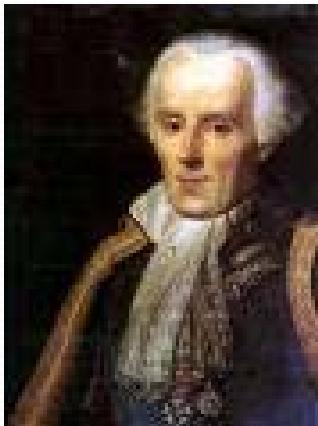


Thermal measurement at low temperature: heat capacity, thermal conductance

History



Pierre Simon de Laplace



Antoine Laurent Lavoisier

- Treatise on calorimetry published in 1782
- First experiment of heat capacity
- Ice calorimeter (temperature reference)

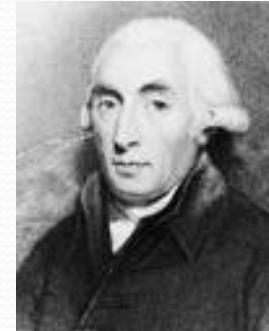


L (latent heat
ice/water at $T = 0^\circ\text{C}$)
 $= 334 \text{ J/g} \approx 80 \text{ cal/g}$

L (latent heat
water/water vapour
at $T = 100^\circ\text{C}$) $= 2260 \text{ J/g} \approx 539 \text{ cal/g}$

Heat/Matter/Temperature

- Concepts of chemistry/physics late XVIIIth cent.
- Heat and temperature (Joseph Black 1728-1799)
- Heat transfer law (Joseph Fourier 1768-1830)
- Matter and energy(James Joule 1818-1889)
- Pierre-Eugène Berthelot (1827-1907) first modern calorimeter



Joseph Black



Joseph Fourier

Théorie analytique de la chaleur (1822)



James Joule



Pierre Eugène Berthelot

Signal treatment/measure using temperature modulation

- Focus on electrical based measurement
- Problems related to measurement using continuous signal
- Preamplification of the signal before measurement
- AC thermal measurements: proposed since 1910 by O. Corbino
- Measure by lock-in amplifier
- Differential geometry



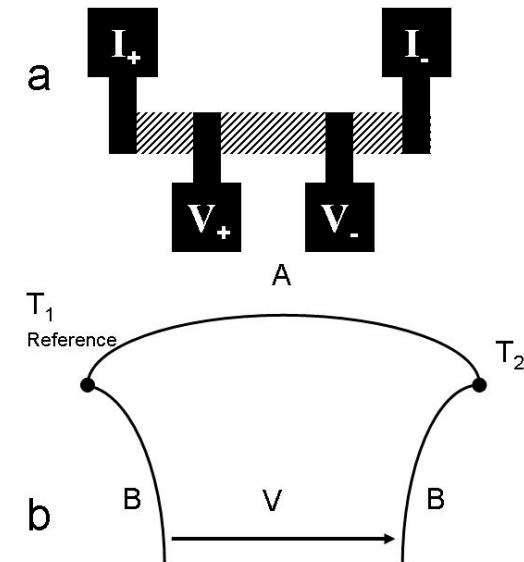
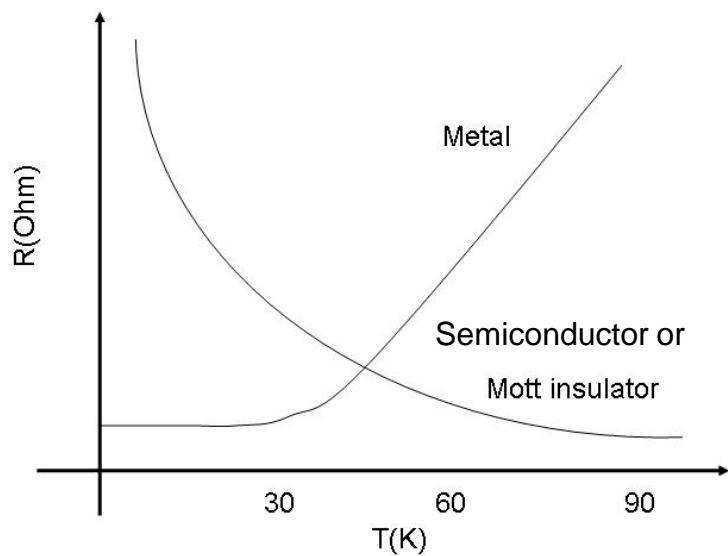
Orso Mario Corbino (1876-1937)

O.M. Corbino, Phys. Z. **11**, 413 (1910), *ibid.* **12**, 292 (1911)

Thermometry and measurement

- Sensitivity of the measurement: temperature coefficient
- Resistive Thermometry (films minces)
- Thermocouple (High temp.)
- 4 probe measurement(4 pointes) separate the voltage leads from the current leads.

$$\alpha = \frac{1}{\Psi} \frac{\partial \Psi}{\partial T}$$



Warning : Limitation of resistive thermometry at low temperature

- Measurement of the electron temperature
- Under low electric field

$$T_{e^-} - T_{ph} = \frac{P_{e^-}}{K_{e^-/ph}} = \tau_{e^-/ph} \frac{V^2}{RC_{e^-}}$$

- High electric field

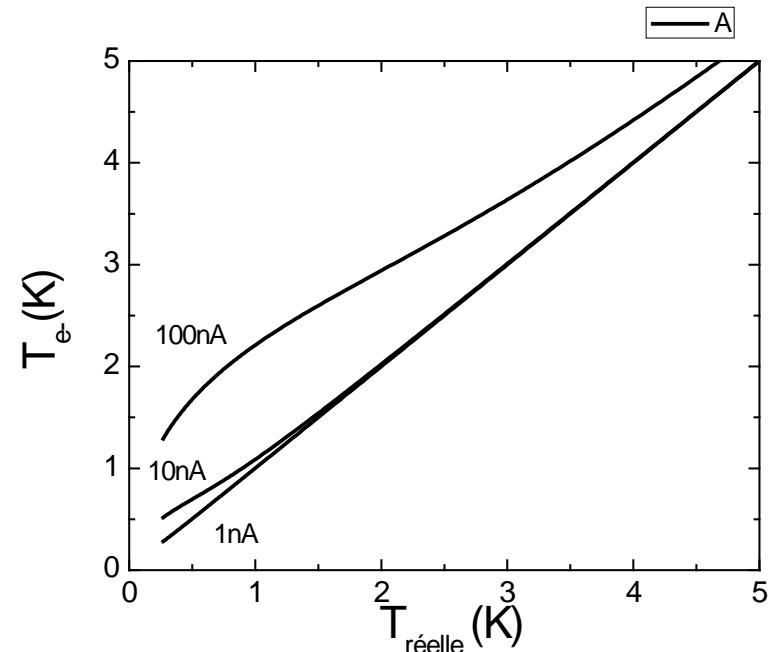
$$T_{e^-}^n - T_{ph}^n = \frac{P_{e^-}}{Vg_{e^-/ph}}$$

- Work with high impedance thermometer

e-phonon coupling constant (g_{e^-}/ph)
1000W/K⁵cm³

Exercise: thermometer of 50nm, 100nm large, 10mm long

R=200kOhm, I_{ac}=1nA, T=0.5K, ΔT=??

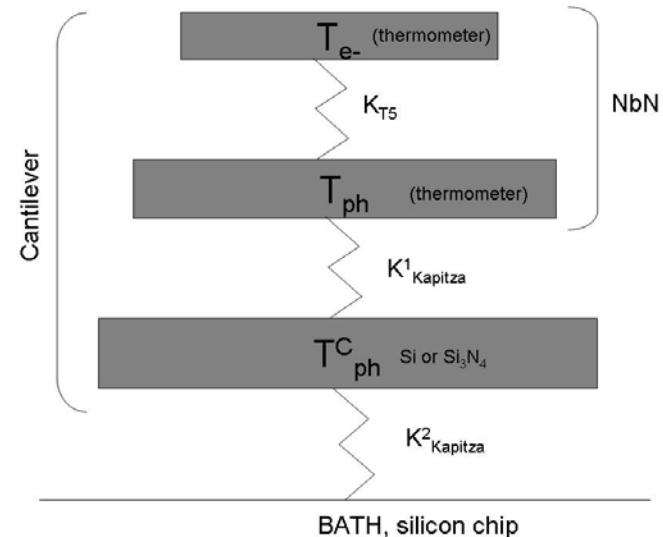


Example : silicon nanowire

Answer 4mK

Measurement of downscaled system

- Sensitive measurement of the temperature
- Temperature measurement as close as possible to the system
- Kapitza resistance
- Acoustic Mismatch Model (AMM) and Diffusive Mismatch Model (DMM)

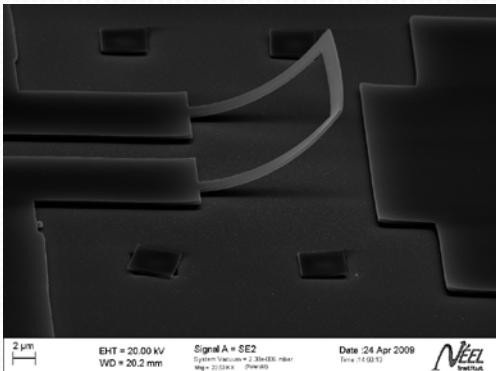


$$K_{Kapit}^1 = \frac{T^3}{12} \text{ W/Kcm}^2$$

See Swartz and Pohl *Rev. Mod Phys* **61**, 605 (1989)

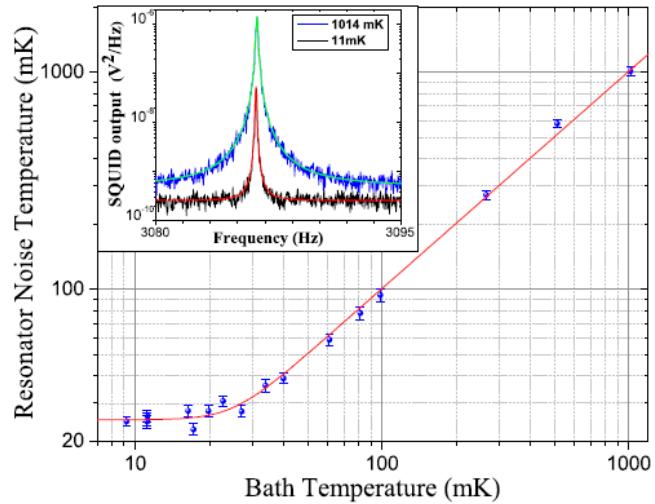
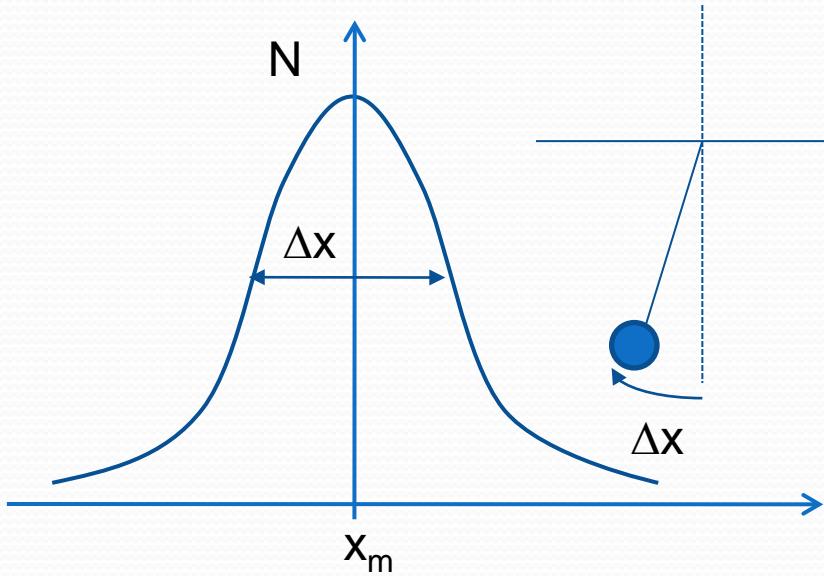
Temperature measurement from nanomechanics

- Measurement of thermal noise
- Vibration of a cantilever : NEMS
- No thermometer needed, universal thermometry FWHM $\sim k_B T$
- Macroscopic measurement of the global temperature of a beam



$$\frac{1}{2}k\langle\Delta x^2\rangle=\frac{1}{2}k_B T$$

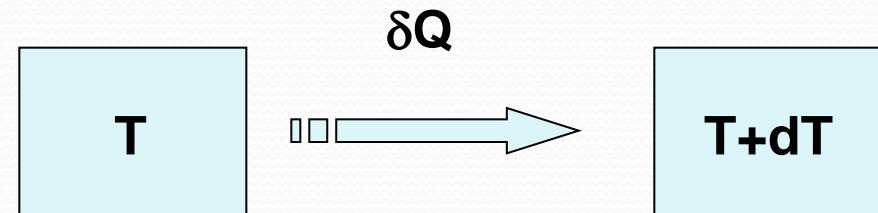
$$\langle\Delta x^2\rangle=\frac{k_B T}{m\omega_0^2}$$



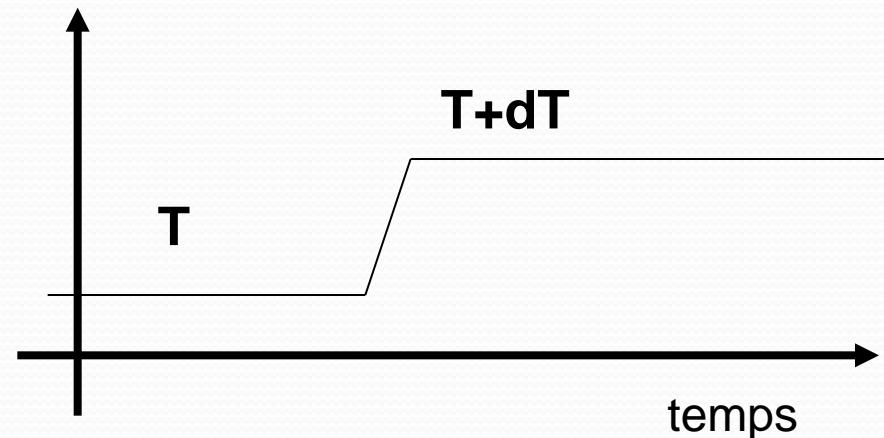
APPLIED PHYSICS LETTERS 98, 133105 (2011)

Heat capacity measurement

- Experimental notion of specific heat
- Adiabatic calorimetry
- Sample infinitely thermally isolated
- Real system coupled to the heat bath through K:

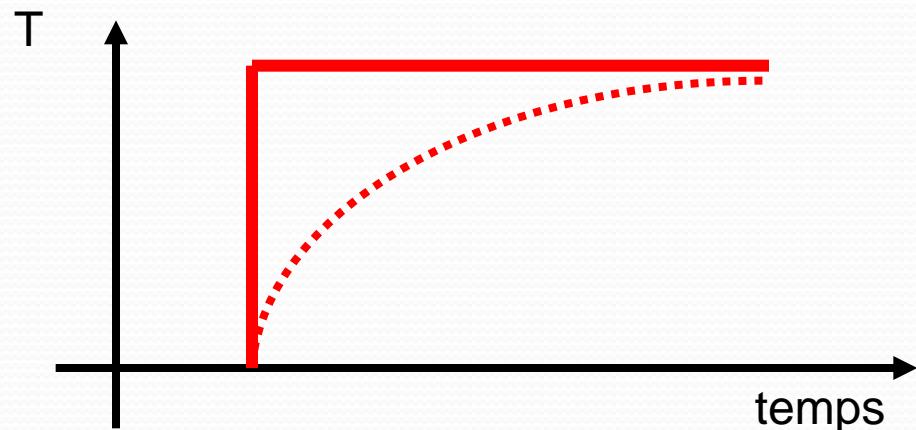
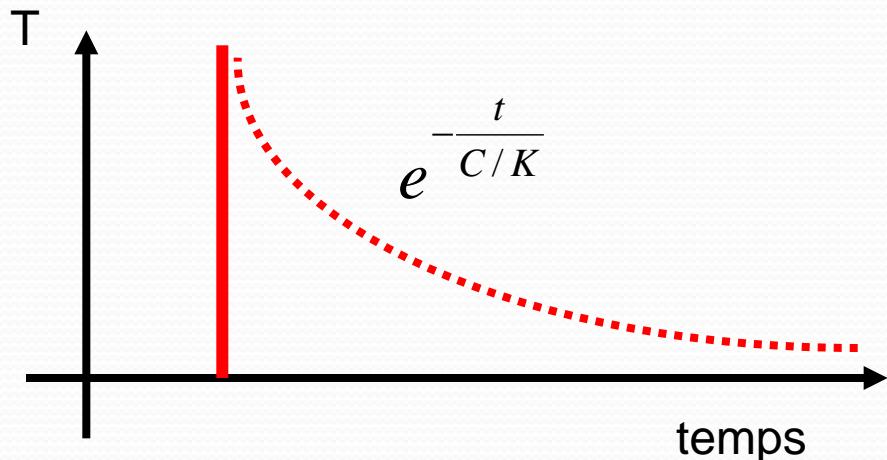
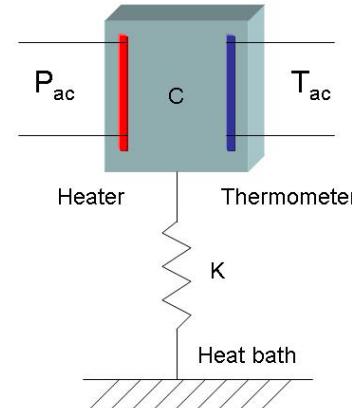


$$\tau = \frac{C}{K}$$



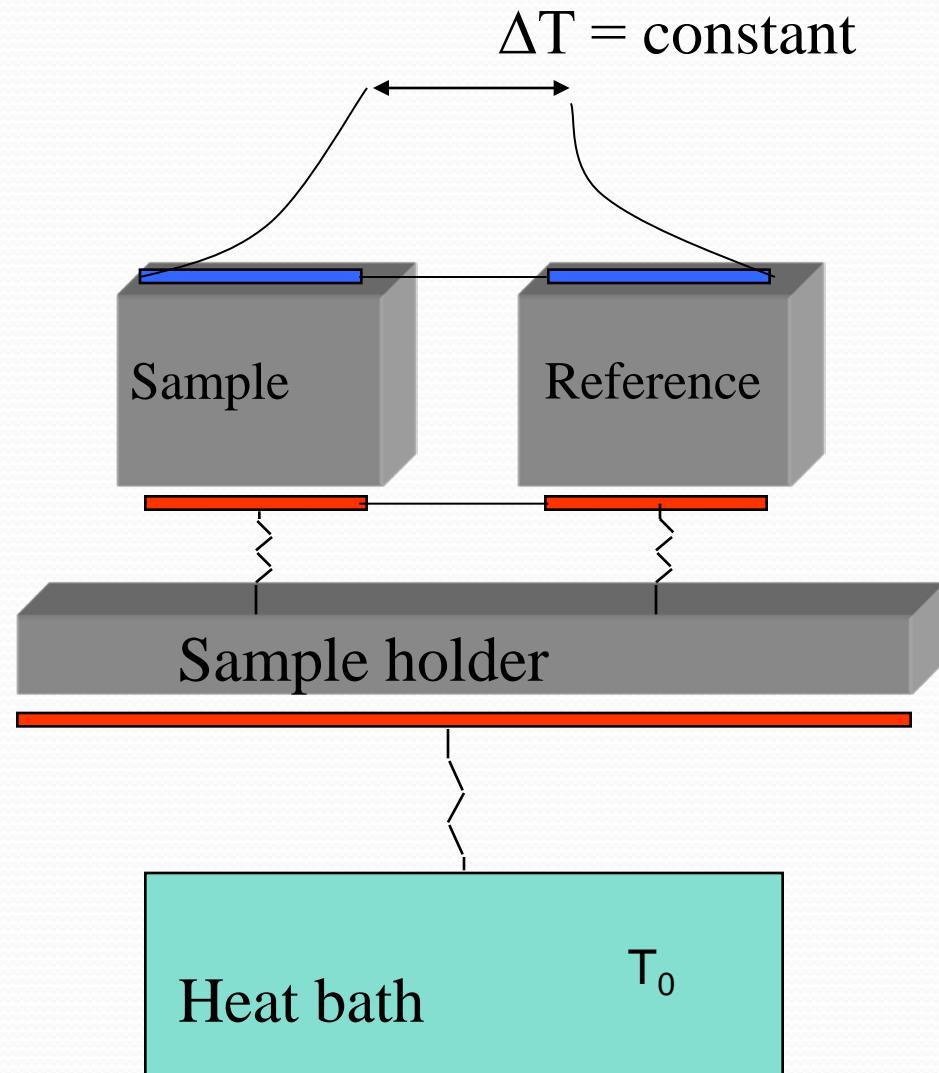
Relaxation calorimetry

- Highly coupled to the heat bath
- Heat Pulse: exponential decrease of the temperature
- Heat step function: measurement of the thermalization of the system
- C/K: knowledge of K is necessary
- Limited by thermal drift



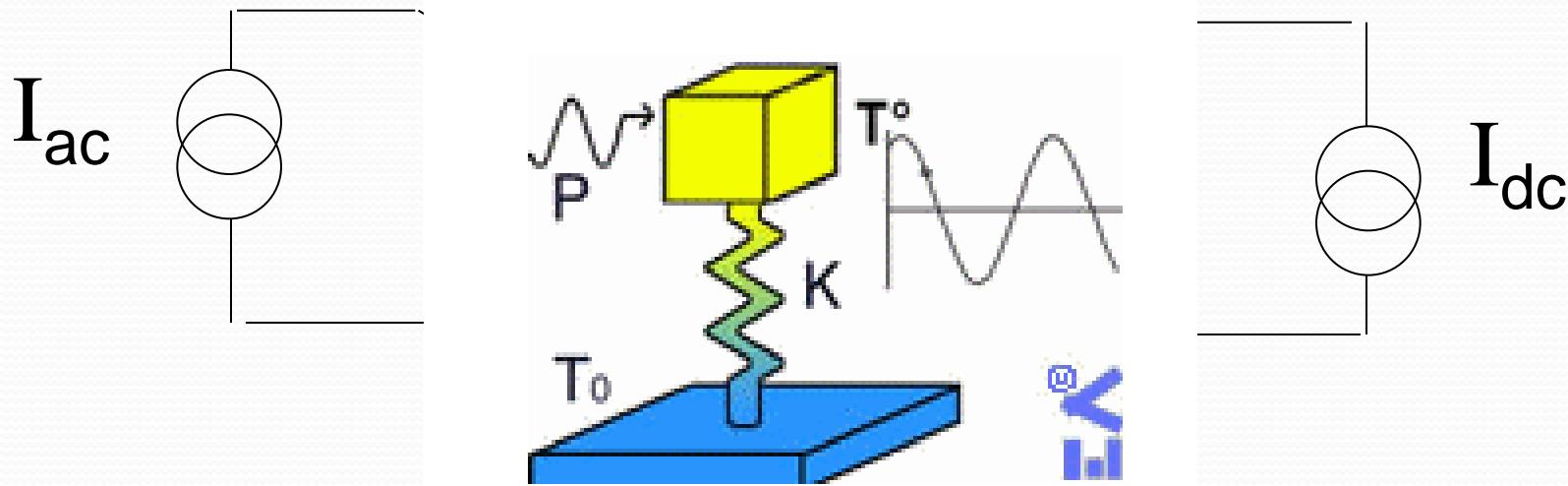
Differential Scanning Calorimetry (DSC)

- Not very used at low temp.
- Sensitivity related to the temperature scanning rate
- Get rid of temperature drift



Calorimetry by temperature oscillation (AC calorimetry)

- Principle: ac current in the heater at the frequency f
- Oscillation of temperature measured on the thermometer at $2f$
- Not sensitive to the thermal drift of the heat bath



P.E. Sullivan, G. Seidel, Phys. Rev. 173, 679 (1968)

Some equations...

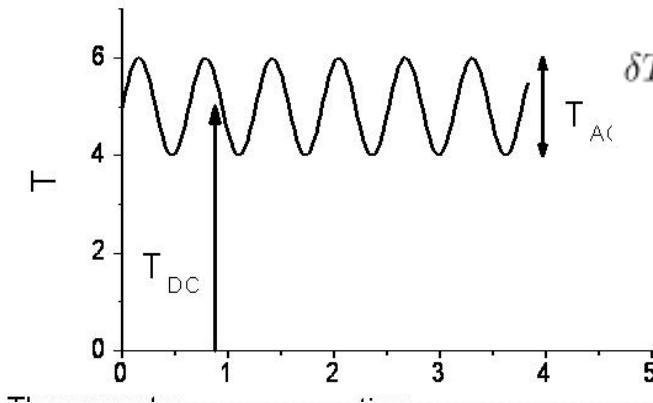
- Thermal balance
- Restriction on the working frequency:

$$1/\tau \ll f_{meas} \ll 1/\tau_{diff}$$

$$C \frac{dT}{dt} = P_{Heat} - K(T - T_B)$$

$$P_{Heat} = RI_0^2/2(1 + \cos(2\omega t))$$

$$T(t) = T_B + \frac{RI_0^2}{2K} + \delta T_{ac}^{Heater} \quad \text{avec} \quad \delta T_{ac}^{Heater} = \frac{RI_0^2}{2\omega C} \left(1 + \frac{1}{(\omega\tau)^2}\right)^{-1/2} \cos(2\omega t + \varphi)$$



$$\delta T_{ac}^{thermo} = \frac{RI_0^2}{2\omega C} \left(1 + \frac{1}{(\omega\tau)^2} + (\omega\tau_{diff})^2\right)^{1/2} \cos(2\omega t + \varphi)$$

$$\delta T_{acRMS}^{thermo} = \frac{RI_0^2}{2\omega C}$$

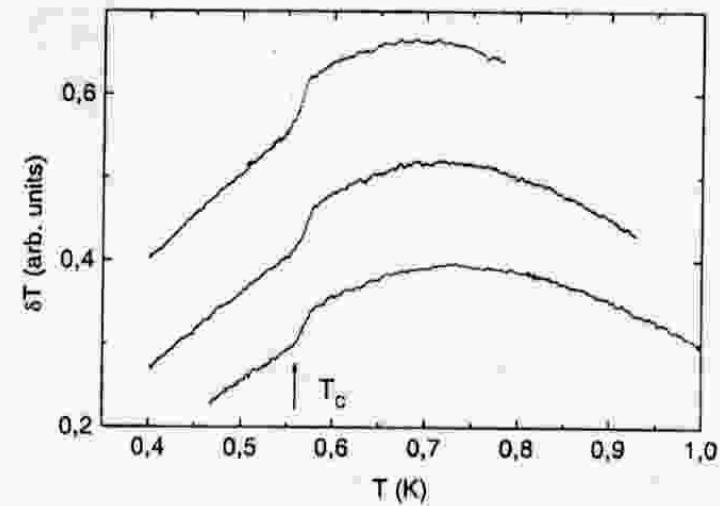
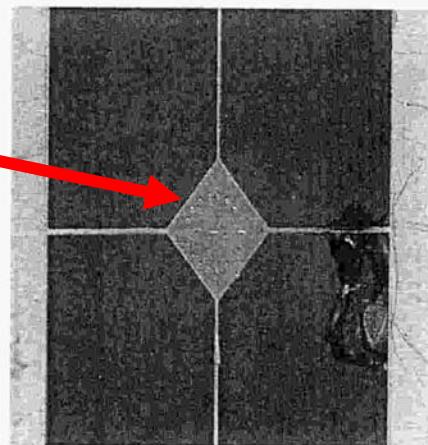
ac calorimetry on membrane

- Integration at the micro scale
- Very high sensitivity (thermometer)
- Adapted for thin films and micro-crystal
- Silicon, silicon nitride, diamond, AsGa

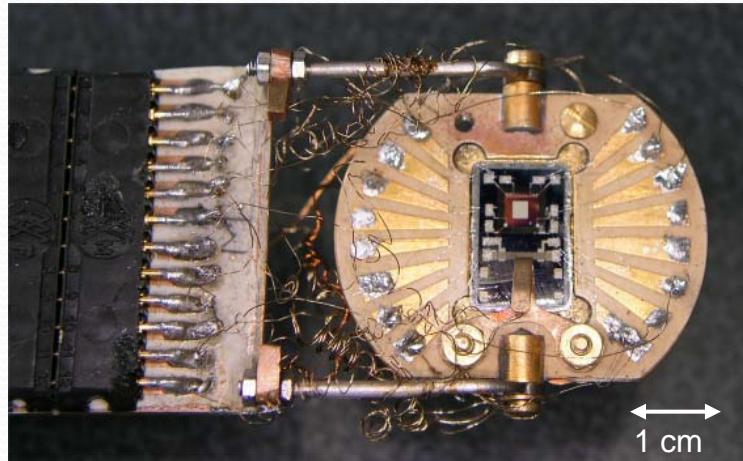
Measurement Jukka Pekola group by ac calorimetry (University of Jyväskylä)

- Previous experiment of specific heat measurements on mesoscopic samples:
A. Lindell *et al.* Physica B 284-288, 1884 (2000) and theoretical calculations (Heat capacity of mesoscopic superconducting disks, P. Singha Deo *et al.* Europhys. Lett. 50, 649 (2000)).

