$$\alpha = \frac{k}{\rho C_p}$$

$$E = \sqrt{k \rho C_p}$$

Thermal diffusivity and effusivity of thin layers based on thermoreflectance with femtosecond laser pulse

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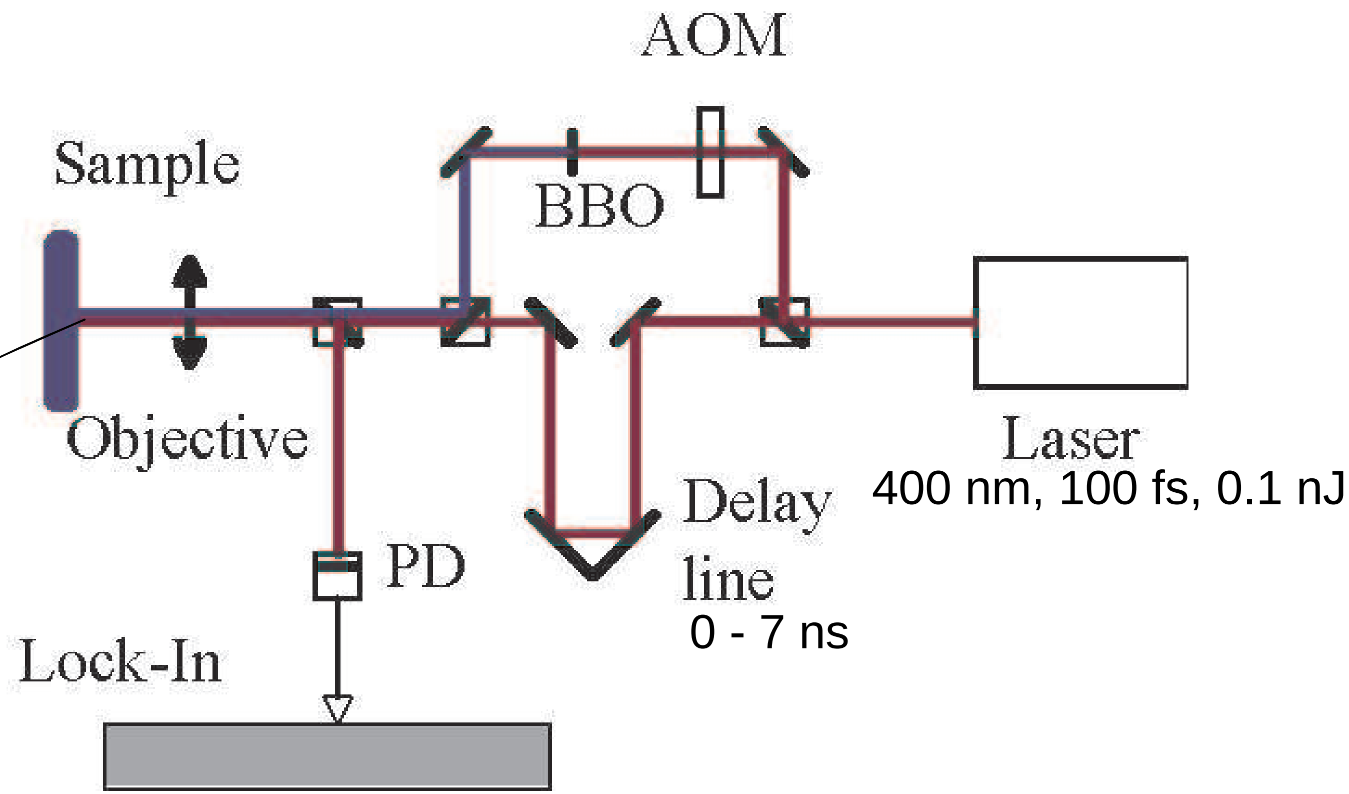
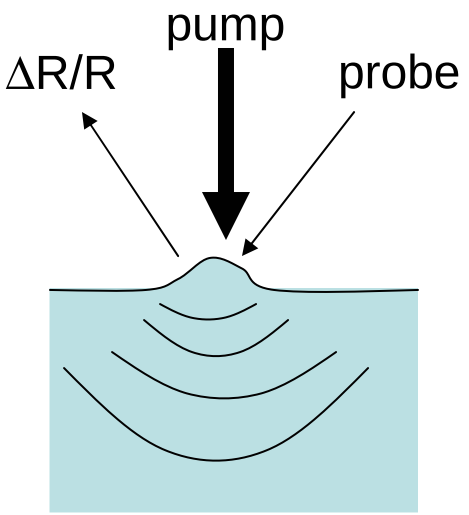
TDTR experimental setup

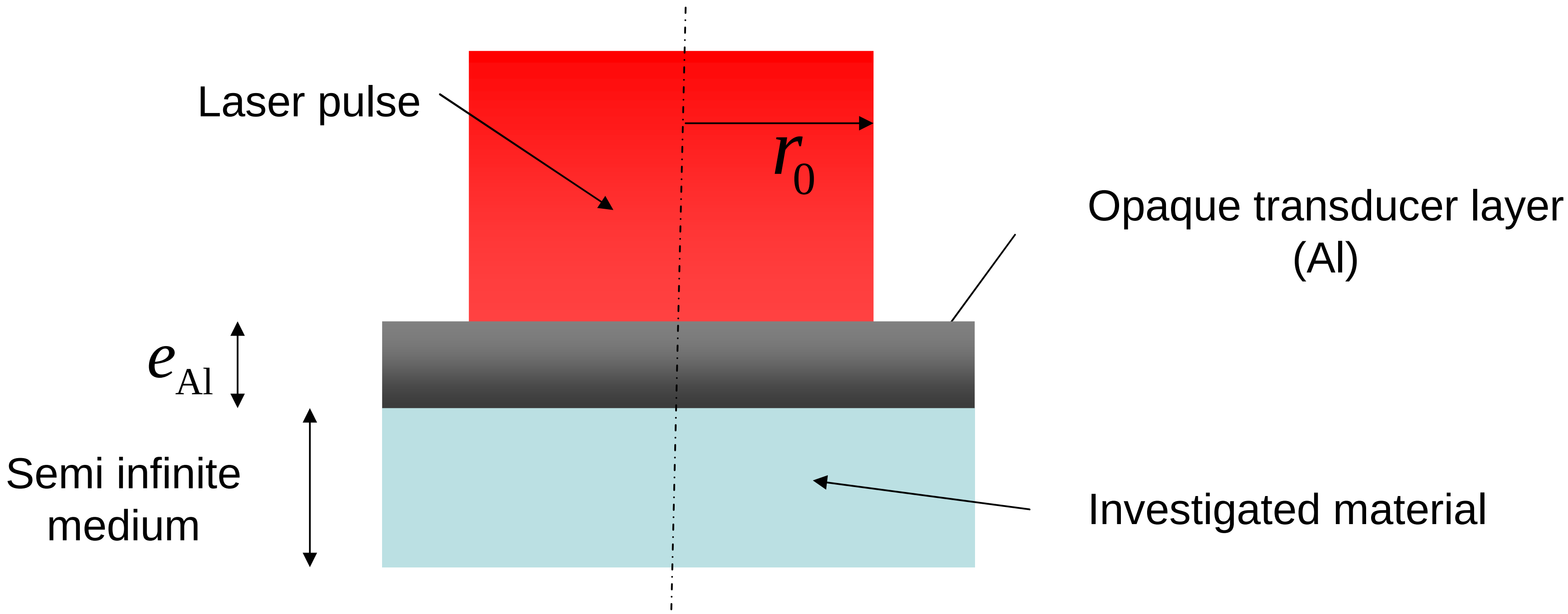
Rossignol *et al*., Phys. Rev.B, 2002





Goal: measuring the thermal properties of thin films using the TDTR experiment <u>without the need of measuring the absolute temperature rise.</u>

Experimental configuration



The material is capped with a metal film

Laser pulse

r_0

Opaque transducer layer (Al)

e_{Al}

Semi infinite medium

Investigated material

$$\overline{\text{TDTR}} = \frac{\Delta R}{R} = a_e \Delta T_e + a_l \, \Delta T_l$$

