Phonon transport

Samy Merabia
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Outline

I. Introduction - phenomenology

II. Vibrations

III. Bulk thermal conductivity

IV. Phonon transport at interfaces

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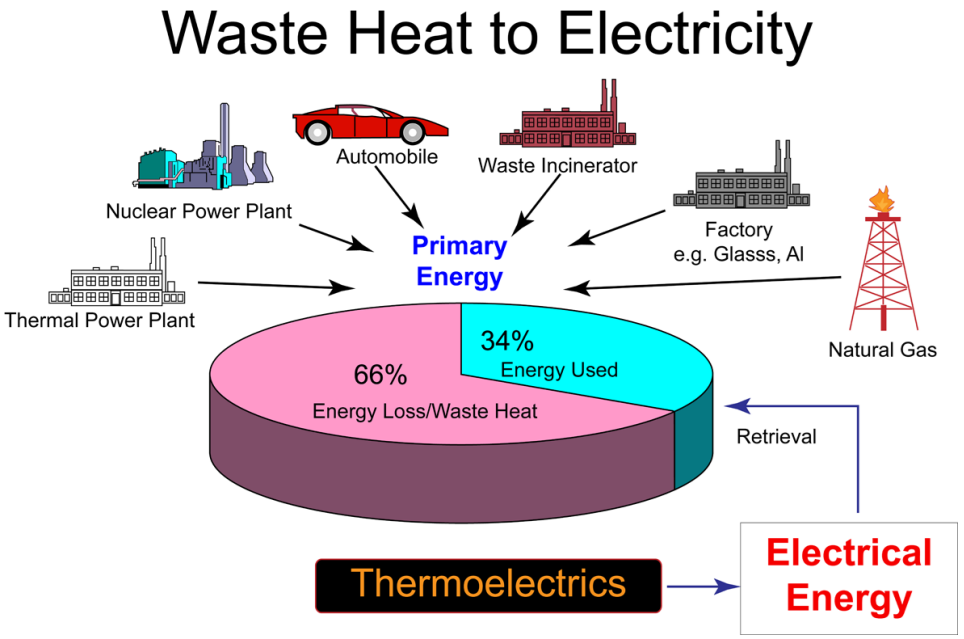
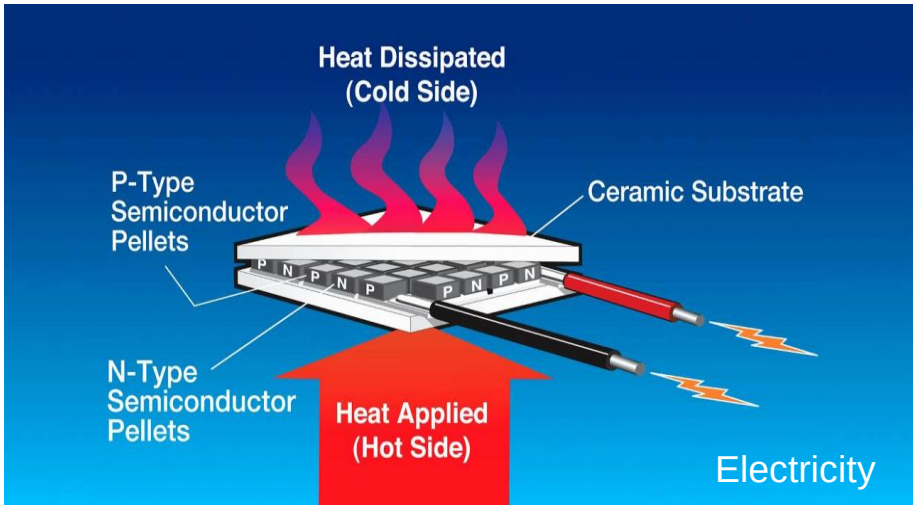
Waste heat recovery and thermoelectricity





-Urgent need to covert waste heat in recoverable form of energy

-The ideal thermoelectric material is a good electronic conductor, but a poor thermal conductor

Thermoelectricity and thermal conductivity

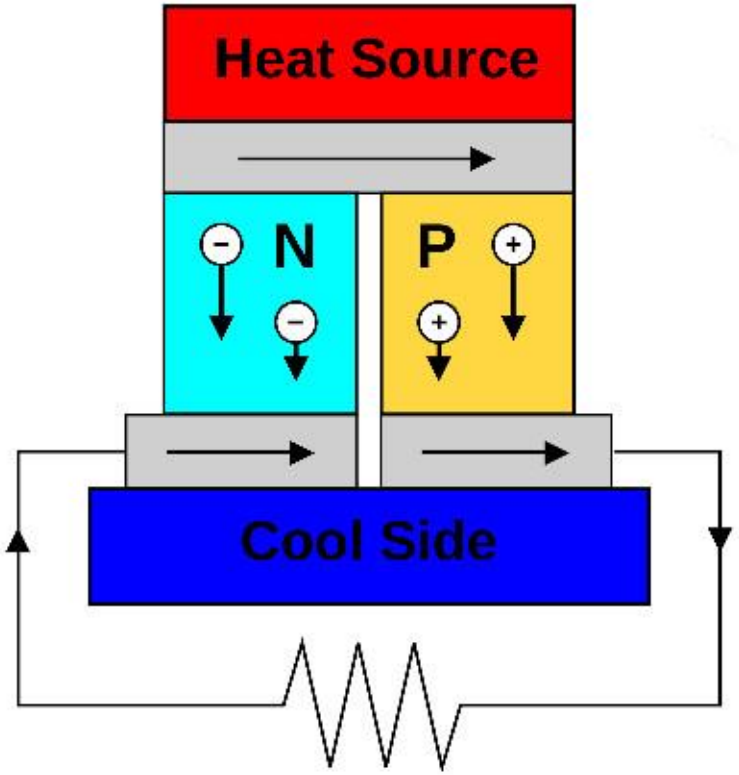


Figure of merit

$$Z = \frac{S^2\sigma}{\lambda_{ph} + \lambda_e}$$

S Seebeck coefficient

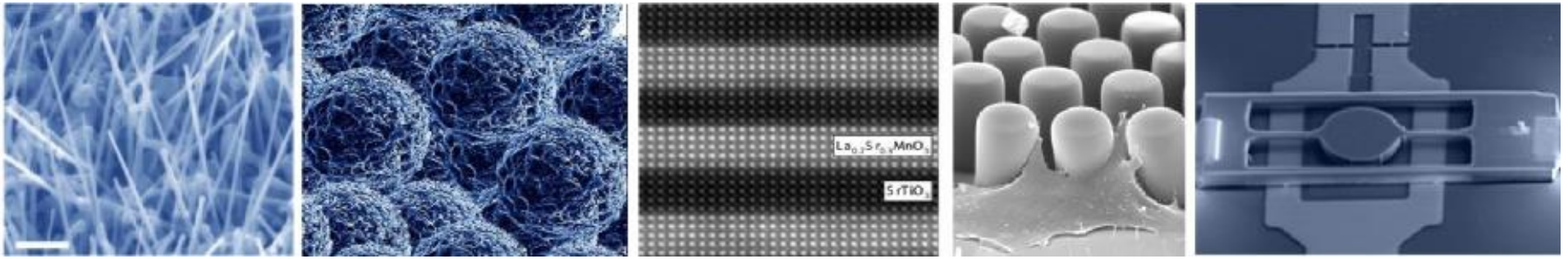
σ Electronic conductivity

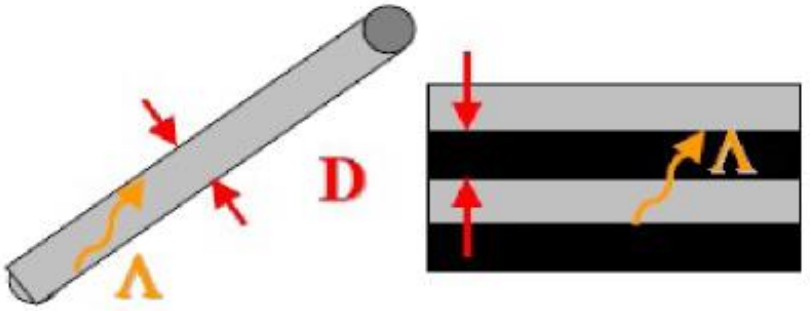
λ Thermal conductivity

Property	Metals	Semiconductors	Insulators
S (μVK^{-1})	~ 5	~ 200	~ 1000
σ ($\Omega^{-1}cm^{-1}$)	~ 10^6	~ 10^3	~ 10^{-12}
Z (K^{-1})	~3×10^{-6}	~2×10^{-3}	~5×10^{-17}

Strategies to reduce the thermal conductivity

Nanostructured materials





Two main strategies :

-boundary scattering (nanowires)

-interface scattering (superlattices)

$D >> \Lambda$, **Diffusive regime**

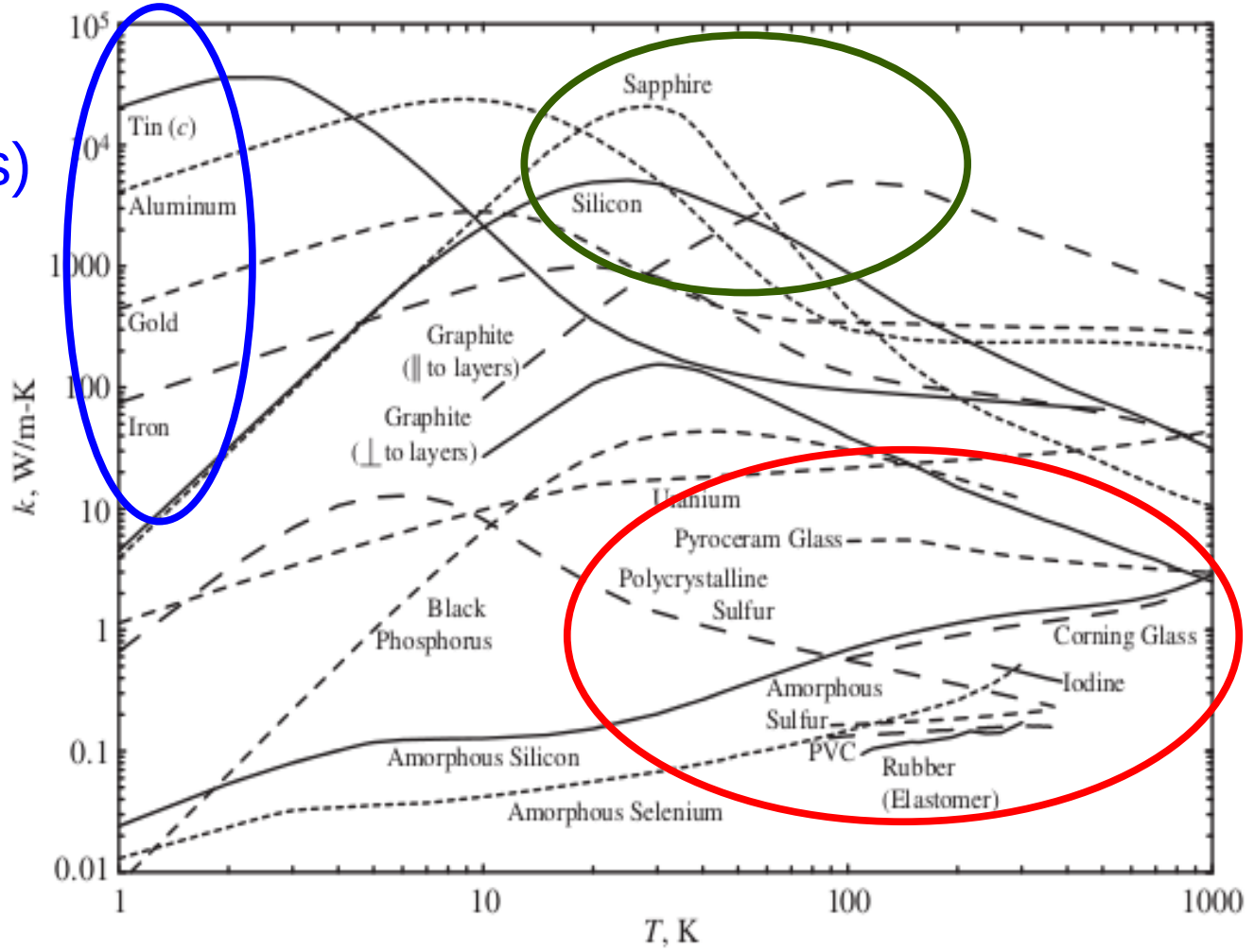
$D \approx \Lambda$, **Ballistic regime**

Si @300 K : $\Lambda > 300nm$

Thermal conductivity

Semi conductors
(phonons)

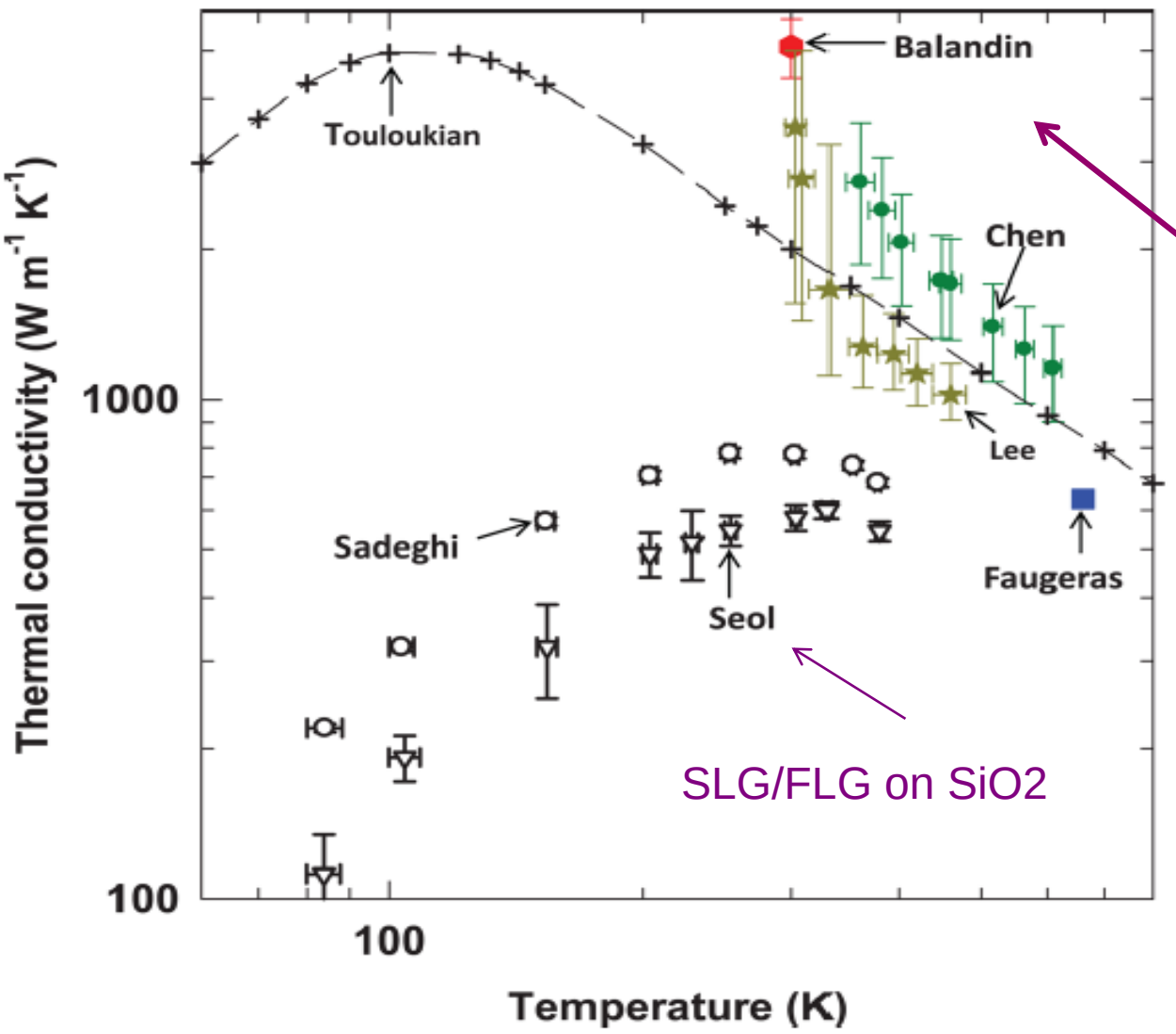
Metals
(electrons)



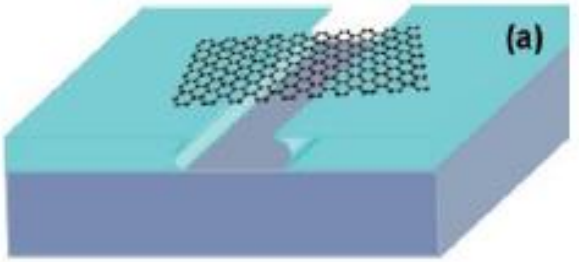
Polymers &
Amorphous
materials
(diffusons)

Y. Touloukian, *Thermophysical properties of matter*

Thermal conductivity



2000's



Suspended graphene

Nanoscale thermal transport II, *Appl. Phys. Rev.*, 2014

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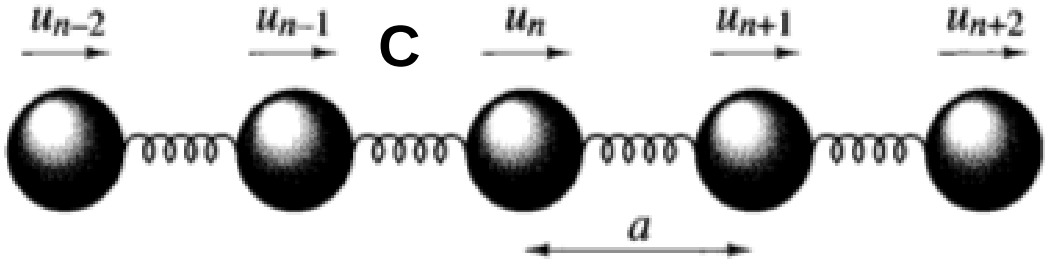
Dynamics of a monoatomic chain

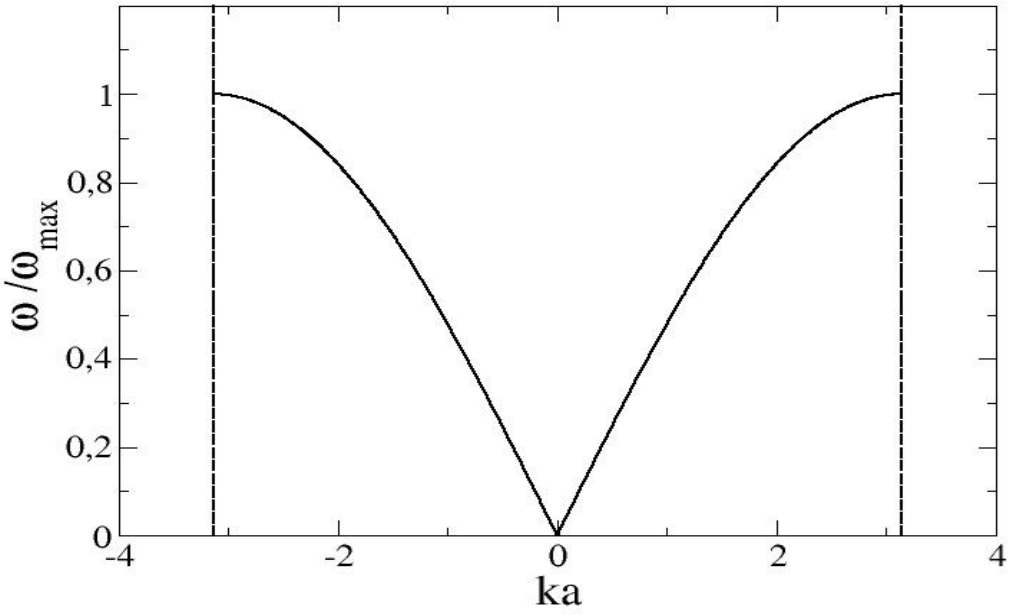


Equations of motion : $m\frac{d^2u_n}{dt^2} = C(u_{n+1} + u_{n-1} - 2u_n)$

Solutions : $u_n(t) = q_k \exp\big(i(kna - \omega(k)t)\big)$ $\quad k = \frac{2\pi p}{N}\,(p\,integer)$

Dispersion relation :

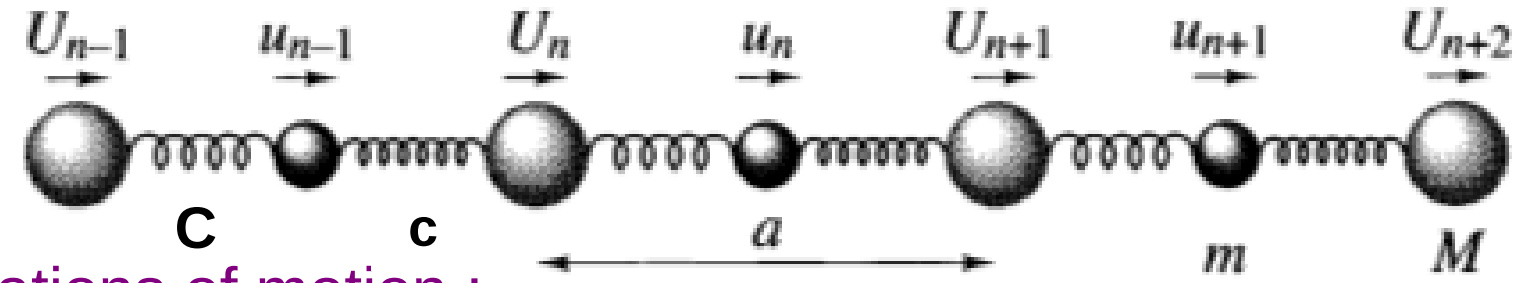
$$\omega(k) = \left(\frac{4C}{m}\right)^{1/2} \left|\sin\left(\frac{ka}{2}\right)\right|$$



Group velocity

$$v_g(k) = \left(\frac{C}{m}\right)^{1/2} a\cos\left(\frac{ka}{2}\right)$$

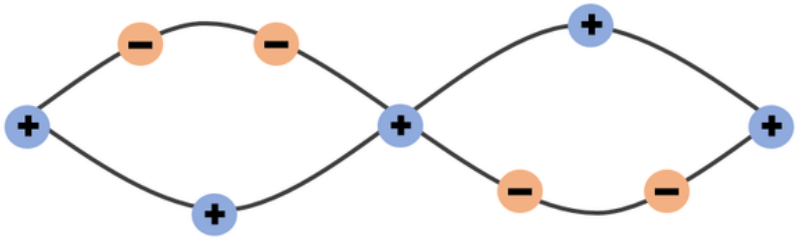
Dynamics of a diatomic chain



Equations of motion :

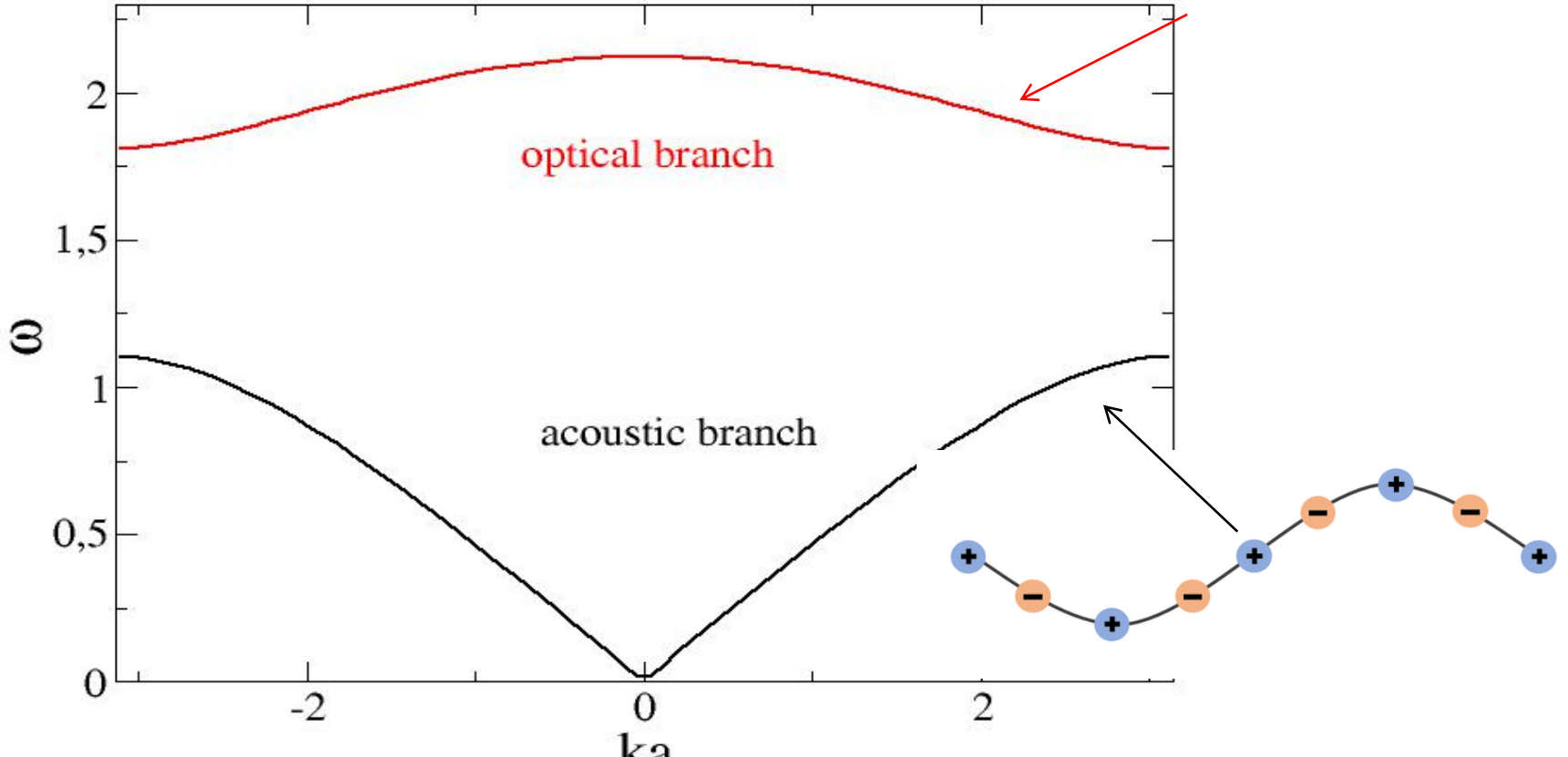
$$M\frac{d^2U_n}{dt^2} = -C(U_n - u_n) - c(U_n - u_{n-1})$$

$$m\frac{d^2u_n}{dt^2} = -c(u_n - U_{n+1}) - C(u_n - U_n)$$



Optical Phonon

K



Acoustic Phono

K

Three dimensional dispersion curve

Harmonic Hamiltonian : $H = H_0 + \frac{1}{2}\sum_{i,j} \Phi_{ij}^{\alpha\beta} u_i^{\alpha} u_j^{\beta}$ $\quad \alpha, \beta \in x, y, z$

