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Thermal Conductivities at High Temperatures from First Principles

In spite of significant research efforts, a parameter-free first-principles determination of the thermal conductivity at high temperatures, e.g, above 70 of the melting temperature, has remained elusive. Techniques that rely on the harmonic approximation or on small perturbations thereof [1] are questionable in this temperature regime. Similarly, methods based on non-equilibrium molecular dynamics (MD) require enormous temperature gradients along the computationally feasible supercell, and hence lead to undesired non-linear effects [2]. The Green-Kubo method, which does not suffer from these shortcomings, involves the assessment of the thermal conductivity from the auto-correlation of the heat flux via equilibrium MD. This method has hitherto been discarded in first-principles simulations both due to its numerical cost and due to conceptual difficulties in the definition of the heat flux, the computation of which requires a partitioning of the interactions onto the individual atoms.

In this work, we show that the non-convective contribution to the heat flux can be assessed via the analytical stress tensor, a quantity that is directly accessible in density functional theory calculations and that we recently implemented in FHI- aims [4]. By means of the virial theorem, this approach leads to a unique definition of the microscopic heat flux in solids that does not rely on any partitioning scheme. To demonstrate the general applicability of this technique, we use the Green-Kubo formalism to investigate the heat transport of zirconia and silicon, two materials that feature particularly low / high thermal conductivities. We critically discuss the details of our implementation and present strategies to reduce the numerical effort associated with such ab initio MD simulations.

References:

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