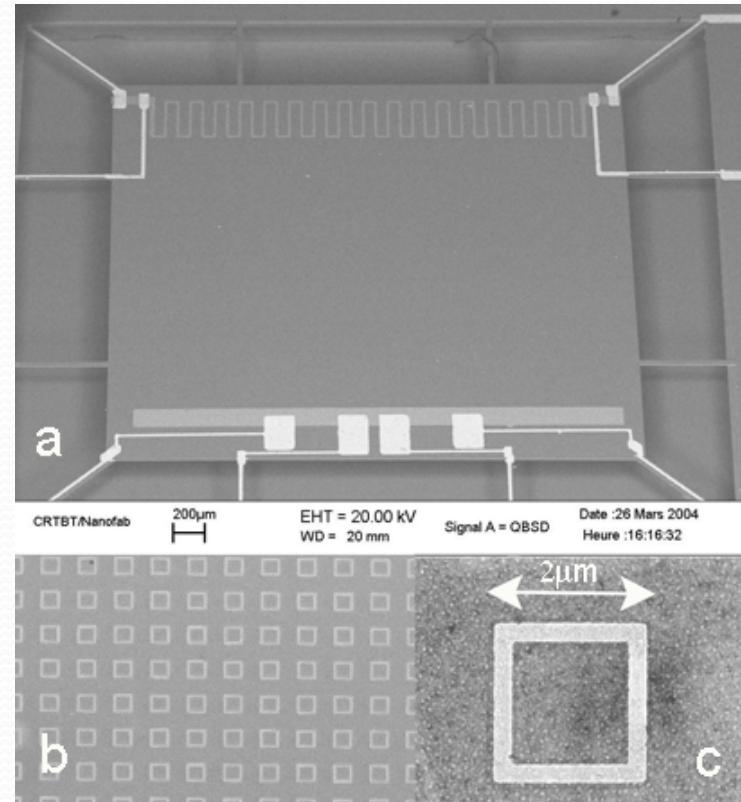
14 Titanium disks, on a silicon nitride membrane



AttoJoule calorimetry



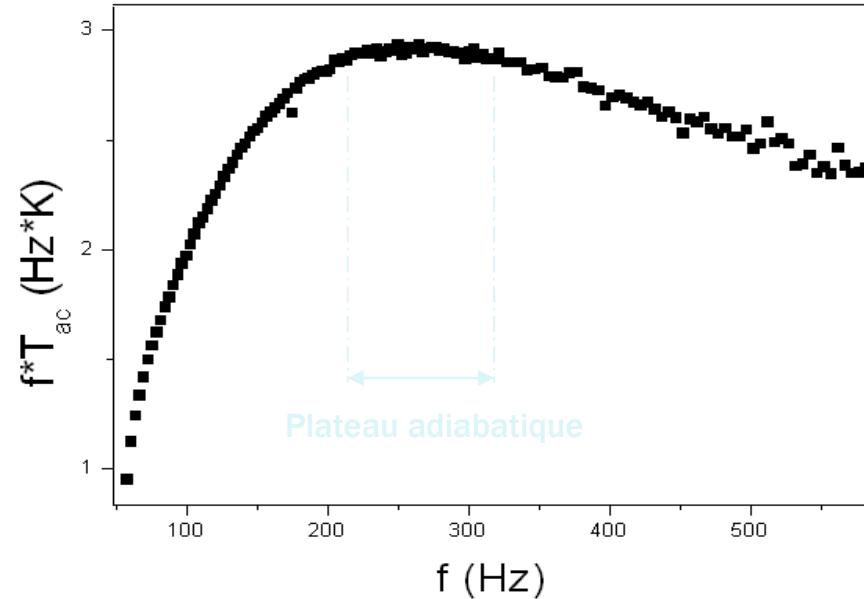
- Thickness of the membrane : few microns.
- 4mm² available surface.
- Between 5x10⁵ rings of radius 1mm or 10⁷ rings of radius 100nm.
- Ebeam lithography of a high number of independent system : 10⁵/10⁷.



O. Bourgeois, F. Ong, S. Skipetrov and J. Chaussy, PRL 94 057007 (2005)

Working Frequency and performance

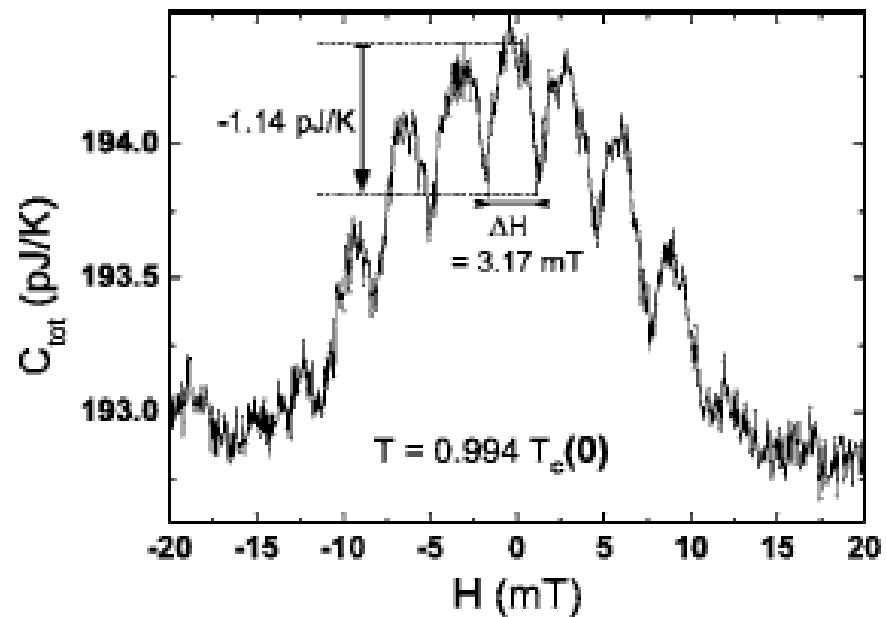
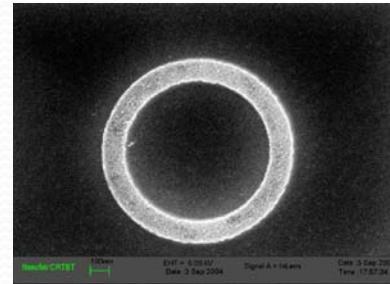
- Finding the working frequency ?
- Performances:



- AC calorimetry, oscillation de température: $\Delta T=10\text{mK}$.
- Resolution: 5×10^{-5}
- Sensitivity en C_p : 10^{-15} J/K
- **Sensitivity/ μm^2 :** $5 k_B$.

C_p of mesoscopic systems

- Observation of the magnetic flux quantization in a supraconducting mesoscopic ring
- Thermal signature of vortex entry in the ring
- “Little Parks effect” in the specific heat

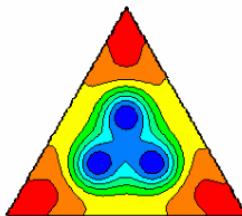
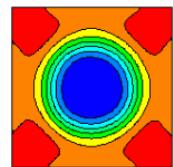
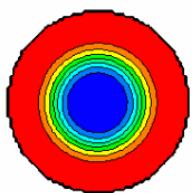


Avalanche of specific heat jump in superconducting disks

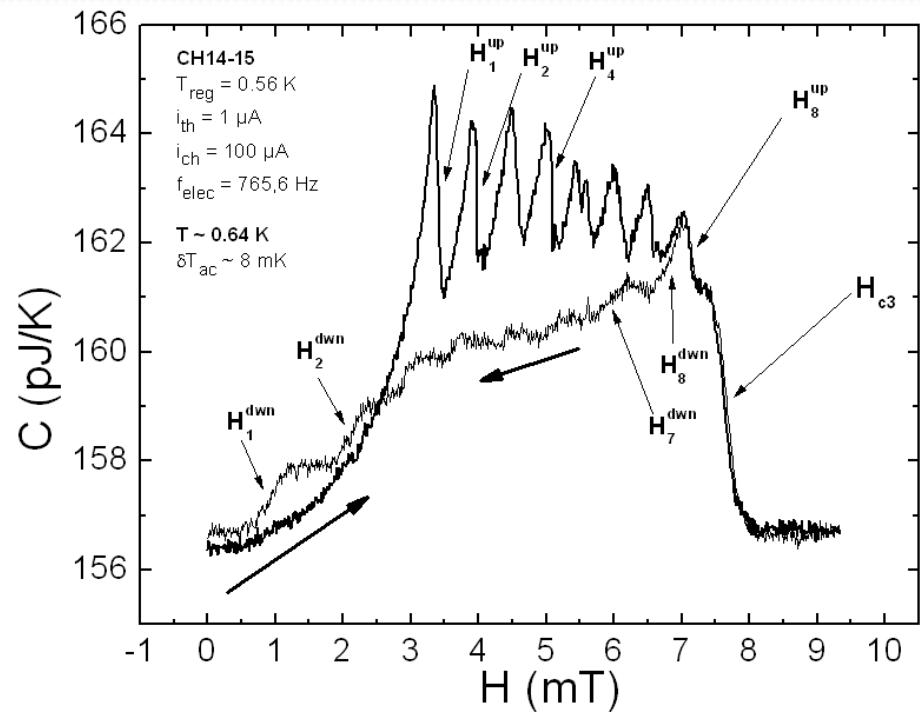
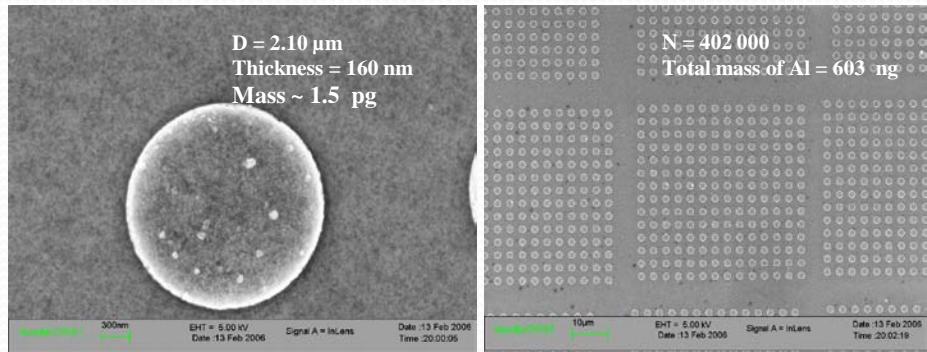
- Cp jump (2nd order phase transition) depend on the topology of the mesoscopic system
- Induced by the application of an external magnetic field (vortex)

Vortex configurations in disks, squares and triangles

(Surface $S = \pi R^2$, $R = 4.0\xi$, $d = 0.1\xi$, $\kappa = 0.28$)



Condensed Matter Theory
University of Antwerp (UIA)



F.R. Ong and O. B, *Topology effect on the heat capacity of mesoscopic superconducting disks*, Europhys. Lett. **79**, 67003 (2007).



Silicon nitride membrane Roukes group in Caltech

- Relaxation calorimetry, amplitude of the heat pulse: $\Delta T=100\text{mK}$.
- Resolution: 10^{-4}
- Sensivity: $5 \times 10^{-19} \text{ J/K}$
- Sensivity / μm^2 : $100 k_B$.

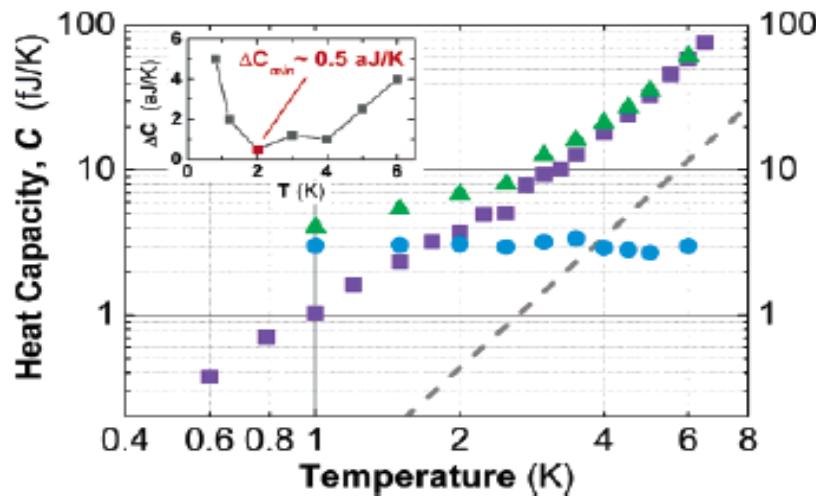
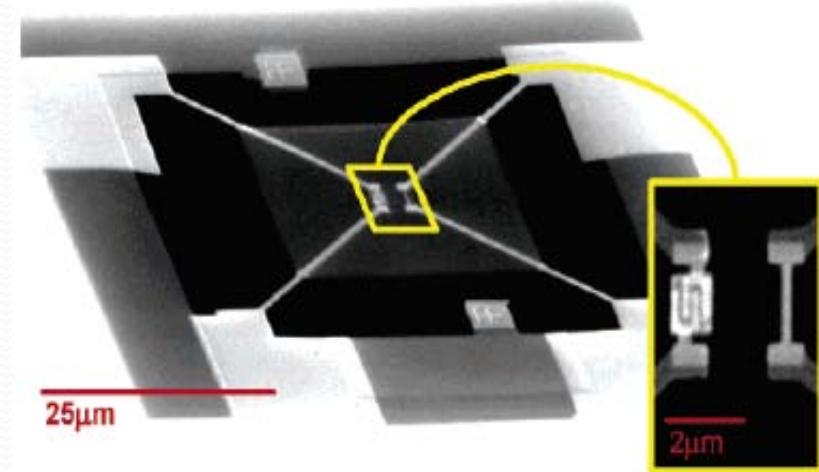


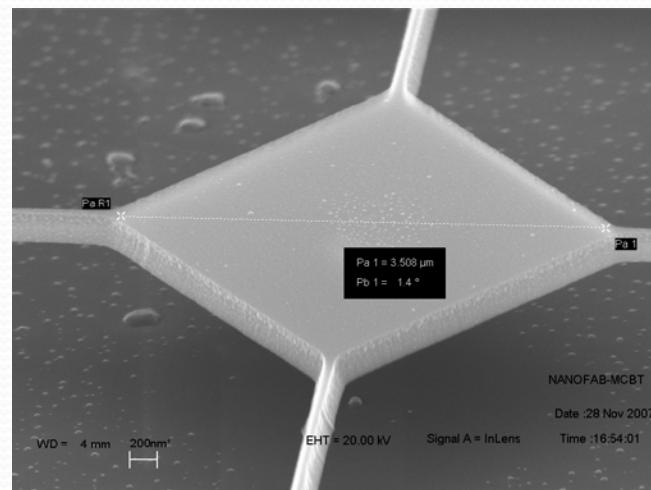
Figure 4. The heat capacity of the calorimeter (violet), the heat capacity of calorimeter with adsorbed He gas film (green), and the heat capacity of the He gas film (blue). The He gas coverage is ~ 0.16 monolayer over the device. The dashed gray line represents the estimated Debye phonon heat capacity of the calorimeter. Inset: Measurement resolution attained at various temperatures by the calorimeter for 5 s averaging time. Highest resolution, $\Delta C \sim 0.5 \text{ aJ/K}$ ($\sim 36000 k_B$), is attained at 2 K in these experiments.



