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T01 - ARTRITH Nongnuch

Efficient training of ANN potentials by including atomic forces via Taylor expansion and application to water and a transition-metal oxide

T02 - BHATTACHARYA Swastika

Composition of the Earth's inner core

T03 - DANGIĆ Đorđe

Lattice thermal conductivity of highly anharmonic materials: Beyond the Boltzmann transport approach

T04 - IBAÑEZ AZPIROZ Julen

Ab initio calculation of the shift photocurrent by Wannier interpolation

T05 - JACOB David

Mott Metal-Insulator Transition from Steady-State Density Functional Theory

T06 - MARRAZZO Antimo

Kane-Mele and emergent topology in jacutingaite

T07 - MAZZOLA Guglielmo

Machine learning physics of dense hydrogen

T08 - MONACELLI Lorenzo

Time-Dependent Self Consistent Harmonic Approximation: Anharmonic nuclear quantum dynamics and time correlation functions

T09 - MOUSTAFA Hadeel

Investigation of one-dimensional materials

T10 - RODRIGUES MIRANDA Caetano

Materials discovery for biomedical applications: from machine learning and first-principles calculations to finite elements

T11 - ROYO VALLS Miquel

First-principles calculation of the bulk flexoelectric tensor

T12 - STENGEL Massimiliano

Long-range dielectric screening and interatomic force constants in two dimensions

T01

Efficient training of ANN potentials by including atomic forces via Taylor expansion and application to water and a transition-metal oxide

Artificial neural network (ANN) potentials enable the efficient large-scale atomistic modeling of complex materials with near first-principles accuracy. For molecular dynamics simulations, accurate energies and interatomic forces are a prerequisite, but training ANN potentials simultaneously on energies and forces from electronic structure calculations is computationally demanding. Here, we introduce an efficient alternative method for the training of ANN potentials on energy and force information, based on an extrapolation of the total energy via a Taylor expansion. By translating the force information to approximate energies, the quadratic scaling with the number of atoms exhibited by conventional force-training methods can be avoided, which enables the training on reference datasets containing complex atomic structures. We demonstrate for different materials systems, clusters of water molecules, bulk liquid water, and a lithium transition-metal oxide that the proposed force-training approach provides substantial improvements over schemes that train on energies only. Including force information for training reduces the size of the reference datasets required for ANN potential construction, increases the transferability of the potential, and generally improves the force prediction accuracy. For a set of water clusters, the Taylor-expansion approach achieves around 50% of the force error improvement compared to the explicit training on all force components, at a much smaller computational cost. The alternative force-training approach thus simplifies the construction of general ANN potentials for the prediction of accurate energies and interatomic forces for diverse types of materials, as demonstrated here for water and a transition-metal oxide.

T02

Composition of the Earth’s inner core

The composition of the Earth’s inner core is highly controversial. It is now well accepted that the inner core consists mostly of Fe and some amount of Ni. However, the crystal structure of the Fe-Ni alloy is a matter of debate. On the other hand, seismic studies indicate the presence of some amount of light elements in the inner core. However, the nature and incorporation mechanism are still not established. In this presentation, I will be discussing about the present status of research in this area and also discuss our investigations in this regard. Our calculations suggest that there may be more than one light element that may be present in the inner core. We find that doping of a few wt% of Si in Fe7C3 carbide phase, which is found to dynamically as well as thermodynamically stable at inner core pressure-temperature conditions, leads to marked improvement in the agreement of seismic parameters, Poisson's ratio and density with respect to the Preliminary Reference Earth Model (PREM).

T03

Lattice thermal conductivity of highly anharmonic materials: Beyond the Boltzmann transport approach

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Abstract

Phonon spectral functions of highly anharmonic materials show distinctively non-Lorentzian shapes. Utilizing the Boltzmann transport equation (BTE) for the calculation of the lattice thermal conductivity κ is not justified for these materials. Here we present a method for calculating κ that incorporates the full information about phonon spectral functions combining the Green-Kubo theory and traditional lattice dynamics in the semi-perturbative approach (i.e. the temperature-dependent effective potential method [1]) [2]. Two distinct transport regimes are recognized corresponding to diagonal and non-diagonal contribution in phonon branches. We show that the diagonal part reduces in the limit of small anharmonicity to the BTE result with a relaxation time approximation. The non-diagonal part describes tunneling between modes with the same wavevector and different branch index, which might be the dominant transport mechanism in highly disordered structures [3]. We test this approach for the case of germanium telluride close to the ferroelectric phase transition. We show that the BTE consistently underestimates κ compared to the new method. Overall this underestimation increases linearly with temperature and peaks at the phase transition. We elucidate the reason behind this intriguing result.

