Collective Behavior in Phonon Thermal Transport on Semiconductor Devices

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Abstract - The Kinetic-Collective model is presented to calculate the thermal conductivity of several semiconductor materials. This model is an alternative method to Callaway-like models to account for the role of normal collisions in the thermal transport. Its more rigorous treatment of the collision term in the Boltzmann Transport Equation provides a more accurate prediction of thermal conductivity. Within this model, the thermal conductivity is explained as a combination of two phonon behaviors leading to a kinetic and a collective phonon flux with significantly different expressions. The main difference between these regimes of behavior is that, in the kinetic one, relaxation-time depends on the modes independently while in the collective term relaxation-time is the same for all phonons. From this approach, very accurate thermal conductivity predictions for bulk and nanostructured samples of group IV semiconductors are obtained.

Keywords: Phonons, Heat transport, Thermal conductivity, Kinetic regime, Collective regime.

1. Introduction

Unveiling the physics underlying phononic transport in semiconductors is crucial to tailor thermoelectric devices, and it represents a central topic in current nanotechnology research, where the control of the thermal conductivity is relevant in problems related to thermoelectrical energy conversion or nanorefrigeration. Within this framework, a general and simple model able to predict the phonon contribution to thermal conductivity of different materials and device sizes in a wide temperature interval would simplify the numerical complexity needed to predict a device thermal conductivity in the first steps of their design and could be technologically helpful for a fast exploration of a wide variety of nanosystems, allowing to identify the most promising designs.

The usual starting point for thermal conductivity modeling is the Boltzmann Transport Equation (BTE), but deriving a general predictive solution is a tremendous task that at the moment has not been completely achieved. Here, we present a model that from a novel mathematical treatment of phonon interactions leads to establish two different regimes in phonon thermal transport. The role of normal scattering is crucial to understand the phonon behavior in each regime. Finally, our model is tested on several bulk semiconductors providing very good agreement with experimental data.

2. Kinetic-collective Model

According to DeTomas et al. (2014a), in the kinetic-collective model the trade-off between non-resistive and resistive scattering events will determine the behavior of the phonon collectivity. This leads to establish two limiting behaviors: kinetic and collective regime.

The kinetic regime is established when resistive scattering dominates over non-resistive scattering. In this regime each phonon contributes independently to the heat flux, this is, each phonon participating in thermal transport collides individually, without affecting the remaining phonons. Then, the thermal conductivity in this regime κ_{kin} is obtained as the usual kinetic equation

$$\kappa_{\mathrm{kin}} = \frac{1}{3} \int \hbar\omega\tau_{R,\omega} v_\omega^2 \frac{\partial f_\omega^0}{\partial T} D_\omega d\omega \tag{1}$$

where ω is the phonon frequency, f_ω^0 the equilibrium phonon distribution given by the Bose-Einstein distribution function, T temperature, D_ω density of states, v_ω group velocity, and $\tau_{R,\omega}$ is the total resistive relaxation time accounting for all the resistive scattering events obtained according to the Matthiessen rule. Note that here, and onward, although the phonon branch index is omitted to simplify notation, the sum over all phonon branches must be performed in the integrals.

The collective regime is established when non-resistive or normal scattering dominates over resistive scattering. In this regime phonons behave as a collectivity, since the main role of normal scattering is redistributing the conserved momentum among all the phononic modes. Then, any collision (resistive or not) suffered by a phonon of the collectivity affects the remaining phonons. This way, the thermal conductivity in this regime κ_{coll} is

$$\kappa_{\mathrm{coll}} = \frac{1}{3} \frac{\left(\int v_\omega q_\omega \frac{\partial f_\omega^0}{\partial T} D_\omega d\omega \right)^2}{\int \frac{q_\omega^2}{\hbar\omega} \frac{1}{\tau_{R,\omega}} \frac{\partial f_\omega^0}{\partial T} D_\omega d\omega}. \tag{2}$$

where q is the phonon wavevector.

Finally, the total phonon thermal conductivity κ accounts for the transition from one limiting regime to the other, going through all possible intermediate regimes, and it is given by

$$\kappa = \kappa_{\mathrm{kin}}(1 - \Sigma) + \kappa_{\mathrm{coll}} F(L_{\mathrm{eff}})\Sigma. \tag{3}$$

where

$$\Sigma = [1 + \langle\tau_N\rangle/\langle\tau_R\rangle]^{-1} \tag{4}$$

is a switching factor weighting the relative importance between resistive and normal scattering. Σ takes values in the range $[0, 1]$ depending on the averages of normal $\langle\tau_N\rangle$ and resistive $\langle\tau_R\rangle$ relaxation times. The averaged relaxation times are calculated as

$$\langle\tau_i\rangle = \frac{\int \hbar\omega\tau_{i,\omega} \frac{\partial f_\omega^0}{\partial T} D_\omega d\omega}{\int \hbar\omega \frac{\partial f_\omega^0}{\partial T} D_\omega d\omega}, \tag{5}$$

with the subindex i indicating resistive R or normal N relaxation time. Note that the denominator in Eq.(5) is in fact the total heat capacity. If resistive processes dominate over normal scattering $\langle\tau_N\rangle \gg \langle\tau_R\rangle$, then $\Sigma \to 0$ and $\kappa \to \kappa_{\mathrm{kin}}$. Otherwise, normal scattering dominates and $\langle\tau_R\rangle \gg \langle\tau_N\rangle$, then $\Sigma \to 1$ and $\kappa \to \kappa_{\mathrm{coll}}$. Note that in spite of the effect of the normal scattering, thermal conductivities κ_{kin} and κ_{coll} include only resistive collisions through τ_R and normal scattering is included only in the switching factor Σ. Then, the main difference between κ_{kin} and κ_{coll} does not lie in the used relaxation times but in the way they are

averaged in each term. In Eq. (3) it also appears the form factor $F(L_{\mathrm{eff}})$ depending on the effective size of the sample L_{eff}. This factor accounts for the role of boundary scattering on the collective flux, it is obtained under hydrodynamics considerations (Alvarez et al. 2007) and it is valid for any geometry, entering the appropriate L_{eff} (Zhang 2007).

