A First Principles Approach to Thermal Transport in Nanomaterials

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A predictive theory for evaluating thermal conductivity is essential for the design of new materials for efficient thermoelectric refrigeration and power generation and it could help in understanding heat dissipation in micro- and nano-electronics devices.

Model

A flux of heat established by a Gradient of Temperature $\nabla T$ can be written in terms of phonon energies $h\omega_{qj}$ phonon group velocities $c^\alpha_{qj}$ and perturbed phonon population $n_{qj}$ with vector $q$ and branch index $j$:

$$\frac{1}{N_0 \Omega} \sum_{qj} h\omega_{qj} c^\alpha_{qj} n_{qj} = -k_{\alpha\beta} |\nabla T|_\beta$$

Knowledge of perturbed phonon population allows the evaluation of thermal conductivity.

The perturbation due to the gradient of temperature is balanced by the scattering processes:

$$-c_{qj} |\nabla T| \left( \frac{\partial n_{qj}}{\partial T} \right) + \frac{\partial n_{qj}}{\partial T_{\text{scatt}}} = 0$$

Boltzmann Transport Equation

All the fundamental ingredients are obtained from the harmonic and anharmonic interatomic force constants.

In this work we evaluated them fully ab initio by using a new implementation$^3$ of the 2n+1 theorem in Quantum Espresso$^4$

Main Goal

Accurate prediction of thermal conductivity in nanostructured materials by using Density Functional Perturbation Theory and solving the Boltzmann Transport Equation.

RESULTS

BTE solutions

- Approximated solution: Single Mode Approximation (SMA)
- Exact (iterative) solutions: Omini-Sparavigna$^5$
  - Fugallo et al.

Lattice thermal conductivity for pure, isotopically enriched [0.07-0.1]$^{13}$C and naturally occurring 1.1$^{13}$C$^{12}$ diamond. Comparison with Experimental results.$^6$.

Diamond lattice thermal conductivity at 100K and 300K as a function of different C$^{13}$ percentages.

3. L. Paulatto et al., to be published.
6. G. Fugallo et al., to be published.