MM 27: Transport in Materials: Diffusion, Charge or Heat Conduction

Machine Learning, Quantum Theory

Time: Thursday 10:15–13:00 Location: H22

MM 27.1 Thu 10:15 H22

Modelling Heat Transport in Metal-Organic Frameworks with Machine Learned Potentials — •Martin Klotz[1], Florian Lindner[1], Sandro Wieser[2], and Egbert Zojer[1] — [1]Institute of Solid State Physics, Graz University of Technology, Austria — [2]Institute of Materials Chemistry, TU Wien, Austria

Many of the envisioned applications of porous metal-organic frameworks (MOFs), like gas storage or catalysis, involve exothermal processes. This requires the materials to efficiently dissipate heat. It is, thus, vital to gain a fundamental understanding of how structural and chemical modifications impact heat-transport in such systems. Here, we chose MOF-74/Zn as parent system for which we calculated the anisotropic thermal conductivity using approach to equilibrium molecular dynamics simulations. For these, we applied machine-learned force field potentials, as they surpass DFT simulations by orders of magnitudes in terms of speed and classically parametrized force field potentials in terms of accuracy. Subsequent to thoroughly benchmarking and testing the employed methodology, we studied structure-to-property relationships for MOF-74 derivatives, systematically varying the node metal and the linker structure.

MM 27.2 Thu 10:30 H22

Predicting 2D conventional superconductors — Thalis H. B. da Silva[1], Théo Cavignac[2], Tiago F. T. Cerqueira[1], •Hai-Chen Wang[2], and Miguel A. L. Marques[2] — [1]CFisUC, Department of Physics, University of Coimbra, Rua Larga, 3004-516 Coimbra, Portugal — [2]Universitätsstr. 150

We perform a large-scale search for two-dimensional (2D) superconductors by using electron-phonon calculations with density-functional perturbation theory combined with machine learning models. In total, we screened over 140k 2D compounds from the Alexandria database. Our high-throughput approach revealed a multitude of 2D superconductors with diverse chemistries and crystal structures. Moreover, we find that 2D materials generally exhibit stronger electron-phonon coupling than their 3D counterparts, although their average phonon frequencies are lower, leading to an overall lower transition temperature (T_c). Despite this, we discovered several out-of-distribution materials with relatively high T_c. In total, 105 2D systems were found with $T_c > 5$ K. Some interesting compounds, such as CuH_2, NbN, and V_2NS_2, demonstrate high T_c values and good thermodynamic stability, making them strong candidates for experimental synthesis and practical applications. Our findings highlight the critical role of computational databases and machine learning in accelerating the discovery of novel superconductors.

MM 27.3 Thu 10:45 H22

Modelling complex proton transport phenomena - Exploring the limits of fine-tuning and transferability of foundational machine-learned force fields — •Christian Dressler[1], Malte Grunert[2], Jonas Hänseroth[1], Max Grossmann[2], and Erich Runge[2] — [1]TU Ilmenau, Institute of Physics, Theoretical Solid State Physics — [2]TU Ilmenau, Institute of Physics, Group of Theoretical Physics 1

The solid acids CsH_2PO_4 and $Cs_7(H_4PO_4)(H_2PO_4)_8$ pose significant challenges for the simulation of proton transport phenomena. In this talk, we present the use of the recently developed machine-learned force field MACE to model proton dynamics on nanosecond timescales for these systems and compare its performance with long-term ab initio molecular dynamics (AIMD) simulations. The MACE-MP-0 foundation model shows remarkable performance for all observables derived from molecular dynamics simulations, but minor quantitative discrepancies remain compared to the AIMD reference data. However, we show that minimal fine-tuning - fitting to as little as 1 ps of AIMD data - leads to full quantitative agreement between the radial distribution and autocorrelation functions of MACE force field and AIMD simulations. Long-time AIMD simulations are unable to capture the correct qualitative trends in diffusion coefficients due to their inherent time scale limitations. In contrast, we demonstrate that accurate and convergent diffusion coefficients, consistent with experimental data, can only be reliably achieved through multi-nanosecond molecular dynamics simulations utilizing machine-learned force fields.

