Photo thermal radiometry method development and application for nuclear technologies

Alexandre Semerok, Sergey V. Fomichev, Christian Grisolia, Sang Pham Tu Quoc, Antonin Poisson, Guy Cheymol, Fernando Lomello, Alexandre Michau, Frédéric Schuster, George Hamaoui, et al.

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PHOTOTHERMAL RADIOMETRY METHOD DEVELOPMENT AND APPLICATION FOR NUCLEAR TECHNOLOGIES

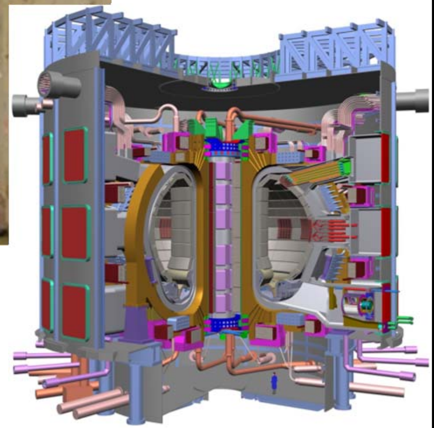
A. Semerok [1], S.V. Fomichev [2,3], C. Grisolia [4], S. Pham Tu Quoc [1]
A. Poisson [1], G. Cheymol [1], F. Lomello [1], A. Michau [1], F. Schuster [5]
G. Hamaoui [6], N. Horny [6], M. Chirtoc [6] and H. Maskrot [1]

1. Den-Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, F-91191, Gif sur Yvette, France
2. National Research Center "Kurchatov Institute", 123182 Moscow, Russia
3. Moscow Institute of Physics and Technology, 141700 Dolgoprudny, Moscow region, Russia
4. CEA, IRFM, F-13108, Saint Paul Lez Durance, France
5. CEA Cross-Cutting Program on Materials and Processes Skills, F-91191, Gif sur Yvette, France
6. Multiscale Thermophysics Lab, GRESPI, Université de Reims Champagne Ardenne, URCA, Moulin de la Housse, BP 1039, Reims, 51687, France

10 July 2019

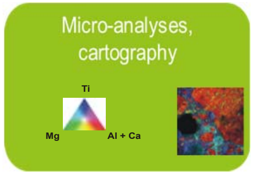
Development of Innovated Laser Methods in DEN/DANS/DPC/SEARS CEA Saclay

- Surface Decontamination:
  - paint on concrete;
  - metals;
  - laser detritiation for ITER (heating and ablation);

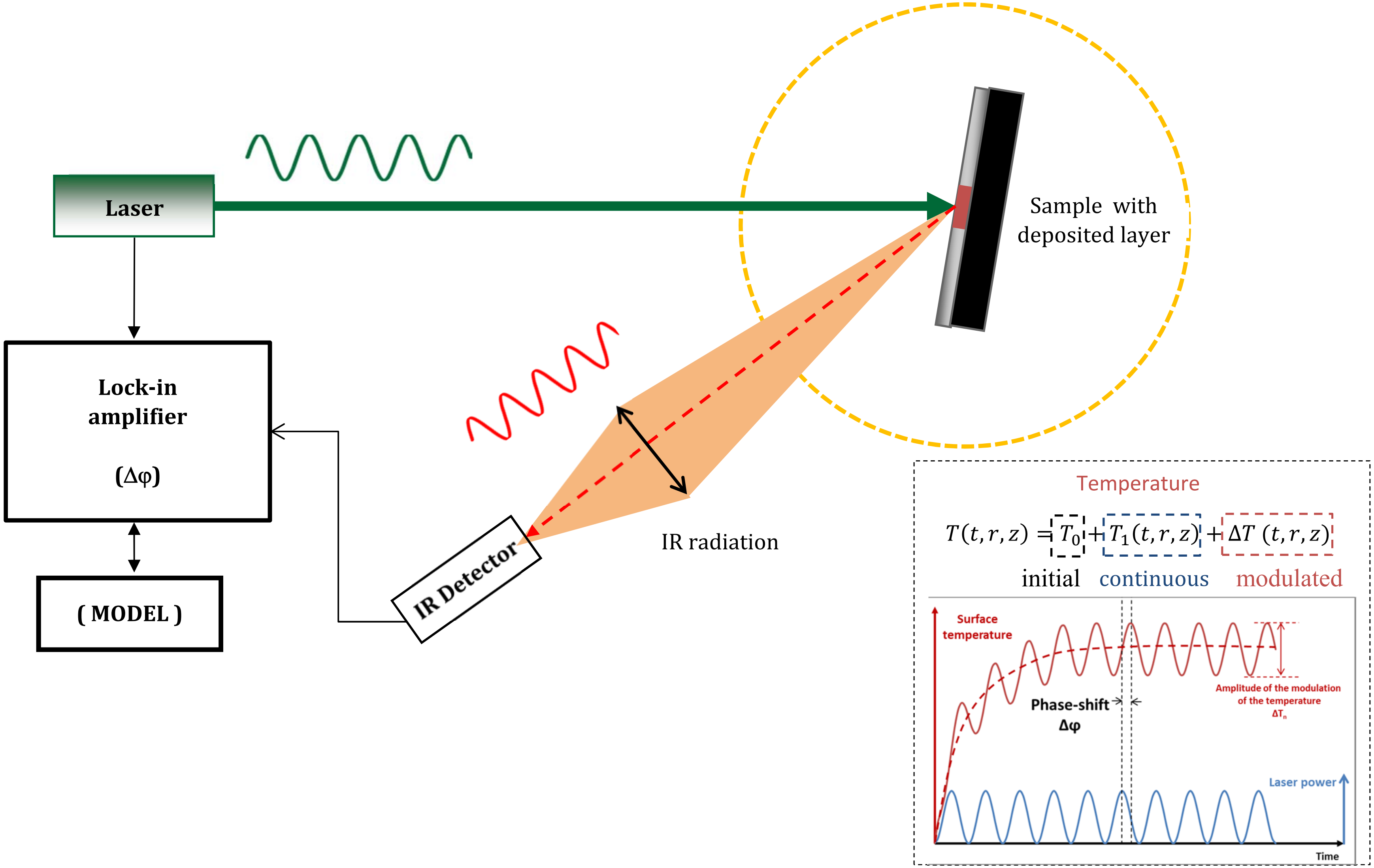


- Surface Improvement and Micro Processing:
  - material durability by rapid laser heating (corrosion resistance);
  - laser projection of powder;
  - micro gravure (microsystems);

