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Title: Thermal transport from first-principles

Abstract: Unlike electronic properties, the calculation of the lattice thermal conductivity from first-principles is not that straightforward. We have developed a methodology to perform these calculations [1,2], and have applied it to several materials including Si, GaAs, PbTe, PbSe, and ZrCoSb. We intend to pursue this path and also calculate the thermal transport properties of new thermoelectric materials such as GeSe.

Research on thermoelectric materials was revived in the 90's [3] after realizing that in low-dimensional systems, the sharp variations in the density of states can lead to a large Seebeck coefficient. For energy conversion using thermoelectric materials, or in thermal barrier coatings used in aerospace industry, one needs to have a low thermal conductivity material. For optoelectronics applications, on the other hand, high thermal conductivity materials are required so that the generated heat from currents can be quickly evacuated. Management of heat is therefore of paramount importance for technological applications. In semiconductors used in thermoelectric devices, heat is mostly carried by phonons. One can interpret the standard Boltzmann formula within the relaxation time approximation, as the thermal conductivity coming from different phonon modes, each carrying an amount of heat proportional to the mean free path (MFP), group velocity and heat capacity of that mode. Accordingly, acoustic modes carry more heat than optical one because of their larger group velocity and larger MFP. The latter is inversely proportional to the phonon scattering rate, itself determined from the Fermi's golden rule (FGR). At high temperatures, phonons scattering is dominated by 3-phonon processes. The probability of such processes depend on the strength of the phonon-phonon coupling dictated by the cubic force constants. We have used first-principles density functional theory (FP-DFT) to compute accurately the harmonic and cubic force constants (FCs). The former is used to obtain the phonon frequencies and group velocities, and the latter provides the phonon lifetimes through FGR. The methodology to extract the FCs from FP-DFT calculations in a supercell is detailed in [1] while the calculation of the phonon lifetimes and thermal conductivity is described in [2]. We applied this methodology to Si [2], PbTe, GaAs and half-heusler compounds such as ZrCoSb. The results were discussed in [4]. In figure 1 we show the calculated thermal conductivities of these materials as compared with experiments. We can note that the agreement is within 10% or less in the studied cases. Figure 2 shows the contribution of the phonons of different MFP to the total thermal conductivity.

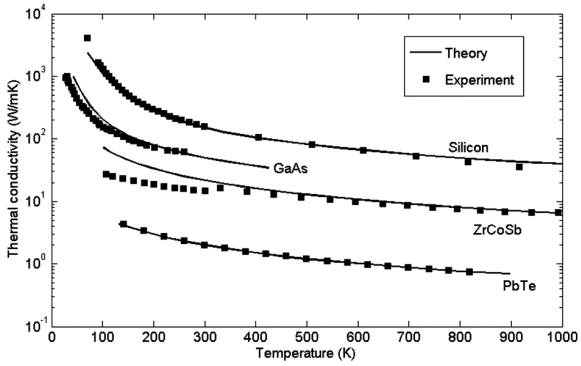

Fig. 1 Thermal conductivities of several materials versus T.

It can be seen that in Si for example, phonons of MFP more than 1000 nm contribute almost to half of the thermal conductivity at room temperature. This information is very useful in designing nanostructured materials of desired thermal properties. In Si, if the nanostructure size is100 nm, the thermal conductivity will almost be reduced by a factor of 4, while this size does not affect much thermal transport in PbTe or ZrCoSb.

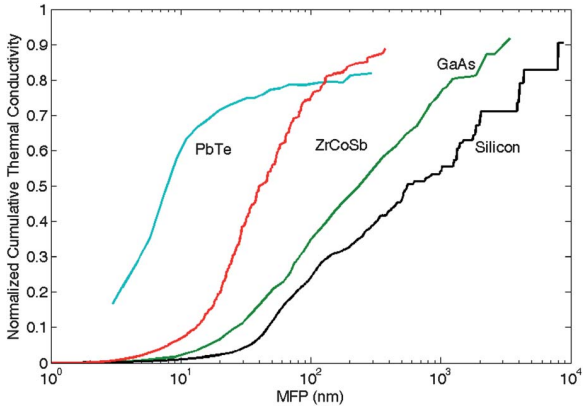

Fig 2. Accumulated thermal conductivity versus the mean free path for the same materials as in Fig 1. at T=300K

References

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Key Words

Thermal transport, thermoelectricity, phonon mean free paths

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