Exploring Lattice Thermal Transport in Methane Hydrates: Insights from Deep Neural Network-Enhanced Interatomic Potentials

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Molecular dynamics (MD) serves as a valuable tool for unraveling the atomistic intricacies governing thermal transport in methane hydrates, particularly in light of the challenges posed by experimental investigations. Despite its utility, conventional empirical interatomic potentials employed in MD simulations are constrained by limitations in precision and the comprehensive representation of material physics and chemistry. Addressing this gap, the advent of machine-learning-based algorithms has proven pivotal in crafting accurate potentials for MD simulations. This study takes a step further by leveraging MD simulations based on deep neural network (DNN) interatomic potentials, trained through ab initio molecular dynamics (AIMD) simulations. Our investigation utilizing the developed DNN potential focuses on unraveling the impact of temperature, pressure, and the filling ratio of guest methane molecules on thermal conductivity. This exploration brings to light the pivotal roles played by anharmonic phonon scattering, guest molecule-clathrate scattering, and structural stability in shaping the thermal transport properties of clathrate hydrates. Additionally, we extract phonon dispersion relations and lifetimes from atomic velocities derived from our DNN-potential based MD simulations. Employing spectral energy density (SED) analysis, we rigorously examine the influence of temperature and filling ratio on phonon lifetime and the overall bandstructure. This comprehensive approach sheds new light on the intricate interplay of factors governing thermal transport in methane hydrates, offering valuable insights for both fundamental understanding and practical applications.