- Analytical Methods :
  - LIBS (elemental macro and µ-analysis):
    - ✓ in nuclear environment (portable LIBS)
    - ✓ in situ purity control of liquid sodium;
    - ✓ on line analysis of liquid glasses (wastes);
    - ✓ elemental µ-cartography;
  - ChemCam for the Mars exploration;
  - Active laser pyrometry (layer thickness, resistance, conductivity,….);
  - Lock-in Thermography for characterization of nuclear materials;



LOCK-IN THERMOGRAPHY METHOD



INSTALLATION EXPÉRIMENTAL DU LISL

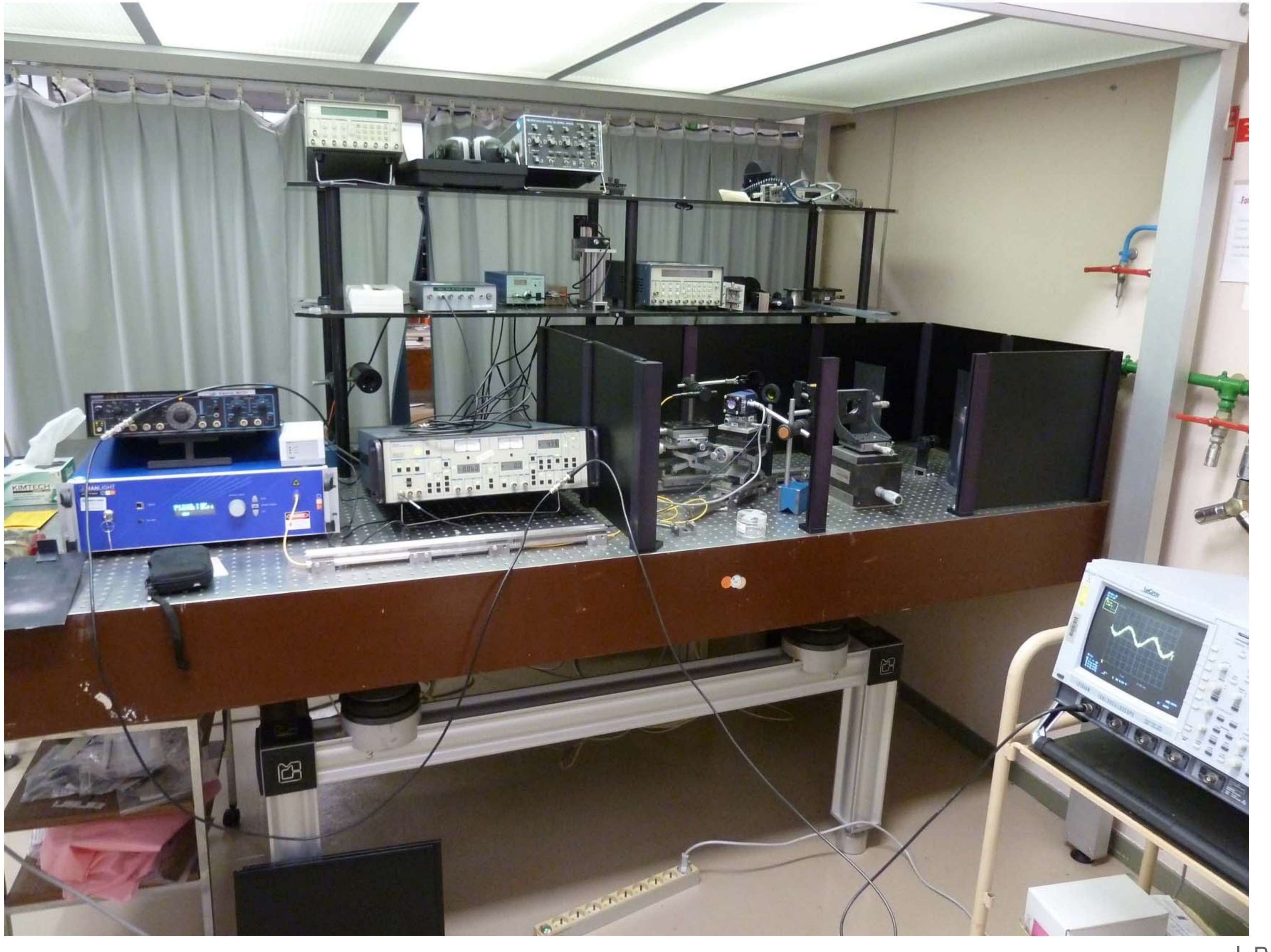
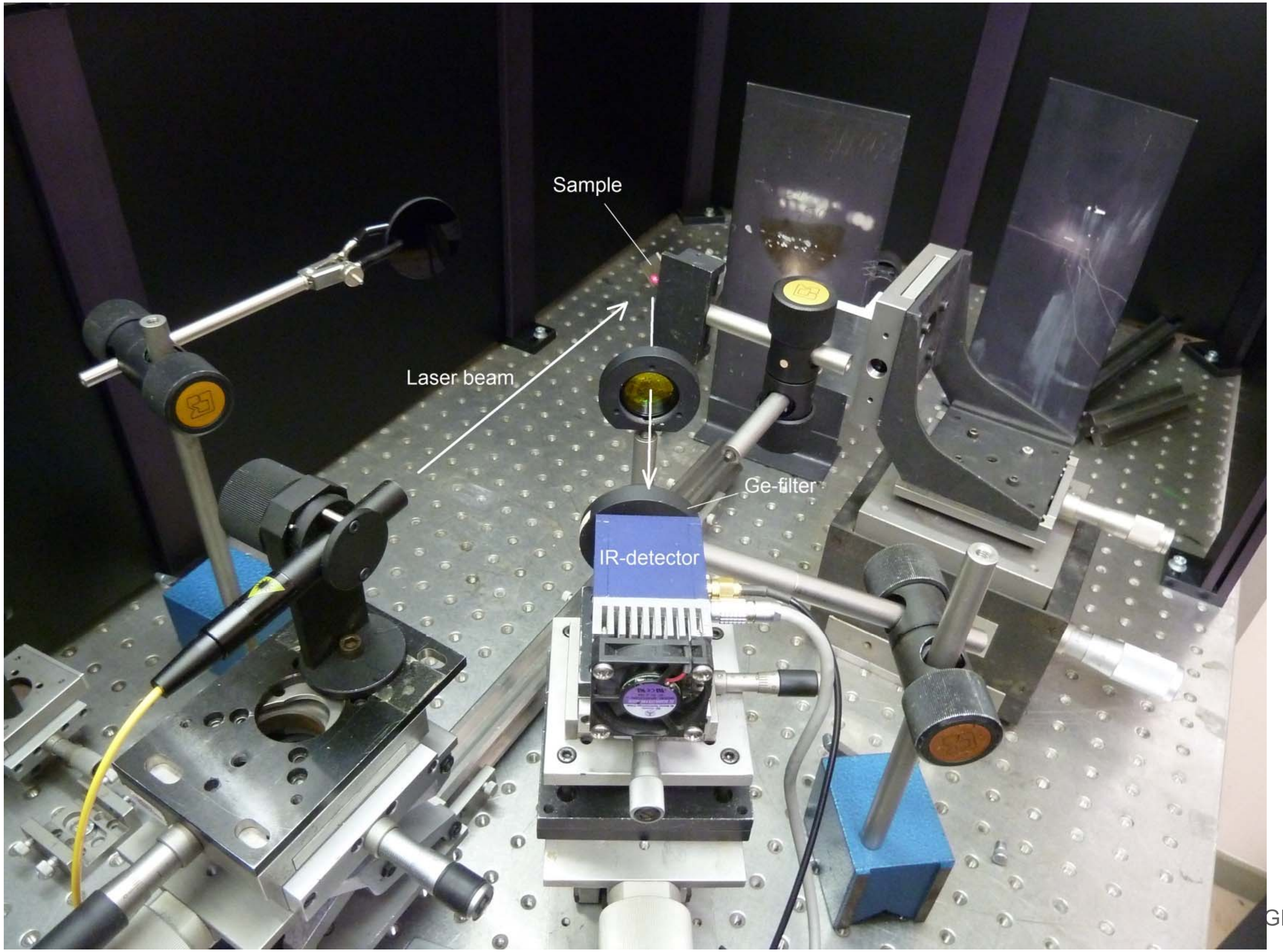


