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Polytelluride square planar chain induced anharmonicity results in ultralow thermal conductivity and high thermoelectric efficiency in Al_2Te_5 monolayers

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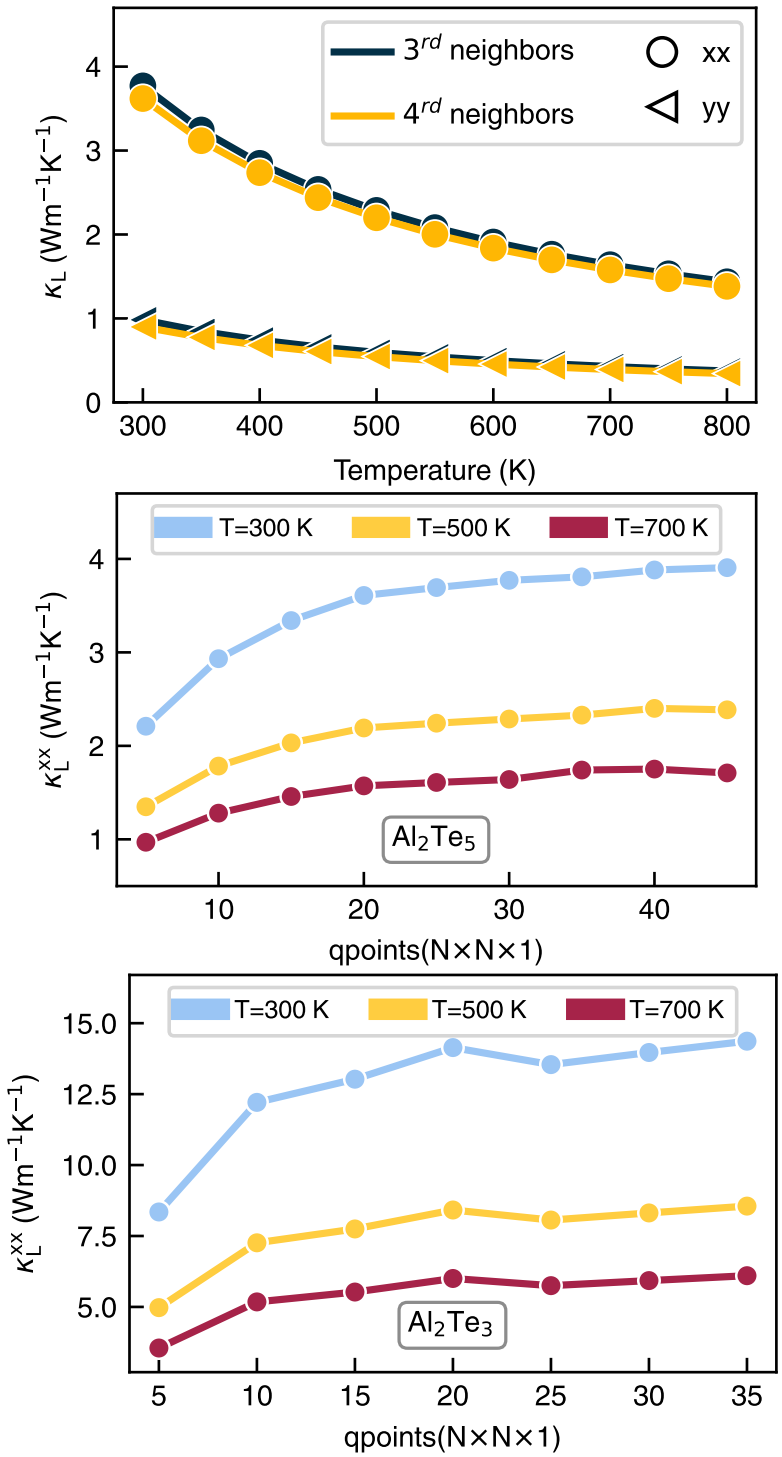

Fig S1: Convergence test for κ_{L} as function of q-point grid and interaction cutoff.