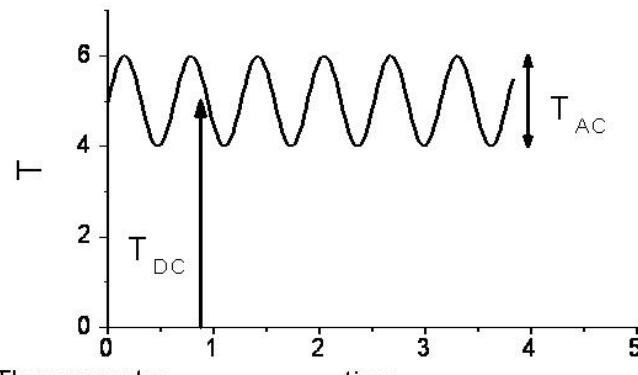
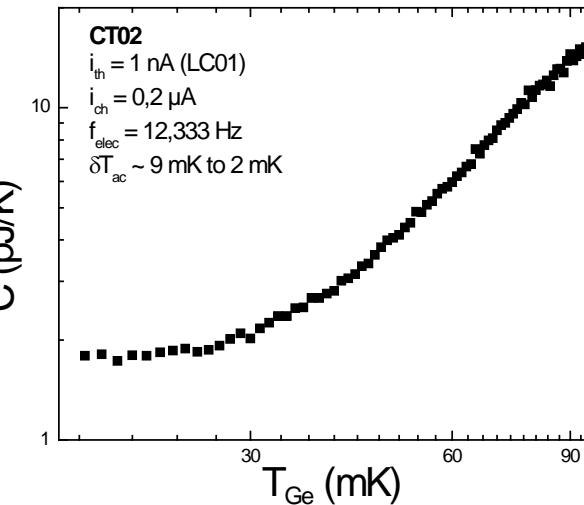
Fon et al., NanoLetters 2005

Specificity of Cp measurement at dilution fridge temp. $T < 100\text{mK}$

- Problem of significant ΔT_{DC} (can be a limitation)
- Thermometer and heater (by ebeam litho)
- SOI: thickness of the membrane 200nm
- Possibility of measurement of single object
- $C = 10^{-20}/10^{-21}\text{J/K}$
- Sensitivity close to 1 k_B (10^{-23} J/K)

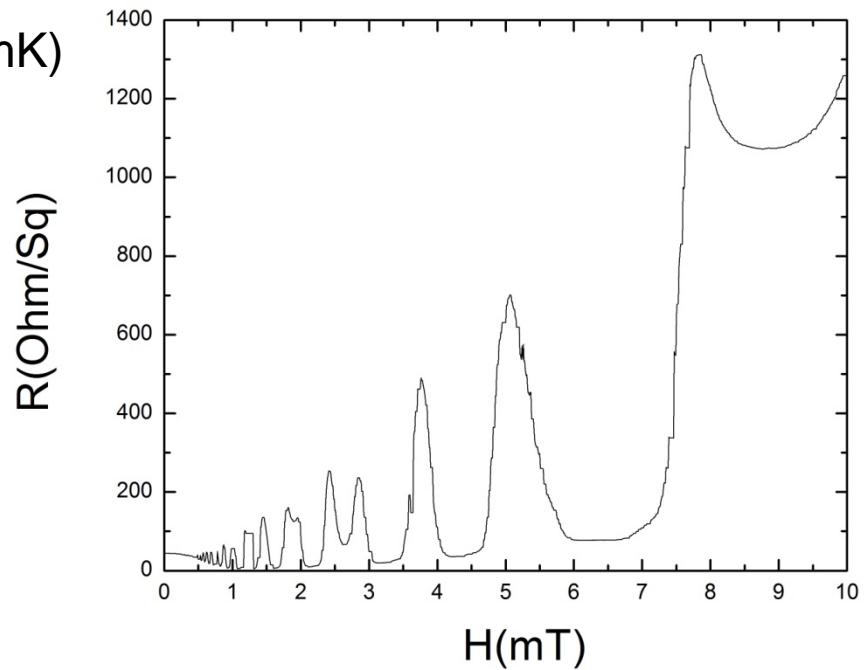
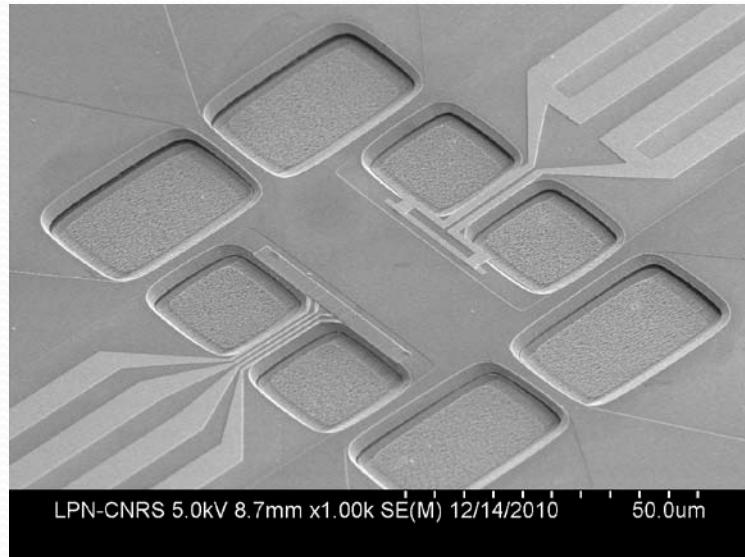


(a)



Measurement on AsGa heterostructure: 2DEG

- Even suspended the 2DEG has very good characteristic: mobility $4 \times 10^5 \text{ cm}^{-2}$
- Allow thermal measurement of one layer (superlattices)
- C in the frac., int. QHE (12T below 100mK)



With D. Kazazis and U. Gennser (LPN)

At high temperature : thermal sensor based on polymer membrane

A. Lopeandia, ...O. B. , Rev. Sci. Instrum. **81**, 053901 (2010)

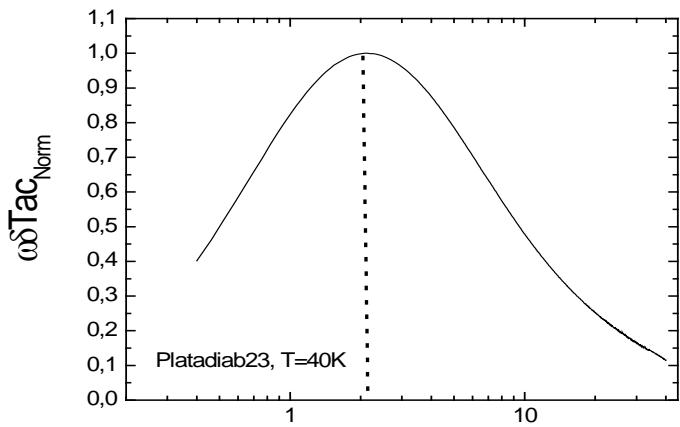
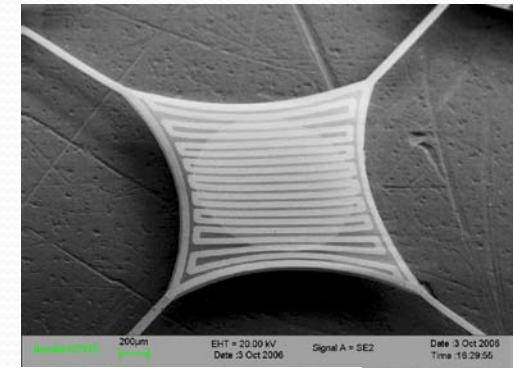
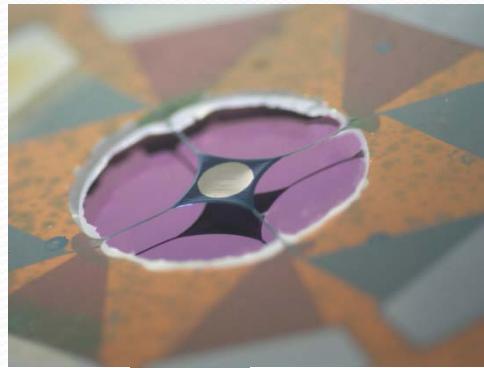


Fig. 1. Plateaux quasi-adiabatique

Resolution

$$\frac{\Delta C}{C} \approx 10^{-4} \approx 0,01\%$$

$$C = 1 \mu J / K$$

Course nanothermal Phys

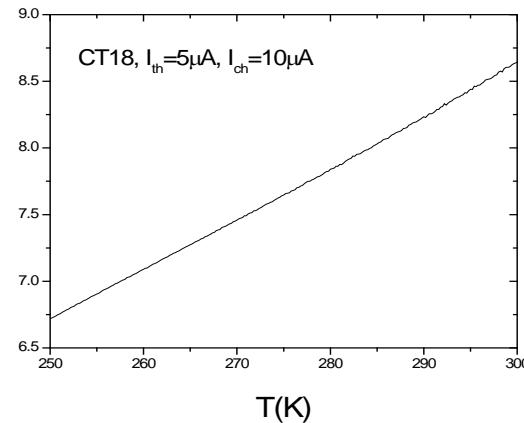


Fig. 2. Specific heat measurement versus temperature

$$\Delta C \approx 5 \cdot 10^{-9} J / K$$

$$\Delta E = 10^{-12} J = 1 pJ$$

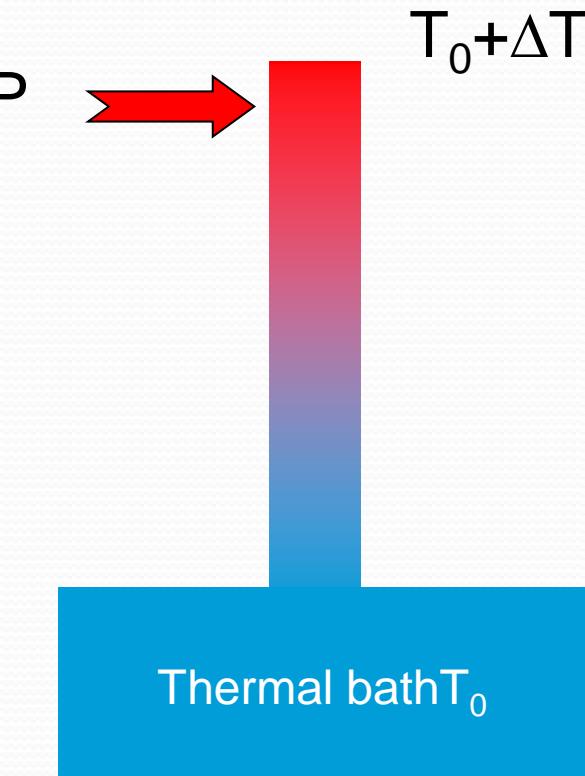
Sensitivity

Measurement of thermal conductance

Thermal conductance

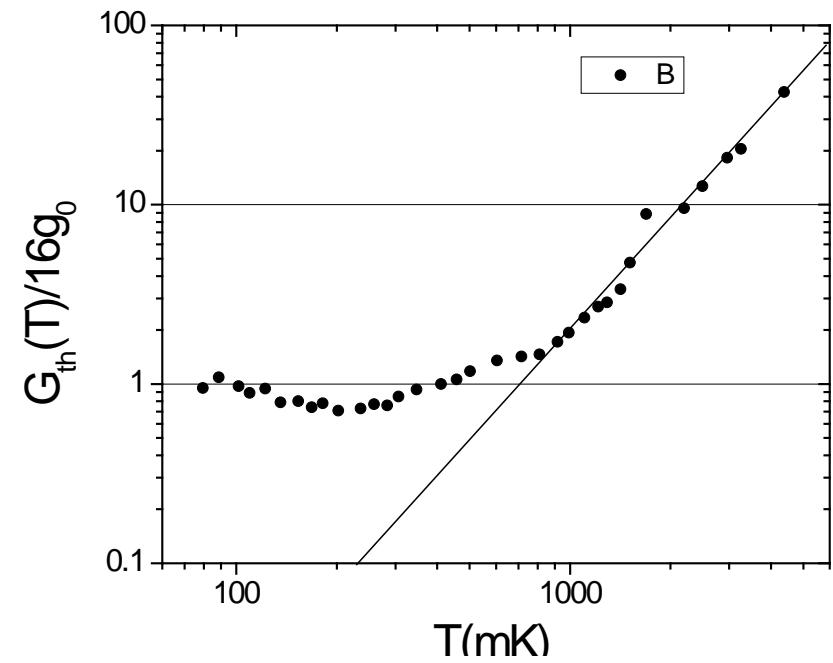
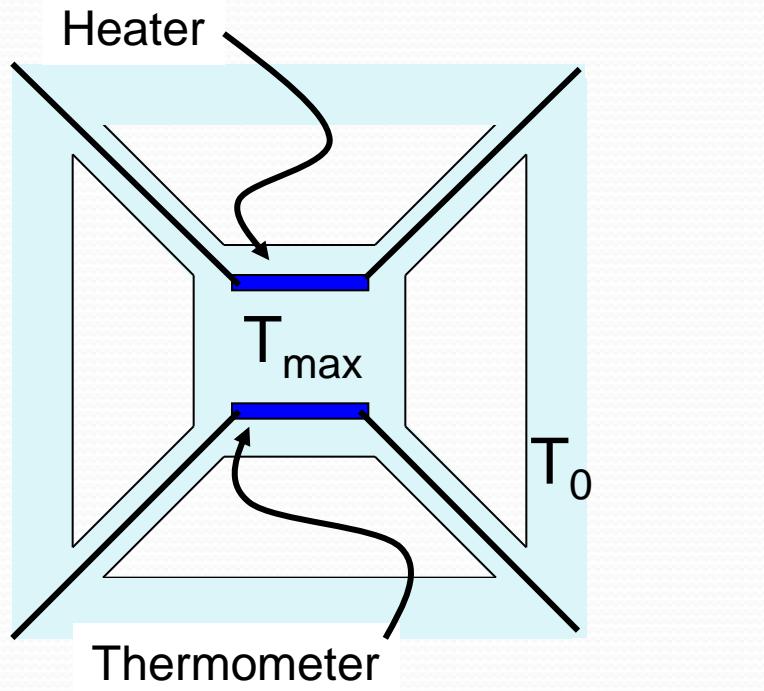
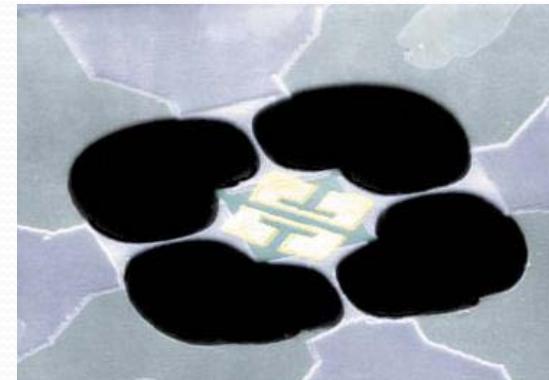
- Known power
- Measure of ΔT
- K is a mean value integrated over ΔT
- Dynamic measurement by modulating P
- Nanowire ??
- Connection to heat bath and to thermometer
- Thermal contact