SCHÉMA OPTIQUE DE MESURE PONCTUELLE



3D+1 MODEL OF HEATING

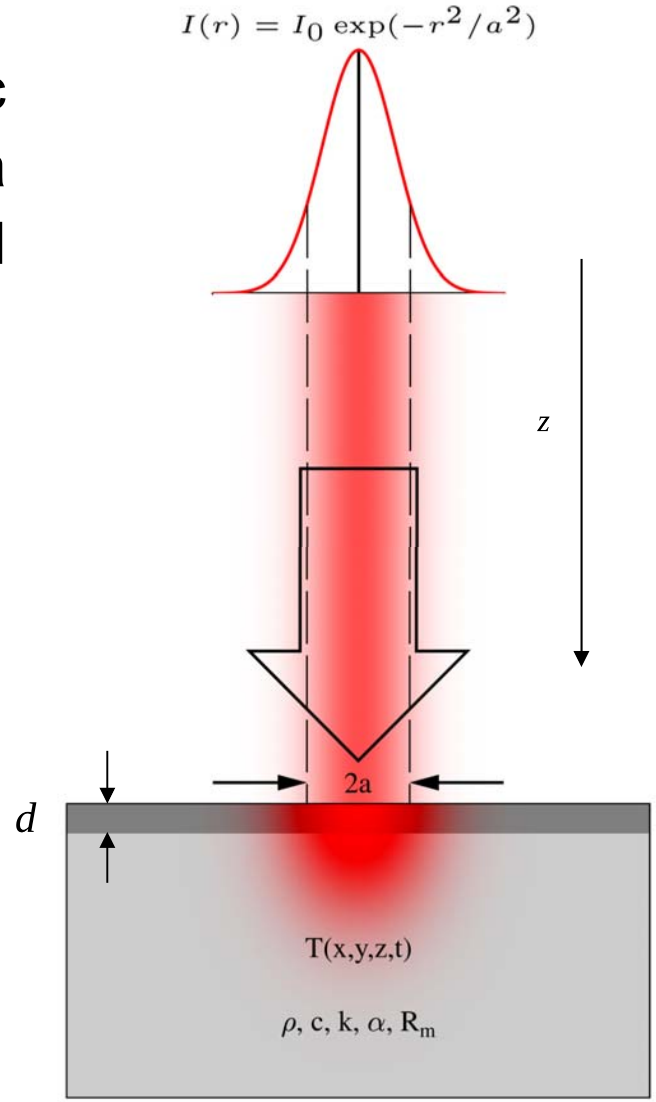
Heated surface comprises a micrometric (1 – 1000 µm) layer of one substance on a substrate with different thermo-physical and optical properties. Deposit adhesion.

Complex surfaces:

- friable deposited graphite layer on technical graphite substrate in the thermo-nuclear tokamak installations;
- oxide layer on metal
- deposit on mirror or window
- paint on a concrete, etc.

Laser beam:

- CW, single pulse or multi-pulses;
- *I((r)*, Gaussian or top-hat;
- Fixed or moving.



