

Materials Science and Technology Division
"Model calculations of thermal
conductivity of skutterudites"

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Abstract:

The skutterudites are crystals with the CoAs_3 crystallographic space group and they have promising thermoelectric properties. They have cage-like structures and, at least in the cases studied to our knowledge, their thermal conductivity depends sensitively on whether or not the cages are filled: Generally filled skutterudites have a substantially lower lattice thermal conductivity than unfilled ones.

While the lattice thermal conductivity of the skutterudites has been of great interest it had not been calculated before within a microscopic theory. Here a central force, Guggenheim-McGlashen, model with parameters largely extracted from first principles calculations and spectroscopic data specific to $\text{LaFe}_4\text{Sb}_{12}$ or CoSb_3 is employed in a Green-Kubo/molecular dynamics calculation of thermal conductivity as a function of temperature. We find that the thermal conductivity of a filled solid is more than a factor of two lower than that of an unfilled solid (assuming the "framework" interatomic force parameters are the same between filled and unfilled solids) and that this decrease is almost entirely due to the cubic anharmonic interaction between filling and framework atoms.

In addition, partially as a test of our models, we calculate thermal expansivity and isotropic atomic mean square displacements using both molecular dynamics and lattice dynamics methods. These quantities are in reasonable agreement with experiment, increasing our confidence in the anharmonic parameters of our models. We also find an anomalously large filling-atom mode Gruneisen parameter that is apparently observed for a filled skutterudite and is observed in a clathrate.

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