Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Electronic supplementary information for High thermoelectric figure of merit in *p*-type $Mg_3Si_2Te_6$: Role of multi-valley bands and high anharmonicity

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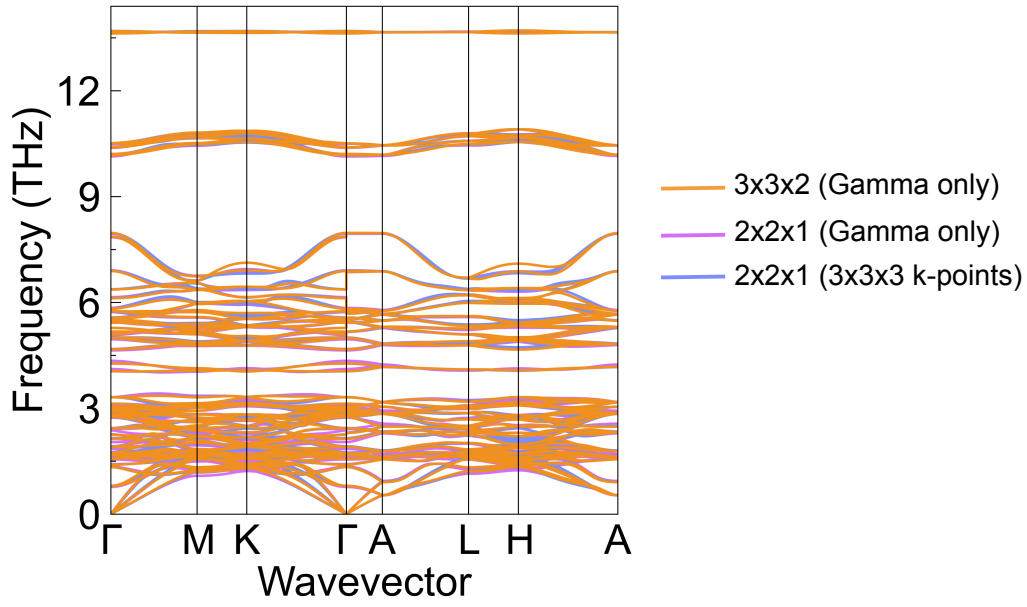

FIG. S1. Convergence of phonon dispersion with respect to the supercell size. Phonon frequencies are well converged on a 2×2×1 supercell.

