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19th International Workshop on Computational Physics and Material Science: Total Energy and Force Methods

9 - 11 January 2019

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BOOK OF ABSTRACTS

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19th International Workshop on Computational Physics and Material Science: Total Energy and Force Methods

9 - 11 January 2019
Trieste, Italy

Further information:
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The "Total Energy" Workshop has been held traditionally in Trieste every two years since 1987. This year, the Workshop is about recent progress in electronic structure methods and their applications. The aim is a critical discussion of methods and of possible combinations and challenging applications. It has become one of the most popular regular events of the international ab-initio electronic structure community. This event also has the ambition to reach out to countries where the electronic structure community is not traditionally as strong as in Europe, the USA and Japan, in coherence with the missions of its traditional host institution, the ICTP.

Description:

The Workshop focuses on recent progress in electronic structure methods and their applications to a steadily increasing range of materials and systems.
The most widespread approaches used in our community are DFT, TDDFT, GW, DMFT, and quantum Monte Carlo methods, which provide the foundation for computing many physical and chemical properties of solids, liquids, and low-dimensional systems. There are numerous challenging applications for which the levels of approximation used in these implementations, or their technical limitations, do not yet permit accurate quantitative predictions of some properties of materials. The goal of the Workshop is to give an in-depth analysis of a selection of scientific cases and applications. Cutting edge topics such as the use of multiscale modeling and machine learning will also form part of the programme.

Topics:

- Quantum Materials: Topology and Correlation;
- Strong Correlations in Quantum Chemistry;
- Excited-State Phenomena: New Approaches and Applications;
- 2D Materials and Heterostructures;
- Dynamics at Complex Interfaces;
- Advances in Molecular Dynamics for Bridging Scales;
- Machine Learning for Methods and Applications.

How to apply:

Online application:
http://indico.ictp.it/event/8658/

Female scientists are encouraged to apply.

Grants:

A limited number of grants are available to support the attendance of selected participants, with priority given to participants from developing countries. There is no registration fee.

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Deadline:

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ABSTRACTS

OF

INVITED TALKS

(in talk order)

Axion coupling in magnetoelectric and topological materials

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I will provide an introduction to the theory of the Chern-Simons axion coupling, which is of growing interest in the context of magnetoelectric and topological materials. The axion coupling can be expressed in terms of a phase angle θ which plays a role in 3D analogous to that of the Berry-phase ϕ describing electric polarization in 1D. In particular, like the Berry phase, θ is only well defined modulo 2π. In ordinary magnetoelectric materials $\theta \ll 1$ and its branch choice is obvious. In some classes of topological materials, however, the symmetry is such that θ might be expected to vanish, but instead $\theta = \pi$ exactly. In such cases, a half-quantized anomalous Hall conductivity (AHC) is present at any gapped surface. This is the case for strong topological insulators, "axion insulators" (in which the topology is protected by inversion), antiferromagnetic topological insulators, and some topological crystalline insulators. In this talk I will give an overview of recent developments in this field, including some of our own recent work on a real-space decomposition of the anomalous Hall conductivity[1,2] and our use of this formalism to study the surface AHC for models of axion insulator.[3]

1. T. Olsen, M. Taherinejad, I. Souza, and D. Vanderbilt, *Surface theorem for the Chern-Simons axion coupling,* Phys. Rev. B **95**, 075137 (2017).
2. T. Rauch, T. Olsen, D. Vanderbilt, and I. Souza, *Geometric and nongeometric contributions to the surface anomalous Hall conductivity*, Phys. Rev. B **98**, 115108 (2018).
3. N. Varnava and D. Vanderbilt, *Surfaces of axion insulators,* arXiv:-1809.02853.

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Standard model for crystalline materials: beyond the elementary particles and the 10-fold classification of non-interacting topological phases

Abstract:

One of the research directions in string theory is the separation of important theoretical problems into distinct classes based on their similarities. Electronic structure problem is usually not considered to be important in the string theory community. In this talk I will show that the electronic structure theory in fact allows not only for theoretical analysis of problems in quantum field theory and general relativity, but also for their cheap (on the LHC scale) experimental tests, and also provides many hints to other problems in physics, often considered to be of bigger importance than the study of material properties. In particular, I will show that even weakly-interacting crystalline materials realize a collection of topologically-protected quasiparticle excitations that can either be direct analogs of relativistic elementary particles, or due to the absence of Lorentz-symmetry constraint realize completely novel quasiparticles not present in the high-energy standard model. Materials that host such quasiparticles exhibit special transport properties. I will give a detailed description of several families of such materials. Finally, I will show that even the simplest elemental compounds hide physical phenomena that provide very accessible analogies to complicated theoretical physics theories, and illustrate that the current understanding of even the simplest non-correlated crystalline materials is far from complete.

Understanding strong-correlation effects in spin-orbit t_{2g} materials

Eva Pavarini

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Forschungszentrum Jülich, Germany

Strong-correlation effects in systems with low symmetry, non-spherical Coulomb vertex and spin-orbit interaction are particularly difficult to describe. Recent advances make it possible to investigate these phenomena via QMC-based LDA+DMFT calculations [1]. In this talk, I will use this technique to explain the electronic properties of representative t_{2g} materials [1-5]. For the unconventional superconductor Sr_2RuO_4 i will clarify the key role played by spin-orbit coupling and tetragonal Coulomb terms in determining the topology of the Fermi surface. For the Mott insulator Ca_2RuO_4, I will show that the spin-orbit interaction plays a little role in the metal-insulator transition. For the magnetic phase, two different scenario have been proposed, the local spin-moment ($S = 1$) scenario and the zero total-angular-momentum (or van-Vleck) picture. I will show that magnetic order and spin-wave spectra are well described in the perturbative spin-orbit limit, thus excluding the van-Vleck scenario.

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Dynamical susceptibilities and spontaneous symmetry breaking with dynamical mean-field theory

Abstract:

Combination of dynamical mean-field theory (DMFT) with ab initio band structure methods provided quantitive description electronic correlations in real materials. With spontaneous symmetry breaking being one of the most prominent correlation effects, DMFT allows access to various physical observables across phase transitions driven by temperature or other external parameters. For practical as well as fundamental reasons DMFT calculations focused largely on one-particle dynamics so far. In particular, the behavior of collective modes associated with spontaneous symmetry breaking has been unexplored with DMFT. Here we present a computational study of dynamical susceptibilities across excitonic condensation transition in two-band Hubbard model. We demonstrate that DMFT correctly captures the Goldstone modes associated with breaking of continuous symmetries and investigate the smooth evolution from gapped Goldstone mode to a truly amplitude Higgs mode when symmetry-breaking terms of increasing strength are added to the Hamiltonian. Specific to the excitonic condensation, we observe a quantitative change in the dynamical spin susceptibility at the transition, which may serve as the experimental probe of the condensate. I will also discuss the computational limitations and feasibility of the present approach to real materials.

Geffroy et al. arXiv:1808.08046

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Electronic Structure of Transition Metal Compounds by Variational Cluster Approximation

Abstract:

A famous theorem by Luttinger and Ward states that the grand canonicalpotential of an interacting Fermion system can be represented as a functionalof its self-energy and that this functional is stationary at the exact self energy. The variational cluster approximation is an application of this theorem, whereby trial self-energies are generated by exact diagonalization of small clusters. In applying this method to transition metal compounds it is important to treat the Coulomb interaction between electrons in the d-shell by means of multiplet theory. Applications of the method to 3d transition metal oxides and the spin-state transition in LaCoO3 will be discussed.

Modeling Point Defects for Quantum Information Science

Chris G. Van de Walle

Materials Department, University of California, Santa Barbara, California, USA

Defects in wide-band-gap semiconductors have emerged as promising single-photon emitters and solid-state qubits [1]. The nitrogen-vacancy (NV) center in diamond has been widely studied as an individually-addressable quantum system that can be initialized, manipulated, and measured with high fidelity at room temperature. The success of the NV center stems from its nature as a localized "deep-center" point defect. We have performed in-depth first-principles analyses of the NV center in order to elucidate its properties and to predict which centers in other materials might exhibit similarly favorable properties [2, 3, 4]. I will present an overview of the physics of deep centers, focusing on the characteristics that are key to their performance as "NV-like" centers. Building on the general methodology for performing point-defect calculations [5], we have developed the capability to predict transition energies and lineshapes associated with the optical transitions that play a central role in the functionality of the defect. Our methodology rigorously addresses the coupling between electrons and phonons during an optical transition, leading to an excellent description of the luminescence band [6, 7]. We also model nonradiative transitions [8]. The developments will be illustrated with examples for III-Nitride semiconductors including AlN and BN.

Work performed in collaboration with A. Alkauskas, D. Awschalom, C. E. Dreyer, L. Gordon, A. Janotti, G. Kresse, M. Mackoit, J.-X. Shen, M. Turiansky, J. Varley, J. Weber, L. Weston, D. Wickramaratne, and Q. Yan, and supported by DOE and NSF.

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Talk Abstract for IAS Focused Program on Quantum Simulations: From Chemistry to Materials Science

Many-body effects in oxygen transport proteins: a dynamical mean-field theory study

Cedric Weber

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Title:

Abstract:

Phenomena that are connected to quantum mechanics, such as magnetism, transport, and the effect of impurity atoms and disorder, and their relation to material design and energy needs are important for almost every branch of the industry. Density functional theory (DFT) was successful at making accurate predictions for many materials,in particular compounds which have a metallic behaviour.

DFT combines high accuracy and moderate computational cost, but the computational effort of performing calculations with conventional DFT approaches is still non negligible and scales with the cube of the number of atoms. A recent optimised implementation of DFT was however shown to scale linearly with the number of atoms (ONETEP), and opened the route to large scale DFT calculations for molecules and nano-structures. Nonetheless, one bottleneck of DFT and ONETEP, is that it fails at describing well some of the compounds where strong correlations are present, in particular because the computational scheme has to capture boththe band-like character of the uncorrelated part of the compound and the Mott-like features emerging from the local strongly correlated atoms. A recent progress has been made in this direction by the dynamical mean-field theory (DMFT), that allows to describe the two limits (metal and insulator) in a remarkable precise way when combined with DFT. The ONETEP+DMFT implementation and strategies to overcome the main bottlenecks of this type of calculations will be discussed, and its applications on oxygen transport proteins, e.g. Haemoglobin and Hemocyanin, will be demonstrated. The role of many body effects for the binding of transition metal complexes will be discussed.

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PRL 108, 256402 ‘ 12
PRL 110, 106402 ' 13
PNAS 111, 5790 '14

Using light as a topological switch

Luis E. F. Foa Torres
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Light-matter interaction is at the center of intriguing phenomena and has led to many practical applications like, for instance, Raman spectroscopy. But beyond characterization, several studies have gone deeper into actually using light to modify the electrical properties of a material. This can be done, for example, by using light to switch off the conduction in graphene [1,2] (or other materials [3]), thereby allowing to tune the material's response by optical means, or even inducing tunable topological states in materials that would otherwise lack them [1,2,4,5,6,7,8] (i.e. a *Floquet topological insulator* [4]). The latter would expand the playground of topological insulators to a broader set of materials. Recent studies have reported the experimental observation of laser-induced bandgaps at the surface of a topological insulator [3], and the observation of a light-induced Hall response [9], thereby adding much interest to this area

In this talk I will provide a brief overview of our works in this field with a focus on the generation of Floquet chiral edge states in graphene [6,10-11] and other systems/materials [8,12]. I will also comment on their Hall response [13] and laser-induced one-way transport of charge and valley [14].

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Hongjun XIANG

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Theoretical studies on new mechanisms of ferroelectricity and multiferroicity

Abstract:

Ferroelectric (FE) and multiferroic (coexisting ferroelectricity and magnetism) materials have attracted numerous interest over several decades due to the exciting possibility of novel applications such as non-volatile memory. To design high-performance ferroelectrics and multiferroics, it is highly desirable to discover new mechanisms of ferroelectricity and multiferroicity. Recently, we made some progress in this direction:

(1) It is generally believed that the oxygen octahedral rotation (an AFD mode) and FE mode in ABO_3 perovskites tend to compete and suppress each other. We reveal a dual nature of the FE—AFD coupling, i.e., it turns from competitive to cooperative as the AFD mode strengthens. We provide a unified model of such a dual interaction by introducing novel high-order coupling terms, and explain the atomistic origin of the resulting new form of ferroelectricity in terms of universal steric mechanisms [1].

(2) We discover that the ferroelectricity in SnTe and perovskite oxides thin films might increase with the decrease of the film thickness, in sharp contrast to the usual trend [2,3].

(3) It was known that either charge order (CO) or orbital order (OO) can lead to ferroelectricity. In some case where the individual CO or OO cannot ensure the appearance of ferroelectricity, we find that ferroelectricity may arise due to the simultaneous presence of CO and OO. On the basis of this new mechanism, we reveal that that $(CrBr_3)_2Li$ is a 2D ferromagnetic FE multiferroic with the asymmetric Jahn-Teller distortions [4].

(4) We propose that the hydroxide may act as a new knob to simultaneously control the magnetization and polarization. This may pave a new way to realize ultimate high-density memory using a single isolated hydroxide to represent one bit [5].

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Total Energy and Forces in Africa

Abstract:

Despite the scarcity of computational power in Africa, some research and training in the calculation of the electronic structure of different systems are ongoing. This talk will highlight the work of researchers in certain regions of Africa as well as showcase some successful training efforts and collaborations in the computational determination of total energy and forces of various systems. Collaborations are important in Africa as there is only a small percentage of researchers who can carry out high-level calculations of large systems withoutÂ linking up with groups outside their home countries. At the East Africa Institute for Fundamental Research (EAIFR), which is an ICTP Partner Institute, we are teaming up with others to create infrastructure for such high-level calculations as well as developing methods that can be used to carry out calculations on large systems with the accuracy of high-level first-principles approaches. At the same time, we are building human capacity that can make significant impact in Africa using these infrastructure and methods. Some of our work at EAIFR on determining the excited states of large quantum dots, with more than 10,000 electrons, will also be presented.

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Interatomic potentials from first principles

Abstract:

There is a new renaissance of interatomic potentials, they are now interpreted as surrogate models of the quantum mechanical potential energy surfaces. I will show how to define suitable representations of atomic coordinates and use regularised regression techniques, popular in machine learning nowadays, to achieve unprecedented accuracy in modelling hard and soft materials across a range of phases, including a wide variety of defects, allowing the study of phenomena previously unreachable using first principles approaches.

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Physics-Based Machine Learning for Atomistic Modelling

Abstract:

Statistical regression techniques have become very fashionable as a tool to predict the properties of systems at the atomic scale, sidestepping much of the computational cost of first-principles simulations and making it possible to perform simulations that require thorough statistical sampling without compromising on the accuracy of the electronic structure model.

In this talk I will argue how data-driven modelling can be rooted in a mathematically rigorous and physically-motivated framework, and how this is beneficial to the accuracy and the transferability of the model. I will also highlight how machine learning - despite amounting essentially at data interpolation - can provide important physical insights on the behavior of complex systems, on the synthesizability and on the structure-property relations of materials.

I will give examples concerning all sorts of atomistic systems, from semiconductors to molecular crystals, and properties as diverse as drug-protein interactions, dielectric response of aqueous systems and NMR chemical shielding in the solid state.

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Application of accurate wavefunction based methods to solids and surfaces

Abstract:

Theoretical studies on solids and surfaces have been dominantly based on density functional theory for many decades and its successes and failures have been well documented. In quantum chemistry, post Hartree-Fock methods have been developed that systematically converge to the exact solution of the many particle problem. The methods (mostly based on coupled cluster theory first proposed in the context nuclear physics) have the disadvantage that their rigorous application shows very unfavorable scaling with system size (7th power of the molecular size). However, great progress has been made in the past ten years and approximation methods have emerged that reduce the scaling down to linear while retaining 99.9% of the accuracy of the parent method. This allows for systematically accurate results to be obtained on systems with hundreds of atoms. That opens the possibility to apply the same methods to large cluster models of solids and surfaces in conjunction with embedding approaches. In order to validate the validity of the cluster approach, the strategy is two-fold: (a) monitor the convergence of the results with respect to cluster size extension and (b) for a converged cluster compare the results from the cluster and a true periodic DFT calculation. Agreement indicates that the wavefunction based calculation then provides a realistic representation of the solid or surface. We present results for binding energies, band gaps and X-ray spectra at the metal K- and L-edges all of which suggest that wavefunction based methods deliver an accuracy that is superior to DFT.

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Low-scaling GW calculations using Gaussian basis functions

Abstract:

The *GW* approximation of many-body perturbation theory is an accurate method for computing electron addition and removal energies of molecules and solids. In a canonical implementation, however, its computational cost is O(N^4) in the system size N, which prohibits its application to many systems of interest. I present a full-frequency *GW* algorithm in a Gaussian-type basis, whose computational cost scales with N^2 to N^3. The implementation is optimized for massively parallel execution on state-of-the-art supercomputers and is suitable for nanostructures and molecules, using either pseudopotentials or all electrons. The accuracy of the algorithm is validated on the *GW*100 molecular test set, finding mean absolute deviations of 35 meV for ionization potentials and 27 meV for electron affinities. Furthermore, the length-dependence of quasiparticle energies in armchair graphene nanoribbons of up to 1734 atoms in size is studied, and the local density of states across a nanoscale heterojunction is computed.

Optical and core spectroscopy from many-body perturbation theory

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First-principles methods based on density-functional theory and many-body perturbation theory are the state-of-the-art approaches to study light-matter interaction at the nanoscale. All-electron implementations, like the one provided in the **`exciting`** code [1], offer the additional advantage to treat on the same footing optical and core excitations [2]. After introducing the underlying methodology, I will discuss the role of correlation effects that are essential for a quantitative description of light-absorption phenomena and yet act rather differently in transitions to unoccupied states from valence and core electrons. To illustrate this point, I will present selected examples from my recent research, addressing optical and core spectroscopy in inorganic [2,3], organic [4-6], and hybrid materials [7,8].

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Optoelectronic properties at finite temperature

Abstract:

The optoelectronic properties of materials underlie a plethora of technological applications including solar cells, light emitting diodes, and flat pannel displays. Optimising the performance of these devices requires a detailed understanding of the microscopic quantum mechanical processes regulating light absorption and emission in semiconducting materials. Furthermore, as these devices need to optimally operate around room temperature, their microscopic description using first principles methods would benefit from the inclusion of finite temperature effects.

In this talk I will describe our approach to include the effects of temperature in the study of optoelectronic properties of semiconductors, including both thermal expansion and electron-phonon interactions. Our approach is based on finite difference methods, exploiting mathematical tricks such as the mean value theorem for integrals to accelerate the calculations [1,2]. I will describe the finite temperature properties of band alignments between semiconductors [3], optical absorption across both indirect [4] and dipole forbidden gaps [5], and luminescence driven by exciton-phonon coupling [6].

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[6] arXiv:1807.11797

A sounded journey from nano to macro into everyday materials through multiscale molecular simulations

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Abstract:

Recent advances in nanostructured systems have opened a wide range of new multifunctional materials with promising potential to control interfaces and flow at nanoscale. Furthermore, the unusual physical properties of confined fluids at nanoscale can play an important role in a plethora of chemical, geochemical and environmental processes. In this talk, I will summarize some of the activities in our group at IFUSP based on multiscale molecular simulations to explore the applications of nanoscience for fluid flow through nanoporous media on everyday materials (cement, rocks, oil and water). By using an integrated bottom-up multiscale computational approach ranging from first principles calculations based on Density Functional Theory, classical molecular dynamics and Lattice Boltzmann modeling, we have been able to systematically model, characterize and investigate the effects of the interfacial and wetting properties on fluid behavior of everyday materials over scales. This approach will be illustrated with two cases: (1) water confined at cement and (2) enhanced oil recovery processes based on surface drive flow. The mobilisation of hydrocarbons trapped at the pore scale can be favored by controlling by the chemical environment through “Wetability modifiers”, such as functionalized NPs and surfactants. The methodological challenges and the potential applications in the industry will be discussed. Additionally, we have applied sonification techniques to molecular simulations, which help us to improve our perception of the phenomena at nanoscale, but also it can be used as an inspiration for music composition and enhance immersive virtual reality experiences.