Small times
< psec

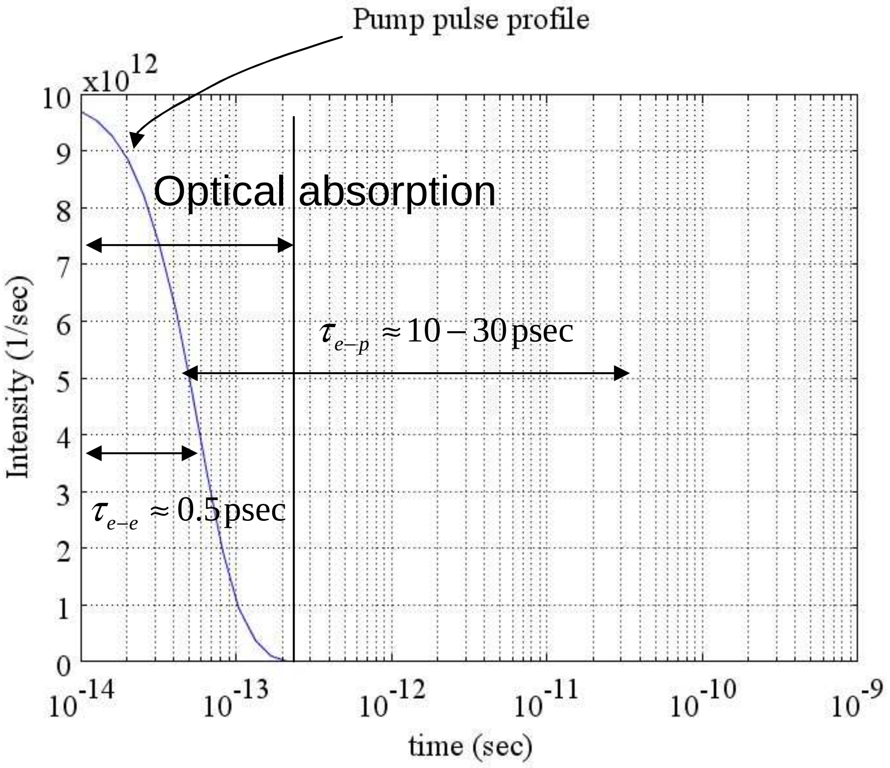
Long times
>10 psec

Physical phenomena

- The optical absorption depth of the laser depends on the laser wavelength and on the extinction coefficient of the medium (imaginary part of the refractive index)

- In case of a dielectric material, heat diffuses from the lattice vibrations (phonon)

- In case of a metal or a semiconductor, electron gas temperature increases rapidly, leaving the lattice temperature unchanged (electron-electron collisions). The lattice temperature increases until it reached the electronic temperature: this is the thermallization process (electron-phonon collisions). Finally heat diffuses inside the medium.



$$I(t) = I_0 \frac{1}{\tau_l \cosh\left(1.76\, t/\tau_l\right)}$$

Thermal effusivity of the material

Capinski et al., Phys. Rev B **59**, 8105 (1999)

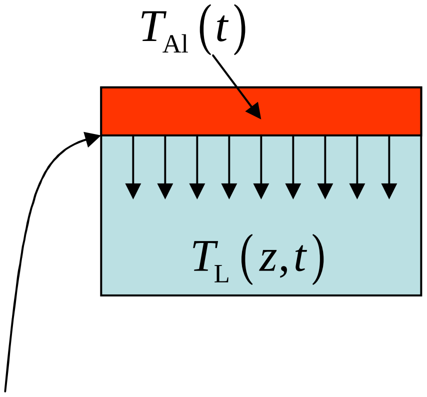
Capped layer configuration, the aluminium film is considered as thermally thin

1D heat diffusion problem
$r_h >> \alpha t_\mathrm{d}$

$$C_L \frac{\partial T_L(t,z)}{\partial t} = k \frac{\partial^2 T_L(t,z)}{\partial z^2}, \quad z > 0,\ t > 0$$

No heat loss, the rate of
energy diffuses in the layer

$$C_\mathrm{Al}\, e_\mathrm{Al} \frac{\partial T_\mathrm{Al}(t)}{\partial t} = k \frac{\partial T_L(t,z)}{\partial z}, \quad z = 0,\ t > 0$$



No thermal (Kapitza) resistance at the Al-Layer interface

$$\boxed{\overline{\mathrm{TDTR}} = \frac{T_\mathrm{Al}(t)}{T_\mathrm{Al}(0)} = e^{\alpha^2 t}\,\mathrm{erfc}\left(\alpha\sqrt{t}\right)}$$

$$\alpha = \frac{\sqrt{k\, C_L}}{e_\mathrm{Al}\, C_\mathrm{Al}} = \frac{E}{e_\mathrm{Al}\, C_\mathrm{Al}}$$

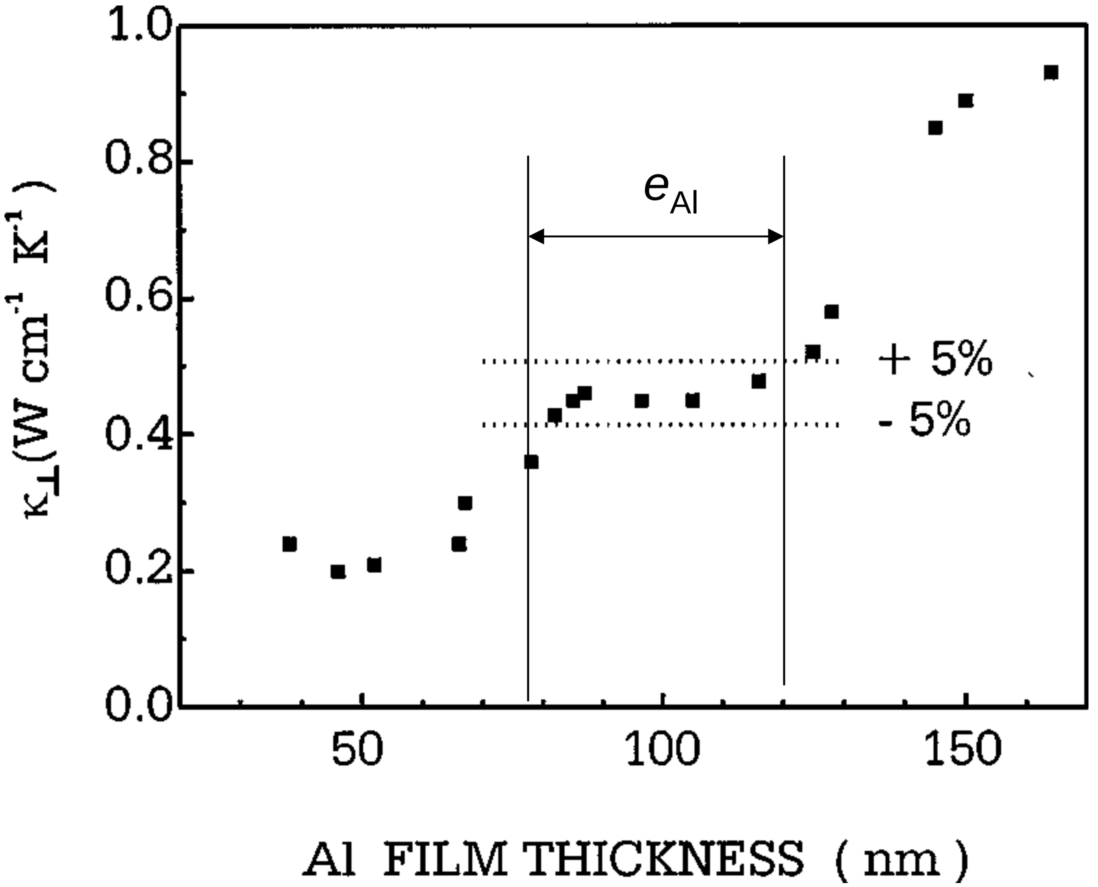
Identifying α on the TDTR response lead to E

$$\boxed{E = \hat{\alpha}\, e_\mathrm{Al}\, C_\mathrm{Al}}$$

$$e_\mathrm{Al}\ ?$$

Experimental calibration for e_{Al}

Capinski et al., Phys. Rev B **59**, 8105 (1999)



