BIPM Workshop
on Challenges in Metrology for Dynamic Measurement

Dynamic measurements of thermophysical properties for material metrology standards

Measurement by impulse heating and analysis by response functions and transfer functions

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BIPM Workshop on Challenges in Metrology for Dynamic Measurement

- Session 1 Dynamic Mechanical Quantities
  Force, Torque, Vibration, etc
- Session 2 Dynamic Fluid and Flowmetry
  Pressure, temperature, and volume of fluid
- Session 3 Thermo Physical Quantities
  Thermal properties, material properties, etc
  Temperature

Dynamic response of sensor, measurement

Frequency response, Step response, Impulse response

Mathematical presentation

Impulse response function
Transfer function

Outline of the presentation

1. Introduction
   - Needs for thermophysical quantities
   - Common interest with dynamic measurement of mechanical quantities, fluid and flowmetry
2. Pulsed light heating methods
   - Laser flash method for thermal diffusivity measurements
   - Ultra fast laser flash method for thin films measurement
3. Analysis by impulse response function and transfer function
   - Analysis of heat diffusion
   - Analysis of finite response time of temperature detection
   - Transfer function and areal heat diffusion time
4. Summary

Needs for thermal metrology 1

- Energy production:
  - Electric power generator
  - Nuclear power plant
  - Natural energy
    - Geothermal power generation
    - Solar power generation

Needs for thermal metrology 2

- Energy saving:
  - Industrial sector
    - Production line
    - Plants
    - Furnaces, kilns, heat treatment
      - Steel making, ceramics industry
  - Transportation sector
    - Automobiles
    - Trains, ships, aircrafts
  - Commercial and residential sector
    - Buildings and houses

Needs for thermal metrology 3

- Electronics:
  - Thermal design and management
    - PC, server, data center, projector
    - Home electric appliance
      - LED illumination
      - Flat panel display
  - Device
    - CPU
    - Memory and storage
      - Phase change memory
      - Hard disk (Heat assisted)
      - Organic EL

CMC service categories covered by WG9

6. Thermophysical properties

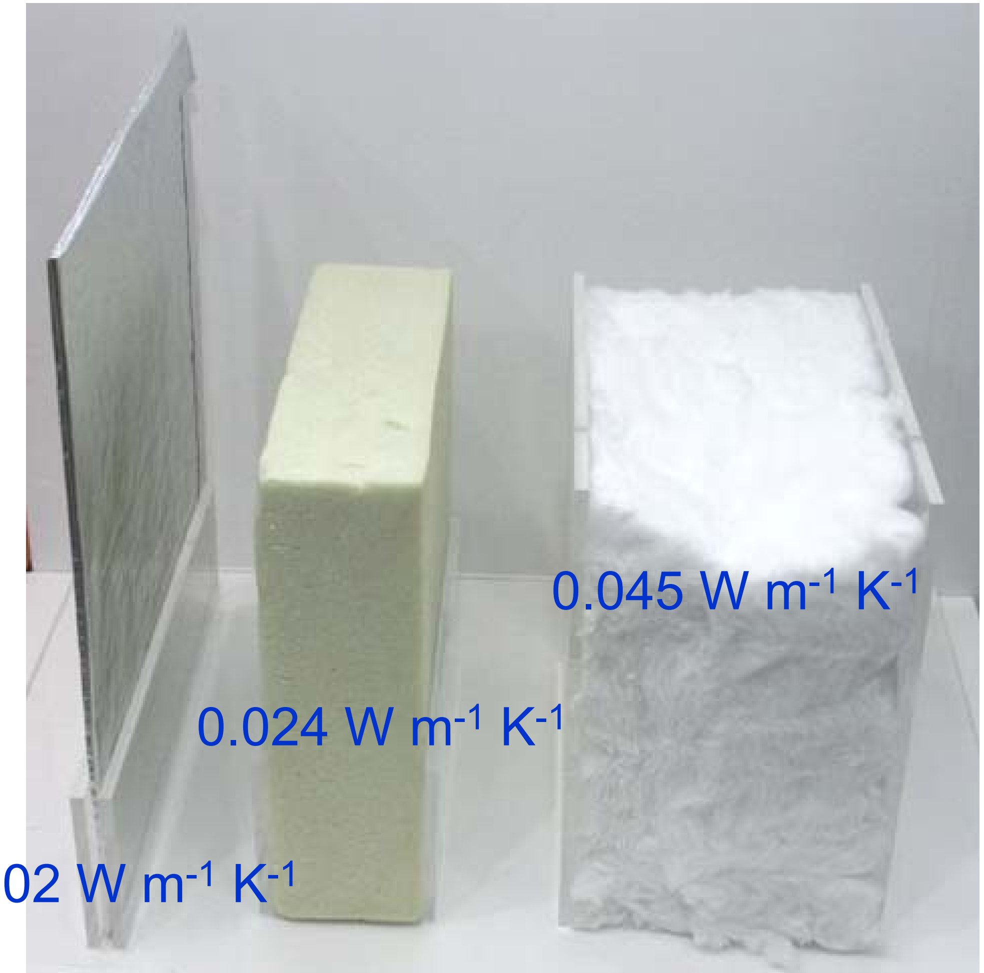
6.1 Thermal transport property
- 6.1.1 Thermal conductivity
- 6.1.2 Thermal diffusivity

6.2 Caloric property
- 6.2.1 Specific heat capacity
- 6.2.2 Heat of fusion
- 6.2.3 Calorific value

6.3 Radiative property
- 6.3.1 Spectral emissivity
- 6.3.2 Total emissivity

6.4 Thermo-mechanical property
- 6.4.1 Thermal expansion coefficient

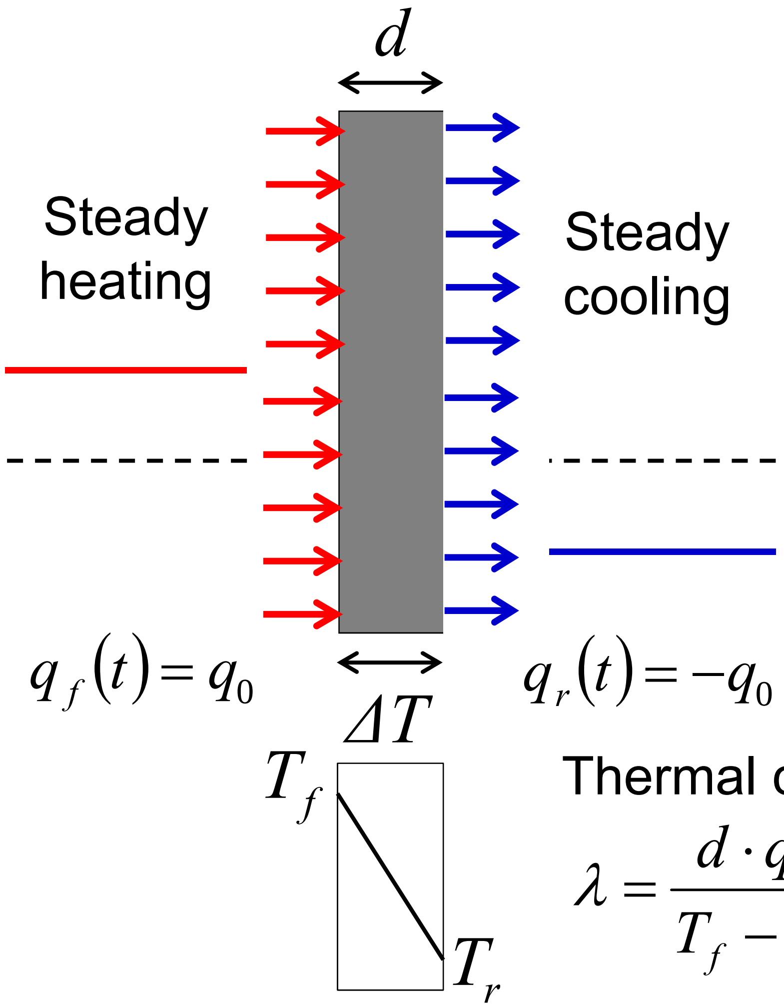
Vacuum insulation panel (VIP)



VIP   Urethane form   Glass wool

Measurement of thermal conductivity

Thermal conductivity, λ, measurement by steady method



Thermal conductivity

$$\lambda = \frac{d \cdot q_0}{T_f - T_r} = \frac{d \cdot q_0}{\Delta T_{fr}}$$

Three pilot studies organized by CCT WG9

CCT-P01 (CCT-S2)

Thermal conductivity of insulating materials

Temperature range	0 - 100 ° C
Measurement technique	guarded hot plate method
Pilot institute	LNE, France

CCT-S2

Information

Metrology area, branch	Thermometry, Thermophysical quantities
Description	Thermal conductivity
Time of measurement	2007 - 2010
Status	Report in progress, Draft A
Reference(s)	No references available
Measurand	Thermal conductivity: 0.01 $Wm^{-1}K^{-1}$ to 0.1 $Wm^{-1}K^{-1}$
Parameter(s)	Temperature: 10 °C, 23 °C and 40 °C
Transfer device(s)	Two thermal insulating materials
Comparison type	Supplementary comparison
Consultative Committee	CCT (Consultative Committee for Thermometry)
Conducted by	CCT (Consultative Committee for Thermometry)