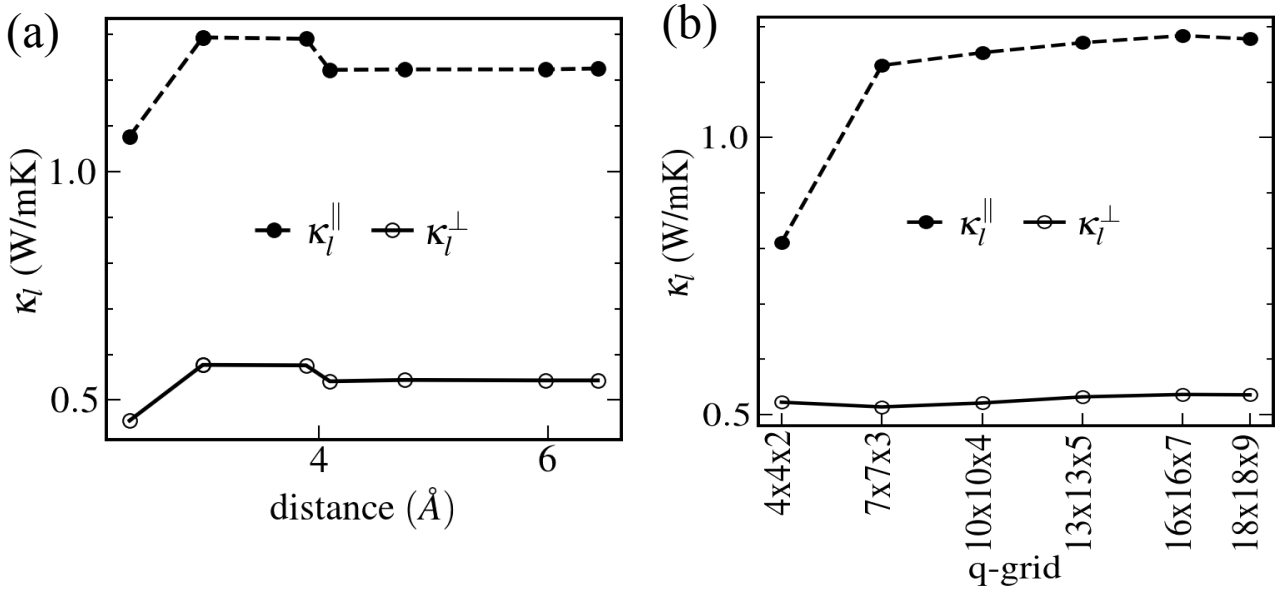

FIG. S2. Convergence of the room-temperature lattice thermal conductivity with respect to (a) nearest-neighbor cutoff used in 3rd order inter-atomic force constants, and (b) integration grid. Lattice thermal conductivity is well converged at interaction cutoff >5 , and integration grid of 13×13 ×5.

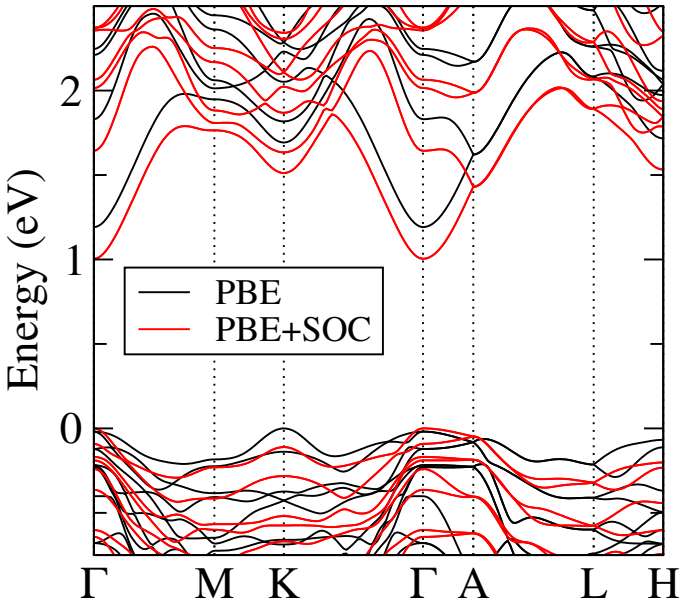

FIG. S3. Comparison of $Mg_3Si_2Te_6$ band structure calculated without (PBE) and with spin-orbit coupling (PBE+SOC).

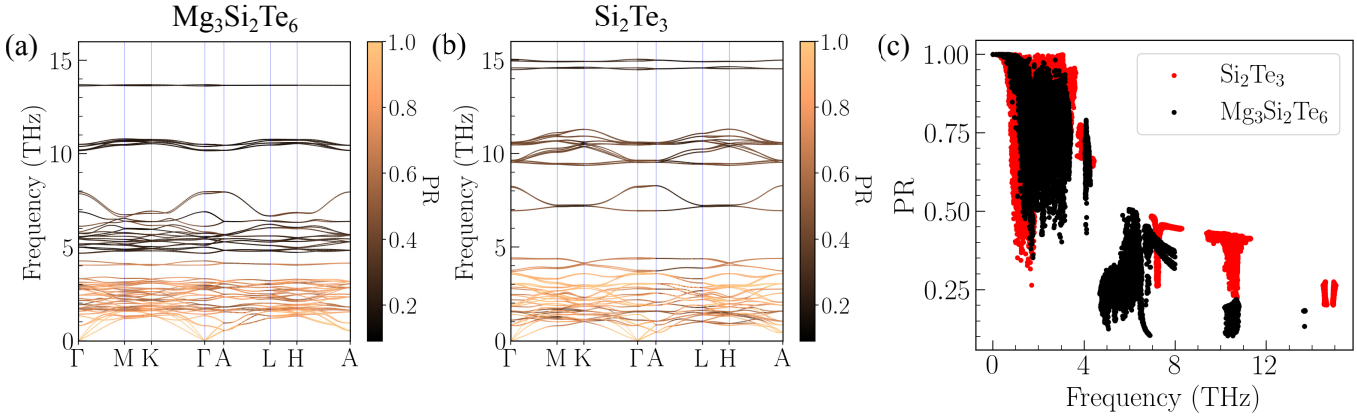

FIG. S4. Calculated participation ratio (PR) along the high symmetry path, in (a) $Mg_3Si_2Te_6$ and (b) Si_2Te_3. (c) Comparison of PR in both compounds in the entire Brillouin zone. Lower PR value in $Mg_3Si_2Te_6$ indicates that phonon modes in this compound are more localized. The most of localization is provided by Mg atoms.

