MM 34: Development of Calculation Methods

Atomic Structure, Quantum Effects

Time: Friday 10:15–12:15 Location: H10

MM 34.1 Fri 10:15 H10

Enhancing 3D Volume Reconstruction in Atom Probe Tomography through Curvature-Based Tip Shape Analysis — Sebastian Eich and •Guido Schmitz — Department for Materials Physics, Institute for Materials Science, University of Stuttgart

Atom Probe Tomography (APT) provides chemical analysis of nanometric volumes with single-atom sensitivity in 3D. Traditionally, reconstruction assumes a hemispherical tip shape, which can lead to significant distortions, especially due to local magnification effects.

Instead of relying on in-situ correlative microscopy, we propose a numerical method to extract the emitter shape from the event density statistics on the 2D detector plane. This method is based on the fundamental relationship between event density and the local Gaussian curvature of the tip surface, with no mathematical restrictions other than convexity. By knowing the curvature and assuming suitable boundary conditions, the surface profile can be uniquely reconstructed, bypassing the need for a hemispherical assumption.

The method is implemented as an easy-to-use Python module, which will be demonstrated using various simulated and experimental datasets involving complex tip shapes. This approach significantly reduces the local magnification effects at material interfaces with contrasting evaporation thresholds and is expected to improve the accuracy of atom probe reconstructions.

MM 34.2 Fri 10:30 H10

Sum frequency generation from real-time simulation in 2D crystals — •Mike Nico Pionteck[1], Myrta Grüning[2,3], Simone Sanna[1], and Claudio Attaccalite[3,4] — [1]Institut für Theoretische Physik and Center for Materials Research (LaMa), Justus-Liebig-Universität Gießen, Germany — [2]School of Mathematics and Physics, Queen's University Belfast, United Kingdom — [3]European Theoretical Spectroscopy Facilities (ETSF) — [4]CNRS/Aix-Marseille Université, Centre Interdisciplinaire de Nanoscience de Marseille UMR 7325 Campus de Luminy, France

Sum and difference frequency generation (SFG, DFG) are powerful experimental techniques which involve the interaction of two laser with frequency ω_1 and ω_2, generating an output beam with frequency $\omega = \omega_1 \pm \omega_2$ as second-order nonlinear response. These techniques are widely used to study 2D materials, providing complementary insights to those obtained from infrared and Raman spectroscopy.

We present an implementation of SFG and DFG within the Yambo code [1], based on real-time time-dependent adiabatic GW (TD-aGW). To account for local field effects and electron-hole interactions, our approach reduces to the Bethe-Salpeter equation (BSE) in the linear limit regime. We demonstrate this framework by calculating SFG and DFG signatures in h-BN and MoS_2. Furthermore, the method enables the extraction of higher-order response functions (e.g. TPA). This work establishes a first-principles approach to nonlinear optics, enabling investigations of optical responses of bulk materials with high versatility. [1] D. Sangalli *et al.*, J. Phys. Condens. Matter **31**, 325902 (2019).

MM 34.3 Fri 10:45 H10

Ab initio calculations of longitudinal electrical conductivity using a Wannier-based coherent potential approximation — •Shota Namerikawa and Takashi Koretsune — Department of Physics, Tohoku University, Sendai, Japan

We present a longitudinal electrical conductivity calculation method for disordered alloys applicable from a wide range of density functional theory (DFT) codes based on the first-principles Wannier-based coherent potential approximation (Wannier-CPA). For evaluation of electrical conductivity, we employ two complementary methods; the Kubo-Greenwood formula and numerical analytic continuation of the current-current correlation function. We apply the developed method to Ag-Pd alloys and find that the results obtained by the Wannier-CPA reasonably reproduce previous studies by the well-established CPA implementation based on the Korringa-Kohn-Rostoker Green*s function method (KKR-CPA).

MM 34.4 Fri 11:00 H10

Ab-initio-based analysis of phonon contributions to negative thermal expansion of a-tin — •Petr Čípek[1,2], Jana Pavlů[2], Martin Friák[1], and Alena Michalcová[3] — [1]Inst. Phys. Mater., Czech Acad. Sci., Brno, Czech Rep. — [2]Dept. Chem., Masaryk Uni., Brno, Czech Rep. — [3]Dept. Met. Corr. Eng., Uni. Chem. Technol. Prague, Czech Rep.