Ceramics and compound semiconductors

AlN

SiC

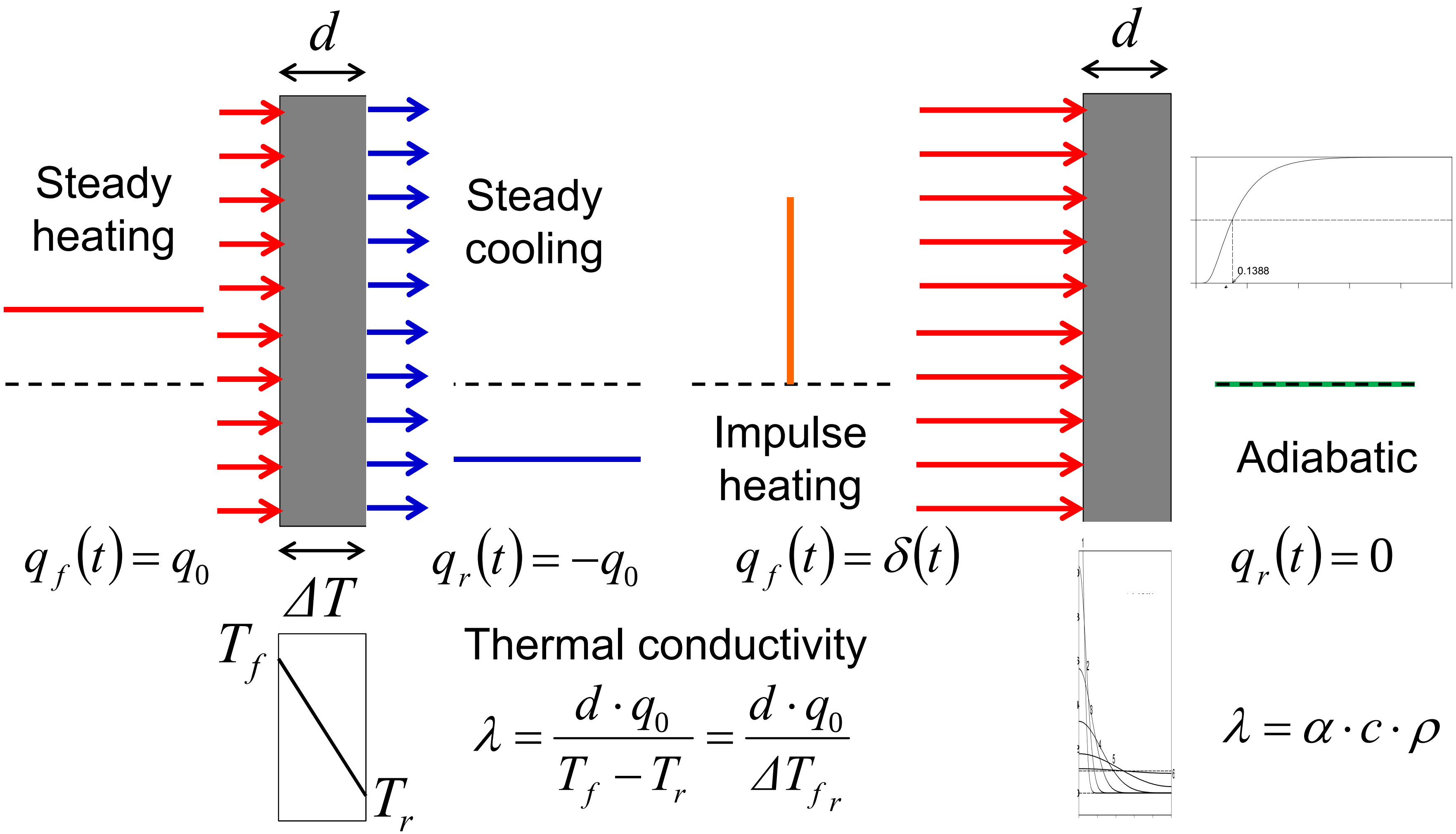
Si_3N_4

GaN

Measurement of thermal conductivity and thermal diffusivity

Thermal conductivity, λ, measurement by steady method

Thermal diffusivity, α, measurement by dynamic method

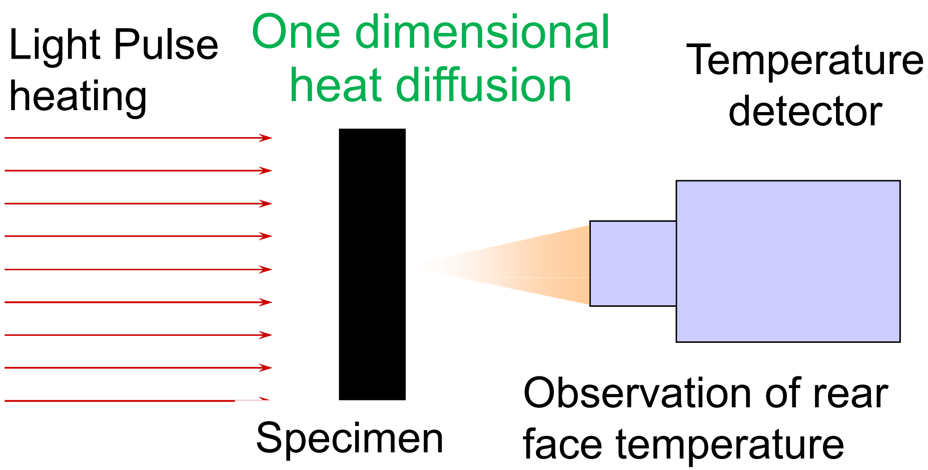


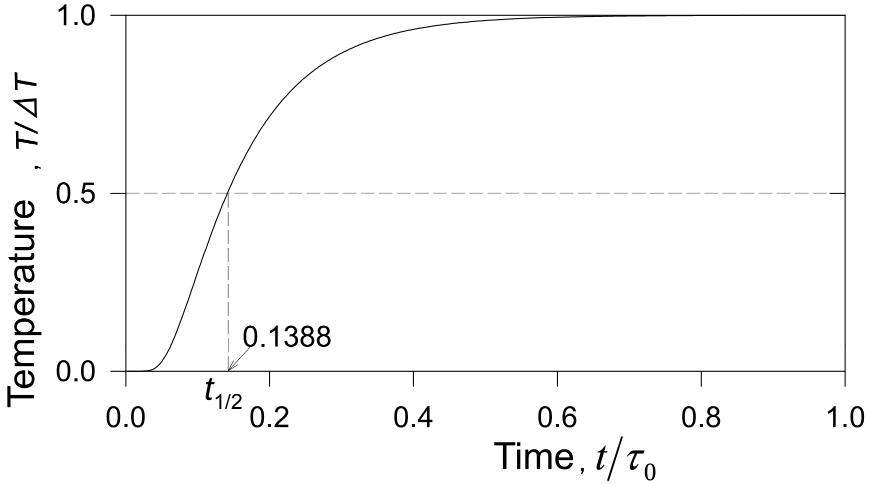
Thermal conductivity

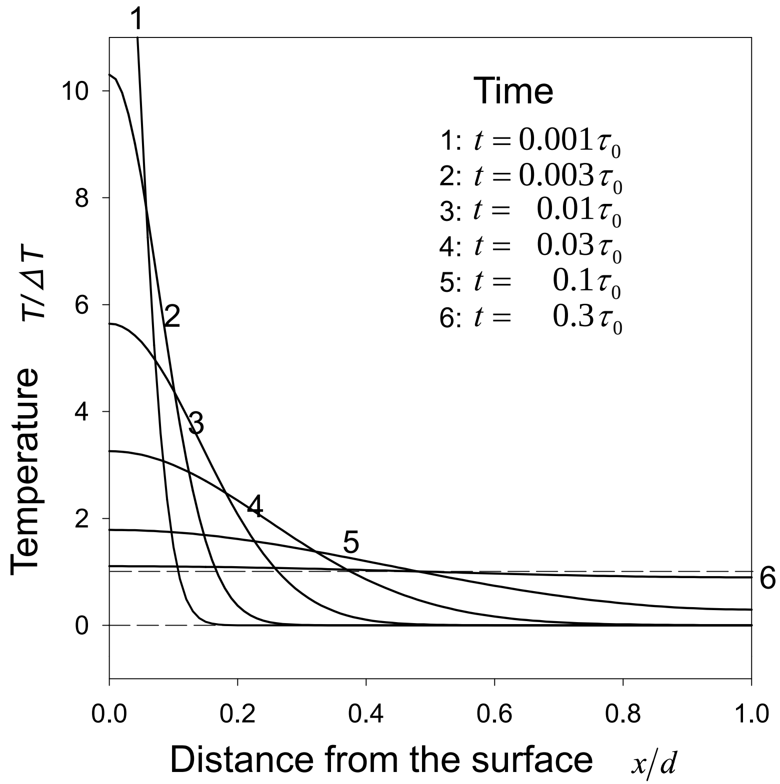
$$\lambda = \frac{d \cdot q_0}{T_f - T_r} = \frac{d \cdot q_0}{\Delta T_{fr}}$$

$$\lambda = \alpha \cdot c \cdot \rho$$

Laser flash method





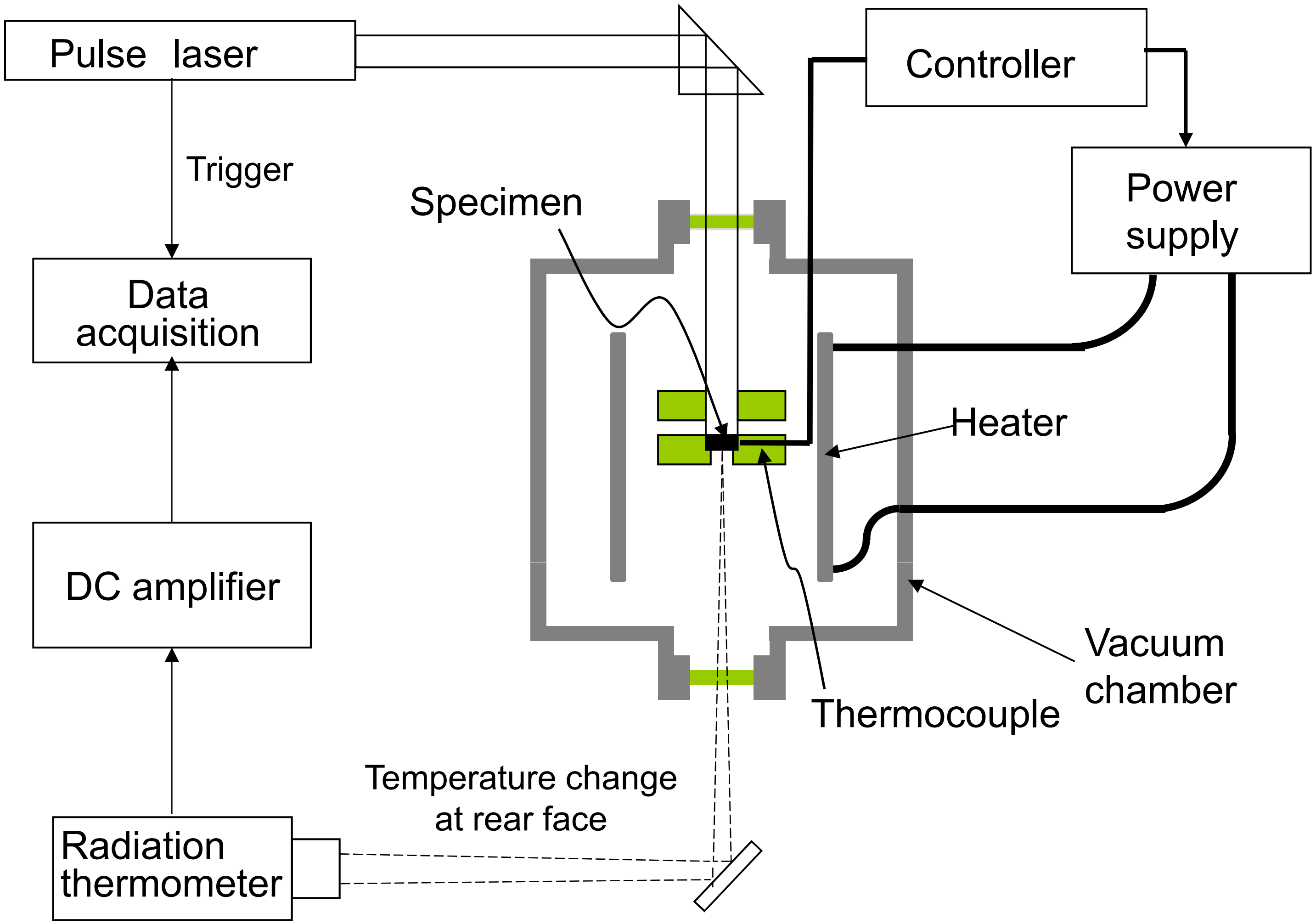


Thermal diffusivity

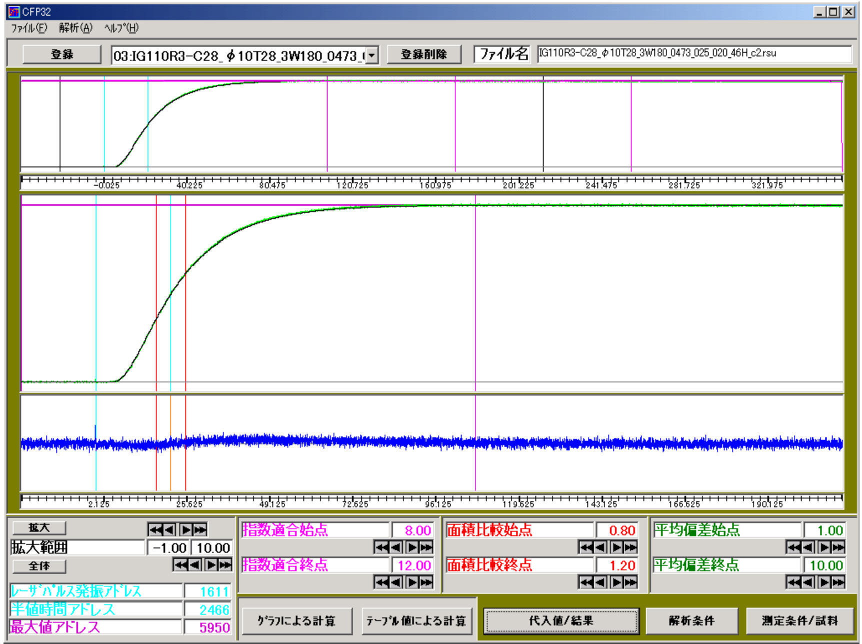
$$\kappa = \frac{d^2}{\tau_0} = 0.1388\frac{d^2}{t_{1/2}}$$