3. Results and Discussions

As shown by De Tomas et al. (2014a), the kinetic-collective model leads to excellent results for silicon thermal conductivity in a very wide range of temperatures [0-1000K] and effective sizes (from bulk sizes down to $\propto$ 30nm), the shown samples also present different geometries, like thin-films and nanowires. Here, we have tested the model on other well-known semiconductor materials: germanium and diamond. To perform the thermal conductivity calculations one have to consider two things: first, full dispersion relations should be used entering Eq. (3) to avoid any error induced from a dispersionless model, for this purpose we have calculate the dispersion relations of the materials considered here with the bond-charge model (Weber 1974), and second, appropriate expressions for the phonon relaxation times should be used. For all the materials considered here, we have taken into account 4 main scattering mechanisms: boundary scattering τ_B, mass-defect scattering τ_I, umklapp scattering τ_U and normal scattering τ_N. Their functional forms used here are given in De Tomas et al. 2014. Although τ_B and τ_I do not contain free parameters, τ_U and τ_N include three adjustable parameters in total. However, they can also be calculated from theoretical expressions (Morelli 2002, De Tomas et al. 2014b).

In Fig. 1 we show the thermal conductivity calculated within the kinetic-collective model from Eq. (3) for naturally occurring bulk samples of silicon, germanium and diamond. Our predictions are in very good agreement with experimental data (Onn 1992, Inyushkin 2004, Asen-Palmer 1997). In the plot, it can be observed that in the temperature range where boundary scattering is dominating the transport, the total thermal conductivity is purely kinetic. As the temperature rises, the other scattering mechanisms begin to participate in the thermal transport. Therefore, the total thermal conductivity begins to separate from the pure kinetic regime, tending to a collective regime as normal scattering increases its dominating role in the transport. Note that these materials seem to have proportional trends in the curves. Since all of them have in common the same diamond-like crystal structure, this may be inducing similar behavior in phonon-phonon interactions.

4. Conclusion

We have presented a novel way of understanding phonon transport by considering two different regimes: one where phonons moves independently (kinetic regime) and another where phonons moves as a collectivity (collective regime). The trade-off between these extremed limits yields the net propagating phonon flux. Within this model, we have obtained the phonon thermal conductivity of several semiconductors: silicon, germanium and diamond, in very good agreement with experimental data. These materials have a wide variety of applications in nanotechnology and our model has revealed as a useful tool to characterize their thermal conductivity in a very simple and general way. This model may be extended to similar semiconductor materials with promising results.
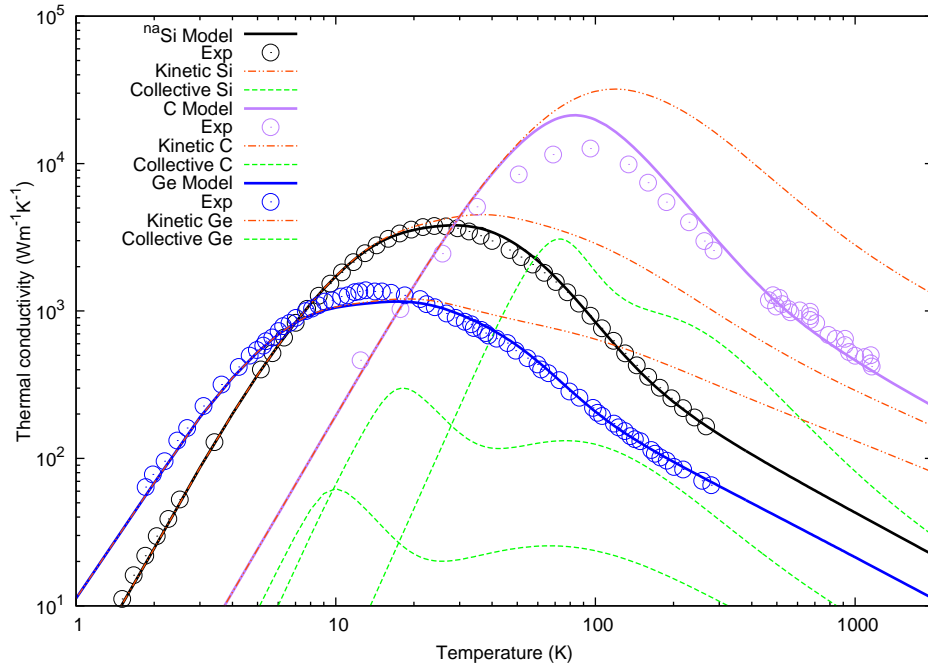
Fig. 1. Phonon thermal conductivity of naturally occurring silicon, germanium and diamond calculated with the kinetic-collective model (solid lines) in comparison with experimental data (Onn 1992, Inyushkin 2004, Asen-Palmer 1997) (symbols). For each material we show in dashed-dot red lines the kinetic regime and in dotted green lines the collective regime. Note that highest lines correspond to diamond, medium lines to silicon and lowest lines to germanium.

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