Thermal Conductivities in Solids from First Principles: Accurate Computations and Rapid Estimates

CHRISTIAN CARBOGNO, MATTHIAS SCHEFFLER, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

— In spite of significant research efforts, a first-principles determination of the thermal conductivity $\kappa$ at high temperatures has remained elusive. Boltzmann transport techniques that account for anharmonicity perturbatively become inaccurate under such conditions. \textit{Ab initio} molecular dynamics (MD) techniques using the Green-Kubo (GK) formalism capture the full anharmonicity, but can become prohibitively costly to converge in time and size. We developed a formalism that accelerates such GK simulations by several orders of magnitude and that thus enables its application within the limited time and length scales accessible in \textit{ab initio} MD. For this purpose, we determine the effective harmonic potential occurring during the MD, the associated temperature-dependent phonon properties and lifetimes. Interpolation in reciprocal and frequency space then allows to extrapolate to the macroscopic scale. For both force-field and \textit{ab initio} MD, we validate this approach by computing $\kappa$ for Si and ZrO$_2$, two materials known for their particularly harmonic and anharmonic character. Eventually, we demonstrate how these techniques facilitate reasonable estimates of $\kappa$ from existing MD calculations at virtually no additional computational cost.