Vienna Ab-initio Simulation Package (VASP) and Applications

Book of Abstracts

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Opening Talk

Machine-learning-assisted determination of the global zero-temperature phase diagram of materials (IT)

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Crystal-graph attention neural networks have emerged recently as remarkable tools for the prediction of thermodynamic stability. The efficacy of their learning capabilities and their reliability is however subject to the quantity and quality of the data they are fed. Previous networks exhibit strong biases due to the inhomogeneity of the training data. Here a high-quality dataset is engineered to provide a better balance across chemical and crystal-symmetry space. Crystal-graph neural networks trained with this dataset show unprecedented generalization accuracy. Such networks are applied to perform machine-learning-assisted high-throughput searches of stable materials, spanning 1 billion candidates. In this way, the number of vertices of the global $T = 0$ K phase diagram is increased by 30% and find more than ≈150 000 compounds with a distance to the convex hull of stability of less than 50 meV atom^{-1}. The discovered materials are then accessed for applications, identifying compounds with extreme values of a few properties, such as superconductivity, superhardness, and giant gap-deformation potentials.

Day 1 - Session 1

High throughput first-principle prediction of tribological properties of solid-solid interfaces using VASP (CT)

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The tribological properties of natural and artificial solid-solid interfaces become relevant whenever two materials come into contact while moving laterally to each other. They are thus key quantities in a variety of fields ranging from geology to nanotechnology including, of course, energetic efficiency.

An ab-initio determination of the tribological properties of solid-solid interfaces is crucial because it is the specific chemistry of the interface that primarily governs adhesion, a measure of the strength by which two surfaces bind together. It also governs the generalized stacking fault energy surface which describes the variation of adhesion between two surfaces as a function of their relative lateral position, ultimately determining the theoretical shear strength, i.e. the interface resistance to a shear load.

The atomistic simulation of heterogeneous interfaces is challenging, as computations can quickly become very expensive when two different lattices must be matched together in the same supercell. To face these challenges our group developed TribChem [1], a software specifically designed to perform the high-throughput study of solid-solid interfaces. The software is composed of three main units for the study of bulks, surfaces, and interfaces. It is entirely written in Python and uses different packages from the Materials Project [2]. To perform the density functional theory (DFT) calculations, TribChem presently relies on the Vienna Ab initio Simulation Package (VASP) [3, 4].

In this work I will introduce the theoretical framework in which Tribchem is developed, and starting from the original version of the code [4] l will give an overview of its current structure and of the general workflow that is implemented. I will also show how Tribchem has been applied for the high-throughput determination of the adhesion energy of around more than a hundred metallic heterostructures, ranging from transition to noble metals [5].

These results are part of the SLIDE project that has received funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation program (Grant agreement No. 865633).

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MPS_3 and MPS_4 - The interplay between structure and magnetic ordering (CT)

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Magnetic van der Waals materials, such as transition metal phosphorous chalcogenides MPS_3 and MPS_4 (M = Mn, Fe, Co, and Ni), offer potential for coupled spintronic and optoelectronic applications due to their wide band gap (1.2 - 3.5 eV) [1] and efficient light absorption. While both belong to the C2/m space group, they differ in magnetic ordering, with MPS_3 preserving antiferromagnetic (AFM) ordering [1] and $CrPS_4$ changing from AFM to ferromagnetic (FM) in the monolayer limit [2]. However, the mechanism and role of exchange interaction still need to be clarified, especially in the case of MPS_4. This work investigates and compares the magnetic states that arise in MPS_4 to the MPS_3 class.

We studied the stability and electronic properties of the MPS_4 and MPS_3 families using DFT, focusing on magnetic properties. We used VASP with PAW and PBE+D3, and optB86-vdW functionals, as well as the SCAN functional. We used the GGA+U approach with Dudarev formalism [3] for the magnetic structure, testing different values for U. The exchange parameters for a Heisenberg Hamiltonian were then derived with the TB2J Python package by downfolding the DFT bands in the vicinity of the Fermi energy employing Wannier functions.

First, we studied $CrPS_4$ and found an FM ground state with a magnetic moment of 3.0 μB/magnetic atom, in agreement with the literature [4]. The exchange parameters of $CrPS_4$ were analyzed in detail, providing insight into the competition between different exchange-interaction mechanisms. We explain why this material, different from MPS_3, has an FM order in the monolayer.

