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Advanced Workshop on Energy Transport in Low-Dimensional Systems: Achievements and Mysteries

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Thermal and Electrical Transport and Thermoelectric Figure-of-Merit in Low-Dimensional Nanostructures

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Model for the thermal transport in 3D, 2D and 1D materials

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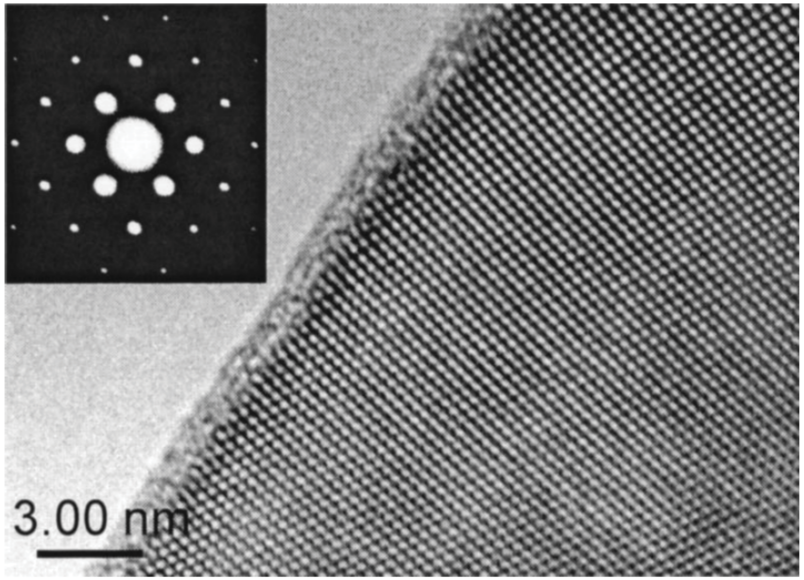
Outlook

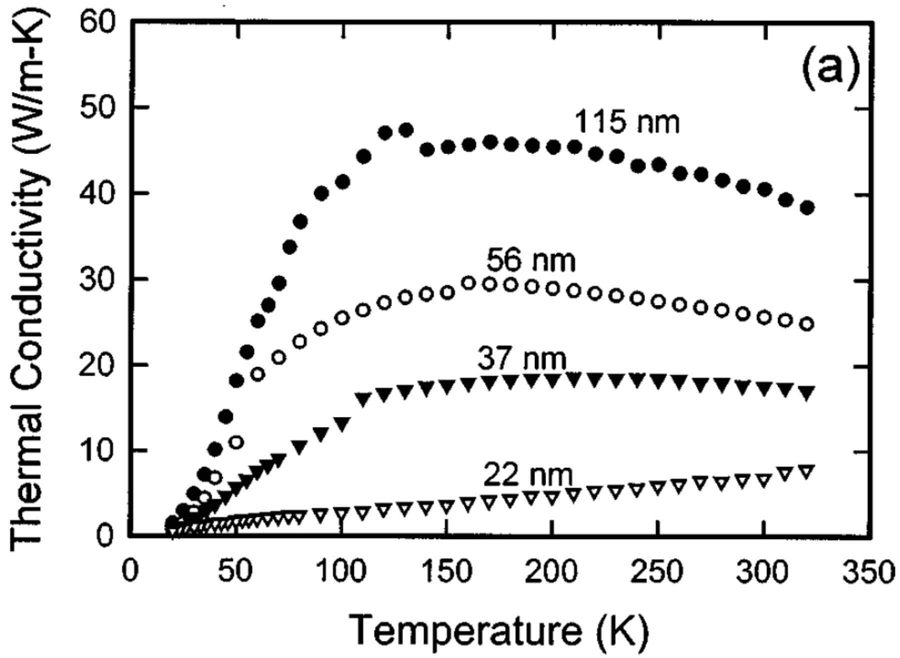
- Motivation
- Starting point
- Boltzmann equation
- Callaway model
- Gruyel-Krumshansl model
- Modified model GKM
- Results
- Conclusions

