MM 41: Development of Calculation Methods II

Time: Friday 10:15–12:45 Location: SCH/A251

MM 41.1 Fri 10:15 SCH/A251

Atomistic Framework for Glassy Polymer Viscoelasticity Across 20 Frequency Decades — •Ankit Singh[1], Vinay Vaibhav[1,2], Caterina Czibula[3], Timothy W. Sirk[4], and Alessio Zaccone[1] — [1]Department of Physics “A. Pontremoli", University of Milan, Via Celoria 16, 20133 Milan, Italy — [2]Institut für Theoretische Physik, University of Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany — [3]Institute of Bioproducts and Paper Technology, Graz University of Technology, Inffeldgasse 23, 8010 Graz, Austria — [4]US Army DEVCOM Army Research Laboratory, Aberdeen Proving Ground, Maryland 21005, United States

Glassy polymers are central to engineering applications, yet their viscoelastic response over broad frequency and temperature ranges remains difficult to characterize. We extend non-affine deformation theory by incorporating a time-dependent memory kernel within the Generalized Langevin Equation for atomistic non-affine motions, yielding frequency-dependent mechanical response. Applied to poly(methyl methacrylate) (PMMA), the method captures the shear modulus and relaxation spectrum across more than twenty decades in frequency, from hundreds of terahertz down to the millihertz regime, thus bridging polymer mechanics from ordinary to extreme scales. Our predictions show quantitative consistency with independent estimates from oscillatory-shear molecular dynamics, Brillouin scattering, ultrasonic spectroscopy, Split-Hopkinson testing, and dynamic mechanical analysis (DMA), demonstrating a unified theoretical computational route for multiscale characterization of polymer glasses.

MM 41.2 Fri 10:30 SCH/A251

Clarifying the existence of a miscibility gap in platinum-palladium alloys via compositional fluctuations — •Florian Groll, Sebastian Eich, and Guido Schittz — Institute of Materials Science, Stuttgart, Germany

In materials science, the Gibbs free energy is of central importance for calculating phase diagrams. CALPHAD assessments require extensive experimental measurements and data obtained from well-equilibrated samples. For alloy systems with extremely low diffusion coefficients, achieving such equilibrium can be very time-consuming or practically impossible. Because this approach relies only on nanometer-scale equilibration, it provides a significantly more efficient route to the determination of CALPHAD parameters for systems with extremely slow diffusion. The palladium-platinum system is a well-known example for those kind of systems with unknown miscibility gap. In this study, we determine the Pd*Pt phase diagram by extracting the curvature of the Gibbs free energy from local composition fluctuations in the solid state using fluctuation theory. The experimental data needed for applying the local fluctuation theory are obtained from atom probe tomography, and the phase diagram is subsequently calculated.

MM 41.3 Fri 10:45 SCH/A251

Exploring the magnetic phase behaviour of MnAl with a novel sampling method, beginning from first principles — •Georgia A Marchant[1,2], M Nur Hasan[1], Rafael Vieira[1], Julie B Staunton[2], and Heike Herper[1] — [1]Uppsala University, Uppsala, Sweden — [2]University of Warwick, Coventry, United Kingdom

The demand for powerful magnetic materials is ever-increasing, as they dominate our modern technological landscape. Addressing rare earth (RE) criticality is crucial, so the search for viable permanent magnets that are RE-free and RE-lean continues. MnAl, with around 55% Mn content, has been a popular candidate for research in this regard. It transforms from the paramagnetic epsilon phase to the ferromagnetic tau phase when annealed appropriately, but the transition rate enhances under a surprisingly modest magnetic field. To shed light on this, we have computationally predicted the magnetic properties of MnAl across the transition.

The method uses data from first-principles calculations to build efficient atomic models of the phases' magnetic interactions. Thermodynamic properties are then determined with nested sampling (NS). Recently, NS has proven to be a powerful alternative to Markhov chain Monte Carlo sampling (MCMC). While NS has been used extensively in non-magnetic physical chemistry, this work represents its first application to magnetic materials. We present these results to demonstrate the power of NS; compare its effectiveness to MCMC; and unveil previously-unknown magnetic phases of MnAl.

MM 41.4 Fri 11:00 SCH/A251

A Demonstration of the Finite-Temperature Upside-Down Adiabatic Connection with the Asymmetric Hubbard Dimer — •Vincent Martinetto[1,2,3], Juri Grossi[1], and Aurora Pribram-Jones[1] — [1]University of California, Merced, CA 95343, United States — [2]Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany — [3]Center for Advanced Systems Understanding, 02826 Görlitz, Germany

The finite-temperature upside-down adiabatic connection is an integral expression for the decorrelation free energy, a quantity of interest when considering finite-temperature strictly correlated electrons. This approach smoothly connects a thermal ensemble of strictly correlated electrons to an ensemble with realistic interaction strength while requiring the density of the system to remain fixed across all interaction strengths. As the strictly correlated limit is the natural opposite of the Kohn-Sham limit, it can provide new insights into the difficult regime of strong correlation in finite-temperature systems. An exact demonstration of the upside-down finite-temperature adiabatic connection will be performed with the asymmetric Hubbard dimer, along with a discussion of strong correlation in finite-temperature systems.

