Poster session VASP workshop – Monday June 30 2025

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P16 *Machine Learning assisted approach for prediction of the g factors of TMDs heterobilayers* - Galazka Karol

P17 *Impact of a Si(001) substrate on the electronic reconstruction and two-dimensional electron gas formation at $LaTiO_3$ /$SrTiO_3$ (001)* - Darmawan Andri

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P19 *Theoretical Investigation of Intrinsically Patterned 2D Transition Metal Halides: Defects, Structure, and Magnetic Phenomena* - Bisht Neeta

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Doping and topochemical modification of (oxy)chalcogenide photocatalysts

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Keywords; photocatalysts, hydrogen, chalcogenides, band gap

Abstract

Mixed anion compounds have proven great potential in designing new functional materials and enhancing numerous properties [1], including photocatalytic reactions such as water-splitting by shifting them into the visible range. They offer the possibility to tune the band gap and the local structure with the contribution of several anions, with impact on the optical absorption and the charge carriers' properties which are crucial aspects.

More particularly, oxy-chalcogenides appeared in recent years as stable and tuneable alternatives for solar water-splitting photocatalysis [2]. In the meantime, several studies have recently highlighted the interesting role of lone pair cations and how they influence the photocatalytic properties [3]. In that context, this communication will be dedicated to the impact of lone pair cations insertion into two families of (oxy)chalcogenide water-splitting photocatalysts: α-LaOInS$_2$ [4] (doping with Bi^{3+}) and NaInS$_2$ [5].

For the latter, topochemical cationic exchange of Na$^+$ by Sn^{2+}/□ have been carried out as demonstrated in other compounds [3]. The impact on the local structure, the electronic structure (band gap) and the properties will be presented and supported by DFT calculations and crystallochemical analysis to address the structure-properties relationships.

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STUDY OF LITHIUM DIFFUSION IN $Li_xMn_{12}Ni_4O_{32}$ SPINELS

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In lithium-ion battery technologies, the cathode material plays a crucial role in the cell's voltage, capacity and lifetime. The spinel LixN i0.5M n1.5O4 (LNMO) appears as a promising alternative to cobalt-based cathode materials due to its high operating voltage (4.7 V vs. Li+/Li), cobalt-free composition, and relatively low manufacturing cost [2]. The substitution of cobalt with nickel not only reduces dependence on critical resources but also ensures good electrochemical stability of the battery [3].

This spinel exists in two structural forms: an ordered phase with space group P4332, obtained by annealing at 700°C, and a so-called "disordered" phase with space group Fd3m, formed at high temperature (> 800 °C). During battery operation, ionic and electronic conductivity are closely coupled, jointly influencing the kinetics of Li+ insertion and extraction [1]. The performance of LNMO as a cathode material remains strongly linked to the lithium diffusion mechanisms within it. These mechanisms are not yet well understood.

In this study, we focus on the ordered structure of LNMO. We use density functional theory calculations to examine the lithium diffusion mechanisms in the three thermodynamically stable phases: the lithiated phase (x=1), the half-lithiated phase (x=0.5), and the delithiated phase (x=0). Our initial results reveal that at 0 K, in the lithiated and delithiated phases, single motion of lithium is the most energetically favorable mechanism, while in the half-lithiated phase, the collective motion of four lithium ions is preferred. However, diffusion is globally easier in the lithiated phase. These results provide valuable insights for the design of optimized cobalt-free cathode materials, highlighting the importance of crystal structure and lithiation state on lithium mobility.

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Magnetooptical properties of two-dimensional magnetic semiconductors and their heterostructures

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The exploration of two-dimensional magnetic semiconductors (2DMSs) has shown great promise and interest in tuning the magnetic and electronic properties as well as studying magneto-optical effects. Recent studies show that the ordering of atomic magnetic moments in 2DMSs can be tuned by external factors and leads to significant changes in their optical spectra. We employ Density Functional Theory (DFT) and DFT-based Bethe-Salpeter Equation (BSE) to theoretically investigate the magnetooptical properties of 2DMSs, their alloys and heterostructures, with regard to their experimental spectra.

We investigate the impact of the spin direction on the optoelectronic properties of transition metal phosphorus trichalcogenides (MPX_3, M = Mn, Ni, Fe; X = S, Se) exhibiting various antiferromagnetic (AFM) arrangements within the 2D limit. Our analysis reveals large exciton binding energies (up to 1.1 eV), exceeding the values of transition metal dichalcogenides (TMDs). We determine the optically active band-edge transitions, revealing that they are sensitive to in-plane magnetic order. We predict the anisotropic effective masses and the type of linear polarization as important fingerprints for sensing the type of magnetic arrangements. We identify the spin-orientation-dependent features such as the valley splitting, the effective mass of holes, and the exciton binding energy. In particular, we demonstrate that for X=S, Se, a pair of nonequivalent K+ and K− points exists yielding the valley splittings that strongly depend on the direction of AFM aligned spins. Notably, for the out-of-plane direction of spins, two distinct peaks are expected to be visible below the absorption onset, whereas one peak should emerge for the in-plane configuration of spins. We propose a strategy for how the spin valley polarization can be realized in 2D AFM within a honeycomb lattice [1].

Within DFT+U approach we qualitatively explain the origin and the position of the experimentally observed mid band-gap states in layered $MnPS_3$, and corresponding peaks visible in the alloyed systems $Mn_xZn_{1-x}PS_3$. Accordingly, emission at 1.3 eV in all alloyed compounds results from recombination from a $^4T_{1g}$ Mn(II) excited state to a hybrid p-d state at the valence band, as turns out from the analysis of calculated projected densities of states [2].

