techniques to systematically screen the infinite chemical space of plausible metal hydrides, aiming to design and synthesize materials with improved hydrogen storage properties.

In this work, we have built a suite of machine learning models to predict key hydrogen storage properties such as hydrogen storage capacity, (HYST)[1,2], enthalpy of hydride formation (THOR)[1], and equilibrium plateau pressure (EquiP)[3]. We have also built a model capable of predicting the Pressure-Composition-Temperature isotherms (MH-PCTpro)[4]. PCT isotherms provide crucial information, like reversible storage capacity and equilibrium plateau pressure. These models are validated across diverse alloy families, agreeing with experimental results. Further, we employed these models to predict hydrogen storage properties of 6.4 million unseen compositions. In this poster, I will present these models and provide a detailed analysis of the predicted hydrogen storage properties for 6.4 million novel compositions. I will also present the experimental results for the compositions identified by these models.[5] Additionally, I will discuss the active machine learning approach we're using, where model predictions strategically guide experimental design, and experimental results immediately update and improve the model predictability.[6]

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# Magnetic Phase Transition in $LaVO_3$ thin films

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In transition metal oxide perovskites, strongly interacting d-orbital electrons can give rise to a variety of exotic properties. Over the past two decades, two-dimensional materials like graphene and transition-metal dichalcogenides have demonstrated that reducing a bulk crystal to the monolayer limit gives rise to a diverse range of electronic and magnetic properties. Similarly, monolayer transition metal oxide perovskite could unlock a rich spectrum of exotic 2D correlated phases. In this work using first principles calculations, we investigate the magnetic ground state of LaVO3 thin films of different thickness. While bulk LaVO3 has a C-type AFM structure, monolayer prefers a Stripe-AFM phase. There is a phase transition in in-plane configuration from Stripe to AFM as we move from monolayer to bilayer and another phase transition in interlayer coupling from AFM to FM as we go from bilayer to trilayer. The final configuration is same as bulk. We further aim to understand the reason behind the phase transition and how to manipulate it via strain and doping. We also plan on looking at LaVO3 based heterostructures and superlattices.

# Theoretical Investigation on the Magnetic and Spintronic behavior of 2D MXenes

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We have investigated the magnetic properties of oxygen and sulfur-terminated $Cr_2C$ MXene using Density Functional Theory (DFT)[1]. We have calculated parameters like spin magnetic moment, magnetic anisotropy energy, band structure, density of states, and Gilbert damping to understand the magnetic ordering. We have examined the effects of doping on the magnetic properties of $Cr_2C$ MXene with different transition metals. We have found that the magnetic properties of $Cr_2C$ depend on the surface terminations and reflect in terms of magnetic moment and magnetic anisotropy energy. Our results reveal that the Mn-doped $Cr_2CO_2$ gives semi-metallic ferromagnetic behavior with high spin magnetic moment, while Co-doped $Cr_2CO_2$ is antiferromagnetic. Moreover, Mn-doped $Cr_2CS_2$ demonstrates significant magnetic anisotropy energy. This study's findings may help design next-generation spintronic devices, magnetic sensors, and data storage systems[2].

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# Orbital Free Density Functional Theory Applied On Solids And Finite Systems

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In the light of recent progress of Orbital-Free Density Functional Theory (OFDFT) based on approximation of non-local (NL) Kinetic Energy Density Functionals (KEDFs), one can foresee the prospects that lies ahead in terms of computational efficiency and dealing with large scale systems. It is important to mention that, accuracy of OFDFT depends on the approximations made for KEDFs. Till now, the most accurate KEDFs are based on NL kernels constructed from the linear response theory of homogeneous electron gas (HEG). In this work, we explore beyond the HEG by employing a more general kernel based on the jellium-with-gap model (JGM).

We propose a new NL-KEDF that incorporates several new features[1], such as (i) having the correct low momentum(q) limit of the response function for metals and semiconductors without any modelling term, (ii) the underlying kernel is density-independent, and most importantly, (iii) parameter-free. The accuracy and efficiency of the proposed JGM NL-KEDF have been demonstrated for several semiconductors and metals. The encouraging results indicate the utility and predictive power of the JGM kernel for NL KEDF developments. This approach is also physically appealing and practically useful as we have presented a general formalism to incorporate the gap kernel in all existing Lindhard-based functionals. In this work, we extend the jellium-with-gap model (JGM) kernel by making it density-dependent, enabling it to effectively model localized systems such as molecular clusters. Benchmark calculations and analysis of the Pauli potential—which satisfies several critical constraints—demonstrate exceptional agreement with results from the state-of-the-art Orbital-free methods such as Huang and Carter (HC) functional. Additionally, we calculate optical properties using our modified approach. By enhancing the accuracy of NL-KEDFs for finite systems, our work advances nanomaterial design and a deeper understanding of nanoscale phenomena.

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# Application of machine learning for solving materials science problems

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The integration of machine learning (ML) into materials science has revolutionized our ability towards materials exploration, unlocking new frontiers in material optimization and innovation. This proposed poster explores applications of machine learning in solving materials science problems. In the first example, a machine-learned force field is developed for MAX compound $Ti_2AlC$ to explore its non-linear elastic behavior, the underlying mechanism and the effects of vacancies in the aforementioned elastic behavior [1]. The next work involves semiconductor heterostructures, particularly, its type derived from the bandstructures of the constituent semiconductors [2]. In another set of work, we have delved into the domain of nanoscale binary metal alloys to investigate the driving factors determining their core-shell preference using a ML scheme[3], and catalytic properties in Pd containing nanoalloys [4]. In the final example we have utilized ML in developing a microscopic understanding of formation of atomic wires of appreciable length by using experimentally obtained data in training [5]. Using a supervised learning scheme, we have found optimal conditions for Au chain formation. We have further applied an unsupervised learning scheme for the classification of individual breaking traces, thus identifying junction structures associated with longer chains. Our findings are corroborated by ab-initio molecular dynamics simulations.

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# Atomistic and AI-Driven Insights into Ferroelectric Switching in Hybrid Improper Double Perovskite Oxides

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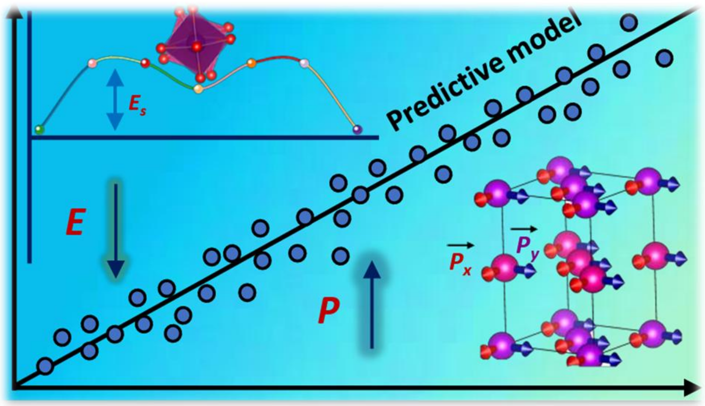
Computational design of multiferroic double perovskite oxides necessitates identifying materials that exhibit robust polarization, magnetization, and strong coupling between these properties. A promising class of such materials follows the general formula $AA'BB'O_6$, derived from $ABO_3$ single perovskite oxides with a centrosymmetric *Pnma* structure. By introducing A/A′ layered ordering and B/B′ rocksalt cation arrangements, the symmetry reduces to a non-centrosymmetric, polar $P2_1$ phase, enabling ferroelectric behavior. A systematic compositional exploration across diverse chemical spaces, combined with polarization evaluation, can reveal new candidates with enhanced ferroelectric properties. In this framework, Our atomistic simulations reveal that polarization switching in hybrid improper ferroelectric double perovskites is driven by structural distortions, particularly out-of-phase octahedral rotations [1]. To expedite the discovery of materials with tunable polarization and switching barriers, we propose a predictive framework that integrates atomistic simulations with machine learning (ML) models trained on first-principles calculations. Our approach identifies key geometry-driven features such as charge states, cationic radii, and structural mode-based order parameters that are crucial for predicting both polarization and switching barriers. These structural modes enable accurate polarization component predictions, offering a pathway to discovering double perovskite oxides with high polarization and low switching barriers. Our findings highlight promising candidates for future spintronic and memory device applications [2].



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# AAPALI PSI-K 2025

# Thermoelectric Performance of *LiCaB* Half-Heusler Alloys in Presence of Aliovalent Doping and Co-doping

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Half-Heusler (HH) alloys have gained significant attention for their promising properties, particularly in thermoelectrics, offering potential solutions to the energy crisis. The efficiency of a thermoelectric material is characterised by high figure of merit (*ZT*) and thus its improvement has proven to be a challenging task. Defect engineering has emerged as a key technique for boosting the thermoelectric efficiency of HH alloys. [1-4]. In the present work, we have introduced divalent (*Mg*) and trivalent (*In*) atoms in the X-site of *LiCaB* at various doping concentrations of 3.125%, 6.25%, 12.5%, 25%, 50% and 75%. Additionally, *Mg* and *In* atoms have been co-substituted in the equivalent doping site. The thermoelectric properties of $Li_{1-x}M_xCaB$ (M=*Mg, In*) and $Li_{0.5}Mg_{0.25}In_{0.25}CaB$ systems have been evaluated within the temperature range of 200-800K. Positive values of Seebeck coefficient (*S*) and Hall carrier density ($n_H$) for $Li_{1-x}M_xCaB$ (M=*Mg, In*) and $Li_{0.5}Mg_{0.25}In_{0.25}CaB$ indicate *p*-type nature of the alloys. Aliovalent doping is effective in lowering the lattice thermal conductivity ($\kappa_l$) for *LiCaB* system. The effect is more pronounced for heavier dopant *In* that introduces larger mass fluctuation than *Mg* enhancing the point defect phonon scattering of the system. More precisely, $Li_{0.75}In_{0.25}CaB$ exhibits the most prominent reduction of $\kappa_l$ by ~79% (1.25 *W/mK*) compared to the pristine alloy (5.94 *W/mK*) at 800K. The $\kappa_l$ of *LiCaB* could be further reduced significantly by ~91% for $Li_{0.5}Mg_{0.25}In_{0.25}CaB$ (0.55 *W/mK*). This results in notable decrement in $\kappa_{tot}$ for the considered doped alloys favouring improved thermoelectric performance. Accordingly, the total figure of merit ($ZT_{tot}$) of pristine *LiCaB* (0.56) could be uplifted by ~5% in $Li_{0.75}In_{0.25}CaB$ (0.59) as well as up to ~30% in $Li_{0.5}Mg_{0.25}In_{0.25}CaB$ (0.73). Thus, defect engineering through doping and co-doping with appropriate concentration provides valuable insight for improving thermoelectric efficiency of HH alloys.

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# Stochastic $G_0W_0$ study on silicon nanocrystals

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Silicon often serves as a prototypical semiconductor due to the vast number of theoretical and experimental studies conducted on it. Defects in silicon play a crucial role in determining its properties and potential applications of silicon nanocrystals in photovoltaics. However, defect studies have typically been carried out using the supercell approach, as the nanocrystal sizes required for converged formation energy calculations were computationally prohibitive.

In this work, we employ the stochastic $G_0W_0$ method, as implemented in the Pseudopotential Algorithm for Real-Space Electronic Calculations, to study neutral and charged single vacancies in silicon nanocrystals. The formation energy is expressed as the sum of the quasiparticle and relaxation energy of the vacancy. The stochastic $G_0W_0$ method is an efficient approach for determining quasiparticle energies in large systems containing thousands of atoms.

Our stochastic $G_0W_0$ implementation has been benchmarked against the GW20 dataset using results from the NanoGW code. Our findings show excellent agreement with the NanoGW results. Leveraging this benchmarked code, we have computed the quasiparticle energies for silicon nanocrystal systems containing approximately 2000 atoms.

# Developing fully ab initio classical force fields for 2D materials

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Graphene and hexagonal boron nitride (hBN) are prominent two-dimensional (2D) materials, which are increasingly finding use in a variety of applications. These 2D materials are weakly bound layered materials, held together by van der Waals interactions. There is a lack of fully ab initio classical molecular dynamics (MD) force fields that accurately capture the electrostatic and van der Waals interactions between the layers of 2D materials. We used a completely first-principles approach to parameterise force fields for these materials. Density functional theory (DFT) based ab initio molecular dynamics simulations (AIMD) were used to parameterize the bonded force field interactions. On the other hand, the semi-local exchange-correlation functionals used in DFT calculations do not sufficiently account for the non-local van der Waals interactions in these layered materials. Moreover, dispersion corrections to DFT overestimate van der Waals interactions. Thus, we employed random phase approximation (RPA) calculations based on the adiabatic-connection fluctuation dissipation theorem to obtain the distance-dependent binding energy of these 2D materials. The nonbonded force-field parameters were obtained by fitting to such binding energy curves. The resulting fully ab initio force fields are expected to provide an accurate description of the mechanical and interfacial properties of 2D materials.

# Spatial distribution of Mayer bond order in periodic systems

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**Abstract**: We present a scheme to partition the total charge of a given periodic system into charges shared among atoms and charges retained by atoms, and plot their spatial distribution using Mayer bond order (MBO) as the primary quantity, in the basis of hybrid atomic Wannier orbitals optimally directed towards coordination. As an application of this approach, we explore two distinct scenarios. First, we quantify the extent to which different orbitals contribute to the Born effective charge of Ti in $BaTiO_3$, providing deeper insight into the electronic structure and polarization mechanisms in this material. Second, we investigate the evolution of metavalent bonding in GeTe across its crystallographic phases(Rock Salt and R-3m), shedding light on the interplay between bonding characteristics and structural transitions. Our findings offer a more detailed and quantitative understanding of bonding and charge transfer in these technologically significant materials.

# Enhanced Thermoelectric Performance of $BaCuGdTe_3$ due to Strong Acoustic Phonon Suppression

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Thermoelectric materials have great potential to mitigate environmental damage by converting the wasted heat generated from fossil fuel combustion into electrical energy [1]. Their efficiency depends on achieving high electrical conductivity while minimizing phononic transport. The occurrence of low phonon transport and high electrical transport is a rare phenomenon in crystalline semiconductors [2]. Hence, various strategies have been explored to modulate lattice thermal conductivity ($\kappa_l$) including nano-crystallization [3], introducing crystal defects [4], inducing structural disorders [5], and incorporating rattling guest atoms [6]. However, these approaches often compromise electronic transport properties. Hence, finding crystalline semiconductors with intrinsically low $\kappa_l$ is a good choice for designing high-performance thermoelectrics.

In this work, using first-principles density functional theory calculations, Boltzmann transport theory and explicit consideration of phonon-phonon and electron-phonon interactions, we study $BaCuGdTe_3$, a layered quaternary chalcogenide semiconductor, which exhibits exceptionally low $\kappa_l$ and enhanced electrical transport properties. This ultralow $\kappa_l$ ($< 0.2$ W/m.K at room temperature) is due to the layered structure and local distortions present in the compound that strongly suppress acoustic phonon modes, reducing phonon group velocities and enhancing phonon scattering rates. Additionally, the large amplitude vibrations of weakly bound Ba atoms create rattling-like modes that cause avoided crossings between acoustic and optical phonon modes, further increasing the scattering channels. The weak chemical bonding within the crystal also softens the acoustic phonons, lowering their group velocities and contributing to the overall suppression of thermal transport. Furthermore, we showed that this compound has excellent electrical transport properties due to strong covalent bonding within its layers. These characteristics make $BaCuGdTe_3$ a highly promising candidate for thermoelectric applications with a high figure of merit at modest carrier concentrations and temperatures.

