Eugene Wigner Colloquium

event of SFB 910

Christian Carbogno

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Habilitation colloquium

"The Key Role of Lattice Anharmonicity for Heat and Charge Transport in Solids: Fundamental Concepts, Novel Methods, and Relevant Applications"

The Colloquium will take place online via Zoom. For information on how to access the event, please contact: henning.reinken@itp.tu-berlin.de

Thursday, 12.11.20 · 16:15h

Technische Universität Berlin · Institut für Theoretische Physik · Hardenbergstraße 36 · 10623 Berlin

www.itp.tu-berlin.de/sfb910

Eugene Wigner Colloquium

event of SFB 910

The Key Role of Lattice Anharmonicity for Heat and Charge Transport in Solids: Fundamental Concepts, Novel Methods, and Relevant Applications

Heat and charge transport play a key role in materials science and thus for many technological applications that are key to establish a sustainable energy economy and ecology. Examples include improving the fuel-efficiency of aeronautic turbines [**1**], for developing efficient thermoelectric devices able to recover useful voltage from otherwise wasted heat [**2**], and for designing novel battery materials for advancing e-mobility [**3**]. To understand the microscopic mechanisms that drive or hinder heat and charge transport in the required functional materials under actual operational temperatures above 300 K, a quantitatively reliable description of the interatomic interactions is essential. For this purpose, we explore material space using a recently developed measure for anharmonicity [**4**], i.e., a metric for the strength of those interatomic interactions that are neglected in the harmonic approximation. This reveals that largely harmonic materials are the exception to the rule under realistic conditions and that strong anharmonic effects that cannot be accounted for within the harmonic picture – not even by perturbative corrections on top of it – are not uncommon at all. Clearly, *ab initio* molecular dynamics (aiMD) is the computational technique of choice to overcome these hurdles, since it correctly accounts for all orders of anharmonicity.

To exemplify, we discuss how the derivation of a gauge-independent vibrational heat flux definition enables to exploit the fluctuation-dissipation theorem and to perform *ab initio* Green-Kubo calculations of lattice thermal conductivities up to the melting point [**5**]. In turn, this allows a qualitative understanding of the mechanisms active under these thermodynamic conditions and opens up opportunities for tailoring them, e.g., to improve the insulation of thermal-barrier coatings in airplane turbines [**6**]. Furthermore, we discuss how the presented concepts can be generalized so to not only capture vibrational couplings between nuclei, but also vibronic ones, i.e., the ones between electronic and nuclear degrees of freedom. As demonstrated for the perovskite $SrTiO_3$, perturbative expansions also break down for the description of vibronic effects, leading to a severe overestimation of the electronic band-gap at elevated temperatures [**7**]. Eventually, we discuss the implications of these findings for the assessment of charge transport and explore routes towards a non-perturbative theory of electronic charge and heat conductivities in semiconductors. This then enables a fully parameter-free, quantitative theory of thermoelectricity.

[**1**] A. G. Evans, D. R. Clarke, and C. G. Levi, *J. Eur. Ceram. Soc.* **28**, 1405 (2008).
[**2**] G. J. Snyder and E. S. Toberer, *Nature Materials* **7**, 105 (2008).
[**3**] Y. Liu, Y. Zhu, and Y. Cui, *Nature Energy* **4**, 540 (2019).
[**4**] F. Knoop, T. A. R. Purcell, M. Scheffler, and C. Carbogno, *Phys. Rev. Mat.* **4**, 083809(2020).
[**5**] C. Carbogno, R. Ramprasad, and M. Scheffler, *Phys. Rev. Lett.* **118**, 175901 (2017).
[**6**] C. Carbogno, C. G. Levi, C. G. Van de Walle, and M. Scheffler, *Phys. Rev. B* **90**, 144109(2014).
[**7**] M. Zacharias, M. Scheffler, and C. Carbogno, *Phys. Rev. B* **102**, 045126 (2020).