Supporting Information: Harnessing the Unusually Strong Improvement of Thermoelectric Performance of $AgInTe_2$ with Nanostructuring

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Nanostructuring is a well-established approach to improve the thermoelectric behavior of materials. However, its effectiveness is restricted if excessively small particle sizes are necessary to considerably decrease the lattice thermal conductivity. Furthermore, if the electrical conductivity is unfavorably affected by the nanostructuring, it could cancel out the advantages of this approach. Computer simulations predict that silver indium telluride, $AgInTe_2$, is unique among chalcopyrite-structured chalcogenides in requiring only a mild reduction of particle size to achieve a substantial reduction in lattice thermal conductivity. Here, ab-initio calculations and machine learning are combined to systematically chart the thermoelectric properties of nanostructured $AgInTe_2$, in comparison with the Cu-based counterpart, $CuInTe_2$. In addition to the temperature and doping carrier concentration dependence, ZT is calculated for both materials as functions of the polycrystalline average grain size, taking into account the effect of nanostructuring on both phonon and electron transport. It is shown that the different order of magnitude between the mean free path of electrons and phonons disentangles the connection between the power factor and lattice thermal conductivity when reducing the crystal size. ZT values up to 2 are predicted for p-type $AgInTe_2$ at 700 K when the average grain size is in the affordable 10-100 nm range.

I. MEAN FREE PATHS

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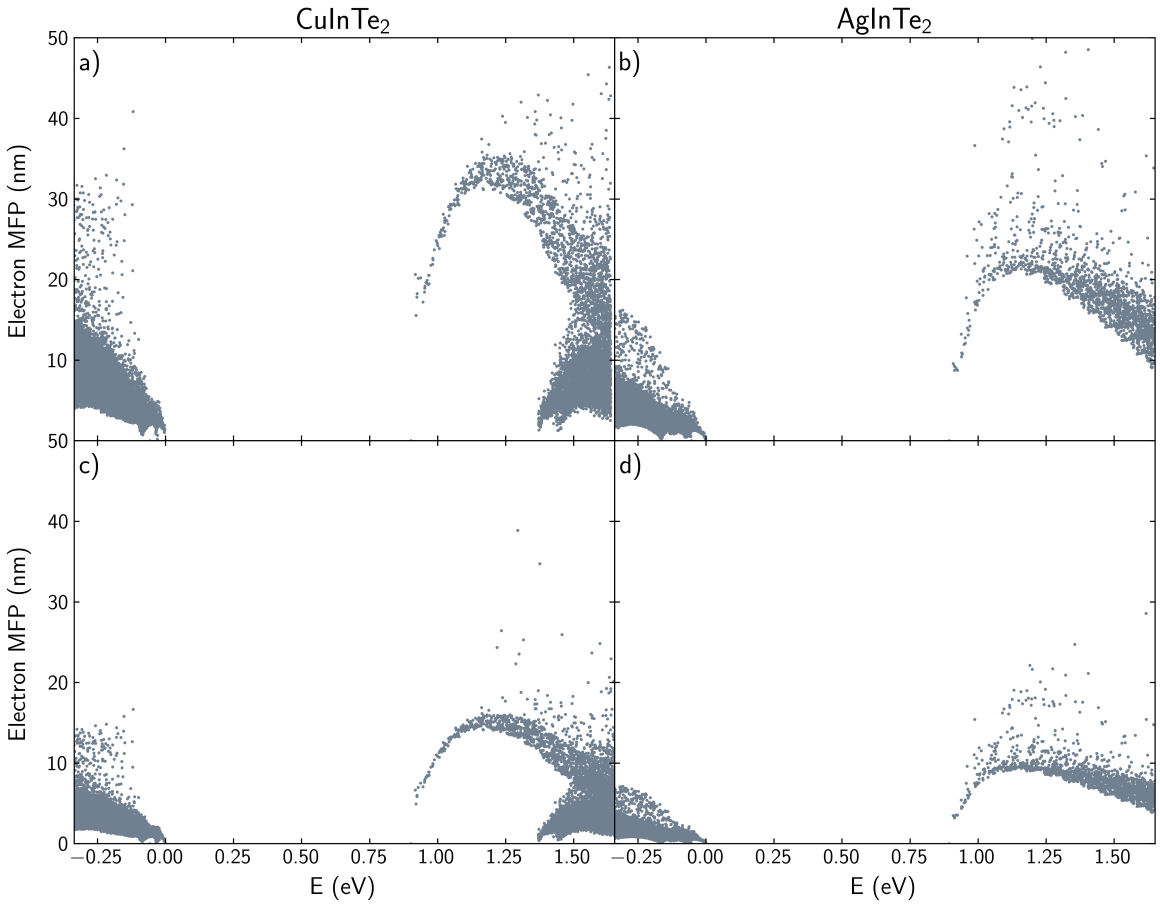

FIG. 1. Electron mean free path for $CuInTe_2$ and $AgInTe_2$ at 300 and 700 K.