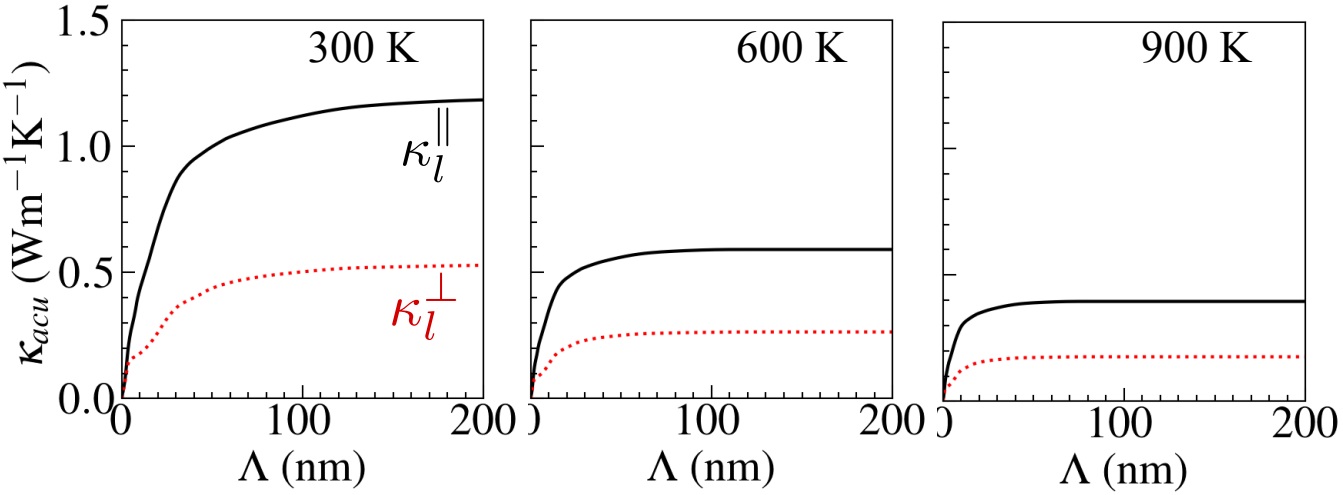

FIG. S5. Cumulative lattice thermal conductivity of $Mg_3Si_2Te_6$ as a function of the mean free path at 300 K, 600 K, and 900 K. For mean free path in the range 10-20 nm, κ_l can be reduced by ∼50 %.

TABLE S1. $Mg_3Si_2Te_6$ parameters used for the electronic transport property calculations within AMSET code. **C** is the elastic tensor in Voigt notation. $\boldsymbol{\epsilon}_s$ and $\boldsymbol{\epsilon}_\infty$ are the static and high-frequency dielectric constants, respectively. ω_{PO} is the effective polar phonon frequency.

C (in GPa) =	$\begin{bmatrix} 68.14 & 19.39 & 11.57 & 0 & -2.0 & 0 \\ 19.39 & 68.14 & 11.57 & 0 & 2.0 & 0 \\ 11.57 & 11.57 & 39.20 & 0 & 0 & 01 \\ 0 & 0 & 0 & 13.58 & 0 & 2.0 \\ -2.0 & 2.0 & 0 & 0 & 13.58 & 0 \\ 0 & 0 & 0 & 2.0 & 0 & 24.33 \end{bmatrix}$
$\boldsymbol{\epsilon}_s$ (in ϵ_0) =	$\begin{bmatrix} 15.45 & 0 & 0 \\ 0 & 15.45 & 0 \\ 0 & 0 & 12.37 \end{bmatrix}$
$\boldsymbol{\epsilon}_\infty$ (in ϵ_0) =	$\begin{bmatrix} 9.95 & 0 & 0 \\ 0 & 9.95 & 0 \\ 0 & 0 & 7.26 \end{bmatrix}$
ω_{PO}(in THz) =	5.85