W. J. Parker, et. al. *J. Appl. Phys.*, vol.32, **1961**, pp1679–84

Bloch diagram of the laser flash method



Thermal diffusivity measurement of carbon by the laser flash method



Three pilot studies organized by CCT WG9

CCT-P02 (CCT-S3)

Thermal diffusivity of dense materials

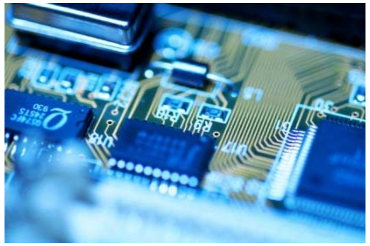
Temperature range	RT - 1000 °C
Measurement technique	laser flash method
Pilot institute	NMIJ, Japan

CCT-S3

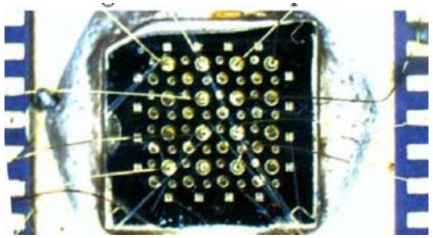
Information

Metrology area, branch	Thermometry, Thermophysical quantities
Description	Thermal diffusivity
Time of measurement	2007 - 2008
Status	Report in progress, Draft A
Reference(s)	No references available
Measurand	Thermal diffusivity in m^2s^{-1}
Parameter(s)	Temperature: 300 K to 1200 K
Transfer device(s)	Dense materials
Comparison type	Supplementary comparison
Consultative Committee	CCT (Consultative Committee for Thermometry)
Conducted by	CCT (Consultative Committee for Thermometry)
Comments	Measurements using laser flash method for dense materials

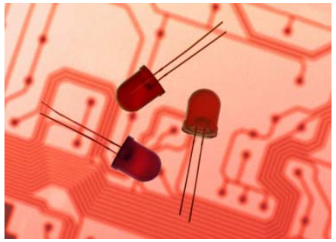
Needs for thermophysical property data by electronics industry



LSI



Power device

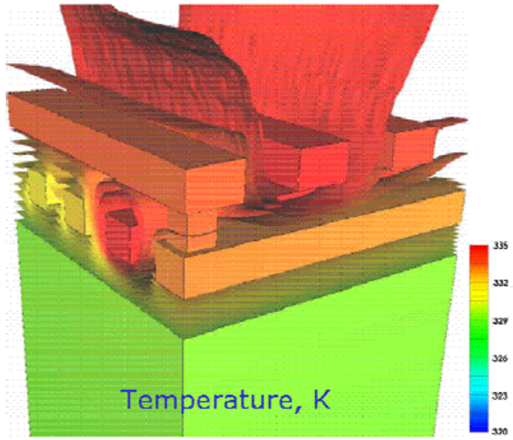


Lighting

Flat panel display

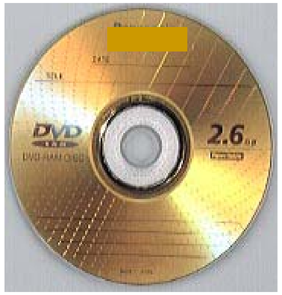
Low K in LSI , AlN and diamond for heat spreader,
GaN for LED, ITO and IZO in FPD

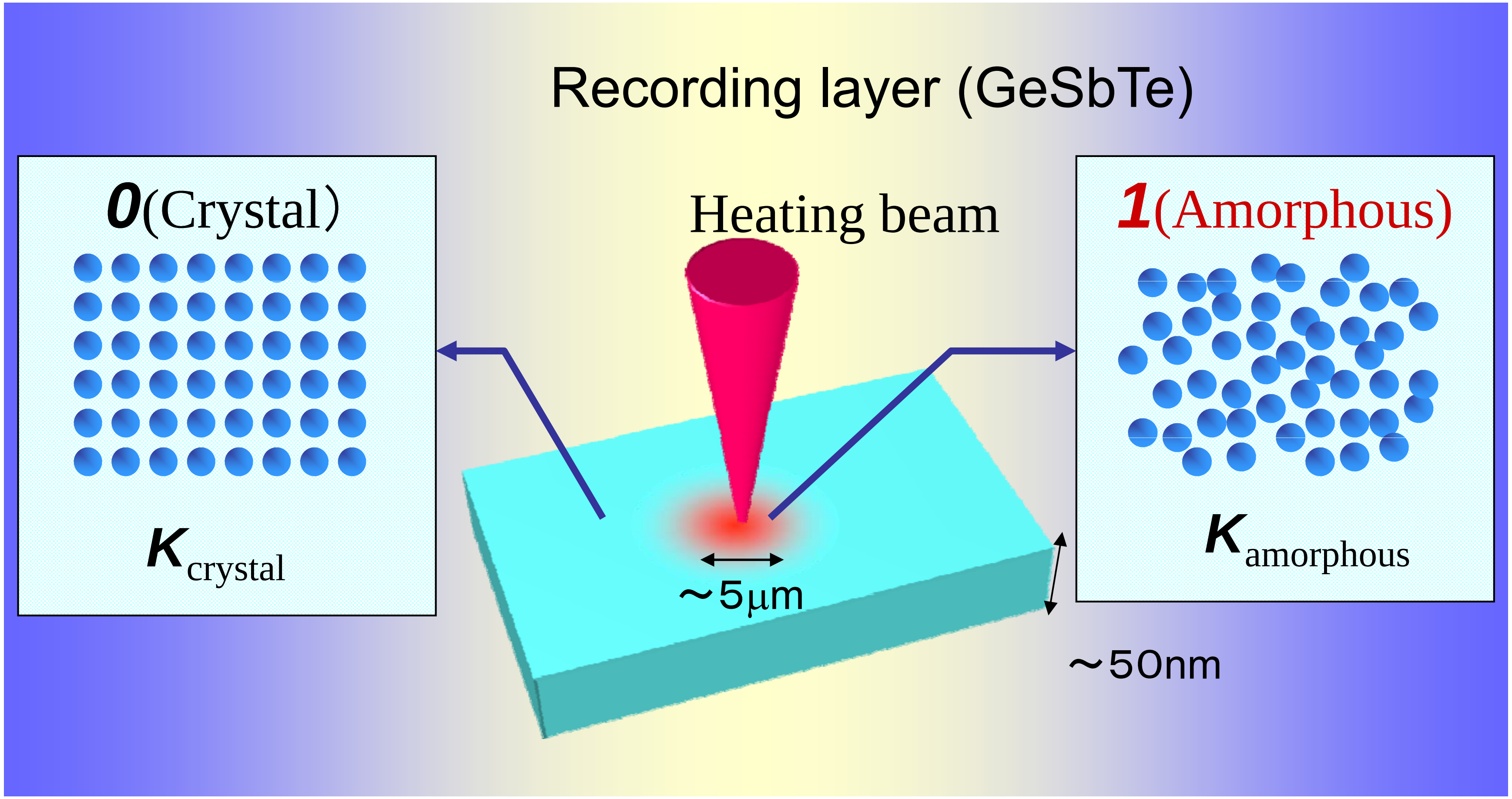
Thermophysical property data
- Thermal conductivity
- Thermal diffusivity
- Specific heat capacity



Thermal design and simulation

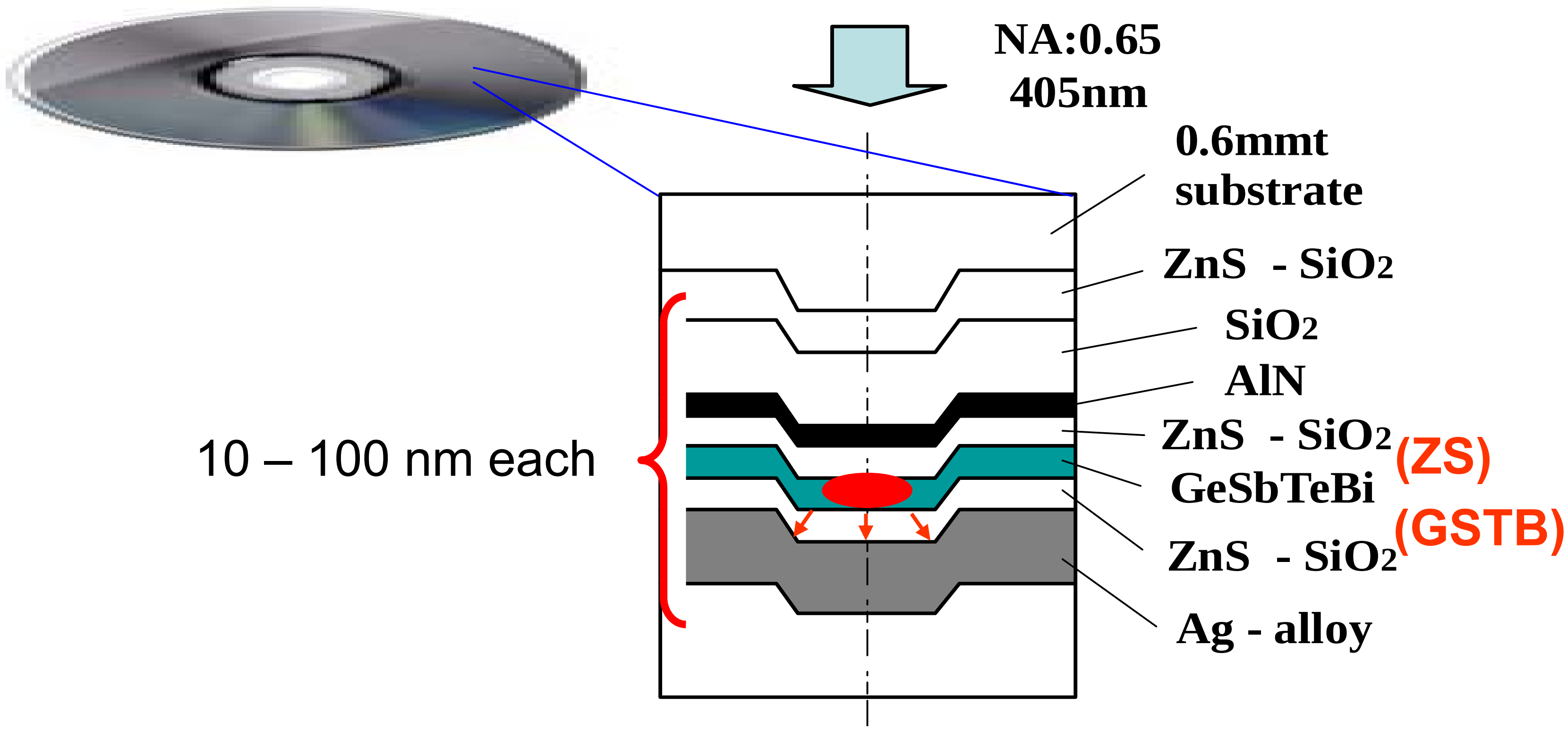
Recording by phase change in optical recording media





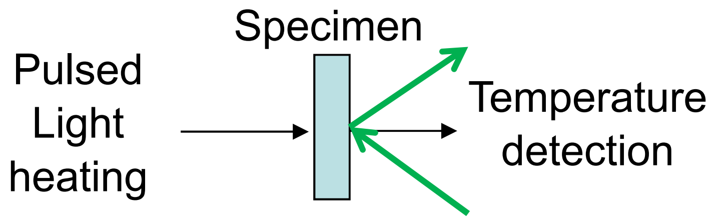
The Structure of Thin Film Stack : HD DVD-ARW Medium

Reliable values are especially needed for ZS, GSTB and boundary resistance between them.



