O 82: Poster Session VI: Poster to Mini-Symposium: Frontiers of electronic-structure theory III

Time: Wednesday 13:30–15:30 Location: P

O 82.1 Wed 13:30 P

Diagrammatic Monte Carlo study of the acoustic lattice polaron — •Thomas Hahn[1], Andrey Mishchenko[2,3], Naoto Nagaosa[2], and Cesare Franchini[1,4] — [1]Faculty of Physics, Center for Computational Materials Science, University of Vienna, A-1090 Vienna, Austria — [2]RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama 351-0198, Japan — [3]National Research Center Kurchatov Institute, 123182 Moscow, Russia — [4]Dipartimento di Fisica e Astronomia, Università di Bologna, 40127 Bologna, Italy

The Diagrammatic Monte Carlo (DMC) method is used to study an electron interacting with acoustic phonons via the deformation potential. For the first time, we obtain unbiased results for the acoustic polaron in a realistic condensed matter discrete lattice model and resolve self-contradictory conclusions from previous studies based on the continuum approximation. We present accurate numerical results for the ground state energy, effective mass, quasiparticle weight and the structure of the phonon cloud of the polaron. The most interesting data is obtained for excited states in the parameter range of the transient region between the weak and strong coupling regime. The unique structureless shape of the incoherent part of the spectral function and the flat background of the optical conductivity allows us to follow the behavior of excited states, whose studies are hindered in optical polaron models. We show that the behavior of the excited states in the spectral function is different from what is seen in the optical conductivity. This confirms the different nature of the excited states observed in these different probes of the excited spectra of the polaron.

O 82.2 Wed 13:30 P

First principles study of spin spirals in the multiferroic $BiFeO_3$ — •Sebastian Meyer[1], Bin Xu[2,3], Matthieu Verstraete[1], Laurent Bellaiche[2], and Bertrand Dupé[1,4] — [1]Nanomat/Q-mat/CESAM, University of Liège, Belgium — [2]Physics Department and Institute for Nanoscience and Engineering, University of Arkansas, USA — [3]Jiangsu Key Laboratory of Thin Films, School of Physical Science and Technology, Soochow University, China — [4]Fonds de la Recherche Scientifique (FNRS), Bruxelles, Belgium

We carry out density functional theory (DFT) calculations to explore the antiferromagnetic (AFM) spin spiral in multiferroic $BiFeO_3$. We calculate the spin spiral energy dispersion $E(\mathbf{q})$ along the high symmetry directions of the pseudo-cubic unit cell, for four different structural phases: *cubic*, $R\bar{3}c$, $R3m$ and $R3c$. In all cases, we find a large exchange frustration. The comparison provides detailed insight into how polarization and octahedral anti-phase tilting affect the different magnetic interactions and the magnetic ground state in $BiFeO_3$. For the $R3c$ structural ground state, we find an AFM spin spiral ground state with a periodicity of ∼80 nm in good agreement with experiments and previous findings. This spin spiral is driven by a Dzyaloshinskii-Moriya interaction stemming from the Fe–Bi ferroelectric displacement. The spiral appears to be stable because the anisotropy energy in $R3c$ $BiFeO_3$ is too small to enforce the collinear order. For all the four phases, we discuss the magnetic ground state and identify its stabilization mechanisms.

O 82.3 Wed 13:30 P

Influence of electronic excitations on defect formation on GaAs — •Daniel Muñoz-Santiburcio[1], Natalia Koval[1], Fabiana Da Pieve[2], and Emilio Artacho[1,3] — [1]CIC Nanogune, Tolosa Hiribidea 76, 20018 San Sebastián, Spain — [2]Royal Belgian Institute for Space Aeronomy, Av Circulaire 3, 1180 Brussels, Belgium — [3]Theory of Condensed Matter, Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom

Solar cells in spacecrafts are subject to high energy ions of solar and cosmic radiation, which promote the formation of defects in their different layers, notably affecting their performance.

The formation of defects in such materials has been abundantly studied, but usually assuming that the system stays in its electronic ground state during the whole process. On the other hand, when the target is irradiated with high energy particles, its electronic subsystem is significantly excited as a consequence of the projectile' passing. These electronic excitations may greatly alter the process of defect formation in the material in comparison to the ground-state conditions.

Here we present a study via first-principles Molecular Dynamics of the influence of the electronic excitations on the formation of defects on GaAs. We simulate the formation of different types of defects in the material for excitations of varying extent. We will show that the electronic excitations have a significant impact on the defect formation, changing the threshold displacement energy and causing different types of defects, even promoting local phase changes in the target material.

O 82.4 Wed 13:30 P

Optimized effective potentials to increase the accuracy of approximate proton transfer energy calculations in the excited state — •Pouya Partovi-Azar and Daniel Sebastiani — Institute of Chemistry, MLU Halle-Wittenberg, Halle (Saale), Germany

In various systems, acidic properties emerge when the system is electronically excited. Although the time scale attributed to the dynamics of the electrons is usually on the order of femtoseconds, the electronic excitations can in general trigger much slower processes.

