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(in alphabetical order of presenting author)

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Thermoelectric transport properties with non-parabolicity, degeneracy and multiplicity of band edges: The case of anisotropic p-type SnSe

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Efficient *ab initio* computational methods for the calculation of materials thermoelectric (TE) transport properties, is of great avail for energy harvesting technologies. The `BOLTZTRAP` code [1], has been largely used towards this aim. However, its current version that is publicly available is based only on the constant relaxation time (RT) approximation within the Boltzmann transport equation. Here, we extended the implementation of `BOLTZTRAP` code by incorporating realistic **k**-dependent RT models of the temperature dependence of main scattering processes, namely, screened polar and nonpolar scattering by optical phonons, scattering by acoustical phonons, and scattering by ionized impurities with screening. Our RT models are based on a smooth Fourier interpolation of Kohn-Sham eigenvalues and its derivatives, taking into account non-parabolicity (beyond the parabolic or Kane models), degeneracy and multiplicity of the energy bands on the same footing, with very low computational cost. In order to test our methodology, we calculated the anisotropic TE transport properties of *Pnma* phase of p-type SnSe. Our results present quantitative agreement with experimental data, concerning the evolution of anisotropic TE coefficients with both temperature and chemical potential [2,3]. Hence, from this picture, we also obtained the evolution and understanding of the main scattering processes involved. Moreover, we estimated the evolution of ionized vacancies formation and its corresponding anisotropic vibrational entropies with temperature. Our results show a correlation between peaks in the anisotropic Lorenz function with anisotropic crossover behavior in the vacancy formation around 680K, which can be directly correlated with the increase of mode softening that leads into the structural phase transition.

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Consequences of phonon anharmonicity on structural, thermal and vibrational properties of 2D materials

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Anharmonic effect is usually very strong in two dimensional (2D) crystals and it becomes more pronounced at high temperatures. The conventional lattice dynamics methods fail to capture the strong anharmonic nature of these materials. To accomplish this task, we developed a spectral energy density (SED) based method, and have been extensively used to study the temperature dependent structural stability, frequency shift, linewidth and coupling of normal modes of vibrations of graphene [1], 2D h-BN [2] and monolayer (ML)-MoS_2 [3]. The results obtained show significant differences from quasi-harmonic studies due to the inclusion of higher order phonon-phonon coupling processes, which were absent in quasi-harmonic approximation. The structural stability analysis envisages that the inclusion of higher order phonon-phonon coupling is a must to stabilize the graphene and 2D h-BN sheet at finite temperatures with the accompanying crumpling of the sheets observed in experiments. Unlike graphene and 2D h-BN, in ML-MoS_2, its finite thickness counteracts against the membrane effects and makes the sheet stable and prevents the crumpling transition. To understand the temperature dependent phonon frequency shift and linewidth of optic phonon modes, the mode resolved phonon spectra are computed at Γ point. This study shows that higher order phonon-phonon coupling processes have significant role in determining the temperature dependent peak shift and broadening of optic phonon modes in all 2D materials studied in this context. Thermally excited ripples are inevitable in 2D crystals and they can affect their thermal expansion properties. We have delineated the role of such ripples on the thermal expansion properties of aforementioned 2D materials by explicitly carrying out 2D and 3D MD simulations [4]. Ripples affect the thermal expansion properties of graphene and 2D h-BN significantly, while its effect is marginal in ML-MoS_2. The above discrepancy is attributed to the special S-Mo-S sandwich structure of ML-MoS_2, which reduces the rippling behavior considerably.

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Realistic Description of Competing Interactions in Metallic TMDCs

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Two-dimensional transition metal dichalcogenides constitute a prominent showplace for competing many-body instabilities such as superconductivity [1], charge-density waves [2] and magnetism [3]. In this study, we show that even though the observed phase diagrams are complex, the underlying mechanisms are captured by a compact unifying theoretical framework. We apply the constrained random-phase approximation (cRPA) [4] and constrained density-functional perturbation theory (cDFPT) [5] to the metallic monolayers H-MX_2 with $M \in \{\mathrm{V}, \mathrm{Nb}, \mathrm{Ta}\}$ and $X \in \{\mathrm{S}, \mathrm{Se}\}$ and summarize the material specifics with a small number of representative Coulomb and electron-phonon interaction parameters. Both cRPA and cDFPT imply a separation of the electrons into a correlated subspace, here an isolated metallic band, and the rest. We find that all relevant physics emerges from interactions within this subspace. Beyond that, the materials can be well described by very similar tight-binding and mass-spring models.

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Electron-phonon scattering in n-type PbTe from first principles calculations

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In PbTe, one of the most efficient thermoelectric materials, the main scattering mechanism for electrons has been attributed to acoustic phonons to explain the temperature dependence of the mobility [1]. However, a recent study suggested that longitudinal optical (LO) phonon scattering is important in PbTe [2]. In order to determine the dominant scattering mechanism, we build accurate models of electronic and phonon bands, and electron-phonon scattering solely from first principles. We show that it is necessary to go beyond standard density functional theory to accurately describe the electronic states and deformation potentials of PbTe near the band gap. Our calculations show that acoustic phonon scattering in n-type PbTe is much weaker than previously thought [2]. We found that LO scattering dominates the electronic transport in PbTe over a large range of temperatures and carrier concentrations. At higher doping concentrations, screening weakens LO scattering, making acoustic and LO scattering comparable. We further calculate thermoelectric transport properties by solving the Boltzmann equation in the generalized transport relaxation time approximation. Our calculated values of the electronic mobility and power factor of PbTe are in very good agreement with experiment [3]. Our results may stimulate future search for new polar thermoelectric materials with weak acoustic phonon coupling.

