**Evaluation of the Virtual Crystal Approximation**

Accurately predicting the thermal conductivity of a dielectric or semiconducting material requires the properties of phonons from the entire Brillouon zone. Accurate predictions of phonon properties for bulk systems can be made with anharmonic lattice dynamics 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 which 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, bond strength, etc.). The “virtual crystal” approximation is evaluated and examined using two model alloyed 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 (non-perturbative). These two alloyed systems have different thermal conductivity spectra (with respect to 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. In contrast to Stillinger-Weber silicon, high frequency modes are dominant in Lennard-Jones argon's thermal conductivity spectrum. For Lennard-Jones argon, it is shown that the virtual crystal approximation using anharmonic lattice dynamics underpredicts the high frequency phonon lifetimes, which is responsible for underpredicting the thermal conductivity. Caution must be used when applying the virtual crystal approximation, especially when using with *ab initio* calculations to compare with experiment.