HL 28: Focus Session: Heat transport at the nanoscale: theory meets experiment

Tailoring heat transport is fundamental in the design of semiconducting devices. Over the last decades, tremendous progress has been made in understanding the atomistic mechanisms that determine heat transport at the macro- and the nanoscale, as well as at interfaces. For the latter case, low-dimensional and non-epitaxial materials can lead to ultra-low, directional or topological phonon transport. Largely, these advances were enabled by the development of novel experimental nanoscale measurements and improved first-principles techniques covering the different transport regimes. The focus session brings together international experts that have been driving this progress to discuss their most recent advancements.

Organized by Christian Carbogno, Saskia Fischer, and Markus Wagner

Time: Wednesday 9:30–13:00 Location: EW 203

Invited Talk HL 28.1 Wed 9:30 EW 203

Green-Kubo lattice dynamics approach to thermal transport in strongly anharmonic materials — •Ivana Savić — King's College London, UK

Over the last 15 years, there has been a great progress in the development of theoretical and computational tools to describe lattice thermal conductivity in realistic materials from first principles. Standard approaches are based on the phonon Boltzmann transport equation and a perturbative description of phonon-phonon interactions, including only third order anharmonicity. As a result, they are appropriate only for weakly anharmonic materials. In this talk, I will present a new method to simulate lattice thermal transport in strongly anharmonic materials, based on the Green-Kubo formalism and a non-perturbative treatment of phonon-phonon interactions [1]. I will also present the application of this method to understand the lattice thermal conductivity of a well-known thermoelectric material, GeTe, near the ferroelectric phase transition.

[1] D. Dangic, O. Hellman, S. Fahy, and I. Savic, npj Comp. Mater. 7, 57 (2021)

Invited Talk HL 28.2 Wed 10:00 EW 203

Hybrid crystal-glass heat conduction and radiative effects in disordered solids — •Michele Simoncelli — Theory of Condensed Matter Group of the Cavendish Laboratory, University of Cambridge

At ordinary temperatures, crystals and glasses exhibit opposite thermal conductivities upon heating: decreasing in the former and increasing in the latter. At extreme temperatures, instead, experiments in crystalline and glassy polar dielectrics show a qualitatively similar strong enhancement in their conductivities, which departs from predictions obtained using state-of-the-art heat-conduction theories. Here, we employ and extend the Wigner formulation of thermal transport to shed light on the microscopic physics determining heat transfer in crystals and glasses over a broad temperature range. First, we show that at ordinary temperatures, the magnitude and trend of the conductivity can be engineered through the degree of disorder in the atomistic bond topology, geometry, or composition of a solid, allowing for the emergence of hybrid crystal-glass conductivity trends. Second, we extend the Wigner formulation to account for phonon-photon couplings; relying on such framework, we show from first principles that the conductivity enhancement in polar dielectrics at extreme temperatures originates from an interplay between conductive and radiative heat transfer and is regulated by atomistic disorder.

Invited Talk HL 28.3 Wed 10:30 EW 203

Engineering and probing phonons and thermal transport — •Ilaria Zardo[1,2], Begoña Abad[1], Chaitanya Arya[1], Giulio de Vito[1], Yashpreet Kaur[1], Dominik M. Koch[1], Grazia Raciti[1], Aswathi K. Sivan[1], Jose M. Sojo[1], and Johannes Trautvetter[1] — [1]Department of Physics, University of Basel, Basel, Switzerland — [2]Swiss Nanoscience Institute, University of Basel, Basel, Switzerland

The recently growing research field called "Nanophononics" deals with the investigation and control of vibrations in solids at the nanoscale. Phonon engineering leads to a controlled modification of phonon dispersion, phonon interactions, and transport. However, engineering and probing phonons and phonon transport at the nanoscale is a non-trivial problem.

In this talk, we discuss how phononic properties and thermal transport can be engineered and measured in nanowires and the challenges and progresses in the measurement of the thermal conductivity of nanostructures and low dimensional systems. We experimentally show that a controlled design of the nanowires' phononic properties can be decided à la carte by tuning the superlattice period. We also investigated thermal rectification in semiconducting gallium arsenide nanowires with an abrupt change in diameter, also called telescopic nanowires. We measured rectification values ranging from 2 to 7% at a range of ambient temperatures. Finally, Raman thermometry is used to probe the temperature profile in nanostructures upon application of a thermal gradient, enabling the differentiation between different thermal transport regimes.