Modelling electrochemical solid/liquid interfaces by first principles calculations

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Processes taking place at solid-liquid interfaces are at the heart of many present day technological challenges related to the improvement of battery materials, electro-catalysis, fuel cells, corrosion and others. Describing and quantifying the underlying fundamental mechanisms is equally challenging for experimental and theoretical techniques.

Ab initio modelling using DFT has proven immensely successful in providing atomistic insight into various questions of materials science. A challenge in applying these methods to electrochemical problems is the presence of the solvent or constraints imposed by the periodic boundary conditions common to many DFT codes. Utilising concepts from semiconductor physics allowed us to overcome these obstacles [1, 2]. Key ideas of the underlying methods will be discussed by showing two prototypical applications: Selective stabilisation of polar ZnO(0001) surfaces by an aqueous environment [3] and corrosion on Mg surfaces [4].

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Descriptors from Small Data: Simple Yet Successful Descriptors for Self Assembly of Organic Molecules on Surfaces

Abstract:

Self-assembly is probably the most promising route to constructing devices at the nanoscale. The challenge is to predict the geometries of self-assembled architectures by utilizing only the properties of the individual molecular components. Working jointly with experimentalists, we have explored the feasibility of such an approach for a model set consisting of three host molecules (carboxylic acid derivatives of phenyleneethynylene) and five guest molecules (napthalene, phenanthrene, benzo-c-phenanthrene, benzo-ghi-perylene and coronene), self-assembled on graphene.

Using insights gained from scanning tunneling microscopy experiments and density functional theory calculations, we have formulated simple descriptors that can successfully predict the geometries of the host-guest architectures self-assembled from our palette of organic molecules. A structure map can be constructed using host and guest descriptors, with structures of the same type clustering in descriptor space. Though these descriptors can be evaluated at essentially zero computational cost, they correctly reproduce experimental observations, including the structural transitions exhibited by host assemblies upon introducing certain guest molecules. The descriptors are validated by their success in predicting not just the ground state geometry but also the energetic difference between competing structures for molecules that did not form part of of the training set used when formulating the descriptors.

This work suggests a novel approach toward the rational design of self-assembled nanostructures. It was performed in collaboration with Sukanya Ghosh at JNCASR, and Pratap Zalake and K. George Thomas at IISER Thiruvananthapuram.

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Computational materials discovery: good data vs big data

Abstract:

I'll present our approach and case studies in the broad area of computational design and discovery of novel materials, with a special focus on novel two-dimensional materials and Li-ion solid-state conductors. In the process, I'll highlight the needs and the infrastructure we built to manage massive amounts of calculations arising from such efforts, ensure the full reproducibility and provenance of the data, and allow the entire community to access, share, and store reliable and robust computational workflows and the resulting curated and raw data.

Electronic screening of quasiparticle excitations by atomically thin substrates

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The weaker screening response within two-dimensional (2D) materials has manifested in novel physical phenomena, with strongly bound excitons being a well-known example. Here, we study the screening properties in reduced dimensions by employing first principles many-electron GW calculations, focusing on the electronic screening of point charge perturbations adjacent to a 2D material. We find that this is an excellent approximation for predicting the HOMO-LUMO gaps of benzene adsorbed on 2D materials, without expensive GW calculations for the full system. Interestingly, by comparing the screening response of ~15 2D and 3D substrates to point charge perturbations above the substrate, we find that both 2D and 3D substrates have a screening response that obeys the *same* approximately linear relation with their quasiparticle gaps. This is in contrast to the much weaker screening response of 2D materials to excitations *within* the material, and implies that 2D materials are effective atomically thin dielectrics. Our results can be attributed to the fact that most of the induced charge responding to the perturbing potential is located within ~2-3 Å of the surface atomic plane in both 2D and 3D substrates. We further develop a GW embedding model that enables the treatment of large supercells of organic monolayers interfaced with 2D materials, and apply the method to a prototypical system – an ordered herringbone structure of PTCDA molecules on a monolayer WSe_2 substrate. These results show that the non-locality of the screened Coulomb interaction and polarizability of the layer result in reduced screening in the PTCDA monolayer compared to that in a monolayer of smaller benzene molecules. Our work contributes to the ongoing effort in making GW calculations more tractable for large interface systems, and uncovers new insights into the unusual physics of screening in reduced dimensions.

On-surface synthesis of graphene nanoribbons from a computational perspective

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Graphene nanoribbons (GNRs) are strips of graphene with nanoscale widths, meaning widths below 100 nm. Besides the width limit, the term GNR imposes no hard geometrical constraints in terms of length, aspect ratio or edge shape, and thus applies to a diverse set of graphene-derived nanostructures. Atomically precise GNRs promise to combine the outstanding electronic properties of graphene with an electronic band gap that is sufficiently large for digital-logic applications at room temperature.

It has been shown that atomically precise GNRs can be fabricated by an on-surface synthesis approach [1]. This versatile method has been successfully applied to the fabrication of armchair GNRs (AGNRs) of different widths -and thus different band gaps- as well as more complicated structures like chevron GNRs or heterojunctions [2]. More recently, it has also been extended to afford the fabrication of GNRs with zigzag edges (ZGNRs) [3], which are predicted to exhibit spin-polarized edge states

I will briefly review the on-surface synthesis approach to AGNRs from a simulation point of view, and discuss some recent additions to the family of GNRs such as atomically precise 6-ZGNRs [3]. I will then focus on the present challenges that simulations encounter in characterizing nanographenes on metallic substrates [4, 5] when they have to compare to experiments. Finally, I will discuss a family of zigzag edge-extended AGNRs hosting topological electronic phases. It will be shown that variations of the AGNR backbone width and the zigzag edge segment spacing drive this family of GNR structures into trivial, metallic and topological insulating phases [6].

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POSTER. SESSION I

WEDNESDAY, 9 JANUARY 2019

GROUP A - L

Exploring the impact of hydrostatic pressure on the Structural, Electronic and Mechanical properties of ZrNiPb Half-Heusler alloy: A DFT approach

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The structural, electronic, elastic and mechanical properties of ZrNiPb half-Heusler alloy under pressure ranging from 0 to 25 GPa has been studied using density functional theory within the generalized gradient approximation(GGA).The results of ambient condition were in good agreement with the available theoretical and experimental data. Our electronic structure and density of state results show that ZrNiPb is an indirect band gap semiconductor half-Heusler alloy with a narrow energy gap of 0.375eV. Based on the calculated elastic constants(C11 ,C12 and C44), Young's modulus(E), Poisson's ratio(ν), Shear modulus(G), Zener anisotropy factor(A) and brittle-ductile behaviours under pressure have been discussed. The calculated Poisson's ratio shows that ZrNiPb undergoes a relatively small volume change during uniaxial deformation. We show that the chemical bonds in ZrNiPb are stronger due to the high value of C11 .

First-principles study of the electronic structure of Na_2KSb for photocathode applications

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In the search for novel materials for electron sources with enhanced quantum efficiency and minimised intrinsic emittance, multi-alkali antimonides are particularly promising in light of their low band gap and electron affinity (~1 eV) corresponding to emission in the near-infrared and visible region [1]. In order to fully exploit their potential, it is essential to reliably characterise and understand the electronic structure of these materials. First-principles methods such as density-functional-theory (DFT) and many-body perturbation theory (MBPT) are particularly suited for this purpose. Here, we employ DFT to investigate the electronic structure of sodium potassium antimonide (Na_2KSb), which remains relatively unstudied compared to other heavier multi-alkali antimonides, particularly those involving caesium [2]. Calculating the band structure and density of states to identify the character of the electronic states is the first step towards accurately studying the electronic and optical excitations based on MBPT (GW and the Bethe-Salpeter Equation). For this purpose, we use exciting [3], an all-electron full-potential code implementing DFT and MBPT in view of modeling and understanding the photoemission process of Na_2KSb.

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Phonon Collapse and Second-Order Phase Transition in Thermoelectric SnSe

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Since 2014 the layered semiconductor SnSe in the high-temperature Cmcm phase is known to be the most efficient thermoelectric material. Making use of first-principles calculations we show that its vibrational and thermal transport properties are determined by huge non-perturbative anharmonic effects. We show that the transition from the Cmcm phase to the low-symmetry Pnma is a second-order phase transition driven by the collapse of a zone border phonon, whose frequency vanishes at the transition temperature. We calculate the lattice thermal conductivity obtaining good agreement with experiments only when non-perturbative anharmonic scattering is included. Our results suggest that the good thermoelectric efficiency of SnSe is strongly affected by the non-perturbative anharmonicity. We expect similar effects to occur in other thermoelectric materials.

Excitonic insulator in high pressure in 2H-MoS$_2$

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and Daniele Varsano[1]

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We study the pressure induced electronic phase transition in bulk 2H-MoS$_2$, an indirect gap layered material. By using accurate ab initio calculations based on the combined many body perturbation theory and density functional theory we calculate the exciton dispersion at different applied pressures showing that an excitonic instability appears at the pressure of 34 GPa, where the indirect electronic band gap is almost closed and the lowest indirect exciton present a binding energy of about 30 meV.

The calculated pressure induced electronic band gap closure is well in line with the estimated metallisation onset (about 35-40 GPa) reported in previous theoretical and experimental results for this material. Our results give promising insights about the occurrence of a pure electronic instability and the realisation of an excitonic insulator for a pressure slightly above the occurrence of the the pressure induced transition to metallic phase of bulk 2H- MoS$_2$.

High-Throughput investigation of close packed Transition Metal surfaces for electrocatalytic reduction of Oxygen

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In Fuel Cells, the anode reaction involves the decomposition of fuel (often Hydrogen), which is carried out fast enough on metal surfaces. The main issue in the development of fuel cells is the cathode reaction, where reduction of Oxygen molecules take place. This reaction is performed slowly due to the low kinetics of the reduction reaction, thereby reducing the efficiency of the fuel cell [1]. As a result, the main challenge in this topic is to find a catalyst for increasing the cathode reaction rate. Comprehensive research is being done on increasing the rate of the cathode reaction through catalytic processes. Platinum (Pt) is known as the most important Oxygen reduction catalyst in fuel cells. Considering different surface morphology or various alloys of platinum, catalysts with better performance can be obtained, which results in increasing the rate of reducing reaction.

In this study we are trying to perform a High-Throughput investigation of close packed Transitional Metal surfaces available in Crystallography Open Database (COD)[2]. Mainly we are focusing on Noble metals which are the metals of groups VIIb, VIII, and Ib of the second and third transition series of the periodic table. Workflows for all multinary alloys of close packed structure and electronic properties of their (111) surfaces are handled by AiiDA platform and Quantum ESPRESSO package respectively. Metal/aqueous work functions and potentials of zero charge [3,4] are calculated in the first step as a characteristic of these alloys. To get more realistic results on adsorption energies of Oxygenated species, the effect of electrode potential is modeled based on a work by Norskov [5,6] in which explicit electron and proton pairs (hydrogen atoms) are inserted simultaneously in metallic and aqueous phase respectively to build the desired potential difference keeping the electro-neutrality of the whole system at the same time without need for homogeneous background charge or counter electrode treatments.

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Finding the best/efficient way to predict mobility in semiconductors

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In this Poster, we will summarize our experience with the most recent ab-initio packages that deals with thermoelectric properties calculations beyond relaxation time approximation. Models within the framework of the Boltzmann transport equation (BTE) rely on constant relaxation time makes them unreliable for first time prediction, especially in semiconductors systems. Scattering effects such as phonon, dislocation and ionized impurity scattering, are a major aspect in defining the right mobility values; by calculating electron and hole mobility using codes such as aMoBT, Abinit, BoltzTrap2 and trans-optic, for a well known systems. We will reveal how finding the dominating scattering mechanism could drastically improve the results.

Electronic Structure and Stability of the Phases of Halide Perovskite RbSnBr3 :

A Density Functional Theory Study

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The most commonly used and studied halide perovskite is ABX_3, where A stands for Rb, B for Sn, and X for Br. A perovskite with high stability and ideal electronic band structure would be of essence, especially considering the materials used for solar cells. In this work, we have considered the three phases (cubic, orthorhombic, and triclinic) of $RbSnBr_3$. the three structures are studied to understand how the crystal structure influences the material's properties.

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Calculation of zero-field splitting for high-spin defects in solids

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For high-spin defects in solids, zero-filed splitting (ZFS, also known as magnetic anisotropy) is one of the key spectroscopic signatures addressable by electron paramagnetic resonance. Due to the complex nature of the ZFS, its comprehensive interpretation often has to rely on a combination of the experiment and the first-principles theory [1]. Here, we report on the recent progress in developing a general and efficient framework for density functional theory (DFT) based calculation of the ZFS within the supercell approach and projector augmented wave formalism [2]. As justified by benchmarking tests, our implementation [3] into the Quantum ESPRESSO software [4, 5] provides all-electron accuracy with the efficiency of the pseudopotential based approach.

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Stacking stability of C_2N bilayer nanosheet

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In recent years, a 2D graphene-like sheet: monolayer C_2N was synthesized via a simple wet-chemical reaction.[1] Here, we study the stability and electronic properties of bilayer C_2N . According to previous a study, a bilayer may exist in one of three highly symmetric stacking configurations, namely as AA, AB and AB'-stacking.[2] For the AA-stacking, the top layer is directly stacked on the bottom layer. Furthermore, AB- and AB'-stacking can be obtained by shifting the top layer of AA-stacking by a/3-b/3 along zigzag direction and by a/2 along armchair direction, respectively, where a and b are translation vectors of the unit cell. By using first-principles calculations, we calculated the stability of AA, AB and AB'-stacking C_2N and their electronic band structure. We found that AB-stacking is the most favorable structure and has the highest band gap, which appeared to agree with previous study. Nevertheless, we furthermore examine the energy landscape and translation sliding barriers between stacking layers. From energy profiles, we interestingly find that the most stable positions are shifted from the high symmetry AB-stacking. In electronic band structure details, band characteristic can be modified according to the shift. In this present work, the interlayer shear mode close to local minimum point was determined to be roughly 2.02×10^{12} rad/s

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Effective spin gating with van der Waals heterostructures

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We consider a possibility of devising an efficient spin current gate based on ferromagnetic metal slab - graphene interface [1]. Such a device should significantly change the spin polarization of the current passing through it when a small external electric field is applied to it. Graphene is chosen for this application because of its small density of states (DOS) around the Fermi level, which should cause large shifts in the Fermi level, and thus the conductivity of the structure, when the field is applied, while it consisting of only one layer would mean that the magnetic proximity effect should make the response weakly but significantly magnetic, in contrast to any nonmagnetic 3D structures added to ferromagnetic slab.

We show that laying graphene directly on cobalt surface results in chemical binding, which destroys the electron structure of graphene and leaves graphene DOS insensitive to the applied field [1]. As a solution to this problem a single layer of hexagonal boron nitride is inserted between graphene and cobalt so that it binds chemically to the slab, and then graphene binds to it by van der Waals forces, which largely preserve the electronic structure. The desired response is obtained in such a structure, although it is too weak because of strong doping of graphene when the chemical potential in structure is equilibrated [1]. To remove this unwanted doping, we explore a possibility of adding an additional platinum layer, platinum being known to absorb the electrons from graphene in pure platinum - graphene interface [2].

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Alternative Materials For Plasmonics and Hyberbolic Metamaterials

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By using first principles approaches, we study the optoelectronic and plasmonic properties of two classes of materials, namely metal-oxides and metal-nitrides, proposed as alternative to standard noble metals for plasmonic applications. We first investigate the origin of near-infrared plasmonic activity of several metal-oxides: transparent conducting oxides (TCO), such as Al-ZnO and Ta-TiO2, and phases change materials (PCM), e.g. VO2. In the former case, we investigate the microscopic effects of metal doping (e.g. Al, Ta) [1] and defects (e.g. vacancies) [2] on the optical and electronic properties of TCOs and how this reflects on the plasmonic response of surface-plasmon polaritons or layered hyperbolic metamaterials, in connection with other dielectric media. In the latter case, we focus on stacked heterostructures resulting from the coexistence of metallic and semiconducting phases of VO2. This joint-phase combination, which has been experimentally realized, gives rise to a natural hyperbolic metamaterial, which supports the propagation of volume-plasmon-polariton TM waves [3]. In the second part of the presentation, we will discuss the plasmonic properties of refractory metal nitrides (e.g. TiN, ZrN). We first investigate the plasmon dispersion relations of TiN bulk [4] and we predict the stability of surface-plasmon polaritons at different TiN/dielectric interfaces proposed by recent experiments. Finally, by combining first-principles theoretical calculations and experimental optical and structural characterization techniques, we study the plasmonic properties of ultrathin TiN films (2-10 nm) at an atomistic level for the realization of ultrathin metasurfaces with plasmonic nonlinear properties [5].

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Theory of phonon-assisted luminescence in solids: application to hexagonal boron nitride

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Here we provide a study of luminescence of hexagonal boron nitride (hBN) by means of non-equilibrium Green's functions plus finite-difference electron-phonon coupling. We derive a formula for light emission in solids in the limit of a weak excitation that includes perturbatively the contribution of electron-phonon coupling at the first order. Such a formula is applied to study luminescence in bulk hBN. This material has attracted interest due to its strong luminescence in the ultraviolet region of the electromagnetic spectrum[1]. The origin of this intense luminescence signal has been widely discussed, but only recently a clear signature of phonon mediated light emission emerged from the experiments [2]. By means of our new theoretical approach we provide a clear and full explanation of light emission in hBN.

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Interplay between interlayer exchange and stacking in CrX_3 (X = Cl, Br, I)

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The growing field of 2D materials has been enriched by the recent discovery of 2D ferromagnetic insulators since it adds magnetism to the already long list of their diverse physical properties: insulators (h-BN), semiconductors (MoS_2), conductors (graphene) and superconductors ($NbSe_2$). The family of CrX_3 (X =Cl,Br,I) is representative of this new materials. Bulk $CrCl_3$ is an antiferromagnet, whereas $CrBr_3$ and CrI_3 are bulk ferromagnets. In contrast, few-layer $CrBr_3$ and CrI_3 present an AFM interlayer coupling. [1] The bulk phases undergo a crystallographic phase transitions with temperature and there is evidence of an strong interplay between the crystallographic and magnetic degrees of freedom, both for the bulk and the few layer systems. [2]

In this study, we address the interplay between stacking and interlayer exchange coupling for CrX_3 2D crystals using first-principles calculations and an effective interlayer coupling model. Our results shed light on the magnetic behavior of the CrX_3 bulk and 2D crystals.

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Modelling graphene growth on stepped ni(001) surfaces

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Evidences of steps bunch opening due to graphene growth over Ni(100) stepped surface is shown by experimental STM images, which allow to identify the mono-atomic nature of the steps after graphene carpeting. Graphene smoothly follows the surface modulation without breaking, forming a moiré pattern with a periodicity ~1.4 larger with respect to the one on a flat Ni(100) surface [1]. Some relevant stepped surfaces configurations have been identified and studied through ab-initio simulations, in absence and presence of graphene. The ab-initio simulations provide a rationale of the characteristic periodicity of the graphene grown on stepped as well as on flat (001)Ni surfaces, in excellent agreement with the experimental evidences.

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Homogeneous Electron Gas Beyond GW Approximation

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Despite its simplicity the interacting homogeneous electron gas (HEG) is a paradigmatic test case in the study of the electronic structure of condensed matter. Beside being a model for valence electrons in simple metals, it also provides the basic ingredient for key electronic-structure theories. Here we propose to study it with many-body perturbation theory (MBPT), including diagrams beyond GW, to improve on the description of its spectral function. A novel numerical implementation of MBPT for the 3D non-relativistic HEG has been developed, with special focus on the treatment of the full-frequency dependence of the Green's function and self-energies.

Phase diagram of molecular and non-molecular phases of low-Z elements and compounds at extreme pressure and temperature conditions

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The phase diagram of solid Oxygen [1] and Carbon Dioxide at terapascal and gigapascal pressures, respectively, and several hundred Kelvin has been studied with ab initio density functional theory within the quasi-harmonic approximation for the vibrational free energy. Our work on Oxygen extends previous theoretical studies [2] done at zero temperature and shows that temperature has a dramatic effect on the sequence of phases. On the other hand, the proposed boundaries in the phase diagram of Carbon Dioxide provides a new perspective on the current phase diagram [3, 4, 5] by means of the relatives Gibbs free energy of the different molecular and non-molecular phases of CO_2.

