

22nd International Workshop on Computational Physics and Materials Science: Total Energy and Force Methods | (SMR 4050)

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Cubic antimony, a parent material for metavalent compounds

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Higher chalcogenides, such as GeTe, GeSe and Sb₂Te₃, have been recently proposed as a new class of materials, called metavalent, which has distinct properties compared to metallic and covalent compounds [1].

Metavalent materials share an unusual combination of functional properties such as reversible amorphous to crystalline phase change, large optical absorption, ferroelectricity, efficient thermoelectric and topological properties [1,2].

In order to shed light on the peculiar properties of metavalent materials and better understand their origin, we have compared cubic antimony, the simplest compound of the family, with lead telluride in the rocksalt structure. While cubic antimony is unstable and metallic, lead telluride is stable and a small gap semiconductor. Still, their band structures reveal strong similarities.

We have analysed the origin of the instability of cubic antimony by calculating phonon properties, its static response function using time-dependent density-functional theory and making the link to the nesting properties of its Fermi surface [3].

Ongoing research is dealing with the dynamical response function which can be measured by electron energy loss spectroscopy.

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Theoretical study of bound excitons in monolayer SnS_2

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SnS_2 is a layered material with visible spectrum absorption and energy level alignment suitable for hydrogen fuel cell electrodes [1]. It exemplifies group IV chalcogenides, increasingly studied for second harmonic generation (SHG) response [2]. We undertake a thorough theoretical investigation of bound excitons in monolayer SnS_2 , employing many-body perturbation theory [3] atop DFT-PBE electronic structure calculations [4]. Our study reveals a richer structure of bound excitons than previously reported [5], likely due to overscreening from the interlayer interaction in the previous work. The calculated first bright-exciton energy of 2.3 eV closely matches the experimental optical gap of 2.23 eV. We offer a detailed analysis of the composition and nature of the excitonic states. Future research will extend to non-centrosymmetric group IV chalcogenide systems such as SnS and $SnSe$ layered structures, also exploring their SHG response [6].

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First principles calculations of elastic constants and Curie temperature under high pressure in iron

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Phase diagram of iron is a long standing challenge for first principles calculations, because of the important role of electronic interactions. Under pressure, however, it is expected that their role should decrease. First, we use first principles DFT+DMFT calculations to discuss the evolution of electronic structure under pressure in both the α and ϵ phases[1, 2]. Secondly, we evaluate the variations under pressure of two quantities, namely the α phase Curie temperature and elastic constants of the ϵ phase. We discuss evolution of Curie temperature with measurements, previous calculations and thermodynamical relations. Elastic constants are validated in comparison to recent experimental measurement on single crystal ϵ phase[2]. Then it is used to evaluate the crystalline anisotropy in -Fe under densities typical of Earths inner core. It allows to interpret seismological observations that indicate anisotropic behavior within the solid inner core.

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Phonnier: Wannier and Long-Range Interactions for Phonons.

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In this work, we present *Phonnier*: a code that simulates phonon systems using a tight-binding model within the Wannier functions formalism. We address the challenge of long-range interactions in phonon simulations and showcase calculations for different systems where LO-TO splitting may play a significant role. We present the problem of long-range interactions when simulating phonon systems and showcase the calculation of different topological landmarks, such as Weyl nodes, surface states and Wilson loops.

Theory of local \mathbb{Z}_2 topological markers for finite and periodic systems

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Topological invariants are global properties of the ground-state wave function, typically defined as winding numbers in reciprocal space. Over the years, a number of topological markers have been introduced, allowing to probe the topological order locally in real space even for disordered and inhomogeneous systems [1]. Here, we address time-reversal symmetric systems in two dimensions and introduce two local \mathbb{Z}_2 topological markers [2]. The first formulation is based on a generalization of the spin-Chern number [3] while the second one is based solely on time-reversal symmetry [4]. Then, we introduce a formulation of the local Chern marker for extended systems with periodic boundary conditions [5, 6], and we extend it to the aforementioned \mathbb{Z}_2 markers [7]. Finally, we show numerical simulations to validate the approach, including pristine disordered and inhomogeneous systems, such as topological/trivial heterojunctions.

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Relativistic Dynamics of Electron Transport in Chiral Molecular Systems

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The Chirality-Induced Spin Selectivity (CISS) effect allows chiral molecules and crystals to favor spin-polarized current transmission, a discovery made in 1999. While this effect shows significant promise for spintronics and electron transfer, its mechanisms are not yet fully understood. The effect is thought to stem from enhanced spin-orbit coupling (SOC) in chiral molecules, but the required SOC strength greatly exceeds typical values for light atoms involved. Using the DIRAC code's relativistic DFT, we analyze how molecular chirality affects electron chirality, its distribution under an external electric field, and spin polarization decay in proximity to magnetic molecules. Finally, the Landauer-Imry-Büttiker approach helps us explore how spin-dependent transmission varies with twist angle. Though aligning qualitatively with experiments, our findings underscore the need for advanced exchange-correlation functionals to better capture a generalized SOC definition, potentially incorporating spin current density.

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Charge transport mechanisms at the Cathode-Electrolyte Interfaces and Discharge Products of Non-Aqueous Rechargeable Sodium-Air Batteries: A DFT study

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Sodium–air batteries have superior theoretical specific energy to existing secondary batteries, even compared to state-of-the-art secondary lithium-ion batteries (LIBs). They exhibit less than 200 mV discharge and recharge overpotentials and above 90% electrical energy efficiency drawn at fairly excessive current densities. However, like lithium–air batteries, sodium–air batteries also suffer from poor rechargeability, low capacity, and dendrite formation. In this study, charge transport (ionic conductivity) studies of the cathode–electrolyte interfaces ($\text{Na}_x\text{O}_2@ \text{Na}_2\text{CO}_3$, $x = 1$ and 2) and discharge products (NaO_2 , Na_2O_2 , and Na_2CO_3) of non-aqueous secondary sodium–air/ O_2 batteries are presented using the density functional theory framework. The results revealed that the sodium-ion diffusion is taking place too rapidly with an activation barrier of less than half an eV despite the bandgaps of these materials exceeding 4 eV. The ionic conductivity study is mediated by negative sodium vacancies (V_{Na^-}). Interfaces revealed magnificent ionic conduction; the sodium-ion diffuses with little or no activation or thermodynamic barrier. Thus, the interfaces may provide an alternative pathway for rapid ion transfer during the battery operation, which might enhance the conductivity and eventually boost the accessible capacity and performances of secondary non-aqueous sodium–air batteries.

Structural phase transitions in monolayer TMDs with a neural-network interatomic potential

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Among the wide family of layered materials, transition metal dichalcogenides (TMDs) constitute a broad class of systems exhibiting diverse properties depending on chemical composition, temperature, pressure, and dimensionality. In the single-layer limit, some TMDs, such as tungsten ditelluride (WTe₂) and molybdenum ditelluride (MoTe₂), exhibit different polymorphs. These materials showcase two dynamically stable phases characterized by a small energy difference, both of which can be experimentally observed. Here, we develop a neural-network interatomic potential, trained on density-functional theory simulations, to investigate, at the atomistic level, the structural transition between the two polymorphs separated by a high energy barrier. Through molecular dynamics simulations, we examine the role of defects and interfaces in driving the structural change from the metastable to the most stable phase at high temperatures.

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Microscopic origin of gray tracks in KTiOPO_4 from *ab initio* calculations

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Ferroelectric potassium titanyl phosphate (KTiOPO_4 , KTP) is widely applied in (nonlinear) optical devices, e.g., for second harmonic generation (SHG), as a waveguide material, or as an electro-optical modulator. Unfortunately, the application of high-intensity laser light or strong electric fields is a limiting factor for the crystal. In fact, they provoke the formation of so-called gray tracks, i.e., dark spots showing a high absorption for application-relevant wavelengths, whereby the operation upon damaged crystals can lead to their catastrophic failure. [1] The microscopic origin of gray tracks is usually attributed to the reduction of titanium atoms (i.e., the formation of so-called Ti^{3+} centers) as the result of the charge compensation of various defects: One of these centers is thermally stable, and has already been attributed to an oxygen vacancy [2]. Oxygen vacancies spontaneously form within KTP crystals, since they charge compensate for potassium vacancies. For this, it is not surprising that treatments reducing the number of potassium vacancies [3] or the potassium-ion diffusivity (e.g., by a small rubidium doping [4]) are very beneficial in order to prevent the formation of gray tracks. Nevertheless, the properties of oxygen vacancies and related Ti^{3+} centers have not been systematically investigated, yet. Thus, indicating these Ti^{3+} centers as the only reason for gray tracking seems to be provide a simplistic picture for the phenomenon.

In the present DFT study, we model oxygen vacancies in rubidium-doped KTP (RKTP), and in potassium-deficient KTP [5,6]. The focus of our investigation thereby lays on the systematic analysis of their energetics as well as their electronic and magnetic properties. We find that the properties of Ti^{3+} centers do not strongly depend on the chemical environment. In addition, they are not only energetically more favorable, but do also provide a better agreement with experimental hyperfine tensors [2], if they are located at a near TiO_6 polyhedron rather than at an undercoordinated TiO_5 moiety caused by the oxygen vacancy itself, whereas the Ti^{3+} centers are stabilized by displaced potassium ions. Notably, potassium interstitials rather than oxygen vacancies appear to be directly related to origin of gray tracks. Thus, our results strongly hint that the current gray-tracking model has to be partially revised.

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Koopmans functionals for excitonic properties in materials

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Exciton characterization is crucial for several materials science applications, ranging from energy transport/storage technologies to photocatalysis, plasmonic, sensing. The state-of-the-art approach is ab-initio many-body perturbation theory (MBPT), in particular the Bethe-Salpeter equation (BSE) [1]. This is usually built on top of computationally demanding GW quasiparticle band structures (BSE@GW approach). In this work, we demonstrate how it is possible to construct the BSE Hamiltonian starting from Koopmans functionals [2] eigenvalues as the main ingredient (BSE@Koopmans), obtaining optical spectra with comparable accuracy with respect to the BSE@GW but at reduced computational cost. Preliminary developments on automated workflows to compute BSE@Koopmans from scratch (i.e. Koopmans+BSE) are provided within the koopmans package [3], the AiiDA workflow engine [4] and the Yambo code [5].

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Origin of flat bands in three-dimensional coupled kagome lattices

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Two-dimensional kagome lattices are known to host a flat band (FB) and a Dirac point due to the destructive quantum interference of electronic wavefunctions [1-3]. In this work, we have introduced an exact analytical decimation transformation scheme to explore the coexistence of FB and Dirac point in three-dimensional coupled kagome systems. Our proposed technique allows coarse-graining of the parameter space, which maps the original system to an analogous low-level lattice [4]. The decimated system facilitates defining a descriptor that controls the appearance of a FB and provides a definite criterion for absolute flatness. We confirm our predictions on the emergence of the FB and Dirac points for a class of materials employing material databases in conjunction with density functional theory calculations. The present work explores the intricate electronic properties of coupled kagome lattices and provides an analytical formalism that can efficiently investigate the rich physics of such lattice models.

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Double Counting Correction Free DFT+U-type Functionals from the Flat Plane Condition

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The DFT+ U method is an extremely popular tool for modeling Mott-Hubbard and Charge-Transfer insulators, thanks to its semi-local DFT cost. DFT+ U -type functionals however have been traditionally developed from the Hubbard model with use of ad-hoc double-counting correction schemes.

We derive a DFT+ U -type functional named BLOR [1] and its many-body generalisation mBLOR [2], to explicitly enforce the flat plane condition on localized subspaces, dispensing with the need to invoke the Hubbard model or a double-counting correction scheme. BLOR by design corrects for Many-Electron Self-Interaction Error (MSIE) and Static Correlation Error, two of the most notorious errors that plague practical DFT calculations.

We use multireference homonuclear s -block molecular test systems to compare the performance of BLOR to traditional functionals, with U & J parameters computed using the linear response methodology. We find that DFT(PBE) and DFT+ U (using Dudarev's functional) yields relative energetic errors as high as 8% & 20.5%, respectively, while BLOR yields errors below 0.6%. Similar improvements in the total energy are achieved for multireference homonuclear p -block molecular test systems with the use of the mBLOR functional.

Intriguingly, the BLOR and mBLOR functionals exhibit explicit derivative discontinuities with respect to subspace occupancy, which can contribute to the fundamental band-gap but not the Generalised Kohn Sham (GKS) gap. An additional derivative discontinuity correction term has been developed to incorporate this contribution to the fundamental band-gap within the GKS eigenspectrum. Armed with this derivative discontinuity correction term, BLOR and mBLOR are found to be the only DFT+ U -type functionals to open the GKS gap of the stretched neutral homo-nuclear dimers without the aid of unphysical spin-symmetry breaking.

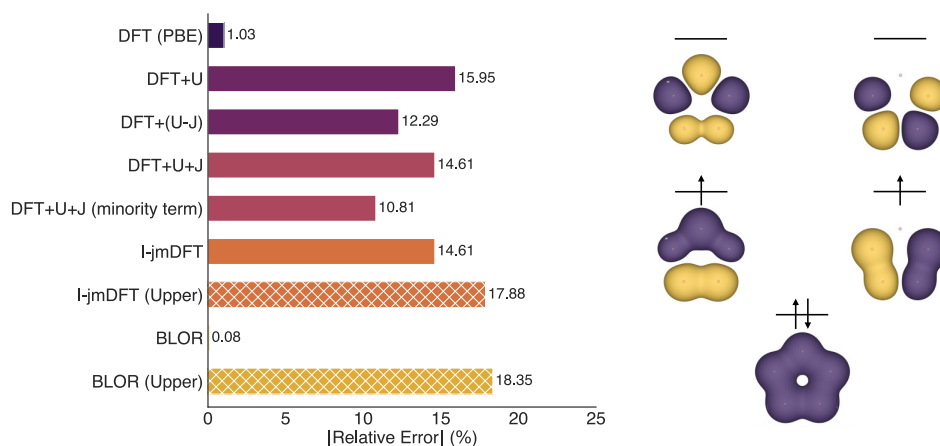


Figure 1: Bar chart of the relative errors in the total energy of dissociated H_5^+ at an internuclear separation of $8a_0$ using different corrective functionals.

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Defect complexes and charge compensation in Ta-doped TiO₂ transparent conductor

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The stability and behavior of point defects and complexes of Ta-doped TiO₂ (TaTO) transparent conducting oxide (TCO) and their effect on its structural, electronic, optical, and transport properties are comprehensively investigated through state-of-the-art first principles simulations and experimental measurements, under various synthesis conditions [1]. Our results show that O vacancies and Ti interstitials, which form relatively easily under O-poor conditions, promote n-type conductivity together with Ta doping; while clustering of donor intrinsic defects is irrelevant both energetically and electronically, Ta clustering is inhibited. In contrast, O interstitials and especially Ti vacancies, which form more easily under O-rich conditions, induce gap states and act as deep acceptors heavily compensating the donor effect thus reducing both conductivity and transparency; their relative positions does not affect the electrical character of the system, although configurations where defects and Ta dopant are neighbors are favored. Interactions between dopants and multiple native defects introduce nonlinear charge-compensation effects, which reduce the quasi-free charge but can mitigate visible transmittance loss in a trade-off mechanism, supported by experimental observations, able to sustain and fine-tune TC properties. The compensation effects of various defects modulate the effective injected charge, thereby influencing the crossover energy position as determined by our simulations. Overall, this study provides valuable insights into the properties of TaTO and defect engineering to optimize it as TCO material for suitable applications in transparent electronics and optoelectronic devices. L. Bursi acknowledges financial support from project PNRR–M4C2INV1.5, NextGenerationEU-Avviso 32772021-ECS_00000033-ECOSISTER-sp6.

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Tailorable materials for plasmonics in extreme conditions

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We investigate and discover refractory materials with tunable optical properties in the near-IR/vis range and high mechanical and thermal resistance for plasmonics applications in extreme and harsh conditions. By using ab initio simulations based on (TD)DFT, we first studied the role of composition and structural disorder of transition-metal nitrides [1-3], and we provided an efficient strategy to engineering stable, easy-to-grow hyperbolic metamaterials [4], with extraordinary mechanical properties. Finally, by combining computational thermodynamics and first principles electronic structure techniques [5], we proposed high-entropy transition-metal carbides [6], which yielded plasmonic properties from room temperature to 1500K. This new class of plasmonic materials may foster previously unexplored optical/mechanical applications (e.g. aerospace and security systems) in extreme conditions.

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Tunable volume plasmon polariton modes in hyperbolic metamaterials based on III-V semiconductors

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Hyperbolic metamaterials (HMMs) have gained particular attention because they allow for the propagation of electromagnetic waves with arbitrarily large wavevectors, which would be evanescent in ordinary materials. These high-k modes, called volume plasmon-polaritons (VPPs), enable advanced applications such as superlenses, sub-wavelength imaging and super-Planckian thermal emission. HMMs are characterized by extreme optical anisotropy, as they behave as metals in one direction and as dielectrics in the perpendicular direction. They are most often realized by alternating layers of metallic and dielectric systems, where plasmonic resonances are used to couple light with the metallic component of the stack. When regular metals are employed, the resulting metamaterials are generally hyperbolically active in the visible-UV range, because of their high electron density. However, HMMs working in the IR-to-THz electromagnetic range would be of great technological interest for applications in infrared optoelectronics. Recent experimental reports [1] have shown the feasibility of fabricating multilayer HMMs working in the infrared spectrum using some III-V semiconductors, where intrinsic and doped compounds serve as dielectric and metallic layers respectively.

In this work [2], we undertake a comprehensive investigation of volume plasmon polaritons within hyperbolic metamaterials. Initially, we integrate ab initio atomistic simulations based on DFT+U and the effective medium approximation to theoretically demonstrate the capability of realizing hyperbolic metamaterials using only III-V semiconductors. The obtained results are in excellent agreement with the experimental findings. However, this approach provides insights only into the first-order VPPs, so we expand beyond the limitations of effective medium theory by employing the scattering matrix method for electrodynamics simulations. This allows us to explore volume plasmon polaritons of higher orders and to predict not only the resonance peaks of transmission and reflectivity but also the electromagnetic field within the metamaterial. Moreover, by leveraging the study of the photonic band structure of the metamaterial, we systematically identify optimal excitation conditions for these resonances, independently of external environmental factors.

Several parameters can be adjusted to control the energy at which these resonance modes appear. For instance, changing the semiconductor doping, modifying the filling factor, or incorporating nanoscale structures like quantum wells can all influence the energy position of Volume Plasmon Polaritons in the spectrum. This approach enables the efficient design of hyperbolic metamaterials, allowing us to effectively tune and harness these plasmonic resonances.

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Algorithmic inversion on sum over poles to embed interacting many-body systems

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Quantum embedding methods are powerful techniques to study interacting correlated electrons beyond mean-field theories. An established approach is dynamical mean-field theory (DMFT), which tackles the problem by mapping strongly correlated electrons into an Anderson impurity model. Here, we start from a different approach meant to solve Dyson-like equations – the algorithmic inversion on sum over poles [1] – to provide an embedding formulation valid at zero or finite temperature and based on exact diagonalization. We demonstrate the approach on the one-dimensional Hubbard ring, performing self-consistent calculations on the real axis, ensuring the accurate computation of both spectral and thermodynamic quantities.

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Cross comparing ab-initio methods for estimating the Hubbard U : constrained random-phase approximation vs. linear response

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Many quantum materials are strongly correlated systems, where interactions of localized d or f orbitals near the Fermi level significantly impact their properties. To capture this behavior accurately, density functional theory (DFT), typically in its local (LDA) or generalized gradient (GGA) approximations, is often combined with static mean-field Hubbard corrections (DFT+U) or dynamical mean-field theory (DFT+DMFT). A crucial aspect of these methods is determining an accurate value for the screened Coulomb interaction parameter, U .

Two widely used methods for determining the U parameter are the constrained random-phase approximation (cRPA) and linear response theory. In cRPA, typically used in DFT+DMFT, the Kohn-Sham states are divided into correlated and uncorrelated subspaces, with screening computed only for the uncorrelated states. Linear response theory, commonly applied in DFT+U, instead defines U based on the spurious curvature of the energy functional with respect to atomic site occupations, quantifying the error introduced by standard approximations such as LDA or GGA.

