Transport Properties of Thermoelectric Materials from First Principles

G. Samsonidze,[1] B. Kozinsky,[1] C.-H. Park,[1] J. Garg,[2] D. Volja,[2]
N. Bonini,[3] D. Wee,[4] M. Fornari,[5] and N. Marzari[6]

[1] *Research and Technology Center North America, Robert Bosch LLC, Cambridge, MA 02142, USA.*
[2] *Massachusetts Institute of Technology, Cambridge, MA 02139, USA.*
[3] *King's College, London, UK.*
[4] *Ewha Womans University, Seoul, South Korea.*
[5] *Central Michigan University, Mount Pleasant, MI 48859, USA.*
[6] *Ecole Polytechnique Federale de Lausanne, Lausanne, Switzerland.*

Thermoelectric effect offers an effective solution for waste heat recovery. Optimization of thermoelectric materials reqiures description of the electrical and thermal transport properties from first principles. Thermal conductivity is governed by the phonon lifetime and it is important to determine the influence of microscopic effects due to alloying, nanostructuring, and grain boundaries on the macroscopic transport coefficients of the system. We show how density-functional theory, with the help of the Boltzmann transport formalism and quasi-harmonic analysis, can deliver predictive accuracy in calculating thermal conductivity of single-crystal, alloyed and nanostructured materials. Our method combines first-principles determination of harmonic force constants with the use of model potentials for calculating higher order anharmonic response terms. The latter gives the main contribution to phonon scattering at high temperatures. We validate our scheme by the comparison of the heat conductivity in SiGe alloys with available experimental data. Analysis of the grain size effects is carried out by introducing boundary scattering terms and using Matthiessen rule. We apply this methodology to analyze grain size effects in binary skutterudites $CoSb_3$. We also calculate the electrical conductivity and Seebeck coefficient of $CoSb_3$ using a constant electron lifetime approximation. These transport coefficients are sensitive to the electronic band structure, and we stress the importance of using the GW quasiparticle energies as opposed to the DFT Kohn-Sham eigenvalues.

Contact: Georgy.Samsonidze@us.bosch.com