$$K = \frac{P}{\Delta T}$$



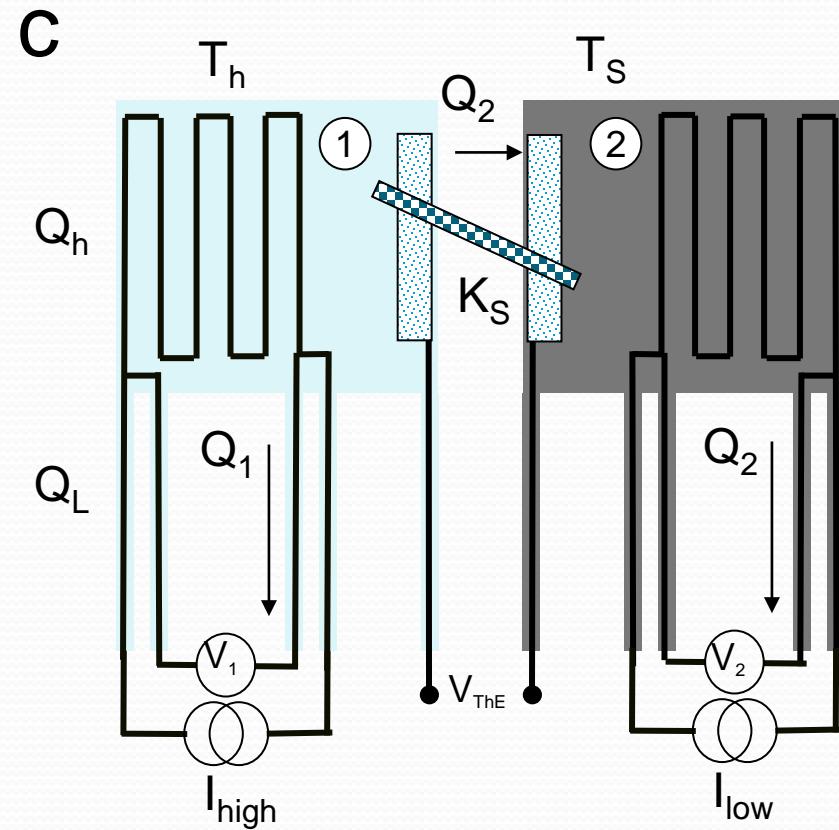
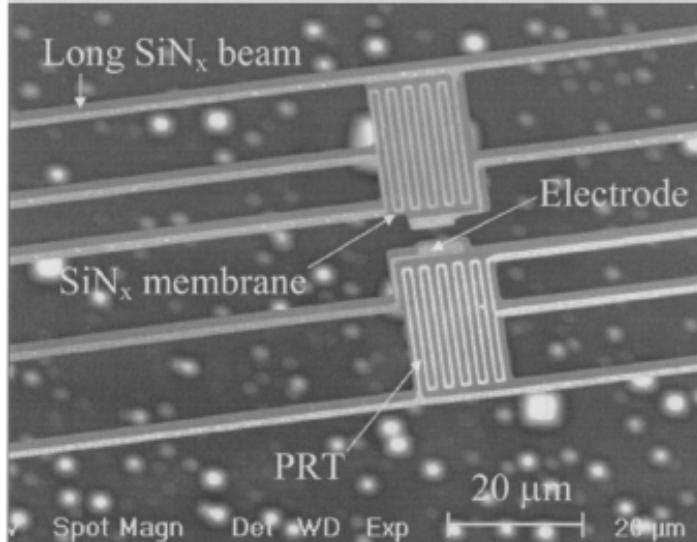
Universal thermal conductance

- Continuous measurement based on a thermal gradient
- Beautiful experiment (not yet reproduced)
- Transmission coefficient optimized through profiled contact (See Rego and Kirczenow)
- Superconducting leads ??



L.Shi/A. Majumdar method

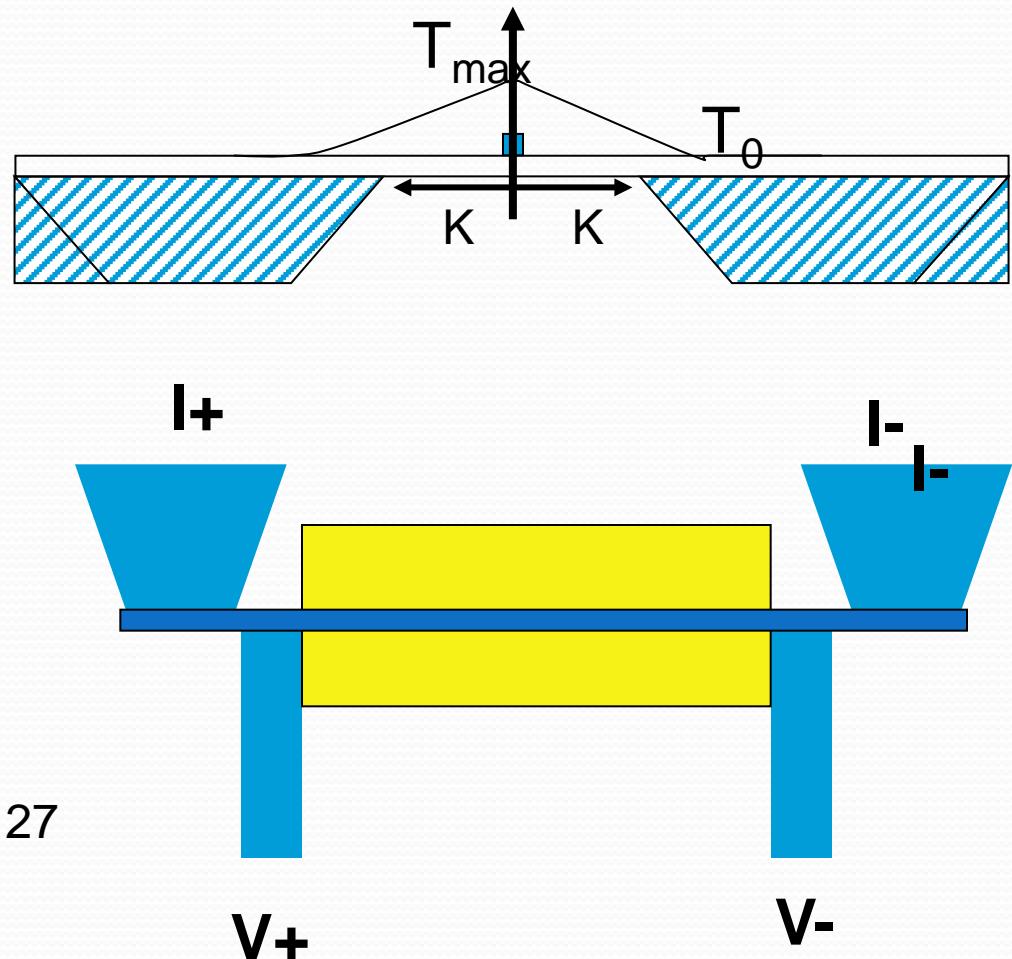
- Measure of K and C and of the thermoelectric power
- Adapted to self grown nano-objects
- Major difficulty: the thermal contact between the nanowire and the heat bath



L. Shi,, P. Kim, A. Majumdar, Journal of Heat Transfer **125**, 881 (2003)

Hot wire method: for thin film and membrane

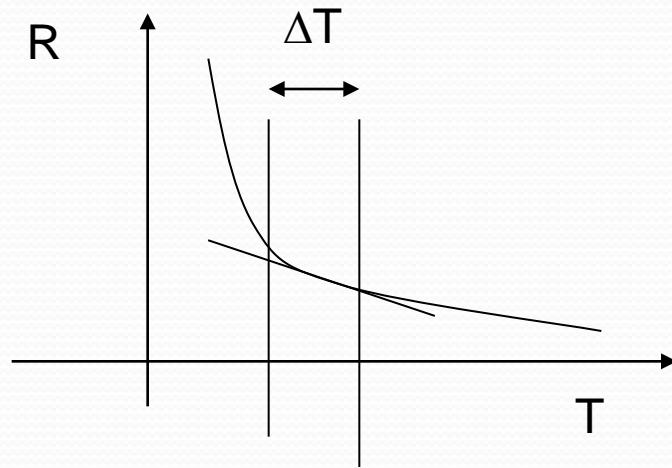
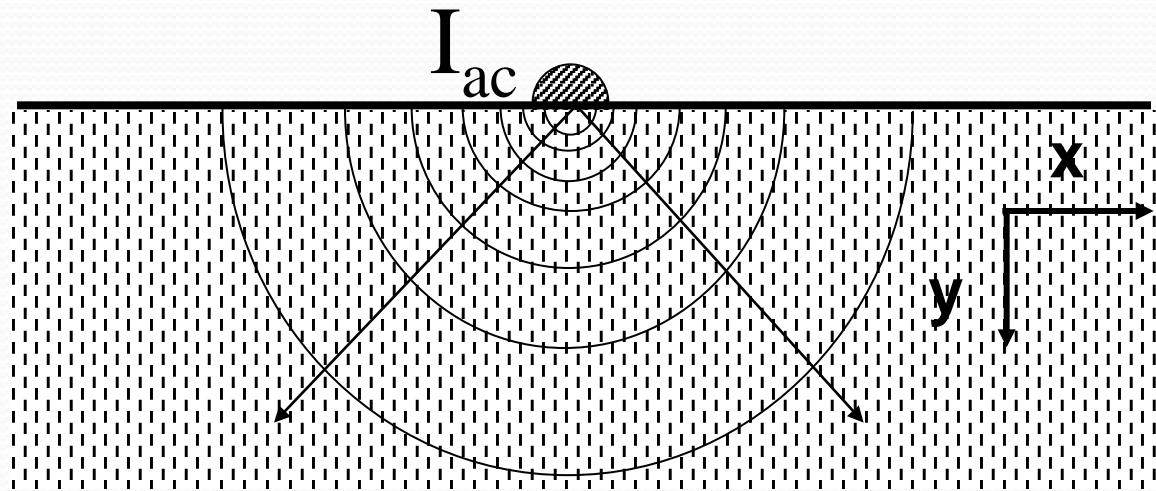
- In plan thermal conductance measurement
- Diff. method 3ω (Cahill)
- Silicon nitride membrane



F. Völklein, Thin solid Films **188**, 27
(1990).

3ω method heat transport perpendicular to the plane (radial)

- Heater=thermometer
- Thermal conductance measurement at low frequency (Cahill RSI 1990)
- Heat capacity measurement at high frequency (Birge & Nagel (RSI 58, 1464 (1987)))



- $I_{ac} \sim 1\omega$
- $T_{ac} \sim I^2 \sim 2\omega$
- $R \sim T \sim 2\omega$
- $V_{3\omega} \sim I_{ac}R \sim 3\omega$

D. G. Cahill, Rev. Sci. Instrum. **61**, 802 (1990)

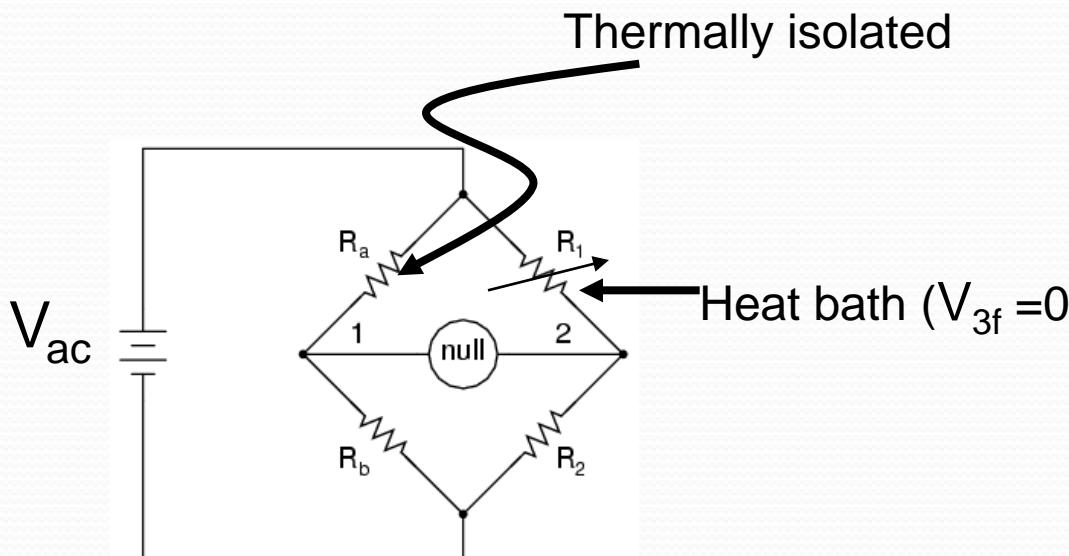
J.-Y. Duquesne, Phys. Rev. B. **79**, 153304 (2009)