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# Ligand-Driven Hot-Carrier Dynamics in Metal-Rich CdSe Quantum Dots: Insights from First-Principles and Nonadiabatic Molecular Dynamics

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Metal-rich semiconducting quantum dots (QDs) are promising materials for optoelectronic applications due to their tunable optical properties and potential for efficient hot-carrier utilization. In this work, we investigate the structural, electronic, and dynamic properties of passivated $Cd_{35}Se_{20}X_{30}$ QDs (X = F, Cl, Br, and I) using first-principles calculations and nonadiabatic molecular dynamics. The nature of the halide ligands significantly influences surface-ligand interactions, thereby modulating the electronic structure and charge carrier dynamics. Our analysis reveals that electron–phonon interaction strength varies with ligand electronegativity, directly impacting nonradiative recombination pathways and carrier relaxation lifetimes. Systems passivated with chlorine exhibit the strongest electron–phonon coupling, resulting in enhanced nonradiative losses, whereas bromine ligands extend hot-carrier lifetimes due to suppressed phonon-mediated relaxation. These findings provide critical insights into ligand-controlled hot-carrier dynamics in metal-rich QDs, paving the way for their potential use in hot-carrier photovoltaics, ultrafast photodetectors, and quantum light sources.

# Hot Electron Transfer from Nanoparticle to PFAS Systems: A Size-Dependent Study

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## Abstract:

Plasmonic metal nanoparticles (NPs) are widely explored in photocatalysis and photovoltaics due to their ability to generate hot carriers and transfer them to nearby molecules or semiconductors.[1,2] Our recent work[3] demonstrated direct hot electron transfer (DHET) from silver nanoparticles to perfluoroalkyl substances (PFAS), promoting their degradation and offering a potential route for environmental remediation. In this study, we investigate how the size of icosahedral silver NPs influences plasmonic behavior and hot carrier dynamics in complexes with two PFAS molecules: perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). Using real-time time-dependent density functional theory (RT-TDDFT), we analyze the evolution of localized surface plasmon resonance (LSPR), hot carrier generation, and electron transfer processes.[4,5] Our results show that while the LSPR peak position remains unaffected mainly by PFAS adsorption, the DHET probability decreases with increasing NP size. Smaller NPs exhibit charge oscillations indicative of reversible electron transfer, whereas larger NPs show more stable and directional transfer behavior. These findings provide fundamental insights into the role of NP size in plasmon-driven charge transfer and support the design of optimized nanomaterials for PFAS degradation and related environmental applications.

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# Saturation of Magnetic Exchange Coupling with Increasing Cumulene Coupler Length in Diarylmethyl Diradicals

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## Abstract

Organic magnetic materials are promising candidates for spintronic applications due to their tunable magnetic interactions and long spin coherence times. In this study, we computationally investigate the effect of cumulene coupler length on the magnetic exchange coupling constant ($J$) in seven diarylmethyl diradicals using Density Functional Theory (DFT) with B3LYP, MN12SX, and BP86 functionals. To understand the effect of conjugation on the magnetic nature of the diradicals structural parameters such as bond length have also been investigated. The molecular orbital analysis like the distribution of MOs and their energy gap have also been studied. The distribution of spin density on the coupler and radicals have been analyzed and further confirms that the cumulene bridge actively facilitates magnetic exchange, with spin delocalization shifting from the radical centres to the coupler. The bond length alternation increases with an increase in the length of the coupler. All the functionals give consistent ferromagnetic interactions for all the designed diradicals. The MO analysis shows that the saturation of magnetic exchange comes from the reduced helical nature of LUMO with increasing coupler length. These findings provide key insights into the role of cumulenes in organic magnetic materials, offering a pathway for designing efficient molecular spintronic devices.

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# *Ab-inito* study of biexcitons using exciton product basis

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Biexcitons are bound states of two electrons and two holes. We employ a first-principles many-body theory that uses the exciton product basis to describe biexcitons. This basis offers an advantage over the conventional four-particle approach, as it inherently includes strong electron-hole interactions within the exciton, leaving only the weaker exciton-exciton interactions to be addressed. This leads to faster convergence of the biexcitonic energies. The interactions among the biexciton's constituent fermions are described using an effective many-body Hamiltonian [1], which incorporates screening. We have used the many-body theory of cobosons [2] to construct the biexciton Hamiltonian using the exciton product basis. Using this method, we have calculated biexciton energies for molecules, and our results are in good agreement with other calculations.

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# Adsorbate-Adsorbate Interaction Model-based Prediction of Oxygen Coverage on Molybdenum Carbide Catalysts and Work Function Correlations

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Molybdenum carbide ($Mo_2C$) has emerged as a promising non-noble metal catalyst for alkane dehydrogenation to alkenes due to its Pt-like characteristics. Understanding the adsorption behaviour of oxygen formed from $CO_2$ dissociation on $Mo_2C$ surfaces during $CO_2$-assisted oxidative dehydrogenation ($CO_2$-ODH) of alkanes is crucial for enhancing its alkene selectivity[1]. We employed a model-based approach guided by the work of Prabhu and Choksi (2024)[2] to predict coverage-dependent surface free energies for oxygen adsorption on $Mo_2C$(101). The model, which incorporates adsorbate-adsorbate lateral interactions, enabled us to construct accurate ab initio phase diagrams to predict the maximum possible oxygen coverage, with minimal computational expense compared to traditional Density Functional Theory (DFT) methods. Our calculations revealed strong site-dependency for oxygen adsorption, with the Mo-top sites dominating at low coverages while Mo-C bridge sites became competitive at higher coverages. Under steady-state ODH conditions, 1 monolayer (ML) oxygen coverage emerged as most stable. Work function calculations demonstrated pronounced dependence on oxygen coverage, adsorption site type, and surface structure, with significant increases at low coverages but minimal changes at higher coverages. Different correlation patterns between work function and adsorbate-adsorbate interactions, expressed in terms of interaction energy which depended on oxygen coverage, were observed depending on surface structure, surface termination, and site types. These electronic modifications may significantly impact catalytic performance of $Mo_2C$ in ODH reactions. Understanding this relationship provides critical insights into how oxygen coverage modulates electronic structure of $Mo_2C$, affecting subsequent adsorbate interactions and reaction pathways. These findings offer a theoretical foundation for rational design of $Mo_2C$ catalysts with optimized oxygen coverage for enhanced alkene selectivity.

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# Exploring Metal-Doped $C_2N$ Catalysts for $CO_2$ Reduction to $C_2$ Products via an Alternative C-C Coupling Pathway

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Renewable energy-driven electrocatalytic $CO_2$ reduction ($CO_2RR$) offers a sustainable route to transform $CO_2$ into valuable feedstocks and fuels. In particular, multi-carbon ($C_2^+$) products such as ethylene, ethanol, and n-propanol exhibit higher volumetric and gravimetric energy densities—and thus greater market value—than typical $C_1$ products like methane, CO, or formic acid. However, efficient production of these multi-carbon products is hampered by the inherent challenges of activating the chemically inert $CO_2$ molecule, suppressing competing hydrogen evolution (HER), and facilitating robust C–C bond formation.

In our study, we systematically explore monomeric, dimeric, and trimeric configurations of transition metal-doped 2D $C_2N$ ($TM_n@C_2N$)—a promising catalyst support noted for its high stability and nanoporous structure—to identify optimal metal sites for enhanced $CO_2RR$ performance. Our computational investigations demonstrate that $Mn_3@C_2N$ is an exceptionally effective catalyst. It not only dual-activates $CO_2$ and promotes alternative C–C coupling pathways beyond the conventional CO dimerization mechanism but also selectively produces $C_2$ products while effectively suppressing both HER and CO evolution. These findings mark a significant advancement in catalyst design, paving the way for sustainable conversion of $CO_2$ into high-value multi-carbon products.

# Machine Learning Potential Based Study of Borate Glasses

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Borate glasses are widely used in various industries to create everyday instruments [1]. Although these glasses have been studied experimentally for a long time, their amorphous network structure makes it difficult to understand how their physical properties arise from their microscopics. Molecular dynamics simulations can provide detailed insights; however, the available classical parameter-based force fields do not adequately capture the coordination transformations and ring statistics of these glasses. First-principles-based molecular dynamics can be beneficial, but they are handicapped in this context due to the viscous nature of supercooled liquids and glasses [2].

To strike a balance between accuracy and speed in molecular simulations, Machine Learning Potentials (MLP) can be very effective. MLP can deliver results that are as accurate as first-principles calculations while being up to 1,000 times faster than Ab Initio Molecular Dynamics. This capability allows us to use MLP for molecular dynamics simulations of borate glasses [3].

The smallest structural unit in $B_2O_3$ glass is a trigonal BO3/2 one. Yet, higher order structures such as the hexagonal B3O3 boroxol units can exist, but their concentration in the glass has been a matter of debate. Raman and NMR spectroscopy suggest that approximately 75% of boron atoms exists in boroxol units [4], while molecular simulations (using either force fields or AIMD) indicate the fraction to be only 30%. This conflict may be resolved by using an accurate force field such as the MLP, along with the adoption of conservative quench rates. Additionally, for $B_2O_3$, there is no crystallization observed at normal pressure; however, crystallization occurs when pressure is applied [5]. Multiple polymorphs too can exist in the P-T phase diagram. The work covers the development of the MLP and explores the network structure of the borate glass and its rich equilibrium phase diagram.

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# Intriguing topological signatures in novel Dumbbell $C_3NX$ (X = C, Si, Ge) and its quasi-1D derivatives

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Two-dimensional carbon nitride materials have garnered significant attention due to their versatile applications in energy harvesting, environmental remediation, and nanoelectronics. Their broad functional scope, coupled with feasible synthesis, has positioned this material family as a compelling research focus, though their fundamental analytical aspects remain underexplored. Recently, leveraging first-principles methodologies, we introduced a family of six distinct $C_3NX$ structures (Jana et al. Journal of Physical Chemistry C 2023) featuring a unique dumbbell-shaped morphology, building upon the synthesized $C_3N$ monolayer (Zhang et al. Nature 2025, Yang et al. Adv. Mater. 2017). In this study, we systematically investigate the non-trivial topological phases of semimetallic dumbbell $C_3NX$ sheets and nanoribbons. The spin–orbit coupling (SOC)-induced gap at the Fermi level, its tunability via an external electric field, the manifestation of band inversion through Berry curvature mapping, and the determination of the $Z_2$ topological index using the Wannier charge center (WCC) method collectively establish the material's topological character. Furthermore, real-space decimation and Green's function techniques are employed to analyze spectral properties and transport characteristics. Remarkably, these quasi-1D systems exhibit intriguing features within the Su-Schrieffer-Heeger (SSH) model, where different twisted phases reveal distinct topological signatures, even in a low atomic mass system such as dumbbell $C_4N$.

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# Machine learning Eigen-values, Eigen-functions and variational parameters

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In this work, we explore the integration of supervised machine learning techniques with quantum mechanics to analyse spherically symmetric potentials. We solve the Schrödinger equation variationally using basis set expansions which can be linear combination of Gaussian or Slater type functions with non zero centres. Initially, our solver was validated against known analytical potentials to ensure its precision and reliability. Subsequently, we applied the solver to a class of spherically symmetric potentials which can also be expressed as linear combinations of Gaussian and Slater type functions. The solver primarily evaluates the energy expectation value using a variational ansatz and then uses Nelder-Mead algorithm to optimise the variational parameters.

Recognizing the strong correlations between the parameters defining these potentials and the resulting wave functions, we employed supervised machine learning algorithms to model and predict eigen-energies, densities and variational parameters. By training our models on the computed data and employing data engineering, we demonstrated that machine learning can effectively capture the underlying physical trends, thereby providing an alternate tool for the analysis and prediction of quantum mechanical properties.

# Mechanistic Insights into the Early Stages of Copper Oxidation: The Role of O-O Repulsion and Substrate-mediated Effects

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Copper-based catalysts play a crucial role in industrial oxidation reactions. It is for this reason that extensive studies have been reported in literature on first-principles simulations of catalytic oxidation processes on copper. While most studies consider copper to be metallic, it is well documented in experimental literature [1,2] that copper can undergo oxidation at ambient conditions forming a passivating copper oxide layer. Given this background, the mechanistic aspects of the onset of copper oxidation need to be understood. Although the oxidation of copper has been thoroughly explored through experimental techniques such as scanning tunnelling microscopy (STM) and low-energy electron diffraction (LEED), the atomistically-resolved copper oxidation mechanism at the flat terraces remains poorly understood. Using density functional theory (DFT) calculations, we show that substrate mediated O-Cu-O adsorbate interactions play a key role. Multiple adsorbed O atom clusters trigger localized surface restructuring, enabling the extraction of Cu atoms from the surface layer, which enables subsurface oxygen diffusion - this constitutes the first step in oxide layer growth. Nudged elastic band (NEB) calculations reveal that subsurface oxide formation can be practically barrierless once such surface arrangements are achieved. These insights provide a foundation for developing a more accurate model for copper catalysis.

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# Material-specific investigations of strongly correlated electron systems through DFT+DMFT

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Strongly correlated electron systems (SCES) have traditionally been studied using effective low-energy many-body Hamiltonians, such as the Hubbard and periodic Anderson models. While these toy models provide valuable insights into phenomena like the Mott metal-insulator transition and heavy fermion physics, they lack material specificity. The emergence of dynamical mean-field theory (DMFT) three decades ago, followed by quantum cluster theories, enabled predictive modeling of SCES when combined with density functional theory (DFT).

In this poster, I will review key advances achieved through the DFT+DMFT approach. I will then present our work on the development and application of a fast and computationally inexpensive, albeit perturbative, impurity solver. This solver operates directly on the real-frequency axis and scales polynomially with the number of orbitals. Finally, I will discuss its potential applications in large-unit-cell systems, including multi-orbital materials, disordered systems, and Moiré lattices [1].

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# Acoustic phonon restricted four-phonon interactions: Impact on lattice thermal conductivity in 2D h-NbN

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Materials with large acoustic-optical (a-o) phonon band gaps exhibit a complex interplay between three-phonon and four-phonon scattering, significantly influencing the lattice thermal conductivity ($\kappa_l$). In this study, we employed first-principles calculations and Boltzmann transport theory to investigate the lattice thermal conductivity of hexagonal niobium nitride (h-NbN) monolayers. Including four-phonon scattering reduced $\kappa_l$ at room temperature by 52% compared to considering three-phonon scattering alone. In monolayers, reflection symmetry restricts flexural phonons in three-phonon processes but not in four-phonon processes[1,2]. Despite the absence of reflection symmetry in h-NbN, four-phonon scattering is as significant as three-phonon scattering due to the large a-o band gap, flexural (ZA) modes, and non-dispersive acoustic band. Notably, the four-phonon interactions are primarily limited to acoustic phonons, unlike the typical interactions between acoustic and optical phonons. Furthermore, under tensile strain, the ZA phonon dispersion transitions from quadratic to linear, impacting $\kappa_l$. Additionally, the electronic transport properties and thermoelectric figure of merit (ZT) under ambient conditions and with strain suggest that h-NbN is a promising semiconductor with low to moderate thermal conductivity and moderate thermoelectric performance at high temperatures.

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D2-035

# Optimising the electronic transport of Cu-based quaternary chalcogenide through first-principles study

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Transition metal-based quaternary chalcogenides have emerged as promising candidates for renewable energy applications, particularly in thermoelectrics (TE). However, their complex structures and low symmetry have posed challenges in achieving high thermoelectric performance, primarily due to low power factors. In this study, we employed first-principles calculations combined with energy- and momentum-dependent relaxation time[1] to explore the TE properties of n- and p-type $Cu_2ZnSiTe_4$, which exhibits favorable electronic transport characteristics in both cases[2]. The presence of the heavy chalcogen 'Te' results in a relatively low bandgap of 0.58 eV due to spin-orbit coupling (SOC), contributing to a high power factor of 3.95 $mWm^{-1}K^{-2}$ (n-type) and 3.06 $mWm^{-1}K^{-2}$ (p-type) at 900 K, along with a favorable electronic band topology. Furthermore, the 'Te' atoms' contribution to the mixing of the optical and acoustic phonon branches results in low lattice thermal conductivity (about 0.7 $mWm^{-1}K^{-1}$) at high temperatures. The compound's TE figure of merit yields promising results, with ZT values of approximately 2.67 (n-type) and 2.11 (p-type) at 900 K. Based on our study, we believe that $Cu_2ZnSiTe_4$ is a viable option for TE applications, and we hope that our findings will inspire experimentalists to explore this compound further.