Harmonic force constants

Equations of motion : $m_i \frac{d^2 u_i^{\alpha}}{dt^2} = -\sum_j \Phi_{ij}^{\alpha,\beta} u_j^{\beta}$

Plane waves solutions : $u_i^{\alpha} = \frac{Q_{\vec{k}}}{\sqrt{m_i}} e_{\alpha}(\vec{k}) \exp\left(i(\vec{k} \cdot \vec{r_i} - \omega t)\right)$

Polarisation vector

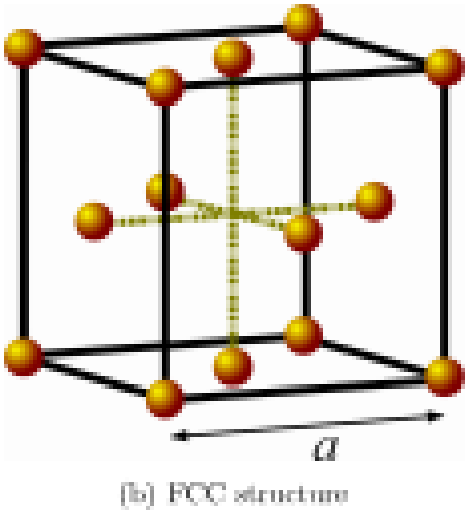
Dynamic matrix : $D_{ij}^{\alpha,\beta} = \frac{1}{\sqrt{m_i m_j}} \Phi_{ij}^{\alpha\beta}$

$$D_{\alpha\beta}(\vec{k}) e_{\beta}(\vec{k}) = \omega^2(\vec{k}) e_{\alpha}(\vec{k})$$

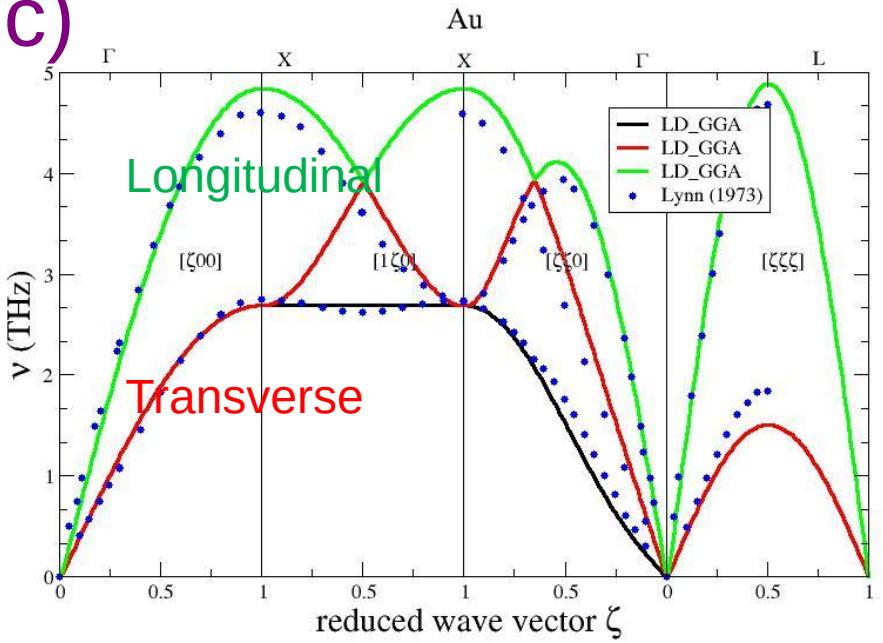
$$D_{\alpha\beta}(\vec{k}) = \sum_j D_{ij}^{\alpha\beta} \exp\left(i\vec{k} \cdot (\vec{r_j} - \vec{r_i})\right)$$

Dispersion curves

Gold (faced centered cubic)



(b) FCC structure



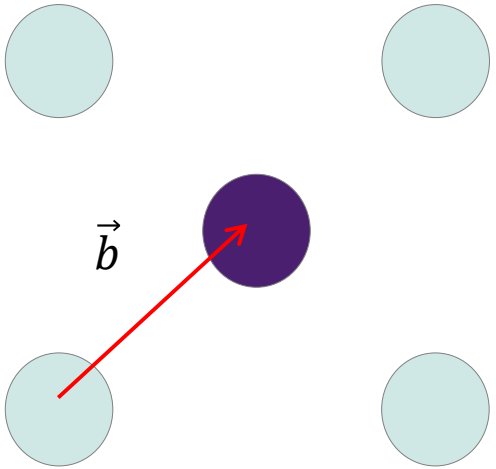
A. Alkurdi thesis

Dispersion relations for complex crystals

Eigenmodes :

$$u_i^{\alpha}(\vec{b}) = \frac{Q_{\vec{k}}}{\sqrt{m_i}} e_{\alpha}(\vec{k},\vec{b}) \exp\left(i\left(\vec{k}\cdot\vec{r_i}(\vec{b}) - \omega t\right)\right)$$

vector defining the position in the primitive cell



$$\omega_s^2(\vec{k}); e_{\alpha,s}(\vec{k},\vec{b})$$

Eigenvalues :

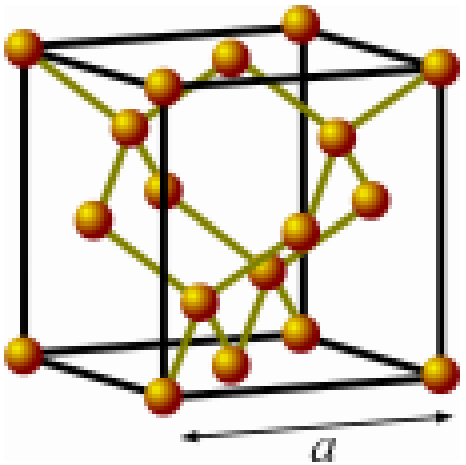
$$\sum_{\vec{b}'} D_{\alpha\beta}(\vec{k};\vec{b},\vec{b}') e_{\beta}(\vec{k},\vec{b}') = \omega^2(\vec{k}) e_{\alpha}(\vec{k},\vec{b})$$

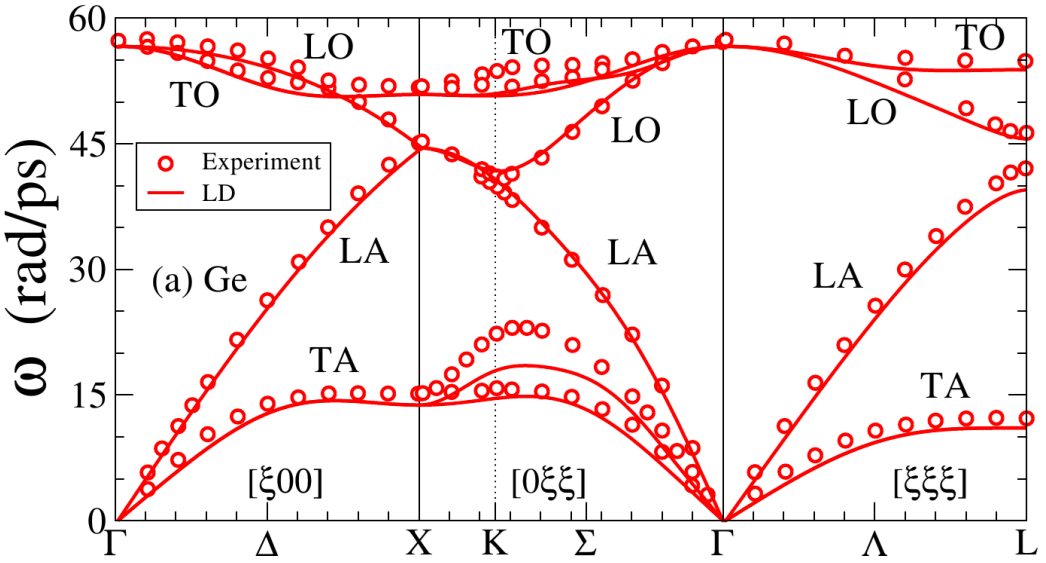
Three dimensional crystal : index s
Total = 3*p branches with p = number of atoms in the primitive cell

3 acoustic modes
3*(p-1) optic modes

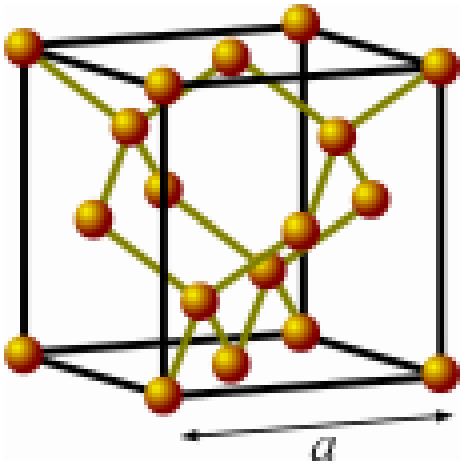
Dispersion curves

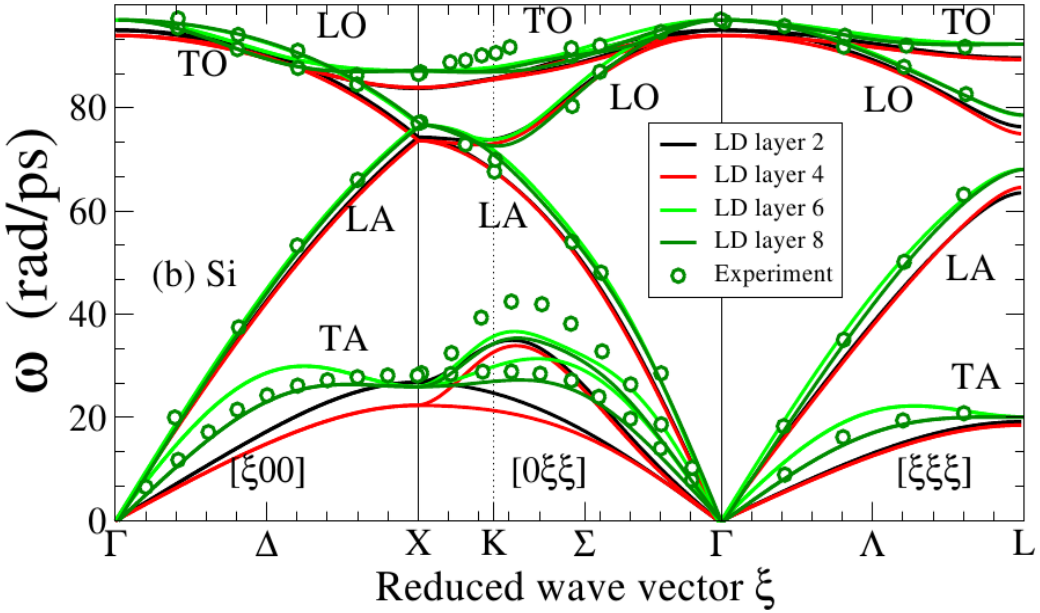
Germanium (diamond)





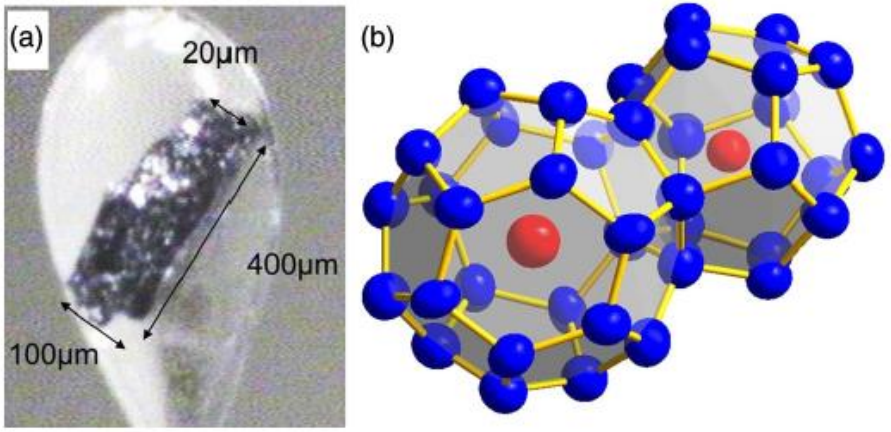
Silicon (diamond)

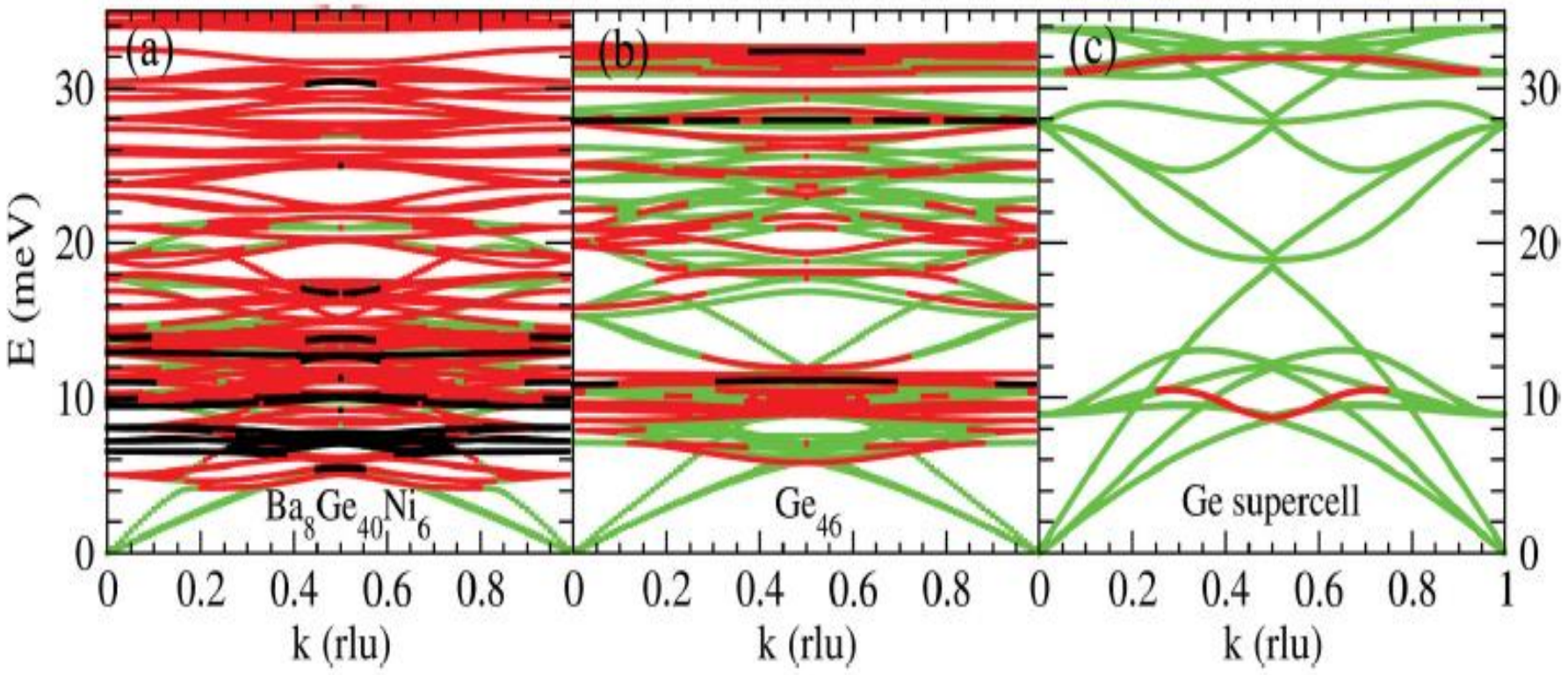




Dispersion in complex crystals

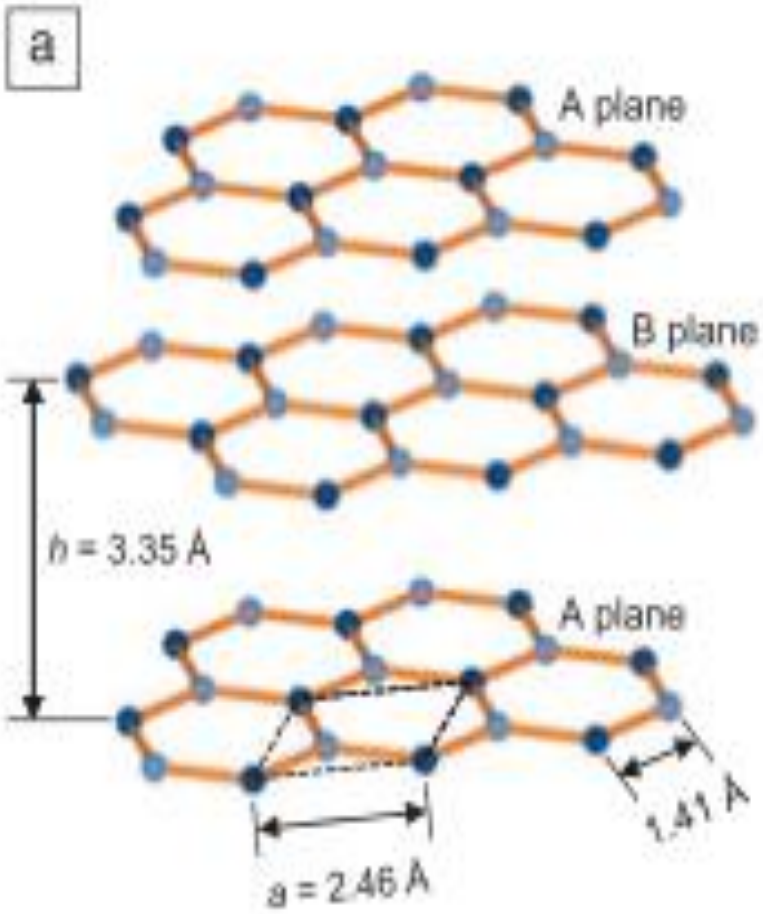
BaGeNi clatrate
(Pailhès et *al*., ILM)

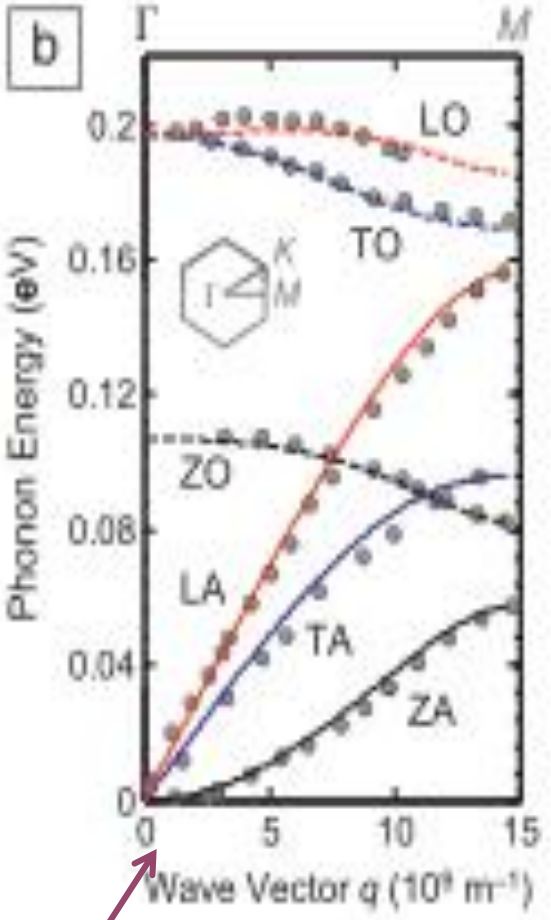




Euchner et *al.*, *Phys. Rev. B* 2012

Phonon dispersion in graphene





Dispersion of the bending modes

$\omega \sim q^2$

2D material

$$g(\omega) = \frac{\omega}{v_\phi(\omega) v_g(\omega)}$$

Bending modes dominant at low q

E. Pop, V. Varshney and A.K. Roy, *MRS Bull.*, 37 (2012) 1273

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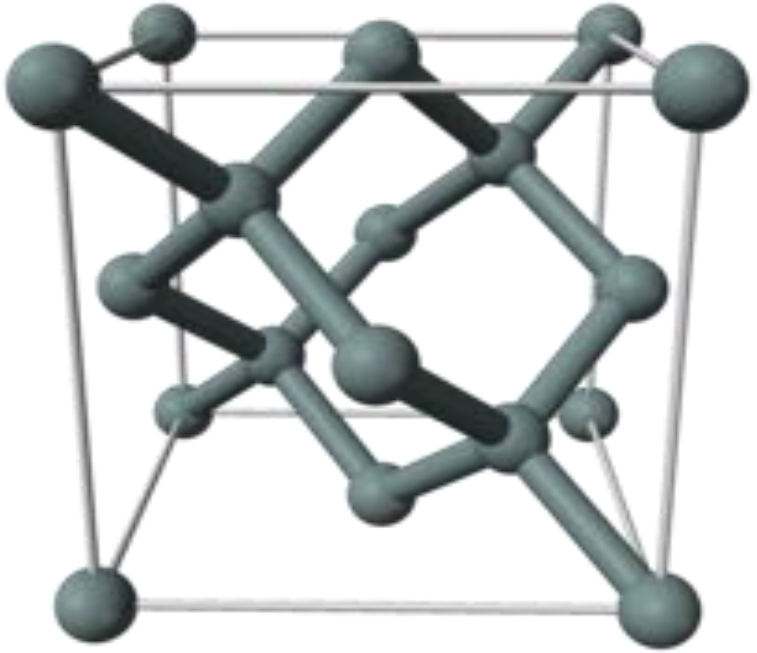
Molecular dynamics

-> see Evelyne Martin's talk

Molecular dynamics : $m\frac{d^2\vec{r_i}}{dt^2} = -\frac{\partial \Phi}{\partial \vec{r_i}}$ + thermal bath

Interatomic potential :

$$\Phi = \sum_{i<j} V_2\left(\vec{r_{ij}}\right) + \sum_{i<j<k} V_3\left(\vec{r_i}, \vec{r_j}, \vec{r_k}\right)$$



Stillinger-Weber potential (Silicon) :

$$\Phi = \sum_i \sum_{i>j} \nu_1(r_{ij}) + \sum_i \sum_{j\neq i} \sum_{k>j} \nu_2(r_{ij}, r_{ik}, \theta_{ijk})$$

$$\upsilon_1(r_{ij}) = A\epsilon\left(B\left(\frac{\sigma}{r_{ij}}\right)^p - 1\right)e^{\frac{\sigma}{r_{ij}-a\sigma}}$$

$$\upsilon_2(r_{ij}, r_{ik}, \theta_{ijk}) = \lambda\,\epsilon\left(\cos\theta_{ijk} - \frac{1}{3}\right)^2 e^{\frac{\sigma\gamma}{r_{ij}-a\sigma}} e^{\frac{\sigma\gamma}{r_{ik}-a\sigma}}$$

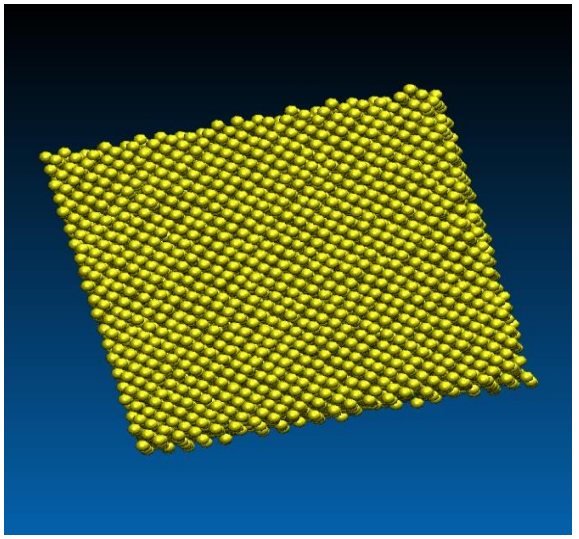
Thermal conductivity in MD

(workshop MD part II)

Essentially, three methods :

1. the « direct » method

Apply a temperature gradient or energy flux
and calculate the conductivity with Fourier law



2. Green-Kubo method

Probe the fluctuations around equilibrium
of the energy flux vector

3. « approach to equilibrium »

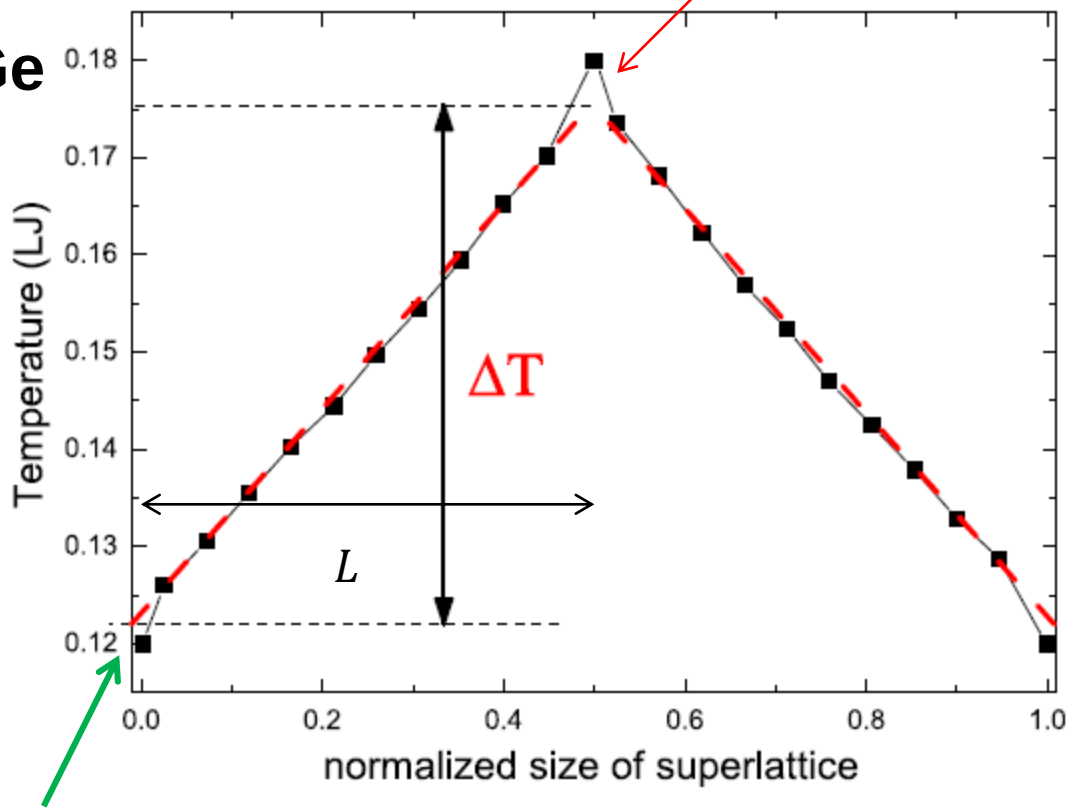
Probe the relaxation of a temperature profile
-> see Evelyne Martin's talk