In this work, we introduce maximally localized Wannier functions, generated via Wannier90, as localized projectors within Quantum ESPRESSO [1]. This unified approach enables both constrained random-phase approximation (cRPA) and linear response theory to be applied consistently on the same Wannier functions, facilitating a direct comparison of these methods [2]. We analyze the similarities and differences between the two approaches, identifying specific cases where cRPA becomes ill-defined while linear response remains robust, as demonstrated by results for two materials: KCuF_3 and Sr_2FeO_4 [2].

As an outlook, this framework also enhances the potential for transferring the U parameter across computational codes, as maximally localized Wannier functions depend only on the Kohn-Sham bands. This development further bridges the DFT+U and DFT+DMFT communities, providing a common ground for more consistent calculations across different methods and codes.

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A dynamical Hubbard approach to the Mott-Hubbard/charge-transfer series of transition-metal monoxides

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Electronic correlations beyond static mean-field theories are of fundamental importance in describing complex materials - such as transition-metal oxides – where the low-energy physics is driven by highly localized d or f orbitals. Here, we generalize our recently introduced dynamical Hubbard approach [1] to the spin-polarized and multi-site case, making it possible to study the spectral properties of the prototypical monoxide series of MnO, FeO, CoO, and NiO in their antiferromagnetic phase. We find excellent agreement with experiments and state-of-the art DFT+DMFT, both for the density of states and the spectral function - including band renormalization and spectral weight transfer - paving the way to a numerically efficient and physically transparent treatment of dynamical correlations in realistic systems.

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Engineering Spin States & Molecular Configurations at the Nanoscale

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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 deposited on metal substrates. Our Density Functional Theory (DFT) calculations reveal that the system is a spin-crossover system where the bridging Fe atoms can be in two different spin states. Scanning Tunneling Microscopy measurements and DFT calculations also reveal that the molecular building blocks can be in one of two stable configurations within the coordination polymers, depending on the surface orientation and spin state. This system presents an ideal platform for studying the interplay between spin states, molecular structure and substrate effects. Further, since the spin state can, in principle, be tuned by external perturbations, it can be used for a variety of technological applications.

Simulating Materials With Ephemeral Data Derived Potentials

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Dynamic compression techniques have allowed new and interesting materials to be probed at extreme pressures. However, the transient nature of the pulse limits the information that can be extracted from a given experiment. Simulations can provide a useful tool to aid interpretation of this experimental data, but capturing the full complexity of the experiment requires large system sizes and time-scales of nano-seconds, all at a quantum mechanical level of accuracy. Classical simulations using data derived potentials have been shown to maintain this ‘quantum accuracy’ at a fraction of the computational cost, allowing much larger length and time-scales to be explored. We present results from the recently developed ephemeral data derived potential (EDDP[1, 2]) approach that has been integrated into the LAMMPS simulation package. Using a range of example systems, we demonstrate that the EDDPs are a versatile tool for fast and accurate simulations at high pressure.

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Quantum Monte Carlo benchmarked hydrogen equation of state at planetary conditions

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Hydrogen is the lightest and most abundant element in the Universe and plays a pivotal role in gas giant planets such as Jupiter. Despite its elemental simplicity, hydrogen exhibits a complex phase diagram due to strong electron-electron correlations. Accurately modeling the interior of gas giant planets requires an equation of state (EOS) for hydrogen with an error on the order of 1% [1]. This requires calculating pressure, energy, and entropy over a pressure range from 1 bar to 10 MBar. However, current experimental techniques fall short of achieving the necessary pressures, while state-of-the-art density functional theory (DFT) calculations provide discrepancies in the EOS up to 30% depending by the choice of Exchange-Correlation functional employed. While the full computation of the EOS with a theory better suited to treat electron-electron correlation, such as Quantum Monte Carlo (QMC), is still not computationally affordable, it is possible to use QMC as a benchmark on uncorrelated atomic configurations sampled from DFT Molecular Dynamics at the temperatures and density (T - ρ) of interest. Meta-GGA functionals with van der Waals correction, and in particular SCAN-vv10 [2], shows remarkable performance against QMC both in term of internal energy and pressure and reproduces the first order Liquid-Liquid transition in perfect agreement with state-of-the-art QMC simulations [3, 4]. To cover the vast range of T - ρ needed for the interior model of a gas giant planet the SCAN-vv10 EOS has been coupled with an EOS based on the chemical picture for the low T - ρ regime [5]. In order to accurately compute absolute entropy it is important to preserve thermodynamic consistency between the two different theories and to enforce it we perform thermodynamic integration [6] on each component of the EOS *separately*, and then interpolate the resulting free energy function. In this way, one can then derive the pressure and energy near the interpolation boundaries by taking partial derivatives, which are naturally consistent by construction [7]. Two reference points are used to incorporate accurate values of the absolute entropy into each separate phase region: an experimental value for the low T - ρ regime and the absolute entropy value from the SCAN-vv10 EOS computed within the framework of Targeted Free Energy Perturbation [8] and flow matching [9] comparing the state of interest to one with tractable thermodynamic results. The resulting EOS improve the current results both in term of Pressure-Density relation and absolute Entropy, and will help clarifying the longstanding inconsistencies between Jupiter interior models and measurements from spacecraft missions.

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An alternative GPU acceleration for a pseudopotential plane-waves density functional theory code with applications to metallic systems

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We present an alternative GPU acceleration for plane waves pseudopotentials electronic structure codes designed for systems that have small unit cells but require a large number of \mathbf{k} points to sample the Brillouin zone as happens, for instance, in metals. We discuss the diagonalization of the Kohn and Sham equations and the solution of the linear system derived in density functional perturbation theory. Both problems take advantage from a rewriting of the routine that applies the Hamiltonian to the Bloch wave-functions to work simultaneously (in parallel on the GPU threads) on the wave-functions with different wave-vectors \mathbf{k} , as many as allowed by the GPU memory. Our implementation is written in `CUDA Fortran` and makes extensive use of kernel routines that run on the GPU (`GLOBAL` and `DEVICE` routines). We compare our method with the CPUs only calculation and with the approach currently implemented in `Quantum ESPRESSO` that uses GPU accelerated libraries for the FFT and for the linear algebra tasks such as the matrix-matrix multiplications as well as `OpenACC` directives for loop parallelization. We show in a realistic example that our method can give a significant improvement in the cases for which it has been designed. [1]

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Excitons in bilayer WTe₂

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Bilayer WTe₂ is a remarkable two-dimensional metal, since it exhibits a macroscopic out-of-plane electric dipole in spite of the presence of charge carriers that screen the electrostatic forces between ions [1]. At low temperature the system develops a narrow transport gap, like its monolayer counterpart where the appearance of the gap has been attributed to condensation of excitons [2]. The similarities between transport measurements in mono and bilayer structures suggest that similar excitonic physics is at play in the bilayer. Moreover, contrary to other known bilayer excitonic insulators, in which electrons and holes are spatially separated, in WTe₂ interlayer tunneling is significant. This might impact the observable features of the putative exciton condensate, giving rise to a coherent contribution to the ferroelectric dipole. In this work we investigate bilayer WTe₂ both from first principles and with a model built upon the symmetries of the system, focusing on the characterisation of excitons by means of the Bethe-Salpeter equation. The ultimate goal is to assess the instability against exciton condensation, as well as to predict its possible experimental fingerprint.

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Slip properties of graphene-water interfaces: atomistic simulations

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The reduction of aero/hydrodynamic resistance in automotive and aerospace components is central to a sustainability strategy aimed at enhancing efficiency and reducing energy consumption. In recent years, there has been growing interest in coatings that exhibit low-friction/low-resistance interactions between surfaces and air or water. Notably, although its hydrodynamic properties remain under explored, graphene has been proposed as a potential coating material to mitigate aerodynamic and hydrodynamic drag. A microscopic understanding of the interfacial dynamics between water and graphene-coated surfaces could support the design and optimization of new low-resistance materials for various types of vehicles.

We focus here on hydrodynamic resistance and we report on non equilibrium molecular dynamics calculations simulating a water flow parallel to a graphene surface. At variance with previous studies [1, 2, 3, 4], we introduce a *fixed-velocity zone* for molecules far from the surface to model the interaction with a steady flow. Using simulation cells containing some thousands of water molecules, we obtain velocity profiles that are linear as a function of the surface distance, which is in agreement with classical hydrodynamics and allows us to calculate the slip length and slip velocity. These two quantities are of key in assessing drag reduction, and more in general the out-of-equilibrium properties at the solid-liquid interface [5].

This method shows to be efficient, allowing to obtain results with reasonable statistical accuracy through simulations relatively short compared to other known methods. From simulations based on different cell sizes, we obtain slip length values of about 20 nm, in good agreement with the values reported in literature [1] from similar molecular dynamics simulations, confirming graphene's ability to enable significant water slip over its surface (high slip systems).

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Coupling Many Body Perturbation Theory (MBPT) with Polarizable Continuum Models (PCM)

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The theoretical description of quantum systems embedded in external environments represents a challenge for what concerns the application of MBPT approaches to the study of the electronic excitations.

Different level of accuracy can be used to treat the actual system, for which a quantum mechanical description is employed, and its surrounding environment, that can be approximated through a simplified quantum model, a molecular mechanical approach or a continuum model.

We will present a theoretical framework in which the MBPT treatment of the quantum system is coupled to a PCM description of its surrounding environment. The formalism has been implemented within the YAMBO computational platform [1,2] by exploiting the ENVIRON package [1,2] available withing the QuantumEspresso suite of codes [4,5].

Preliminary results of the methodology will be presented for a selected class of systems.

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2D Transition-metal dichalcogenides-based bilayer heterojunctions for efficient solar cells: A first-principles DFT and G_0W_0 -BSE study

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This work presents a first-principles density functional theory (DFT) and many-body perturbation-based G_0W_0 -BSE study of the optoelectronic properties of vertically stacked bilayer heterostructures composed of 2D transition-metal dichalcogenides (TMDs). We aim to propose these TMD heterostructures for potential applications in solar cells. The four TMD monolayers, MoS₂, WS₂, MoSe₂, and WSe₂ are considered to form heterojunctions due to their favorable band gaps, high carrier mobility, robust absorption in the visible region, and excellent stability. The electronic structures of the considered heterostructures (WS₂/MoS₂, MoSe₂/MoS₂, MoSe₂/WS₂, WSe₂/MoS₂, WSe₂/MoSe₂, and WSe₂/WS₂) are initially calculated at the DFT level [1, 2] employing PBE and HSE06 functionals and then using G_0W_0 approximation. The significantly large quasi-particle gaps obtained from the G_0W_0 results suggest strong correlation effects. All six heterostructures exhibit type II band alignment (crucial for efficient solar cells) at the DFT level, but according to G_0W_0 approximation, MoSe₂/WS₂ does not exhibit type II band alignment. The substantial intrinsic electric field values resulting from the horizontal mirror asymmetry in the heterostructures, and small electron and hole effective masses would assist charge carrier separation, resulting in a lower recombination rate. The optical properties are calculated by solving the Bethe-Salpeter equation (BSE) [3] and a superior optical response with low exciton binding energies is obtained, compared to the constituent monolayers. The power conversion efficiencies are calculated using the method proposed by Scharber *et al.* [4], at both the HSE06 and G_0W_0 -BSE levels to assess the quality of our design heterojunction solar cells. The maximum PCE of our designed solar cells can reach up to 20% for the WSe₂/WS₂ and MoSe₂/MoS₂ heterojunctions according to the HSE06 results.

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Competing magnetic phases in the topological Weyl semimetal EuAuSb

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In the past decade, topological semimetals with non-trivial magnetic order have generated considerable interest as they exhibit a variety of exciting quantum phenomena coupled with emergent Dirac or Weyl Fermions which has helped gain a deeper understanding of fundamental Physics. Among such materials, compounds exhibiting different magnetic phases have received special attention due to their tunability enabling practical applications.

In this work, we report a combined theoretical and experimental study of topological and magnetic properties of the ternary compound EuAuSb. First, we show the competition of magnetic phases in EuAuSb from *ab-initio* density functional theory calculations. Among the studied magnetic phases is an altermagnet, a fundamentally new type of magnetic order similar to a collinear antiferromagnet with a vanishing net moment but showing non-relativistic spin-splitting as a consequence of crystalline symmetries. All the studied magnetic orderings break the combined spatial inversion and time-reversal symmetries giving rise to a Weyl semimetallic phase in EuAuSb with topologically non-trivial surface Fermi arcs as obtained from our theoretical calculations. Our experimental study on single crystals of EuAuSb reveals it to be an antiferromagnet with a low Néel temperature ($T_N \sim 3.5$ K). Magnetization measurements show the presence of magnetic anisotropy and metamagnetic transitions in EuAuSb whereas transport measurements show chiral anomaly and topological Hall signals. This work is published in Physical Review B [1].

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Adaptive Machine Learning for Accurate Band Gap Prediction in Next-Generation Double Perovskite Oxides

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The band gap's value and nature (direct or indirect) are essential in defining a material's electronic properties and suitability for various technological applications. In perovskites, this property is crucial for light-harvesting efficiency, which directly influences the performance of optoelectronic devices, including solar cells, LEDs, and photodetectors [1]. However, predicting band gaps using first-principles simulations is computationally expensive. To address this challenge, machine learning (ML) offers a promising and efficient alternative approach [2-4]. In this study, we employ ML techniques to predict the band gap characteristics of double perovskite oxides ($A_2BB'O_6$), considering essential attributes like elemental composition, ionic radius, ionic character, and electronegativity. We have constructed a dataset with chemical information and band gap values for 598 double perovskite oxides from the Materials Project database [5]. Various machine learning models such as support vector regression (SVR), random forest (RF), k-nearest neighbours (KNN), and gradient boosting (GB) have been applied to this dataset, to predict the band gap of double perovskite oxides. The predictive performance of these models has been assessed using metrics such as mean absolute error (MAE), root mean square error (RMSE), and the coefficient of determination (R^2). Further, these models have been utilized to predict the band gap of some newly designed double perovskite oxides such as Ba_2FeBiO_6 , Ba_2FeMnO_6 and Ba_2FeCrO_6 . Moreover, the predicted band gap has been validated with the band gap obtained using the density functional theory (DFT) approach and experimentally measured data. Our approach eliminates the need for complex and time-consuming DFT calculations, providing a rapid and cost-effective method for screening electronic band gaps.

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Thermal backflow and rectification from phonon hydrodynamics for next-generation heat management devices

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Recent experiments have provided compelling evidence supporting theoretical predictions of phonon hydrodynamics in graphene and other high thermal conductivity materials. Unlike diffusive heat transport, this hydrodynamic regime resembles viscous fluid behavior and is governed by normal phonon-phonon scattering. Here, we apply the viscous heat equations (VHE) [1]—a thermal analogue to the Navier-Stokes equations (NSE) in the laminar regime—to investigate 2D devices. By decoupling the steady-state VHE in Fourier space, we examine how thermal vorticity and compressibility distinctly characterize viscous heat transport. Furthermore, we establish a direct link between thermal viscosity and a measurable negative thermal resistance, or heat backflow from cooler to warmer regions, and propose a setup for its detection. Additionally, we identify the NSE for the electron fluid as the incompressible limit of the compressible phonon VHE, bridging fluid and thermal transport theories in these systems. Lastly, we propose designs for microdevices capable of exhibiting negative thermal resistance and thermal rectification in experimental settings, paving the way for a new paradigm in solid-state heat management.

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Exploring two-dimensional crystalline phases of boron oxide on Pt(111)

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Over the last 20 years the study of low-dimensional systems has been extremely prolific, giving rise to new technological applications and new developments for fundamental research [1]. Despite the great diversity of two-dimensional materials synthesized so far, only a handful of systems with large open structures, such as the silica bilayer [2], are known. This class of systems is deeply fascinating, as they can form either a crystalline or an amorphous layer depending on the substrate and the growth conditions [3]. The presence of large openings in their structures and their stability under ambient conditions make these materials suitable for numerous possible applications, such as filtrations, anticorrosive coatings and templates for the growth of complex functionalized structures [2].

This work focuses on a first principles density functional theory (DFT) investigation of a novel boron oxide compound recently synthesized on Pt(111) at CNR-IOM (Trieste, Italy). By comparing experiments and simulations, we characterize the system and identify its atomic scale structure, discriminating among different possible compositions and models: it is a one-atom thick material with formula unit B_2O_3 , composed of boroxine rings connected by oxygens, resulting in a structure with low density and large openings. Moreover, the oxygen bridges between boroxine rings can be easily bent, which lead to a Young's modulus an order of magnitude lower than that of graphene [4]. The material interacts with the substrate through a weak van der Waals interaction, therefore we expect that the system can be easily exfoliated, as was done for the silica bilayer [5]. Our simulations allow us to gain a deep understanding of the interplay between bond flexibility and pore size. These results shed some light on the properties of the curious class of two-dimensional zeolite-like materials.

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Stability Enhancement of Cubic CsSnCl₃ as Solid Electrolyte - A Computational Approach

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The intermittency of wind and solar power – the solely sustainable energy sources which are considered to be abundantly available – leaves only one consequence: For the transition towards renewable energy systems, efficient and reliable storage technologies are needed. Batteries are one of the most widely used storage devices, but current technology based on the transfer of Li-ions faces several challenges including their dependence on critical materials with respect to both, scarcity and toxicity. [1]

In our contribution, we will present atomic-scale investigations of potential future battery materials carried out using a combination of density functional theory (DFT) and machine learning interatomic potential (MLIP) calculations. We employed a high-throughput approach in order to evaluate potential dopants for the well-known Cl-ion conductor CsSnCl₃; a solid electrolyte material for chloride ion batteries (CIBs) which is ascribed the capability to fully exploit the potential of this alternative battery type. [2,3] The investigated dopants were chosen based on a dual doping strategy: Cation doping aims at enhancing the stability of the material while the introduction of mobile species, i.e., Cl vacancies/interstitials, balances the formal charge of the system and aims at improving the Cl-ion conductivity.

Through such in silico investigations we significantly narrow down the potential materials space for our experimental coworkers and, thereby, contribute to finding green, cheap and reliable devices for energy storage.

Furthermore, we demonstrate that the chosen computational methodology is very well suited for the problem at hand as by combinatorics, the introduction of dopants and vacancies/interstitials leads to a number of inequivalent supercells in the order of thousands per material, while the chemistries of the various supercells which correspond to each individual material are very similar and a MLIP can be trained to sufficient accuracy with a reasonable amount of DFT calculations.

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Electron-phonon scattering in thermoelectric materials: Generalised Herring and Vogt and machine learning deformation potential approach

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High valley degeneracy in electronic bands enhances a material's thermoelectric efficiency, quantified by the figure of merit (ZT). We investigate the role of valley engineering in improving ZT of p-type PbTe. By aligning the valence band maxima at the L and Σ points in the Brillouin zone, a phenomenon known as "valley convergence," significant enhancements in the figure of merit (ZT) have been observed. However, the impact of inter-valley scattering between L and Σ valleys on ZT remains uncertain. To address this, we have developed a first-principles model to analyze electron-phonon scattering in p-type PbTe [1]. Our model accurately reproduces experimental thermoelectric properties at room temperature [2]. By incorporating temperature-dependent band structure, we can quantify the influence of L and Σ valley convergence on ZT at elevated temperatures (~620 K) [3].

Two-dimensional transition metal dichalcogenides, such as MoS₂, are promising candidates for thermoelectric applications due to their high Seebeck coefficient and low thermal conductivity. We employ a multiscale approach, combining first-principles calculations and machine learning, to accurately compute electron-phonon scattering rates and thermoelectric transport properties in single-layer MoS₂. Our models capture scattering from each phonon mode, including out-of-plane homopolar and in-plane modes, efficiently handling the 2D Fröhlich interaction. Our findings, which align well with experimental data [4], reveal that longitudinal acoustic phonon and the Fröhlich interaction are the primary scattering mechanism in MoS₂.

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Beating the Egg-box Effect in Plane Wave DFT Simulations

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The "egg-box effect" is a known challenge in Density Functional Theory (DFT) calculations which arises from the discretization of continuous quantities, e.g. the electron density. This effect is observed as an unphysical change in the system's total energy, reminiscent of an egg-box, as the system is moved relative to the discrete grid, violating translational invariance. This can cause unphysical results such as geometry relaxations finding incorrect crystal symmetries or negative phonon modes in vibrational spectra calculations. The egg-box effect can be mitigated by using finer grids to represent all continuous quantities however this greatly increases computational cost.