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# Six-Fold Degenerate Phonons in the $M_3Sn$ (M = V, Nb): A density functional theory calculations

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Topology has become one of the most extensively studied domain of condensed matter physics over the last decade. Its exotic surface phenomena excite researchers to explore novel quantum properties. From topological insulators to semimetals, materials leverage topology for unique electronic properties. With rich applications in spintronics and dissipation less electron transport, electronic topology is now a well-established topic among researchers. Beyond fermionic topology, phononic topological material garnered significant attention owing to their applications in thermoelectric with suppressing the thermal conductivity while maintaining high electrical conductivity and acoustic waveguides with the robust and scattering-free phonon transport [1].

In this work, we explore the presence of sixfold degenerate points (SDPs) in experimentally synthesized $M_3Sn$ (M = V, Nb) compounds. These compounds belong to the $Pm\bar{3}n$ space group (SG-223) which exhibit multifold band degeneracies in their electronic and phononic band structures [2,3]. All calculations were performed using density functional theory as implemented in VASP code. In the electronic band structure of the compounds, we observe the six-fold band degeneracy at the R-point. The structural, mechanical, and thermal stability of these materials was confirmed through elastic constants and AIMD simulations. We observed sixfold degenerate points at the R high-symmetry point of the Brillouin zone in electronic as well as phononic band structures. Additionally, we analyzed the topological surface states associated with these SDPs, confirming their topological nature. The presence of topological phonons facilitates dissipation less phonon transport and has potential applications in thermoelectric and phononic devices. Moreover, we hope our study will inspire experimentalists to investigate this using inelastic x-ray scattering and newly introduced HREELS and STEM-EELS methods.

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# Superconductivity and CDW phases in Ca intercalated bi-Layer silicene

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Superconducting quantum materials are becoming indispensable components for quantum devices, encompassing various applications, ranging from quantum computers to power transmission systems. In this study, we investigate Ca-intercalated bilayer silicene ($Si_2CaSi_2$) as a potential superconductor using first-principles calculations. Our findings reveal that superconductivity in this system is driven by strong electron-phonon coupling, primarily facilitated by the hybridization of Ca-3d and Si-3$p_z$ orbitals. This hybridization forms a distinctive flower-like interlayer Fermi surface at the $\Gamma$ point, which couples effectively with low-energy out-of-plane phonon vibrations enabled by the buckled structure of silicene. We predict a superconducting transition temperature ($T_c$) of 12.5 K [1], placing $Si_2CaSi_2$ among promising covalent superconductors.

Additionally, phonon softening at specific symmetry points and new Fermi surface features indicate a possible CDW phase in this system. The interplay between superconductivity and CDW order suggests that $Si_2CaSi_2$ shares characteristics with other correlated quantum materials, such as dichalcogenides and kagome metals[2], but with distinct underlying mechanisms. These findings establish $Si_2CaSi_2$ as a compelling candidate for tunable superconducting phases in silicon-based quantum materials, offering new opportunities for next-generation electronic and quantum device applications.

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# An Effective Spin Hamiltonian: Renormalization by Vibration

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Quantum Spin Liquids (QSLs) are exotic phases of matter characterized by fractionalized excitations, long-range quantum entanglement, and the absence of conventional magnetic order. Two crucial theoretical frameworks proposed to describe QSL behavior are the *Resonating Valence Bond* (RVB) state conjecture and the *Kitaev model*, both involving spin-$\frac{1}{2}$ degrees of freedom and exhibiting fractionalized excitations. We formulated a Hamiltonian describing spin-vibration coupling between two spin-$\frac{1}{2}$ sites and derived an effective spin Hamiltonian by integrating out vibrational degrees of freedom, capturing the spin-singlet of the RVB state. Now, we are trying to extend this formalism to a 2D honeycomb lattice to compare the effective spin Hamiltonian with the Kitaev Hamiltonian. Through this comparative study, we intend to establish a connection between the RVB state conjecture and the Kitaev model, for a broader understanding of QSLs. Our investigation is expected to provide insights that will guide future developments in realizing these quantum phases using first principles methods, with potential applications in quantum computing, high-temperature superconductivity, spintronics, and other emerging quantum technologies.

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# Intermediate band formation and delocalization in second-row transition metal (Y, Zr, Nb, and Mo) doped $Cu_2O$ for photovoltaic applications: GGA+U study

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Intermediate band photovoltaics aim to exceed the efficiency limits of single-junction solar cells. Luque and Marti demonstrated a theoretical efficiency of 63.1% for a material with an intermediate band within the bandgap. The proposed bandgap for such a material is 1.95 eV. A partially filled intermediate band is preferable, as it reduces non-radiative recombination compared to a completely filled band [1]. Cuprous oxide ($Cu_2O$) is a p-type material with a bandgap of 2.17 eV, close to the value proposed by Luque and Marti, but it has not yet been explored as an intermediate band material.

In this work, yttrium (Y), zirconium (Zr), niobium (Nb), and molybdenum (Mo) were substituted for Cu in $Cu_2O$ at a dopant concentration of 3%, and their electronic and optical properties were studied to assess intermediate band formation. The defect level possessing delocalized behavior spread across the entire crystal is essential for reducing recombination. To determine the nature of this defect intermediate state, the dopant concentration was increased to 6%, and the partial charge density was analyzed. Yttrium substitution introduces an occupied level approximately 1.17 eV below the conduction band minimum. Zr and Nb introduce a doubly degenerate intermediate level in the host material, while Mo creates a triply degenerate manifold [2]. With the exception of yttrium, substituting Zr, Nb, or Mo for Cu in cuprous oxide results in the formation of partially filled intermediate bands within the forbidden region, making these materials suitable for improving photovoltaic efficiency.

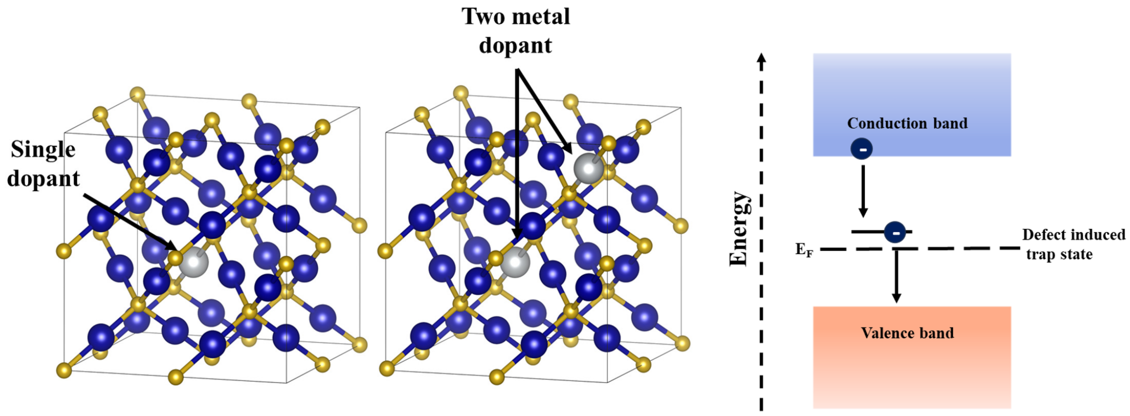

Figure 1: The crystal structures of a transition metal (Y, Zr, Nb, and Mo) doped $Cu_2O$ with two different dopant concentration and schematic representation of trap induced recombination.

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# PAW-DFT-FE: A fast and accurate real-space finite-element based method for large-scale ab-initio material modelling

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Gaining *ab initio* insights into complex materials problems involving nanometers in length-scale and nanoseconds of time-scale demand enormous computational resources owing to the stringent accuracy requirements of density functional theory (DFT) calculations compounded with the cubic scaling complexity of DFT with number of atoms. This study presents for the first time a new approach that utilizes a local real-space formulation discretized with finite-element (FE) basis to enable rapid and scalable electronic structure calculations employing the projector augmented wave (PAW) method. To achieve this, we have developed efficient computational techniques that leverage modern GPU architectures. These techniques incorporate low-rank perturbation of identity, block-diagonal representation of inverse PAW overlap matrix using spectral finite-element basis, a residual based Chebyshev Filtered subspace iteration approach in conjunction with mixed-precision arithmetic and subspace locking strategies to solve the underlying FE discretized PAW generalized eigenproblem. Our framework (PAW-DFT-FE) significantly reduces the degrees of freedom compared to finite-element based norm-conserving pseudopotential calculations while ensuring the necessary chemical accuracy and accommodating generic boundary conditions, thereby enabling faster and accurate large-scale DFT simulations (>50,000 electrons) than possible today. Furthermore, our approach demonstrates a considerable computational advantage over state-of-the-art plane-wave codes for system sizes greater than 5000 electrons. Notably, our framework represents the first real-space PAW approach capable of competing with plane-wave methods in modern GPU architectures. We illustrate the effectiveness of PAW-DFT-FE for addressing large-scale problems involving tens of thousands of electrons, which are beyond the capabilities of existing DFT codes. Finally, leveraging PAW-DFT-FE we demonstrate few case-studies of fast and scalable DFT calculations with generic boundary conditions that push the boundaries of length scales accessible to *ab initio* calculations.

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# Toxic Gas Sensing Mechanisms on 2D TMD Heterostructures: A First-Principles Approach.

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Two-dimensional (2D) transition metal dichalcogenide (TMD) heterostructures have garnered significant attention due to their exceptional structural versatility, tunable electronic properties, high surface sensitivity, and robust adsorption capabilities[1]. In this study, we systematically investigated the adsorption behaviour of hydrogen fluoride (HF) toxic gas on the surface of a $WSe_2$-$MoSe_2$ TMD heterostructure using first-principles calculations within the framework of density functional theory (DFT). The structural stability of the $WSe_2$-$MoSe_2$ heterostructure was rigorously validated through phonon dispersion analysis, confirming the absence of imaginary frequencies and ensuring dynamic stability. The adsorption of HF molecules on the TMD heterostructure was quantitatively assessed by calculating the adsorption energy ($E_{ad}$), which indicates a favourable adsorption interaction, suggesting its implications for sensor sensitivity and recovery behaviour. To elucidate the underlying interaction mechanisms, charge transfer phenomena were examined using Bader charge analysis and electronic potential surface mapping. Electronic structure analysis, including the density of states (DOS), partial density of states (PDOS), and electronic band structure, revealed significant electronic modifications upon HF adsorption, highlighting changes in bandgap and electronic states near the Fermi level. These alterations are indicative of enhanced electronic response upon gas exposure. Additionally, the system demonstrated a suitable recovery time ($\tau$), signifying its potential for real-time sensing applications. Our results provide critical insights into the sensing mechanism, emphasizing selectivity, sensitivity, and rapid response-recovery characteristics. The study underscores the potential of engineered 2D TMD heterostructures as high-performance HF gas sensors, offering promising applications in industrial safety systems and environmental monitoring technologies.

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AAPALI PSI-K 2025

# Symmetry induced novel spin textures in non-centrosymmetric systems

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In non-centrosymmetric systems, the gradient of the electrostatic potential generates a momentum-dependent magnetic field in the rest frame of the electron. This field locks the electron's spin to its momentum, lifting spin degeneracy of the bands and creating complex spin textures in reciprocal space, primarily enforced by symmetries. Depending on the symmetry, the system may exhibit Rashba, Dresselhaus, Weyl (radial), persistent, or other complex spin textures. Using first-principles electronic structure calculations based on density functional theory (DFT) and supplemented with ***k.p*** low-energy model Hamiltonian, we have identified diverse spin textures in nonpolar half-Heusler alloys [1] and nonpolar chiral systems [2]. Our analysis highlights the key role of little group symmetries at specific k-points and the orbital character of the associated bands in defining the spin textures [3]. We observe linear Dresselhaus effect, Rashba effect with both linear and nonlinear terms, as well as nontrivial Zeeman spin splitting and vanishing spin polarization in nonpolar half-Heusler alloys. We show in nonpolar chiral systems, bands with specific orbital characters near a high-symmetry point with a $D_2$ little group may give rise to a single spin-dependent term in the low-energy ***k·p*** Hamiltonian, resulting in a persistent spin texture. These diverse spin textures offer promising opportunities for spintronic and spin-valleytronic applications.

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# Exploring and Elucidating the $CO_2$ Reduction Mechanisms on the Surface of Two-Dimensional Nitrogen-Vacancy ($V_N$) Hexagonal Boron Nitride

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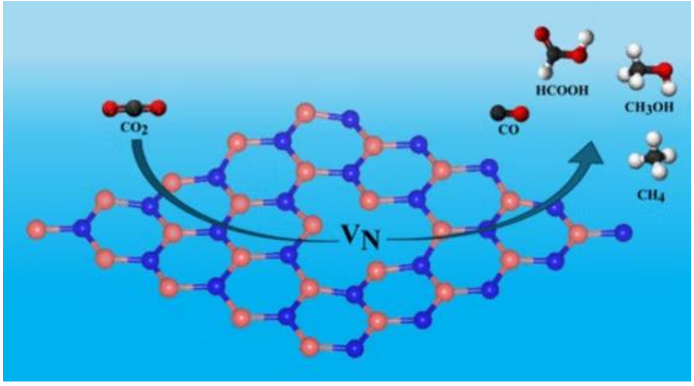

The conversion of waste $CO_2$ gas into valuable products and fuels through an electrocatalytic $CO_2$ reduction reaction ($CO_2RR$) is a promising approach. The sluggish kinetics of the $CO_2RR$ require developing novel strategies for the electrocatalyst design. 2D materials emerge as promising candidates for the $CO_2RR$ due to their distinctive electronic and structural properties. We follow the first principles-based DFT-D method to examine the electrocatalytic competencies of the defective two-dimensional boron nitride monolayer (d-BN) material toward the $CO_2RR$.[1] Introducing a particular defect with nitrogen vacancies in 2D single-layer pristine hexagonal boron nitride ($V_N$_d-BN) can efficiently activate the $CO_2$ molecules for hydrogenation by reducing the electronic band gap of pristine hBN from 6.23 to 3.0 eV.[1,2] Therefore, the $V_N$_d-BN material can act as a large band gap semiconductor. Our findings demonstrate that the defective regions in 2D monolayer $V_N$_d-BN serve as active sites (boron) for both the adsorption and activation of $CO_2$ molecules. The subsequent hydrogenation steps occur sequentially once the $CO_2$ molecule is adsorbed on the catalytic surface. Our results indicate that the OCHO* path is the most favorable for $CH_4$ production. Hence, the 2D monolayer $V_N$_d-BN material holds great promise as a cost-effective catalyst for the $CO_2RR$ and presents a viable alternative to expensive platinum (Pt) catalysts.

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D2-044

# TITLE: Computational study of oxide-based spinel material for the application as cathode in Energy storage

Abstract:

Spinel based cathode materials for single cells in battery systems can operate over 4.5 V vs. Li/$Li^+$ using reversible extraction of lithium from them. In these systems $Li^+$ fully occupies tetrahedral sites and can, therefore, move through the well-known tetrahedral site-empty octahedral site pathway; Transition Metals (TM) occupies octahedral sites, and exhibits mixed-valence states, so that fast electronic transport is expected to occur between neighboring $TM^{3+}$/$TM^{4+}$. In all the spinels most, studied structure is $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO), in this work we've replaced Mn in LNMO with Cr, making $LiNi_{0.5}Cr_{1.5}$O4 (LNCrO) having same oxidation state as Mn. In LNCrO the electrochemical process is attributable to $Ni^{2+/4+}$. In the family of spinel cathodes there are many candidates for high voltage category among them LNCrO will have highest voltage of 5.02 V, this will be very much useful in next-generation lithium batteries.

# Trends of magnetic interactions in transition-metal trilayers under electric-fields

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Magnetic skyrmions are topologically protected swirling spin structures which are proposed as the building blocks of next-generation spintronic devices due to their nanoscale size and robust stability arising from the topological protection. Traditionally, skyrmions are manipulated via electric current which generates heat and destabilizes the skyrmionic bits. The electric-field driven control offers an elegant way to manipulate the skyrmions while avoiding the heating problem. Inspite of its potential, theoretical and experimental studies on the electric-field assisted creation and annihilation of skyrmions remain limited and a comprehensive investigation is required.