Two-temperatures model

Anisimov *et al*., 1974.

$$\boxed{\begin{gathered} C_e\left(T_e\right)\frac{\partial T_e}{\partial t} = \nabla_{r,z}\cdot\left[k_e\left(T_e,T_l\right)\overrightarrow{\nabla_{r,z}}\,T_e\right] - g\left(T_e - T_l\right) + S \\ C_l\,\frac{\partial T_l}{\partial t} = g\left(T_e - T_l\right) \end{gathered}}$$

Heat source term $S = \frac{T_\lambda}{\beta_o \pi r_h^{\,2}} e^{-z/\beta_o}\, e^{-\left(r/r_h\right)^2}\, I_0\, f\left(t\right)$

Time profile for the laser pulse $f\left(t\right) = \frac{1}{\tau_l \cosh\left(1.76t/\tau_l\right)^2}$

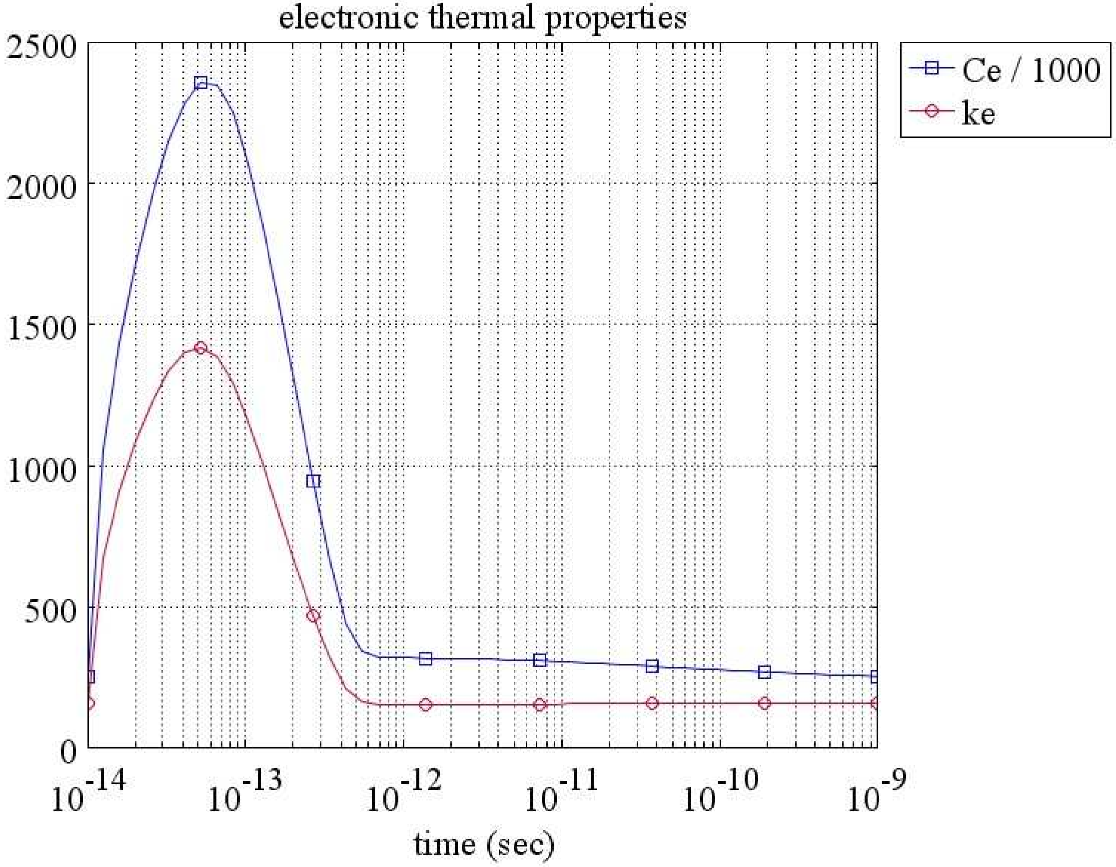
Optical absorption depth in the film $\beta_o = \frac{4\pi\kappa_\lambda}{\lambda}$

g is the electron-phonon coupling factor

Electronic thermal diffusivity

$$\begin{tikzcd} k_e = k_0 \frac{T_e}{T_l} \arrow[dr] & \\ & \alpha_e = \frac{k_e}{C_e} = \frac{k_0}{\gamma\, T_l} \\ C_e = \gamma\, T_e \arrow[ur] & \end{tikzcd}$$

But… the electronic <u>thermal diffusivity</u> depends on the lattice temperature



Electron-Phonon coupling factor g [W m^{-3} K^{-1}]

Allen, Phys. Rev. Lett. **59**, 1460 (1987)

$$g \approx \frac{3\hbar}{\pi k_B} \lambda \left\langle \omega^2 \right\rangle$$

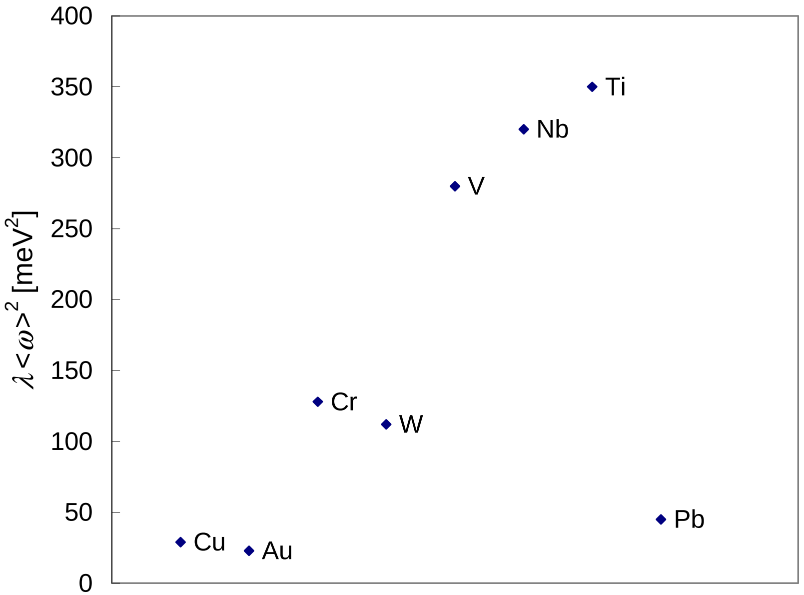
λ: coupling constant
$<\omega^2>$: second moment of the phonon spectrum

Brorson et al. , Phys. Rev. Lett. **64**, 2172 (1990)

