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

\[ \alpha = \frac{k}{\rho C_p} \]

\[ E = \sqrt{k \rho C_p} \]

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TDTR experimental setup

**Goal:** measuring the thermal properties of thin films using the TDTR experiment without the need of measuring the absolute temperature rise.

Diagram:
- Laser: 400 nm, 100 fs, 0.1 nJ
- Sample
- Objective
- BBO
- Delay line 0 - 7 ns
- PD
- Lock-In
- ΔR/R
- pump probe
Experimental configuration

The material is capped with a metal film

Laser pulse

Opaque transducer layer (Al)

Investigated material

$e_{\text{Al}}$

Semi infinite medium

$\Delta R = \Delta T_e + a_l \Delta T_l$

Small times $< \text{psec}$

Long times $>10 \text{ psec}$
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 thermalization process (electron-phonon collisions). Finally heat diffuses inside the medium.

\[
I(t) = I_0 \frac{1}{\tau_i \cosh(1.76t/\tau_i)}
\]
Thermal effusivity of the material


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

1D heat diffusion problem

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

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

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

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

\[
\text{TDTR} = \frac{T_{Al}(t)}{T_{Al}(0)} = e^{\alpha^2 t} \text{erfc}(\alpha \sqrt{t})
\]

\[
\alpha = \sqrt{\frac{kC_L}{e_{Al} C_{Al}}} = \frac{E}{e_{Al} C_{Al}}
\]

Identifying \( \alpha \) on the TDTR response lead to \( E \)

\[ E = \hat{\alpha} e_{Al} C_{Al} \]

\[ e_{Al} \]
Experimental calibration for $e_{\text{Al}}$

Two-temperatures model
Anisimov et al., 1974.

\[
C_e (T_e) \frac{\partial T_e}{\partial t} = \nabla_{r,z} \cdot \left[ k_e (T_e, T_l) \nabla_{r,z} T_e \right] - g (T_e - T_l) + S
\]

\[
C_l \frac{\partial T_l}{\partial t} = g (T_e - T_l)
\]

Heat source term

\[S = \frac{T_\lambda}{\beta_o \pi r_h^2} e^{-z/\beta_o} e^{-(r/r_h)^2} I_0 f (t)\]

Time profile for the laser pulse

\[f (t) = \frac{1}{\tau_l \cosh (1.76 t/\tau_l)^2}\]

Optical absorption depth in the film

\[\beta_o = \frac{4 \pi \kappa_\lambda}{\lambda}\]

\(g\) is the electron-phonon coupling factor
Electronic thermal diffusivity

\[ k_e = k_0 \frac{T_e}{T_l} \]

\[ C_e = \gamma T_e \]

\[ \alpha_e = \frac{k_e}{C_e} = \frac{k_0}{\gamma T_l} \]

But... the electronic thermal diffusivity depends on the lattice temperature
Electron-Phonon coupling factor
\[ g \ [W \ m^{-3} K^{-1}] \]


\[ g \approx \frac{3\hbar}{\pi k_B} \lambda \langle \omega^2 \rangle \]
\[ \lambda: \text{coupling constant} \]
\[ \langle \omega^2 \rangle: \text{second moment of the phonon spectrum} \]


\[ g \approx \frac{\pi^2 m n v_s^2}{6} \]
\[ m: \text{electron mass} \]
\[ n: \text{electron density} \]
\[ v_s: \text{sound velocity} \]

The coupling factor depend on the microstructure (grain size, defects,...)
FEM simulation - configuration

3D-axi configuration

$r_m = r_h = 1\mu m$

Lagrange – quadratic elements

Solver: Conjugate Gradients with Incomplete LU factorization preconditioner
Simulated average electronic and lattice temperature

\[ \langle T_e (z = 0, t) \rangle = \frac{2}{r_m^2} \int_0^{r_m} r T_e (r, z = 0, t) \, dr \]

\[ \langle T_l (z = 0, t) \rangle = \frac{2}{r_m^2} \int_0^{r_m} r T_l (r, z = 0, t) \, dr \]

20 psec < \tau < 40 psec
Influence of $g$
Influence of $S(t)$

The thermalization time decreases with $I_0$. $e_{Al}$ and $\beta$ depend on $I_0$. 

$\beta_0$
$T_e(z)$ and $T_l(z)$ at the end of thermalization

\[ \beta = 50 \text{ nm} \]

\[ \xi(z) = \frac{1}{\cosh(z/\beta)} \]
1 T heat transfer model

At the end of e-p thermalization

\[ T_e = T_l = T \]

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

\[ C_i \frac{\partial T}{\partial t} = \nabla \cdot \left[ k_0 \nabla T \right] + S \]

Heat source term

\[ S = \varepsilon \xi(z) g(r) \delta(t) \]

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

\[ g(r) = e^{-(r/r_h)^2} \]
Application on aluminium layer

1- Experimental result

\[ \tau = 20 \text{ psec} \]

Aluminium layer 6 µm

\[ \text{TDTR}_{t_\infty} \propto \frac{C}{\sqrt{t}} \]
Application on aluminium layer

2- comparison with the model

Aluminium

$\beta = 50 \text{ nm}$

normalized TDTR

measured

1T analytic 1D

1T FEM 3D

time (sec)

$\beta = 50 \text{ nm}$
Thermal diffusivity of the material


\[
\alpha = \frac{\beta^2}{\pi \zeta^2}
\]

\[
\beta = 50 \times 10^{-9} \text{ m}
\]

\[
\tilde{\alpha} = 5.87 \times 10^{-5} \text{ m}^2 \text{ sec}^{-1}
\]

\[
\alpha = 6.58 \times 10^{-5} \text{ m}^2 \text{ sec}^{-1}
\]

\[
\frac{\Delta \alpha}{\alpha} = 10\%
\]

\[
y = 3.680 \times 10^{-6} x + 4.324 \times 10^{-2}
\]

\[
R^2 = 0.995
\]

TDTR: \[ \tau_{\infty} = \zeta \frac{1}{\sqrt{t}} \]
Application on Al-Si$_3$N$_4$

Al layer is deposited on a Si$_3$N$_4$ layer (200 nm)

- $e_{Al} = 50$ nm
- $k_{SiN} = 1.5$ W m$^{-1}$ K$^{-1}$
- $\rho_{SiN} = 2700$ kg m$^{-3}$
- $C_{pSiN} = 300$ J kg$^{-1}$ K$^{-1}$
- $R_K = 3 \times 10^{-8}$ K m$^2$ W$^{-1}$

$E_{est} = 2.033 \times 10^3$ W s$^{-1/2}$ m$^{-2}$ K$^{-1}$
$E_{exp} = 1.122 \times 10^3$ W s$^{-1/2}$ m$^{-2}$ K$^{-1}$
Conclusions

Theoretical part

• The \textit{thermal diffusivity} \( \frac{k}{\rho C_p} \) and \textit{effusivity} \( \sqrt{\frac{k}{\rho C_p}} \) 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 \textit{one temperature model}, when thermalization between electron and phonon is reached.

• Using the \textit{two temperature model} it is shown that the thermalization time extensively depends on the heat source magnitude.

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

\[
\tilde{\alpha} = \frac{\beta^2}{\pi \zeta^2}
\]

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

\[
E = \tilde{\alpha} e_{\text{Al}} C_{\text{Al}}
\]
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. Kapitza’s resistance 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 $\tau$. How relating the measure of $\tau$ with $\beta$?....