Using first-principles density functional theory (DFT), we have performed a systematic study to understand the effect of electric-field on magnetic interactions, including the higher-order exchange interactions, in various 4d/3d/5d unsupported trilayers. We observed a linear variation of the interaction constants with the applied electric-field and explained this linear behavior from the underlying electronic structures. This study allows us to identify promising unsupported trilayers that, when placed on a heavy metal surface, can serve as an ideal platform to realize isolated skyrmions.

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# Effect of higher-order processes on the first-principles driven phonon thermal transport in $ThO_2$

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$ThO_2$ is a promising nuclear fuel material that can potentially replace the conventionally used uranium-based fuels in nuclear reactors. First-principles driven thermal energy transport studies in $ThO_2$ have received much attention, whereas most previous studies were limited to lower-order thermal transport theory [1]. As such, in this work, the effect of inclusion of higher-order thermal transport physics (viz. temperature-dependent interatomic force constants, phonon renormalization, and four-phonon scattering) on the computation of phonon frequencies and lattice thermal conductivity of $ThO_2$ is explored, employing LDA, PBE, and PBEsol exchange-correlation functionals [2].

It is found that, upon renormalization, the frequencies are stiffened for the optical phonon modes, whereas the acoustic modes remain unchanged. The thermal conductivity computed using LDA and PBEsol functionals is within 5% of the experimentally measured values at 300 K, whereas that obtained using PBE functional results in an underprediction of 25%. The temperature-dependent force constants and renormalized phonon frequencies significantly affect the computed lattice thermal conductivity at higher temperatures (40% difference at 1000 K), whereas four-phonon processes have minimal effects (only 10% at 1000 K). [2].

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# Understanding the local electronic properties of ZrTe2 and ZrSeTe

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TMDCs (Transition Metal DiChalcogenides) have garnered significant attention due to their exceptional optical, electronic, & topological properties. Among them, zirconium ditelluride (ZrTe2) stands out for its semimetallic nature, strong spin-orbit coupling, and potential topological states. However, its susceptibility to oxidation presents both challenges and opportunities for device applications. This thesis provides a detailed investigation into the oxidation dynamics of ZrTe2, alongside its structural, surface, and electronic properties. A comprehensive study of ZrTe2 oxidation was conducted using Atomic force microscopy (AFM), High-Resolution Transmission electron microscopy (HRTEM), and Raman spectroscopy. These analyses revealed that oxidation predominantly initiates at the edges and surfaces, leading to the formation of amorphous ZrO2 and tellurium aggregation. This transformation significantly alters the electronic properties of ZrTe2, impacting its potential applications in nanoelectronics. Given that ZrO2 is a high-dielectric ($\kappa = 25$) material widely used in MOSFETs, understanding this oxidation process is critical for the controlled integration of ZrTe2 into electronic devices. Additionally, the rapid and spontaneous oxidation behavior suggests potential use in low-cost oxygen sensors. To complement these findings, STM and spectroscopy (STS) were employed to investigate the atomic-scale surface and electronic properties of ZrTe2 and its derivative ZrSeTe. Ultra-high vacuum low-temperature STM (UHV LT-STM) measurements provided insights into the surface, the density of states, and electronic band structures. The experimental observations were compared with theoretical predictions from density functional theory (DFT). These insights offer a pathway for the controlled utilization of ZrTe2 in future electronic, sensing, and quantum device applications.

D2-048

# Brightening of momentum dark exciton via tensile strain induced valley convergence in $WS_2$ monolayer

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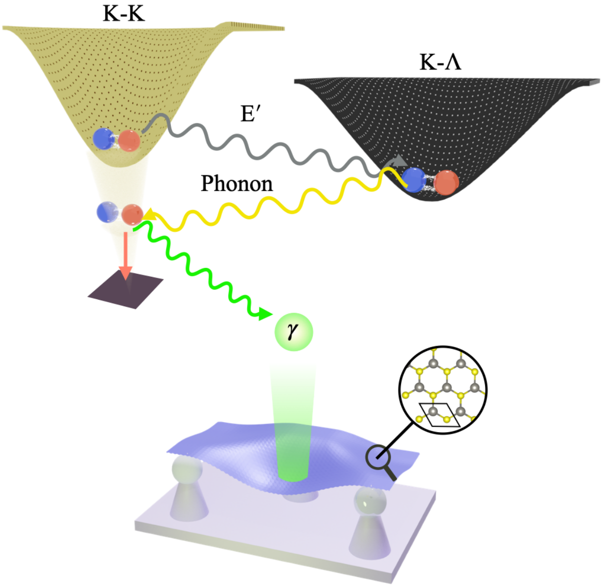
Transition-metal dichalcogenides (TMDs) host tightly bound electron-hole pairs—excitons—which can be either optically bright or dark based on spin and momentum selection rules. In tungsten-based TMDs, a momentum-forbidden dark exciton is the energy ground state, and therefore, it strongly affects the emission properties. In this work, we brighten the momentum-forbidden dark exciton by placing monolayer tungsten disulfide on top of nanotextured substrates, which imparts tensile strain, modifying its electronic band structure. This enables phonon-assisted exciton scattering between momentum valleys, thereby brightening momentum-forbidden dark excitons. In addition to offering a tuning knob for light-matter interactions in two-dimensional materials, our results pave the way for designing ultrasensitive strain-sensing devices based on TMDs.



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# DAY 2 : 20 MAY 2025

# Finite-element methods for noncollinear magnetism and spin-orbit coupling in real-space pseudopotential density functional theory

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We present an efficient finite-element approach[1] for large-scale real-space pseudopotentials density functional theory (DFT) calculations incorporating noncollinear magnetism and spin-orbit coupling effects. The approach, implemented within the open-source DFT-FE[2] computational framework, fills a significant gap in real-space DFT calculations using finite element basis sets, which offer several advantages over traditional DFT basis sets. In particular, we first derive the finite-element (FE) discretized governing equations involving two-component spinors, leveraging a local reformulation of DFT electrostatics. To evaluate the widely used GGA exchange-correlation potentials in these governing equations under the locally collinear approximation, we devise a numerical strategy that avoids the computation of ill-defined gradients of magnetization direction near zero magnetization. Additionally, we utilize an efficient self-consistent field iteration approach based on Chebyshev filtered subspace iteration procedure exploiting the sparsity of local and non-local parts of FE discretized Hamiltonian to solve the underlying nonlinear eigenvalue problem based on a two-grid strategy. Further, we propose using a generalized functional within the framework of noncollinear magnetism and spin-orbit coupling with a stationary point at the minima of the Kohn-Sham DFT energy functional to develop a unified framework for computing atomic forces and periodic unit-cell stresses. Validation against plane-wave implementations shows excellent agreement in ground-state energetics, vertical ionization potentials, magnetic anisotropy energies, band structures, and spin textures. The proposed method achieves significant speed ups for semi-periodic and periodic systems with 5000-7000 electrons in terms of minimum wall times compared to widely used plane-wave implementations on CPUs in addition to exhibiting notable computational advantage on GPUs for material systems with as many as 20,000 electrons. Furthermore, extensions of the afforementioned computational framework within the projector augmented wave formalism[3] to further reduce the computational cost will also be discussed, highlighting the potential applications to studying TMD bilayers with low twist angles incorporating spin-orbit coupling effects. The proposed approach offers a fast, scalable, and systematically convergent framework for large-scale DFT calculations accounting for noncollinear magnetism and spin-orbit coupling, enabling more complex material simulations and extending the range of ab-initio studies.

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# Phonon dynamics in ultrahigh thermal conductivity materials: accelerating the solution of first principles based Boltzmann transport equation

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Thermal management of rapidly miniaturizing electronic devices requires handling high heat flux at micro- to nanometer lengthscales and nano- to picosecond timescales. These devices are mainly constituted of semiconductor materials, where heat is predominantly carried by the quanta of lattice vibrations called phonons. Further, the physics of thermal transport changes as we change the lengthscale of the crystal. In the bulk crystals, thermal transport is a diffusion phenomenon governed by the Fourier's law, involving frequent phonon scattering events; however, at the nanometer lengthscales, the thermal transport takes place in a non-Fourier, nearly scatter-free manner in most materials called the ballistic transport regime.

Besides these transport regimes, there exists another exciting non-Fourier thermal transport regime called the hydrodynamic second sound. In this regime, heat flows through a collective motion of phonons, which is much faster than the usual diffusion regime, and therefore, this regime is important from the thermal management point of view. However, the hydrodynamic second sound is a rare phenomenon as it occurs in a very narrow window of experimental conditions and is observed only in a handful of materials like sodium fluoride [1] and graphite [2] till date, due to the lack of a fast predictive computational framework that can accelerate the materials discovery for phonon hydrodynamics.

This predictive computational framework involves solving the governing equation for phonon transport - the linearized Peierls-Boltzmann equation (LPBE). However, solving LPBE is computationally expensive - it requires years of computational time and terabytes of memory because of the huge and dense collision matrix - whose element represents the strength of the phonon scatterings [3]. Here, we present a data-driven method to accelerate the solution of the LPBE by implementing a low-rank representation of the phonon collision matrix without compromising accuracy [4]. Our findings indicate that only the eigenvectors of the collision matrix associated with small eigenvalues play a significant role in thermal transport, thus allowing us to reduce the problem's dimensionality by excluding the less important eigenvectors without affecting the solution. This technique enables efficient exploration of both Fourier and non-Fourier transport regimes, particularly at low temperatures, where the required dimension of the problem to achieve convergence of the solution is prohibitively large. We demonstrate the effectiveness of this approach by investigating the above-discussed thermal transport regimes at 100 K in diamond, the material with the highest known thermal conductivity.

This work is supported by the Prime Minister's Research Fellowship (02-01036), the Science and Engineering Research Board's Core Research Grant No. CRG/2020/006166 and the Mathematical Research Impact Centric Support Grant No. MTR/2022/001043.

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# High-throughput and data-driven search for stable optoelectronic $AMSe_3$ materials

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The rapid advancement in emerging optoelectronic technologies demand highly efficient, affordable, and ecofriendly materials. In this context, ternary chalcogenides especially ternary selenides show early promise as a material class due to their stability and remarkable electronic, optical, and transport properties. In this work, we integrate first-principles-based high-throughput computations with machine learning (ML) techniques to predict the thermodynamic stability and optoelectronic properties of 920 valency-satisfied selenide compounds. Through investigating polymorphism, our study reveals the edge-sharing orthorhombic *Pnma* phase ($NH_4CdCl_3$-type) as the most stable structure for most ternary selenides. High-fidelity supervised ML models are trained and tested to accelerate stability and band gap predictions. These data-driven models pin down the most influential features that dominantly control key material characteristics. The multistep high-throughput computations identify the ternary selenides with optimal direct band gaps, light carrier masses, and strong optical absorption edges. The extensive materials screening considering phase stability, toxicity, and defect tolerance, finally identifies the seven most suitable candidates for photovoltaic applications. Two of these final compounds, $SrZrSe_3$ and $SrHfSe_3$, have already been synthesized in single-phase form, with the latter showing an optically suitable band gap, aligning well with our findings. The non-adiabatic molecular dynamics reveal sufficiently long photoexcited charge carrier lifetimes (on the order of nanoseconds) in some of these selected selenide materials, indicating their exciting characteristics. Overall, our study demonstrates a robust in-silico framework for screening large material datasets, offering a scalable approach for discovering promising photoactive materials.

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# Data-Driven Design of Electroactive Spacer Molecules to Tune Charge Carrier Dynamics in Layered Halide Perovskite Heterostructures

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Crafting rational heterojunctions with nanostructured materials is instrumental in fostering effective interfacial charge separation and transport for optoelectronics. Layered halide perovskites (LHPs) that form heterojunctions between organic spacer molecules and inorganic metal halide layers exhibit tunable photophysics owing to their customizable band alignment. However, controlling photogenerated carrier dynamics by strategically designing layered perovskite heterojunctions remains largely unexplored. We combine a data-driven approach with time-domain density functional theory (TD-DFT) and non-adiabatic molecular dynamics (NAMD) to screen and select electronically active spacer dications (A′) that introduce a type-II heterojunction in the lead iodide-based Dion–Jacobson phase LHPs. The composition–structure–electronic property correlations reveal that the number of nitrogens in aromatic heterocycles is the key factor in designing electron-accepting spacers in these perovskites. The detailed atomistic simulations validate the design strategy further by modeling (A′)$PbI_4$ perovskites, which incorporate three different screened electroactive A′ spacers. The computed excited charge carrier dynamics illustrate the phonon-mediated ultrafast interfacial electron transfer from the inorganic conduction band edge to the lower-lying unoccupied orbitals of spacers, exhibiting photoluminescence quenching in these (A′)$PbI_4$ perovskites. The spatially separated electrons and holes at the type-II heterojunction interface prolong the excited charge carrier lifetime, boosting the carrier transport and exciton dynamics. Our work illustrates a robust in silico approach for designing LHPs with exciting optoelectronic properties originating from their fine-tuned heterojunctions.[1]

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# Structural Evolution of $FeCl_2$ under High Pressure: A First-Principles Investigation

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The observed terrestrial depletion of heavy halogens by roughly an order of magnitude relative to their expected abundances based on volatility suggests a sequestration mechanism, possibly involving the Earth's core. The strong chemical affinity between Fe and these halogens may form iron halides within the core, potentially affecting its physical properties and long-term evolution [1]. In this study, we investigate the high-pressure behaviour of ferrous iron chloride ($FeCl_2$) using *first-principles* based calculations. Our results confirm that the $R\bar{3}m$ phase of $FeCl_2$ stabilizes under ambient pressure conditions. The first phase transition is observed at 0.4 GPa, where the structure transforms into the $P\bar{3}m1$ phase. This structure remains thermodynamically stable up to 14 GPa, beyond which it transits to the orthorhombic *Pnnm* phase, which is stable up to 28.5 GPa. At higher pressure, another structural phase transition is observed, leading to the formation of the cubic $Pa\,\bar{3}$ phase. The study focuses on the newly predicted intermediate phase *Pnnm* of $FeCl_2$ which was not correctly identified in experiments [2,3]. These findings enhance our understanding of chlorine transport and storage from the middle-lower mantle to the outer core.

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# Electrical and Optical Properties of BSe and Twisted Bi-Layer Graphene Heterostructure using DFT First principles Calculation

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Bilayer graphene systems tend to possess interesting characteritics when we introduce inter-layer twisting such as exotic topological[1] and correlational effects[2] are observed at magic angles and close to it. We also observe change in thermal conductivity[3] , change in optical properties[4][5] etc. We have made an attempt to modify the electrical and optical properties for twisted graphene by adding a layer of BSe on top of it. In this work we have focussed on electrical and optical properties of such a heterostructure consisting of a layer of BSe on top of twisted bi-layer graphene with a Relative Rotation Angle(RRA) of $21.79°$ between the two graphene layers using Density Functional Theory. The lattice mismatch for the unit cell is within appropriate limit. The structural stability has been verified through AIMD simulations at room temperature with timestep of 2 fs for a total of 5 ps. The band-gap for the Heterostructure is obtained as 2 meV at the high symmetry point in the Brillouin Zone. The optical properties reveal the material to be quite active in the UV region with a peak in absorption coefficient obtained at 6.6 eV hence the material could be for UV filtering applications as well as for UV based devices in it's range of activity.

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# Metal Oxide-Based Hydrogen Sensors Beyond the Adsorbed-Oxygen Model: A Pt–ZnO Case Study

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## Abstract

Conventional gas sensor research has long centered on the adsorbed-oxygen pathway as the primary detection mechanism in metal oxide-based sensors. However, recent experiments demonstrate that lattice oxygen removal can also drive sensing [1, 2]. Here, we integrate experimental findings with density functional theory (DFT) calculations on Pt–ZnO, examining eight possible surface reactions involving O, $O_2$, H, $H_2$, OH, and $H_2O$. We analyze the reaction energetics of various sensing pathways, including adsorbed oxygen, lattice oxygen, OH formation, and OH auto-reduction, on the polar (002) plane. Our computational insights confirm the experimental results, highlighting the role of Pt in enhancing hydrogen sensitivity by activating multiple reaction pathways. Additionally, we establish the thermodynamic feasibility of the lattice oxygen pathway, identifying the O-terminated surface as the most active for $H_2$ sensing. This systematic study, combining robust experimental design with first-principles modeling beyond traditional binding energy and DOS calculations, provides a deeper understanding of reaction thermodynamics in metal oxide sensors. Such an approach could accelerate the design of next-generation metal oxide based sensors.