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Koopmans-compliant functionals: A reliable and efficient tool for the prediction of spectroscopic properties

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Commonly used approximate density functionals produce total energies that do not exhibit the expected piecewise-linear behavior as a function of the particle number, leading to a discrepancy between total and partial electron removal/addition energies and poor predictive capabilities of ionization potentials. Koopmans-compliant functionals [1, 2, 3] enforce a generalized criterion of piecewise linearity in the energy of any approximate density functional with respect to the partial removal/addition of an electron - i.e., with respect to charged excitations - from/to any orbital in the system. When used to purify approximate density functionals, Koopmans corrections lead to orbital-density dependent functionals and potentials that are able to deliver accurate spectroscopic properties. As an example, ionization potentials of a large set of molecules (the GW100 test set), photoe- mission spectra of organic donors and acceptors [4] and band gaps of 35 semiconductors and insulators [5] are presented, showing very good agreement with experiment or higher- order theories. Being this a functional framework, the straightforward advantages are that forces and other derivatives are also readily accessible, that the computational costs are much reduced, and the numerical parameters are those typical of DFT calculations.

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Implementation of non-collinear spin-constrained DFT calculations in SIESTA with a fully relativistic Hamiltonian

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An accurate and efficient general method to constrain the magnetization of individual atoms or groups of atoms within a fully relativistic non-collinear spin density functional theory formalism is presented and implemented within the SIESTA code This approach can be applied to study a variety of complex magnetic configurations and to build effective magnetic hamiltonians for multiscaling micromagnetic simulations. As an example, the method is applied to obtain constrained magnetic states for a Fe_3 structure, and for a S=1/2 kagome layer (vanadium oxyfluoride $V_7O_6F_{18}$). Of paramount importance in spintronics is the control and manipulation of magnetic interactions between constituent species, characterized mainly by the pair–wise magnetic exchange tensor J_{ij}. By constraining the atomic magnetizations of an infinite Fe linear chain, the total selfconsistent energy values are mapped to a generalized Heisenberg model, obtaining not only the diagonal terms of J_{ij} but also the off–diagonal contributions due to the explicit presence of the Spin–Orbit coupling in the formalism. The diagonal values of J_{ij} promote short ranged ferromagnetic alignment whilst the non–zero off–diagonal values can lead to the formation of the spiral states in the chain, as expected from theory.

Geometrical Indicators for Magnetism in Pt Clusters

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"There is plenty of room at the bottom" has been a motto that has driven the research in nanoscale physics for many years [1]. Feynman points clearly at the nanoscale where new and exciting phenomena can take place, in particular when the behaviour of "a few" can influence that of "many" particles. In particular, in Nanoparticle physics, the lack of periodicity energetically stabilises various isomers whose morphology would be impossible in crystalline bulk. At the same time, the physical and chemical properties of a mono-metallic nanocluster do depend strongly on its shape and size. For example, catalytic properties rely on the presence of specific and diverse adsorption sites. Clearly, by playing with size, shape, and chemical composition and ordering of the nanocluster itself, we can modify its properties towards tailored applications. Here, we show that the magnetism arising in nano-sized Pt objects is due to an effect present in the second coordination shell [2] and how it might depend on the local morphology. We are also going to demonstrate how magnetism is affected by the surrounding environment in the case of small Pt- nanoclusters embedded in zeolite pores [3]. Hopefully, we can identify specific geometrical indicators that can help in designing nano-clusters with given physical responses.

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Band structure of semiconductors and insulators from Koopmans-compliant functionals

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Koopmans-compliant functionals [1, 2, 3] provide a novel orbital-density-dependent frame- work for an accurate evaluation of spectral properties by imposing a generalized piecewise- linearity condition on the total energy of the system with respect to the occupation of each orbital.

Because of the orbital-density-dependent nature of the functionals, minimization of the total energy leads to a ground-state set of variational orbitals that are localized. Within such representation the Bloch translational symmetry is broken and we are forced to use the supercell method.

As a proof of principle, here we show the Koopmans-compliant band structure of selected semiconductors and insulators, obtained by unfolding and interpolating the Gamma point only calculations. We finally present some considerations to address Bloch's theorem and overcome the limitations of the supercell method.

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Abstract

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We perform a fully relativistic study of electronic surface states and resonances of Ru(0001) and compare them to the recently measured photoemission band structure data. Our calculations are carried out in the plane-wave pseudopotential formalism of density-functional theory employing a fully relativistic ultrasoft pseudopotential. Surface states and resonances are identified by calculating the projected bulk band structure and by systematically evaluating the states' weight of the probability density at the surfaces. New surface states and resonances are found with respect to the previous studies. Finally, the role of the spin-orbit coupling in the Ru(0001) electronic structure is discussed. The largest spin-orbit splitting is found for the surface states at the K point, in agreement with the recent photoemission data.

Structural and electronic properties of SnS_2 stacked nanosheets: An ab-initio study

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We present an ab-initio study of the structural and electronic properties of SnS_2 stacked nanosheets using the standard LDA and GGA functionals as well as the newly developed variants of the non-local van der Waals (vdW) exchange correlation functionals, namely vdW-DF-revPBE and vdW-DF2-C09. We have examined different stacking configurations of the two, three and four SnS_2 layers. The GGA-PBE functional fails to describe the interlayer binding energies and interlayer spacing of SnS_2 nanosheets, while a good agreement is observed between the calculated and available experimental values when the van der Waals corrected functionals are used, mostly the vdW-DF2-C09. It is found that the interlayer interactions in the SnS_2 films are not only vdW type but, the overlap of wave functions of neighboring layers have to be taken into account. We have observed a systematic reduction in the band gap with the increase in the number of stacked layers. This can be another way of controlling the band gap of SnS_2 nanosheets as required for electronic devices.

Crystallization in elemental antimony: an Artificial Neural Network potential study

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Phase-change materials based on chalcogenide alloys are of great interest due to their ability to undergo reversible and fast transitions between the amorphous and crystalline phases upon heating. This property is exploited in rewritable optical disks (DVD) and phase-change non-volatile memories (PCM)[1]. Recently, elemental Sb has also been proposed as a suitable candidate for electronic PCM when confined in ultrathin film configurations [2]. Despite its extreme proneness to crystallization in the bulk, amorphous Sb can be stabilized up to hours at 300 K when grown in 3-nm-thick films capped with a dielectric material. The reasons behind the stabilization of the amorphous phase in this confined geometry is, however, unclear. Large scale atomistic simulations can provide useful insights on several effects that might concur to such stabilization. To this end, we have generated a high-dimensional Artificial Neural Network interatomic potential [3] for elemental Sb. Training is performed on a dataset of highly-converged DFT-PBE total energies and forces of approximately 15000 configurations generated from molecular dy- namics runs. The potential is first validated by computing structural properties of the bulk crystalline, amorphous and liquid phases. Then it is used to investigate the crystallization kinetics of Sb in bulk and confined geometries. Possible effects of the capping layer on the atomic mobility and of the stress present in the films on the crystallization kinetics are discussed.

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Ab Initio Simulation of Heat Transport in Silica Glass

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Thermal conductivity of glasses, and in particular of amorphous silica (a-SiO_2), is of crucial importance for many technological applications, ranging from thermal insulation to semiconductor fabrication, and for the interpretation of laser damage of optical glasses. Vitreous silica serves also as the basis of multicomponent silica glasses, which are usually characterised by a complex chemistry that is difficult to model with classical force fields, and that are widely used, e.g., for the safe disposal of nuclear wastes. Many classical MD studies have been performed in the past and have demonstrated their ability to account quite well for all the structural properties of a-SiO_2, but remarkably lack a proper description of its vibrational spectrum, that requires a treatment from first principles.

The methods usually adopted to study heat transport in crystalline solids, such as the Boltzmann transport equation, cannot be applied to glasses, were the disorder makes the quasi-particle picture of heat carriers break down. Instead, the Green-Kubo (GK) theory of linear response can be straightforwardly applied to obtain the thermal conductivity from the fluctuations of the heat current at equilibrium. Nonetheless, until very recently, the GK approach was not deemed compatible with quantum simulation techniques based on density functional theory because the concepts of energy density and current, which are essential ingredients of the former, are not well defined in the latter. Besides, the study of transport coefficients using the GK theory is known to require very long molecular dynamics (MD) simulations, thus making ab initio techniques unaffordable in practice.

We discuss how to overcome these two hurdles thanks to a paradigm shift based on the concept of gauge invariance of transport coefficients [1, 2], and by using a novel data analysis technique based on the so-called cepstral analysis of stationary time series [3]. These theoretical and methodological advances make the quantum simulation of heat transport in liquids and amorphous solids finally possible, using equilibrium ab initio (AI) MD simulations.

By means of classical MD simulations we study the dependence of the thermal conductivity of a-SiO_2 on the sample size and the quenching rate used to prepare the glass sample, we show that relatively short trajectories are needed to obtain an accuracy of the order of 10% on the thermal conductivity, and we prepare glass models of appropriate size that will be subsequently simulated using AIMD. Finally, one sample of a-SiO_2 is simulated with AIMD at four different temperatures. The resulting thermal conductivities show fairly good agreement with experimental data, and a substantial improvement with respect to classical MD results.

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CDW temperature in bulk and monolayer transition metal dichalcogenides from first-principles

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A charge-density wave (CDW) transition is a lattice dynamical instability that induces a modulation of the charge density. Transition metal dichalcogenides (TMDs) show a very interesting and diverse phase diagram where CDW and superconducting states might appear.

The interest on TMDs has attracted a great deal of attention since it was shown that these layered materials could be exfoliated down to the monolayer and provide a wide of range of 2D materials[1]. Whether the dimensionality reduction affects the CDW is not clear at the moment as contradictory results have been recently reported: while optical and electrical transport measurements suggest the CDW temperature is strongly enhanced in the monolayer[2], scanning tunnelling microscopy suggests it is practically unaffected[3]. This apparent contradiction calls for a theoretical first-principles study of the CDW temperature in both bulk and monolayer TMDs.

A CDW transition is a second-order structural phase transition in which the frequency of a phonon mode of the high-temperature phase is strongly suppressed with lowering the temperature, collapsing to 0 at the CDW transition temperature. This triggers the CDW state. Thus, in order to determine the CDW temperature, ab initio methods need to calculate the temperature dependence of the phonons at the high temperature phase and see at which temperature the frequency of the CDW mode vanishes. This, unfortunately, is a daunting task, as the standard harmonic approximation predicts that the phonons of the high-temperature phase are unstable and cannot explain any temperature dependence of the phonon frequencies. Moreover, any perturbative approach is not viable and a non-perturbative treatment of anharmonicity becomes necessary, not a trivial task for first-principles approaches.

Making use of the stochastic self-consistent harmonic approximation (SSCHA)[4, 5, 6] that we have developed, we can accurately calculate for the first time the temperature dependence of the phonons even in the non-perturbative anharmonic regime and deter- mine the CDW transition temperature. Here, we present what to our knowledge is the first fully ab initio calculation of a CDW temperature both in bulk and monolayer NbSe2 and NbS2. Our calculations are crucial to understand the interaction of the CDW with the dimensionality, and shine light into the experimental controversy.

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Tribochemical conversion of Methane to Graphene on Sliding Nickel Surfaces

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Graphene and other carbon-based nanostructures are known to provide remarkable friction and wear performance but without continuous replenishment, they wear out, and thus high friction and wear prevail again. We show dissociative extraction of graphene and disordered carbon from methane molecules on the sliding surfaces of Ni. In a steel on steel sliding experiment it is observed, by means of Raman spectroscopy and tunnel electron microscopy, that methane is converted to a tribofilm consisting of graphene and other carbon nanostructures if steel is coated by a Ni-VN nanocomposite. The carbon based tribofilm is shown to reduce the friction coefficient by nearly 50% [1]. By means of ab-initio Molecular Dynamics simulations we identified the initial stage of tribologi- cally induced formation of 2D carbon nanostructures from CH_4 molecules. The process is initiated by CH_4 dehydrogenation, which is highly accelerated by the tribological conditions: the process is endothermic at the open surface in static conditions, while it occurs almost instantaneously at the tribological interface. The detached H atoms diffuse into the Ni bulk, and the interfacial C atoms become interconnected within chains that form an amorphous film of low density. The load and shear stresses applied to the carbon film sandwiched between the Ni surfaces promoted the rehybridization of C atoms and the formation of sp 2 planar structures with the same lattice parameter of Ni(111). The newly formed hexagonal rings constitute the first nucleation centers for graphene growth. Eventually the amorphous film self-organizes into two separate graphene flakes adsorbed on the sliding surfaces.

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An ab initio perspective on the nature of reversible martensitic transformations in metals

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The peculiarity of shape memory materials is to exhibit extremely reversible martensitic transformations (MTs) when temperature or stresses are applied. Modelling these materials from first principles is not trivial, since static calculations are usually not sufficient to explore the most important aspects of the MTs, such as the Ehrenfest order of the phase transitions, their temperature hysteresis and their free energy barrier. In this poster, a fully ab initio strategy to characterize MTs in alloys is presented. The temperature dependent order parameters of a MT are calculated with ab initio molecular dynamics; then, the free energy of the MT, from which all the relevant thermodynamic properties of the MT can be derived, is modelled with a Landau-like polynomial expansion. This approach is applied to the high-temperature shape memory Ti-Ta alloy, for which exceptionally small values for the free energy barrier and temperature hysteresis are observed.

Ab-initio investigation of a novel photocathode: bulk and surface properties of $CuFeO_2$

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The urgent need to transition from fossil fuels to renewable energy sources is driving several avenues of intense research. Photoelectrochemical (PEC) devices offer the possibility to convert solar radiation into chemical fuels, mimicking the natural process of photosynthesis. Recent experiments have highlighted the delafossite $CuFeO_2$ (CFO) as a promising candidate in the role of the photocathode [1, 2], yet in spite of these encouraging results, improvement in the catalytic activity and charge separation is required. An adequate theoretical characterization is currently not available to provide insight that can direct further experimental investigations.

In this work, we present the first fully self-consistent Hubbard U investigation of CFO using the linear response approach introduced and refined by Cococcioni, De Gironcoli, Kulic, Scherlis and Marzari [3, 4]. On this basis we characterize both the geometric and the electronic properties of the bulk CFO for a range of magnetic configurations, with results suggesting that, like other Fe-oxides, the antiferromagnetic arrangement of spins is favored.

The absolute position of the CFO band edges relative to the HOMO and LUMO levels of H_2O at the CFO/H_2O interface is fundamental for PEC applications. To this end it is necessary to go beyond the bare bulk properties, investigating the energetics of the surfaces and how they interact with water. Given the novelty of the application as a photocathode, little experimental characterization of the surface and interface is available, it therefore remains for us to establish the most stable surface.

The coupling of Density Functional Theory and Ab-Initio Thermodynamics allows us to make comparisons between a range of pristine surfaces, and also those surfaces in the pres- ence of (up to know charge neutral) defects. Based on this thermodynamic screening we have been able to focus our investigation on the two most energetically favored surfaces: a $(11\bar{2}0)$ cut with Fe vacancies and a (0001) cut with a complete Fe/O termination. We report on the electronic and geometric properties of these surfaces exposed to the vacuum and H_2O.

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Quantum Magnetism in Osmates Double Perovskites

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The interplay between electron correlation, structural distortions and spin-orbit coupling is among the most challenging aspect of condensed matter physics. These energy scales are simultaneously active in 5d transition metal oxides, which represent a rich playground for discovering new quantum states of matter. In particular, strong relativistic effects can lead to the formation of complex noncollinear magnetic orderings, whose origin can- not be understood within a standard Heisenberg picture. Instead, one needs to adopt a pseudospin approach and consider multipolar superexchange interactions. In this contri- bution, we study the ground state of the $5d^1$ osmate-based double perovskite Ba_2NaOsO_6 (BNOO) and decipher the driving mechanism that leads to the onset of the observed canted antiferromagntic (AFM) pattern. The structural, electronic and magnetic proper- ties of BNOO are computed using fully relativistic and magnetically constrained DFT+U, where the on site Coulumb interaction parameter U is estimated fully ab initio within the constrained random phase approximation. We find that the magnetic energy landscape (in particular the canting angle) depends critically on the cooperative Jahn-Teller distor- tions and on the strength of U. In order to acquire additional information on the quantum origin of the canted AFM state we map the first principles total energies onto an extended pseudospin Hamiltonian, and found that quadrupolar and octupolar terms are the key interactions that drive the stabilization of the noncollinear ground state. Furthermore, as a preliminary step for understanding the effect of electron-doping in the system, we will show the results obtained for a full chemical substitution of Na with Ca, resulting in the $5d^2$ double perovaskite Ba_2CaOsO_6.

Floquet theory for the electronic stopping of projectiles in solids

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The problem of the electrons in a solid being perturbed by a projectile, the electronic stopping problem, has been extensively studied for over a century. It is traditionally approached and it is well understood in the context of linear response[1], but there are regimes in which the perturbation cannot be considered small in any sense, and the response is far from linear. A fully non-linear theory[2] was proposed four decades ago based on a change to the projectile's reference frame for jellium as the target solid, which provided an important paradigm for the understanding of electronic stopping processes in simple metals, but left unexplained many observed qualitatively different behaviours in e.g. insulators (with an observed threshold effect at low projectile velocity), or noble and transition metals (with an unexplained change in the low-velocity dissipative force). More recently, more quantitative simulations stated to be performed from first principles using time dependent density functional theory, which are not limited to simple metals, and have shown reasonably predictive accuracy, including for the qualitatively different effects mentioned, but which remain computationally expensive, and, more importantly, do not provide a paradigm for the intuitive understanding of the stopping process. Moreover, it has always been assumed, when performing such calculations, that a stationary state is achieved while the velocity does not change appreciably. It appears naturally in the linear-response and jellium theories, but there is no formal validation of this assumption for the general case.

We propose here a general (single-particle) stationary theory for the electronic exci- tation in crystalline solids by a constant-velocity projectile. It is based on the Floquet formalism for time-periodic systems[3]. It arises from the observation that when a projec- tile travels at a constant velocity through a crystal in a direction such that the crystalline potential is periodic, there is a discrete translational symmetry in space-time, namely, the simultaneous translation by nL in space, n being any integer, and L the repetition length along the trajectory, and by nL/v in time, v being the projectile velocity. One can then use Floquet-Bloch theory along that space-time diagonal, or purely Floquet's in the projectile's reference frame, generalising to any crystalline solid the theory of Ref. [2]. We show that, in this formalism, stationary (stroboscopic) solutions are explicitly allowed and appear naturally as Floquet states. Non-trivial effects such as the mentioned low-velocity threshold effect can be analysed in this framework and one can see that such a threshold for the electronic stopping power is not to be understood in a strict threshold sense, but the substantial depression of electronic stopping is understood, confirming what had been recently advanced from the analysis of the results of first-principles simulations. This framework can be useful not only for electronic stopping calculations, but it can be relevant also for the study of ultra-cold atomic gases, where charged impurities might be added to a neutral gas, and, in general, for any system in which an impurity moves within a periodic lattice.

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High-throughput DFT calculationsto explore structures of AB_2 type monolayers

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Recently two-dimensional (2D) monolayers, which have peculiarities and novel properties, have attracted great interest. For example, graphene and silicene are well-known materials as atomically thin monolayers. Boron nitride and silicon carbide are known as AB type monolayers. In addition, various kinds of monolayers of transition metal (A=Mo, W, etc.) dichalcogenides (B=S, Se or Te) with a formula AB_2, which are arranged in tri-atomic layers, have been recently synthesized experimentally for producing an AB_2 type 2D transition metal chalcogenide library [1]. These kinds of investigations have revealed interesting phenomena such as two-dimensional superconductivity and quantum spin Hall effect. However, since the combination of the elements is diverse even only for the AB_2 composition, it can be considered that there is a room for exploring unknown stable monolayers which have not been synthesized experimentally yet.