We theoretically address the recent observation of remarkably strong magneto-excitonic effects in CrSBr, which exhibits optical transitions above the fundamental exciton energy with a colossal spectral shift approaching 100 meV under applied magnetic fields - an order of magnitude larger than previously observed magneto-excitonic responses. DFT-based BSE calculations quantitatively explain the microscopic origin of the observed red- and blueshifts [3].

Motivated by recent optical measurements in magnetic field, we study a $NiPS_3/WSe_2$ heterostructure with various magnetic orderings and stackings. Our DFT+U calculations reveal the presence of interlayer excitons coupled with in-gap 3d states of Ni, as well as an exotic spin texture effecting from Rashba spin-orbit coupling and exchange interactions [4].

Acknowledgments

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Calculating the vibrational spectrum of the GeV defect in Diamond.

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Keywords: DFT, vibrational spectrum, machine learning

As quantum information science advances, the demand for novel devices and materials capable of manifesting quantum effects becomes increasingly important. Diamond color centers are considered excellent candidates as single photon emitters. The interest in the GeV center and other group-IV color centers arises mainly from their intense zero-phonon line and small phonon sideband. [1,2] However, further research is needed to characterize the GeV center under experimental conditions, such as strain, which commonly arises during diamond growth and is known to alter the ZPL wavelength significantly. [3] To better understand these influences, a thorough examination of the phonon sideband is needed to characterise defects and zero-phonon line (ZPL) properties. Density Functional Theory (DFT) calculations provide valuable insights into the vibrational structure of the color center and serve as a powerful complement to experimental studies. [4,5] However, achieving precise vibrational calculations requires tremendous computational resources, often beyond what is available.

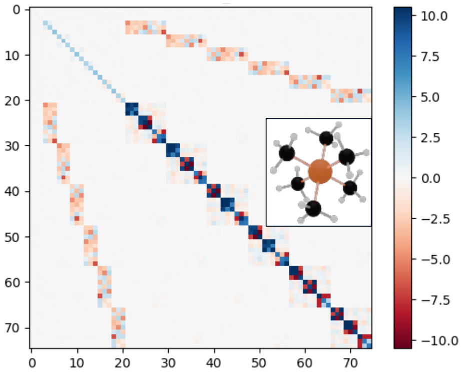

Figure 1: Dynamical matrix of GeC$_6$H$_{18}$ cluster

In this work, we aim to predict vibrational spectra with Density Functional Theory (DFT) enhanced Machine Learning (ML). By combining these methodologies, we aim to address the critical need for accurate vibrational calculations in a computationally efficient manner. As a first step, we focus on the QM9 dataset, which consists of small organic molecules, to develop and benchmark a machine learning framework that can later be extended to solid-state systems. This test set with small molecules is used to develop a machine learning protocol that can be transferred to solids. Our approach relies on redundant internal coordinates (RICs) [6], which describe molecular or solid structures through bond lengths, bond angles, and dihedral angles. The Hessian matrix, initially calculated in Cartesian coordinates, is transformed into the RIC representation. The elements of this transformed Hessian matrix serve as the prediction targets. For each matrix element, we construct a feature vector that incorporates geometric information—such as bond lengths, angles, and dihedrals—along with descriptors of the local chemical environment. Using this setup, we train several machine learning models, including linear regression, random forest, and support vector machines, to learn the relationship between structural features and vibrational properties. The ultimate goal is to transfer this protocol to solids, enabling accurate and efficient vibrational spectra predictions for complex systems like color centers in diamond.

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Optimized Computational Workflow for Exploring Electronic and Structural Properties in 2D TMDs Alloys

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Two-dimensional (2D) materials, with their distinctive electronic, optical, and mechanical properties, have significantly influenced semiconductor research. Among these, transition metal dichalcogenides (TMDs), such as molybdenum disulfide (MoS_2) and tungsten diselenide (WSe_2), exhibit a layer-dependent band structure, strong spin–orbit coupling, and a transition from an indirect bandgap in bulk form to a direct bandgap in the monolayer limit. Alloying—wherein one transition metal is substituted for another—provides a powerful method to tailor properties such as bandgap energy, carrier effective mass, and phase stability. As a result, TMD alloys have gathered attention for next-generation optoelectronic (e.g., photodetectors and light-emitting devices) [1], spintronics (due to spin–valley coupling)[2], and catalytic applications (such as hydrogen evolution reactions)[3], where fine-tuning electronic and optical properties is critical for device performance.

Herein, we present an improved computational workflow for investigating the electronic and structural physical quantities of 2D TMD alloys, with a particular focus on the $Mo_{1-x}W_xS_2$ system. By combining density functional theory (DFT) calculations, symmetry-based reduction of alloy configurations, and band-structure unfolding techniques, our approach significantly lowers computational overhead while retaining high accuracy. A central feature is the use of symmetry analysis to eliminate equivalent dopant configurations, thus providing comprehensive coverage of dopant distributions without exhaustive enumeration of all combinatorial variants.

Applying this methodology to a 3×3 supercell reveals that the doping concentration is the dominant factor in determining key material parameters—most notably the bandgap energy and charge-carrier effective masses. We observe that the bandgap remains direct at the K point for all alloy compositions, exhibiting a pronounced non-linear dependence on the doping fraction, characterised by a clear bowing effect. In addition, subtle correlations emerge between atomic arrangements, total energy, and bandgap values, as well as between dopant distribution and slight anisotropies in carrier effective masses. Nonetheless, our results show that while different alloy configurations display minor variations in their properties, none are significantly more energetically favourable than the others. The approach outlined here provides a systematic and efficient method to explore the composition–property relationships in 2D materials, facilitating the rational design and optimization of next-generation of optoelectronic devisces.

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MoS_2 Nanoparticles Crystallization Modeling: The Molecular Dynamics Study With The Machine-Learning Potential

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Keywords: machine-learning potentials, van der Waals materials, femtosecond laser ablation, heterogeneous nucleation, molecular dynamics.