1. Thermal conductivity : the direct method

Steady state temperature profile

Heat source

Example Si/Ge



Heat sink

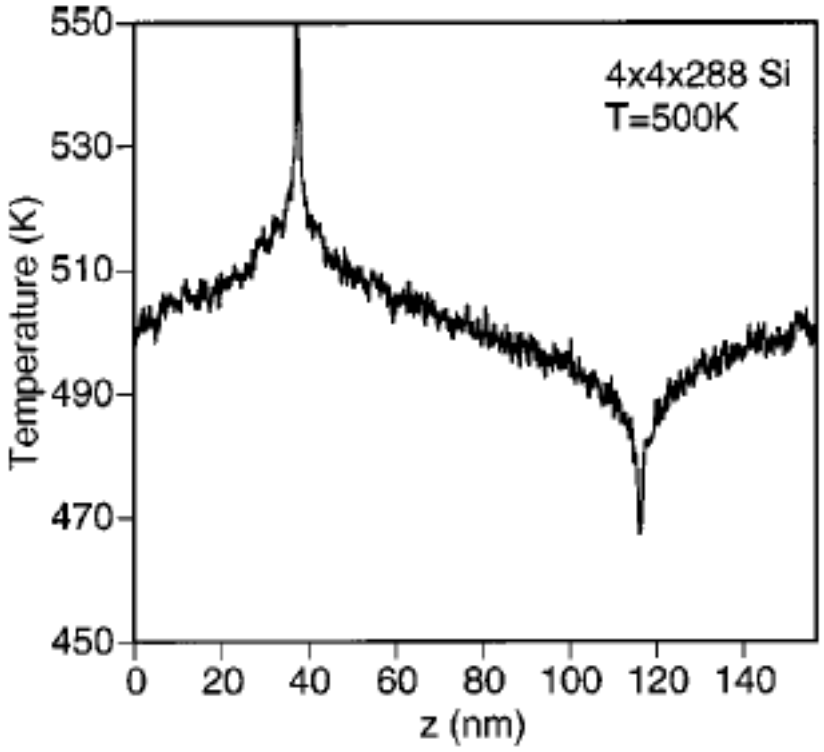
Fourier law : $J = -\lambda \frac{\Delta T}{L}$

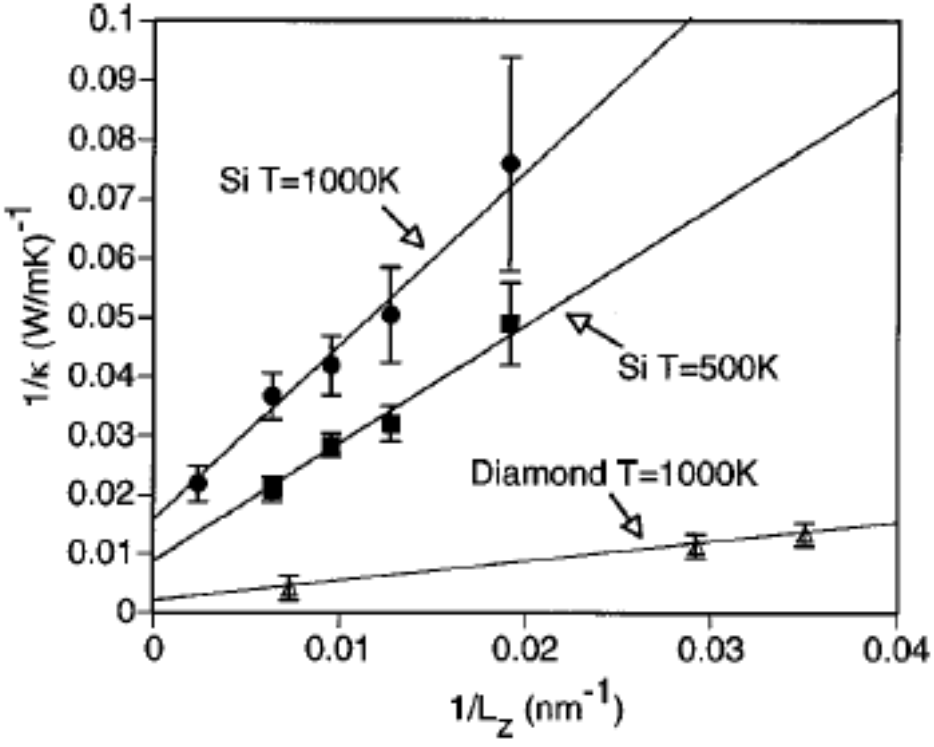
Molecular Dynamics Simulations and Thermal Transport at the Nano-Scale

Konstantinos Termentzidis[1,2] and Samy Merabia[3]

1. Thermal conductivity : the direct method

Strong finite size effects !





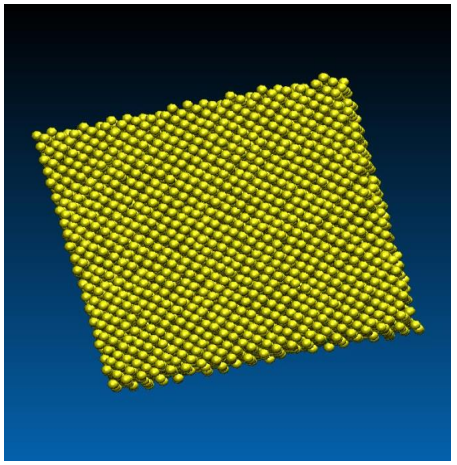
« Bulk » thermal conductivity

$$1/\Lambda(L) = 2/L + 1/\Lambda(L \to \infty)$$

Schelling, Phillpot, Keblinski, *Phys. Rev. B* (2002)
Selan et al. *Phys. Rev. B* 2014

II. Thermal conductivity : Green-Kubo equilibrium simulations

Green-Kubo formula : $$\lambda_{\alpha,\beta} = \frac{1}{V k_B T^2} \int_0^{+\infty} \langle J_\alpha(t) J_\beta(0) \rangle \, dt$$



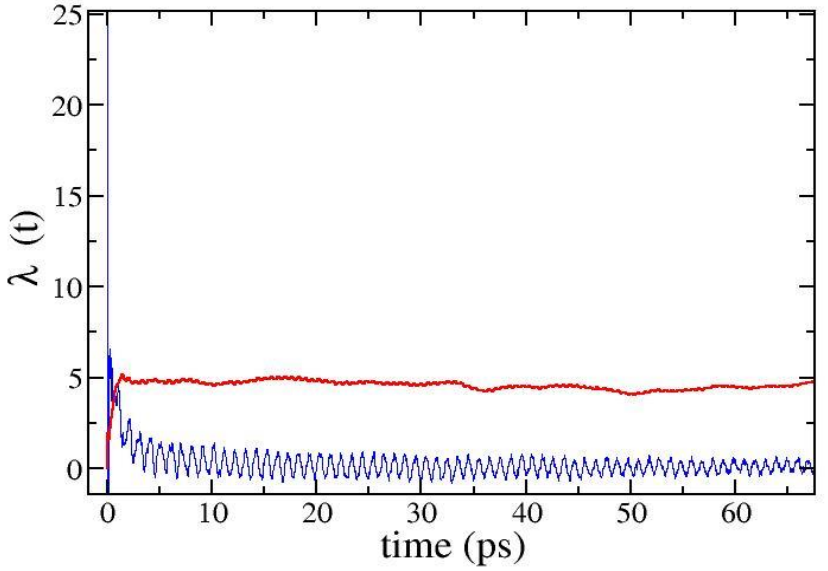
Heat flux vector : $$\vec{J} = \frac{d}{dt}\left(\sum_i E_i \vec{r_i}\right)$$

Two body potentials : $$\vec{J} = \sum_i E_i \vec{v_i} + \frac{1}{2}\sum_{i \neq j} \overrightarrow{r_{ij}} \overrightarrow{F_{ij}} \cdot \left(\vec{v_i} + \vec{v_j}\right)$$

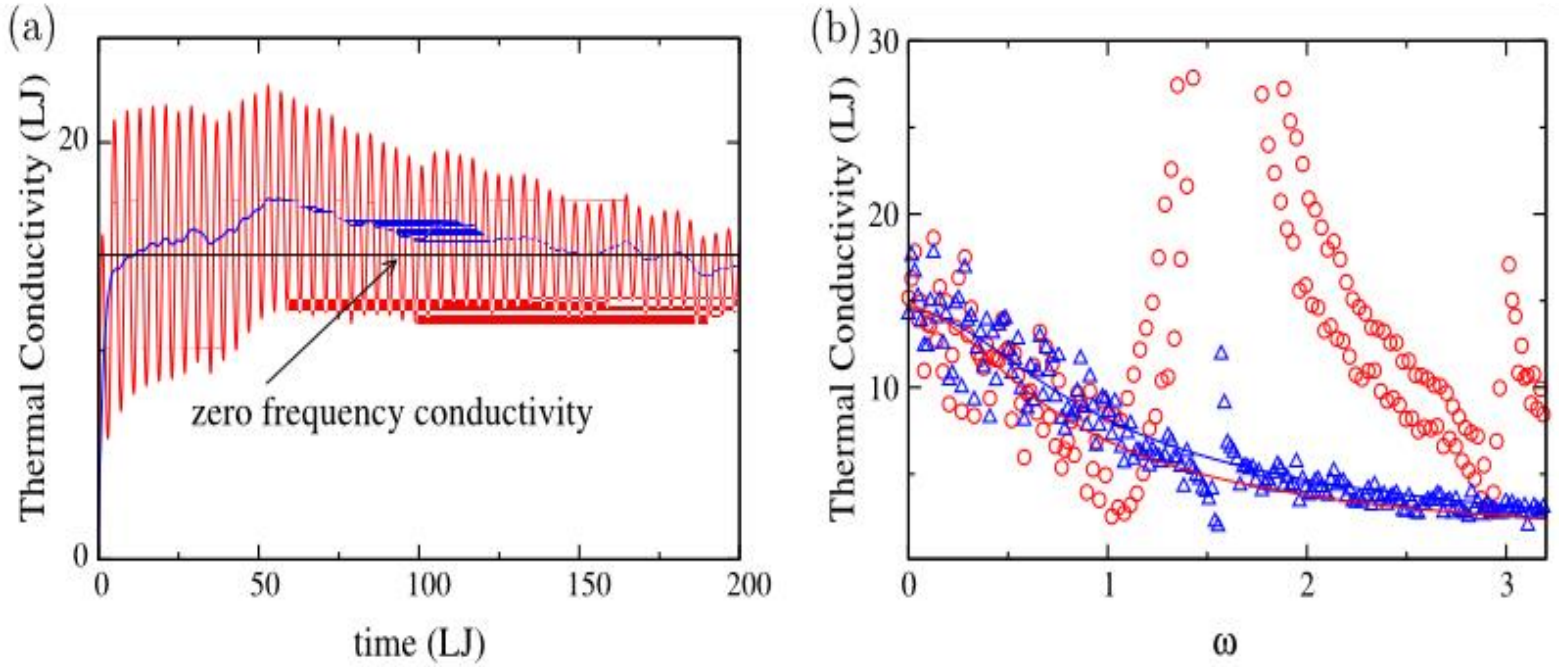
Many body potential :

$$\overrightarrow{J_{pot}} = \frac{1}{2}\sum_{i \neq j} \overrightarrow{r_{ij}} \frac{\partial U_j}{\partial \overrightarrow{r_{ij}}} \cdot \left(\vec{v_i} + \vec{v_j}\right)$$

Hardy's formula (1962)



Filtering optical modes



Heat flux vector :

$$\overrightarrow{J_{pot}} = \frac{1}{2}\sum_{i\neq j} \overrightarrow{r_{ij}}\,\overrightarrow{F_{ij}} \cdot \left(\overrightarrow{v_i} + \overrightarrow{v_j}\right)$$

$$\overrightarrow{J_{pot}} \simeq \frac{1}{2}\sum_{i\neq j} \overrightarrow{r_{ij}^{eq}}\,\overrightarrow{F_{ij}} \cdot \left(\overrightarrow{v_i} + \overrightarrow{v_j}\right)$$

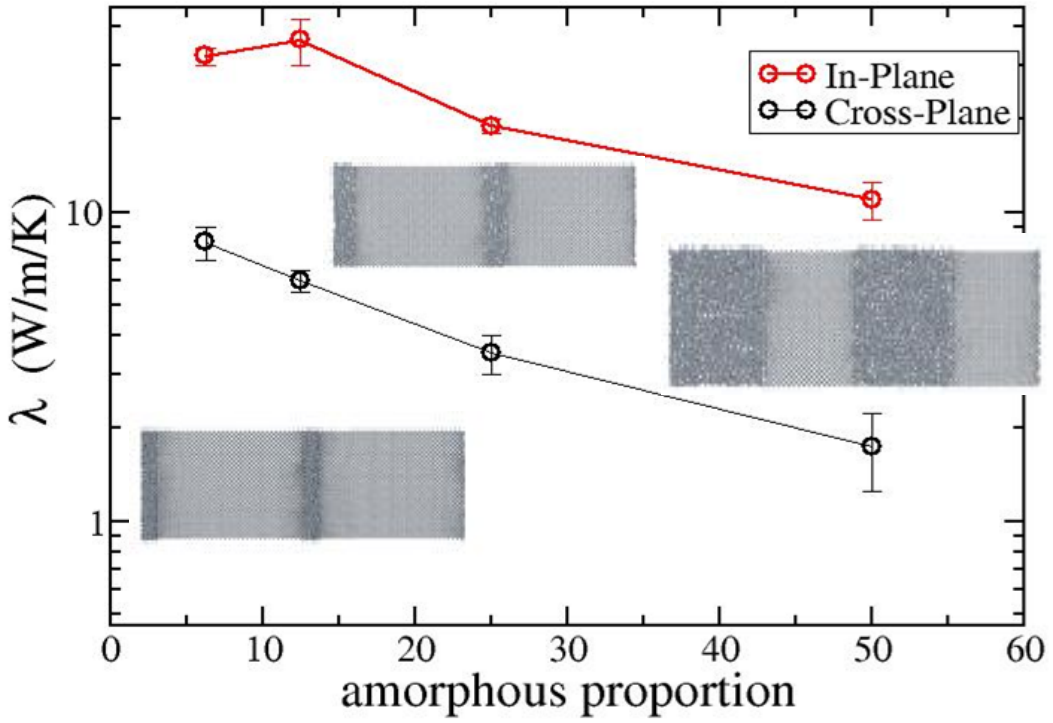
Spectral thermal conductivity :

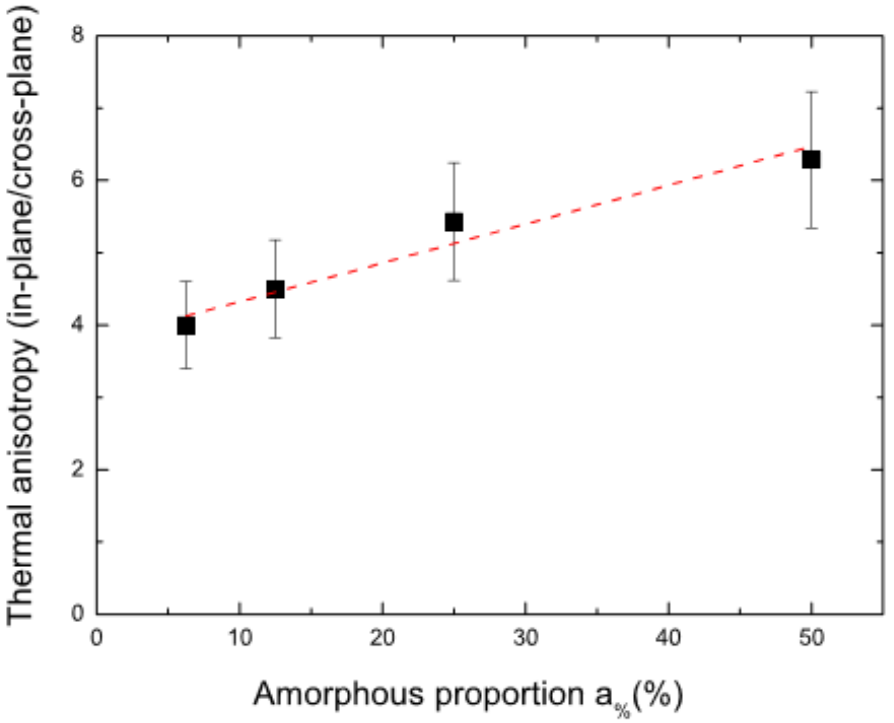
$$\lambda(\omega) = \frac{1}{Vk_BT^2}\int_0^\infty \langle J(t)J(0)\rangle \exp(i\omega t)dt$$

Termentzidis, Merabia, Chantrenne *IJHMT* (2011)

Anisotropic systems

Green-Kubo formula : $$\lambda_{\alpha,\beta} = \frac{1}{V k_B T^2} \int_0^{+\infty} \langle J_\alpha(t) J_\beta(0) \rangle \, dt$$





Lannord, Merabia, Albaret, Lacroix, Termentzidis, JPCM (2014)

Thermal conductivity : NEMD vs Green-Kubo equilibrium simulations

NEMD

Advantages :

-easy to implement
-computation of the thermal boundary resistance

Inconvenients :

-check of the linear regime
-severe finite size effects !

Equilibrium Green-Kubo

Advantages :

- less severe finite size effects
- full thermal conductivity tensor

Inconvenients :

-need to run several independent simulations
-plateau is difficult to identity

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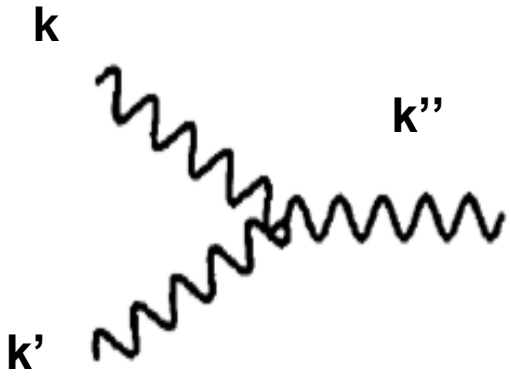
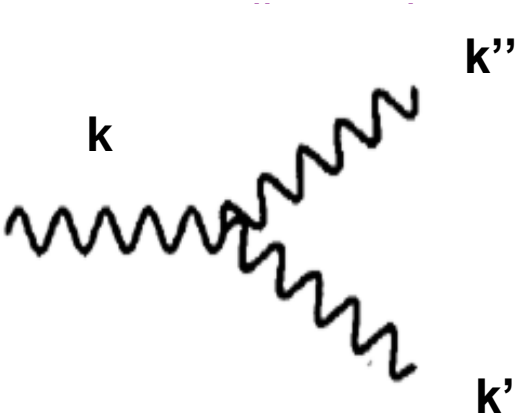
Relaxation times : phonon-phonon scattering



phonon fission



phonon fusion

Three phonons processes (Klemens formula) : $\tau^{-1}(\omega, T) = \gamma^2 \frac{k_B T}{M v^2} \frac{\omega^2}{\omega_D^2}$

Grüneisen parameter

Crystal Hamiltonian $H = \frac{1}{2}\sum \Phi_{ij}^{\alpha,\beta} u_{i,\alpha} u_{j,\beta} + \frac{1}{6}\sum \mathrm{X}_{ijk}^{\alpha,\beta,\gamma} u_{i,\alpha} u_{j,\beta} u_{k,\gamma}$

Phonon dispersion Phonon scattering

P.G. Klemens, « Solid State Physics » Academic Press New-York 1958

Thermal conductivity

Matthiessen's rule

$$\frac{1}{\tau} = \frac{1}{\tau_{3ph}} + \frac{1}{\tau_b} + \frac{1}{\tau_m} + \frac{1}{\tau_d} + \frac{1}{\tau_g}$$

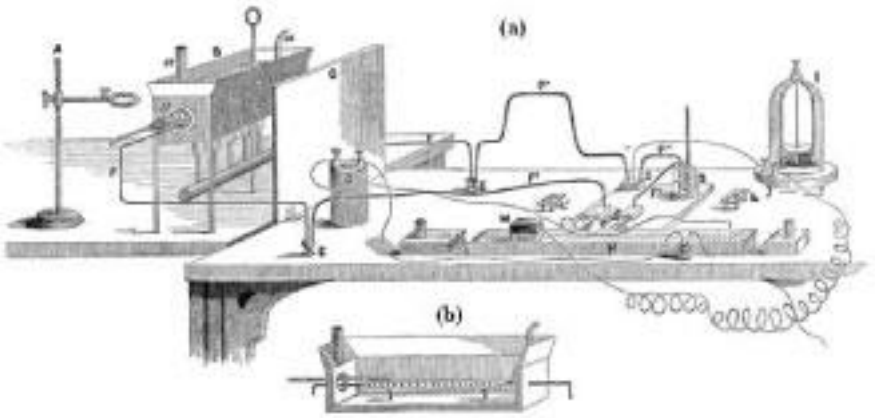
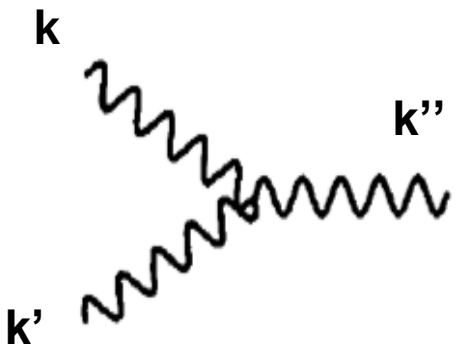
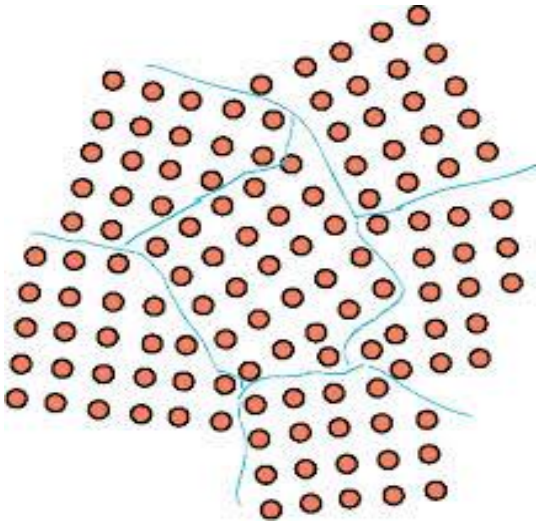


Figure 7 Matthiessen's apparatus. Credit: Ref. [32].

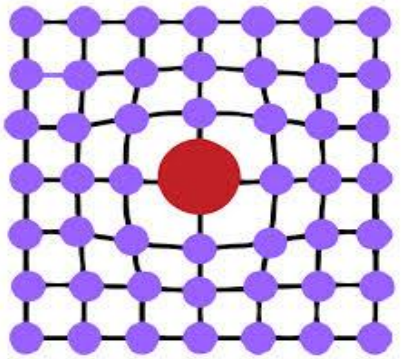
Phonon scattering



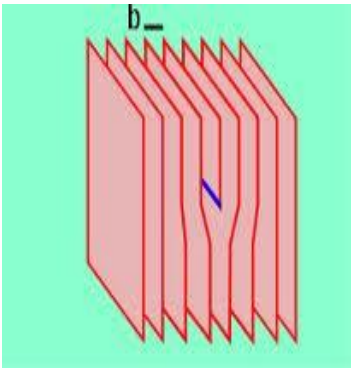
Grain boundaries



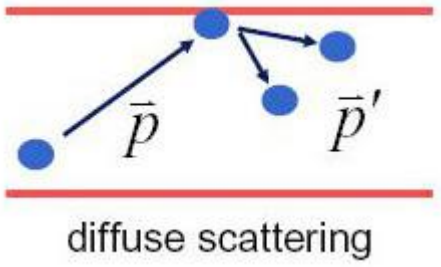
Defect scattering



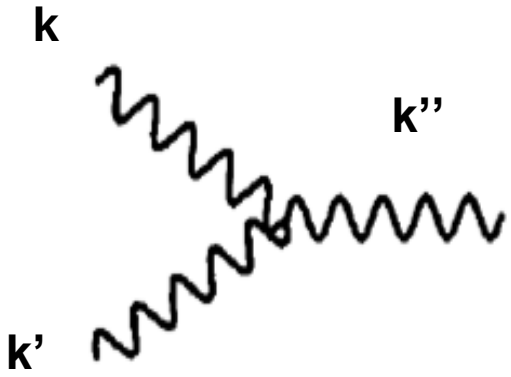
Dislocations



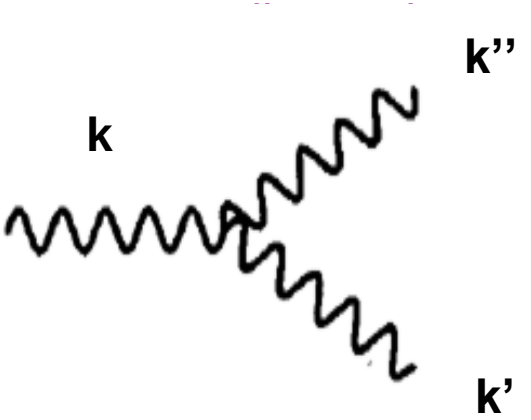
Boundary scattering



Relaxation times : phonon-phonon scattering



phonon fission



phonon fusion

Three phonons processes (Klemens formula) : $\tau^{-1}(\omega, T) = \gamma^2 \frac{k_B T}{M v^2} \frac{\omega^2}{\omega_D^2}$

Grüneisen parameter

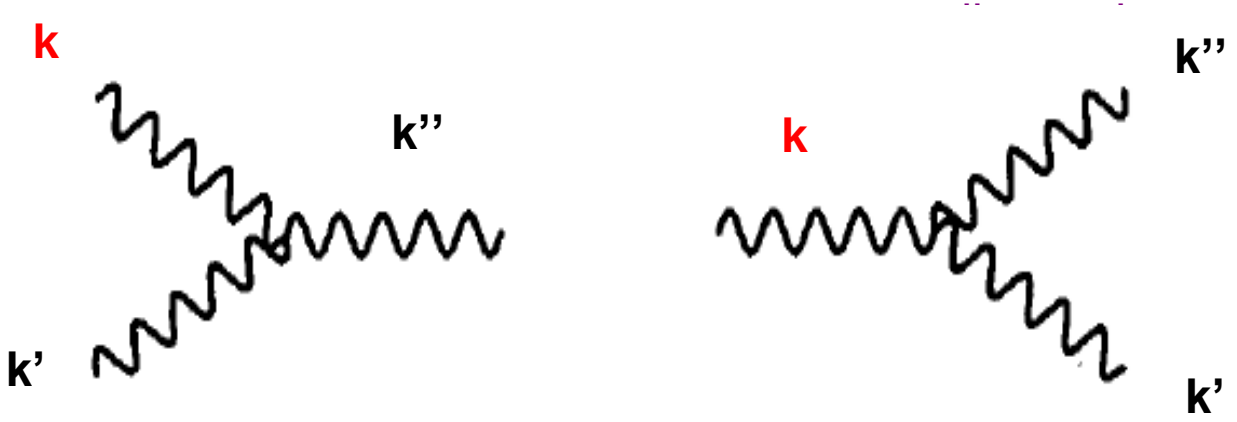
Crystal Hamiltonian $H = \frac{1}{2}\sum \Phi_{ij}^{\alpha,\beta} u_{i,\alpha} u_{j,\beta} + \frac{1}{6}\sum \mathrm{X}_{ijk}^{\alpha,\beta,\gamma} u_{i,\alpha} u_{j,\beta} u_{k,\gamma}$

Phonon dispersion Phonon scattering

P.G. Klemens, « Solid State Physics » Academic Press New-York 1958

Boltzmann transport equation

Boltzmann Transport Equation : $\frac{\partial n(\vec{k},s)}{\partial t} + \overrightarrow{v_g} \cdot \vec{\nabla} n = \sum_{\vec{k}\prime,\vec{k}\prime\prime} C\left(\vec{k},\vec{k}',\vec{k}''\right)$



Collision operator

$C = \Gamma(\vec{k}) + \Lambda(\vec{k},\vec{k}',\vec{k}'')$

Depends on k,k',k'' !