Length and time scale of pulsed light heating methods

Heat diffusion time $\boxed{\tau = \dfrac{d^2}{\alpha}}$ Thickness / Thermal diffusivity



Laser flash method
Bulk materials, 1mm → 10ms-10s

Observation of radiation

Nanosecond TR method
Thin films, 1µm

Change of reflectivity as a function of surface temperature

10ns-10µs

$$R = R_0 + \frac{dR}{dT}\Delta T$$

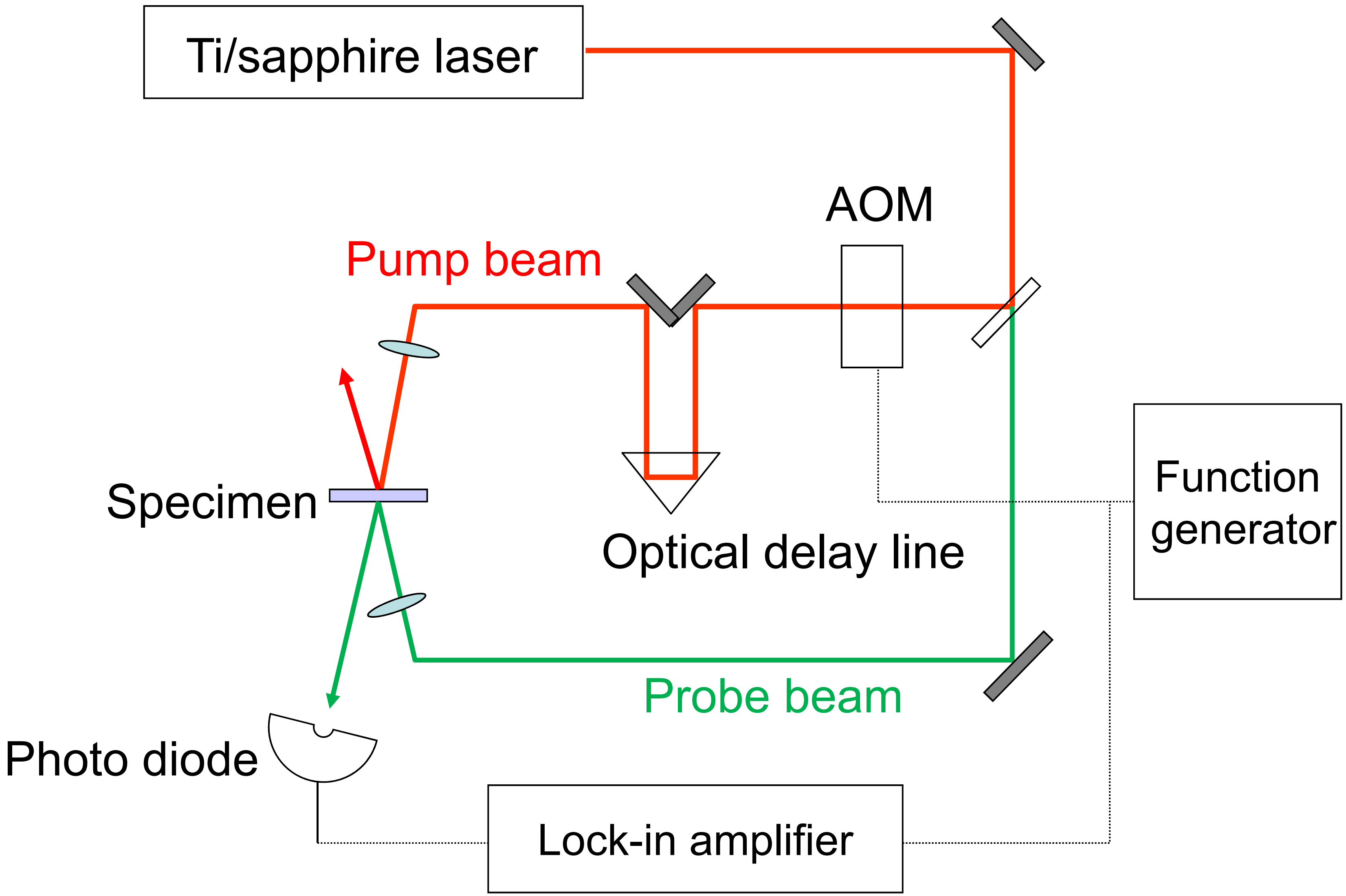
$$\frac{1}{R}\frac{dR}{dT} = 10^{-4} \sim 10^{-5}$$

Thermoreflectance method

Picosecond TR method
Thin films, 30nm

10ps-10ns

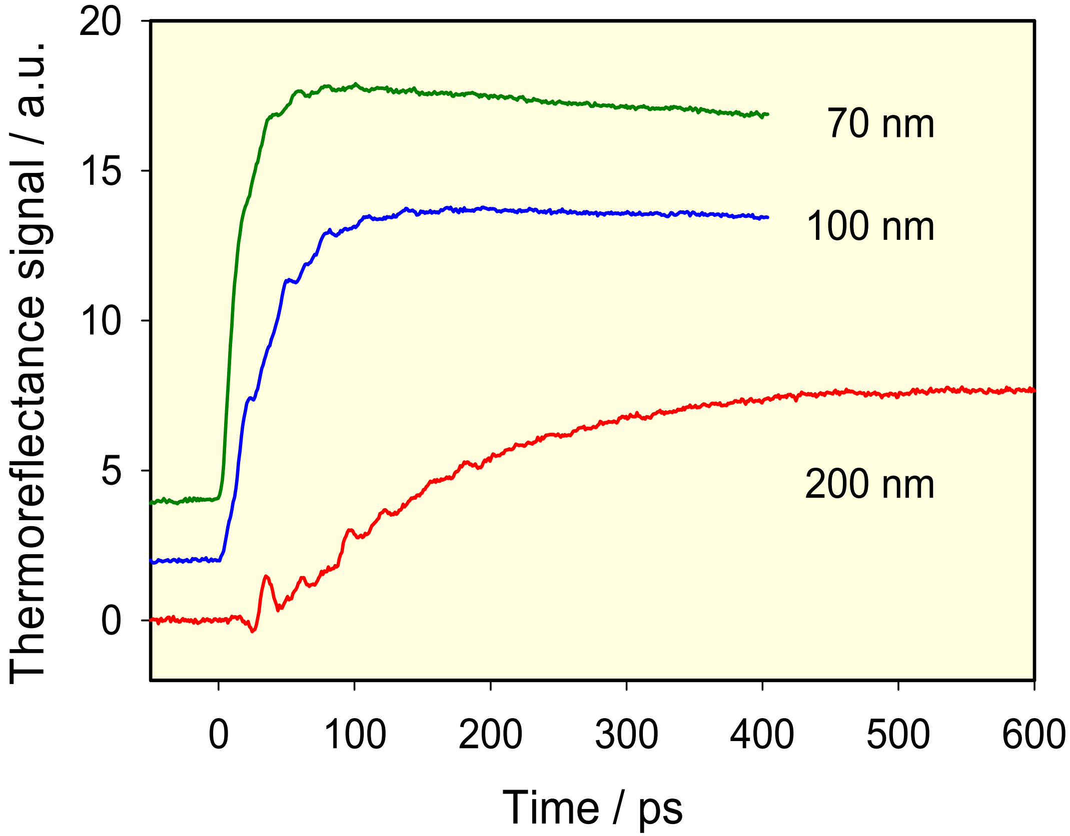
Block diagram of measurement system by the picosecond thermoreflectance method

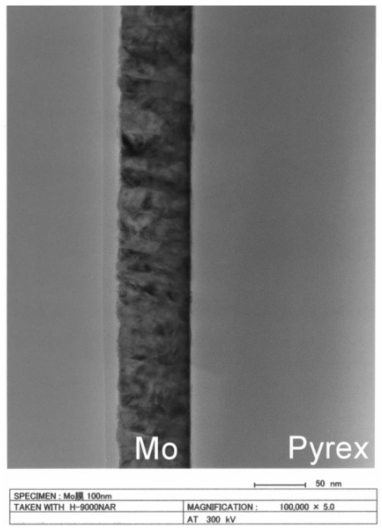


Thermal diffusivity of molybdenum thin films measured with the picosecond thermoreflectance method