**Keywords:** Hydrogen sensing, Zinc oxide, Pt decoration, Lattice oxygen, Adsorbed oxygen, Metal oxides.

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# Inverse design of cathode materials for metal-ion batteries using Variational Auto-Encoder neural network architecture

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The demand for lithium-ion batteries has grown in recent years, driven by the drastic growth of the electric vehicle (EV) industry and portable electronics. However, lithium's limited abundance and high cost have given rise to a shift toward sodium-ion batteries as a more sustainable alternative. Regardless of the chemistry, discovering novel electrode materials, particularly cathode materials with high energy density and extended lifecycle, remains a significant challenge. Although first principles (ab initio) calculations are widely employed for accelerating materials discovery, their high computational cost and time requirements make them inefficient for large-scale exploration. Deep neural network models offer a promising alternative in this context, combining predictive accuracy with efficiency. Recognizing their potential, we developed a pipeline for the inverse design of cathode materials for Li- and Na-ion batteries using Variational Auto-encoders (VAEs) [1] coupled with Density Functional Theory (DFT).

Our VAE model, trained on a diverse dataset of cathode materials, generates potential candidates by sampling a property-conditioned latent space. These candidates exhibit high accuracy in structure, composition, and properties. The predictions predominantly include layered transition metal oxides of lithium and sodium featuring one or more transition metals. The designed compounds were structurally relaxed through spin-polarized GGA+U [2] DFT calculations with appropriate Hubbard 'U' values for transition metals. Additionally, Density of States (DOS) calculations validated the band gaps of the optimized structures, yielding promising results for future applications.

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# $CO_2$ Hydrogenation on Orthorhombic Molybdenum Carbide: A Computational Study using DFT and ReaxFF

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The significant rise in anthropogenic carbon dioxide ($CO_2$) emissions presents a critical environmental challenge, necessitating the development of efficient mitigation strategies. One promising approach is the catalytic conversion of $CO_2$ into value-added chemicals and fuels. Among various candidates, molybdenum carbide ($Mo_2C$) has emerged as a potential catalyst for $CO_2$ conversion due to its tunable microstructure, ordered morphology, and favourable catalytic properties. A key advantage of $Mo_2C$ is its resistance to carbon-deposited deactivation, a common issue in $CO_2$ conversion processes [1]. However, a detailed understanding of its catalytic activity, product selectivity, and long-term stability remains limited, owing to the complexity of competing reaction pathways and susceptibility to surface oxidation [2]. Density functional theory (DFT) is a powerful tool for studying heterogeneous reaction mechanisms at the atomic level, enabling detailed insights into adsorption configurations, reaction energetics, and transition states.

In this study, we employed DFT calculations to investigate the reaction network of $CO_2$ hydrogenation to methane on $Mo_2C$ (100), considering both Mo-terminated (Mo-$Mo_2C$) and C-terminated (C-$Mo_2C$) surfaces. Our DFT results reveal that $CO_2$ adsorbs in a bent configuration on both terminations, with binding energies of -60 kJ/mol on C-$Mo_2C$ and -137 kJ/mol on Mo-$Mo_2C$. $CO_2$ readily dissociates into CO* and O* on both surfaces, with low activation barriers of 20 kJ/mol on Mo-$Mo_2C$ and 57 kJ/mol on C-$Mo_2C$, indicating that CO formation is favoured over direct hydrogenation during $CO_2$ activation step. Subsequent CO activation proceeds via hydrogenation of CO* to HCO* formation, with an activation barrier of 173 kJ/mol and 111 kJ/mol on C- and Mo-terminated surfaces, respectively. However, further hydrogenation is kinetically limited on C–$Mo_2C$, particularly the CH* to $CH_2$* step, which requires 254 kJ/mol. In contrast, Mo–$Mo_2C$ exhibits significantly lower barriers for $CH_x$ hydrogenation (96 kJ/mol for CH* to $CH_2$*), highlighting its superior catalytic activity. These results align with experimental findings that identify Mo-rich surfaces as more active for $CO_2$ hydrogenation [3]. While DFT provides atomistic insights into the reaction mechanism and energetics, it does not capture temperature-dependent dynamics or surface restructuring under operating conditions. To bridge this gap, we developed a ReaxFF potential for the Mo/C/H/O system, trained against quantum mechanical data, enabling large-scale, reactive molecular dynamics simulations under realistic conditions. By integrating static DFT calculations with ReaxFF simulations, this work aims to establish a comprehensive mechanistic framework for $CO_2$ hydrogenation on $Mo_2C$, ultimately guiding the rational design of advanced $Mo_2C$-based catalysts with improved activity, selectivity, and long-term stability.

Key words: $CO_2$ reduction, Reax-FF.

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# Substituted Thiocoumarins Under Solvation and Confinement: A First-Principles Approach to Photophysics and Photochemistry

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Coumarins are utilized as probes to monitor solvation dynamics, with the excited singlet state acting as their reactive state. In contrast, their sulfur counterparts, 2-thioxo-coumarins (TCs), achieve nearly perfect quantum yields for intersystem crossing (ISC) from the excited singlet state to the triplet state. Notably, one of the key findings is that TCs which are otherwise not soluble in water, become soluble in octa acid (OA) capsule, which is a well-known synthetic host that forms capsular assemblies with guest molecules. Under such environment TCs show many intriguing observations such as room temperature phosphorescence (RTP).[1] This indicates efficient ISC and a more prominent role of spin-orbit coupling (SOC).[2] The higher ISC also suggests that the phototriggering mechanisms of TCs are likely different from those of their oxygen analogues. Despite the non-polar environment within the OA capsules, water solubilized TCs can undergo heterolytic photocleavage. In this case while the influence of a nucleophile (water) has been acknowledged, the exact role of it in the overall mechanism is still not very clear.

As a first step to understand all these interesting observations, we employ time-dependent density functional theory (TD-DFT) calculations to understand the nature of states of our molecules of interest and to also identify the stable intermediates that would arise upon photocleavage of such systems.[3]

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# Rare-earth halide based two-dimensional multiferroics

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The coexistence of multiple physical properties in a single material is a way to keep the advancements in the field of electronics going. Multiferroics are such materials where there exist two or more seemingly mutually exclusive ferroic orders simultaneously. We predict, using first principle calculations, a 2D rare-earth halide-based triferroic that showcases three ferroic orders viz. ferroelectricity (FE), ferromagnetism (FM) and ferroelasticity (FA). The ferroelectricity and ferroelasticity are coupled, leading to possible external control of polarization via external strain. The mechanism responsible for FE and FA is found to be bond-centered charge ordering (BCO)[1].

Other such apparently contraindicative properties are conductivity and polarity, as the existence of conduction electrons is expected to screen the long-range Coulomb interactions required for spontaneous polarization or polarity in a material. Using first-principle calculations, we have shown the possibility for the existence of such a polar metal in a Gd-based halide. We also suggest a way to make the ferroic orders in the material coupled so as to make the control of the spin orientation using external forces possible[2].

After charge and spin, another important degree of freedom is valley. The materials having a spontaneous valley polarization are called ferrovalley materials. Using first-principle calculations, we propose a rare-earth-based ferromagnetic-ferrovalley-ferroelectric material where the valley degree of freedom is decided by the direction of electric polarization, i.e., the ferroelectricity and ferrovalleytricity are coupled. Thus, the physical properties associated with the valley degree of freedom, i.e., the Berry phase effect and orbital magnetic moment, can be controlled by an external electric field. This is unlike the conventional ferrovalley materials, where an external magnetic field is required to switch the valley polarization.

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# A theoretical insight on $H_2$ adsorption over two-dimensional materials

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Two theoretical methods: van der Waals density functional theory (vdW-DFT) and kinetic Monte Carlo (kMC) simulation have been combined to understand the adsorption and evolution of $H_2$ molecules over different monolayer materials, such as hexagonal ZnO [1], BN [2, 3], $SnS_2$ and $GeO_2$, in both pristine and defect-induced form. The repulsive lateral interaction going on between two $H_2$ molecules determine the maximum number of molecules that can be packed inside one monolayer unit cell. The output of the vdW-DFT calculations have been fed to the kMC code to study the adsorption, desorption, diffusion of gas molecules along with the overall surface coverage at different temperature and pressure. Our calculations indicate that, for BN substrate, the gravimetric density of $H_2$ molecules has the highest value of 7.5 weight percentage (wt.%) which surpasses the prescribed value of 6.0 wt.% by US Department of Energy for practical fuel cell applications of a host material. Moreover, it has been shown that the increase in layer number (that is, moving towards bilayer/ trilayer from monolayer) or, the formation of a bilayer heterostructure does not have much impact on the $H_2$ adsorption energy, since the adsorption is mainly dependent on the atoms present at the surface.

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# Comparative Study of Photocatalytic Water Splitting and $CO_2$ Conversion Over Infrared and Visible Active Janus MXenes

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Water splitting reactions through photocatalysis is an efficient and sustainable technique for the generation of green energy. The photocatalyst's ability to effect simultaneous generation of hydrogen and oxygen, along with efficiency in utilisation of charged carriers, conversion of solar energy to hydrogen, fast migration, and low recombination rates of carriers, are the parameters to decide its suitability in water splitting. In literature, comprehensive calculation and analysis of all these performance parameters for a potential photocatalyst are rare. In this work, we have performed first-principles-based computations to find new efficient photocatalysts from the family of Janus MXenes and assessed their performance parameters. Strain engineering has been invoked in search of new materials. Out of 14 studied materials, We find that IR active $Zr_2COS$, $Hf_2COS$ at equilibrium and low strain along with 6% strained $Hf_2COSe$ have absorbance, carrier utilisation, carrier separation, exciton binding, and solar-to-hydrogen conversion efficiency that are far superior than the well known 2D photocatalysts for water splitting reactions. Among the visible-active photocatalysts, $Sc_2COS$ cannot meet the benchmark of 10% solar-to-hydrogen conversion efficiency while the performance parameters of $ZrHfCO_2$ are, at best, moderate.

We also investigated the photocatalytic $CO_2$ reduction reaction ($CO_2$RR) on these MXenes. Band edge alignment analysis identified $Sc_2COS$ and $ZrHfCO_2$ as naturally suitable for $CO_2$RR, while $Zr_2COS$, $Hf_2COS$, and $Hf_2COSe$ required strain tuning. High H-coverage was found to favor $CO_2$RR over hydrogen evolution (HER). Among the MXenes, $ZrHfCO_2$ was more HER-active, while $Sc_2COSe$ exhibited the highest $CH_4$ production efficiency. Free energy calculations revealed that $CO_2$ converts to $CH_4$ via -OCHO and -HCOOH intermediates, with conditions favoring $CH_4$ desorption near ambient temperature and pressure. $Sc_2COSe$, particularly at pH 14, demonstrated superior $CO_2$RR selectivity and performance under both dark and illuminated conditions. Other MXenes required additional external potential, making them suitable for photoelectrocatalysis.This comprehensive investigation highlights $Sc_2COSe$ as a standout material for $CO_2$RR under diverse conditions, with the potential for practical applications in $CO_2$ conversion technologies.

This study not only provides a deeper understanding of the mechanisms driving $CO_2$ reduction reactions ($CO_2$RR) in Janus MXenes such as $M_2COT$(M=Sc, Zr, Hf; T=S/Se) and $ZrHfCO_2$ but also offers valuable insights for designing MXene-based photocatalysts with enhanced activity and selectivity. To our knowledge, this is the first comprehensive work demonstrating the utility of Janus MXenes as catalysts in photo-assisted water-splitting and $CO_2$ reduction reactions.

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# Electric field induced Rashba state in topological layered intermetallics

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Emergence of composite phenomena in a quantum material is always facinating from the prespective of the interplay of different degrees of freedom. In the same line of thought, we have investigated using DFT based electronic structure method, a newly synthesized layered ABX type intermetallic compound where A=K, B=Cd, X=Bi, in contrast to the previously reported same class of structure KZnBi. Going from Zn to Cd, hexagonal to tetragonal structural change can be understood from the radius of B site elements. Electronic structure calculation reveals that in absence of spin orbit coupling (SOC) gapless metallic band structure that has 2-types of crossing: $C_{2v}$ symmetry protected band crossing close to the Fermi energy and non-symmorphic symmetry protected 4 fold-Dirac crossing. Incorporation of SOC leads to open up a gap ~ 17-146 meV close to Fermi energy and produced electron-hole-like pockets, whereas non-symmorphic symmetry enforced gapless-crossing still remain gapless close to the energy ~0.7eV below Fermi level. Topological study concludes the compound to be a strong topological $Z_2$ semimetal hosting linear crossed surface state. We have extended our inquisition with the 2D monolayer of the easily cleavable layered structure. The 2D structure is also metallic in absence of SOC similar to that of bulk 3D, but having completely gapped bands with SOC throughout the brillouin zone around the Fermi level. The 2D monolayer is completely gapped in contrast to that of bulk because there is an enhancement of anisotropy in going from bulk 3D to monolayer 2D structure, which is magnified more in presence of SOC along the direction of dimensional confinement. The 2D monolayer structure also retains its non-triviality hosting non-trivial edge state. On application of electric field in the 2D monolayer, Rashba states appear and there is an enhancement of Rashba coefficient with increasing applied electric field. In topological perspect, there is a transition from topologically non-trivial insulating Rashba state below the critical field to nontrivial metallic state without Rashba above the critical field.

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# Mechanistic Insights and Design Principles for Efficient $NH_3$-SCR Catalysts: A DFT Study on $Fe_2O_3$ and NiO Systems

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The selective catalytic reduction (SCR) of NO using $NH_3$ is a crucial approach for managing $NO_x$ emissions. This study utilises density functional theory (DFT) simulations to explore $Fe_2O_3$- and NiO-based catalysts, focusing on pristine, doped, and partially reduced surfaces to reveal structure-activity relationships. In the case of $Fe_2O_3$, the oxidation of NO to $NO_2$, followed by $NH_2$ coupling, led to the formation of $NH_2NO_2$. However, the high N–O bond cleavage barrier in the NHNO intermediate (2.58 eV) limited catalytic activity and favoured the production of $N_2O$. Introducing Mo doping altered the reaction pathway by inhibiting NO oxidation and facilitating efficient N–O bond cleavage, lowering the barrier to 1.44 eV and selectively generating $N_2$. A calculated turnover frequency (TOF) of $1.9\times10^{-3}$ $s^{-1}$ corresponded well with experimental observations, thus validating the proposed mechanism. The presence of Mo improved surface reducibility and Lewis acidity, thereby optimising catalytic performance. On the NiO catalyst, stoichiometric surfaces depended on NO-assisted $O_2$ dissociation, leading to high activation barriers (up to 1.95 eV) and resulting in $N_2O$ formation. In contrast, partially reduced NiO surfaces allowed for direct $O_2$ activation, significantly reducing barriers to 0.89 eV and enabling selective $N_2$ formation. Ru doping further enhanced reducibility and decreased the N–O cleavage barrier. These findings offer valuable insights for developing efficient SCR catalysts to comply with stringent $NO_x$ emission standards in lean combustion environments.

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# Unveiling heterogeneously active HER sites by surface reconstruction via termination groups in MXenes

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The evaluation of catalytic activity of any catalyst highly depends on the description of its surface and details of the active sites. Catalytic activity varies depending on the various parameters like pH, applied potential, temperature, and electrolyte due to changes in catalyst surface by the process of surface reconstruction. The present work explores materials from the 2D class known as MXene[1] for their catalytic activity using density functional theory[2]. This work delves deeper to understand the complex adsorbate-adsorbate interaction between surface termination groups over the MXene surface. We have systematically determined the equilibrium coverage of the termination group over the MXene surface using Monte Carlo simulation and the cluster expansion method by considering various adsorbate-adsorbate interactions of the termination group. The catalytic activity was probed by hydrogen evolution reaction (HER) over pristine, fully terminated surface and surface with equilibrium coverage of termination groups. The heterogeneously active sites were identified over the MXene surface with excellent HER activity as compared to its pristine or fully surface terminated form. It is observed that the type of surface termination groups, their distribution over MXene surface, and their equilibrium coverage are key factors that control the HER activity of MXene.