Single relaxation time approximation (SRTA): $C = -\frac{\left(n(\vec{k},s) - n_{eq}(\vec{k},s)\right)}{\tau(\vec{k},s)}$

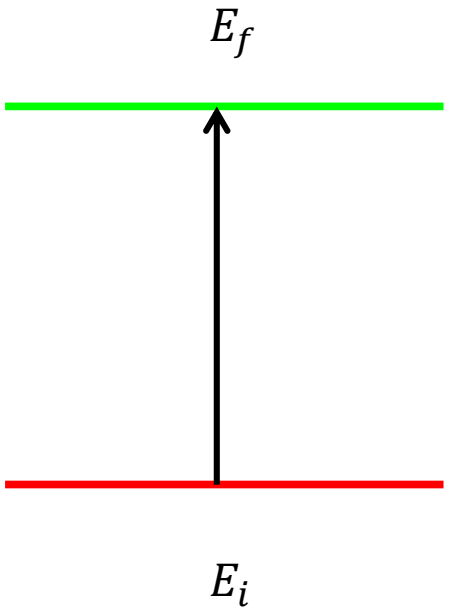
Boltzmann equation in the SRTA: $\frac{\partial n(\vec{k},s)}{\partial t} + \overrightarrow{v_g} \cdot \vec{\nabla} n = -\frac{\left(n(\vec{k},s) - n_{eq}(\vec{k},s)\right)}{\tau(\vec{k},s)}$

Thermal conductivity (SRTA)

$$\lambda = \frac{1}{3}\sum_{\vec{k},s} c_v \left|\overrightarrow{v_g}\right|^2 \tau(\vec{k},s)$$

Phonon-phonon scattering :perturbation theory

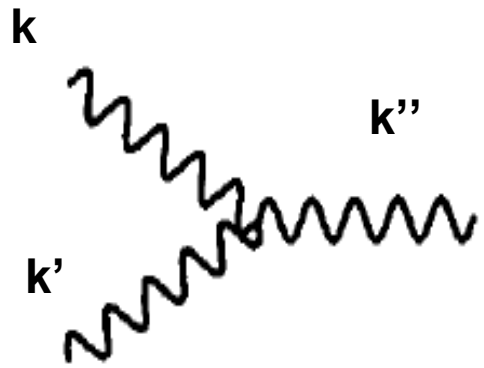
Hamiltonian : $H = H_0 + H_1$



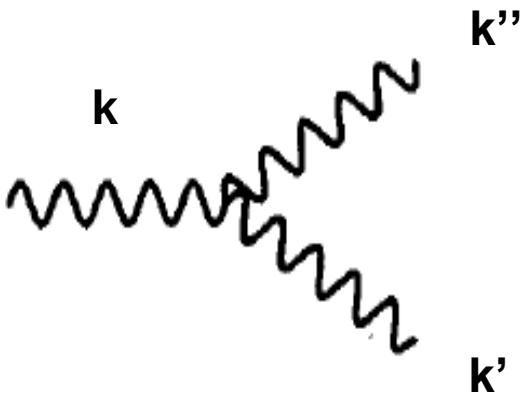
$$H_0 = \frac{1}{2}\sum \Phi_{ij}^{\alpha,\beta}\, u_{i,\alpha} u_{j,\beta}$$

$$H_1 = \frac{1}{6}\sum \mathrm{X}_{ijk}^{\alpha,\beta,\gamma}\, u_{i,\alpha} u_{j,\beta} u_{k,\gamma}$$

$$P_i^f = \frac{2\pi}{\hbar}|\langle i|H_1|f\rangle|^2 \rho(E_i)\delta\left(E_f - E_i\right)$$



Initial state Final state

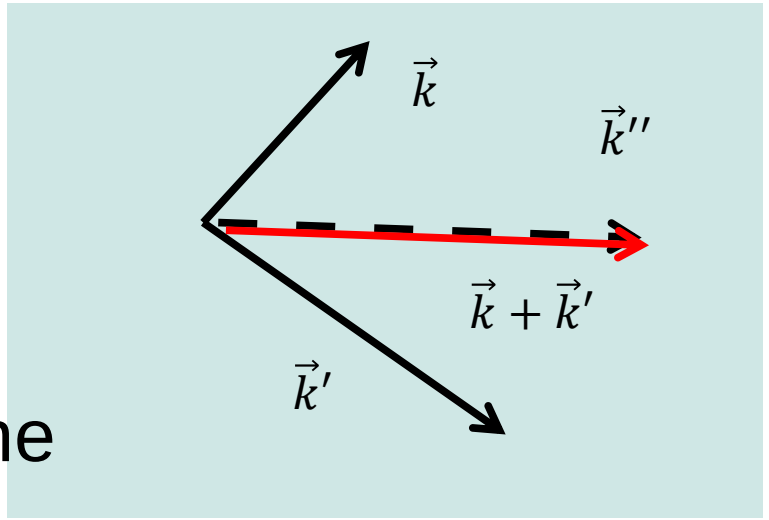


Initial state Final state

E. Fermi

Normal and Umklapp processes

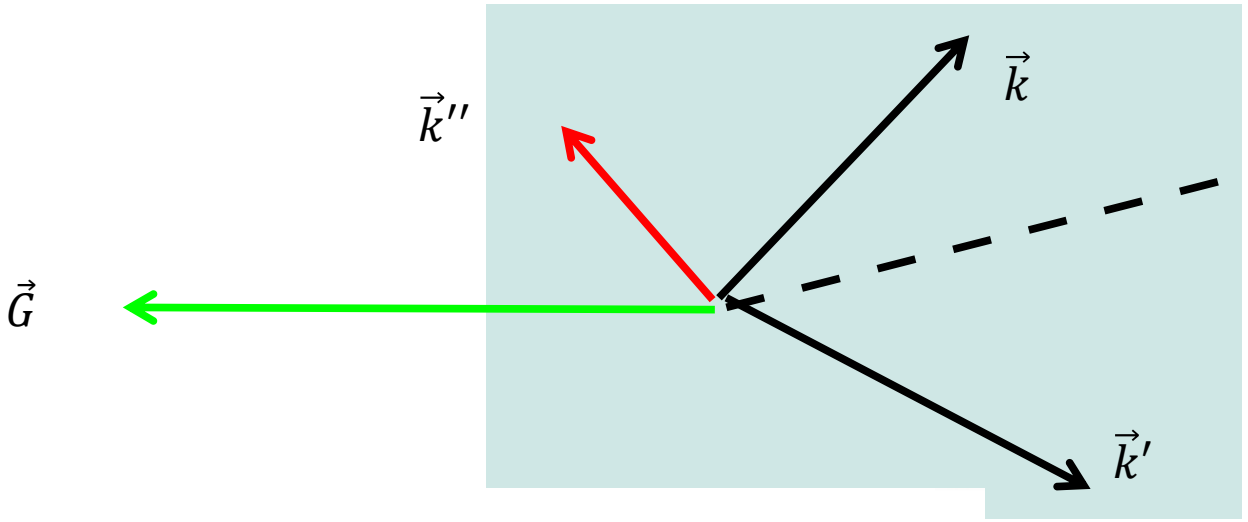
Normal processes



$$\vec{k} + \vec{k}' = \vec{k}''$$

Brilllouin zone

Umklapp processes



$$\vec{k} + \vec{k}' = \vec{k}'' + \vec{G}$$

Demystifying umklapp vs normal scattering in lattice thermal conductivity

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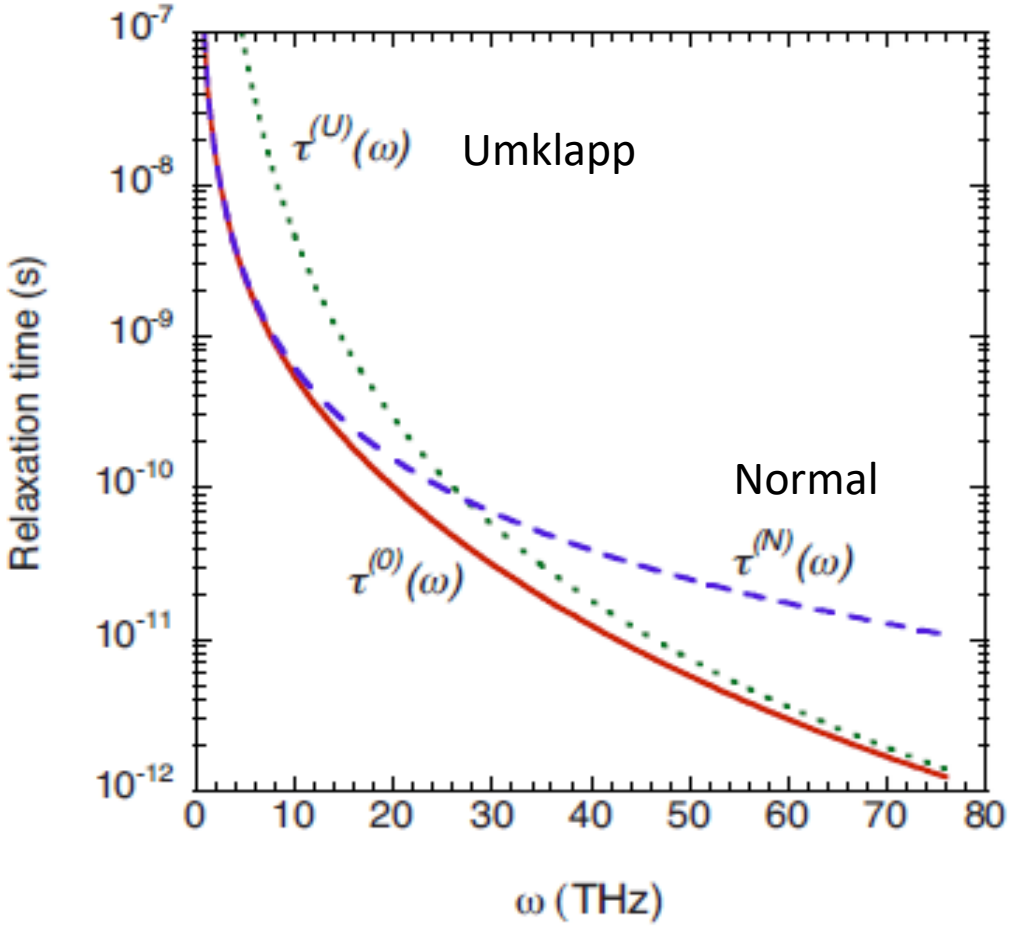
(Received 17 October 2013; accepted 28 July 2014)

PHYSICAL REVIEW B 81, 085205 (2010)

Intrinsic phonon relaxation times from first-principles studies of the thermal conductivities of Si and Ge

A. Ward and D. A. Broido
Department of Physics, Boston College, Chestnut Hill, Massachusetts 02467, USA
(Received 23 December 2009; published 4 February 2010)



DFT calculations Si,Ge

-Importance of N processes

$\tau_N \sim \omega^{-2}$ Limit thermal

transport as $\tau_N \sim \omega \to 0$

$\tau_U \sim \omega^{-4}$ Limit thermal

Transport at high frequencies /high temperatures

PHYSICAL REVIEW B 81, 085205 (2010)

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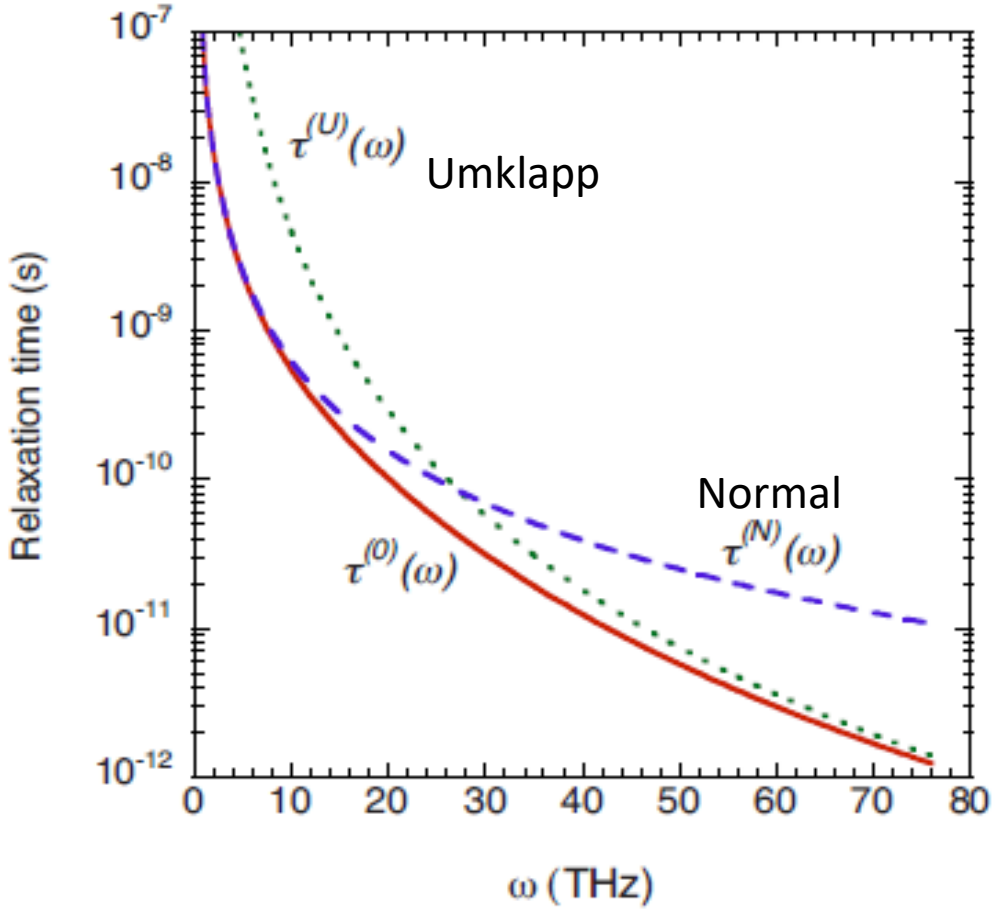


DFT calculations Si,Ge

-Importance of N processes

-Optical modes participate in phonon scattering :

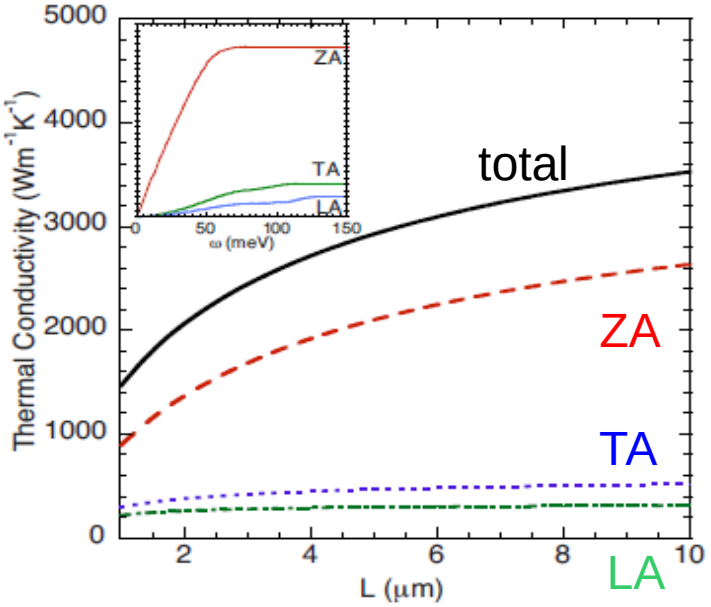
>50 % of the scattering processes Include optical phonons

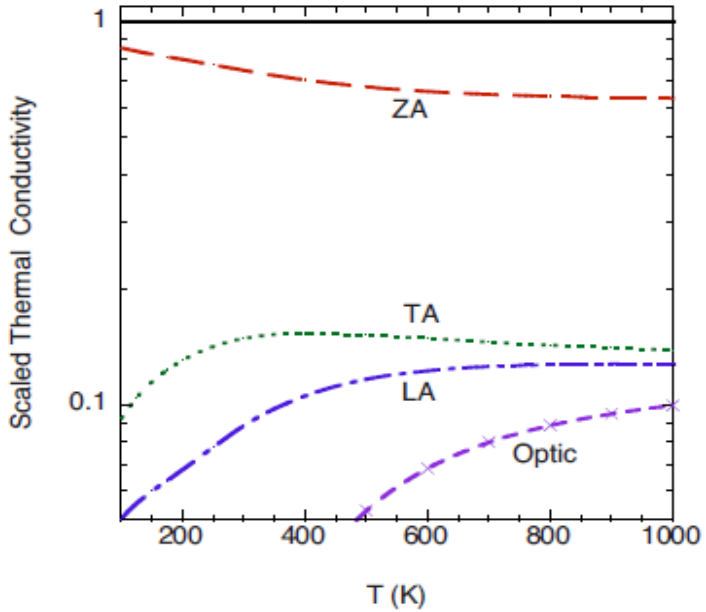
With optical : $\lambda = 150\, W/M/K$ @$300K$

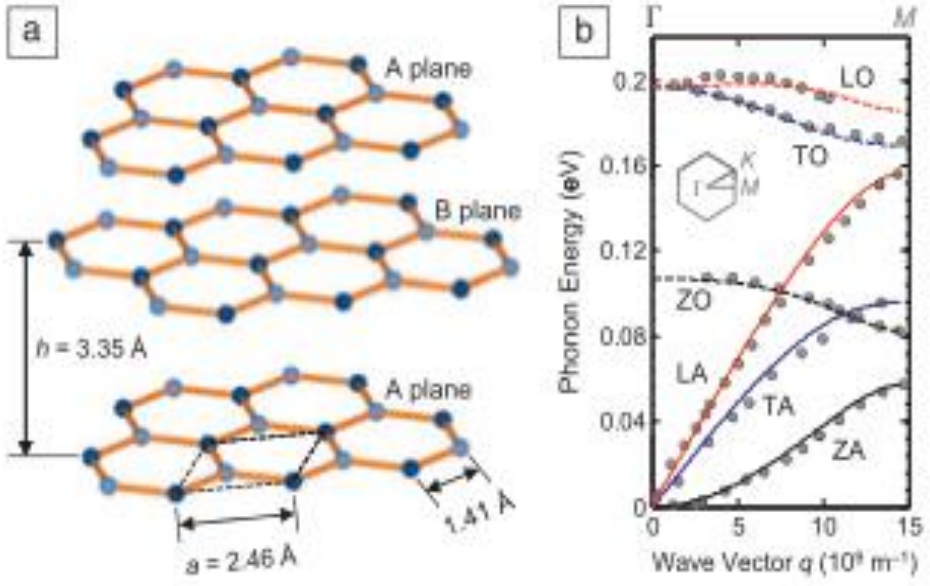
Without $\lambda = 500\, W/M/K$

-single relaxation time Approximation OK for Si,Ge

Scattering processes in graphene







Predominance of ZA modes

Constraints on ZA modes
Even number of ZA modes
(symmetry reasons)

Ex : ZA + TA → ZA
ZA + LA → TA forbidden !!

PHYSICAL REVIEW B **82**, 115427 (2010)

Flexural phonons and thermal transport in graphene

L. Lindsay,[1,2] D. A. Broido,[1] and Natalio Mingo[3,4]

Collective excitations of graphene : Failure of the single relaxation time approximation

Thermal Conductivity of Graphene and Graphite: Collective Excitations and Mean Free Paths

Giorgia Fugallo,*[†,‡] Andrea Cepellotti,[‡,¶] Lorenzo Paulatto,[‡] Michele Lazzeri,[†] Nicola Marzari,[‡,§] and Francesco Mauri[†]

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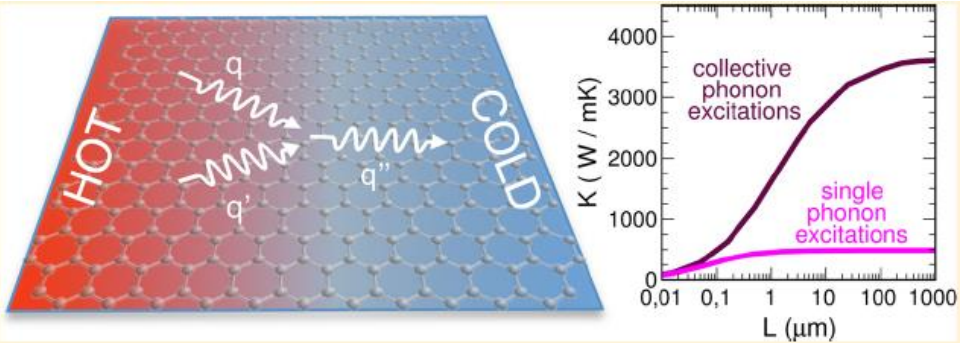
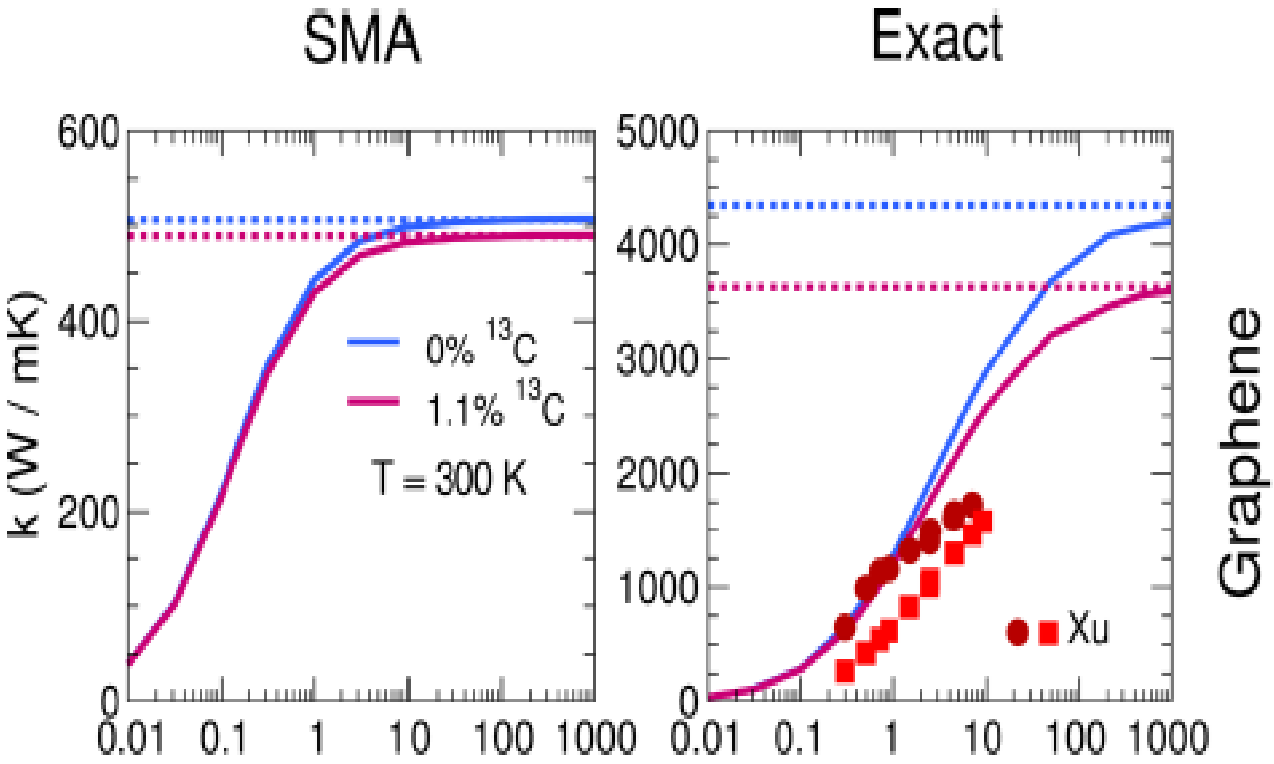
[‡]Theory and Simulations of Materials (THEOS), École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland
[§]National Center for Computational Design and Discovery of Novel Materials (MARVEL), École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland





SRTA underestimates

by one order of magnitude

the thermal conductivity

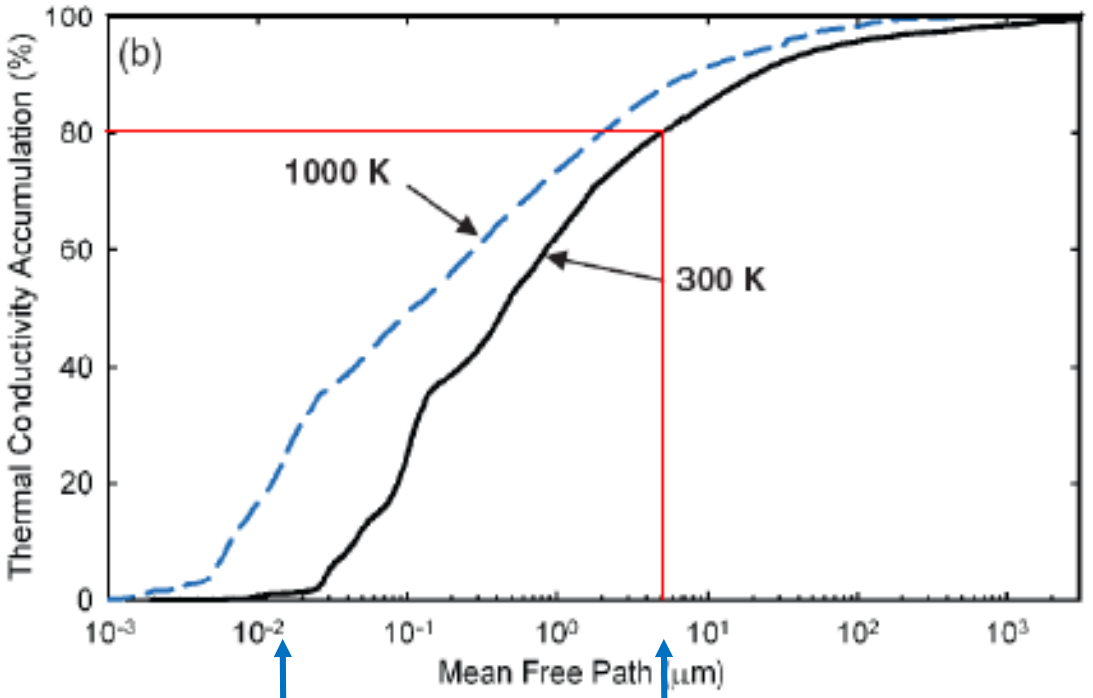
and the maximal mean free

paths

Accumulation function : phonon spectroscopy

MD simulations Si, Henry and Chen

$$\lambda(\omega) = \int_0^{\omega} c_v(\omega) v_g(\omega) \Lambda(\omega) d\omega$$



