HL 49: Energy materials: Thermoelectrics

Time: Tuesday 14:00–16:00 Location: POT 051

HL 49.1 Tue 14:00 POT 051

Silicene: A Promising Thermoelectric Material — •Xiaoliang Zhang[1] and Ming Hu[1,2] — [1]Institute of Mineral Engineering, Division of Materials Science and Engineering, Faculty of Georesources and Materials Engineering, RWTH Aachen University, 52064 Aachen, Germany — [2]Aachen Institute for Advanced Study in Computational Engineering Science (AICES), RWTH Aachen University, 52062 Aachen, Germany

Thermoelectric materials can directly convert waste heat to electricity for improving the efficiency of energy utilization. Silicene, the silicon-based counterpart of graphene, has shown its great potential for thermoelectric applications. First of all, from electronic structure point of view ab initio calculations suggest that silicene is equivalent to graphene, i.e., the electrical conductivity of silicene is as high as that of graphene. Second, in contrast to graphene, silicene has a buckled atomic structure, leading to non-zero energy gap and enhanced Seebeck coefficient. Third, by conducting molecular dynamics simulations, we confirm that the thermal conductivity of silicene is extremely low, suggesting silicene as a potential candidate of high efficiency thermoelectric materials. Moreover, our molecular dynamics simulation results suggest that the thermal conductivity of silicene can be further reduced by hydrogenation and oxidation implying enhanced thermoelectric performance. Compared with pristine silicene, the hydrogenated and oxidized silicene structures are more stable and thus can be used for energy conversion at high temperatures.

HL 49.2 Tue 14:15 POT 051

Resonant tunneling energy harvesters — Andrew N. Jordan[1], Björn Sothmann[2], •Rafael Sánchez[3], and Markus Büttiker[2] — [1]Department of Physics and Astronomy, University of Rochester, Rochester, U. S. A — [2]Département de Physique Théorique, Université de Genève, Genève, Switzerland — [3]Instituto de Ciencia de Materiales de Madrid, CSIC, 28049, Madrid, Spain

An important task in condensed matter physics is to find new ways to harvest ambient thermal energy, particularly at the smallest length scales where electronics operate. To achieve this goal, there is on one hand the miniaturizing of electrical devices, and on the other, the maximization of either efficiency or power the devices produce. We present the theory of nano heat engines able to efficiently convert heat into electrical power. We propose a resonant tunneling quantum dot engine that can be operated either in the Carnot efficient mode, or maximal power mode. The ability to scale the power by putting many such engines in a "Swiss cheese sandwich" geometry gives a paradigmatic system for harvesting thermal energy at the nanoscale[1].

Alternative configurations based on resonant tunneling through quantum wells provide a comparable thermoelectric performance with the advantage of being easier to construct[2].

[1] A. N. Jordan, B. Sothmann, R. Sánchez, M. Büttiker, Phys. Rev. B 87 (2013) 075312.

[2] B. Sothmann, R. Sánchez, A. N. Jordan, M. Büttiker, New J. Phys. 15 (2013) 095021.

HL 49.3 Tue 14:30 POT 051

Hybrid Si/AlO$_x$ thin films of the electron crystal-phonon glass type — •Markus Trutschel[1,2], Jens Glenneberg[2], Stefan Ebbinghaus[2], Peter Werner[1], and Hartmut S. Leipner[2] — [1]Max-Planck-Institut für Mikrostrukturphysik, Halle (Saale) — [2]Martin-Luther-Universität, Halle (Saale)

Heat in modern integrated circuits is produced concentrated in small areas. Active cooling may enhance the performance of such devices. Materials for thermoelectric cooling require a high electrical conductivity and Seebeck coefficient together with a low thermal conductivity. Thus, suitable materials are those of the electron crystal-phonon glass type. Composites may provide nontoxic, cheap and available materials with a good thermoelectric performance to be integrated in silicon technology.

