First-principles simulation of phononic thermal transport in strongly anharmonic solids

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Lattice thermal conductivity (LTC) has been the subject of intense theoretical/computational studies in recent years because of its importance for optimizing the thermoelectric figure-of-merit ZT. Owing to the recent development of the first-principles methods to compute third-order interatomic force constants, it is now rather straightforward to predict LTC of bulk materials based on the Boltzmann transport theory (BTE). However, the conventional BTE method often fails to predict LTC of emergent thermoelectric materials with ultralow LTC because it employs the harmonic lattice dynamics wave-function as the ground state and neglects the temperature dependence of phonons associated with large atomic displacements.

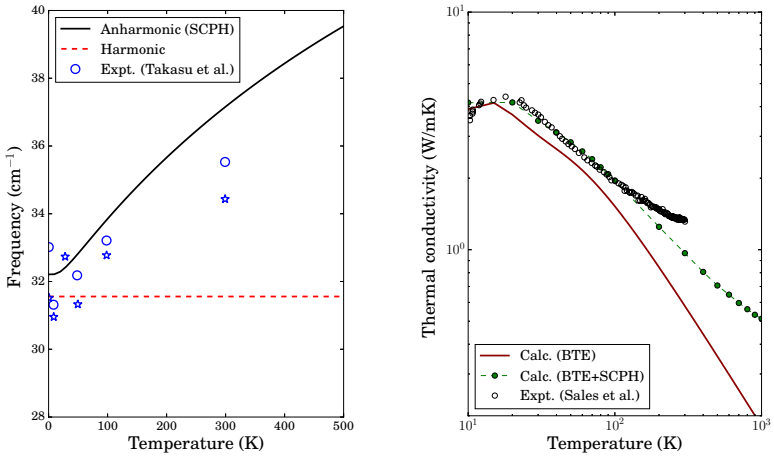

Fig. 1. Temperature dependence of the Raman shift (left) and the lattice thermal conductivity (right) in type-I clathrate $Ba_8Ga_{16}Ge_{30}$

To overcome the limitation of the BTE-based method for severely anharmonic materials, we have developed an efficient first-principles method to compute temperature-dependent phonon properties [1]. The method is based on the self-consistent phonon (SCP) theory, and the frequency renormalization due to the quartic anharmonicity is calculated at the mean-field level. By using the present method, we can predict the temperature dependence of the Raman shift with good agreements with experimental data (**Fig.1** left). In addition, the accuracy of LTC prediction can be improved by solving the BTE on top of the SCP equation (**Fig.1** right). We will demonstrate the validity of our BTE+SCP approach for type-I clathrate $Ba_8Ga_{16}Ge_{30}$ and other severely anharmonic materials.

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