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This work investigates the magnetic properties of an intrinsically disordered hyperhoneycomb lattice compound $Na_{0.5}Yb_{0.5}WO_4$. Low temperature magnetization studies and specific heat analysis shows no sign of magnetic transition in the system, for temperatures as low as 0.4 K. Further, μSR studies confirm a dynamic magnetic ground state in the system. In order to explain this exotic phenomena observed experimentally, we used a combined first-principles and Monte Carlo simulations approach. The ground state of the system was approximated using an iterative minimisation procedure, following which the finite temperature behaviour was numerically simulated using Monte Carlo simulations. We found that the nearest neighbour interactions are ferromagnetic, while the second neighbour interactions are antiferromagnetic and almost half of the nearest neighbour exchange. Our simulated spin structure factor, which is the Fourier transform of spin-spin correlation function, shows a broad diffuse peak, signalling at a spiral order in the system. Thus our studies reveal that the existence of competing exchange interactions combined with geometric frustration leads to a dynamic magnetic ground state in $Na_{0.5}Yb_{0.5}WO_4$ .

# Electrically tunable valleytronics in 2D ferrovalley/ferroelectric heterostructure

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## Abstract

The sign inversion of anomalous Hall conductivity (AHC) and spontaneous valley polarization offers an unprecedented insight into the electronic structure of condensed matter systems. Herein, we report the presence of intrinsic valley polarization with sign inversion in anomalous transport features because of valley dependent Berry curvature in ferrovalley/ferroelectric vdW heterostructure. The ferrovalley and ferroelectric monolayer are interfaced considering vdW interaction which breaks both time-inversion symmetry resulting in sizable Berry curvature. The robust characteristic of valley splitting is realized in the heterostructure system due to magneto-valley coupling (MVC) effect, which is flexible towards external stimuli. The valley polarization in the heterostructure system is found to be 93.2 meV at no electric field. At zero electric field, we find the AHC value to be 155.32 S/cm at the Fermi energy region and net negative Berry curvature peaked at K and K′ valleys. From the real-time device perspective, the sign inversion facet of AHC at Fermi energy region is of vital interest as it removes conventional approach of adding dopants and defects. Alongside AHC sign inversion, we observe the phase change from semiconductor to half metallicity at a field of -0.2 V/Å and beyond due to presence of MVC effect in heterostructure system under the electric field. These finding suggest that the electric field driven AHC sign inversion and valley dependent characteristics can act as a promising candidate for instantaneous low power consumption quantum devices.

# What drives the distortions in 2D hybrid perovskites?

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Structural distortions in halide perovskites play a crucial role in determining their electronic and optical properties. The distortions that take place in inorganic perovskites are well understood and are primarily driven by the size of the A-site cation. While these ideas have been borrowed into the discussion of structural distortions in three dimensional hybrid perovskites, other factors enter the discussion. Specifically, hydrogen bonding between the hydrogens of the functional groups attached to the A-site cation and the anions of the inorganic framework can lead to significant structural modifications[1]. These hydrogen bonds can restrict the rotational freedom of the A-site cation and lead to the octahedral distortions of the inorganic network[1].

However, in low-dimensional (2D) halide perovskites, the origin of the structural distortions remains less explored. Using first-principles density functional theory (DFT) calculations, we systematically investigate the driving forces behind distortions in 2D halide perovskites. We analyze the role of organic spacer cations in modulating the inorganic framework. Our findings provide insight into how these factors influence octahedral tilting and lattice deformations, thereby affecting the material's stability and electronic properties.

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# Bimetallic Molecular Model Inspired Designing of M–Zn–N–C (M = Fe, Co, Ni, Cu) type Catalysts for Enhanced HER and OER Performance

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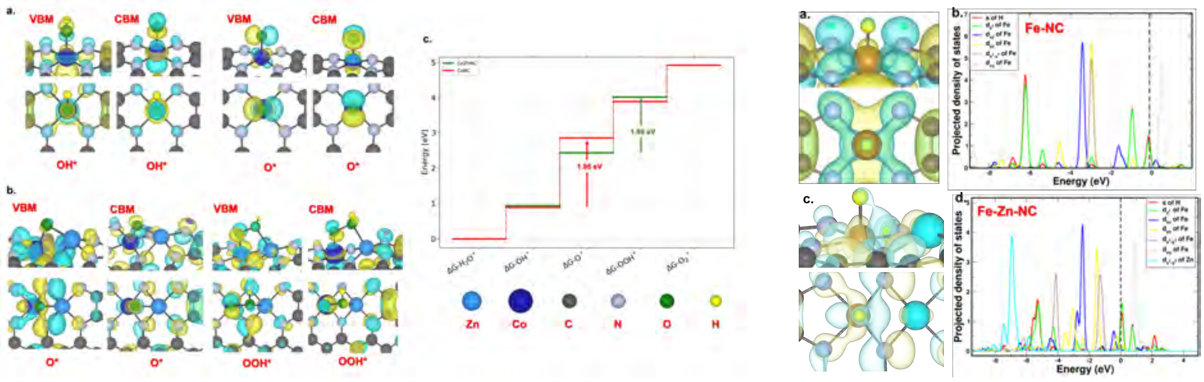
Fuel cells and metal-air batteries [[1]] offer high efficiency and zero emissions, driving interest in bimetallic M-N-C (MNC) catalysts for HER and OER in water electrolysis[[2]] and fuel cells. The catalytic activity is mainly controlled by the environment surrounding the active site, where the chemical reaction occurs. Modulations of the environment in which the catalysts are placed can be designed to adjust the electronic and geometric properties with the precision of the active site, thus greatly increasing the catalytic activity and selectivity. This study presents a comprehensive computational investigation of bimetallic M-Zn-N-C (M = Fe, Co, Ni, Cu) catalysts designed to enhance electrocatalytic performance in hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). Using density functional theory (DFT) calculations, we explore the impact of incorporating zinc into single-atom M-N-C catalysts to create dual-atom catalysts (DACs) inspired by a bimetallic molecular catalyst (BMC) model. Our findings demonstrate that zinc incorporation significantly modifies the electronic structure of transition metals, activating low-lying $d_{x^2-y^2}$ and $d_{z^2}$ orbitals and optimizing interactions with adsorbed species [[3]].

For OER, zinc enhances the interaction between the d-orbitals of transition metals and the $p_z$ orbitals of oxygen, reducing overpotential values. The CoZn catalyst exhibits the best OER performance with an overpotential of 0.70 V in the BMC model and 0.36 V in the MNC model, a substantial improvement over the pure Co catalysts (2.36 V and 0.72 V, respectively) [[3]]. The order of OER activity is determined to be CoZn $>$ FeZn $>$ NiZn $>$ CuZn.



(a) Interaction of orbitals at the reaction barrier in Co-Zn-NC and Co-NC, Effect of Zn doping.

(b) Comparative orbital interaction plot for the HER reaction between Fe-NC and Fe-Zn-NC catalysts.

Figure 1: Effect of Zn incorporation through orbital interaction analysis.

For HER, zinc modifies the interaction between the $d_{z^2}$ orbital of transition metals and the s-orbital of hydrogen, optimizing adsorption energies and lowering reaction barriers. The FeZn catalyst shows the highest HER activity, with hydrogen adsorption energy ($\Delta E_{H*}$) of -0.09 eV in the BMC model and 0.07 eV in the MNC model, compared to -0.81 eV and -0.56 eV for pure Fe catalysts. The order of HER activity is found to be FeZn $>$ CoZn $>$ CuZn $>$ NiZn.

Ab initio molecular dynamics simulations confirm the stability of these bimetallic catalysts at room temperature. The study provides crucial insights into the structure-activity relationships of M-Zn-N-C catalysts, demonstrating the potential of BMC-inspired design principles for enhancing the catalytic activity of MNC-type catalysts. These findings offer a promising pathway for designing efficient and sustainable electrocatalysts for renewable energy applications, particularly in water splitting processes.

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# Coupled electron-phonon dynamics in polar semiconductors: A first-principles Boltzmann transport approach

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The energy transport properties of materials such as the thermal conductivity, the electronic mobility and the Seebeck coefficient are strongly dependent on the interactions among elementary energy carriers - electrons and phonons. The physics of coupled electron-phonon transport is of primary interest to study various phenomena such as superconductivity, thermoelectricity, phonon drag and hydrodynamic transport [1]. Here, we present an efficient computational tool to obtain a full numerical solution of the coupled electron-phonon linearized Boltzmann transport equation (BTE) to predict the energy transport properties in polar semiconductors such as GaAs, GaN and BAs. We focus on low temperatures, where the momentum-conserving collisions among phonons and electrons - the Normal processes - dominate over the momentum-dissipating collisions - the Umklapp processes, thus necessitating the development of an iterative scheme to fully solve the coupled BTE beyond the commonly used relaxation time approximation (RTA). In our approach, we allow the phonon and the electron system to go out of equilibrium, unlike many of the previous approaches that assume that the phonons remain in equilibrium while calculating the transport coefficients [2]. We address the computational complexity involved in these calculations by constructing two different discretizations of the Brillouin zone for electrons and phonons respectively, that enabling us to selectively refine the regions of the electronic and the phononic Brillouin zones of interest, depending on the temperature and the number density of electrons under consideration. From our calculations, we find that the effect of phonon drag on the phonon thermal conductivity is relatively weak in GaAs, GaN and BAs at all temperatures. However, there is a giant enhancement in the absolute value of the Seebeck coefficient at low temperatures, driven by the phonon drag effect, in all of these materials. Our approach enables a rapid, yet accurate evaluation of thermal, electrical and thermoelectric transport coefficients in doped semiconductors at a fraction of the computational cost, compared to the approaches available in the literature [3,4].

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D3-022

# Unveiling different structural orderings in Fe$_{5-x}$GeTe$_2$

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We investigate the metallic van der Waals itinerant ferromagnet Fe$_{5-x}$GeTe$_2$ with atomic scale, spatially resolved low-temperature scanning tunneling microscopy (STM), and spectroscopy (STS). STM images unveil a new structural order $2a \times 1a$ along with the known order $\sqrt{3}a \times \sqrt{3}a$ manifesting as non-uniform domains[1]. STS shows spatial and energy resolved local density of states that reveal the crucial influence of Fe(1) site occupancy on the system's electronic interactions. Our magnetization measurements show magnetic anomalies at lower temperatures and identify a Curie temperature ($T_c$) surpassing room temperature. Collectively, our results elucidate the intricate nature of Fe$_{5-x}$GeTe$_2$ and underscore its potential for tunability of spintronics and high-temperature magnetic applications.

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# AAPALI PSI-K 2025

# Non-trivial Topology, Hyperferroelectricity and Giant Rashba Spin-splitting in Non-centrosymmetric LiZnAs Compound

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The exploration of coexistence of multiple quantum phenomena within a single material enables the study of physics associated with relativistic electrons in solids [1]. These compounds are known as composite quantum compounds (CQCs) [2]. Despite being distinct phenomena, Hyperferroelectricity (HyFE), Rashba spin physics and topological non-triviality can coexist in a CQC platform [2]. HyFE is an intriguing phenomenon in which polarization persists under open circuit boundary conditions (OCBC) [3]. Using density functional theory (DFT), we investigate the hexagonal LiZnAs compound with surface-induced asymmetry for its HyFE, Rashba effect, and topologically non-trivial states using VASP 6.3.2 and WIEN2k 23.2. Phonon spectra of paraelectric phase under OCBC indicates that $A_{2u}$(LO) is soft with imaginary frequency of $51i$ cm$^{-1}$ which is drastically different from $A_{2u}$(TO) with $91i$ cm$^{-1}$ indicating HyFE. Recently, it has been observed that soft LO at zone centre may not be hallmark of HyFE [4] and one must employ more rigorous approach of evaluating electric free energy under OCBC. HyFE solid must exhibit non-zero polarization at the minimum of free energy under OCBC. This was validated by our calculations for LiZnAs compound. The in-plane lattice expansion in LiZnAs leads to topological phase transition (TPT) with intermediate Weyl semimetal phase. The orbital band inversion after critical point indicates the topological insulating phase. Topological surface states and $Z_2$ invariants are calculated to validate the non-trivial topology. The switching of buckling atomic planes leads to reversed Rashba spin-texture ensuring the change in polarization direction of LiZnAs. This paves the path towards the electrical way to control and manipulate spin-textures. The calculated giant Rashba coefficient are 4.43 eV Å at ambient and 2.42 eV Å after TPT. This facilitates in spintronics, spin-orbitronic and current-induced switching. Our findings can open avenues for interplay between Rashba effect, HyFE, and topological phenomena with advanced theoretical methods.

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# Concerted rattler dynamics induce glass-like thermal conductivity in crystalline TlAgTe

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Crystalline semiconductors possessing glass-like thermal conductivity are fundamentally interesting since they bridge the gap between the particle-like ballistic propagation of phonon wave packet in crystalline materials and diffusion of localized vibrational modes in disordered systems such as glass. Such compounds are also technologically important with applications such as thermoelectric, thermal barrier coating, and data storage devices.

In this work, we show how the concerted rattler dynamics of Tl and accompanying large anharmonicity lead to ultralow and nearly temperature-independent glass-like thermal conductivity mediated by phonons. Combining state-of-the-art first-principles density functional theory, Peierls-Boltzmann transport equation and considering high-order anharmonicity while treating both phonon frequencies and scattering rates, we show that lattice thermal conductivity becomes ultralow which remains almost independent of temperature. Our analysis reveals significant coherent contributions to the lattice thermal conductivity due to hopping among localized vibrations, that is typically found to be insignificant in crystalline system. The calculated lattice thermal conductivity is strongly affected by high-order quartic anharmonicity, which is evident in the strong renormalization of phonon dispersion at finite temperature and the large four-phonon scattering rates. Our calculations agree well with the experimental observations. Our work sheds light on the role of concerted rattling mechanisms on inducing glass-like thermal conductivity, which can help in novel materials design and engineering.

# Magnetism in a two-dimensional honeycomb antiferromagnet $BaNi_2(XO_4)_2$; (X=P, V & As): A combined DFT+U and Monte-Carlo simulations

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We present a comprehensive theoretical investigation of the electronic and magnetic properties of the quasi-two-dimensional honeycomb antiferromagnets $BaNi_2(XO_4)_2$ (X = P, V, As) using density functional theory (DFT) combined with Hubbard U (DFT+U) corrections, incorporating spin-orbit coupling (SOC). Our calculations reveal that the lowest-energy magnetic state exhibits a Mott-Hubbard insulating character with a sizable band gap, highlighting the strong electronic correlations in these materials. A detailed analysis of the magnetic interactions indicates that the orbital moment is significant, and SOC plays a crucial role in shaping the overall magnetic behaviour. We systematically compute the interatomic exchange interactions, including both isotropic and anisotropic contributions such as Dzyaloshinskii-Moriya (DM) interactions and symmetric exchange anisotropy. Using these computed parameters, we construct a complete spin model and perform large-scale classical Monte-Carlo simulations to investigate the finite-temperature magnetic properties. Our results provide a microscopic understanding of the experimentally observed Néel-type antiferromagnetic ground state in this class of materials.

Additionally, we explore possible routes to realize alternative magnetic phases. We discuss how tuning key interaction parameters, such as exchange anisotropy and frustration, could stabilize noncollinear spin structures, including spiral magnetism, or drive the system towards a potential quantum spin-liquid ground state. Our findings offer valuable insights into the interplay of spin-orbit coupling, electronic correlations, and magnetic frustration in honeycomb antiferromagnets, paving the way for future experimental and theoretical studies.

