Phonons and thermal physics at the micro and nanoscale

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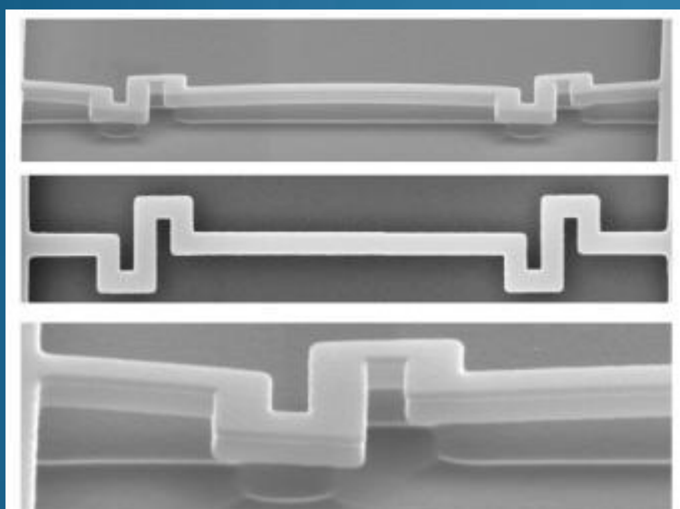
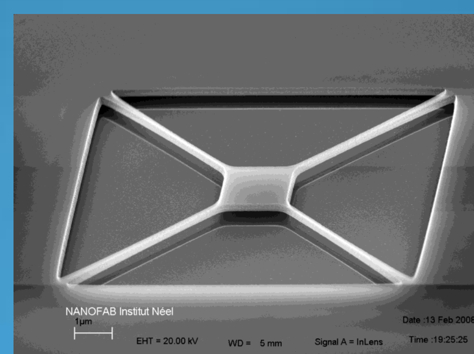
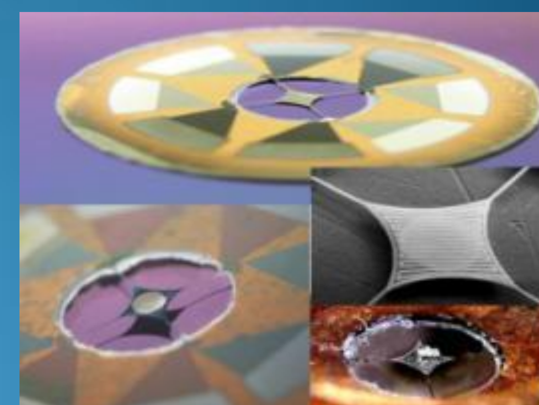






Outline

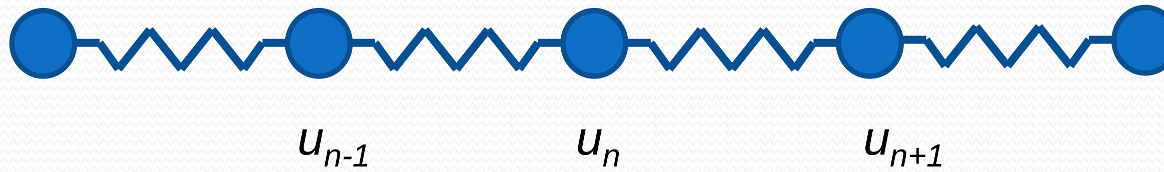
- Introduction: necessary concepts: phonons in low dimension, characteristic length
- Part 1: Transport and heat storage via phonons
  - Specific heat and kinetic equation
  - Thermal conductance, transport of phonon at the nanoscale, mean free path
  - Casimir model, Ziman model and beyond
- Part 2: Dynamic method for thermal investigation
  - History
  - Corbino contribution
  - Ac calorimetry, membrane based measurements
- Part 3: Measurement method for phonon transport
  - General method (3ω, pump probe experiment)
  - At the nanoscale and at low temperature
  - Some example (nanowire, membrane experiment)
- Conclusive remarks

Phonon : linear chain

- Linear monoatomic chain
- Periodic conditions: Born-von Karman
- Quantization of the vibrational modes
- Dispersion relation
- Group velocity

$$E = \left(n_q + 1/2\right)\hbar\omega$$

- Elementary excitation: phonon

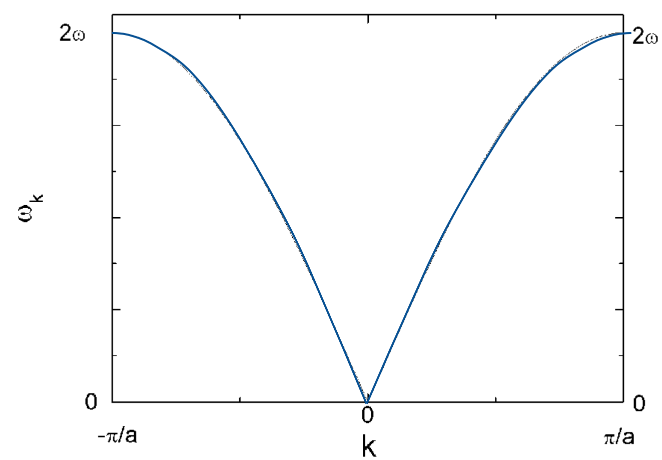


$$m\frac{d^2u_n}{dt^2} = K(2(u_{n+1} - u_n) + (u_n - u_{n-1}))$$

$$u_n = \exp(i(kna - \omega t))$$

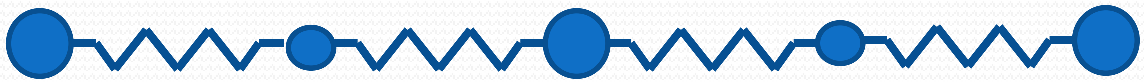
$$\omega = \sqrt{\frac{K}{am}}\left|\sin\left(\frac{ka}{2}\right)\right|$$

$$v = \frac{\partial\omega}{\partial k}$$



Phonons and low dimensions

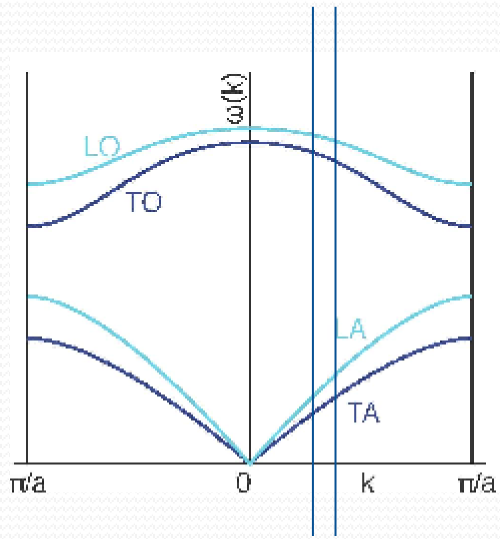
- Diatomic chain
- Acoustic and optical modes
- Spatial extension of a phonon ?
- Phonon=Wave packet
- Propagating modes TA and LA (related to *v*)



$$\Delta x \Delta k > 2\pi$$

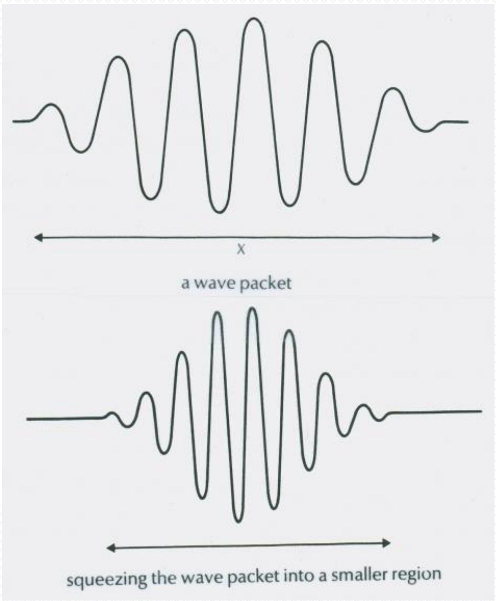
$$\Delta k < \frac{\pi}{10a} \qquad \Delta x > 10a$$

