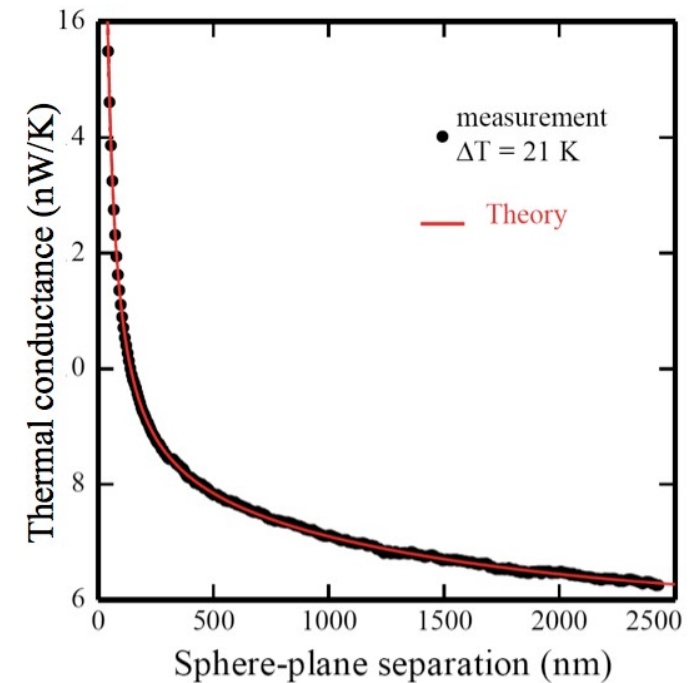


# Some details about radiative heat transfer measurements at the nanoscale

---

E. Rousseau

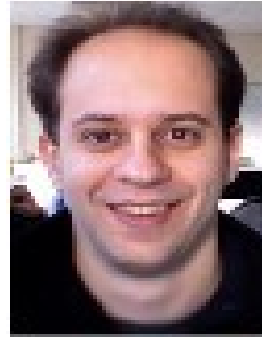
*Now at  
Laboratoire Charles Coulomb  
CNRS- Université Montpellier 2*



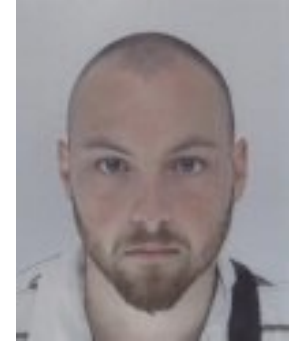
# Main Protagonists

---

Pierre-Olivier Chapuis  
*Centre de Thermique de Lyon  
Lyon*



Alessandro Siria  
*Institut Lumière Matière  
Lyon*



Sebastian Volz  
*Laboratoire Energétique  
macroscopique et moléculaire,  
Combustion  
Châtenay-Malabry*



Joël Chevrier  
*Institut Néel  
Grenoble*



Jean-Jacques Greffet  
*Laboratoire Charles Fabry  
Palaiseau*



## 1- Two ingredients along this talk:

*The heat transfer coefficient*

*The thermal conductance*

## 2-Plane-Plane experiments:

*Is there a good temperature to work at?*

*Measuring  $1/d^2$  law*

## 3- Sphere-Plane experiments

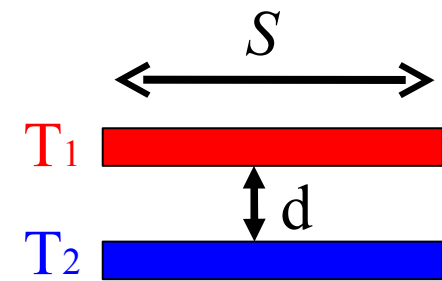
*A sensitive flux-meter*

*The experimental setup*

*Connecting the measurements to the theory*

# Ingredients: *heat transfer coefficient*

*Two infinite planes separated by  $d$*



$$\delta\varphi = \int d\omega K(\omega, \varepsilon_i, d, \dots) (L(\omega, T_1) - L(\omega, T_2)) S$$

$$\delta\varphi = \int d\omega K(\omega, \varepsilon_i, d, \dots) \frac{\partial L(\omega, T)}{\partial T} \delta T S$$

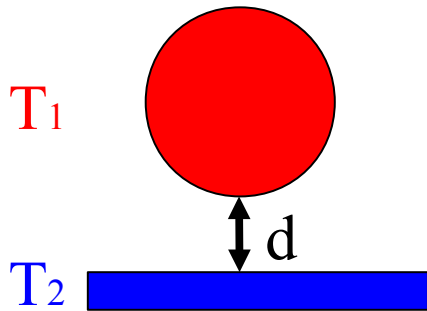
$$\delta\varphi = \int d\omega h(\omega, \varepsilon_i, d, T) \delta T S$$

$$\delta\varphi = h(\varepsilon_i, d, T) \delta T S \quad W.m^{-2}.K^{-1}$$

From Joulain et al., Surface Sciences Reports **57**, 59 (2005)

# Ingredients: *thermal conductance*

*Heat transfer between a sphere and a plane separated by  $d$*



$$\delta\varphi = G(\varepsilon_i, d, T)\delta T \quad W.K^{-1}$$

In my talk: from the Derjaguin approximation

Now: from exact theory

## 1- Two main ingredients:

*The heat transfer coefficient*

*The thermal conductance*

## 2-Plane-Plane experiments:

*Is there a good temperature to work at?*

*Measuring  $1/d^2$  law*

## 3- Sphere-Plane experiments

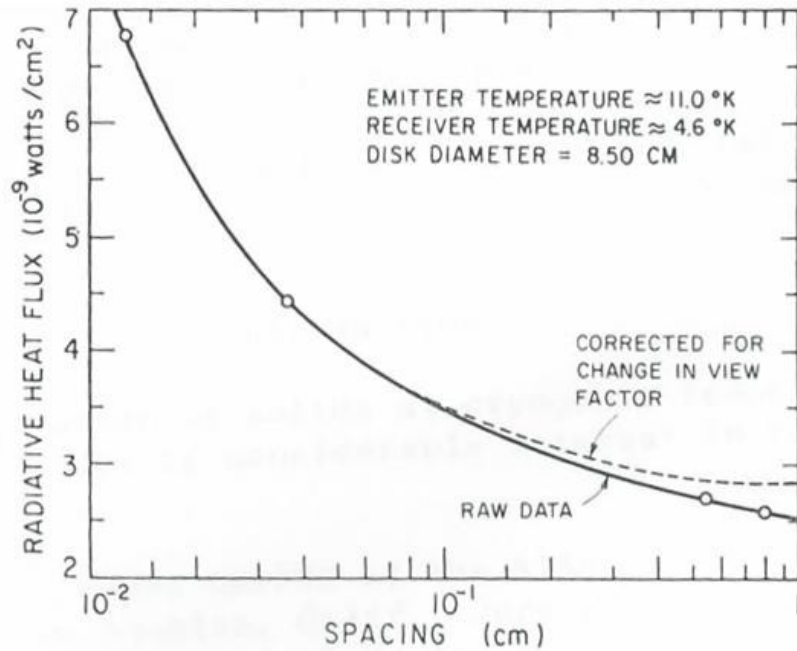
*A sensitive flux-meter*

*The experimental setup*

*Connecting the measurements to the theory*

# Measurements at low temperature $\sim 10$ K

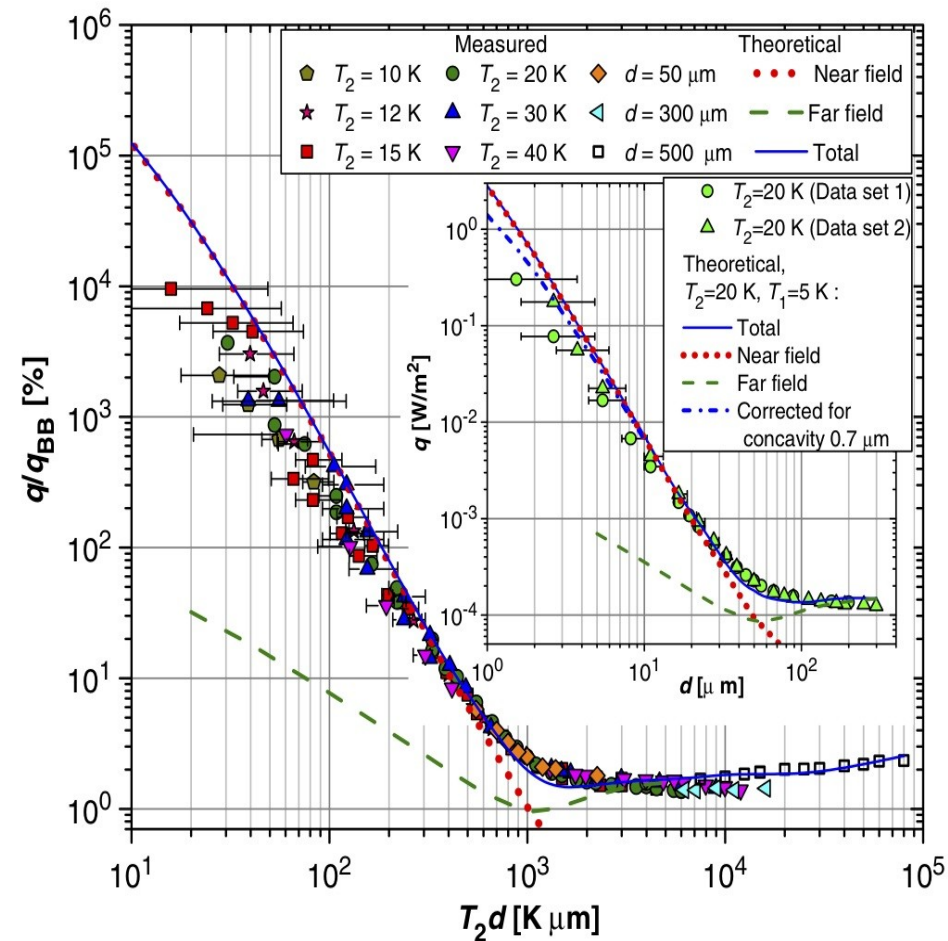
## Between metals



Radiative heat flux vs spacing between parallel copper disks.

From E.G. Cravalho et al. , *Progres in Aeronautics and Astronautics* **22**, 531 (1968)

## Between metals



From Kralik et al. *PRL* **109**, 224302 (2012)

## Between metals

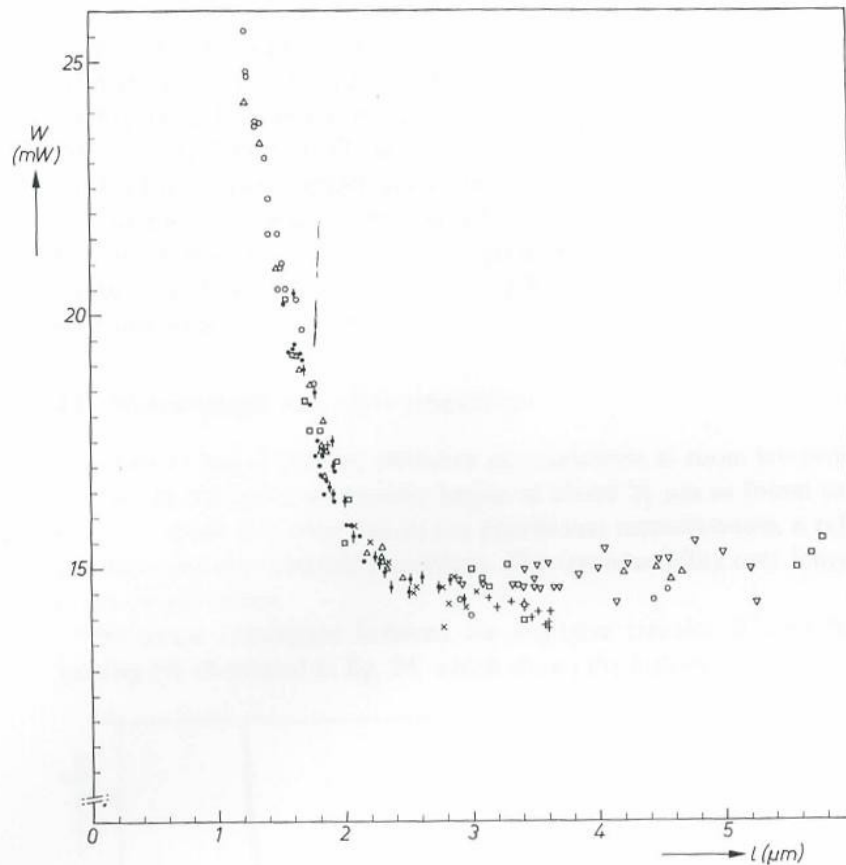
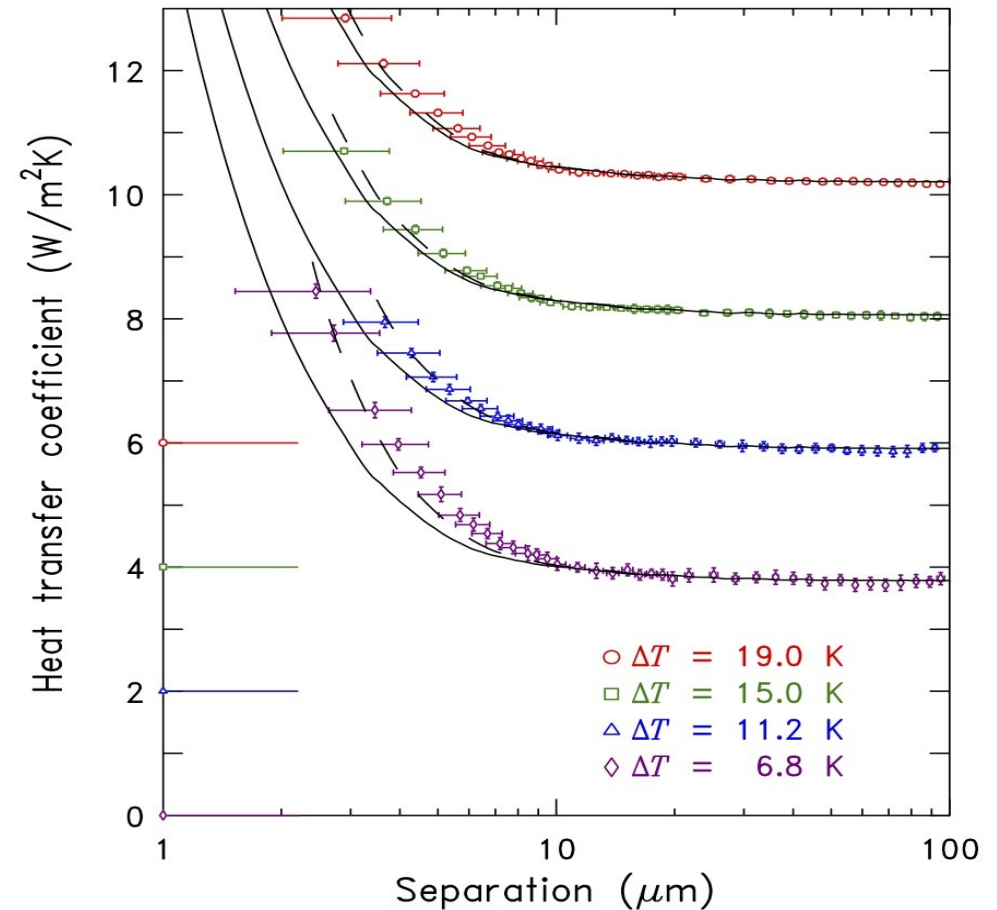


Fig. 22. Provisional measurements <sup>79)</sup> of the radiative transfer  $W$  between parallel metal surfaces as a function of the spacing  $l$ . Mean temperature  $T = 315$  K,  $\Delta T = 17$  K.

From Hargreaves et al. Phys. Lett. A **30A**  
491 (1969)

## Between dielectrics



From Ottens et al. PRL **107**, 014301 (2011)

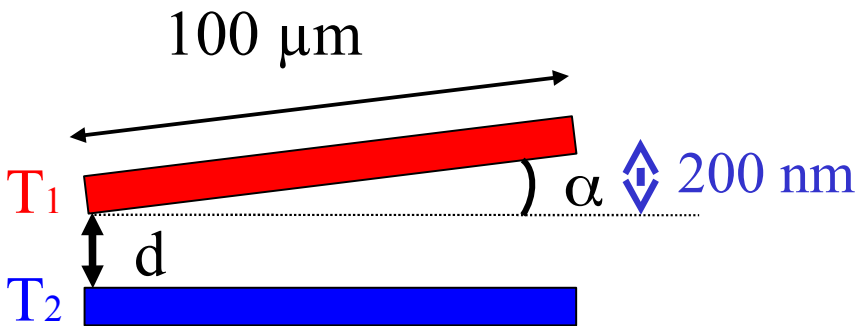


Plane –plane experiments are:

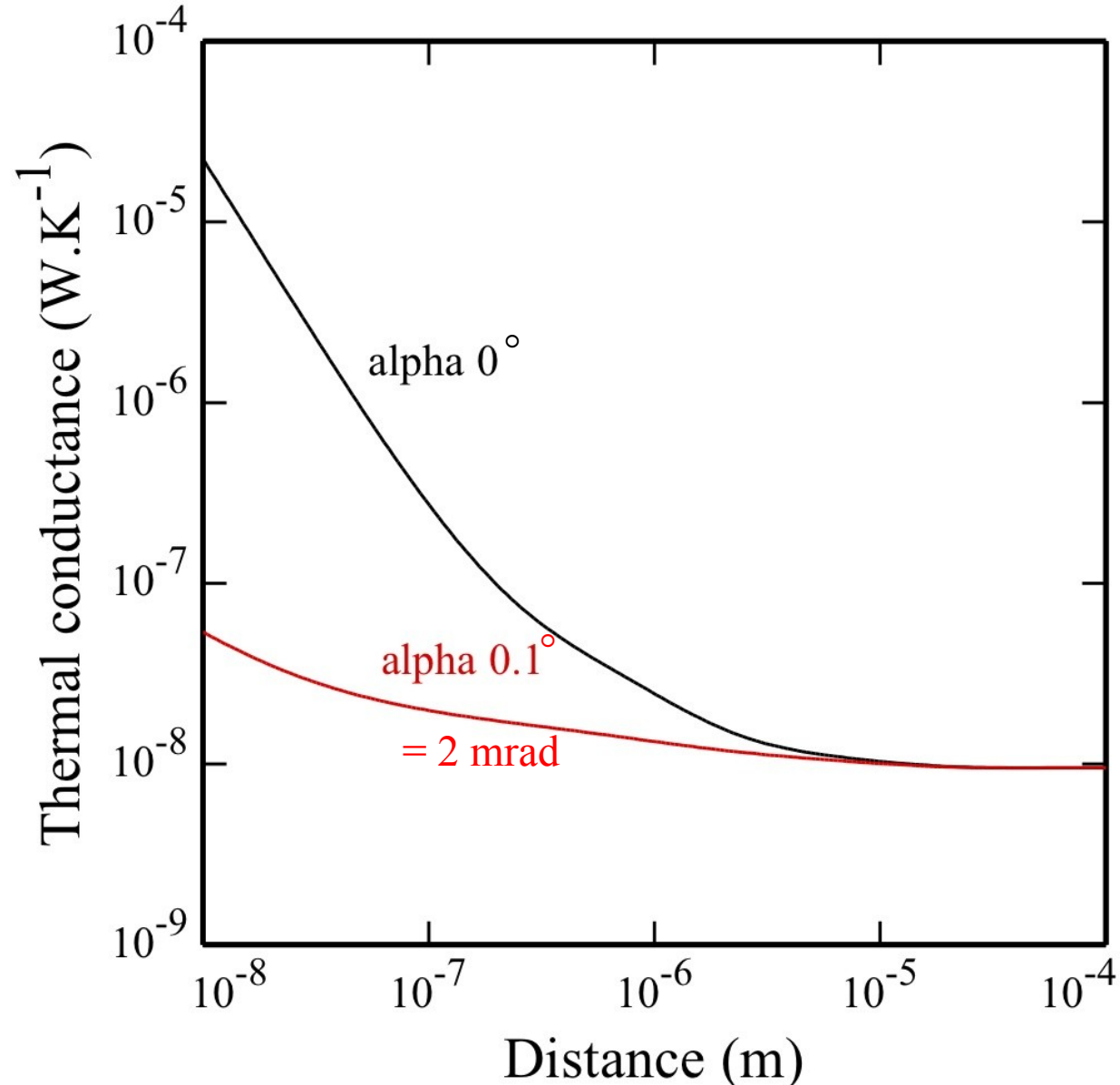
- important: easily compared to the theory
- interesting from an experimental point of view:  
*large area = large flux even if surface fluxes are not so high*
- difficult: because parallelism is a difficult task

# Misalignment consequences...

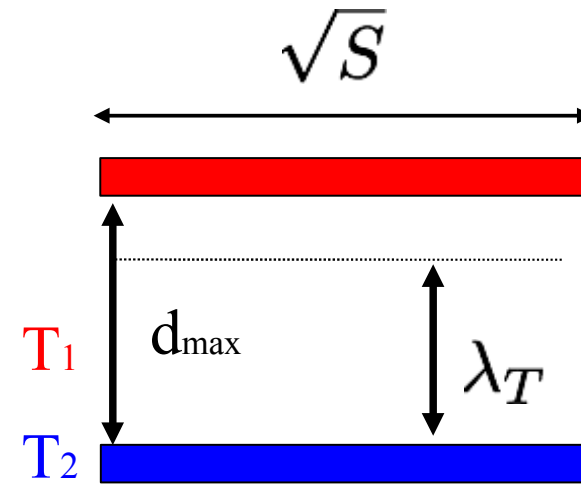
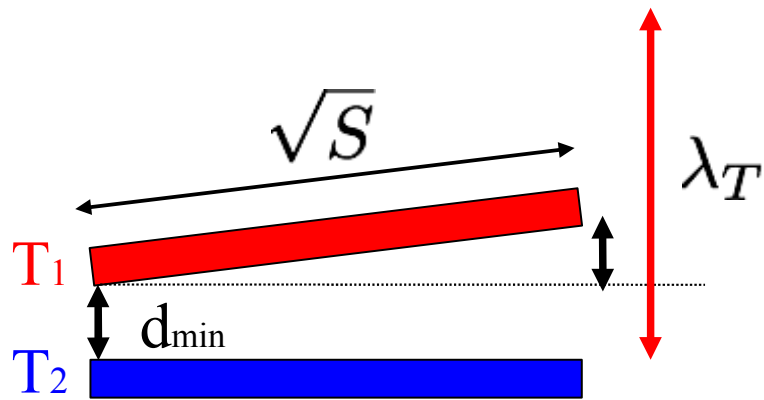
Crude estimation with Proximity Approximation



Surface modes gap-dependence ( $1/d^2$ ) cannot be measured if angle are not extremely well control



# At which temperature should I work?



$\sqrt{S}$  Characteristic lateral scale

$d_{\max}$  Maximum achieved gap  $\sim 10 \mu\text{m}$

$d_{\min}$  Minimum achieved gap  $\sim 100\text{-}10 \text{ nm}$

Maybe what I would like to measure will fix the temperature?