Rapid method of phase shift calculations

$$\Delta T_n(z=0,r=0)=\frac{\alpha_l(1-R_l)I_n}{k_l}\int_0^{+\infty}\frac{\tilde{\Phi}(\tilde{\xi})}{\tilde{\xi}^2/r_0^2-\alpha_l^2-2\pi i n\nu_L\rho_l C_l/k_l}$$
$$\times\left[\frac{2\alpha_l e^{-\kappa_{1n}d}}{\kappa_{1n}D}\left\{\left(e^{-\alpha_l d}-e^{-\kappa_{1n}d}\right)\left(1+\frac{k_s\kappa_{2n}}{h}\right)+\left(e^{-\kappa_{1n}d}-\frac{\kappa_{1n}}{\alpha_l}e^{-\alpha_l d}\right)\frac{k_s\kappa_{2n}}{k_l\kappa_{1n}}\right\}+\left(1-\frac{\alpha_l}{\kappa_{1n}}\right)\right]d\tilde{\xi}.$$

With $\tilde{\xi}=\xi r_0$, $\tilde{\Phi}(\tilde{\xi})=0.5\tilde{\xi}\exp(-\tilde{\xi}^2/4)$ for the Gaussian laser beam and

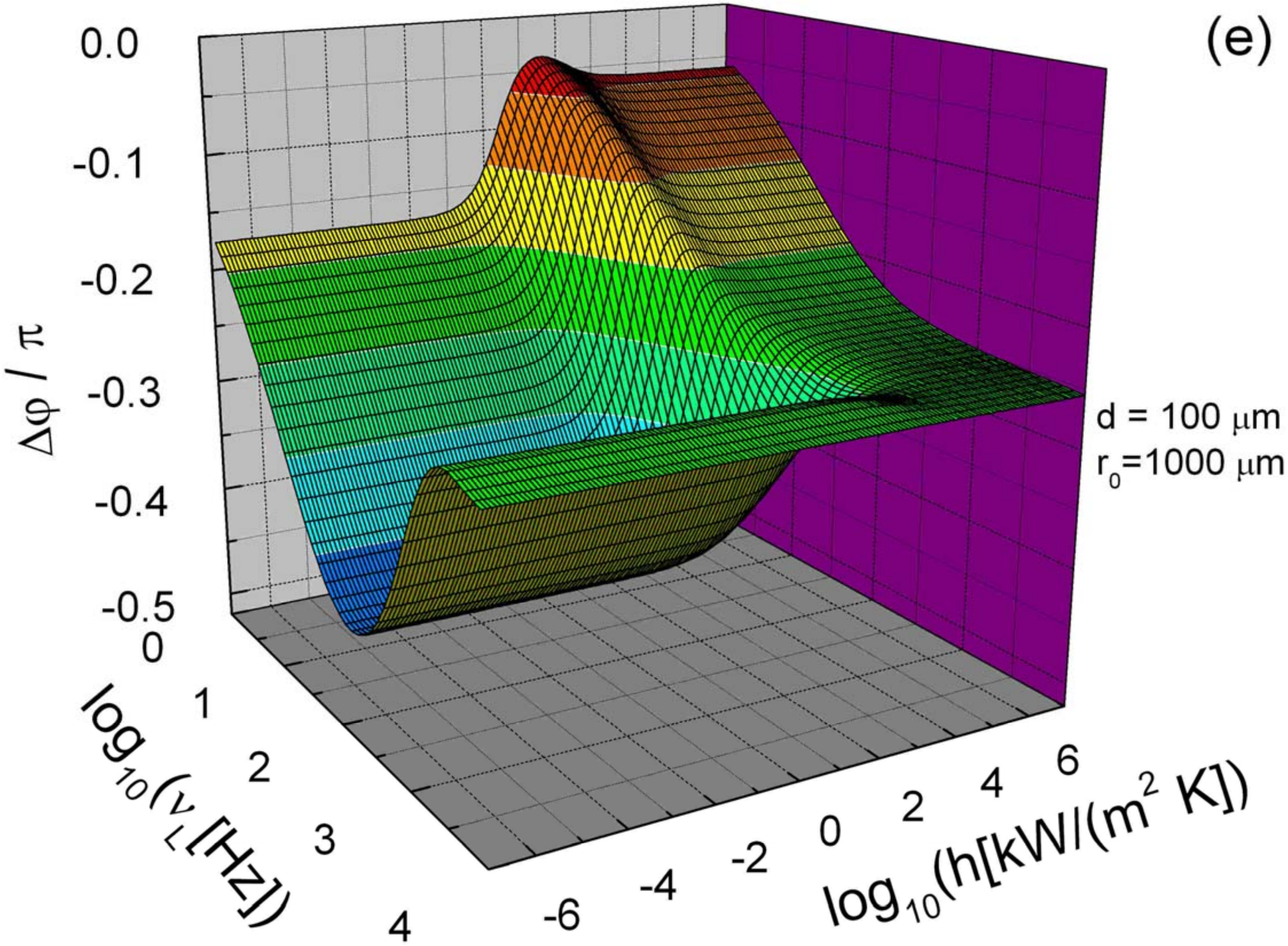
$$\kappa_{1n}=\sqrt{\tilde{\xi}^2/r_0^2-2\pi i n\nu_L\rho_l C_l/k_l}\qquad \kappa_{2n}=\sqrt{\tilde{\xi}^2/r_0^2-2\pi i n\nu_L\rho_s C_s/k_s}$$

$$D=\left(1+e^{-2\kappa_{1n}d}\right)\frac{k_s\kappa_{2n}}{k_l\kappa_{1n}}+\left(1-e^{-2\kappa_{1n}d}\right)\left(1+\frac{k_s\kappa_{2n}}{h}\right)$$

The Fourier contribution ΔT_1 corresponding to n = 1 (laser repetition rate frequency).