15 min. break

Invited Talk HL 28.4 Wed 11:15 EW 203

Challenges and opportunities of thermally anisotropic materials — •Sebastian Reparaz — Materials Science Institute of Barcelona (ICMAB)

The study of the thermal conductivity (or diffusivity) tensor (κij) in bulk and low dimensional materials has gained considerable momentum in recent years. A large number of experimental methods to study the out-of-plane components of the thermal conductivity have been developed and successfully demonstrated using different methodologies, e.g., based on electrical or optical methods. On the other hand, the study of in-plane thermal transport is comparatively more challenging due to the lack of sensitivity to this component of most developed methods, among other reasons. I will discuss two complementary experimental approaches recently developed with enhanced sensitivity to thermal anisotropy and, in particular, to in-plane thermal transport, which are based on using a 1D heat source with uniform power distribution along its long axis. I will show the application of these recently developed methodologies to study the thermal properties of a large variety of samples, with special focus on determining the thermal conductivity tensor elements. In particular, I will address the following list of materials: β-Ga2O3; highly oriented pyrolytic graphite (HOPG); suspended silicon and polymer membranes with different thicknesses; bismuth, silicon, glass, AlN, GaN, ZnO, and ZnS substrates; and several Van der Waals materials such as PdSe2, hence, demonstrating their excellent performance and rather simple data analysis procedure.

HL 28.5 Wed 11:45 EW 203

Thermal transient grating, measuring thermal anisotropies on ultra-short length scales — •Luca Sung-Min Choi[1], Moritz Meissner[1], Alwin Wüthrich[1], Kai Xu[2], Riccardo Mincigrucci[3], Laura Foglia[3], Danny Fainozzi[3], Filippo Bencivenga[3], Sebastian Reparaz[2], and Markus R. Wagner[1,4] — [1]Technische Universität Berlin, Berlin, Germany — [2]Institut de Ciència de Materials de Barcelona, Barcelona, Spain — [3]Elettra Sincrotrone Trieste, Trieste, Italy — [4]Paul-Drude-Institut für Festkörperelektronik, Berlin, Germany

Optical transient thermal grating (TTG) is a contactless method to determine anisotropies of thermal transport and acoustic phonons. These physical properties can be observed by fluctuations of the complex diffraction index. Using extreme UV emission of free electron lasers (FELs), ultra-short grating lengths ranging from 110 to 26 nm can be realized. In order to illustrate the possibilities of this method using FELs as emission sources, we exemplary introduce our investigation of the strong anisotropy in the beta-phase of gallium oxide. The experiments were conducted at the Fermi Free Electron Laser [Elettra Synchrotron Trieste, dedicated endstation EIS-TIMER] on a (001)-$\beta - Ga_2O_3$ sample.

HL 28.6 Wed 12:00 EW 203

Ballistic phonon transport in ß-Ga2O3 — •Rüdiger Mitdank[1], Robin Ahrling[1], Andreas Popp[2], Jana Rehm[2], Arub Akhtar[2], Zbigniew Galazka[2], and Saskia Fischer[1,3] — [1]Novel Materials Group, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — [2]Leibniz Institut für Kristallzüchtung, 12489 Berlin, Germany — [3]CSMB, Humboldt-Universität zu Berlin, 12489 Berlin, Germany

The anisotropic thermal conductivity and the phonon mean free path (mfp) in monoclinic ß-Ga2O3 single crystals and homoepitaxial films of several micron were determined using the 3-ω method in the temperature range from 10 K-300 K. Analysis of the phonon mfp shows a dominance of phonon-phonon-Umklapp scattering above 80 K, below which the influence of point-defect scattering is observed. Below 30 K the phonon mfp increases and the dominance of boundary effects and the crossover from resistive to ballistic phonon transport is observed. The measured effective thermal conductivity reaches a maximum of 1000-2000 W/(mK) and decreases with T^3 below 25 K. The resistive and ballistic phonon transport regimes in ß-Ga2O3 are discussed corresponding to the models of Callaway (resistive transport - Fourier limit) and Majumdar (phonon radiative transport - Casimir limit), respectively. The Casimir limit is achieved in very pure Ga2O3 single crystals and homoepitaxial layers.