MM 41.5 Fri 11:15 SCH/A251

Towards self-consistent electrostatic corrections for charged defects in periodic boundary conditions — •Ermanno Botti[1,2,3], Davide Campi[3], Nicéphore Bonnet[2], and Nicola Marzari[1,2] — [1]PSI, Switzerland — [2]EPFL, Switzerland — [3]UniMiB, Italy

Charged point defects in semiconductors and insulators play a pivotal role in determining a wide range of physical properties, significantly impacting materials behavior and device performance. First principles calculations have become an essential tool to better characterize defects properties and to speed up technological advancements. However, some key defect properties, such as formation energies in the dilute limit, are highly sensitive to finite size effects in periodic boundary conditions DFT simulations. These issues arise from long range Coulomb interactions between periodic replicas and the neutralizing background needed to prevent the divergence of the electrostatic potential. To reduce these effects, corrections are required to remove these spurious electrostatic interactions. In this work we extend an a-posteriori correction scheme to achieve self-consistency in the open access package Quantum Espresso and test it on simple systems.

15 min. break

MM 41.6 Fri 11:45 SCH/A251

Tessellation-free full charge density method for atom-centered basis sets — •Franco Moitzi[1], Andrei V. Ruban[2,3], Vsevolod I. Razumovskiy[1], and Oleg E. Peil[2] — [1]Christian Doppler Laboratory for Digital material design guidelines for mitigation of alloy embrittlement, Materials Center Leoben Forschung GmbH, Vordernberger Straße 12, A-8700 Leoben, Austria — [2]Materials Center Leoben Forschung GmbH, Vordernberger Straße 12, A-8700 Leoben, Austria — [3]Department of Materials Science and Engineering, Royal Institute of Technology, 10044 Stockholm, Sweden

Accurate Poisson solvers are essential for reliable total energies and forces in Density Functional Theory (DFT) calculations. Atom-centered basis methods, such as Korringa-Kohn-Rostoker methods, often rely on Voronoi tessellations to decompose space, which complicate Poisson solutions in the interstitial region due to the high l-cutoffs required for the nearly flat charge there and the boundary discontinuities, leading to numerical instabilities as atoms move. We introduce a tessellation-free method that avoids these issues by decomposing the charge into non-overlapping atomic spheres and an interstitial region.

Following Andersen *et al.*, the interstitial charge is expanded in screened spherical waves—Helmholtz-equation solutions with special boundary conditions—and smoothly extended from the atomic spheres, allowing the Poisson equation to be solved analytically. Their analytical properties allow accurate evaluation of the Hartree potential, electrostatic, and exchange-correlation energies, enabling precise and efficient DFT calculations with atom-centered basis sets.

MM 41.7 Fri 12:00 SCH/A251

Machine-Learning Augmented Electronic Structure Calculations via Adaptive DFTB Parameters

— •Yihua Song, Christoph Scheurer, Karsten Reuter, and Chiara Panosetti — Fritz Haber Institut der MPG

As a semi-empirical DFT approximation, Density Functional Tight Binding (DFTB) combines reduced computational cost with accessible electronic structure, providing a compromise between the full physicality of *ab initio* methods and the speed of machine-learning interatomic potentials. Yet, its accuracy in realistic systems remains limited due to intrinsic approximations and dependence on parametrization quality. To this end, we propose that the precalculated Slater-Koster (SK) electronic interaction tables in DFTB should be adapted to local atomic environments and demonstrate proof-of-principle studies showing the improved electronic structure description. Building on the observed smoothness of SK integrals across chemical and spatial environments, we use machine learning based on atomic descriptors to continuously adapt the parametrization beyond discrete atomic types. As a prototype, we introduce the DFTB Orbital Verity Engine (DOVE), a machine-learning framework that learns subtle discrepancies in electronic parameters across diverse local environments from reference electronic structures. We successfully validate it against complicated systems with multiple coordination states. This machine-learning framework for environment-adaptive tight-binding parameterization enables scalable electronic structure simulations with enhanced fidelity, efficient speed, and physical interpretability.

MM 41.8 Fri 12:15 SCH/A251

Detecting topologically-protected surface states in doped systems using the coherent potential approximation

— •Artem Chmeruk, Dylan Jones, and Liviu Chioncel — University of Augsburg, Augsburg, Germany

The signature of the non-trivial electronic band structure is the existence of gapless conducting states at interfaces where the topological invariant changes. This change signals a topological phase transition, which in real systems can be accomplished through doping. Modeling doping effects can be done by combining density functional theory and coherent potential approximation, which gives the possibility to model a continuous range of concentrations, and provides a sensitive probe to detect the emergent electronic states. Here, we analyse the topologically-protected surface states in $Bi_{2-x}Sb_xSe_3$.

MM 41.9 Fri 12:30 SCH/A251

Defect-induced vibrational modes in graphene

— •Iskander Mukatayev, Guillaume Radtke, and Michele Lazzeri — Sorbonne Université, CNRS UMR 7590, MNHN, IMPMC, 4 place Jussieu, 75252 Paris, France

We present a general framework for investigation of the vibrational properties of point defects in graphene, motivated by recent advances in vibrational EELS that enable phonon spectroscopy with atomic resolution. Our approach combines DFT and density-functional perturbation theory with a force-matching procedure [1-2], in which interatomic force constants (IFCs) are computed independently for pristine graphene and defect-containing supercells and then merged to dynamical matrices of very large systems(up to 20000 atoms). This strategy enables the modeling of isolated defects without defect-defect interactions while retaining full ab initio accuracy.

We investigate a wide range of point defects including substitutional dopants (B, N, Si, P), single, double vacancies and Stone-Wales reconstruction. Depending on the details of defect-induced bond reconstruction, different behaviours are observed ranging from quasi-localized (or resonant) states falling in the continuum of the bulk to high-frequency fully localised modes. From these examples we draw conclusions of the conditions of occurrence of both types vibrations.

References

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2. G. Radtke, S. Klotz, M. Lazzeri, P. Loubeyre, M. Krisch, A. Bossak, Phys. Rev. Lett. 132, 056102 (2024)