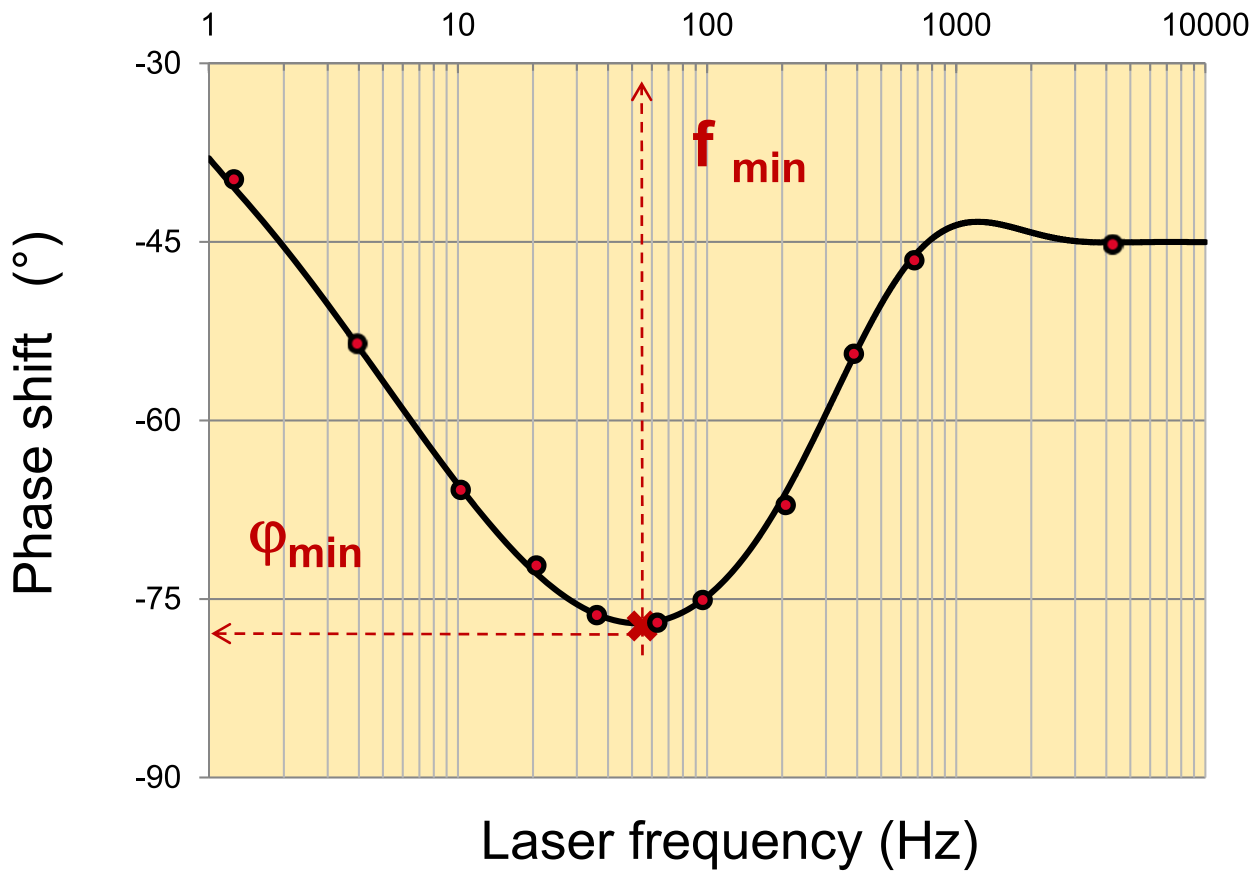
$$\Delta\varphi^{(1)}\equiv\varphi_{LH}^{(1)}-\varphi_{LP}^{(1)}=\mathrm{Re}\left\{i\ln\left(\Delta T_1(z=0,r=0)/\tilde{I}_1\right)\right\}$$

Rapid method of phase shift calculations



φ_{min} and f_{min} application

SS 304L plate (400 µm thickness)

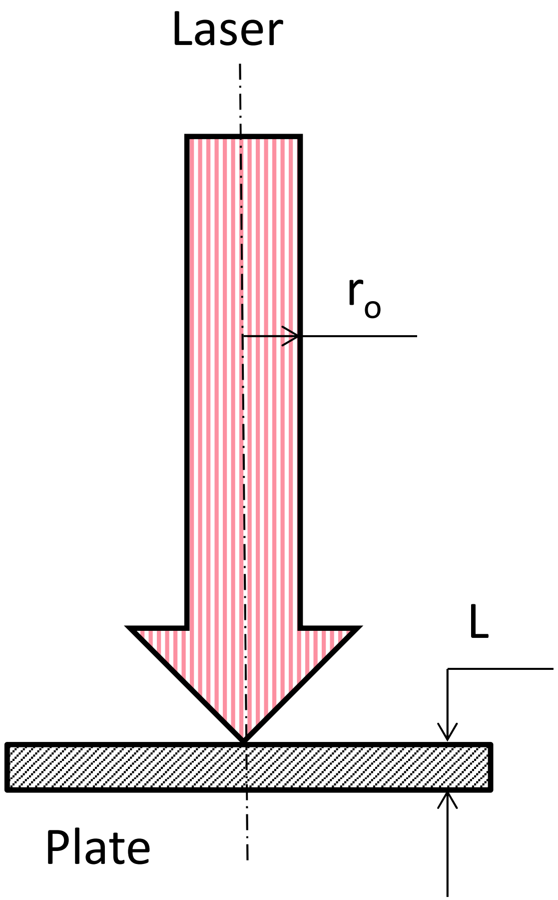


Simplified rapid measurements of sample thickness L and diffusivity D

$$\mathbf{L}\,[\mu m] \cong (3/2) \times \mathbf{r_o}\,[\mu m] \times \ln\,(90°/\varphi_{\mathbf{min}})$$