Second, we compared the exchange interaction in $CrPS_4$ and $NiPS_3$. For $NiPS_3$, it is already established that the distance between neighbors is crucial [5]; our results showed that the essential interaction is the AFM superexchange with the third neighbor that overcomer all the others because of the alignment of sulfur atoms in a plane that offers a favored path for the interaction which is in agreement with previous studies [5]. Regarding $CrPS_4$, the long-range interaction is also meaningful; however, the structural differences lead to FM order.

In conclusion, our study of $CrPS_4$ confirmed the FM ground state and magnetic moment found in previous literature. Besides that, we can now explain why $CrPS_4$ is FM, and we envision that this can now be influenced by targeted modification.

Automated workflows for materials discovery (IT)

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Automated ab initio calculations have emerged as a powerful tool for computational materials science. Automated workflows offer many benefits over traditional manual approaches, including reproducibility for complex multi-step calculation procedures, scalability across tens of thousands of compounds, and usability through well-tested default values. Atomate2 is a library of over 100 computational materials science workflows and is the infrastructure that powers the Materials Project database. Atomate2 supports a range of density functional theory packages (VASP, CP2K, ABINIT, FHI-aims) and integrates with common machine learning force fields, AMSET for electron-phonon and electronic transport calculations, phonopy for finite temperature properties, Lobsterpy for bonding analysis, and many other codes. This talk will introduce atomate and the benefits and best practices for users wishing to get started with automated workflows.

2D transition metal dichalcogenides based bilayer heterojunctions for efficient solar cells (CT)

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In this work we present a first-principles investigation on the optoelectronic properties of vertically-stacked bilayer heterostructures composed of 2D transition-metal dichalcogenides (TMDs). Our aim is to propose these TMD heterostructures for potential applications in solar cells. The TMD monolayers comprising the heterojunctions considered in this research are MoS_2, WoS_2, $MoSe_2$, and WSe_2 due to their favorable band gaps, high carrier mobility, robust absorption in the visible region, and excellent stability. These four TMD monolayers provide the basis for a total of six potential heterostructures. However, not all of the considered heterostructures meet the essential criterion of type II band alignment, a critical factor in extending carrier lifetime. Consequently, we have exclusively examined the structural, electronic, and optical properties of five heterostructures (WS_2/MoS_2, $MoSe_2/WS_2$, WSe_2/MoS_2, $WSe_2/MoSe_2$, and WS_2/WSe_2) that fulfill the type II band alignment condition. These chosen heterostructures exhibit superior optoelectronic properties compared to their respective isolated monolayers. The effective masses are also calculated for the considered TMDs heterojunctions. Remarkably lower electron and hole effective masses are obtained which is important to oppose the further recombination of photogenerated charge carriers. The lower probability of carrier recombination is desirable for enhancing the performance of solar cells. The built-in electric field is also obtained which results in due to horizontal mirror asymmetry in the considered heterostructures. Thus, by carefully designing Type II heterostructures using TMD monolayers, it is possible to tailor the energy bandgap and absorption properties, allowing for the efficient absorption of a broader spectrum of sunlight. The quality of these heterojunction solar cells is estimated by computing their power conversion efficiencies (PCE). The maximum PCE of our designed solar cells can reach up to 21.23% for the WS_2/WSe_2 heterojunctions.

Insights from first principles calculations into low lattice thermal conductivity materials for thermal energy applications (CT)

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The present study employed temperature dependent effective potential (TDEP) in conjunction with first principles Born-Oppenheimer molecular dynamics simulations and Boltzman transport theory to systematically investigate the impact of mass contrast and tensile lattice strain on lattice dynamics and phonon transport properties. We predict that low-lying optical phonons contribute considerably to lattice thermal conductivity (kL) in materials with a mass ratio near to unity (for example, SrSe, BaTe) compared to materials with a large mass ratio (for example, BaO, SrO). Materials that have a mass ratio near to one exhibit lower scattering rates, which in turn leads to longer phonon lifetimes and an increase in kL. Anomalies in kL are seen for both alkali halides and alkaline-earth chalcogenides. In addition, the application of lattice strain leads to a further decrease in kL. A deeper understanding of the role that mass contrast and tensile lattice strain play in lattice dynamics and phonon transport properties is provided by the detailed study on binary systems. This understanding is essential for the design and development of functional materials for applications that involve sustainable thermal energy management.