Henry and Chen, J. Comp. Theo. Nanosci 5, 1 (2008)

Message 1 : wide distribution of mean free paths

10 nanometers

10 microns

Accumulation function :phonon spectroscopy

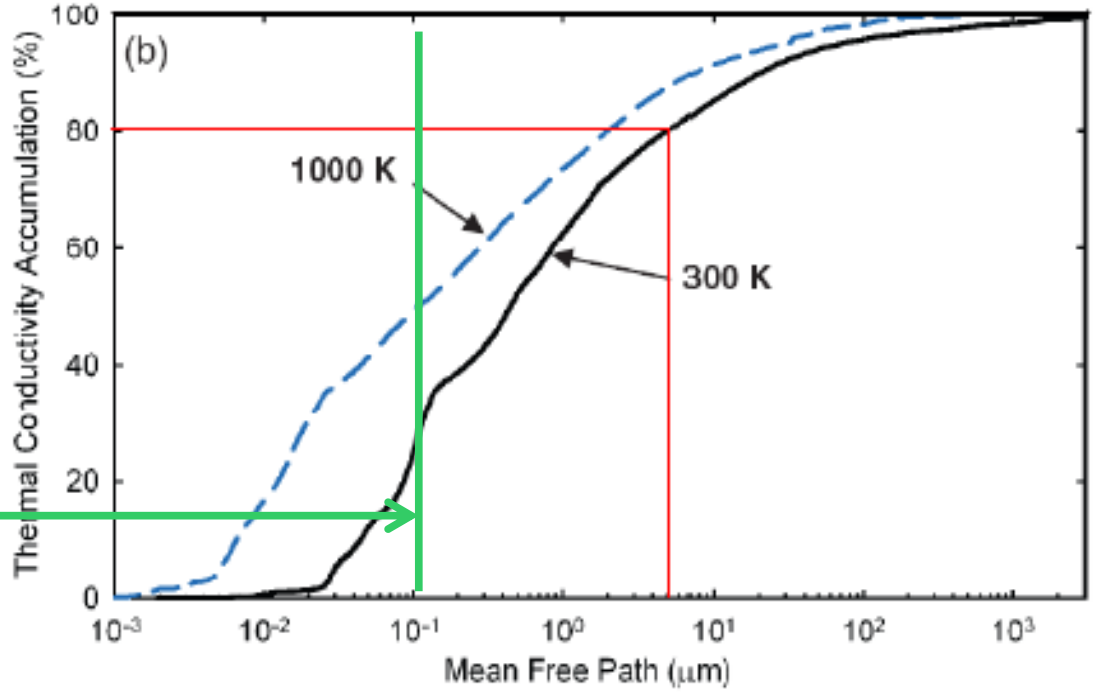
$$\lambda(\omega) = \int_0^{\omega} c_v(\omega) v_g(\omega) \Lambda(\omega) d\omega$$

MD simulations Si, Henry and Chen

Kinetic theory: $\lambda = \frac{1}{3} v_s c_v \Lambda$

$\lambda = 150\, W/m/K \; @300K$

$\Lambda = 100nm$



Henry and Chen, J. Comp. Theo. Nanosci 5, 1 (2008)

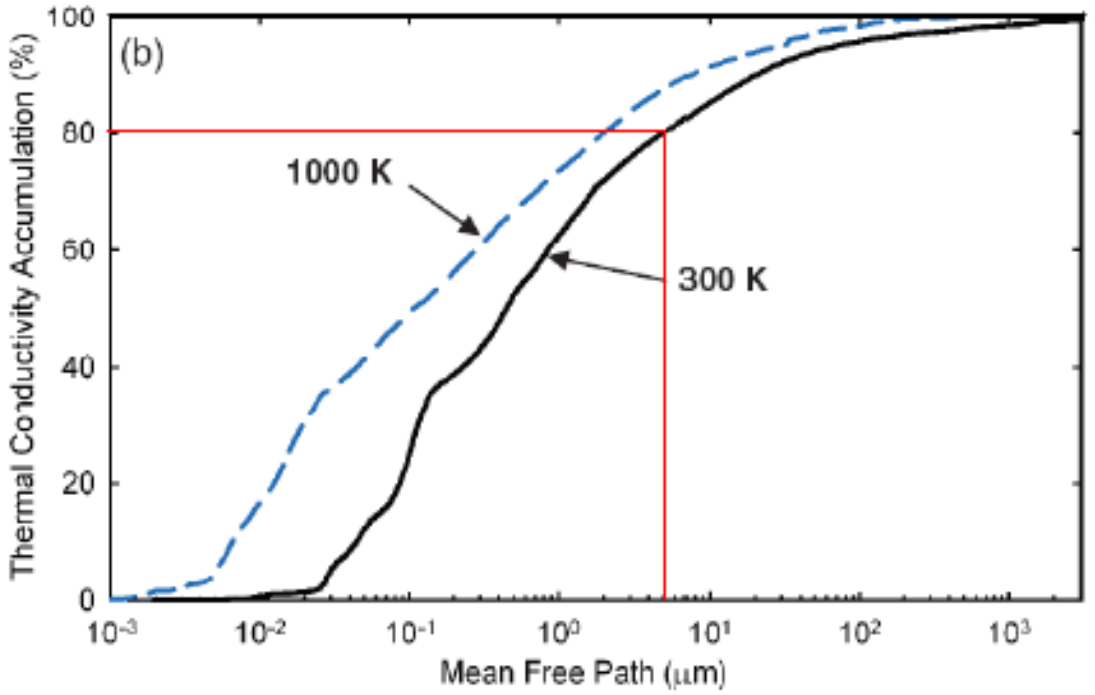
Message 1 : wide distribution of mean free paths

Message 2 : kinetic theory severely underestimates mean free paths

Accumulation function : phonon spectroscopy

MD simulations Si, Henry and Chen

$$acc(\omega) = \int_0^{\omega} c_v(\omega) v_g(\omega) \Lambda(\omega) d\omega$$



Henry and Chen, J. Comp. Theo. Nanosci 5, 1 (2008)

Message 1 : wide distribution of mean free paths

Message 2 : kinetic theory severely underestimates mean free paths

Message 3 : boundary scattering reduces significantly conductivity

Outline

I. Introduction - phenomenology

II. Vibrations

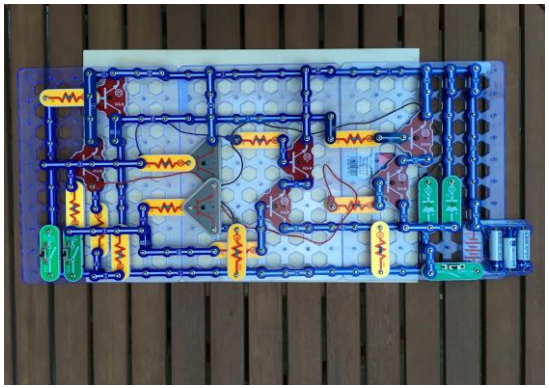
III. Thermal conductivity

IV. Phonon spectroscopy

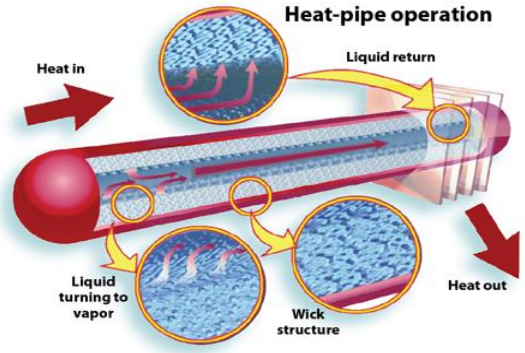
V. Phonon scattering at interfaces

Importance of interface phonon scattering

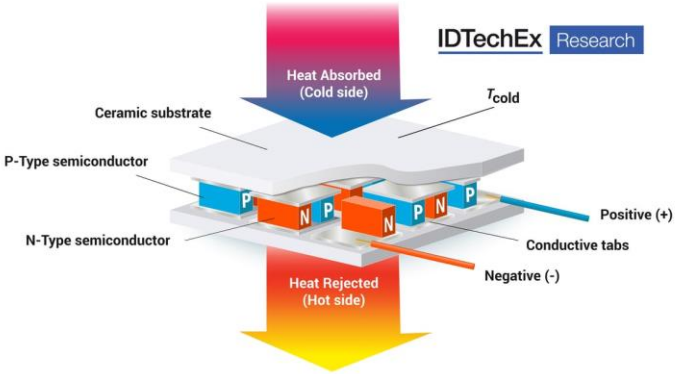
Logic circuits



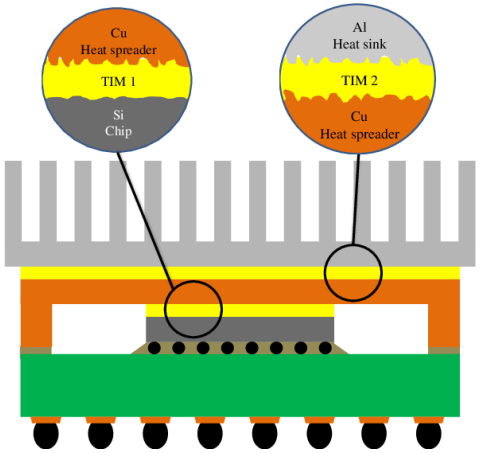
Liquid-solid coolers

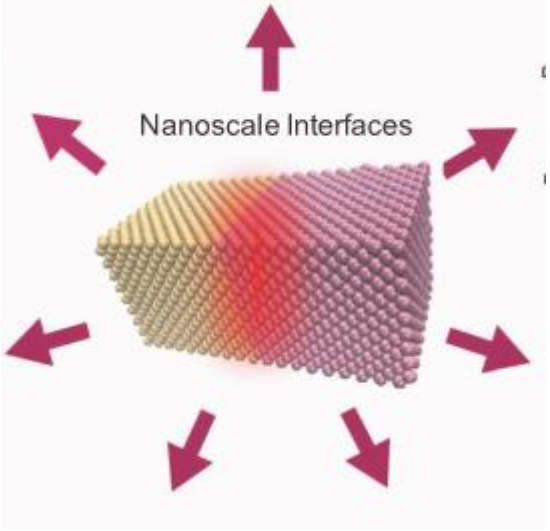


Thermoelectric devices

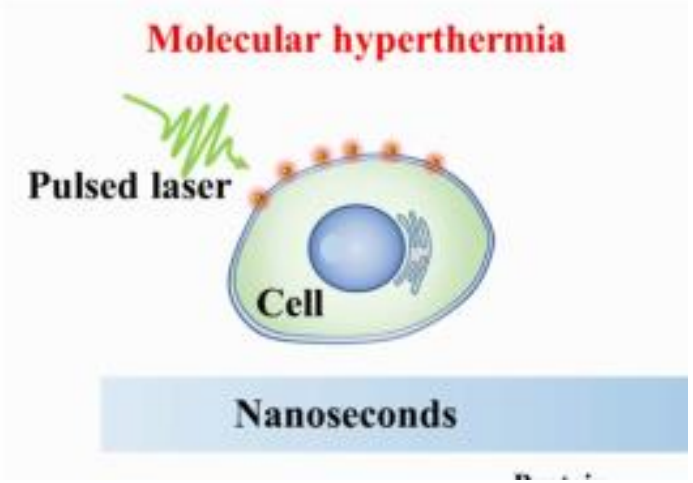


Thermal management of laptops

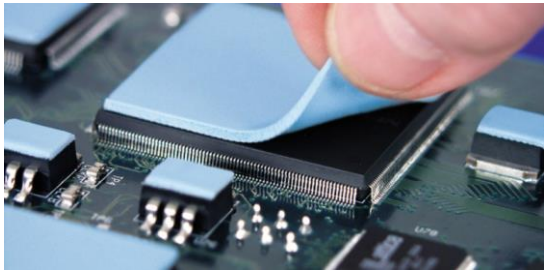




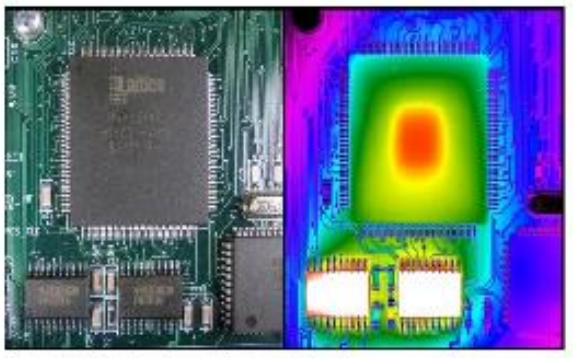
Nanoparticle based phototherapies



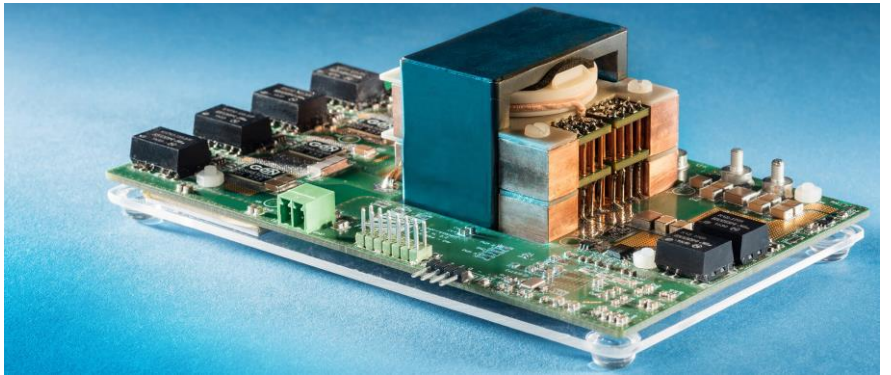
Thermal interface materials



Thermal management hot spots



High power electronics

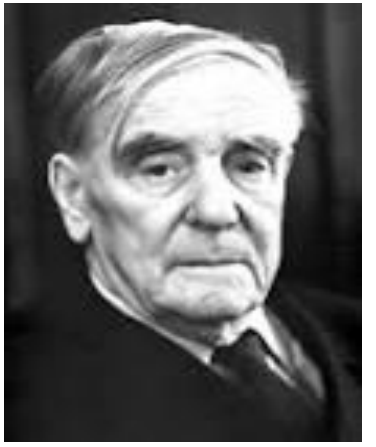


Thermal Boundary Resistance (TBR)
Kapitza resistance

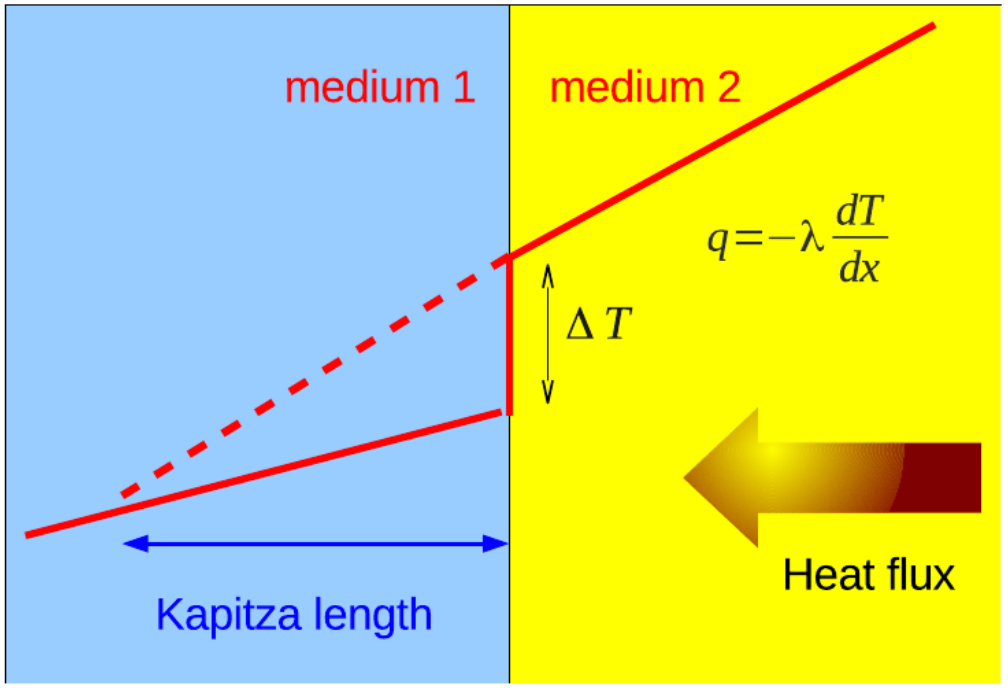
1941 : Kapitza's measurements of the temperature drop near the boundary liquid helium/solid

Kapitza resistance :

$$R = \frac{1}{G} = \frac{(T_2 - T_1)}{q}$$



Kapitza



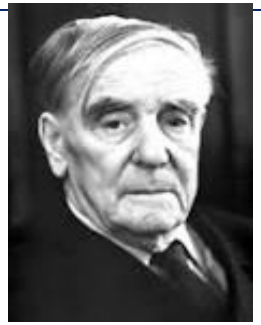
Kapitza

$$l_K = \lambda / G$$

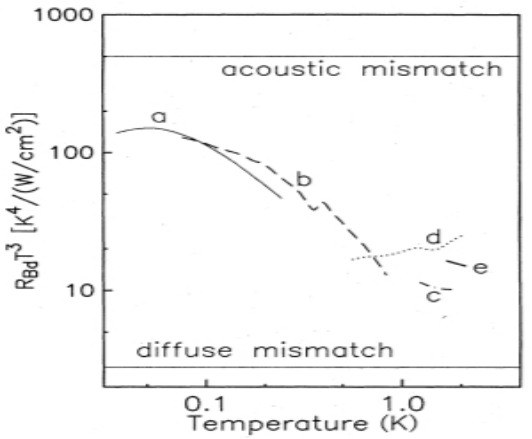
l_K ~ 1 -100 nm

Some elements of history

First measurements Helium 4-metal
Kapitza 40's



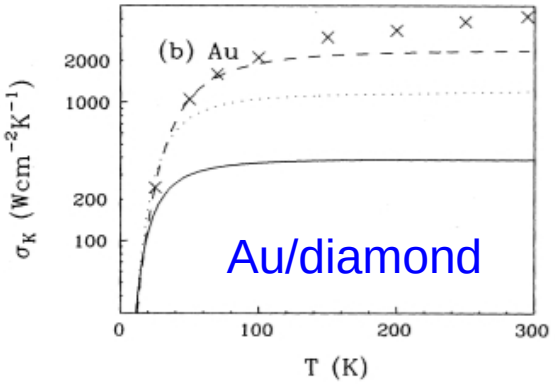
Kapitza, P. L., 1941, "The study of heat transfer in helium II," Zh. Eksp. Teor. Fiz. **11**, 1 [J. Phys. (USSR) **4**, 181 (1941)]; also in *Collected Papers of P. L. Kapitza Vol. 2*, edited by D. ter Haar (Pergamon, Oxford, 1965), p. 581.



Metal-solid interfaces

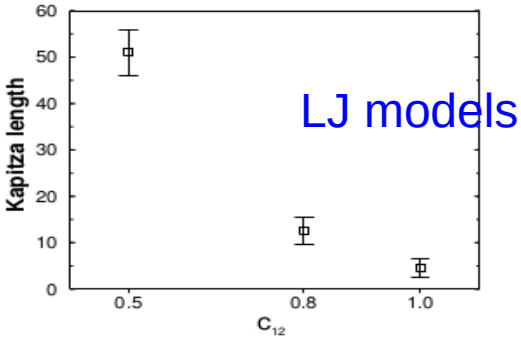
Stoner and Maris, *Phys. Rev. B (*1993)

Blowing activity in the 2000's : Norris, Hopkins, Cahill, etc



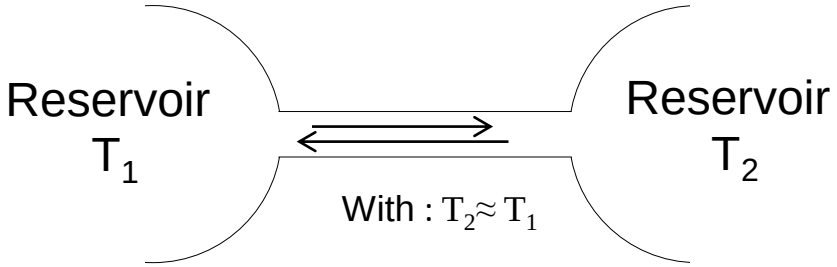
Liquid-solid interfaces

Barrat and Chiaruttini, *Mol. Phys.* (2003)
Xe and Keblinski, *J. Chem. Phys. (*2003)



Computational methods

Landauer formalism : Transport is viewed as a transmission process between two reservoirs at equilibrium.



$$G_{\mathrm{eq}} = \frac{1}{V} \sum_{p,\vec{k}}^{+} \hbar \omega v_{1x} t_{12} \frac{\partial f_{\mathrm{eq}}}{\partial T}.$$

equilibrium distribution function

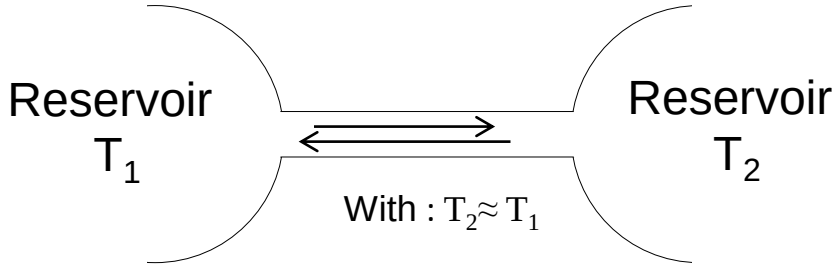
Non equilibrium effects :

Landry and McGaughey, Phys. Rev. B (2009)

Merabia and Termentzidis, Phys. Rev. B (201

Computational methods

Landauer formalism : Transport is viewed as a transmission process between two reservoirs at equilibrium.



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equilibrium distribution function

Non equilibrium effects :

Landry and McGaughey, Phys. Rev. B (2009)
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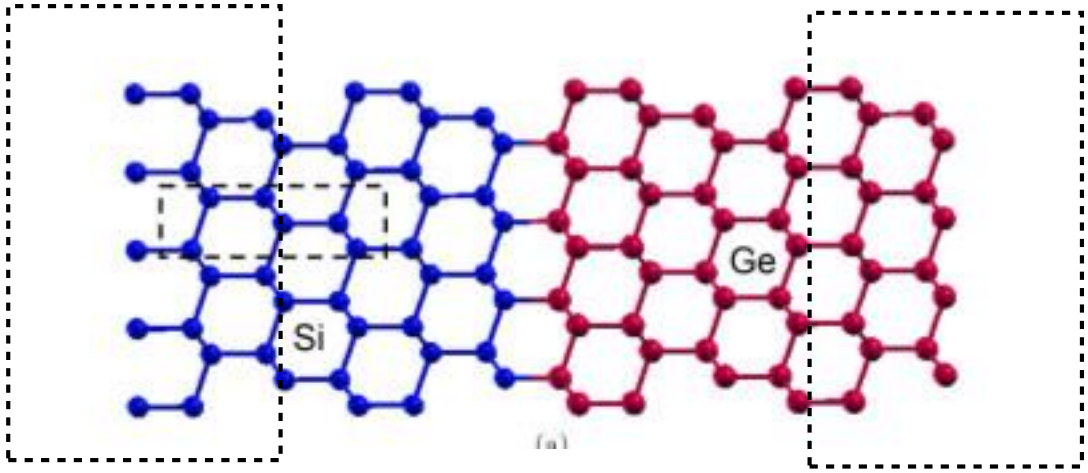


-Phonon NEGF
(Non equilibrium Green's functions

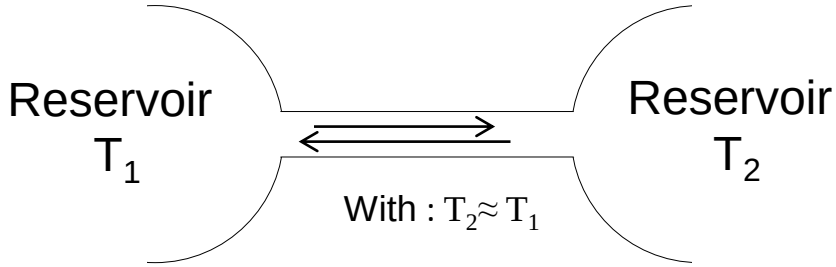
$$t_{12}(\omega) = Tr\,(\Gamma_1 G\, \Gamma_2\, G^{+})$$

$$G(\omega) = (\omega^2 I - H_d - \mathrm{T}\Sigma_1 - \Sigma_2)^{-1}$$

Mingo and Yang, Phys. Rev. B (2003)
Guo et al., Phys. Rev. B (2020)

Computational methods

Landauer formalism : Transport is viewed as a transmission process between two reservoirs at equilibrium.

