Four-phonon scattering significantly reduces thermal conductivity of boron arsenide, silicon and diamond

T.L. Feng[1], L. Lindsay[2], and X.L. Ruan[1]

[1]School of Mechanical Engineering and the Birck Nanotechnology Center, Purdue University, West Lafayette, Indiana 47907-2088, USA
[2]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

Recently, first principles-based prediction of lattice thermal conductivity κ from perturbation theory has achieved significant success. However, it has only included three-phonon scattering due to the assumption that four-phonon and higher-order processes are generally unimportant. Also, directly evaluating the scattering rates of four-phonon and higher-order processes has been a long-standing challenge. We have developed a formalism to explicitly determine quantum mechanical scattering probability matrices for four-phonon scattering in the full Brillouin zone [1]. By mitigating the computational challenge we can rigorously calculate four-phonon scattering rates and the resulted intrinsic phonon thermal resistance from four-phonon scattering processes using first principles Boltzmann transport methods. Fundamental questions concerning the role of higher order scattering at high temperature and in systems with otherwise weak intrinsic scattering are answered. Using diamond and silicon as benchmark materials, the predicted thermal conductivity including intrinsic four-phonon resistance gives significantly better agreement with measurements at high temperatures than previous first principles calculations. In the predicted ultrahigh thermal conductivity material, zinc blende BAs, four-phonon scattering is strikingly strong when compared to three-phonon processes, even at room temperature, as the latter have an extremely limited phase space for scattering. Including four-phonon thermal resistance reduces the predicted thermal conductivity of BAs from ~2200 W/m-K to ~1400 W/m-K.

[1] T.L. Feng and X.L. Ruan, *Phys. Rev. B.* **93**, 045202 (2016).