In this study, high-throughput calculations based on the density functional theory were performed by using OpenMX[2] to create a structure map for AB_2 type monolayers. At first, we classified the AB_2 structures into planar, 1T (trigonal) phase (ex. ZrS_2 [1]), 2H (hexagonal) phase (ex. MoS_2 [1]), memory structure and the others. Here, the group of "planar" means B atoms form almost honeycomb structures and A atoms are on the center of the honeycombs. The group of "memory structure" means that the structure is similar to the planar, but A atoms are shifted from the plane formed by B atoms. Since the positions (up or down) of A atoms can represent binary digits, we call the buckled structures "memory structure" here. This structure can be a candidate for a data storage application with an extremely high areal density. Next, we prepared the three kinds of 2×2 supercell AB_2 initial structures (1T, 2H and planar) for selected compounds (62 elements×62 elements=3844 compounds), where compounds of noble gases, lanthanoids and actinoids are excluded. After geometry relaxations and variable cell optimizations were carried out, the most stable converged structures were summarized on the structure map for AB_2 type monolayers.

The structure map predicted that some of the structures can be the memory structure. In addition, some NEB calculations for these memory structures indicate that it may be possible to control them as binary digits storage applications. In this presentation, we report the details of the high-throughput calculations and show some obtained interesting structures.

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Relative stability and magnetic ground state of all stacking patterns in bilayer chromium trihalides

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Chromium trihalides are layered magnetic materials that have recently attracted considerable attention owing to their easy exfoliability and the ensuing possibility to isolate magnetic 2D materials. Single layers of CrX_3 (with X = Cl, Br, I) are ferromagnetic, with a magneto-crystalline anisotropy that favour spins to lie in the plane of the layer for $CrCl_3$ and out-of-plane for $CrBr_3$ and CrI_3. This difference is reflected also in distinct interlayer interactions when layers are stacked into their bulk form: the interlayer cou- pling is antiferromagnetic for $CrCl_3$, while it is ferromagnetic for $CrBr_3$ and CrI_3. Still, multilayers of CrI_3 seem to behave as antiferromagnets, whereas $CrBr_3$ remains ferromag- netic. Here, we address this controversy by studying bilayers of these materials using density functional theory thanks to the recently developed spin-polarized van-der-Waals functionals. We enumerate all possible stacking patterns for bilayers with the smallest commensurate unit cell and investigate their relative stability. We show that, depending on the stacking order, the magnetic ground state can be different. In particular, we recover that the lowest-energy configuration corresponds to the bulk stacking and it is correctly ferromagnetic for CrI_3 and $CrBr_3$, although there exist other low-lying metastable con- figurations that are antiferromagnetic and that could be obtained during the exfoliation process. Based on our findings, we speculate on how the apparent controversy could be resolved.

Efficient nonparametric n-body force fields from machine learning

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In the last decade there has been a proliferation of successful applications of Machine Learning (ML) algorithms to complex physical problems, a good example being the use of Gaussian Process (GP) regression to approximate interatomic force fields at DFT level of accuracy.

The quality of a GP regression is entirely dictated by the quality the its kernel function, which encapsulates the prior knowledge on the system studied. We developed a general scheme to build GP kernels that fully encode physical prior information while maintaining a controllable level of complexity [1].

The procedure extends and clarifies well known previous approaches (e.g., [2] and [3]) and lies in a systematic expansion of the approximating force field into n-body interactions via the design of n-body kernels. The problem of choosing the order n, best suited to describe a given system can be then approached in a principled way through Bayesian model selection.

GP predictions coming form the chosen n-body kernels can finally be mapped exactly onto explicit basis functions, giving rise to nonparametric classical potentials. These are as fast as standard classical potentials [4] (and thus crucially orders of magnitude faster than typical ML force fields), but provide substantially lower errors with respect to DFT forces. Furthermore, they can be generated automatically with no need of complex non-linear parametrisation and optimisation.

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Multiscale simulations of critical interfacial failure in carbon nanotube-polymer composites

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Computational investigation of interfacial failure in composite materials is challenging because it is inherently multi-scale: the bond-breaking processes that occur at the covalently bonded interface and initiate failure involve quantum mechanical phenomena, yet the mechanisms by which external stresses are transferred through the matrix occur on length and time-scales far in excess of anything that can be simulated quantum mechanically. In this work, we demonstrate and validate an adaptive quantum mechanics / molecular mechanics (QM/MM) simulation method that can be used to address these issues and apply it to study critical failure at covalently bonded carbon nanotube (CNT)-polymer interface. In this hybrid approach, the majority of the system is simulated with a classical forcefield, while areas of particular interest are identified on-the-fly and atomic forces in those regions are updated based on QM calculations. We demonstrate that the hybrid method results are in excellent agreement with fully-QM benchmark simulations and offers qualitative insights missing from classical simulations. We use the hybrid approach to show how chemical structure at the CNT-polymer interface determines its strength and we propose candidate chemistries to guide further experimental work in this area.

Keywords: Multiscale, QM/MM, CNT, polymer, composite, interface

Computational insight on the relation between structure and color expression in anthocyanin-based natural dyes and their metal complexes

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Anthocyanins are among the most interesting organic dyes in nature, which, depending on the chemical environment (such as acidity and co-pigmentation), give rise to a variety of colors and shades and are responsible for most of the natural hues in the red-purple-blue gamut [1]. The presence of metal also plays a very important role. In spite of the ubiquity and potential applications of anthocyanins in industry, the molecular mechanisms underlying their exceptional photophysical versatility is not yet fully understood.

I will present the application of use a newly developed multiscale modeling protocol [2, 3, 4] to reveal the intricate relationship existing between the electronic and geometrical factors that determine the optical properties and hence the color function of anthocyanins in a broad pH range (1 to 9) of cyanidin-3-glucoside in water solution, as a prototypical model system. The control on the optical properties is then obtained by addressing not only the charge state (positive flavylium cation, neutral and negative bases) of the relevant chemical species, but also the different tautomers of each charged state. For each species, detailed optical information is obtained by addressing the properties of different conformers, characterized by the slow dynamics of a few internal degrees of freedom. Our study unambiguously reveals that a subtle combination of different structural and electronic traits controls color expression, such as dihedral angles between aromatic moieties, the nature of the bond connecting the two, and the overall charge state of the molecule [5]

Besides, I will introduce some more recent results on metal complexes. Aluminium complexes of cyanidin-3-glucoside are known to form in solution rendering bluer shades, especially at large pH. By computer simulation, we can gain important understanding about the mechanism for such color properties. We are also able to provide a qualitative picture about the chemical species present at different pH, that were unprecedently reported [6].

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Peierls distorion in the blue bronze: a quantitative study based on ab initio density functional theory

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Most of the quasi-one dimensional materials undergo a metal to insulator phase transition followed by a periodic lattice distortion[1]. The anisotropy of their electronic structures reflects in their temperature dependent behaviour; for instance, by studying the nesting vectors which couple the different regions of the Fermi surface, one can often reveal different mechanisms that drive electronic instabilities leading to changes in the electronic properties of these materials. Density functional theory calculations can provide plenty of information about these processes, and just a look at the Fermi surface can provide very valuable insight. Nevertheless, the Lindhard response function provides a more quantitative description. Here, we provide a direct confirmation for the interband nesting mechanism which drives the Peierls distortion in the quasio-one dimensional blue bronze, $K_{0.3}MoO_3$, by calculating the Lindhard response function. This procedure allows us to provide precise values of the nesting vectors of this material and thus we can make a comparison with the available experimental data. Finally, we provide evidence for the proposed weak electron-phonon coupling scenario in the Peierls transition of this material.

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Electronic Stopping of Protons in Graphite and Prototypical Polymers

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Ions shooting through condensed matter have been studied for over a century [1] in different contexts, both fundamental (particle discovery and target system characterisation) and applied (relevant to radiation damage processes of materials of interest to the nuclear and aerospace industries as well as to medical applications). At projectile velocities typically beyond 1% of the speed of light the projectiles are mainly stopped by electrons, in processes that are strongly non-adiabatic and far from equilibrium, with electronic stopping powers from a few eV/Ang (light projectiles) to many keV/Ang. Recent advances in the first principles calculation of these processes using time-dependent density-functional theory have allowed their detailed and relatively accurate simulation in a variety of systems, including e.g. protons in wide band-gap insulators [2] and noble metals [3], and heavy projectiles in transition metals [4].

In this work we focus on target systems with a less homogeneous particle density in order to explore the expected larger impact-parameter dependence and anisotropy of the electronic stopping processes. Results will be presented for the electronic stopping power for protons in graphite and in crystalline polyethylene and polyacetylene, which represent prototypical cases of two-dimensional and one-dimensional covalently bonded systems, respectively, the sheets or chains being weakly bound to each other by dispersion inter- actions, with interstitial regions of low electron density. In addition to anisotropy, the proximity of the trajectory to carbon atoms is found to be more significant than expected, conforming to assumptions of local density dependence of the stopping power.

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Graphene and *h-BN* based symmetric and non-symmetric magnetoresistive junctions

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ABSTRACT

Using first principles investigations combined with the nonequilibrium Green's function are performed to understand the spin-polarized transport in magnetotunnel junctions (MTJs) consisting of an out-of-plane graphene (Gr) and h-BN single and bilayer sheets as a barrier in between two ferromagnetic Transition metals (TM). Our investigation is not restricted to only symmetric junctions, i.e., with the same metal on both sides of the spacer layer *Ni(111)* but non-symmetric, two different (*Ni* & *Co(111))* metallic ferromagnetic electrodes. In spite of low magnetic exchange couplings in all junctions, the spin filtering is quite sensitive to ferromagnetic metal and the thickness of the semiconductor. Remarkably, a significant spin polarization is observed in bilayers Gr(BN)that is robust and independent of the junction type as compared to single layer junctions. Interestingly, the TMR ratio is higher for symmetric junctions rather than non-symmetric junctions. The highest 50% TMR ratio is achieved for bilayer graphene while it is 43% for bilayer h-BN with symmetric *Ni(111)* electrode. Our present work reveals that the bilayer sheets of semiconductors are promising candidate for the spacer of MTJs which may be used for the design of next-generation spintronics applications.

Bridging Scales: Ethylene Epoxidation on Silver from Ab Initio Calculations to Reactor Level Simulations

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Ethylene epoxidation is one of the most important selective oxidations in industry. For a controlled oxidation of ethylene into epoxide, silver is the only viable catalyst. Usu- ally, silver is doped with alkali or noble metals and chlorine to achieve better conversion and selectivity. However, even pure silver exhibits very different selectivity, activity, and turnover frequency if different facets are examined. We studied this reaction at various time- and length-scales. With density functional theory (DFT) calculations, we elucidated the reaction mechanism.

On Ag(1 1 1), effects of alkali metals and chlorine on the reaction mechanism were investigated. We showed that electropositive dopants induce electric dipoles on the surface, which improve the activity and selectivity of the reaction. Chlorine stabilizes the alkali metals and increases the selectivity. [1] Comparison of the activation barriers, reaction energies and entropy on different facets (Ag(1 1 1), Ag(1 1 0), and Ag(1 0 0)) showed that Ag(1 0 0) should be much more active and selective than Ag(1 1 1). With kinetic Monte Carlo (kMC) simulations, we confirmed this effect. [2] For two most active surfaces, mean-field concepts – micro-kinetics and computational fluid dynamics (CFD) – were used to simulate how the synthesis proceeds in a reactor. [3] We investigated the effects of pressure, temperature, and particle composition, size and shape on the performance. We showed how multiscale modelling can be used to bridge gaps between different scales of the theoretical description.

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Ab initio calculation of the shift photocurrent by Wannier interpolation

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The bulk photovoltaic effect (BPVE), also known as the shift-current effect, is a nonlinear optical response that yields a net photocurrent in noncentrosymmetric crystals. Although the theory of the BPVE is well established, its practical implementation is challenging because it involves a subtle Berry-phase-like quantity [1]. Here we calculate the BPVE using a Wannier-interpolation scheme as shown in Ref. [2] and implemented in the context of the Wannier90 package [3]. Our method provides two main advantages over previous numerical schemes. First, it avoids summing over a complete set of virtual states, and hence it is free from truncation errors. Second, it allows for an efficient interpolation of the Brillouin-zone integrand onto a fine k -point mesh, providing high accuracy and conver- gence. We test the Wannier interpolation of the BPVE in benchmark systems including GaAs [1] and single-layer monochalcogenide GeS [4]. We also show how our formalism relates to the tight-binding-based approach to the BPVE [5].

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Predicting the defect thermodynamic properties induced at low temperature in 2D TMDs

E. Igumbor and E B Lombardi

Density functional theory with the aid of Quantum espresso was used to modelled the electronic, thermal and optical properties of defect levels induced in a monolayer MoS2, CrS2 and WS2 2D TMDs. We have studied the most stable non-interacting and complex defect (VS and VTM) of the listed 2D TMDs. While for the WS2 and MS2, the S vacancy is the most energetically favorable for the non-interacting defect, the double S vacancies under equilibrium conditions is the most energetically favorable for the complex defects. Our results shown that the double vacancies of S and TMD are always stable and dissociation can only occur at the expense of higher energy. Defect levels where induced in the band gap of the 2D TMDs as the Fermi energy is varied from the valence band maximum to the conduction band minimum.

Thermal conductivity in disordered systems

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In disordered systems the typical phonon mean free paths are so short that the quasi-particle picture of heat carriers breaks down and the kinetic theory of thermal transport mathematically embodied in the phonon Boltzmann transport equation (BTE) is no longer applicable.

In this work we show that by introducing a quasi-harmonic description of a solid, both crystalline and disordered, where normal modes are characterized by harmonic frequencies and finite lifetimes, the Green-Kubo expression for the heat conductivity can be cast in a general form that comprises both the BTE expression in the mode-resolved relaxation-time approximation for crystals and the Allen-Feldman model for heat transport in disordered system. In the latter case, the normal-mode inverse lifetimes provide physically motivated values for the broadening factor that was introduced by Allen and Feldman as a mathematical device to turn a discrete spectrum into a continuous one in a purely harmonic framework, which does not apply to crystals.

Our approach is applied to crystalline and amorphous silicon using harmonic normal-mode frequencies and lifetimes computed using the classical limit of the Fermi's golden rule, and thoroughly validated by comparing its predictions with those of standard equilibrium molecular-dynamics simulation.

Berry-phase-mediated Thermoelectric Effects in Magnetic Materials

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Electron spins can act as interconverter of electricity, light, sound vibration, and heat [1]. Here we are interested in making high thermoelectric conversion efficiency by effectively manipulating electron-spin degree of freedom. One way to achieve that is to control Berry-phase-mediated thermoelectric effects, namely the contribution of the anomalous Hall conductivity (AHC) to thermoelectric power. Our target is the anomalous Nernst effect (ANE), which is a heat-to-electricity conversion observed in magnetic materials and directly related to AHC. We discussed AHC mainly driven by an effective magnetic fields inside materials, Berry curvature, induced by spin-orbit coupling and/or spin chirality. The magnitude of ANE originate in two mechainism as follows: (i) asymmetry of the AHC along the energy axis; (ii) product of Seebeck coefficient and Hall angle which is related to the ratio of off-diagonal to diagonal elements of electrical conductivity tensor.

In this presentation, we will present our recent first-principles computations [2] on topological magnet (skyrmion crystals [3, 4] and meron crystals), and half-Heusler [5] and related compounds. We found that the effective magnetic fields indeed generates large ANE. This behavior was clearly understood by the chemical potential dependence of AHC. Based on the gained knowledge, we will discuss how to enhance such Berry-phase-mediated thermoelectric effects. We will also discuss thermoelectric figure of merit ZT by evaluating thermal conductivity and power factor in magnetic materials.

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First-principles ultrafast charge carrier dynamics at the hybrid F4TCNQ:H-Si(111) interface

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Organic donor/acceptor molecules adsorbed on inorganic semiconductors have been used to tune the electronic properties of the latter in a controlled way [1]. Upon interaction with light, the resulting hybrid inorganic/organic systems exhibit unique optical properties, typically dominated by charge-transfer excitations [2]. Here, we consider an interface formed by an strong electron accepting molecule 2,3,5,6-Tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4TCNQ), adsorbed on the hydrogenated Si(111) surface and we investigate the charge-carrier dynamics induced by an ultrafast laser pulse. The linear- absorption spectrum of the system exhibits two maxima in the visible region corresponding to transitions between electronic states hybridized across the inorganic and components. In the framework of RT-TDDFT, as implemented in the octopus code [3], we set the laser frequency in resonance with the energy of these excitations and follow the resulting charge-carrier dynamics. Our results offer relevant insight to understand the earliest-stage formation of the optical excitations in hybrid inorganic/organic interfaces.

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Ordering phenomena in graphene like C-B-N 2D alloys -

Monte Carlo studies

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Two-dimensional materials and their alloys remain to be under interest of scien- tists, due to their very appealing and possibly easily tunable properties. Graphene-like $C_{1-x-y}B_xN_y$ layered alloys constitute intriguing class of materials, including C-doped hBN, B- and/or N- doped graphene, mixtures of graphene and hBN, or CBN structures with long-range order. Many reports on synthesis of such alloys are available, as well as numerous theoretical predictions for, mostly quasi-ordered configurations of constituent C, B, N atoms. However, the deep understanding of the equilibrium morphology, stabil- ity, and phase diagrams of these interesting two-dimensional compounds is still lacking. These issues are addressed in the present theoretical computational research in the frame- work of Monte Carlo simulations with the valence force field like potentials to account for the interaction between atoms. In particular, we search for short- and long range ordering phenomena, and further on try to determine the influence of these effects on the electronic structure and further materials properties. We employ empirical, bond- order Tersoff potentials enabling calculation of energy of systems containing thousands of atoms and perform Monte Carlo simulations to find most energetically favorable distri- bution of atoms [1]. Simultaneously optimization of the systems geometry is performed. This approach avoids artificial periodicity that is often introduced in ab initio simula- tions performed with supercells of very limited size. We find that all investigated systems deviate strongly from random alloys, exhibit short-range ordering, and have tendency to form carbon (graphene) and h-BN domains. Moreover, we investigate the impact of edges and various structural defects, such as vacancies, 57 Stone-Wales defects, and grain boundaries, on distribution of constituent atoms, and observe local changes in ordering in the area of abovementioned imperfections in honeycomb lattice. Furthermore, we ex- tend our research by using more accurate reactive charge optimized many body potentials.

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Doping Effects on the Migration of Oxygen Vacancies in Lanthanum Strontium Manganite Compounds

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Complex manganese oxides have been the subject of intense study for decades due to the discovery of both intrinsic and extrinsic properties such as colossal magnetoresistance, ferroelectricity, and metal-insulator transitions, among others. They are all modulated by factors such as the amount of doping, structural strain, crystal defects and the presence of surfaces and interfaces. This variety of properties promises several technological applications for manganites. Recently, this material has been extensively used as cathode in solid oxide fuel cells (SOFCs), a clean energy generation technology which takes advantage of particular characteristics regarding the chemical and structural stability, ionic-electronic conductivity and catalytic activity of manganites.

In this work, we present a study about the diffusion of oxygen vacancies, focusing on the role of Sr-doping in the formation energies and migration barriers in $La_{(1-x)}Sr_xMnO_{3-\delta}$ (LSMO) compounds by means of the Density Functional Theory (DFT) and the Nudged Elastic Band (NEB) method. The distribution of Sr ions within the crystal structure is obtained using the Special Quasirandom Structures (SQS) approach, this allow us to explore Sr-rich and Sr-poor environments. We focus on a composition range of $0.20 < x < 0.50$ which includes the doping concentrations most frequently used in SOFCs.

We find that the effect of local and global doping influence both the electronic bandstructure and the oxygen vacancy formation energies leading to two distinct regimes, displaying either a deep defect state or a shallow band-like state close the conduction band. Regarding the oxygen ion transport, the diffusion is enhanced at the surface mainly for LaSrO terminal planes. These results contribute to optimize the LSMO cathode performance operating at lower temperatures.