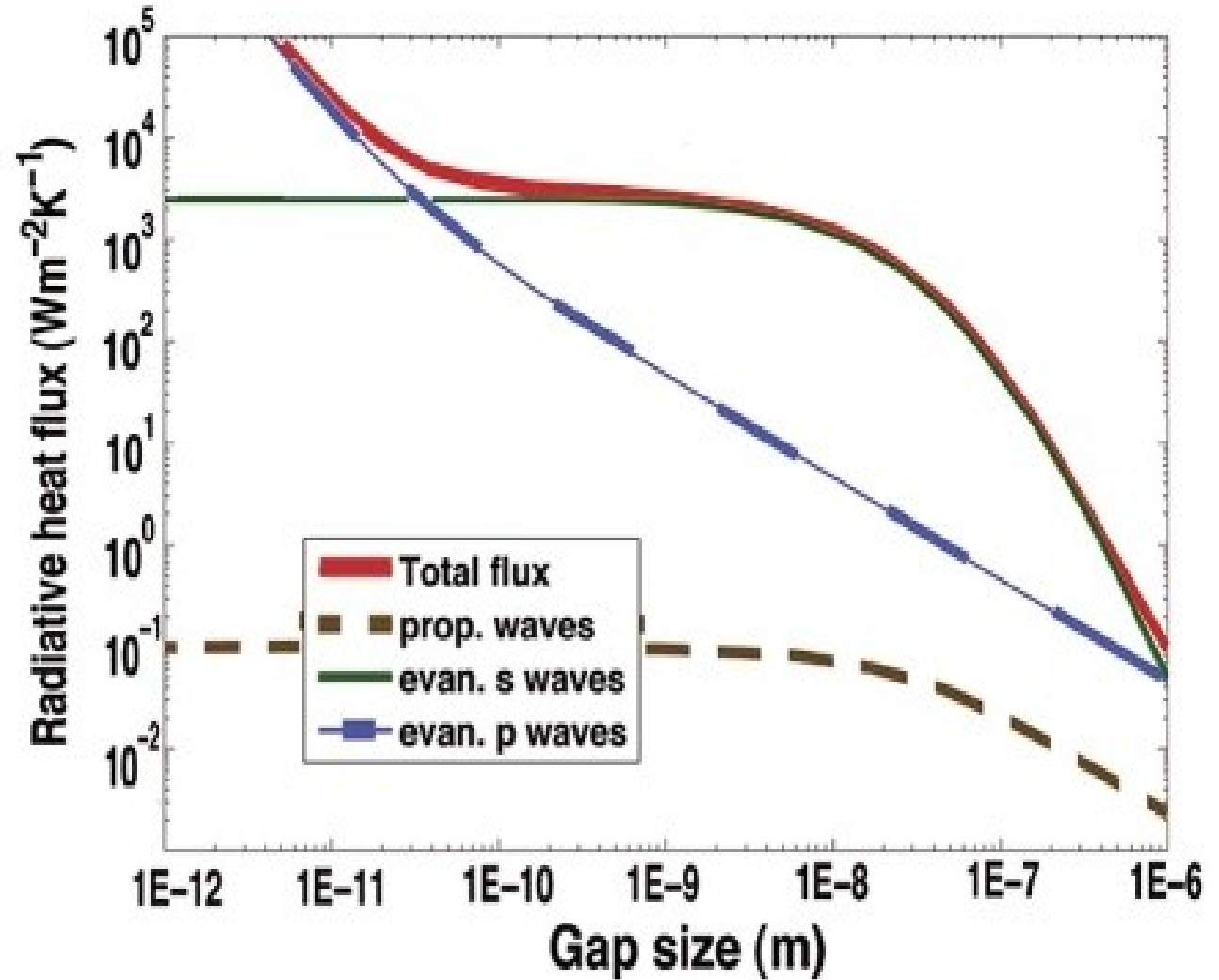
# What have not been shown yet?

Metallic materials:  
Induction zone

Surface modes  
coupling:  
Unavailable at  
reasonable  
temperature

Saturation:  
after skin depth  
*Not shown yet*

Between two Gold plates at 300 K



From Chapuis et al. PRB 77 035431 (2008)

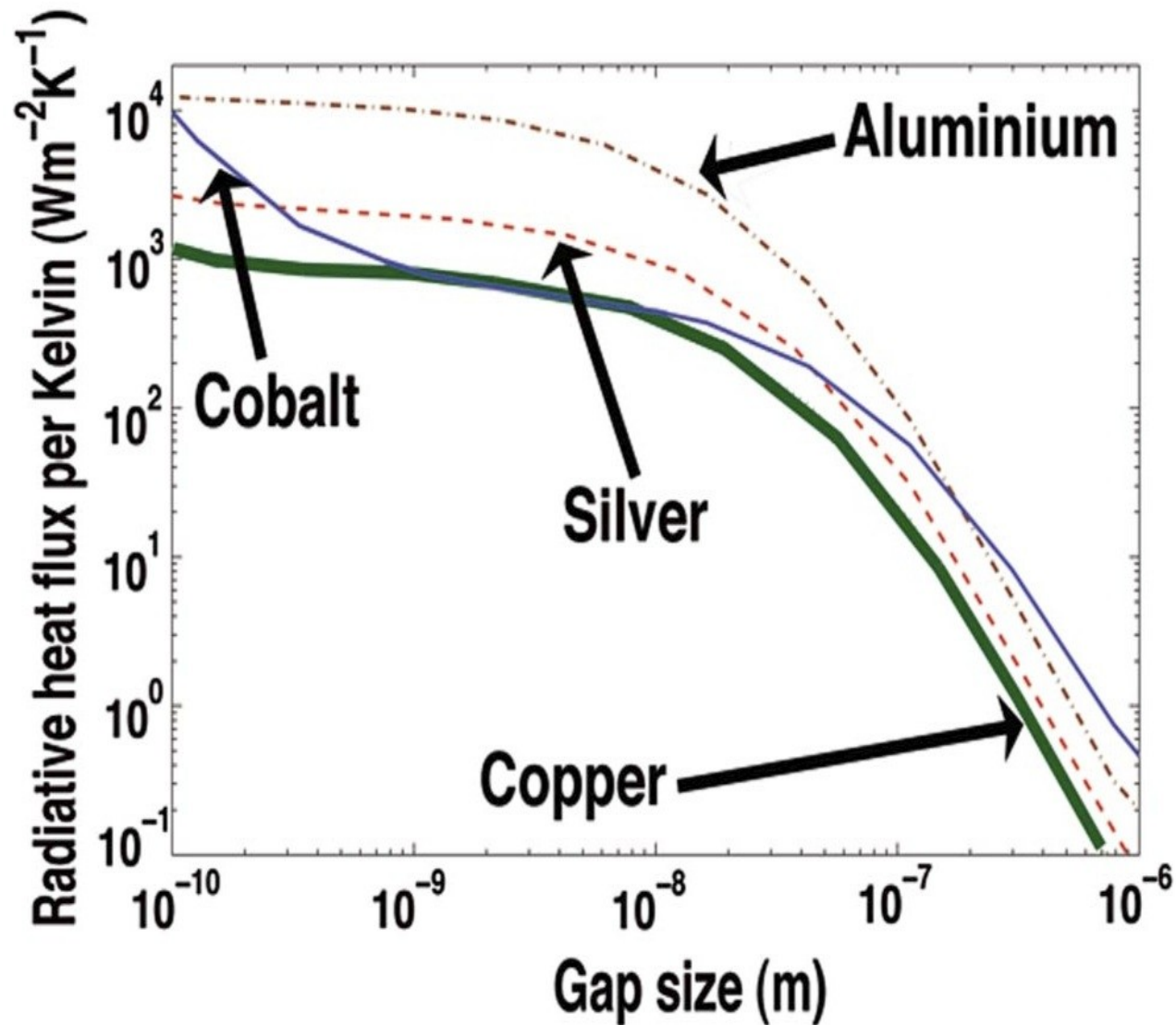
# What have not been shown yet? *For metals*

Distance of saturation:

Material dependent

But

Temperature independent



From Chapuis et al. PRB 77 035431 (2008)

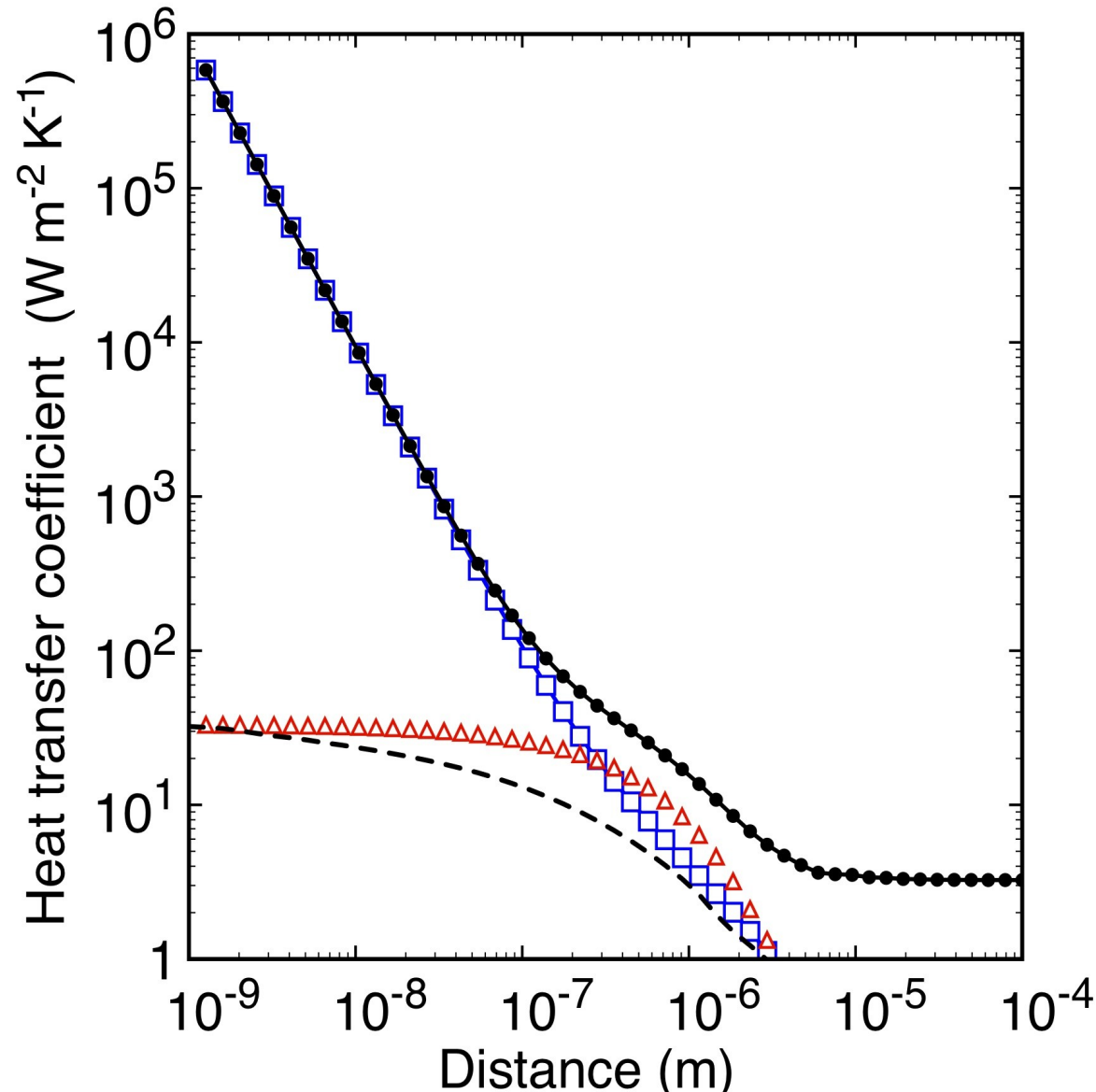
# What have not been shown yet?

For materials supporting surface modes in the IR:  
(Dielectric polar materials or doped silicon)

Intermediate regime: contribution of frustrated reflexions

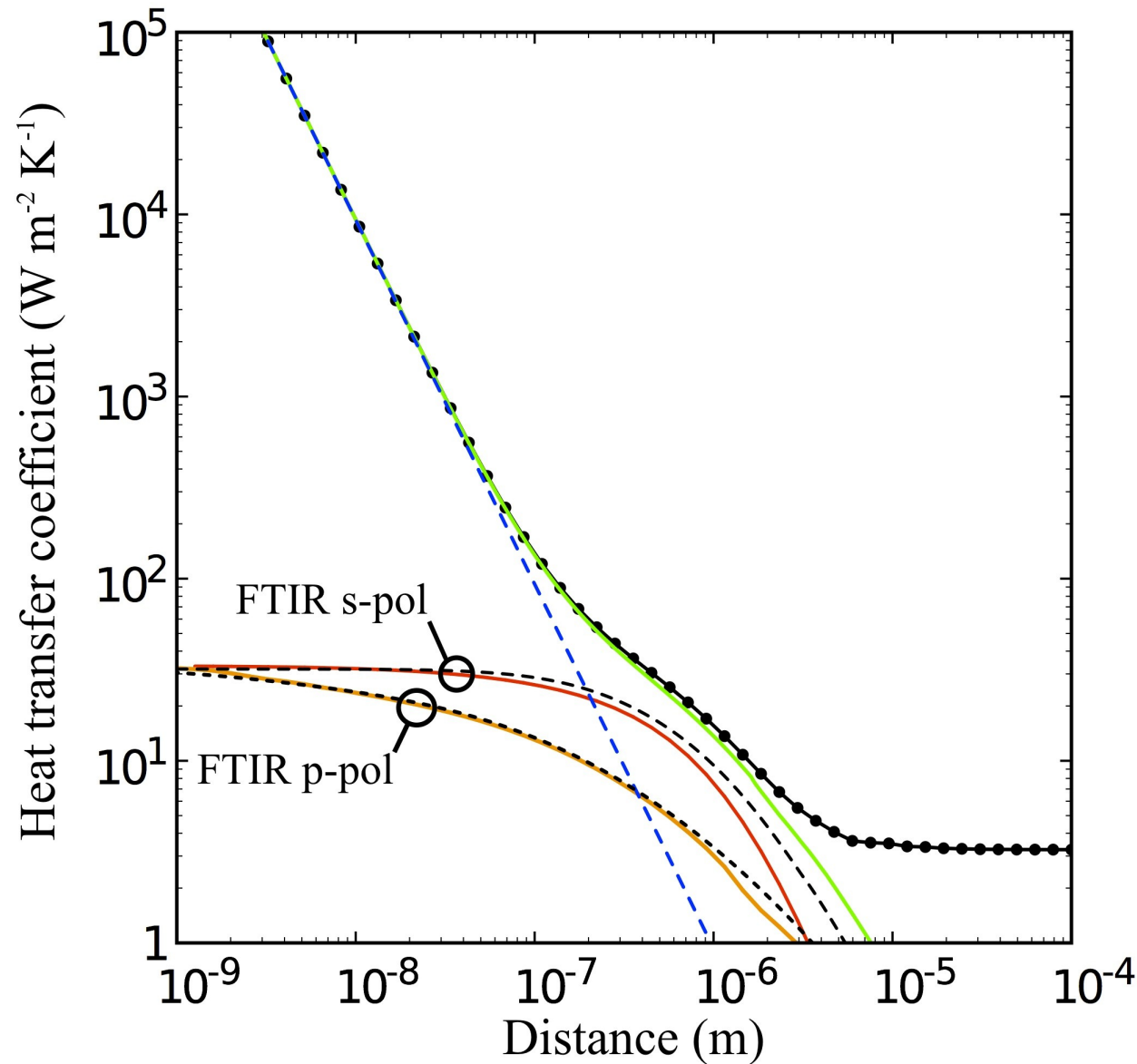
Surface modes contribution: electrostatic approximation  $1/d^2$   
*Not shown yet*

*What is the best temperature to work at?*



# Some asymptotic results

It is possible to measure the  $1/d^2$  law  
when SPP contribution  
dominates



From Rousseau et al. JAP 111 014311  
(2012)

# Some analytical results

$$h_{spp}(d, T) = \frac{\delta G(T)}{d^2}$$

Starting from stochastic electrodynamics

TM polarisation evanescent contribution:

$$h_{evan}^p(u, d, T) = \frac{3}{2\pi^3} \frac{g_0}{\lambda_T^2} h^0(u) \times$$

$$\int_0^\infty \tilde{\gamma} d\tilde{\gamma} \frac{4\text{Im}(r_{31})\text{Im}(r_{32})e^{-2k_T\tilde{\gamma}ud}}{|1 - r_{31}r_{32}e^{-2k_T\tilde{\gamma}ud}|^2}$$

$$u = \frac{\hbar\omega}{k_B T}$$



# Some analytical results

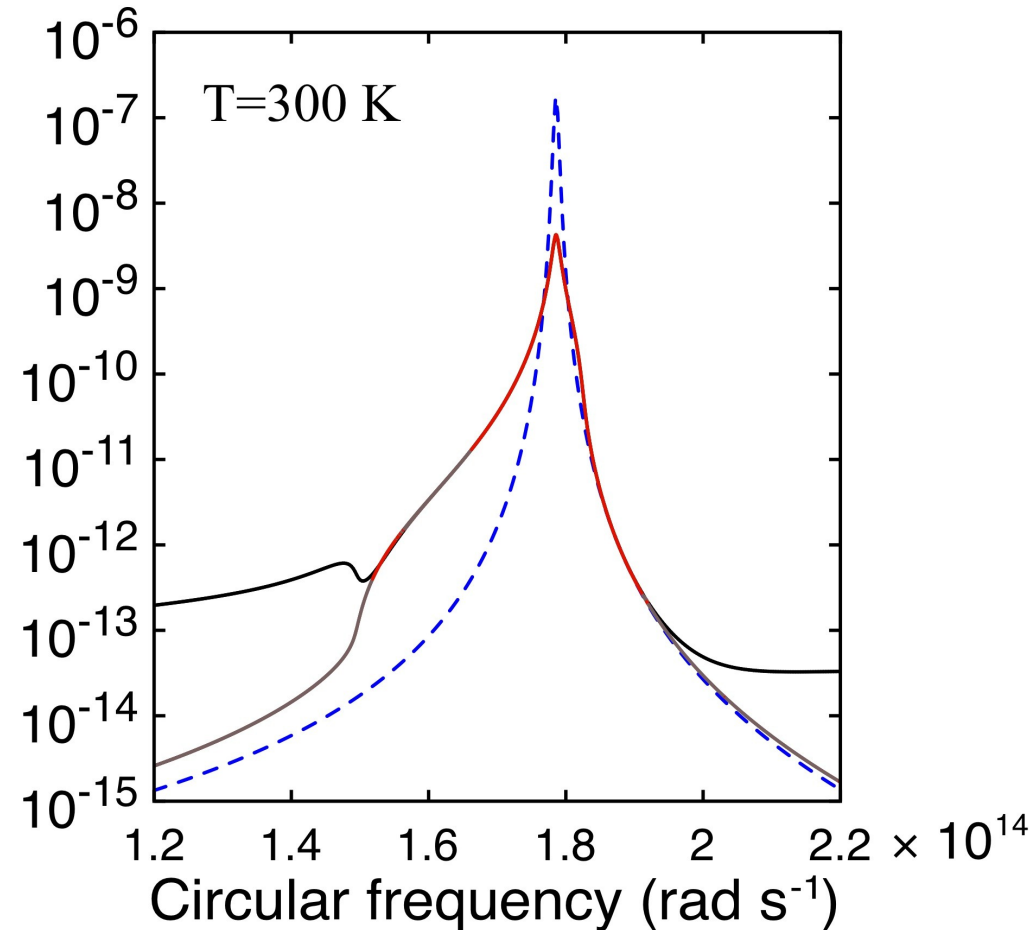
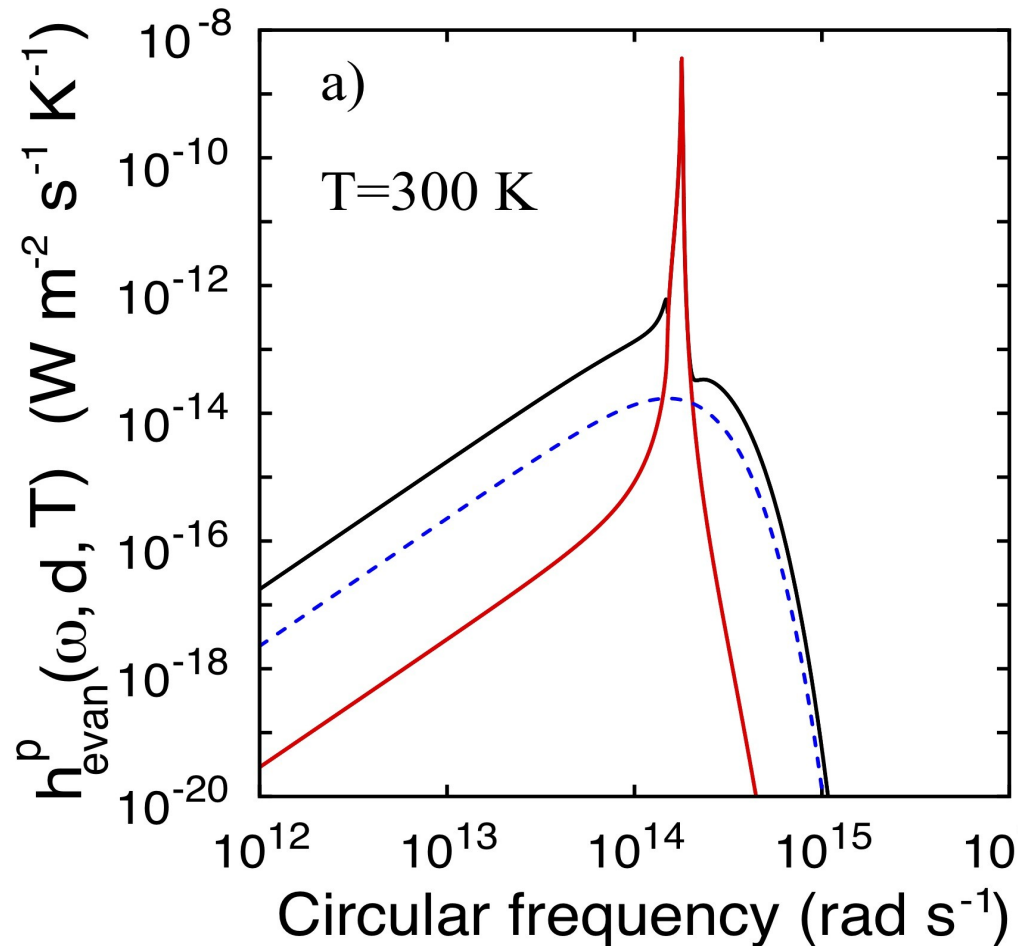
$$h_{evan}^p(u, d, T) = \frac{3}{2\pi^3} \frac{g_0}{\lambda_T^2} h^0(u) \times \int_0^\infty \tilde{\gamma} d\tilde{\gamma} \frac{4\text{Im}(r_{31})\text{Im}(r_{32})e^{-2k_T\tilde{\gamma}ud}}{|1 - r_{31}r_{32}e^{-2k_T\tilde{\gamma}ud}|^2}$$

Electrostatic approximation: integration over wavevectors

$$h_{spp}(u, d, T) = \frac{3}{2\pi^3} \frac{g_0}{d^2} \frac{h^0(u)}{u^2} \times \frac{\text{Im}(\tilde{r}_{31})\text{Im}(\tilde{r}_{32})}{\text{Im}(\tilde{r}_{31}\tilde{r}_{32})} \text{Im}[Li_2(\tilde{r}_{31}\tilde{r}_{32})]$$

# Some analytical results

Two SiC slab separated by 10 nm



This asymptotic expression capture the dispersion relation of the two-plates geometry

# Some analytical results

---

$$h_{spp}(u, d, T) = \frac{3}{2\pi^3} \frac{g_0}{d^2} \frac{h^0(u)}{u^2} \times$$
$$\frac{\text{Im}(\tilde{r}_{31})\text{Im}(\tilde{r}_{32})}{\text{Im}(\tilde{r}_{31}\tilde{r}_{32})} \text{Im}[Li_2(\tilde{r}_{31}\tilde{r}_{32})]$$

In the complex plane:

From Rousseau et al. JAP 111 014311  
(2012)

# Some analytical results

$$h_{spp}(u, d, T) = \frac{3}{2\pi^3} \frac{g_0}{d^2} \frac{h^0(u)}{u^2} \times$$

$$\frac{1}{f(u)} = \frac{\text{Im}(\tilde{r}_{31})\text{Im}(\tilde{r}_{32})}{\text{Im}(\tilde{r}_{31}\tilde{r}_{32})} \text{Im}[Li_2(\tilde{r}_{31}\tilde{r}_{32})]$$

In the complex plane:

$$h_{spp}(d, T) = \frac{\delta G(T)}{d^2}$$

$$\delta G(T) = \frac{3}{2\pi^2} g_0 \times$$

$$\sum_{u_i} \frac{1}{f'(u_i)} u_i^2 \frac{e^{u_i}}{(e^{u_i} - 1)^2} \text{Re}[Li_2(\tilde{r}_1(u_i)\tilde{r}_2(u_i))]$$

# Some analytical results

$$h_{spp}(d, T) = \frac{\delta G(T)}{d^2}$$

$$u = \frac{\hbar\omega}{k_B T}$$

$$\delta G(T) = \frac{3}{2\pi^2} g_0 \times$$

$$\sum_{u_i} \frac{1}{f'(u_i)} u_i^2 \frac{e^{u_i}}{(e^{u_i} - 1)^2} \text{Re}[Li_2(\tilde{r}_1(u_i)\tilde{r}_2(u_i))]$$

are zeros of

$$f(u) = \frac{\text{Im}(\tilde{r}_{31})\text{Im}(\tilde{r}_{32})}{\text{Im}(\tilde{r}_{31}\tilde{r}_{32})}$$

