Supplementary

Anharmonic atomic dynamics and thermal transport in SnTe with stoichiometric defects

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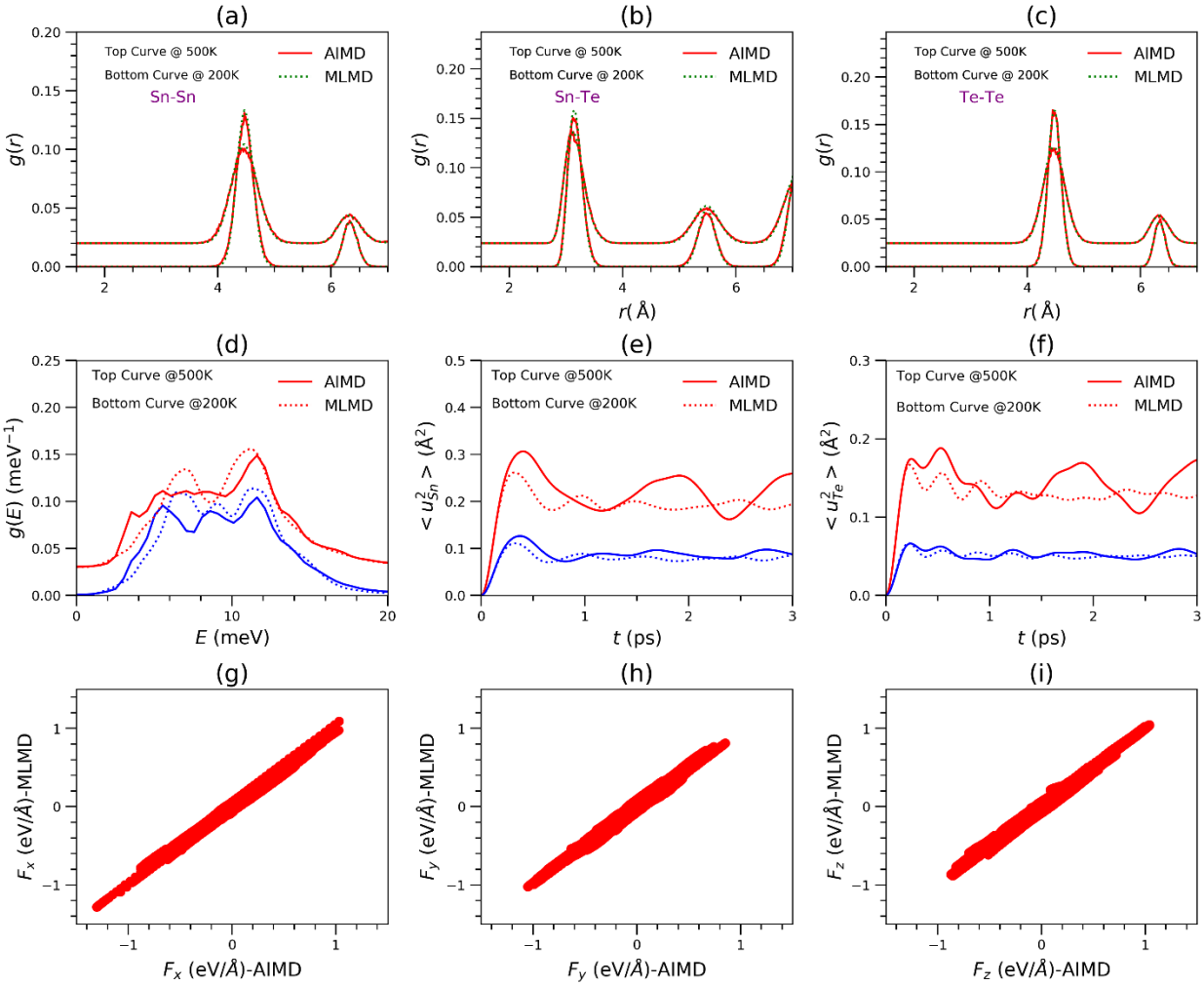

Figure S1. The benchmarking of Machine Learned Force Field by comparing the calculated (a-c) PDF, (d) PDOS and (e-f) MSD of SnTe using AIMD (solid line) and MLMD (dash line) at 200 K and 500 K, showing good overall agreement between the AIMD and MLMD results. (g-i) The computed forces from AIMD vs MLMD force-field on various configurations validate the model.