MM 27.4 Thu 11:00 H22

Understanding thermal transport in organic semiconductors using machine learned force fields — •Florian Unterkofler[1], Lukas Reicht[1], Lukas Legenstein[1], Sandro Wieser[2], Michele Simoncelli[3], and Egbert Zojer[1] — [1]Graz University of Technology, Austria — [2]TU Wien, Austria — [3]Columbia University, New York (USA)

Organic semiconductors (OSCs) are key materials for optoelectronic devices such as solar cells and organic light-emitting diodes (OLEDs). While the properties related to charge transport of OSCs are relatively well understood, we still lack an understanding of the fundamentals of the heat transport in those materials. To study the atomistic origins of heat transport, we developed a strategy for calculating the thermal conductivity of complex organic crystals employing non-equilibrium molecular dynamics (NEMD) simulations with highly accurate, system-specific, machine-learned Moment Tensor Potentials (MTPs). These MTPs are trained on ab initio data obtained from on-the-fly active-learning molecular dynamics simulations.[1]

We then simulated the thermal transport in pentacene with NEMD to analyze the heat conduction in real space at an atomistic level and to identify heat-transport bottlenecks. Alternatively, we also use the MTPs to accurately calculate thermal conductivities arising from the particle-like propagation and the wave-like tunneling of phonons in reciprocal space. Both approaches are consistent and agree with available experiments.

[1] npj Comput Mater 10, 18 (2024)

MM 27.5 Thu 11:15 H22

Influence of Defects and Layer Twisting on Phonon Dynamics in Bilayer Graphene and MoS_2 Using Machine Learned-Force Field Calculations — •Sabuhi Badalov[1,2] and Harald Oberhofer[1,2] — [1]Department of Physics, University of Bayreuth — [2]Bavarian Center for Battery Technology, Bayreuth, Germany

In crystalline materials, thermal, mechanical and even electronic properties are often described in terms phonon spectra and dynamics. These, can, to a large degree be influenced by even minute structural changes, such as through defects or by altering the relative alignment of a material's layers. By means of a state-of-the-art machine learning-augmented force field approach, we carry out extensive phonon calculations to understand how the defect density and layer configurations influence the phonon spectra in bilayer graphene and MoS_2. Our results highlight an interplay of phononic phenomena with the materials' Moiré patterns with major consequences on the transport properties of these materials. Moreover, we explore the possibility of phonon-mediated superconductivity through the electron-phonon coupling in topologically nontrivial phonon states. While this is still ongoing research, our results so far not only further the understanding of phonon dynamics in 2D materials, but also offer a solid foundation for future investigations into harnessing topological phonons for advanced electronic and thermal control technologies.

15 min. break

MM 27.6 Thu 11:45 H22

Thermoelectric quantum transport simulations via the time-linear nonequilibrium Green's function method — •Riku Tuovinen[1] and Yaroslav Pavlyukh[2] — [1]Department of Physics, University of Jyväskylä, Finland — [2]Institute of Theoretical Physics, Wroclaw University of Science and Technology, Poland

Thermoelectric transport focuses on understanding charge and heat flow in quantum systems. While measuring electron current is relatively straightforward, nanoscale heat flux remains challenging to quantify [1]. Heat and charge exhibit fundamental differences, as highlighted by electron transport analysis within the nonequilibrium Green's function theory [2]. In the time-linear formulation, based on the generalized Kadanoff-Baym ansatz (GKBA), open system dynamics are described using an embedding correlator, enabling the calculation of time-dependent currents via the Meir-Wingreen formula [3].

However, calculating heat currents presents challenges, particularly within the wide-band limit approximation (WBLA), which can result in divergent energy integrals. Besides the mathematical issues, the WBLA's reliability depends on the physical properties of the leads. To address these limitations, we present thermoelectric quantum transport simulations using the time-linear GKBA method without relying on the WBLA [4].

[1] J. P. Pekola and B. Karimi, Rev. Mod. Phys. 93, 041001 (2021).
[2] M. Ridley et al., J. Phys. A: Math. Theor. 55, 273001 (2022).
[3] R. Tuovinen et al., Phys. Rev. Lett. 130, 246301 (2023).
[4] R. Tuovinen and Y. Pavlyukh, in preparation (2024).

