Modeling heat transport in amorphous $Ge_2Sb_2Te_5$ with a deep neural network interatomic potential

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$Ge_2Sb_2Te_5$ is a phase change compound deployed in non-volatile electronic memories and neuromorphic devices, which rely on transitions between its crystalline and amorphous phases via Joule heating. The lattice thermal conductivity of crystalline materials can be calculated with precise ab initio computational methods, since the use of symmetry can significantly reduce the required computational effort, and anharmonic effects play a limited role. In the case of amorphous materials, the absence of crystal symmetry and the presence of large anharmonicities, preclude the use of high precision ab initio methods. In this case, molecular dynamics simulations become one of the best available options since it accounts for all levels of anharmonicities intrinsicaly. However, in order to simulate large amorphous samples, molecular dynamics requires efficient interatomic potentials, which often suffer from low accuracy and poor transferability when compared to ab initio methods. In this work we employ a recently developed machine-learned neural network potential [1] to calculate the lattice thermal conductivity of amorphous $Ge_2Sb_2Te_5$ via non-equilibrium molecular dynamics simulations at different temperatures. We also employ the Allen and Feldman theory [2] to compute the lattice thermal conductivity of $Ge_2Sb_2Te_5$ in the harmonic limit, which relies on force constants computed with the neural network potential, and determine the effect of anharmonicities by directly comparing the Allen-Feldman conductivity to the molecular dynamics results. Finally, we employ the neural network potential to compute the lattice thermal conductivity of $Ge_2Sb_2Te_5$ within the recently developed Wigner formulation of thermal transport in solids [3], which encompasses the emergence and coexistence of the particle-like phonon wave packet propagation discussed by Peierls for crystals, and the wave-like interband conduction mechanisms discussed by Allen and Feldman for harmonic glasses. The Wigner formulation allows one to describe the thermal conductivity of crystalline systems, where it is equivalent to the linearized Boltzmann transport equation, and of disordered glasses, where it generalizes Allen-Feldman theory accounting for anharmonicity. More importantly, the Wigner formulation also accurately describes the thermal conductivity of materials with intermediate characteristics between crystals and amorphous glasses, where both particle-like and wave-like heat conduction mechanisms are relevant. Our complete framework, employing molecular dynamics simulations, Allen-Feldman theory and the Wigner formulation, all based on interatomic forces described by a novel machine learned deep neural network potential, provides a complete picture of heat conduction in amorphous $Ge_2Sb_2Te_5$ which is fundamental to its application in novel electronic devices.

References

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