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Electronic properties and lattice dynamics studies of the iron-based superconductor – ThFeAsN

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Abstract: Recently, Wang *et al.* [1] synthesized a new superconductor ThFeAsN, which undergoes a superconducting transition at a rather high transition temperature (T_c) of 30 K. The emergence of superconductivity in ThFeAsN without extrinsic doping and in the absence of a spin-density wave anomaly implies that a different superconducting mechanism, as compared to the other Fe-based superconductors, could be at work. We perform theoretical studies of the electronic and lattice properties of ThFeAsN using density functional theory. In the nonmagnetic state, the electronic properties show similarities to other Fe-based superconductors (e.g., LaOFeAs). The lattice dynamical properties are used for our aim, which is to find the superconductivity mechanism in ThFeAsN.

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Ab-initio Studies on Novel Properties of Two-Dimensional Materials

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The fascinating physical properties associated with graphene have motivated researchers to search for new graphene-like two-dimensional (2D) materials made up of other elements. Several candidates based on elements from group III (borophene, aluminene), IV (silicene, germanene, stanene, plumbene), V (phosphorene, arsenene, antimonene, bismuthene), and their binaries have been proposed. Silicene, germanene, stanene and borophene have already been grown. Phosphorene and few layers of arsenene and bismuthene have been manufactured by exfoliating their bulk. Recently, devices based on silicene and phosphorene have also been made [1,2]. These experimental successes caused a recent flourish of studies on 2D materials.

Using first-principles calculations, we have predicted the physical properties of three new stable 2D monolayers namely, aluminene, arsenene, and CSe. These 2D monolayers exist in different geometries such as planar, buckled and puckered configurations depending upon the type of hybridization between the constituent atoms. Group IV monolayers show buckled geometry and they are all semi-metal [3,4]. Our results show that geometry of aluminene is a planar which shows metallic character [5]. On the other hand, Arsenene and IV-VI binary counterparts of phosphorene exhibit both buckled and puckered geometric configurations. Puckered CSe and arsenene posses direct and indirect band gaps respectively [6,7]. Interestingly, it is possibility to make an indirect-to-direct gap transition in arsenene and few other group IV-VI monolayers by applying mechanical strain. Specifically, 1% strain is enough to transform puckered arsenene into a direct-gap semiconductor [6]. Few interesting results on silicene multilayers and hybrid silicene/BN will also be discussed [8-10].

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Temperature effects on the relative stability of ultra-high pressure phases of oxygen

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The crystal structure of solid oxygen in the terapascal regime has been investigated with Density Functional Theory and the Quasi-Harmonic Approximation. We have performed finite temperature calculations that provide information about a dissimilar contribution to the entropy term of the free energy for various known phases, originated in differences in the vibration degrees of freedom of the system, on top of an also important energy contribution coming from the zero-point quantum motion. Our calculations at this level suggest that the regime of stability of molecular and non-molecular forms at temperatures of relevance for planetary sciences, differs in important ways from the results predicted at zero temperature.

Electronic structures of 2D Nanomaterials:Transport and Other properties

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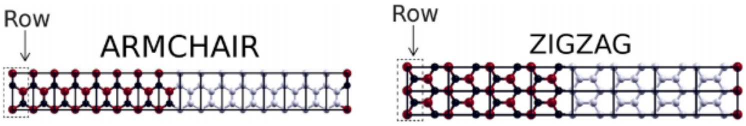
Graphene and its structural analog, a single sheet of hexagonal boron nitride (h-BN) comprising of alternating boron and nitrogen atoms in hexagonal ring, provide prototype models for the study of two-dimensional (2D) systems. Besides being interesting from fundamental physics point of view, they offer technological importance for possible applications in the field of nanoelectronics. We present some of our efforts to study the transport and thermodynamic properties of such 2D systems.



Using combination of Density Functional Theory and Monte Carlo simulation, we study the phase stability and electronic properties of two dimensional hexagonal composites of boron nitride and graphene, with a goal to uncover the role of the interface geometry formed between the two. Our study highlights that preferential creation of extended armchair interfaces may facilitate formation of solid solution of boron nitride and graphene within a certain temperature range. We further find that for band-gap engineering, armchair interfaces or patchy interfaces with mixed geometry are most suitable. Extending the study to nanoribbon geometry shows that reduction of dimensionality makes the tendency to phase segregation of the two phases even stronger. Our thorough study should form a useful database in designing boron nitride-graphene composites with desired properties.

The phonon dispersion, density of states, Grüneisen parameters, and the lattice thermal conductivity of single-layer and multilayer boron nitride were calculated using first-principles methods. For the bulk h-BN we also report the two-phonon density of states. We also present simple analytical solutions to the acoustic vibrational mode-dependent lattice thermal conductivity. Moreover, computations based on the elaborate Callaway-Klemens and the real-space supercell methods are presented to calculate the sample length and temperature-dependent lattice thermal conductivity of single-layer and multilayer hexagonal boron nitride which shows good agreement with experimental data.

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Ultrafast thermalization dynamics of hot electrons in titanium nitride

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Titanium nitride (TiN) is a hard material with a high melting point and good electrical and thermal conductivity making TiN attractive for applications in hot carrier devices. We study the ultrafast thermalization dynamics of excited carriers in TiN due to their interaction with phonons. For this, we use the two-temperature model with parameters determined from ab initio density-functional theory calculations. We find that lifetimes of hot carrier populations in TiN are shorter than one picosecond. We also study the effect of oxygen defects and nitrogen vacancies on hot carrier thermalization.

