**Evaluation of the Virtual Crystal Approximation for Predicting Thermal Conductivity**

Accurately predicting the thermal conductivity of a dielectric or semiconducting material requires the properties of phonons from the entire Brillouin zone. Accurate predictions of phonon properties for bulk systems can be made with anharmonic lattice dynamics theory using *ab initio* calculations. However, computational costs limit the size of unit cells in *ab initio* calculations to be less than 100 atoms, making it difficult to directly incorporate the effects of disorder. Alternatively, theory that treats disorder as a harmonic perturbation can be used to estimate the reduction in phonon lifetimes due to disorder scattering without the use of a large unit cell. Under this approximation, the disordered crystal is replaced with a perfect “virtual crystal” with properties equivalent to an averaging over the disorder (e.g. mass or bond strength).

In this work, the virtual crystal approximation for mass disorder is evaluated by examining two model alloy systems: Lennard-Jones argon and Stillinger-Weber silicon. In both cases the perfect crystal is alloyed with a heavier mass species up to equal concentration . These two alloyed systems have different ranges of phonon frequencies, lifetimes, and mean free paths. For Stillinger-Weber silicon, the virtual crystal approximation predicts phonon properties and thermal conductivity in good agreement with molecular dynamics-based methods. For Lennard-Jones argon, the virtual crystal approximation underpredicts the high frequency phonon lifetimes, leading to an underpredicting of its thermal conductivity.