Synthesized by magnetron DC sputtering
Substrate：Corning 7740 glass

Temperature response curve of Mo thin films



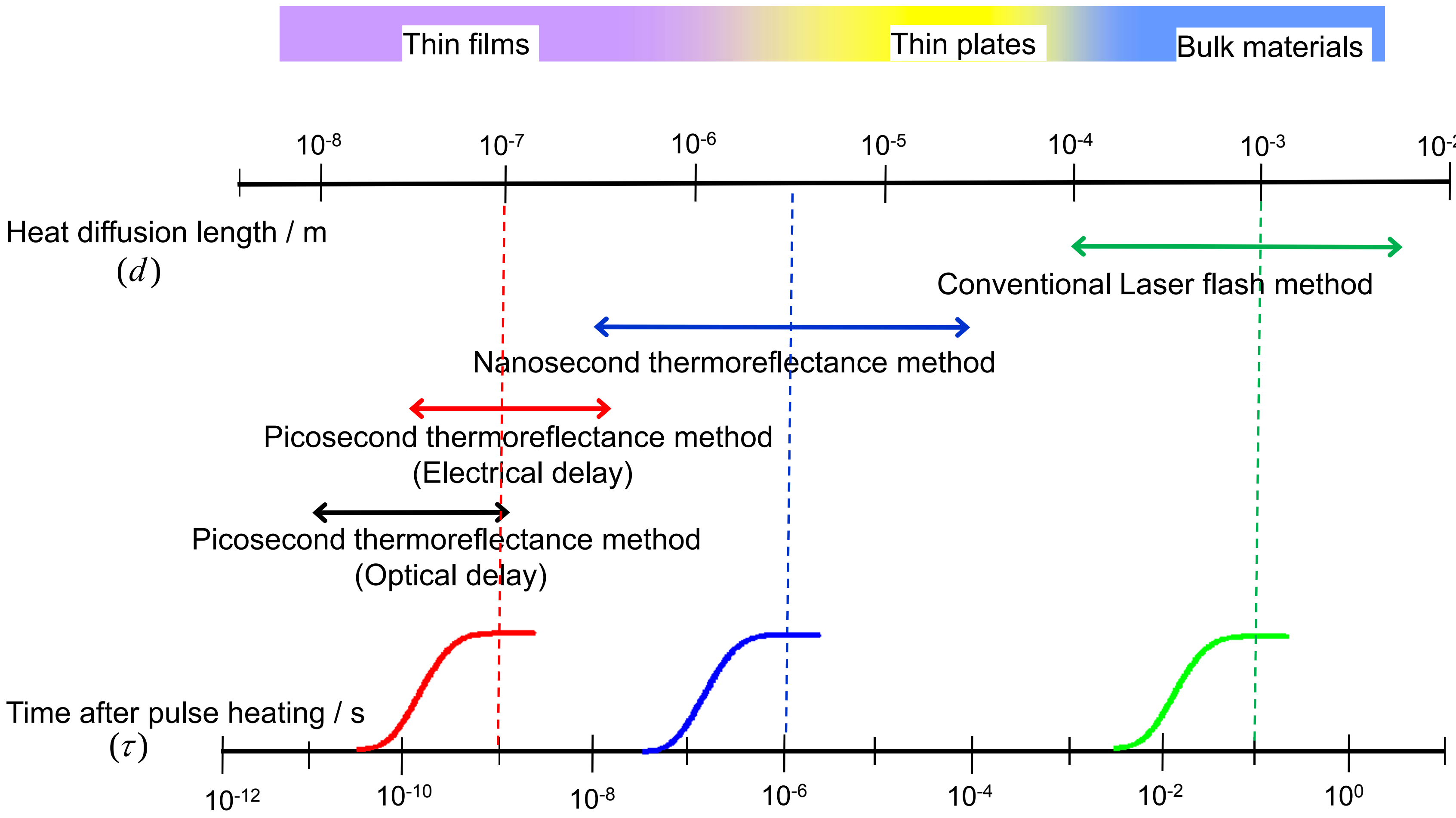


Thermal diffusivity of molybdenum thin films measured with the picosecond thermoreflectance method

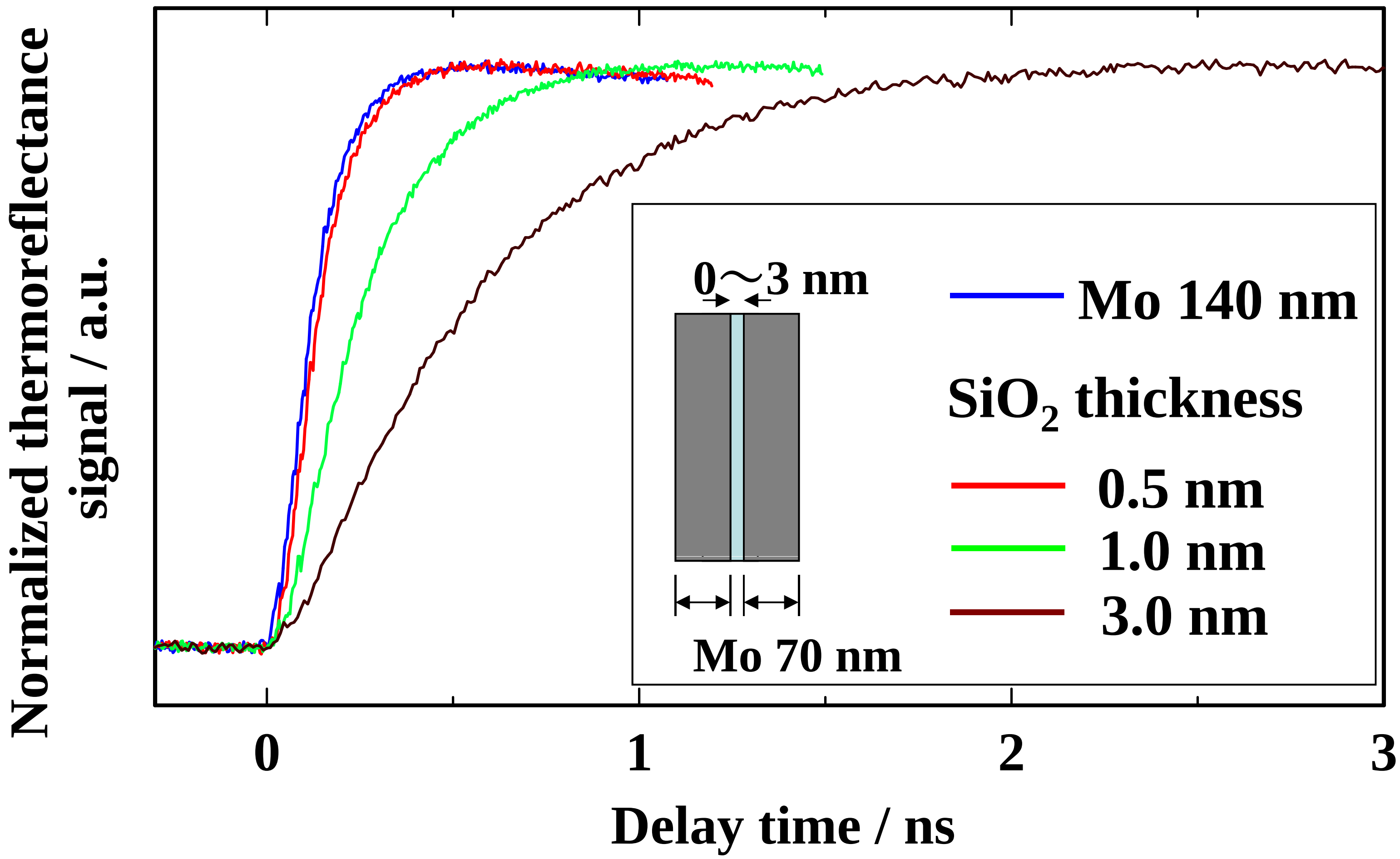
Synthesized by magnetron DC sputtering
Substrate：Corning 7740 glass

Nominal thickness nm	Measured thickness nm	Absorption coefficient x $10^{7} m^{-1}$	Thermal diffusivity $10^{-5} m^{2}s^{-1}$
70	65.4	5.8	3.0
100	94.6	5.4	3.6
200	191.0	5.0	3.0
Bulk	−	5.5	5.4

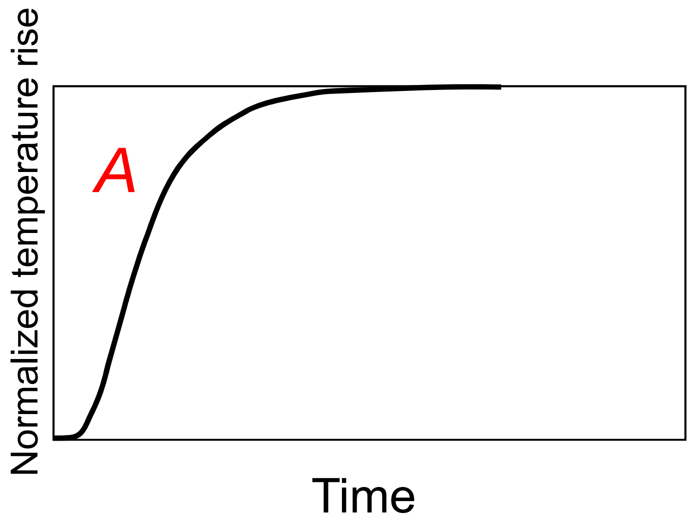
Thermal diffusivity measurements from bulk materials to thin films by pulsed light heating methods



(Thermal diffusivity, $\alpha = 1\times10^{-5}\ \mathrm{m^2s^{-1}}$, $\tau = d^2/\alpha$)


Normalized thermoreflectance signal / a.u.
0
1
2
3
Delay time / ns
0～3 nm
Mo 70 nm
Mo 140 nm
SiO2 thickness
0.5 nm
1.0 nm
3.0 nm

Definition of areal heat diffusion time



Areal heat diffusion time

$$A = \int_0^\infty \left[1 - T_r(t)/T_{\max}\right]dt$$

$$= \lim_{\xi \to 0} \int_0^\infty \exp(-\xi t)\left[1 - T_r(t)/T_{\max}\right]dt$$

$$= \lim_{\xi \to 0}\left[\frac{1}{\xi} - \boxed{\tilde{T}_r(\xi)}/T_{\max}\right]$$

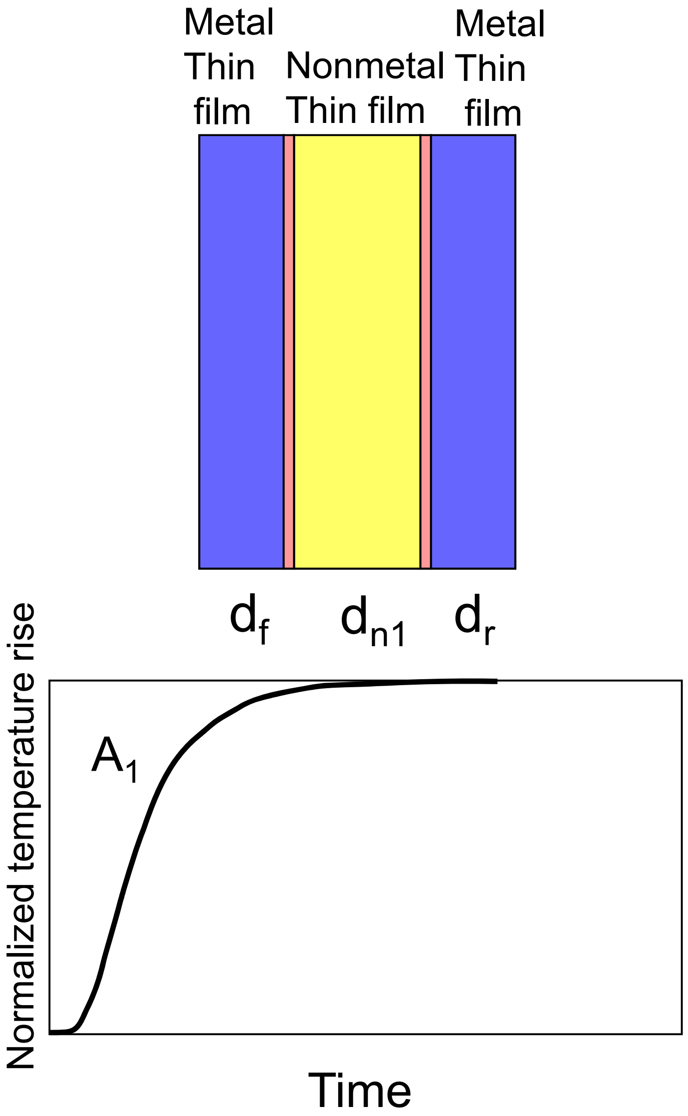
$$= \lim_{\xi \to 0}\left[\frac{1}{\xi} - \boxed{\tilde{R}_{rf}(\xi)}/T_{\max}\right]$$