Wheatstone bridge/differential bridge

- Limit $V_{if} \gg V_{3f}$
- R_a is thermally isolated and R_1 is the reference Wheatstone Bridge (cannot be done in the radial configuration)
- differential bridge

$$I = I_0 \cos(\omega t)$$

$$V_{Total} = (R_0 + \left(\frac{dR}{dT} \right) (T(t) - T_0)) I_0 \cos(\omega t)$$



$$\Delta T(r) = \frac{RI_0^2}{l\pi K} J_0\left(\frac{r}{\lambda_h}\right)$$

Method of measurements

3ω method

Metal line heated with $I_{AC} = f(\omega) = A \sin(\omega t)$

-> Dissipated power: $R I_{AC}^2 = f(2\omega)$

-> Resistance variation = $f(2\omega)$

-> Voltage variation = $f(2\omega) \cdot f(\omega)$

-> Measured voltage: ohmic part $V(\omega)$ + thermal part $V(3\omega)$

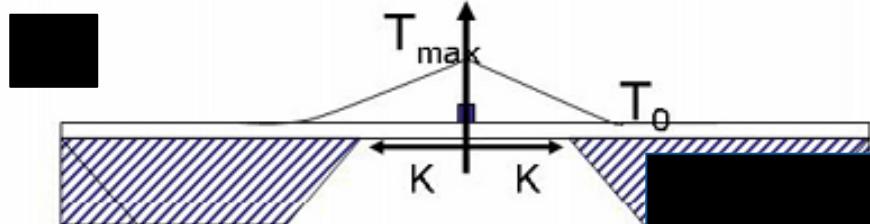
thermometer/heater

Sample

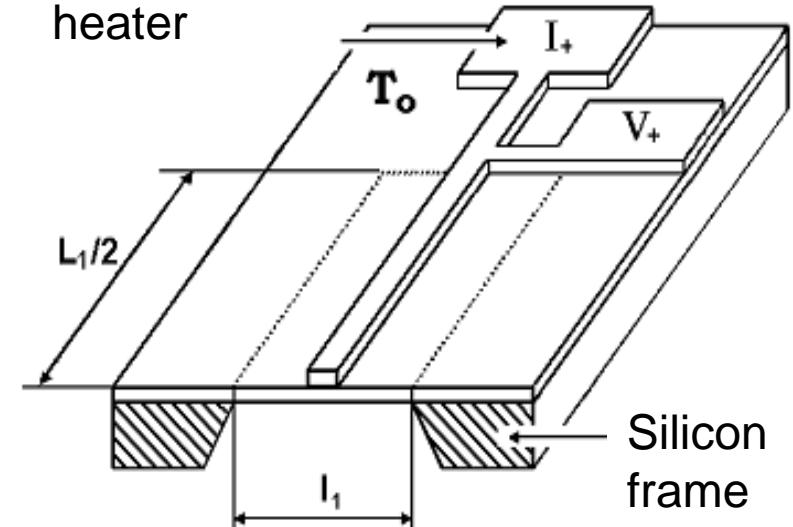
substrate

Völklein method

-Direct current



heater



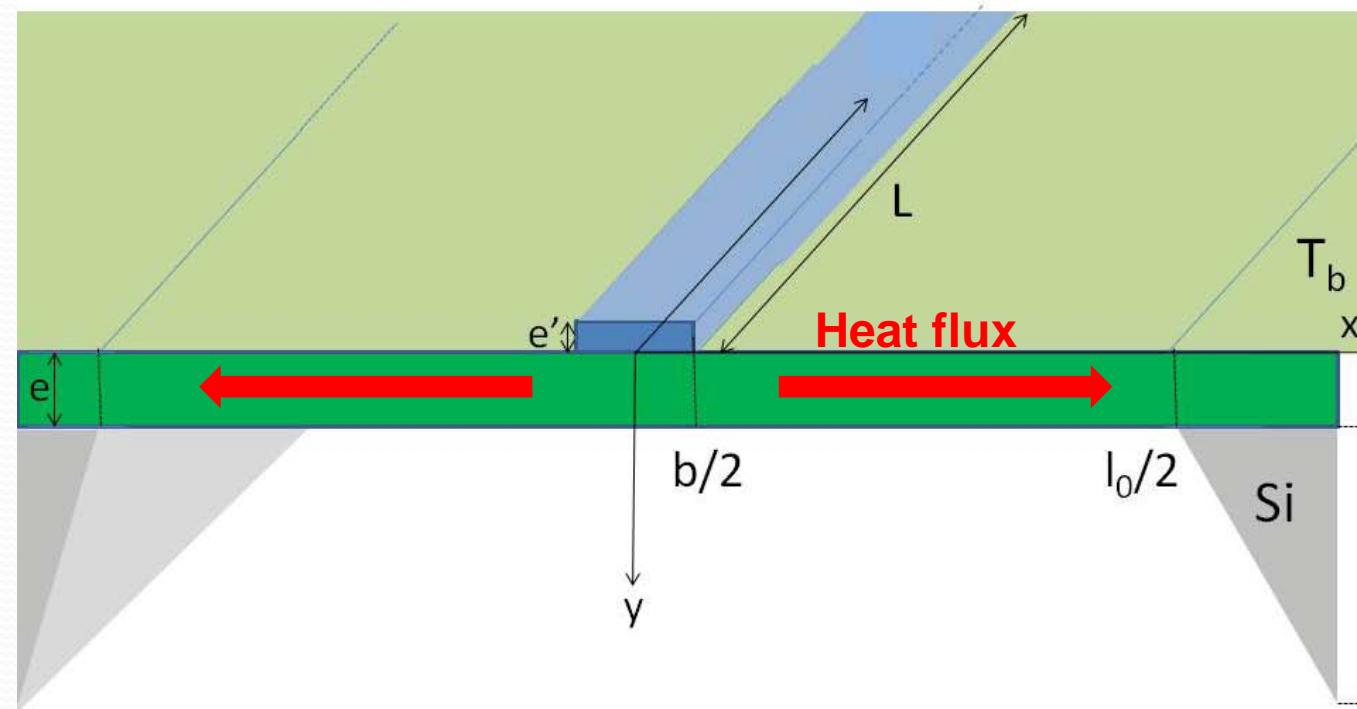
F. Völklein, Thin solid Films 188, 27 (1990).

A. Jain and K. E. Goodson, Journal of Heat Transfer 130, 102402 (2008)

Heat balance: measurement of the thermal conductance in the plane

3ω / Völklein method: 3ω on membrane

$$V(3\omega) = f(K, C)$$



$$\begin{aligned} e &= 100/400 \text{ nm} \\ e' &= \sim 50 \text{ nm} \\ b &= \sim 20 \mu\text{m} \\ L &= \sim 1 \text{ mm} \\ l_0 &= 150/500 \mu\text{m} \end{aligned}$$

NbN thermometer: high sensibility, TCR can be tailored for a temperature range.
Bourgeois et al., Review of Scientific Instrument 77 2006

A. Jain and K. E. Goodson, Journal of Heat Transfer **130**,
102402 (2008).

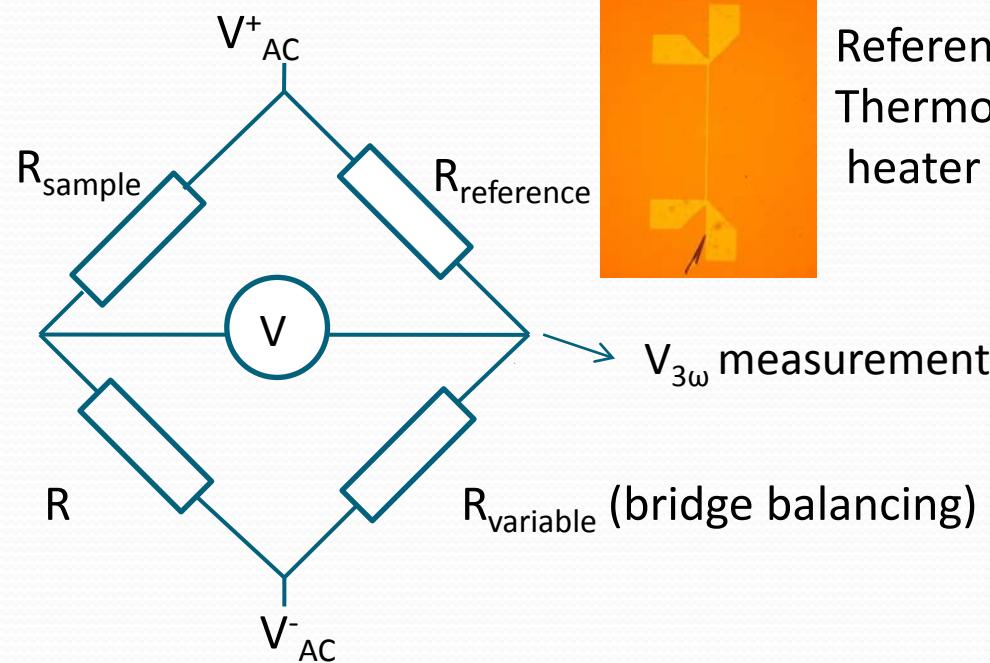
Signal treatment: Wheatstone bridge

Problem: $V(\omega) \gg V(3\omega)$, $V(3\omega)/V(\omega) \approx 3 \times 10^{-4}$



Minimisation of $V(\omega)$ voltage thanks to a **Wheatstone bridge**

Membrane +
NbN
Thermometer/
heater

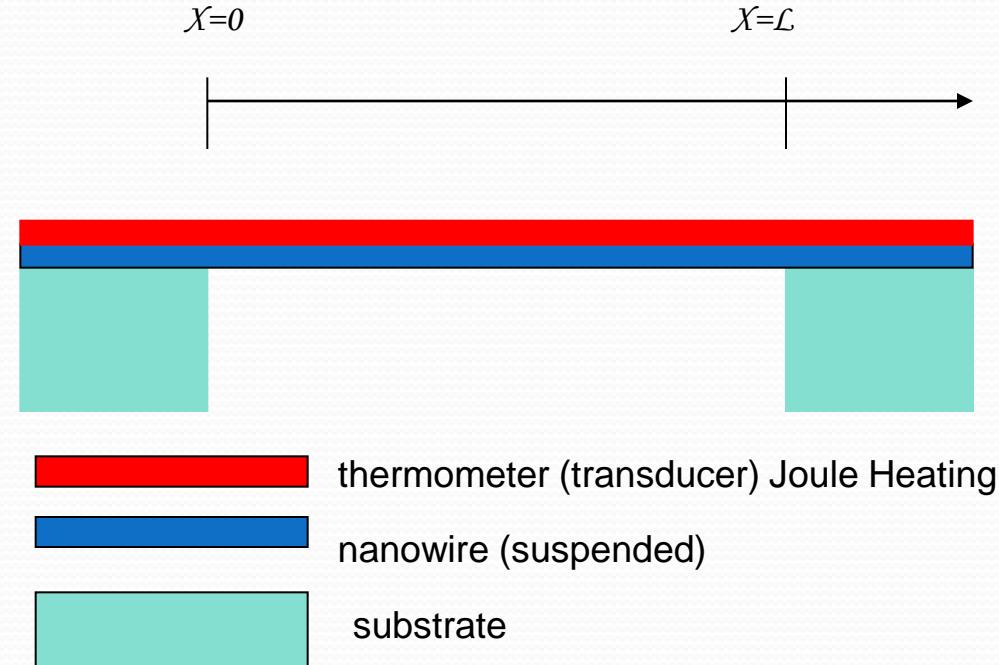
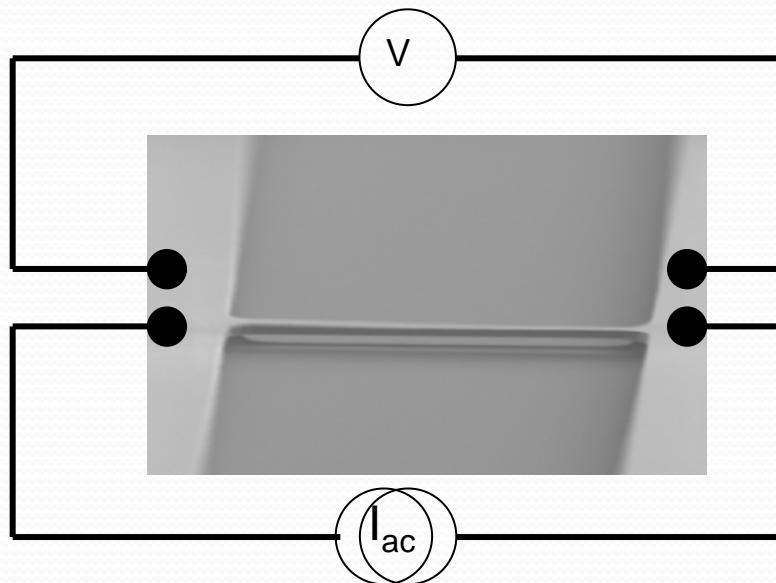


Reference:
Thermometer/
heater NbN

After balancing: $V(3\omega)/V(\omega) \approx 4 \times 10^{-1}$ (2V)

3ω method Measure in plane (longitudinal)