Abstract

Van der Waals materials, particularly MoS_2 [1], are promising for various technological applications due to their nonlinear optical properties [2, 3]. All simulations were performed using the LAMMPS package [4] with the machine-learning MLIP-2 potential [5], trained on 3,250 configurations. Force, energy, and stress calculations for the training set were conducted using the VASP software package [6], achieving a training error of 16 meV/atom.

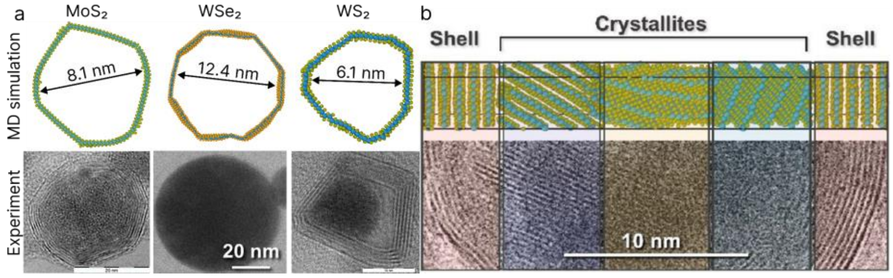

Figure 1. Simulated and experimental images of cylindrical and polygonal nanoparticles (a), and MoS_2 core-shell structures (b).

As cooling rates increase, the size of the core structure and the average crystal size decrease, the shell structure increases. MoS_2 and WSe_2 exhibit high bending moduli, allowing them to sustain significant curvature. In contrast, WS_2 exhibits a lower modulus and critical bending angle, resulting in polygonal nanoparticles as observed in Fig. 1a [1, 2]. This work proposes a hypothesis for the formation of spherical nanoparticles (MoS_2) versus polygonal nanostructures (WS_2), illustrated in Fig. 1b. Authors acknowledge financial support from Russian Science Foundation No. 24-73-10055.

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Amino Acid Functionalized MOF-808 for CO_2 Capture: Unraveling the Host-Guest Interaction via INS Spectroscopy and DFT Calculations

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Abstract

In recent years, the exploration of different materials and technologies for carbon dioxide (CO_2) capture has gained substantial attention in pursuit of sustainable and efficient methods to mitigate anthropogenic CO_2 emissions. The concentration of CO_2 in the atmosphere was 421 parts per million (ppm) until 2022 [1], which is an alarming figure considering that this concentration was 280 ppm at the onset of the Industrial Revolution and the safe level is 350 ppm [2]. The combustion of fossil fuels, such as coal, oil, and natural gas, for energy production is a major source of anthropogenic CO_2 emissions. It is the primary cause of climate change and the greenhouse gas effect [3]. Currently, there are two routes to reduce CO_2 emissions. The first one is direct air capture (DAC) and sequestration [4], and the second one is the reduction of CO_2 emissions from stationary and mobile sources after combustion when the concentration of CO_2 is between 10 and 15%. An appropriate adsorbent with good kinetics, selectivity at trace concentrations, and uptake would make DAC a hands-on methodology to address this global issue [5]. MOFs are promissory solid adsorbents for DAC in terms of selectivity, production costs, and CO_2 uptake that, with a convenient assembly of the building blocks, it could reach adsorption capacities and selectivity features not typically found in traditional CO_2 solid porous adsorbents obtained as a result of unique affinity towards CO_2 molecule. MOF-808 is composed of zirconium metal nodes and trimesic acid as the organic linker. These components form its periodic structure with a characteristic pore of 14 Å. The aim of this work was to functionalize the pore of MOF-808 by employing a post-synthesis procedure with the amino acid Lysine (Lys^{+1}) to enhance carbon dioxide (CO_2) capture. It was observed a noticeable enhancement in adsorption for the MOF-808@Lys^{+1} compared to the bare one, particularly within the pressure range relevant for industrial applications. The adsorption mechanism between the CO_2 and the functionalized MOF was unraveled through inelastic neutron scattering (INS) spectroscopy and DFT calculations. The results indicated that the adsorption mechanism is governed by chemisorption processes where the CO_2 forms carbamic acid with the lateral amine present in the lysine.

Keywords: Inelastic Neutron Scattering, DFT Calculations, Metal-Organic Frameworks, Greenhouse Gases Adsorption

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Molecular Dynamics study of the liquid Fe-O system using a machine learning interatomic potential

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Keywords: molecular dynamics, machine learning, density functional theory, iron oxide, metal oxidation.

Abstract

Predicting the properties of materials under external conditions where experiments are challenging or existing data are ambiguous is one of the primary goals of computational methods such as density functional theory (DFT) [1] and molecular dynamics (MD). In this work, we calculate the thermodynamic and transport properties of the iron–oxygen system at temperatures ranging from 2000 to 3000 K and atmospheric pressure. A wide range of compositions is explored, sufficient to reconstruct a significant part of the phase diagram. The system dynamics is described using a machine-learning interatomic potential [2] trained on data obtained from DFT calculations. The region of immiscibility in the liquid Fe–O system was investigated using molecular dynamics simulations, and the positions of the binodal branches were determined. For homogeneous compositions, density, enthalpy, diffusion coefficients, and viscosity were calculated. The results are in good agreement with experimental data and complement existing knowledge about the phase behavior of the system.

Additionally, the oxidation process of liquid iron was investigated in more detail. MD simulations were carried out to investigate how an oxygen molecule adsorbs onto the surfaces of liquid iron slabs with various compositions. From these simulations, the mass and thermal accommodation coefficients were extracted. Furthermore, the time evolution of oxygen distribution within the system was analyzed, providing insights into the kinetics and spatial characteristics of the oxidation process. This allowed us to characterize the growth rate of the

oxide film and solubility behavior of oxygen in the liquid phase under different thermodynamic conditions.