Phononic and electronic transport in nanostructures and complex materials

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Approximately two-thirds of all used energy is lost as waste heat [1]. Thermoelectrics are promising materials for waste-heat recovery. Efficient thermoelectric materials have very low thermal conductivity, high electrical conductivity (σ), and high Seebeck coefficient (S). Significant advances have been made to increase the efficiency of thermoelectrics via nanostructuring and band engineering. Nanostructuring promotes phonon scattering across a wide range of the spectrum, and can thus contribute significantly to lowering the thermal conductivity of materials. At the electron level, nanostructuring can be used to modulate electron transport, thus increasing the power factor (σS^2) [2,3]. Band engineering can also improve the performance of complex materials such as Heuslers, oxides, chalcogenides, etc., and provide guidance for rationally identifying novel chemical structures with desirable properties.

In our group we explore electronic and thermal transport at various length scales, from nano- to macro-, using various transport techniques that range from classical (e.g. molecular dynamics) and semi-classical (e.g. Monte Carlo) to fully quantum mechanical (e.g. non-equilibrium Greens functions). In all these, especially in the case of materials with complex bandstructures and complex geometries, accurate electronic properties, phonon properties, and electron-phonon scattering need to be considered. We have begun exploring the electronic properties of Heusler compounds using density functional theory, with an eye on next-generation thermoelectric materials, and using equilibrium molecular dynamics to study phonon transport in hierarchical structures. We focus here on introducing a map of the work we're performing on the latter topic, including some preliminary results for bulk crystalline Si with nanovoids, performed using the Green–Kubo formalism; and a qualitative analysis of how individual acoustic and optical modes are scattered, computed by means of a wavepacket approach.

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Magnetic transition, nature of spiral state, and absence of electric polarisation in Sr doped $YBaCuFeO_5$: A DFT + QMC study

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Contradictory results on the ferroelectric response of $YBaCuFeO_5$, in its incommensurate phase, has of late, opened up a lively debate. There are ambiguous reports on the nature of the spiral magnetic state. Using density functional theory (DFT) calculations for the parent compound within LSDA+U+SO approximation, the multiferroic response and the nature of spiral state is revealed. The helical spiral is found to be more stable below the transition temperature as spins prefer to lie in the ab plane. Dzyaloshinskii-Moriya (DM) interaction turns out to be too small to facilitate cycloidal spiral. The spin current mechanism rules out an electric polarisation in the helical spiral state. These findings are in very good agreement with the recent single-crystal experiment. We also investigate the magnetic transition in $YBa_{1-x}Sr_xCuFeO_5$ for the entire range ($0 \leq x \leq 1$) of doping. The exchange interactions are estimated from first-principles calculations as a function of doping. Later, a Quantum Monte Carlo (QMC) calculation on an effective spin Hamiltonian shows that the paramagnetic to commensurate phase transition temperature increases with doping till $x = 0.5$ and decreases beyond.

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Electron-Phonon Interactions Using Wannier-Fourier Interpolation Inside the PAW Framework

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An ab-inito method for describing electron-phonon interactions inside the projector augmented-wave (PAW) framework using Wannier-Fourier Interpolation and finite differences is presented. The quantities ot interest, such as the electron-phonon matrix element, have been derived from the theoretical framework of Allen, Heine and Cardona using second-order perturbation theory. Each such quantity can be expressed as a sum of matrix elements involving only the PAW Hamiltonian, the PAW Overlap, derivatives thereof and the unperturbed electronic pseudo wave functions. The rapid decay of these matrix elements as a function of distance in real space enables the use of Wannier-Fourier interpolation to effectively achieve very fine sampling of the Brillouin zone for both electron and phonon momenta. By employing large super cells, derivatives with respect to individual atomic displacements can be calculated directly in a basis spanned by atom-centered Wannier functions using finite displacement for a small subset of atoms. Results are presented for the phonon-induced electron self energy up to second order.

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Four-spin ring interaction as a source of unconventional magnetic orders in orthorhombic perovskite manganites

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We use *ab initio* electronic structure calculations in combination with Monte Carlo simulations to investigate the magnetic and ferroelectric properties of bulk orthorhombic $\mathrm{HoMnO_3}$ and $\mathrm{ErMnO_3}$. Our goal is to clarify the inconsistencies in the results of the experimental studies of the magnetism in orthorhombic $R\mathrm{MnO_3}$ with small R cations. Moreover, we address the contradictions between directions and amplitudes of the electric polarizations observed in these systems by different experimental groups. We report several exotic magnetic orders (so-called w-spiral, H-AFM and I-AFM) which can be stabilized in o-$R\mathrm{MnO_3}$ and whose presence can resolve the contradictions in the measurements of their magnetic and ferroelectric properties. We show that these orders emerge due to strong four-spin ring exchange interactions in these materials.

Optoelectronic Properties of Methylammonium Lead Iodide

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Perovskite solar cells are emerging as one of the most promising photovoltaic technologies, having exceeded the performance of thin-film silicon devices (beyond 21%) in only 5 years of development. This performance is exclusively due to the optimum optoelectronic properties of the prototypical organic-inorganic lead-halide perovskite, methylammonium lead-iodide ($CH_3NH_3PbI_3$), and related lead halides. The continued development of perovskite solar cells relies on elucidating the fundamental optoelectronic properties of organic-inorganic lead-halide perovskites and identifying practical routes to tune and control these properties so as to improve the performance of photovoltaic devices.

In this work we present a theoretical study of the optoelectronic properties of $CH_3NH_3PbI_3$ [1,2,3], calculated within density functional theory (DFT) and the GW approximation. Firstly, we calculate the quasiparticle band gap of $CH_3NH_3PbI_3$ within the GW approximation, and demonstrate the importance of self-consistency in the calculation of the quasiparticle correction [1]. Furthermore, we use Wannier interpolation to calculate the quasiparticle band structure and charge carrier effective masses, obtaining very good agreement with experimental measurements [2,3]. Finally, we analyze the parabolicity of the conduction and valence band edges by calculating the joint density of states and optical absorption spectrum close to the absorption onset [3]. From a joint theoretical and experimental study we show that the valence and conduction band edges of $CH_3NH_3PbI_3$ are parabolic up to 0.3 eV from the optical absorption onset, and that the optical absorption line-shape up to 0.1 eV of the onset can be modelled using Elliot's theory [4]. Our overall findings converge to the conclusion that the hybrid organic-inorganic lead-halide perovskite, $CH_3NH_3PbI_3$, exhibits very similar optoelectronic properties to those of conventional inorganic semiconductors, such as GaAs.

