A First Principles Approach to Thermal Transport in Nanomaterials

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PROJECT: ACCATTONE

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[1].

Model

A flux of heat established by a Gradient of Temperature ∇T can be written[2] in terms of phonon energies $\hbar\omega_{\mathbf{q}j}$ phonon group velocities $c^{\alpha}_{\mathbf{q}j}$ and perturbed phonon population $n_{\mathbf{q}j}$ with vector q and branch index j

$$\frac{1}{N_0\Omega}\sum_{\mathbf{q}j}\hbar\omega_{\mathbf{q}j}c^{\alpha}_{\mathbf{q}j}n_{\mathbf{q}j} = -k_{\alpha\beta}|\nabla T|_{\beta}$$

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[2]

$$-\mathbf{c}_{\mathbf{q}j}|\nabla T|\left(\frac{\partial n_{\mathbf{q}s}}{\partial T}\right) + \left.\frac{\partial n_{\mathbf{q}j}}{\partial t}\right|_{scatt} = 0$$

Boltzmann Transport Equation

All the fundamental ingredients are obtained from the harmonic and anharmonic intermatomic 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]**

Linearized BTE

$$n_{\mathbf{q}j} \simeq \bar{n}_{\mathbf{q}j} + \bar{n}_{\mathbf{q}j}(\bar{n}_{\mathbf{q}j}+1)\Psi_{\mathbf{q}j}$$

P : scattering rates

$$-\mathbf{c}_{\mathbf{q}j}\cdot\nabla T\left(\frac{\partial\bar{n}_{\mathbf{q}j}}{\partial T}\right) = \sum_{\mathbf{q}'j',j''}\left[\tilde{P}^{\mathbf{q}''j''}_{\mathbf{q}j,\mathbf{q}'j'}(\Psi^{j}_{\mathbf{q}}+\Psi^{j'}_{\mathbf{q}'}-\Psi^{j'}_{\mathbf{q}'}+\frac{1}{2}\tilde{P}^{\mathbf{q}'j',\mathbf{q}''j''}_{\mathbf{q}j}(\Psi^{j}_{\mathbf{q}}-\Psi^{j'}_{\mathbf{q}'}-\Psi^{j''}_{\mathbf{q}''})\right] + \sum_{\mathbf{q}'j'}P^{imp}_{\mathbf{q}j,\mathbf{q}'j'}(\Psi^{j}_{\mathbf{q}}-\Psi^{j'}_{\mathbf{q}'}) + P^{be}_{\mathbf{q}j}\Psi^{j}_{\mathbf{q}}$$

Anharmonic –three phonon scattering : **Intrinsic resistivity** | Isotopic impurities | Border Effects

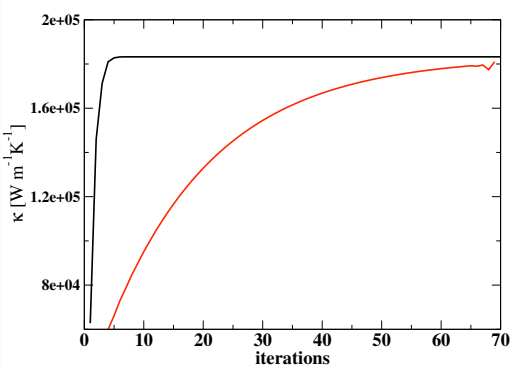
Main Goal

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

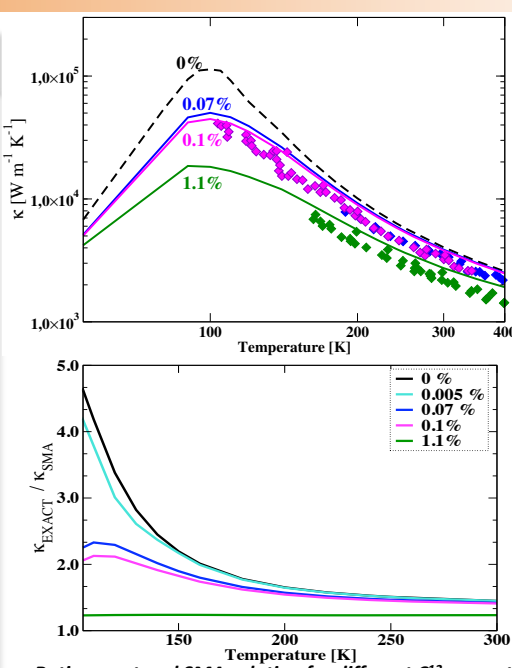
RESULTS

BTE solutions

- **Approximated solution**: Single Mode Approximation (SMA)
- **Exact (iterative) solutions:** Omini- Sparavigna[5] *Fugallo et al*[6]

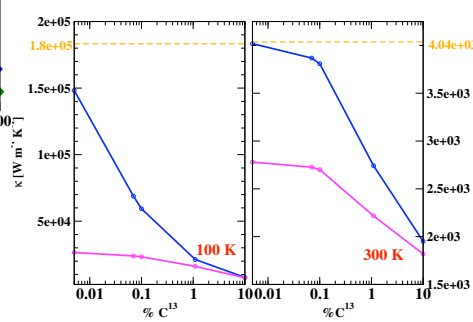


Diamond lattice thermal conductivity at 100 K



Ratio: exact and SMA solution for different C^{13} percentages

Lattice thermal conductivity for pure, isotopically enriched [0.07-0.1] C^{13} and naturally occurring 1.1 C^{13} diamond. Comparison with Experimental results[7,8]



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

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