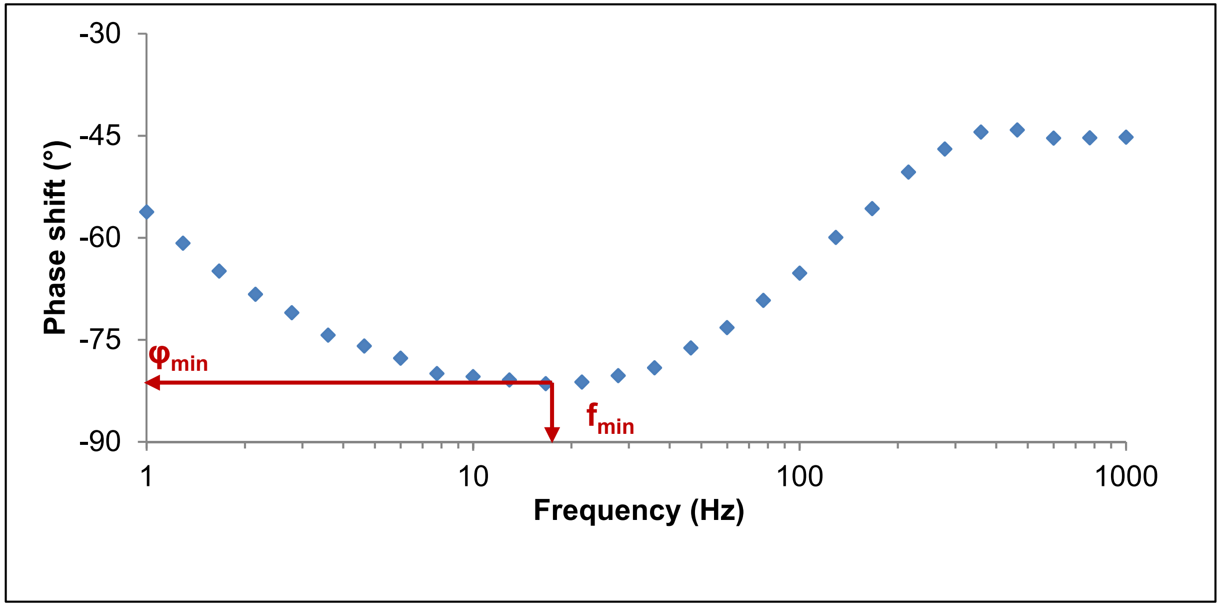
$$\mathbf{D}[\mu m^2/s] \cong 2 \times \mathbf{r_o}\,[\mu m] \times \mathbf{L}[\mu m] \times \mathbf{f_{min}}\,[Hz]$$

$$2 \le \frac{r_0}{L} \le 20$$



Two patents of the methods

Application for titanium plate

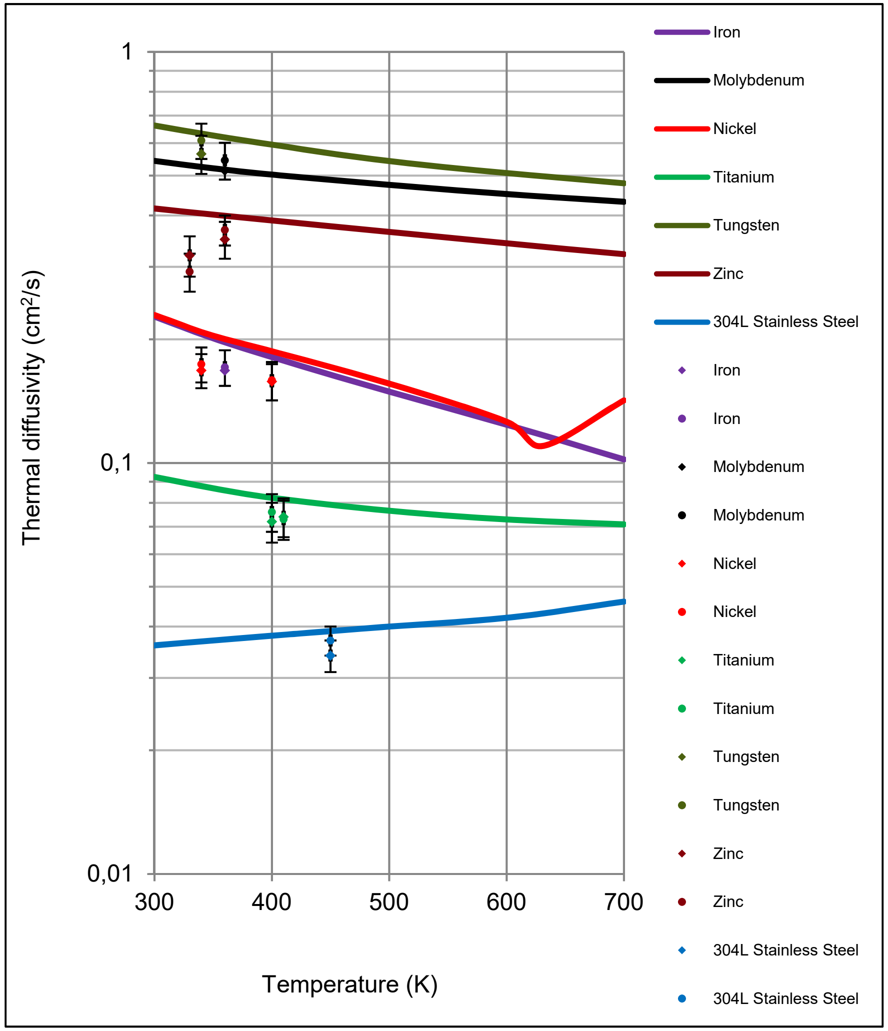


r_0 (µm)	φ_{min} (°)	L measured (µm)	L_{ref} (µm)	Deviation L_{mes} - L_{ref}	f_{min} (Hz)	D measured (cm^2/s)	D_{ref} (cm^2/s)	Deviation D_{mes} - D_{ref}
1740 ±30	- 81 ± 0.5	121 ± 7	127 ± 2.5	- 6 µm - 5%	18.5 ± 1.9	0.072 ± 0.009	0.082 ± 0.008	- 0.01 cm^2/s - 12%
		$L_{mes} = \frac{1}{\zeta_\varphi} r_0 \ln\left(\frac{90}{\lvert\Delta\varphi_{min}\rvert}\right)$				$D_{mes} = \frac{1}{\zeta_f} f_{min} r_0 L_{mes}$		

L_{ref} = 121±7 µm : measured plate thickness

D_{ref} : Y. S. Touloukian, R. W. Powell, C. Y. Ho, and M. C. Nicolaou, *Thermal Diffusivity* (IFI/Plenum, New York, 1973).