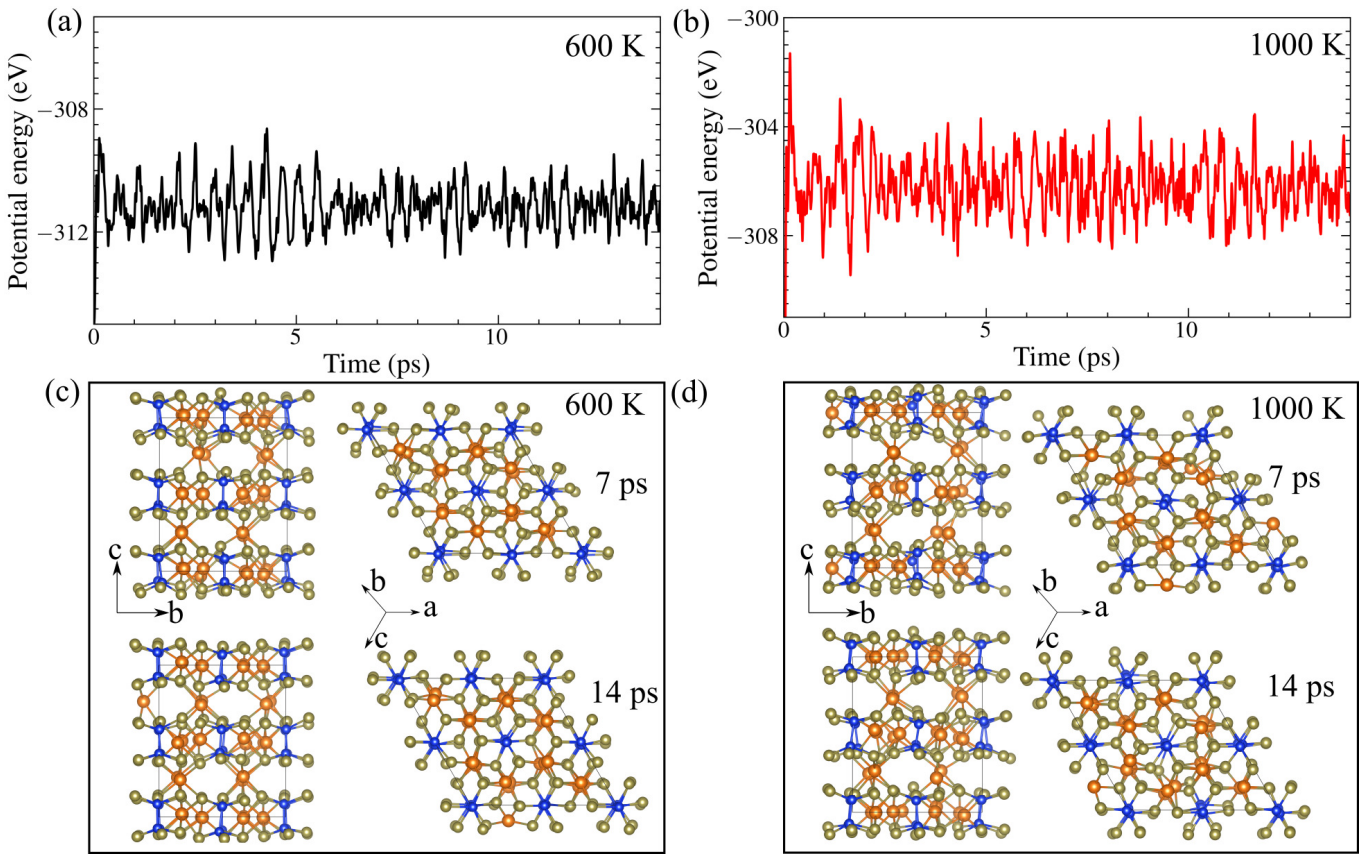

FIG. S6. (color online) Potential energy profiles of $Mg_3Si_2Te_6$ during the AIMD simulation at (a) 600 K and (b) 1000 K, respectively. The corresponding 600 and 1000 K snapshots of atomic configurations at 7 and 14 ps are also shown in (c) and (d), respectively.