The origin of the effect in plane-wave DFT is in the evaluation of the Exchange and Correlation (XC) energy. We present a novel technique for estimating size of the violation of translational invariance, providing an estimate for the uncertainty. This is done by Fourier interpolating the ground state density to a set of shifted grid positions and re-evaluating the change in XC energy at these grid offsets. This is found to be in very close agreement to the change in total energy from the egg-box effect. Our results also show that the numerical behavior of XC approximations [1] is strongly linked to the magnitude of the violation of the translation invariance. The more numerically ill-behaved XC functionals, such as the more advanced GGAs and meta-GGAs functionals, exhibit violations of translation invariance that are orders of magnitudes worse than less advanced functionals. Performing this analysis at an early stage of a workflow can inform the user about the expected accuracy of subsequent calculations.

Further, our results demonstrate that by selectively computing the XC energy and its corresponding potential on a finer grid, via Fourier interpolation, violations of translation invariance can be reduced and the egg-box effect can be eliminated from calculations altogether. Coupled with the uncertainty quantification method, egg-box related inaccuracies can be avoided more conveniently than just increasing all the grids until inaccuracies appear to have been converged out. This work offers a promising pathway towards mitigating the egg-box effect in a diverse range of materials modelling applications.

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Perovskite Molecular Dynamics Enhanced by LATTE: Machine Learning Potentials Using Local Atomic Tensors

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Perovskites are crucial for many applications, such as solar cells. Computational studies of these materials using ab initio modeling techniques are limited due to constraints on feasible system sizes and length scales. A complementary approach is molecular dynamics simulation. However, traditional interatomic potentials often struggle to capture the complex interactions within perovskites, especially under varying temperature and pressure conditions [1].

In this work, we employ the Local Atomic Tensor Trainable Expansion (LATTE) descriptor, recently developed by Pellegrini et al.[2], to create a machine-learning-based force field for CsPbI₃ perovskite. Our results show that the LATTE-trained force field achieves a lower loss compared to results reported for ACE on the same dataset. These findings indicate that LATTE can provide highly accurate machine-learning potentials, holding significant promise for advancing perovskite-based applications.

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Pressure And Magnetic Field Control Of The Topological Phase In Antiferromagnetic Bilayers

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Antiferromagnetic bilayers are said to be A-type when spins align ferromagnetically within each layer but point in opposite directions in the two layers, resulting in a zero net magnetization. The opposite orientation of majority spins in the two layers strongly suppresses interlayer hopping and the electronic structure is not affected significantly by a change in interlayer distance. Nonetheless, an external magnetic field can drive a metamagnetic transition into a fully ferromagnetic state, where spins in both layers are parallel, so that interlayer hopping is possible and energy bands are sensitive to the separation between the layers. This difference can be exploited to manipulate the energy bands of an A-type antiferromagnetic bilayer by means of the combined effect of pressure and magnetic field. Here we consider bilayer CrSBr as a prototypical example and show using first-principles simulations that pressure affects the interlayer distance, enhancing the interlayer hopping in the ferromagnetic state, and eventually closes the energy gap, inducing a topological phase transition. Remarkably, depending on the magnetization direction it is possible to tune the system either in a quantum anomalous Hall insulating state when spins are out-of-plane or into a half Chern-Weyl semimetallic phase when spins are in-plane, with the emergence in both cases of topological edge states. We expect this phenomenon to be general to A-type antiferromagnetic bilayers, opening interesting perspectives on the manipulation of their topological character towards applications in spintronics and quantum computation.

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Perpendicular magnetic anisotropy in H-VTe₂ monolayer by hole doping

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Two-dimensional materials with large perpendicular magnetic and high Curie temperature offer great potential to improve low-dimensional spintronic devices. It has been shown that these properties can be modulated by strain engineering or by carrier doping in some materials. Of these materials, vanadium dichalcogenides have sparked interest for their versatile band structure and tunable magnetic properties [1, 2]. Their monolayers are stable in either the octahedral T-phase or the trigonal prismatic H-phase. While theory and experiments have shown that the T-phase is the ground state of vanadium ditelluride (VTe₂), it is metallic with a charge density wave that inhibits ferromagnetism. On the other hand, the H-phase is an indirect band-gap semiconductor with a ferromagnetic ordering at room temperature [3]. We perform a density functional theory study to determine the effect of charge doping on the electronic and magnetic properties of the H-phase of VTe₂ monolayer. The pristine H-VTe₂ has an in-plane easy magnetization axis, but upon hole doping, we have observed a switching of the magnetic anisotropy energy to easy out-of-plane magnetization. This perpendicular magnetocrystalline anisotropy (MCA) is in the order of milli-electron volts even for small hole doping. As the hole doping concentration is increased, the MCA and the magnetic moment also increase. The effect of both hole and electron doping is explained in the framework of the rigid band model. The switching of MCA upon hole doping is enabled by the spin-orbit splitting of the valence band edge which depends on the magnetization direction.

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Nearly Quantized Born Effective charges as probes for the topological phase transition in the Haldane and Kane-Mele models

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A new approach to study the topological phase transitions, based on the assessment of the vibrational resonances in infrared spectra, is here proposed [1]. In the prototypical models for 2D Chern and Quantum Spin Hall topological insulators, the Haldane and Kane-Mele one respectively, the Born effective charges, quantifying the intensity of vibrational resonances, are nearly quantized, with a discontinuous jump associated with the topological phase transition, corresponding to a finite value in the trivial phase and a null one in the nontrivial one. This result is explained by the connection between Born effective charges and electronic Berry curvature at the band edges [2]. Finally, at the topological phase transition of the Haldane model, due to the time-reversal symmetry breaking, a nearly quantized jump of the chiral splitting of the phonon frequencies is observed ([3, 4, 5]).

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Comprehensive Screening of Plasma-Facing Materials for Nuclear Fusion

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Plasma-facing materials (PFMs) represent one of the most significant challenges in the design of future nuclear fusion reactors [1]. Inside the reactor, the divertor experiences the harshest material environment: intense neutron and plasma bombardment coupled with extremely large heat fluxes. Tungsten is the material chosen for ITER due to its properties, though it still faces several challenges in a reactor environment. This work provides a comprehensive screening of candidate PFMs based on experimentally known materials [2]. The methodology to identify the most promising PFMs combines peer-reviewed data of inorganic crystals from the Pauling File database and first-principles DFT calculations of two key PFMs properties, namely the surface binding energy and the formation energy of hydrogen interstitials. The materials that meet the necessary thermal performance to withstand heat loads are critically compared with the state-of-the-art literature, defining an optimal subset where to perform first-principles calculations. Well-known PFMs - such as W, Mo, and carbon-based materials - are captured by this screening, along with less familiar refractory materials that warrant further investigation.

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Functional theory of the occupied spectral density

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We address the problem of interacting electrons in an external potential by introducing the occupied spectral density as fundamental variable. First we formulate the problem using an embedding framework [1] and prove a one-to-one correspondence between a spectral density and the local, dynamical external potential that embeds it into an open quantum system. Then, we use the Klein functional to (i) define a universal functional of the spectral density, (ii) introduce a variational principle for the total energy, and (iii) formulate a non-interacting mapping suitable for numerical applications. The resulting equations, which involve local and dynamical potentials, are then solved by using the algorithmic inversion method [2, 3, 1] based on a sum-over-poles to represent propagators.

At variance with time-dependent density-functional theory, this formulation aims at studying charged excitations and electronic spectra with a functional theory, although an explicit and formally correct description of all electronic levels could also lead more naturally to accurate approximations for the total energy.

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Effects of colored disorder on the heat conductivity of SiGe alloys from first principles

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Semiconducting alloys, particularly SiGe, have been employed for several decades as high-temperature thermoelectric materials. Devising strategies to reduce their thermal conductivity may substantially improve their thermoelectric performance, especially at lower temperatures [1]. We have carried out [2] an ab initio investigation of the thermal conductivity of SiGe alloys with random and spatially correlated (“colored”) mass disorder, employing the Quasi-Harmonic Green-Kubo (QHGK) theory [3] with force constants computed by density functional theory.

Leveraging QHGK and hydrodynamic extrapolation [4] to achieve size convergence, we obtained a detailed understanding of lattice heat conduction in SiGe and demonstrated that colored disorder suppresses thermal transport across the acoustic vibrational spectrum, in agreement with recent theoretical findings [5]. Remarkably, this suppression leads to a fourfold enhancement in the intrinsic thermoelectric figure of merit.

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Structural models of amorphous C-based materials from machine learning interatomic potentials

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Carbon-based materials, such as activated carbon (AC), find extensive application in storing natural gas and hydrogen, as well as in selective filtration or CO₂ capture through post-functionalization. The high specific surface area of AC (thousands of m²/g) provides numerous active sites where gas molecules can interact with the support, facilitating significant gas storage. This study focuses on developing realistic models of amorphous carbonaceous materials that accurately replicate known experimental descriptors. Given the structural complexity of these materials, both static and kinetic simulations employ precise and efficient interatomic potentials, such as machine learning interatomic potentials (MLIPs). This approach marks a paradigm shift, replacing physics-based functional forms of potentials with highly complex mathematical forms based on neural networks. These networks, trained on reference ab-initio datasets, can reproduce high-level quantum mechanical calculations with negligible errors, achieving accuracies comparable to experimental uncertainties. State of the art MLIP software like NequIP [1] and Allegro [2] have been trained on available carbon-based ab-initio datasets. Trained models have been tested against ab-initio and experimental data, and compared to available interatomic potentials foundation models [3].

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Crystal Structure Prediction with Anharmonic Lattice Dynamics

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Crystal structure prediction (CSP) is a well-established tool in computational condensed matter physics and materials science, mainly for identifying stable phases on potential energy surfaces (PES). However, traditional CSP methods may miss configurations that are stabilized by quantum fluctuations and anharmonicity, such as superconducting hydrides. The stochastic self-consistent harmonic approximation (SSCHA) [1] can treat anharmonic lattice dynamics, though its computational cost limits the exploration of a broader range of crystal structures. Here, we integrate a machine learning interatomic potential (MLIP), specifically MACE [2], with CSP and SSCHA, making this combination computationally feasible. Using H₃S as a case study, we train a potential starting from the MACE foundation model [3] iteratively on configurations generated by CSP. This trained potential supports extensive structure searches and following SSCHA relaxations under various pressures. The pretrained foundation model reduces the dataset requirement, and the fine-tuned potential correctly predicts the stability sequence of known phases, even when some of them are not included in the dataset. While MLIP-guided searches find numerous artificial minima on the PES, SSCHA relaxations significantly filter these. This work demonstrates the possibility to incorporate anharmonicity and temperature effects into CSP, advancing the search for high-temperature superconducting hydrides at lower pressures or even under ambient conditions.

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INTW code: Unifying Electron-Phonon Coupling Analyses Across LCAO and Plane-Wave Basis

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Over the past few decades, first-principles calculations have been essential for enhancing our understanding of many important problems in physics and materials science. Density Functional Theory (DFT) has played a paramount role in this area, often providing the theoretical framework for examining a wide array of phenomena related to electron-electron and electron-phonon interactions using ab-initio, which primarily rely on the output generated by DFT codes as their input, it is important to note that these options are mainly limited to plane-wave DFT codes.

Here we introduce the INTW package, a new modular environment designed for conducting many-body calculations from first-principles. A key feature of INTW is its compatibility with two popular DFT codes: Quantum Espresso and SIESTA. This flexibility allows users to select either a plane-wave or an LCAO basis, tailored to the specific needs of their research. Furthermore, INTW prioritizes efficiency by using crystal symmetries, deriving all electron states, phonon dynamical matrices, and induced potentials through symmetry operations from their self-consistently calculated counterparts in the irreducible wedge of the Brillouin zone.

Due to its modular design, INTW provides a wide range of capabilities for post-processing DFT calculations. Among others, it has been successfully applied to problems involving the calculation of electron-phonon coupling, Eliashberg functions, self-energy functions, and spectral functions [1, 2]. Notably, its application has extended to solving the anisotropic non-linear Eliashberg equations of superconductivity [3] and determining the spin lifetime in magnetic adatoms [4].

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Surprising excitons: from solid helium to the homogeneous electron gas

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Excitons are interacting electron-hole pairs due to the attractive screened Coulomb interaction. They dominate absorption spectra of semiconductors and low-dimensional materials, which are usually successfully described by the ab initio solution of the Bethe-Salpeter equation (BSE) in the GW approximation [1].

Here, we discover and explain surprising excitonic effects at large momentum transfers in two opposite limits: solid helium, an insulator with a gap larger than 20 eV, and the homogeneous electron gas (HEG), i.e., the prototypical model for metals.

In helium, excitons are tightly bound, as expected. However, they show a striking anisotropy for spectra at momentum transfers around Bragg reflections [2]. Infinitesimal momentum-transfer changes entirely suppress or create intense exciton peaks. We explain this giant effect in terms of the electron-hole exchange interaction and make the link with the anomalous anisotropy of plasmons in graphite that was understood in terms of crystal local field effects [3].

In the HEG, excitonic effects are supposed to be weak due to the perfect screening of the Coulomb interaction. In contrast to these expectations, we show that, besides the classical high-energy plasmons, in the low-density regime, where static screening turns its sign becoming negative, low-energy excitonic collective modes also exist [4] that are due to reduced screening at short distances [5]. The standard BSE fails to capture these excitonic effects, which require to eliminate the GW self-polarization error that overscreens the electron-hole interaction. Understanding this exotic regime is crucial for (photo)doped semiconductors and interfaces.

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Enhancing thermoelectric figure of merit of half Heusler alloys through hierarchical bonding and disorder: Some theoretical predictions

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Thermoelectric (TE) materials are used in devices to convert waste heat to energy. Their efficiency is measured by a dimensionless quantity called figure of merit ($zT = \frac{S^2\sigma}{(\kappa_e + \kappa_l)}$). Designing efficient thermoelectric materials are challenging because the transport properties that determine their efficiency are interdependent and in a reverse way. For example, to increase zT both electrical conductivity (σ) and Seebeck coefficient (S) needs to be enhanced. While σ can be enhanced by increasing carrier concentration, the latter is detrimental for Seebeck coefficient (S). Hence there are lot of efforts to tune the transport properties of materials to enhance zT . Moreover, a large family of materials are being explored for applications at different temperatures.

Amongst these half Heusler (HH) alloys are plausible candidates for high temperature thermoelectric applications. HHs, comprising of early (X) and late (Y) transition metals and p-block elements (Z) and with chemical formula XYZ, are highly stable at high temperatures and have excellent electronic transport properties. However, their applications are limited by very high lattice thermal conductivity (κ_l), the reason for which can be attributed to their highly symmetric cubic structure. Hence there are efforts to reduce the lattice thermal conductivity by designing HHs with more complex structures. This is typically achieved by introducing disorder in the X, Y and Z positions. Though there are lot of experimental studies and high throughput computational studies to design novel HHs with low lattice thermal conductivity, there are very few studies on the microscopic understanding of how disorder affects κ_l , i.e., is the lattice thermal conductivity reduced due to mass fluctuations induced by the disorder or there is changing in the chemical bonding within the lattice?

In this work, using density functional theory and semi-classical Boltzmann transport theory based calculations, we have studied double HH and high entropy HH alloys to discern the role of chemical bonding and disorder on lattice thermal conductivity of these material. In particular, we studied thermoelectric properties of $X_2FeNiSb_2$ (X=Ti, Zr and Hf), Nb_2Co_2ZSb (Z=In and Ga) and $ZrHfCoNiSnSb$ HHs. Through our calculations we show that the disorder results in heterogeneity in chemical bonding. Some of the bonds become more ionic while others become more covalent compared to the parent HHs thereby introducing hierarchical bonding in the lattice. As a consequence, we observe that the lattice becomes anharmonic, thereby showing reduced lattice thermal conductivity. Further, the mass fluctuations due to the variation in the atomic masses of the constituent elements scatter the heat carrying optical phonons, thereby further reducing κ_l . Our calculations predict that these materials have a $zT > 1$ around 900 K and at moderate doping concentration ($\sim 10^{21}/\text{cm}^3$).

High temperature and pressure thermoelasticity of hcp metals from ab initio quasi-harmonic free energy calculations: the beryllium case

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We present a systematic ab-initio study of the temperature- and pressure-dependent thermoelastic properties of hcp beryllium within the quasiharmonic approximation (QHA). The accuracies of the zero static internal stress approximation (ZSISA) and of the volume-constrained ZSISA that are widely applied in ab initio thermodynamic calculations are quantified. Particularly, the effect of ZSISA on the calculation of C_{11} and C_{12} is compared with a numerical approach which minimizes the free energy with respect to the atomic positions at each strain. In beryllium, minor deviations are found within ZSISA, which gives elastic constants (ECs) in good agreement with the full free energy minimization. A substantial difference is found between the QHA and the quasistatic approximation (QSA), with the former closer to experiments. Within the QSA, we compare the ECs obtained by interpolating from a set of geometries along the “stress-pressure” isotherm at 0 K (within the constant-volume ZSISA) with a more general interpolation on a two-dimensional grid of crystal parameters, which allows the calculation of the ECs along the 0 kbar isobar. This paper provides a practical approach for the investigation of the thermoelastic properties of hcp metals at extreme conditions. [1]

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Automated Workflows and Machine Learning models for X-ray spectra simulations: applications to Li-ion battery materials

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In the framework of material science, core-level spectroscopies are established strategies to probe the electronic structure and chemical environment of materials. While these techniques provide valuable information, their interpretation in complex systems is not straightforward, emphasizing the importance of theoretical insights from ab-initio approaches. On the other hand, the time required for computing an X-ray spectrum increases with the number of non-equivalent atoms, becoming prohibitively expensive for complex amorphous materials. This drawback can be addressed by employing a surrogate model that combines the precision of ab initio methods with computational efficiency [1]. We trained a machine-learning (ML) model based on Kernel Ridge Regression (KRR) [2], using atom-density descriptors for predicting X-ray Photoelectron Spectroscopies (XPS), by using core-electron Binding Energies (BE) as the target quantity. A comprehensive automated AiiDA workflow [3], integrating first-principles XPS simulation with sample sub-selection via Farthest Point Sampling (FPS), was employed to generate the critical amount of data needed for the training process. The ML models were trained on a representative dataset comprising about 250 lithiated Si-based structures, previously obtained through ab initio molecular dynamics (MD), DeePMD and grand canonical Monte Carlo simulations [4]. Validation on a dataset of around 50 structures demonstrated an accuracy of 0.1 eV, aligning with typical XPS experimental resolution. We leveraged the model to construct a stoichiometry map in order to identify the Li_xSi phases that form at various potentials in Si-based anodes.

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Tuning of ferroelectric polarization in a polar metal: the case of bilayer VTe₂

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In polar metals [1] ferroelectricity and metallicity coexist. While polar metals are rare in general, the class of two-dimensional metallic van der Waals materials offers a number of candidates. In such systems the weak coupling between layers prevents efficient electron screening of the ionic polarization perpendicular to the layers, allowing net polarization in that direction. The ferroelectric polarization can appear as a consequence of relative sliding of the van der Waals layers in some of the systems. Two-dimensional transition metal dichalcogenides (TMDs) are ideal playground for investigating sliding ferroelectricity [2]. In particular, vanadium ditelluride, VTe₂, displays a number of peculiar properties, such as charge-density waves [3], a multitude of stable magnetic configurations, strong spin-orbit coupling and ferroelectricity in one or more of its phases. The ground state of the bilayer VTe₂, in the charge-density wave T' structure, is metallic and multiferroic. In this work, we search for effective ways to tune the ferroelectric polarization of the ground-state bilayer T'-VTe₂. Specifically, we examine the effects of doping, strain, and defects, such as vacancies and chalcogen atom substitutions, as well as the impact of layering VTe₂ with different TMDs. We analyze and generalize our results to other van der Waals bilayers.

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Using *ab initio* methods to identify an optimum catalyst formulation: ethylene epoxidation

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Ethylene epoxidation is a reaction of great industrial significance, where silver serves as a catalyst under oxygen. While traditionally optimized through empirical methods, the advent of *ab initio* density functional theory (DFT) has enabled an atomistic understanding of the reaction mechanisms involved. Here, we focus on the role of DFT in elucidating the catalytic properties of silver and its alloys in ethylene epoxidation, emphasizing the predictive power of these quantum mechanical approaches.