Halogen modification of SACs for CO_2 reduction reaction (CT)

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Electrochemical CO_2 Reduction Reaction (CO_2RR) is one of the most promising strategy to relieve the atmospheric CO_2 concentration whilst producing useful fuels/chemicals such as formic acid, methanol, and methane as reaction products. However, the CO_2RR process constitutes of multiple proton-electron transfer reactions competing with the Hydrogen evolution reaction (HER). It is desirable to develop energy efficient heterogeneous electrocatalysts for CORR based on low-cost and earth-abundant metals. In this regard, the third-abundant metal, Manganese based electrocatalysts are shown to have better activity for CO_2RR. Also, the minimal use of metal in catalyst with enhanced activity can be drastically bring down the cost of the catalyst. This is possible if the metal atoms are stabilized as single atom active sites in support materials. The graphitic carbon nitride (g-C_3N_4) is theoretically shown to be suitable for stabilization of a single atoms of V, Cr, Zr and Mn at the center of the voids available between the heptazine units. Interestingly, the metal atoms present in the voids coordinate with six N atoms of surrounding heptazine motiffs. We done a detailed study on Mn@g-C_3N_4, as it shows good absorbance towards CO_2 comparing to other metals using first principle calculations. We tried to modify this active site, so it effectively inhibits HER. Our study shows doping of halides strongly inhibits HER, as there is a downward shift in d-band center of Mn atom when halides are attached. The first protonation happens at C atom of CO_2, which prohibits the CO formation, and the primary reaction pathway is through formate intermediate. Interestingly the Gibbs free energy calculations for product formic acid would be formed without any applied potential. Methanol and methane can be produced as secondary products with subsequent protonation on HCOOH, when applied low potential (0.32-0.56 V). Thus, this study opens an opportunity for improving the designing of metal-nitrogen-halide active site for CO_2RR on SACs.

Modelling the infrared and Raman fingerprint of phosphate adsorption on ceria surfaces (IT)

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We will consider the case of cerium dioxide, an important material for catalyses such as water gas shift reactions, three-way catalysis, soot oxidation, and enzyme mimetic activity (nanozymes). In such catalytic applications, the performance of the catalyst is dependent on the species adsorbed at the surfaces. For example, effective nanozymes need to retain their activity independently of their surface speciation in biological environments. Characterizing surface speciation is therefore crucial to understanding and controlling nanozymatic capabilities. Adsorbed phosphates impact the nanozymatic activity to convert phosphate prodrugs into therapeutics, and define the early stages of phosphate-scavenging activity that leads to the transformation of active cerium oxide into inactive cerium phosphate. In this work, we demonstrate that ab initio lattice-dynamics calculations can be effective to study the interaction of phosphates with the three most stable surfaces of cerium oxide and to predict the infrared (IR) and Raman spectral signatures of adsorbed phosphate species. We find that phosphates adsorb strongly onto cerium oxide surfaces in a range of stable binding configurations. 5-fold coordinated P species in a trigonal bipyramidal coordination are also present at the surface as stable configurations and may represent a stable intermediate in the early stages of phosphate scavenging. We find that the phosphate species show characteristic spectral fingerprints between 500 and 1500 cm^{-1}, whereas bare cerium oxide surfaces show no active modes above 600 cm^{-1}, and the 5-fold coordinated P species in particular show potential diagnostic P–O stretching modes between 650 and 700 cm^{-1} in both IR and Raman spectra. This provides an important step toward the experimental characterization of phosphate speciation and, ultimately, control of its impact on the performance of cerium oxide nanozymes.

Day 2 - Session 1

VASP-based simulation of scanning tunneling microscopy (IT)

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Understanding and engineering scanning tunneling microscopy (STM) image contrasts and scanning tunneling spectroscopy (STS) is of crucial importance in wide areas of surface science and related technologies, ranging from magnetic surfaces to molecular structures. In the talk a few electron tunneling theories are introduced for the purpose of high-resolution simulation of STM and STS. These theoretical methods rely on electronic structure data obtained from first principles, and are implemented in two STM program packages: BSKAN [1,2] and 3D-WKB-STM [3]. A practical introduction to the usage of these STM simulation codes based on VASP data is briefly given, followed by selected examples on different STM tip effects on the STM imaging mechanism: (i) the role of tip-orbital interference within the revised Chen's derivative rules [4], and (ii) asymmetric/tilted tip orientation effects [5]. Going beyond scalar charge current in spin-polarized STM, a combined tunneling electron charge and vector spin - longitudinal spin current and spin transfer torque - transport theory is presented, with application to magnetic skyrmions [6].