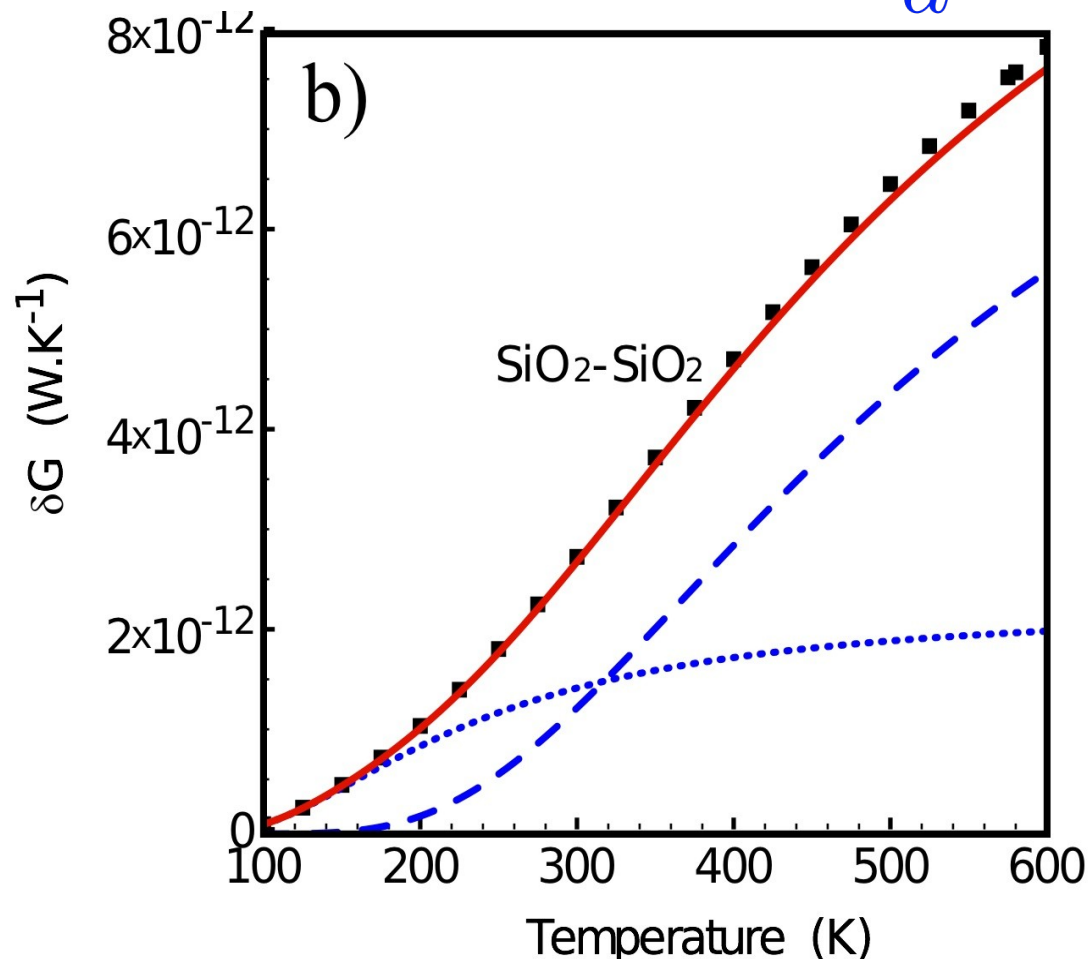
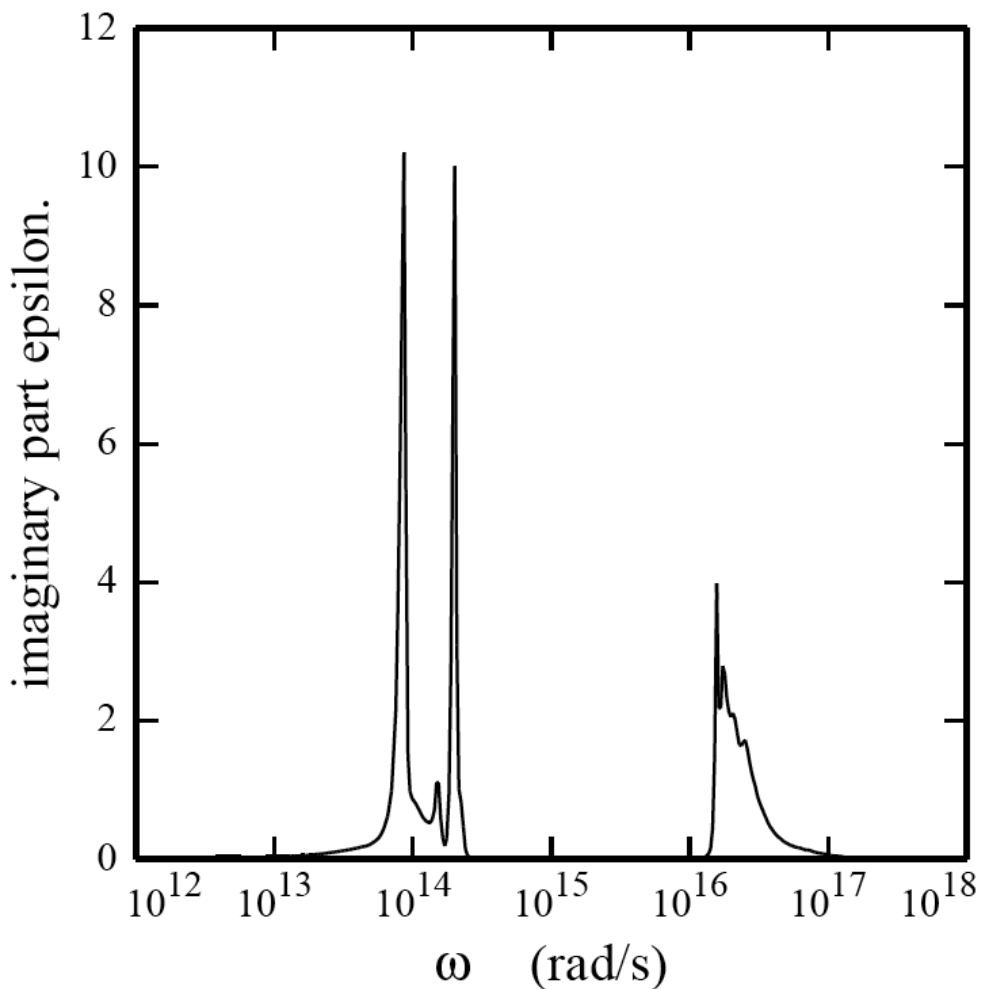
with  $f'(u) < 0$

Can be found  
directly from  
experimental  
data

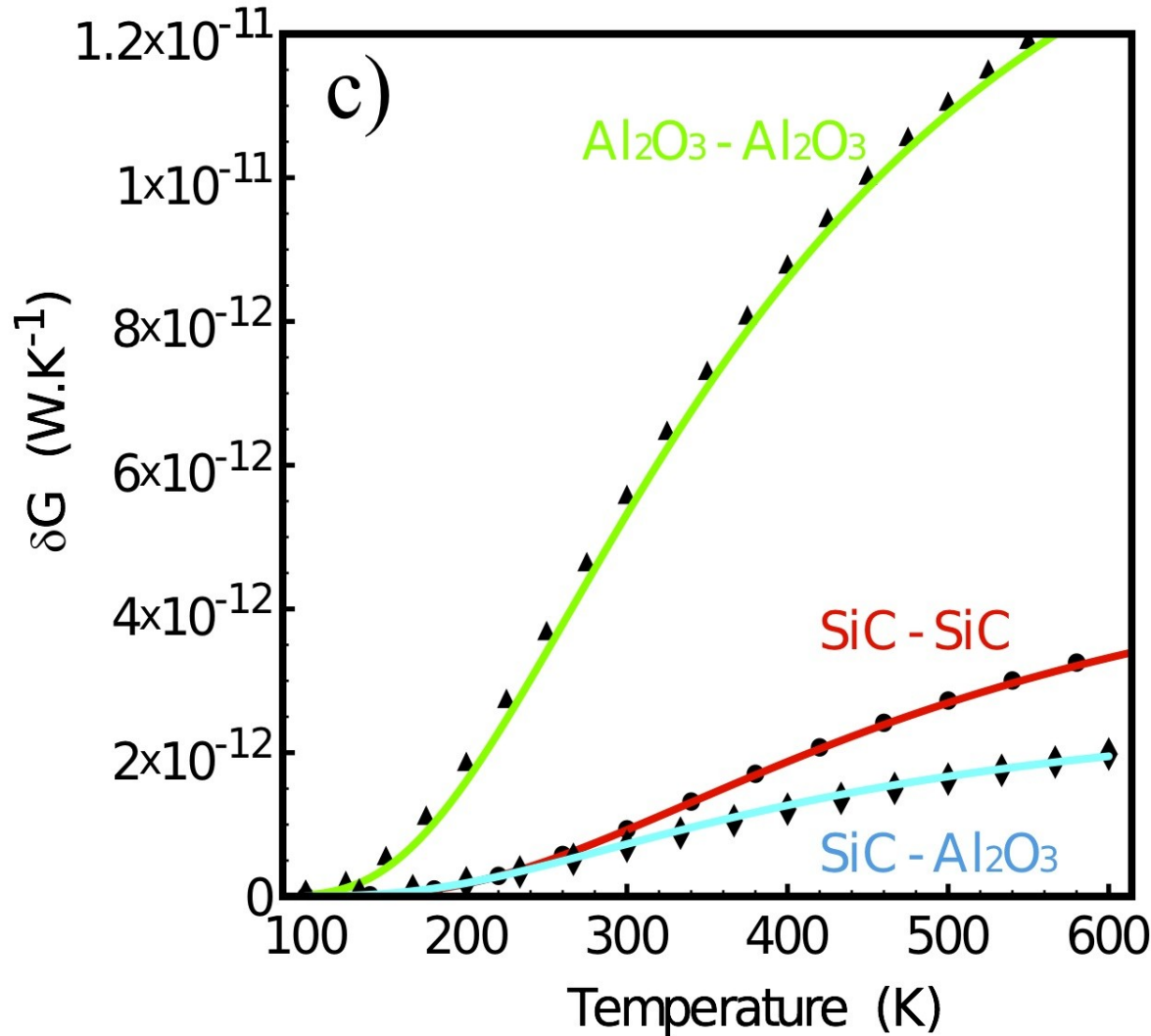
# Some analytical results

Two surface modes for silica

$$h_{spp}(d, T) = \frac{\delta G(T)}{d^2}$$



# Some analytical results



$$h_{spp}(d, T) = \frac{\delta G(T)}{d^2}$$

Different materials

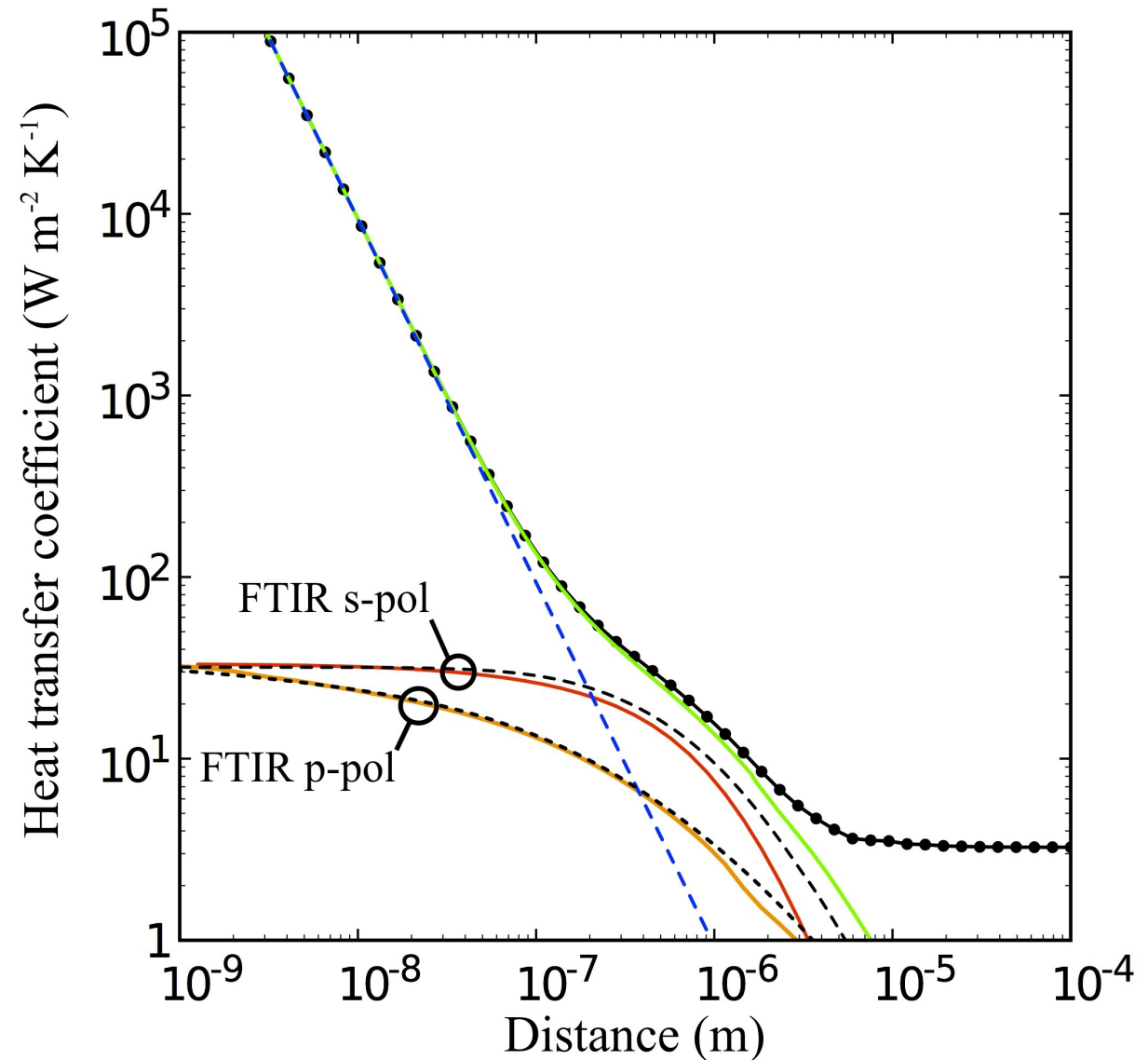
Similar or not

# SPP contribution

Surface modes coupling  
Contribution OK

Frustrated Total Internal  
Reflections

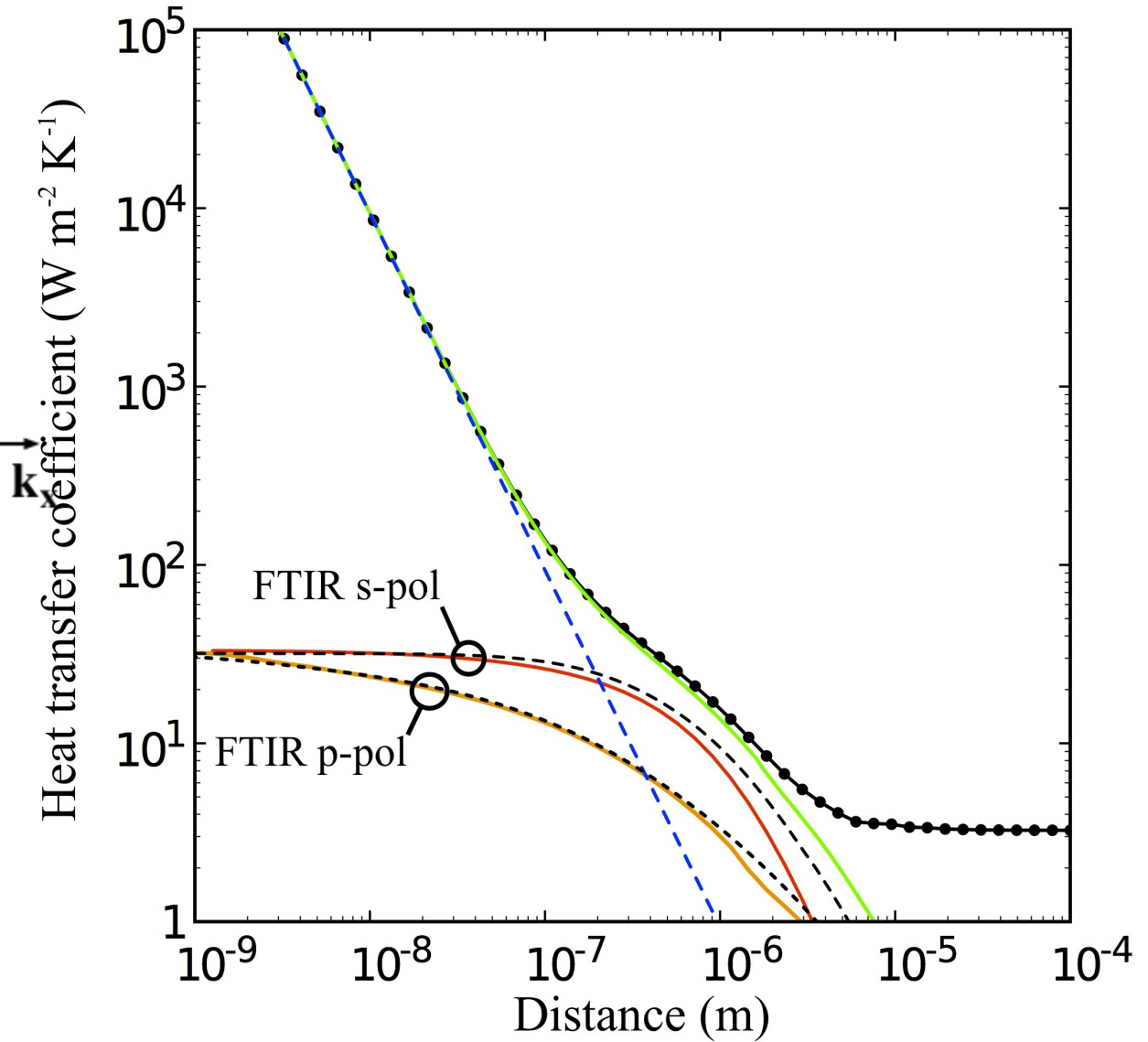
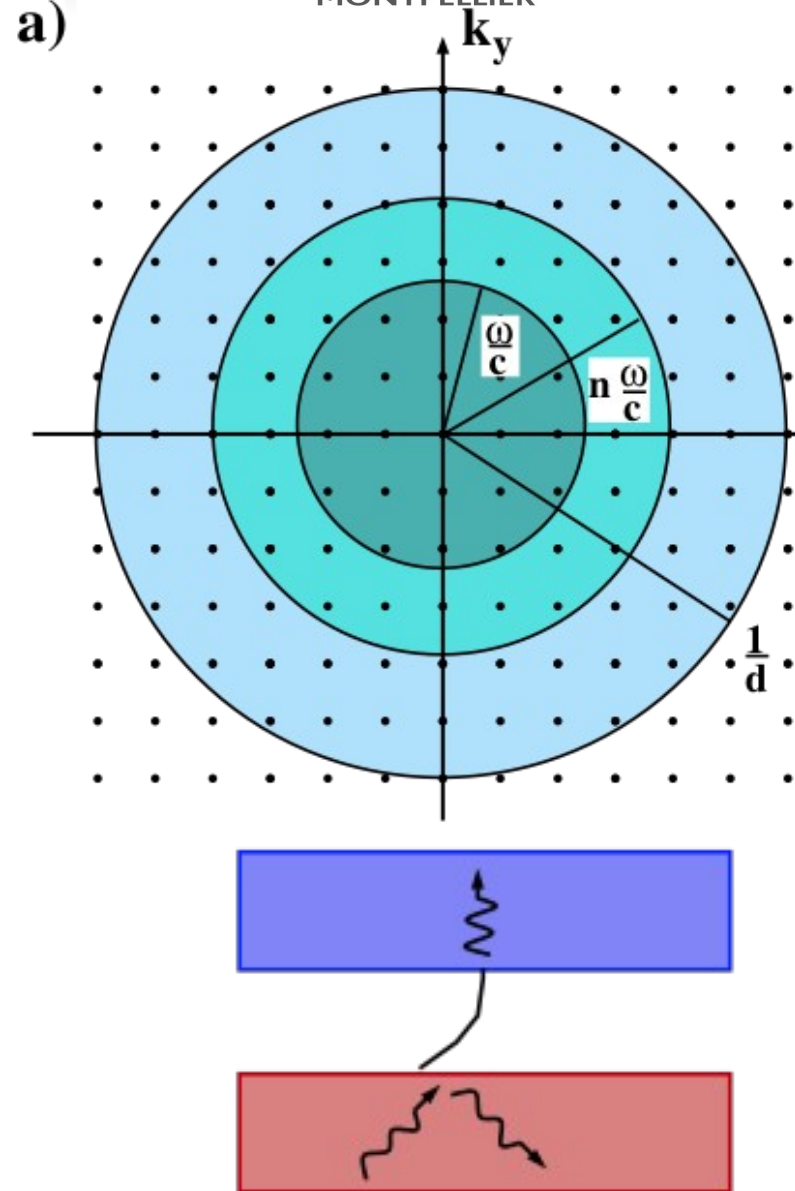
In a Landauer point of view



From Rousseau et al. JAP 111 014311  
(2012)



# FTIR contribution

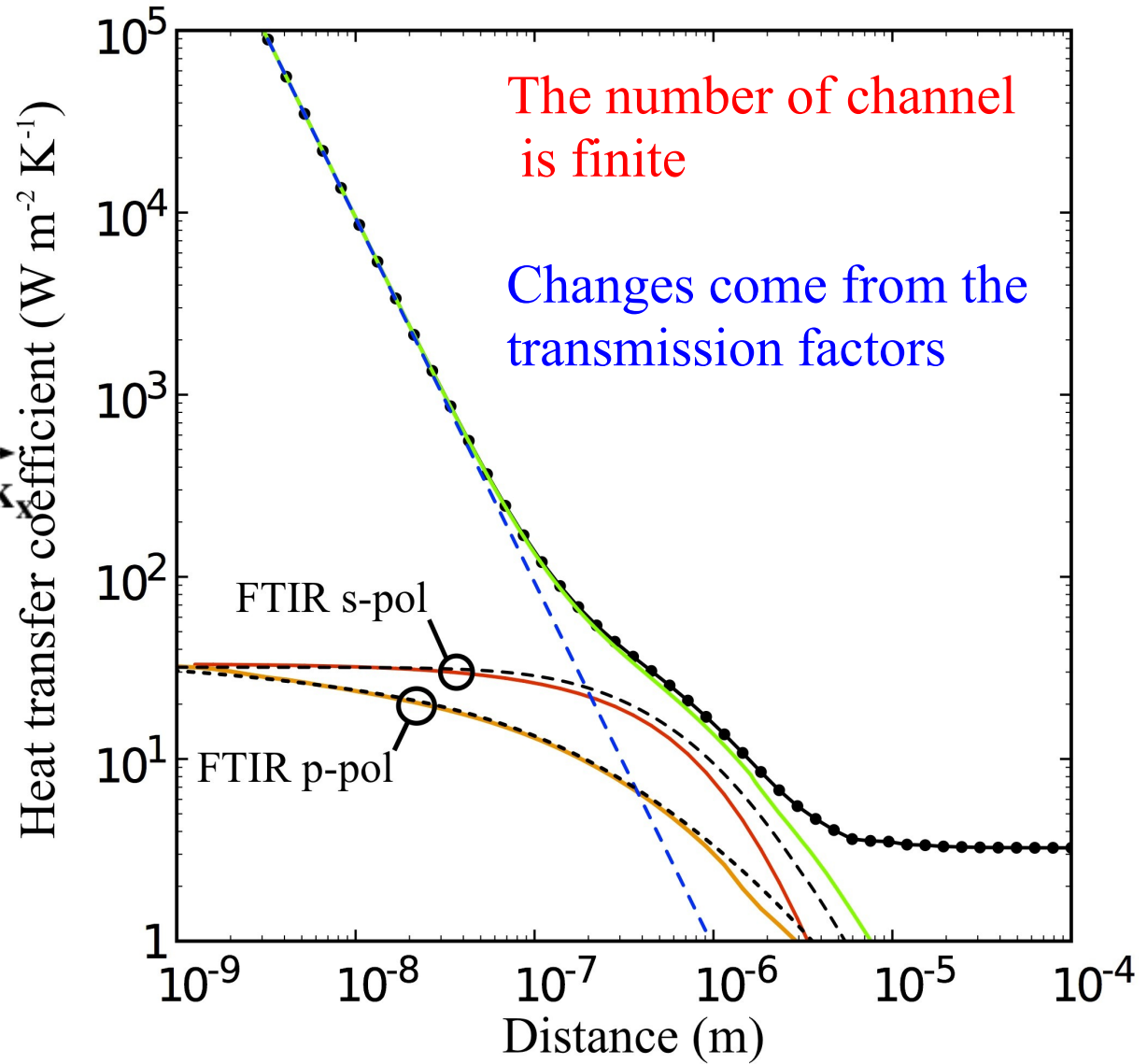
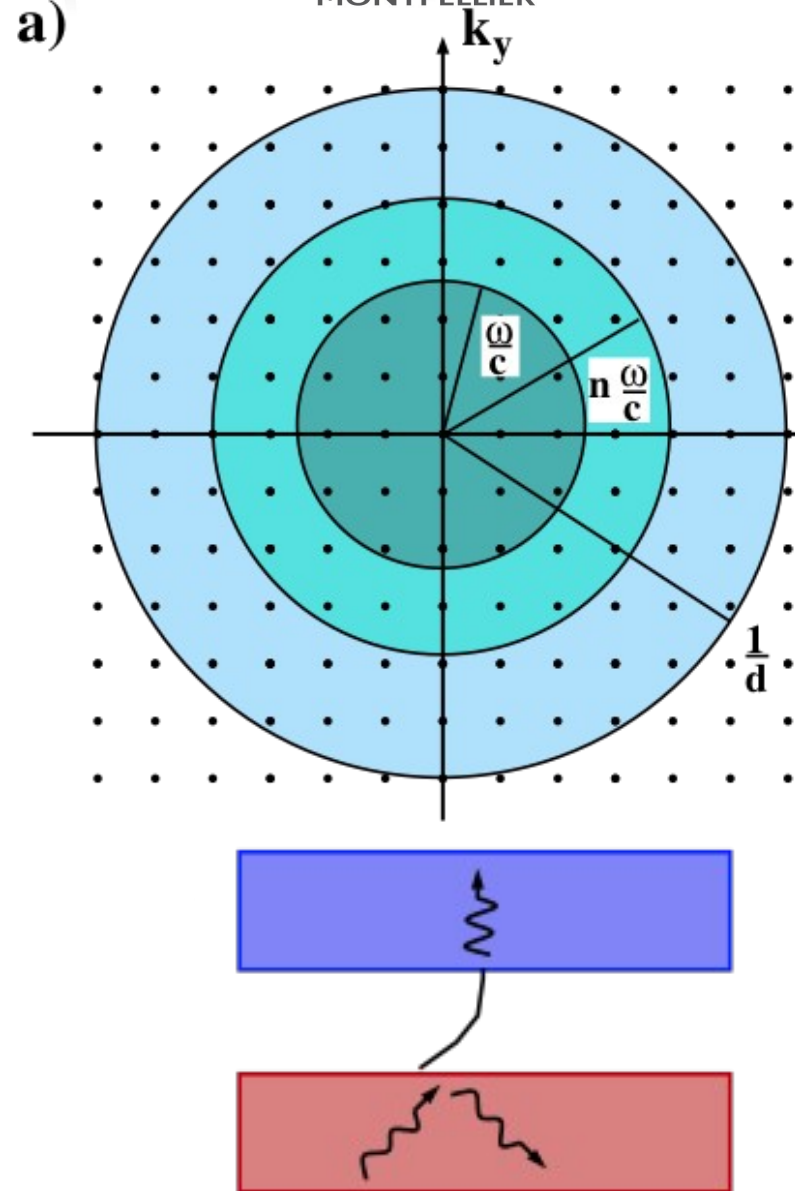