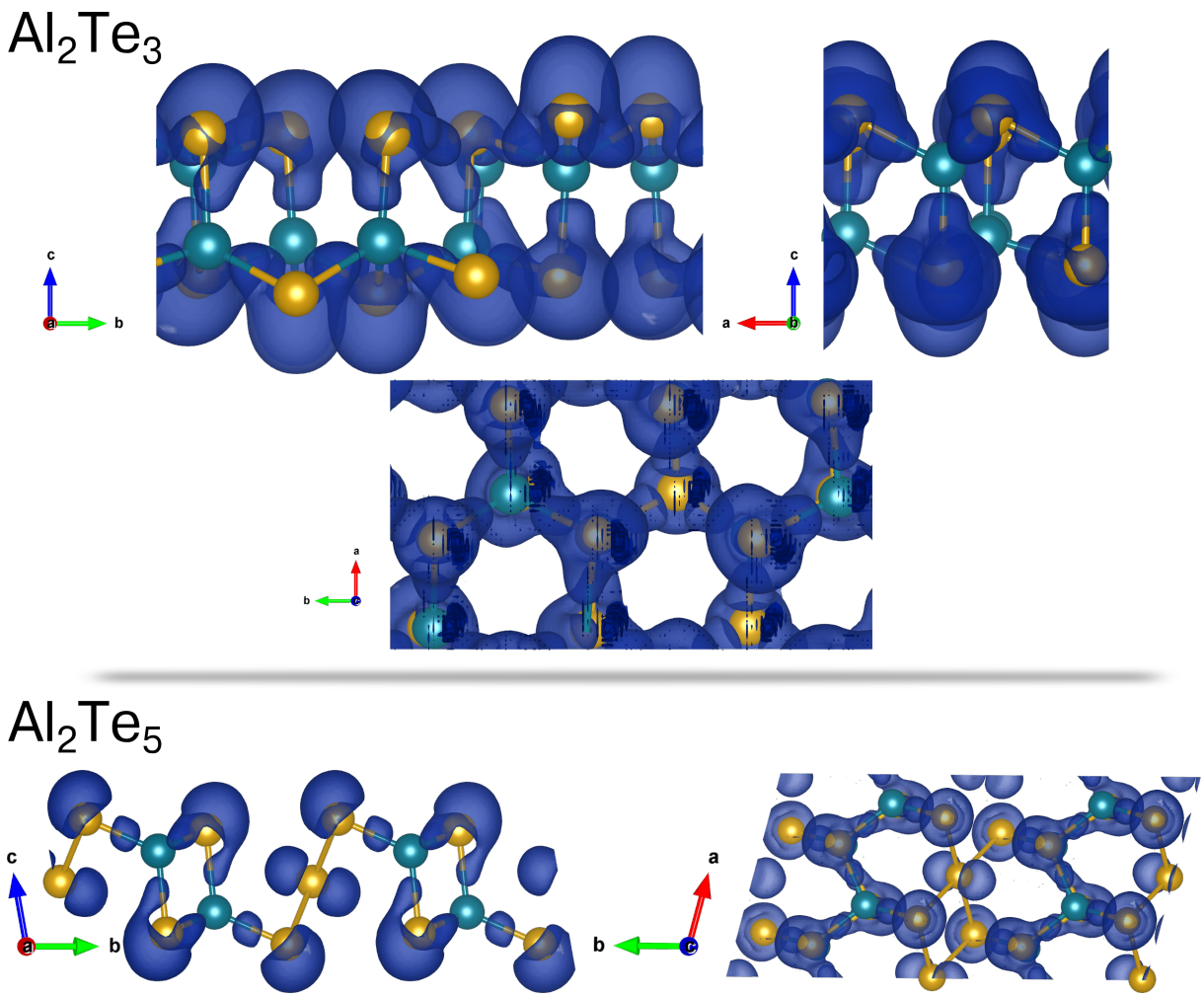

Fig S 2: Electron localization function (ELF) plots of Al_2Te_3 and Al_2Te_5 from different viewpoints.

