Lattice Thermal Conductivities of Group-IV, V, VI based Semiconductors

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Since the lattice thermal conductivity is a key metric for the materials' applications into thermoelectrics and power electronics, the last decade has witnessed the rapid increase of demand in accelerating the theoretical prediction of lattice thermal conductivities of materials from first-principles calculations and Boltzmann transport equation. Figure 1(a) presents the comparison between our calculated lattice thermal conductivities and the experimental values for 15 compounds. We can find a good agreement is reached in our calculations with respect to the experiments. However, the high computational cost of obtaining cubic and higher-order anharmonic force constants has affected the high throughput evaluation rate of lattice thermal conductivity. Since we often expect the interactions of atoms are weakened rapidly with increasing the distances among atoms at higher order force constants, it is a usual idea to introduce a cutoff distance to omit the computation of force constants of the atoms whose mutual distances are larger than the cutoff distance. However, it has not been carefully investigated how much this approach is reliable for a variety of compounds. In this study, we performed systematic calculations of lattice thermal conductivities of 91 compounds with respect to cutoff distances and show the convergences of the lattice thermal conductivities in comparison to those without using the cutoff distances.

By screening the binary and ternary non-metallic compounds from Phonon database at Kyoto university [1] we have studied the lattice thermal conductivities of 91 dynamically stable compounds with cubic or tetragonal lattice systems. The quadratic and cubic force constants were obtained by the supercell approach with finite atomic displacements of 0.03 Å. The density functional theory (DFT) calculations were performed by using the VASP code [2, 3]. The exchange-correlation functional, cutoff energy, *k*-mesh, and the supercell size are in line with those in Phonon database [1]. The calculations of lattice thermal conductivities were carried out by Phono3py code [4, 5] within the framework of relaxation-time approximation. The elements of third-order force constants were set to zero if all pairs of atoms among three atoms have larger distance than the cutoff distance. Figure 1(b) shows the calculated lattice thermal conductivities at different cutoff distances for the 91 materials. The multiple cutoff distances were collected from pair distances of atoms in each compound, by which each convergence was examined. The values of lattice thermal conductivities, as shown by the black squares in panel (b), are all normalized with respect to the rightmost values (red triangles) that are calculated by using the full elements of third-order force constants in the corresponding supercells. Thus, the values of all rightmost points are 1. The relative errors of normalized lattice thermal conductivities for all compounds are found smaller than 25%, in particular, 88 compounds show relative error smaller than 10%, indicating effectiveness of the use of cutoff distance when these errors are tolerated. Using cutoff distance can reduce a large number of DFT calculations of supercells. Take tetragonal $DyAgTe_2$ (ID: mp-4024 in both Phonon database and

Materials project) as an example, in contrast to 4737 calculations of 128-atom supercells (2×2×4 supercell size) for full elements of third-order force constants, using the smallest cutoff distance reduces 97% of computational costs for DFT calculations with the relative error of 10% in lattice thermal conductivity.

Our study shows that using cutoff distance can significantly accelerate the quantitative prediction of lattice thermal conductivities of materials with marginal error, which is of great importance to high throughput screening of ultra-low or ultra-high lattice thermal conductivity materials.

References

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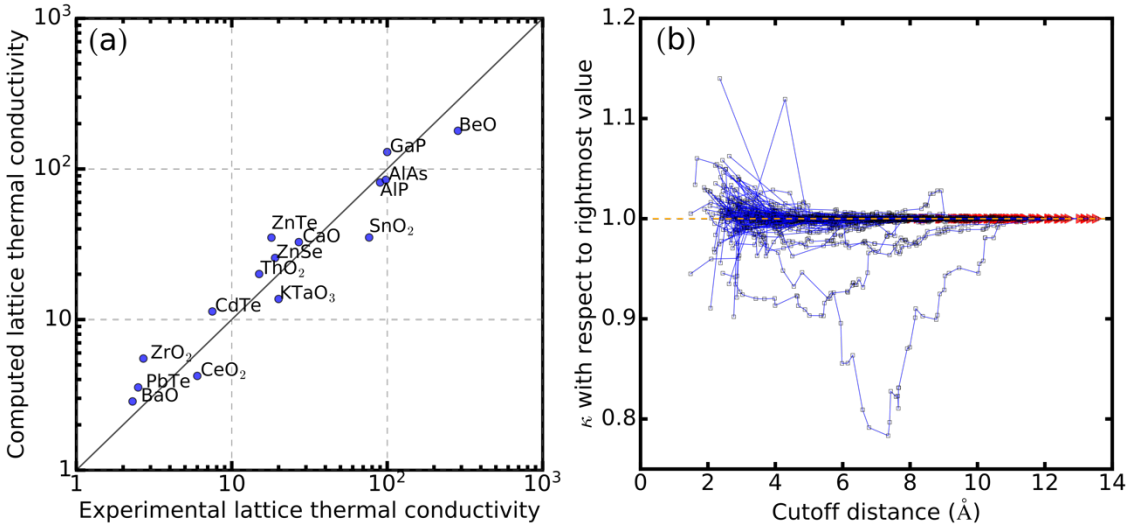

FIG. 1. (a) The comparison between computed lattice thermal conductivities without using cutoff distances (blue circles) and experimental values at room temperature. Note that a logarithmic scale is used in both horizontal and vertical axes. (b) The calculated lattice thermal conductivity (black squares) with respect to the rightmost value (red triangles) at room temperature. The blue lines are only used to guide the eye.

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