FTIR contribution



# FTIR contribution

LABORATOIRE  
CHARLES  
COULOMB  
MONTPELLIER



From Rousseau et al. JAP 111 014311  
(2012)

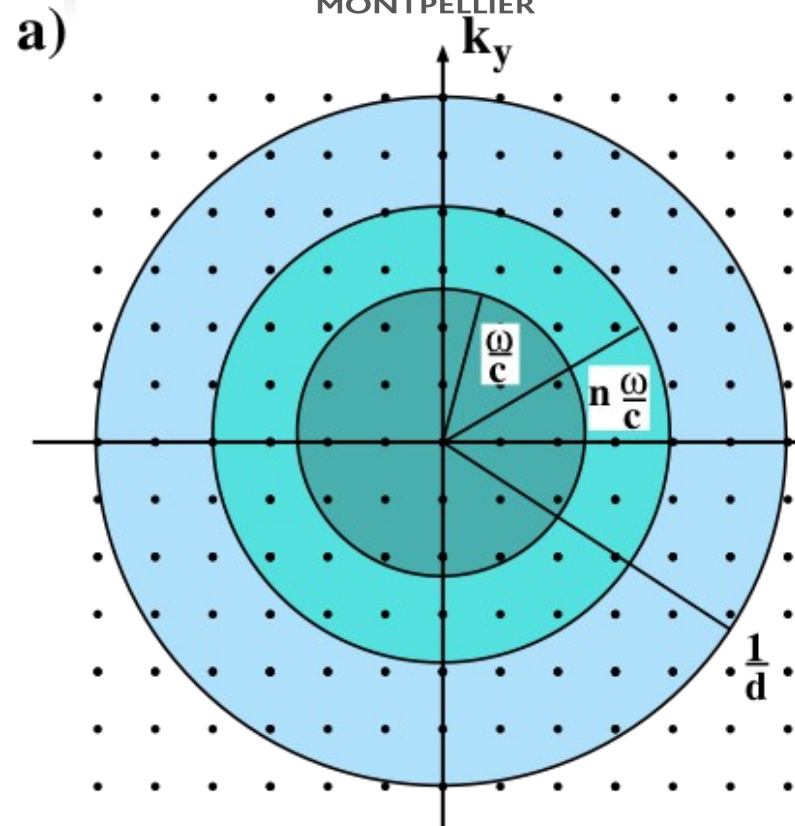
Emmanuel.rousseau@univ-montp2.fr



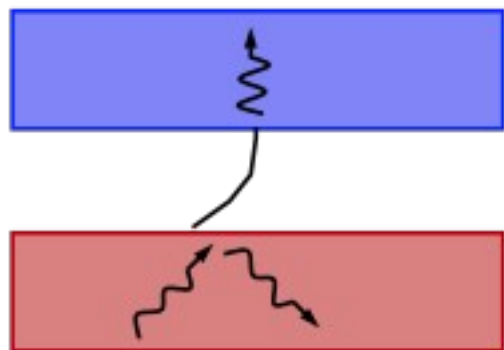


# FTIR contribution

LABORATOIRE  
CHARLES  
COULOMB  
MONTPELLIER



$$h_{evan}^p(u, d, T) = \frac{3}{2\pi^3} \frac{g_0}{\lambda_T^2} h^0(u) \times \int_0^{\gamma_{\max}} \tilde{\gamma} d\tilde{\gamma} \frac{4\text{Im}(r_{31})\text{Im}(r_{32})e^{-2k_T\tilde{\gamma}ul}}{|1 - r_{31}r_{32}e^{-2k_T\tilde{\gamma}ul}d|^2}$$



$$T = \frac{4\text{Im}(r_s)^2 e^{-2\gamma''d}}{|1 - r_s^2 e^{-2\gamma''d}|^2} \leq 1$$

$$T = \frac{4\text{Im}(r_s)^2 e^{-2\gamma'' d}}{|1 - r_s^2 e^{-2\gamma'' d}|^2} \leq 1$$

The trick  $T = 1 - \varepsilon(d)$

Rousseau et al. JAP 111 014311 (2012)

$$h_f^s(\omega, d, T) \simeq h^0(\omega) \frac{n_r^2 - 1}{2} \times$$
$$\left\{ 1 + \left(\frac{k_0 d}{2}\right)^2 (\varepsilon_r - 1) \ln \left[ \left(\frac{k_0 d}{2}\right)^2 (\varepsilon_r - 1) \right] \right\}$$

$$h_f^s(\omega, d, T) \simeq \boxed{h^0(\omega) \frac{n_r^2 - 1}{2}} = 4\sigma T^3 (n_r^2 - 1) \left\{ 1 + \left(\frac{k_0 d}{2}\right)^2 (\epsilon_r - 1) \ln \left[ \left(\frac{k_0 d}{2}\right)^2 (\epsilon_r - 1) \right] \right\}$$

See also

Rousseau et al. APL **95** 231913 (2009)

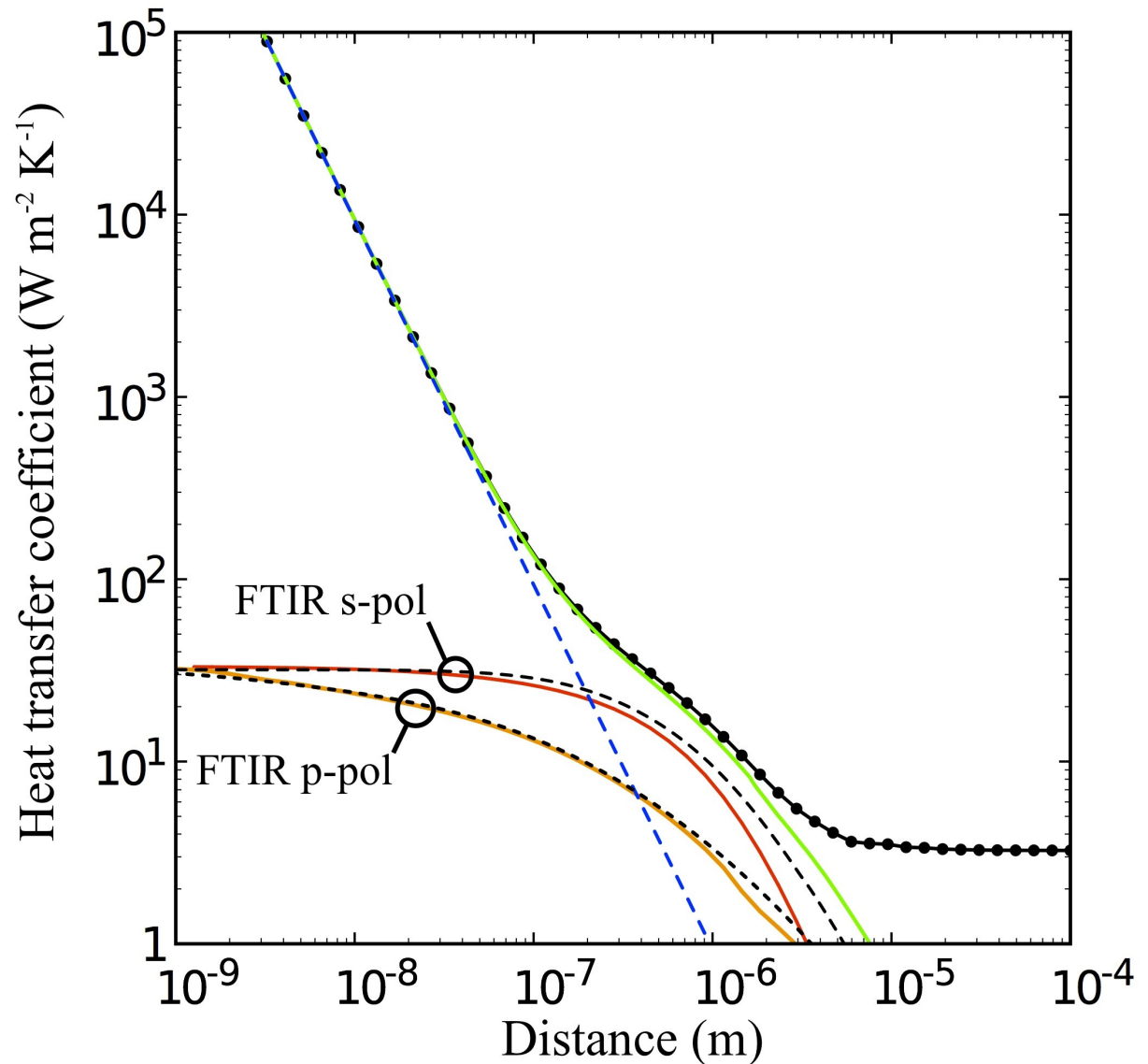
Rousseau et al. Journal of Quantitative Spectroscopy and Radiative heat transfer **111** 1005 (2010)

# Characteristic distance

Surface modes coupling  
Contribution OK

Frustrated Total Internal  
Reflections OK

Now I can estimate when  
SPP dominates the heat  
transfer



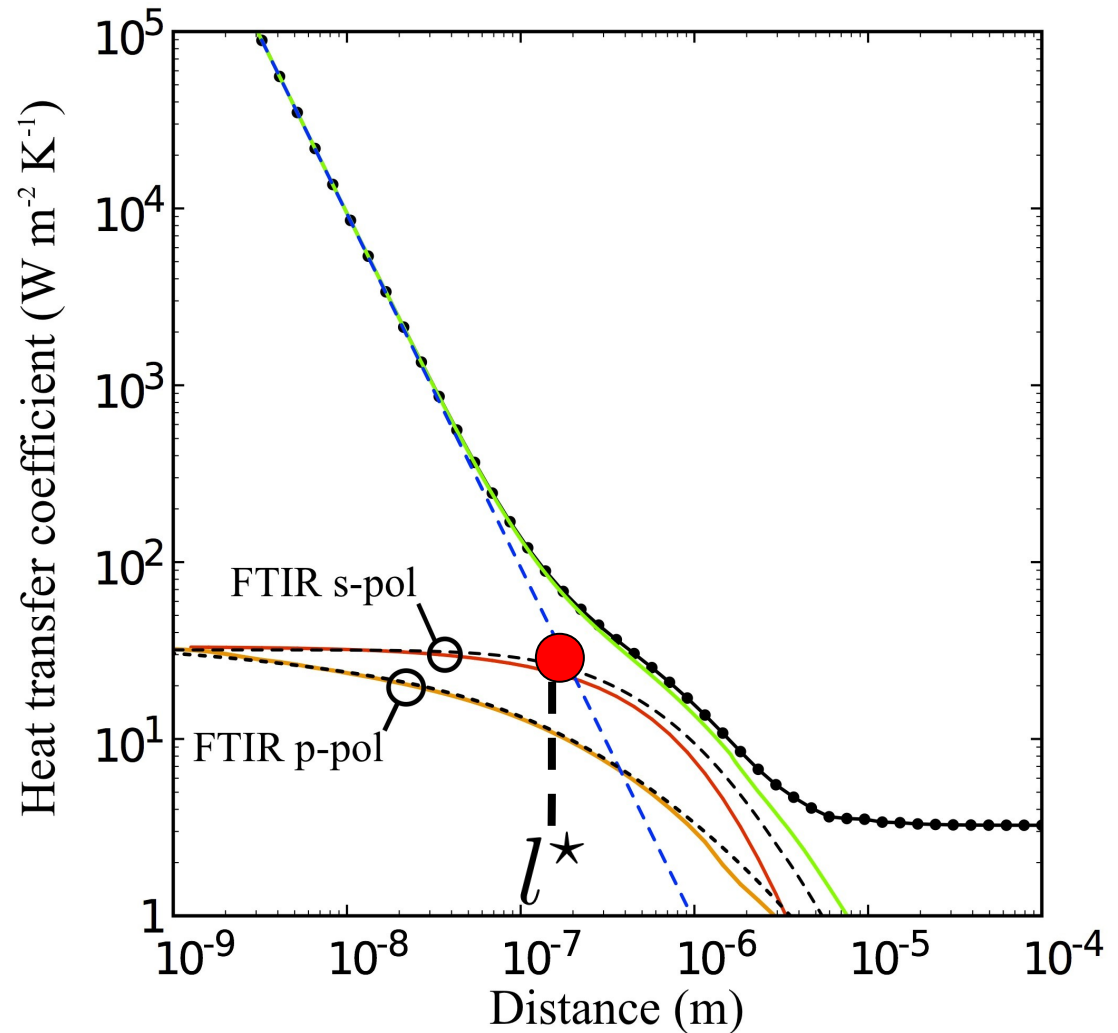
From Rousseau et al. JAP 111 014311  
(2012)

# Characteristic distance

$$l^* = \sqrt{\frac{\delta G(T)}{4\sigma T^3(n_r^2 - 1)}}$$

$T$	$\lambda_T$	$l^*$
100 K	29 $\mu\text{m}$	19 nm
300 K	10 $\mu\text{m}$	170 nm
500 K	6 $\mu\text{m}$	110 nm

For SiC-SiC



Better for  $\text{Al}_2\text{O}_3$   $l^* \sim 400\text{nm}$  @ 300 K

Choosing temperature:

Total displacement *vs* angle requirement

Not really temperature requirements for metals

To measure the  $1/d^2$  law:

Room temperature is more suitable for materials supporting resonance near  $10\ \mu\text{m}$

*But in any cases nanometer gaps have to be reached*



## 1- Two main ingredients:

*The heat transfer coefficient*

*The thermal conductance*

## 2-Plane-Plane experiments:

*Is there a good temperature to work at?*

*Measuring  $1/d^2$  law*

## 3- Sphere-Plane experiments

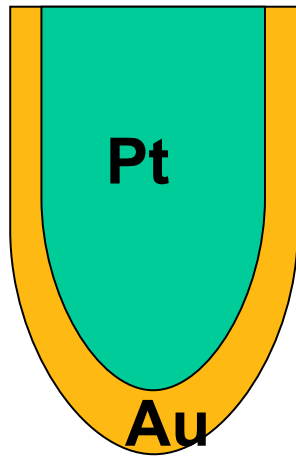
*A sensitive flux-meter*

*The experimental setup*

*Connecting the measurements to the theory*

# Previous non plane-plane geometries explored

SEM  
Thermocouple

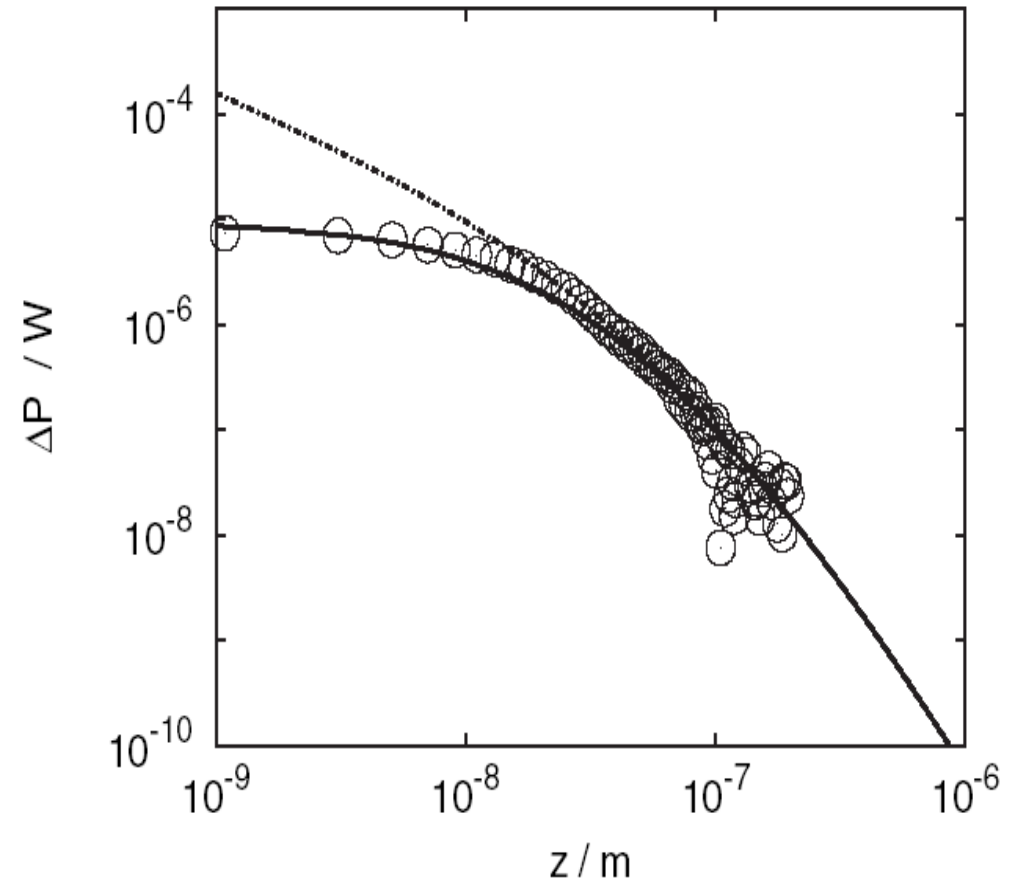


$R_c \sim 50\text{nm}$

Au

$T \sim 100\text{ K}$

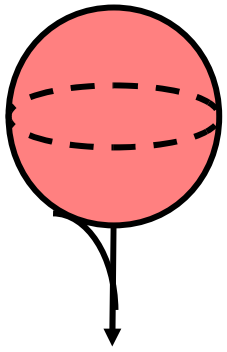
Radiatif transfer vs distance



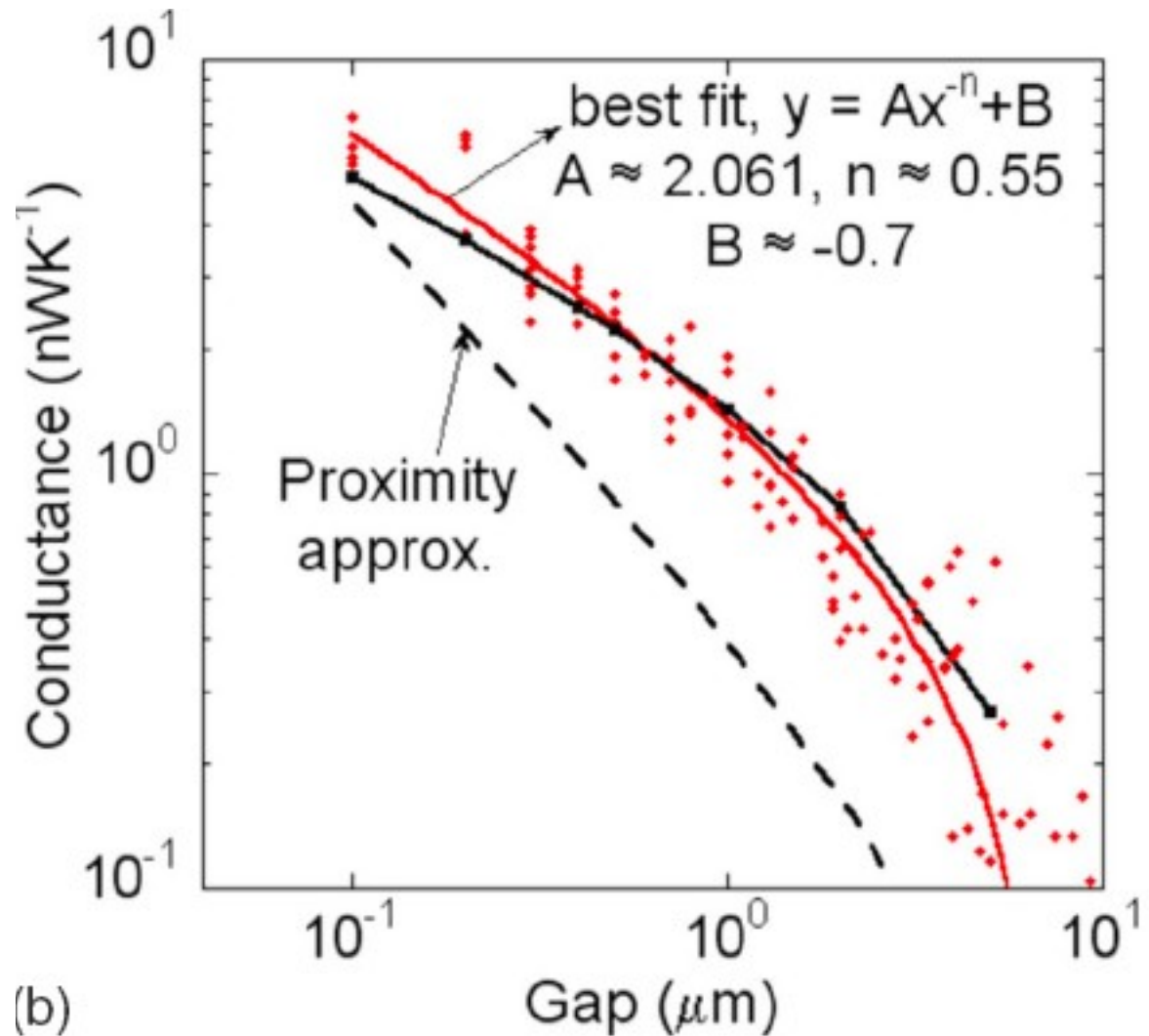
*Kittel et al. , PRL 95 p 224301 (2005)*