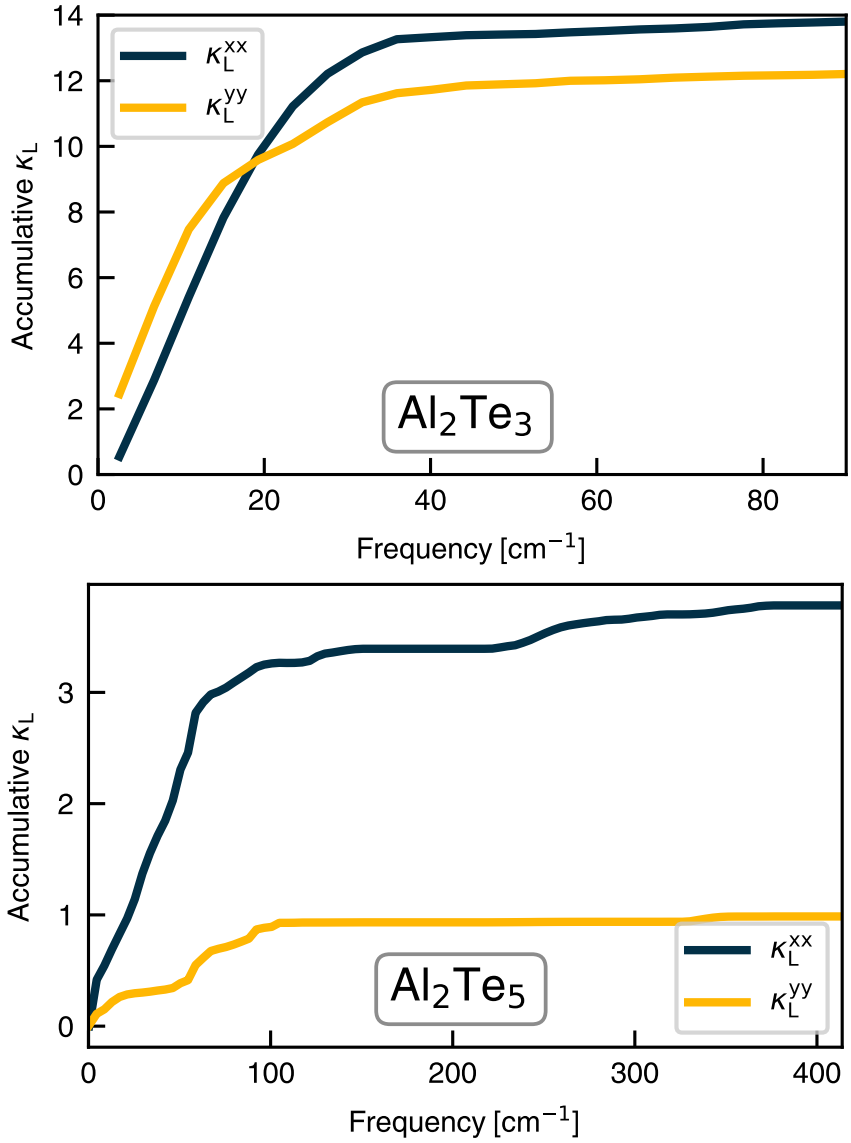

Fig S 3: Accumulative κ_L versus frequency for Al_2Te_3 and Al_2Te_5 at 300K. The figure reveals that lattice thermal transport is dominated by phonon modes with frequencies less than 40 cm^{-1} and 100 cm^{-1} for Al_2Te_3 and Al_2Te_5, respectively.