We first present a brief review of the contributions of DFT calculations to understanding the surface chemistry of silver catalysts, particularly with respect to adsorption energetics, transition state structures, and reaction pathways. Additionally, we highlight how *ab initio* models have provided insights into the electronic and geometric effects of dopants and co-catalysts, which have historically been optimized empirically. [1, 2, 3]

Recent advances in computational resources have enabled the development of increasingly accurate DFT models. These models can better capture complex catalytic behaviour under reaction conditions, including surface oxidation and the role of intermediates. [4]

Finally, we report on a high-throughput computational screening of metals using DFT-based simulations, which identified several promising silver alloys, such as Ag/CuPb, Ag/CuCd, and Ag/CuTl, as superior catalysts. These findings were experimentally validated, demonstrating improved catalytic performance and scalable synthesis protocols. Importantly, we incorporate realistic reaction conditions and address common pitfalls in computational catalysis, such as the neglect of side reactions and catalyst surface dynamics. By integrating DFT calculations with kinetic Monte Carlo simulations and microkinetic modeling, we provide a comprehensive, first-principles understanding that goes beyond traditional steady-state approximations. [5]

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First-principles study of layer-resolved anomalous Hall conductivity in thin films and heterointerfaces

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Recent studies have highlighted the anomalous Hall effect (AHE) for its distinct characteristics observed at surfaces, interfaces and within the bulk of a diverse range of material systems, including topological magnetic insulators and metallic interfaces. Analyzing the contributions of individual layers to the AHE is essential for understanding the electronic and topological properties of complex materials. In this work, we extended computational methods for AHE analysis using a local Berry phase approach[1] in combination with hybrid Wannier functions. This approach enables us to resolve layer-specific contributions in magnetic topological insulators as well as magnetic metal thin films. Notably, our method successfully predicted a half-quantized AHE at the surface of Axion insulators. We further investigate the AHE at surfaces and interfaces in systems such as Fe(001)/W thin films and oxide heterostructures such as SrRuO₃/SrIrO₃, revealing how topological electronic states impact transport properties. This presentation will cover our findings on the layer-resolved AHE in various materials, demonstrating how a layer-by-layer analysis reveals new topological insights and enhances our understanding of transport phenomena in complex systems.

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Optical Properties of Cementitious Oxides for Design of Photonic Metaconcrete – an *Ab initio* study

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Abstract

Concrete is the single most commonly produced material by humans. The downside of large-scale production of cement, the main ingredient in concrete, is a massive CO₂ footprint, responsible for about 10% of global emissions. In addition, **buildings** account for approximately **40% of total electricity consumption**. There is therefore an urgent need to reduce the environmental impact of concrete throughout its lifecycle. One potential solution is to reduce the energy consumption of buildings by exploiting the **radiative cooling phenomenon** [1]. The **MIRACLE** (Photonic Metaconcrete with Infrared Radiative Cooling capacity for Large Energy savings) project is pioneering a new generation of green concrete capable of passive cooling to reduce energy consumption in buildings [2]. Radiative cooler dissipates heat within the atmospheric transparency window (ATW), a range of infrared frequencies to which the atmosphere is permeable. The heat emitted within the ATW escapes the atmosphere and dissipates into space. This allows the object to thermally couple to an environment that is significantly colder than its surroundings and effectively cool down. In practice the research aims to develop photonic concrete by employing structured metamaterials [3] to manipulate the absorption and emission spectrum of concrete.

To engineer effective daytime radiative coolers, we need to find materials whose emissivity matches the ATW. Additionally, to minimize heat absorption from the sun, the metaconcrete should exhibit high reflectivity at frequencies outside the ATW range. Both absorbance and reflectance are inherent material characteristics linked to their optical dielectric properties, which originate at the atomic and electronic level and can be accurately predicted by first principles calculations. To advance the ideas of photonic concrete, we provide an overview of the **optical properties and excitonic spectra of relevant oxides**. In our research, we compute the frequency-dependent dielectric function using various levels of approximation [4, 5]. The electronic structure based on the Kohn-Sham (KS) approach often suffers from significant inaccuracies in band gaps and band structures. A correction can be made **using the GW approximation**, which treats electrons and holes as screened quasiparticles [5]. Nevertheless, to accurately simulate experimental spectra, we must consider the electron-hole interaction [6], which significantly influences the spectra, especially around the band gap energy. This interaction can be accounted for by **solving the Bethe-Salpeter equation** (BSE) [6].

Our particular focus is on understanding the excitonic properties of basic oxides such as CaO, SiO₂, Al₂O₃ and Fe₂O₃ which serve as building blocks for complex cement oxides like alite (Ca₃SiO₅) and belite (Ca₂SiO₄). Additionally, we investigate other clinker phases, including gypsum (CaSO₄·2H₂O) and calcium sulfate (CaSO₄), as well as hydrated phases like portlandite (Ca(OH)₂) and tobermorite (Ca₅Si₆O₁₆(OH)₂·4H₂O/ Ca₅Si₆O₁₆(OH)₂·7H₂O), among others. The dielectric spectra obtained from these studies are used to examine the light scattering properties and serve as input to machine learning software aimed at predicting the ideal composition, shape, and distribution of the studied metamaterial. From a computational point of view, we use GGA [7] and SCAN meta-GGA [8] exchange-correlation ground state calculations, combined with the G₀W₀ and BSE approaches, as implemented in the VASP package [9].

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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.

Theoretical study of cyanoacrylated terthiophene bonded to ZnO (10-10) surface for dye-sensitized solar cells

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Cyanoacrylated thiophenes linked to ZnO surface have been proposed [1] as a promising combination for Dye-Sensitized Solar Cells, that is, a design for the electron-transfer layer after light capture. We present a theoretical study of the ZnO (10-10) surface functionalized by a cyanoacrylated terthiophene molecule (3TC) using Density Functional Theory, and also optical properties for the isolated molecule through the Bethe-Salpeter formalism. We find that 3TC displays a strong optical excitation around 2.7eV mostly due to the HOMO→LUMO transition, with the HOMO localized on the 3T part and the LUMO on the C termination (which will be the bonding link to the surface). Regarding the 3TC@ZnO, we obtain a molecule-surface binding energy indicating stable chemical bonding, and the electronic structure results show energy levels alignment suitable for light-induced charge transfer, i.e., the molecular HOMO energy in the middle of the ZnO gap, and the LUMO in the conduction band. Importantly, the HOMO state is concentrated on the molecule while a molecule-to-surface charge extension of the LUMO was found, indicating a real possibility of photo-excited charge transfer from 3TC to the ZnO surface. The authors thank HPC-USP, CENAPAD-SP, INCT-INEO and CNPq.

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Periodic All-Electron Δ SCF Calculations of Core Electron Binding Energies in Solids

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Theoretical methods for predicting core electron binding energies are important for the analysis of experimental results from X-ray Photoelectron Spectroscopy (XPS). The Δ -Self-Consistent-Field (Δ SCF) method is a well-known technique for calculating core electron binding energies in free molecules, but the application of this approach to periodic solids is challenging.

In this poster, recent progress in *ab initio* calculations of core electron binding energies is presented [1,2]. Solutions to problems such as charge compensation, energy referencing, and the elimination of finite size effects are demonstrated, and an innovative method that combines the Δ SCF method for core electron binding energies with the GW approach for predicting the position of the valence band maximum is described. Finally, the performance of current state-of-the-art methods is evaluated by comparing the theoretical results to experimental data.

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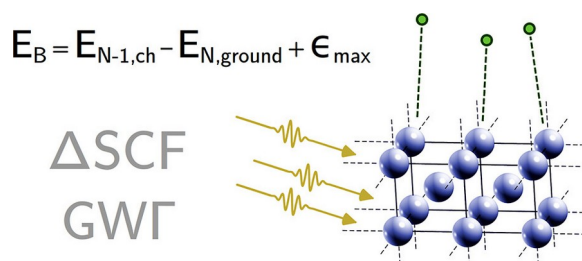


Fig. 1 Graphical abstract from reference [2]. The GW and Δ SCF methods can be combined for predicting core electron binding energies in insulating solids, when the position of the valence band maximum (VBM) is used as the zero of the binding energy scale.

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Impurity diffusion in solids plays a crucial role in various important phenomena, including material aging [1]. In metals, the diffusion of small chemical elements (such as H, B, C, N and O) occurs through an interstitial mechanism [2]. Among these elements, nitrogen is particularly significant in iron alloys, as it is the most effective additive for martensitic steel. This type of steel is widely utilized in industrial and technological applications due to its superior strength compared to other materials [3]. Notably, the presence of neighboring impurities greatly influences the diffusion process. However, experimental techniques are insufficient to study these short-range impurity interactions, making theoretical approaches essential for accurate estimations.

We achieved the investigation of nitrogen diffusion in the presence of other nitrogen atoms using DFT study via VASP code and then Kinetic Monte Carlo (KMC) techniques. We considered all possible nitrogen-nitrogen pair configurations that correspond to local energy minima, with N-N distances extending up to 6 Å. Employing the Nudged Elastic Band (NEB) method, we identified all migration paths and their associated saddle points between these local minima (Fig. 1 a/b). This *first-principles* energy database was subsequently integrated into KMC simulations to calculate the thermodynamic and kinetic parameters.

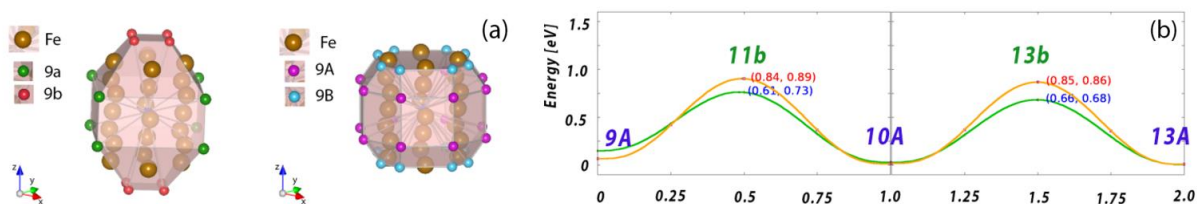


Figure 1. Stable N-N configurations (a) are used to find migration saddle points during diffusion (b).

Our simulations reveal that the repulsive interactions between N-N pairs result in several significant effects: (1) the alterations of order/disorder transition conditions based on nitrogen concentration and temperature; (2) a notable increase in nitrogen diffusivity, particularly along the Zener ordering direction, which can be up to a factor of ten; and (3) significant kinetic correlations. The results indicate that the diffusion coefficient in the direction perpendicular to the Zener ordering direction (D_x) is only slightly affected by N–N interactions, with D_x increasing by approximately 10% at a nitrogen concentration of 4 at.% when these interactions are considered. In contrast, the diffusion coefficient along the Zener ordering direction (D_z) is significantly influenced by N–N interactions, increasing by an order of magnitude at the same nitrogen concentration when these interactions are considered [4]. These results are constantly compared with previous results from our C-C interaction studies [5].

These findings underscore the importance of considering these effects for accurately predicting low-temperature aging in ferritic and martensitic Fe–N alloys.

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First-principles phase diagram and electronic structure properties of $\text{KCu}_3(\text{Se,S})_2$ alloys for photoelectrodes

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A recent high-throughput study of copper-based semiconductors has identified potassium-based copper chalcogenides as optimal light absorbers in PV/PEC devices. In this work, we investigate the applicability of materials in the homologous series $\text{KCu}_3\text{Se}_{2(1-x)}\text{S}_{2x}$ ($0 \leq x \leq 1$) as proton reduction photocathodes. We first calculate the temperature-composition phase diagram of $\text{KCu}_3\text{Se}_{2(1-x)}\text{S}_{2x}$ through solid solution models and Monte Carlo simulations based on cluster expansion. Our calculations predict that the alloy forms a solid solution in the monoclinic structure over the entire composition range although ordered ground states are present at low temperatures. On the bases of the stable structure of the alloys, we calculate the electronic structure properties via DFT. Unlike the bandgap bowing typical of highly mismatched alloys, predicted by band-anticrossing (BAC) model the electronic band gap of potassium-based copper poly-chalcogenide alloys obey the simple rule of mixtures. The optical absorption coefficient, mobility, and band alignment of the alloy with water redox potentials suggest its effectiveness as a photocathode.

First Principle Studies Of Ternary Hexafluorides as Scintillator for Medical Imaging Technologies

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Scintillators are materials that convert ionizing radiation into detectable low-energy radiation and are crucial in applications such as medical imaging technologies like computed tomography (CT) and positron emission tomography (PET) [1]. In time-of-flight PET (TOF-PET), the signal-to-noise ratio and spatial resolution are directly related to the coincidence time resolution of scintillator detectors [2]. The discovery of new scintillators with higher yield and shorter response times would enable the development of more efficient and accurate detectors, potentially allowing for higher-resolution images and reduced radiation doses to patients. However, experimental testing of scintillator materials is time-consuming and requires expensive specialized equipment, making computational identification of promising candidates highly desirable [3].

In this study, we investigate inorganic hexafluoride scintillators (K_2GeF_6 , K_2SiF_6 , $BaGeF_6$ and $NaGeF_6$) using first-principles modeling to determine their fundamental operating principles. In this poster, we present the results of our preliminary investigations of the electronic structures of these ternary hexafluorides based on Density Functional Theory (DFT), and compare them to the available experimental data from measurements of optical absorption, luminescence, and photoemission.

In the future, we plan to further examine quasiparticle energy levels and electronic excitations in these materials using the GW approximation and the Bethe-Salpeter equation, and to study the role of phonons in the optical spectra of these materials.

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Linear response DFT+U+J produces accurate band-gaps and defect properties on transition metal oxides

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Computationally fast density functional theory(DFT) methods such as PBE frequently underestimate the band gap of materials such as transition metal oxides (TMO's). Hubbard U corrections are often used to reduce this level of band gap underestimation without incurring significant computational cost. However this requires the selection of a U parameter for the level of correction, which has often been chosen on an arbitrary basis or fixed to experiment. In response to this, methods such as linear response DFT+U+J have been proposed, which calculate both hubbard U and Hunds J parameters from first principles and implements them seperately as energy corrections¹.

In our research², a standardised first-principles procedure is implemented for calculating U and J for both metal and oxygen elements using the linear response to an applied potential. This procedure is benchmarked with five different TMO materials to assess the effect of DFT+U+J on the band gap, effective mass, bond lengths and unit cell volume.

Our results showed that over this test set, DFT+U+J predicted band gap accuracy at a level that was significantly greater than regular PBE, and had similar performance to literature averages for the much costlier HSE06 and PBE0 hybrid functionals. This was accomplished with little geometric distortion, producing cell volumes that were more accurate than PBE, and minimal electronic distortion, with effective masses that were similar to uncorrected PBE.

Following on from this, we extended this analysis to the use of defect formation energy calculations, using TiO₂, ZrO₂ and HfO₂ as candidate materials. This introduced new challenges, as in the DFT+U+J method O atoms of different coordination have different U parameters, which introduces significant problems with formation energy calculations. We tested three potential methods to alleviate this, and found the "differing potentials method" to produce the outcomes most in line with literature values. This method involves simulating seperate reference oxides with corresponding U and J parameters applied.

Using the corrected methods, we found O vacancy defect formation energies transition energies, and that were qualitatively in line with hybrid results for ZrO₂ and HfO₂, although TiO₂ was less accurate, possibly due to it's shallow defect level. Overall we show there is significant potential for DFT+U+J to be used for fast, first principles predictions of TMO properties.

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Predicting electronic screening for fast Koopmans spectral functionals

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Koopmans spectral functionals are a powerful extension of Kohn-Sham density-functional theory (DFT) that can predict spectral properties with state-of-the-art accuracy in a functional framework [1]. The success of these functionals relies on capturing the effects of electronic screening through scalar, orbital-dependent parameters, which have to be computed for every calculation, making Koopmans spectral functionals more expensive than their DFT counterparts [2]. In this work, we present a machine-learning model that — with minimal training — can predict these screening parameters directly from orbital densities calculated at the DFT level [3]. We show on two prototypical use cases that using the screening parameters predicted by this model, instead of those calculated from linear response, leads to orbital energies that differ by less than 20 meV on average. Since this approach dramatically reduces run-times with minimal loss of accuracy, it will enable the application of Koopmans spectral functionals to classes of problems that previously would have been prohibitively expensive, such as the prediction of temperature-dependent spectral properties. More broadly, this work demonstrates that measuring and correcting violations of piecewise linearity (i.e. curvature in total energies with respect to occupancies) can be done efficiently by combining frozen-orbital approximations and machine learning.

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Delta-learning interatomic potential for bulk nickel oxide based on multi-reference methods

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Conventional density functional theory (DFT) often fails qualitatively to describe the electronic structure of strongly correlated transition metal oxides. On the other hand, wavefunction methods [1] and dynamical mean-field theory [2] have been applied successfully to mitigate the problem, however, their application to bulk materials is still preliminary [3, 4]. Here we focus on using delta-learning to correct DFT energy and derivatives to the level of multi-determinant theory for bulk NiO. We use complete active space self-consistent field (CASSCF) calculations to capture on-site strong correlation in clusters representative of the bulk environment, and then train a machine learning interatomic potential (MLIP) on the difference of the two energy landscapes. The Δ -machine learning scheme is necessary for two purposes: 1) to isolate long-range interaction captured by DFT and the localized correlation 2) to reduce the complexity of the energy landscape and therefore the number of expensive calculations needed [Nandi et al. 2021]. We also design atomic descriptors including both structural and electronic information and automate CASSCF workflow to facilitate the training procedure. This Delta-MLIP leverages the efficiency of the density function theory and the accuracy of wavefunction methods and is applicable to a variety of systems with localized multi-determinant correlations.

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A Deep-Learning-Driven Hybrid Monte Carlo Approach for Predicting Phase Relations in Solid Solutions

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Phase diagrams of solid solutions are essential for understanding structural transformations under varying pressure and temperature conditions, serving as the foundation for guiding the design of alloys and composites with targeted properties. These diagrams are also crucial for interpreting geophysical observations related to Earth's deep interior. Atomistic simulations, particularly those based on *ab initio* methods, have shown significant success in accurately predicting phase diagrams for pure and ordered solid phases. However, these simulations often struggle to maintain *ab initio* accuracy while capturing the complex atomic disorder characteristic of solid solutions at finite temperatures. To address this challenge, we developed a methodology that integrates a deep-learning-based interatomic potential derived from *ab initio* calculations [1] with efficient sampling through thermodynamic integration and hybrid Monte Carlo algorithms [2]. This approach enables a rigorous and simultaneous treatment of atomic configurational and positional disorder. We validated this methodology by computing the phase diagram of the (Mg,Ca)O binary alloy [2], which shows excellent agreement with experimental data. When applied to the Fe-Si and Fe-Si-O alloys under Earth's core conditions, our method indicates that, at Earth-relevant concentrations of Si and O, the solid Earth's inner core is likely to exist in the body-centered cubic (bcc) structure, rather than the hexagonal close-packed structure observed for pure Fe. Furthermore, we found that the stability of the bcc phase is critically dependent on the presence of short-range ordering in Si and O [3]. The demonstrated accuracy and efficiency of our approach open new avenues for determining the free energies of solid solutions with *ab initio* accuracy, thereby advancing both materials design and our understanding of geophysical processes.

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Modeling Molecular Crystals with Machine Learning Interatomic Potentials

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Molecular crystals are a common and important class of crystalline materials. However, modelling molecular crystals based on first principles (eg. with density functional theory) is often difficult due to the size of a typical unit cell. Therefore, high-throughput calculations for the discovery of useful properties are rare. In this presentation, I will show how machine-learned interatomic potentials can enable accurate and fast calculations of mechanical and thermal properties of molecular crystals enabling an understanding of experimental observations as well as high-throughput search for materials with the desired properties [1,2,3]. In principle, to train machine learning potential one would need to create a sufficiently large database of molecular crystals calculated with the desired accuracy. This is also a very challenging task and we will show how to avoid this step using transfer learning and existing databases of small systems.