** The work has been carried out in collaboration with Dr. Swarup K. Panda, Bennett University.*

# Strain induced electronic and magnetic transition in S=3/2 ferromagnetic spin chain compound SbCrSe$_3$

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The study of two-dimensional (2D) magnetic materials and their pressure driven electronic and magnetic transitions are growing research field in modern condensed matter physics. In this context, recently ferromagnetic Cr-based compounds such as CrGeTe$_3$, CrI$_3$, have been investigated experimentally and theoretically for their possible spintronics applications. Here, we theoretically studied the electronic and magnetic properties of a relatively less explored Cr-based chalcogenide, namely SbCrSe$_3$ where 2D layers of magnetic Cr$^{3+}$ ions form a triangular lattice. In this work, we employed density functional theory including Hubbard *U* (DFT+*U* approach) in conjunction with constrained random-phase approximation (cRPA) where the later was used to estimate the strength of *U*. Our findings at ambient pressure show that the system exhibits a semiconducting ferromagnetic ground state with a gap of 0.5 eV and large Cr moments that corresponds to nominal S=3/2 spin-state. To understand the nature of magnetism, we calculated the inter-site magnetic exchange coupling strength using magnetic force theorem. The exchange interaction calculation shows, 1st nearest-neighbor (NN) interatomic exchange coupling ($J_1$) is found to be weak ferromagnetic (FM), while 2nd NN coupling ($J_2$) are relatively strong ferromagnetic (FM), making this system a candidate for 1D non-frustrated ferromagnetic spin-chain family of materials. Based on orbital resolved interactions, we demonstrated the reason behind two different types of interactions among 1st and 2nd NN despite their very similar bond lengths. We observe a significant spin-orbit coupling effect, giving rise to a finite magneto crystalline anisotropy, and Dzyaloshinskii-Moriya (DM) interaction. Further, we found that by applying uniaxial strain along crystallographic a and b-axis, SbCrSe$_3$ exhibits a magnetic transition to a semi-conducting AFM ground state and give rise to novel phenomenon angle dependence of exchange coupling. We also showed that the Neel temperature of AFM ground state could be significantly enhanced by applying compressive strain. Thus, our findings can enrich the versatility of SbCrSe$_3$ and make it a potential application.

**This work has been carried out in collaboration with Dr. Swarup Kr. Panda (Bennett University, Greater Noida, India).*

# Abstract for AAPALI PSI-K 2025

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Exploring efficient electrocatalysts for the splitting of water to generate hydrogen and oxygen is essential for development of renewable energy sources, especially considering the detrimental environmental impacts of fossil fuels. Single-atom catalysts (SACs) have emerged as highly promising candidates for both hydrogen and oxygen evolution reactions. Significant amount of research has been done on hetero atom doped carbon-based SACs, such as graphene nanosheets, nanorods, and other carbon allotropes. However, the potential of fullerene, another allotrope of carbon, for electrocatalytic applications has not been extensively studied. In this work, we investigate transition metal (TM)-doped (Fe, Co, Ni, Ru, Rh, Pd, Os, Ir and Pt) fullerene as electrocatalyst for HER and OER using density functional theory (DFT). The ab initio results show that fifth-row TMs, such as Ir and Pt, exhibit excellent catalytic activity for HER, while fourth-row TMs, e.g. Ru and Pd are identified as superior catalysts for OER. The superior catalytic activities of the fourth-row TMs (Ru, Pd) for OER and the fifth-row TMs (Ir, Pt) for HER are explained through the electronic structure of the catalysts, interaction between the involved intermediates and the catalytic surface, the exchange current density related to catalytic activity and the detailed effects of band centres of the TM-doped fullerene systems.

AAPALI PSI-K 2025

# Crucial role of Fe-Fe magnetic interactions in layered triangular antiferromagnet in dictating Li-ion cathode material efficiency

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The efficiency of the Li-ion battery mostly understood from the redox reactions capacity. Incidentally one of the most prospective Li-ion cathode materials belongs to the class of polyanionic compounds with Fe. If Fe is in +3 or +4 state, it has partially filled 3*d*-shell, with unpaired spin configuration, which can lead to the exotic magnetic ground state. However, very little effort have been made in the direction to understand the role of the magnetism in deriving the observed redox reaction capacity in the context of the Li-ion battery. In this work we are doing density functional theory based electronic structure investigation of the Fe based Li-ion cathode material to understand the role of the Fe magnetic moment and Fe-Fe magnetic exchange interaction in the electrochemistry of the battery with the support from experiments. We found that in the charging process when the Li-ion taking out from the cathode, the magnetic moment of the Fe decreases monotonically. The calculated magnetic exchange interactions clearly show the anti-ferromagnetic ordering of the triangular Fe sublattice. As a results strong magnetic frustration appears in the system. In the charging process (reduction of Li-ion) the magnetic interactions vary non-monotonically. Initial calculated results indicated the possibilities of the mixed valent Fe state and we are trying to understand the evolution of the charge state and spin state during the charging cycle through NEB calculations.

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# M-$N_x$B SAC Catalysts with CO gas as Reactant Selective Towards $C_2$ Product

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Abstract:

Obtaining $C_1$ or $C_2$ products in CORR or $CO_2$RR is a challenge still demanding. SAC catalysts can obtain $C_1$ products in CORR or $CO_2$RR, while parallel hydrogen evolution reaction is highly probable[1]. The formation of C-C bond[2,3] as an essential step for obtaining $C_2$ products, requires two active sites which challenge the basic geometry of SAC. Moreover, obtaining any desired product in such reaction depends on key factors like active site as well as the neighborhood. In case of $C_2$ products formation multiple key intermediates and parallel pathways further intricate the analysis. DAC can procure $C_2$ products, but these catalysts are wreckable and challenging to synthesize. In this work, we propose in the presence of CO gas the SAC with N-B neighborhood to obtain ethylene and ethanol as desired $C_2$ products. The innovative addition of B helps to promote CORR or $CO_2$RR while suppressing HER or $C_1$ production. We also identified the important role of CO gas to promote C-C bond formation in CORR and CO/$CO_2$RR. We have identified Fe, Co, Rh, Ni as possible active center atom in M-N-B SAC can produce $C_2$ product with low over potential as low as -0.66 V ($FeN_3B$) in CORR and CO/$CO_2$RR. Our findings indicate that the presence of CO gas can simultaneously suppress the HER and shift the selectivity from $C_1$ to $C_2$ products in $MN_xB$ single-atom catalysts (SACs) during CORR and CO/$CO_2$RR. This opens up the possibility of future investigation on in SAC for $C_2$ products in CORR beyond Cu.

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# Generative AI Model for Designing Magnetic Materials

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Designing new materials with targeted properties is a fundamental challenge in materials science, as the vast chemical space make traditional discovery approaches inefficient. Conventional methods, which rely on high-throughput density functional theory (DFT) calculations and human-guided modifications, are computationally expensive and often constrained by heuristics. Recent advancements in generative machine learning provide a promising pathway to accelerate materials discovery by exploring diverse structural and compositional spaces beyond human intuition.

In this work, we develop a diffusion model [1], a powerful generative AI approach, for materials design, with a focus on discovering novel rare-earth-free permanent magnets (PMs). PMs are essential for energy-efficient technologies such as electric vehicles and wind turbines, but high-performance PMs typically rely on rare-earth (RE) elements, which are expensive and subject to supply constraints.

Previously, we designed RE-free PMs using a conditional variational autoencoder (cVAE) [2], which demonstrated the potential of generative models in materials discovery. However, a key limitation of the cVAE was its bias toward generating low-symmetry crystal structures, which may not always exhibit optimal magnetic properties. In this work, we overcome this drawback by employing a diffusion model, which enables more diverse and physically meaningful crystal generation.

To identify promising candidates, we first generate materials using the trained diffusion model and screen them with machine learning models that predict formation energy and saturation magnetization ($M_s$). The most promising candidates are further validated via DFT calculations, incorporating magnetocrystalline anisotropy energy ($K_1$) to assess their potential as high-performance PMs.

By integrating generative modeling, property prediction, and first-principles verification, our approach enables the systematic discovery of RE-free PMs with high $M_s$ and $K_1$. This framework provides a scalable and data-driven strategy for accelerating the design of advanced materials across various domains.

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# A Computational Methodology for Kinetic Energy Density Dependent Meta-GGA Functionals in Finite-element Based DFT Calculations with Accelerated Self-consistent Field Iterations

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First principles modelling methods have become a ubiquitous tool for studying various properties of materials in diverse applications, with Kohn-Sham density functional theory (DFT) being the frontrunner. While Kohn-Sham DFT is theoretically exact, the specific form of exchange-correlation energy remains elusive and is approximated using different levels of theory with increasing complexity. Kinetic energy density dependent meta-GGA ($\tau$-mGGA) functionals like SCAN that incorporate kinetic energy density alongside local electron density and gradient of electron density, offer improved accuracy but pose computational challenges. To this end, the regularized-restored SCAN (r$^2$SCAN) exchange-correlation functional represents a promising compromise between numerical stability and accuracy, meeting the most known constraints of density functional approximations. The current work introduces finite-element discretization and an efficient numerical implementation of $\tau$-mGGA functionals in the open-source DFT-FE code[1] to conduct large-scale electronic structure calculations. Despite improved accuracy, the usage of $\tau$-mGGA functionals is somewhat limited due to the numerical instability[2] and slow convergence nature of the self-consistent field (SCF) iterations associated with these functionals. To mitigate this, we propose a new preconditioned mixing scheme within the aforementioned finite-element framework to accelerate the convergence of SCF iterations when using $\tau$-mGGA functional, leveraging a low-rank approximation of the dielectric matrix.[3] We assess the robustness and efficiency of aforementioned computational methods across a range of system sizes and material systems highlighting the applicability of the framework to generic boundary conditions.

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# Semi-universal Solution of Thomas-Fermi Equation for Jellium Spheres

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**Motivation:** Find the exact and universal solution of the Thomas-Fermi equation for jellium spheres.

## 1. Abstract

The Thomas-Fermi model of electronic structures provides a universal solution for neutral atoms. Are there other systems where such universal solutions can be formed? In this work, we look for answer to this question and show that for neutral jellium spheres, a semi-universal solution of the Thomas-Fermi equation exists. We demonstrate that our solution depends on a parameter, and for a group of jellium spheres, this parameter is unique. Thus, one parameter determines the properties of many neutral jellium spheres with different numbers of electrons and bulk density parameters.

# Exploring Titanium-Doped $V_{n+1}C_nO_2$ MXenes as Efficient Urea Adsorbents for Wearable Artificial Kidneys

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Chronic kidney disease (CKD) is a major global health issue, particularly in developing countries like India, where it significantly contributes to mortality. In 2021, about 4.7 million people required kidney replacement therapy (KRT) , with challenges including high treatment costs and shortages of haemodialysis supplies. Portable dialysis machines offer a practical solution by allowing patients to manage their renal health at home, reducing hospital visits and easing the burden on healthcare systems.To maximize the effectiveness of these devices, there is a pressing need to develop advanced adsorbent materials with high surface area, stability, affordability, biocompatibility, and toxin selectivity.Cost-effective, high-performance adsorbents are essential to improve care for the growing number of CKD patients.Wearable Artificial Kidneys (WAKs) represent a promising solution for blood purification in patients with chronic kidney disease. For these devices to function effectively, it is crucial to identify materials that can efficiently regenerate dialysate, remove harmful substances, and restore electrolyte balance while maintaining optimal levels of creatinine and urea in the blood. In recent years, Ti3C2, a 3-2 MXene, has shown great potential for WAKs due to its ability to effectively adsorb creatinine and urea. However, the adsorption properties of other MXenes remain largely unexplored in this context. In this work, we investigate the adsorption capabilities of Titanium doped $V_{n+1}C_nO_2$ MXenes. The Titanium atoms on the surface of the material reacted with oxygen to form tiny particles of TiO2 (titanium dioxide), but it doesn't affect the vanadium parts. The TiO2 particles also create many active sites on the surface. These are spots where chemical reactions can happen more easily. This is useful for adsorbing (sticking to) and removing toxins like urea. By selectively oxidizing the titanium atoms and retaining the vanadium atoms, we enhance the material's ability to remove toxins (like urea) from dialysate. This makes the material more efficient and effective for use in a Wearable Artificial Kidney (WAK).

# Accelerating the Search for Superconductors Using Machine Learning

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Predicting the critical temperature ($T_c$) of superconductors remains a significant challenge in condensed matter physics. While BCS theory explains superconductivity in conventional superconductors, it does not provide a framework to predict critical temperature for the unconventional superconductors. Quantum Structure Diagrams have been successful in establishing structure-property relationship from chemical composition [1, 2]. In this work, we explore whether such chemical composition-based features, leveraged through machine learning, can predict critical temperature universally across different compound classes. Earlier studies generally rely on datasets with inconsistencies and incomplete information, leading to suboptimal predictions [3]. To address this, we introduce a data cleaning workflow that enhances the quality of superconducting material datasets [4] by eliminating redundancies and resolving inconsistencies. Principal Component Analysis (PCA) further uncovers clustering of different classes of superconductors. With this improvised database, we apply a supervised learning approach and develop a Random Forest model to predict critical temperature. Our model is demonstrated to generalize effectively, to predict the critical temperature of compounds outside the database with reasonable accuracy. We use our machine learning model to predict that $\mathrm{Ba_6Ca_6Tl_5Cu_9O_{29}}$ show superconductivity with a critical temperature of 105 K. Our approach effectively identifies promising candidates, demonstrating the potential of machine learning in discovering new superconductors. As our machine learning model predicts critical temperature from merely chemical composition, we expect it to be effective and powerful in prediction of new superconductors.

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# Large electro-mechanical actuation in hydrogenated Xenes leading to topological transition

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We report from first principles, the possibility of reversible onset of topological phase in hydrogenated Xenes (H-Xenes) beyond Silicane, namely, Germanane and Stanane, exclusively through electro-mechanical actuation. It is found possible to systematically induce robust uniaxial strain through non-uniform application of in-plane electric field, as possible through application of in-homogeneous bias at gates of realizable length-scales. Electrically induced strain causes substantial lowering of band-gap trivially across all H-Xenes, eventually evolving topologically beyond a threshold amplitude in Germanane and Stanane promisingly within the range sustained by the monolayers. In case of nano-ribbons of these H-Xenes, with bias applied at gates parallel to them but covering them only partially, switchable emergence of topologically protected chiral interface states can be possible in sufficiently wider ribbons. The demonstrated electro-mechanical actuation and the associated trivial to topological tuning of band-structure should be possible in the broader class of two dimensional covalent networks in general.

[1] Sujith N. S., Nabendu Mondal and Joydeep Bhattacharjee. [under review]

# AAPALI PSI-K 2025

## Exploring the Foundations: Prerequisites for Direct Epoxidation

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Catalysts are fundamental to the chemical industry, driving extensive experimental and theoretical research aimed at designing and optimizing catalysts for a wide range of chemical reactions. Among these, the ethylene epoxidation reaction has attracted significant attention due to its industrial importance, with selectivity being a primary focus of research. While several factors influence selectivity, considerable efforts have been directed toward understanding the nature of oxygen species involved in selective epoxidation. However, a clear consensus on the precise nature of electrophilic oxygen species responsible for promoting epoxidation is yet to be established. This study uses density functional theory (DFT) to investigate the interaction between ethylene and oxygen species adsorbed on the Ag(100) surface under varying oxygen coverages. Structural and electronic analysis identified three oxygen species: atomic oxygen, dissociatively adsorbed molecular oxygen ($O_2$-dis), and $O_3$. All three exhibit electrophilic character, enabling epoxidation. Direct epoxidation occurs when ethylene approaches symmetrically, while asymmetric interactions lead to intermediates such as oxametallacycles or dioxo-metallacycles, hindering selectivity. The results demonstrate that epoxidation selectivity depends not only on the oxygen species but also on surface reconstruction and ethylene orientation, indicating that oxygen species alone do not dictate the outcome.