# The first sphere-plane experiments

Silica sphere



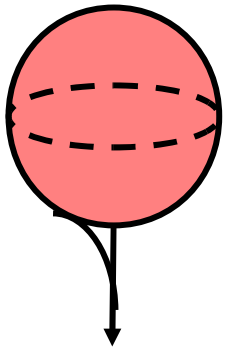
Silica plate



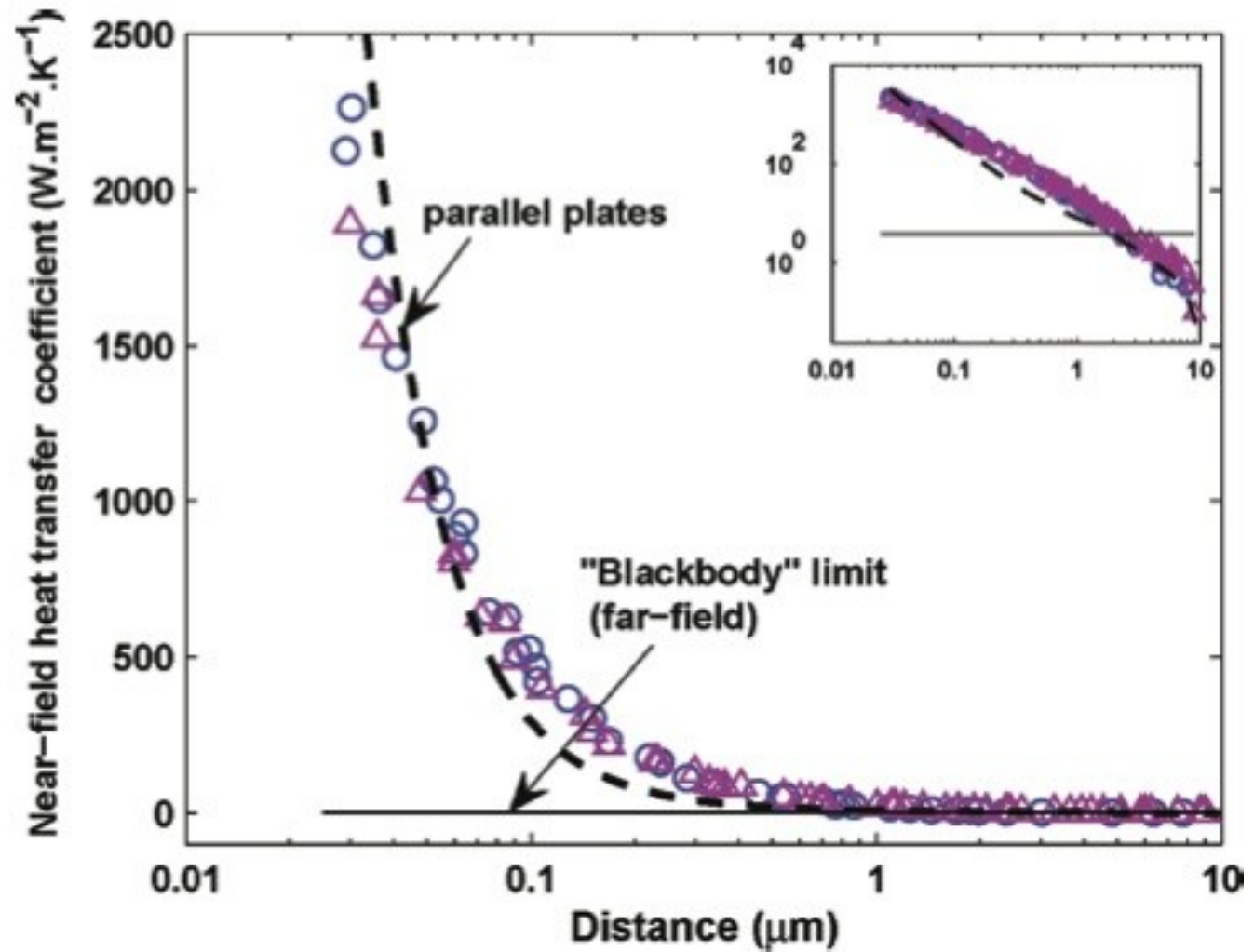
From Narayanaswamy A, G. Chen PRB **78**, 115303 (2008)

# The first sphere-plane experiments

Silica sphere

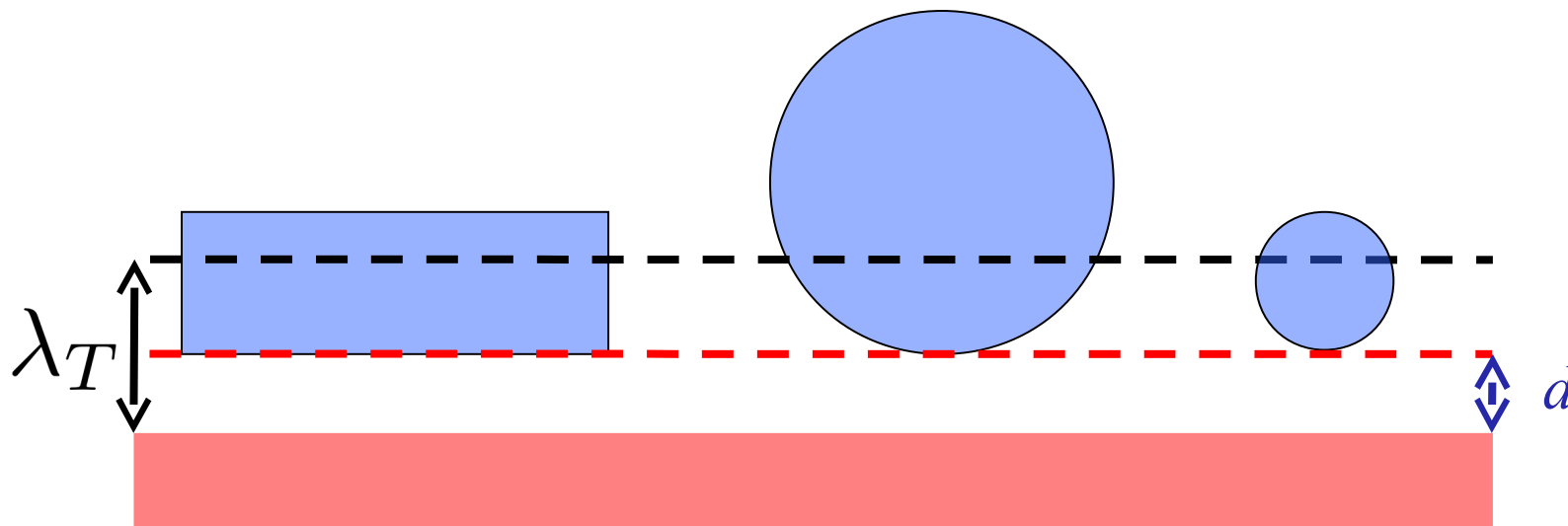


Silica plate



From S. Shen et al. NanoLetters 9, 2909 (2009)

Sphere radius should not be too large



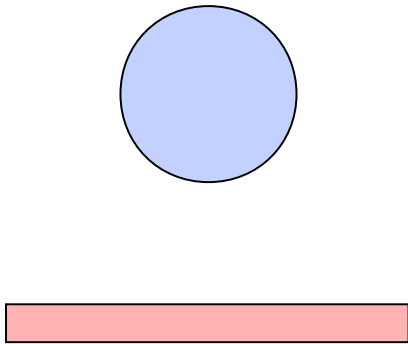
# Flux estimate

## Far-field

$$\varphi = 4\pi R^2 F_{s-p} \varepsilon \sigma (T_h^4 - T_c^4)$$

$$\varphi = 2\pi R^2 4\varepsilon \sigma T^3 \Delta T$$

$$G = 2\pi R^2 4\varepsilon \sigma T^3$$



$$R \sim 20 \mu m$$

Flux in the order of  $\sim 100$  nW

Not a too small sphere

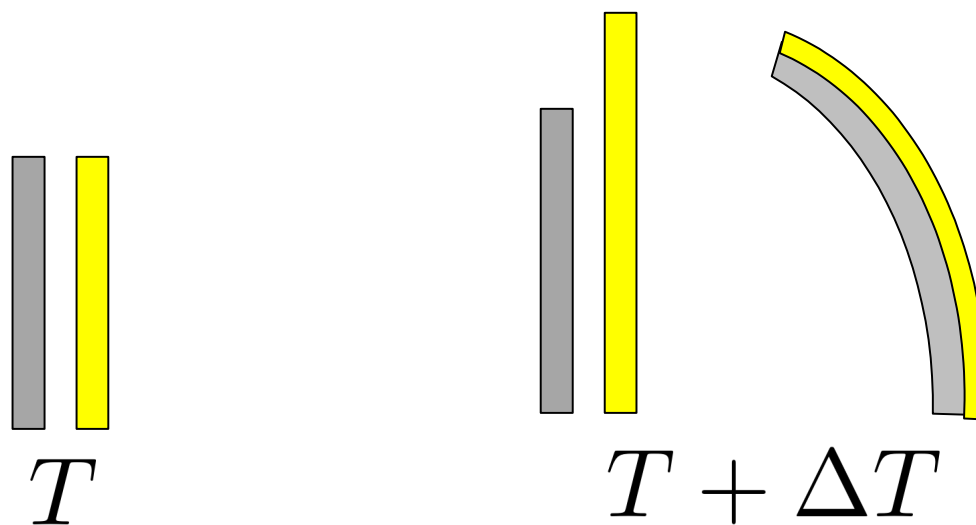
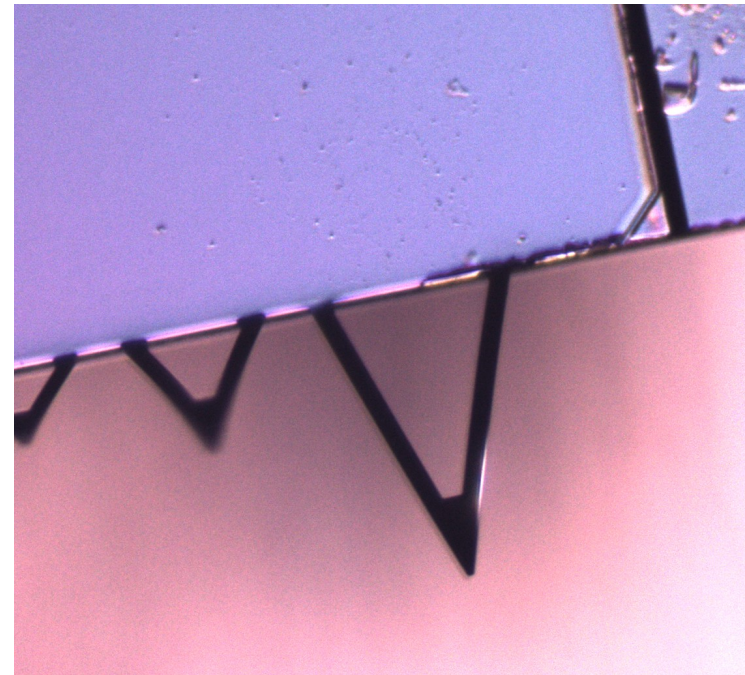
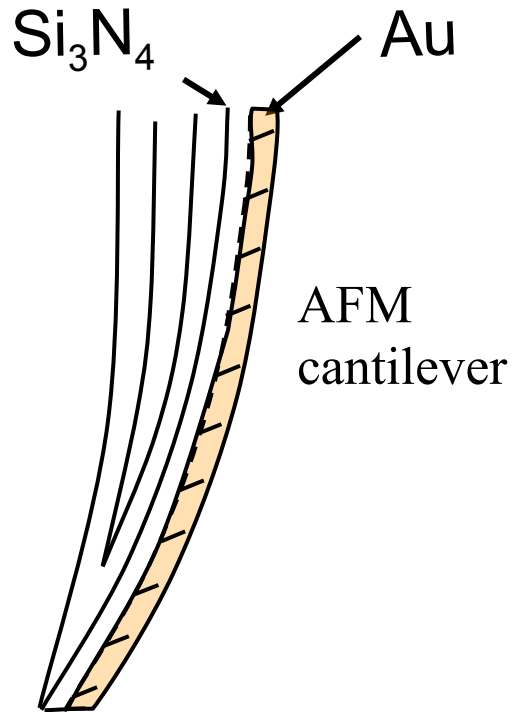
Materials supporting surface modes (glass  $\sim$  silica )

A very sensitive fluxmeter



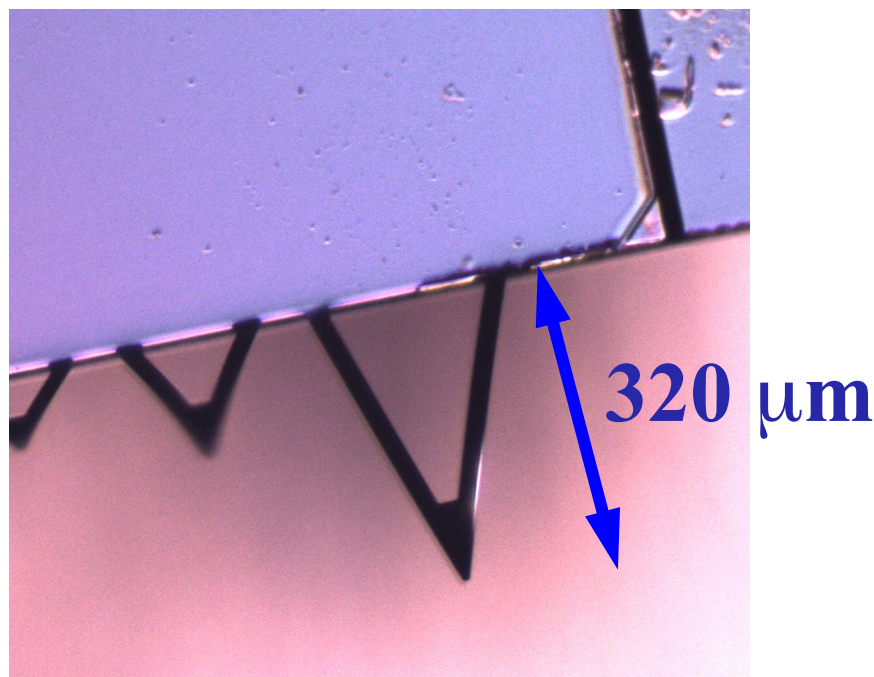
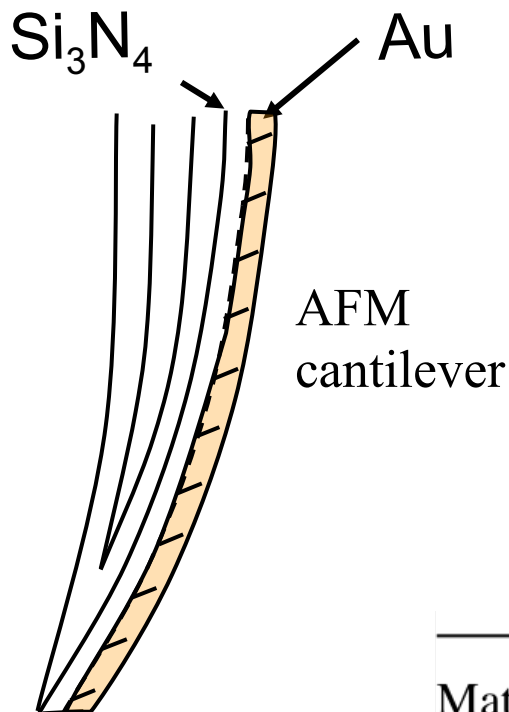
LABORATOIRE  
CHARLES  
COULOMB  
MONTPELLIER

# A very sensitive fluxmeter: a bimorph based on an AFM cantilever





# A very sensitive fluxmeter: a bimorph based on an AFM cantilever

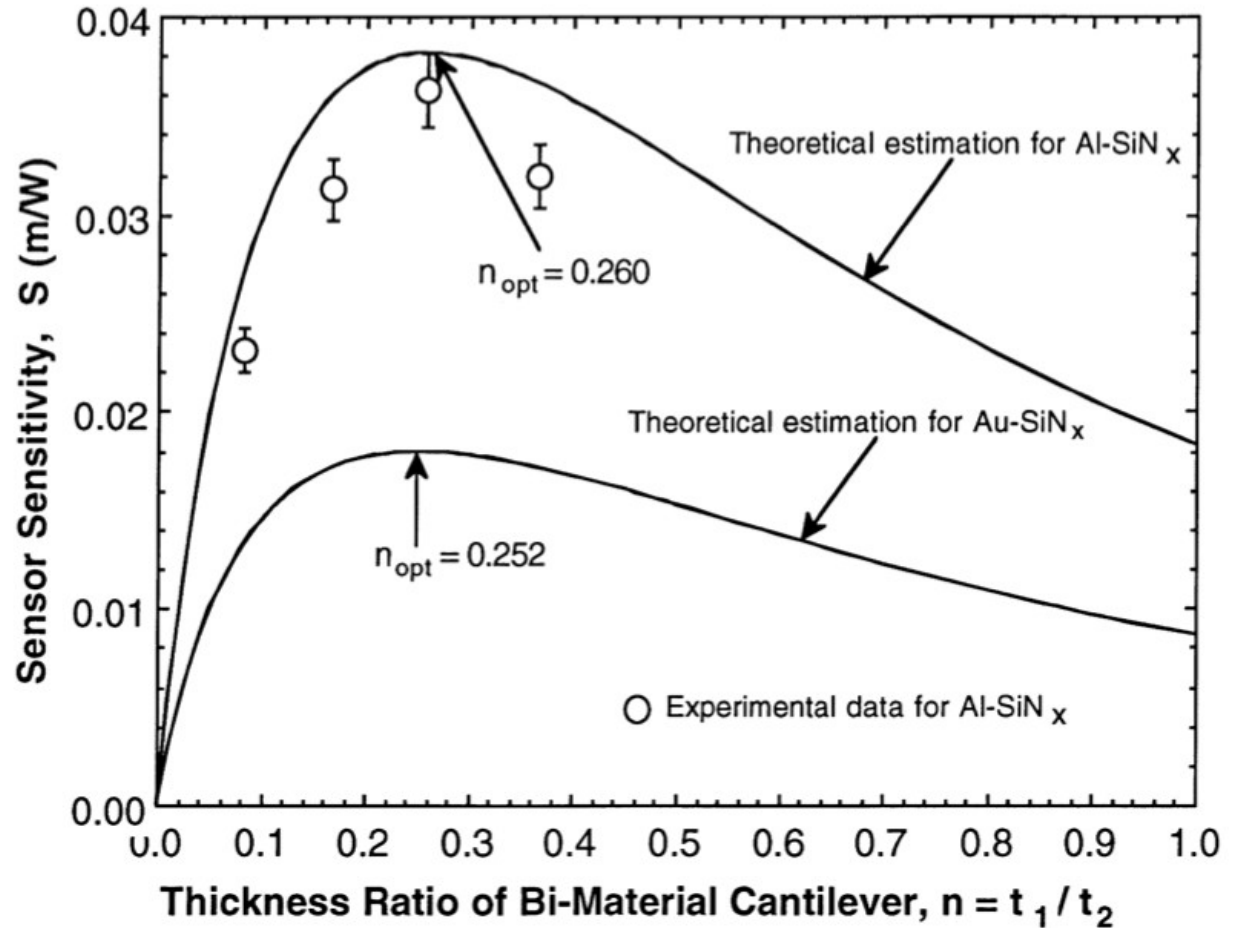
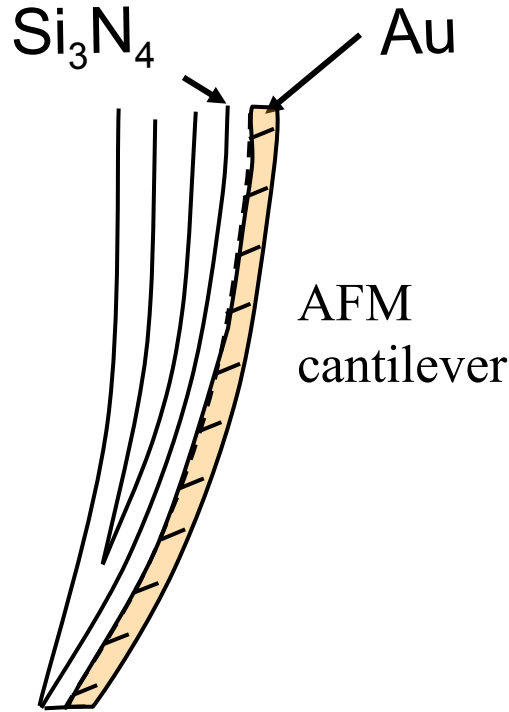


Material	Elastic modulus, $E$ [ $10^{11}$ N m <sup>-2</sup> ]	Thermal expansion coefficient, $\alpha$ [ $10^{-6}$ K <sup>-1</sup> ]
Silicon nitride	1.80	0.8
Silicon	1.00	2.6
Gold	0.73	14.2
Aluminum	0.80	23.6





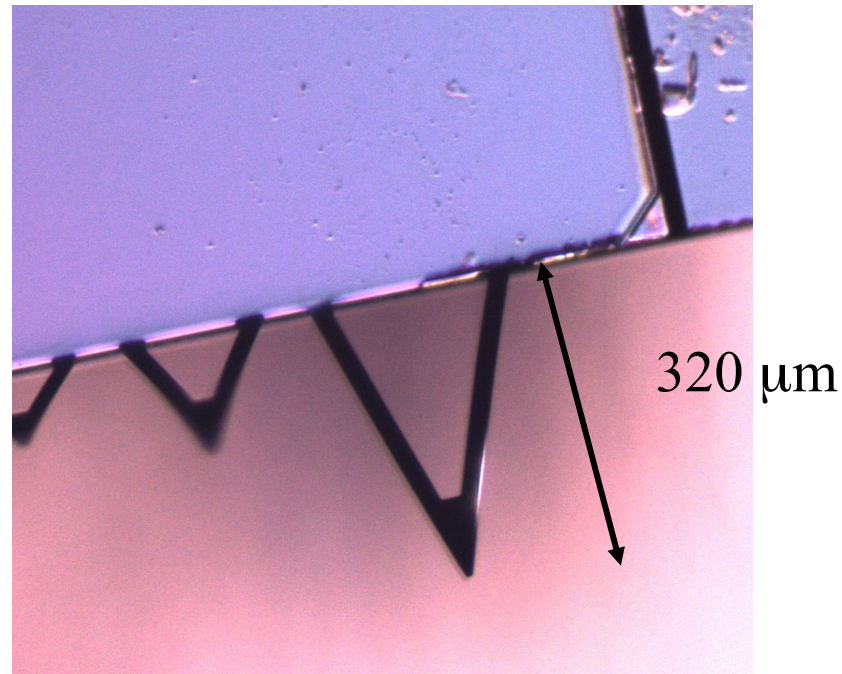
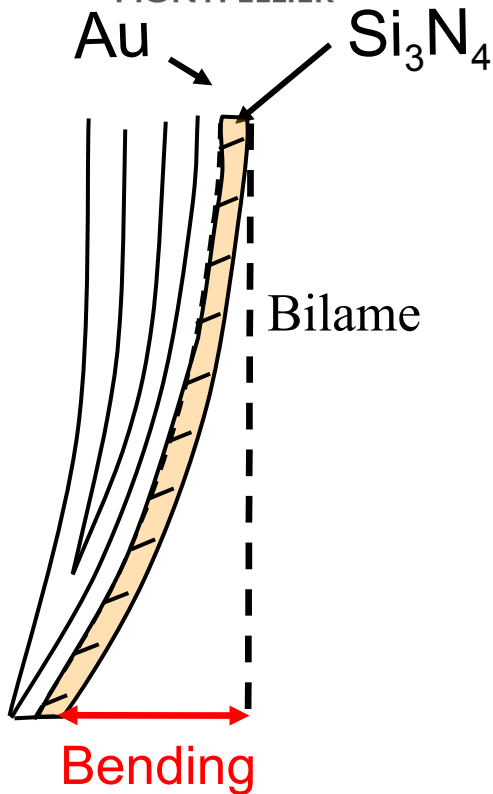
# A very sensitive fluxmeter: a bimorph based on an AFM cantilever



From Lai et al. Sensors and Actuators A 58 113 (1997)

Optimisation is difficult because of SiN<sub>x</sub>  
Commercially available SiN<sub>x</sub>/Au