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A theoretical method for angle-resolved photoemission of two-dimensional systems based on repeated-slab projector-augmented wave functions (CT)

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We report the development of a computational method for angle-resolved photoemission spectra of two-dimensional systems based on the electronic wave functions from VASP. We obtained the photoelectron wave functions by matching the eigenstates of a slab with free wave states in the vacuum. The constructed wave functions fulfill the desired boundary conditions. We have applied the method to graphene and analyzed the normal emission spectra for p-polarized light varying the photon energy. We calculated the photoelectron wave functions within the projector-augmented wave approach and compared the pseudo-waves and all-electron waves. The photoemission intensity calculated with the all-electron waves agrees well with the one obtained with the multiple scattering method. We found a strong photon energy dependence on the photoelectron intensity. In particular, our calculations predict a resonance for the π-band normal emission. The resonance is due to 2D interband transitions and highlights the importance of the final state effects beyond the plane-wave approximation.

Topological phase transition in isostructural $SnPbSe_2$: A first-principle study (CT)

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The relationship between topological band theory and accurate band structure calculations plays an important role in searching topological phases in real materials. Thallium based III-V-VI series of ternary chalcogenides i.e., $TlBiX_2$ and $TlSbX_2$ (X=S, Se, Te) have shown to exhibit topological non-trivial nature [1-2]. Here, we used first principles calculations and the green function approach to study an isostructural material i.e., $SnPbSe_2$ of the Thallium chalcogenides. We used TB-mBJ [3] functional with spin-orbit coupling (SOC) to analyse the topological behaviour of this material at ambient conditions and under hydrostatic pressure. We predicted that $SnPbSe_2$ shows a trivial to non-trivial transition with single band inversion at 18 kbar of hydrostatic pressure. This non-trivial topological nature is also verified by presence of a single Dirac cone in the surface states. Further, we calculated the Z_2 topological invariants using the product of parities analysis at time-reversal invariant momenta (TRIM) points in Brillion zone (BZ). The non-zero values of first Z_2 invariant also supports our claim. The increase in hydrostatic pressure to 23 kbar shows an even number of band inversions which can be seen as two Dirac cones in the surface states. This makes the material topological trivial at 23 kbar pressure. So, this material behaves as a topological insulator between 18 to 23 kbar of pressure.

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2D BN-Biphenylene: Structure stability and properties tunability with DFT perspective (CT)

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In process of exploring the counterparts of an existing structure using Density functional theory and ab initio molecular dynamics, we created BN-analog of a recently synthesized 2D Biphenylene (BPh) sheet. For the theoretical validation of a novel structure, we first need to check its stability. We employed four different approaches in our works: cohesive energy, dynamical, thermal, and mechanical stability. These have been confirmed by: (i) comparing cohesive energy of other similar material, (ii) calculating phonon spectrum, (iii) ab-initio molecular dynamics (AIMD), (iv) satisfying Born criteria. Phonon-spectrum and AIMD show all positive frequencies and negligible variations in structure (bond length), kinetic, and total energy for 5ps, respectively. Having found stable BN-BPh, we analyzed the electronic, mechanical, optical, and vibration properties of BN-BPh and compared them with those of BPh. We found the following: (1) Electronic properties (density of states and band structure) show that BN-equivalent of metallic BPh opens a large band gap of 3.12 eV and 4.48 eV with PBE and HSE06 DFT functional, respectively, similar to graphene BN-equivalent (hBN) with a band gap of 6.1eV. (ii) The mechanical strength of BN-BPh is close to that of BPh with a difference of ~15 N/m and ~10 N/m along x and y-directions, and its Young's modulus indicates BN-BPh is stiffer than BN-graphyne. (iii) The optical absorption coefficient of BN-BPh shows the first peak at 4.48 eV (3.12eV) for HSE06 (PBE) functional. (iv) By substituting external atoms such as P, Al, As, C, Li, Mg, S, and Si to B and N atoms in BN-BPh, we could tune its electronic properties to semiconducting, semi-metallic, and metallic.

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Day 2 - Session 2 - Posters

Study of magnetic and magnetocaloric properties of Gd_3In material (P)

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This abstract presents a synthesis of my graduation project conducted at the Laboratory of Condensed Matter and Interdisciplinary Sciences, Faculty of Science (FSR), University Mohammed V-Rabat, Morocco. The primary aim of this research is to conduct an Ab initio study on the magnetic and magnetocaloric properties of Gd_3In material utilizing the Quantum Espresso, Wien2k code, and VASP.

Gd_3In is distinguished by a remarkable magnetocaloric effect, a crucial physical attribute underlying the cooling or heating of specific materials when subjected to a magnetic field. One notable application of this effect is in magnetic refrigeration, an emerging technology for cold production that serves as a cleaner alternative to traditional methods employing environmentally harmful refrigerants.