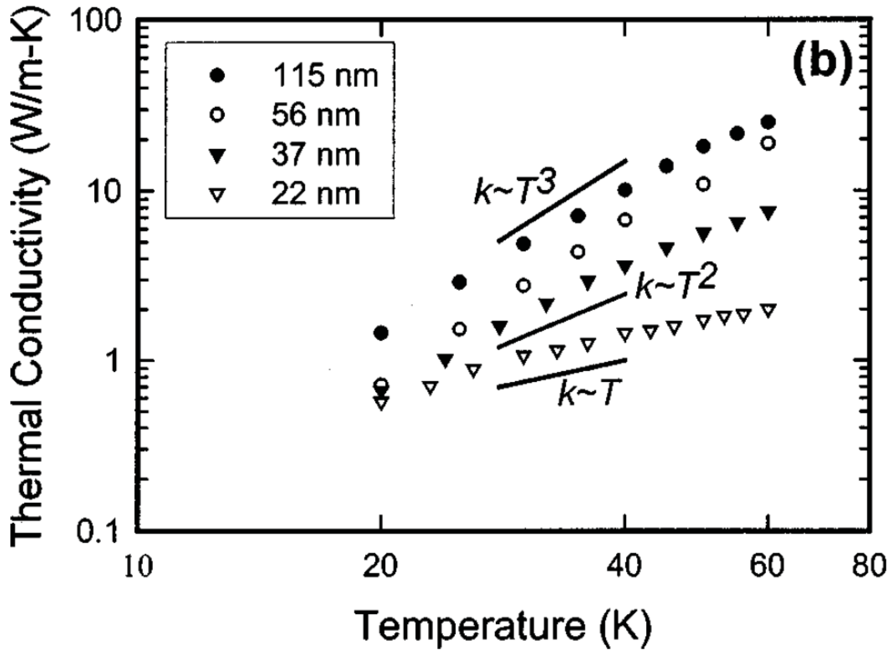
Motivation

- The thermal conductivity of silicon nanowires found in several experimental works
- Unclear explanation proposed by several authors based on the BTE and several expressions for the different relaxation times involved
- Several authors have tried to explain the thermal conductivity using classical molecular dynamics, even for the thinnest wires and low temperature
- In some of the theoretical papers found in the literature, N-processes are not considered at all



High-resolution TEM image of a 22 nm single crystal Si NW.





(a) Measured thermal conductivity of different diameter Si nanowires. The number beside each curve denotes the corresponding wire diameter.

(b) Low temperature experimental data on a log scale.

D. Li et al, APL 83, 2934 (2003)

Calculation of the contribution of the surface/boundary to the thermal conductivity

$$\Delta\kappa_l^{\text{wire}} = \frac{24}{\pi}\left(\frac{k_B}{\hbar}\right)^3 \frac{k_B}{2\pi^2 V} T^3 \int_0^{\theta_D/T} \frac{\tau_C x^4 e^x}{(e^x-1)^2} G(\eta(x),p)\,dx$$

The relaxation time has several contributions:

$$\frac{1}{\tau_C} = \frac{1}{\tau_U} + \frac{1}{\tau_M} + \frac{1}{\tau_B} + \frac{1}{\tau_{\text{ph-}e}}$$