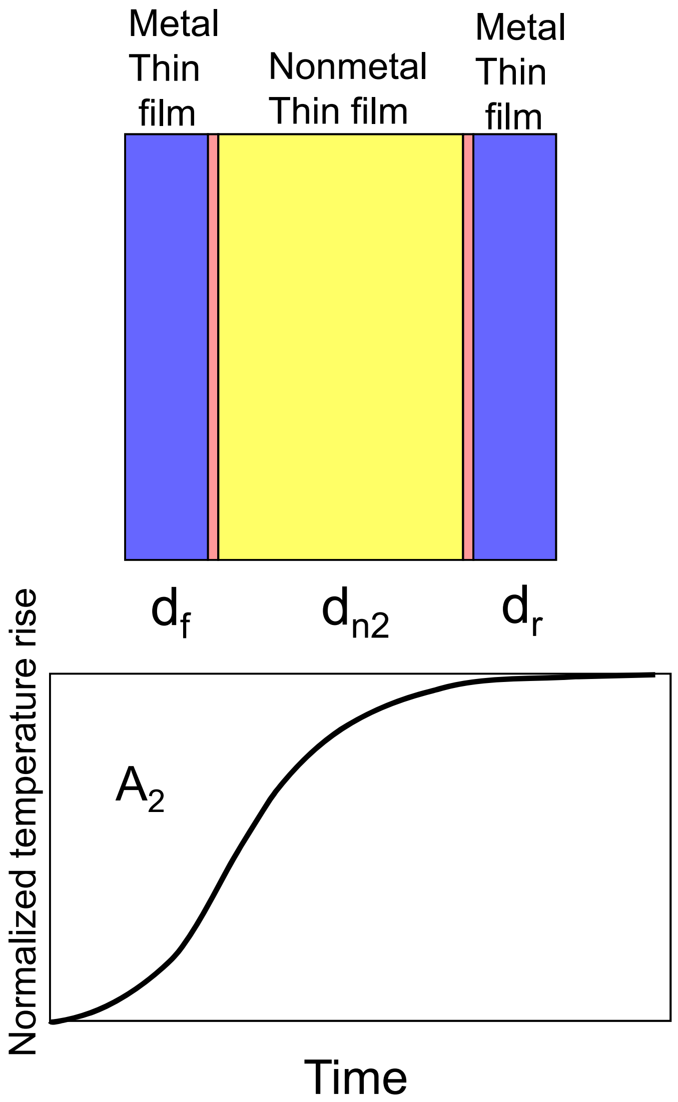
Transfer function

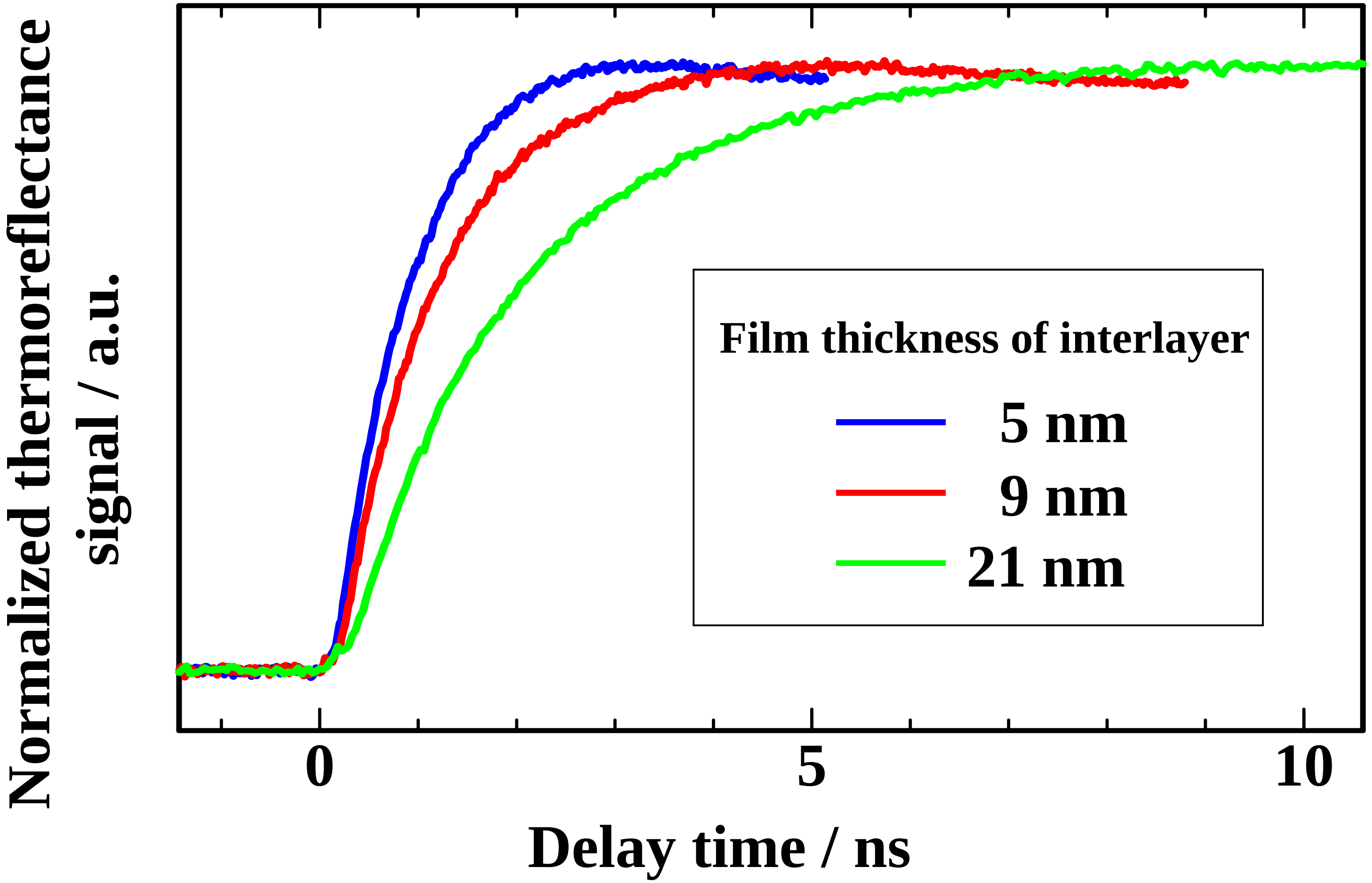
Laplace transformation

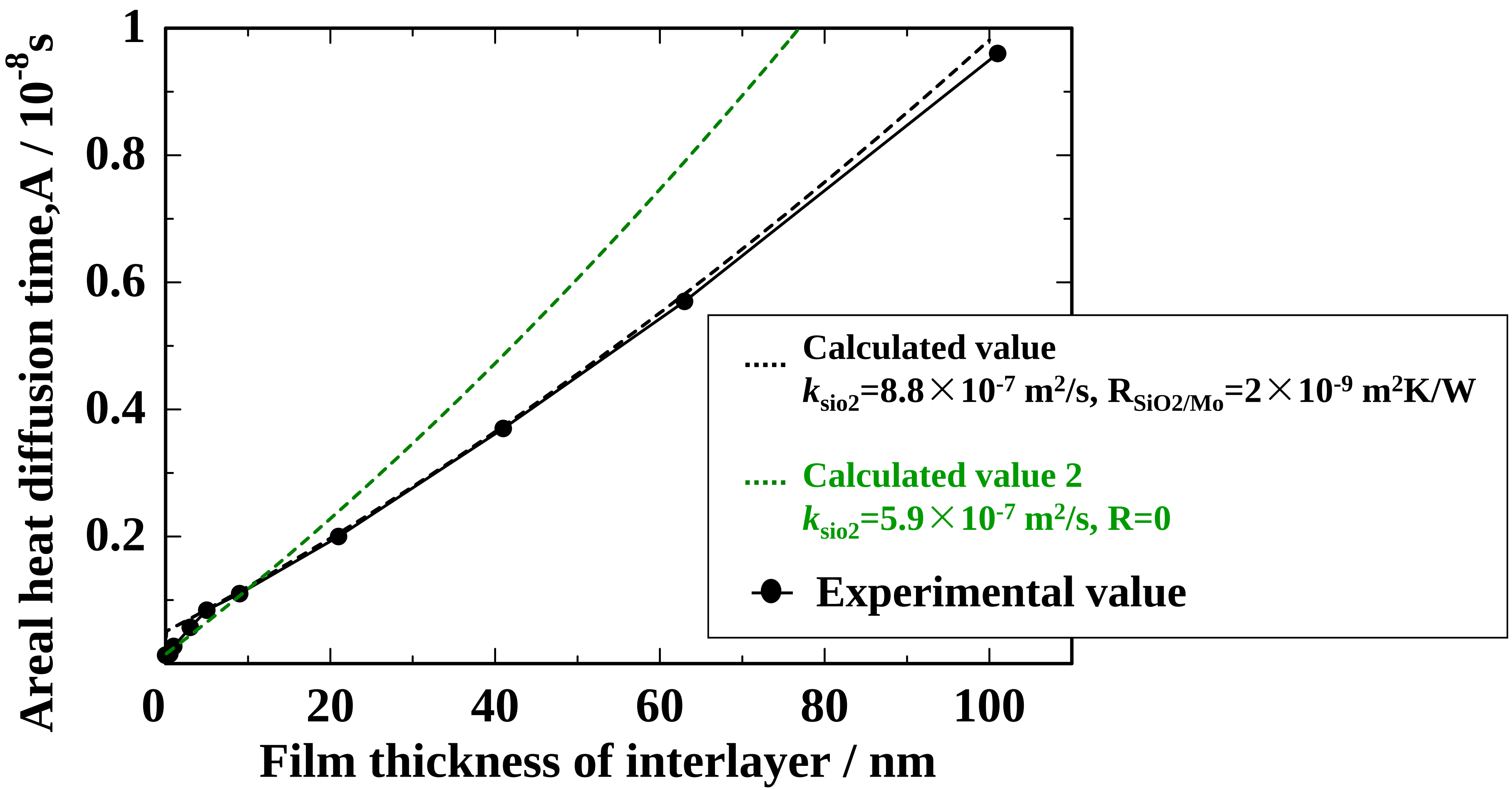
$$\tilde{f}(\xi) = \int_0^\infty \exp(-\xi t) f(t)dt$$

Temperature history curves of 3 layer specimens with different thickness of nonmetal thin film






Normalized thermoreflectance signal / a.u.
Delay time / ns
0
5
10
Film thickness of interlayer
5 nm
9 nm
21 nm

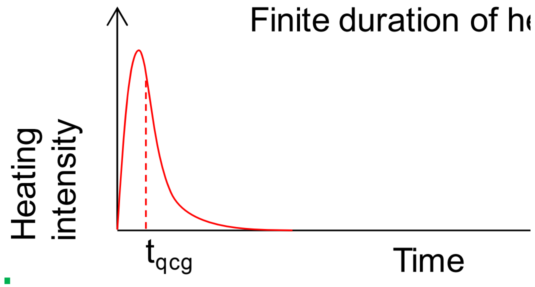

Areal heat diffusion time,A / 10-8 s
1
0.8
0.6
0.4
0.2
0
20
40
60
80
100
Film thickness of interlayer / nm
Calculated value
ksio2=8.8×10-7 m2/s, RSiO2/Mo=2×10-9 m2K/W
Calculated value 2
ksio2=5.9×10-7 m2/s, R=0
Experimental value

Thin films and boundary thermal resistances measured by this study

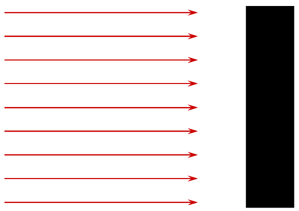
- Metals: Al, Mo, W, Pt, etc.
- Oxides: SiO_2, Al2O_3,In_2O_3, ZnO, etc.
- Nitrides: TiN, AlN, etc.
- Compounds composed of 3 elements: $Mg(OH)_2$, GeSbTe, etc
- Compounds composed of 4 elements: AgInSbTe, etc
- Boundary thermal resistance between metal layer and nonmetal layer
  Mo/SiO_2, Mo/Al2O_3,Mo/In_2O_3, etc.