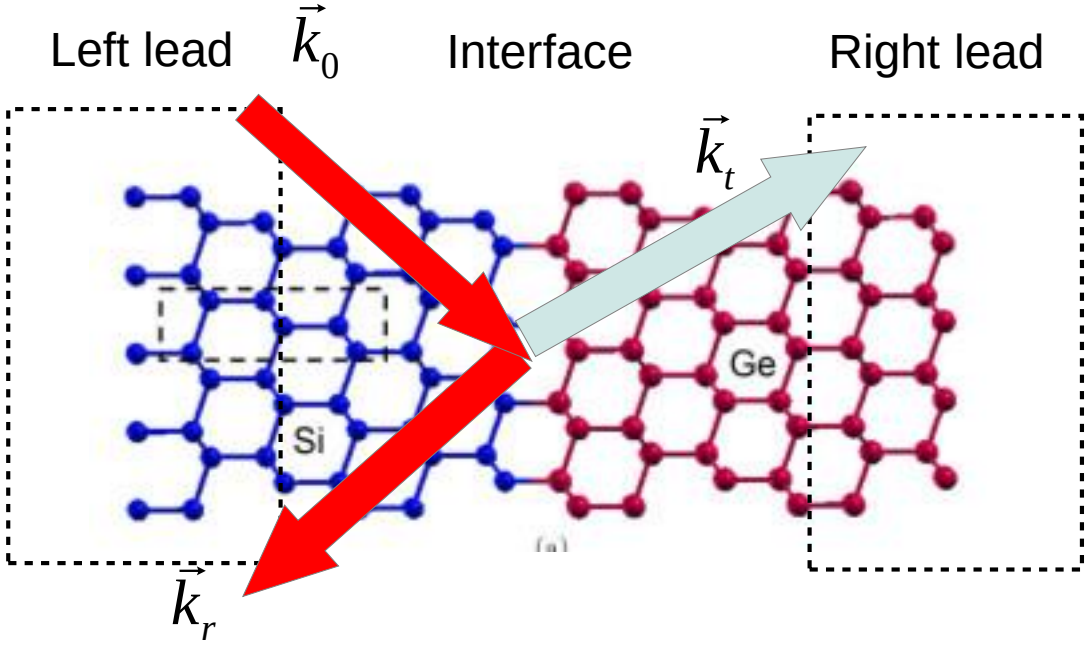


$$G_{\mathrm{eq}} = \frac{1}{V} \sum_{p,\vec{k}}^{+} \hbar \omega v_{1x} t_{12} \frac{\partial f_{\mathrm{eq}}}{\partial T}.$$

equilibrium distribution function

Non equilibrium effects :

Landry and McGaughey, Phys. Rev. B (2009)
Merabia and Termentzidis, Phys. Rev. B (201



Stoner and Maris, Phys. Rev. B (1993)
Zhao and Freud, J. Appl. Phys. (2005)
Alkurdi and Merabia, Appl. Phys. Lett. (2017)

-Phonon NEGF (Non equilibrium Green's functions

$$t_{12}(\omega) = Tr\,(\Gamma_1 G\, \Gamma_2\, G^{+})$$

$$G(\omega) = (\omega^2 I - H_d - \mathrm{T}\Sigma_1 - \Sigma_2)^{-1}$$

Mingo and Yang, Phys. Rev. B (2003)
Guo et al., Phys. Rev. B (2020)

-Lattice dynamics

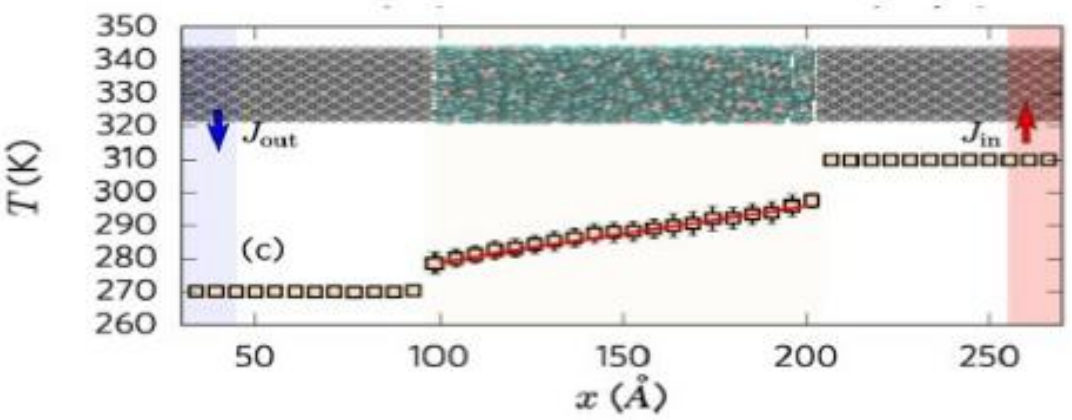
$$\left(\Phi^{AA}_{\alpha,\beta} - m_A \omega^2 \delta_{\alpha,\beta}\right) u^A_\beta + \Phi^{AB}_{\alpha,\beta} u^B_\beta = 0$$

$$\Phi^{AB}_{\alpha,\beta} u^A_\beta + \left(\Phi^{BB}_{\alpha,\beta} - m_B \omega^2 \delta_{\alpha,\beta}\right) u^B_\beta = 0$$

Molecular dynamics

Steady state non equilibrium simulations

$$R = \frac{1}{G} = \frac{(T_2 - T_1)}{q}$$



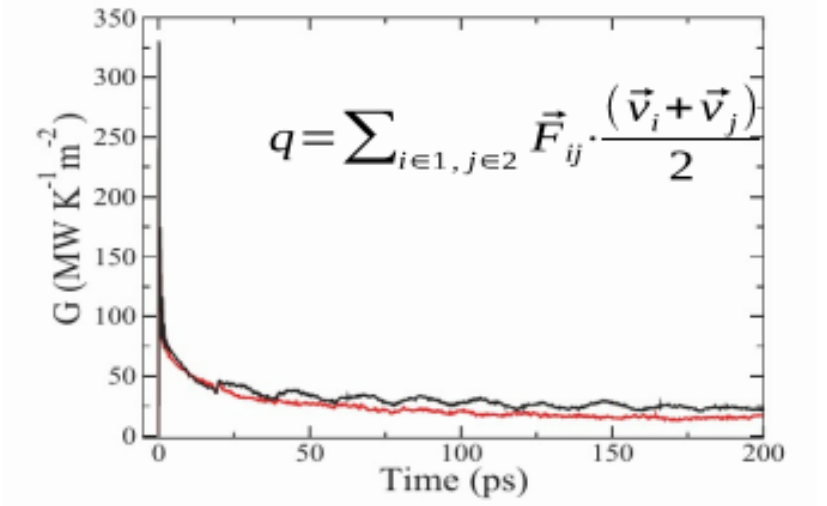
Barrat and Chiaruttini, *Mol. Phys.* (2003)
Landry and McGaughey, Phys. Rev. B (2009)
Merabia and Termentzidis, *Phys. Rev. B* (2012)

Green-Kubo simulations

$$G = \frac{1}{Ak_BT^2}\int_0^{+\infty} \langle q(t)\cdot q(0)\rangle\, dt$$

Analogous to the Green-Kubo formula for the thermal conductivity

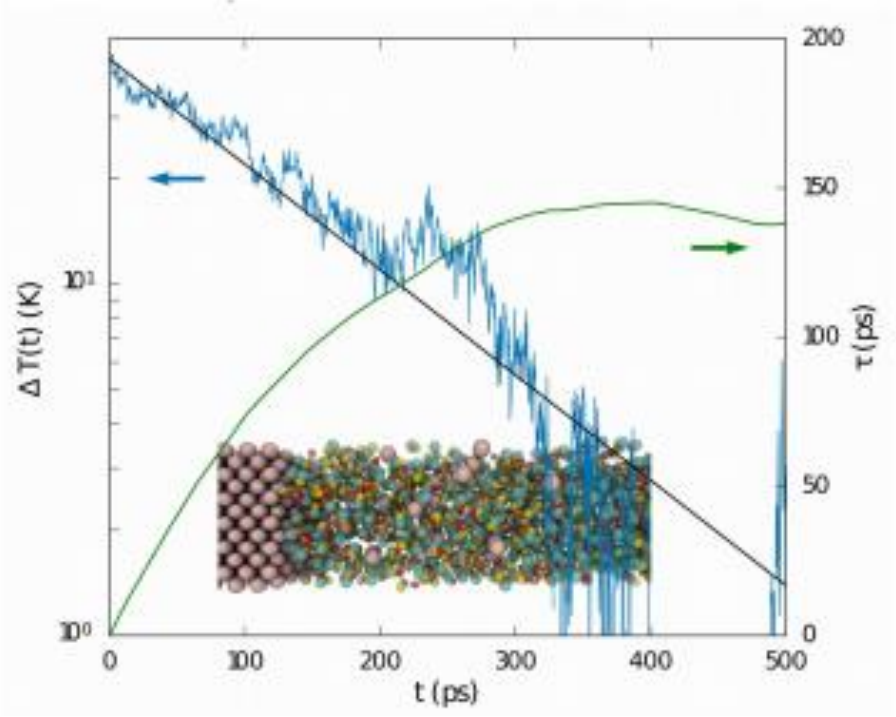
$$G = R = \frac{\mathrm{A}}{k_BT^2}\int_0^{+\infty} \langle T(t)\cdot\ T\ (0)\rangle\, dt$$



Barrat and Chiaruttini, *Mol. Phys.* (2003)
Merabia and Termentzidis, *Phys. Rev. B* (2012)
Chalopin et al., *Phys. Rev. B* (2012)
Rajabpour et al., *J. Chem. Phys.* (2019)

Transient simulations

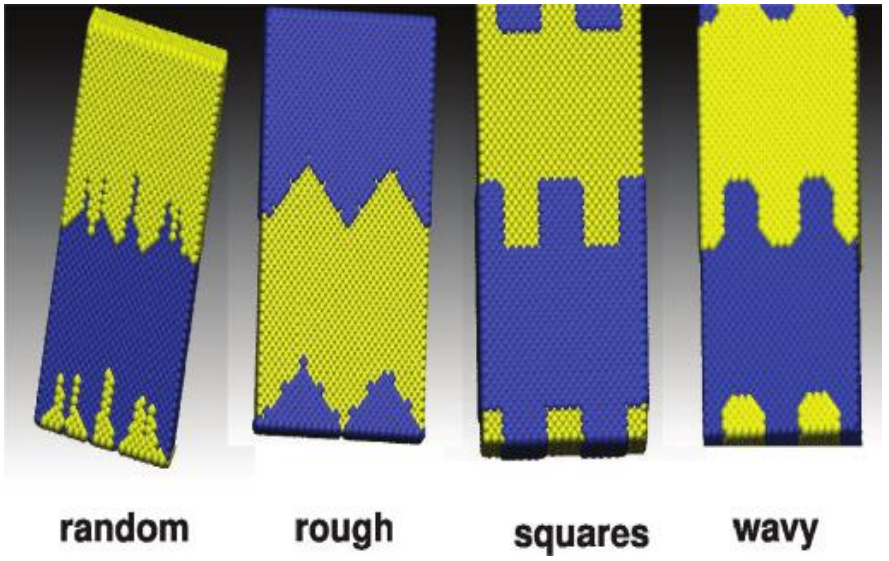
Temperature relaxation



$$\Delta T(t) \propto \exp(-t/\tau)$$

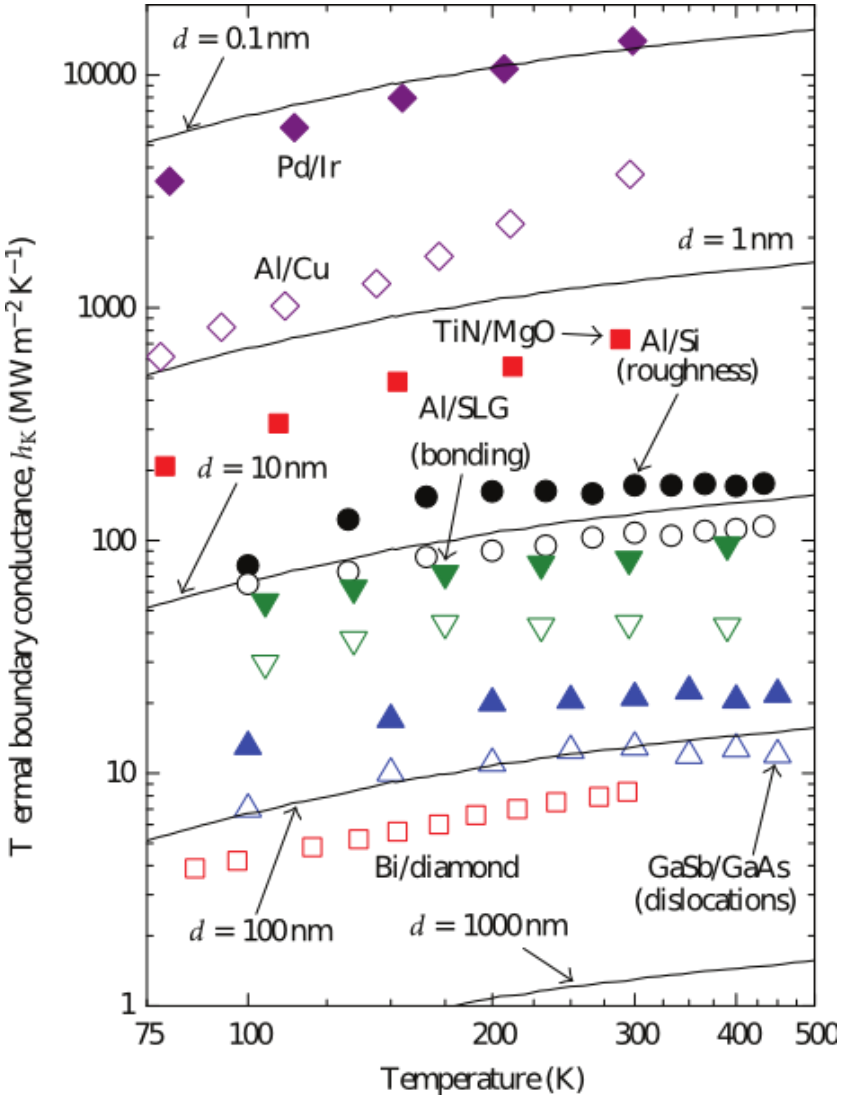
$$G = C_v/(A\tau)$$

Well adapted to irregular interfaces !

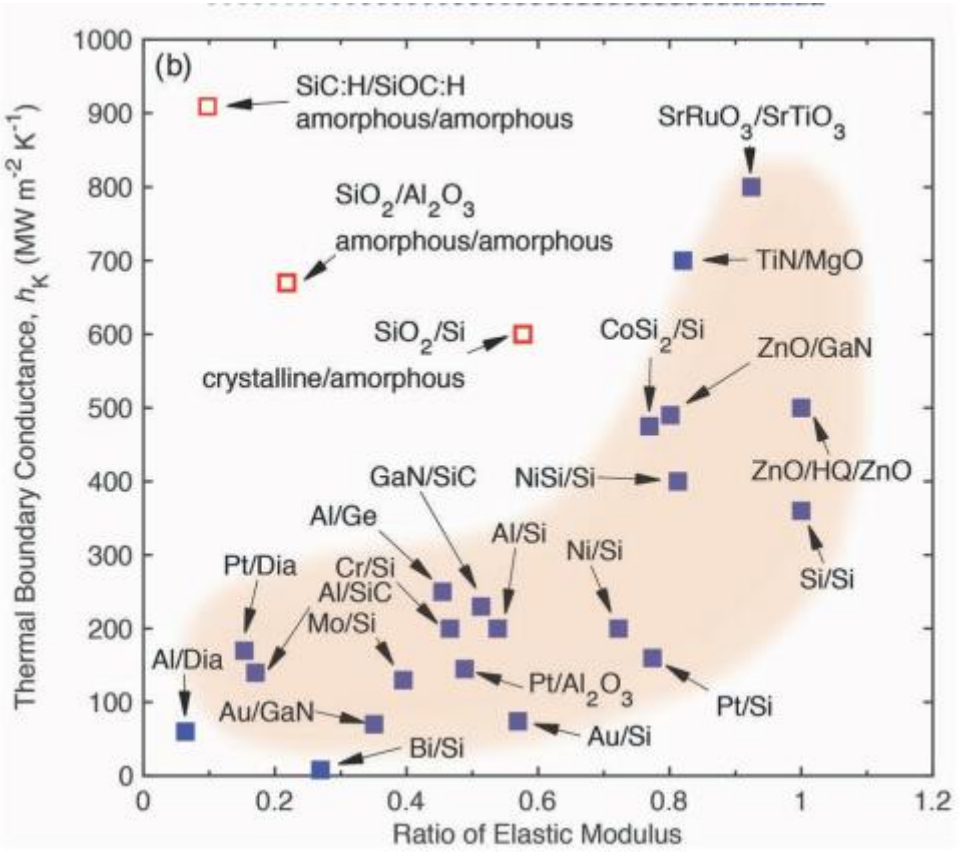


S. Shenogin et al., *J. Appl. Phys.* 2004
E. Lampin et al., *Appl. Phys. Lett.* 2012
S. Merabia and Termentzidis, *Phys. Rev. B* 2014
H. Han, S. Merabia, F. Müller-Plathe, *Nanoscale* 2017

Effects influencing thermal boundary conductance



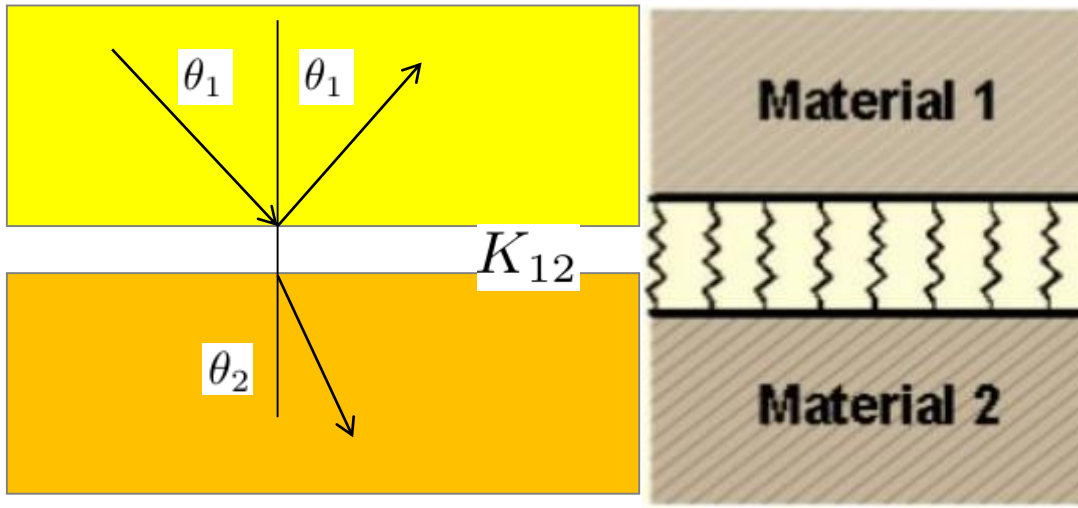
Hopkins, *ISRN Mech. Eng* (2013)



Giri and Hopkins, *Adv. Func. Mat*. (2019)

Acoustic Mismatch Model (AMM)

- Continuum acoustics
- Acoustic impedances
- Specular scattering



Phonon transmission :

$$t_{12}(\theta_1, \theta_2) = \frac{4Z_1 Z_2 \cos\theta_1 \cos\theta_2}{(Z_1 \cos\theta_1 + Z_2 \cos\theta_2)^2}$$

Khalatnikov (1952)

Generalized AMM model :

$$t_{12}\,(\theta_1, \theta_2) = \frac{4Z_1 Z_2 \cos\theta_1 \cos\theta_2}{(Z_1 \cos\theta_1 + Z_2 \cos\theta_2)^2 + \frac{\omega^2}{K_{12}^2}(Z_1 Z_2 \cos\theta_1 \cos\theta_2)^2}$$

Interface spring stiffness : $K_{12} = \frac{d^2 V_{12}}{dr^2} = 72/2^{1/3} n_s\, \varepsilon_{12}/\sigma_{12}^2$

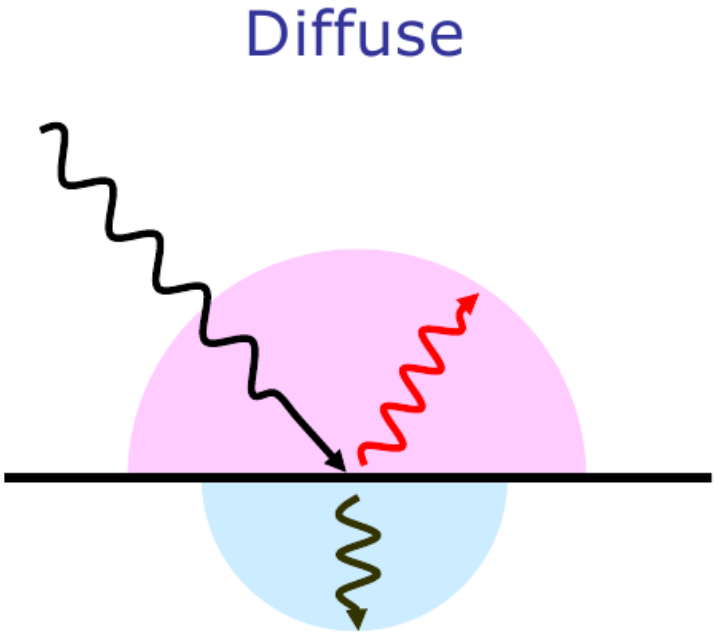
Prasher, *Appl. Phys. Lett.* 94 (2009) 041905
Merabia et al., *Int. Jour. Heat Mass Transf.* 100 (2016) 287

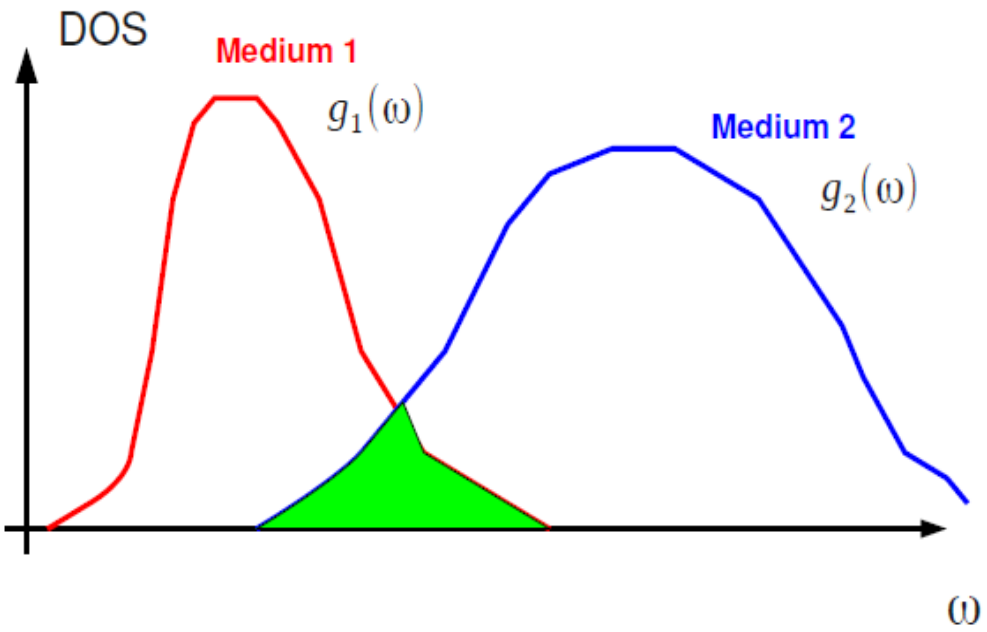
Diffuse Mismatch Model (DMM)

The transmission coefficient is given by detailed balance

Phonon transmission :

$$t_{12}(\omega) = \frac{c_2 g_2(\omega)}{c_1 g_1(\omega) + c_2 g_2(\omega)}$$

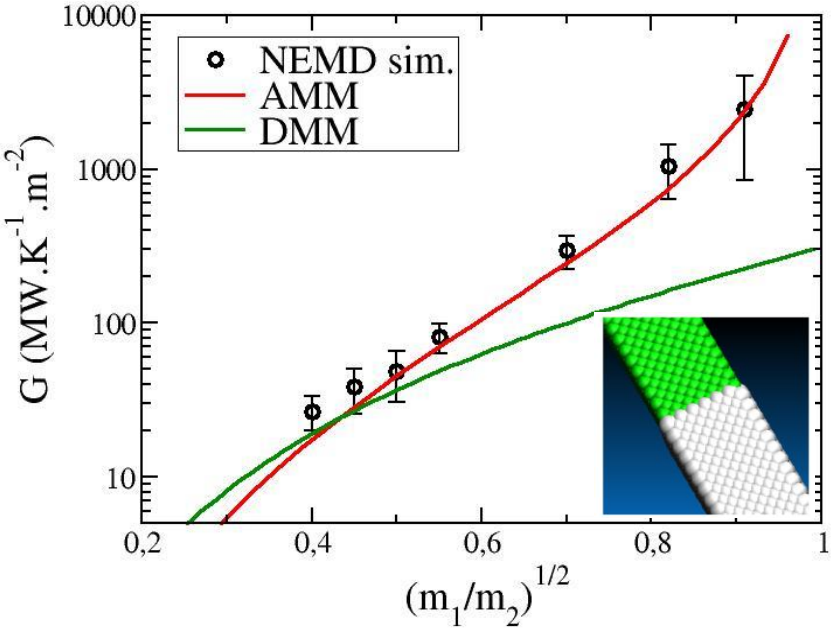




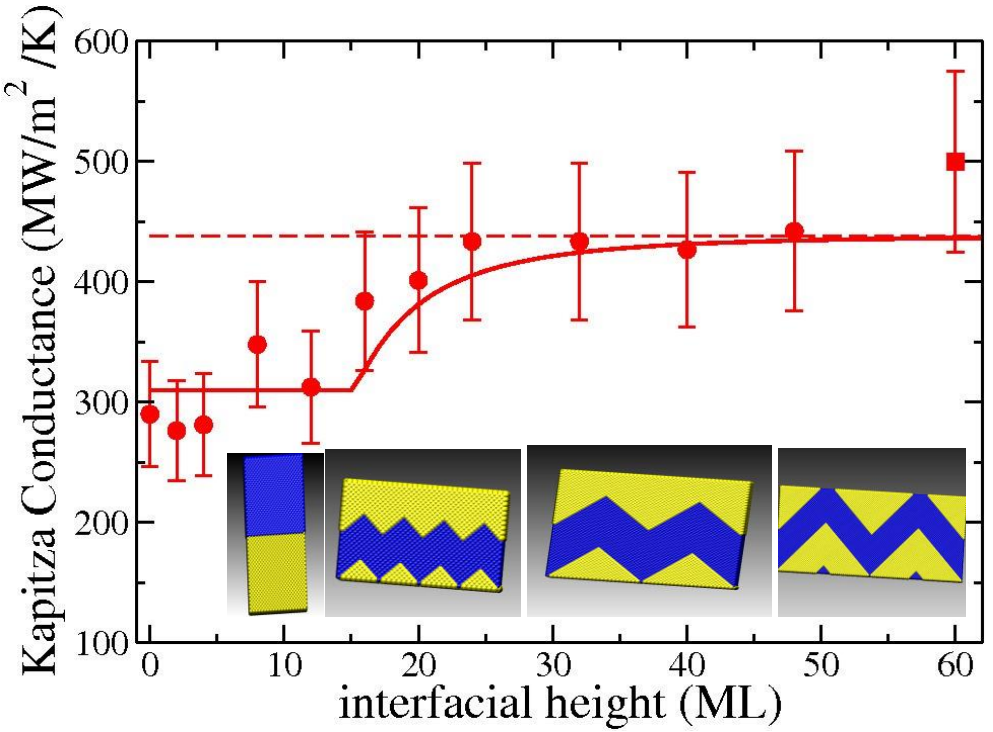
The transmission coefficient is given by detailed balance

