Transport properties of thermoelectric materials

The effect of disorder on transport properties of alloys

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In Short

- Complex clathrate compounds are promising candidates for thermoelectric applications
- Understanding of how their ordering phenomena affect the electronic transport coefficients
- Study of finite-temperature transport coefficients
- Developing and applying new concepts and methodology for reliable *ab-initio* calculations of transport coefficients

Thermoelectric materials allow for transforming waste heat into electricity. The search for materials with large thermoelectric efficiency is a prerequisite for profitable applications and concern a hot topic in materials research. The efficiency of a material depends on the electrical conductivity, the Seebeck coefficient, and the thermal conductivity, which determine the figure of merit zT (a large zT implies a large efficiency). In order to achieve reliable calculations of these transport coefficients, improved theoretical methods are needed.

We tackle this topic using different theoretical approaches, beginning with two well established methods - the linearized Boltzmann equation [1] in the relaxation-time approximation and Kubo's linear response theory [2]. These methods will be implemented in `exciting` [3] and applied to clathrate compounds, which are promising candidates for thermoelectricity [4].

The clathrate compounds have a complex unit cell. An example for $Ba_8Al_xSi_{46-x}$ is shown in Fig. 1(left)). At a single composition x, a combinatorially exploding number of configurations results from different arrangements of the Al atoms in the cage structure. These configurations can differ considerably in their structural, as well as in their electronic properties. To study structural-dependent properties of the clathrates, we have employed an iterative cluster-expansion approach [5]. With this procedure, we found the ground-state configuration for $Ba_8Al_xSi_{46-x}$ and $Sr_8Al_xSi_{46-x}$ $(6 \leq x \leq 16)$ [6]. For the charge-compensated composition $(x = 16)$, the density of states for different configurations is shown in Fig. 1(right). The ground-state configuration (black line) is found to be semiconducting, while

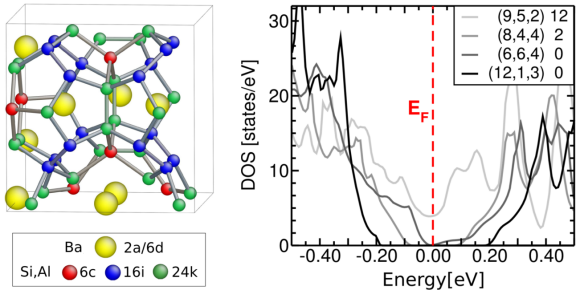

Figure 1: *The clathrate compound $Ba_8Al_xSi_{46-x}$. Left: Unit cell with the cage structure consisting of 46 atoms (Al or Si atoms) sitting at the Wyckoff sites $24k$ (green), $16i$ (blue) and $6c$ (red) and the guest atoms (Ba atoms) sitting at the Wyckoff sites $2a$ and $6d$. Right: Density of states (DOS) of four configurations of the charged compensated composition $x = 16$.*

configurations high in energy are metallic (*e.g.* the density of states of the configuration shown with the light grey line). Further investigations confirmed that this semiconductor-to-metal transition is driven by a structural order-disorder transition upon temperature. The closing of the band gap clearly affects the Seebeck coefficient. This emphasizes the need to take substitutional ordering into account for the reliable calculation of the transport properties.

WWW

http://sol.physik.hu-berlin.de

More Information

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Project Partners

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