MM 27.7 Thu 12:00 H22

Sublinear in temperature transport in kagome metals: interplay of Dirac cones and Van Hove singularities — •Nikolai Peshcherenko[1], Ning Mao[1], Claudia Felser[1], and Yang Zhang[2,3] — [1]Max Planck Institute for Chemical Physics of Solids, 01187, Dresden, Germany — [2]Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996, USA — [3]Min H. Kao Department of Electrical Engineering and Computer Science, University of Tennessee, Knoxville, Tennessee 37996, USA

Kagome metals are known to host Dirac fermions and saddle point Van Hove singularities near Fermi level. With the minimal two-pocket model (Dirac cone + Van Hove singularity), we propose a semiclassical theory to explain the experimentally observed sublinear resistivity in Ni_3In and other Kagome metals. We derive the full semiclassical description of kinetic phenomena using Boltzmann equation, and demonstrate that internode electron-electron interaction leads to sublinear in T scaling for both electrical and thermal transport at low temperatures. At higher temperatures above the Dirac node chemical potential, thermal and electric currents dissipate through distinct scattering channels, making a ground for Wiedemann-Franz law violation.

MM 27.8 Thu 12:15 H22

Phonon induced heat transfer between gold nanogap electrodes — •Yuki Hanamura, Kazuma Kishimoto, Mizuki Tada, Ryo Yamada, and Hirokazu Tada — Graduate School of Engineering Science, Osaka University, Japan

Recent studies have observed anomalous increases in thermal conductance in nanogaps narrower than a few nanometers [1]. Theoretical models attribute this phenomenon to electron tunneling effects and propagation of lattice vibrations due to mechanical interactions between the electrodes, i.e., phonon transport [2]. Experimental difficulties in the nanoscale have prevented us from sufficiently understanding the heat transfer mechanisms.

We have developed a device to measure both thermal and electrical conductance across nanogap electrodes. The device comprises a suspended structure with Micro Electro Mechanical System (MEMS) based actuators to tune the gap distance. We observed increased thermal conductances in sufficiently separated nanogaps, even in the absence of electron tunneling, suggesting phonon transport as the dominant mechanism [3]. We will show the effects of electrode gap distance and temperature on heat transfer across the nanogap.

[1] M. Pascale et al., Appl. Phys. Lett. 122, 100501 (2023).
[2] T. Tokunaga et al., Phys. Rev. B 104, 125404 (2021).
[3] Y. Hanamura et al., Nanoscale, in press.

MM 27.9 Thu 12:30 H22

Hydrodynamics of Lorentz symmetric systems: a quantum Monte Carlo study — •Adrien Reingruber[1], Kitinan Pongsangangan[2], Fakher Assaad[1], and Maksim Ulybyshev[1] — [1]Universität Würzburg, Würzburg, Germany — [2]Mahidol University, Bangkok, Thailand

We present a study on the hydrodynamic behavior of charge current in a Lorentz symmetric sys- tem: graphene at charge-neutrality. The momentum flow is completely decoupled from the charge current in this regime, since the electrons and holes propagate in opposite directions with exactly equal distribution functions. Instead of Navier-Stokes equations for the velocity field, we derive sim- ilar equations directly for the charge current. This eliminates the need for any coupling between the velocity field and charge current to explain the experimentally observed hydrodynamic flow profiles in graphene at half-filling. In this framework, the current diffusion coefficient replaces viscosity. To support this, we performed an extensive quantum Monte Carlo study, directly simulating samples with disordered edges using the underlying microscopic interacting quantum Hamiltonian. For the first time, we observe hydrodynamic behavior of the charge current in such simulations, extract- ing current profiles and a current diffusion coefficient whose temperature dependence qualitatively agrees with predictions from Boltzmann transport theory.

MM 27.10 Thu 12:45 H22

Transport coefficients of Weyl semimetals: the contribution of plasmons — •Kitinan Pongsangangan — Mahidol University, Bangkok, Thailand

This work investigates the contribution of plasmon, a collective mode arising from the dynamical screening of the long-range coulomb interaction, to thermo-electric responses as well as shear viscosity of Weyl semimetals using Boltzmann-equation approach. We find that plasmons make a noticeable contribution to the thermal conductivity and shear viscosity in an appropriate temperature window. We propose that this effect could be potentially observed in, for example, TaAs and NbAs.