Comparison of the measured thermal diffusivities with the referenced values



Application to Zy4 cladding

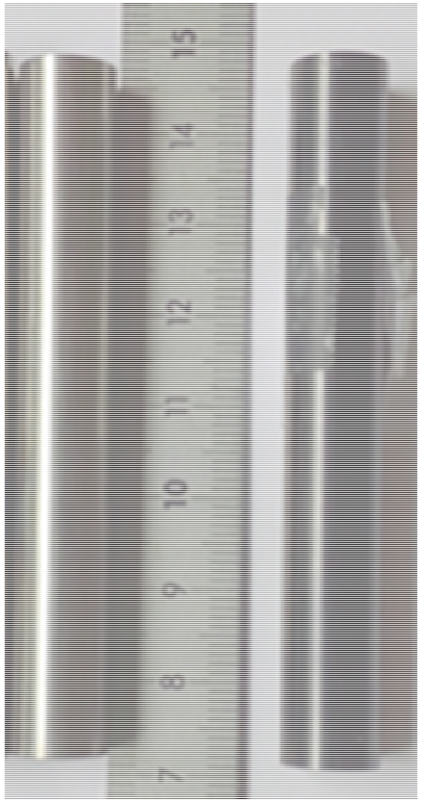
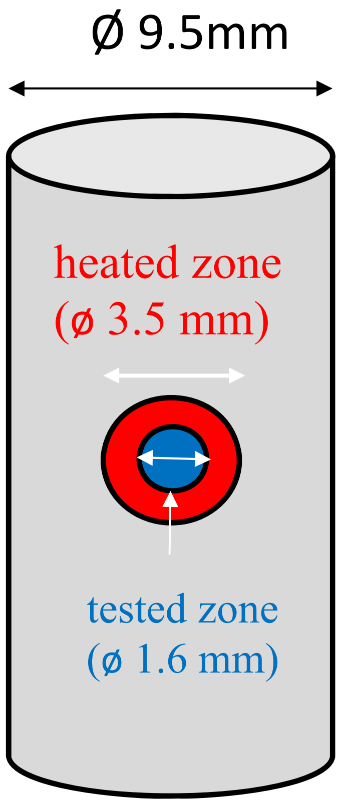
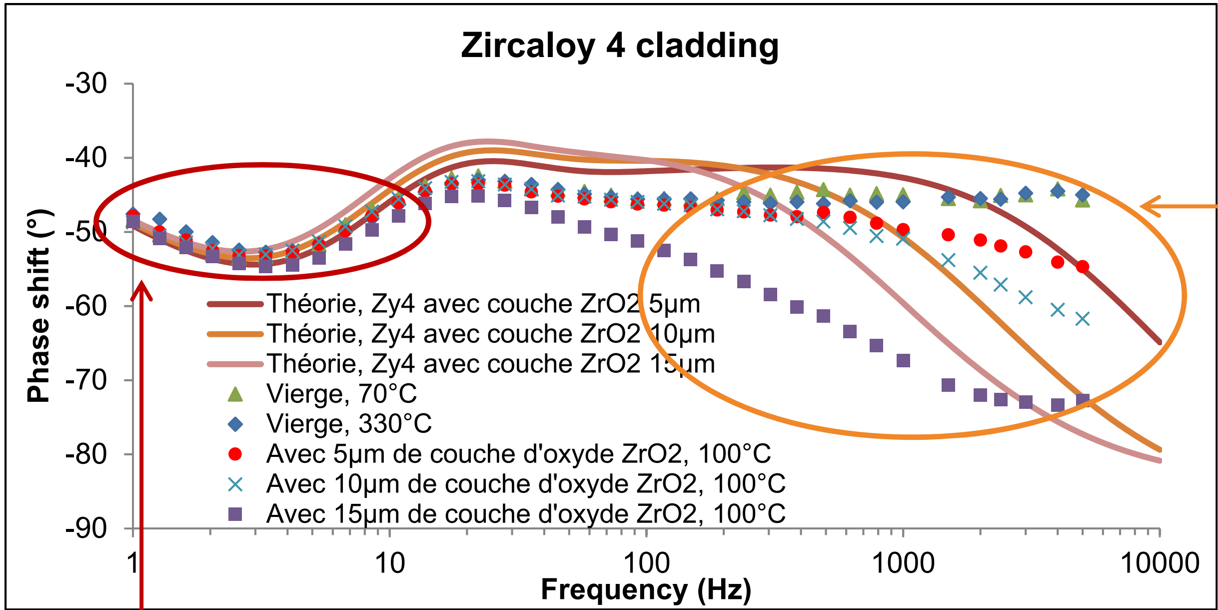


Photo of Zy4 claddings. Some Zy4 claddings were artificially oxidized.



Schematic comparison of the Zy4 cladding diameter and those of the heated zone (in red) and of the tested zone on it (in blue).

