Predicting Vibrational Mean Free Paths in Disordered Systems

Understanding thermal transport in crystalline systems requires detailed knowledge of phonons, which are the quanta of energy associated with atomic vibrations. By definition, phonons are non-localized vibrations that transport energy over distances much larger than the atomic spacing. For disordered materials (e.g., high concentration alloys, amorphous phases), with the exception of long wavelength modes, the vibrational modes are localized and do not propagate like phonons. The Einstein model assumes that the mean free path of these localized vibrations is the average interatomic distance and that their group velocity is equal to the speed of sound. The Cahill-Pohl model assumes that the mean free path of the localized modes is equal to half of their wavelength. While these approach can be used to estimate the thermal conductivity of disordered systems, they only provide a qualitative description of the vibrations that contribute to the lattice thermal conductivity. Using lattice dynamics calculations and molecular dynamics simulations on Lennard-Jones argon and Stillinger-Weber silicon crystalline, alloy, and amorphous systems, we predict and characterize the contributions from phonons and localized vibrations to lattice thermal conductivity. Of particular importance is predicting a representative group velocity for vibrational modes in disordered systems, which allows the prediction of thermal conductivity mean free path spectra.