In silico screening of experimental structural repositories for new ionic conductors

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A holistic screening of structural databases for ionic conductors by means of atomistic simulations can lead to new candidates for next-generation solid-state lithium-ion batteries and deepen our understanding of the underlying processes governing ionic diffusion in the solid state. The task is ambitious because no known modeling technique is predictive for a large number of systems at reasonable computational cost. To model ionic diffusion more efficiently without substantial loss of accuracy or generality, we apply physically motivated approximations to the Hamiltonian of Kohn-Sham density functional theory, arriving at a novel hybrid quantum/empirical model that can be used in molecular dynamics for solid- state diffusion [1]. The model is the key to a high-throughput screening approach relying on the AiiDA materials informatics [2] platform. We will present the different screening stages and show how high-level workflows can be used to automatize and optimize the calculation of transport coefficient. We can show first results of the screening, including first experimental evidence of the ionic conductors discovered.

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Electron-phonon coupling in semiconductors within the GW approximation

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The magnitude of the renormalization of the band gaps due to zero-point motions of the lattice is calculated for 18 semiconductors, including diamond and silicon. This particular collection of semiconductors constitute a wide range of band gaps and atomic masses. The renormalized electronic structures are obtained using stochastic methods to sample the displacement related to the vibrations in the lattice. Specifically, a recently developed one-shot method is utilized where only a single calculation is needed to get similar results as the one obtained by standard Monte Carlo sampling. In addition, a fast real-space GW method is employed and the effects of G_0W_0 corrections on the renormalization are also investigated. We find that the band-gap renormalizations inversely depend on the mass of the constituting ions, and that for the majority of investigated compounds the G_0W_0 corrections to the renormalization are very small and thus not significant.

Benchmark of density functional theory for superconductors in elemental materials

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The first-principle calculation of the superconducting properties such as the transition temperature (T_C) and the gap function is of great interest to explore new materials as well as to understand the mechanism of known superconductor. Density functional theory for superconductors (SCDFT) [1] is one of the framework for such calculations; this method enables us to perform fully non-empirical simulation in the superconducting phase within reasonable computational cost. In this method, we can treat the electron-phonon interac- tion, the electron-electron repulsion, and the spin-fluctuation mediated interaction [2] in the first-principles manner. However, the accuracy of the current approximated functional of SCDFT and the effects of the spin-orbit interaction (SOI), spin-fluctuation, and the phononic anharmonicity has not been verified systematically although such a verification is highly desirable before we apply this method to the wide range of materials. Such a benchmark is also useful to find the guideline for improving the superconducting density functional. For this purpose, we started the benchmark calculation of SCDFT with our newly developed first-principles program package Superconducting-Toolkit (SCTK) [3]. In this presentation, we show benchmark results of superconducting properties calculated by SCDFT for 32 elemental materials together with computational details, and discuss accuracy of the predicted T_C and the effect of the spin-orbit interaction up on T_C.

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The role of surface defects in the methanol synthesis pathway on Cu-type catalysts: DFT and kMC study

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The nature of the reaction sites on surfaces is an open question of the current theoretical modelling studies of catalytic processes [1, 2]. Complex reaction pathways, such as methanol synthesis from CO_2 hydrogenation, are particularly sensitive to the geometry of active sites, their chemical structure, and the size of the interaction area [3, 4]. Properties of the surface reaction sites were studied on Cu(533) surface exhibiting a step representing a surface defect. First-principles multiscale modelling using ab initio density functional theory (DFT) and mesoscopic kinetic Monte Carlo (kMC) simulations is employed to probe the atomistic-scale quantum chemistry and meso-scale kinetics of the carbon dioxide activation for methanol synthesis. We focus on how the surface defects affect the methanol synthesis pathway, the catalyst selectivity and activity. The theoretical study and comparison of the same pathway on Cu(111) surface show the advantages of surface defects and their catalytic role in the methanol production. Stepped Cu(533) surface enhances selectivity and activity of the methanol synthesis, with selectivity up to $\sim 80\%$ and turn-over frequency (TOF) up to $10^{-4}\ s^{-1}$, which is over 4 orders of magnitude better as for Cu(111) surface. It is found that the surface defect, compared to Cu(111) surface and other Cu/metal oxide alloys from the literature [5, 6], triggers the H_2COH hydrogenation reaction, resulting in higher total methanol yield.

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Ab initio modelling of solvated Mg^{2+} at Mg electrode

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Understanding the nature of electrochemical electrode/electrolyte interfaces is a crucial step in the optimization of systems for energy conversion and storage, as the interface properties and changing electrode potential govern the electrochemical reactions. However, there is no generally accepted method to consider varying electrode potentials in first-principles calculations, and modelling interfaces remains one of the biggest challenges for the density functional theory (DFT) community.

It has been shown on lithium system that the use of implicit solvent model implemented in some DFT codes [1, 2] together with the inclusion of explicit solvent molecules reproduces the results that are in agreement with experimental values [3]. To confirm the validity and efficiency of the method on divalent systems, we study Mg^{2+} solvated in monoglyme (DME) and in ethylene carbonate (EC).

First we determine the explicit first solvation shell of Mg^{2+} in DME and EC using molecular DFT (Gaussian). In both solvents the implicit solvent model was used to model the surroundings of the solvated Mg^{2+} complex. The results are then used in plane wave DFT calculations (VASP). In both solvents we obtain the free electrochemical energies, equilibrium potentials and differential capacitance as a function of potential [4] in excellent agreement with experimental results. The method is further developed to test the stability of the solvents at the interface. We show that although free solvent molecules have a large potential stability, the stability is drastically modified in the presence of Mg^{2+}. The method presented thus allows reproduction of correct range of potential and provides insights into why some solvents are not appropriate for Mg battery systems despite their apparent stability.

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Fundamental Study of Hydrogen Storage in Ca Decorated MOF Designed with Graphyne Linker

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The increasing demand for energy and dwindling supply of presently available fossil fuels, their adverse effects of CO_2 emissions have raised the interest in sustainable, clean fuels such as hydrogen. But the storage of hydrogen for onboard application is a major problem in attaining this goal.[1]

In this study, metal-organic framework (MOF) designed with graphyne linker decorated with Ca metal and explored their hydrogen storage properties.[2] Applying density functional theory employing the GGA-PBE functional with double numeric basis method, the structural stability, and physicochemical properties have been analyzed. It is found that each graphyne linker strand can bind two Ca atoms from across the linker by Dewar coordination. On saturation with hydrogen, each Ca adsorb 6 H_2 molecules in molecular form in the Ca decorated MOF. The H_2 and metal interact by Kubas-Niu mechanism with average H-H bond length elongated from free H_2 bond length of 0.741 Å, which reveal that all the adsorbed H_2 molecules are physisorbed in metal decorated MOF. The average H_2 adsorption energy and sequential desorption energy is found in the range between 0.2 to 0.6 eV which indicates the fast kinetics under moderate temperature and pressure conditions required by the US DOE. The hydrogen storage capacity is found to be 7.9 wt. % for hydrogen saturated Ca decorated MOF with graphyne. The energetics and storage capacity meet the ultimate DOE target 2020 which makes them sustainable hydrogen storage material.

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Inherent Ambiguity of Pressure-Induced Structural Phase Transformations in ReO_3

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We revisit the question of pressure-induced structural phase transitions in ReO_3 (cubic primitive structure, Pm-3m,Z = 1), which, so far, has been rather controversial. Previously reported phases observed between 0.5 GPa and 12 GPa include tetragonal (P4/mbm,Z =2),cubicscutterudite(Im-3,Z =8),andmonoclinicMnF_3-type(C2/c,Z = 12); beyond 12 GPa a rhombohedral VF_3-type structure (R-3c,Z = 6) is adopted. We performed the DFT calculations (LDA, plane waves, PAW, vasp) of energies, pressures, enthalpies, and vibrational frequencies, at various volumes, for the structures suggested by experiment, leaving provisorily aside only the monoclinic MnF_3 type structure (48 atoms, 20 parameters to optimize). The conspicuous feature which differentiates ReO_3 from other solids is in the relative positions of the $E(V)$-equations of state (EOS). While the usual EOS map consists of shifted and intersecting $E(V)$'s, the four EOS in ReO_3 nestle around the common energy minimum $E(V_0)$: at volumes close to V_0 the $E(V)$'s of all four phases are degenerate (undistinguishable within the accuracy of the DFT calculations). – In the hindsight this behavior should not surprise: all four can be viewed as (deformed) superstructures deriving from the same cubic phase. – As a consequence, also the $P(V)$ equations of state and the enthalpy variations $H(P)$ are degenerate, up to $P \approx 3$ GPa. That means that upon compressing the simple cubic structure the system has (at $T = 0$) a “free choice” between 3 continuous paths leading to denser phases: its preference for one or another structure is not dictated by the enthalpies.

This fairly unusual behavior explains the unexpected ambiguity observed in the x-ray diffraction experiments (Ref. [1], pp. 58, 60): a second-order transition Pm-3m → P4/mbm was established on one sample, while Pm-3m → (a fleeting P4/mbm) → Im-3 observed on another sample. Upon further compression a first-order transformation brings the system to the R-3c structure on both samples (either straight from Im-3 or via an intermediate monoclinic phase C2/c). The existence of two different pathways is further supported by Raman experiments on the same samples (Ref. [1], p. 65). The ambivalence in the system's choice of structure explains the apparent controversies encountered in the literature. The existence of different and competing second-order transition paths eluding the control by enthalpies makes the structural evolution of ReO_3 extremely sensitive to other 'external' factors – which are presently neither clearly identified nor understood: the detailed crystal nature of the samples (e.g. powder versus single crystals), possibly energy barriers between the structures. In absence of a better understanding we refer to them by the all-encompassing term “sample dependence”. This peculiar behavior appears to be an inherent and farther reaching property of ReO_3: we notice that in a completely different context, viz. in studies of anomalous thermal expansion, a “sample dependence” of $\alpha(T)$ was witnessed as well [2] – without, in that context, any explanation on hand.

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DFT + GW Study of Charge Transition Levels of Point Defects in Monolayer and Bilayer Phosphorene

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Phosphorene has emerged as an important material in the family of two-dimensional layered materials with several potential applications including field effect transistors, photo- voltaic p-n junctions etc. This is due to the fact that it not only possesses a direct band gap ranging from 0.3–2.1 eV (depending on the number of layers) but also a high carrier mobility (1000 cm^2/Vs for the holes). Native point defects [1, 2] can induce states within the electronic band gap of phosphorene and influence its electrical, optical and magnetic properties. The puckered structure of phosphorene accommodates different kinds of na- tive point defects. We have studied the stability and electronic properties of vacancy and self-interstitial defects in mono- and bilayer phosphorene. We calculate the forma- tion energies and charge transition levels of these defects using the combined formalism of density functional theory and GW. Our calculations show that these defects have low formation energies making them easy to form. Furthermore, phosphorus vacancy shows acceptor type behaviour which could explain the p-type conductivity in phosphorene. On the other hand, interstitial defect can act as both acceptor and donor type defect.

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Computation of Tautomeric Equilibria in Anthocyanins

Sara Laporte – SISSA, Trieste October 21, 2018

We present a method to compute equilibrium constants between prototropic tautomers (i.e. two molecules differing from each other only in the position of a proton). In water at ambient conditions, entropic effects need to be taken into account, hence the need for accurate free energy calculations involving explicit solvent. We use thermodynamic integration in ab-initio molecular dynamics, with an ’alchemical’ integration parameter enabling us to gradually switch the proton from one site to another in a series of parallel simulations. The integral of the energy with respect to the parameter is computed from its analytical form on-the-fly, removing the need for obtaining numerical derivatives, thus improving accuracy. The method is applied to cyanin-3-glucoside (C3G), a common natural dye found in many flowers, fruits and vegetables. C3G is also a pH indicator, varying in colour due to gradual deprotonation with increasing pH. The order in which various sites lose a proton is not completely agreed upon in the literature, and it is likely that at neutral to high pH, several tautomers may coexist in solution. The case of 4’ vs 7 deprotonation in C3G is simulated within our framework, and convergence for the difference in free energy of the two forms was obtained in as little as 4 ps after equilibration. This method could be further applied to other relevant systems such as nucleic acid bases, in which this type of proton transfer reaction has been linked to the appearance of DNA and RNA mutations.

Prediction of MGa_3 phases (M= transition metals)

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The discovery of new crystal structures by numerical simulations is a significant challenge in materials science. Indeed, the exercise is far from being simple : How to reach the lowest stable structure as fast as possible among the millions of possible structures knowing only the chemical composition? An effective and strong method, USPEX, tries to answer this problem. It is based on the concepts of the Darwinian theory of the evolution. In the study, we look at the structural and electronic properties of MGa_3 phases with M = Cr, Mo,W, Fe, and Ir, which could potentially develop interesting thermoelectric properties. Our USPEX study confirms that the arrangement of type $FeGa_3$ for group 6 metals is thermodynamically stable and semi-conductor. Our results have uncovered a new structural phase for $IrGa_3$ (orthorombic Pnma, n°62) instead of the proposed $FeGa_3$-type structure (quadratic $P4_2/mnm$, n°136). Its dynamical stability has been verified by phonon calculations. This thermodynamically stable $IrGa_3$ phase is metallic, and therefore does not present thermoelectric properties.

First-principles study of dielectric permittivity in $(C_4H_9NH_3)_2PbI_4$ using atomic basis functions

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To study a large-scale system by first-principles calculations, the atomic basis functions are usually adopted to reduce the number of basis functions in comparison with, for example, a plane-wave basis. To further access the excitation properties of such a system, the corresponding tight-binding representation can be utilized since the tight-binding Hamiltonian is straightforwardly obtained after finishing the self-consistent calculation. In the poster, we will present how to calculate the optical conductivity and the Kohn-Sham response function using the tight-binding representation. For the momentum matrix elements needed to deliver the conductivity in the Kubo-Greenwood formula, the position matrix elements are required[1]. For calculating the Kohn-Sham response function, the formula is similar to the conventional one. Both formulae will be given in detail. For a case study, we will illustrate the dielectric permittivities of $(C_4H_9NH_3)_2PbI_4$, which has been shown to exhibit hyperbolic dispersion experimentally[2], and discuss how to improve the result to approach better agreement with experiments.

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Study of Oxygen adsorption on Co(0001) and Co/Ir(111) surfaces

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In recent years great interest arose for graphene (Gr) layers grown on non-commensurate transition metal (TM) substrates leading to a moir´e patterning, to be further used as tem- plates for the adsorption of e.g. magnetic molecules [1, 2]. The presence of the graphene interlayer can then result in a partial molecule-substrate decoupling at the electronic level, while maintaining the magnetic coupling. A practical realization of this structure has been achieved recently by adsorbing TM-phthalocyanine molecules on Gr/Co/Ir [1]. Depending on how many Co layers are intercalated on Gr/Iridium [1, 2], Gr can be lat- tice matched (multi Co layers) or not (eg when only one Co layer is intercalated it is arranged morphologically to the Ir surface lattice) [2]. The magnetic configuration of Co intercalated under graphene can be tuned as a function of Co thickness [2]. Oxygen can play a role similar to Gr for the decoupling. It has been shown experimentally that these systems can adsorb oxygen stably under Gr, as a way to control Co magnetization, but the mechanisms and structural details of the adsorbed oxygen are unclear [3, 4].

Computationally, the adsorption of O on thick Co (0001) films has been studied in Ref. [5]. In this work we perform total energy calculations within density functional theory at PBE level to compare the adsorption of O on Co (0001) and Co/Ir(111) with a one monolayer of Co. We have studied a number of different adsorption configurations for O, with coverage ranging from 0.25 and 0.5ML, and including inequivalent O atoms in the unit cell. Our results show that, for all configurations, the strained Co/Ir system absorbs oxygen with binding energies larger (0.25-0.51 eV) than those obtained for O@Co(0001) thick film. Our results can be used to support the interpretation of experimental data concerning the kinetics of Oxygen intercalation and diffusion.

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Elastocaloric Effect in Carbon Nanotubes and Graphene from first principles

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Carbon nanotubes are famous for their many extraordinary properties. We use a thermodynamical approach, experimental data from the literature, and atomistic simulations to reveal one more remarkable property of the carbon nanotubes that has so far been overlooked. Namely, we predict the existence of very large elastocaloric effect that can reach up to 30 K under moderate loads. Potentially even larger values could be achieved under extreme loads, putting carbon nanotubes in the forefront of caloric materials. Other remarkable features of the elastocaloric effect in carbon nanotubes include linearity of elastocaloric temperature change in applied force (compressive or stretching), very weak dependence on the temperature, and an absence of hysteresis. Such features are extremely desirable for practical applications in cooling devices. Moreover, a similarly large elastocaloric effect is predicted for the graphene. The prediction of a large elastocaloric effect in carbon nanotubes and graphene sets forward an unconventional strategy of targeting materials with moderate caloric responses but the ability to withstand very large loads.

Beyond the quasiparticle approximation: Fully self-consistent GW calculations

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We present quasiparticle (QP) energies from fully self-consistent GW (scGW) calculations for a set of prototypical semiconductors and insulators within the framework of the projector-augmented wave methodology [1]. This is based on our previous cubic-scaling GW code [2], but goes beyond the quasiparticle approximation. To obtain converged results, both finite basis-set corrections and k-point corrections are included, and a simple procedure is suggested to deal with the singularity of the Coulomb kernel in the long-wavelength limit, the so called head correction. It is shown that the inclusion of the head corrections in the scGW calculations is critical to obtain accurate QP energies with a reasonable k-point set. We first validate our implementation by presenting detailed results for the selected case of diamond, and then we discuss the converged QP ener- gies, in particular the band gaps, for a set of gapped compounds and compare them to single-shot G_0W_0, QP self-consistent GW, and previously available scGW results as well as experimental results.

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Thermosalient (”jumping”) crystals

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Thermosalient materials when heated or cooled are characterized by a fast and energetic phase transition in which crystals experience macroscopic change in dimensions and jump to distances much larger than their own dimension. Since the first systematic study [1], there has been a rising intrest for these interesting, and potentially useful, acutating materials. As a one example, N-2-propylidene-4-hydroxybenzohydrazide shows irreversible thermosalient phase transition from Form I to Form II and reversible thermosalient phase transition from Form II to Form III. We combine experiments and first-principles electronic structure calculations to elucidate this interesting and technologically useful phenomenon [2].

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POSTER. SESSION II

THURSDAY, 10 JANUARY 2019

GROUP M - Z

Temperature dependent atomic B-factors: an ab-initio calculation

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The Debye Waller factor describes the effect of temperature in X-ray or neutron diffrac- tion pattern intensities. It is defined in terms of the B-matrix: its elements $B_{\alpha\beta}$ are mean square atomic displacemens in different directions. These quantities are important to describe the effects of temperature and quantum fluctuations and have been used in sev- eral contexts. We compute B-factors ($8\pi^2 B_{\alpha\beta}$) for different elemental crystals: silicon, ruthenium, magnesium, and cadmium. We compute them both by direct summation in the Brillouin zone or using generalized phonon densities of states. Both methods have been implemented in the thermo pw software [1]: a driver of Quantum ESPRESSO routines that calculates thermodynamic properties of materials. We compare the predictions of the harmonic and quasi-harmonic theories with the available experimental data.

[1] See https://dalcorso.github.io/thermo pw/.

Computational study of complexes of antioxidant acylphloroglucinols with a Cu^{2+} ion. An overview of results

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Acylphloroglucinols [1] are a large class of compounds structurally derivative from 1,3,5- trihydroxybenzene and characterised by the presence of a COR group. Many of them are of natural origin and possess a variety of biological activities. Nearly all of them are characterised by the presence of an intramolecular hydrogen bond (IHB) between the sp^2 O of COR and a neighbouring phenol OH. Several of them contain substituents with groups that can form additional O–H···O IHBs or O–H···π IHBs.

Complexes with a Cu^{2+} ion have been calculated for selected acylphloroglucinols having antioxidant activity, and also for some others structurally-related molecules, to evaluate their ability to coordinate and reduce the Cu^{2+} ion [2, 3], which is also relevant to the antioxidant activity [4, 5]. The complexation entails the transfer of an electron from the molecule to the ion. Calculations were performed at the DFT level with the B3LYP functional and the 6-31+G(d,p) basis set for the C, O and H atoms and the LANL2DZ pseudopotential for the Cu^{2+} ion. All the sites to which the ion can bind were considered for each molecule. The results show that the ion prefers to simultaneously bind to two or three geometrically-suitable sites. When it binds to only one site, it prefers sp^2 O atoms or π bonds. The complexation causes changes in the parameters of the IHBs (above all in the bond lengths) and in their red shifts, and, for O–H···O IHBs, may also cause the transfer of the proton to the other O atom. The changes depend on the site/s to which the Cu^{2+} ion binds.