$$\Delta x > 5nm$$



Δk

$$v = \frac{\partial \omega}{\partial k}$$



Phonon identity card

- Boson (Bose-Einstein distribution)
- Number of phonons not conserved
- Chemical potential =0
- Quasi-particle
- At low temperature : only large wave length phonon are excited (low energy)
- No optical phonon (only acoustic modes)
- Planck black body radiation (for infinitely rough surfaces)

$$n_q = \frac{1}{\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1}$$

High temperature

$$n_q \approx \frac{k_B T}{\hbar\omega}$$

Low temperature

$$n_q \approx \exp\left(-\frac{\hbar\omega}{k_B T}\right)$$

Introduction : at low temperature

Two kind of heat carriers **(radiation neglected)**

T<10K

Electrons		Phonons
Λ_{e-}~nm	•Mean free path	Λ_{ph}~mm
λ_F~0.1nm	•Relevant wave length	λ_{ph}~100nm
Lφ~10μm	•Coherence length	Not coherent
T	•Temperature dependence (K, C)	T^3
diffusive	•Transport	ballistic
fermions	•statistic	bosons

Thermal conductance (heat transport/out of equilibrium) **Heat capacity (heat storage/equilibrium)**

Low temperature specificities

- Competition between electrons and phonons for the thermal transport
- No Umklapp process (k too small)
- Large mean free path for phonons… transport limited by boundary scattering
- Importance of interface resistance ($T<1K$)
- Quantum effect (size effect): quantization of energy level, effects of phase coherence etc…

“Micro-Nano” problematic

- Loss of the bulk behavior, competition between surface and volume.
- Significant characteristic length (dominant phonon wave length, phase coherent length, mean free path, , etc…)
- New condensed matter state (new phase transition)
- Specific thermal behavior at small length scale (universal thermal conductance, definition of temperature …)
- Systems under study need to be thermally isolated : membrane, suspended structure (nanowire, graphene sheet, sensitive sensors)
- Development of new experimental tools using nanotechnology adapted to very small thermal signals and adapted to very small mass samples of the order of zepto (10^{-21}J) or yoctoJoule (10^{-23}J).

Thermal study based on Electrical measurements

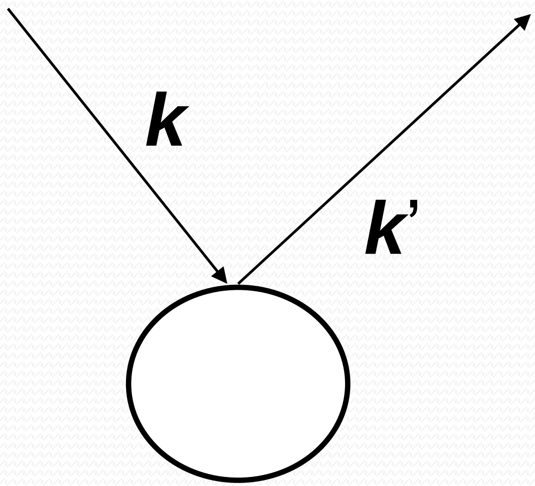
- Understanding the thermal physics in extreme conditions (ultra low temperature, small mass sample, nanostructured materials)
- Development of adapted technology (instrumentation, sensors,
- Bolometry, MEMS and NEMS
- Nano ElectroThermal System (NETS)
- Thermal decoherence for quantum information

Understand and master thermal properties at low temperature and low dimensions

•Heat pulse technique (Pump probe experiment, thermometer) Optical method
•Phonon spectroscopy (MHz to GHz)
•Ect…

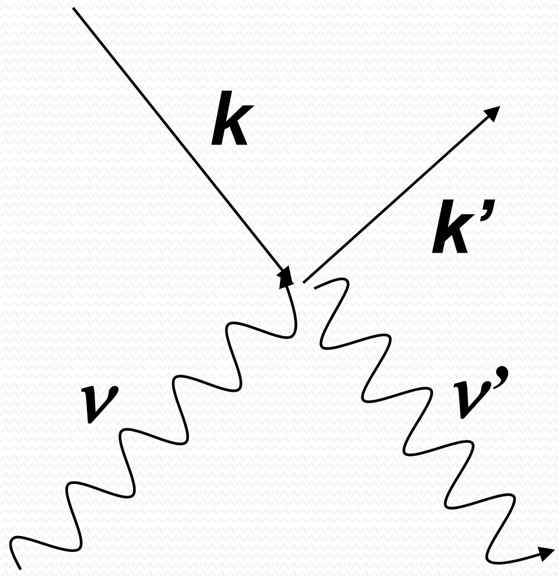
Concept of Temperature at low dimensions

Mean free path: elastic versus inelastic



Phonon elastic scattering (impurity)

$$|k| = |k'|$$



Inelastic scattering

$$|k| \neq |k'| \quad \text{and} \quad |\upsilon| \neq |\upsilon'|$$

Different scattering processes

- Scattering on dislocation(static imperfection)
- Anharmonic scattering (three phonons, Umklapp processes)
- Boundary scattering (finite size effect)
- Electron-phonon interaction (doped semiconductor)

$$\frac{\partial f}{\partial t} + \frac{d\,\vec{r}}{dt}.\vec{\nabla} f + \frac{d\,\vec{p}}{dt}.\vec{\nabla} f = \left(\frac{\partial f}{\partial t}\right)_{coll}$$

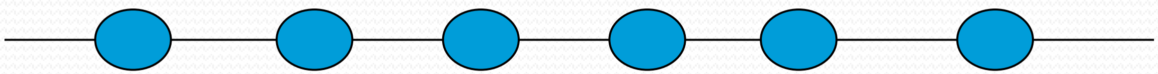
$$\left(\frac{\partial f}{\partial t}\right)_{coll} = -\frac{f - f_0}{\tau}$$

Mathiessen rule:

$$\tau^{-1} = \sum_{i=1}^{n} \tau_i^{-1}$$

$$\Lambda_{ph} = v_{ph}\tau_{scatt}$$

Temperature at the nanometer scale ?



What is the minimum size to define a temperature?

Existence of Temperature on the Nanoscale

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(Received 30 December 2003; published 19 August 2004)

We consider a regular chain of quantum particles with nearest neighbor interactions in a canonical state with temperature T. We analyze the conditions under which the state factors into a product of canonical density matrices with respect to groups of n particles each and under which these groups have
inimum group size $n_{\min}$ depends on the
ur analysis to a harmonic chain and find
ature and $n_{\min} \propto T^{-3}$ below.