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Electronic structures of ferromagnetic Heusler alloys Ni_2MnX (Al, Ga, In) using quasiparticle self-consistent *GW*

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Keywords: magnetic shape memory, Heusler alloys, quasiparticle self-consistent *GW*, generalized suscpetibility

Abstract

The essence of the magnetic shape memory (MSM) effect remains unexplained despite all efforts. This effect combines ferromagnetism and ferroelasticity and can reach up to 11% magnetically induced strain in Ni-Mn-Ga, the Heusler alloy in which it was first discovered. The phase transition between austenite and martensite is an essential aspect of the MSM effect. Non-modulated as well as several modulated martensitic phases exist in Ni-Mn-Ga, but only 10M and 14M modulated phases exhibit the MSM effect, suggesting the importance of modulation. An ability to predict a modulation formation may facilitate the search for other MSM materials. However, the MSM effect does not occur in other similar compounds, such as Ni-Mn-Al and Ni-Mn-In, which also form modulated phases, indicating that an additional essential factor enables the MSM effect in Ni-Mn-Ga.

Comparison of the electronic structures of these compounds can provide necessary insight into the differences among them, resulting in the different experimental behavior and, subsequently, the fundamental mechanism of the MSM effect. The electronic structures of the austenite (high-temperature phase) of the ferromagnetic Heusler alloys Ni_2MnX (X = Al, Ga, In) were calculated using the density functional theory (DFT), DFT with Hubbard U parameters (DFT+U), and the quasiparticle self-consistent GW (QSGW) method implemented in ecalj package [1]. One of the studied properties is the generalized susceptibility $\chi(\mathbf{q})$, which can predict the lattice instabilities and the probability of the modulation formation. The generalized susceptibility identifies wave vectors $\mathbf{q}$ where electron scattering is enhanced, indicating possible atomic modulations that require low energy due to Fermi surface nesting. The

generalized susceptibility $\chi(\mathbf{q})$ calculated by the QSGW method, which improves the electron correlation description, exhibits significantly different peaks compared to DFT calculations of Ni_2MnGa [2,3]. A potential instability is present in all studied Heusler alloys Ni_2MnX (Al, Ga, In) [3]. However, the Fermi surface of Ni_2MnIn differs significantly from the other two systems, suggesting that Ni2MnAl could behave similarly to Ni_2MnGa if the $L2_1$ phase is stabilized in Ni-Mn-Al.

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Selective flipping of electronic spins in $BiFeO_3$ via chiral *d-d* excitations

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Keywords: Light-matter interaction, oxides, excitons, spins, optical control, ab initio

Abstract

BiFeO3 is a multiferroic material featuring ferroelectricity and noncollinear antiferromagnetism. Definitive and efficient control of the characteristic spin texture of $BiFeO_3$ is attractive for emerging quantum devices. In this regard, crystal-field *d-d* excitations localized on Fe atomic sites in $BiFeO_3$ provide an avenue for manipulation of the spin texture as they induce a complex interplay among the spin, charge, and lattice degrees of freedom. In this work, the ab initio GW-BSE method is used to characterize these excitations within an excitonic picture. We find that the *d-d* transitions appear as strongly bound, chiral, spin-flip excitons deep within the electronic band gap as a result of the intricate competition between the lattice potential, the antiferromagnetic ordering, the spin-orbit coupling, and the electron-hole interaction. Most crucially, these excitons are composed of electron-hole pairs with opposite spins that constitute almost all of their $\pm\hbar$ total angular momentum. These excitons of specific angular momentum can be selectively excited using circularly polarized light, consequently modulating the local magnetic moment.

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Predicting Band Gaps of Chromium-Based 2D Materials: Accuracy and Pitfalls of GW Methods

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Abstract

Reliable prediction of fundamental gaps in 2D materials is essential for advancing flexible electronic and photonic devices. In Kolos et al. (2025), we assess several GW-based approaches, including self-consistent QPGW, applied to chromium-based 2D systems. We demonstrate that the widely used single-shot G_0W_0@PBE method can yield consistent and accurate gap values when carefully converged, even for complex materials. In contrast, G_0W_0@HSE06 tends to overestimate the gap, especially for strongly correlated or magnetic materials. Striking differences in charge density between PBE and HSE06—specific to chromium and absent in other 2D materials—further highlight the limitations of hybrid functionals as GW inputs in such cases. Our findings emphasize the critical importance of input wavefunction choice and cautious interpretation of GW results for transition-metal-based 2D materials.

Keywords: band gap, 2D materials, GW, PBE, HSE06

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Magnetism and Optoelectronic Response in 2D MXenes: Theoretical Exploration

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Abstract

MXenes, an emerging family of two-dimensional (2D) carbides, nitrides, and carbonitrides, exhibit a diverse array of intriguing properties, rendering them promising for various technical applications. Over 20 MXenes have been experimentally synthesized, and theoretical predictions suggest the existence of numerous others. The properties of 2D MXenes are highly sensitive to compositional variations, termination groups, and external conditions, resulting in a rich spectrum of phases, including metals, semiconductors, ferromagnets, antiferromagnets, topological insulators, and excitonic insulators.

In this work, we focus on *ab initio* prediction and characterization of magnetic MXenes, with emphasis on their magnetic phase stability, anisotropy, and optoelectronic responses. First, we investigate the magnetic and optical properties of the semiconducting oxygen-terminated manganese-based MXene Mn_2CO_2 [1]. We identify a unique ground-state antiferromagnetic (AFM) spin solution consistent with the precursor MAX phase [2], with a preferred magnetization direction along the out-of-plane axis. The optical response, calculated within the advanced many-body Bethe–Salpeter equation (BSE) framework, reveals a remarkably high absorptance (20–30%) in the visible and near-UV range, indicating potential for optoelectronic applications.