1. A. Majumdar, Journal of Heat Transfer, 115, 7, 1993; 2. Callaway, J, Phys. Rev. 113, 1046, 1959; 3. H. Casimir, Physica, 5, no. 6, 495, 1938

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HL 28.7 Wed 12:15 EW 203

anisotropic thermal conductivity studied with the *ab initio* Green-Kubo approach: the example of Ga_2O_3 — •Shuo Zhao, Thomas A. R. Purcell, Kisung Kang, Matthias Scheffler, and Christian Carbogno — The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin

Anisotropic heat transport is important in thermoelectric materials and electronic devices since the efficiency of heat dissipation depends on the crystal axis. [1] The anisotropic thermal conductivity is often addressed in the phonon picture, but the role of (strong) anharmonicity is rarely discussed. To clarify this question, we employ the *ab initio* Green-Kubo method, [2] which accounts for all anharmonic effects through *ab initio* molecular dynamics. By extending its applicability to anisotropic transport, we discuss the thermal conductivity for the β, α, and κ-phase of Ga_2O_3, promising candidate materials for field-effect transistors. Our calculations show reasonable agreement with the experiment and allow us to explain the observed anisotropic transport. In particular, we discuss the role of strong anharmonic effects beyond the phonon picture at higher temperatures. In this regard, we also analyze the role of the employed exchange-correlation functional. This study paves the way for exploring the anisotropic, strongly anharmonic systems.

[1] C. Chang *et al.*, *Science* **360**, 778 (2018).
[2] C. Carbogno, R. Ramprasad, and M. Scheffler, *Phys. Rev. Lett.* **118**, 175901 (2017).

HL 28.8 Wed 12:30 EW 203

Computing Green-Kubo Thermal Conductivities with Semi-Local Machine-Learning Potentials — •Marcel F. Langer[1,2,3,4], Florian Knoop[4,5], J. Thorben Frank[2,3], Christian Carbogno[4], Matthias Scheffler[4], and Matthias Rupp[6] — [1]COSMO Laboratory, EPFL, Lausanne, Switzerland — [2]BIFOLD, Berlin, Germany — [3]ML Group, TU Berlin, Germany — [4]NOMAD Laboratory at the FHI of the Max Planck Society and IRIS Adlershof of HU Berlin, Germany — [5]Theoretical Physics Division, IFM, Linköping University, Sweden — [6]Materials Research and Technology Dept., Luxembourg Institute of Science and Technology, Luxembourg

The Green-Kubo method is a rigorous framework for heat transport simulations in materials, but requires an accurate description of the potential-energy surface and converged statistics. In this context, machine-learning potentials can achieve the accuracy of first-principles methods while allowing to reach well beyond their simulation time and length scales at a fraction of the cost. Recently developed potentials can include equivariant semi-local interactions through message-passing mechanisms and use automatic differentiation to obtain derivatives. We explain how to define and efficiently implement the heat flux for such potentials [1]. Based on this, we present a framework for running GPU-accelerated Green-Kubo calculations with machine-learning potentials, and demonstrate its use through the calculation of the thermal conductivity of several solid semiconductors and insulators.

[1]: M.F. Langer et al., Phys. Rev. B 108, L100302 (2023); M.F. Langer et al., J. Chem. Phys. 159, 174105 (2023)

HL 28.9 Wed 12:45 EW 203

Quantitatively accurate description of heat transport in metal-organic frameworks — Sandro Wieser, Florian Lindner, Lukas Legenstein, Lukas Reicht, and •Egbert Zojer — Institute of Solid State Physics, Graz University of Technology, Graz, Austria

Metal-organic frameworks (MOFs) comprise a highly porous class of materials consisting of metal-oxide nodes and organic linkers. They are envisioned for a wide variety of applications, many of which involve the generation or consumption of thermal energy. Therefore, understanding their heat-transport properties is of crucial importance. Unfortunately, MOFs are typically so complex (containing dozens or even hundreds of atoms in their unit cells), that an ab initio-based simulation of thermal transport appears impossible. Also conventional, transferable force fields are not suitable for the task, as they yield thermal conductivity values far from experiments. In the current contribution we show that the situation can be resolved employing on-the-fly-trained, machine-learned potentials (MLPs), which enable an essentially ab initio-quality description of phonon properties of MOFs at computational costs reduced by many orders of magnitude. Interestingly, with accurate MLPs at hand, various applied methodologies (NEMD, AEMD, Green-Kubo MD, as well as lattice dynamics) yield equivalent results in quantitative agreement with experimental, single crystal data. A similar situation is encountered for polymers and molecular crystals. The results pave the way for a reliable, atomistic understanding of heat transport in the said materials.