Swartz et Pohl, *Appl. Phys. Lett.* (1987)

Comparison with AMM

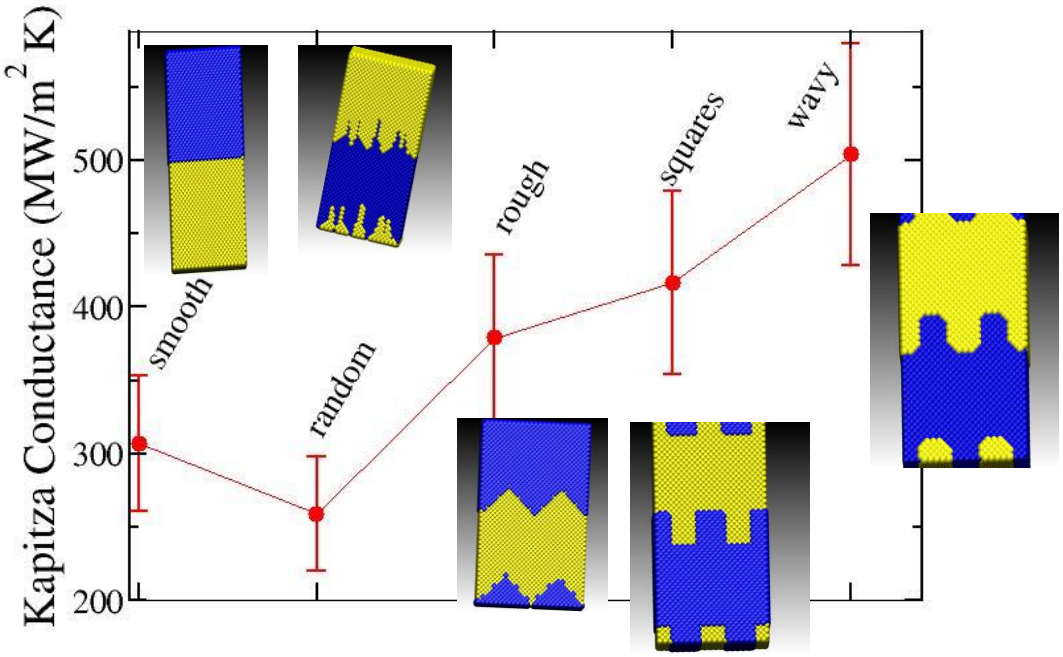


S. Merabia and K. Termentzidis,
Phys. Rev. B 2012



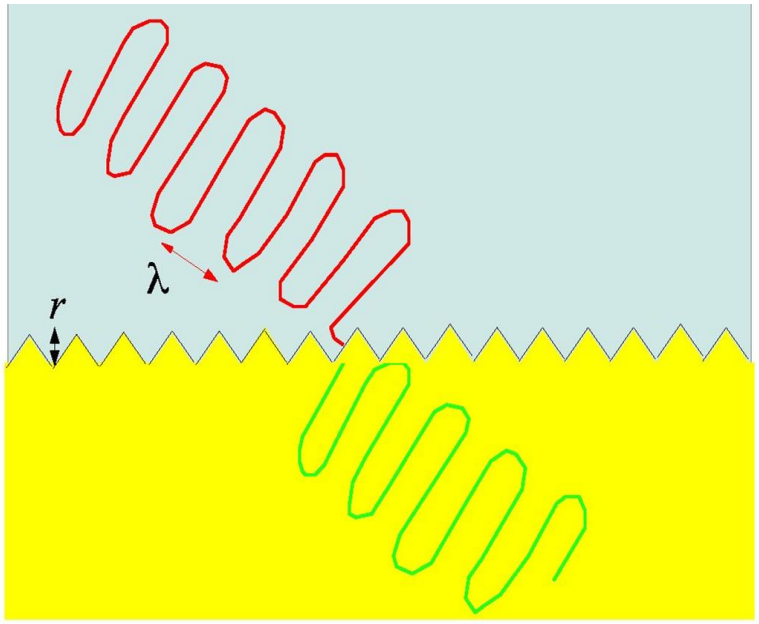
S. Merabia and K. Termentzidis,
Phys. Rev. B 2014

Effect of the roughness



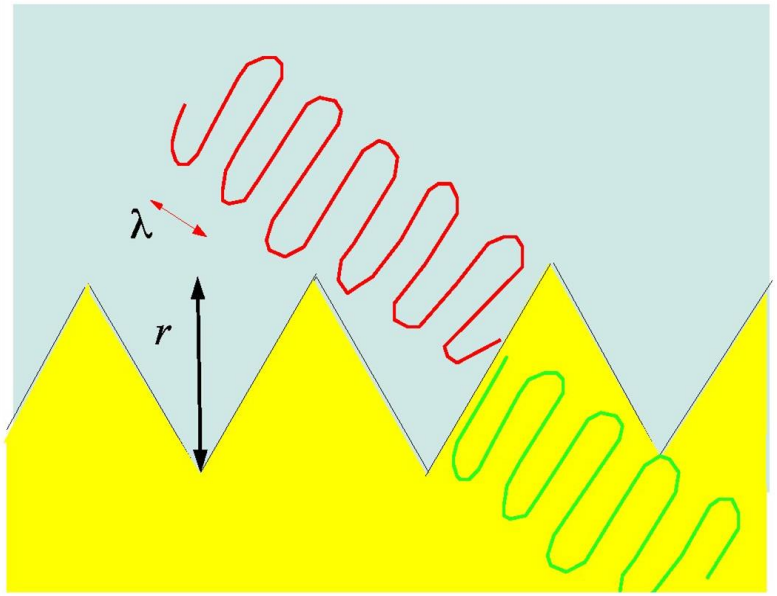
S. Merabia and K. Termentzidis,
Phys. Rev. B 2014

Small roughnesses



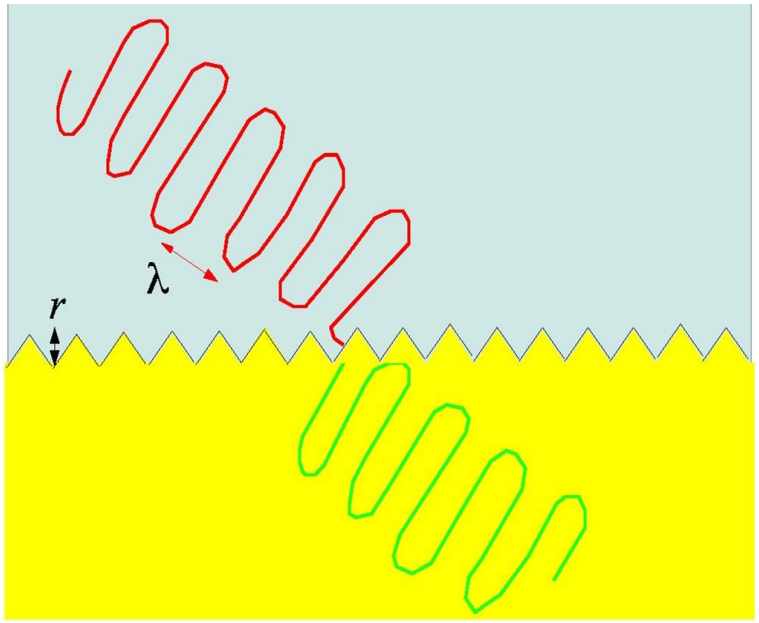
Heat flux propto the **projected** interfacial area

Large roughnesses



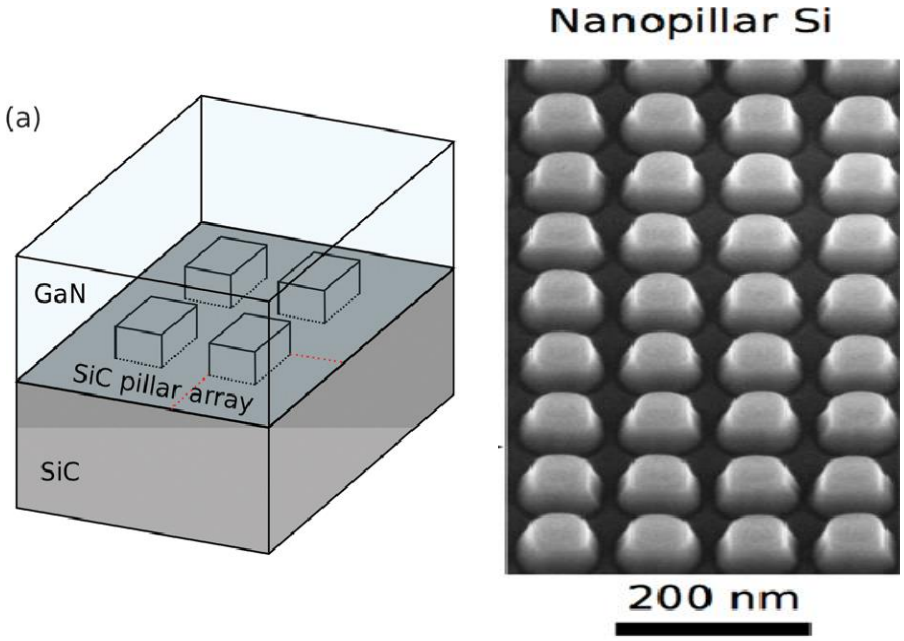
Heat flux propto the **true** interfacial area

Small roughnesses



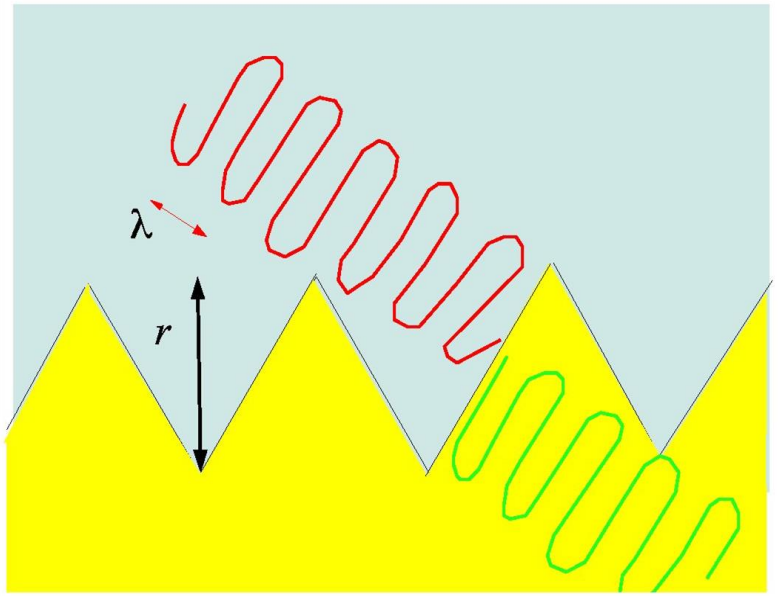
Heat flux propto the **projected i**nterfacial area

Experimental confirmation



Lee et *al.*, ACS ami (2016)

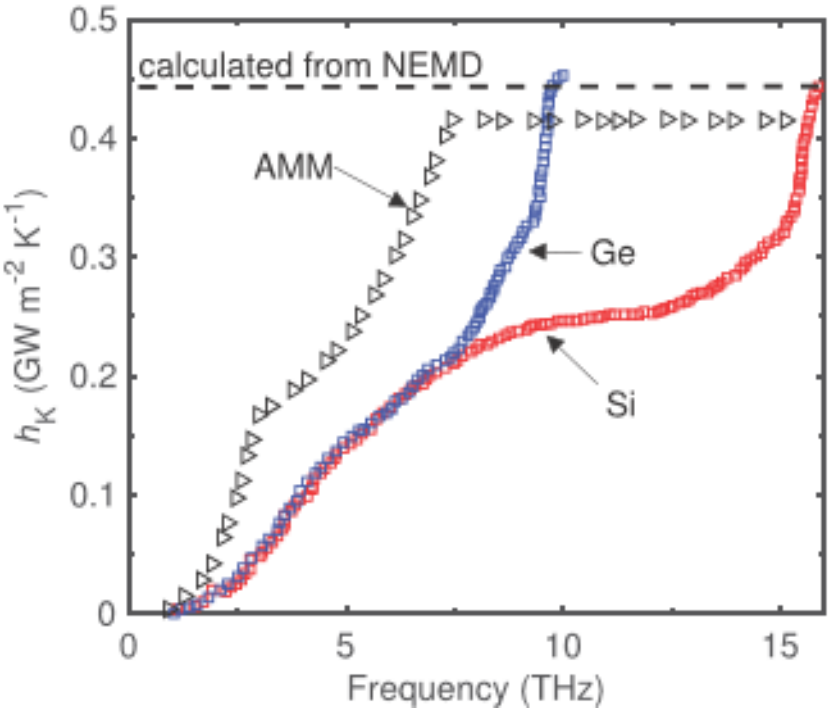
Large roughnesses



Heat flux propto the **true** interfacial area

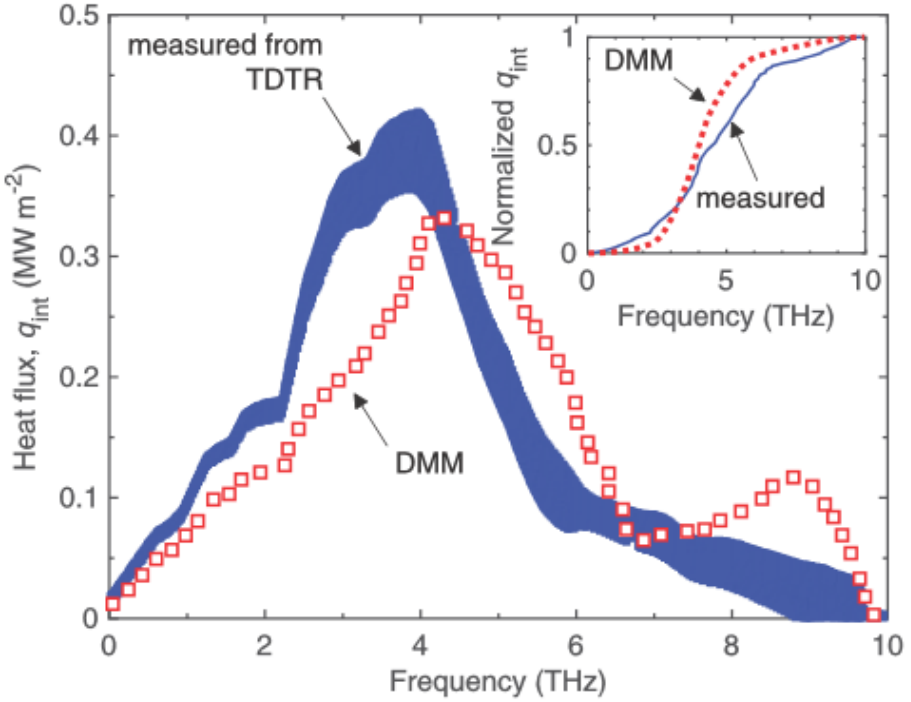
Comparison with the models : spectral transmission

MD system Si/Ge



T. Feng et al., *Phys. Rev B* 2019

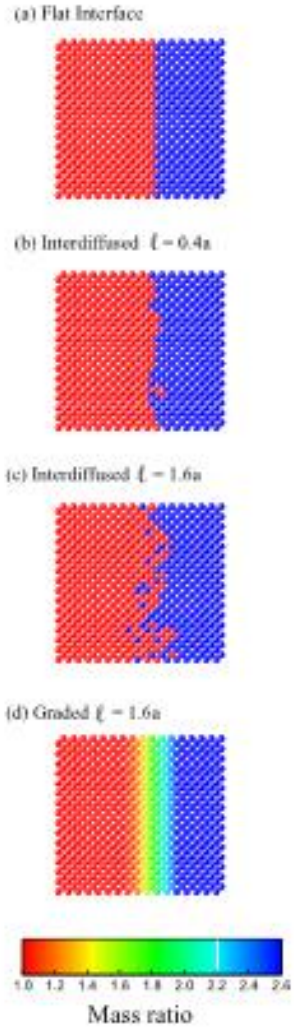
Experiments Al/Si

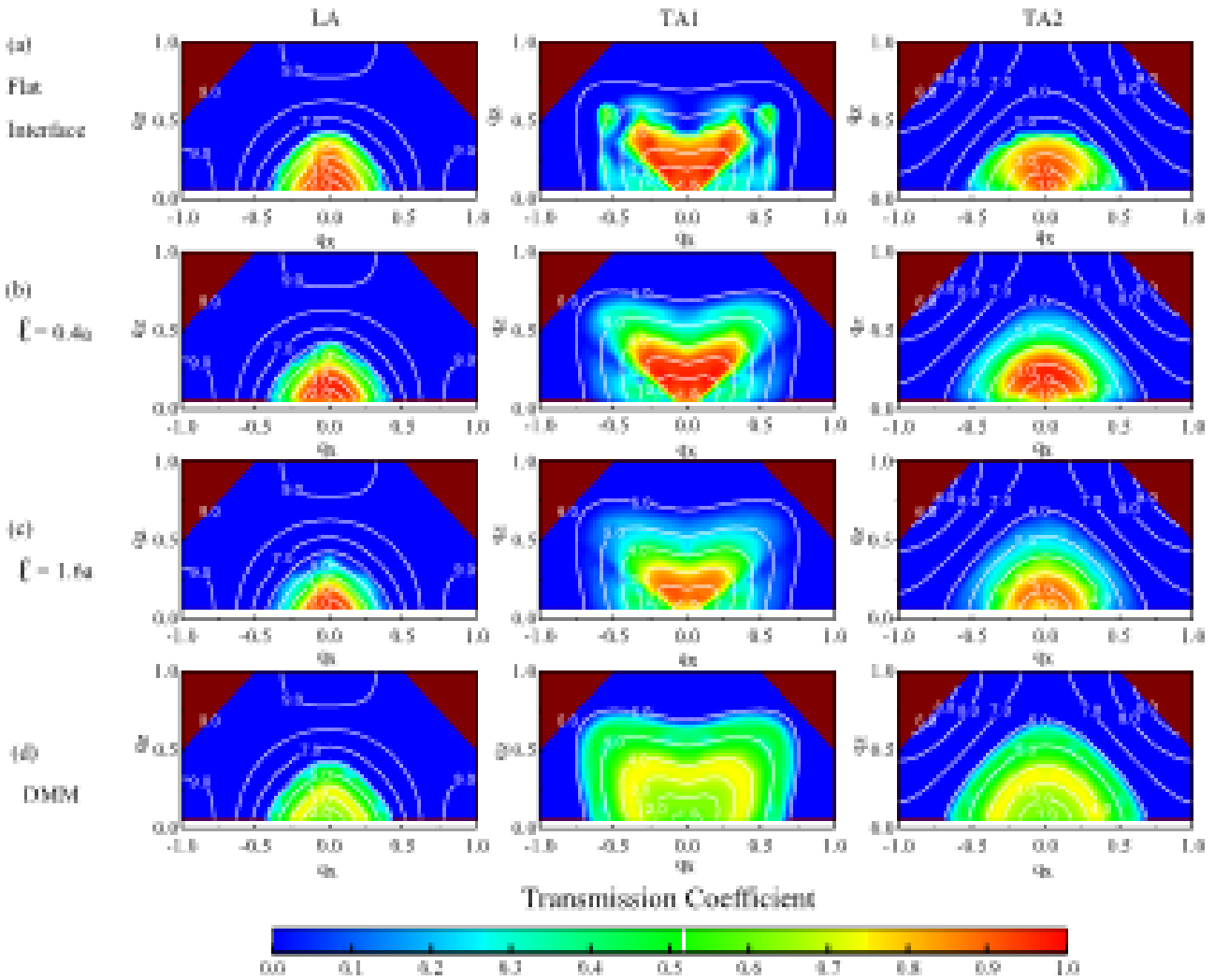


C. Hua et al., *Phys. Rev B* 2017

Comparison with DMM model

Highly disordered interfaces





Modes are not found to loose memory of their polarization and orientation

Kadlokar and Feser, J. *Appl. Phys.* 2017

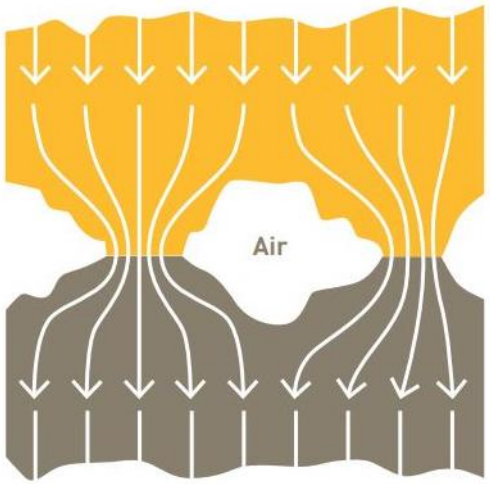
Comparison with the models: conclusions

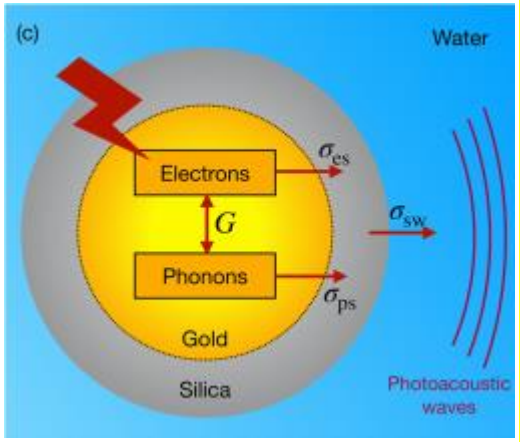
1. AMM and DMM models may provide a good description of the thermal boundary conductance

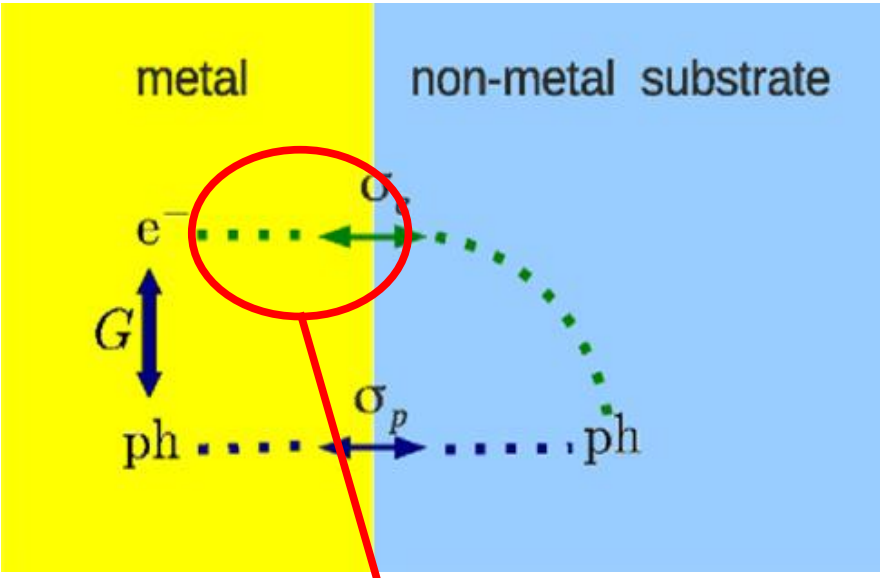
2. However, the spectral transmission is in general not in agreement with simulations/experiments

3. In conclusion, they are phenomenological models which may help understand the physics of phonon scattering. Accurate predictions should rather rely on atomistic models such as NEGF or lattice dynamics with ab-initio force constants.

Metal-semiconductor interfaces







Alkurdi, Lombard, Merabia, *Phys. Rev. Appl.* (2020)

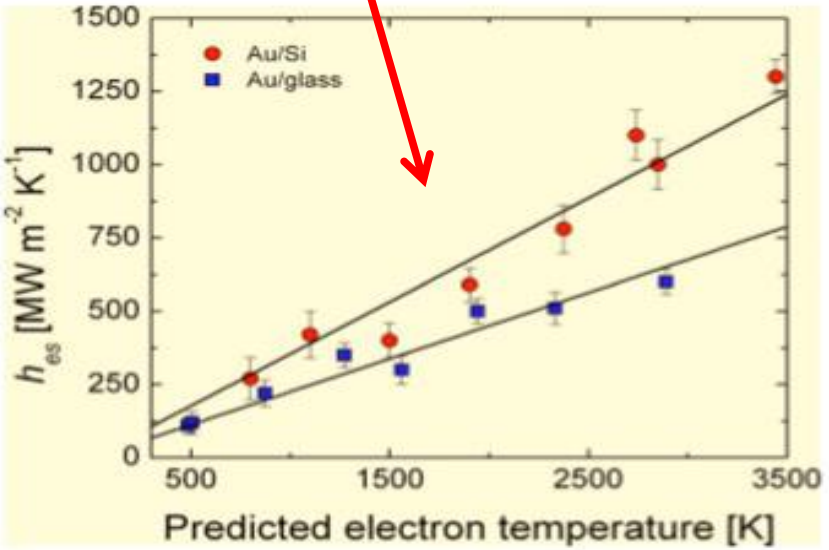
Metal electrons

Interfaces ?