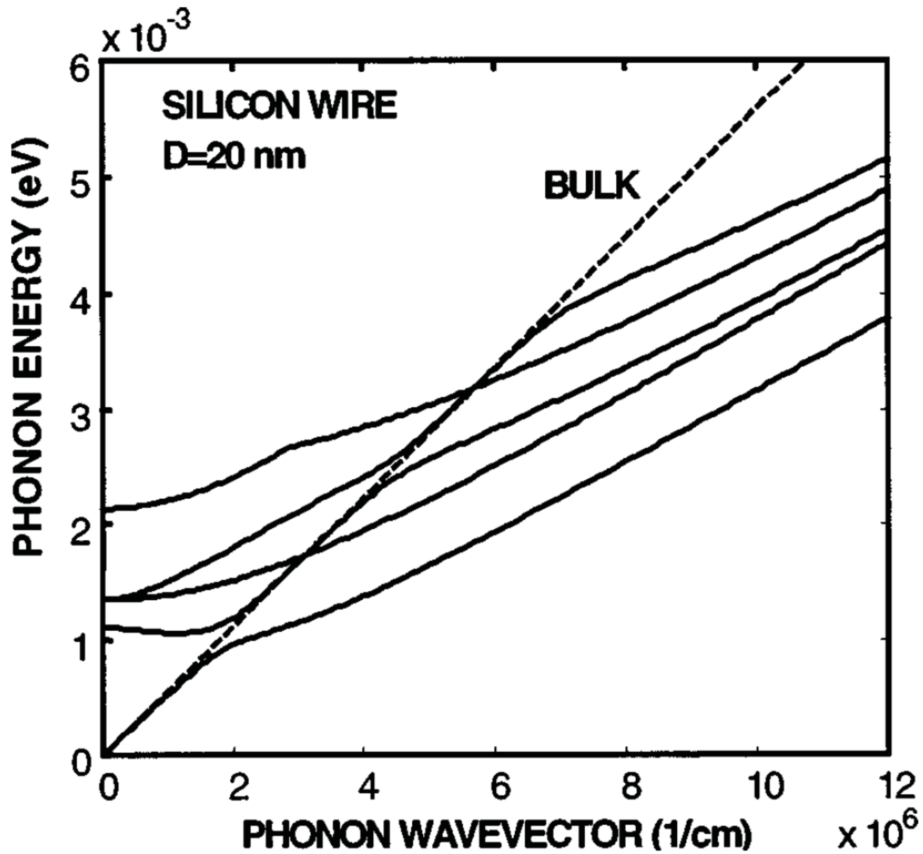
For U-processes, $$\frac{1}{\tau_U} = 2\gamma^2 \frac{k_B T}{\mu V_0} \frac{\omega^2}{\omega_D}$$

For mass-defect scattering $$\frac{1}{\tau_M} = \frac{V_0 \Gamma \omega^4}{4\pi V^3}$$

For boundary scattering $$\frac{1}{\tau_B} = \frac{V}{D}(1-p)$$

Phonon-electron interaction

$$\frac{1}{\tau_{\text{ph-}e}} = \frac{n_e \epsilon_1^2 \omega}{\rho V^2 k_B T} \sqrt{\frac{\pi m^* V^2}{2 k_B T}} \exp\left(-\frac{m^* V^2}{2 k_B T}\right)$$



J. Zou & A. Balandin, JAP89, 2932 (2001).

VII. DECAY OF OPTICAL PHONONS INTO ACOUSTIC PHONONS

M. Kazan et al add a term to account for the decay of optical phonons in acoustic phonons, even at 0K!

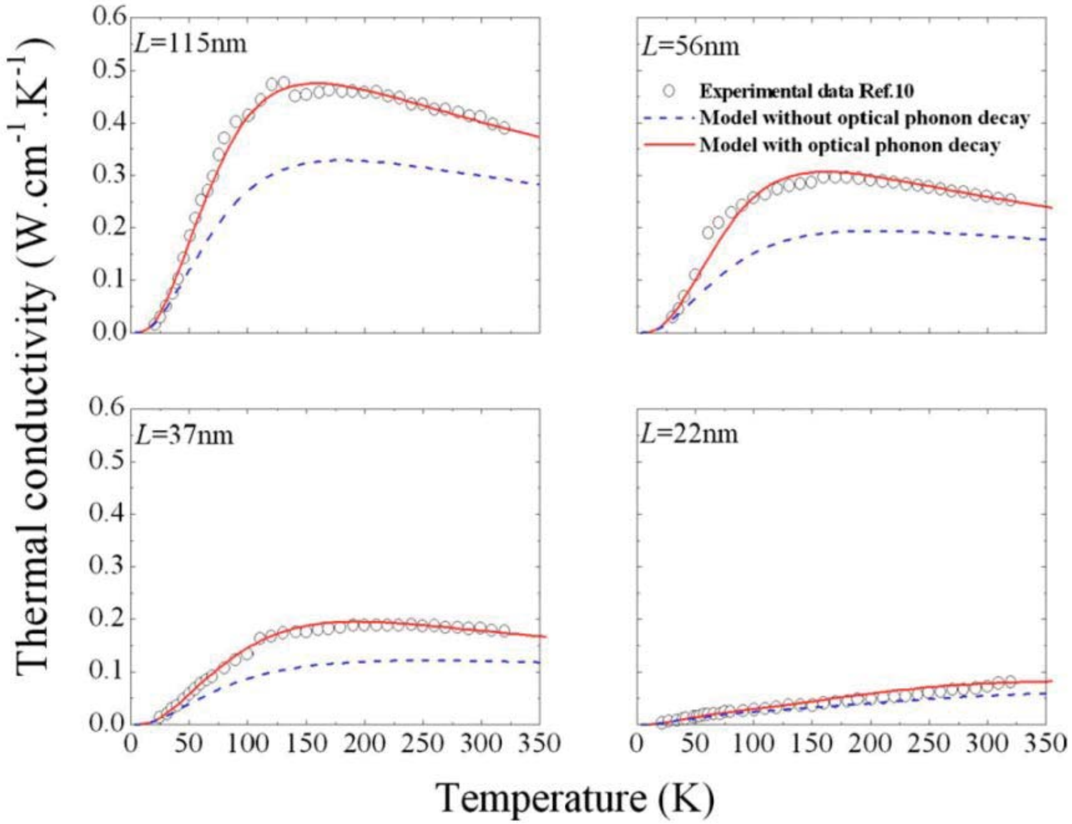
$$\tau^{-1} = -\frac{1}{\tilde{N}_{OP}} \frac{d}{dt}(\tilde{N}_{OP})$$