To achieve the project's objectives, a comprehensive set of tools was employed. The Quantum Espresso, Wien2k, and VASP codes were utilized for calculating structural, electronic, and magnetic properties. Furthermore, thermodynamic properties were analyzed through Monte Carlo simulation, culminating in the calculation of magnetocaloric properties. This research contributes to the understanding of Gd_3In's unique characteristics and potential applications in innovative cooling technologies.

Predicting novel 2D AsBiX_3 (X = S, Se, and Te) auxetic monolayers with favorable optical and photocatalytic water-splitting properties (P)

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The design of two-dimensional multifunctional materials is highly desirable for nanoscale device applications. In this study, we report the structural, electronic, mechanical, and photocatalytic properties of chalcogenide-based monolayers AsBiX_3 (X = S, Se, and Te) using first-principles calculations. The stability of these monolayers is confirmed through energetic and mechanical analyses, as well as ab initio molecular dynamics simulations. The analysis of mechanical properties reveals significant mechanical anisotropy and a bidirectional in-plane negative Poisson ratio in the monolayers. Additionally, the computed electronic band structures, obtained with and without spin–orbit coupling, indicate that these monolayers are indirect-gap semiconductors. At the Heyd–Scuseria–Ernzerhof level, the values of the band gap are determined to be 1.91 eV for $AsBiS_3$, 1.66 eV for $AsBiSe_3$, and 1.32 eV for $AsBiTe_3$. These monolayers have a very high absorbance on the order of $\sim 5 \times 10^5$ cm^{-1} in the visible and ultraviolet regions with considerable anisotropy. We also found that monolayers hold a high mobility anisotropy. The predicted solar-to-hydrogen efficiency of all monolayers surpasses the critical value (>10%) for the economical production of hydrogen from photocatalytic water splitting. Notably, $AsBiS_3$ and $AsBiSe_3$ monolayers have appropriate band-edge positions that perfectly match the conditions for photocatalytic water splitting at pH = 0, and the band gap and band-edge positions can be adjusted through strain engineering. With these outstanding properties, AsBiX_3 (X = S, Se, and Te) monolayers present themselves as promising candidates for applications in optoelectronics, mechanics, and photocatalytic water splitting.

Synthesis, photocatalytic activity and electric dipole formation in bismuth oxychloride $Bi_xO_yCl_z$: Experiment and theory (P)

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Layered bismuth oxyhalides $Bi_xO_yCl_z$ are known to have outstanding visible-light photocatalytic activity was studied in this paper. Our investigation indicated that the Bi_3O_4Cl, $Bi_{24}O_{31}Cl_{10}$, $Bi_{12}O_{15}Cl_{16}$ form dipolar coordination, with $Bi_{12}O_{15}Cl_6$and $Bi_{24}O_{31}Cl_{10}$ having the highest tetrahedral local environment. Our calculated visible light absorption and optical spectra show good agreement with the experiment. There is small difference in the values of indirect optical gap in the case of the optical spectra. The theoretical values are greater than the experiment for about 0.103eV and 0.169eV in Bi_3O_4Cl and BiOCl respectively. Whereas for the $Bi_{24}O_{31}Cl_{10}$ a difference of 0.104eV was observed, with the experimental value exceeding calculated one. These dipole arrangements could provide insight into photocatalytic activities in these compounds. The calculated optical gap and electronic bandgap compares well with the experimental reported values. The direction of the increase photo response performance is as follow: $Bi_{12}O_{17}Cl_2$ > $Bi_{24}O_{31}Cl_{10}$ > Bi_3O_4Cl > $Bi_{12}O_{15}Cl_6$ > BiOCl for the experiment. Whereas for the three structures in the theoretical results; it shows: Bi_3O_4Cl > $Bi_{24}O_{31}Cl_{10}$ > BiOCl. The overall trend here indicates that with increase in the BiOC content. The photocatalytic activity of $Bi_xO_yCl_z$ (x=1, 3, 12, 24; y=1, 4, 15, 31; z=1, 1, 6, and 10) was measured by photocatalytic degradation of RhB under visible light ($\lambda > 420$ nm) irradiation. RhB dye is chosen as a model organic contaminant due to its high stability and extensive applicability. The degradation rate is estimated to be around 0.005 min^{-1} via the first-order reaction kinetic fitting.