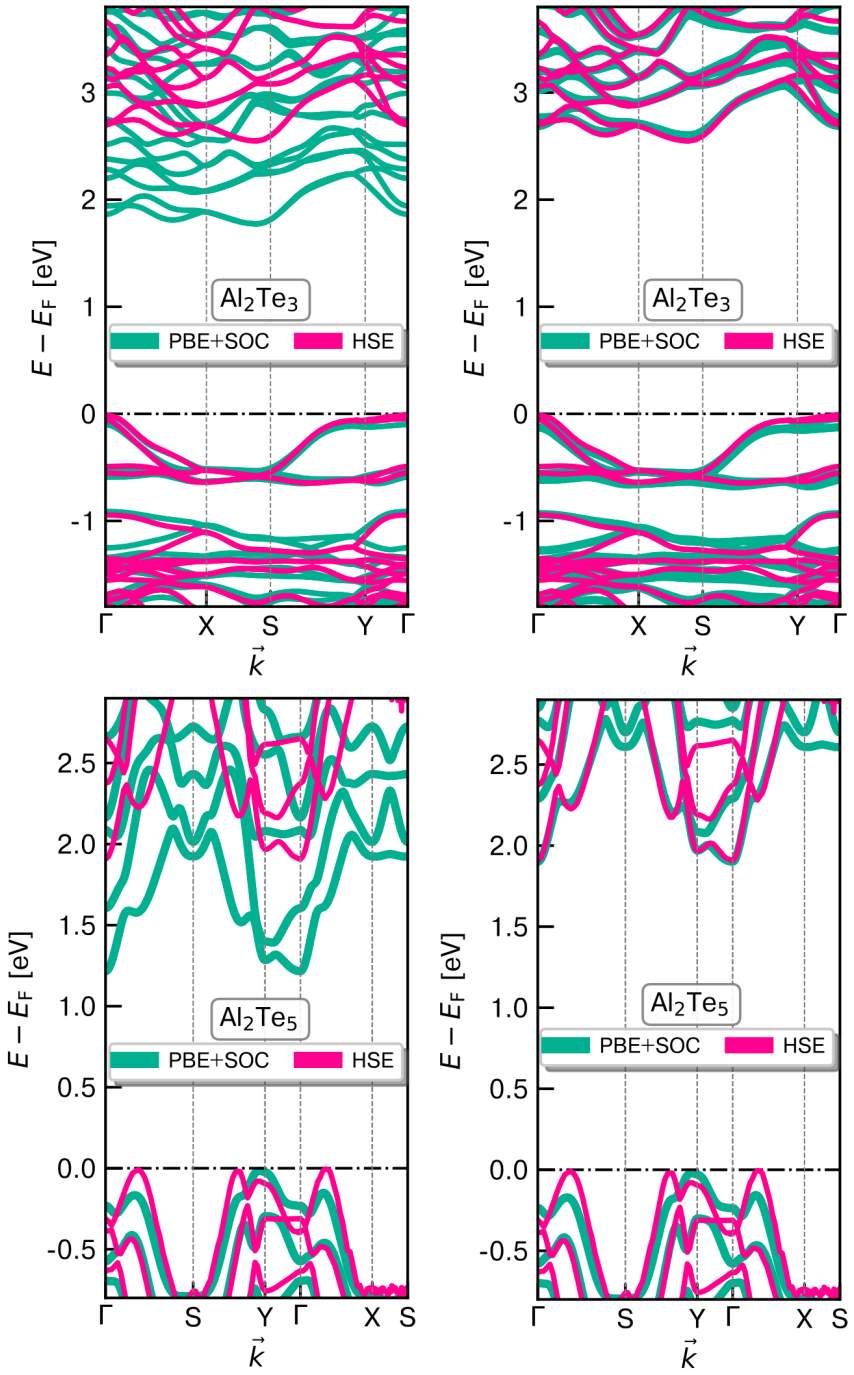

Fig S4: Overlay of the HSE and PBE+SOC band structures of Al_2Te_3 and Al_2Te_5. To simplify the comparison, the right panel shifts the PBE band gap by 0.697 eV for Al_2Te_3 and 0.651 eV for Al_2Te_5. The HSE-calculated band structure of Al_2Te_5 exhibits enhanced valley degeneracy and changes in band curvature, while its CB and also that of Al_2Te_3 remains similar to the PBE-calculated one.

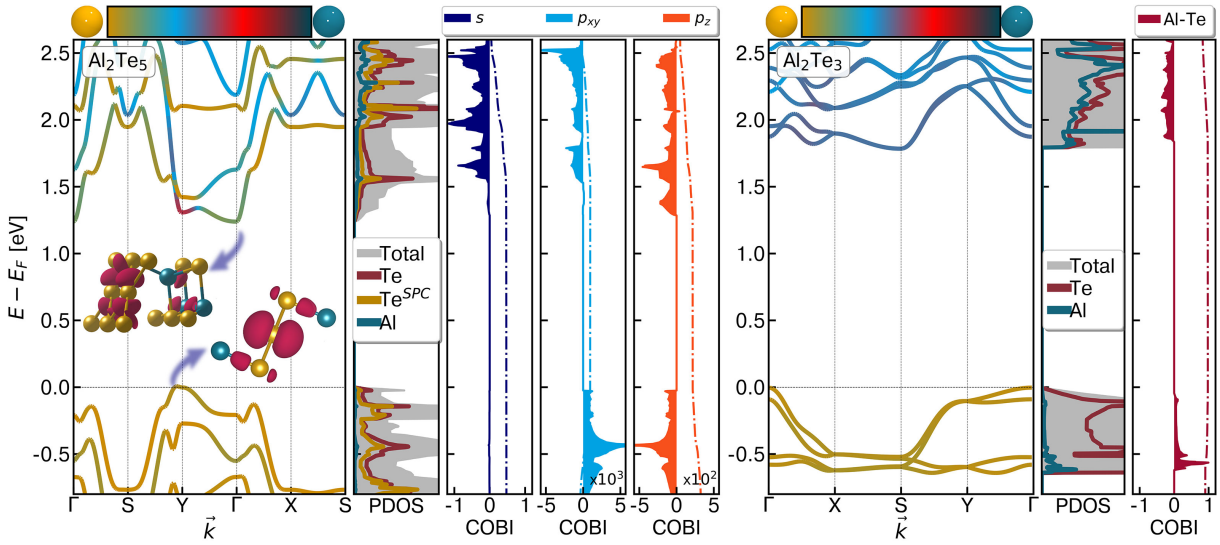

Fig S5: PBE+SOC functional atom-projected band structures, density of states, and COBI plots for Al_2Te_5 (left panel) and Al_2Te_3 (right panel). The inset in the band structure plot of Al_2Te_5 depicts the charge decomposition of the valence band maximum (VBM) and conduction band minimum (CBM).