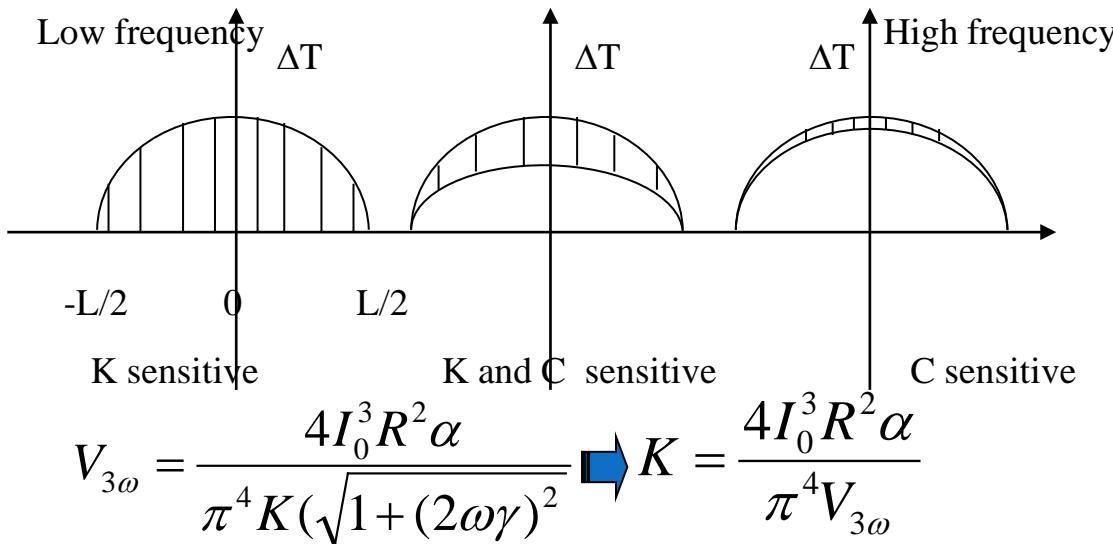
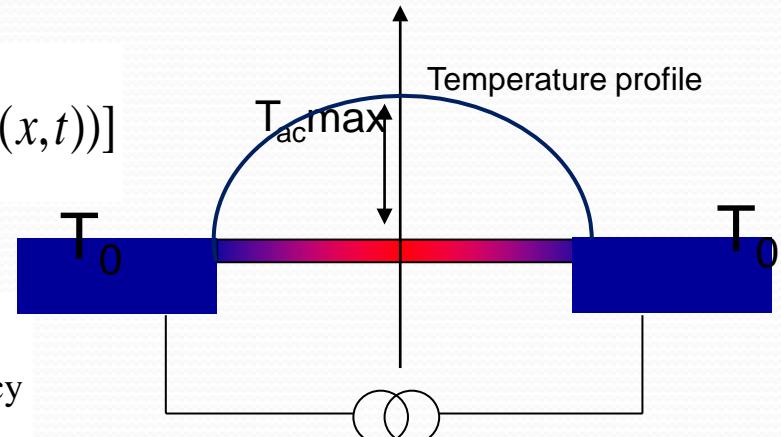
- Proposée par Lu et coll.
- Adapté pour nanofils suspendue
- Limite basse fréquence



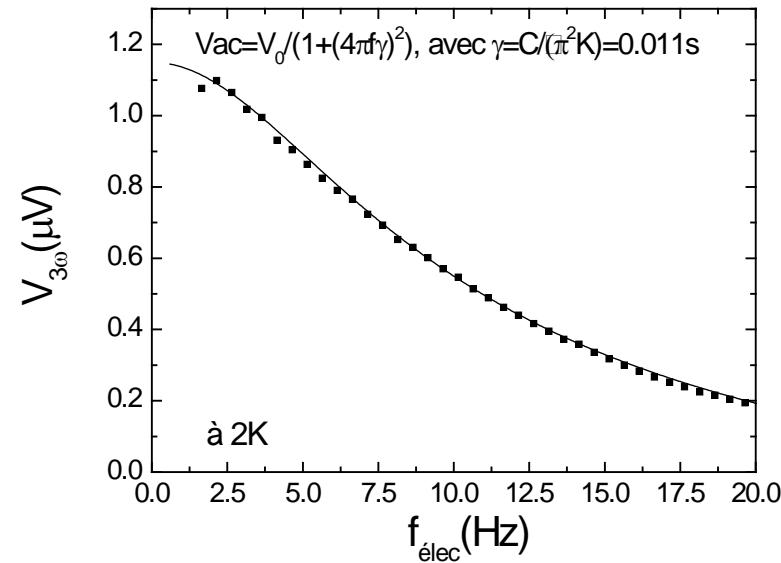
Lu L, Yi W and Zhang DL 2001 *Rev. Sci. Instrum.* **72** 2996

Principle of the measurement

$$\rho C_p \frac{\partial}{\partial t} T(x,t) - k \frac{\partial^2}{\partial x^2} T(x,t) = \frac{I_0^2 \sin^2(wt)}{LS} [R + \left(\frac{dR}{dT}\right)_{T_0} (\Delta T(x,t))]$$

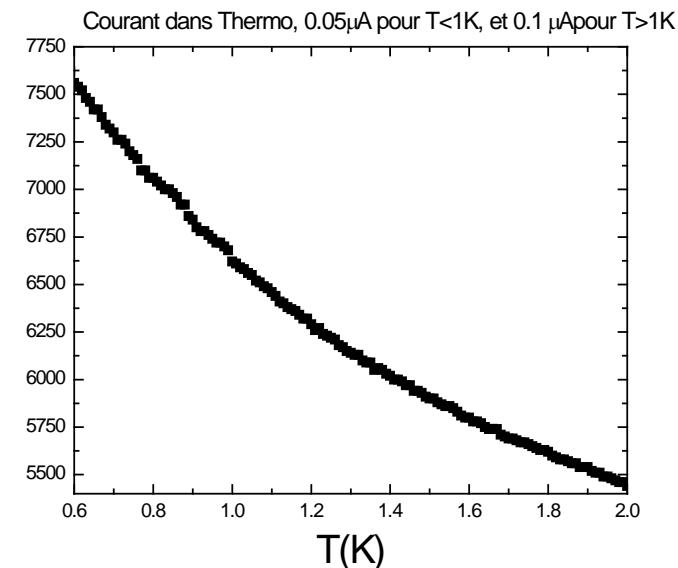
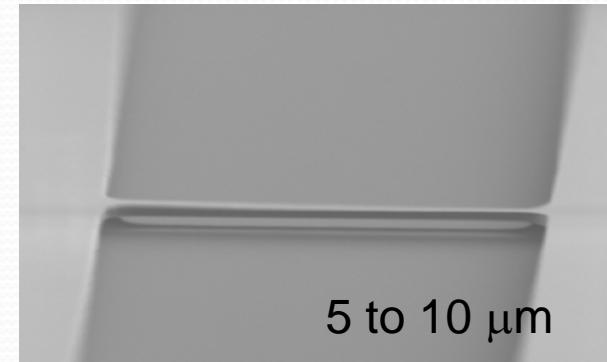
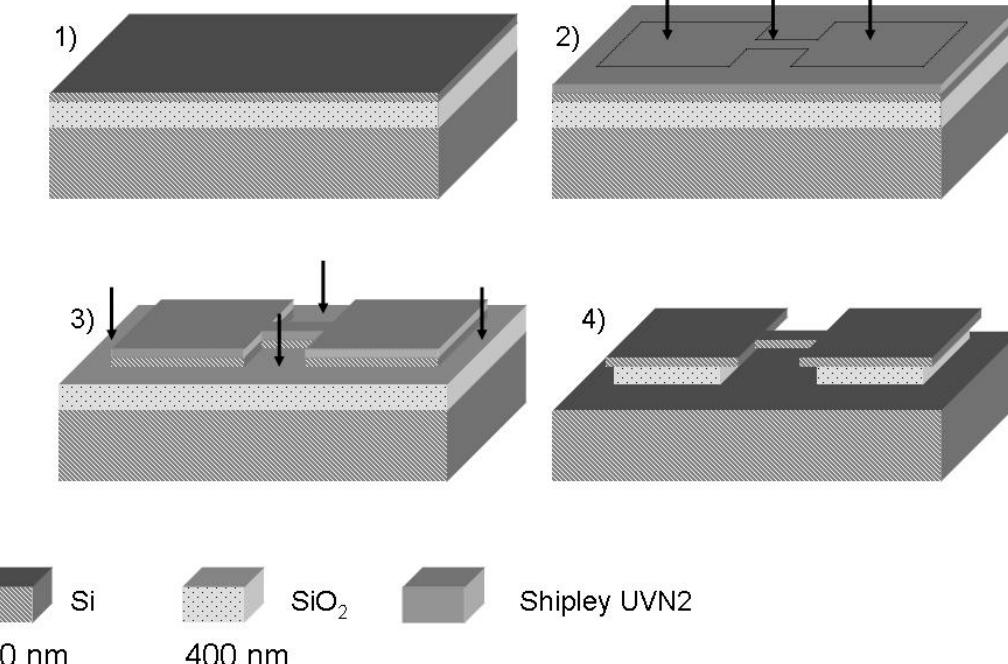


O. Bourgeois, Th. Fournier and J. Chaussy, J. Appl. Phys, **101**, 016104 (2007)



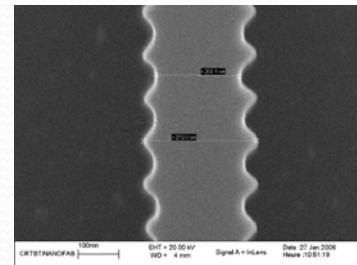
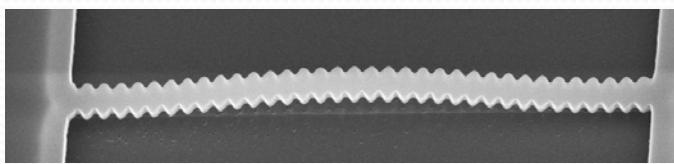
Fabrication of suspended nanowire

- The fabrication of the silicon nanowire are realized by e-beam lithography at the LETI on SOI substrate.
- Numerous different wires can be built from 50 to 200 nm of cross-section by 100 nm and 130 nm thick.
- A NbN transducer is deposited on top.



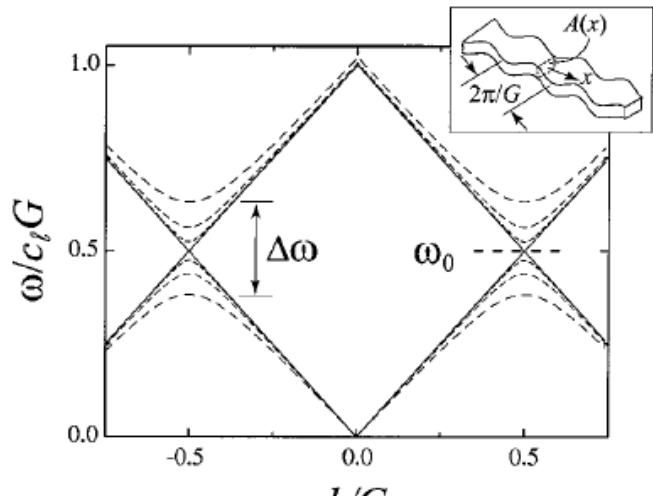
Phononic crystal

Specific geometries can modify the conductance:



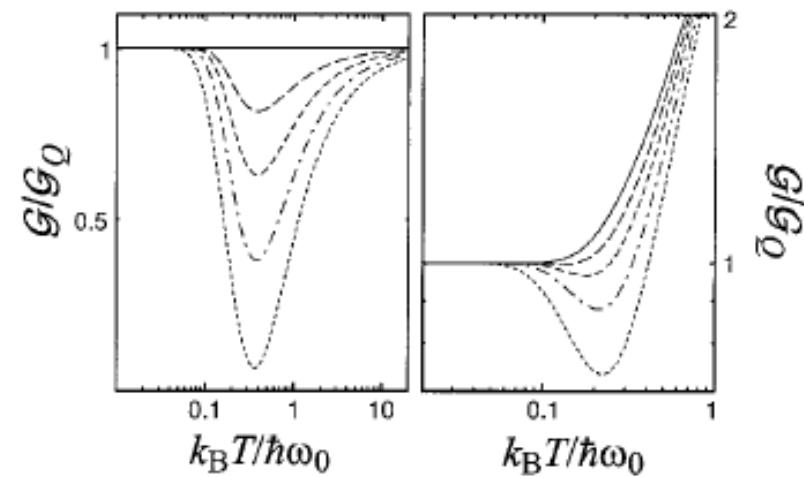
With sinusoidal profile, opening of a GAP in the dispersion relation for the acoustic phonons, so the conductance exhibit a minimum at a well defined temperature (λ_D =modulation)

From Thermal conductance of nanostructured phononic crystals, Cleland et al., PRB 2001



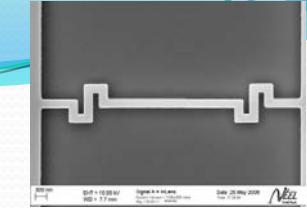
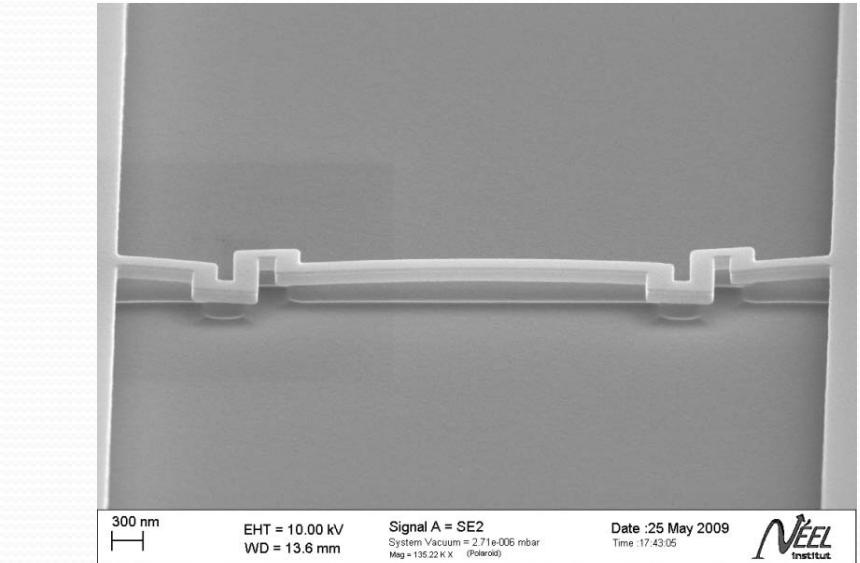
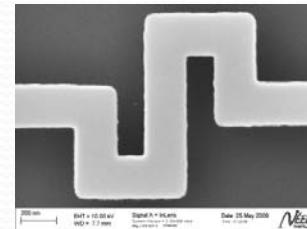
(a)

(b)