Furthermore, we explore the magnetic properties of the mixed-metal MXene $Cr_2TiC_2T_x$, the first experimentally confirmed magnetic MXene [3]. Using density functional theory (DFT) calculations, we map out the relative stability of competing magnetic configurations, including collinear and non-collinear phases. The magnetocrystalline anisotropy is shown to vary significantly with the type of surface functionalization, altering the easy axis of magnetization. In addition, we compute the optical absorptance spectra of the semiconducting AFM phase using the Bethe–Salpeter equation formalism.

Our study provides fundamental insight into the design and control of magnetism in 2D MXenes and emphasizes their potential as a versatile platform for future quantum and spintronic technologies.

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Spin-State Modification and Charge Redistribution at the Co(111)–H_2Pc Interface: A DFT+U Investigation

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Keywords: Density functional theory, charge transfer, hybridization, metallo-molecular interfaces

Abstract

This study investigates the electronic structure, charge redistribution, and magnetic properties of a cobalt (Co) metal slab interacting with a hydrogenated phthalocyanine (H_2Pc) molecule using spin-polarized density functional theory (DFT+U) calculations. The central aim is to elucidate how molecular adsorption influences the surface electronic and magnetic characteristics of Co, with implications for molecular spintronics and interface engineering. A four-layer Co(111) slab was modelled, with and without an adsorbed H_2Pc molecule, to analyze interface-induced effects. Projected and total densities of states (DOS) and Bader charge analysis were used as diagnostic tools to interpret changes in orbital hybridization, magnetic moments, and electronic structure. Our DFT+U approach (U = 0 eV) provides an inexpensive, robust population analysis for the Co-H_2Pc interface, capturing essential physics relevant to experimental observations. We find that adsorption of the molecule induces significant hybridization between Co *d*-states and molecular *p*-orbitals consistent with scanning tunneling spectroscopy and XPS measurements of phthalocyanine–metal interfaces, which show strong molecular–orbital overlap and hybridization effects [1,3]. A strong increase in minority spin electron density near the Fermi level is observed in the Co-H_2Pc system (fig.1a), which leads to a reduction in the local magnetic moment of top-layer Co atoms. This spin polarization is in qualitative agreement with experimental magnetic moment suppression at such interfaces [1,2]. Bader charge analysis reveals a clear pattern of charge redistribution upon molecular adsorption. Surface Co atoms exhibit a charge depletion of $\delta q > +0.4$ e, indicating electron transfer from the Co surface to the molecule. A cumulative charge loss of +4.0 e across all Co layers highlights the significant electronic restructuring due to molecule-surface interaction (see fig.1b). Interestingly, the second and bottom Co layers also exhibit charge perturbations, implying long-range electronic effects [3]. In conclusion, our simulations

demonstrate that H_2Pc adsorption on Co(111) not only modifies magnetic properties via spin-dependent hybridization but also induces significant charge redistribution. These findings are of direct relevance to tailoring molecule-metal interfaces for applications in spin-based devices and molecular electronics.

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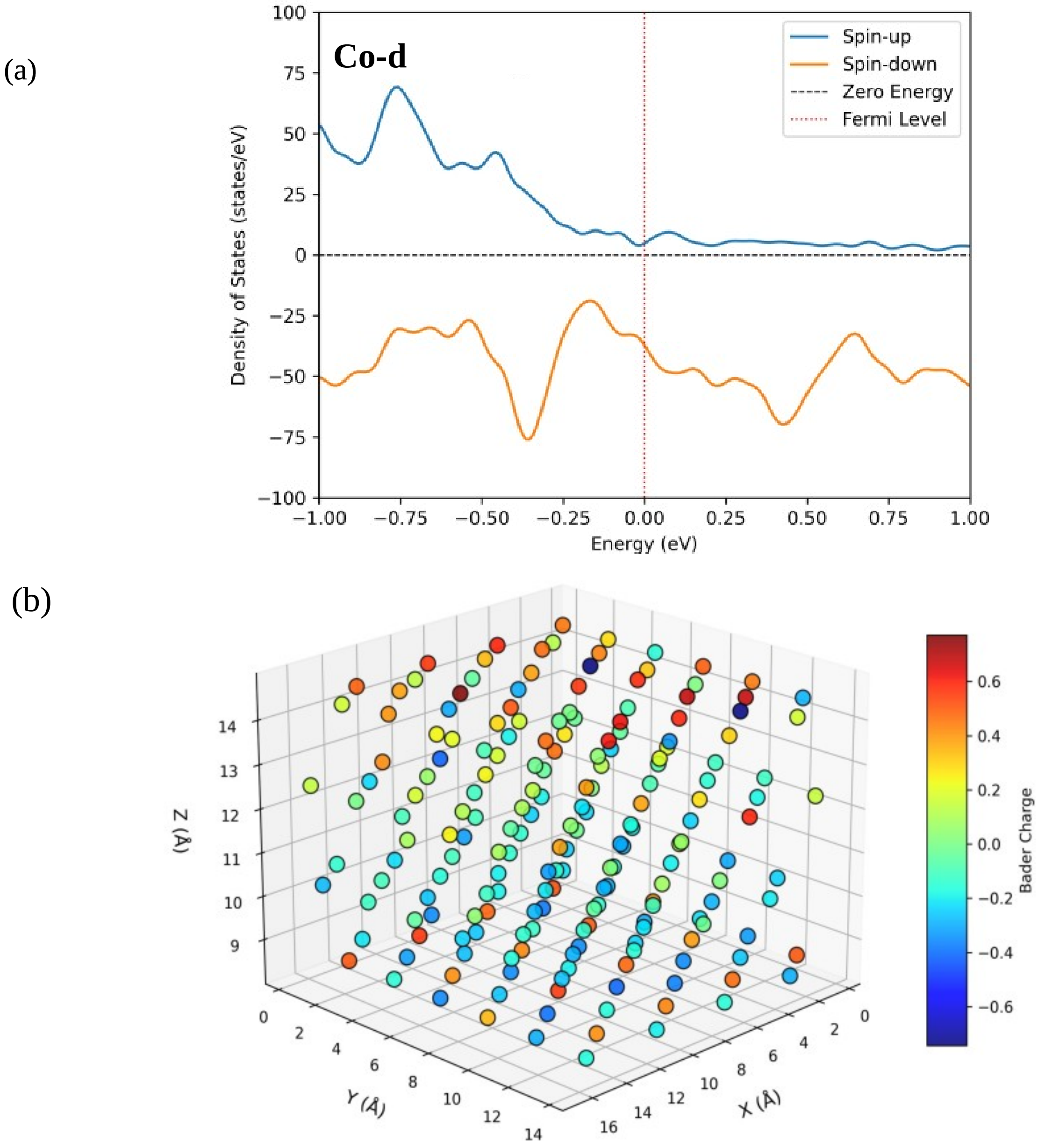