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Ab-initio study of magnetic properties of molecular magnets

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Based on first-principles calculations, we explored the nature and strength of the magnetic interactions in particular molecular magnetic systems, with annular shape, Cr_8 and V_8 , that are very promising for a variety of technological applications, i.e. in the quantum-computing research field [1, 2].

Calculations were performed within Hubbard-corrected non-collinear spin density functional theory (DFT) implemented in the Quantum ESPRESSO package. The systematic study consisted in the determination of the electronic structure, in the estimation of the exchange coupling parameter and in the analysis of the magnetic anisotropy, by comparing various spin arrangements and thorough comparison of their energies. The Cr_8 ring magnet, consistent with the existing literature, has an antiferromagnetic ground state (AFM) with numerically dominant first neighbor interactions and with contained anisotropy. The V_8 ring, on the other hand, is ferromagnetic (FM) with fairly important near-second exchange interactions and slightly more pronounced anisotropies.

We used a chain model to easily calculate the one-site (U) and inter-site Hubbard interaction (V) which would be difficult to access if evaluated by the annular systems. The more accurate description of the electron localization obtained in DFT+U+V calculations allows to improve the agreement between the calculated spin coupling constants and those extrapolated from experimental results [3] and thus to improve the predictivity of our ab-initio calculations also from the quantitative point of view.

This approach is particularly useful for clarifying the origin of Dzyaloshinskii-Moriya (DM) interactions, their relationship with the structure and symmetry of the considered molecule, and their effects on system behavior, especially in view of potential applications.

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Abstract template for 22nd International Workshop on Computational Physics and Materials Science: Total Energy and Force Methods

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An emerging class of technology based on spin-crossover (SCO), a mechanism long observed and understood macroscopically, exhibits high potential for sustainable sensing and storage applications like carbon capture and spintronic devices. Yet, without an accurate microscopic description of their adiabatic energetics and electronic structure properties, we are unlikely to design and/or discover new SCO materials that optimize and tailor functionality while simultaneously minimizing, if not eliminating, impracticalities.

Materials scientists and quantum chemists increasingly look to density functional theory (DFT) for this task, especially as (semi-)local density functional approximations (DFAs) expand their realms of achievable accuracy to include coarse-grained spin-based properties. In particular, fully first-principles DFT+U+J, equipped with *in situ* Hubbard U and Hund's J parameters to address self-interaction and static-correlation errors, has demonstrated its ability to restore correct band structures without increasing computation time. It is currently unknown whether Hubbard-like corrective functionals, specifically those incorporating the Hund's J, in spin-DFT can achieve accurate adiabatic energy differences.

Our recently established international collaboration executes a detailed study designed to answer this query, first by calculating and analyzing trends of the Hubbard U and Hund's J, determined via linear response for all valence subspaces in a series of four octahedrally-coordinated Fe(II) SCO molecules that span the ligand-field strength spectrum. We then methodically apply these parameters via a range of Hubbard functionals, both common, state-of-the-art, and experimental, in search of the simplest combination to yield reliable spin-state energetic properties with respect to those obtained by CASPT2/CC[1, 2] or DMC[3, 4] reference methods.

We discuss the potency of basis set projector functions on the values of the *in situ* Hubbard U and Hund's J parameters calculated via linear response in a variety of molecular subspaces. Following this, we examine the somewhat counterintuitive failure of the Hund's J in furthering DFT+U's already robust capacity to obtain accurate adiabatic energy differences. This investigation maps previously uncharted limitations of the method and precisely highlights the areas for improvement therein.

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Polaritons and electron scattering from plasmons and phonons in Van der Waals heterostructures within an ab-initio framework: the case of BN-capped graphene

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Plasmons and polar phonons are elementary electrodynamic excitations of matter. In two dimensions and at long wavelengths they act as polaritons. They also dictate the scattering of charged carriers. In Van der Waals heterostructures, excitations from different layers are coupled via long-range Coulomb interactions, which determine their dispersion and their scattering strength with electrons. Even when the excitations do not couple directly, they are still influenced by the screening from all layers, leading to complex dynamical interactions between electrons, plasmons, and polar phonons. We develop an efficient ab initio model to determine the electrodynamic excitations of the Van der Waals heterostructures, accompanied by a formalism to extract relevant spectroscopic and transport quantities. Notably, we obtain electronic scattering rates originated by the remote phonons of the heterostructure. We apply our developments to BN-capped graphene, in which polar phonons from BN couple to plasmons in graphene. We study the nature of the coupled excitations, their dispersion and their coupling to graphene's electrons. Regimes driven by either phonons or plasmons are identified, together with a truly hybrid regime corresponding to the plasmon-phonon-polariton at long wavelengths. Interestingly, the dynamical screening of the coupling between BN's LO phonons and graphene's electrons crosses over from inefficient to metal-like depending on the relative value of the phonons' frequency and the energetic onset of interband transitions. While the coupling is significant in general, the associated scattering of graphene's carriers is found to be negligible with respect to the particularly large one coming from intrinsic phonons in the context of electronic transport.

Evidence of ferroelectric features in low-density supercooled water from ab initio deep neural-network simulations

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A persistent query revolves around whether supercooled liquid water may experience a first-order transition, resulting in the coexistence of two distinct liquid phases with different densities, known as high-density liquid (HDL) and low-density liquid (LDL). Recently, molecular dynamics simulations performed using deep neural-network force fields trained to accurate quantum mechanical data have confirmed this scenario, providing clear evidence of the existence of a metastable critical point in the deeply supercooled region [1]. Building on these findings, we show that the resulting low-density phase displays a strong propensity towards spontaneous polarization [2]. In addition, our results suggest that the kinetic transition from the LDL phase involves a collective reorientational angular jumps of water molecules in the hydrogen-bond network that triggers disorder in the system before annihilating the polarization in the system and moving to the HDL phase. These new features, besides offering new insights into the molecular structure and relative stability of the two coexisting phases, holds the potential to unveil novel strategies for detecting and characterizing the long-sought-after transition.

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Materials Design and Discovery with Generative Deep Learning

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The design and discovery of new functional materials have traditionally relied on a trial-and-error approach, which is both time-consuming and costly. Advances in high-throughput computation, open material databases, and machine learning-based property prediction models have enabled rapid screening of thousands of materials to identify the most promising candidates [1]. However, these screening-based methods are fundamentally limited by the existing material databases and are not well-suited for efficiently directing the search towards materials with specific properties.

The ultimate challenge in materials research lies in inverse design, where materials are directly generated to meet desired property constraints. In this regard, deep learning generative models (GMs) hold significant promise. Since GMs learn the underlying probability distribution of materials, they are highly efficient at inverse designing entirely new materials.

We have developed a deep generative model, *MagGen* [2], based on a variational autoencoder (VAE), to inverse design stable magnetic materials. During training, the model is conditioned simultaneously by two target properties - formation energy and saturation magnetization. The property-embedded latent space of the model is analyzed using graph theory. Remarkably, about 96% of the generated materials are predicted to satisfy the target properties. This is a substantial improvement over approaches that do not condition the latent space of the model by target properties or do not account for the connectivity of parent materials from which new materials are generated.

This impressive feat is achieved using a straightforward real-space representation of materials that can be directly derived from material CIF files. Finally, model predictions are validated by density functional theory (DFT) calculations on a randomly chosen subset of materials. The performance of *MagGen* is comparable or superior to previously reported models, and is particularly applied in designing rare-earth-free permanent magnets with promising results. This approach represents a substantial step forward in the directed design of materials with targeted functionalities.

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AiiDA-trainspot: automated learning of neural-network interatomic potentials

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Neural-network interatomic potentials (NNIPs) have transformed atomistic simulations, particularly in molecular dynamics, by enabling near ab initio accuracy with reduced computational costs and improved scalability, provided a rich, diverse training dataset is available. Recent advancements have exploited the expressive power of equivariant frameworks to develop pre-trained foundational models, expanding NNIP applicability across varied chemical and structural environments. Despite these advances, crafting high-precision NNIPs remains a complex task, demanding specialized expertise in both machine learning and electronic structure calculations.

Here, we introduce an automated, open-source, and user-friendly workflow that streamlines the creation of accurate NNIPs, minimizing the need for manual intervention. Our approach integrates density-functional theory (DFT) with classical molecular dynamics to systematically explore the potential energy landscape using random distortions, strain, interfaces, neutral vacancies, and molecular dynamics trajectories at varied temperatures. Leveraging an active learning strategy, a committee of potentials is trained, with on-the-fly calibration of committee disagreement against true errors to ensure reliability. This entire process is orchestrated by the AiiDA materials informatics platform, supporting seamless execution and scaling to massive datasets.

Our results demonstrate the data efficiency and workflow automation of this tool, providing molecular dynamics practitioners across fields—from materials science to chemistry and biophysics—with accessible ab initio accuracy. This platform democratizes NNIP development, empowering users to achieve high-precision simulations with minimal human intervention.

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Impact of non-adiabatic effects on the temperature-dependent Kohn anomalies in metallic nanowires

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Kohn anomalies [1], which appear as dips in phonon dispersion, are particularly prominent in low-dimensional materials and represent a common scenario where the Born-Oppenheimer approximation fails, indicating departures from adiabatic electron-nuclei coupling [2, 3]. In this work, we investigate the impact of non-adiabatic effects on Kohn anomalies in one-dimensional metals. We develop a theoretical model to analyze how the renormalized phonon frequency is influenced by intrinsic physical properties of the system, such as bare phonon frequency, electron effective mass, and electron-phonon coupling strength. Additionally, we identify the conditions under which an instability may arise, leading to an imaginary phonon frequency. We then validate our model through first-principles studies of selected metallic atomic chains.

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Ab initio method for time-dependent linear response functions considering electron-phonon self-energy

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Cooperative phenomena in solid-state physics have gained significant attention due to their scientific and technological importance. This relevance was officially acknowledged in the Ginzburg Nobel Lecture in 2004, which emphasized the pursuit of high-temperature superconductivity as a key challenge for the 21st century. Recent technological advancements have enabled a more profound exploration of these phenomena, gathering instances where electronic cooperation is mediated by phonon interactions. However, this research faces experimental challenges that hinders sample characterization. Vibrational spectroscopy plays a key role in sample characterization, especially infrared reflectivity measurement in case of high-pressure environment where the diamond-anvil-cell apparatus is required [1].

However, in order to complete a sample characterization, the material signature found experimentally should be compared with an accurate theoretical prediction. Our work provides an ab initio DFPT framework that can accurately describe the linear response functions of materials with strong electron-phonon interactions by developing an ab initio method for time-dependent linear response functions and then empower it with a many body approach for considering the electron-phonon self-energy effects. We apply this method to the calculation of the optical properties of high-temperature superconductors H₃S, and we find that the electron-phonon self-energy plays a crucial role in determining the properties of these materials. Our results provide new insights into the role of electron-phonon interactions in high-temperature superconductors and pave the way for a more accurate theoretical description of these materials[2].

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Impact of dimensionality on the topological magnons in Mn_5Ge_3 from first principles

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Topological magnons, collective spin excitations with non-trivial topological properties often characterized by protected edge states and robust transport, hold significant potential for applications in spintronics and quantum information processing. Recently, despite the material's centrosymmetric nature, bulk Mn_5Ge_3 was identified as an unexpected host for topological magnons induced by the Dzyaloshinskii-Moriya interaction [1]. A distinctive feature of topological physics is the gap opening at the K-point, which can be closed upon the rotation of magnetization. In this study, we explore the robustness of this gap in reduced dimensions by investigating thin films of Mn_5Ge_3 and examining how the spin-wave spectrum is affected. Our findings clarify the potential applications of topological magnons within Mn_5Ge_3 for spintronics and magnonics.

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Dynamical screening of the Electron–Phonon interaction

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The electron–phonon interaction (EPI) is ubiquitous in several research fields. It enters in the definition, among others, of the phonon and electronic self-energies and plays a key role in a wealth of physics applications: the calculation of electronic corrections to the phonon states or of phonon corrections to the electronic levels, thermal transport, out-of-equilibrium dynamics and more.

Despite the crucial importance of the EPI its definition is still actively debated. This is due to the fact that the EPI is not a natural ingredient of an Hamiltonian treatment but, rather, is the result of a manipulation of the electron–nuclei interaction. More importantly the final form of the EPI is dynamically screened by the electronic fluctuations.

Most of the calculations performed up to now have used a statically screened EPI [4]. Still the accuracy of such approach is not clear. Indeed, if such an approach has been fueled by the historical use of model Hamiltonians [3] clear mathematical demonstrations are still lacking. Indeed some authors have proposed an approach based on Time–Dependent Density Functional Theory [1, 2] that, however, has been recently showed to be not theoretically valid [6]. Even in the Many–Body community there is still not an agreement as demonstrated by the recent works of Stefanucci et al.[7] and myself[5].

In this talk I will present a formal and accurate derivation of the dynamical screening of the EPI by using MBPT. I will discuss the limitations of the variational approaches introduced by Calandra et al. [1, 2] and also the subtle theoretical aspects raised by Stefanucci et al. in [7] and in my recent work[5]. I will, in particular, demonstrate that it is possible to derive an effective vertex correction function that can be easily defined using simple ingredients and used to amend calculations based on the statically screened approximation.

I will support the theoretical derivations by exact results of a three dimensional homogeneous electron gas and some preliminary result on MgB₂.

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Diffusion behavior of $\text{La}_2\text{NiO}_3\text{F}_2$ Ruddlesden-Popper perovskite

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Ruddlesden-Popper (R-P) perovskites represent an interesting crystalline framework to model and design materials with desirably high diffusion properties to be applied to energy storage solutions. The prototypical structure shows alternate stacking of perovskite and rocksalt sublattices and the stacking order can be tuned during synthesis to obtain different geometries and incorporate different transition-metal and rare-earth ions [1].

In our ongoing work we explore the diffusion dynamics of the $\text{La}_2\text{NiO}_3\text{F}_2$ R-P perovskite with classical Molecular Dynamics methods, based on the MACE Machine-Learned Interatomic Potential (MLIP) [2]. We calculate the migration barriers for fluoride (F^-) anions with the ab-initio Nudged Elastic Band (NEB) method [4]. The MACE architecture has been also applied to the calculation of the Pair Distribution Function (PDF) [3] for the material under investigation and showed qualitative agreement with the experimental data.

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A novel neural network potential to determine the melting temperature of crystals: the case study of CaO

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The thermodynamic properties of multicomponent liquids at high pressure and temperature (HP-HT) are of paramount importance to shed a light on the melting and crystallization phase relations in geological systems. At the required extreme conditions of high pressure and high temperature, though, experiments suffer from uncertainties of hundreds of K and rarely predict the ultimate nature of melting [1]. Ab initio molecular dynamics (AIMD) already gave useful insights on the structure-energy properties of solid and liquid phases up to very HP-HT conditions, but the calculated melting temperature (T_m) depends critically on the simulation protocol and the computational cost increases with the number of atoms to process.

In this work we focus on the melting curve of CaO, not only because is a key phase in material and geological sciences, but also because the thermodynamic properties of the liquid phase are still controversial due to the very high melting point of the crystal [1,4].

We employ a novel software package for the generation of neural network interatomic potentials called PANNA[9] (Properties from Artificial Neural Network Architecture) implemented with the most recent descriptor LATTE[10] (Local Atomic Tensors Trainable Expansion) to extract a neural network potential for this system, trained on a dataset of more than 11,000 different structures.

We simulate the melting process of CaO with classic molecular dynamics, employing the LAMMPS code [5]. We tested three different methods, which gave insights both on the melting temperature of the crystal and on the enthalpy of fusion (ΔH_f).

The two-phase solid-liquid coexistence method consists of running a solid-liquid interface at constant enthalpy [3,6]. The melting temperature is determined as the average equilibrium temperature.

The void-nucleated method exploits holes in the crystal structure to initiate the melting process, thus decreasing the unrealistic high melting point resulting from homogeneous heating simulations by introducing a defect [2].

Finally, to calculate the enthalpy of fusion, the temperature of the crystal is increased up until the solid melts, and then the temperature is lowered again until the liquid phase recrystallizes. The difference in energy between the liquid and the solid curves at the melting point represents the enthalpy of fusion, ΔH_f [2].

The values of T_m and ΔH_f obtained with these methods by the means of classic MD on CaO are found to be consistent with experimental data available in literature [7,8].

Employing the two-phase solid-liquid coexistence method, we also calculated the high-pressure melting curve of CaO up to 30 GPa, a result which represents a starting point for the construction of a completely theoretically predicted HP-HT phase diagram of the system.

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Study of the exfoliation of two-dimensional Cu-doped TiSe₂

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Materials with thicknesses ranging from a few nanometers to a single atomic layer present unprecedented opportunities to investigate matter properties restricted to the two-dimensional plane. One of the most studied two-dimensional materials are the so-called transition metal chalcogenides (TMD). Among these materials, titanium diselenide (TiSe₂) and its derivatives stand out due to their well-known physical properties. However, certain aspects of their behavior still pose challenges to current theoretical knowledge, leading to difficulties or delays in their application. Jurelo et al. [1] studied through density functional theory (DFT) calculations the effect of copper (Cu) intercalation on the structural, vibrational and electronic properties of TiSe₂, observing that it exhibits properties akin to those of a high-temperature superconductor. In general, the synthesis of two-dimensional TiSe₂ is performed by chemical vapor deposition (CVD), mechanical exfoliation, or liquid phase exfoliation. To overcome the complexity and cost of these processes Rosa et al. [2] developed a novel method that consists of synthesizing and isolating single 2D sheets from 3D Cu_xTiSe₂ crystals, through a solvothermal exfoliation process using hydrazine (N₂H₄) as a solvent. It was observed that N₂H₄ facilitates exfoliation while preserving the structure of Cu_xTiSe₂ [2].

With the goal of understanding how N₂H₄ influences the exfoliation of TiSe₂, we perform DFT simulations on TiSe₂, Cu_xTiSe₂ systems and their interaction with N₂H₄. The calculations were performed with the Quantum Espresso software package using the Perdew–Burke–Ernzerho (PBE) functionals within the conjugate gradient approximation (GGA) and with PAW Pseudopotentials. In addition to analyzing the electrical and structural properties at the atomic level, we perform nudged elastic band (NEB) calculations were performed to study the activation energy for the diffusion of Cu adsorbed on TiSe₂. Our results, consistent with those previously reported [1], reveal a favorable interaction between the N atoms of N₂H₄ and the Se atoms in TiSe₂. We also observed that the intercalation of N₂H₄ increases the separation between the TiSe₂ sheets, which decreases the van der Waals interaction between them and facilitates the exfoliation process in the 3D material. These effects are enhanced by the presence of copper in the Cu_xTiSe₂ structure, since N₂H₄ also interacts favorably with Cu atoms. NEB calculations provide crucial information on the dynamics and mobility of Cu atoms in the material, contributing to a deeper understanding of doping transport and diffusion mechanisms.

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A unified quantum framework for simulating electrons and ions: The self-consistent harmonic approximation on a neural network curved manifold

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The numerical solution of the many-body problem of interacting electrons and ions beyond the adiabatic approximation is a key challenge in condensed matter physics, chemistry, and materials science. Traditional methods to solve the multi-component quantum Hamiltonian tend to be specialized electrons or ions and can suffer from a methodological gap when applied to both electrons and ions simultaneously[1]. In addition, ionic techniques often limited as $T \rightarrow 0$ K, whereas electronic methods are designed for 0 K. Thus, efficient strategies that simultaneously address the thermal fluctuations of ions at ambient temperature without struggling to describe the electronic quantum state from first principles are missing.

This work extends the self-consistent harmonic approximation[2] for the ions to include also the electrons. The approach minimizes the total free energy by optimizing an *ansatz* density matrix, solving a fermionic self-consistent harmonic Hamiltonian on a curved manifold[3], which is parametrized through a neural network. We demonstrate that this approach, designed initially for a flat Cartesian space to treat quantum nuclei at finite temperatures, can efficiently tackle both the ground and excited state properties of electronic systems, thus paving the way to a unified quantum description for electrons and atomic nuclei. Importantly, this approach preserves an analytical expression for entropy, enabling the direct computation of free energies and phase diagrams of quantum materials.

We benchmark the numerical implementation in several prototypical cases, proving that it captures quantum tunneling, electron-ion cusps, excited states energies, and static electronic correlations in the dissociation of H_2 , where other mean-field approaches fail[4].