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# Monte Carlo Simulated Annealing and Machine Learning Interatomic Potentials for High-Entropy Materials Design

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High-entropy materials (HEMs) are a class of advanced functional materials with multi-element compositions, offering exciting opportunities in energy storage, catalysis, and beyond [1-2]. However, determining atomic arrangements in these compositionally complex systems is challenging, as density functional theory (DFT) calculations become prohibitively expensive for large configuration spaces.

To address this, we developed a Monte Carlo Simulated Annealing (MCSA) code within the Atomic Simulation Environment (ASE), that supports any potential energy model with an ASE interface, including several modern machine learning interatomic potentials (MLIPs).

As a case study, we applied this code to high-entropy oxyfluoride cathodes (HEOFs) for Na-ion batteries [3]. We benchmarked MLIP-predicted energies for a large number of HEOF configurations against DFT, using general-purpose models such as MACE, ORB, SevenNet, FAIR-Chem, CHGNet etc. We also compared MLIP-predicted lattice parameters of HEOFs at various desodiation states with DFT results. This analysis helped identify MLIP models that most accurately reproduce DFT energy and structural trends.

We are currently extending the MCSA framework presented here with a smart move strategy, in which lightweight machine learning models are trained to predict the quality of atomic swaps based on local environments. This is expected to guide Monte Carlo sampling more efficiently toward low-energy configurations.

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# Data-driven Discovery of Novel High-performance Quaternary Chalcogenide Photovoltaics

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Conversion of light to useful electricity is mediated by the interactions of light with matter in photovoltaic materials, which offer a sustainable and cost-effective solutions to energy generation. Combining data-driven approaches with state-of-the-art first-principles calculations based on static and time-dependent density functional theories, and nonadiabatic molecular dynamics simulations, we predict four high-performance photo-absorber materials ($SrCuGdSe_3$, $SrCuDyTe_3$, $BaCuLaSe_3$ and $BaCuLaTe_3$) belonging to a novel family of known semiconductors. Analysis of the crystal structure reveals that Gd/La/Dy and Cu cations possess octahedral and tetrahedral coordination environments, respectively, suggesting them to exhibit properties (e.g., optical activity, defect tolerance, stability) complementary to the known solar absorbers with either tetrahedral (e.g, Si, GaAs, CdTe) or octahedral (e.g., $MAPbI_3$) coordination chemistries. Chemical bonding analysis reveals anti-bonding states in the valence bands which is an indicator of strong defect tolerance. In addition to being thermodynamically stable, these compounds also exhibit dynamical stability as revealed by the lattice dynamical calculations. Hybrid density functional theory calculations including spin-orbit coupling reveal that these compounds possess direct band gaps, which lie in the close vicinity of optimal range of band gap ideal for visible light absorption. As a consequence, the calculated optical absorption coefficient ($\alpha(E)$) and spectroscopic limited maximum efficiency for these compounds become comparable or larger than crystalline silicon, GaAs, and methylammonium lead iodide ($MAPbI_3$). Temperature-dependent dynamics of structure and electronic band energies reveal relatively low fluctuation, signifying their high stability and small electronic-ionic sub-lattice coupling, signifying robust mechanisms behind their high conversion efficiencies. We further determined defect formation energies in these compounds, which showed that despite the likely formation of cation vacancy and interstitial defects, mid gap states remain absent, making them non detrimental to carrier recombination. Our work warrants experimental verification of our prediction and invites further theoretical investigations of other compounds in this family.

# Impact of oxygen and ozone exposer on the reaction kinetics of two-dimensional $C_2N$ monolayer: A First Principles Study

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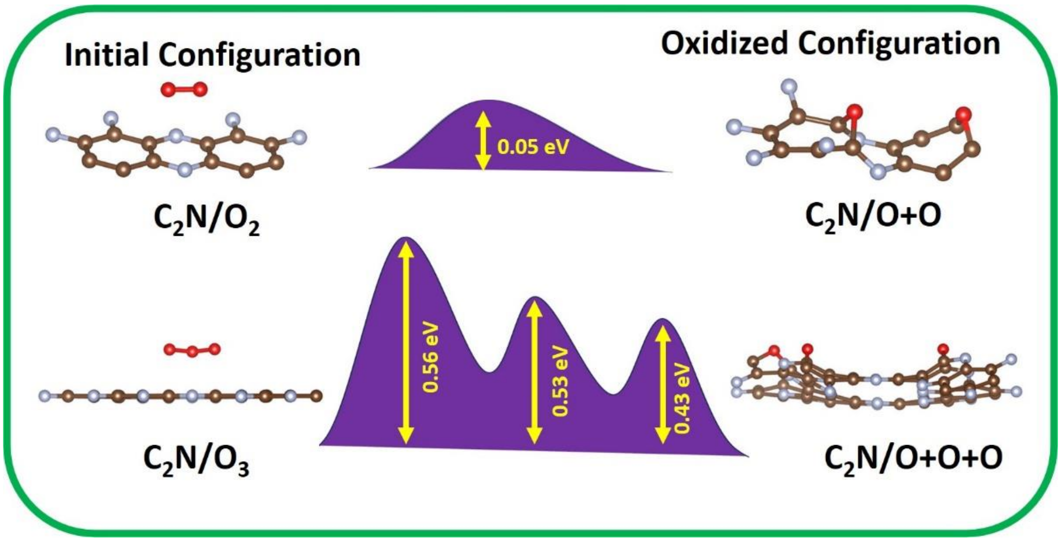
## Abstract

The process of environmental oxidation is pivotal in determining the physical and chemical properties of two-dimensional (2D) materials. Its impact holds great significance for the practical application of these materials in nanoscale devices functioning under ambient conditions. This study delves into the influence of $O_2$ and $O_3$ exposure on the structural and electronic characteristics of the $C_2N$ monolayer, focusing on the kinetics of adsorption and dissociation reactions. Employing first-principles density functional theory calculations alongside climbing image nudged elastic band calculations, we observe that the $C_2N$ monolayer exhibits resistance to ozonation, evidenced by energy barriers of 0.56 eV, respectively. These processes are accompanied by the formation of C–O–C groups. Furthermore, the dissociation mechanism involves charge transfers from the monolayer to the molecules. Notably, the dissociated configurations demonstrate higher bandgaps compared to the pristine $C_2N$ monolayer, attributed to robust C-O hybridization. These findings suggest the robustness of $C_2N$ monolayers against oxygen/ozone exposures, ensuring stability for devices incorporating these materials.



# Inorganic Antiperovskite Derivatives: Next-Gen Materials for Optoelectronics

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Inspired by the stellar success of halide perovskites in photovoltaics, their derivative semiconductors have emerged as exciting contenders for optoelectronic applications. However, they still struggle to match the photovoltaic prowess of their parent compounds. The key challenge lies in structural deficiencies at the B-site or X-site, which break the seamless connectivity of the vital [$BX_6$] octahedral framework, hindering performance. Herein, we introduce a novel class of antiperovskite-derivative materials [1, 2] with the formula $X_3BA_3$ and unveil their intriguing optoelectronic, excitonic, and polaronic properties. Leveraging cutting-edge computational techniques—including density functional theory (DFT), density functional perturbation theory (DFPT), and many-body perturbation theory [GW and Bethe-Salpeter equation, (BSE)]—we provide deep insights into their potential for next-generation optoelectronic applications. Our findings demonstrate the thermodynamic, dynamical, and mechanical stability of these materials. GW and BSE calculations unveil the true potential of these compounds, revealing direct optimal bandgaps, strong light absorption, moderate exciton binding energy, and balanced polaronic mobility—making them prime contenders for next-gen optoelectronics. This has been further validated by calculating the spectroscopic limited maximum efficiency, which is estimated to be approximately 18.2%–29.1% for these materials.

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# Manipulating van der Waals Interaction with Strong Light-matter Coupling

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van der Waals interactions play important roles in diverse scientific fields ranging from DNA base stacking to 2D material interlayer interactions. Manipulating such interactions in the regime of strong light-matter coupling (inside of an optical cavity) can open new paths for controlling the properties of condensed phase systems. However, theoretical modeling of such interactions is highly challenging inside the cavity due to spatial non-locality. As demonstrated by a perturbation analysis, the van der Waals interaction in the cavity has a slow $R^{-3}$ decay and it does not decay to 0 when R approaches infinity, where R is the inter-molecular distance[1]. We have designed a machine learning approach to faithfully model these weak interactions with deep neural networks. However, a straightforward training does not generate the accurate enough results. We found out that incorporation of correct physics with our machine learning approach can significantly improve the accuracy and the generalizability of the model, which is crucial for modeling thermodynamics of many molecules in the cavity with molecular dynamics simulations. Importance of these results in predicting structural and thermodynamic properties of fluid $H_2$ will be discussed.

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# AAPALI PSI-K 2025

## Retrieval augmented generation for building datasets from scientific literature

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In this work, we show that employing Retrieval Augmented Generation (RAG) with a Large Language Model (LLM) enables us to extract accurate data from scientific literature and construct datasets. The rapid growth in publications necessitates the automation of extraction of structured data as it is crucial for training machine learning (ML) models. The pipeline developed is simple and can be adjusted accordingly with natural language as input. Quantization enables us to run LLMs on consumer hardware and remove the reliance on closed-source models. Both Llama3-8B and Gemma2-9B with RAG give structured output consistently and with high accuracy as compared to direct prompting. Using the newly developed protocol, we created a data set of metal hydrides for solid-state hydrogen storage from paper abstracts. The accuracy of the generated dataset was > 88 % in the cases tested. Further, we demonstrate that the generated dataset is ready-to-use for ML models by testing it with HYST to predict the H2wt% at a given temperature. Thus, we demonstrate a pipeline to create datasets from scientific literature at minimal computational cost and high accuracy.

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# Toplogical Properties of CdX (X = S, Se and Te) Nanoribbons

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We have studied the electronic structure of free standing Sn doped cadmium chalcogenide (CdSnX, X = S, Se and Te) nanoribbons of varying widths with distorted arm-chair edge structures. The nanoribbons are derived from free-standing hexagonal buckled monolayers. The unsatisfied bonds of X and Sn atoms at the edges cause non-zero values of the magnetic moment. This introduces an exchange field leading to inverted bands. The band structures exhibit tilting of bands on inclusion of spin-orbit coupling (SOC) in the calculations. Moreover, different types of spin splittings are observed for different X atoms due to different values of spin-orbit interaction. This work has established the fascinating electronic properties of CdSnX nanoribbons viz., Weyl-like [1] semi-metallic behavior, Zeeman-type spin splitting and signature of Rashba [2] spin splitting along with Zeeman-type [3] spin splitting for X = S, Se and Te respectively. The gap openings at the band crossings near the Fermi level are mainly due to SOC of Sn atoms and are responsible for the electron and hole pockets making the system topologically exotic. Anomalous Hall conductivity (AHC) [4], non zero value of $Z_2$ index, unclosed Fermi arcs [5] and edge states are some of the hallmark criteria of a system to attend its topologically nontrivial version.

In this study we have explored all the above mentioned properties in distorted arm-chair CdSnX ( X = S, Se and Te) nanoribbons of varying widths. All the nanoribbons show $Z_2$ index value 1. We have studied the Berry curvature and its variations due to the presence of Weyl, Zeeman and Rashba type of band splitting, Fermi arcs and their spin texture in these nanoribbons. We get maximum AHC value of −34.01 S per cm in distorted CdSnTe nanoribbon. All the rest of the nanoribbons show non-zero AHC value. Distorted arm-chair edge CdSnTe nanoribbon of width 5*m* (*m* is the lattice constant of the monolayer unit cell) has proved to be very strong topologically nontrivial nanoribbon which shows Weyl, Zeeman and Rashba type of spin splitting of the bands near the Fermi level along with multiple edge states with unclosed Fermi arcs.

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# Exploring the Potential of Cadmium Sulphide (CdS) via First-Principles Calculations: Electronic, Optical, and Thermoelectric Perspectives

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**Abstract:** Cadmium sulfide (CdS), a widely studied II–VI semiconductor, holds strong potential for optoelectronic and thermoelectric applications owing to its favorable electronic structure and mechanical robustness. In this study, a comprehensive theoretical investigation of bulk CdS is performed using density functional theory (DFT) with the Quantum ESPRESSO package (QE) [1]. Structural optimization confirms the stability of the wurtzite phase, and the electronic band structure reveals a direct bandgap of 2.42 eV [2]. The projected density of states shows that the valence band maximum is primarily derived from sulfur (S) 3p orbitals, while the conduction band minimum is dominated by cadmium (Cd) 5p orbitals, offering insights into the nature of electronic transitions. Optical calculations, including the dielectric function, absorption coefficient, and refractive index, indicate strong absorption in the visible region, supporting CdS's applicability in light-harvesting devices [3]. Thermoelectric properties are assessed via the Boltzmann transport equation under the constant relaxation time approximation, yielding key parameters such as the Seebeck coefficient, electrical conductivity, and power factor [4]. The lattice thermal conductivity is estimated to be 3.84 W/m·K, suggesting moderate phonon transport. Mechanical properties derived from the Thermo_PW package (QE) [5] yield a bulk modulus ($B_H$) of 53.80 GPa and shear modulus ($G_H$) of 19.10 GPa, along with longitudinal, transverse, and average sound velocities of 4146 m/s, 2035 m/s, and 2240 m/s, respectively. The Debye temperature is calculated to be 229.7 K, and the Grüneisen parameter is 2.06, indicating moderate anharmonic effects. These findings confirm the mechanical stability and anisotropic transport characteristics, highlighting the multifunctional nature of CdS and its suitability for future integrated optoelectronic and thermoelectric device applications.

**Keywords:** Cadmium sulfide (CdS), Density Functional Theory (DFT), Electronic structure, Optical properties, Thermoelectric properties.

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# AB INITIO STUDY OF $CO_2$ CONVERSION ON $MOS_2$ MONOLAYERS WITH S VACANCIES: 2H V/S 1T V/S 1T′ PHASE

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**Abstract.** Chemical conversion of $CO_2$ to fuels and chemicals is an effective strategy to get rid of excess $CO_2$ from the atmosphere. A very high thermodynamic stability of the $CO_2$ molecule (750 kJ/mol C=O bond dissociation energy) makes this conversion process very difficult, demanding very high temperature routes. However, a photocatalytic pathway can lower the energy demands for this conversion using suitable photocatalysts. $MoS_2$, a prototypical transition metal dichalcogenide (TMD), has gained significant attention in catalysis and energy applications. 2H $MoS_2$ monolayers with S vacancies are potential catalysts, while the 1T (and 1T′) phase is rarely explored for $CO_2$ reduction. This study explores the photocatalytic conversion of $CO_2$ on $MoS_2$ monolayers with S vacancies, giving a comparison between 2H, 1T and 1T′ phases. The thermodynamics of $CO_2$ reduction have been studied using DFT based methods. The obtained computational results are in good agreement with the experimental findings [1].

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# Role of lateral Di-Sulfur Defects in Achieving High Mobility in $MoS_2$ Monolayers

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Defects in semiconductors play a crucial role in modifying their electronic structure and transport properties. In transition metal dichalcogenides (TMDs), atomic chalcogen vacancies are among the most common intrinsic defects. While their influence on electrical transport has been widely explored, their precise role remains not fully understood. In this work, we investigate the impact of chalcogen vacancies in monolayer $MoS_2$ grown by chemical vapour deposition, employing a combination of optical spectroscopy, low-temperature electrical transport measurements, scanning tunnelling microscopy (STM), and first-principles density functional theory (DFT) calculations. We specifically focus on di-sulfur vacancies and demonstrate their significant role in modulating the electronic properties of MoS2. These vacancies introduce shallow donor states near the conduction band edge, which facilitate electron hopping conduction—a mechanism supported by both low-temperature transport data and spatially resolved STM measurements. DFT simulations further reveal that the defect-associated states are relatively delocalized, enabling n-type doping and enhanced conductivity. This defect-assisted transport mechanism accounts for the high field-effect mobility observed in these samples, often exceeding 100 $cm^2 V^{-1}s^{-1}$. Our results provide a clearer picture of how native defects influence charge transport in 2D materials and highlight the potential of defect engineering as a powerful strategy to tailor the performance of TMDs for electronic and optoelectronic applications. [1]

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