We studied structural, electrical and thermoelectric properties of silicon particles formed in an aluminum oxide matrix. The samples were synthesized by means of a physicochemical process. Thin films of aluminum were deposited on silicon oxide layers followed by a thermal annealing step in argon atmosphere. The silicon oxide was reduced to silicon during the heat treatment, whereas the aluminum was oxidized. For annealing temperatures between 540°C and 600°C, the size, shape and distribution of the silicon particles formed was found to be interesting for technical applications. We present the structural characterization of hybrid Si/AlO$_x$ films along with the results of the electrical and thermoelectric properties.

HL 49.4 Tue 14:45 POT 051

Phononic thermal transport in nanostructured silicon membranes — •Sanghamitra Neogi, Luiz F. C. Pereira, and Davide Donadio — Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany

The interest in energy harvesting using thermoelectric devices and thermal management in nanostructures has heightened the necessity of understanding the phononic thermal transport in nanoscale materials. Nanostructuring can result in the reduction of the thermal conductivity of crystalline semiconductors while preserving their electronic properties, leading to an improved TE efficiency in these systems [1].

We use harmonic lattice dynamics and classical molecular dynamics to investigate the nature of phononic thermal transport in ultra-thin silicon membranes, with thicknesses up to 20 nm. We show that dimensionality reduction has a significant effect on the phonons of the membranes and leads to a 4-fold reduction in the thermal conductivity of the membranes. Nanostructuring Si membranes by means of pattern formation and surface oxidation, in combination with dimensionality reduction, results in a 25-fold reduction in the thermal conductivity of the membranes with respect to the bulk, implying a 25-fold enhancement of the thermoelectric figure of merit at room temperature. Such figures make nanostructured silicon membranes viable materials for thermoelectric units.

[1] M. S. Dresselhaus et al, Adv. Mater., 22, 3970 (2010).

Acknowledgment: This project is funded by the program FP7-ENERGY-2012-1-2STAGE under contract number 309150.

HL 49.5 Tue 15:00 POT 051

Thermoelectric transverse voltage by Joule heating — a Boltzmann equation approach beyond quasi-equilibrium — •Stephan Rojek, Alfred Hucht, and Jürgen König — Theoretische Physik, Universität Duisburg-Essen and CENIDE, 47048 Duisburg, Germany

We consider structures within a two-dimensional electron gas where a thermoelectric transverse voltage is measured perpendicular to an input current. The latter gives rise to Joule heating. An external potential induced by surface gates breaks the transverse symmetry. To realize tunable rectification devices [1] or to get insight into thermoelectric transport properties of the electron gas [2] are two of various applications. Heat diffusion and finite size effects lead to a non-trivial temperature profile, which have to be taken into account if the device's dimensions are of the order of the energy diffusion length.

We employ a Boltzmann equation approach beyond the quasi-equilibrium description with an effective chemical potential and temperature. An expansion in moments of the distribution function allows for a systematic calculation of all non-equilibrium contributions.

[1] A. Ganczarczyk, S. Rojek, A. Quindeau, M. Geller, A. Hucht, C. Notthoff, J. König, A. Lorke, D. Reuter, and A. D. Wieck, Phys. Rev. B **86**, 085309 (2012).

[2] W. E. Chickering, J. P. Eisenstein, and J. L. Reno, Phys. Rev. Lett. **103**, 046807 (2009).

HL 49.6 Tue 15:15 POT 051

Thermoelectric Coefficients from First Principles — •Karsten Rasim, Christian Carbogno, and Matthias Scheffler — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

In spite of significant efforts, an accurate assessment of the electronic transport coefficients, i.e., the electrical conductivity σ, the Seebeck coefficient S and the electronic heat conductivity κ_{el}, still constitutes an open challenge for first-principles theory. Even the most involved approaches [1] treat the nuclear dynamics in the harmonic approximation and thus become inaccurate at elevated temperatures. To overcome this limitation, we use the *Kubo-Greenwood formalism* [2] to assess σ, S, and κ_{el} from the thermodynamic equilibrium fluctuations of the electronic structure and of its dipole transition matrix elements.

