Supplementary materials

# Novel insights into lattice thermal transport in nanocrystalline $Mg_3Sb_2$ from first principles: The crucial role of higher-order phonon scattering

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## Convergence test

Regarding the convergence test of $\kappa$ at 300 K *versus* different **Q**-points mesh along in- and cross-plane directions depicted in Figure S1, we observe that considering only three phonon scattering requires a 25×25×25 **Q**-point mesh to produce converged values, while the inclusion of three- and four-phonon scatterings only requires a 13×13×13 **Q**-point mesh to converge. Meanwhile, we also perform calculations on the above convergence **Q**-points mesh with scalebroad=1.0. Furthermore, it can be clearly seen that both $\kappa_{xx}$(3+4ph) and $\kappa_{zz}$(3+4ph) represent two convergence trends that is similar to $\kappa_{xx}$(3ph) and $\kappa_{zz}$(3ph), respectively.

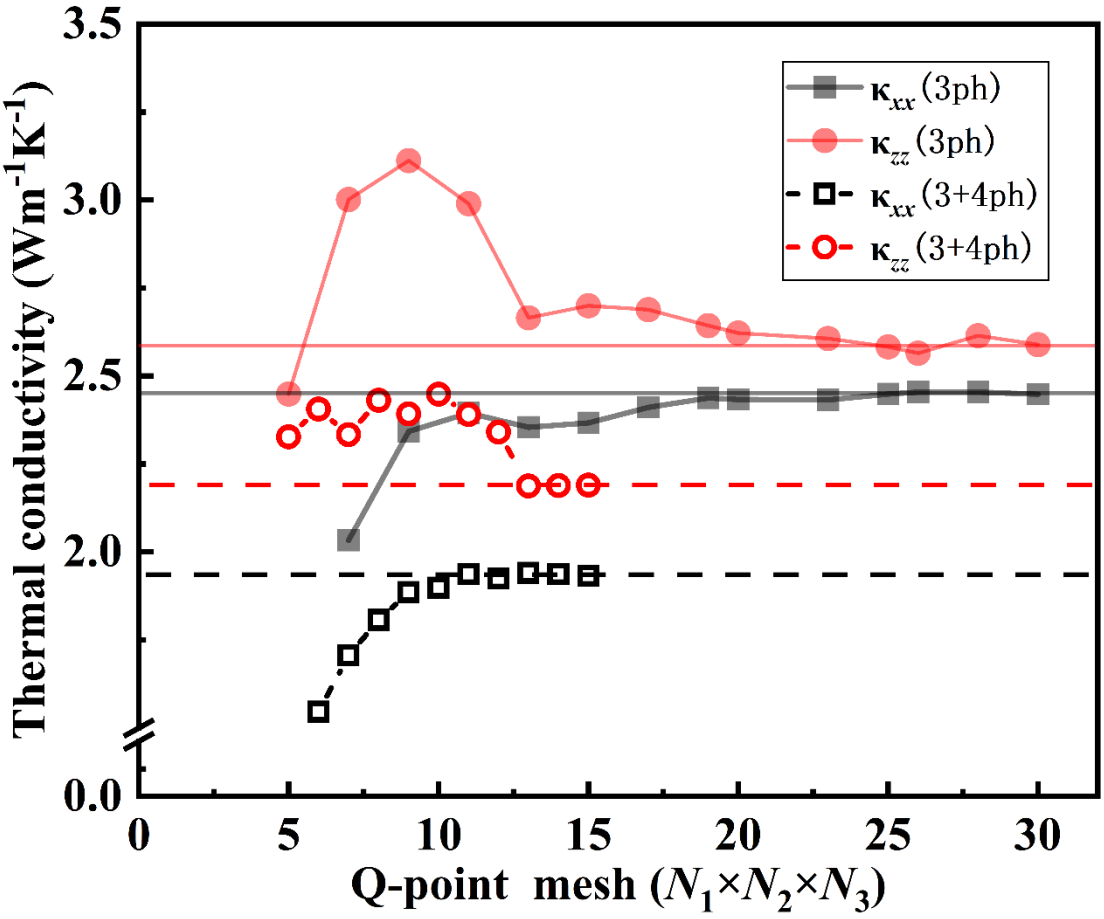


Figure S1. Variation of the computed lattice thermal conductivity ($\kappa$) of $Mg_3Sb_2$ at 300K considering only three-phonon scattering (3ph) and both three- and four-phonon scatterings (3+4ph) along in- and cross-plane directions with respect to different **Q**-points mesh.

