MAR17-2016-000507

Abstract for an Invited Paper
for the MAR17 Meeting of
the American Physical Society

Car and Parrinello meet Green and Kubo: simulating atomic heat transport from equilibrium ab initio molecular dynamics[1]

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Modern simulation methods based on electronic-structure theory have long been deemed unfit to compute heat transport coefficients within the Green-Kubo formalism. This is so because the quantum-mechanical energy density from which the heat flux is derived is inherently ill defined, thus allegedly hampering the use of the Green-Kubo formula. While this objection would actually apply to classical systems as well, I will demonstrate that the thermal conductivity is indeed independent of the specific microscopic expression for the energy density and current from which it is derived. This fact results from a kind of *gauge invariance* stemming from energy conservation and extensivity, which I will illustrate numerically for a classical Lennard-Jones fluid. I will then introduce an expression for the adiabatic energy flux, derived within density-functional theory, that allows simulating atomic heat transport using equilibrium *ab initio* molecular dynamics. The resulting methodology is demonstrated by comparing results from ab-initio and classical molecular-dynamics simulations of a model liquid-Argon system, for which accurate inter-atomic potentials are derived by the force-matching method, and applied to compute the thermal conductivity of heavy water at ambient conditions. The problem of evaluating transport coefficients along with their accuracy from relatively short trajectories is finally addressed and discussed with a few representative examples.

[1]Partially funded by the European Union through the MaX Centre of Excellence (Grant No. 676598)