Design of spin-orbital-textures in ferromagnetic/topological insulator interfaces (P)

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The interfaces of materials are crucial in the construction of devices as they connect a particular effect to the observed signal. Topological materials have received significant attention due to their surface states with spin-momentum locking, providing new possibilities for designing quantum devices, particularly for potential applications in spintronics [1]. Currently, increasing interest is directed towards the study of topological/magnetic interfaces because of the presence of charge/spin conversion phenomena, such as direct or inverse Rashba-Edelstein [2] or spin-orbit torque [3,4], which enables the control of magnetization direction. In this study [5], we combined Density Functional Theory calculations with effective Hamiltonians to investigate the interfacial coupling between the surface states of a 3D topological insulator (Bi_2Se_3) and the magnetic moments of a 2D insulator ferromagnetic (CrI_3), with a focus on a complete characterization of the dispersive image of the interface topological states, especially regarding their spin texture behavior. We discovered that modulating different degrees of freedom of the system, such as controlling the finite size effects on the topological material, the magnetization direction of the ferromagnetic, as well as on which surfaces of the topological insulator the ferromagnetic is deposited, enables the attainment of a rich family of both in-plane spin texture and energy dispersion near the Fermi level. Furthermore, we observed efficient switching between the magnetization direction and the orientation of the spin texture. The aim of this work is to understand how this system can be adjusted to promote efficient control of the emerging phenomena of surface states, to enhance charge/spin conversion mechanics for spintronics applications, as well as to obtain a fundamental understanding of how the behavior of in-plane spin texture can influence the transport and scattering properties of these states.

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Influence of transition metal elements on phase stabilities, electronic and elastic properties of the MnV alloys: Ab initio calculations (P)

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Rare-earth intermetallic compounds are attractive materials from both fundamental and industrial perspectives and have a wide range of applications in engineering systems. In 2003, K. Gschneidner reported on the discovery of some ductile rare earth intermetallic compounds with the CsCl type-B2 structure[1-3]. They are characterized by high melting point, high specific strength, good ductility, and excellent corrosion resistance. The most recent interest in them is due to their high ductility and high fracture toughness at room temperature, as well as chemical stability. In this work, we propose to study the structural, electronic, and mechanical properties of MnV ductile intermetallic as a candidate to replace these rare earth intermetallics which crystallize in the B2-CsCl type structure using the pseudo-potential method based on the density functional theory (DFT) implemented in the VASP code. We also report the influence of substituted transition metals (Al, Ti, Cr, Co, Ni, Cu, Zn, Zr, Nb, Mo, Ag) on the crystallographic sites Mn and V to understand their effects on the structural, electronic and mechanical properties of MnV alloys.

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Effect of lattice dynamics on thermoelectric performance of Li-based transition metal nitrides (P)

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High thermoelectric performance of a material requires a high power factor and a low thermal conductivity. Previous studies have found that substitution of a heavy element may be an effective strategy for reducing lattice thermal conductivity of the material. However, our investigations based upon first-principles calculations reveals that substituting the material with heavy element may not always reduce the thermal conductivity. This study investigates the lattice dynamics of Li_2TMN_2 (TM= Zr and Hf) to unveil the factors contributing to their anomalous thermal transport properties. Both compounds are energetically and dynamically stable and their lattice parameters are in good agreement with the previous experimental study. Also, the transport properties exhibit a strong anisotropic behaviour for both materials. Li_2ZrN_2 has low lattice thermal conductivity of 1.52 $Wm^{-1}K^{-1}$ along a-axis at 1000 K owing to the significant reduction in phonon lifetime caused by the rattling behaviour of Zr. The figure of merit for Li_2ZrN_2 ~1 at 1000 K along a-axis. Therefore, Li_2ZrN_2 exhibits relatively good thermoelectric properties, rendering it an excellent high temperature thermoelectric material. This research provides insights into reducing thermal conductivity in transition metal nitrides for energy harvesting applications.