They subtract the decay involving three or four phonon processes to the remaining resistive processes:

$$\tau_{R,j}^{-1} = (\tau_{B,j}^{-1} + \tau_{I,j}^{-1} - \tau_{G(3),j}^{-1} - \tau_{G(4),j}^{-1}) + \tau_{U,j}^{-1}$$

and gives the following expression for the N-processes

$$\tau_{N,L}^{-1} = B_{N,L}\omega^2 T^3 \qquad \tau_{N,T}^{-1} = B_{N,T}\omega T^4$$



Dashed line: without phonon decay. Solid line: including phonon decay.

- M. Kazan et al, JAP107, 083503 (2010)

Starting point

- When the diameter of the NWs is reduced, the boundary or surface effects (classical) are more important
- Below some diameter, quantum effects may play a role
- In the 20 to 100 nm range, it is still not clear if quantum effects could be important
- Several works including physical engineering have been proposed in the literature

What we propose:

- We plan to analyze here the range where classical effects are enough to explain the thermal conductivity
- Our approach will be based on the BTE to account for the phonon transport
- We use a lattice dynamics model to obtain the bulk dispersion relations (bond charge model)
- The calculation of the thermal conductivity will be done using the Gruyer-Krumhansl model (GKM) instead of the previously used, the Callaway-Holland model (CM).

Callaway model

Callaway starts from the Boltzmann equation and takes for the collision integral

$$\left(\frac{\partial N}{\partial t}\right)_c = \frac{N(\lambda)-N}{\tau_N} + \frac{N_0 - N}{\tau_u}$$

and writes the classical expression for the thermal conductivity

$$\kappa = \frac{3}{(2\pi)^3}\int c^2 \cos^2\theta\, \alpha(k) C_{\mathrm{ph}}(k) d^3k$$

He called U-processes to all resistive processes, including impurity or boundary

$$\tau_u^{-1} = A\omega^4 + B_1 T^3 \omega^2 + c/L$$

and gives the following expression for the N-processes

$$\tau_N^{-1} = B_2 T^3 \omega^2$$

The expression for the thermal conductivity is:

$$\kappa = \frac{c^2}{2\pi^2}\int \tau_c \left(1 + \frac{\beta}{\tau_N}\right) C_{\mathrm{ph}} k^2 dk$$

Gruyer-Krumhansl model

In the GKM the BTE is written as:

$$\left[\frac{\partial}{\partial t} + \boldsymbol{v} \cdot \boldsymbol{\nabla} - \boldsymbol{C}\right] f(\boldsymbol{r}, \boldsymbol{q}, t) = 0$$

By introducing the drift operator,

$$\boldsymbol{D} = \frac{\partial}{\partial t} + \boldsymbol{v} \cdot \boldsymbol{\nabla}$$

the BTE can be written in the following form:

$$\boldsymbol{D} f(\boldsymbol{r}, \boldsymbol{q}, t) = (\boldsymbol{R} + \boldsymbol{N})\, f(\boldsymbol{r}, \boldsymbol{q}, t)$$

where the collision operator has been divided into two components, one related to the resistive processes and a second component related to N-processes

In order to symmetrized the collision operator, in the GKM the following basis is used:

$$f^*(\boldsymbol{r}, \boldsymbol{q}, t) = f(\boldsymbol{r}, \boldsymbol{q}, t) 2 \sinh \frac{\hbar \omega_{\boldsymbol{q}}}{2 k_B T}$$

- R. A. Gruyer and J. A. Krumhansl, PR148, 766 (1966).