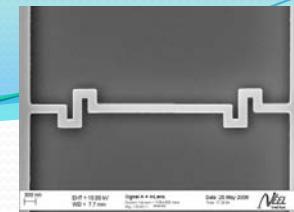


Blocking phonons at the nanoscale

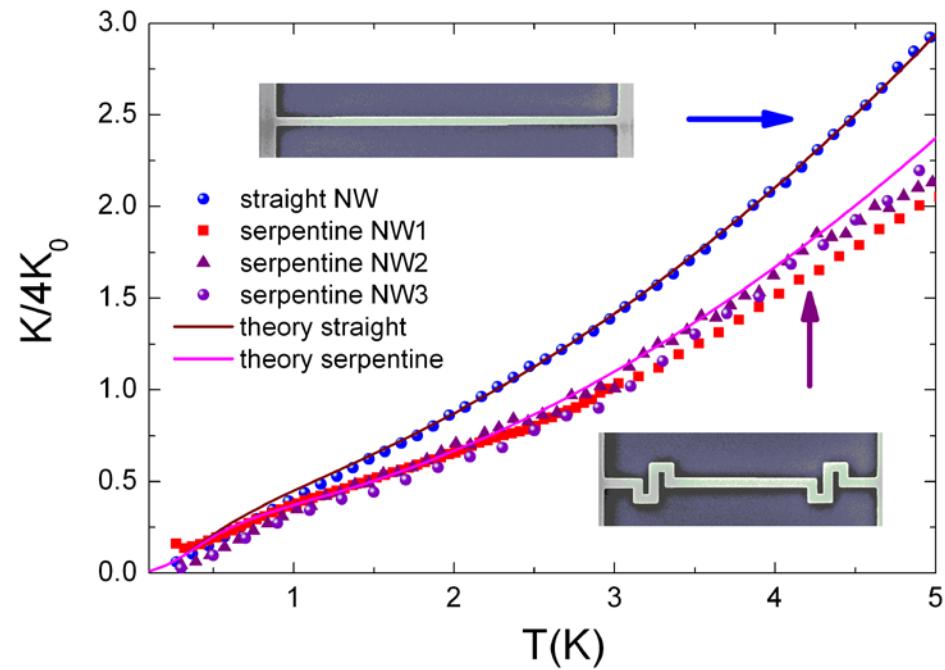
- Introducing of a serpentine nanostructure in the suspended nanowire ($5\mu\text{m}$ long)
- Length scale 200nm
- Blocking only the ballistic phonons
- Reduce the thermal conductance



Strong reduction of thermal conductance



- Reduction of up to 40% of the thermal conductance
- Model this system by transmission function analysis
- Very good agreement between the model and the data
- Concerning ballistic phonons the reduction is of the order of 80%



J-S. Heron, C. Bera, T. Fournier, N. Mingo, and O. Bourgeois, *Blocking phonons via nanoscale geometrical design*, Phys Rev B **82**, 155458 (2010)

Transmission function analysis : why phonons are blocked

$$\sigma(T) = \int_0^\infty T(\omega) \hbar\omega \frac{df}{dT} \frac{d\omega}{2\pi}$$

$$T = \frac{A}{4\pi^2} \sum_{\alpha=1}^3 \int_0^{\omega/c_\alpha} \tilde{t}^\alpha(\vec{k}_\perp, \omega) d^2k_\perp \equiv \frac{A}{4\pi} \sum_{\alpha=1}^3 t^\alpha(\omega) \frac{1}{c_\alpha^2} \omega^2.$$

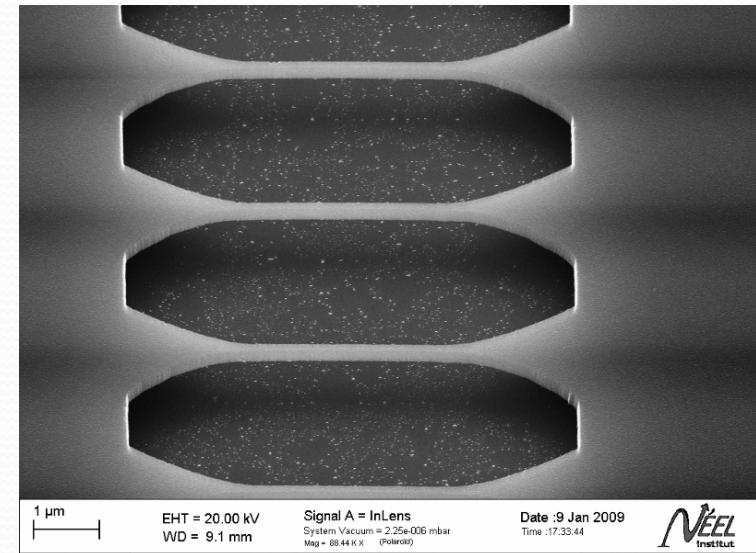
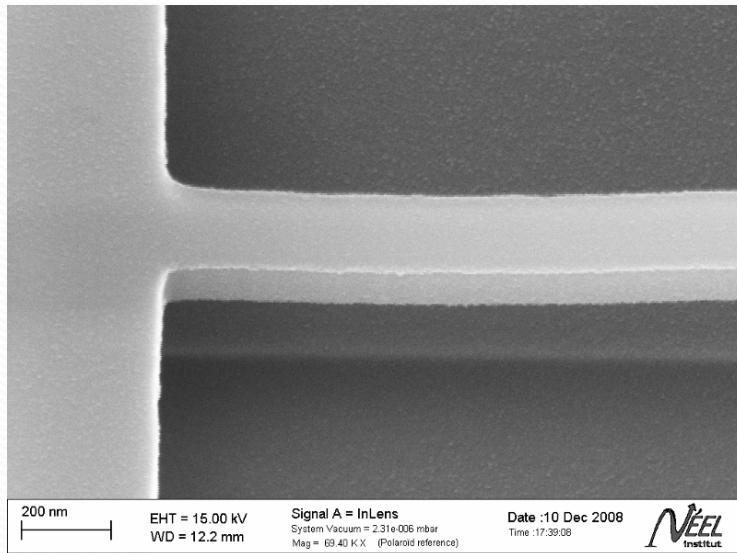
$$t(\omega) \simeq \frac{1}{t_{obs}^{-1} + \frac{3L}{4\lambda(\omega)}}. \quad \lambda(\omega) \simeq \frac{p(\omega) + 1}{p(\omega) - 1} 1.12\sqrt{A}.$$

For the double bend nanowire we have to put $t_{obs}=0.2$

This means a reduction of 80% of the thermal conductance for the ballistic phonons.

Phys Rev B **82** (2010) J-S. Heron, C. Bera, T. Fournier, N. Mingo, and O. Bourgeois, *Blocking phonons via nanoscale geometrical design*, (nov 2010).

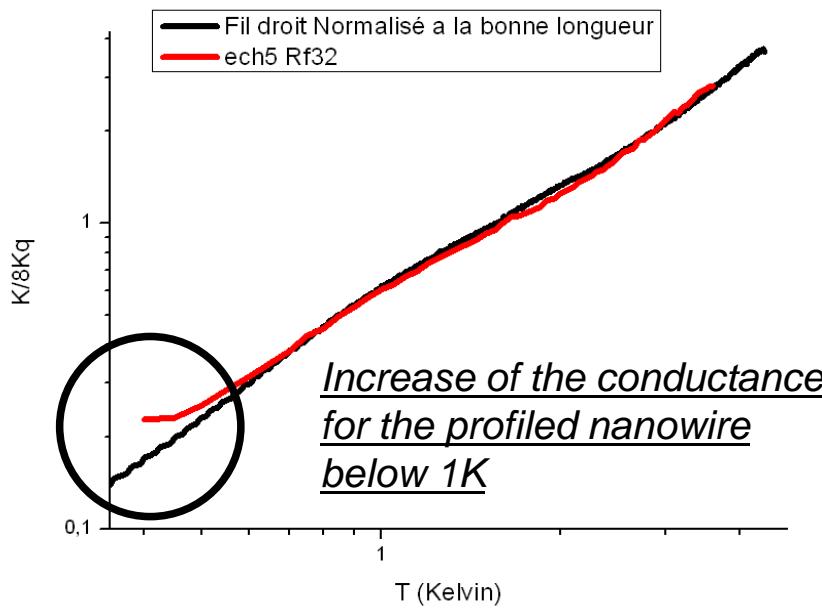
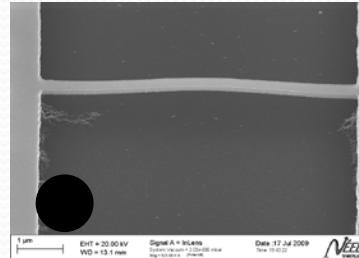
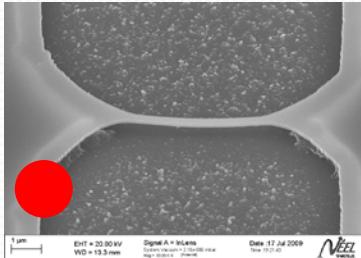
Thermal resistance of the wire/reservoir junction at very low temperature



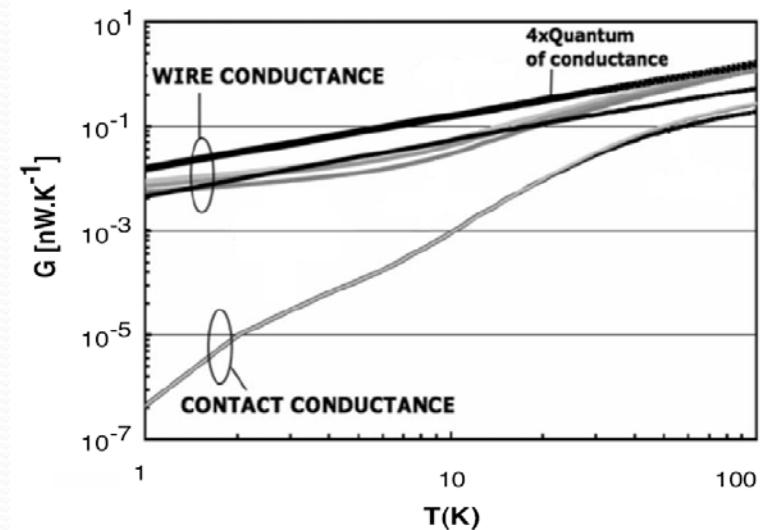
The mismatch between the density of states of the phonons of the 3D reservoir and the 1D wire implies an important thermal resistance which becomes dominant below 0.5K, with a cubic temperature dependence

Chalopin, Y;Gillet, J.-N.; Volz, S. Phys. Rev. B **77**, 233309 (2008).

Thermal contact resistance



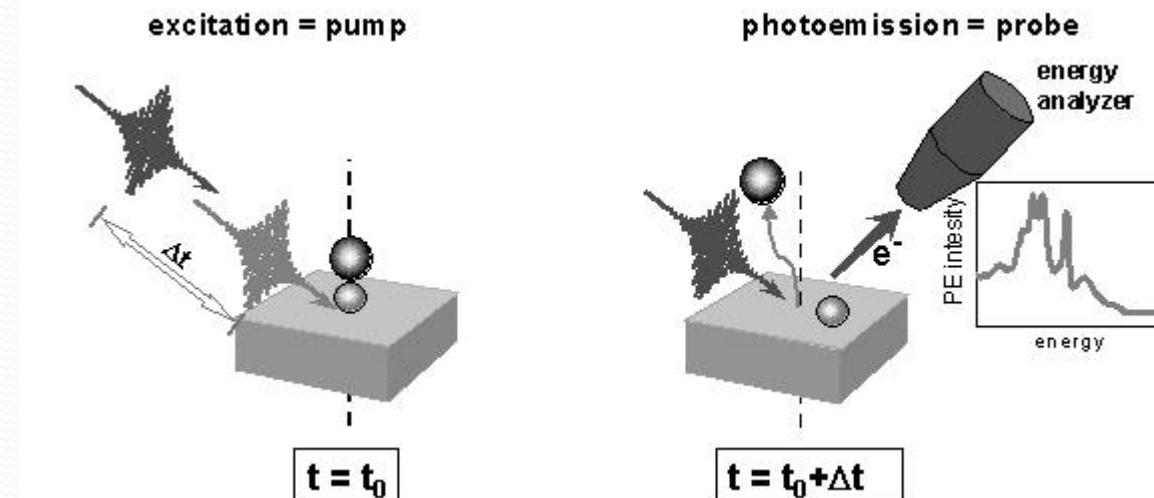
1. Contact resistance at the interface for ballistic phonons
2. Section of Nanowire 100nmx100nm
3. Observation of an increase of thermal conductance for the profiled contact



Yann Chalopin, Jean-Numa Gillet, and Sebastian Volz,
Predominance of thermal contact resistance in a silicon nanowire on a planar substrate, PHYSICAL REVIEW B, 77, 233309 (2008)

Other method (not based on electrical meas.)

- Pump-probe experiment (laser)
- Laser flash method (high temperature)
- Thermoreflectance spectroscopy
- Difficult to use on nanosystems
- Difficult to implement at very low temperature
- Major advantage: time resolved exp. down to 0.1 picoseconde
- probe the phonon exchange at the femtoseconde



See Cahill J. Appl. Phys 2003 (Review)

Conclusive comments

- Electrical measurement opens up tremendous possibilities for the study of thermal properties of micro and nanostructure at LT
 - Cp: relaxation calo and ac calo
 - K: 3w method, hot wire, thermal gradient
 - Very few experiment exists
-
- Breakdown of the concept of thermal conductivity (MFP larger than the size of the system)
 - Quantum regime: universal behavior of K (dom. phonon WL is bigger than the size of the system)

- N.W. Ashcroft and N.D. Mermin *Solid State Physics* (Saunders College Publishing, New York, 1976)
- H.M. Rosenberg *Low temperature solid state physics* (Clarendon Press, Oxford, 1963)
- F. Pobell *Matter and methods at low temperatures* (Springer Verlag, Berlin, 1992)
- J.M. Ziman *Electrons and phonons* (Clarendon Press, Oxford, 2001)
- E.S.R. Gopal *Specific heats at low temperatures* (Clarendon Press, Oxford, 2001)
- A. Tari *Specific heat of matter at low temperatures* (Imperial College Press, London, 2003)
- G. Chen *Nanoscale energy transport and conversion* (University Press, Oxford, 2005)

Book on phonon only :

- 1-G.P. Srivastava *The physics of Phonons* (Taylor & Francis (1990))
2-J.P. Wolfe *Imaging Phonons* (Cambridge University Press (1998))

O. Bourgeois, *Heat Transfer in Low Temperature Micro- and Nanosystems*, Topics in Applied Physics **118**, 537 (2009) in *Thermal Nanosystems and Nanomaterials*, Ed S. Volz, Springer 2009.

J.-L. Garden, H. Guillou, A.F. Lopeandia, J. Richard, J-S. Heron, G.M. Souche, F.R. Ong, O. Bourgeois *Thermodynamics of small systems by nanocalorimetry, from physical to biological nano-objects*, *Thermochimica Acta* **492**, 16 (2009).