Major factors contributing uncertainty of dynamic thermophysical quantity measurement

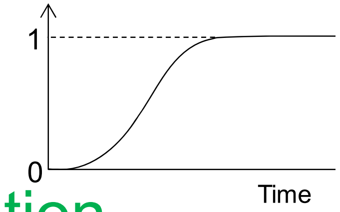
- Heating Heating function
  Impulse, step, sinusoidal, etc.
  Intensity variation in time domain



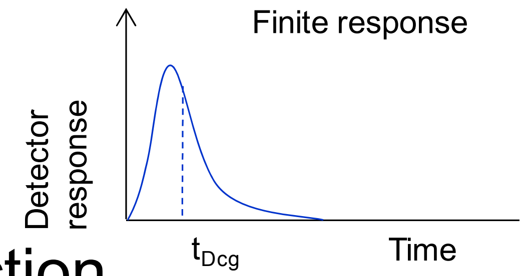
- Geometry Heat conduction equation



  Shape and dimensions of the specimen
  Heating method and position
  Temperature detection method and position



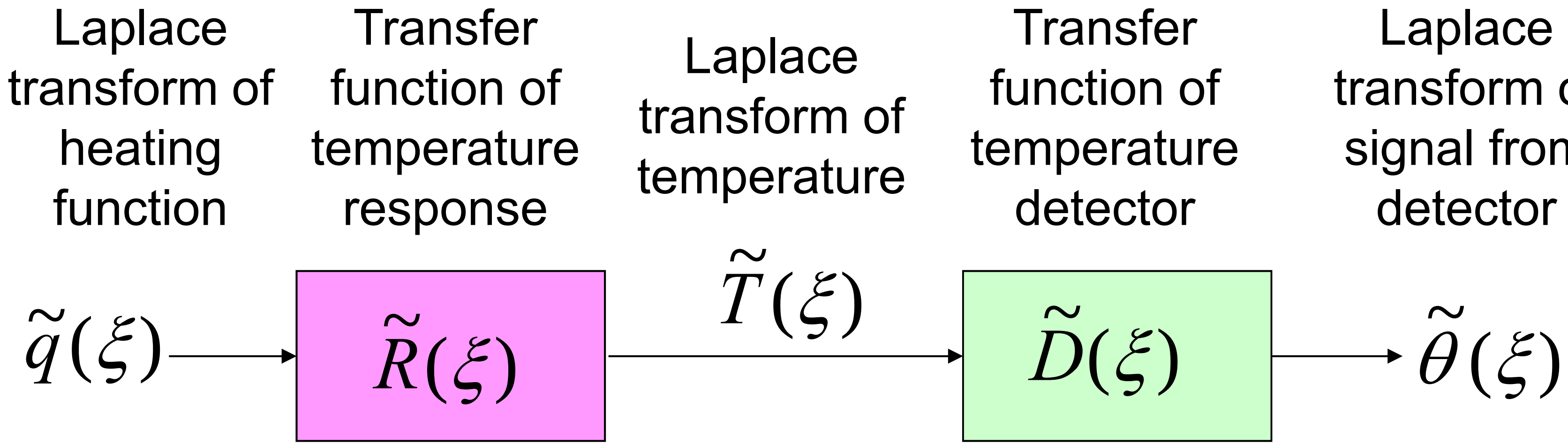
- Response of temperature detection
  Nonlinearity
  Response time



Impulse response function

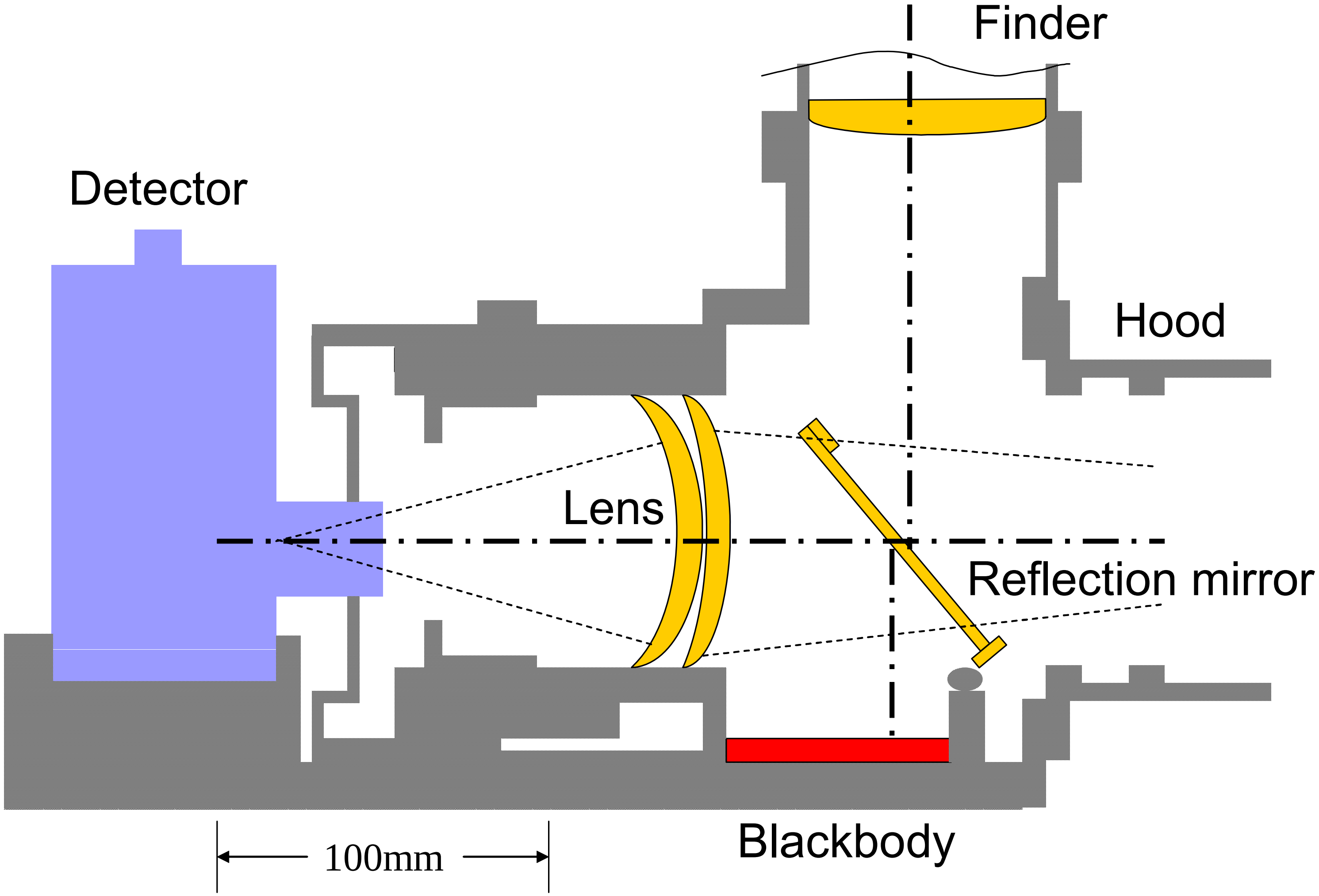
Block diagram of dynamic thermophysical quantity measurement

- Heating
- Heat diffusion
- Response of detector

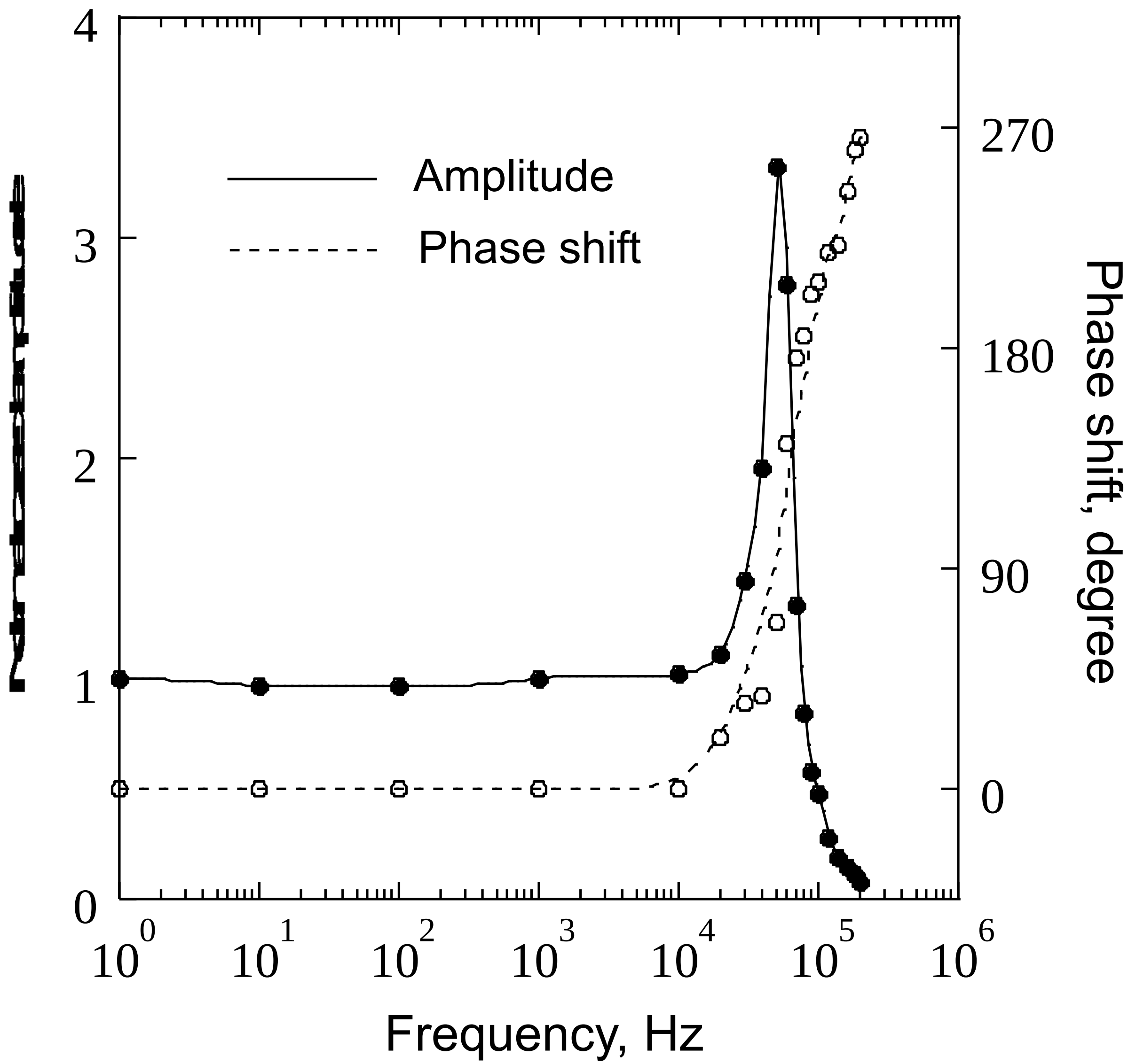


$$\tilde{\theta}(\xi) = \tilde{D}(\xi)\cdot\tilde{R}(\xi)\cdot\tilde{q}(\xi) \qquad \tilde{R}(\xi) = \frac{\tilde{\theta}(\xi)}{\tilde{D}(\xi)\cdot\tilde{q}(\xi)}$$