Here, we propose and benchmark a novel approximate first-principles molecular dynamics simulation idea for increasing the computational efficiency of density functional theory-based calculations of the excite states. We focus on obtaining proton transfer energy at the S_1 excited state through actual density functional theory calculations at the T_1 state with additional optimized effective potentials. The potentials are optimized such as to reproduce the time-dependent density functional theory energy surface, but can be generalized to other more accurate quantum chemical methods. We demonstrate the applicability of this method for two prototypical photoacids, namely phenol and 7-hydroxyquinoline. We show that after optimizing the additional effective potentials for carbon, nitrogen, oxygen, and the acidic hydrogens, both thermodynamics and kinetics of proton dissociation reaction can be well reproduced as compared to reference excited-state calculations. It is found that a good agreement can be reached by only optimizing two effective potential parameters per each species in the photoacids.

O 82.5 Wed 13:30 P

Assessment of the Ab Initio Bethe-Salpeter Equation Approach for the Low-Lying Excitation Energies of Bacteriochlorophylls and Chlorophylls — •Zohreh Hashemi[1] and Linn Leppert[1,2] — [1]Institute of Physics, University of Bayreuth, Germany — [2]MESA+ Institute for Nanotechnology, University of Twente, Netherlands

Bacteriochlorophyll and Chlorophyll molecules are crucial building blocks of the photosynthetic apparatus in bacteria, algae and plants. In this contribution we assess the accuracy of ab initio many body perturbation theory within the GW approximation and Bethe-Salpeter equation (BSE) approach for calculating the electronic structure and optical excitations of seven members of this important family of light harvesting pigments. We compare our calculations with results from time-dependent density functional theory, multireference RASPT2 and experimental literature results. We find that optical excitations calculated with GW+BSE are in excellent agreement with experimental data, with an average deviation of less than 100 meV for the first three bright excitations of the entire family of (Bacterio)chlorophylls. Contrary to state-of-the-art TDDFT with an optimally-tuned range-separated hybrid functional, this accuracy is achieved in a parameter-free approach. Moreover, GW+BSE predicts the energy differences between the low-energy excitations correctly, and eliminates spurious charge transfer states that TDDFT with (semi)local approximations is known to produce.

O 82.6 Wed 13:30 P

microscopic modeling of rare-earth perovskite properties — •alireza sasani[1], jorge iniguez[2,3], and eric bousquet[1] — [1]Physique Théorique des Matériaux, QMAT, CESAM, Université de Liège, B-4000 Sart-Tilman, Belgium — [2]MaterialsResearch and Technology Department, Luxembourg Institute of Science and Technology (LIST), 5 avenue des Hauts-Fourneaux, L-4362, Esch/Alzette, Luxemburg — [3]Department of Physics and Materials Science, University of Luxembourg, Rue du Brill 41, L-4422 Belvaux, Luxembourg

Rare earth perovskites($RFeO_3$,$RCrO_3$ and R a rare earth element) have been studied for a long time due to having interesting magnetic behaviors,i.e magnetization reversal(Mr) and spin reorientation(SR). In this work, we shed some more light onto the magnetic properties of

the rare earth perovskites(RFeO's) by using density functional theory (DFT) to fit a microscopic Heisenberg model that includes the superexchange and the DMI between both Fe-Fe and Fe-R interactions. This model is used to do classical spin dynamics and it is employed as a global general model where the different parameters are tuned to understand their specific role in the (MR) and (SR) transitions. The results are also compared with analytical solutions to confirm the consistency of the spin dynamics solutions. Our works allow to explain the origin of the SR by determining the important parameters for the SR temperature interval and how the R magnetism is affected while it is in its paramagnetic regime. Our study also allows to fully understand the exact microscopic origin of the MR.

O 82.7 Wed 13:30 P

***Ab initio* phonon self-energies and fluctuation diagnostics of phonon anomalies: Lattice instabilities from Dirac pseudospin physics in transition metal dichalcogenides** — •Jan Berges[1], Erik van Loon[1], Arne Schobert[1], Malte Rösner[2], and Tim Wehling[1] — [1]Universität Bremen, Germany — [2]Radboud Universiteit Nijmegen, The Netherlands

We present an *ab initio* approach for the calculation of phonon self-energies and their fluctuation diagnostics, which allows us to identify the electronic processes behind phonon anomalies. Application to the transition-metal-dichalcogenide monolayer TaS_2 reveals that coupling between the longitudinal–acoustic phonons and the electrons from an isolated low-energy metallic band is entirely responsible for phonon anomalies such as the mode softening and associated charge-density waves observed in this material. Our analysis allows us to distinguish between different mode-softening mechanisms including matrix-element effects, Fermi-surface nesting, and Van Hove scenarios. We find that matrix-element effects originating from a peculiar type of Dirac pseudospin textures control the charge-density-wave physics in TaS_2 and similar transition metal dichalcogenides.