Machine Learing Accelerated Explorion of Strongly Anharmonic Thermal Insulators

Exploring Strongly Anharmonic Thermal Insulators with Machine-Learned Interatomic Potential from Active Learning Scheme

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In Short

- Active learning workflow
- Efficient Green-Kubo method from MLIPs
- Exploration of strongly anharmonic thermal insulators

Thermal insulating semiconductors have numerous potential applications, such as thermoelectric generators based on the Seebeck effect, thermoelectric cooling via the Peltier effect, and thermal insulation coatings for thermal banks or thermal barriers. The reliable prediction and atomistic understanding of heat transport in thermal insulators, however, remain challenging. Thermal insulating semiconductors often exhibit stronge anharmonicity, particularly associated with rare events such as defect creation and phase transition precursors [1].

Strongly anharmonic materials exhibit phenomena arising from significant deviations of the potential energy surface (PES) from the harmonic approximation near equilibrium. These deviations are especially pronounced at high temperatures or when multiple local minima exist besides the equilibrium structure. Such conditions invalidate the conventional phonon picture, which relies on harmonic assumptions around equilibrium, rendering perturbative methods, such as the Peierls-Boltzmann transport equation (PBTE) and the Wigner transport method, ineffective for describing heat transport. Non-perturbative methods, such as the Green-Kubo formalism based on molecular dynamics (MD) simulations, allow thermal conductivity calculations without relying on the quasiparticle approximation and are ideal for strongly anharmonic systems. However, these methods are computationally challenging due to the required large systems and long simulation times needed to capture rare events and long-wavelength vibrations.

Advancements in machine-learned interatomic potentials (MLIPs) offer a promising solution by providing near first-principles accuracy at significantly reduced computational expense. However, challenges remain in fully capturing rare events and ensuring MLIP reliability across the potential energy surface because infrequent phenomena are often missed in training data.

In this proposal, we present a integrated framework combining the Green-Kubo formalism with MLIPs based on equivariant neural network, enhanced by a sequential active learning (AL) scheme [2]. The AL method will be applied for reliable MLIP training of the strongly anharmonic materials by exploring unfamiliar configurations during molecular dynamics simulations. Moreover, the implementation of efficient heat flux calculations within MLIPs will enable reliable thermal conductivity results from the Green-Kubo formalism.

Using the integrated framework, we aim to calculate the thermal conductivity of 15 materials that were previously predicted to have ultra-low thermal conductivity by a symbolic regression model [3]. Our demonstrations and results will not only provide reliable thermal conductivity predictions for strongly anharmonic systems but also shed light on the atomistic understanding of the thermal insulating behavior of these semiconductors, which will accelerate exploration and motivate the design of novel thermal insulation materials.

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https://nomad.fhi.mpg.de

More Information

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[3] Purcell, T.A.R., Scheffler, M., Ghiringhelli, L.M. et al. *npj Comput. Mater.* **9**, 112 (2023). doi: 10.1038/s41524-023-01063-y

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