Infrared radiation thermometer for laser flash thermal diffusivity measurements



Frequency response of the infrared radiation thermometer

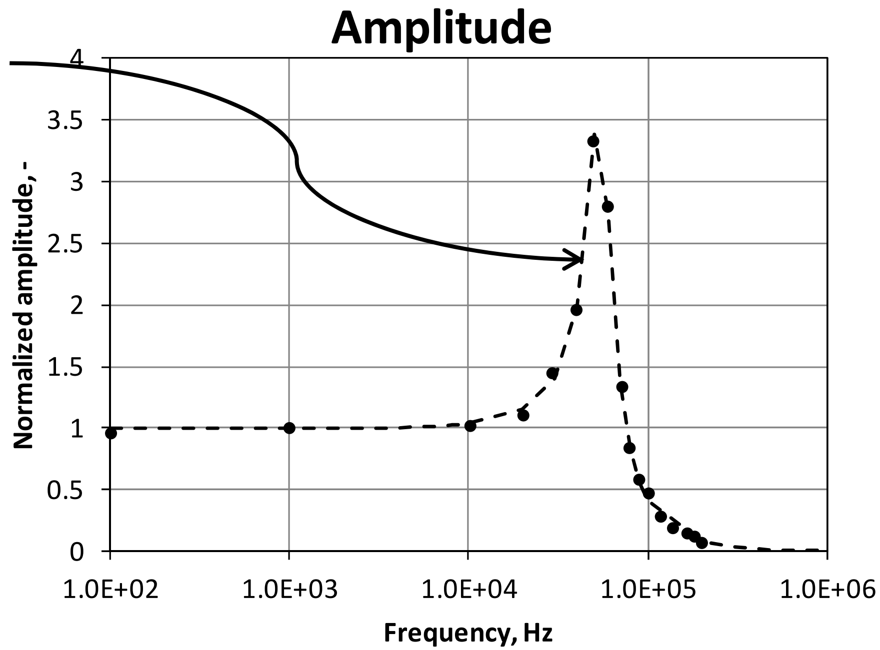


Frequency response of the infrared radiation thermometer

$$F_D(\omega) = \tilde{D}(i\omega) = \frac{\omega_n^{\ 2}}{(i\omega)^2 + 2\eta(i\omega)\omega_n + \omega_n^{\ 2}} = \frac{\omega_n^{\ 2}}{\omega_n^{\ 2} - \omega^2 + 2i\cdot\eta\cdot\omega\cdot\omega_n}$$

$$= \frac{\omega_n^{\ 2}\cdot\left(\omega_n^{\ 2} - \omega^2\right)}{\left(\omega_n^{\ 2} - \omega^2\right)^2 + 4\left(\eta\cdot\omega\cdot\omega_n\right)^2} - i\cdot\frac{2\eta\cdot\omega\cdot\omega_n^{\ 3}}{\left(\omega_n^{\ 2} - \omega^2\right)^2 + 4\left(\eta\cdot\omega\cdot\omega_n\right)^2}$$

$$\left|F_D(\omega)\right| = \frac{\omega_n^{\ 2}}{\sqrt{\left(\omega_n^{\ 2} - \omega^2\right)^2 + 4\left(\eta\cdot\omega\cdot\omega_n\right)^2}}$$



$$\omega_n = 54.6\ \mathrm{kHz}$$

$$\eta = 0.135$$

Transfer function of detector

$$\boxed{\tilde{D}(\xi) = \frac{\omega_n^{\ 2}}{\xi^2 + 2\eta\xi\omega_n + \omega_n^{\ 2}} \qquad \omega_n = 54.6\ \mathrm{kHz} \quad \eta = 0.135}$$

$$\overline{D} = \int_0^\infty D(t)\, dt = \lim_{\xi\to 0}\int_0^\infty \exp(-\xi t) D(t) dt = \lim_{\xi\to 0}\tilde{D}(\xi) = 1$$

$$\boxed{t_{Dcg}} = \frac{\int_0^\infty t\cdot D(t) dt}{\overline{D}} = \lim_{\xi\to 0}\int_0^\infty \exp(-\xi t)\cdot t\cdot D(t)dt = \lim_{\xi\to 0}\left[-\frac{d\tilde{D}(\xi)}{d\xi}\right]$$

$$= \lim_{\xi\to 0}\left[-\frac{d}{d\xi}\left(\frac{\omega_n^{\ 2}}{\xi^2 + 2\eta\xi\omega_n + \omega_n^{\ 2}}\right)\right] = \lim_{\xi\to 0}\left[\frac{\omega_n^{\ 2}\cdot(2\xi + 2\eta\omega_n)}{\left(\xi^2 + 2\eta\xi\omega_n + \omega_n^{\ 2}\right)^2}\right]$$

$$\boxed{= \frac{2\eta}{\omega_n} = \frac{2 * 0.135}{5.46 * 10^4\ \mathrm{Hz}} = 4.9 * 10^{-6}\mathrm{s}}$$

Analysis of observed areal heat diffusion time

$$\boxed{A_\theta} = \int_0^\infty \left[1-\theta(t)/\theta_{\max}\right]dt = \lim_{\xi\to 0}\int_0^\infty \exp(-\xi t)\left[1-\theta(t)/\theta_{\max}\right]dt$$

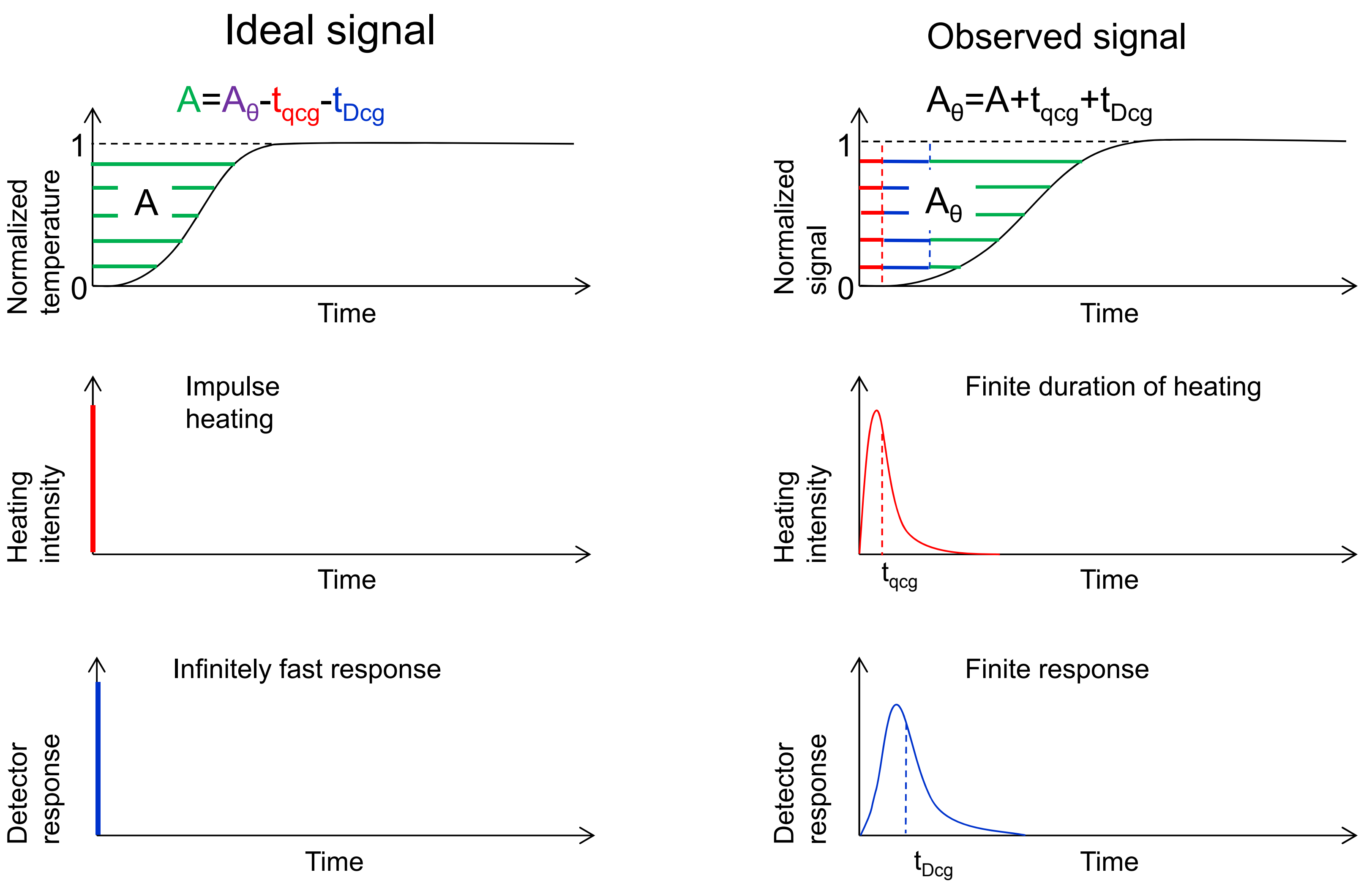
$$= \lim_{\xi\to 0}\left[\frac{1}{\xi} - \boxed{\tilde{\theta}(\xi)}/\theta_{\max}\right] = \lim_{\xi\to 0}\left[\frac{1}{\xi} - \frac{\boxed{\tilde{D}(\xi)\cdot \tilde{\mathrm{R}}_{rf}(\xi)\cdot \tilde{q}_f(\xi)}}{\overline{D}\cdot \mathrm{R}_{rf\,\max}\cdot \overline{q}}\right]$$

$$= \lim_{\xi\to 0}\left[\frac{1}{\xi} - \frac{(1-\xi\cdot t_{Dcg})\cdot \overline{D}\cdot \tilde{\mathrm{R}}_{rf}(\xi)\cdot (1-\xi\cdot t_{qcg})\cdot \overline{q}}{\overline{D}\cdot \mathrm{R}_{rf\,\max}\cdot \overline{q}}\right]$$

$$= \lim_{\xi\to 0}\left[\frac{1}{\xi} - \frac{\tilde{\mathrm{R}}_{rf}(\xi)}{\mathrm{R}_{rf\,\max}}\right] + t_{Dcg} + t_{qcg}\ \boxed{= A_T + t_{Dcg} + t_{qcg}}$$

$$\boxed{A_T = A_\theta - t_{Dcg} - t_{qcg}}$$

Correction of observed areal heat diffusion time for finite duration of the heating function and finite response of temperature detector



Uncertainty evaluation for pulsed light heating methods

1. Heat diffusion time: τ_0 ⟸ Evaluated by transfer function and areal heat diffusion time
   - Heating
   - Heat conduction equation
     Shape and dimensions of the specimen
     Boundary conditions
     Heating method and position
     Temperature detection method and position
   - Temperature detection
     Nonlinearity
     Response time
2. Thickness of the specimen: d
3. Steady temperature of the specimen: T

Thermal diffusivity at T

$$\alpha = \frac{d^2}{\tau_0}$$

Summary

- Light pulse heating methods have been developed to measure thermal diffusivity from bulk materials thicker than 1 mm to films thinner than 100 nm.
- One dimensional heat diffusion can be analyzed by the response function method and the areal heat diffusion time method.
- Uncertainty of finite heating distribution, evaluation of heat diffusion in material and finite response of detector can be universally analyzed by transfer functions and areal heat diffusion time.