The final form of the linearized BTE is a matrix, given by

$$\left\{ \begin{pmatrix} D_{00} & D_{01} & 0 \\ D_{10} & D_{11} & D_{12} \\ 0 & D_{21} & D_{22} \end{pmatrix} - \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & N_{22} \end{pmatrix} - \begin{pmatrix} 0 & 0 & 0 \\ 0 & R_{11} & R_{12} \\ 0 & R_{21} & R_{22} \end{pmatrix} \begin{pmatrix} a_0 \\ \boldsymbol{a}_1 \\ \boldsymbol{a}_2 \end{pmatrix} \right\} = 0$$

and the thermal conductivity can be written as

$$\kappa = \frac{1}{3} C_v v^2 \left[\langle \tau_{RB} \rangle (1 - \Sigma) + \langle \tau_R^{-1} \rangle^{-1} G(R) \Sigma \right]$$

Where the resistive contribution to the relaxation time is

$$\tau_{RB}^{-1} = \tau_I^{-1} + \tau_U^{-1} + \tau_B^{-1}$$

and Σ, called Ziman factor, is given by $\Sigma = \dfrac{1}{1 + \langle \tau_N \rangle / \langle \tau_{RB} \rangle}$

with the usual definition of $\langle \tau \rangle = \dfrac{2}{3kT} \dfrac{\int_0^\infty E \tau dn(E)}{\int_0^\infty dn(E)}$

Hydraulic analogy

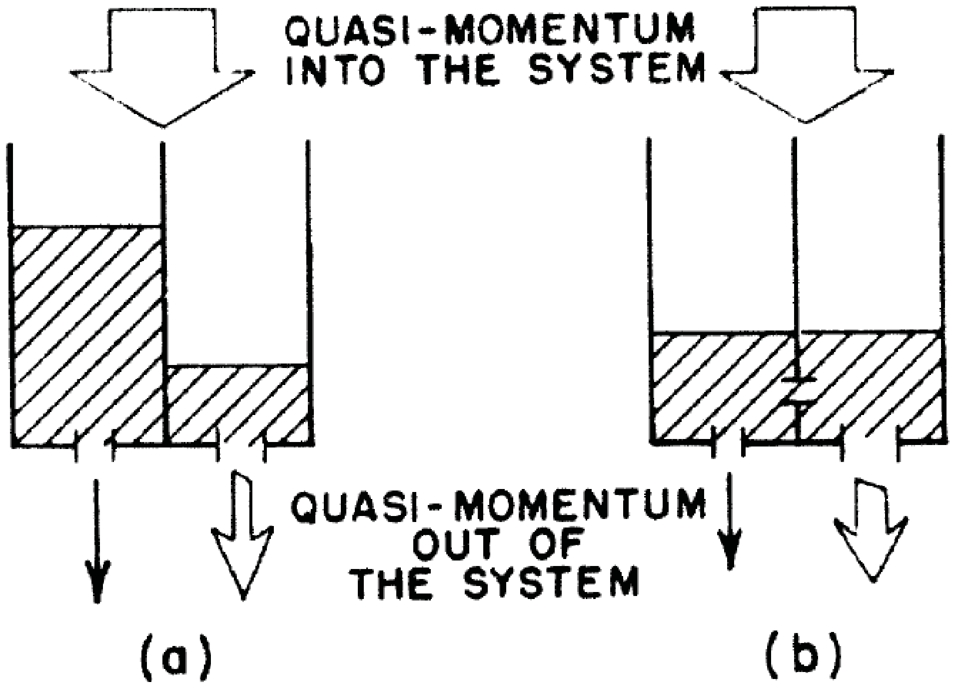


FIG. 1. Schematic analogy of quasimomentum balance in phonon system; (a) represents weak normal processes where various groups of phonons lose momentum individually, while in (b) strong N processes effectively transfer quasimomentum to the strongly scattered state (indicated by the large arrow).