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Machine learning assisted canonical sampling

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Numerical modeling of hydrogen absorption in metal hydrides. One of the main challenges to fully utilize hydrogen as a green and renewable energy vector is its storage. We study the absorption of hydrogen in ternary compounds of type M-Mg-Ni with a combination of ab initio molecular dynamics and classical molecular dynamics using machine learning interatomic potentials (MLIP). Our goal is to accurately predict the enthalpy of absorption, the desorption temperature and the entropy of absorption. We employ the newly developed Machine Learning Assisted Canonical Sampling (MLACS) method to generate on-the-fly interatomic potentials throughout the molecular dynamics simulation.

Viscous current-induced forces on a diatomic impurity in electron liquid

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Electronic viscosity, manifested within time-dependent density functional theory (TDDFT) by the dynamic exchange-correlation (xc) kernel $f_{xc}(\mathbf{r}, \mathbf{r}', \omega)$ [1], is known to contribute, alongside with the single particle scattering, to the stopping power of metals for atomic projectiles [2]. We have recently addressed the problem of the composite impurity motion in a viscous electron liquid [3]. An approach based on linear response TDDFT combined with Ehrenfest dynamics leads to a system of linear algebraic equations, which account for the competing effects of the current-induced force and the electronic friction. We find the coupling between the center of mass motion and that of the nuclei relative to each other, a feature due to the mediation of the two-body interaction by the environment. Starting from the ground state at the equilibrium inter-nuclear distance and applying a current pulse, we observe three phases of the motion: (I) acceleration due to the prevalence of the current-induced force, (II) stabilization upon balancing of the two forces, and (III) deceleration due to the friction after the end of the pulse. At lower, but still metallic, electron densities, the dynamic xc (viscous) contribution to the force significantly affects the acceleration (deceleration) in the first (third) phase of the process. For the Cs density ($r_s \approx 6$ a.u.), this correction amounts to about 40% in the rotational regime. Fundamental implications for electromigration theory are envisaged.

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Multiscale QM/classical models based on the GW approximation coupled with the Fluctuating Charges force field

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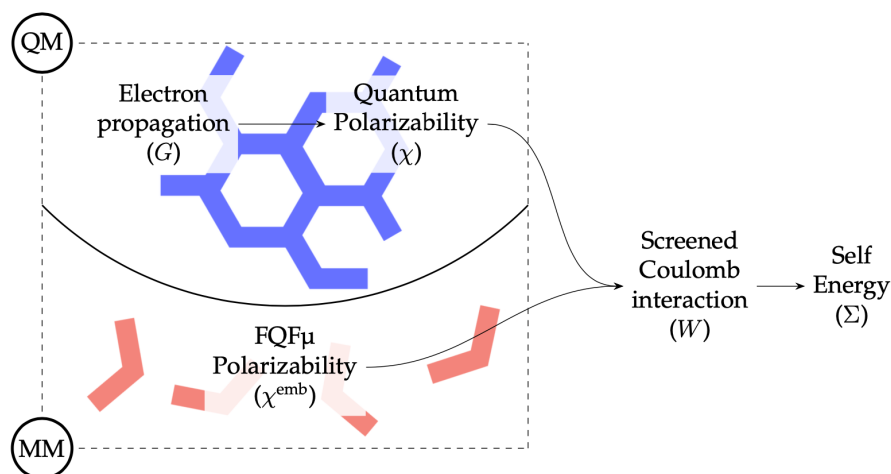


Figure 1: The system is composed of two moieties: the red moiety is treated classically (FQF μ force field), while the blue moiety is treated using the GW approximation. Each component provides a distinct contribution to the GW observables. The combination of all these quantities is necessary to evaluate the total self-energy.

In the context of complex molecular systems, multiscale approaches can recover correct descriptions of the quantum features of the system while remaining computationally feasible.

We propose a novel multiscale QM/classical methodology based on the GW approximation [1, 2], combined with the fluctuating charges (FQ) and fluctuating charges and dipoles (FQF μ) force fields, which have amply been employed in the context of computational spectroscopy of solvated systems [3]. The GW approximation is exploited to capture electron correlation effects, while FQ or FQF μ are used to model the mutual polarization effects between the quantum GW system and its surrounding environment.

The model is validated through test calculations of ionization potentials of selected molecular systems in solution.

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Equivariant Neural Network Potential for Zeolite Topologies

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In our previous work, we developed an invariant neural network potential for pure zeolites via active learning, resulting in a dataset of 16,552 structures from ten silica polymorphs. Using this dataset, we built a model based on an equivariant message-passing neural network architecture. This new potential significantly reduced training errors for both energies and forces. A validation test against properties computed with the invariant-based architecture, including structural, mechanical, thermovibrational properties, phonon dispersions, and density of states, demonstrated considerable improvements in accuracy. These results suggest that a low-cost, invariant architecture can be effectively used to generate high-quality data, which can, in turn, produce more accurate models using more sophisticated and computationally expensive equivariant architectures.

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The Tilted Plane Condition on the exact total energy landscape

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The flat plane condition [1] defines the profile of the ground-state energy landscape with respect to the electron population N and certain restricted values of the spin-magnetisation M . The breach of this exact condition is closely related to the poor performance of standard XC approximations in predicting band gaps, molecular dissociation, and polarizability. We have derived the tilted plane condition [2], a generalisation of the flat plane condition to the exact ground-state energy landscape for *all* values of N and M . Examples of exact energy landscapes, including tilted planes, are shown in Fig. (1). The magnetic analog of the DFT Koopmans' theorem is derived, and the magnetic piecewise linearity error (MPLE) is introduced.

We demonstrate how the tilted-plane condition reveals a new feature of the exact functional, namely derivative discontinuities at non-integer electron counts. It also helps us better understand approximate functionals, where we have identified large errors that go beyond quadratic order in the spin-magnetisation M . From the tilted-plane condition, we show that energy functionals of occupancies must take different forms depending on symmetry-imposed degeneracies. We may also highlight our recent proof of the long-assumed convexity condition of DFT [3].

We will demonstrate the performance our recently developed DFT+U type corrective functional BLOR [4], as well as its multi-orbital generalisation mBLOR [5]. These are derived from, and uniquely defined by, imposition of the tilted plane condition. They are fully detached from the Hubbard model and thereby free of double-counting approximations.

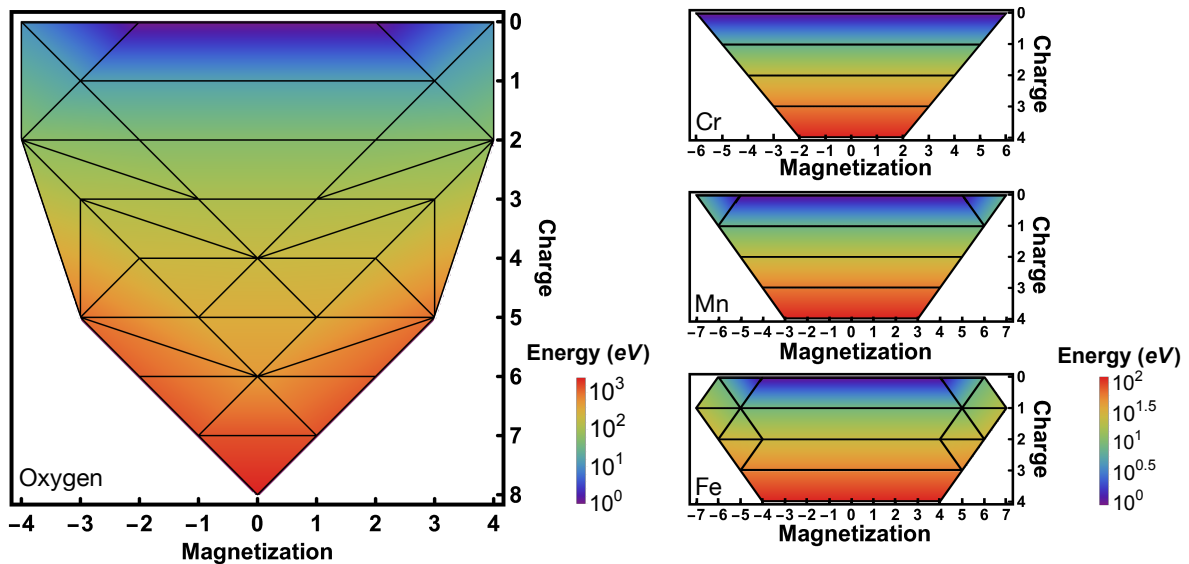


Figure 1: The projection of the $E_v[N, M]$ surface of the oxygen, chromium, manganese and iron atoms onto the $N - M$ plane. Energy values at vertices are based on spectroscopic data.

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Ionic Solutes Slow Down the Dynamics of Solvent Water Molecules: Effects on the Dielectric Response

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Water inherently contains trace amounts of various salts, yet the microscopic mechanisms by which salts influence some of its physical properties remain elusive. Notably, the mechanisms that reduce the dielectric constant of water upon salt addition are still debated. Also, the shift of the primary absorption peak for electromagnetic radiation – commonly used in microwave heating – towards higher frequencies in saline solutions suggests faster water molecular dynamics. This observation, however, contrasts with the simultaneous increase in viscosity and experimental reports that ionic solutes would slow down water molecular motion. In this work, we employ deep neural networks trained to reproduce the interatomic forces and molecular dipoles to compute the dielectric spectra of perchlorate water saline solution, possibly relevant to the recent discovery of liquid water beneath a thick ice crust at the Martian south pole. Our results reveal that both the reduction in the dielectric constant and the absorption peak shift can be attributed to ion-induced changes in the orientational ordering of water molecules. Additionally, we demonstrate that analyzing the self-part of the dipole-dipole correlation function reveals clear signatures of the slowing dynamics within the first cationic solvation shell, as reflected in the dielectric spectrum and consistent with the experimentally observed increase in viscosity.

Automated prediction of Fermi surfaces from first principles

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The Fermi surface of a metal separates occupied from unoccupied electronic states. The electrons in its immediate vicinity are subject to low energy excitations and determine the electronic properties at low temperatures. Knowing the shape of the Fermi surface is crucial to understand various phenomena from Pauli paramagnetic susceptibility to non-conventional superconductivity. Accurate Fermi surface simulations require a very dense sampling of the Brillouin zone, thus direct density functional theory calculations are limited by their computational cost. To obtain a detailed simulation of the Fermi surface we therefore use interpolation from the basis set of maximally localized Wannier functions (MLWFs). Thanks to their spatial localization, they provide a way to efficiently compute accurate Fermi surfaces on grids with k -point distances of a few hundredths of an inverse angstrom corresponding to hundreds of thousands of k -points for small unit cells. Taking advantage of the recently introduced robust algorithm for the automated Wannierisation (projectability disentangled Wannier functions, PDWFs) [1] and of the AiiDA workflow infrastructure for automation [2], we have computed the Fermi surfaces of more than 7'500 inorganic metals. We have also computed de Haas-van Alphen oscillation frequencies that allow for direct comparison with the experimental measurements. The results will be published openly on the Materials Cloud 3D crystals database (MC3D) [3] to provide a reference for the experimentalists and facilitate materials discovery.

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Spectral properties obtained from an efficient multipole representation of the GW self-energy

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We propose an efficient analytical representation of the frequency-dependent GW self-energy via a multipole approximation (MPA- Σ). Similar to the earlier developed multipole approach for the screening interaction W (MPA- W) [1, 2], the multipole-Padé model for the self-energy is interpolated from a small set of values evaluated numerically in the complex frequency plane. As for MPA- W , we show that a good choice of frequency samplings increases computational efficiency and results in a very accurate description of the self-energy. Crucially, MPA- Σ enables a multipole representation for the interacting Green's function G (MPA- G), providing straightforward evaluation of all the spectral properties. Combining the MPA- W and MPA- Σ schemes considerably reduces the cost of the self-energy calculations, especially when targeting spectral band structures. We validate the MPA- Σ approach in bulk Si, Na and Cu, monolayer MoS₂, and the NaCl and F₂ molecules, as prototypical semiconducting and metallic materials of different dimensionality. We also use toy models of the MPA- Σ , and their corresponding MPA- G solutions, to discuss the quasiparticle picture and the renormalization factor in the description of the spectral weights.

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Insulating and metallic phases in the 1D Hubbard-Su-Schrieffer-Heeger model: Insights from a backflow-inspired variational wave function

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The Su–Schrieffer–Heeger model[1] was introduced to describe how electrons deal with lattice distortions; by modeling phonons as quantum harmonic oscillators located on each lattice site and considering electron hopping modulated by the phonon displacements, interesting physics can appear. Specifically, for interacting electrons at half-filling, there is a competition between the Peierls insulator (with finite lattice distortion) and an undistorted Mott insulating phase. The behavior of such a system in the doped regime is still an open problem.

We introduce a new variational wave function[2] for electron-phonon systems. The fermionic part of the variational state depends on the phonon configuration in a backflow-inspired way, thus going beyond simple Jastrow-Slater approaches. We show that this wave function can capture correctly the physical properties of the system at half-filling, showing a transition between Peierls and Mott insulators. In this respect, we also benchmark our results against density matrix renormalization group calculations. Furthermore, studying the hole-doped regime, we report the presence of a conventional Luttinger liquid (gapless in all excitation channels) upon doping the Mott insulating phase; instead, when lightly doping the Peierls insulator, we observe that the resulting metallic phase retains a finite spin gap (Luther Emery liquid[3]).

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Stability of second-order screened-exchange solutions from the algorithmic-inversion method and sum-over-poles representations

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and Andrea Ferretti

In this work we mainly deal with the SOSEX correlation self-energy, in comparison with the HF, GW, and 2B approximations. To do so, we take advantage of the in-house AGWX code, which implements many-body methods on a lattice, where we include all the required extensions to deal with the SOSEX self-energy. Specifically, we exploit a sum-over-poles (SOP) [1-3] representation of propagators, including Green's functions, polarizabilities, and self-energies. This approach is numerically stable and allows for an accurate treatment of dynamical operators, in turn enabling the possibility to perform self-consistent Green's function simulations.

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T. Chiarotti, N. Marzari, and A. Ferretti, Phys. Rev. Research 4, 013242 (2022)

First-principles mobilities with hybrid and Koopmans functionals

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Electron-phonon interactions are crucial to understanding many properties of materials, such as electron and hole mobilities [1]. Accurate modelling of these interactions [2] can lead to innovative applications, going from efficient electronic devices to novel superconducting materials. However, the limitations of traditional DFT in predicting the electronic structure and charged excitations/band structures make it compelling to explore more accurate approaches, such as hybrid or Koopmans functionals [3]. This work focuses on the development of a general framework for calculating electron-phonon matrix elements on a coarse grid with beyond-DFT functionals, requiring only the eigenvalues and eigenfunctions of the relevant Hamiltonians. Performing later Wannier-Fourier interpolation to the dense grid allows us to study transport properties using the Boltzmann transport equation. We demonstrated our approach using hybrid or Koopmans functionals as case studies, showing the effect of these methods on electron-phonon couplings and mobilities for common semiconductors.

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Construction of Machine Learning Interatomic Potential for Twisted Bilayer WSe₂

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Twisted bilayer systems recently attracted significant interest for their unique electronic and structural properties, such as the emergence of unconventional superconductivity [1,2]. Twisted bilayer WSe₂ is one of model systems for exploring electronic properties influenced by Moiré superlattices, which arise from small twist angles between layers. Recent experimental result on twisted bilayer WSe₂ shows the emergence of ultra flat band induced by lattice reconstruction [3]. To understand the properties from its intrinsic part, it is important to simulate the twisted bilayer WSe₂ atomic structures. However, the computational costs are expensive for simulating such large supercell systems. In this study, we constructed machine learning interatomic potential (MLIP) for twisted bilayer WSe₂ based on ab-initio molecular dynamics calculation in OpenMX package [4]. This machine learning approach could enhance the computational efficiency for larger system sizes. The constructed MLIP enables the optimization of the atomic configurations of bilayer WSe₂ for various twist angles, which is crucial for understanding their electronic properties. By reproducing the energy landscape of the twisted bilayer WSe₂, our model may provide the insights to support the investigation of the quantum phenomena and device application driven by machine learning for studying Moiré structures at computational scale.

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Broken symmetry solutions in one-dimensional lattice models via many-body perturbation theory

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In this work we study self-consistent solutions in one-dimensional lattice models obtained via many-body perturbation theory. The Dyson equation is solved in a fully self-consistent manner via the algorithmic inversion method based on the sum-over-pole representation (AIM-SOP) of dynamical operators [1, 2, 3]. We start by validating our self-consistent AIM-SOP implementation by taking as a test case the one-dimensional Hubbard model. We then move to the study of antiferromagnetic and charge density wave solutions in one-dimensional lattice models, taking into account a long-range Coulomb interaction between the electrons. Complementary, by solving the Sham-Schlüter equation, we can compute the non-interacting Green's function reproducing the same charge density of the interacting system. In turn, this allows for the evaluation of the derivative discontinuity in the Kohn-Sham potential, which gives a measure of how the Kohn-Sham gap approximates the many-body fundamental gap of these systems.

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Electric control of spin and valley polarization in two-dimensional altermagnets

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Altermagnets are a special class of antiferromagnets where a zero net magnetization is enforced by symmetries that relate spin-up and spin-down sublattices but that, at the same time, allow for a finite spin splitting between energy bands, even in the absence of spin-orbit coupling. Despite this spin splitting, the presence of sublattice-relating symmetries requires band extrema –and thus valleys– in semiconducting altermagnets to be spin degenerate, although they might occur at different locations in the Brillouin zone for opposite spins. An external electric field can break these symmetries and thus give rise to a finite and controllable spin (and valley) polarization. This is particularly promising in two-dimensional (2D) materials where it is easy to apply a vertical electric field in a double-gate field-effect setup, provided that 2D altermagnets with suitable crystal symmetries are found. Here, by using first-principles simulations, not only we put forward an interesting family of 2D altermagnets that display the correct symmetries, but we also show the electric field effect is sizable in these materials and reaches the requirements needed for applications in spin-valleytronics.

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Thermodynamic stability of the materials in the Materials Cloud three-dimensional crystals database (MC3D)

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Thermodynamic stability of materials, calculated using DFT, is one of the most applied properties for estimating synthesizability of materials and guiding computational materials discovery studies. However, deviations between DFT-predicted and experimentally measured thermodynamic stability arise, especially at the GGA level of DFT. In this work, we present and analyze the thermodynamic stability of the materials in the MC3D [1], a DFT optimized and curated structural database of inorganic crystals, by calculating the formation energies and inspecting their energies above the convex hull. All calculations are managed and driven by the AiiDA [2, 3] workflow engine, allowing to browse the full provenance graph and to share the results in the Materials Cloud [4]. In this way, we can identify stable compounds that are expected to be more likely to exist and be synthesizable under experimental conditions. To improve the agreement between the theoretical and experimental thermodynamic stability, we apply empirical [5] and machine-learning [6] based corrections, and improve upon them, discussing the agreement with experimental data on stability.

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Nonempirical hybrid functional based on metaGGA

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Semi-local density functionals such as PBE typically underestimate experimental band gaps by 50%. Hybrid functionals address this “band gap problem” by admixing a fraction of exact exchange to semi-local exchange. The optimal mixing parameter depends on the specific material and can be identified as the inverse dielectric constant [1]. Here, we show that dielectric constants obtained using the r^2 SCAN metaGGA functional [2] are significantly more accurate than dielectric constants obtained using PBE [3]. This can be understood through the improved treatment of electronic self-interaction within the metaGGA framework [4, 5]. Further, a dielectric-dependent hybrid functional based on r^2 SCAN can outperform the standard PBE based hybrid in terms of band gaps [3]. Particularly impressive improvements are obtained for narrow gap semiconductors such as Ge and InAs, where PBE wrongly predicts a metallic phase, but r^2 SCAN can open a gap.

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Quantum embedding for strong correlation in defects and materials

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To address strong electron correlation, novel quantum embedding schemes are needed for studying both defects and materials. In this approach we combine density functional theory (DFT) with a high-level wave function method to capture correlation within the defect. By partitioning the system into two subsystems, the embedding formalism treats the extended system using DFT, while the local correlated subspace of the defect is treated with wave function-based methods such as FCI or CASSCF. This hybrid (embedding) approach allows for an accurate treatment of multi-reference effects and static correlations that are challenging for DFT. The method relies on a Green's function formulation of embedding, and it is based on the algorithmic inversion method (AIM) for solving Dyson like equations [1]. This quantum embedding scheme has the potential to significantly improve the precision and the accuracy of calculations for systems with defects, impurities, and can also be extended to strongly correlated (periodic) crystals.