Fine-tuning of thermoelectric figure of merit: The role of carrier concentration in HfO_2 (P)

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We examined the thermoelectric (TE) performance of various polymorphs of HfO_2 in experimentally observed carrier concentration range (~1018-1022 cm^{-3}), using semi-classical Boltzmann transport theory and phonon dynamics employed with density functional theory. The absence of negative frequency in phonon band structure of all polymorphs confirm the dynamical stability. At room temperature the observed values of the Seebeck coefficient are 945.27 mV/K, 922.62 mV/K, 867.44 mV/K, and 830.81 mV/K for tetragonal, orthorhombic, monoclinic, and cubic phases of HfO_2, respectively. Among the polymorphs of HfO_2, cubic phase has maximum lattice thermal conductivity. The low lattice thermal conductivity leads to the high values of the figure of merit (ZT) at the optimized carrier concentrations (~1021 cm^{-3}). The least value of ZT observed in cubic phase to enhance the TE performance, the doping of Ti and S at the Hf and O sites was analyzed, respectively. The convex hull formalism was used to examine the stable doping concentrations and thermodynamical stability of the resultant materials. Doping leads to create new trap states and reduces the band gap. The magnitude of the Seebeck coefficient is high in Ti doping as compared to S doping. Further, the effective mass calculation was done to understand the behavior of the Seebeck coefficient. The predicted ZT makes cubic-HfO_2 as a possible high-temperature thermoelectric energy harvester.

Li/Na battery anode materials: A first-principles study of YB MBene using density functional theory (P)

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Transition-metal borides, also known as MBenes, represent a novel class of two-dimensional materials [1,2] that exhibit exceptional physical and chemical properties due to their layered structure, reminiscent of MXenes [3]. In this study, we conducted first-principle calculations to evaluate the electrochemical properties of monolayer Yttrium boride (YB) for its potential utilization as an electrode material in both Li-ion and Na-ion batteries. Our electronic structure calculations affirm that the YB monolayer maintains its metallic character, both with and without Li/Na adsorption. Additionally, our ab-initio molecular dynamics (AIMD) simulations confirm the thermal stability of the monolayer at a temperature of 300K. Our investigations reveal that the most stable adsorption site for Li/Na atoms is located at the top of the B layer. The theoretical specific charge capacities are calculated to be 197mAh/g and 116mAh/g for single Li and Na adsorption, respectively. Despite its promising electrochemical properties, it's worth noting that YB monolayer exhibits a relatively low open circuit voltage of 0.16 V for Li-ion batteries and 0.70 V for Na-ion batteries. Nevertheless, these findings suggest that YB monolayer has potential applications as an efficient anode material for both Li-ion and Na-ion batteries.

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Utilizing tunable hydrogen interstitials in two-dimensional n-type pyrite, a highly selective catalyst for CO_2 reduction (P)

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Density functional theory (DFT) calculations were employed to elucidate the impact of hydrogen interstitial (H_i) defects on the optical and efficacy of iron pyrite (FeS_2) as a selective carbon dioxide reduction catalyst (CO_2RR). Three different charge states were considered, namely H_i^1, H_i^0 or H_i^{-1}. Upon examining the formation energy of the various possible charge states that the H_i can accommodate, it was found that H_i will be incorporated in the structure as an n-type defect in the (+1) state across almost the entire bandgap. However, under heavy n-type growth conditions, it can act as an amphoteric dopant, and can accommodate the 3 states simultaneously. The performance of FeS_2-H in CO_2RR is compared to the pristine counterpart. The calculations show that the addition of H_i facilitates the methanol pathway by prioritizing the attachment of the $COOH^*$ intermediate. Consequently, H_i promotes the formation of C1 compounds with higher performance. This is achieved by inhibiting the competitor hydrogen evolution reaction (HER), leading to a significant improvement in the efficiency.

Day 2 - Session 3

Tuning adhesion of C/Cu interfaces through chemical modifications and surface reconstruction (CT)

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Diamond-like carbon (DLC) coatings are well-known for their exceptional combination of tribological and mechanical properties, such as low friction coefficients and wear rates, together with relatively high levels of hardness and elastic modulus. However, one of the main limitations in the employment of these coatings concerns their spallation from the substrate. In principle, they should strongly bind to the substrate on one side and, at the same time, exhibit a minimum adhesion with respect to the countersurface; it is thus extremely interesting to explore how DLCs adhesion can be tuned through chemical modifications of its surfaces. In this work, we employ *ab initio* simulations, performed with VASP, to study the effect of surface reconstruction and chemical species intercalation (B, P, O, F, N, S) on the adhesion of non-reconstructed and Pandey-reconstructed C(111) surfaces with Cu(111). Since the two selected diamond terminations represent the extreme limits of the diamond surface energy and are indicative of the fraction of sp^3 and sp^2 bonds in DLCs, our results already provide valuable insights for tuning the adhesion of DLCs/diamond coatings in a specific and controlled manner. By studying the effect of surface reconstruction, we found that increasing the sp^3/sp^2 bond ratio causes an increasing of the interface adhesion; while, adatom intercalation leads to adhesion quenching of DLCs with high sp^3 fraction. Fluorine stands out as an unique case, causing an adhesion reduction of almost 100% (adhesion energy value of 0.01 J/m^2), implying that it could serve as a significant element in reducing adhesion at interfaces with diamond. Moreover, an adhesion fine tuning is possible by increasing the fraction of sp^2 bonds and playing with atomic species intercalation. Indeed, both N and especially B increase the adhesion of Pandey-reconstructed C(111)/Cu(111) interface, while S is the most effective in reducing it, leading to the complete passivation of the Cu surface. Insights on the different contributions to the overall interaction happening at the interface are provided by the analysis of the atomic bond order, which suggests that adhesion energy modifications are linked to two key behaviours of the intercalated adatom: whether it acts as a bridge connecting the two surfaces, leaving at the same time a strong residual interaction between them.