The presentation offers an overview of the effects of complexation on the IHBs present in these molecules, highlighting similarities and differences in relation to the characteristics of each molecule, to the type of IHBs present (O–H···O, O–H···π) and to the binding site of the ion. Spin density maps are included to highlight the distribution of the unpaired electron in the molecular ion within the complex.

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Towards the description of hybrid organic-inorganic Nanosystems: simulating dye-metal interactions and optical properties of the combined system.

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Hybrid organic-inorganic Nanoparticles (HNPs) are very interesting and widely studied materials, for their versatile applications in biotechnology and medicine, with high potential in biomedical imaging, gene and drug delivery, and photothermal cancer therapy [1, 2], making them one of the most promising materials for early and accurate cancer diagnosis and effective cancer therapy. However, computing their physico-chemical properties in details proves to be a challenge. While the nature of the organic component of the HNPs necessitates a full quantum chemical treatment, the size of the inorganic component renders this treatment computationally too expensive to be assessed with an homogeneous technique.

For this reason hybrid models have been developed combining a QM level treatment and a classical electromagnetism approach, respectively, for molecules and the inorganic nanostructures upon which they are adsorbed [3]. In particular, the inorganic component, usually a metal, is considered as a continuous body, characterized by its own frequency dependent dielectric function (Polarizable Continuum Model, PCM), while excitation energies due to the energy transfer from the molecule to the metal is evaluated exploiting Time Dependent Density Functional Theory (TDDFT).

After proving that the polarization charges distribution, introduced by PCM, well describe the optical properties of bare inorganic Nanoparticles [4], reproducing experimental spectra [5, 6] of bare gold Nanoparticles using PCM tools, we moved towards the next step: describe the interactions of the organic molecular frame with the Nanoparticles.

In order to reproduce the desired spectroscopical properties for the hybrid system we evaluated on one side the effects introduced by the presence of a Nanoparticle over the energies and the excited state dynamics of the isolated organic dyes (radiative and non radiative deactivation constants), correcting the TDDFT results with the electrodynamic effects that cannot be seen with a simple electrostatic PCM. On the other side we computed how the organic layer of dye and surfactant around the gold Nanoparticle affects the plasmonic excitation of the metallic subsystem adding this layer to the electrodynamic computations.

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First principles studies of XAS/XMCD experiments in 2D heterostructures under steady-state non-equilibrium conditions

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We report on progress in the field of ab initio theoretical spectroscopy for out-of-equilibrium scenarios. This regime can be approached through either pump-probe experimental se- tups, or through the application of a steady-state, time independent but finite external perturbation.

In both cases additional aspects of Green function theory become relevant for a proper description of the physics, and new challenges must be overcome for practical first-principles calculations for realistic materials.

We consider here the case of XAS/XMCD investigations in multi-layer compounds subjected to a finite electric field [1]. These techniques allow for high selectivity in the chemical element and even local environment being targeted. In combination with various kinds of theoretical sum rules, this can lead to fundamental insight on basic electronic structure features of a material.

Our computational framework is based on a fully relativistic multiple scattering (Korringa, Kohn and Rostoker) Green function solution scheme for the basic SCF-DFT problem, and the simulation of various kind of spectroscopy and transport experiments [3]. Inspired by recent investigations that probe the appearance of voltage-controlled, transient magnetic moments [1, 6] on top of static proximity effects in a non-magnetic (NM) element over a magnetic substrate, we put to the test a combination of recent developments for the non-equilibrium extension of this basic formalism [4, 5] with established procedures for the study of x-ray absorption [2] in bulks or 2D heterostructures [7].

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Prediction of a large-gap and switchable Kane-Mele quantum spin Hall insulator from first-principles simulations

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Fundamental research and technological applications of topological insulators are hindered by the rarity of materials exhibiting a robust topologically non-trivial phase, especially in two dimensions. Here, by means of extensive first-principles calculations, we pro- pose a novel quantum spin Hall insulator (QSHI) with a sizeable band gap of $\sim$0.5 eV that is a monolayer of jacutingaite [1, 2], a naturally occurring layered mineral first discovered in 2008 in Brazil [3] and recently synthesised [4]. This system realises the paradigmatic Kane-Mele model [5, 6] for QSHIs in a potentially exfoliable two-dimensional monolayer, with helical edge states that are robust and that can be manipulated exploiting a unique strong interplay between spin-orbit coupling, crystal-symmetry breaking and dielectric response [1]. Finally, we give an update on ongoing experimental efforts in the synthesis and characterisation of bulk and monolayer jacutingaite.

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Oxide- and sulfide-based electrolytes for Li-ion All-Solid-State batteries: a First-Principles Comparative Study of $Li_{10}GeP_2O_{12}$ and $Li_{10}GeP_2S_{12}$ in the LISICON and LGPS phases

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Although high-energy Li-ion batteries are desirable for green energy storage and CO_2 abatement, issues with their safety and non-toxicity are nowadays crucial, and replacing liquid organic with solid state electrolytes would be an effective way to circumvent them. Among the families of known crystalline solid state conductors, thio-LISICON materials, especially in the recently discovered tetragonal phase (called LGPS from $Li_{10}GeP_2S_{12}$) have been extensively studied in the last decades [1, 2]. However, sulfide materials have narrow electrochemical windows and raise toxicity and safety problems that make them unlikely to pursue the environmental goals of Li-ion batteries. In this respect it is appealing to investigate the analogous oxide materials, starting from the prototype $Li_{10}GeP_2O_{12}$ (LGPO) [4]. In this work we address Li diffusion in LGPO and LGPS through Car- Parrinello molecular dynamics through the QUANTUM ESPRESSO package [3], with extra care in achieving a correct thermalization of the heavy species and the light Li ions. Both the (quasi)-orthorhombic (LISICON) [1, 5] and tetragonal (LGPS) [2, 4] phases reported in the literature are studied for the oxide and for the sulfide materials, with the aim of comparing their stability and diffusion with one and only method. The simulations feature extended thermostatted thermalization followed by microcanonical evolution. NPT variable cell dynamics simulations within the Parrinello-Rahman scheme are also performed at a representative temperature to complete exploration of the configurational space, since in these superionic materials there is not a corresponding unique ground state structure. On the basis of the reported theoretical evidences, a thorough analysis of the structure and conducting performance for the oxide and the sulfide is presented.

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Band unfolding made simple

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We present a simple method to unfold the energy bands of supercell calculations. We introduce the "fully unfolded bands" as the local density of states in reciprocal space. This provides an intuitive approach, valid not only for crystals, but also for non periodic systems, like surfaces and liquids. We then refold these fully unfolded bands into the first Brillouin zone of the perfect crystal, thus obtaining the conventional unfolded bands. We have implemented this method in the Siesta package and we explore some of its potential applications by applying it to defective silicon crystals and surfaces, as well as to amorphous silicon.

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Thermodynamic and Kinetic Limitations for Peroxide and Superoxide Formation in Na−O2 Batteries

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ABSTRACT

The Na−O2 system holds great potential as a low-cost, high-energy-density battery, but under normal operating conditions, the discharge is limited to sodium superoxide (NaO2), whereas the high-capacity peroxide state (Na2O2) remains elusive. Here, we apply density functional theory calculations with an improved error-correction scheme to determine equilibrium potentials and free energies as a function of temperature for the different phases of NaO2 and Na2O2, identifying NaO 2 as the thermodynamically preferred discharge product up to ∼120 K, after which Na2O2 is thermodynamically preferred. We also investigate the reaction mechanisms and resulting electrochemical overpotentials on stepped surfaces of the NaO2 and Na2O2 systems, showing low overpotentials for NaO2 formation (ηdis = 0.14 V) and depletion (ηcha = 0.19 V), whereas the overpotentials for Na2O2 formation (ηdis = 0.69 V) and depletion (ηcha = 0.68 V) are found to be prohibitively high. These findings are in good agreement with experimental data on the thermodynamic properties of the NaxO2 species and provide a kinetic explanation for why NaO2 is the main discharge product in Na−O2 batteries under normal operating conditions.

This work has recently published in the Journal of Physical Chemistry Letters, Vol. 9, 2018, 4413- 4419.

Towards realistic DFT predictions of materials at high pressure

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Stimulated by recent state-of-art high pressure experiments, carried out the Advanced Photon Source by collaborators, we focused our research on improving the reality of first principles Crystal Structure Predictions.

The first problem to overcome, is the dependence of the calculated equations of state (EOS) on the choice of the exchange-correlation (XC) functional. To this end, we calculated the EOS of a common perovskite (SrRuO3) with the LDA, PBE, PBEsol and the recently developed, pseudo-hybrid ACBN0 functional [1]. In particular, the ACBN0 functional yields structural parameters at ambient pressure in very good agreement with experiments. Moreover, the band structures computed at the ACBN0 are in good agreement with more expensive, many body GW and DMFT calculations.

The second difficulty stands in the reliable prediction of crystal structures starting from the chemical composition alone, without further input from experiments. To this end we accepted the challenge of predicting the most stable structures of the Y-N system at 50 GPa. High pressure high temperature experiments, showed that metallic Y reacts spontaneously with nitrogen in a laser-heated diamond anvil cell (50 GPa and 2000 K) forming a series of unknown compounds of general formula YN_x. However, the precise refinement of atomic positions from the experimental powder X-Ray diffractograms is very difficult, and an educated ab-initio guess is mandatory.

To this end, we employed the evolutionary algorithms implemented in the USPEX code, coupled with the local-basis code SIESTA [2], in order to reduce the computational cost. We performed the USPEX search at very high pressure (100 GPa) to enhance the “variety” of the structures, and we performed both constant- and variable-composition calculations. Finally, we relaxed the most stable structures from 100 GPa down to ambient pressure, using the plane-wave code Quantum Espresso [3] and determined the most stable polymorphs of YN_2 and YN_3. We found that the yttrium cation is always trivalent and as a consequence the di-nitride system is metallic and shows a charge disproportionation. On the contrary, the $[N_3]^{3-}$ moiety is well described by a pair of resonating Lewis structure, and the system is a band-gap insulator.

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Strong influence of ligand fields to the exchange in Cu^{2+} spin-1/2 systems

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The influence of the structural features like bond angles and distances on the ex- change integrals in cuprate materials has been studied in detail for many years. One of the dominating structural features is the Cu-O-Cu bond angle in chain-like compounds [1, 2]. However, similarly strong influence on the exchange couplings by side groups was observed recently [3, 4]. As an example, we demonstrate the strong influence of hydrogen positions in the crystal structure for O-H groups or crystal water on the leading exchange terms, changing from substantial antiferromagnetic to moderately ferromagnetic. We elucidate the underlying microscopic mechanism based on detailed DFT studies and subsequently derived multi band tight binding models. The results widely improve our understanding of magneto-structural correlations in cuprate compounds.

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How does anharmonicity affect crystal structures? Theory and application to ice and high-pressure phases of hydrogen

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Abstract

Thanks to the new developments recently introduced in the Self-Consistent Har- monic Approximation (SCHA) I will show how it is possible to fully consider the effect of quantum fluctuations and anharmonicity in ab-initio simulation[1]. The total computational effort, even for systems with large cells, is lower than classical ab-initio molecular dynamics. Thanks to the link with the Harmonic theory, and the short-range of anharmonic interactions, it is possible to converge the results in much smaller super-cells than those required by molecular dynamics. A new way of relaxing the unit cell is also introduced and presented. I will discuss these effects in ice and high-pressure phases of hydrogen.

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Thermodynamic properties, Dynamical and Mechanical Stabilities of $ZnSe_2$ and $ZnTe_2$ in pyrite Structure: DFT Study

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Abstract

The present work is the first quantitative theoretical prediction of elastic, dynamical and thermodynamic properties of Zinc diselenide, ZnSe2 and Zinc ditelluride, ZnTe2 based on density functional theory (DFT) plane-wave pseudopotential method. The first-principles calculations were carried out using local density approximation, (LDA) and generalized gradient approximation, (GGA) in pyrite-type structure. The structural properties obtained were compared with existing experimental and theoretical results with related approach and were found to be of reasonable agreement. The elastic constants were calculated by adopting the effective stress-strain approach and thus, the mechanical stability of the compounds was established using the elastic stability criteria. Both compounds were predicted to be ductile with good plasticity according to the Pugh's and Poisson's ratios respectively. The dynamical stability was also determined from the calculations of phonon dispersion spectra. ZnSe2 is predicted to be dynamically stable while ZnTe2 stability is open for verification when experimental result is available, this is because LDA predicts dynamically stable compound while PBE proves otherwise.The thermodynamic properties between the temperature range of 0 to 1000K were calculated using the computed phonon density of state based on harmonic approximation. The result of the internal energies were seen to increase linearly with increasing temperature while the vibrational free energies decreases continually as temperature increases. A sharp increase were observed in specific heat capacity at constant volume between 0 and 400K while at higher temperature CV gradually approaches the asymptotic limit of 300 J.mol-1k-1. The values at ambient temperature were also recorded.

Keywords: Zinc dichalcogenides; Structural properties; Thermodynamic properties; Mechanical stability; Dynamical stability;

Chain Melting Simulations in Potassium

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ABSTRACT

Potassium under compression transforms, like typical simple metals, from BCC to FCC. Above 19 GPa this behaviour subsides, as consistent with other alkali metals, potassium forms a sequence of complex structures. The first of these, **K-III**, is a host-guest (HG) structure comprised of a **host lattice** and an **incommensurate guest lattice**. The guest lattice makes up a linear set of chains exhibiting long range order at low temperature. Upon heating, guest lattice diffraction peaks disappear. The inter- and/or intra-chain de-correlation has been dubbed **"chain melting"**.

Here, we simulate potassium's high-pressure and -temperature phase diagram by ***ab initio* molecular dynamics** (AIMD), using approximants for the incommensurate K-III phase. We then **train a forcefield** on the AIMD data set using **machine learning**. This forcefield can **simulate the entire phase diagram** of potassium up to 60 GPa in quantitative agreement with experiment, including chain melting and full melting. We discuss properties of the chain melted state.

2D Graphene/h-BN/Graphene Lateral Heterostructures for DNA Sequencing: A DFT Investigation

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A solid state device based on a heterostructure of graphene and 2D boron nitride has been proposed for the detection of different DNA nucleotides. The quantum tunneling current flows through the device has been calculated as the characteristic signal for Adenine, Cytosine, Guanine and Thymine. The h-BN insulating region contains a nanopore and is sandwiched between two graphene electrodes. DFT and NEGF formalisms are employed for the calculations. The translocation of nucleotides through the nanopore or on the surface of nanoribbon strongly changes the device current in a way that different nucleotides could be detected. We believe that the information obtained from the proposed devices could lead to a reliable sequencing of DNA nucleotides.

Large-scale DFT calculations on metallic systems using multi-site local orbitals in CONQUEST

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CONQUEST [1] is a linear-scaling (O(N)) DFT code developed jointly by UCL and NIMS. CONQUEST achieves O(N) by using the locality of density matrices with the den- sity matrix minimization method. Local orbitals which are called support functions are used to express the density matrices and Kohn-Sham orbitals. Our recent study shows that the code can employ DFT calculations on million-atom systems.

The computational cost scales cubically to the number of the support functions, both in the O(N) and the conventional diagonalization calculations. Therefore, to reduce the number of support functions without losing accuracy, we have introduced multi-site support functions (MSSF) [2]. MSSFs are the linear combinations of pseudo-atomic orbitals from a target atom and its neighbor atoms in a cutoff region. MSSFs correspond to local molecular orbitals so that the number of required support functions can be the minimal. The linear-combination coefficients are optimized numerically while the initial coefficients are determined by using the localized filter diagonalization method [3]. By using the MSSFs, we have succeeded in reducing the computational time dramatically [2].

The O(N) method is not guaranteed to be applicable for metallic systems. On the other hand, the multi-site method can be applied for metallic systems because the cutoff region in the method is only for the support functions, not for the wave functions of the whole systems. Therefore, we can treat large metallic systems by using the MSSFs even with the conventional diagonalization method. In the present study, we will demonstrate some applications of the multi-site method on the metallic systems containing several thousand atoms [4].

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Supercell effects on the electronic properties of graphene/TMDC’s heterostructures

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In this work we study the effect on the electronic properties related to the construction of supercells as a result of the mismatch between lattice parameters of the bilayer which form the heterostructure, in our case, graphene and one Transition Metal Dichalcogenide (TMDC). Recent works showed that once we put the graphene monolayer on top of another layer material, the electronic properties will depend on the cell used to perform the ab initio calculations, changing the position of its Dirac cone and the fermi energy, which in turns, will also change the transport properties. in this regard we try to address the inclusion of Spin Orbit Coupling (SOC) and propose the mechanisms related to the spin transport allowed by the features of these materials.

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Efficient O(N) divide-conquer method with localized natural orbitals

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An efficient O(N) divide-conquer (DC) method based on localized natural orbitals (LNOs) is presented for large-scale density functional theories (DFT) calculations of gapped and metallic systems [1]. The LNOs are non-iteratively calculated by a low-rank approxi- mation via a local eigendecomposition of a projection operator for the occupied space. Introducing LNOs to represent the long range region of a truncated cluster reduces the computational cost of the DC method while keeping computational accuracy. The novel O(N) method has been implemented into OpenMX DFT code which is based on opti- mized localized numerical orbitals and norm-conserving pseudopotentials [2]. A series of benchmark calculations on computational accuracy and parallel efficiency in a multilevel parallelization clearly demonstrate that the O(N) method enables us to perform large- scale simulations for a wide variety of materials including metals with sufficient accuracy in accordance with development of massively parallel computers.

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Theory and numerical simulation of the thermal conductivity of water

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Despite having a fairly simple chemical structure, water properties are non-trivial and still nowadays not completely understood. The complex properties of water are mainly due to the intricate network of hydrogen bonds that form between molecules. A recently developed model called MB-pol [1, 2, 3] has proven to be very accurate in the description of both static and dynamic properties of water, from the gaseous to the condensed phases [4]. Among other transport properties like the diffusion coefficient, whose experimental value is faithfully reproduced by MB-pol, a feature that was lacking in the computer implementation of this model is a microscopic formula for the heat flux: this is the fundamental ingredient to compute the thermal conductivity via the Green-Kubo formalism [6, 7]. The heat flux requires the distribution of the total energy and the forces among the atoms in the system: an easy task in weakly interacting materials, when the configurational energy is modeled through pair potentials, but a complicated and not completely settled issue in the presence of many-body forces.

The heat flux formula for MB-pol is obtained and implemented in the model, and the thermal conductivity is computed via Molecular Dynamics simulations and Cepstral Analysis [5] of the heat flux time-series.

The results will be discussed and compared with experimental values and with other classical models and ab initio simulations of water.

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Colour prediction of molecules in solvents: pH indicators

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The ability to predicting the colour of a given molecule from first principles would be extremely useful in a variety of industrial applications, but is a complicated problem. Calculating the molecular absorption spectrum is an inherently quantum mechanical problem, but the colour can also be affected strongly by interactions with a solvent. A method based on correcting and averaging over the spectra generated from many individual time- dependent density functional theory (TDDFT) calculations has been shown to give good results for simple solvent-solute combinations previously[?, ?]. In this work, we extend this method to test its efficacy on more complex solutes, including charged molecules, by considering pH indicators. Although the calculations are not yet complete, the method shows further promise, which bodes well for applying it to industrially useful systems in the future.

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Geometric and nongeometric contributions to the surface anomalous Hall conductivity

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The dimensionless axion coupling θ describes the isotropic part of the linear magnetoelectric tensor. In a bulk crystal θ is only defined modulo 2π, and only its space-time gradients enter Maxwell's equations. At surfaces, the spatial gradient of θ gives rise to a surface anomalous Hall conductivity (AHC). In this work, we derive a microscopic expression for the AHC of an insulating surface. We find that in general it comprises not only a geometric contribution that is a property of the occupied states, but also a non-geometric "cross-gap" term. Thus, the surface AHC is qualitatively different from the intrinsic AHC of a free-standing film or slab, which is given entirely by the geometric contribution. By constructing tight-binding models in a slab geometry, we numerically test our analytical results and explore the connection between the surface AHC and the bulk axion coupling. In particular, we illustrate how different insulating surfaces of the same bulk crystal can have AHCs that differ by an integer multiple of e^2/h, and that this difference resides in the geometric term alone.