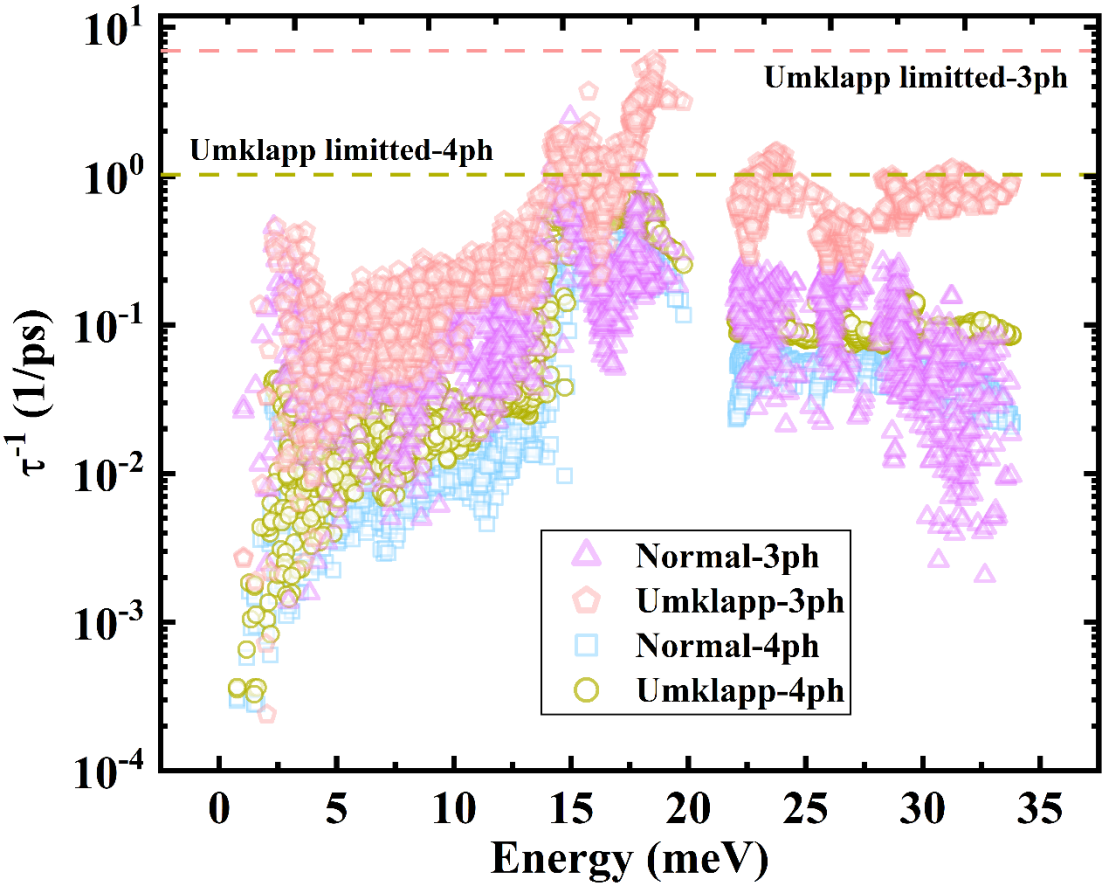


Figure S2. Phonon model level scattering rate decomposed into Normal- and Umklapp-processes in 3ph- and 4ph-scattering interactions.

Table S1. Percentage contribution of different phonon branches to $\kappa$ for occurring $Mg_3Sb_2$ at 300 K and 800 K, respectively.

| Material | $\kappa$ % | 300K-3-ph only | | 300K-3+4ph | | 800K-3ph only | | 800K-3+4ph | |
|---|---|---|---|---|---|---|---|---|---|
| | | xx | zz | xx | zz | xx | zz | xx | zz |
| $Mg_3Sb_2$ | TA1 | 28.54 | 15.59 | 27.54 | 16.35 | 28.25 | 15.52 | 25.97 | 16.38 |
| | TA2 | 25.88 | 38.96 | 26.87 | 36.07 | 25.65 | 38.76 | 26.97 | 36.44 |
| | LA | 22.65 | 35.11 | 21.11 | 36.96 | 22.52 | 35.01 | 20.49 | 35.68 |
| | Optical | 22.93 | 10.34 | 24.48 | 10.62 | 23.58 | 10.71 | 26.57 | 11.50 |