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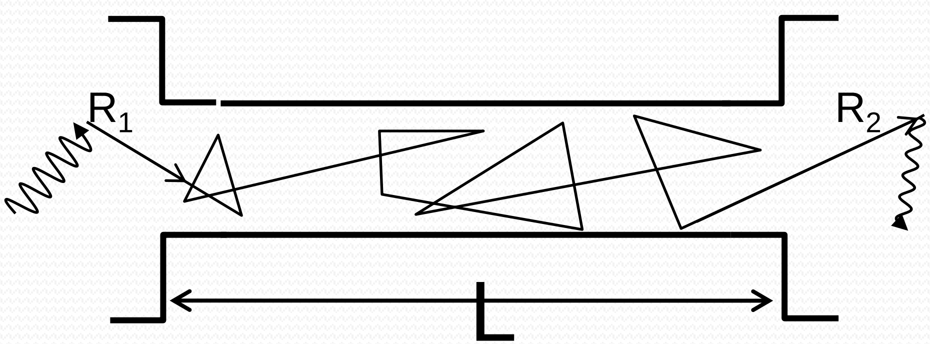
$$n_{\min} \approx \begin{cases} 2\alpha/\delta & \text{for } T > \Theta, \\ (3\alpha/2\pi^2)(\Theta/T)^3 & \text{for } T < \Theta. \end{cases}$$

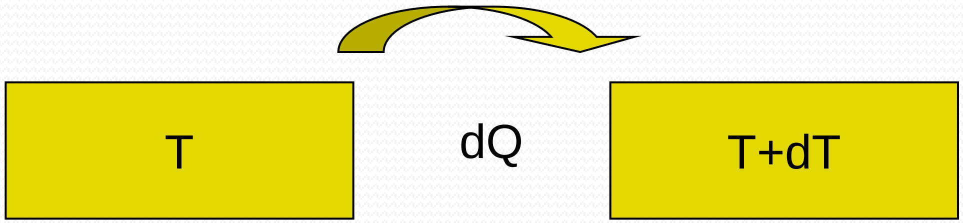
In silicon at 1K, for a Debye temperature of 680K, it is meaningless to speak about temperature for a cube of **50x50x50nm**.



$$\mathcal{V} < \Lambda_{ph}^{3}$$

Specific heat at low dimensions

Heat capacity: heat storage



$$C = \frac{\delta Q}{\delta T}$$

- Physical quantity related to the bulk
- Relating the energy necessary to increase the temperature by one Kelvin
- Degree of freedom of the system (at equilibrium)
- Anomaly: phase transformation, phase transition (1st order and2nd order)

$$\boxed{C = \frac{dU}{dT} = \frac{T\partial S}{\partial T} = -T\frac{\partial^2 F}{\partial T^2}}$$

Calculation of C_p for phonons

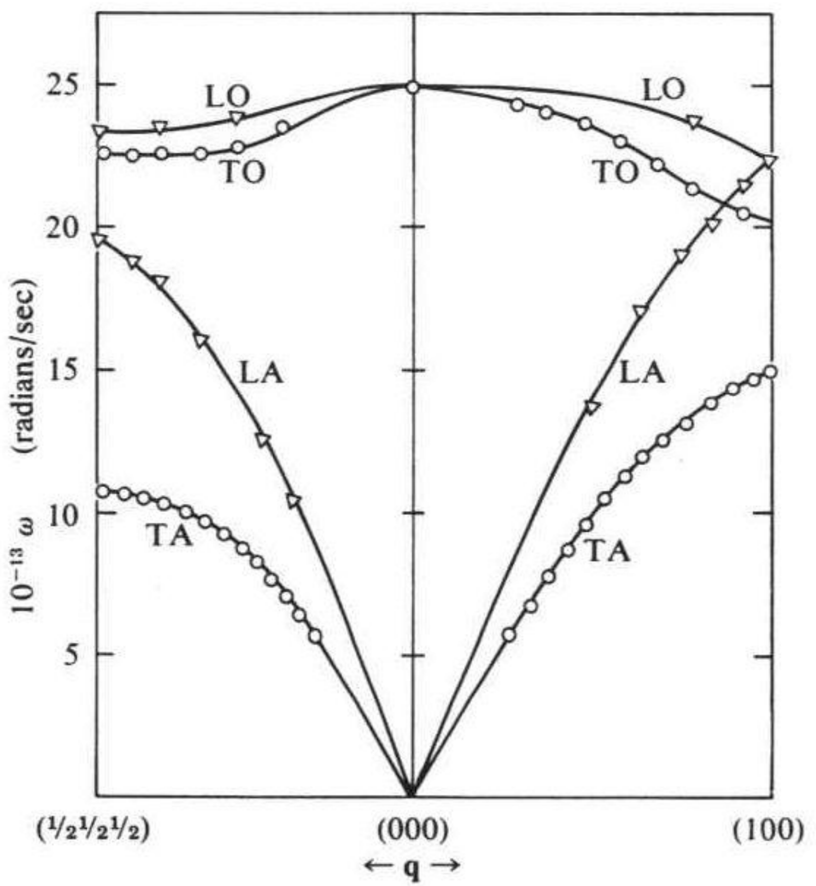
- Lattice considered as a sum of harmonic oscillators
- Only large wave length are excited (kT low)
- Linear dispersion relation $\omega=\alpha k$
- Einstein model improved by Debye (1912)

$$C_p \propto T^d$$

- This will be wrong in the case of materials having reduced dimensions like CNT or graphene (quadratic dispersion relation)

$$\omega \propto k^{\delta}$$

$$C_p \propto T^{\frac{d}{\delta}}$$



Calculation of C_p for phonons

- The crystal lattice is considered as a sum of harmonic oscillator
- Only large wave length are excited (low temperature)
- Linear dispersion relation ω=αk
- Einstein model improved by Debye (1912)

$$C_p \propto T^d$$

- Not correct for specific materials carbon nanotube, or graphene (dispersion relation quadratique) si

$$\omega \propto k^\delta$$

$$C_p \propto T^{\frac{d}{\delta}}$$

$$U = \sum_k \left(\frac{1}{2}\hbar\omega(\boldsymbol{k}) + \frac{\hbar\omega(\boldsymbol{k})}{e^{\left(\frac{\hbar\omega(\boldsymbol{k})}{k_B T}\right)} - 1} \right)$$

$$U = \int_{BZ} \frac{\hbar\omega(\boldsymbol{k})}{e^{\left(\frac{\hbar\omega(\boldsymbol{k})}{k_B T}\right)} - 1} \frac{d\boldsymbol{k}}{(2\pi)^3} = \int \frac{\hbar\omega}{e^{\left(\frac{\hbar\omega}{k_B T}\right)} - 1} D(\omega) d\omega$$

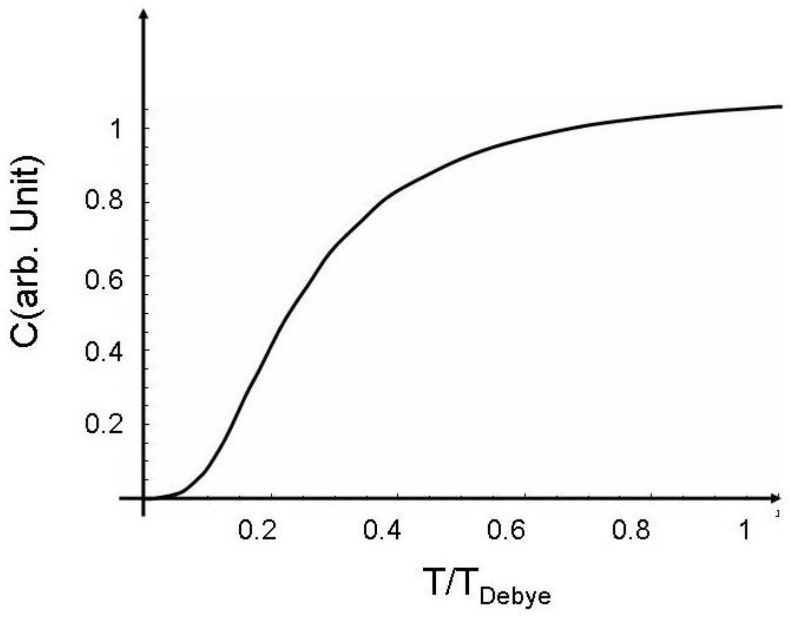
$$U = \frac{6}{\pi^2} \int_0^\infty \frac{\hbar v_s k^3 dk}{e^{\left(\frac{\hbar v_s k}{k_B T}\right)} - 1}$$

Low temperature limit:

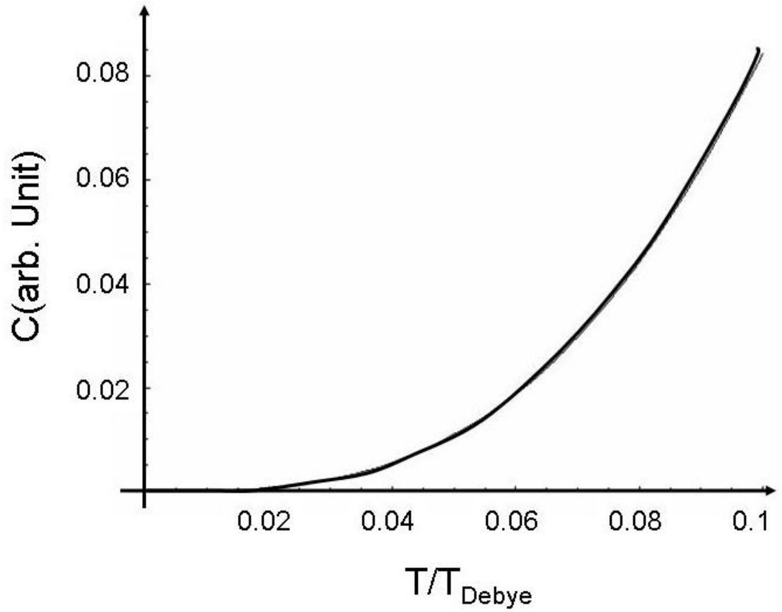
$$C_{ph} = \frac{2\pi^2}{5} \frac{k_B^4 T^3}{\hbar^3 v_s^3}$$

Variation of specific heat versus temperature: Debye model

Debye



T^3 law



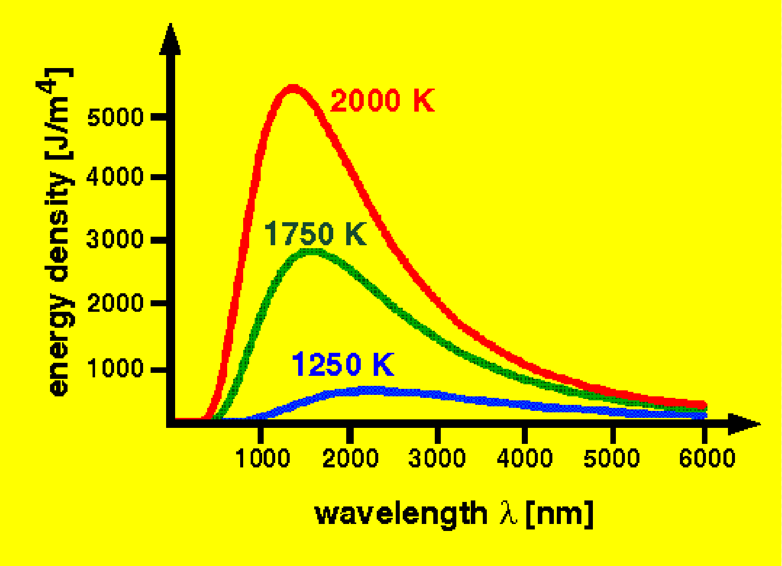
1D, 2D, 3D ?

- What is the relevant characteristic length ?
- The dominant phonon wave length

$$\lambda_{dom} \approx \frac{\theta_{Debye} a}{T} \approx \frac{h v_s}{2.82 k_B T}$$

$$d \leq \lambda_{dom}$$

In silicon at 1K λ_{dom}=100nm
In diamond at 1K λ_{dom}=300nm



Planck law/ Black body radiation

Theoretical calculation of the specific heat in a submicron membrane

- Vibration modes in a suspended phonon cavity
- T^2 behavior for C_p at low temperature

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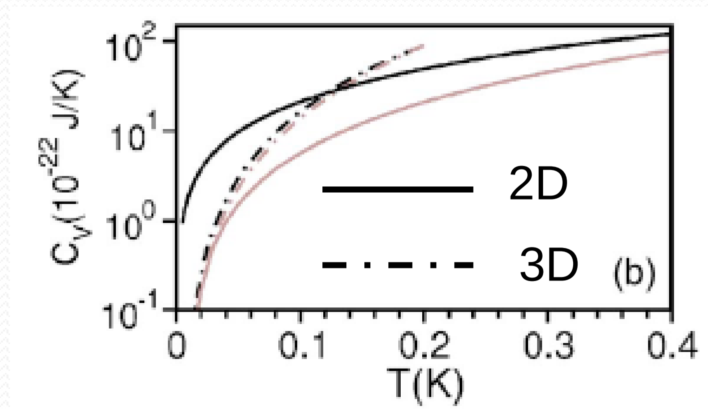
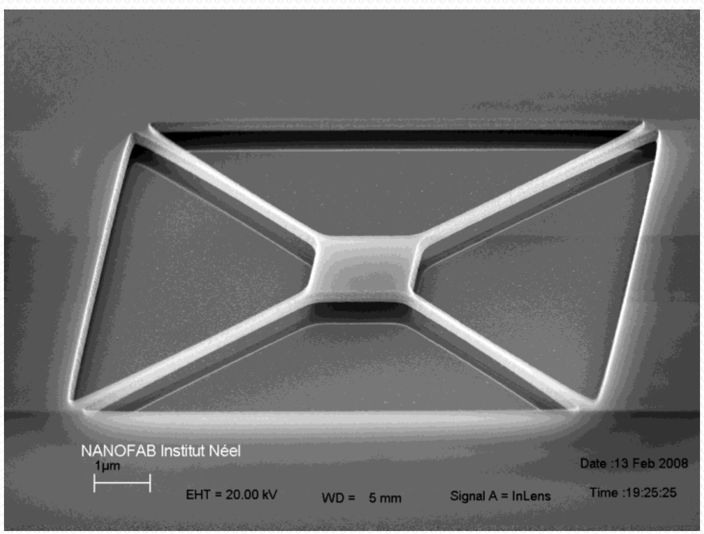
Heat capacity of suspended phonon cavities

A. Gusso[1,2,*] and Luis G. C. Rego[1]





Electronic specific heat

- Linear in temperature
- Dominant at low temperature (as compared to the lattice)
- In case of noble metals
- Hypothesis:
  $k_BT << E_F$
  $D(E) \sim D(E_F)$ (low temp.)

$$U = \int_0^\infty E f(E) D(E) dE$$

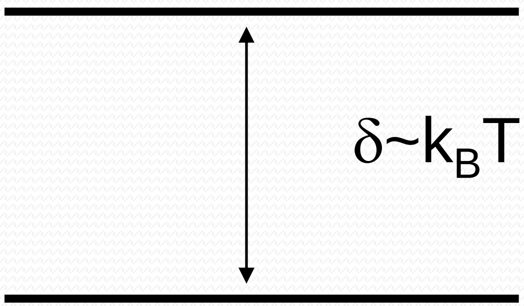
$$f(E) = \frac{1}{\left(1 + \exp\left(\frac{E - E_F}{k_B T}\right)\right)}$$

$$D(E) = 1/2\pi^2 \left(\frac{2m}{\hbar^2}\right)^{3/2} \sqrt{E}$$

$$C_{e^-} = \frac{\pi^2}{3} D(E_F) k_B^2 T$$

Example of finite size consequences on C_p : nanoparticle

- Discrete energy levels: Schottky anomaly at low temperature (two level system)
- Fluctuations
- Superconducting phase transition perturbed when $\delta \sim \Delta$