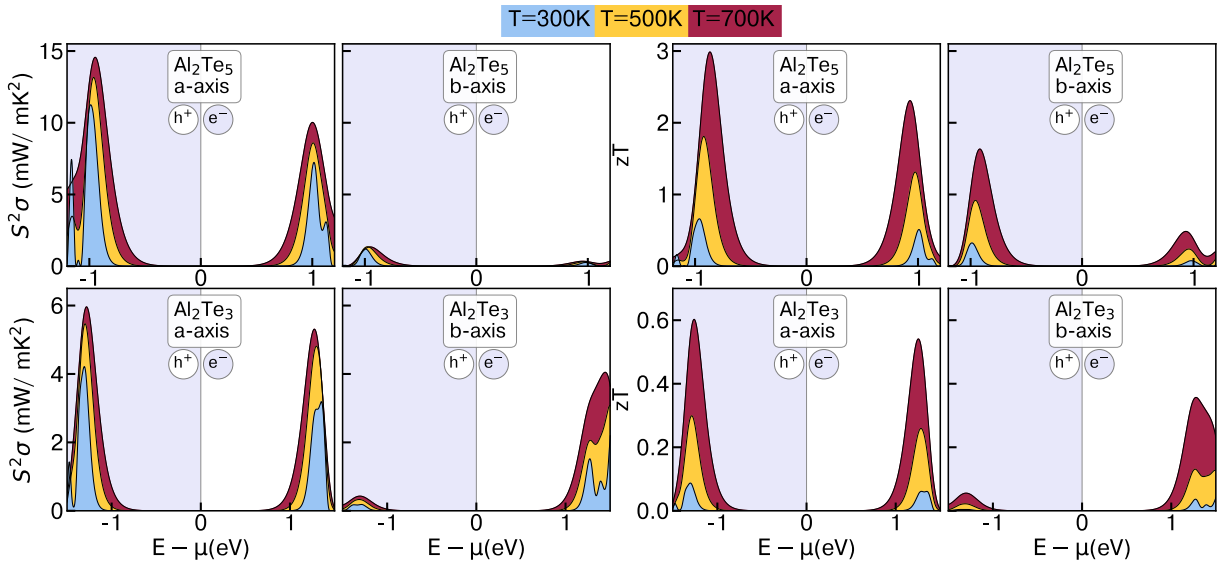

Fig S 6: Thermoelectric properties of Al_2Te_5 and Al_2Te_3 along the a- and b-axes were calculated using the HSE functional and the constant relaxation time approximation (CRTA) with a relaxation time of $\tau = 1\times10^{-14}$s.

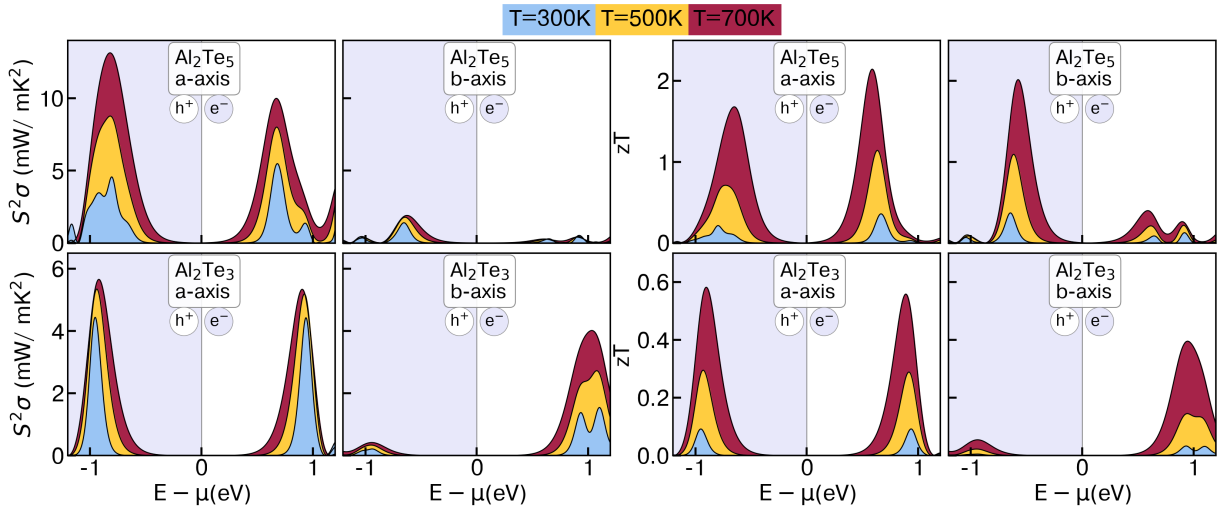

Fig S 7: The electron and hole thermoelectric properties of Al_2Te_5 and Al_2Te_3 along the a- and b-axes were calculated using the PBE+SOC functional and the constant relaxation time approximation (CRTA) with a relaxation time of $\tau = 1\times10^{-14}$s.

Table S 1: The high symmetry k-points coordinates used for representation band structure and phonon dispersion.