References

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T04

Ab initio calculation of the shift photocurrent by Wannier interpolation

The bulk photovoltaic effect, also known as the shift current, is a second-order optical response that has attracted recent interest in various fronts, e.g. by yielding record power-conversion efficiency in ferroelectric insulators [1] and insights into the topology of materials through their photoresponse [2]. From the ab-initio point of view, the practical implementation of the shift current is challenging due to a subtle Berry-phase-like quantity involved in the transition matrix elements [3, 4]. Here we propose a Wannier-interpolation scheme [5] that presents several advantages over existing methods, including avoidance of band-truncation errors, low computational cost, and a correct treatment of the optical matrix elements with nonlocal pseudopotentials. The proposed methodology and its implementation into the WANNIER90 code package [6] can help turn the calculation of the shift current into a fairly routine task, promoting the use of first-principles calculations in the search for novel bulk photovoltaics. [1] J. E. Spanier et. al., Nat. Photonics 10, 611–616 (2016) [2] T. Morimoto and N. Nagaosa, Science Advances 2, 1501524 (2016) [3] J. E. Sipe and A. I. Shkrebtii, PRB 61 , 5337 (2000) [4] S. M. Young and A. M. Rappe, PRL 109 , 116601 (2012) [5] J. Ibañez-Azpiroz, S. S. Tsirkin and I. Souza, PRB 97, 245143 (2018) [6] G. Pizzi et. al., J. Phys. Cond. Matt. 32, 165902 (2020)

T05

Mott Metal-Insulator Transition from Steady-State Density Functional Theory

We present a computationally efficient method to obtain the many-body spectral function of bulk systems in the framework of steady-state density functional theory (i-DFT) using an idealized scanning tunneling microscope (STM) setup. We calculate the current through the STM tip and then extract the spectral function from the finite-bias differential conductance. The fictitious noninteracting system of i-DFT features an exchange-correlation (XC) contribution to the bias which guarantees the same current as in the true interacting system. Exact properties of the XC bias are established using Fermi-liquid theory and subsequently implemented to construct approximations for the Hubbard model. We show for two different lattice structures that the Mott metal-insulator transition is captured by i-DFT.

T06

Kane-Mele and emergent topology in jacutingaite

Recently, we predicted [1] monolayer jacutingaite (Pt2HgSe3, a naturally-occurring mineral) to be the very first large-gap Kane-Mele (KM) quantum spin Hall insulator (QSHI). Soon after [2,3], we discovered that 3D jacutingaite is a dual topological insulator where a weak Z2 topological phase of type (0;001) coexists with a non-trivial mirror Chern number (MCN). In this talk, I will start by discussing the rich physics of monolayer jacutingaite, including its close relationship with graphene and the interplay between spin-orbit coupling, crystal-symmetry breaking, and dielectric response. Then I will focus on 3D jacutingaite crystals, where ARPES experiments [3] report the presence of 001-surface states that we show [3] to be topologically protected and to emerge from a non-trivial interlayer coupling, where KM-QSHI monolayers interact through a strong second nearest-layer hopping: that breaks the standard paradigm of weak topological insulators. We extend the KM model to 3D and explain the origin of the 001-surface states through the viewpoint of a k-dependent Su-Schrieffer–Heeger (SSH) model and its Zak phase [2]. Complemented by this term, the predictions of the Kane-Mele model are in remarkable agreement with recent experiments and first-principles simulations, providing an appealing conceptual framework also relevant for other layered materials made of stacked honeycomb lattices. [1] A. Marrazzo et al., Phys. Rev. Lett. 120, 117701(2018) [2] A. Marrazzo et al., Phys. Rev. Research 2, 012063(R) (2020) [3] I. Cucchi, A. Marrazzo et al., Phys. Rev. Lett. 124, 106402 (2020)

T07

Machine learning physics of dense hydrogen

The phase diagram of hydrogen at Mbar pressures is remarkably complex [1]. In particular, its insulator-to-metal transition, predicted almost one century ago by Wigner and Huntington is crucial in planetary science, and has important technological applications (cfn. the recent discovery of high-temperature superconductivity in dense hydrogen compound [2]). Experiments at such extreme conditions are challenging and often lead to hard-to-interpret and controversial observations, whereas theoretical investigations are constrained by the huge computational cost of sufficiently accurate quantum mechanical calculations. In this talk I will first briefly summarize the current state-of-the art for the hydrogen-phase diagram, and report a new computational discovery obtained with machine learning. [3] We provide evidence for a continuous molecular-to-atomic transition in the liquid, with no first-order transition observed above the melting line. This suggests a smooth transition between insulating and metallic layers in giant gas planets, and reconciles existing discrepancies between experiments as a manifestation of supercritical behaviour. [1] Helled, Mazzola & Redmer. Nature Reviews Physics, volume 2, pages562–574 (2020) [2] Snider et. al. Nature, volume 586, pages 373–377 (2020) [3] Cheng, Mazzola, Pickard & Ceriotti, Nature volume 585, pages217–220 (2020)