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Metallic σ-bonding electrons and possible superconductivity in $LiB_{1+x}C_{1-x}$

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Abstract: The metallic σ-bonding electrons are the origin of the 39K superconductivity of MgB_2. Here we define a descriptor $p_xp_y\%$ to describe the metallic σ-bonding electrons near the Fermi surface in MgB_2-like superconductors and discovere that there is a positive correlation between $p_xp_y\%$ and T_c in MgB_2-like superconductors. We also find thermodynamically stable structures of $LiB_{1+x}C_{1-x}$, which have lower energy that previous predicted structures. By using the descriptor $p_xp_y\%$, we show that there are notable metallic σ-bonding electrons near the Fermi surface in $LiB_{1+x}C_{1-x}$, which may lead to phonon-mediated high-temperature superconductivity.

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Lattice vibrational effects on singlet fission rates from ab-initio many body perturbation theory

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Singlet fission, a process in which a photoexcited singlet exciton rapidly decays to a bi-triplet exciton, is of recent interest for achieving beyond Shockley-Queissier limit solar cells. Although this process was experimentally observed over 50 years ago in molecular crystals, mechanisms of singlet fission are still hotly debated. Recently, a novel first principles reciprocal-space Green's function framework for computing singlet fission rates was developed and shown to predict quantitative singlet fission rates in crystalline Pentacene [1]. In this work, we focus on crystalline Tetracene, and present some preliminary work concerning how the previous framework can be generalized to include lattice vibrations.

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Diagrammatic Monte Carlo study of the Fröhlich polaron dispersion in 2D and 3D

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The Diagrammatic Monte Carlo is a powerful method which has proven to work in many applications for many different systems. For this paper, we have implemented a DMC code based on the Refs. [1, 2] and applied it to the solution of the large polaron Fröhlich Hamiltonian in 3D and 2D. We benchmarked our code with existing DMC results for the 3D case to verify its correctness and then computed polaron ground state energies, effective polaron masses and polaron dispersion curves in 2D and 3D.

Our data confirm that the effect of electron-phonon coupling is enhanced in 2D compared to 3D, and this is reflected in all computed physical quantities. We further show complete polaron dispersion curves. The DMC calculations reproduce very well the different behavior seen in 2D and 3D: in 2D the energy curve approaches the continuum edge asymptotically from below, whereas in 3D it reaches the continuum edge at a finite critical wave vector k_c. The accuracy of the calculated polaron dispersions are tested by comparing them to known upper and lower bounds from Ref. [3].

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Electron-phonon coupling in LED phosphors: The effect on transition energy, luminescence lineshape and thermal quenching

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Abstract

Nowadays, phosphor-converted white light-emission diodes (pc-LEDs) have drawn increasing attention as new-generation light source for general white lighting. Accordingly, the US Department of Energy defined a 2020 target for the green and red-emission converters, which mentioned that the developed phosphors should possess a narrow emission band with high thermal stability. At present, many efforts were devoted to this topic, especially the Eu^{2+}-doped ones. However, most of the research relies on trials and errors with some semi-empirical insights. The quantitative understanding of the optical behavior of these Eu^{2+}-doped phosphors, at the atomic scale, is urgently needed.

To address this problem, here we study from first-principles the luminescence of fifteen Eu^{2+}-doped phosphors through a Constrained Density Functional Theory and the one dimensional configuration coordination diagram (1D-CCD) is used to estimate the electron-phonon coupling in these phosphors. Following such methods, the transition energies and Stokes shift, the full width at half-maximum (FWHM) of the emission spectrum and thermal barrier for the 4f-5d crossover in these fifteen Eu^{2+}-doped phosphors have been estimated. The calculated results give the following assessments: (1) For the fifteen representative set, the calculated absorption and emission energies can match the experiment within 0.3 eV; (2) The 4f-5d crossover cannot be the dominant mechanism for the thermal quenching behavior of Eu^{2+}-doped phosphors; (3) The FWHM from the 1D-CCD analysis yields a 30% mean absolute relative error with respect to the experiment data; (4) The predicting power of the semi-empirical model is more limited in its accuracy and scope than the first-principles method.

Electron-phonon dynamic in hexagonal 2D tin layer

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Collecting carriers before they thermalize is the main goal for carrier's cells. This work presents a study on hot carriers in stanene using density functional and many-body perturbation theories. The present approach is applied to investigate the electron linewidth from electron-phonon interaction at 0K and 300K. It is shown that electron-phonon linewidth in stanene displays an important temperature dependence as a function of electron energy. Two different cases are considered in this report, first for electron state initially at the valence band maximum and second at the conduction band minimum and we found that the electron-phonon interaction is strongly dependent on the phonon wave vector. This orientation dependence can be used as a basis for transport phenomena. The corresponding electron scattering rate at room temperature is projected on the six phonons modes and report that the contribution to this scattering rate is overall dominated by longitudinal optical modes (LO). Another findings is that stanene's hot carriers thermalize at 250 fs, which is faster than in graphene (405fs). Our study paves the way to study hot electrons that is difficult to achieve in experiment.