	Lable	k_x	k_y	k_z
Al_2Te_5	Γ	0.0000	0.0000	0.0000
	S	0.5000	0.5000	0.0000
	X	0.5000	0.0000	0.0000
	Y	0.0000	0.5000	0.0000
Al_2Te_3	Γ	0.0000	0.0000	0.0000
	X	0.5000	0.0000	0.0000
	S	0.5000	0.5000	0.0000
	Y	0.0000	0.5000	0.0000

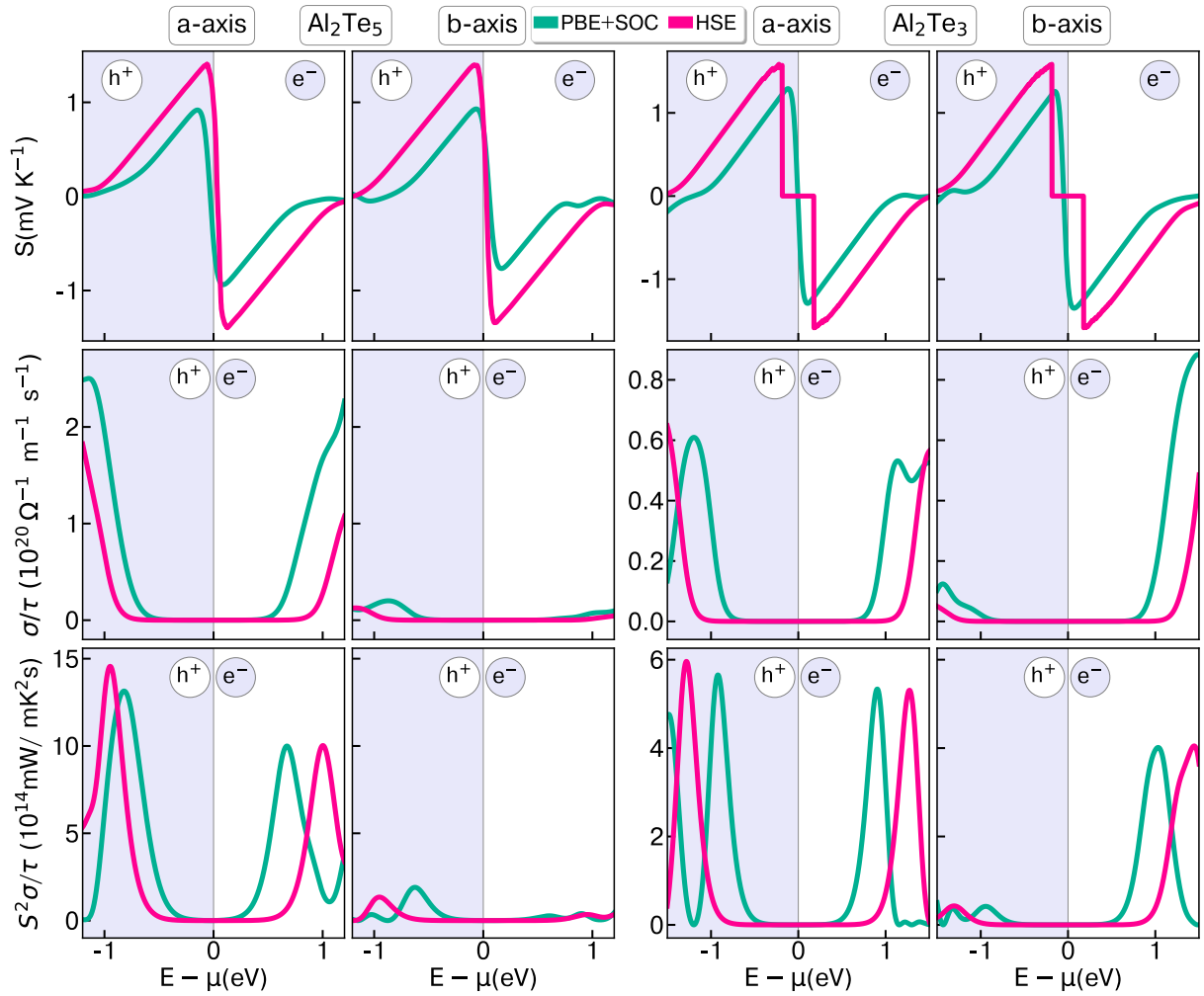

Fig S8: Calculated Seebeck coefficients (S), electrical conductivity (σ), and power factor (PF) of Al_2Te_5 and Al_2Te_3 using PBE and HSE methods at 700K. The HSE-calculated S is larger than that of PBE due to its dependence on the band gap. However, if the band gap widens, akin to the scissor operator, the enhancement of S and σ counterbalances, leaving PF unchanged—a critical factor for determining zT. Al_2Te_3 exemplifies this behavior. However, in Al_2Te_5, HSE alters the band curvature and increases the valley degeneracy of VB, slightly enhancing the p-type PF while leaving the CB and n-type PF unchanged.