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Theoretical prediction of arsenene for the anode of lithium/sodium-ion batteries: A first-principles study

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Being widely used in portable electronic devices, electrical vehicles, and any electric power storage devices, rechargeable batteries have strongly penetrated into our everyday life. Lithium (Li), and other members of alkali metals are ideal materials for most consumer electronics applications. Principally, electrochemical properties of the electrode materials determine important battery performance characteristics such as power capacity and operation voltage. As a traditional anode material, graphite has found a widespread use for Li-ion batteries [1]. However, graphite anode does not intercalate sodium to any appreciable extent [2]. Since 1990, significant research efforts have been devoted to developing high capacity anode materials to replace carbonaceous materials compound, i.e. graphite or graphitable carbons.

Recently, honeycomb structures of other members of the carbon group: silicene, germanene, and stanene that shares almost all the remarkable properties of graphene have attracted attentions. From the pnictogens group, phosphorene with a monolayer puckered structure was recently obtained by exfoliating black phosphorus. The puckered structure of phosphorene is different from planar graphene and buckled silicene. Fostered by recent experimental achievements and flourishing studies of phosphorene, buckled and puckered arsenene, honeycomb monolayers of arsenic have been investigated [3].

In this work, we have performed first-principles calculations to investigate the atomic and electronic structure, electrochemical and diffusion properties of lithium/sodium doped arsenene. We found that both adatoms strongly bind to arsenene indicating that Li/Na could be stabilized on arsenene without clustering. Intercalating of two adatoms on one side and both sides of arsenene, calculations showed a strong repulsion at the adatom-adatom distance of less than 3.93 Å and 4.35 Å for Li and Na, respectively. Moreover, for both low and high adatom concentrations, we witnessed diffusion energy barriers of as low as 37-59 meV and 41-62 meV for Li and Na atoms on the arsenene surface, respectively. This implies that both adatoms possess a high mobility during the charge/discharge process.

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Single atom catalyst: H_2 dissociation on single noble metal at cerium oxide (111) surface

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Single atom catalyst (SAC) is the new frontier of heterogenous catalysis: where the catalyst consists of single metal atoms dispersed on a support [1]. This is the most efficient way to exploit every single metal atom, but the role of the atom in the catalysis is currently unclear [2]. To clarify this aspect, we have studied the effect of the SAC in the activation of the Hydrogen molecule on the CeO_2 (111) surface. The oxidation of H_2 is a very important process because it is the reaction occurring at the anodes of proton exchange membrane fuel cells (PEMFC), which could represent the next generation power sources [3] or a step in the synthesize of chemical products [4].

We have focused on single Ag catalysts, and compared them to Cu and Au. For all the noble metals we have considered different configurations: atom adsorbed on the surface, atom substituting a Ce on both the stoichiometric and the reduced CeO_2(111). Density functional calculations (DFT) have been carried out using the Quantum Espresso Package [5],[6]. The Hubbard correction has been added on the f orbitals of the cerium atoms due to the strong electron correlation in these orbitals. The potential energy profiles along the Minimum Energy Pathway (MEP) have been obtained using the CI-NEB [7].

We have found that when a single nobel metal atom is adsorbed on CeO_2, the activation energies drop with respect to those of pure ceria, and in particular they are slightly lower for Ag than Au and Cu. Instead, when the noble metals substitutes a Ce atom, we have calculated that only for Ag a significative decrease of the barrier energy occurs. If an Oxygen vacancy is present on the surface then both Ag and Au are good catalysts, whereas the activation energy for the reaction is larger for Cu.

Charge transfers play a key role in determing the catalytic noble metal behavior: the noble metal facilitates the oxidation of H_2 if, following the breaking of the molecule, it could reach its favorite oxidation state by acquiring the H_2 electrons. Thus, Ag is the best single atom catalyst in the activation of H_2 on the CeO_2(111) surface: indeed, for all the examined configurations of Ag on the (111) ceria surface, the H_2 activation energy is remarkably lowered.

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Co-doped reduced graphene oxide as novel electrocatalyst for the oxygen and carbon dioxide reduction reaction

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In recent years, the negative impact of anthropogenic CO_2 emissions on our planet's climate has motivated a progressive transition from a global energy scenario heavily re- liant on fossil fuels to one based on environmentally benign, renewable energy sources [1, 2]. Consequently, the past decade has seen considerable growth in the development of materials for fuel cell electrodes that consume H_2 and O_2 as sustainable technologies [4]. Therefore electrochemical reactions such as oxygen reduction reaction (ORR) in these energy converting systems are becoming increasingly important [4]. However, while the long-term solution is abandoning fossil fuels for renewable sources, it has been shown [3] that in the short-to-medium term conventional fuels will still be used and CO_2 emissions will have to be compensated for. To this aim, one of the most interesting strategies is the electroreduction of captured CO_2 (CO_2RR) to value-added fuels and chemicals.

Although very promising, these technologies based on electrochemical reduction re- actions still face several challenges, due to the high reaction barriers, slow kinetics and low selectivity towards a specific target product. To tackle these problems, in the last decade a growing theoretical effort has focused on the quantum modelling of materials and reaction mechanisms in electrochemical cells and, more recently, on CO_2 and O_2 elec- troreduction. Herein we present the recent progress of codoped reduced graphene oxide and its role in electrochemical reduction reactions, such as ORR and CO_2RR. Carbon- based materials are very cheap and have good electric properties, but they hardly show low onset potential and high efficiency. Doping rGO with metal ions has shown to be an effective way to tune its electronic properties and catalytic activity. In our study, we employed Density Functional Theory (DFT) calculations to predict realistic structures of metal-containing rGO co-doped with nitrogen atoms. In particular, we considered several model host-systems involving N-graphitic, N-pyridinic and N-pyrrolic-like defects which are able to coordinate metal (M) ions (M=Mn, Mg, Co, Ni, Zn, Cu etc.). We investigated how the properties of rGO depend on the type of doping elements and identify the most active catalytic sites. Further we identified the reaction steps that lead from CO_2 to the specific final products (value-added chemicals such as formic acid, carbon monoxide, syn- thetic gas and methanol) or from O_2 to H_2O, calculating key quantities, such as reaction barriers and onset potentials, that allow to propose a rational design of new catalysts to be implemented and tested experimentally.

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Thermal transport in one-dimensional carbon chains: a first principle relaxon approach

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The discovery and synthesis of graphene has led to a surge in investigating low-dimensional systems as well as the other allotropes belonging to the carbon family. Among all carbon nano-structures, carbynes are the truly one-dimensional nano-materials and are predicted to exhibit several interesting properties both for basic science and possible applications. As a consequence of the well-known Peierls instability, two possible structures exist of the ideal infinite carbon atom wire: the metallic cumulene, and the semiconducting (bond- alternating) polyyne. In this work we investigate thermal transport in polyyne. We focus on the effects of tensile strain and temperature on thermal conductivity, thus comparing the relaxon picture [1] with the variational method [2] and the well-known SMA approx- imation to solve the Boltzmann Transport Equation (BTE). The former two approaches solve exactly the BTE, providing results in agreement with each other and confirming the failure of the SMA approximation in low-dimensional systems, as previously shown for graphene [1]. The values for the thermal conductivity are the highest among any material studied, and display an interesting and non trivial behavior a a function of strain.

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Deep-center defects in semiconductors

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A renewed interest has been recently devoted to the study of deep-center defects in materi- als for applications in emergent quantum technologies such as quantum sensing, quantum information processing and communication [1]. The typical example of a deep-center de- fect is the nitrogen-vacancy (NV) center in diamond. The spin states of this defect can be optically manipulated at room temperature, which makes it attractive for quantum applications [2].

The present work focuses on the application of NV-center to magnetic sensing at pressures in the Megabar range. The application of such very high pressures induces the closure of the electronic band gap of insulators/semiconductors, and is usually performed in a diamond anvil cell (DAC). The observation of superconductivity induced by the band gap closure in the DAC is experimentally challenging. Implanting NV-centers directly in the diamond anvil cell allows one to use the NV-centers as in-situ magnetic sensors, and to directly detect the Meissner effect. The behavior of the deep level states as a function of pressure is however unknown. The aim of the present work is therefore to settle a reference method to model the electronic states of the NV-center in diamond, investigate their behavior under pressure, and make a link with experiment that are being performed in the THALES Research Center.

Density Functional Theory is widely used for the calculation of the electronic structure of defects [3]. However, the single Slater determinant nature of the DFT wavefunctions does not allow for the calculations of the many-body levels of some defects. In this work I developed a Hubbard model for the NV-center and implemented it in a code written from scratch. The model parameters were calculated from DFT-GGA as implemented in the Quantum Espresso package [4]. I will show that the model enables access to some of the multideterminant singlet defect levels and I will discuss their evolution under pressure.

This work is within a collaboration between ENS-Paris-Saclay, THALES and CEA-DAM. Part of the work is performed in collaboration with Michele Casula, IMPMC, Sorbonne University.

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Energy landscape of ZrO_2 via a neural-network potential through charge equilibration technique

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In the present work, we use a machine learning method to construct an accurate high- dimensional potential for Zirconia using a charge equilibration neural-network technique (CENT)[1]. In order to describe the local environments of each atom, optimized sym- metry functions for machine-learning interatomic potentials of multicomponent systems are used [2]. A training set of stoichiometric ZrO_2 containing clusters, slabs and bulks is prepared in the framework of density functional theory. After training the neural-network potential, the reliability and transferability of the potential are verified by calculations of crystal structures and energies, phonon spectra. Surface energies and structures as well as dynamical properties derived from phonon calculations obtained by the CENT potential are in good agreement with PBE results. We also studied the stability of different surfaces of Zirconia.

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Spatial dispersion properties from DFPT: Dynamical quadrupoles and flexoelectric tensor

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In condensed-matter physics, spatial dispersion refers to the dependence of many material properties on the wavevector q at which they are probed, or equivalently on the gradients of the external field (electric, magnetic, strain...) and/or the response in real space. Remarkable examples of such gradient effects include the natural optical rota- tion, [1] whereby some crystals rotate the polarization plane of the transmitted light, or the flexoelectric tensor, [2] which describes the polarization response to a gradient of applied strain. The former is originated from the first-order wavevector dependence of the dielectric susceptibility tensor, while the latter can be written as the electrical current that is produced by an acoustic phonon at second-order in q. Density-functional perturbation theory (DFPT) appears as the ideal framework to compute these effects from first principles, but the general computational tools to deal with the long-wavelength limit are currently missing.

In this contribution we present two applications of a general formalism [3] based on an analytical long-wavelength expansion of the second-order DFPT energies that enables the direct calculation of spatial dispersion quantities. The latter are directly obtained from the uniform field linear-response functions and at essentially no additional cost. In particular, we elaborate on the specific implementation of the method for the cases of the clamped-ion flexoelectric tensor and the dynamical quadrupoles (the higher-order multipolar counterpart of the Born effective charges). The flexoelectric coefficients calculated for several materials are directly benchmarked against previously published results, [4, 5] whereas the lattice sum of the $PbTiO_3$ quadrupole moments is shown to exactly reproduce its piezoelectric tensor, as predicted by R.M. Martin in his seminal paper. [6]

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Vibrational Fingerprints of Graphene Nanoribbons

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Ultra-narrow stripes of graphene, called graphene nanoribbons, can find several applications in nano- and opto-electronics. Every production method - among the several proposed ones - gives rise to different types of ribbons, in terms of structural quality, width, edge pattern, and type of functional groups. We calculate from first-principles (and compare to experiments) the Raman and IR spectra [1,2] for several prototype graphene nanoribbons as produced by different techniques by focusing on (and rationalizing) the effect of different structural parameters, such as width and edge-patterns.

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Computational Study of Structural-Electronic Interplay in $IrTe_2$

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Metal chalcogenides display a wealth of interesting physical and chemical properties that make them promising candidates for cutting-edge technological applications [1]. Within this family of compounds, $IrTe_2$ has recently attracted a great deal of interest due to the many peculiar Temperature(T)-induced phase transitions exhibited by this material [2]. By doping the material, it is possible to tune the transition T and, for certain compositions, to induce superconductivity [3]. Moreover, $IrTe_2$ phase transitions can be triggered by photoexcitation [4]. These transitions are in fact structural rearrangements that can be traced back to a change in the chemical bonding pattern of the compound. In particular, the low-T phases display the shortening (15-30%) of certain Ir-Ir and Te-Te bonds [3]. Different hypothesis were put forward as to which chemical interaction(s) drive the transitions, but no general consensus has been reached. Because of this intriguing scenario, $IrTe_2$ represents an important test case for elucidating the interplay between electronic and structural degrees of freedom in complex materials with potential technological applications.

In this contribution, we present an in-depth computational investigation of the various phases of bare and doped $IrTe_2$. In particular, we tackle the following issues: i) the role of lattice vibrations: we calculate the phonon spectra of the various phases to check whether it is the entropic contribution (as opposed to the enthalpic one) that stabilize the high-T phases, and whether anharmonic effects play an important role. ii) chemical bonding: we investigate it by exploiting several tools and approaches (Crystal Orbital Hamilton Population, Electron Localization Function distribution, and Density Of States analyses), as well as through specifically designed computational experiments, so as to predictively explain the greater (electronic) stability of low-T phases. iii) effect of doping: we carry out chemical bonding and phonons analysis on the Se- and Pt-(substitutionally)doped material, in order to study how the dopants influence the transition T (*e.g.* electronic vs phonon energy). This latter point will lead to the design of empirical rules to predict how other doping elements can change the stability of the various phases of $IrTe_2$ and possibly of other similar metal chalcogenides.

A number of important insights emerge from our investigation, some examples of which are mentioned in the following. We demonstrate that the interlayer Te-Te interactions are not the driving force for the phase transitions, at odd with what was previously hypothesized [2,5]: in fact, those interactions are destabilizing the low-T phases, although the corresponding energy penalty is (over)compensated by the energy gain associated to intralayer bonds. We confirm that it is the -TS (entropic) contribution that induces the phase transitions upon heating. We show that effect of doping on the relative stability of various phases is quite complex and changes depending on what element (Ir or Te) is being substituted.

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Computational Methods for the Study and Modelling of Graphene Oxide

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Graphene oxide (GO) is a versatile 2D material whose properties can be tuned by changing the type and concentration of oxygen-containing species bonded to its surface. In this sense, a precise knowledge of the relationship between chemo/physical properties of GO is vital to have effective technological applications. To this extent we aim to provide reliable results and simulation procedures that accurately represent reduced Graphene Oxide (rGO). In [1] we studied the relationships between the structure and the electronic properties of rGO, providing guidelines for the assignment of peaks in the C-1s XPS spectra to different oxygen containing species, and introducing a revision to the widely used Lerf-Klinowsky structural model. During this work we also discovered that the ReaxFF [2], one of the most used reactive potential when it comes to perform classical molecular dynamics on rGO, yields unreliable results in describing rGO structure compared to DFT, with forces and bond lengths significantly affected. For this reason, our efforts are focused into developing a new interatomic potential to perform realistic, fast, and very large scale classical moleculardynamics (MD) on rGO.

We are developing a machine learning potential using a very large set of realistic rGO structures for the training of deep neural networks, following the scheme proposed by Wang et al in [3] and Han et al in [4]. In this approach, deep neural networks are trained to represent the potential energy surface (PES) of an arbitrary input atomic structure. A thorough training of a deep neural network potential, will allow to perform MD simulations with a speed comparable to that of classical MD and with an accuracy comparable to that of the method used to build the training set, as discussed in [4]. This in turn will introduce an alternative potential to the ReaxFF, with a comparable computational cost and a much better accuracy. We expect the outcomes of this project to finally open the way for realistic large-scale MD simulations of rGO with potential impact in many technological fields, such as membranes, electrodes and paper-like materials.

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Low-complexity MP2 for solids

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Hartree-Fock plus MP2 is a standard approach in materials physics and quantum chemistry to evaluate the energy of matter. However, this is computationally very demanding since conventional MP2 implementations scale with the fifth power of the system size, O(N^5), and are difficult to parallelize. We present two low-complexity implemen- tations that have a lower scaling and an almost ideal parallelization efficiency. The key concept of both approaches is to eliminate the summation over all combinations of occu- pied and unoccupied states, which can be elegantly achieved in the Laplace transformed MP2 formulation [1]. In this way we attain a quartic scaling high performance algorithm, O(N^4), in the plane-wave basis without introducing further approximations. Moreover, using stochastic HF orbitals, a cubic scaling, O(N^3), can be achieved when a fixed abso- lute error is assumed which turns into a linear scaling, O(N^1), when only a fixed relative error is assumed (e.g. per atom) [2, 3]. Analogously, the approaches could allow us to calculate second-order screened exchange as well as particle-hole ladder diagrams with a similar low complexity.

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Metric-wave approach to flexoelectricity within density-functional perturbation theory

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Theoretical interest on the fundamentals of mechanical deformations has experienced a recent boost due to the surge in popularity of the flexoelectric effect, i.e. the polariza- tion response of a generic insulator to a strain gradient deformation. First-principles approaches to flexoelectricity have revolved around the analysis of long-wavelength acoustic phonons, which are an effective way of dealing with the nonperiodic nature of a strain- gradient perturbation. However, previous calculations of the flexoelectric coefficients were plagued by a number of conceptual and technical difficulties, for example related to the presence of peculiar magnetic-like contributions that are associated with rotation gradi- ents. [1]

In order to overcome such obstacles once and for all, we implement and test a novel "metric wave" response function, within the framework of density-functional perturbation theory (DFPT). It consists in the reformulation of an acoustic phonon perturbation in the curvilinear frame that is co-moving with the atoms. This means that all the perturbation effects are encoded in the first-order variation of the real-space metric, while the atomic positions remain fixed. This can be regarded as the generalization of the approach of Hamann et al. [2] to the case of inhomogeneous deformations and greatly facilitates the calculation of advanced electromechanical couplings, such as the flexoelectric tensor. We demonstrate the accuracy of our approach with extensive tests on model systems and bulk crystals (Si, $SrTiO_3$).

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Enhancement of Magnetic Anisotropy of Cobalt Cluster by Doping

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The rare earth (RE) intermetallics $Nd_2Fe_{14}B$ and $SmCo_5$ are widely used as high performance permanent magnet because they have high magneto crystalline anisotropy (MCA) with reasonable magnetization and Curie temperature. They show large hysteresis and coercivity. They are used in high density memory storage and a variety of industrial and medical applications.

Due to the crisis of RE elements, there is an ongoing global effort to design RE free permanent magnets with properties comparable to those of RE magnets.

MCA is one of the factors (in addition to shape and surface anisotropy) affecting properties of nano-scale magnets. MCA is a result of spin-orbit coupling, a relativistic effect in atoms. It is expected to be large in heavier elements, is usually small in 3d elements but larger in 5d elements.

Surprisingly, a nano-scale material of the composition Co_3C was found to have large MCA [1] much larger than bulk hcp Co. The cobalt carbide nano-magnets with domain size 8 nm exhibit thermally stable long range ferromagnetic order with a Curie temper- ature of $\sim$ 650 K and a blocking temperature 570 K. This blocking temperature is even higher than the operating temperature of $Nd_2Fe_{14}B$ [2]. This nano-magnet has a MCA energy $7.5\times10^5 J/m^3$, much higher than the value of $Nd_2Fe_{14}B$ $5mJ/m^3$ [3].

Islam and Khanna [4] claimed that mixing of Carbon p states with Cobalt d states is responsible for such unusually large anisotropy energies. Motivated by these we have calculated MCA energies of Co_4 clusters doped with C, N, Si, Ge and As atoms. We want to address the question whether other p elements can increase the anisotropy energy further, and whether one can arrive at a principle about increase in anisotropy energy so that a design strategy of RE free permanent magnets can be proposed.