We applied quantum-mechanical calculations to investigate tin's thermodynamic and dynamic stability in its various allotropic forms. Focusing specifically on the a-Sn, our results showed a negative thermal expansion within the temperature range from T = 0 K to T = 45 K. No similar phenomenon appears in spectra of other allotropic modifications of tin. That means that the origin of this behaviour is connected to the diamond structure of alpha tin. We investigated the behaviour of the Grüneisen constant of a-tin in reciprocal space to explain the causes of this phenomenon. There are interesting temperature-dependent contributions of individual phonon modes from different k-points in the reciprocal space. In particular, these contributions are negative at all studied temperatures for the k-points around k-point X (0, 0, 0.5) and positive around k-point Γ (0, 0, 0). Negative thermal expansion of a-Sn at low temperatures is thus related to the behaviour of phonons around k-points X, M and N.

MM 34.5 Fri 11:15 H10

Bayesian Uncertainty Estimates for Spin-Component-Scaled Second-Order Møller-Plesset Perturbation Theory — •Elisabeth Keller and Johannes Margraf — Universität Bayreuth, Bayreuth, Germany

Spin-component-scaled second-order Møller-Plesset perturbation theory (SCS-MP2) improves upon MP2 by separately scaling the same-spin and opposite-spin MP2 contributions, achieving near coupled cluster (CCSD(T)) accuracy in some applications. However, the optimal scaling parameters vary by target domain, limiting the transferability of any given SCS-MP2 parameterization. To address this limitation, we employ a Bayesian multilevel linear regression model to obtain a robust parameterization for SCS-MP2, termed BSCS-MP2, predicting various energetic properties, including total, atomization, reaction, and non-covalent interaction energies. Additionally, the Bayesian model provides credible intervals to quantify the uncertainty of the BSCS-MP2 energy predictions. We evaluate how these uncertainty estimates adjust to data quality and model complexity, and assess their robustness for out-of-sample inference.

15 min. break

MM 34.6 Fri 11:45 H10

pyTTN, an open source toolbox for Quantum Dynamics simulations using Tree Tensor Network states — Lachlan Lindoy, •Daniel Rodrigo-Albert, Yannic Rath, and Ivan Rungger — National Physical Laboratory, Teddington, TW11 0LW, United Kingdom

The simulation of large-scale dissipative quantum systems is a significant challenge arising in several areas of physics and chemistry. In this talk we will discuss pyTTN, our recently developed software package for simulating dynamical properties of open quantum systems. This package makes use of Tree Tensor Network (or equivalently, Multi-Layer Multi-Configuration Time-Dependent Hartree) based representations of the state vector, and features both single- and multi-set ansätze, as well as adaptive bond dimension through subspace expansion techniques. The software has been designed with a focus on performance and ease of setup of new models and wavefunction topologies, including simple preparation of zero- and finite-temperature calculations with general bosonic, fermionic, and spin Hamiltonians. We will demonstrate the capabilities of the package with both unitary dynamics and non-unitary pseudomode-based approaches for the simulation of Anderson impurity models.

MM 34.7 Fri 12:00 H10

Spectral properties from an efficient analytical representation of the GW self-energy within a multipole approximation — •Dario Alejandro Leon[1], Kristian Berland[1], and Claudia Cardoso[2] — [1]Norwegian University of Life Sciences, As, Norway —

[2]S3 Centre, Istituto Nanoscienze, CNR, Modena, Italy

We propose an efficient analytical representation of the frequency-dependent GW self-energy via a multipole approximation (MPA-Σ). Similar to the earlier developed multipole approach for the screening interaction W (MPA-W) [Phys. Rev. B 104, 115157 (2021)], the multipole-Padé model for the self-energy is interpolated from a small set of values evaluated numerically in the complex frequency plane. As for MPA-W , we show that an appropriate choice of the frequency sampling is paramount to guarantee computational efficiency and high level of accuracy in the description of the self-energy. Crucially, MPA-Σ enables a multipole representation for the interacting Green's function G (MPA-G), providing straightforward evaluation of all the spectral properties. Combining the MPA-W and MPA-Σ schemes considerably reduces the cost of full-frequency self-energy calculations, especially when targeting spectral band structures in a wide energy range. We validate the MPA-Σ approach in bulk Si, Na and Cu, monolayer MoS2, the NaCl ion-pair and the F2 molecule, as prototypical semiconducting and metallic materials of different dimensionality. Moreover, toy MPA-Σ models with one and two poles and their corresponding MPA-G solutions, are used to examine the quasiparticle picture in different situations.