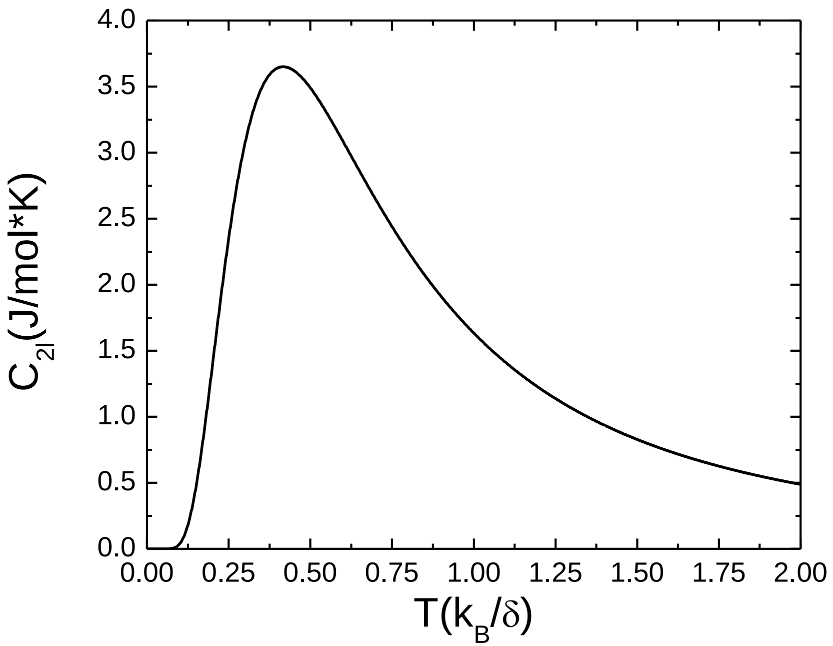
Example of finite size consequences on C_p : nanoparticle

- Discrete energy levels: Schottky anomaly at low temperature (two level system)
- Fluctuations
- Superconducting phase transition perturbed when $\delta \sim \Delta$

$$Z = \sum_{i=1}^{2} e^{\frac{-\epsilon_i}{k_B T}}$$

$$E = \frac{1}{Z}\sum_{i=1}^{2} \epsilon_i e^{\frac{-\epsilon_i}{k_B T}} = \epsilon_1 + \delta \frac{e^{-\delta/k_B T}}{1 + e^{-\delta/k_B T}}$$

$$C_{2l} = \frac{\delta^2}{k_B T^2} \frac{e^{-\delta/k_B T}}{(1 + e^{-\delta/k_B T})^2}$$



W. Schottky Phys. Z, 23, 448 (1922)

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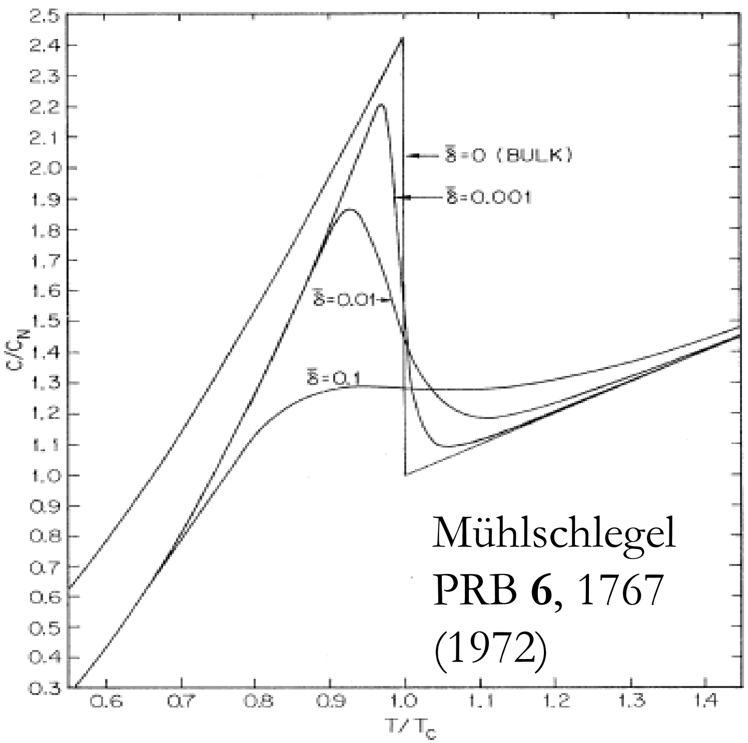
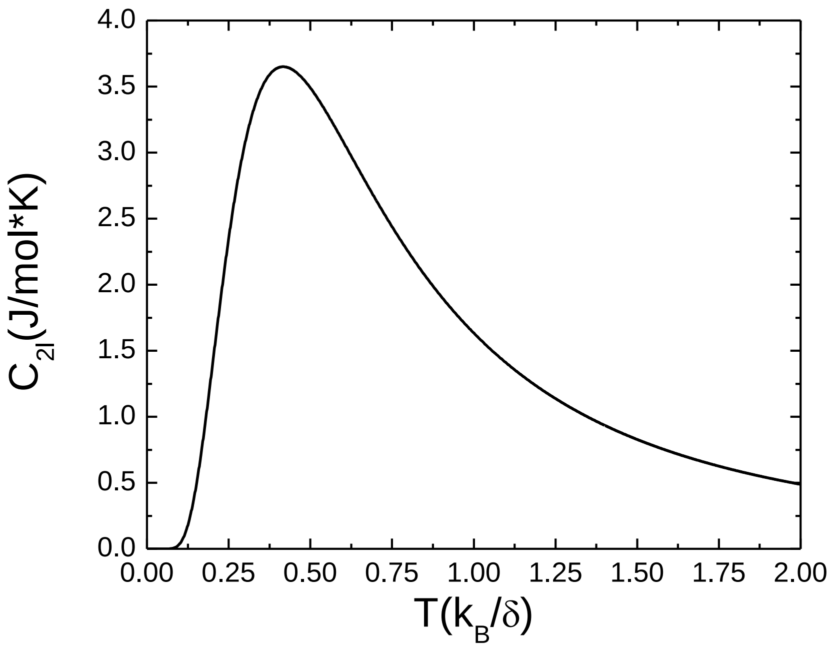


FIG. 2. Normalized specific heat near $T = T_c$ calculated in the GL approach for several values of the parameter $\bar{\delta} = \delta/kT_c$. For reference the bulk BCS limit is also shown.



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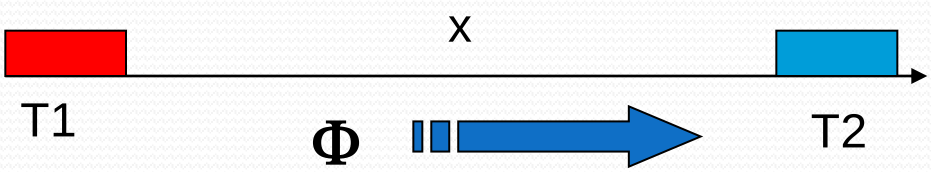
Thermal conductivity or thermal conductance ?