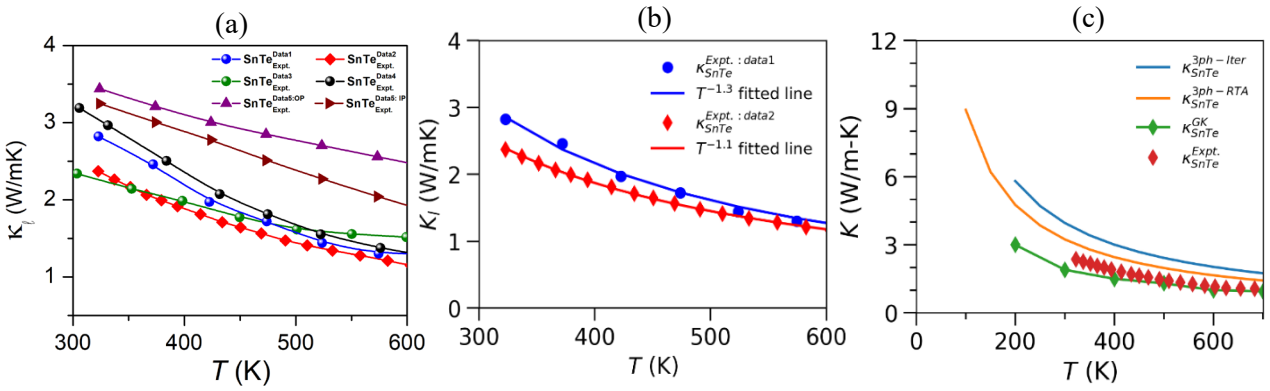

Figure S2.: (a) The measured lattice thermal conductivity of SnTe, reported in literatures [1-5], shows a significant variation in measured lattice thermal conductivity. (b) The temperature dependence of measured lattice thermal conductivity of SnTe [1, 2] show deviation from $1/T$ infers the presence of higher-order anharmonicty. (c) Our calculated lattice thermal conductivity of SnTe using different theoretical methods and compared with measured lattice thermal conductivity[2].

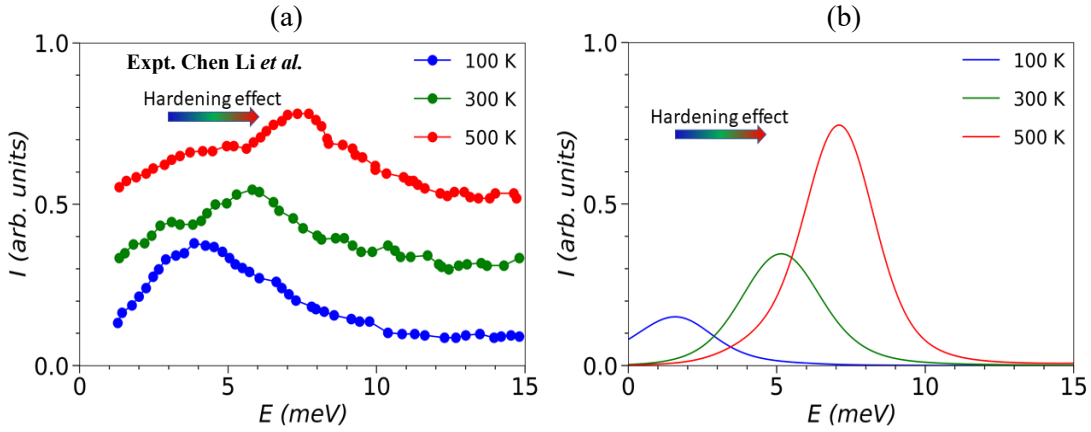

Figure S3: (a) The measured temperature dependence of the lowest zone-center optical phonon mode by Chen Li et al. shows the phonon hardening with temperature[6], inferring the presence of 4th and higher-order phonon scattering. (b) Our calculated temperature dependence of the lowest zone-center optical phonon mode using molecular dynamics trajectories reproduces the hardening of phonon. This confirms that our simulations correctly capture the actual anharmonic effects, including higher-order anharmonicity, and validate the model's robustness.

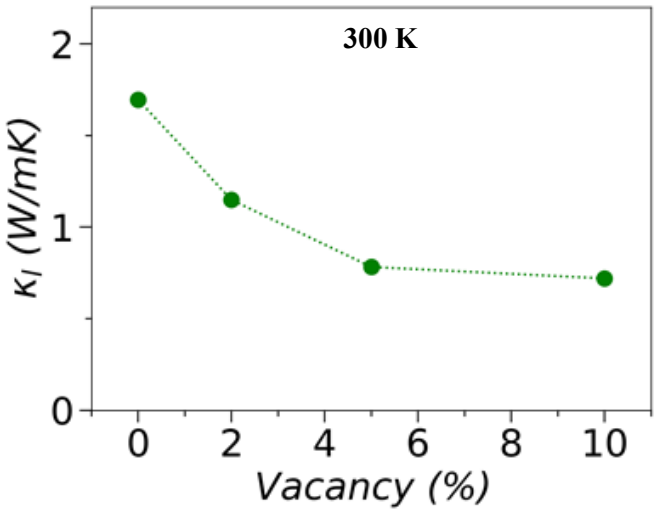

Figure S4: The calculated lattice thermal conductivity with different Sn vacancy concentrations at 300K using the Green-Kubo method. The Sn vacancy concentration plays a significant role in lowering the lattice thermal conductivity.