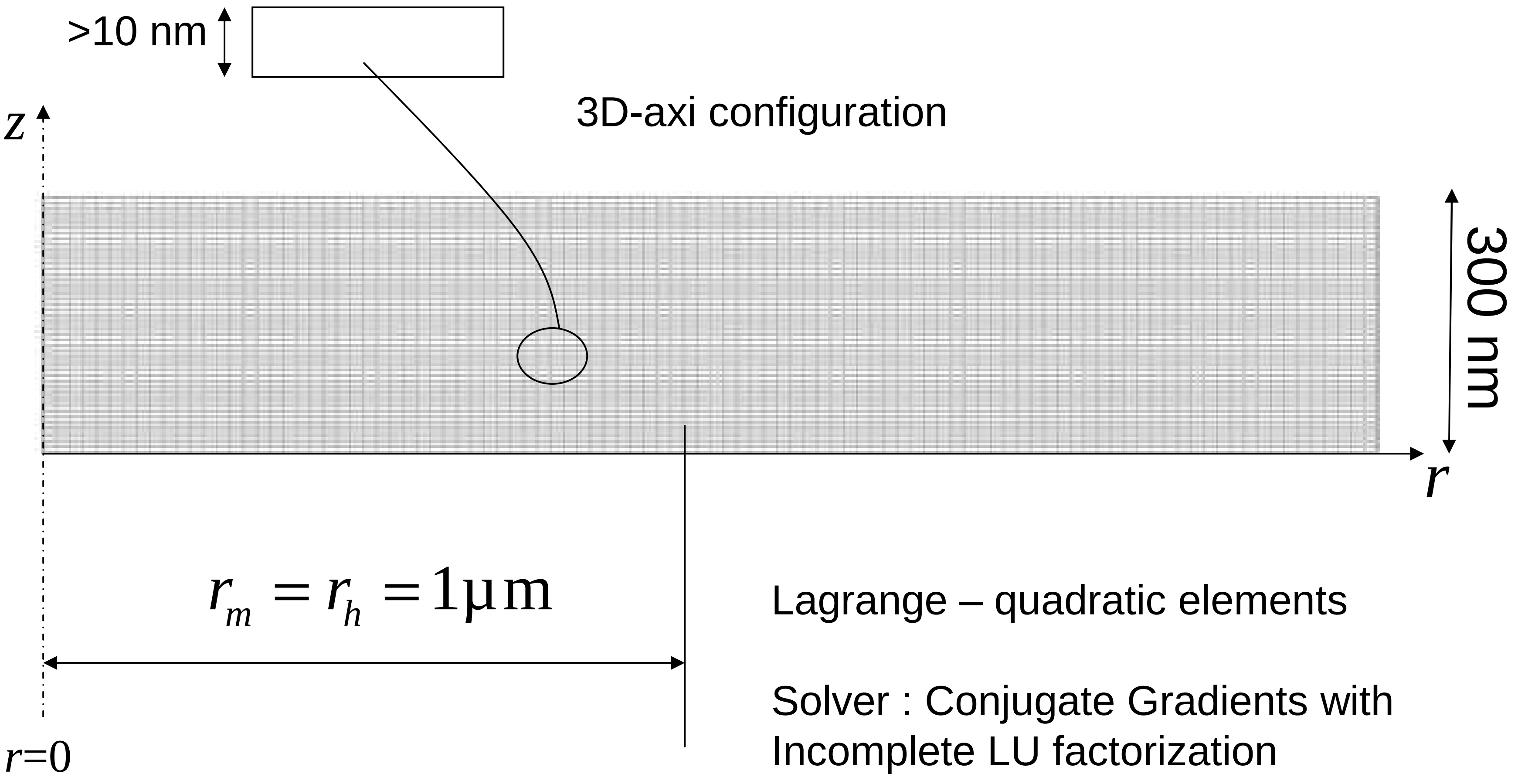
$$g \approx \frac{\pi^2 m n v_s{}^2}{6}$$

m: electron mass
n: electron density
V_s: sound velocity

The coupling factor depend on the microstructure (grain size, defects,…)