Anharmonic effects in the nuclear motion are fully incorporated by the means of *ab initio* molecular dynamics. We discuss the details of our implementation and validate our approach by investigating various direct and indirect band gap semiconductors. Furthermore, we compute the temperature dependent electronic transport coefficients of borcarbides, a material class regarded as promising for thermoelectric applications. We demonstrate that in this case anharmonic effects are crucial to achieve a correct description of the transport coefficients.
[1] P. Boulet and M. J. Verstraete, Comp. Mat.Sci. **50**, 3 (2001)
[2] B. Holst, M. French, and R. Redmer, Phys. Rev. B **83**, 235120 (2011).

HL 49.7 Tue 15:30 POT 051

First-principles Study of Vacancies in Thermoelectric Clathrates — •Amrita Bhattacharya, Christian Carbogno, and Matthias Scheffler — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin

For the development of improved thermoelectrics, one promising material class may be clathrates, i.e., semiconducting host lattices that can encapsulate guest atoms. Even in the simplest clathrates, such as Si_{46} and Ge_{46}, the introduction of guests can result in surprising electronic and structural changes: In the case of Si, the addition of K results in a K_8Si_{46} clathrate that exhibits metallic behavior due to the electrons donated by the guests [1]. Conversely, Ge-based clathrates remain semiconducting, since two spontaneously generated vacancies balance the donated electrons in K_8Ge_{44}. But even more intriguingly, filling the Ge-clathrate with Ba does not generate *four*, but *three* vacancies, whereby the resulting Ba_8Ge_{43} exhibits a metal-semiconductor transition at $\sim$ 280 K [2]. In this work, we use density-functional theory to investigate these effects and to unravel their puzzling mechanism. We compute the formation energies for vacancies and vacancy complexes in Si- and Ge-hosts as function of the filling with K and Ba, whereby we take into account structural disorder as well as geometric and lattice relaxations. Furthermore, we study the dynamics in these compounds (also analyzing *non-harmonic effects*) to clarify the role the vacancies play for the thermodynamic properties.
[1] G. K. Ramachandran, *et al.*, *J. Sol. St. Chem.* **154**, 626 (2000)
[2] U. Aydemir *et al.*, *Dalton Trans.* **39**, 1078 (2010).

HL 49.8 Tue 15:45 POT 051

Thermoelectric power factor of ternary single-crystalline Sb_2Te_3- and Bi_2Te_3-based nanowires — •Svenja Bässler[1], Tim Böhnert[1], Johannes Gooth[1], Christian Schumacher[1], Eckhard Pippel[2], and Kornelius Nielsch[1] — [1]Institute of Applied Physics, Universität Hamburg, Jungiusstrasse 11, 20355 Hamburg, Germany — [2]Max Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle, Germany

Nanowires of bismuth antimony telluride and bismuth telluride selenide ($Bi_{15}Sb_{29}Te_{56}$ and $Bi_{38}Te_{55}Se_7$) are grown by template-based pulsed electrodeposition. The composition and the crystallinity of the nanowires are determined by high resolution transmission electron microscopy. The thermoelectric properties (Seebeck coefficient and electrical conductivity) of single p- and n-type nanowires with a diameter of 80 nm and 200 nm, respectively, are determined as a function of temperature before and during heating in helium atmosphere up to 300 K along the growth direction of the nanowires. After additional annealing in Te atmosphere at 525 K, significantly enhanced transport properties are observed. Bulk-like power-factors are achieved. In $Bi_{38}Te_{55}Se_7$ nanowires the Seebeck coefficients increase up to -115 μV K^{-1} and the thermoelectric power factors up to 2820 μW K^{-2} m^{-1} at room temperature. In $Bi_{15}Sb_{29}Te_{56}$ nanowires Seebeck coefficients of up to +156 μV K^{-1} and power factors of up to 1750 μW K^{-2} m^{-1} are obtained at room temperature.

Reference:
S. Bäßler *et al.* Nanotechnology 24 (2013) 495402