First principles assessment of prospective tunnel barriers at semiconductor/superconductor interfaces (CT)

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Majorana zero modes, with prospective applications in topological quantum computing, are expected to arise in semiconductor-superconductor interfaces. However, this interfacing can lead to detrimental effects such as band bending and the loss of necessary semiconductor properties [1]. We study the electronic properties of semiconductor-superconductor interfaces relevant to Majorana zero mode systems, introducing barriers to control these detrimental effects, via density functional theory (DFT) using the first principles VASP package.

InAs and InSb based devices are popular choices for Majorana zero mode setups due to their large spin orbit coupling and growth properties. Metallic superconductors, such as Al and α-Sn, supply the necessary superconducting effect. To moderate the detrimental effects in the semiconductor from this interfacing, the favorably lattice matched materials of CdTe, ZnTe, and CdSe are explored as tunnel barriers for the semiconductor-metallic superconductor interface.

The PBE+U method is used, with the Hubbard U parameters found via a machine-learned Bayesian optimization algorithm. This, and making use of VASP's highly efficient and parallelizable implementation, allows the simulation of large interface slab models. We use the in-house program OGRE[2] to build the required reconstructed and relaxed interface models.

We study the band offsets and the penetration depth of metal-induced gap states (MIGS) in bilayer and trilayer interfaces. We first consider the semiconductor-semiconductor interface, such as InSb/CdTe and InAs/ZnTe, then semiconductor-metal interfaces, and finally move on to the tri-layer interfaces of InSb/CdTe/α-Sn and InAs/ZnTe,CdSe/Al. We study the effect of increasing the thickness of the barrier layers. We find that 16 atomic layers (3.5 nm) of CdTe can serve as a tunnel barrier, effectively shielding the InSb from MIGS from the α-Sn. This, and insights on the InAs-Al interface, may guide the choice of dimensions of the barrier to mediate the coupling in semiconductor-superconductor devices in future Majorana zero modes experiments.

This work was primarily undertaken at the National Energy Research Scientific Computing Center (NERSC) supercomputing cluster, which is the primary scientific computing facility for the Office of Science in the U.S. Department of Energy.

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Calculation of nonradiative transition rates at defects in semiconductors and insulators (IT)

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Point defects and impurities in semiconductors or insulators can trap free carriers, holding onto them for long times or enabling their recombination without the emission of light. These processes can have a significant impact on the performance of electronic and optoelectronic devices. More recently, defects and impurities have found relevance for quantum applications, where single defects or impurities can be used as qubits, sources for single photons, or as nanoscale sensors. The operation of these so-called quantum defects can depend sensitively on the various nonradiative transitions between their orbitals. Identifying the microscopic origin of the defects or impurities at play is a recurrent problem plaguing many fields, and first-principles calculations based on the framework of density functional theory can be a powerful tool to solve this problem.

In this talk, I will discuss the formalism behind the evaluation of nonradiative transition rates from first principles, as well as the development of our open-source python code Nonrad, which implements the formalism and is interfaced with the output of the VASP code. I will then overview several examples of its application to solve outstanding research questions. Halide perovskite solar cells have achieved remarkable efficiencies despite the fact that defect-mediated nonradiative recombination still poses a problem. I will discuss how our studies of nonradiative recombination in the perovskites provide potential routes for improving device efficiency. In hexagonal boron nitride, bright single-photon emitters have been observed and are promising for applications in quantum information science. I will describe how nonradiative transitions can help to rationalize the optical dynamics of these emitters and suggest that the microscopic origin of the emission is a boron dangling bond. Lastly, I will discuss the role of nonradiative transitions in limiting the efficiency of telecom-wavelength single-photon emitters in cubic boron nitride.

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