FEM simulation - configuration

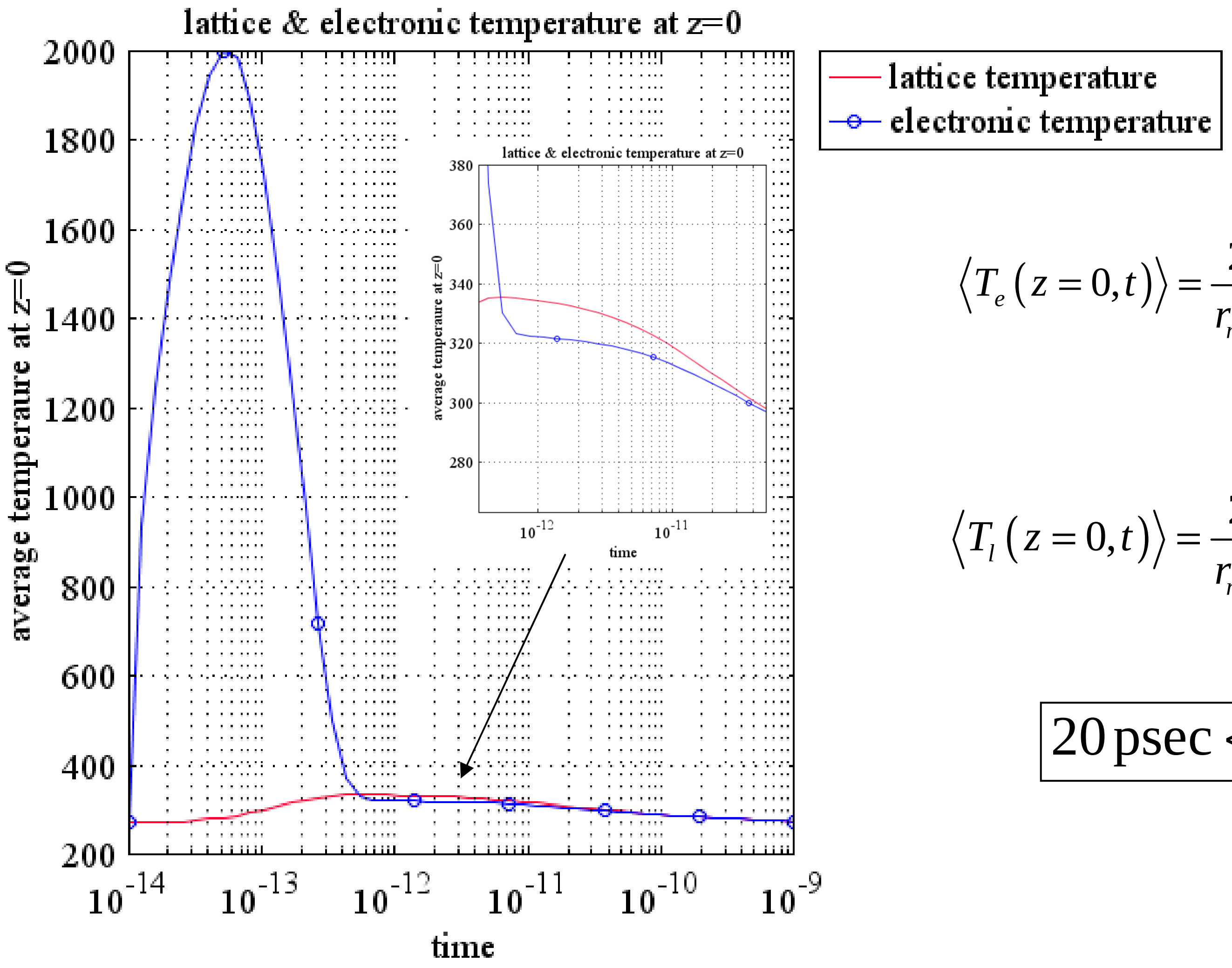


3D-axi configuration

Lagrange – quadratic elements

Solver : Conjugate Gradients with Incomplete LU factorization preconditioner

Simulated average electronic and lattice temperature

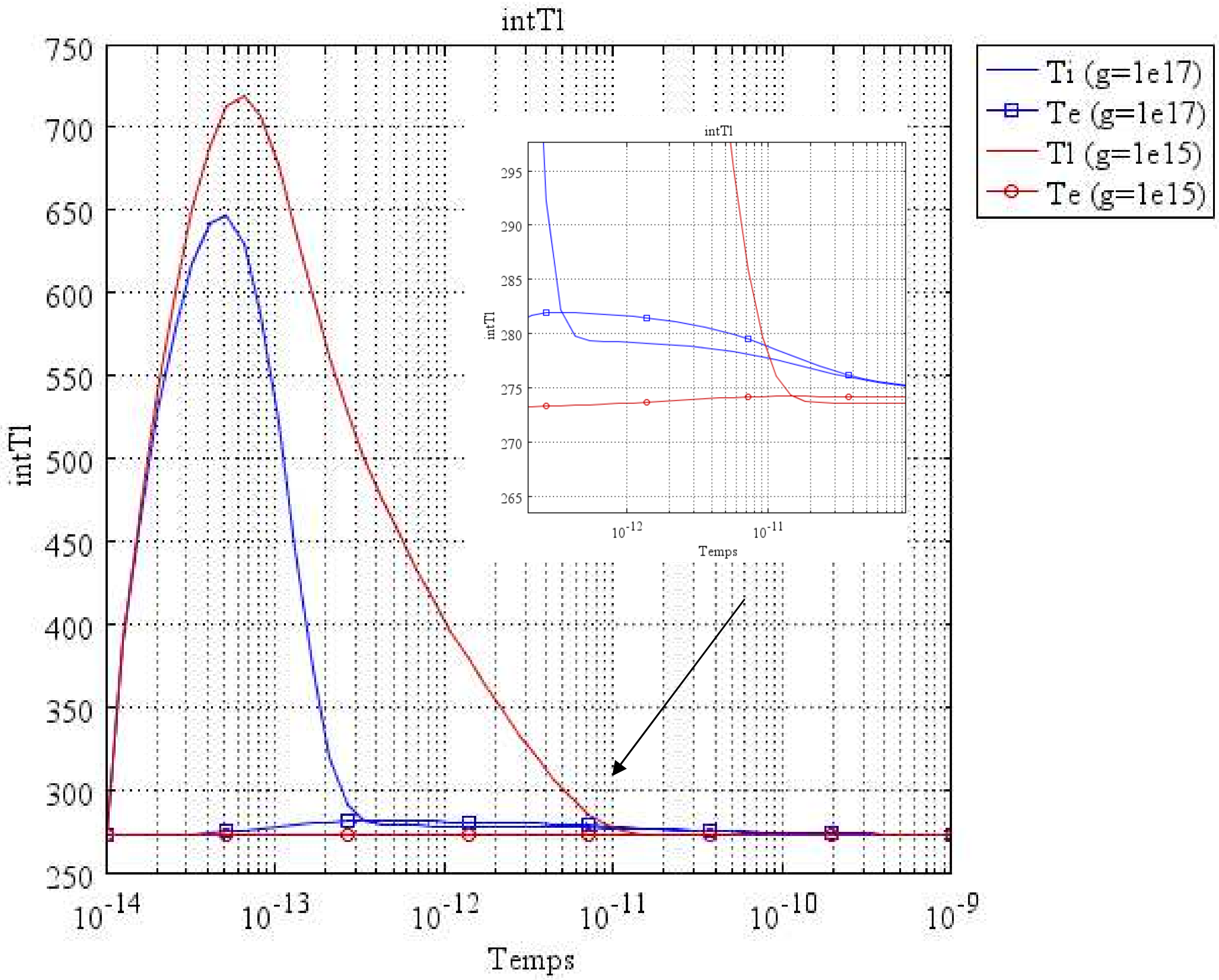


$$\left\langle T_e\left(z=0,t\right)\right\rangle = \frac{2}{{r_m}^2}\int_0^{r_m} r\,T_e\left(r,z=0,t\right)\mathrm{d}r$$

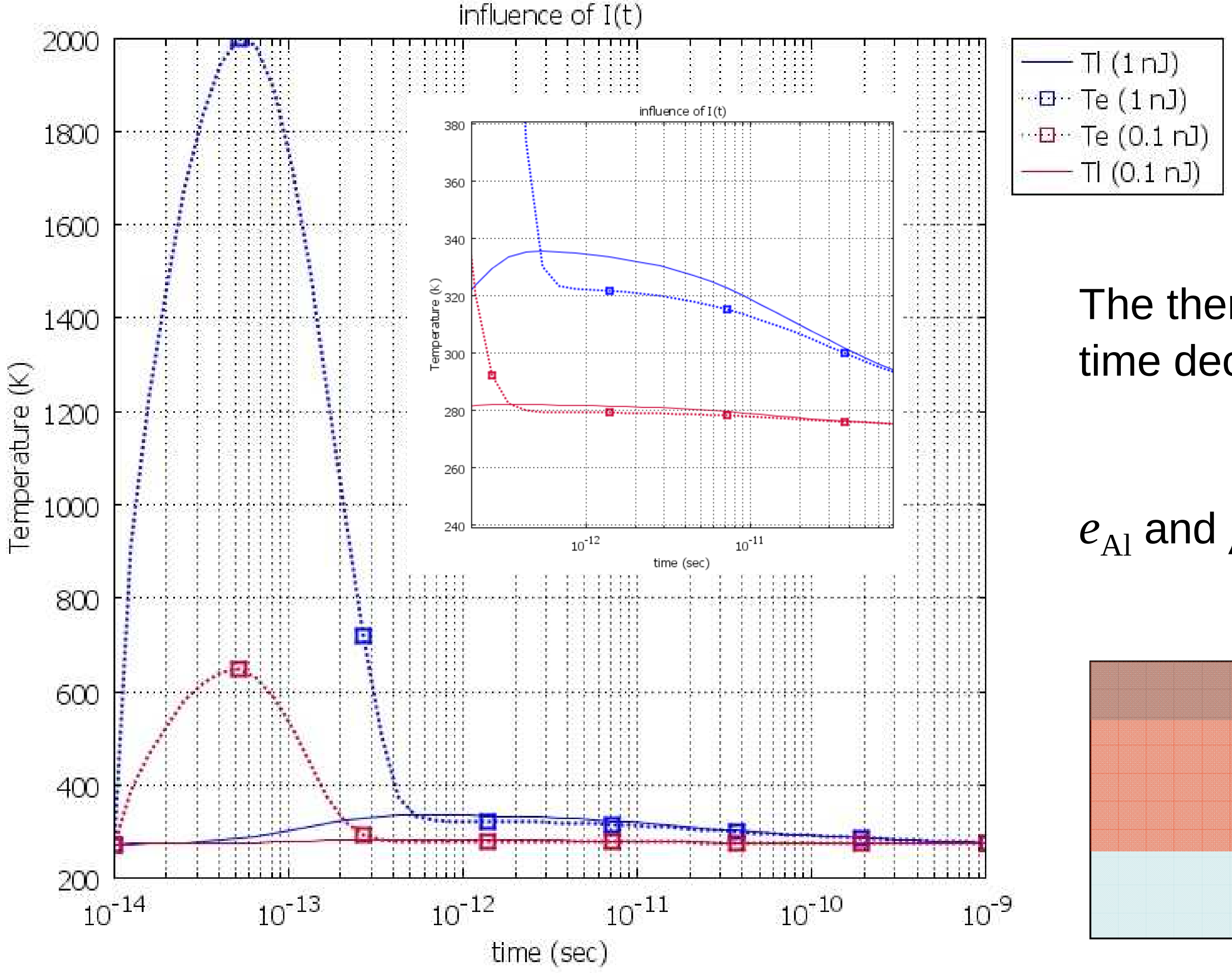
$$\left\langle T_l\left(z=0,t\right)\right\rangle = \frac{2}{{r_m}^2}\int_0^{r_m} r\,T_l\left(r,z=0,t\right)\mathrm{d}r$$