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Ultra-weak interlayer coupling in two-dimensional gallium selenide

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Beyond-graphene two-dimensional (2D) materials are envisioned as the future technology for optoelectronics, and the study of group IIIA metal monochalcogenides (GIIIAMMs) in 2D form is an emerging research field. Bulk gallium selenide (GaSe) is a layered material of this family which is widely used in nonlinear optics and is promising as a lubricant. The interlayer coupling in few-layer GaSe is currently unknown, and the stability of different polytypes is unclear. Here we use symmetry arguments and first-principles calculations to investigate the phase stability, interlayer coupling, and the Raman and infrared activity of the low-frequency shear and breathing modes expected in few-layer GaSe. Strategies to distinguish the number of layers and the β and ε polytypes are discussed. These symmetry results are valid for other isostructural few-layer GIIIAMM materials. Most importantly, by using a linear chain model, we show that the shear and breathing force constants reveal an ultra-weak interlayer coupling at the nanoscale in GaSe. These results suggest that b and e few-layer GaSe show similar lubricant properties to those observed for few-layer graphite. Our analysis opens new perspectives about the study of interlayer interactions and their role in the mechanical and electrical properties of these new 2D materials.

Reference: Phys. Chem. Chem. Phys. 2016, **18**, 25401

Theoretical Predictions for monitoring the applied strain in monolayer Gallium Selenide by means of Raman Spectroscopy

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Monolayer Gallium Selenide, a layered semiconductor, displays naturally desired physical properties for realizing nanoelectronics and nano-optoelectronics. The band gap and carriers effective masses of monolayer Gallium Selenide can be tuned by strain-engineering within wide range, opening new possibilities of applications of this beyond-graphene material. For the success of this tunning technique, it is crucial the assessment of the local strain applied to the nanomaterial.

An approach for this task is to monitor the strain-evolution of the vibrational modes in monolayer Gallium Selenide. Raman spectroscopy is an quick, non-destructive, and sensitive to strain technique. Besides, strain induced anisotropy in the Raman activity may be used to identify the crystallographic orientation of the strained samples. Here we apply first-principles calculations and symmetry analysis to theoretically investigate the strain evolution of the vibrational modes in monolayer Gallium Selenide. Our results many be of great help to experimentalist working on implementation of strain-based enhancement in microelectronics.

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Superconductivity and electron-phonon properties of doped antimonene

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Antimonene is a recently discovered 2D semiconductor: elemental monolayer of Sb atoms with buckled honeycomb structure. The material was successfully obtained experimentally, and posses interesting set of properties. It was shown to have high stability on base of both experimental observations and *ab initio* modeling,and is also predicted to have interesting optical properties and strain tunable band gap. At the same time, superconductivity in doped phosphorene and graphene was recently observed experimentally, which opens new opportunities for the application of 2D materials. However, superconductivity in doped antimonene was not yet studied.

We present the *ab initio* calculations of electron-phonon coupling properties and critical superconducting temperature in both n- and p-doped antimonene at experimentally achievable carrier concentrations. The effect of bias voltage on the critical temperature is also considered. Required quantities are obtained by using density functional theory implementation of electron-phonon Wannier-Fourier interpolation in EPW and QE codes. Critical temperature at various carrier densities is estimated using McMillan-Allen-Dynes equation. We also discuss the role of various contributions to electron-phonon coupling.

The work is a part of the research program "Two-dimensional semiconductor crystals" (prj. 14TWOD01), which is partly financed by the Netherlands Organization for Scientific Research (NWO).

Magneto-transport coefficients in p-doped diamond from first principles

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(Dated: March 2, 2018)

We present a first-principles computational approach to calculate magneto-transport coefficients via the exact solution of the linearized Boltzmann transport equation (BTE). We use DFT and DFPT to describe the electronic and vibrational properties of the system, including electron-phonon interactions; carriers scattering rates entering the BTE are computed using standard perturbation theory. A similar method has been recently used to calculate thermoelectric coefficients for n-doped silicon in absence of magnetic field [1]. We discuss the application of this approach to p-doped Diamond. In particular, we focus on the effect of the magnetic field on the electronic current and on the Seebeck coefficient, including the phonon drag effect, in a range of temperatures and carrier concentrations; our method is valid for any arbitrary strength and orientation of the magnetic field. This approach gives results in good agreement with experimental data and provides a detailed characterization of thermo-magnetic transport properties of diamond, including magneto-resistance, Hall-coefficient and magneto-Seebeck effect. In this perspective, p-doped diamond is a perfect testing case because of the exceptionally high values of its transport coefficients that make diamond appealing for high-temperature electronics. Future developments will involve the application of our new method to metals, where the magneto-transport coefficient are crucially dependent on the shape of the Fermi surface [2, 3], or to semiconductors such as SiC [4], where there are still open questions about the Hall mobility, including its temperature dependence and anisotropy.

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Temperature Dependent Layer Breathing Modes in Two Dimensional Materials

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Relative out of plane displacements of the constituent layers of two dimensional materials gives rise to unique low frequency breathing modes. By computing the height-height correlation functions in momentum space, we show that, the layer breathing modes (LBMs) can be mapped consistently to vibrations of a simple linear chain model. Our calculated thickness dependence of LBM frequencies for few layer (FL) graphene and molybdenum disulphide (MoS_2) are in excellent agreement with available experiments. Our results show a redshift of LBM frequency with increase in temperature, which is a direct consequence of anharmonicities present in the interlayer interaction. We also predict the thickness and temperature dependence of LBM frequencies for FL hexagonal boron nitride (hBN). Our study provides a simple and efficient way to probe the interlayer interaction for layered materials and their heterostructures, with the inclusion of anharmonic effects.