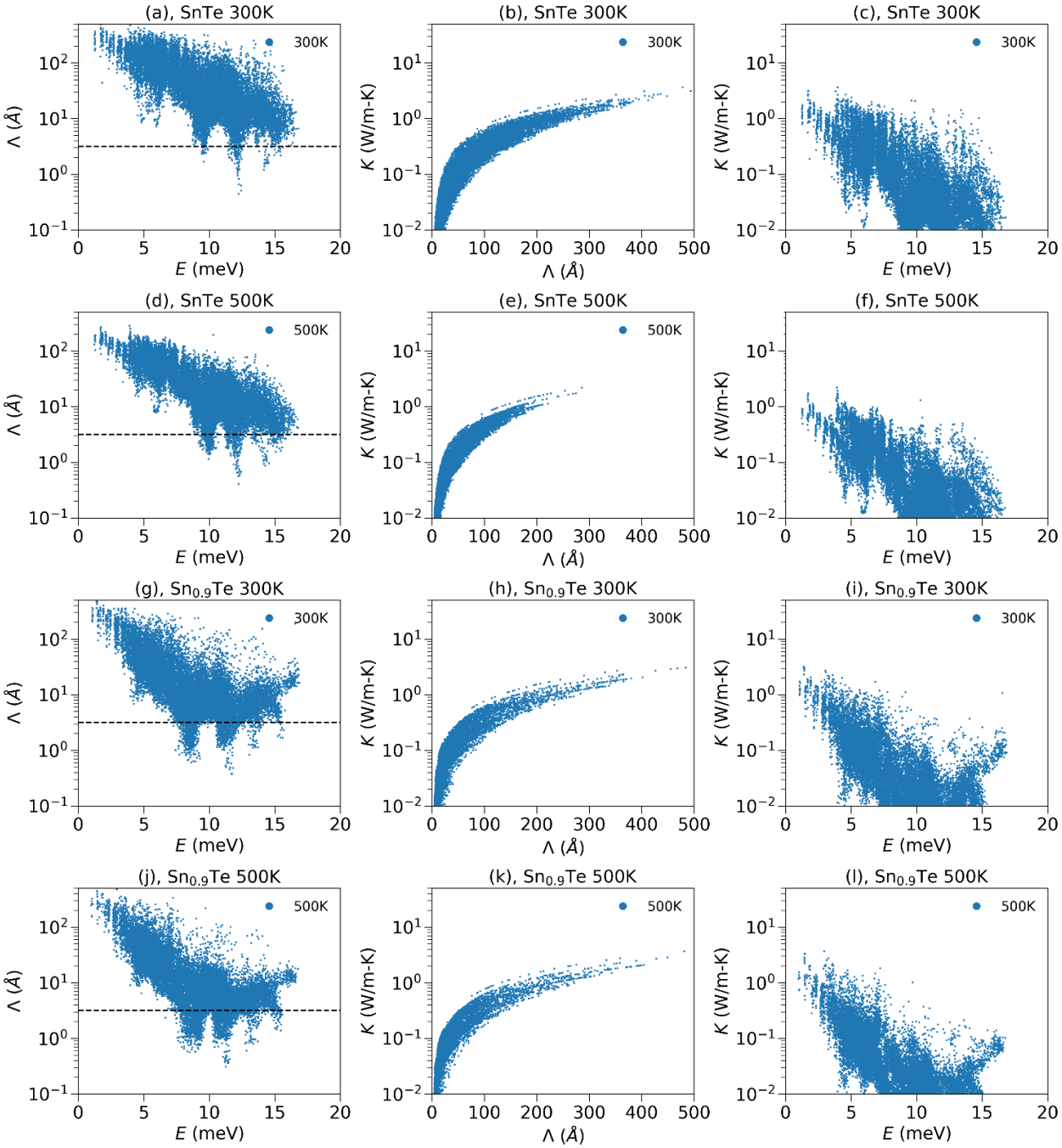

Figure S5: The calculated phonon mean free path (Λ), lattice thermal conductivity (κ) as a function of phonon mean free path, and phonon energies in the entire Brillouin zone using molecular dynamics trajectories at 300 K and 500 K for both (a-f) SnTe and (g-l) $Sn_{0.9}Te$.

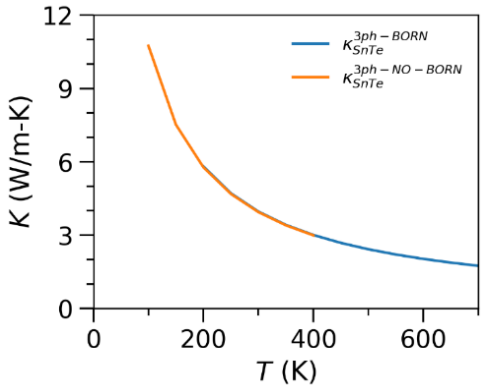

Figure S6: The calculated lattice thermal conductivity with and without including the Born effective charges. The Born-effective charges do not influence the lattice thermal conductivity.

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