$$\boxed{20\,\mathrm{psec} < \tau < 40\,\mathrm{psec}}$$

Influence of *g*



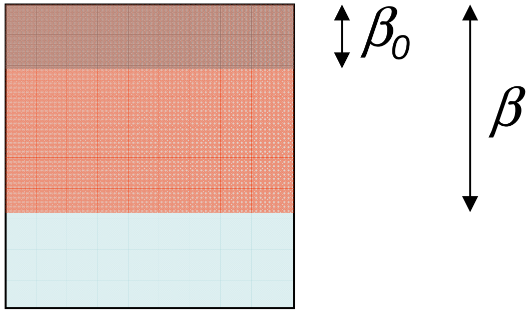
Influence of $S(t)$



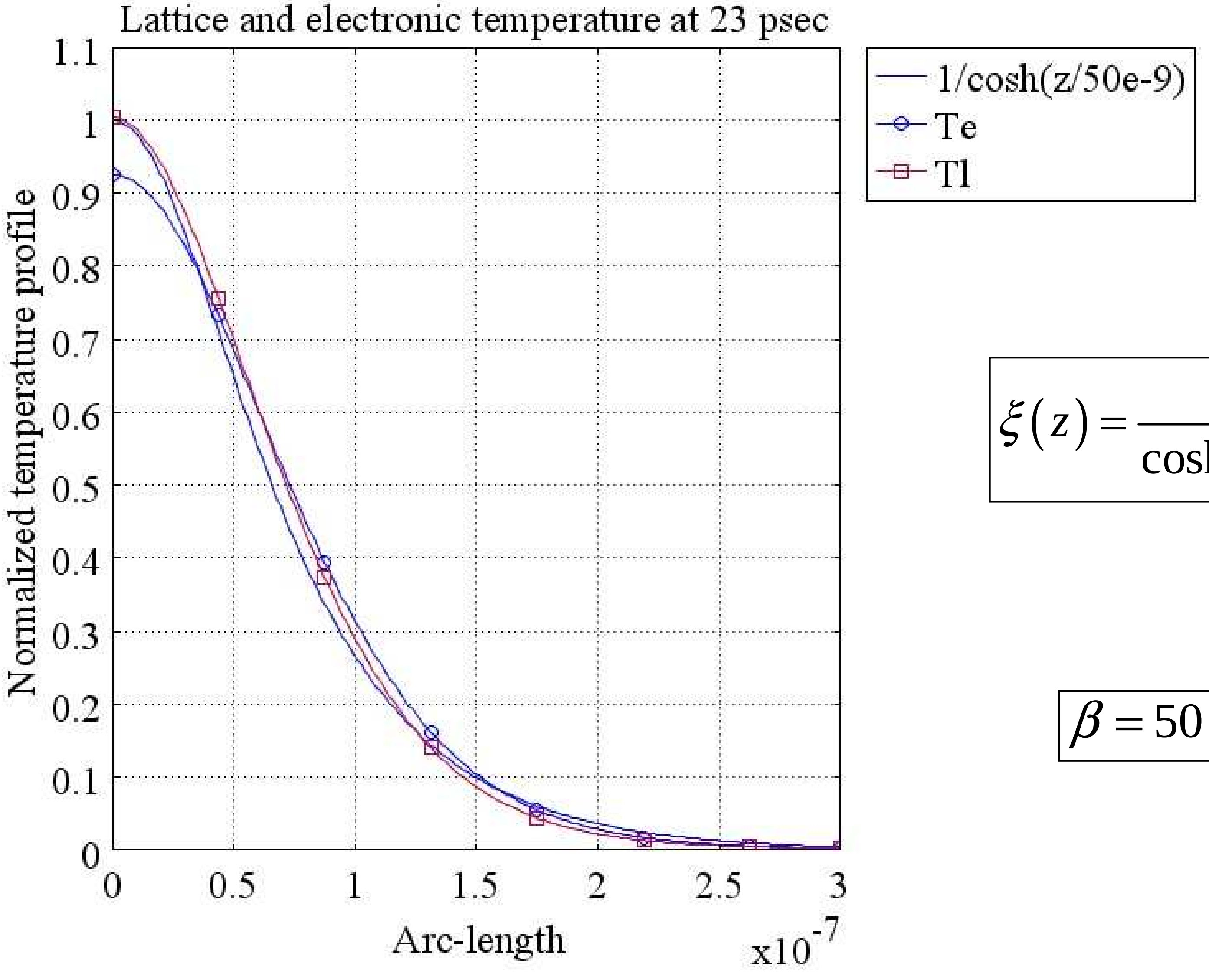
The thermallization time decreases with I_0

↓

e_{Al} and β depend on I_0



$T_e(z)$ and $T_l(z)$ at the end of thermallization



$$\boxed{\xi(z) = \frac{1}{\cosh(z/\beta)}}$$

$$\boxed{\beta = 50\,\mathrm{nm}}$$

1 T heat transfer model

At the end of e-p thermallization

$$T_e = T_l = T$$

Heat diffusion with non uniform initial temperature is mathematically equivalent to:

$$C_l \frac{\partial T}{\partial t} = \nabla_{r,z} \cdot \left[k_0 \overrightarrow{\nabla_{r,z}} T \right] + S$$

Heat source term

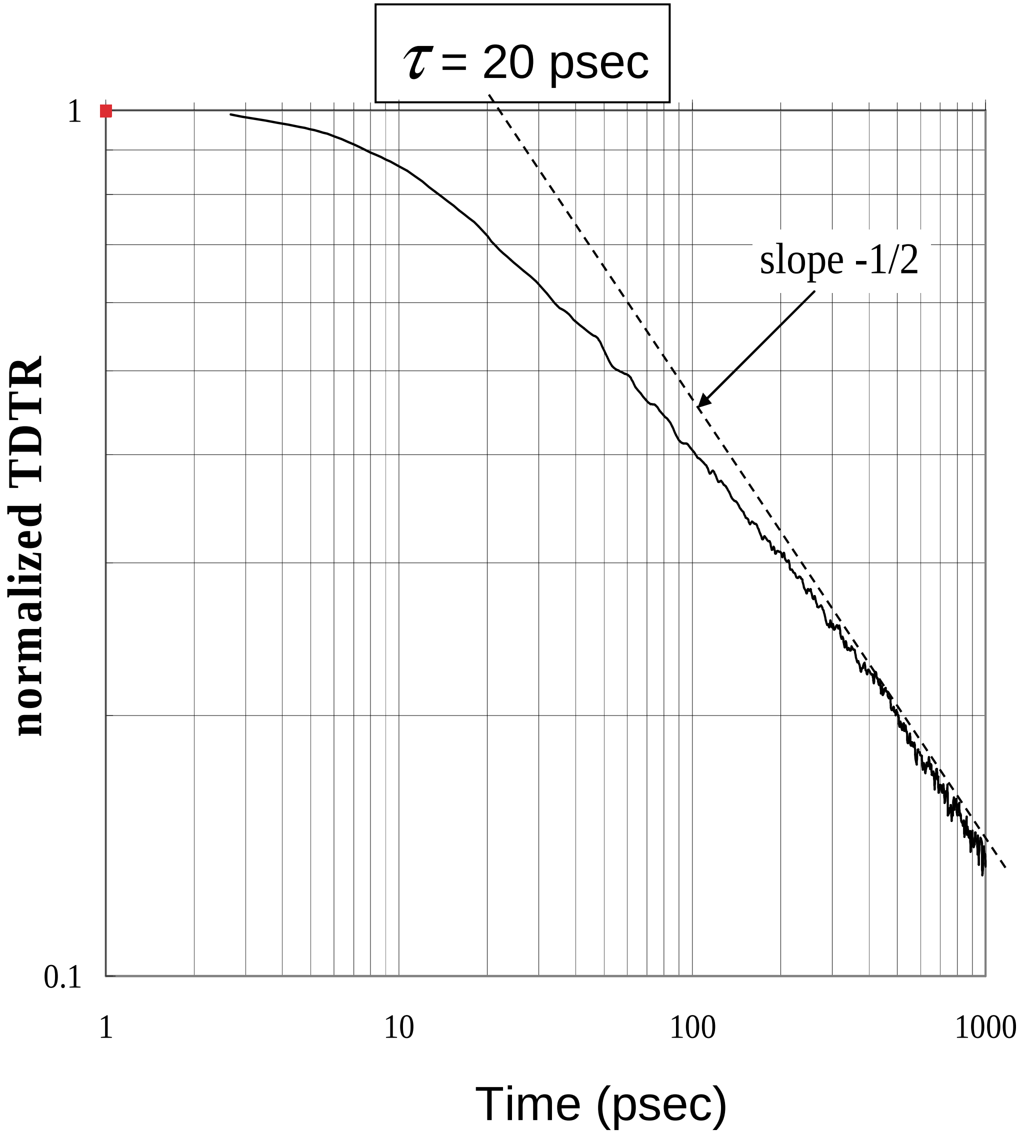
$$\boxed{S = \varepsilon \xi(z) g(r) \delta(t)}$$