The final expression for the thermal conductivity can be written as

$$\kappa = \kappa_k(1-\Sigma) + \kappa_Z F(\ell/L_{\text{eff}})\Sigma$$

with two components, the kinetic component and the Ziman component. The factor F, taking into account the geometry of the system is

$$F(\ell/L_{\text{eff}}) = \frac{1}{2\pi^2}\frac{L_{\text{eff}}^2}{\ell^2}\left(\sqrt{1+4\pi^2\frac{\ell^2}{L_{\text{eff}}^2}}-1\right)$$

This factor has been derived in previous paper and it has been obtained as an asymptotic expansion of a continued-fraction approximation of the thermal conductivity.

When the normal processes are negligible,

$$\tau_N \to \infty \qquad\qquad \Sigma \to 0$$

and the thermal conductivity is dominated by the kinetic part. When the normal processes are very rapid,

$$\tau_N \to 0 \qquad\qquad \Sigma \to 1$$

and the conductivity is dominated by the Ziman part.

F. X. Alvarez and D. Jou, Appl. Phys. Lett. 90, 083109 (2007).

For the scattering by impurities and boundaries we take the standard expressions

$$\tau_I^{-1} = A\omega^4 \qquad \tau_B^{-1} = \frac{v}{L_{\mathrm{eff}}}$$

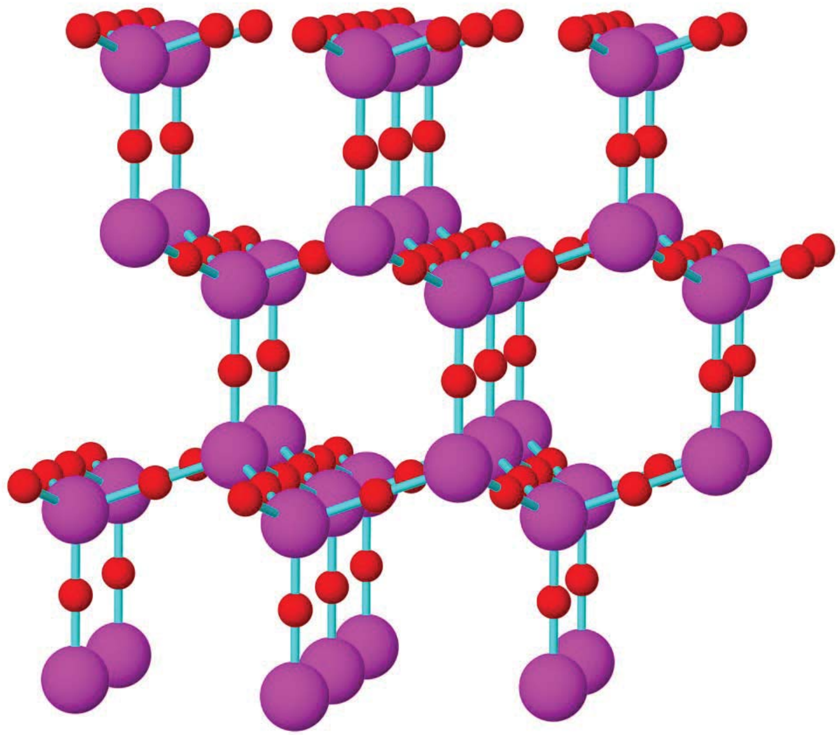
And for the scattering by N and U-processes we used an expression calculated by A. Ward and D.A. Broido by solving the Boltzmann equation with DFT theory

$$\tau_U^{-1} = B_U \omega^4 T[1 - \exp(-3T/\Theta_D)]$$

$$\tau_N^{-1} = \left(\frac{1}{B_N T} + \frac{1}{B_N' T^3}\right)^{-1} \omega^2 [1 - \exp(-3T/\Theta_D)]$$

- A. Ward and D.A. Broido, PRB81, 085205 (2010).