T08

Time-Dependent Self Consistent Harmonic Approximation: Anharmonic nuclear quantum dynamics and time correlation functions

Most material properties of great physical interest are directly related to time-correlation functions of nuclear positions, eg the ionic thermal conductivity, Raman/IR vibrational spectra, the dynamical structure factor, inelastic X-ray, and Neutron scattering. A theory able to compute from first principles these properties, fully accounting for the quantum nature of the nuclei and the anharmonicity in the nuclear energy landscape that can be implemented in systems with hundreds of atoms is missing. Here, we derive an approximated theory for the quantum time evolution of lattice vibrations at finite temperature. This theory introduces the time dynamics in the Self-Consistent Harmonic Approximation (SCHA) and shares with the static case the same computational cost. It is nonempirical, as pure states evolve according to the Dirac least action principle and the dynamics of the thermal ensemble conserves both energy and entropy. The static SCHA is recovered as a stationary solution of the dynamical equations. We apply perturbation theory around the static SCHA solution and derive an algorithm to compute efficiently quantum dynamical correlation functions. Thanks to this new algorithm, we have access to the response function of any general external time-dependent perturbation, enabling the simulation of phonon spectra without following any perturbative expansion of the nuclear potential or empirical methods. We benchmark the method on the IR and Raman spectroscopy of high-pressure hydrogen phase III, with a simulation cell of 96 atoms. The results show an excellent agreement with experimental data and outperform other state-of-art methods.

T09

Investigation of one-dimensional materials

1D materials are an interesting subset of materials with promising applications in batteries, photonic crystals and as electronic interconnects. 1D materials also present the possibility of combining them with other 1D materials or higher dimensional materials to create new hetero-structures with novel physical properties. Another potential application could be in heterogeneous catalysis, where the restricted geometry of 1D materials might lead to new types of atomic sites with different chemical characteristics. We identify potential 1D materials through a screening procedure applied to the inorganic crystal structure database (ICSD) and the crystallography open database (COD). We employ the dimensionality scoring parameter, which is based exclusively on the atomic geometry. The algorithm extract one-dimensional components from periodic three-dimensional crystals. So far around 300 compounds have been studied. Their basic properties like atomic structure, stability (heat of formation and convex hull), band structure, density of states and work function have been calculated. They are furthermore characterized using symmetry and grouped together using a clustering algorithm based on the root-mean-square-distance. In the future we expect to construct new potential 1D materials by element substitution in the constructed database.

T10

Materials discovery for biomedical applications: from machine learning and first-principles calculations to finite elements

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Titanium alloys are widely used as high-performance materials for biomedical applications. The low-modulus Ti-based alloys exhibit bone-like elastic modulus, therefore increasing biocompatibility. New alloys screening is challenging due to overall composition rates. To accelerate the materials discovery process, machine learning (ML) methods were employed to predict the bulk modulus and the shear modulus of optimized Ti-Nb-Zr ternary alloys [1]. Based on the elasticity data available in the Materials Project database [2], compositional-only models were generated, which could predict the resultant Young modulus (E) for all possible compositions in the Ti-Nb-Zr chemical space with variations in the composition of 2 at.%. The ML optimal compositions were further explored with first principles calculations within the Density Functional Theory and quasi-random structures (SQSs) methods to obtain the phase stability and elastic properties [1]. In the phase diagram, the low-modulus sweet spots have been identified, and the most promising candidates for biomedical applications were alloys with 22 Zr at.%. The content of Nb higher than 14.8 at.% helps to favor the bcc **β** phase since the presence of **ω** phase leads to non-desirable materials with higher elastic modulus. Considering the newly discovered Ti-14.8Nb-22Zr alloy by ML and FP, we analyzed through the Finite Element Method, a prosthetic leg design's mechanical behavior using Elmer's multiphysics simulation software. These simulations require the introduction of data concerning the materials' mechanical properties. In summary, we introduce an atom to device proof-of-concept by integrating materials discovery using ML and first-principles calculations to hip-joint implants designed by finite element method.