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Diffusion of Amoxicillin in water

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Abstract:

The knowledge about transport phenomena like diffusion, transport of matter from one part of system to another, has great importance to study about the working mechanism of drugs. In this work, we have performed the classical molecular dynamics simulation to study about the diffusion phenomenon of a system having 2 amoxicillin and 2160 water molecules at different temperature taking SPC/E water model using GROMACS package. We have calculated the self diffusion coefficient of amoxicillin as solute and water as solvent using Einstein's relation and their mutual diffusion coefficient using Darken's relation at temperature 293 K, 298 K, 303 K, 305 K, 310 K and 313 K. Also, we have studied the temperature dependence of diffusion coefficients of solute and solvent; and have calculated the activation energy of amoxicillin and water using Arrhenius formula and effect of system size. The calculated values for solvent are in close agreement with previously reported values. Furthermore, we have also studied the structural properties of the system using Radial Distribution Function (RDF). We further analyze the water rings around the amoxicillin molecule.

Lithium Adsorption on Graphene at Finite Temperature

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The increasing demand for high energy density lithium ion batteries motivates a search for alternative electrode materials. Experimentally obtained graphene based structures have been suggested to replace the state-of-the-art graphitic anode [1]. For a thorough characterization of Li adsorption on graphene, we study the interaction of Li with graphene both at zero and finite temperatures. The zero temperature study was carried out by means of density functional theory (DFT) accounting for van der Waals interactions while the finite temperature behavior was studied by Monte Carlo techniques with DFT-derived Li-graphene interaction potential constructed via cluster expansion method. The zero temperature calculations show that Li-graphene interaction is mainly electrostatic and dispersed Li ions are unstable with respect to bulk Li. At higher temperatures, entropic effects stabilize lower concentrations with respect to bulk Li. At temperatures below 400 K, we found that at any given chemical potential, the formation of 2D Li-clusters is stable over a random distribution of Li on graphene. Our calculations reveal different ordering of Li-clusters such as Li-islands and Li-stripes. The formation of the Li-stripes is however due to the finite size of the simulation cell and therefore, the Li-islands phase is expected to dominate in the thermodynamic limit.

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Unified theory of thermal transport in crystals and disordered solids

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The phonon Boltzmann equation developed by Peierls [1] describes the heat conduction in solids in terms of the dynamics of interacting phonon wave-packets. This picture holds true only in the limit of a perfectly ordered and infinite crystal. Several methods have been recently developed to solve this equation in a numerically exact way, allowing to determine the thermal conductivity of crystalline solids [2, 3].

Introducing disorder, it is possible to reach a point where the phonon wave-packets do not propagate far enough to sample the periodicity of the solid, rendering impossible to attribute them a wave vector or a group velocity. This regime is often described by a harmonic theory introduced by Allen and Feldman [4].

We generalize the Peierls and Allen-Feldman approaches in a unified master equation, which enables reliable first-principles predictions of the thermal conductivity of any insu- lator, ranging from complex crystals to anharmonic glasses. We showcase this approach with an application to a thermoelectric material that displays ultra-low glass-like thermal conductivity and rattling phonon modes.

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Towards a Common Format for Computational Materials Science Data

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Abstract:

Materials science relies on computational methods, to predict and investigate the electronic, transport and mechanical properties of the material. Such studies could drive researchers towards the most promising candidates for a specific application and it is used to assess the performance of known materials or to simulate materials not yet existing. This field of research is extremely popular, and leads to a huge variety of codes, computational methods and amount of data produced by academics and industries all around the world.

The NOMAD (Novel Materials Discovery) Laboratory is a European Centre of Excellence (CoE) that involves eight complementary research groups in materials simulation from all around Europe (Spain, Germany, UK, Finland, Denmark) and four high-performance computing centres. This CoE's main goal is to gather data coming from as many different sources as possible in a repository (NOMAD repository), convert them to common format and make it open access.

The building block of such infrastructure is ``the conversion layer''. This is where raw data from theory and modelling are transformed into a code-independent format and stored in a open access homogeneous data archive. The underlying concept is that all the information contained in inputs and outputs in the repository, e.g. optimised geometries, total energies, atomic forces, is always parsed and converted to a common format. NOMAD accepts entries from over 32 different ab--initio (e.g. CASTEP) and classical (e.g. LAMMPS) codes and the details of how this is achieved, despite the variety of data formats.

Based on this archive a big-data analytics toolkit and a material encyclopaedia have been built. These allow one direct access to the information on materials and properties retrieved from the data pool. Such infrastructure will give academic and corporate users the chance to gain a special insight into materials.

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First-principles theory of spatial dispersion effects in crystals

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Density-functional perturbation theory (DFPT) is nowadays the method of choice for the accurate computation of linear and non-linear response properties of materials from first principles. A notable advantage of DFPT over alternative approaches is the possibility of treating incommensurate lattice distortions with an arbitrary wavevector, q, at comparable computational cost as the lattice-periodic case. In this poster I will show that q can be formally treated as a perturbation parameter, and used in conjunction with established re- sults of perturbation theory (e.g. the "2n+1" theorem) to perform a long-wave expansion of an arbitrary response function in powers of the wavevector components. This provides a powerful, general framework to accessing a wide range of spatial dispersion effects that were formerly difficult to calculate by means of first-principles electronic-structure meth- ods. In particular, the physical response to the spatial gradient of any external field can now be calculated at essentially no cost, by using the response functions to uniform per- turbations (electric, magnetic or strain fields) as the only input. I will also discuss special issues that need to be addressed for the calculation of the flexoelectric tensor, such as the finite-q generalization of the polarization [1] response and of the strain [2] perturbation.

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Phonons transport and structural phase transition in $CsPbBr_3$ from first-principles

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Halide perovskites ABX_3 (A=CH_3NH_3,Cs; B=Pb, Sn; X=Cl, Br, I) have attracted significant attention in recent years for their excellent optoelectronic properties. While the optoelectronic properties of halide perovskites have widely been studied both experimentally and theoretically, their thermal properties have been studied only partially. In particular, first-principles calculation of phonons of halide perovskites is challenging because of the strong phonon anharmonicity, by which the conventional harmonic approximation breaks down.

In this study, we applied the recently developed self-consistent phonon (SCP) scheme [1, 2] to the all-inorganic perovskite $CsPbBr_3$ and calculated the thermal conductivity κ_L and vibrational free-energy from first principles. The SCP can incorporate the anharmonic effect non-perturbatively and enables us to obtain well-defined phonons of high-temperature phases. Also, we can predict the structural phase-transition temperatures T_c of $CsPbBr_3$ by comparing the anharmonic free-energy of the three phases: cubic, tetragonal, and orthorhombic. We will demonstrate the validity of our approach by comparing the predicted κ_L and T_c values with available experimental data.

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On-site and inter-site Hubbard interactions from density-functional perturbation theory

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DFT+U+V is a simple and powerful tool to model systems containing partially-filled manifolds of localized states [1]. However, the Hubbard parameters are often treated semi-empirically, which is a somewhat unsatisfactory approach. Conceptual and practical methods to determine e.g. the Hubbard U parameter from first principles have nevertheless been introduced long ago, based either on the constrained random-phase ap- proximation (cRPA) [2] or on linear-response theory [3]. Nonetheless, these approaches are often overlooked due to their cost or complexity. Here, we introduce a computation- ally inexpensive and straightforward approach to determine on-site U and inter-site V [4], hitherto obtained from the difference between bare and self-consistent inverse electronic susceptibilities evaluated from supercell calculations. By recasting these calculations in the language of density-functional perturbation theory [5] we remove the need of su- percells, and allow for a fully automated determination of susceptibilities and Hubbard parameters. Such developments open the way for deployment in high-throughput studies, while providing the community with a simple tool to calculate consistent values of U and V for any system at hand. Last, the approach has been implemented in the Quantum ESPRESSO package [6] and is showcased with applications to selected transition-metal compounds.

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Heat transport from Deep Neural Network potentials

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Interatomic potentials derived from deep neural networks hold the promise to pro- vide an alternative, or at the very least a powerful complement, to fully ab initio, but much more expensive, approaches based on density-functional theory. In particular, the recently developed Deep Potential–Smooth Edition (DeepPot-SE) model [1, 2], once prop- erly trained, has proved to correctly predict the structural properties of small organic molecules, inorganic clusters and bulk systems, and of liquid water.

Since the model is continuously differentiable and crucially based on a local partition of the total energy into atomic contributions, its has all the ingredients necessary to properly describe heat transport as well. In this work we present a derivation and an implementation of an explicit expression for the energy flux in DeepPot-SE, suitable for the computation of heat-transport coefficients in crystalline solids, as well as in glasses and liquids, using equilibrium molecular dynamics and the Green-Kubo theory of linear response. Our approach is extensively benchmarked and validated against ab initio results for liquid water and for some of its high-pressure/temperature phases relevant to the physics of giant planets, obtained using a recently developed approach based on density- functional theory [3].

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Chiral spin texture in the charge-density-wave phase of the correlated metallic Pb/Si(111) monolayer

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Group IV adatoms on group IV (111) surfaces represent an important research topic. In fact those materials present a lot of exotic phenomena: reversible phase transitions[1],magnetic orderings[2, 3], metal-insulator transitions[4, 5] and also superconductivity[6]. At room temperature 1/3 ML coverage of Pb (or Sn) grown on to of Si(111) (orGe(111))displays a √3 × √3-R30° reconstruction: the host atoms occupy the T4 sites atop the substrate surface in an hexagonal array[4], the so-called α-phase. A free unsaturated electron is left at each T4 site, thus these systems are expected to present a half-filled surface band. This expectation is however in contrast with experimental evidences of an insulating ground state in Sn on Ge(111)[4] and Si(111)[7]. The reason of this behaviour has been attributed to electronic correlations[5]. At low temperature the situation is compli-√√ cated by the presence in most systems of a reversible structural transition from 3 × 3 to 3 × 3 periodicity[1, 8]. Although its origin has been extensively debated, it is still not clear whether this transition is due to the freezing of an out-of-plane phonon mode[9] or produced by long-range electronic correlations[10], both favouring a charge-density-wave having the so-called 1Up-2Down atoms configuration. In this work we study the 1/3 ML α-Pb/Si(111) by STM/STS and by fully-relativistic Density Functional Theory. Experimental results reveal the 3×3 ground-state to be a correlated metal not undergoing a Mott transition. It is characterized by a highly depressed density of states at the Fermi level where well-defined quasiparticles exist. We demonstrate Pb-substrate interaction, non-collinear spin-orbit coupling and correlation effects are all mandatory ingredients to correctly describe the electronic structure of the 1/3 ML Pb/Si(111) surface and its reconstructions. We have found that the low-temperature ground-state of the 3×3 reconstruction is a strongly correlated metal and, by comparing calculated quasiparticle interference with Fourier transform of STS data, we demonstrated the occurrence of two large hexagonal Fermi surfaces with in-plane spin polarization and opposite helicities.

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Density functional perturbation theory with fully relativistic pseudopotentials: the magnetic case

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We extend density functional perturbation theory for lattice dynamics formulated for fully relativistic norm-conserving and ultrasoft pseudopotentials [1] to magnetic materials. Our approach is based on the application of the time reversal operator to the Sternheimer linear system and to its self-consistent solutions, similarly to what proposed in Ref. [2] for the calculation of magnons. Moreover, we discuss how to include in the formalism the symmetry operations of magnetic point groups, which require the time reversal operator. We validate our implementation by comparison with the frozen phonon method applied to Ni, and discuss some applications.

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Towards MBPT for atomic systems: method benchmarking and reference results

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While the GW method has been the subject of an intense work of validation, beyond-GW and higher order many-body perturbation theory (MBPT) methods have received much less attention. In this work we aim at investigating the performance of these beyond-GW MBPT approaches and focus on atomic systems described within the spherical approximation. This allows us to implement a dedicated numerical treatment of MBPT without most of the commonly adopted numerical approximations, involving e.g. space and frequency representation of Greens' functions and dynamical operators in general, the use of pseudopotentials, the lack of self-consistency, etc.

Within our approach, implemented in the AGWX code, we make use of spherical harmonics to represent the angular dependency of space variables and of a B-spline basis set together with the Gauss-Legendre quadrature to handle radial degrees of freedom [1]. We have performed validation and verification analyses of the approach, resulting in excellent agreement with DFT, HF, and linear response calculations performed using Quantum ESPRESSO [4] and YAMBO [5], or from the existing literature [2]. Work is in progress to benchmark well-established approximations in many-body perturbation theory (MBPT), such as GW, and testing others lying at the frontier, such as second order screened exchange (SOSEX).

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Impact of conjugation length on electronic and optical properties of oligothiophene/F4TCNQ charge-transfer complexes

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The creation of charge-transfer complexes (CTC) in organic donors and acceptor organic materials is, along with ion pair formation, the ruling doping mechanism in these systems. Recent observations [1] have pointed out that the appearance of either process critically depends on the donor length. To address this issue, we investigate electronic properties and optical excitations of a series of oligothiophene (nT) molecules doped by the strong acceptor F4TCNQ. In the framework of density-functional theory (DFT), and many-body perturbation theory, including the G_0W_0 approximation and the Bethe-Salpeter equation, we consider CTC formed by four different oligomers with increasing length (4T, 6T, 8T and 10T). Based on the level alignment obtained from DFT adopting the CAM-B3LYP hybrid functional [2] we find that the HOMO and the LUMO have CT character in all complexes. On the other hand, occupied and virtual states above and below them are strongly dependent on the nT length.

In addition, the optical spectra reflect the electronic structure. For all systems, the first bright excitation is dominated by the HOMO-LUMO transition. At increasing donor length, higher-energy peaks exhibit different character, which again points to conjugation-length dependence. The rationale offered by our results contributes to clarify the excitation processes in organic donor/acceptor complexes.

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Dynamical local connector approach for electronic excitations

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Density Functional Theory is an extremely useful tool for dealing with ground state properties such as the density or total energy. Kohn-Sham eigenvalues are often considered as approximated electronic excitations, but the resulting spectra are poor.

We propose a generalization of the Kohn-Sham approach to address in an exact frame- work electron addition and removal spectra. They can be measured by photoemission experiments, and can be evaluated using a computationally expensive non-local Self En- ergy. Our method is instead based on a frequency-dependent local potential [1], which significantly reduces the computing time of an ab-initio calculation.

To find this spectral potential in practice, we propose a jellium-based dynamical local connector approximation (dynLCA): it relates the unknown potential to its homogeneous counterpart, via a non-trivial connector in space and frequency, which is based on phys- ical insight. In this poster I present the achievements and the limits of dynLCA for prototypical real materials [2] and models [3].

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A monolayer transition metal dichalcogenide as a topological excitonic insulator

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Monolayer transition metal dichalcogenides with T? structure might realize the quantum spin Hall effect at room temperature, since they have a large hybridization gap between the inverted p and d valence bands that provides topological order with robustness. Here we demonstrate that T?-MoS2 is unstable against spontaneous exciton generation on the basis of first-principles many-body perturbation theory, as the computed exciton binding energy is larger than the quasiparticle gap. We predict that the true ground state of T?-MoS2 is a novel correlated insulator in which both excitonic and topological orders coexist by reducing the spatial point group symmetry, whereas typically electron-electron interactions tend to disrupt the topological order. A self-consistent mean-field calculation provides us with clear-cut signatures of this topological excitonic insulator, such as an en- hanced bulk quasiparticle gap (and hence increased topological robusteness), spontaneous inversion symmetry breaking, spin-splitting of quasiparticle bands. The phase diagram, in the space whose axes are temperature and strain, includes a second—topologically trivial—excitonic phase that appears with a discontinuos variation of the quasiparticle gap, which surprisingly does not close across the transition.

Hydrodynamic heat transport regime in bismuth: a theoretical viewpoint

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Currently, a lot of attention is devoted to the study of phonon-based heat transport regimes in nanostructures. Of particular interest is the hydrodynamic regime, in which a number of fascinating phenomena such as Poiseuilles phonon flow and second sound occur, and where temperature fluctuations are predicted to propagate as a true temperature wave of the form $e^{i(k.r\omega\ t)}$. Together with solid helium and NaF, bismuth is one of the rare materials in which second sound has been experimentally observed, and regimes of heat transport vary with the increase of the (yet cryogenic) temperature: from heat transport via ballistic phonons, to the regime of Poiseuilles flow with second sound, to the diffusive (Fourier) propagation [1].

In this work [2,3], a major advance consists of accounting for the phonon repopulation by the normal phonon-phonon processes in the framework of the exact variational solution of the Boltzmann transport equation, coupled to the ab initio description of anharmonic- ity: three-phonon collisions turn out to be particularly strong at low temperatures and lead to the creation of new phonons in the direction of the heat flow (normal processes), which enhance the heat transport. This induces time and length scales over which heat carriers behave collectively and form a hydrodynamic flow that cannot be described by independent phonons with their own energy and lifetime.

Our exact calculations predict the occurrence of this Poiseuille phonon flow between 1.5 and 3.5 K, in a sample size of 3.86 and 9.06 mm, consistent with the experimental observations. Hydrodynamic heat flow characteristics are given for any temperature: heat wave propagation length, drift velocity, and Knudsen number. We finally discuss a Gedanken experiment allowing us to assess the presence of a hydrodynamic regime in any isotopically pure bulk material.

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Fully-automated construction of Maximally Localized Wannier Functions for materials: SCDM-k method and the AiiDA workflow

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In the original Wannier90 framework[1, 4], the automation of MLWFs is hindered by the non-trivial step of selecting a set of functions with the correct symmetries and centers to use as initial guess for the optimization.

In a recent paper, by Damle, Lin and Ying [5], the authors describe the SCDM-k method for obtaining a set of localised Wannier functions without the need for an initial guess. The method is also applicable to the case of a set of bands that do not form an isolated manifold (i.e., are entangled).

Based on the information in the DFT states (single-particle density matrix) the method is able to automatically generate localised functions to be used by Wannier90 as initial guesses, with almost no user intervention. For an isolated manifold of bands, e.g., the valence bands of an insulator, the algorithm is parameter-free. In the case of entangled bands only two parameters are required, regardless of the number of Wannier functions sought.

We have implemented this method in the Quantum ESPRESSO code and combined with Wannier90 to generate MLWFs. We have tested the method on a variety of solid- state systems, exploiting the AiiDA [5] workflow and databases, which allows us to au- tomatically generate MLWFs for materials without having to specify initial parameters. This opens the way for high-throughput, user-intervention-free computation of advanced materials properties with Wannier functions.

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The Role of Functional Groups on the Adsorption Characteristics of Self Assembled Carboranethiols on Au(111)

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Self assembled monolayers are favorable in various areas such as molecular electronics, surface wetting, molecular recognition, and biotechnology. [1] Depending on coating density, inter-molecular and molecule-surface interactions are the main factors that de- termine film characteristics. In other words, it is possible to form a film with the required properties. Moreover the work function of the metal electrodes used in the design of organic electronic devices can be controlled by setting the dipole moment magnitude and vector of the SAM molecules. Dicarbacloso-dodecaborane thiols ($C_2B_{10}H_{12}S$, briefly carboranethiols) are highly suitable molecules for this purpose. The electronic properties of carboranethiols can be modifed without distorting their cage geometry by playing with the positions of carbon and sulfur atoms and by attaching different functional groups. This allows to change the dipole moment of the molecules and can be used to control the work function of gold with SAMs. Altering the dipole moment of thiolates also changes the surface geometry which leads to SAMs with diferent structural properties. The chem- ical stability and almost spherical cage structure of carboranethiols make them promising molecules to prepare films that can be used in many different applications.

We have systematically studied both isolated and full monolayer adsorption of functionalized carboranethiols on Au(111) using dispersive corrected density functional theory (vdW-DFT). The vdW interactions have been included using the SCAN+rVV10 func- tional [2] based on our tests and comparisons with the available experimental data. The primary emphasis has been given to the role of various possible radicals (COH, COOH, $CONH_2$, OH, NH_2, NO_2, Cl, F) on the chemisorption characteristics (adsorption sites, binding energies and tilt angles on the gold surface) of eight different variants of carbo- ranethiols. The effect of functional groups on the atomic arrangements and the dipole moments of carboranethiol molecules have been elucidated. The change in the the work function of Au(111) in the presence of SAMs of carboranethiols with different radicals have been discussed.

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