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First principles evaluation of thermal conductivity of UN and ThN

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Abstract

Uranium Nitrides and thorium nitrides are proposed to play a crucial role in the pursuit of accident tolerant nuclear fuels. Compared to presently used nuclear fuel uranium dioxide, one of the significant advantages of the metallic fuels is their high thermal conductivity. In metallic fuels, the major contributor of thermal conductivity is from the electrons. Despite this fact, the theoretical prediction of the electron contribution of thermal conductivity of UN and ThN is not available. Therefore, in this work, we evaluate the electron contribution of thermal conductivity of UN and ThN using the Wiedemann-Franz law. The electrical conductivity (σ) required in the Wiedemann-Franz law was obtained using BoltzTraP code. The Boltztrap code gives the ratio of electrical conductivity over relaxation time (σ/τ) and the relaxation time required to calculate the electrical conductivity is predicted using EPW code.

First principles study of electron mobility of Mg_2Si

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Mg_2Si constitute an important class of materials of technical and fundamental implications in thermoelectrics research for its for medium temperature applications. Notwithstanding decades of extensive study on the experimental side, in-depth theoretical understanding of the phonon transport in Mg_2Si from first principles was attained fairly recently. [1] Meanwhile, there lacks a feasible first-principles calculation methodology of the electrical transport properties of Mg_2Si. We in this work conduct ab initio calculations to study the electrical transport properties of Mg_2Si. The microscopic electrical transport parameters as mobility and mean free path are calculated within the framework of Boltzmann Transport Equation (BTE) [2-7] in conjunction with ab initio calculations of electron-phonon coupling constants. Both the BTE and the relaxation time are solved iteratively. The calculated results are in good agreement with experimental reports. In addition, further manipulation of electrical transport properties via strain engineering is discussed.

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Static exciton-phonon coupling calculations on hexagonal boron nitride

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Abstract

The optical response of layered materials such as hexagonal boron nitride (hBN) is usually dominated by the presence of strongly bound excitons (i.e., interacting electron-hole pairs). Excitons are affected by crystal lattice vibrations, and the contribution of this exciton-phonon coupling (ExPC) to their intrinsic optical properties is garnering recently a lot of attention. The effects of ExPC on the optical response are twofold (in analogy with the electron-phonon coupling in the independent-particles case). First, excitonic states are shifted in energy and acquire a finite lifetime (exciton-phonon renormalization). Second, phonon-assisted transitions (i.e. indirect excitons) are now allowed via phonon emission/absorption. We explore these two aspects in hBN with ab initio many-body perturbation theory, in a static approach.

1) We investigate the exciton-phonon renormalization of the lowest-bound exciton in monolayer hBN using the Williams-Lax theory[1] (which accounts for static, multi-phonon processes) and compare it with an approximate method that includes single-phonon, dynamical effects only at the independent-particle level[2]. We find in both cases that the exciton energy undergoes a strong redshift (>0.5 eV) with respect to the equilibrium value, although its lifetime is only weakly affected. We discuss strengths and weaknesses of both approaches.

2) We investigate phonon-assisted absorption to low-lying indirect excitons in bulk hBN, computing the finite-difference second-order derivative of the excitonic dielectric function with respect to lattice displacements. The displacements are taken along phonon modes with momentum corresponding to the momentum difference of the indirect single-particle band gap. We find that two finite-momentum excitons exist below the direct ones, and we obtain the indirect absorption fine structure due to single-phonon emission. Our results demonstrate the existence of an indirect optical gap in bulk hBN and compare favorably with recent photoluminescence experiments[3,4].

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Electron-Phonon Interaction in Monolayer MgB_2 from the First Principles

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In this study magnesium diboride is explored in low-dimensional limit and here are presented structural, electronic, vibrational properties of MgB_2 as a monolayer. Using density functional theory and the isotropic Eliashberg approach we study the electron-phonon interaction in the single layer of MgB_2 showing it is superconductive, presenting it as a two-dimensional materials with a highest critical temperature. We showed that the low-frequency vibrations contribute more in the low-dimensional structures however not to overcome a decrease of coupling in high-frequency region. The calculated critical temperature of 18K, can be further enhanced with application of biaxial strain or by adding one more layer of boron atoms. Further we compare properties of MgB_2 monolayer with structuraly and electronicaly similar superconducting 2D materials, Li and Sr doped graphene.

Temperature variation of electronic structure of n-type PbTe and its impact on thermoelectric transport

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Some of the most efficient thermoelectric materials are semiconductors with narrow band gaps, such as lead chalcogenides PbX (X=Te, S, Se) [1,2]. However, the small band gaps of these materials make their thermoelectric transport properties very sensitive to the gap variations. Remarkably, PbTe exhibits a large band gap change with temperature, ranging from 0.19 eV at 30 K to 0.38 eV at 500 K [2,3]. An accurate description of the electronic structure as a function of temperature is thus essential for the modelling and design of efficient thermoelectric materials with narrow gaps. In this work, we report a fully *ab initio* calculation of the temperature dependence of the electronic structure of PbTe, using density functional perturbation theory [4] and electron-phonon Wannier [5] approach. We obtain the temperature variation of the direct band gap in PbTe in a very good agreement with experiments [2,3]. Furthermore, we find that the temperature dependence of PbTe's band gap considerably increases the electron-phonon scattering above room temperature. Consequently, the temperature variations of the energy gap reduce the electronic conductivity and the thermoelectric figure of merit of PbTe at high temperatures, resulting in a possible limitation for thermoelectric applications.

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Polaron-mediated surface reconstruction in the reduced Rutile $TiO_2(110)$ surface.

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The role of polarons is of key importance for the understanding of the fundamental properties and functionalities of TiO_2. We use density functional theory with an on-site Coulomb interaction and molecular dynamics to study the formation and dynamics of small polarons in the reduced rutile (110) surface, as reported in our recent publication[1]. We show that excess electrons donated by oxygen-vacancies (V_O) form mobile small polarons that hop easily in subsurface and surface Ti-sites. The polaron formation becomes more favorable by increasing the V_O concentration level (up to 20%) due to the progressively lower energy cost needed to distort the lattice. However, at higher V_O concentration the shortening of the averaged polaron-polaron distance leads to an increased Coulomb repulsion among the trapped charges at the Ti-sites, which weakens this trend. This instability is overtaken by means of a structural 1 × 2 surface reconstruction, characterized by a distinctively more favorable polaron distribution. The calculations are validated by a direct comparison with experimental AFM and STM data. Our study identifies a fundamentally novel mechanism to drive surface reconstructions and resolves a long standing issue on the origin of the reconstruction in rutile (110) surface.

