Ab-initio Thermal Transport Calculations for Crystals and Nanostructures Based on the Boltzmann Transport Equation

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Motivation

Technological progress requires increasingly fine-tuned heat management strategies, be it to keep the operating temperatures of device parts optimized for long lifetimes or to extract higher performance from energy-scavenging cycles. This kind of context imposes new demands on computational materials science, which must be able to deliver predictive thermal transport calculations for new compounds, materials and devices, as opposed to just shedding light on existing solutions.

A predictive atomistic approach

In the last two decades, solutions to the Boltzmann transport equation (BTE) for phonons formulated based on inputs from density functional theory (DFT) have enabled the development of predictive thermal transport calculation workflows in an increasingly diverse catalog of situations dominated by lattice excitations. The first part of this presentation will consist in a guided tour of those developments viewed from a methodological angle but illustrated with relevant examples at each step. The starting point will be the solution for a perfect single crystal in the steady state [1], which serves to introduce most of the important quantities involved. I will then present the extension of this method to more realistic systems containing defects [2], which also affords excellent agreement with experiment, and show solutions for nanostructures, interfaces, models of electronic devices, time-dependent problems [3] and high-temperature phases [4].

The machine learning (ML) revolution

This increase in model complexity can pose formidable computational challenges, which are compounded by the need to scale horizontally as well, i.e., to scan collections of materials or ranges of parameter values while optimizing for a target application. I will illustrate how ML techniques can be of crucial help when exploring chemical and configurational space by detecting and exploiting regularities between systems with similar structures or compositions. ML force fields, regression models for the potential energy and the forces trained on DFT calculations, can decrease the computational cost of the ingredients required for a solution to the phonon BTE by orders of magnitude. One of the keys to enabling this is algorithmic differentiation (AD), a feature of the latest generation of ML frameworks. I will explain how AD opens the door to completely new kinds of workflows [5] of special relevance for thermal transport calculations.

Beyond phonons

To conclude, I will discuss some fundamental limitations of BTE-based approaches when dealing with higher-order anharmonicity and more disordered structures, generalizations [6] and alternatives [7] to treat those systems, and the role that ML techniques can play in making those viable.

References

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