Adiabatic bond charge model



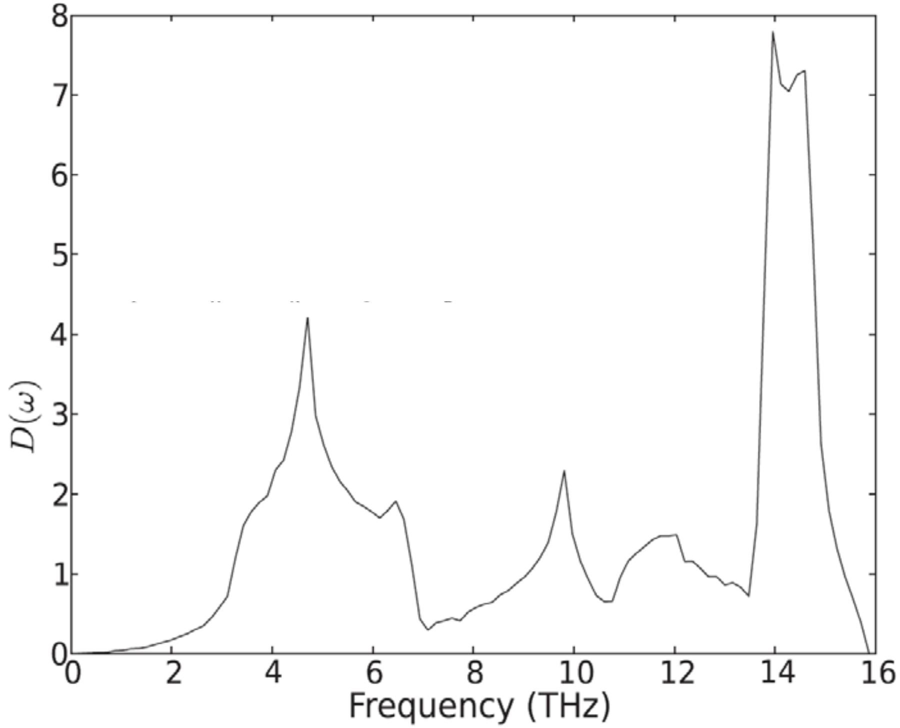
Parameters:

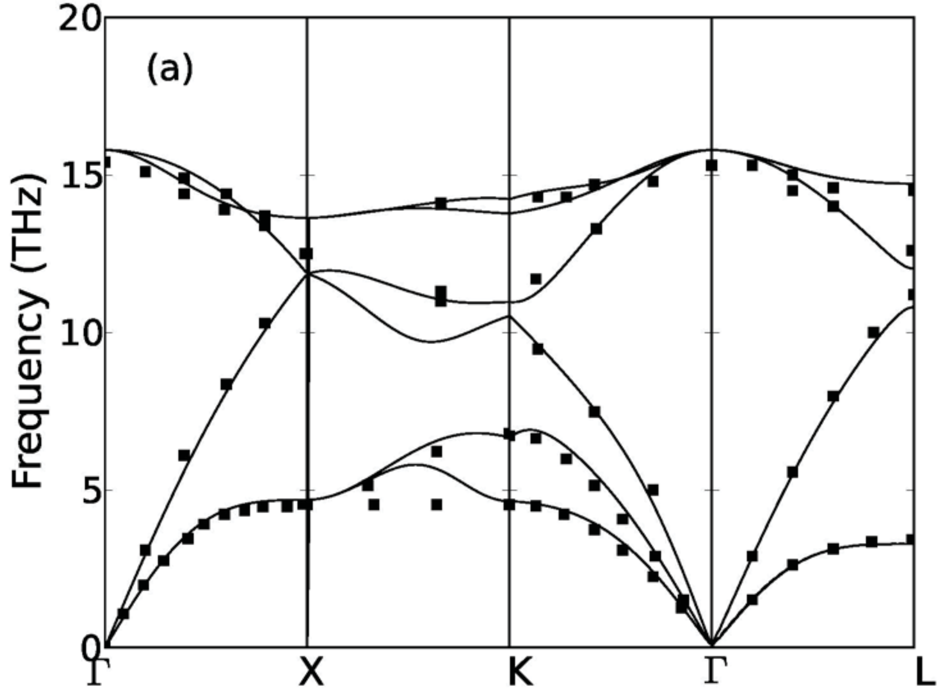
1. Bond stretching and bond-bending between atoms
2. Bond stretching between atoms and bond charges
3. Bond-bending between atoms and bond-charges
4. Effective charge

We used the same parameters as Weber for silicon in his original paper for bulk, thin films and nanowires.

