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Accurate Thermal Conductivities from First Principles

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In spite of significant research efforts, a first-principles determination of the thermal conductivity at high temperatures has remained elusive. On the one hand, Boltzmann transport techniques[1] that include anharmonic effects in the nuclear dynamics only perturbatively become inaccurate or inapplicable under such conditions. On the other hand, non-equilibrium molecular dynamics (MD) methods suffer from enormous finite-size artifacts in the computationally feasible supercells, which prevent an accurate extrapolation to the bulk limit of the thermal conductivity [2]. In this work, we overcome this limitation by performing *ab initio* MD simulations in thermodynamic equilibrium that account for all orders of anharmonicity. The thermal conductivity is then assessed from the auto-correlation function of the heat flux using the Green-Kubo formalism[3]. Foremost, we discuss the fundamental theory underlying a first-principles definition of the heat flux using the virial theorem. We validate our approach and in particular the techniques developed to overcome finite time and size effects, e.g., by inspecting silicon, the thermal conductivity of which is particularly challenging to converge[4]. Furthermore, we use this framework to investigate the thermal conductivity of ZrO_2, which is known for its high degree of anharmonicity. Our calculations shed light on the heat resistance mechanism active in this material, which eventually allows us to discuss how the thermal conductivity can be controlled by doping and co-doping[5].

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