Thermal conductance at low temperature



$$\phi = \frac{1}{V}\sum_k v_x E(k) f(\vec{r},\vec{p})$$

- Thermal flux along the x axis

$$\frac{\partial f}{\partial t} + \frac{d\vec{r}}{dt}.\vec{\nabla} f + \frac{d\vec{p}}{dt}.\vec{\nabla} f = \left(\frac{\partial f}{\partial t}\right)_{coll}$$

- Approximation of the relaxation time

$$\left(\frac{\partial f}{\partial t}\right)_{coll} = -\frac{f - f_0}{\tau}$$

- Kinetic equation

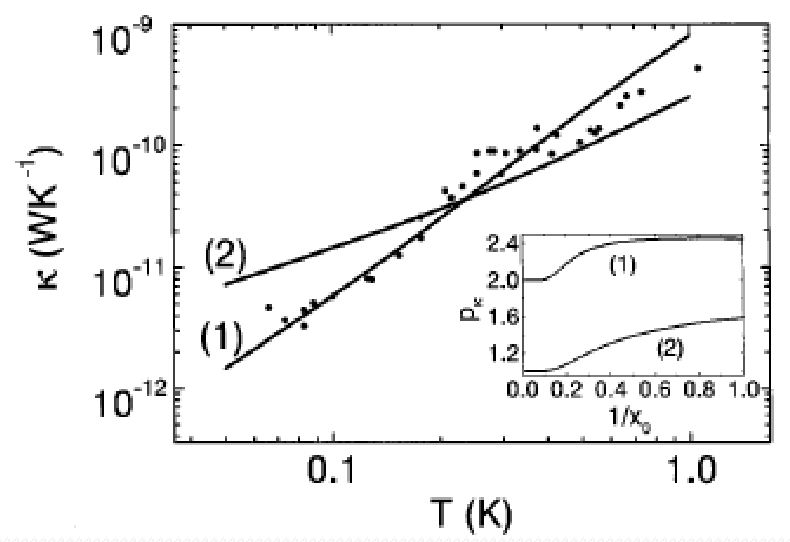
$$\phi = \sum_s \int_0^{\omega_{max}} d\omega \int_0^{2\pi} d\varphi \int_0^{\pi} d\theta v_x \hbar\omega \left(f_0 - \tau \frac{df}{dT}\vec{v}\,.\,\vec{\nabla}T\right)\frac{D(\omega)}{4\pi}$$

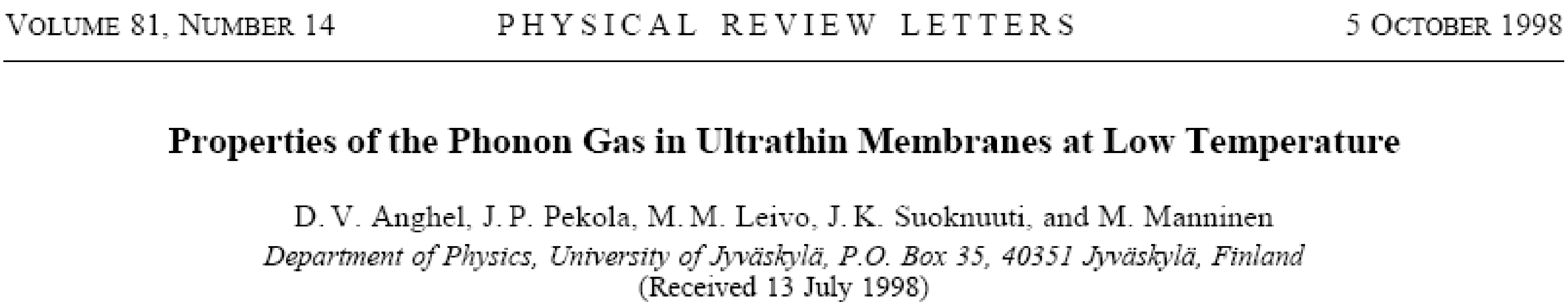
$$\phi = -k_{ph}\frac{dT}{dx} \longrightarrow k_{ph} = \sum_s \int_0^{\omega_{max}} d\omega \int_0^{2\pi} d\varphi \int_0^{\pi} d\theta sin(\theta) cos^2(\theta) \hbar\omega\tau v_x^2 \frac{df_0}{dT}\frac{D(\omega)}{4\pi}$$

$$K_{ph} = \frac{1}{3} C_{ph} v_{ph} \Lambda_{ph} \qquad C_p \propto T^d \qquad K \propto T^d$$

Example of 3D to 2D transition

- Silicon nitride membrane
- Specific theoretical treatment
- Transition from T^3 to T^2 for the thermal conductance

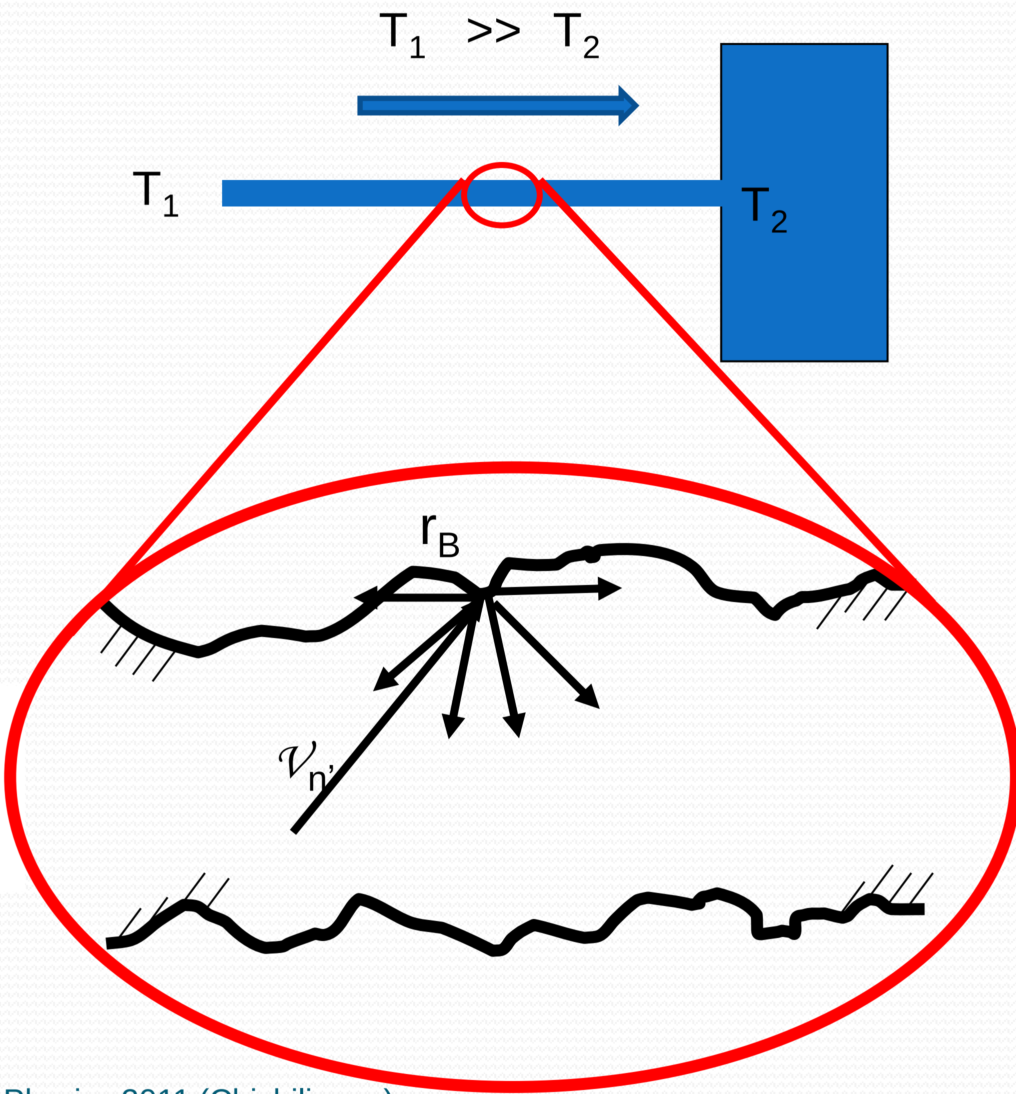




Finite size effect: Casimir theory for phonon transport

- Mean free path Λ_{ph}
- Λ_{Cas}=D (Diameter of the nanowire)
- Boundary scattering: black body radiation for phonons
- Expression for K(T)
- Still diffusive
- Comment on the specific heat (kinetic equation)

$$K(T) = 3.2 \times 10^3 \left(\frac{2\pi^2 k_B^4}{5\hbar^3 v_s^3}\right)^{(2/3)} \frac{S\Lambda_{Cas}}{L} T^3$$