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Combined experimental and theoretical study of the wavelength dependence of resonant Raman scattering in MoSe2 monolayers

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We present preliminary ab initio results of the resonant Raman scattering amplitudes for a $MoSe_2$ monolayer within the independent-particle approximation. Our goal is explaining the wavelength dependence of the resonant Raman scattering amplitudes on $MoSe_2$ in the measurements of Ref.[1]. In order to compute the Raman scattering amplitudes, we use fully relativistic pseudopotentials, compute the phonons, obtain matrix elements of the electron-phonon coupling using the Electron-Phonon-Wannier (EPW) library [2] and combine it with matrix elements of the dielectric-function for non-colinear spins.

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Metal oxide alloys for solar hydrogen production

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Metal oxides have been extensively studied, specifically for solar induced water splitting and hydrogen production. Such devices, called photo-electrochemical cells are intended not only to harness solar energy, but also to store it in the form of chemical bonds. In the specific case, Hydrogen gas, which can later be used in solid fuel cells to produce energy at night or in days of low insulation, for example. In order to effectively convert photons in hydrogen, a semiconductor needs to combine several properties, such as good light absorption, effective charge separation and transport and band edges aligned with the redox potential of hydrogen ions. In this work, using parameter free quantum simulations based on the theory of the density functional, we analyze how some key properties of Bismuth Vanadate (BiVO4) can be tunned by the formation of alloys with Molybdenum and Tungsten.

Thermoelectric Properties of MXene Monolayers

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The newest members of 2D transition metal carbides and nitrides, so-called MXenes, have garnered increasing attention due to their novel electronic and thermal properties that can be tuned for specific applications. In this study, we have investigated the Seebeck coefficients and lattice thermal conductivity values of oxygen terminated M_2CO_2 (where M=Ti, Zr, Hf, Sc) monolayer MXenes in two different functional configurations (Model-II (MD-II) and Model-III (MD-III)), in order to assess their thermoelectric properties. We revealed that the absorption site of oxygen atom has a large impact on the electronic and thermal transport properties of MXenes. The MD-III configuration always has the larger band gap, Seebeck coefficient and zT, and smaller lattice thermal conductivity as compared to the MD-II structure due to larger band gap, highly flat valence band and reduced crystal symmetry in the former. The thermal conductivity, Seebeck coefficient and *zT* coefficient may vary 40% depending on the structural model. For instance, the thermal conductivity is 40.58 W/(mK) for MD-II and 18.42 W/(mK) for MD-III of Ti_2CO_2 at T=300 K. This structural variety provides us an additional degree of freedom for modulating physical and chemical properties of MXenes, that can be exploited to design efficient thermoelectric devices. Among the considered MXenes, Ti_2CO_2 and Zr_2CO_2 in MD-III configuration could be regarded as promising candidates for next-generation thermoelectric applications due to their low thermal-conductivities and large Seebeck coefficients.

A Two-Site Model of Guest Atom Rattling in Type I Silicon Clathrates

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In general, cage compounds exhibit low thermal conductivity which makes them interesting for applications, e.g., as thermoelectric materials [1,2]. A useful conceptual model for understanding the origin of the low thermal conductivity is the phonon-glass electron-crystal (PGEC) [3]. In the PGEC model, the phonon-glass is achieved by the rattling of guest atoms inside oversized cages. Rattling is related to the anharmonic vibrations of the guest atoms [4] but a rigorously quantitative theoretical treatment of the physics is challenging. A common approach is to calculate mode Grüneisen parameters [5] which only capture the quasiharmonic part of rattling physics. From a combination of *ab initio* molecular dynamics (MD) and numerical solution of the vibrational Schrödinger equation, we attempt a full treatment of the problem. We apply the method to the cages in a Type I silicon clathrate, $Rb_8Al_8Si_{38}$. From the results, we propose a two-site model of rattling that appears to capture some of the essential physics involved in guest atom rattling in these compounds.

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Appearance or enhancement of superconducting temperature under non-superconducting point contact.

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While ZrSiS is a 3D Dirac semimetal, it was experimentally observed to superconductor under a macroscopic non-superconducting tip. We have been able to shed light on this experimental observation through first-principles calculation with ZrSiS + Ag-tip superlattice. We show that the Dirac cones are protected under the tip, followed by an enormous increase in the density of states at the Fermi level. We rule out the possibility of tip-induced pressure on the observed superconductivity. Further, electron-phonon coupling and Eliashberg spectral function lead to Tc~15 mK, which is orders of magnitude lower than experimental transition temperature 7.5 K. Thus, we argue that the appearance of superconducting transition in ZrSiS is unconventional, and maybe topological in nature.

Pure Zr superconducts at 0.5 K, however, it is recently found that under non-superconducting metallic tip the transition temperature increases by seven-fold to 3.5 K. We have explained this experimental observation through rigorous calculations of band structure, Eliashberg spectral function, Fermi surface nesting function, electron-phonon coupling, and finally calculating transition temperature. Calculated results are in excellent agreement with the experimental results.

Fully relativistic Green’s function method

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Relativistic effects are important in heavier elements and give rise to a number of interesting phenomena such as magnetic anisotropy, orbital magnetization, etc. A careful treatment of these effects requires solving the Dirac equation. Relativistic form of LMTO method was originally formulated by Ebert [1] and by Solovyev et al [2] for the core states of electrons. We have developed an algorithm for solving the relativistic Dirac equation for the valence-states wavefunctions and applied it to calculating the Green’s functions.