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Study of the mechanism of fluorine-induced magnetism in graphene

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Different types of adsorbed atoms can induce magnetism on graphene. Among all the atoms studied, the magnetism induced by light atoms such as H, F and C on graphene has received great attention because it does not involve d or f orbitals. In the case of an isolated fluorine atom on graphene, the experimental results obtained so far are not sufficient to clarify the induced magnetism [1,2,3]. In this work, we carried out a comprehensive study using Density Functional Theory (DFT) with different functionals (HSE, PBEsol and ACBN0) and also DFT + U + V calculations to investigate the adsorption and magnetic properties of isolated fluorine atoms on graphene. Our results reveal that the gradient corrected functionals erroneously predict a non-magnetic ground state, while more accurate calculations such as hybrid functionals and DFT + U + V calculations predict a magnetic moment of 1 Bohr magneton per cell for the graphene system with adsorbed fluorine. We then develop tight-binding models based on our DFT results, using Wannier functions, that accurately reproduce the observed electronic structure and magnetic order. This allows us to elucidate the physical origins and identify the key factors influencing the magnetic behavior of these covalent systems, paving the way for a deeper understanding of the system and providing a computationally efficient tool to explore the electronic structure and magnetic behavior of the system. Our work demonstrates the crucial importance of an accurate treatment of electronic correlations to capture the magnetic ground state in these systems, contributing to the understanding of light atom-induced magnetism in graphene.

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Anisotropic Lattice Thermal Expansion: First-Principles Methodology for Various Crystallographic Symmetries

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The Quasi Harmonic Approximation (QHA) is a widely used method for calculating the temperature dependence of lattice parameters and the thermal expansion coefficients. However, applying QHA to anisotropic systems typically requires several dozens or even hundreds of phonon band structure calculations, leading to high computational costs. While the ν -ZSISA-QHA method effectively addresses volumetric thermal expansion [1, 2], challenges remain in accurately capturing anisotropic thermal expansion for certain lattice vector components. In this work, we present an efficient implementation of the Zero Static Internal Stress Approximation (ZSISA) within QHA [3, 4], based on the thermal stress concept, enabling its application across a wide range of crystal structures under varying temperature and pressure conditions. By incorporating second-order derivatives of vibrational free energy with respect to lattice degrees of freedom, we significantly reduce the number of required phonon band structure calculations. For hexagonal, trigonal, and tetragonal systems, only six phonon band structure calculations are needed, while 10, 15, and 28 calculations suffice for orthorhombic, monoclinic, and triclinic systems, respectively. This method is tested for a variety of non-cubic materials, from uniaxial ones like ZnO and CaCO₃ to low-symmetry monoclinic or triclinic materials such as ZrO₂, HfO₂, and Al₂SiO₅, demonstrating a significant reduction in computational effort while maintaining accuracy in modeling anisotropic thermal expansion.

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Electromagnons in CrI₃ from nonadiabatic DFPT at constrained magnetic moments

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The terahertz dynamics of CrI₃, a prototypical van der Waals insulating ferromagnet stable down to the monolayer limit, has garnered significant interest due to the potential for externally manipulating spin-wave excitations (magnons) [1, 2], thus opening avenues for low-power computing technologies. The rich magnonic behavior in CrI₃ stems from a complex interplay between spin, lattice, and electronic degrees of freedom, which remains challenging to fully understand at the microscopic level [3, 4]. In this talk, I will present a first-principles investigation of these interactions using a nonadiabatic extension of the density-functional perturbation theory (DFPT) formulated at constrained magnetic moments. By computing frequency-dependent susceptibilities (magnetic, dielectric, and magnetoelectric) with and without spin and ionic relaxation effects, we uncover a key coupling between an optical electromagnon and a nearby polar phonon mode. This coupling leads to a pronounced lattice-mediated local magnetoelectric response and the emergence of electromagnon signatures in the dielectric spectrum, including notable magnetic circular dichroism.

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Electronic transport properties of NbFeSb: an efficient ab-initio approach

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In this study, we investigate how the electronic transport properties of one of the high thermoelectric performance half-Heusler (HH) alloy, NbFeSb, with a p-type TE figure of merit $ZT > 1$, is affected by various scattering mechanisms. We perform a full, detailed and highly accurate investigation of its electronic and thermoelectric power factor properties. We employ our BTE code ElecTra, which takes into account the full energy/momentum/band dependence of the electronic structure and the electron scattering rates [1]. We account for all relevant scattering processes, i.e. with acoustic phonons, non-polar optical phonons (intra- and inter-valley), polar optical phonons (POP), and ionized impurity scattering (IIS). For acoustic and non-polar optical phonon scattering, we extract the acoustic and optical deformation potential values from first-principles calculations [2]. We extract the dielectric constants from first-principles calculations and use those within the Fröhlich formalism to evaluate the POP scattering rates, including the effect of screening. We use the Brooks Herring model for IIS. We show that in this material, like many other HHs, the dominant scattering mechanisms are polar optical phonon scattering and ionized impurity scattering. We also show that screening considerations in the calculation are important and need to be accounted for, despite substantially increasing the computational cost [3]. Our work suggests that the computationally expensive non-polar phonon scattering part (acoustic and non-polar optical) is less important, and focus should be directed towards polar optical phonons and ionized impurity scattering when studying these type of materials. Finally, although we use NbFeSb as an example, the method we employ is generic and can be applied efficiently and accurately for thermoelectric materials in general.

Acknowledgments

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Excess Energy and Countercurrents After a Quantum Kick

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Motivated by the study of non-equilibrium quantum systems, we present some formal results and their implications for systems experiencing a sudden onset of motion. We consider a system of interacting quantum particles under a static external potential, which is described as "kicked" when this potential suddenly begins to move at a constant velocity, \mathbf{v} . This scenario is experimentally relevant to the study of electrons in an atom whose nucleus recoils after being hit by a neutron or dark matter particle, known as Migdal's sudden jolt [1], a well-known effect in particle detection. Here, we present a generalization that serves as a useful reference for time-dependent simulations and could inspire experiments on optical lattices, where defined quantum kicks could be realized. If the system is initially in its ground state, the excess energy at any time after the kick is given by $\mathbf{v} \cdot \langle \mathbf{P} \rangle(t)$, where \mathbf{P} is the total momentum of the system. For a system of mass M , if it remains bound, the long-time average of the excess energy approaches $M \cdot \mathbf{v}^2$, which is twice the excess energy it would have if the motion onset were infinitely smooth. A related expression can be derived for cases involving particle emission. In a macroscopic solid, an electronic current can arise that opposes the motion of the potential and persists on a longer timescale than other thermalization processes. In non-metallic systems, there may exist a threshold velocity below which transient currents persist only on very short time scales. Our results are derived from real-time time-dependent density functional theory calculations implemented in the SIESTA code [2], [3].

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Stress fluctuations and adiabatic speed of sound in liquids: A simple way to estimate c_s from simulations

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Sound propagation and its different mechanisms in liquids and solids are of paramount importance for fundamental many-particle effects in condensed matter, as well as for technologies based on vibrational properties of crystals and disordered systems. One of the fundamental quantities in dynamics of the liquid state, the adiabatic speed of sound c_s , is difficult to predict from computer simulations, especially in simulations where the electronic structure is explicitly solved during the simulation, like the density functional theory-based molecular dynamics (DFTbMD).

Here we derive an expression for the instantaneous correlator of fluctuations of the longitudinal component of stress tensor, which contains c_s along with others quantities easy accessible via computer simulations [1]. We demonstrate the applicability and efficiency of the method using molecular dynamical simulations in the case of Lennard-Jones and soft-sphere simple fluids, Kr-Ar liquid mixture in a simulation with effective pair interactions as well as in liquid Sb, fluid Hg and molten NaCl with DFTbMD simulations. Recently we have extended the range of studies [2].

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Accurate treatment of metallic screening in many-body calculations from first principle

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Many-Body Perturbation Theory calculations in the GW approximation have proven to be a reliable scheme for the computation of quasiparticle (QP) band structures of materials¹. In metallic systems, QP corrections are generally smaller than in semiconductors, but can be required to properly describe spectroscopic properties², especially at low dimensionality.

GW is a computationally expensive method, especially in metals that require an accurate description in both frequency and k-space. Typically, the troublesome long-wavelength limit of intraband transitions is treated by adding a Drude term at the plasmon energy³. The determination of the plasmon energy within the Drude method is generally demanding and in addition may be too simplistic for certain metals and semimetals⁴.

Here, we discuss a more efficient method for performing QP computations in metals. The troublesome screened potential of metals is accurately evaluated through the use of a Monte Carlo integration combined with interpolation techniques^{5,6}. This method was first developed for 2D semiconductors, where it led to a dramatic speed-up of the otherwise slow k-point convergence⁷. We generalise its usage by developing proper interpolation techniques for 3D and 2D metals. In this way, it is possible to capture the effect of intraband transitions without the need for additional parameters and reproduce the QP band structure with reduced k-point grids.

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Causing and Curing Headaches with Quantum Monte Carlo: Accurate Forces for Machine-Learned Force Fields

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Quantum Monte Carlo (QMC) is a powerful method to calculate accurate energies and forces for molecular systems. In this work, we demonstrate how we can obtain accurate QMC forces for fluxional molecules, which are characterized by weak interactions difficult to capture at the density functional theory level. Using ethanol as prototypical case, we assess the excellent performance of our protocol against high-level coupled cluster calculations on a diverse set of representative configurations of the system at room temperature. Then, we train machine-learning force fields on the QMC forces and compare them to models trained on coupled cluster reference data, showing that a force field based on the diffusion Monte Carlo forces can faithfully reproduce coupled cluster power spectra in molecular dynamics simulations. Finally, we extend our method to treat larger molecules, demonstrating the ability of QMC to provides benchmarks when high-level coupled cluster calculations are no longer possible.

First-principles light-driven molecular dynamics through equivariant neural networks

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Recent experiments where materials are driven by coherent THz pulses have shown a rich phenomenology in solids, liquids, and molecules [1]. However, simulation techniques that can describe the nuclear dynamics of these processes for all material classes, without relying on dimensionality reduction, and going beyond perturbation theory, are very challenging. Here we propose an ab initio molecular dynamics (MD) method within the electric dipole approximation that allows a single machine learning model to describe the coupling at diverse field strengths and with time dependence. We train equivariant differentiable neural networks based on MACE [2, 3] to learn the potential energy surface and the dipole of both isolated and periodic systems. Atomic tensor derivatives w.r.t. the nuclear coordinates are obtained through automatic differentiation. We present applications of this machine-learning-assisted MD protocol on liquid water and the ferroelectric LiNbO₃. We show full-dimensional ab initio simulations of the thermal equilibrium dielectric properties of both the systems (even at different constant electric fields), the ferroelectric-paraelectric phase transition, and the excitation of vibrational modes at an ultra-fast time scale in LiNbO₃ and highlight the different nonlinearities of these dynamics.

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Convergence of body-orders in linear cluster expansions

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We study the convergence of a linear atomic cluster expansion (ACE) potential [1, 2] with respect to its basis functions, in terms of the effective two-body interactions of elemental Carbon systems. We build ACE potentials with descriptor sets truncated at body-orders $K = 2, 3, 4$ trained on pure dimers, or on large datasets of different diversities but without any dimers. Potentials trained on a more diverse dataset fare better in validation and result in a nontrivial dimer curve, but still very far from the theoretical two-body interaction calculated by DFT. Moreover, dimer curves between descriptor sets clipped at different K do not seem to converge to a universal function for a given dataset. We conclude that machine learning potentials employing linear cluster expansions optimize losses at low K but fail to generalize and converge properties described by two-body interactions.

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Strain-controlled shift current conductivity in the ferroelectrics α -phase of halide perovskite CsPbI₃

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Ferroelectric perovskite structure with spontaneous polarization and visible light absorption, are recently considered as promising candidates for the application of bulk photovoltaic effect (BPVE) devices based on the shift current mechanisms [1-4]. The α -phase of all-inorganic halide perovskite materials recently attracted huge interest due to their high conversion efficiency and active optical response in the cubic structure [2-4]. However, the cubic-phase is only stable at high temperatures representing the limited operation of halide perovskite in the photovoltaic devices [5]. Recent experimental study reported using quantum dots and biaxial strain on the substrate, can realize the CsPbI₃ thin films stable at room temperature [5-6]. Since the α -phase of halide perovskite is normally a paraelectric phase at high temperature [4-6], it would be interesting for investigating the strain effect in the halide perovskite structure in order to induce room-temperature ferroelectric phase [7]. In this study, we performed the first-principles density functional theory (DFT) calculation [8] of strain effect on the α -phase of cubic structure halide perovskite CsPbI₃. Our calculations found that using both compressive ($\frac{c}{a} > 1$) and tensile ($\frac{c}{a} < 1$) strain, the displacement of Pb-atom induces spontaneous electric polarization along out-of-plane and in-plane direction, respectively. Based on this strain-induced ferroelectric phase, we evaluated the BPVE by computing the shift current conductivity based on the localized Wannier function approach [8-10]. Our results suggested that the magnitude of shift current conductivity can be enhanced through the lattice distortion $\frac{c}{a}$ due to the strain-controlled of electric polarization. This shift current induced by strain effect in the halide perovskite might be expected to realize photovoltaic devices without the need of an external electric field. The present results may open the opportunity for the development of BPVE devices based on the strain-controlled shift current.

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Thermodynamic properties of fcc lead - a scalar and fully relativistic first principle study

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We present the ab-initio thermodynamic properties of fcc-lead studied within the quasi-harmonic approximation (QHA), where the influence of spin-orbit coupling (SOC) and the exchange-correlation functionals is investigated. The calculations were performed using the `Thermo_pw` [1] code, a driver of the Quantum ESPRESSO (QE) routines [2,3]. Both scalar (excludes SOC) (SR) and fully (includes SOC) relativistic (FR) ultrasoft pseudopotential (PP) are considered. Moreover, for each PP, we test the performance of three popular exchange-correlation functionals: Perdew-Burke-Ernzerhof generalized gradient approximation (PBE), PBE modified for dense solids (PBEsol), and local density approximation (LDA). The Helmholtz free energy includes the contribution of lattice vibrations (or phonons) and electronic excitations. From the Helmholtz free energy, the equation of state (at 4 K and 301 K), phonon dispersions (at 100 K and 300 K), mode-Grüneisen parameters ($\gamma_{q\eta}$) (at 100 K), volume thermal expansion coefficient (β), isobaric heat capacity (C_P), bulk modulus (B_T), and thermodynamic average Grüneisen parameter (γ), are determined. For phonon calculations, we use density functional perturbation theory (DFPT) [4] extended to ultrasoft PP [5], and the electronic excitations contribution (EEC) is included within the rigid bands' approximation.

Our study shows that the effect of SOC on the equilibrium parameter is more prominent at higher temperatures for the PBE functional. The difference in the lattice constant's value with and without SOC at 300 K is $\sim 0.3\%$ for LDA and PBEsol and a maximum of $\sim 0.7\%$ in PBE. The phonon dispersion at 100 K indicates that the introduction of SOC is essential to explain the softening in the T branch at **X**. This effect of SOC at **X** is also visible in the mode-Grüneisen parameter $\gamma_{q\eta}$, where the magnitude of $\gamma_{q\eta}$ is significantly higher for the FR case and almost double at **X**. At 300 K, we notice that including SOC substantially decreases the overall phonon frequencies with respect to the experiment and the scalar relativistic functional performs better, except for the T branch at **X**.

The thermodynamic average Grüneisen parameter with temperature increases when SOC is considered, but it remains almost constant for the SR case. We found the results for thermal expansion coefficient, isobaric heat capacity, and phonon dispersion are similar for LDA and PBEsol (for both SR and FR). With increasing pressure, the contributions of SOC in the thermodynamic properties decrease but do not vanish. The pressure-dependent elastic constant-coefficient and Pugh ratio at 0 K indicate the enhancement in the ductility of lead prevails with increasing pressure. This study [6] shows that the electronic excitation contribution in all cases is negligible, and introducing the SOC effect gives results that differ from the SR approach, which is not always in closer agreement with the experiment.

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High-throughput computational screening of fast Li-ion conductor

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We present a high-throughput computational screening to find fast lithium-ion conductors to identify promising candidate materials for application in all solid-state electrolytes (SSEs). Starting with more than 30,000 experimental structures sourced from COD, ICSD and MPDS repositories, we perform highly automated calculations using AiiDA at the level of density functional theory (DFT) to identify electronic insulators. On these ~1000 structures, we use molecular dynamics simulations to estimate Li-ion diffusivities using the pinball model, which describes the potential energy landscape of diffusing lithium at near DFT level accuracy while being 200-500 times faster [1]. Then we study the ~60 most promising unknown fast conductors with full first-principles molecular dynamics (FPMD) simulations at several temperatures to estimate their activation barriers. We discuss the results of FPMD in detail for the 12 fastest conductors at room temperature. We further present the entire screening protocol, including the workflows where the accuracy of the pinball model is improved self-consistently, necessary to automatically running the required calculations and analysing their results.

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Comparative study of magnetic exchange parameters and magnon dispersions in NiO and MnO using different first-principles methods

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Magnons play a crucial role in explaining the behavior of magnetic materials and can lead to key technological applications. However, it is challenging to accurately model these in transition-metal compounds using methods based on density-functional theory (DFT) with (semi-)local functionals, due to the large self-interaction errors for d electrons. Here, we conduct a comparative analysis of the exchange parameters and magnon dispersions in NiO and MnO using three first-principles approaches, all applied to the same DFT+ U ground state with Hubbard U computed from first principles using density-functional perturbation theory [1]. Two of these methods calculate the exchange parameters directly, one via total-energy differences and the other via the magnetic force theorem. From the parameterized Heisenberg Hamiltonian, we compute the magnon dispersions using linear spin-wave theory. The third approach is based on time-dependent density-functional theory with Hubbard correction [2] and probes directly the spin-spin susceptibility. This study contributes to the broader effort of evaluating the accuracy of first-principles methods for magnetic materials.

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Machine learning potentials at hybrids DFT accuracy unravel the transport mechanisms of solid-state electrolytes

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Batteries are a strategic technology for our energy-greedy society. Solid-state electrolytes (SSEs) are particularly attractive due to their advantages over current liquid electrolytes. Among SSEs, Li_3PS_4 shows excellent promise as a Li-ion conductor. However, despite numerous investigations into this material, a comprehensive study of its equilibrium and dynamic properties across all phases is still lacking. To address this gap, we propose to leverage powerful machine learning (ML) models to study Li_3PS_4 across all three known phases and at increasing levels of theory, including PBEsol, SCAN, and PBE0 functionals. We discuss the microscopic origin of the observed superionic behaviour of Li_3PS_4 : the activation of PS4 flipping drives a structural transition to a highly conductive phase, characterised by an increase in Li-site availability and by a drastic reduction in the activation energy of Li-ion diffusion. We showed how a ML at the PBE0 level of theory can obtain very accurate ionic conductivity values [1].

Studying thermal conductivity, we use the Green-Kubo theory to overcome the limitations of lattice methods, which are formally unsuitable for materials where the diffusion of ionic charge carriers generates a lack of equilibrium atomic positions needed for normal-mode expansion. We uncover the different behaviour of the diffuse phases (a glass-like behaviour due to the effective local disorder characterising) and of the γ phase (a low-temperature crystal-like behaviour, typical of heat transport mediated by phonon propagation) [2].

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Ab initio study of point defects in Pu

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At ambient pressure, plutonium exhibits six stable allotropes across various temperature ranges [1]. The thermophysical properties of Pu vary significantly depending on its crystal phase. At room temperature, Pu predominantly exists in its α phase (monoclinic, brittle), yet it can transform to its δ phase (face-centered cubic, ductile) at higher temperatures. Natural self-irradiation in Pu leads to substantial changes in these properties over time; within approximately 10 years, every atom in a Pu sample is displaced from its initial position.