Semi-conductors phonons

Measurements :

Hopkins and Norris. *J. Appl. Phys.* (2009)



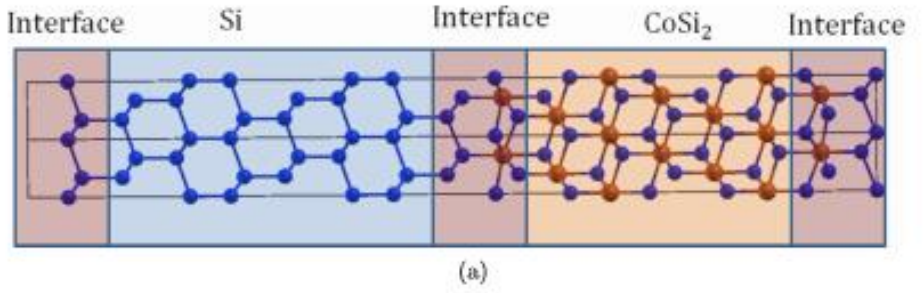
Theory :

Sergeev, *Phys Rev B* (1998)

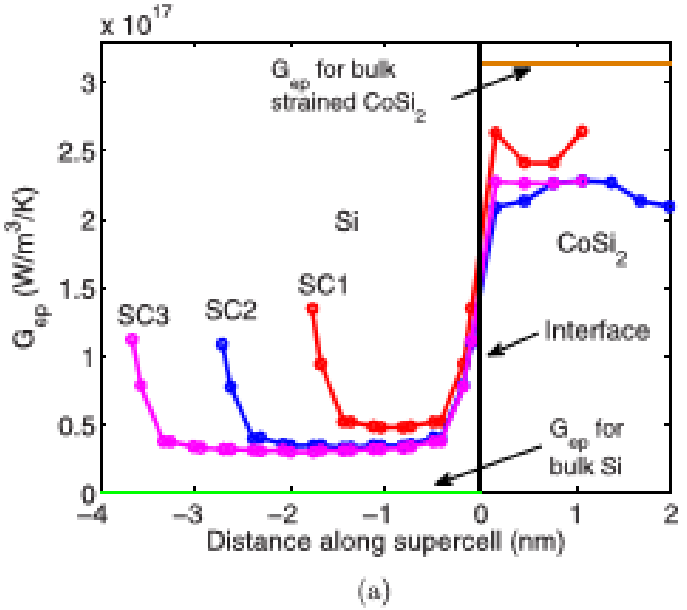
Mahan, *Phys Rev B* (2009)

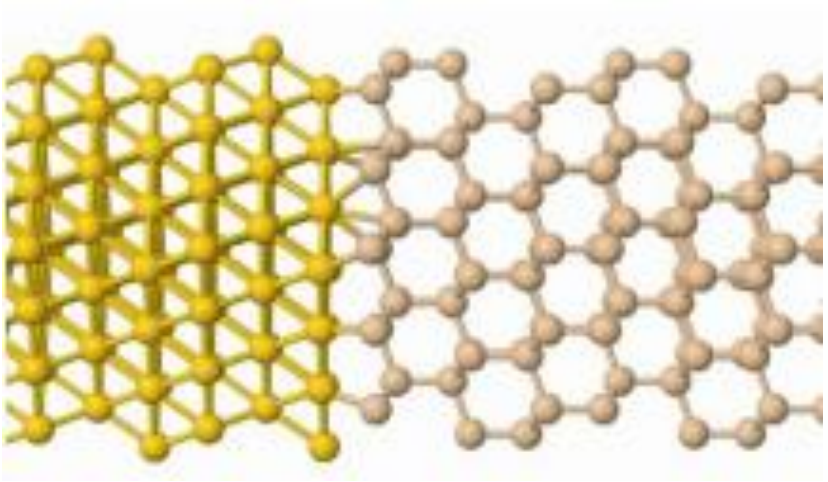
Lombard, Detcheverry, Merabia, *J. Phys. Cond. Mat.* (2015)

Electron-phonon interface thermal transport

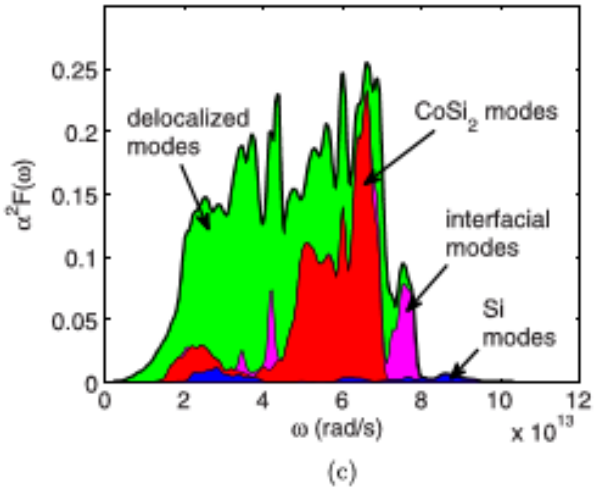


Sadasivam et al. Phys. Rev. B (2017)

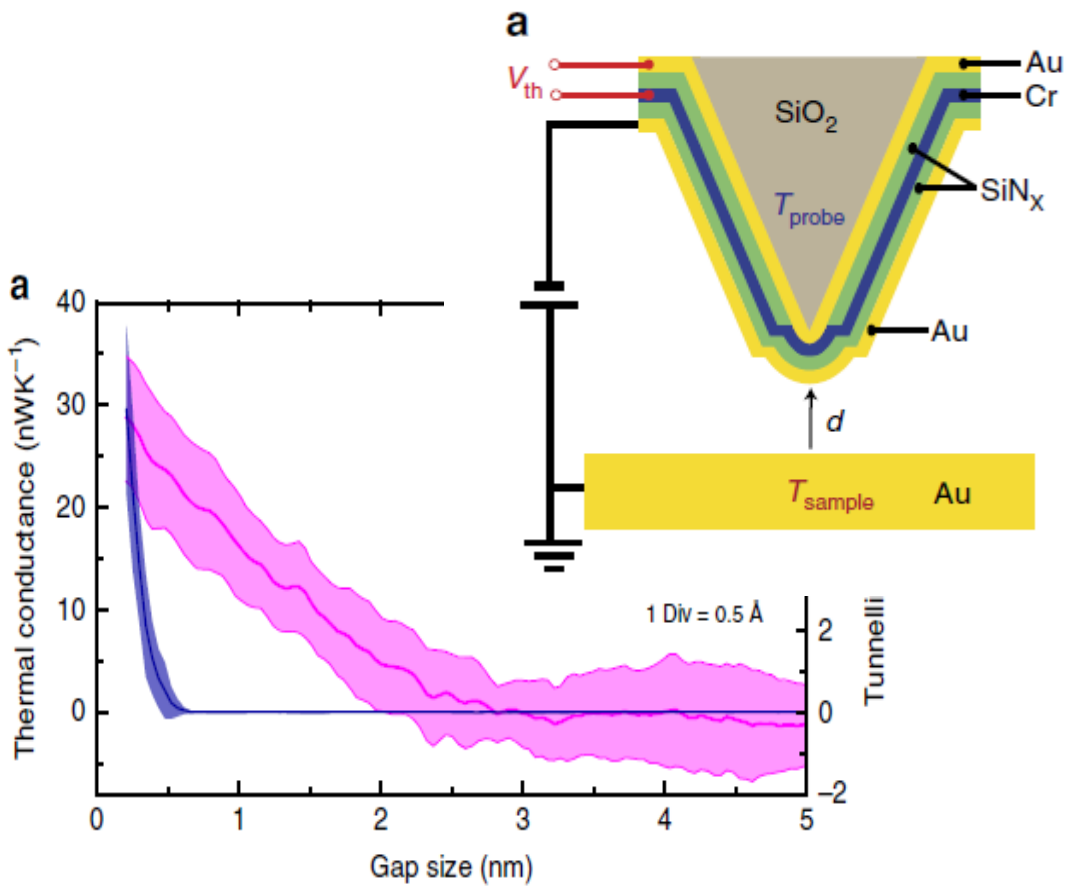


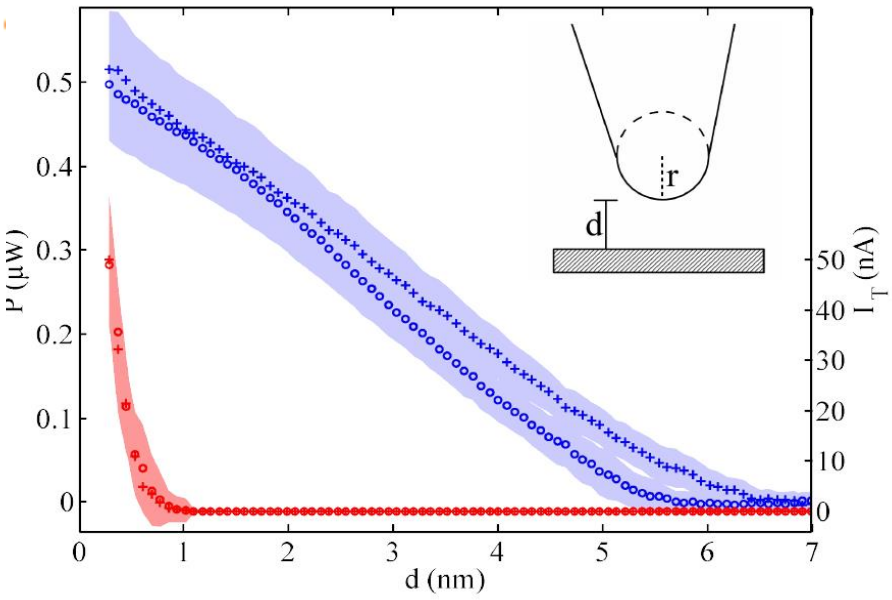


Work in progress (Adessi, De San Feliciano)



Thermal transport across nanoscale vacuum gaps





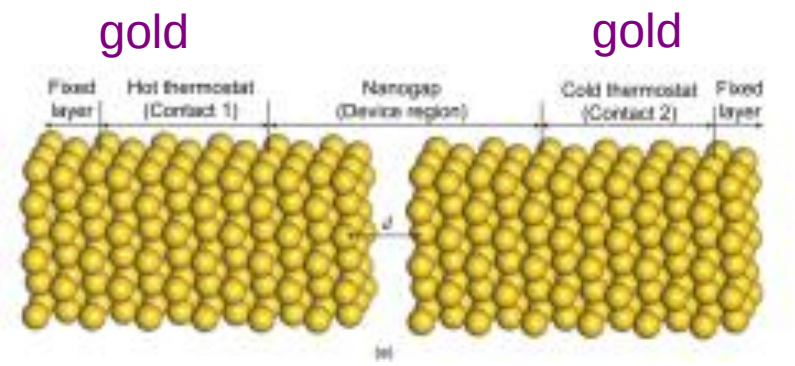
Good agreement with near field radiation theory

Cui et al., *Nat. Com.* (2017)

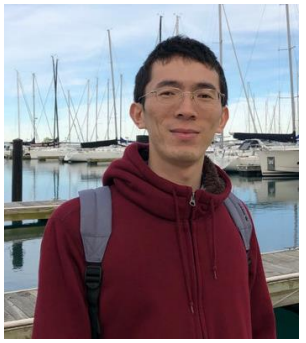
Three orders of magnitude difference with near field radiation theory

Kloppstech et al., *Nat. Com.* (2017)

Thermal transport across gold/vacuum gap/gold

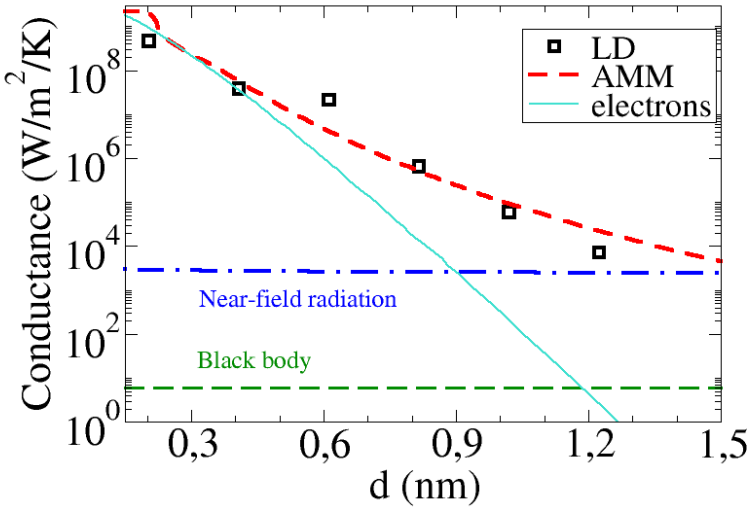


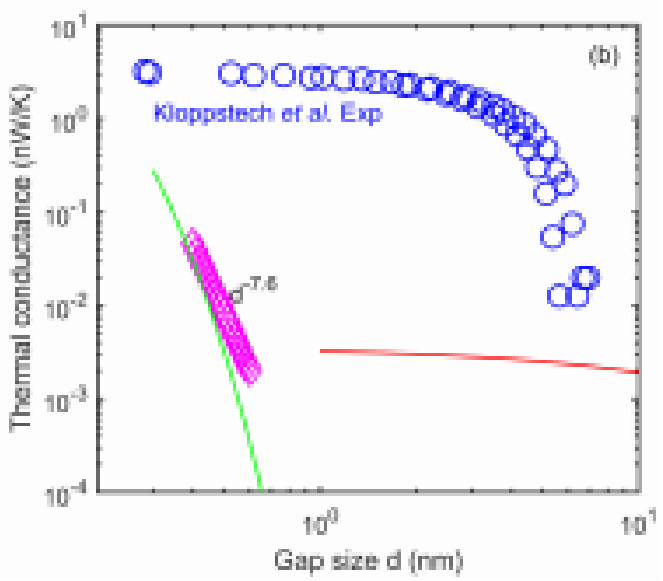
See Yangyu Guo's poster



MD + NEGF

Lattice dynamics

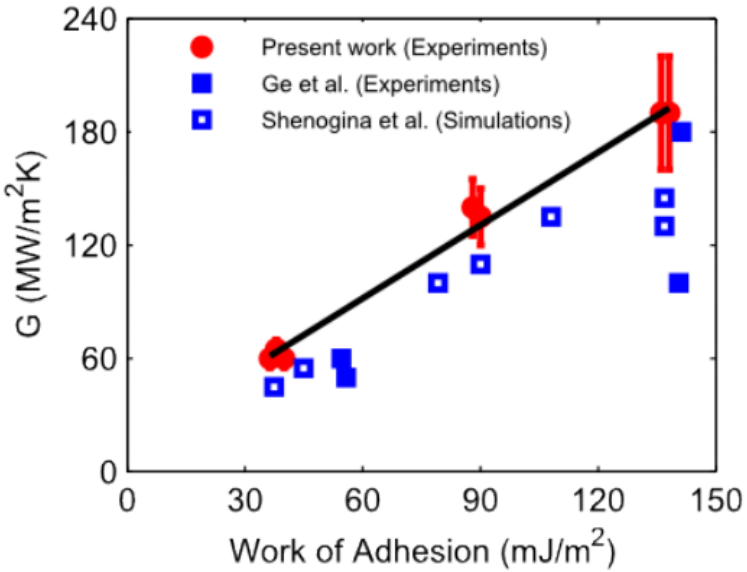


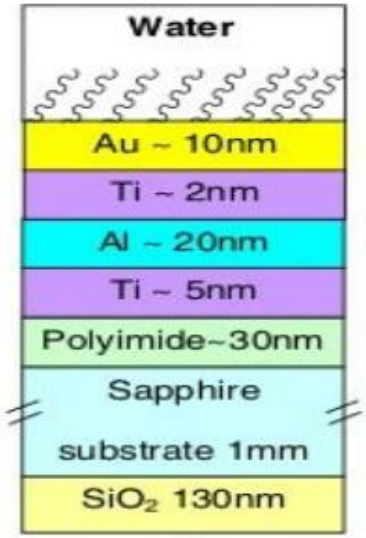


Phonons dominate thermal transport for subnanometer gaps

Alkurdi, Adessi, Tabatabaei, Termentzidis, Merabia, *Int. Jour. Heat Mass Transf.* 158 (2020) 119963
Guo et al., in preparation

Interfacial heat transfer at solid-liquid interfaces

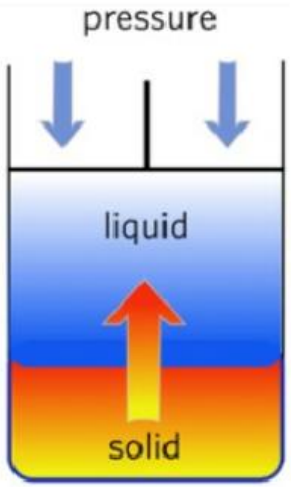


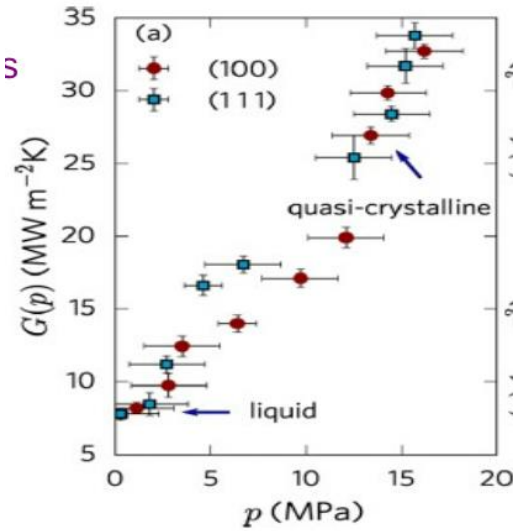


Effect of the wetting angle

Ge et al., *Phys. Rev. Lett.* 2006,
Huxtable, Chen, *Appl. Phys. Lett.* 2013,
Shenogin, Keblinski, Cahill,
Phys. Rev. Lett. 2009
Alvarado et al., *J. Phys. Chem. Lett.* 2016
Merabia, Lombard, Alkurdi, IJHMT 2016

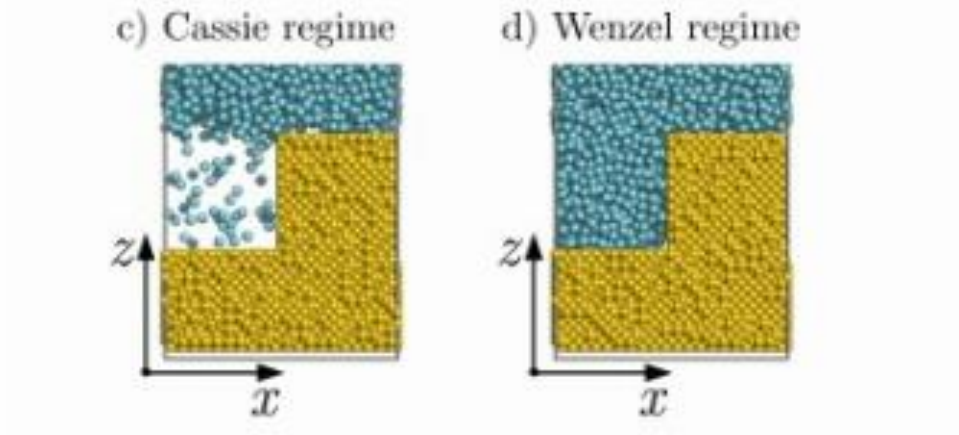
Effect of pressure





Han, Merabia, Müller-Plathe,
J. Phys. Chem. Lett. 2017

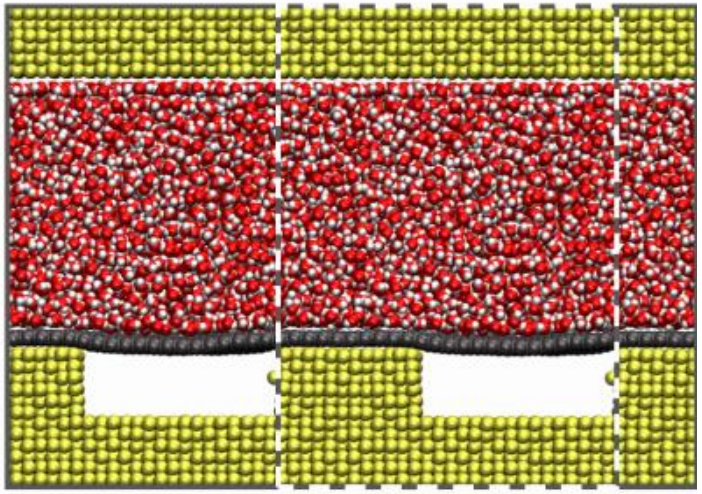
Effect of the roughness

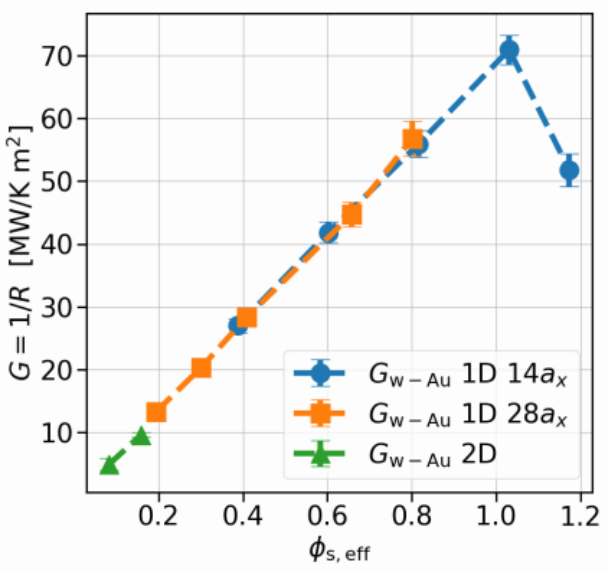


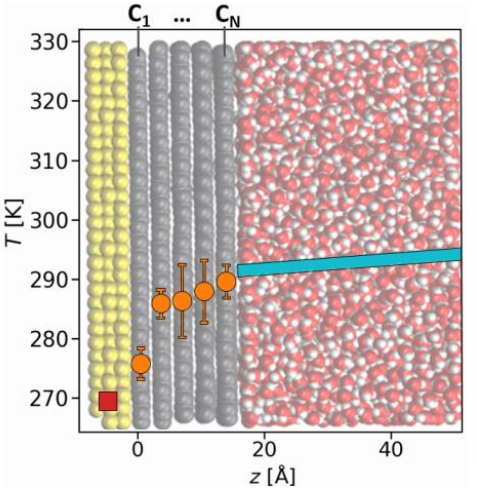
Wang and Keblinski, *Appl. Phys. Lett.* 2006,
Donatas, Ohara, *J. Chem. Phys.* 2019

Heat transport on nanostructured graphene

with C. Herrero and L. Joly





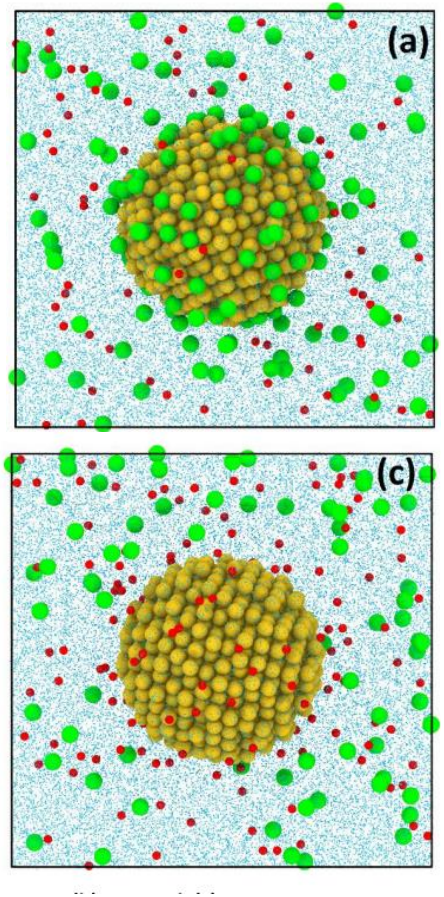


$$R_{w-Au} \simeq R_{C_2-C_1} + \frac{R_{C_1-Au}}{\phi_{s,eff}}.$$

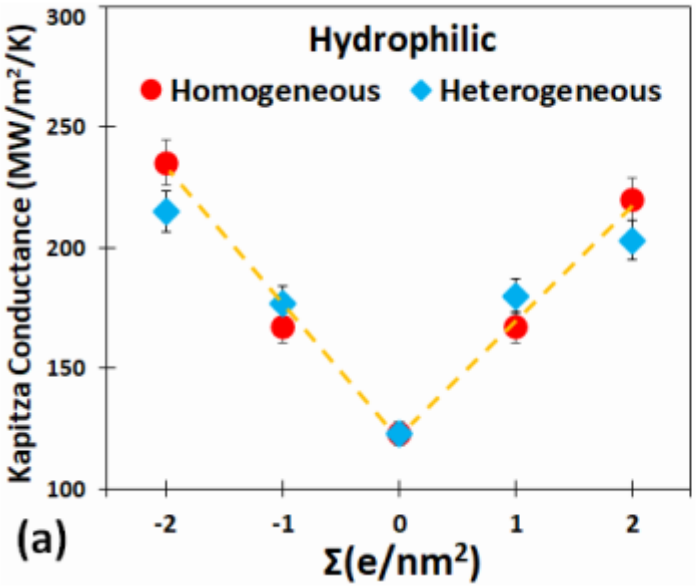
Herrero, Joly, Merabia, *Appl. Phys. Lett.* 2022,

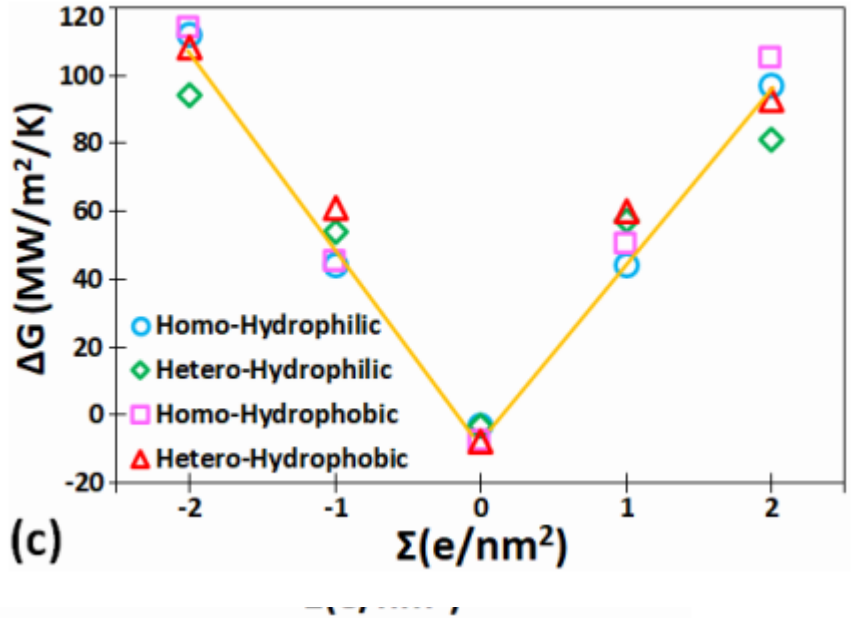
Heat transport at charged nanoparticle interfaces

with **R. Rabani**, L. Joly and A. Rajabpour



$$G^{-1} = R_K = \frac{A}{k_B T_0^2} \int_0^\infty < \Delta T(t) \Delta T(0) > dt,$$





Rabani et *al.*, in prep.

Some useful books and reviews

Textbooks : « The physics of phonons », Srivastava

« Nanoscale energy transport and conversion : a parallel treatment of electrons, molecules, phonons and photons », G. Chen

Review articles : « Nanoscale thermal transport II », *App. Phys. Rev.,* 2014, Cahill et *al.*

« A review of experimental and computational advances in thermal boundary conductance and nanoscale thermal transport », *Adv. Funct. Mater*, Giri and Hopkins

« Interfacial thermal resistance: past, present and future », Chen, Xu, Zhou and Li, *Rev. Mod. Phys.* 2022

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Thank you for your attention !