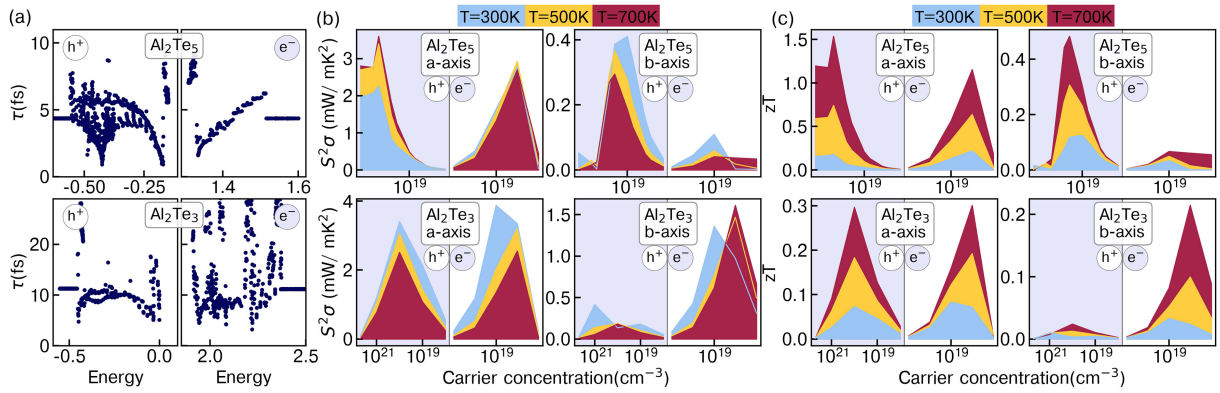

Fig S 9: The electron and hole thermoelectric properties of Al_2Te_5 along the a- and b-axes were calculated using the PBE+SOC functional and accurate relaxation time that contain all scattering rates.

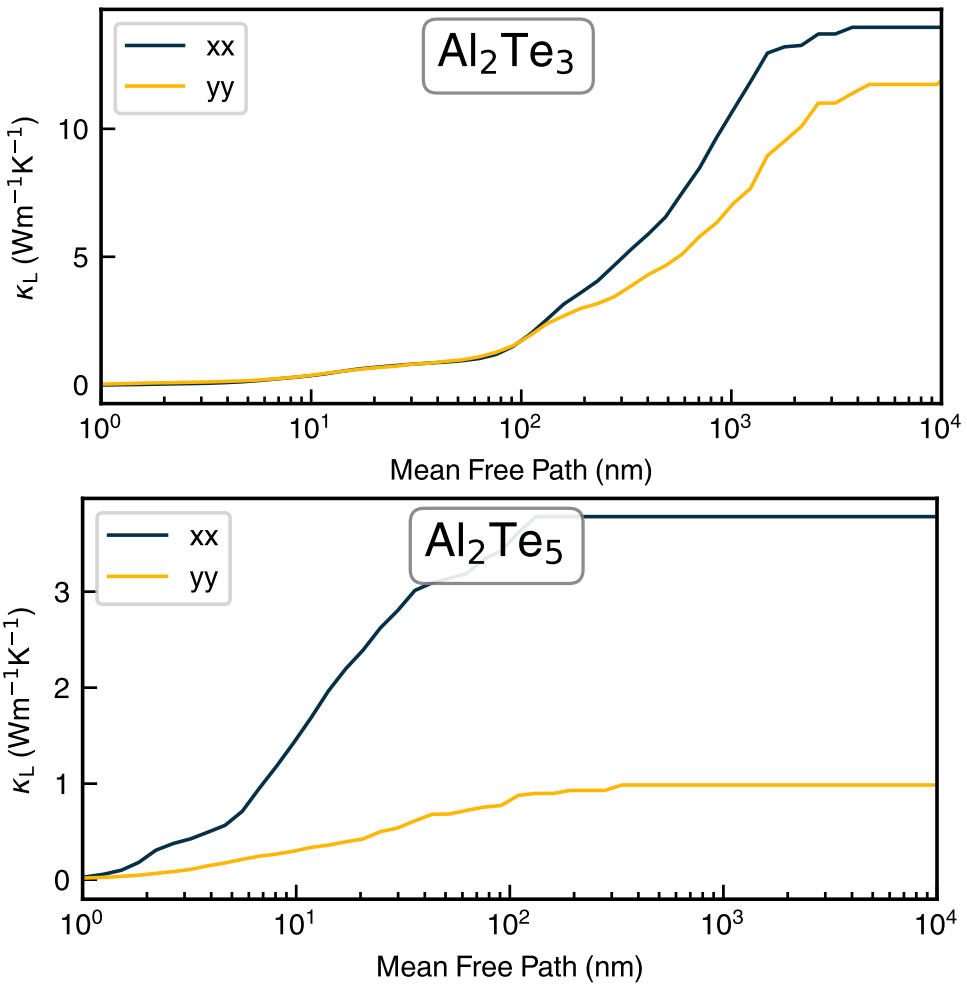

Fig S10: Accumulative κ_L versus mean free path for Al_2Te_3 and Al_2Te_5 at 300K. As the figure illustrates, lattice thermal transport could decrease by half with nanostructuring at sizes of approximately 500 nm and 50 nm for Al_2Te_3 and Al_2Te_5, respectively.