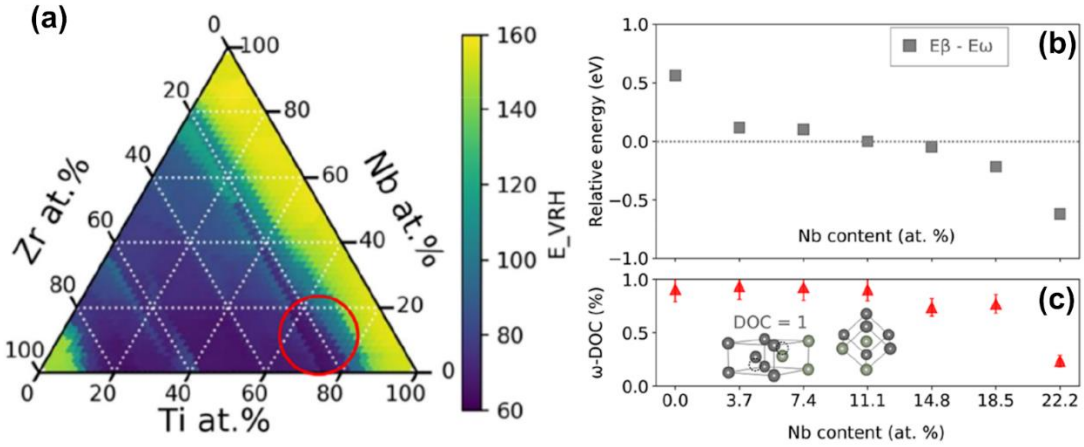

FIG1. Young modulus prediction using Random Forest, the red circle indicates the low-modulus sweet spot; b) relative stability comparison from **β** and **ω** phase; c) phase transition measured in terms of the degree of collapse (DOC).

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T11

First-principles calculation of the bulk flexoelectric tensor

The first-principles theory of flexoelectricity, describing the macroscopic polarization produced by a strain gradient, has made impressive progress since the pioneering works of Resta and Hong et al. in 2010. [1,2] The main difficulty in dealing with the flexoelectric effect, i.e., the breakdown of translational symmetry, has been overcome by reformulating the problem in terms of long-wavelength acoustic phonons. As of early 2020, a complete implementation of the bulk flexoelectric tensor is fully integrated in the latest release of the ABINIT package, [3] which is now publicly available. Additional contributions, that are relevant to the flexural deformation of a finite (quasi-two-dimensional) sample have also been understood, and can be accessed as a by-product of the main linear-response calculation. [4] In this presentation, I will illustrate the underlying theoretical formalism of the implementation, which is based on an analytical long-wave extension of density-functional perturbation theory (DFPT). [5] The latter provides, in close analogy to the third-order response functions module, the intermediate-level spatial-dispersion quantities that are necessary to build the bulk flexoelectric tensor and related materials properties. I will also discuss the physical meaning of the bulk flexoelectric tensor as well as its individual components, and how to use them in order to build reliable models of experimentally observed phenomena. [1] Resta, PRL 105, 127601 (2010). [2] Hong et al., JPCM 22, 112201 (2010). [3] Romero et al., JCP 152, 124102 (2020). [4] Springolo, Royo and Stengel, arXiv:2010.08470. [5] Royo and Stengel, PRX 9, 021050 (2019).

T12

Long-range dielectric screening and interatomic force constants in two dimensions

The foundation of modern theory of lattice dynamics rests on the separation between short-range and long-range interatomic force constants, where the latter are associated with macroscopic electric fields acting in a neighborhood of the Brillouin zone center. In three-dimensional (3D) insulators, the famous dipole-dipole formula was established long ago by Cochran and Cowley; the subsequent work of Pick, Cohen and Martin provided a formal derivation in the framework of first-principles theory. Very recently, we have incorporated higher-order interactions involving, e.g., dynamical quadrupoles, and demonstrated their role in the accurate interpolation of phonon band structures. [1] In this talk, I shall retrace an analogous journey in the context of two-dimensional (2D) crystals, by presenting a rigorous derivation of the long-range screening and interatomic forces in 2D. This enables a systematic generalization of the existing formulas (Sohier et al., Nano Lett. 2017, 17, 6, 3758–3763) to an arbitrary multipolar order. In particular, I will discuss how to incorporate out-of-plane dipoles and dynamical quadrupoles in the long-range part of the dynamical matrix, and how to achieve an optimal representation of the dielectric function. Numerical tests on monolayer BN, SnS2 and BaTiO3 membranes demonstrate a drastic improvement in the description of the long-range electrostatic interactions, with comparable benefits to the quality of the interpolated phonon band structure. [2] [1] M. Royo, K. Hahn and M. Stengel, Physical Review Letters, 125, 217602 (2020). [2] https://arxiv.org/abs/2012.07961