To investigate diffusion mechanisms in δ -Pu, we employed ab initio calculations. This study presents a unified model for δ -Pu that accounts for explicit anharmonic temperature effects [2] as well as the specific structure of point defects at these temperatures. Enabled by machine learning techniques within the MLACS (Machine Learning Assisted Canonical Sampling) framework [3], our approach replaces costly ab initio calculations with a machine learning force field. This optimized potential allows for the efficient exploration of long timescales and rare events.

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How to Obtain High-Quality Wannier Functions Using Topological Quantum Chemistry

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Wannier functions (WFs) have become a popular and powerful technique for studying versatile electronic properties of materials [1]. The dominating code for construction of WFs nowadays is Wannier90 [2], and a rich ecosystem of codes has grown around it [3], one of them being my code WannierBerri [4]. However, the construction of high-quality WFs is still not a straightforward task, often requiring manual intervention and expertise. The crucial part in construction of WFs is the selection of initial projections, which are used as initial guess for the WFs. Traditionally these projections are chosen manually, based on the knowledge of the orbital character of the bands under study (within a frozen window), which requires visual inspection of the band structure. Alternative approaches have been developed recently, such as the Selective Columns of the Density Matrix (SCDM) method [5].

In this contribution I present an approach based on the symmetry indicators of DFT Bloch bands. For that I compute all the irreducible representations (IRs) of the Bloch bands at all points of the *ab initio* grid using IrRep code [6]. Further, for all Wyckoff positions of the crystal I compute the IRs that are generated by placing atomic orbitals on this position. In the terminology of Topological Quantum Chemistry [7] this is called Elementary Band Representations (EBRs). Further, the algorithm searches for such combinations of EBRs that can represent the IRs of the DFT bands, providing a list of possible initial projections. By construction, those projections are compatible with the symmetry of the band structure, and therefore are suitable for construction of symmetry-adapted WFs (SAWF) [8].

It is known that the current implementation of SAWFs in the Wannier90 code has a few limitations which make it unsuitable for many applications. Namely, the code does not support (i) frozen window, (ii) spin-orbit coupling (SOC), (iii) time-reversal and (iv) magnetic symmetries and finally (v) it is compatible only with Quantum Espresso code [9]. Limitations (i)-(ii) have been recently addressed in the SymWannier code [10]. To overcome all the 5 problems, I have implemented the construction of symmetry-adapted Wannier functions within the WannierBerri code, making it compatible with SOC, frozen window, and with multiple DFT codes (VASP, Quantum Espresso, Abinit, etc.) via the interface of IrRep code. In addition, the code supports time-reversal symmetry and magnetic symmetries, which is crucial for the study of (anti-)ferromagnets.

The methods do not require manual inspection of the band structure, and therefore are suitable for high-throughput calculations. The code is open-source and available at [11].

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HubbardML: Self-consistent Hubbard corrections at no cost

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Hubbard corrections have been shown to significantly improve the accuracy of density-functional theory calculations for many strongly self-interacting systems, covering many materials containing transition-metals or rare-earth elements. Using linear-response, the calculation of Hubbard parameters can be integrated into a self-consistent procedure that renders the DFT calculations entirely parameter free, a particularly useful property for screening and materials discovery. However, this typically comes at a computational cost of 80-100 times that of a single DFT calculation. In this work, I present an equivariant machine learning model that can entirely replace linear-response calculations by learning to predict self-consistent Hubbard U and V (Uhrin et al., 2024). As inputs to our neural network, we use the full on-site occupation matrices, which represent the local charge density around each atom. I will discuss how equivariant neural networks make it trivial to express and perform learning on representations of the electronic structure by exploiting concepts from group theory. The resulting model achieves an accuracy of 3% for U and 5% for V parameters across a range of atomic species with very little training data and demonstrates good transferability even when extrapolating to unseen oxidation states. These results open the door to wide-spread use of self-consistent Hubbard parameters, eliminating both the need for domain expertise and access to significant amounts of compute resources.

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New theoretical method to design quasi-atomic systems in the band gap of semiconductors by combining density functional theory (DFT) and the Hubbard effective Hamiltonian: Applications to α boron and to the nitrogen-vacancy center in diamond

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Crystal point defects offer one possible pathway towards the solid-state implementation of some quantum applications. Conditions for such applications are the existence of a **quasi-atomic system** (QAS) in the forbidden band gap, defined as a set of localized in-gap energy levels with the following criteria: (i) the presence of energy levels well localized in the band gap; (ii) some of which are degenerate; and (iii) the capability of occupying the in-gap levels with a number of electrons proper to generate a high-spin state (e.g., a triplet ground state as for the NV⁻ center). Important additional criteria are the possibility to manipulate the spin state by optical excitations and the possibility to control spin selectivity via shelving states.

In the present work, we combine the calculation of total energy in DFT-HSE06 with constrained occupations of the in-gap energy levels, with an in-house Hubbard model fit on the total energy values to describe the many-body energy states of the negatively charged nitrogen-vacancy (NV) center in diamond. We show the need to extend the Hubbard model to spin-spin interactions for the NV center.

We then propose a new theoretical methodology aimed to design QASs similar to the NV center. We introduce the four concepts of primary defect; under-hybridized interstitial impurity; multiple combinations of n primary defects referred to as n -wise combination; and thermodynamic charging. Our methodological stages consist in: (1) choosing the primary defect among simple point defects; (2) establishing a link between the primary defect energy in the reciprocal space and the geometry of n -wise combinations of primary defects in real space with a tight-binding model, thereby positioning defect levels in a way that some of the defect energies are degenerate; (3) thermodynamically charging the defect with an electric charge via modifications of the chemical environment; and (4) obtaining the final sequences of the many-body energy level of promising QASs selected from stage (3), using the effective Hubbard model fit with data from DFT with constrained occupations, to account for in-gap electronic correlations. The effectiveness of the methodology is demonstrated by an application to carbon-based defects in alpha (α) rhombohedral (trigonal) boron.

Calculations have been performed with the Quantum ESPRESSO software and access to HPC resources granted by the IDRIS, CINES, TGCC national centers (GENCI Project 2210), by IPP and DIM SIRTEQ (3Lab cluster), and by the Partnership for Advanced Computing in Europe (Project 2019204962). We acknowledge supports from the BCSi, SADAPTH & 21-CMAQ-002 ANR projects.

Full-potential Multiple Scattering Theory for the Calculation of Total Energy and Forces

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In the multiple scattering theory (MST) with Green function approach [1], also known as the Korringa-Kohn-Rostoker (KKR) method, ab initio density functional theory (DFT) calculations have often resorted to the simplifying assumption of spherically symmetric atomic potentials (muffin tin approximation). More advanced versions of the method account for anisotropy and charge modulation not only atom-to-atom, but also within the Wigner-Seitz cell around each atom - so called “full-potential” treatment [2]. In addition to the subtleties within the space-filling procedure [3], which require careful attention, numerical challenges also exist for the solution of the so-called single-site irregular wavefunction [4]. In this presentation, we report a full-potential MST scheme which does not need to explicitly evaluate this component of the electronic Green’s function. This scheme has been implemented in the open-source MuST all-electron DFT code for first-principles studies of ordered and disordered solids [5]. Beside numerical comparisons in terms of total energy calculation for semiconductors, we benchmark this approach through the calculation of phonon spectra, produced through a new interface to the PHONOPY package [6]. We also discuss other improvements produced by this full-potential scheme for the calculation of core-states, which is an extension of the method for the calculation of the shallow bound states [7] and is expected to produce more realistic results for the ab initio study of specific types of spectroscopy measurements [8].

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Vibrational frequencies and Stark tuning rate with continuum electrochemical models and grand canonical density functional theory

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The electrified solid-liquid interface is extensively studied in electrochemical works, and recently it is also being addressed with electronic-structure methods. Within these simulations, the electrochemical potential introduces a new degree of freedom, leading to grand-canonical or fixed-potential DFT calculations[1]. To account for this additional degree of freedom, a Legendre transformation is applied to translate properties between grand-canonical and canonical conditions. For instance, properties such as total energy, atomic forces, vibrational frequencies, and Stark tuning rates exhibit distinct forms across these boundary conditions. This work focuses on the numerical methods for the continuum model, including implicit solvent and electrolyte effects, and the algorithms for performing grand-potential calculations [2]. We demonstrate that atomic forces in the grand-canonical setting are equivalent to Hellmann–Feynman forces in the canonical framework. However, vibrational frequencies and Stark tuning rates vary between the two conditions. Using a finite displacement method, we find that the Stark tuning rate in the grand-canonical condition closely aligns with experimental data. We also observe that numerical errors can accumulate readily in finite difference calculations.

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Accurate and thermodynamically consistent hydrogen equation of state with flow matching

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Accurate determination of the equation of state of dense hydrogen is crucial for our understanding of planetary interiors [1]. In particular, there is still no consensus on the methods for calculating the entropy, which result in qualitatively different predictions for Jupiter's interior models [2, 3, 4, 5]. Here, we closely investigate various aspects of the entropy calculation of dense hydrogen based on *ab initio* density functional theory molecular dynamics simulations. In particular, we use the recently developed flow matching method [6, 7, 8] to validate the accuracy of traditional thermodynamic integration approach. This method employs a machine learned invertible transformation on the configuration space to directly connect two phase points of interest, thereby avoids any possible errors arising from the interpolation between them. This then lays a solid foundation for us to construct an equation of state over a wide range of temperature and pressure conditions in a fully thermodynamically consistent way, which agrees well with both numerical and experimental data in various phase regions. Compared to previous works, our results are much more reliable and hold the promise to resolve long-standing discrepancies in planetary science applications, as illustrated by a preliminary calculation for the thermal profile of Jupiter [9]. Our work not only represents an essential step toward the accurate description of dense hydrogen and giant planet interiors, but also shows a significant methodological advance in free energy and entropy calculations, an area which has broad interest within the condensed matter physics and material science community.

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Inducing out-of-plane magnetocrystalline anisotropy by chemical doping in 2D multiferroic CuCrP_2S_6

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Perpendicular magnetic anisotropy (PMA) has become a key focus in spintronics due to its inherent stability against thermal fluctuations, scalability, endurance, and low switching current density, making it highly desirable for advanced technological applications. Despite its significance, the ability to efficiently control PMA remains a challenge, prompting ongoing research to explore new ways to tune and obtain PMA in low-dimensional materials [1].

In this study [3], we predict an in-plane to out-of-plane transition in magnetic anisotropy orientation in the two-dimensional (2D) multiferroic material CuCrP_2S_6 (CCPS) [2] upon isovalent doping with indium, where a linear increase in MCA with In concentration in the mixed $\text{CuCr}_{1-x}\text{In}_x\text{P}_2\text{S}_6$ is observed based on first-principles calculations. We attribute this shift in MCA to structural and chemical effects. The chemical effect comprises all the changes caused by the substitution of Cr for In while preserving the pristine CCPS structure. We find it to be related to the decrease in the near-valence-edge density of states of the in-plane majority spin Cr-*d* orbitals through hybridization changes produced by isovalent doping with a nonmagnetic element. The structural effect is analyzed in terms of the changes in the in-plane lattice parameter and the monolayer thickness upon the addition of In. Finally, we extend our findings relative to the tuning of MCA towards more positive values to other non-magnetic dopants, isovalent with Cr^{3+} and of atomic radius larger than that of Cr^{3+} in 2D CCPS and related metal thiophosphates.

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Wannier-based definition of layer anomalous Hall conductivity in magnetic topological insulators

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The Wannier functions can be constructed from the Bloch functions using unitary transformation, and the hybrid Wannier function method is known as a localization method for only one axis [1]. Unlike ordinary Wannier functions, hybrid Wannier functions can be obtained without iterations for the most localized bases. In addition, based on the hybrid Wannier functions, a definition of layer polarization was obtained that corresponds to the contribution of each layer of electric polarization [2]. Thus, the hybrid Wannier function method is a powerful tool for visualizing the layer decomposition of physical quantities.

In this study, we develop a calculation method to decompose the anomalous Hall conductivity (AHC) into the contributions of each layer in layered materials to bring the definition of “layer anomalous Hall conductivity”, which corresponds to the layer-by-layer contribution of AHC, based on the hybrid Wannier functions. We formulated a method for finding the anomalous Hall conductivity at the Wannier center by combining a local Berry phase method [3] with the hybrid Wannier function method [1]. It was reproduced that the even-layered antiferromagnetic MnBi_2Te_4 is an axion insulator exhibiting a surface anomalous Hall effect, where the conductivity is $1/2$ in e^2/h units at the surface. For charge-doped systems of van der Waals antiferromagnets, we also evaluated “layer transverse thermoelectric conductivity” related to the layer-by-layer insight of anomalous Nernst conductivity. Our calculation method for layer anomalous Hall conductivity is expected to be applied to a wide range of systems, including two-dimensional stacking materials and artificial superlattices.

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Development of first-principles calculation method for spin Hall effect using local berry phase

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In the calculation of the spin Hall conductivity (SHC), methods such as the Kubo formula and Maximally Localized Wannier Functions (MLWF) are commonly utilized. However, for materials containing impurities, a supercell-based calculation model is necessary, which leads to significantly high computational costs due to the inclusion of unoccupied states and/or the construction of MLWFs. In this study, we apply the method proposed by Prodan [1] for calculating the spin chern number, combined with methods for calculating the anomalous Hall conductivity using local Berry phases [2,3], to compute the intrinsic spin Hall conductivity. In the presentation, we will present detail the implementation of a method that directly computes the SHC through local Berry phases. We will also present results obtained for Pt and Au and discuss these findings in comparison with previous studies [4].

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Real-space Visualization of Chern Number for Skyrmions

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Skyrmions are topological magnetic textures that emerge as classical objects in most experimental systems. However, the discovery of Skyrmions has generated interest in the quantum field on the nanoscale. Using a tight-binding Hamiltonian [1] $H = \sum_{ij} t^{ij} c_i^\dagger c_j - J \sum \mathbf{n}_i c_i^\dagger \boldsymbol{\sigma} c_i$, we study the two-dimensional Skyrmion lattice containing square lattice, Triangular lattice, and honeycomb lattice. c_i^\dagger (c_i) is the two-component (spin up and spin down) creation (annihilation) operator at the i site, t^{ij} is the transfer integral between nearest-neighbor sites, J is the Hund's coupling strength between the electron spin and background spin texture, $\boldsymbol{\sigma}$ denotes the Pauli matrix. The Chern number (C) can characterize the Skyrmions states as a topological invariant. We calculated the Chern number by the Fukui-Hatsugai method [2] on each occupied energy band. Furthermore, Bianco and Resta [3] have shown that the C can be mapped by a topological marker. The local Chern marker [4] is defined at the lattice site \mathbf{r}_i , $\mathfrak{C}(\mathbf{r}_i) = -4\pi \text{Im} \left[\sum_{\mathbf{r}_j} \langle \mathbf{r}_i | \hat{x}_Q | \mathbf{r}_j \rangle \langle \mathbf{r}_j | \hat{y}_P | \mathbf{r}_i \rangle \right]$. We compute the local Chern marker in the unit cell of different Skyrmion lattices and compare the topological properties of Skyrmion with periodic and open boundary conditions. To study more realistic skyrmion materials, we will present the implementation of this approach to density functional theory code, OpenMX.

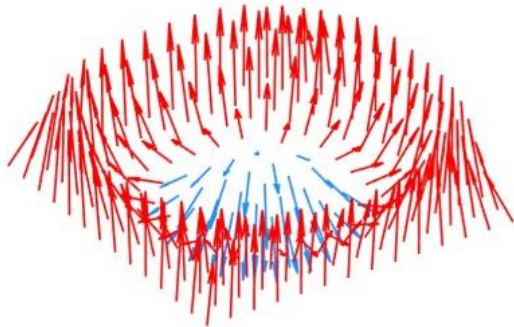


Fig.1: A schematic of the spin configuration of skyrmion.

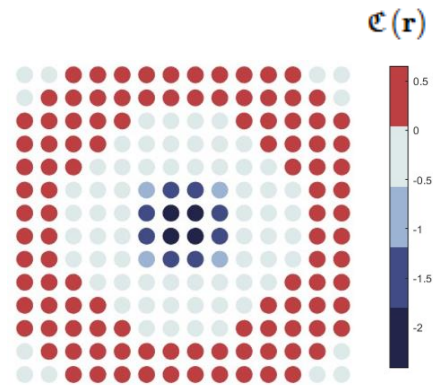


Fig.2: Local Chern marker $\mathfrak{C}(\mathbf{r})$ of the Skyrmion.

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Structure and dynamics of water at feldspar surfaces

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Over the last decade, significant evidence has been gathered that points to feldspar minerals as the most important ice nucleating particles in our planet's atmosphere [1]. There have been efforts to understand the formation of ice at feldspar surfaces using experimental tools such as optical microscopy [2], electron microscopy [3], and atomic force microscopy (AFM) [4], as well as computational tools such as density-functional theory (DFT) calculations [5], and molecular dynamics (MD) simulations [6, 7]. In spite of these efforts, the microscopic characteristics of the nucleation site and the atomic-scale mechanism of ice nucleation at feldspar surfaces have not yet been fully elucidated.

Here, we develop a machine-learning potential (MLP) to model the interactions at the water/microcline feldspar interface, using a dataset generated from density-functional theory (DFT) calculations based on the SCAN exchange-correlation functional[8]. The MLP accurately reproduces the energies and forces obtained from DFT calculations. We then performed MD simulations, driven by the MLP, to investigate the structural and dynamic properties of water on various fully hydroxylated terminations of the (100), (010), and (001) surfaces of microcline feldspar. With these simulations, we explore the effects of ions and surface structure on ice nucleation. Our results provide important insights into the effects of solid-liquid interface on the structure and dynamics of liquid water. These insights contribute to unravelling the microscopic mechanism of ice nucleation on feldspar.

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Multiscale model of photocatalytic ammonia synthesis described by the inclusion of excited states

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Photocatalytic ammonia synthesis is a sustainable and promising alternative to the conventional Haber-Bosch process, which results in notable carbon footprint and requires high energy input to activate the inert and stable nitrogen molecule. Multiscale modelling of photocatalytic reactions is still in its infancy, as atomistic calculations mostly neglect excited states, leading to incomplete results. In our study, we investigated a multiscale model of photocatalytic ammonia synthesis over TiO₂ photocatalyst by combining first-principles calculations at the atomistic level and microkinetic modelling at the meso level of modelling [1,2].

We employed density functional theory (DFT) calculations to study the ground state nitrogen dissociation mechanism, while time-dependent density functional theory (TD-DFT) was used to account for excited states, which are crucial for studying the system under photocatalytic conditions. First-principles studies were carried out using GPAW [3] and VASP [4] software. All the calculations were performed using the plane-wave basis set, the PBE functional, the Grimme-D3 correction to account for Van der Waals interactions and the Hubbard-U correction to consider on-site Coulomb interactions. After obtaining the dissociative reaction mechanism at the atomistic level of modelling, we entered the parameters obtained from the first-principles modelling into a microkinetic model to investigate kinetic-related properties.

Our results showed that it is necessary to consider excited state calculations to study the photocatalytic process, while the calculations performed in the excited states showed lower activation and reaction energies of all elementary steps during the full reaction mechanism. These results confirmed the feasibility of photocatalytic reactions that allow the study of a desired reaction under mild conditions.

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Single-layer boron phosphide on metallic surfaces: screening of promising substrates by first-principles

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Single-layer hexagonal boron phosphide (h-BP) is a predicted graphene-structured semiconductor exhibiting interesting properties for photovoltaic and photocatalytic applications [1] and is the most stable [2] between its proposed allotropes [3].

We investigate by ab-initio calculations the ground-state properties of h-BP on (111) surfaces of different d-shell metals of groups X, XI and XII, with the aim of identifying suitable substrates for its synthesis.

Due to the lattice parameter mismatch with the substrates, the h-BP overlayer generates Moiré patterns that we describe using minimal simulation cells with a tolerance of about 3% for tensile or compressive strain applied to the overlayer.

Adhesion energy, separation, and charge transfer between substrate and overlayer, projected density of states indicate that the least interacting substrate is Ag, closely followed by Au. The epitaxial h-BP monolayer assumes a buckled structure, with a buckling which is minimum on Ag and maximum on Au.

We complete the investigation of h-BP on Ag(111) by considering also the formation of flakes of finite, small size, that are typically more strongly anchored to the substrate through their edges, zig-zag or Klein type.

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