Figure.1 : Hydrogenated phthalocyanine molecule on Co(111) slab ; (a) spin-resolved density of states of Co-d orbitals of top-layer Co atoms, (b) Bader charges of Co atoms and H_2Pc molecule.

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Non-empirical Hybrid Functional for Accurate and Efficient Band Gap Prediction in 3D and Layered Halide Perovskites

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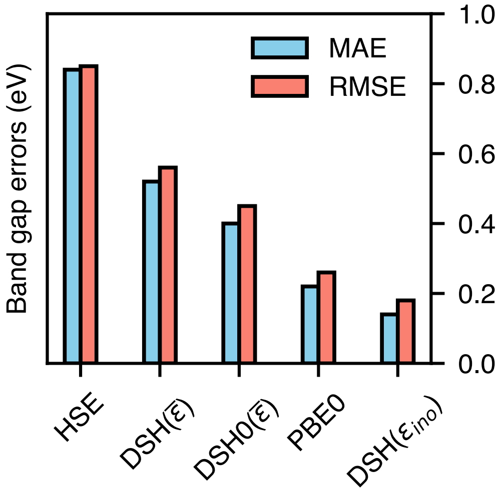
Three-dimensional (3D) and layered halide perovskites have emerged as versatile materials with tunable structural, electronic, and optical properties [1]. An important factor determining the performance and efficiency of materials is the precise control of their band gaps, where accurate predictive simulations can play a crucial role. Hybrid functionals within density functional theory (DFT) are a computationally cost-efficient approach for predicting band gaps of relatively large systems. However, in the case of layered (or quasi-2D) halide perovskites, standard hybrid functionals, like PBE0 and HSE06, can significantly underestimate the experimentally observed band gaps. The usual solution of adjusting the screening parameters based on experimental results undermines their predictive capabilities. Here, we evaluate the performance of the doubly screened dielectric-dependent hybrid (DSH) functional for band gap predictions in Sn- and Pb-based 3D and hybrid halide perovskites [2]. We demonstrate that the DSH functional, which employs material-dependent mixing parameters derived from macroscopic dielectric constants [3], provides accurate band gap predictions for 3D halide perovskites when accounting for structural local disorder in the structures [4,5]. For layered hybrid halide perovskites, we propose using the *ab initio* calculated dielectric constants of their 3D counterparts to define the screening, which leads to accurate results in perovskites with various organic spacers and multilayered structures, such as $BA_2(MA)_{n-1}Pb_nI_{3n-1}$ with n = 1, 2, 3. HSE functional significantly underestimates band gaps compared to PBE0 in both 3D and layered halide perovskites due to the absence of non-local long-range screening. The computational method proposed in this work provides an accurate and efficient framework for investigating 3D and 2D perovskites, enabling the exploration of 3D/2D heterostructures that are currently out of reach within existing *ab initio* frameworks.



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Ab initio investigation of electronic properties of Cl-doped CrSBr.

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CrSBr is known for its strong excitonic effects and pronounced magneto-optical phenomena [1]. The excitons in CrSBr are highly sensitive to both the in-plane anisotropy and the magnetic order of the layers. By alloying, the balance between intralayer exciton recombination and interlayer charge-transfer (CT) exciton contributions can be tuned [3]. For instance, Cl substitution has been demonstrated to modify the extent of electron–hole wavefunction overlap between adjacent layers, thereby influencing the CT character of the excitons [2]. This manipulation is crucial because even if the optical bandgap remains nearly constant, the oscillator strengths, binding energies, and radiative lifetimes of excitons can be dramatically altered.

Moreover, since the magnetic and optical properties in CrSBr are strongly coupled, alloying-induced structural changes can modulate magnetic anisotropy and exchange interactions [2,3]. These changes can be probed by the magneto-optical measurements, such as the magnetic field-induced exciton shift observed in photoluminescence experiments. Thus, alloying serves as an effective tool not only for fine-tuning optical transitions but also for indirectly controlling magneto-optical response.

In this study, we theoretically investigate particular Cl concentrations and various non-equivalent positions of Cl atoms at Br sites within the framework of density functional theory (DFT). Our results reveal that Cl substitution induces anisotropic lattice compression, most notably along the a- and c-axes. Additionally, the orbital contributions to band-edge transitions are affected, which could lead to modifications in excitonic properties. As a result, the energy levels of excitonic transitions could be subtly shifted, enabling precise control over photoluminescence and absorption spectra.