$$\xi(z) = \frac{1}{\cosh(z/\beta)}$$

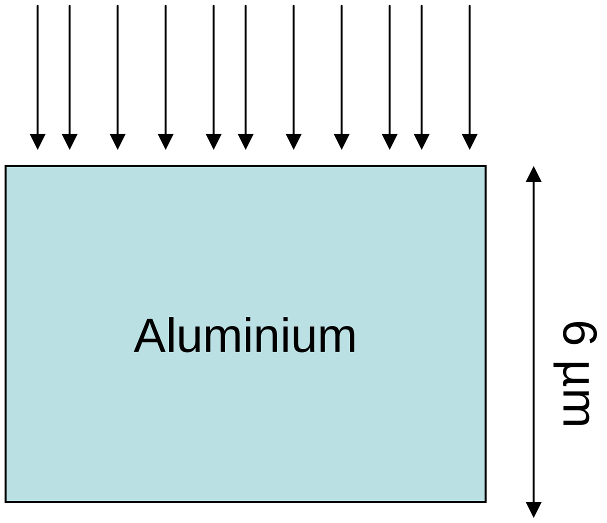
$$g(r) = e^{-(r/r_h)^2}$$

Application on aluminium layer

1- Experimental result



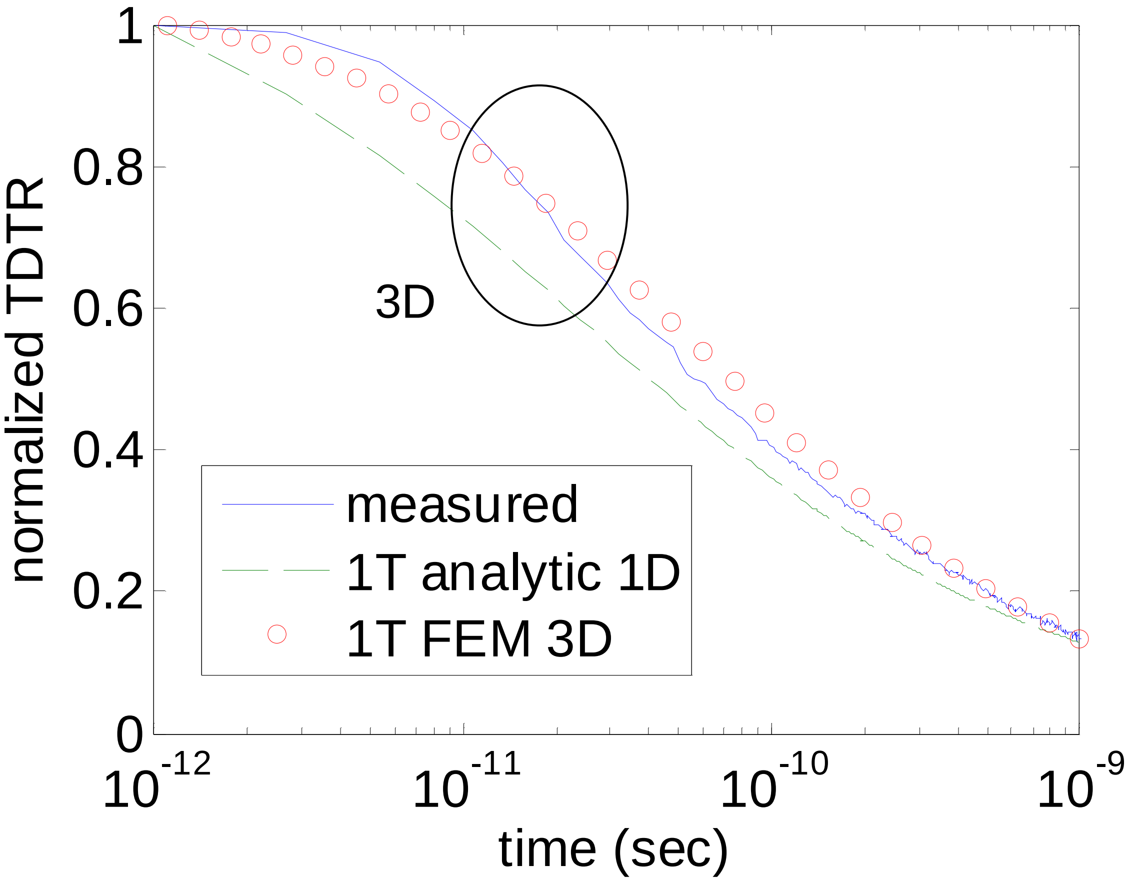
Aluminium layer 6 µm

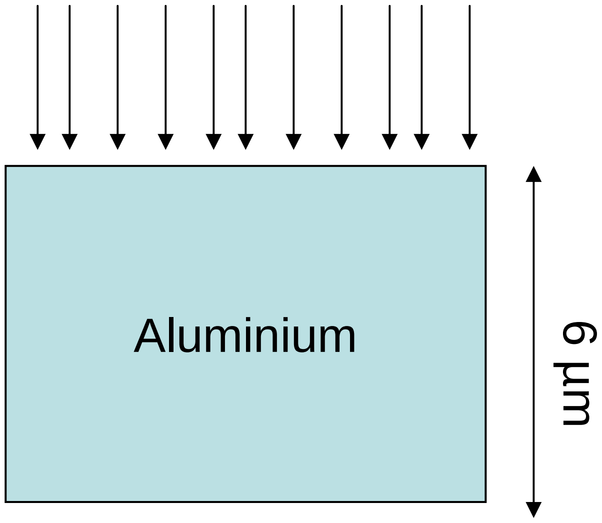


$$\mathrm{TDTR}_{t_\infty} \propto \frac{C}{\sqrt{t}}$$

Application on aluminium layer

2- comparison with the model

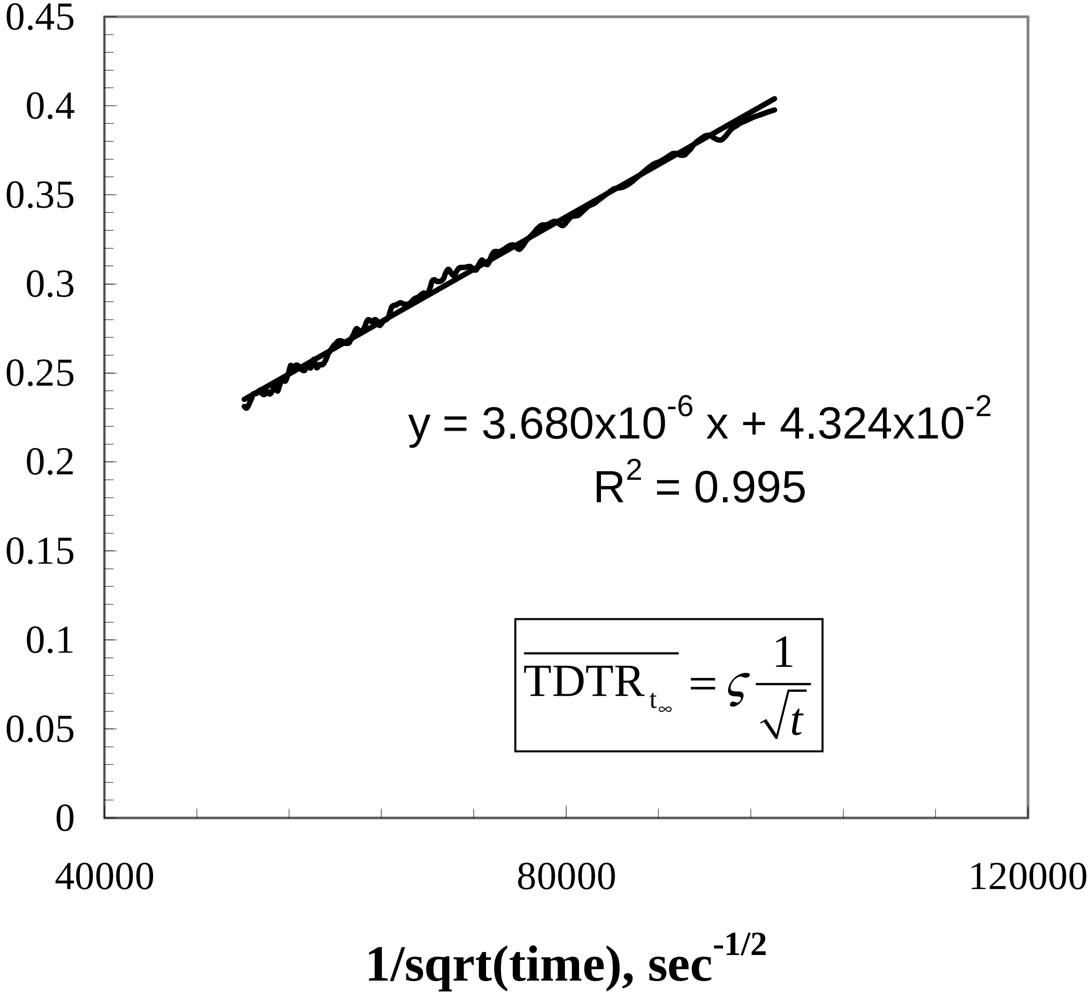




β=50 nm

Thermal diffusivity of the material

J.-L. Battaglia *et al.*, Phys. Rev. B, 76, 184110 (2007)



$$\overline{\mathrm{TDTR}_{t_\infty}} = \varsigma \frac{1}{\sqrt{t}}$$

$$\tilde{\alpha} = \frac{\beta^2}{\pi \varsigma^2}$$

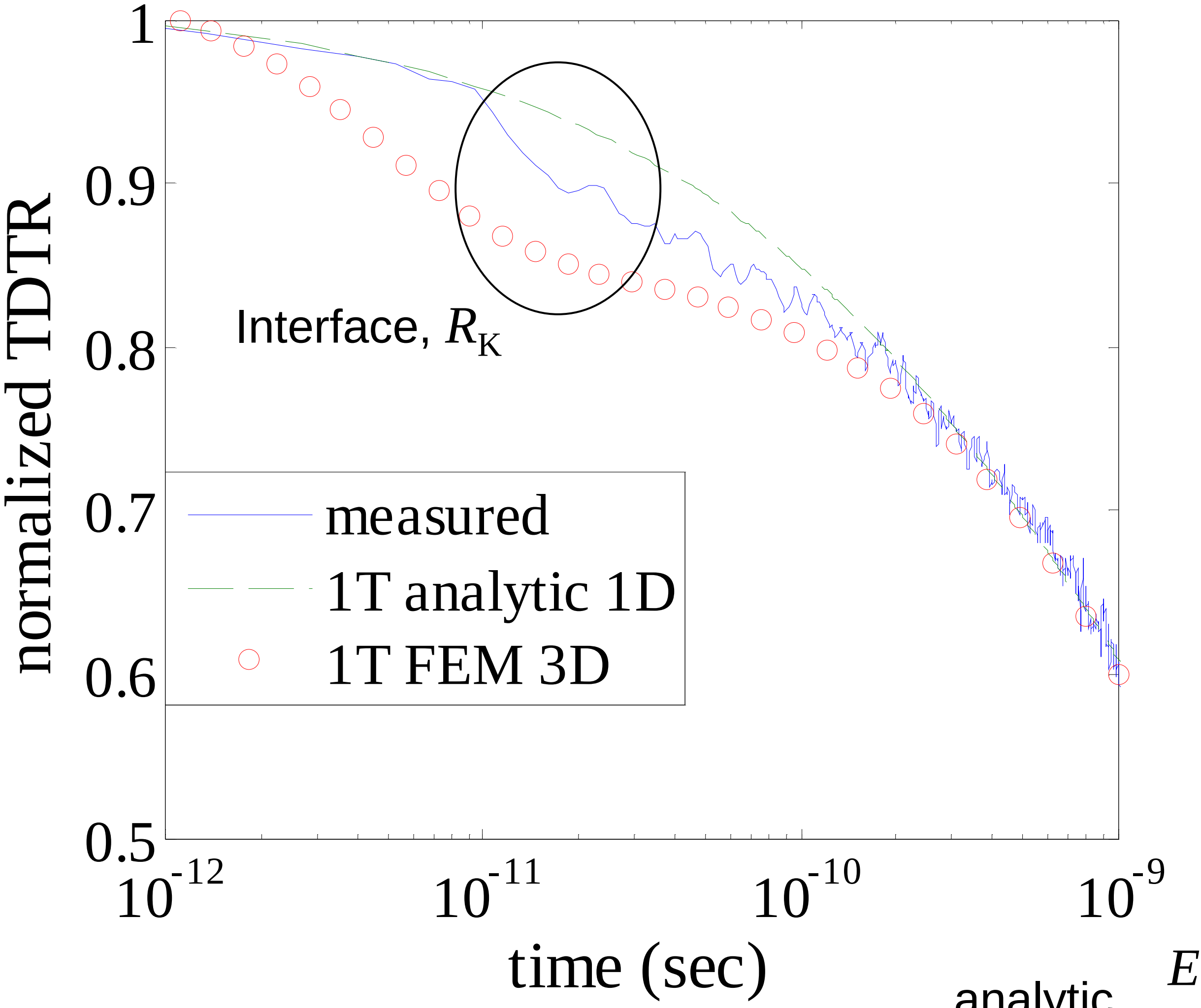
$$\beta = 50 \times 10^{-9}\ \mathrm{m}$$

$$\tilde{\alpha} = 5.87 \times 10^{-5}\ \mathrm{m}^2\,\mathrm{sec}^{-1}$$

$$\alpha = 6.58 \times 10^{-5}\ \mathrm{m}^2\,\mathrm{sec}^{-1}$$

$$\frac{\Delta\alpha}{\alpha} = 10\%$$

Application on Al-Si_3N_4



Al layer is deposited on a Si3N4 layer (200 nm)

e_{Al}=50 nm

k_{SiN}=1.5 W m^{-1} K^{-1}

ρ_{SiN}= 2700 kg m^{-3}

C_{pSiN}=300 J kg^{-1} K^{-1}

R_K=3x10^{-8} K m^2 W^{-1}

analytic

E_{est}=2.033x10^3 W $s^{-1/2}$ m^{-2} K^{-1}
E_{exp}=1.122x10^3 W $s^{-1/2}$ m^{-2} K^{-1}

Conclusions
Theoretical part

- The <u>thermal diffusivity</u> ($k / \rho C_p$) and <u>effusivity</u> (sqrt($k \rho C_p$)) of a thin layer can be measured from the picoseconds thermoreflectance technique.

- It can be done without measuring the absolute temperature in case of the <u>one temperature model</u>, when thermallization between electron an phonon is reached.

- Using the <u>two temperature model</u> it is shown that the thermallization time extensively depends on the heat source magnitude.

- The thermal diffusivity is estimated from the non capped layer configuration.

$$\boxed{\tilde{\alpha} = \frac{\beta^2}{\pi \varsigma^2}}$$

- The thermal effusivity is estimated with the capped layer (by Al) configuration.

$$\boxed{E = \hat{\alpha} e_{\mathrm{Al}} C_{\mathrm{Al}}}$$

Conclusions experimental part

- In the capped layer configuration, the Al thickness is calculated in order to be considered as thermally thin.
The thickness is then derived from the two temperature model simulation according to the heat source (pump) magnitude.
<u>Kapitza's resistance</u> at the interface must be taken into account.

- In the non capped layer configuration, the heat penetration depth at the end of the thermallization process is not known (in case of a metal or a semiconductor).
The thermallization time can be deduced from the measurement by representing the long time behaviour, **slope=-1/2** in the log-log representation. It crosses the x axis at τ.
How relating the measure of τ with β?....