MM 45: Computational Materials Modelling VI - Thermal Conductivity and Transport

Time: Thursday 10:15–11:45 Location: TC 006

MM 45.1 Thu 10:15 TC 006

Thermal Conductivities at High Temperatures from First Principles — •Christian Carbogno[1], Rampi Ramprasad[2], and Matthias Scheffler[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4–6, 14195 Berlin, Germany — [2]Chemical, Materials & Biomolecular Engineering, University of Connecticut, Storrs, USA

In spite of significant research efforts, a first principles determination of the thermal conductivity at *high temperatures* has remained elusive. Under such conditions, techniques that rely on the harmonic approximation become questionable, while non-equilibrium MD methods require huge temperature gradients that lead to undesired non-linear effects. The Green-Kubo method, which does not suffer from these shortcomings, involves the assessment of the thermal conductivity from the auto-correlation of the heat flux via equilibrium MD. This method has hitherto been disregarded in first-principles simulations since the computation of the heat flux requires the energy contributions from the individual atoms, a quantity that is not directly accessible in first-principles schemes. We show that the Green-Kubo approach can be reformulated in terms of the *energy density* [1], which is directly accessible in density functional theory calculations. This approach leads to a unique definition of the heat flux that does not rely on any partitioning scheme for the total energy. To demonstrate the capabilities of this technique, we investigate the thermal conductivity of ZrO_2, a material that is widely used in industrial high-temperature applications.

[1] N. Chetty and R. M. Martin, *Phys. Rev. B* **45**, 6074 (1992).

MM 45.2 Thu 10:30 TC 006

Thermal conductivity of minerals at the Earth's core-mantle boundary from equilibrium molecular dynamics — •Volker Haigis[1], Mathieu Salanne[2], and Sandro Jahn[1] — [1]GFZ German Research Centre for Geosciences, Telegrafenberg, 14473 Potsdam, Germany — [2]UPMC Université Paris 06 and CNRS, UMR 7195, PECSA, 75005 Paris, France

The thermal conductivity at the Earth's core-mantle boundary is an important geophysical parameter which governs the heat flux across the boundary and thus influences the dynamics of both core and mantle. However, conductivities at the relevant temperatures and pressures cannot be measured with present-day experimental techniques. Hence a computational approach is desirable. We report thermal conductivities of fcc MgO and $MgSiO_3$ in the perovskite and the post-perovskite structure at conditions representative of the Earth's lowermost mantle, obtained from equilibrium molcular dynamics. Using an advanced ionic interaction potential [1], the full conductivity tensor was calculated by means of the Green-Kubo method, and the conductivity of $MgSiO_3$post-perovskite was found to be significantly anisotropic. Assuming an iron-free mantle composition with $x_{MgSiO_3} = 0.66$ (in the post-perovskite structure) and $x_{MgO} = 0.34$, we predict the average thermal conductivity at the core-mantle boundary to be (24.3 ± 1.8) W/(mK). Based on experiments [2], we expect that a realistic amount of iron impurities reduces the conductivity to (12 ± 1) W/(mK).

[1] S. Jahn, P. A. Madden, Phys. Earth Planet. Int. **162**, 129 (2007)
[2] G. M. Manthilake *et al.*, PNAS **108**, 17901 (2011)

MM 45.3 Thu 10:45 TC 006

Thermal Conductivity in Ge-based Clathrate Systems — •Daniel Schopf and Hans-Rainer Trebin — Institut für Theoretische und Angewandte Physik, Universität Stuttgart

Intermetallic clathrates are currently actively investigated due to their special thermoelectric properties. They are composed of periodically arranged cages, formed by host atoms, which enclose single guest atoms. The low thermal conductivity of these structures has been attributed to the scattering of the phonons on the local vibration modes ("rattling") of these guest atoms.

For computational studies of dynamic properties long simulation times and large samples are required. This makes first principle calculations of these structures, even with high performance computers, very unfeasible. Classical molecular dynamics, however, can meet the requirements. The potentials needed for these MD simulations can be obtained from ab-initio calculations with the force-matching method. It uses large numbers of reference data to fit an effective potential that reproduces the forces, energies and stresses of the ab-initio calculation.

To model the strongly directional atomic interactions in clathrates, angular dependent potentials are required. An analytic potential will be presented. With this potential and the Green-Kubo method, the thermal transport in different Ge-based clathrate systems has been investigated.

MM 45.4 Thu 11:00 TC 006

Ab initio characterization of thermoelectrical materials — •Matthieu Verstraete — Nanomat unit, Physics Department University of Liege, Belgium

Energy production, storage, and efficiency have become pressing issues in society, science, and materials in particular over the past few decades. Thermoelectrical materials are a classical technology which may come into its own thanks to advanced nanostructuring and controlled alloying. Understanding the microscopic mechanisms which can improve on efficiencies of bulk materials is crucial. We will present the ab initio DFT characterization of thermoelectrics with very different mechanisms, behaviors, and relevant temperature ranges, in particular Mg2Si and FeSb2. Specific unsolved issues in the theoretical treatment of thermoelectrics will be raised and discussed.

MM 45.5 Thu 11:15 TC 006

Transport properties of nanoconfined water — •Akinlolu Akande[1], Ivan Rungger[1,2], Clotilde Cucinotta[1], Konstantinos Gkionis[2], Udo Schwingenschlögl[2], and Stefano Sanvito[1] — [1]School of Physics and CRANN, TCD Dublin, IRELAND — [2]KAUST, Saudi Arabia

Understanding the electronic transport properties of nanoconfined systems under wetting conditions is essential for many applications, ranging from molecular nano-junctions to nano-electronics. Here we focus on the transport properties of water confined between a gold STM tip and a gold surface. We perform calculations for the molecular dynamics of the water and quantum transport simulations for a randomly selected set of time-snapshots. These are at the level of the non-equilibrium Green function method, as implemented in the SMEAGOL code. We calculate the dependence of the conductance in liquid water on the separation between the confining surfaces, and find good agreement with experimental findings.

Within the electron tunnelling regime we study the dependence of transport properties of the nanoconfined system on liquid water, also comparing it with the transport in vacuum. The dependence is explained further in terms of the complex band structure of water, which determines - together with the interface structure - the decay of the conductance with thickness. This study allows us to unravel the relationship between conductance and the microscopic structure of nanoconfined water and explores the possibility to detect the phase of a water sample by current measurements.

MM 45.6 Thu 11:30 TC 006

Quantum transport simulations in metallic carbon nanotubes with metal contacts — •Andreas Zienert[1], Jörg Schuster[2], and Thomas Gessner[1,2] — [1]Center for Microtechnologies, Chemnitz University of Technology, 09126 Chemnitz, Germany — [2]Fraunhofer Research Institution for Electronic Nano Systems, 09126 Chemnitz, Germany

Carbon nanotubes (CNTs) are one dimensional conductors with promising applications in future microelectronic devices and sensors. Due to their high thermal and mechanical stability and their ability to carry high current densities, they are discussed as novel interconnect materials, partially replacing copper metallization in future microelectronic devices. However, the performance will strongly depend on the quality of contacts between CNTs and the environment [1].

The geometric structure of CNT-metal contacts is modeled using density functional theory. A simple approach for contact geometry optimization will be presented. Non-equilibrium Green's function methods in combination with DFT are used to study electronic transport through metal–CNT–metal devices. Results for different metals will be interpreted in terms of electronic properties of the bulk materials and the contact area.

[1] Zienert et al. Phys. Stat. Sol. B 247, 3002 (2010)