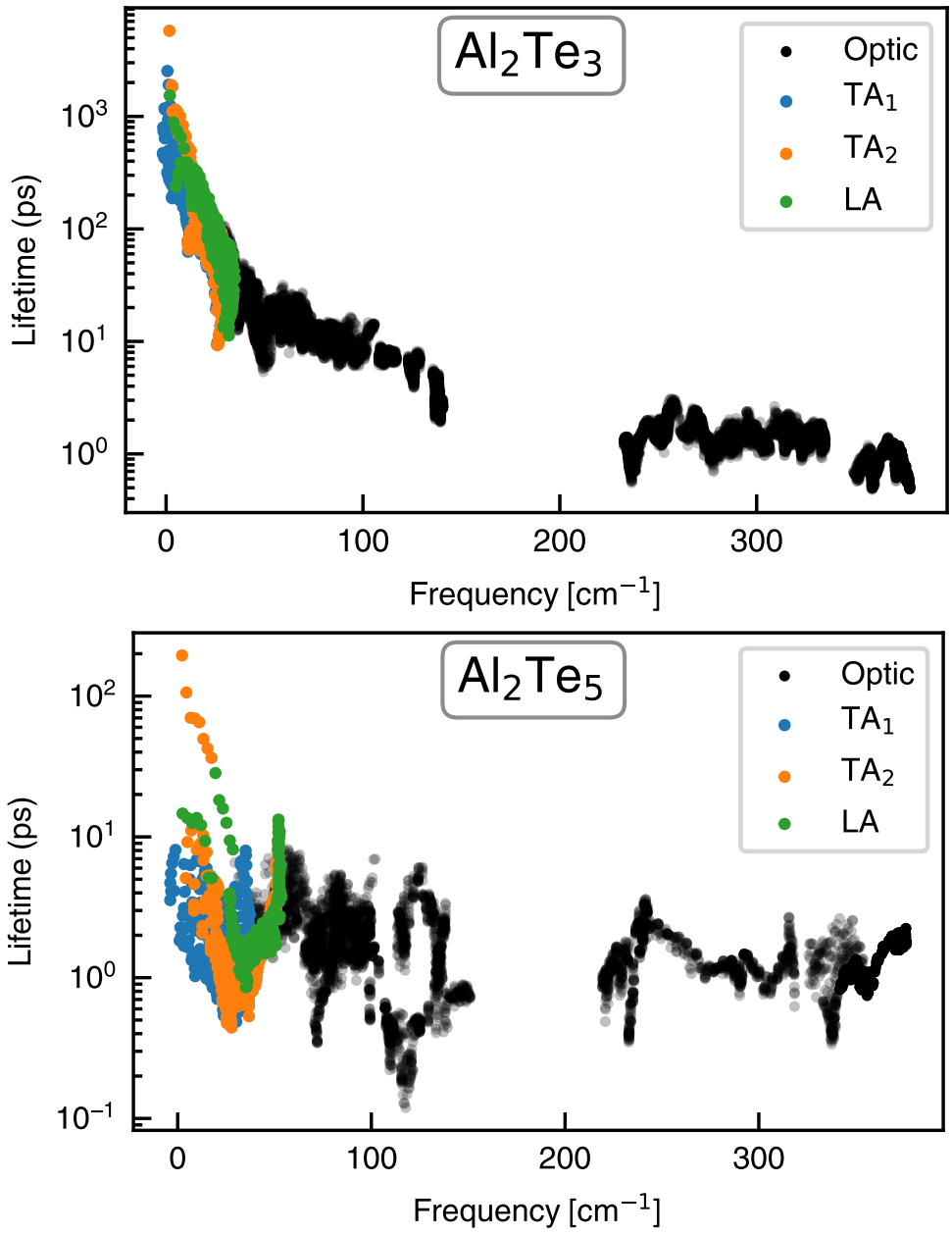

Fig S11: The calculated phonon lifetimes correspond to the phonon frequency at 300K for both materials. As the figures clearly depict, the phonon lifetime for Al_2Te_5 is almost one order of magnitude lower than that for Al_2Te_3, resulting in higher scattering rates and lower phonon thermal transport in Al_2Te_5.