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Machine Learning assisted approach for prediction of the *g* factors of TMDs heterobilayers

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Continued progress of technology and industry necessitates the development of tailor-made materials for specific purposes. In recent years the vast class of two-dimensional (2D) van der Waals (vdW) layered materials have been extensively studied. Among them, Transition Metal Dichalcogenide (TMD) semiconductors have been subjects of particular interest in electronics and optoelectronics. [1]

In principle desired properties could be obtained by vertically stacking various types of monolayer (ML) crystals with arbitrary material sequence and twist angle between layers, owing to their synergistic effects. [2,3] Although the interest in this pursuit is continuously increasing, the lack of high-throughput techniques for fabricating high-quality vdW heterostructures (HTs) hinders its research. The speed of the process could be expedited with the use of theoretical calculations to predict promising structures with desired properties. The *g* factor that quantifies the response of excitons to external magnetic field is one such property, that has been widely studied in the framework of the density functional theory for MLs of TMDs. [3] However, *g* factor research for HTs is limited due to requirement of large number of conduction bands to achieve convergence. [3] The stacking layers and twist angles dependence further complicate the research.

This high computational demand and labor intensive process of prediction limits the pace of progress in the field. Many TMD monolayer phenomena have been extensively examined in relation to the *g* factor. Namely, experimental studies of exciton complexes, excited Rydberg states etc., and theoretical computations based on electronic structure features. The idea is to investigate and utilise their predictive power using Machine Learning Methods (MLMs), applying this knowledge to TMD vdW HTs.

At first an experimental database was constructed, where all available data from publications was collected and considered. It was then cleaned, all damaged or incomplete records were removed or completed and descriptors of records were established. Next a variety of Decision Tree Regressor based algorithms were fitted to the data. This yielded unsatisfactory results, owing mainly to large inconsistencies between publications and an insufficient amount of published data. Hence, another approach is investigated by creating a dataset based on high throughput *ab intio* computations of TMDs-based alloys.

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Impact of a Si(001) substrate on the electronic reconstruction and two-dimensional electron gas formation at $LaTiO_3$ /$SrTiO_3$ (001)

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The two-dimensional electron gas (2DEG) formed at oxide interfaces e.g. between the band insulator $SrTiO_3$ and the Mott insulator $LaTiO_3$ has attracted a lot of attention [1-3]. However, despite the high carrier density at the interface, the carrier mobility is lower compared to semiconductor materials at room temperature [4]. A strategy to overcome this shortcoming is the integration of the oxide system on a semiconductor substrate [5]. Based on density functional theory calculations with a Hubbard U term we modeled $LaTiO_3$/$SrTiO_3$(001) with and without a Si(001) substrate. We explore systematically the effect of the termination to Si(001) as well as the effect of a $SrTiO_3$ capping layer on the electronic reconstruction at $LaTiO_3$/$SrTiO_3$(001) interface. The comparison between the systems with and without a Si(001) substrate indicates lower effective masses and consequently higher mobility of the 2DEG at $LaTiO_3$/$SrTiO_3$/Si(001).

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Tunable Skyrmion Formation via Interfacial Dzyaloshinskii–Moriya Interaction in Two-Dimensional Heterostructures

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Abstract

Two-dimensional magnetic heterostructures have emerged as a versatile platform for engineering novel spin textures such as magnetic skyrmions, which are promising candidates for low-power spintronic devices. In this work, we investigate the electronic and magnetic properties of SnC/MnSeX (X = Se, Te) heterobilayers using first-principles density functional theory and atomistic spin simulations, with a particular emphasis on the stabilization and tunability of Néel-type skyrmions [1]. The inclusion of a SnC monolayer significantly breaks inversion symmetry in the system, leading to a remarkable enhancement of the interfacial Dzyaloshinskii-Moriya interaction (DMI). The computed DMI values reach up to ±3.05 meV, comparable with values observed in well-known skyrmion-hosting systems like Co/Pt and Fe/Ir, enabling the formation of compact and controllable skyrmions. We find that magnetic anisotropy, exchange coupling, and skyrmion radius can be further modulated via external electric and magnetic fields. Among the studied configurations, SnC/MnSe$_2$ exhibits half-metallicity and a significantly increased Curie temperature (~229 K), along with the highest skyrmion collapse energy barrier, making it especially suitable for practical implementation. The |D/J| ratios in all systems exceed the critical threshold for skyrmion formation, with phase diagrams confirming skyrmion lattice and isolated skyrmion states at accessible field and temperature ranges. These results highlight the promise of SnC-based magnetic heterobilayers for robust, tunable topological spin textures, paving the way for their integration into next-generation spintronic technologies, including high-density data storage and neuromorphic computing.

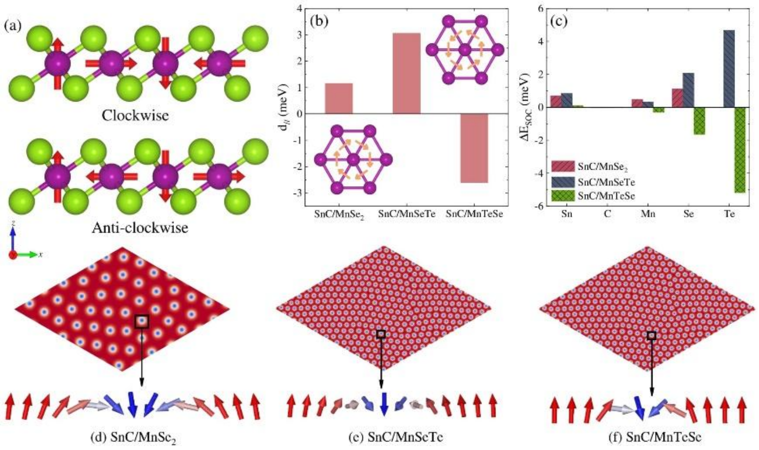

Figure Néel-type skyrmions stabilized by interfacial Dzyaloshinskii–Moriya interaction in SnC/MnSeX heterobilayers.

