Predicting Vibrational Mean Free Paths in Disordered Systems

Understanding thermal transport in crystalline systems requires detailed knowledge of phonons, quanta of energy associated with atomic vibrations. By definition, phonons are propagating vibrations that transport energy over distances much larger than the atomic spacing. With the exception of long wavelength modes, the vibrational modes of strongly disordered materials (e.g., high concentration alloys, amorphous phases) do not propagate. The Einstein model assigns such modes a mean free path equal to the interatomic distance and takes their group velocity to be the speed of sound. The Cahill-Pohl model assumes that the mean free path of these modes is equal to half their wavelength and also uses the speed of sound. While these approaches can be used to estimate the thermal conductivity of disordered systems, they only provide a qualitative description of the properties of the vibrational spectrum.

In this work, we use lattice dynamics calculations and molecular dynamics simulations to predict and characterize the contributions of propagating and non-propagating vibrational modes to the lattice thermal conductivity of Lennard-Jones argon and Stillinger-Weber silicon crystalline, alloy, and amorphous systems. Of particular importance is predicting a representative group velocity for propagating vibrational modes in the disordered systems in order to allow for the generation of thermal conductivity mean free path spectra and accumulation functions.

Jason: my thinking is as follows. First, separate propagons and diffusions. Second: for propagons, challenge is specifying the velocity in order to get a mfp; for diffusions, use Allen Feldman. Diffusons do not have a mfp and make the thermal conductivity accumulation start at a finite value, as opposed to zero for a perfect crystal.

I agree.

For diffusons, one can still assign a mfp by the following:

D\_AF = computed by AF theory

sqrt( D\_AF / \tau\_{NMD/ALD} ) = \Lamda\_{AF}

typical calculations demonstrate that the lower limit to D\_AF should be:

D\_AF = 1/3 v\_s a

where v\_s = sound speed and a = interatomic spacings. So, \Lambda\_{AF} >= a, or something like that.