Now we can use the relativistic treatment to obtain a number of quantities, such as magnetic susceptibility and exchange. At equilibrium susceptibility $\chi(\mathbf{r}-\mathbf{r}') = \partial m(\mathbf{r}')/\partial B_{ext}(\mathbf{r})$. We adopt the Heisenberg model: collection of localized, rigid spins interact through exchange parameters $J_{R,R'}$, $H = -\sum_{RR'} J_{R,R'} S_R \cdot S_{R'}$ and Heisenberg J is $\chi^{-1}(\omega = 0)$. First *ab initio* determination of $J_{R,R'}$ by linear response was done by A.I. Lichtenstein et al [3] with multiple scattering theory and the ASA. LDA descriptions of magnetic exchange most commonly originate from Lichtenstein formula (multiple scattering theory) $J_{ij} = \frac{1}{\pi}\int^{\varepsilon_F} d\varepsilon \, \mathrm{Tr}_L \, \mathrm{Im}\left\{ p_i T_{ij}^{\uparrow} p_j T_{ji}^{\downarrow} \right\}$.

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Dimensionality of excitons in stacked van der Waals materials: The example of hexagonal boron nitride

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With the example of bulk hexagonal boron-nitride, a prototypical van der Waals (vdW) crystal, we demonstrate that the electronic and optical properties of these materials can be tuned by layer patterning. By modifying the stacking, energy, intensity, and character of the electron-hole (e-h) pairs can be selectively modulated. Depending on the specific layer arrangement, lowest-energy excitons are localized within a single layer or delocalized in the three-dimensional space. Only in specific stackings charge-transfer e-h pairs appear above the absorption onset, reflecting the layer selectivity of the electronic states involved. Our results, obtained form first-principles study based on many-body perturbation theory (MBPT, including GW approximation [1-2] and the Bethe-Salpeter equation [3-4]), can be extended and generalized to other vdW materials in view of understanding and predicting their optical behavior and designing novel heterostructures and interfaces with tailored opto-electronic properties.

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Nontrivial contribution of Fröhlich electron-phonon interaction to lattice thermal conductivity of wurtzite GaN

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Abstract

The macroscopic thermal transport is fundamentally determined by the intrinsic interactions among microscopic electrons and phonons. In conventional insulators and semiconductors, phonons dominate the thermal transport, and the contribution of electron-phonon interaction (EPI) is negligible. However, in polar semiconductors, the Fröhlich electron-phonon coupling is strong and its influence on phononic thermal transport is of great significance. In this work, the effect of EPI on phonon dispersion and lattice thermal conductivity of wurtzite gallium nitride (GaN) is comprehensively investigated from the atomistic level by performing first-principles calculations. Due to the existence of relatively large electronegativity difference between Ga and N atoms, the Fröhlich coupling in wurtzite GaN is remarkably strong. Consequently, the lattice thermal conductivity of natural wurtzite GaN at room temperature is reduced by 24%–34% when including EPI, and the resulted thermal conductivity value is in better agreement with experiments. Furthermore, the scattering rate of phonons due to EPI, the intrinsic phonon-phonon interaction (PPI) as well as isotope disorder is computed and analyzed. It shows that the EPI scattering rate is comparable to PPI for low-frequency heat-carrying phonons. This work attempts to explore the mechanism of thermal transport beyond intrinsic PPI for polar semiconductors, with a great potential of thermal conductivity engineering for desired performance.

Carrier Delocalization in Two-Dimensional Coplanar p-n Junctions of Graphene and Metal Dichalcogenides

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With the lateral coplanar heterojunctions of two-dimensional monolayer materials turning into reality, the quantitative understanding of their electronic, electrostatic, doping, and scaling properties becomes imperative. In contrast to traditional bulk 3D junctions where carrier equilibrium is reached through local charge redistribution, a highly nonlocalized charge transfer (trailing off as $1/x$ away from the interface) is present in lateral 2D junctions, increasing the junction size considerably. The depletion width scales as p^{-1}, while the differential capacitance varies very little with the doping level p. The properties of lateral 2D junctions are further quantified through numerical analysis of realistic materials, with graphene, $\mathrm{MoS_2}$, and their hybrid serving as examples. Careful analysis of the built-in potential profile shows strong reduction of Fermi level pinning, suggesting better control of the barrier in 2D metalsemiconductor junctions. [1]

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Mechanochemistry of Aromatic vs. Aliphatic Thiolate-Au Bond is Different!

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The interest in gold-based hybrid organic/inorganic interfacial materials has dramatically increased over the last decade due to their potential applications in diverse fields, such as heterogeneous catalysis, molecular electronics, as well as molecular biology and biomedicine, just to mention a few examples. Much of these applications involve the tailored modification of the metallic surface by incorporating molecules that provide new properties to the hybrid interface. In particular, due to the strength of the S-Au bond, thiolates on gold have been widely used for this purpose, such as in the synthesis and use of thiol-protected gold nanoclusters [1,2]. The understanding of the S-Au interaction, thus, becomes highly relevant from a technological point of view.

Mechanical stability is often neglected in the design of molecule-based devices at the nanometer scale. However, force-induced degradation and detachment processes have been proven to be key aspects for self-assembled monolayers and molecular nanojunctions of various alkyl thiolates on nobel metals [3].

Conspicuously absent from previous studies are aromatic thiolate ligands, where it is conceivable that the mechanochemistry of their S-Au bond could be tuned by using chemical substitution on the aromatic ring. In this work, we present a a systematic study of the thermal and mechanical desorption of two prototypical aliphatic thiolates and a series of substituted-*p*-methylthiophenolates adsorbed on gold surfaces.

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