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Theoretical Investigation of Intrinsically Patterned 2D Transition Metal Halides: Defects, Structure, and Magnetic Phenomena

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Abstract

In the quest for complex structured functional materials, defect engineering and patterning in two-dimensional (2D) systems are critical for tuning material properties and enabling new functionalities[1]. Herein, we report on intrinsically patterned 2D transition metal dihalides (TMDs) on a gold surface, featuring periodic halogen vacancies in the upper and bottom halide layers that result in alternating coordination of the transition metal atoms throughout the film.

Employing Density-Functional Theory (DFT) through the Vienna Ab-initio Simulation Package (VASP), we explore the formation pathways leading to periodic halogen vacancies and their role in modifying the electronic and magnetic structure of TMDs. The defect-engineered vacancy lattice not only stabilizes the structure but also provides atomic-level control over the material's properties. Our calculations, enhanced by VASP's robust handling of non-collinear magnetic textures and its accurate simulation of magnetic anisotropy energies, show that Br vacancies are energetically favorable, and the resulting vacancy lattice significantly reduces the lattice mismatch with the underlying Au(111) surface[2]. The structure formation of the 2D $FeBr_2$ on the surface and the presence of defects are further analyzed using STM simulations. The excellent match between the experimental findings and the DFT calculations, facilitated by VASP, confirms the intrinsic vacancy lattice. The electronic band structure accurately captures the Br and Fe contributions in both the pristine and halogen vacancy lattices.

Moreover, our spin-polarized DFT calculations predict the emergence of unconventional magnetic textures, driven by the interplay of defect-induced strain and transition metal coordination. The calculated magnetic anisotropy energy for pristine $FeBr_2$ favors an out-of-plane orientation,

while the introduction of vacancies shifts the preferred magnetization to an in-plane direction, demonstrating that vacancies can tune the magnetic properties of the films. By coupling our theoretical results with experimental observations, we provide a comprehensive framework for understanding the structure formation, and electronic and magnetic properties of 2D materials, advancing the design of complex structured materials with tunable properties.

Keywords: 2D, TMDs, DFT, STM, VASP, $FeBr_2$

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VASP Workshop 2025

Rennes, France, from June 30th to July 4th, 2025

The Structural, Mechanical and Dynamical stabilities, Electronic, Lattice thermal Conductivity and Optical properties of CsPbI3: Theoretical Study

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The structural, mechanical, electronic, lattice thermal conductivity and optical properties of the orthorhombic perovskite phase of $CsPbI_3$ have been investigated using density functional theory (DFT). Elastic parameters bulk modulus B, Young's modulus E, shear modulus G, Poisson's ratio v and anisotropy value A have been calculated by the Voigt-Reuss-Hill averaging scheme. Phonon dispersions of the structure were investigated using a finite displacement method. The relaxed system is dynamically stable, and the equilibrium elastic constants satisfy all the mechanical stability criteria for orthorhombic crystals, showing stability against the influence of external forces. The lattice thermal conductivity was calculated within the single-mode relaxation-time approximation of the Boltzmann equation from first-principles anharmonic lattice dynamics calculations. Our results show that lattice thermal conductivity is anisotropic, and the corresponding lattice thermal conductivity at 300 K was found to be 0.153, 0.354, and 0.106 $Wm^{-1}\ K^{-1}$ in the *a*, *b* and *c*-directions. Our DFT calculations showed that $CsPbI_3$ possess DFT direct band gaps within the range of (2.36 to 2.52) *eV*, depending on the exchange-correlation approximation used. Many-body perturbation theory at the G_0W_0 level of approximation gave a fundamental band gap of 3.67 *eV*. In order to obtain optical spectra, we carried out Bethe-Salpeter equation calculations on top of a non-self-consistent G_0W_0 calculations. Our calculated optical band gap shows anisotropy with an absorption edge of 2.59 *eV* in the *a*-direction, 2.39 *eV* in the *b*-direction and 2.49 *eV* in the *c*-direction. Our result shows that $CsPbI_3$ compound has a wide band gap with potential applications in multi-junction solar cells.

Integrating Machine Learning and Density Functional Theory for Predicting and Understanding Ion Doping in Hydroxyapatite

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Hydroxyapatite (HAp) is a crucial biomaterial for bone implants, requiring precise control over its crystallinity and surface stability. This study integrates Machine Learning (ML) for crystallinity prediction and Density Functional Theory (DFT) for understanding the thermodynamic stability of doped HAp. Utilizing a dataset of 63 data points, ML models (KNN, CatBoost, XGBoost, ANN) predicted HAp crystallinity under the influence of various ion dopants (Sr^{2+}, Zn^{2+}, Ag^{+}, F^{-}, Ni^{2+}, Fe^{3+}, Er^{3+}, B^{3+}, Al^{3+}, Ba^{2+}, W^{6+}, Mg^{2+}), sintering temperature, and time. XGBoost showed the best prediction performance with an R^2 of 95.6%, effectively handling the dataset's bimodal distribution. ML feature analysis indicated that sintering temperature and time were the most dominant factors controlling crystallinity, yet ion doping consistently influenced crystallinity (e.g., Mg^{2+} significantly reducing it) while playing a vital role in osteoconductivity. In parallel, ab initio DFT calculations on HAp doped with Mg^{2+}, Zn^{2+}, Al^{3+}, and Si^{4+} revealed a thermodynamic stability order: $Si^{4+} < Al^{3+} < Mg^{2+} < Zn^{2+}$, with Si^{4+} doping yielding the most stable system. This stability also implies enhanced adhesion energy with titanium, consistent with the literature. The integration of both approaches provides a holistic understanding: ML predictions are validated and enriched by DFT's mechanistic explanations. This study advances HAp biomaterial design by providing predictive tools and clarifying fundamental relationships between ion doping, crystallinity, and thermodynamic stability.

Keyword: Hydroxyapatite, Ion doping, Machine Learning, Density Functional Theory, Crystallinity, Ground State Energy