# A very sensitive fluxmeter

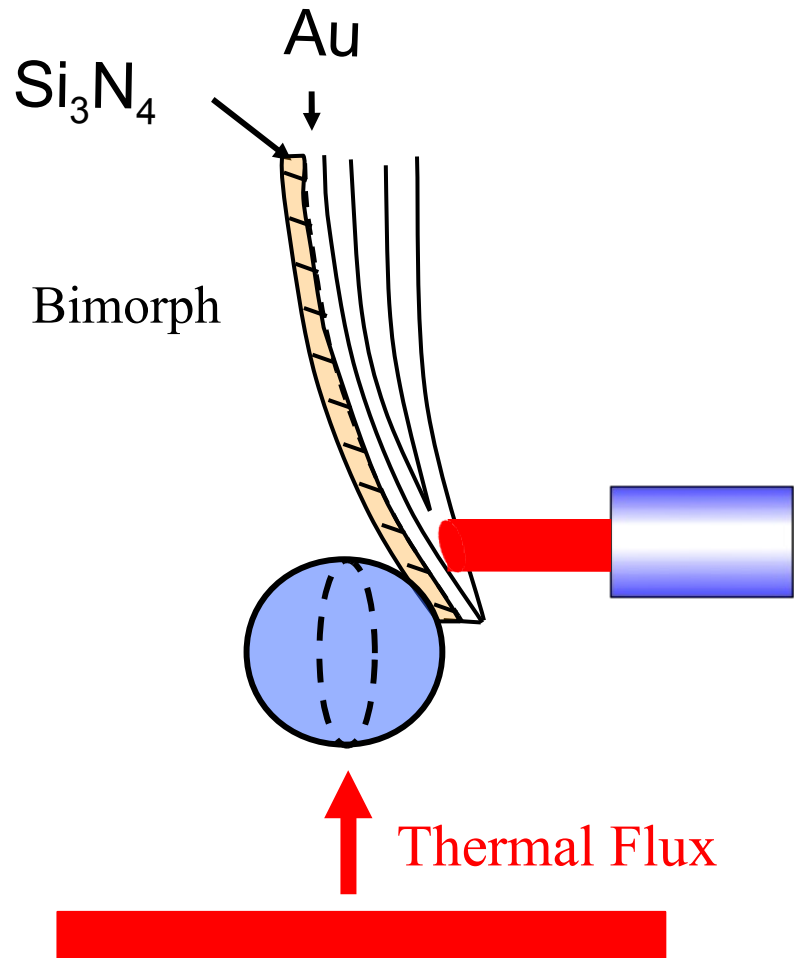


Cantilever bending is proportional to the thermal flux through the bimorph

Flux measurement => bending measurement

Sensitivity: 0.1 nm ♥ T~10 μK ♥ ~10 pW

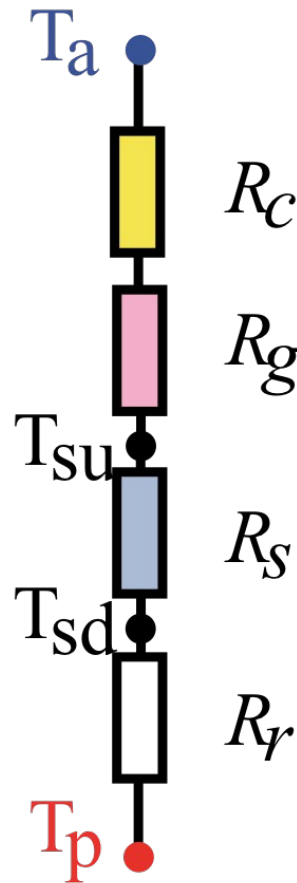
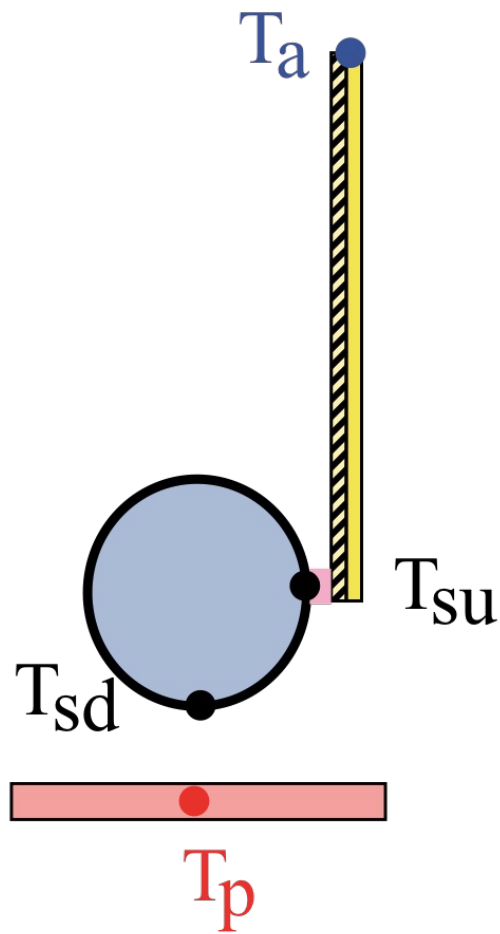
# Sketch of the experiment



Experimental difficulties taken into account:

- 1- Glue a sphere at the end of the cantilever. **Work in vacuum!**
- 2- bring a hot plate closer and closer
- 3- measure the bending of the bimorph

# Thermal circuit



$$R_r = 160 \cdot 10^6 \text{ K/W} \quad @ 10 \mu\text{m}$$

$$R_r = 53 \cdot 10^6 \text{ K/W} \quad @ 50 \text{ nm}$$

$$R_g = 0.6 \cdot 10^6 \text{ K/W}$$

$$R_s = 40 \cdot 10^3 \text{ K/W}$$

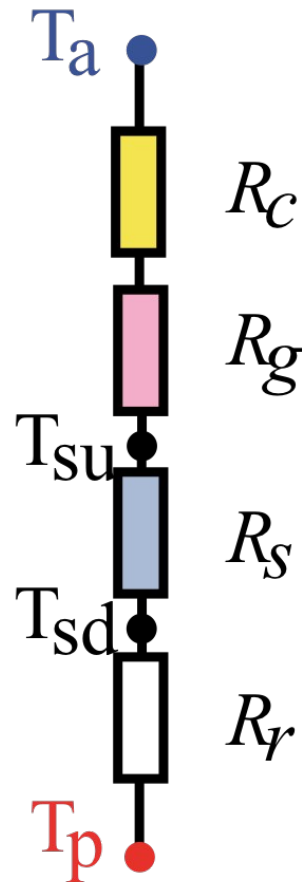
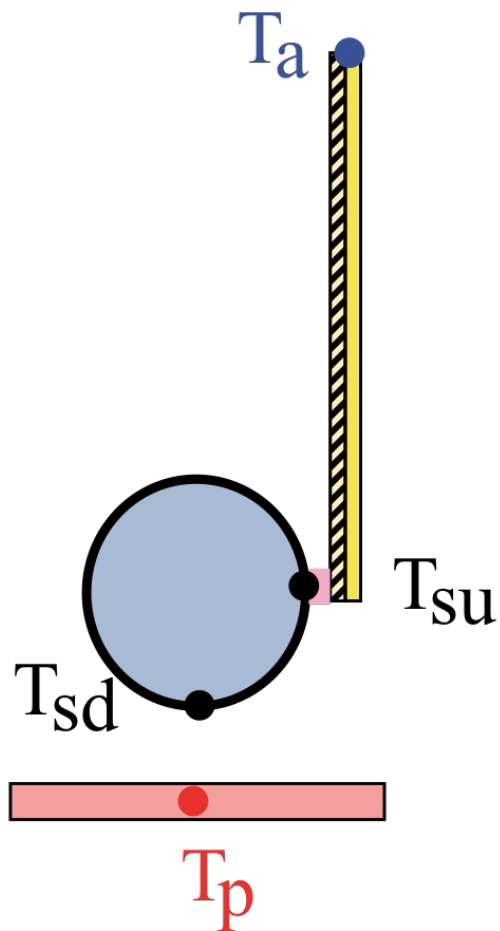
$$R_c = 0.4 \cdot 10^6 \text{ K/W}$$

$$\frac{T_{sd} - T_p}{T_a - T_p} \geq 0.98$$

$$\Delta T_{can} \sim 5 \text{ mK}$$

$$T_{sd} - T_p = \frac{R_r}{R_r + R_s + R_g + R_c} (T_a - T_p)$$

# Thermal drift- Time constant



$$\Delta T_{can} \sim 5 \text{ mK}$$

Room temperature  $T_a$  should be highly stable  
**during measurement**

Measurements are done fast:  
100ms between two data points

Diffusion time in sphere  $\sim 0.5 \text{ ms}$   
Diffusion time in cantilever  $\sim 2 \text{ ms}$

Less than 2 min for one acquisition curve

# The measurement chamber

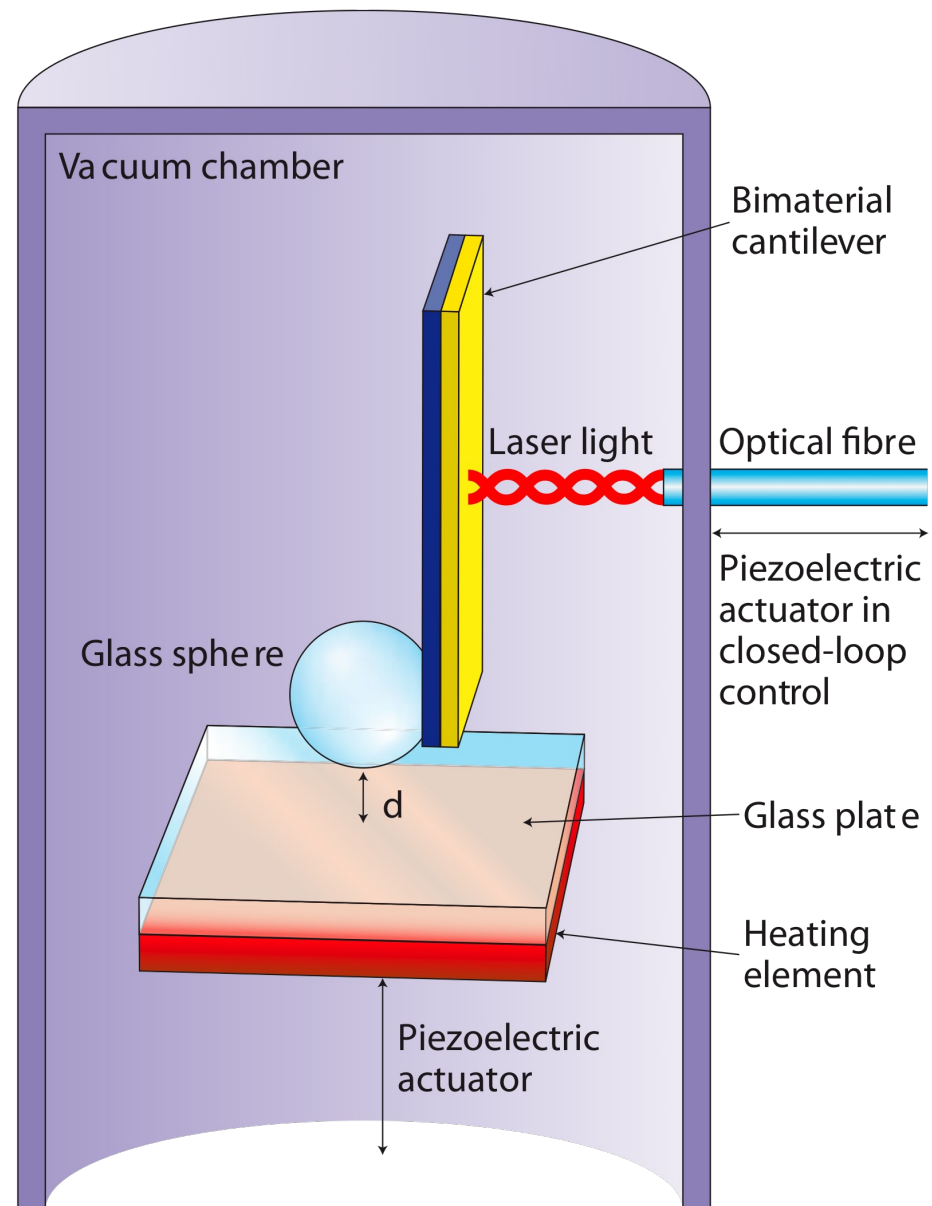
Vacuum condition  $P \sim 10^{-6}$  mbar

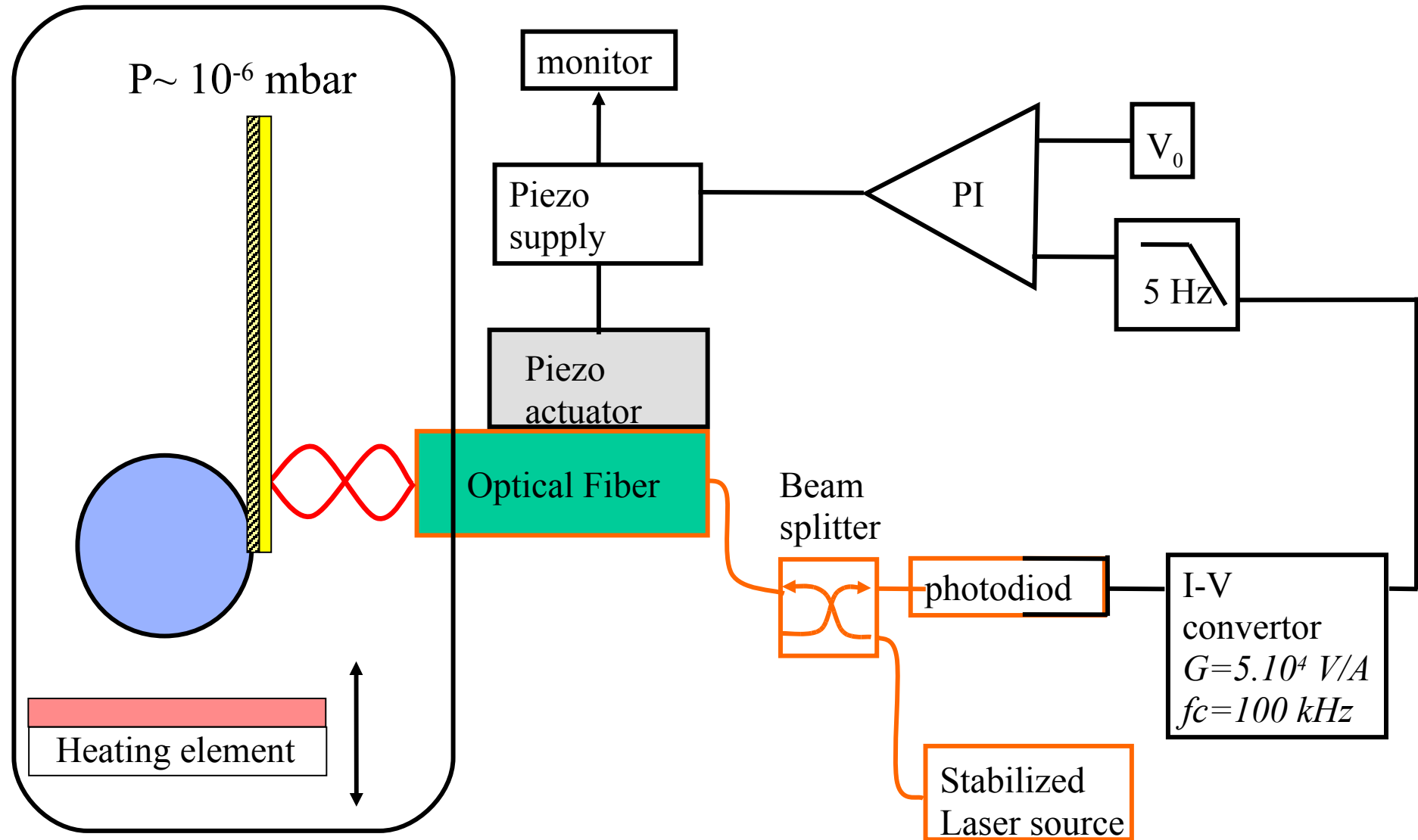
Vertical geometry to prevent bending from electrostatic forces

Plate heated by a Peltier element

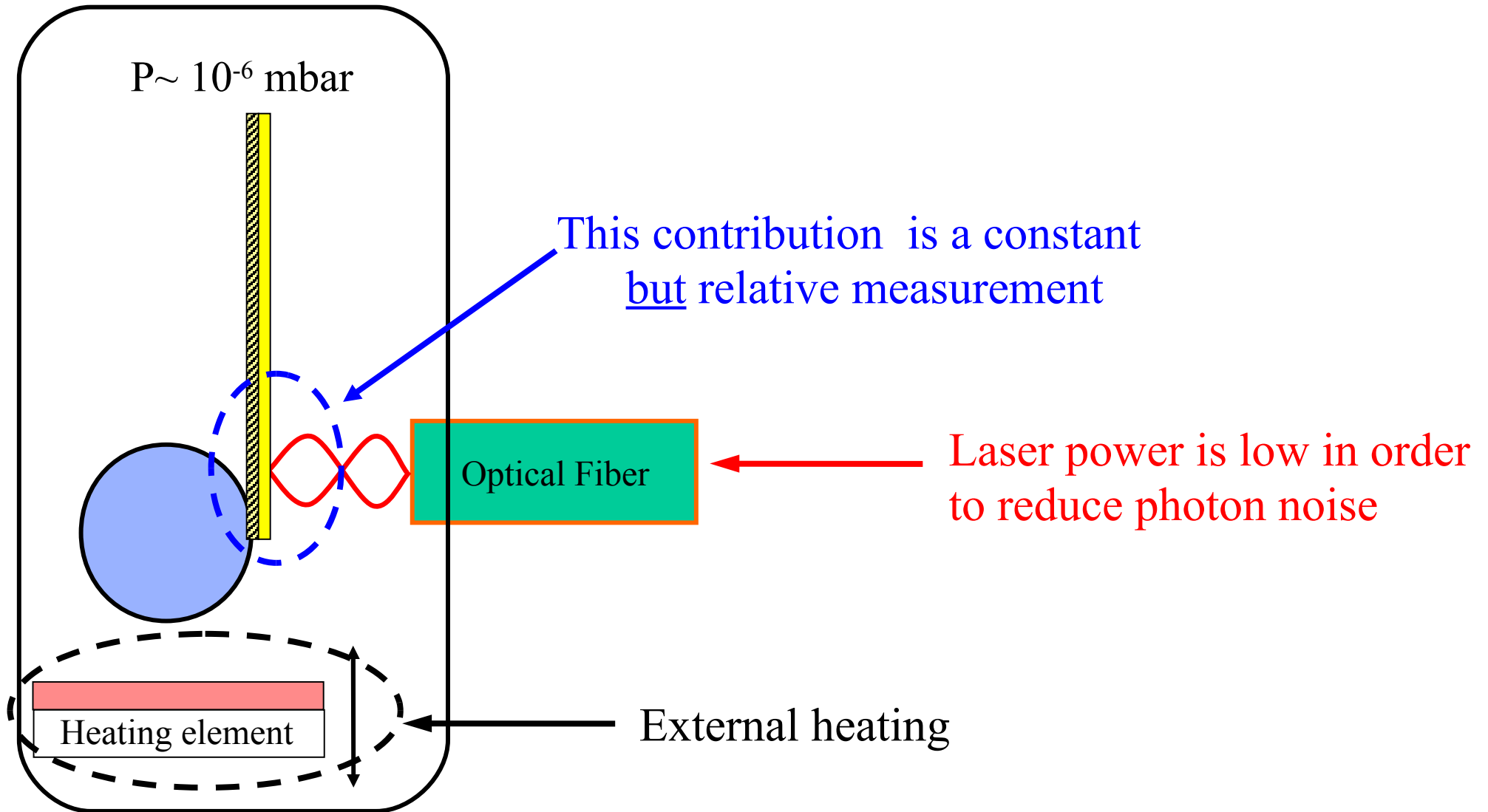
Plate is risen to the sphere with nanometric precision (step 7 nm)

Sphere-plate distance change from  $5 \mu\text{m}$  to contact



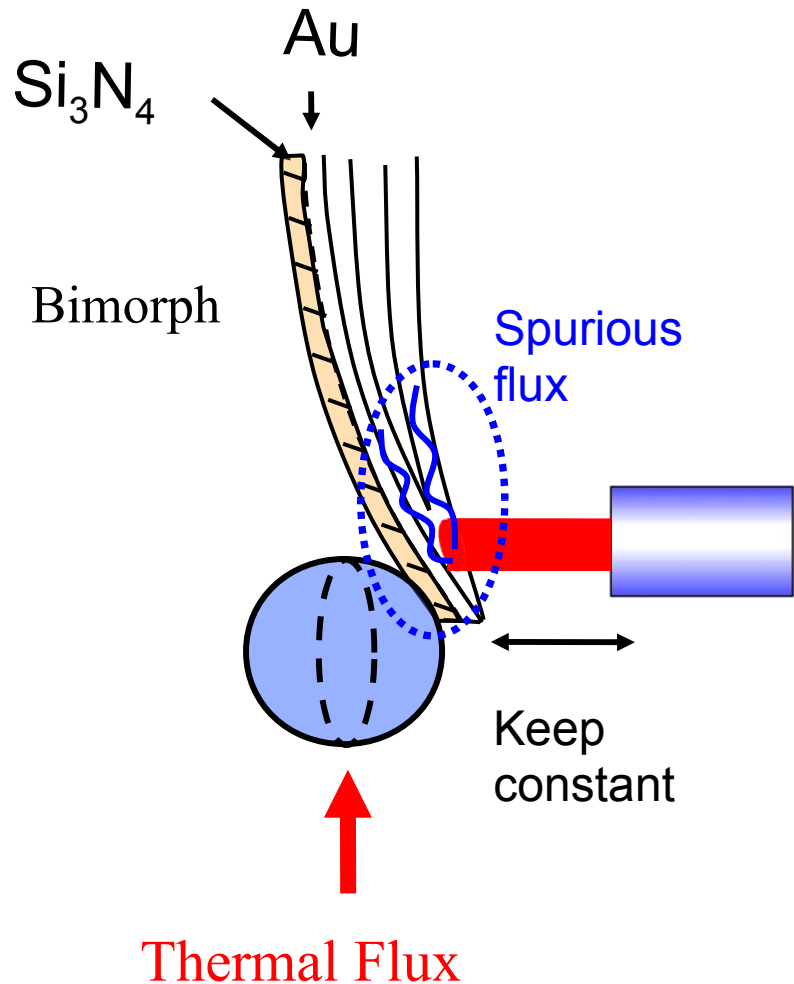


# Some precautions





# Some precautions



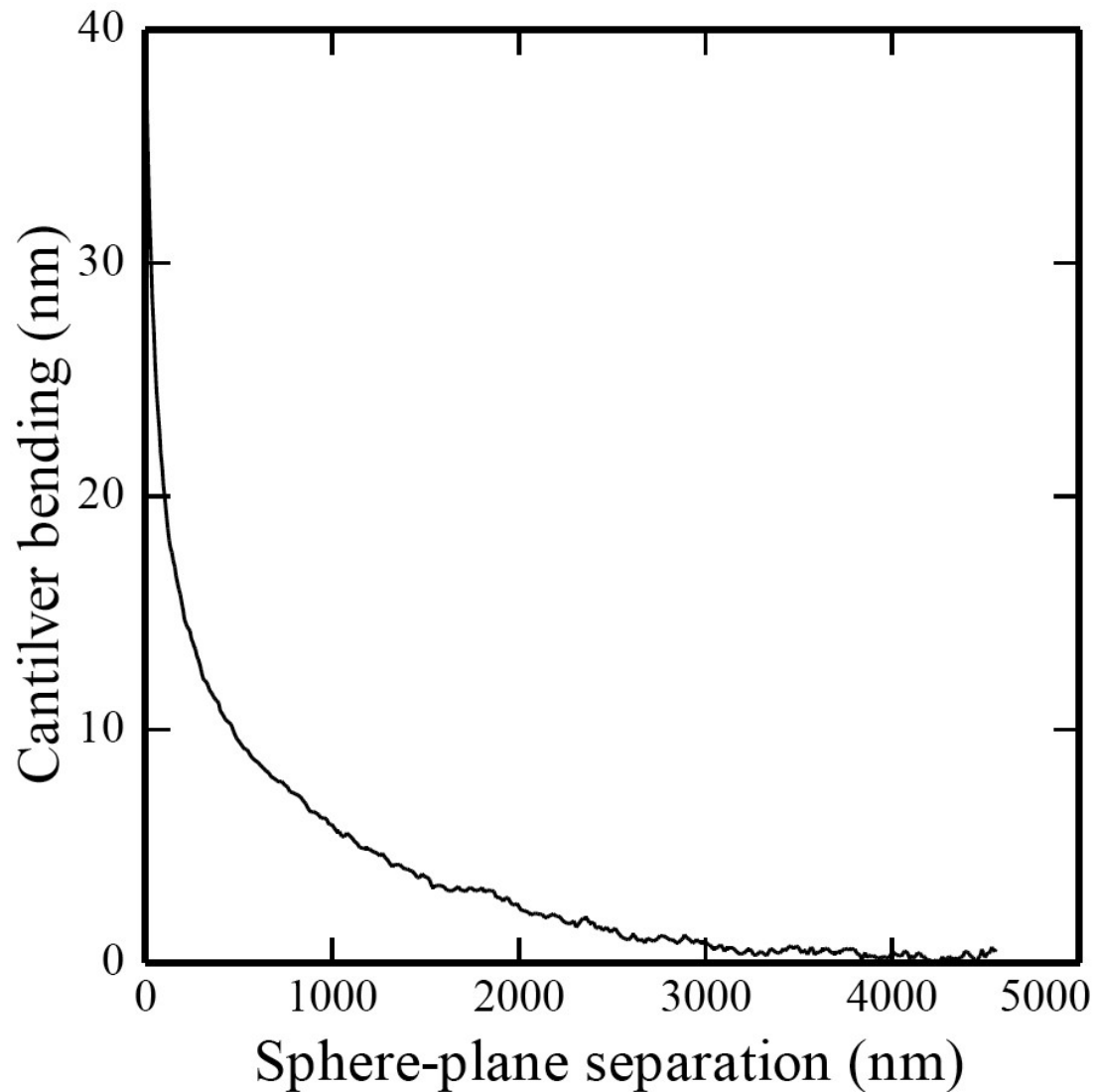
Experimental difficulties taken into account:

1- thermal drift: 1 nm/min

2- spurious flux due to laser. Low laser power to reduce heating and photon noise

⇒ closed-loop keeps the distance bimorph-fibre (i.e. spurious flux) constant: we move the fibre

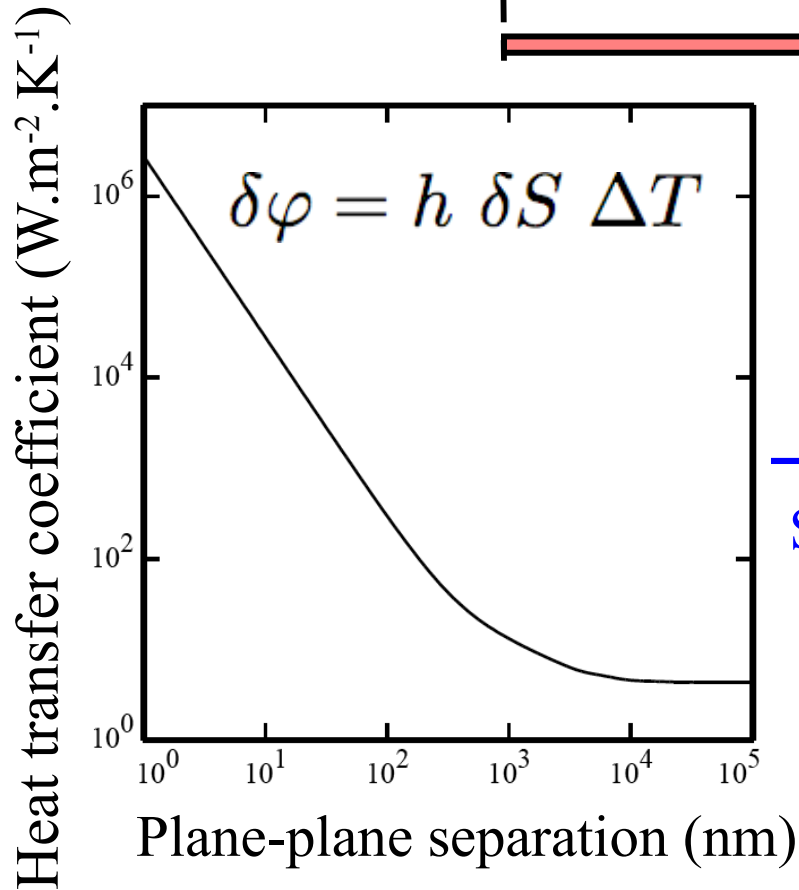
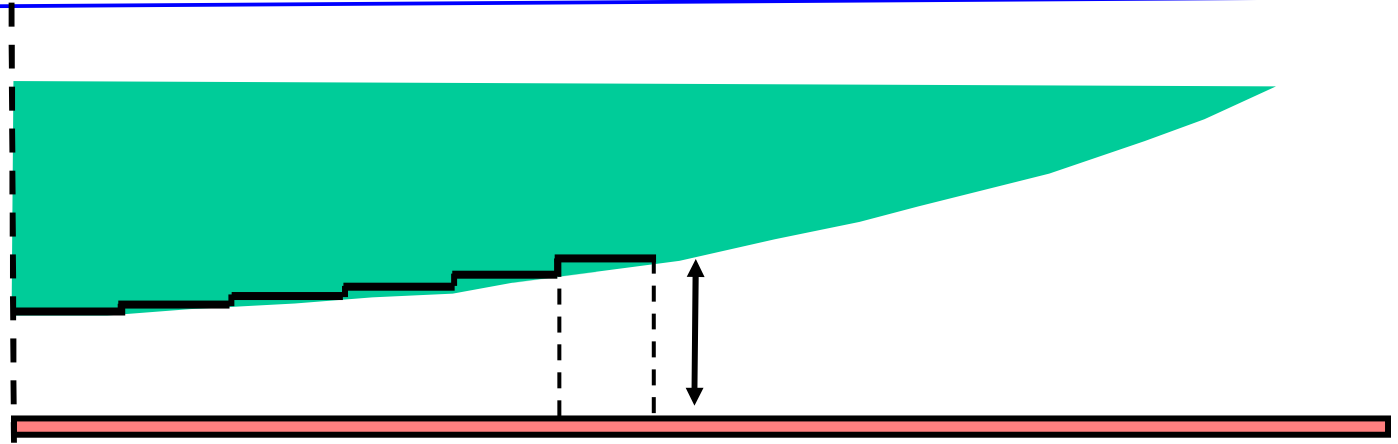
⇒ relative measurement



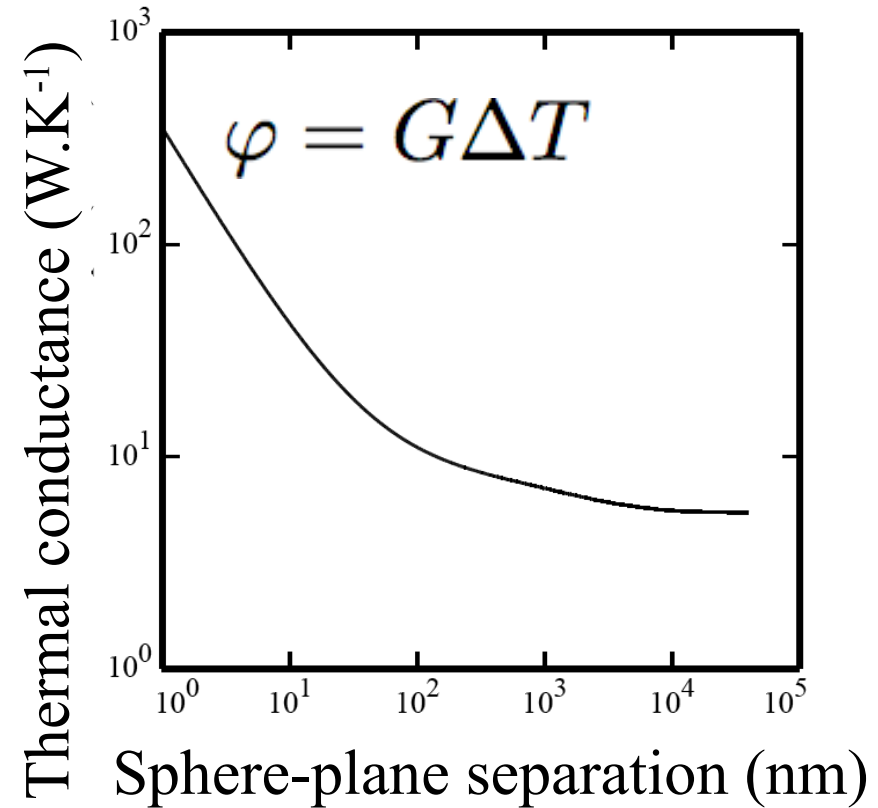
Bending vs sphere-plane distance.

Comparison with theory.

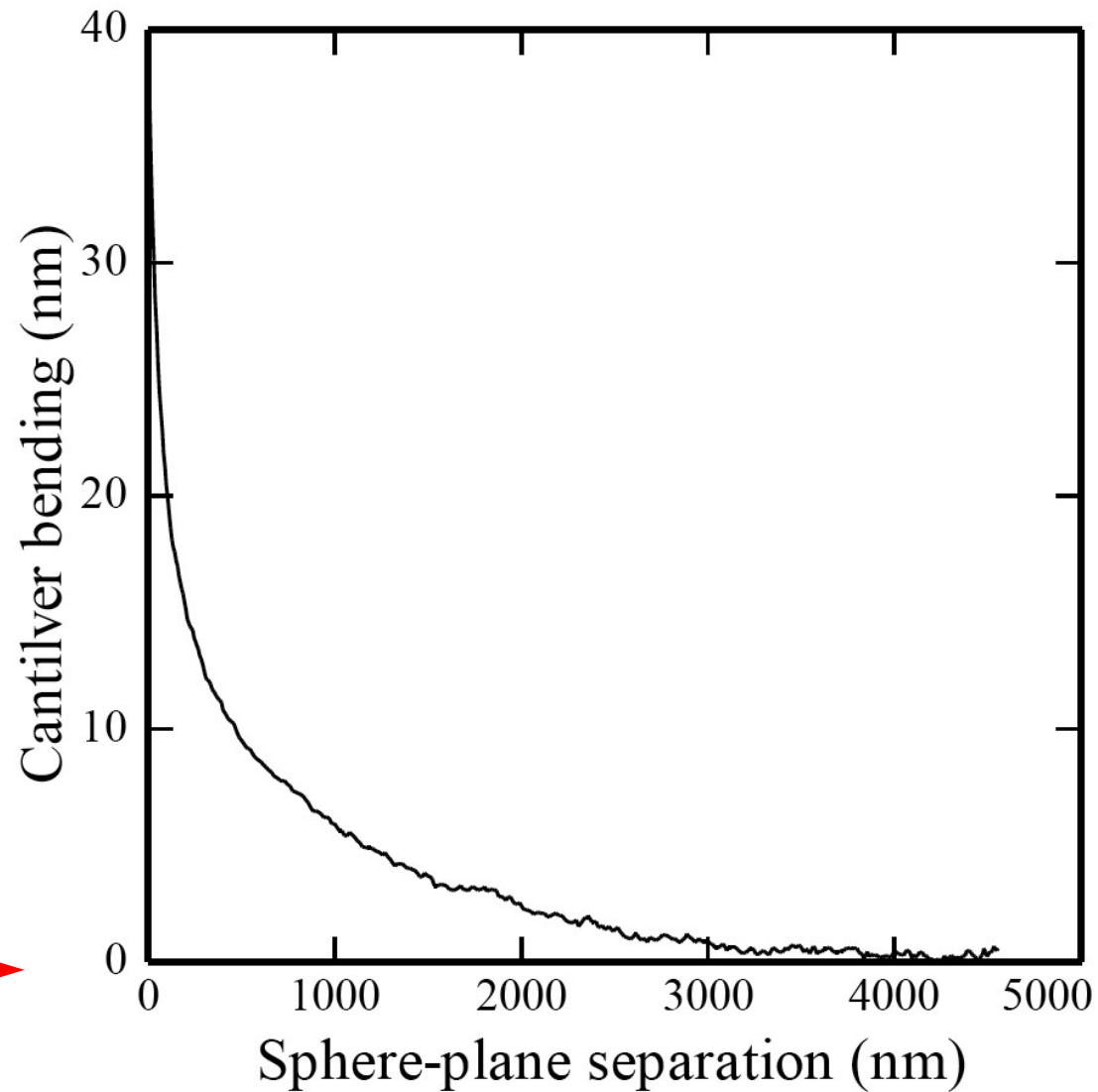
# Derjaguin approximation



Summation



# 1- Y-axis: Far-field contribution.



Zero flux?

No!

Relative measurement!

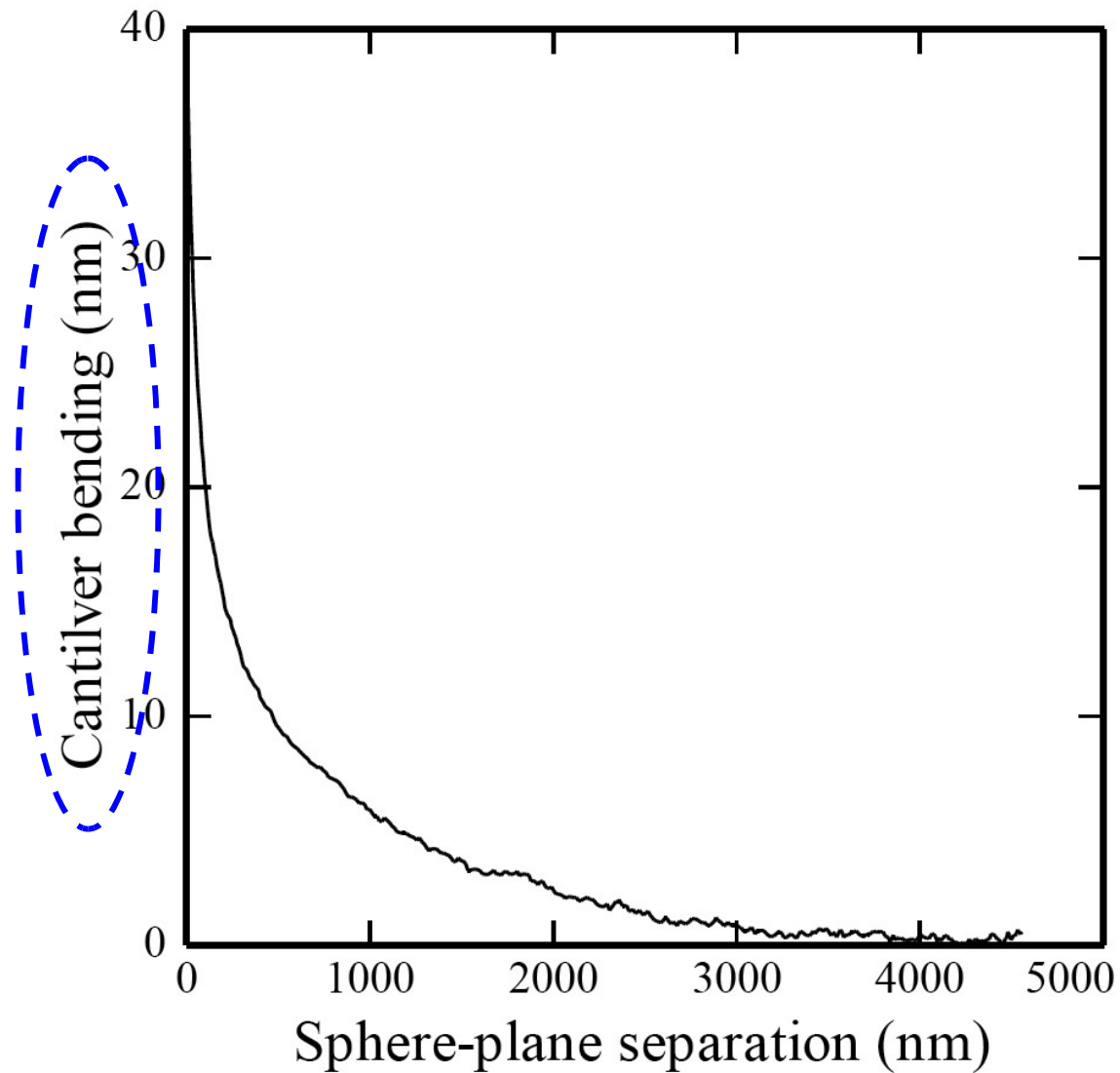


Add far-field contribution

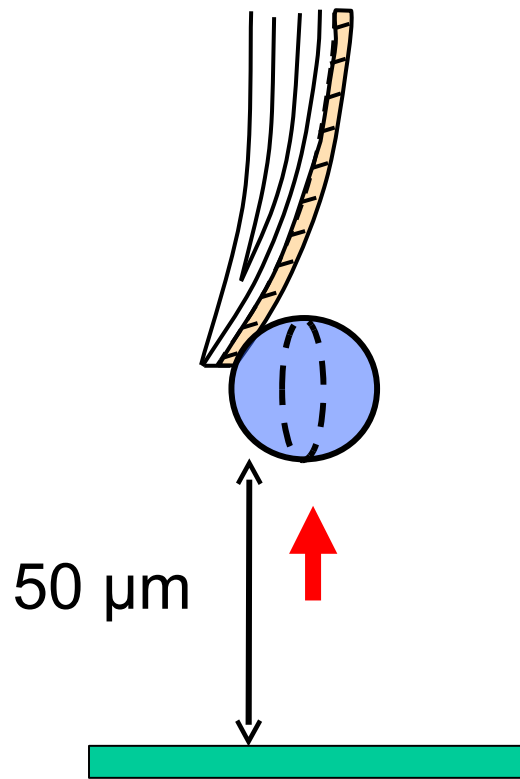
## 2- Y-axis: calibration

$$\phi = H\delta(d)$$

nW  $\longleftrightarrow$  nm

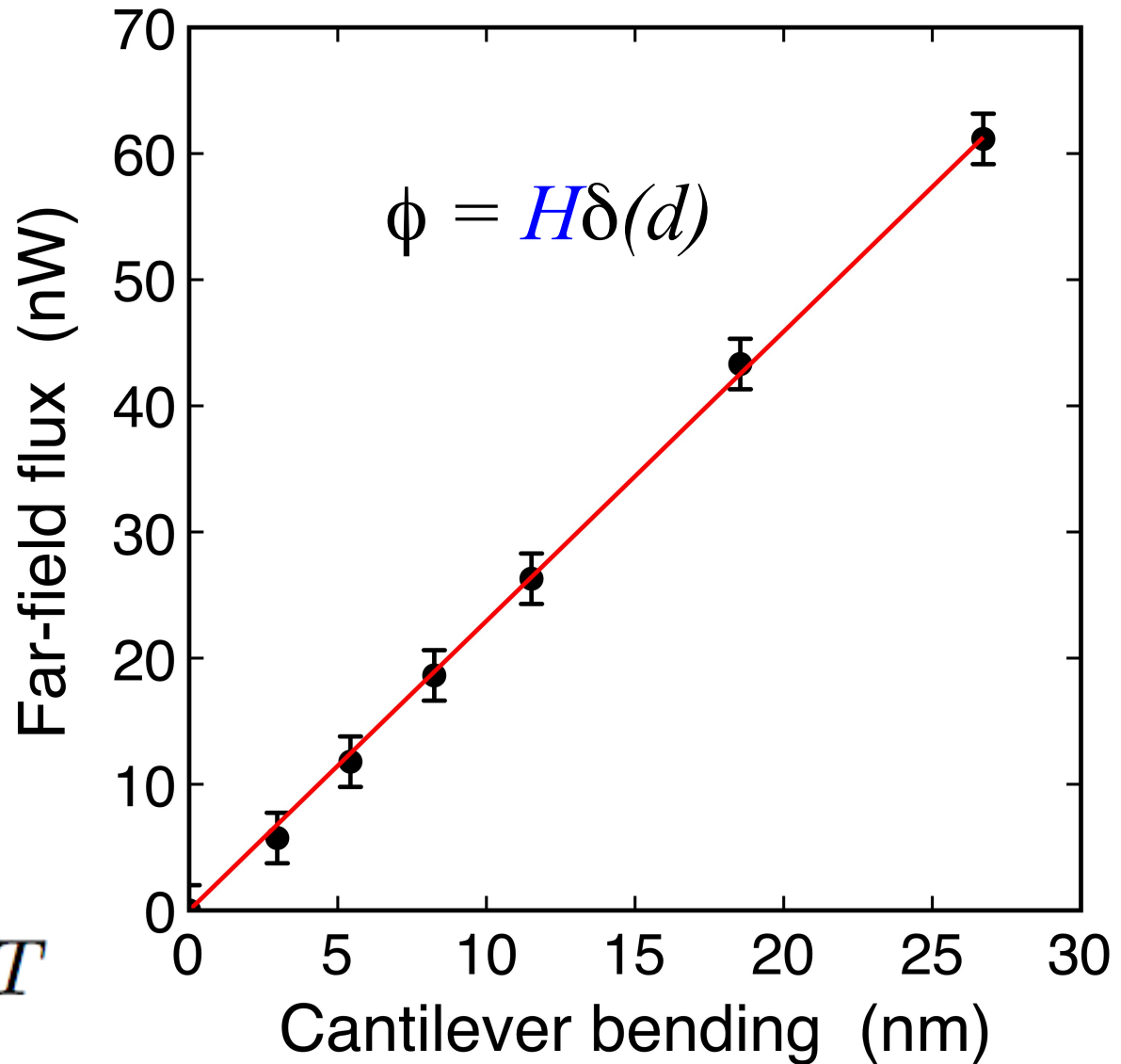


## 2- Y-axis: can be calibrated in far-field



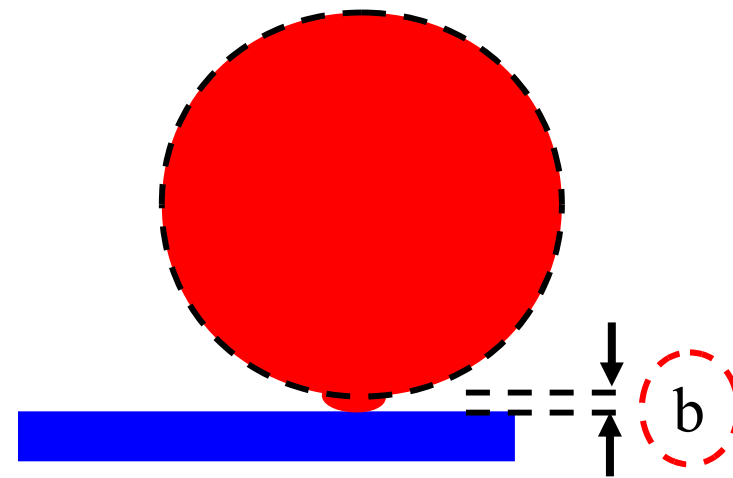
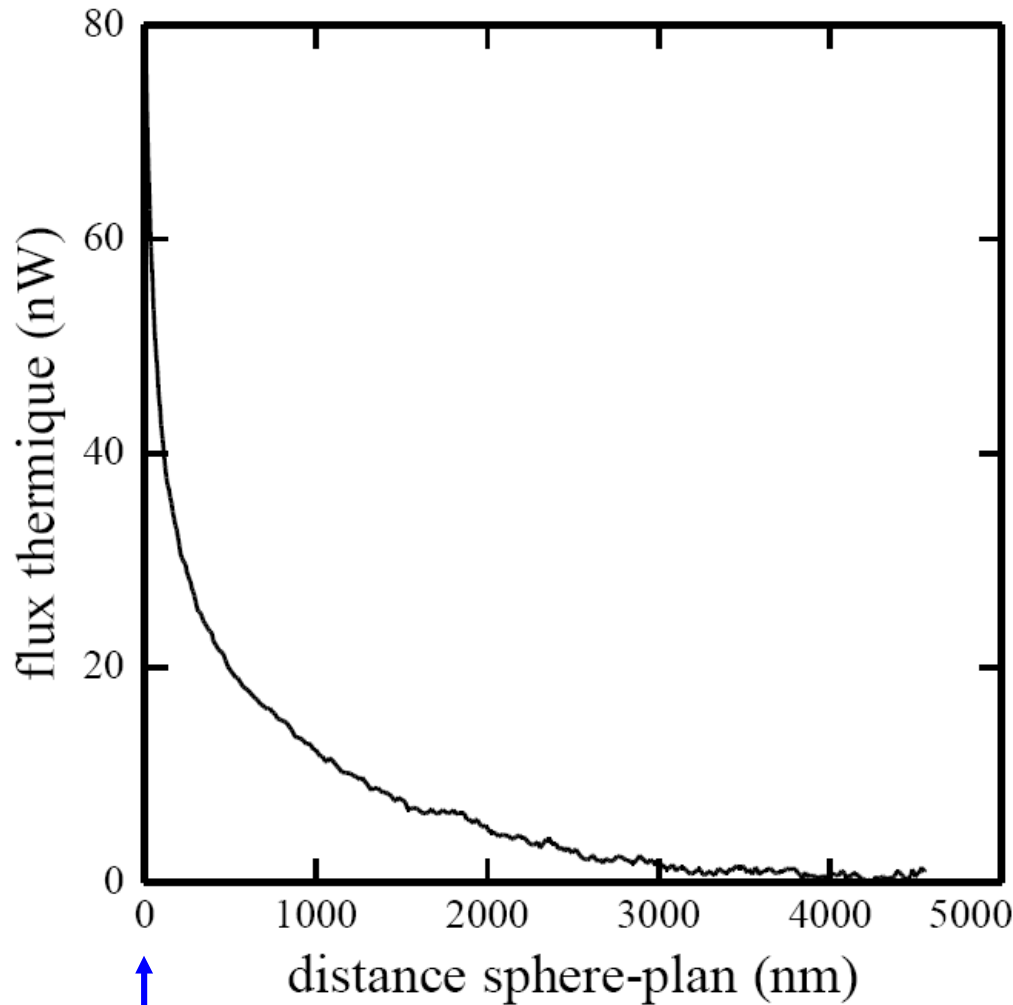
$$\varphi = 2\pi R^2 4\epsilon\sigma T^3 \Delta T$$