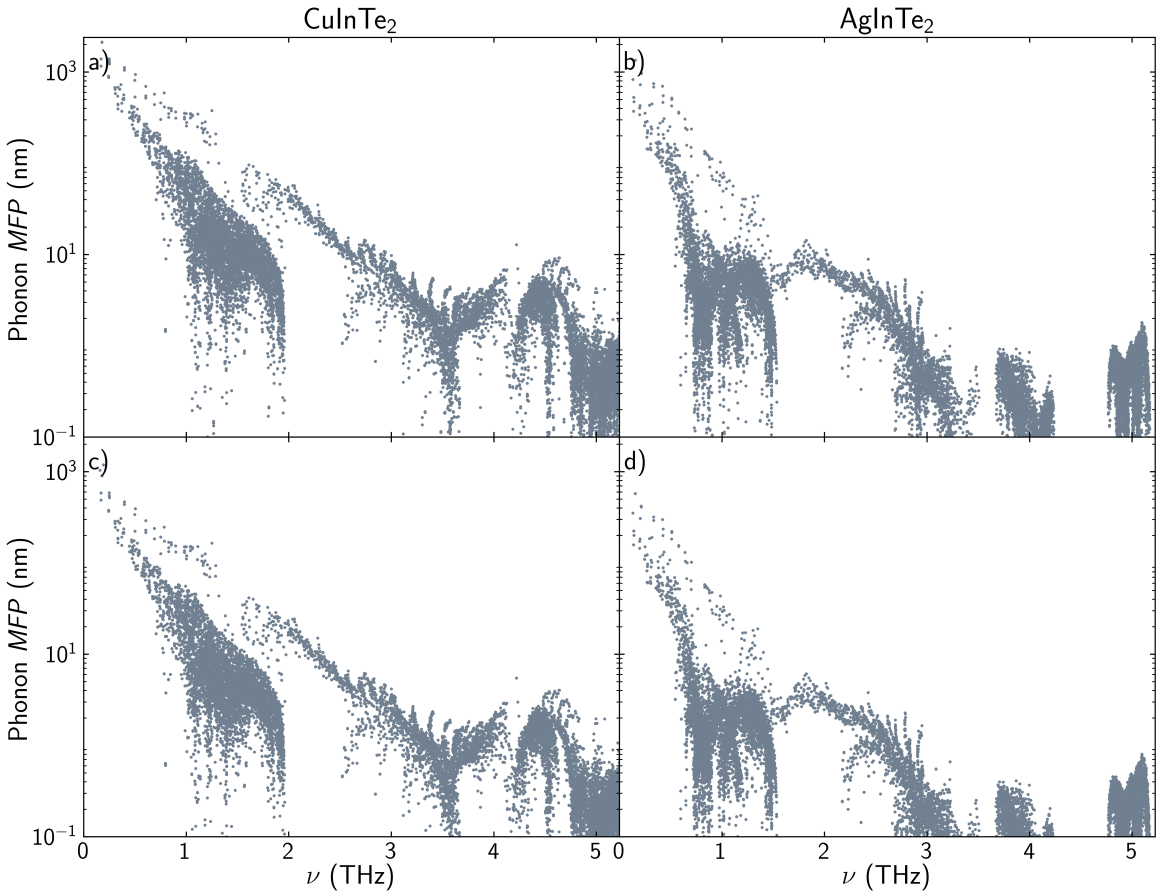

FIG. 2. Phonon mean free path for $CuInTe_2$ and $AgInTe_2$ at 300 and 700 K.

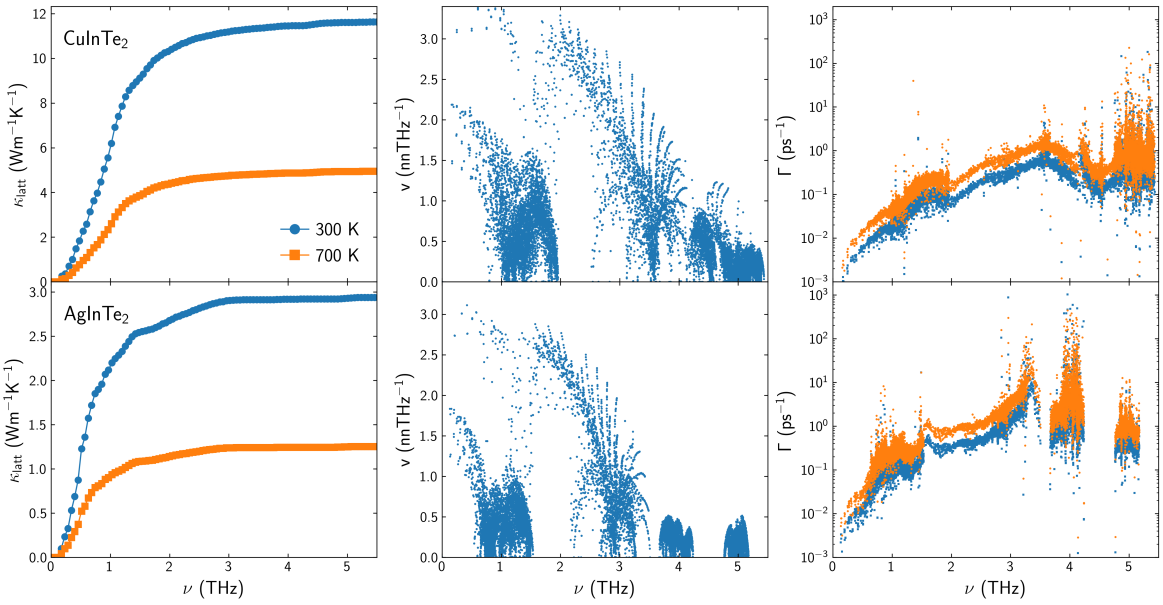

FIG. 3. Left column: Cummulative lattice thermal conductivity for $CuInTe_2$ and $AgInTe_2$ at 300 (blue) and 700 K (orange). Mid column: Group velocities for $CuInTe_2$ and $AgInTe_2$. Right Colum: Scattering rates for $CuInTe_2$ and $AgInTe_2$ at 300 (blue) and 700 K (orange)