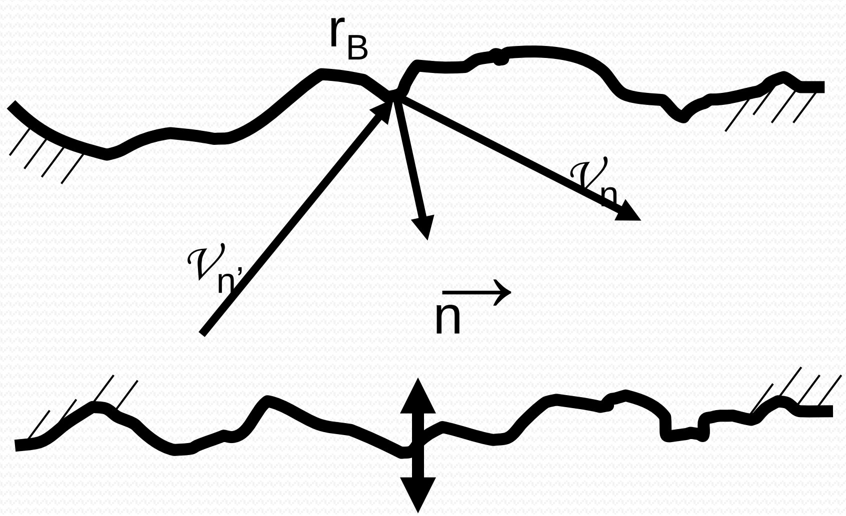


Breakdown of the concept of thermal conductivity

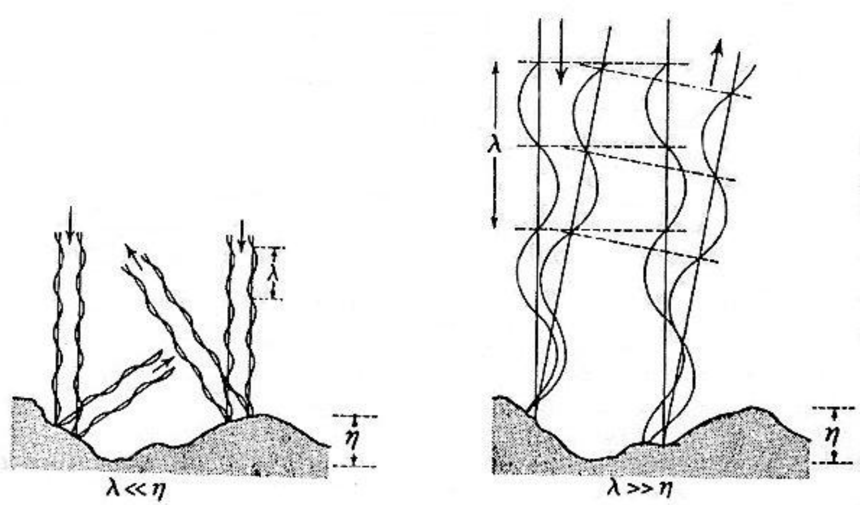
Casimir theory and beyond

- At low temperature, the dominant phonon wave length is increasing:
- Probability of specular reflection $p(\lambda_{dom})$ depending on λ_{dom} (phenomenological parameter)
- $p(\lambda_{dom})=0$ (perfectly rough surface) $\lambda_{dom} \ll \eta_0$ Casimir model
- $p(\lambda_{dom})=1$ (perfectly smooth surface) $\lambda_{dom} \gg \eta_0$

$$\lambda_{dom} = \frac{hv_s}{2.82k_BT}$$



η_0 is the root mean square of the asperity



J.M. Ziman *Electrons and phonons* (Clarendon Press, Oxford, 2001)

Casimir Theory and beyond : the Ziman model

- Ziman-Casimir model

$$\Lambda_{ph} = \frac{1+p}{1-p}\Lambda_{Cas}$$

where *p* probability of specular reflection

If *p*=0 transport is diffusive (Casimir), if *p*=1 ballistic transport

$$p(\lambda) = \int P(\eta) e^{\frac{-16\pi^3\eta^2}{\lambda_{dom}^2}} d\eta$$

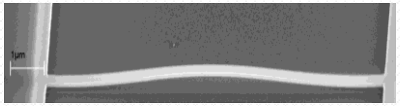
Probability distribution of asperity

$$P(\eta) = \frac{1}{\eta_0} e^{-\eta/\eta_0}$$

$$K(T) = 1.35 \times 10^{-5} \left(\frac{2 - e^{-4\pi\lambda_{dom}(T)/\eta_0}}{e^{-4\pi\lambda_{dom}(T)/\eta_0}} \right) \Lambda_{Cas} T^3$$

Ziman model of phonon transport: ballistic contribution

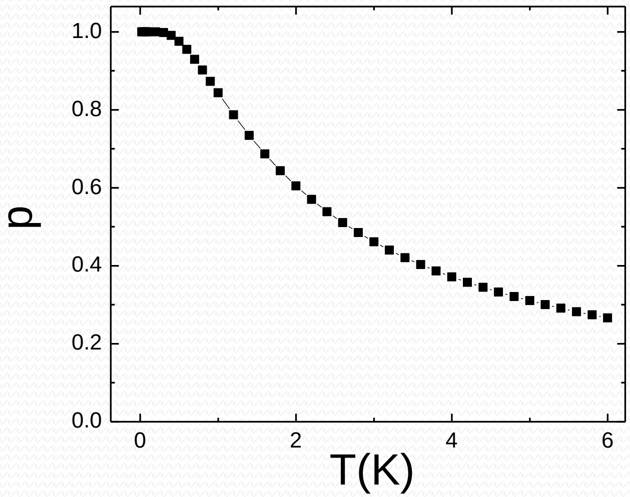
$$K = 3.2\times10^{-3}\left(\frac{2\pi^2 k_B^4}{5\hbar^3 v_s^3}\right)^{2/3}\frac{S\Lambda_{eff}}{L}T^3$$