Emissivity from literature



$$H = 2.30 \pm 0.05 \text{ nW/nm}$$

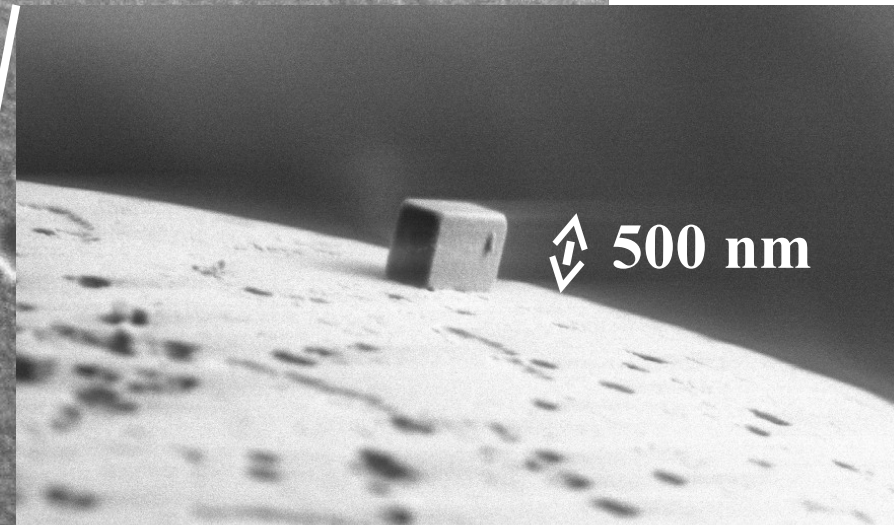
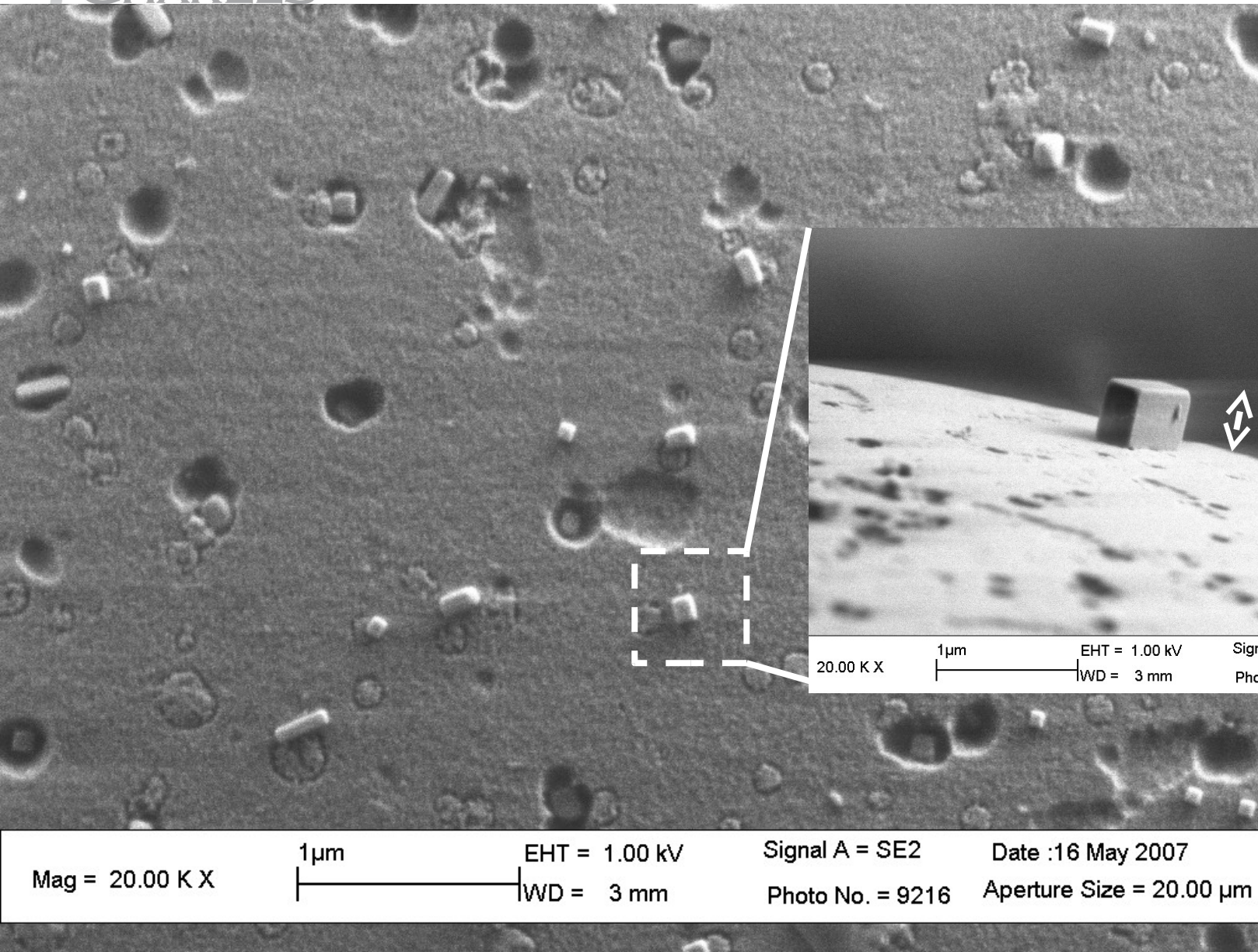
# 3- X-axis: distance mean-surface-plate



No way to be measured.

Contact? No!

# Sphere surface

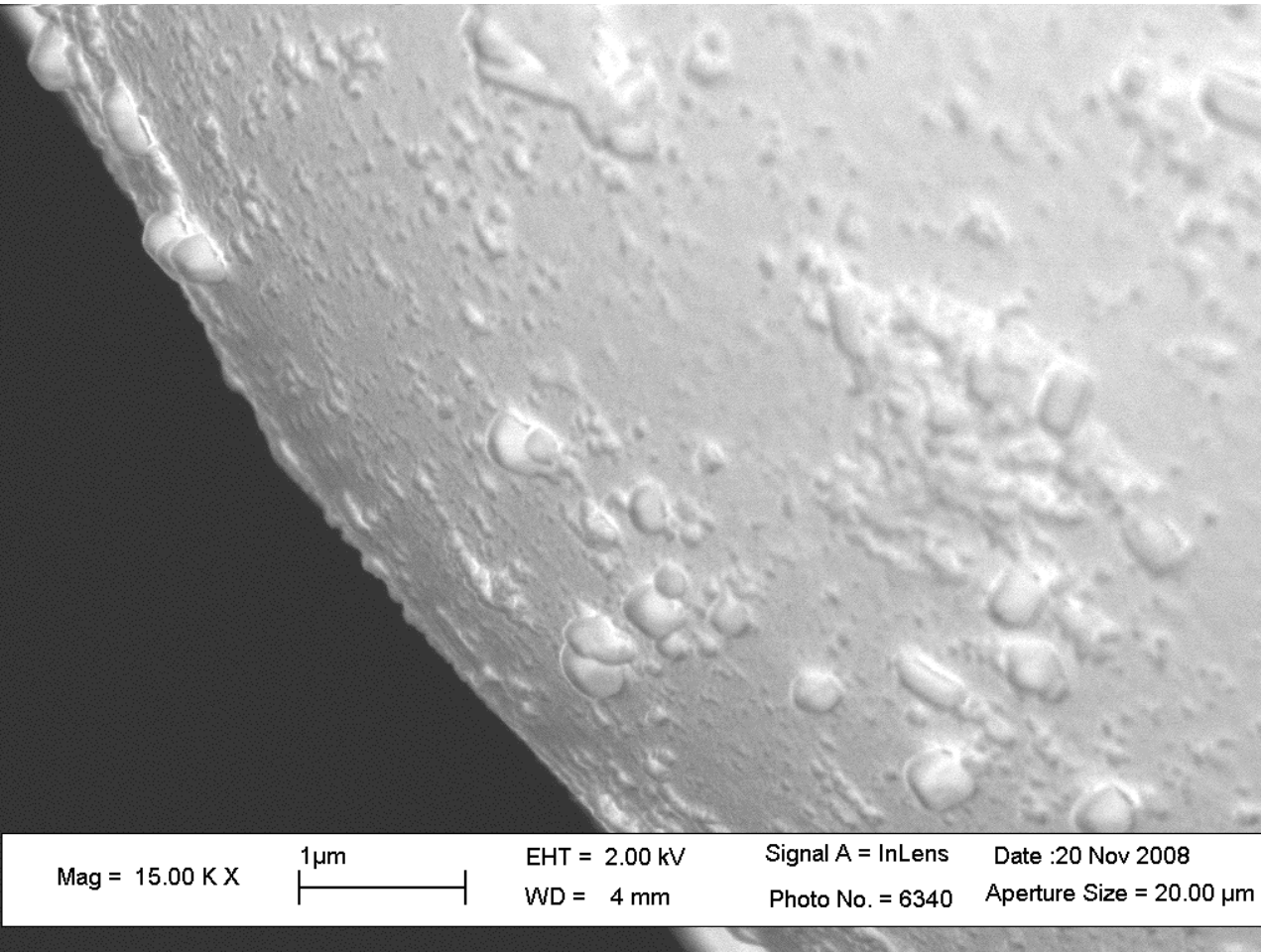


20.00 K X      1 μm      EHT = 1.00 kV      Signal A = SE2      Date :16 May 2007  
 W D = 3 mm      Photo No. = 9218      Aperture Size = 20.00 μm

Mag = 20.00 K X      1 μm      EHT = 1.00 kV      Signal A = SE2      Date :16 May 2007  
 W D = 3 mm      Photo No. = 9216      Aperture Size = 20.00 μm



# Another sphere surface



Asperity size

Up to 150 nm

# Fitting the data

Measured with Thermocouple

$$G_{exp} = G_{ff} + H\delta(d+b)/\Delta T$$

$$G_{ff} = 2\pi R^2 4\sigma\epsilon T^3$$

Compared with

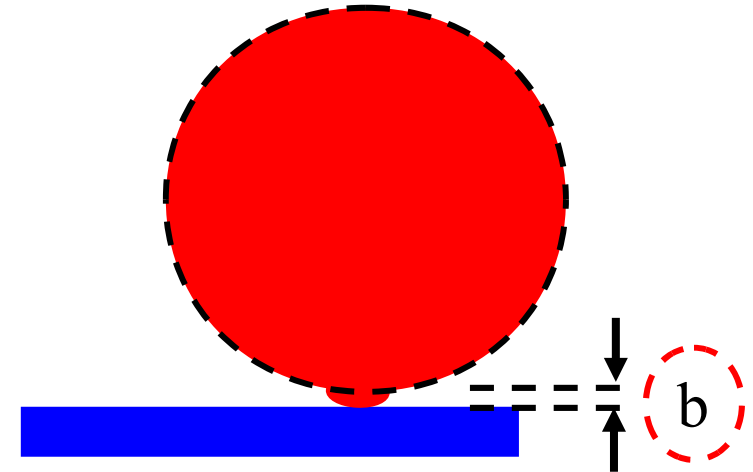
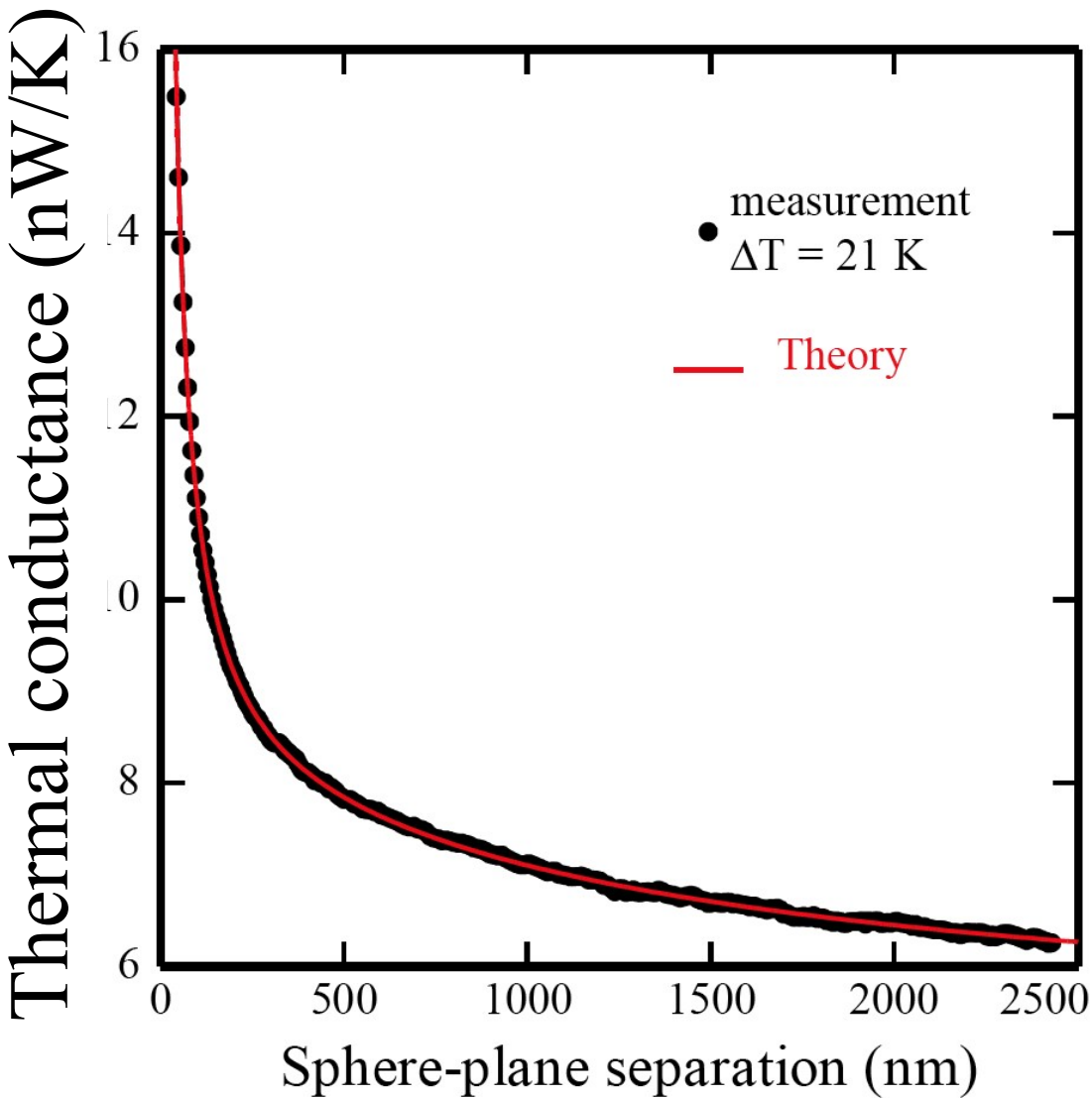
Conversion factor

Asperity size

Fitting  
parameters

From the Derjaguin approximation

# Comparison experiments-theory



$$b = 31.8 \pm 0.2 \text{ nm}$$

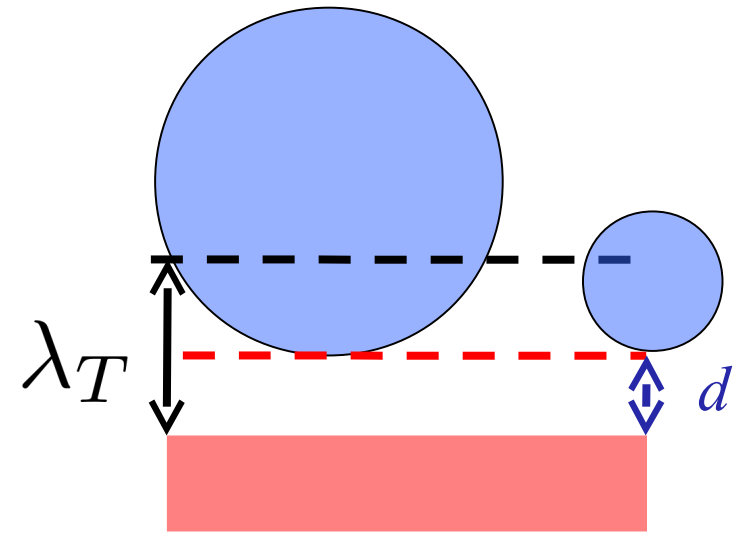
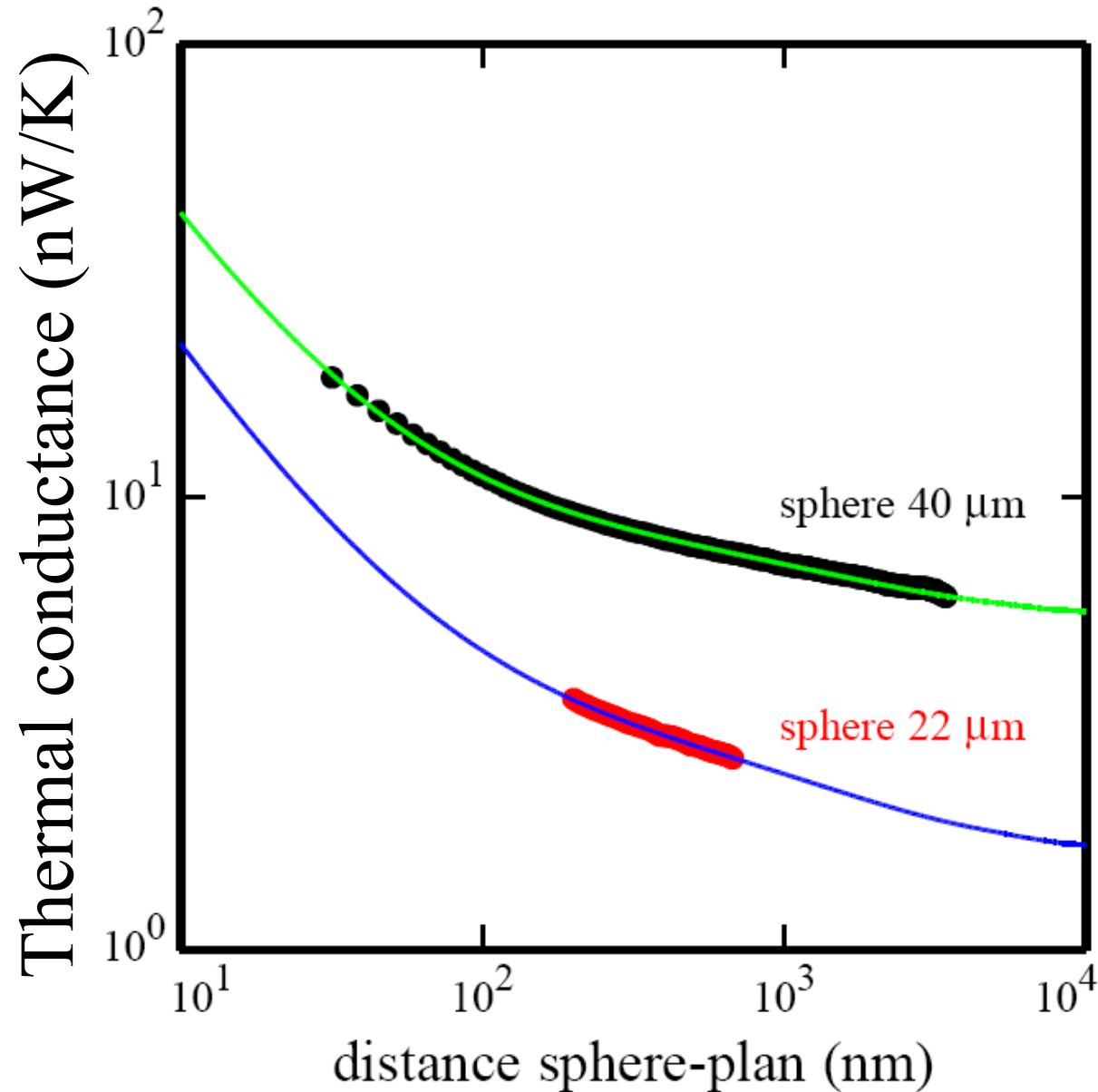
$$H = 2.162 \pm 0.005 \text{ nW/nm}$$

From calibration

$$H = 2.30 \pm 0.05 \text{ nW/nm}$$

From Rousseau et al. Nature Photonics **3** 514 (2009)

# Sphere with smaller radius

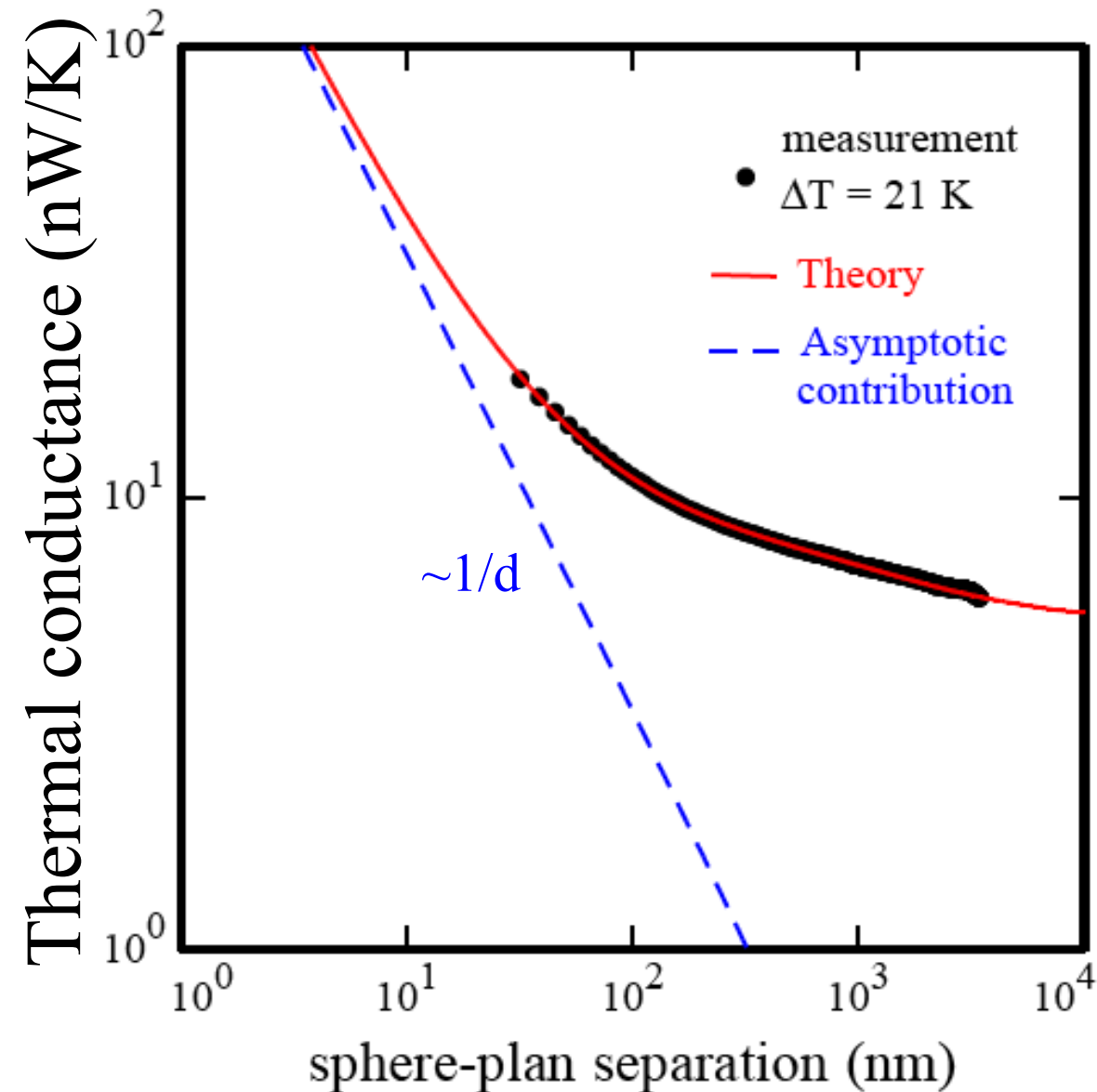


Unfortunately

$b \sim 150 \text{ nm}$

From Rousseau et al.  
Nature Photonics 3 514 (2009)

# Surface mode contribution ?



Increased by a factor 3  
*several order plane/plane*

SPP distance dependance  
*1/d<sup>2</sup> @ 200 nm*

*For a sphere, the asymptotic regime is reached below 10 nm*

*See Joël Chevrier talk*