- W. Weber, Phys. Rev. Lett. 33, 371 (1974).

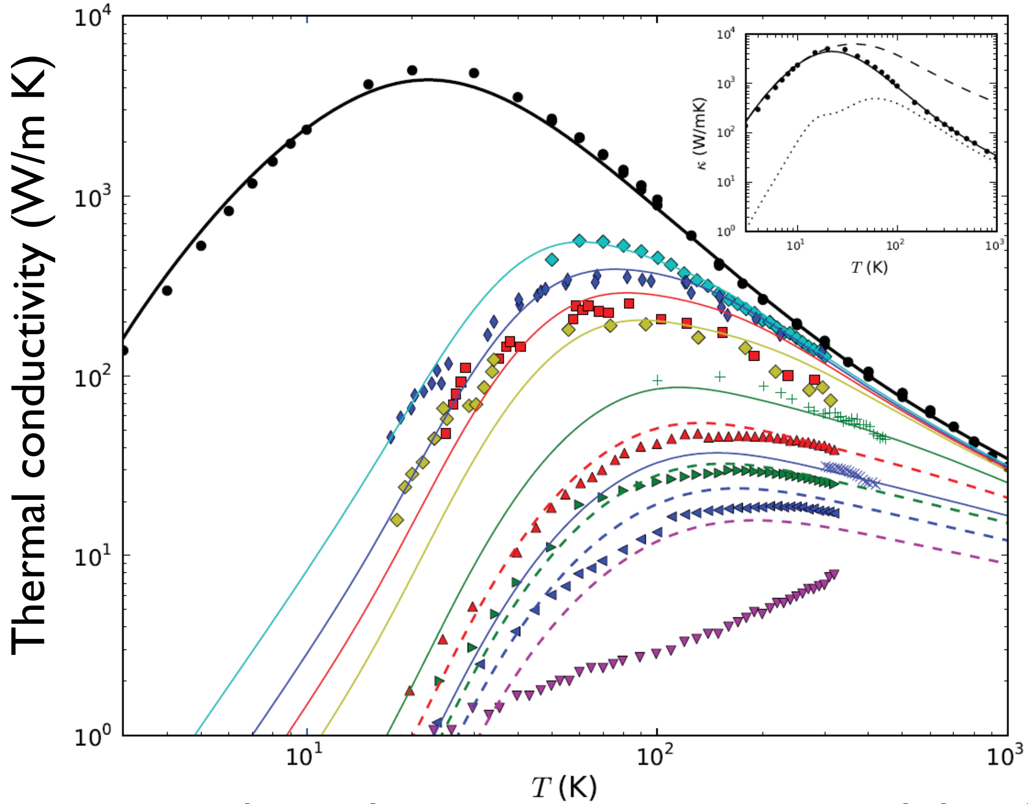
Bond charge model for silicon





Phonon DOS and relation dispersion for silicon with the parameters obtained from W. Weber.

Comparison with experiment



The symbols in the corresponding colors represent experimental data from different references: Bulk (circles), micro thin films (squares and diamonds), nano thin films (crosses), and nanowires (triangles). The curves represent the thermal conductivity predicted from our model as a function of *T* for different Si samples. The dashed and dotted curves in the inset stand for the contributions *of the kinetic* and *Ziman terms,* respectively.

Conclusions

- We have analyzed the phonon thermal conductivity of bulk Si, thin films and Si nanowires within the classical limit
- A modifield Gruyer-Krumhansl model, taking into account a geometrical factor obtained previously through a continuous-fraction approx. of the thermal conductivity
- The reason of using the GKM is that the N-processes are better treated and they are important in Si even at RT
- For the lattice dynamics, the bond charge model proposed by Weber has been used because it has a clear physical meaning and a small number of parameters
- We show that above 30 nm, a classical theory can fit the experimental data with the bulk parameters