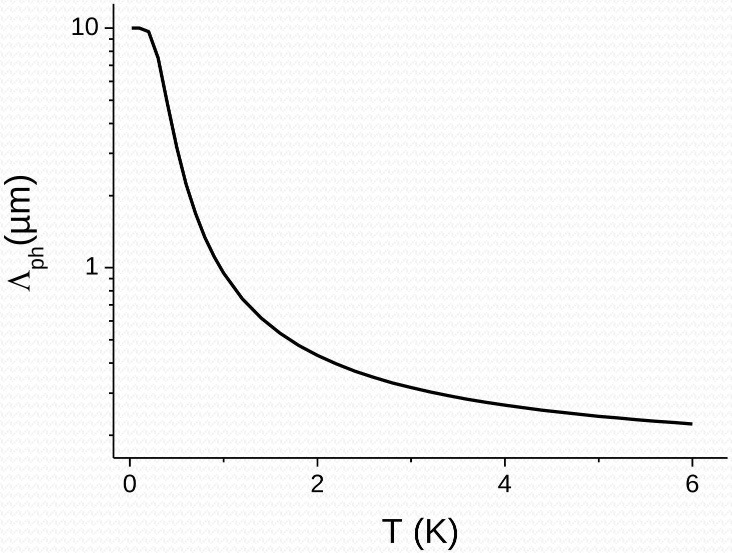


$$\lambda_{Dom} = \frac{hv_S}{2.82k_B T}$$

$$\Lambda_{eff} = \frac{1+p}{1-p}\Lambda_{Cas}$$

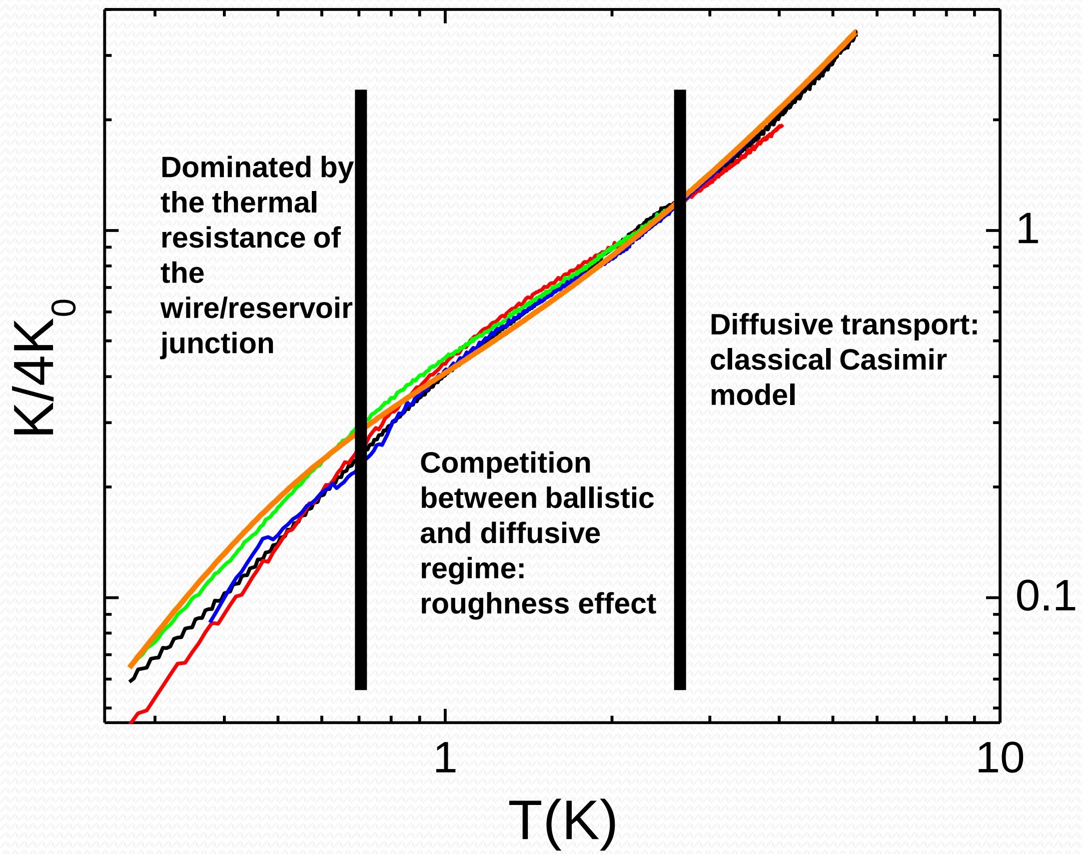
$$\Lambda_{ph}^{-1} = \Lambda_{eff}^{-1} + L^{-1}$$

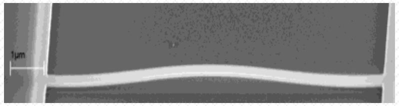




J.-S. Heron, T. Fournier, N. Mingo and O. Bourgeois, Nano Letters **9**, 1861 (2009).

Evidence of contribution from ballistic phonons





Fitting parameter:

- Roughness h=4nm
- Speed of sound 9000m/s
- Contribution of the contact (Chang, C.; Geller, M. *Phys. Rev. B* **2005**, *71*, 125304.)

J.-S. Heron, T. Fournier, N. Mingo and O. B, Nano Letters **9**, 1861 (2009).

Implications: thermalization, nanothermoelectricity

- Ballistic phonon->no local temperature
- ~~Thermal conductivity~~ ->thermal conductance (driven by the size of the systems)
- Play with the phonons: phonon focusing, phonon blocking etc..
- Application to thermoelectricity: phonon scattering at the nanoscale with clusters, nanoparticles, superlattice etc…

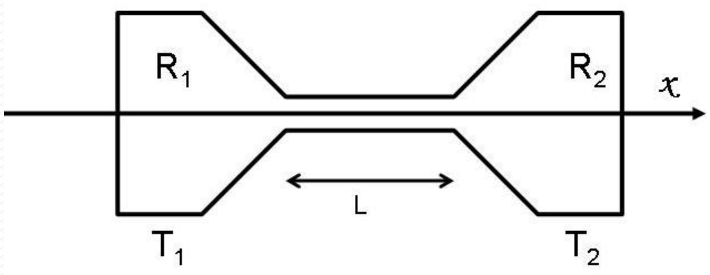
$$ZT = \frac{S^2 T\sigma}{\left(k_{elec} + k_{ph}\right)} \qquad \eta_{\max} = \eta_c \frac{\sqrt{ZT+1}-1}{\sqrt{ZT+1}+1}$$

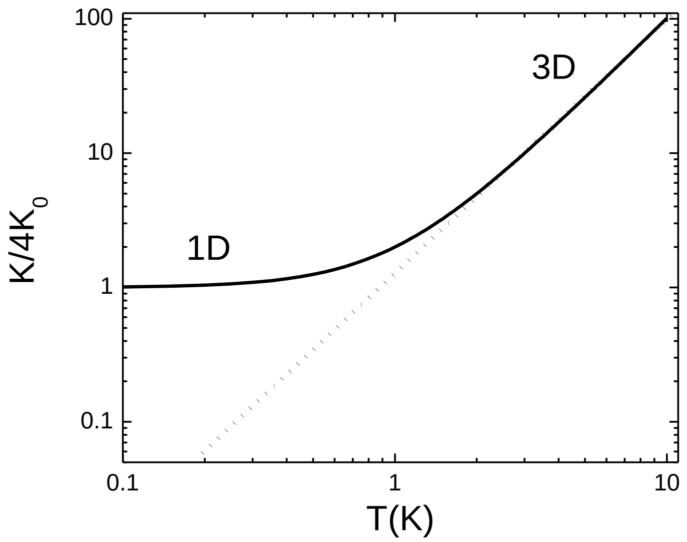
Limit at low dimensions and low temperature: universal thermal conductance

- Λ>>d
- **λ_{dom}>>d**
- 4 acoustic phonon modes

$$K_0 = \frac{4\pi^2 k_B^2 T}{3h}$$

- Conduction channel (Similar to the Landauer model of electrical conductance)
- Not dependant on the materials
- Valid whatever the heat carrier statistic
- Pendry, Maynard: flow of entropy or information





J.B. Pendry, J. Phys. **16**, 2161 (1983)

R. Maynard and E. Akkermans, Phys. Rev. B **32**, 5440 (1985)

L.G.C. Rego and G. Kirczenow, Phys. Rev. Lett. **81** 232 (1998)

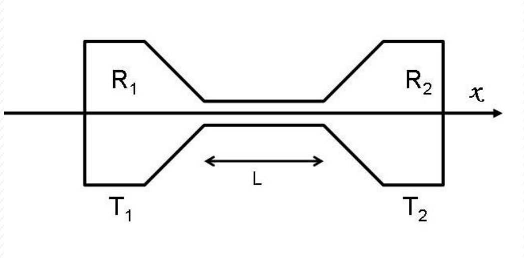
Thermal conductance of electrons

$$k_{e^-} = \frac{1}{3} C_{e^-} v_F \ell_{e^-}$$

- k linear in temp.
- Wiedemann-Franz law

$$k_{e^-} = \frac{\pi^2}{9} D(E_F) v_F \ell_{e^-} k_B^2 T$$

$$\sigma = \tfrac{1}{3} D(E_F) v_F^2 \tau_{e^-} \longrightarrow \frac{K}{GT} = \frac{\pi^2 k_B^2}{3e^2} = L_0$$



$$G = \frac{2e^2}{h}$$

Quantum of electrical conductance!!