PERSPECTIVES



Important influence of oxyde layer

Determination of thermal propeties and thickness of the oxyde layer

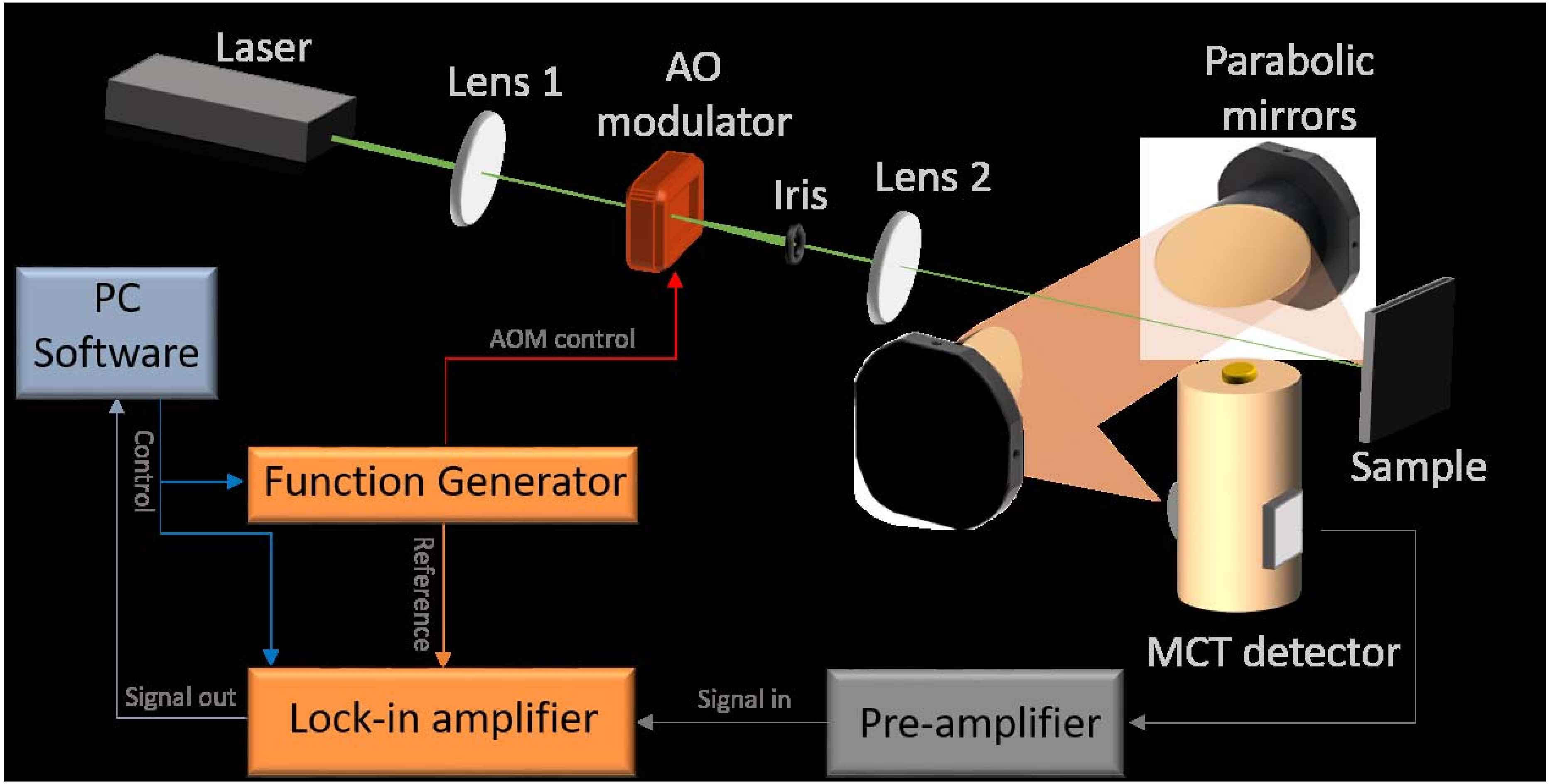
Theoretical model? Semi-transparente or transparente layer

Low dependence of the oxyde layer

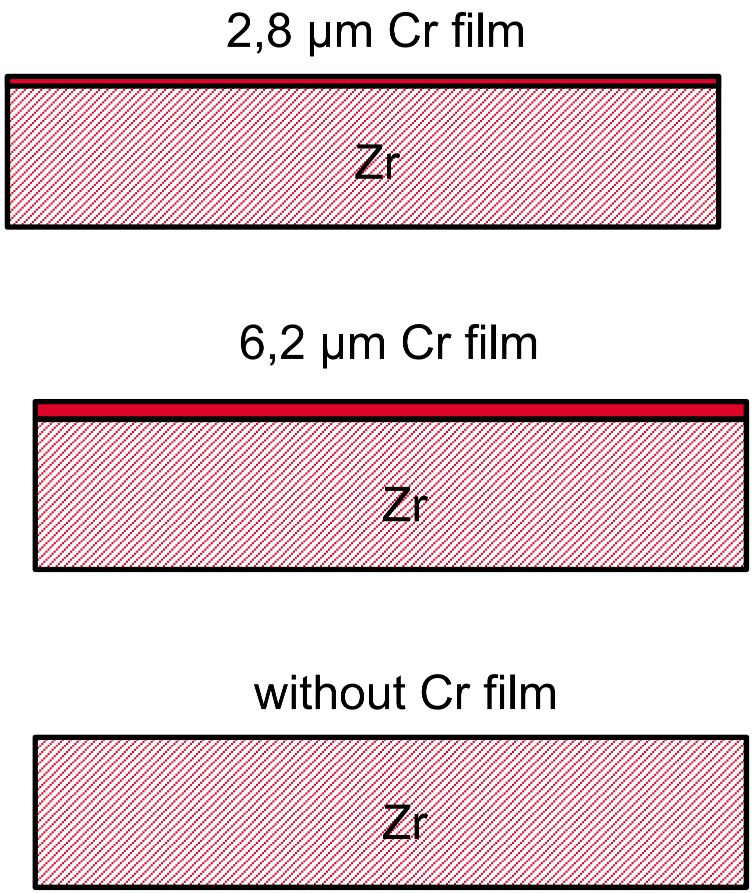
Determination of thickness and thermal diffusivity of the cladding

		Zy4 with 5µm oxyde	Zy4 with 10µm oxyde	Zy4 with 15µm oxyde	Reference (cm^2/s)	Deviation
Temperature		100°C	100°c	100°C		
Thickness	$L_{mes} = \frac{r_0}{\zeta_\varphi} ln\left(\frac{90}{\lvert\Delta\varphi_{min}\rvert}\right)$	594 µm	598 µm	564 µm	570 µm	de -6µm à +24µm
Thermal diffusivity	$D = \frac{1}{\zeta_f} f_{min} \cdot L \cdot r_0$	0.068 cm^2/s ± 0.006 cm^2/s	0.069 cm^2/s ± 0.006 cm^2/s	0.071 cm^2/s ± 0.006 cm^2/s	0.073 cm^2/s ± 0.003 cm^2/s	de -0.002 cm^2/s à -0.005 cm^2/s

Diagram of the experimental setup of high-frequency photothermal radiometry system 1



Zr/Cr Samples



Sample 1

Sample 2

Sample 3

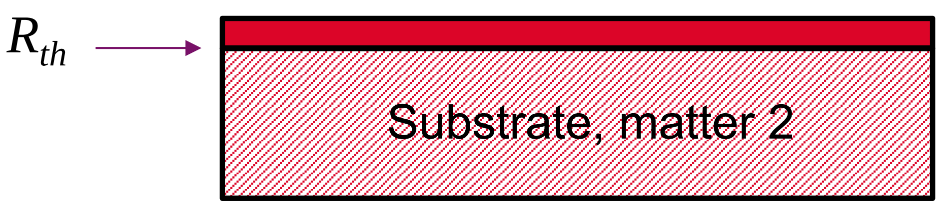
!!!
Surface rugosity for all samples is very high: ± 3 µm

!!!
Precision of film thickness? ± 3 µm?

A quadrupole method is used for the fitting of the experimental data.

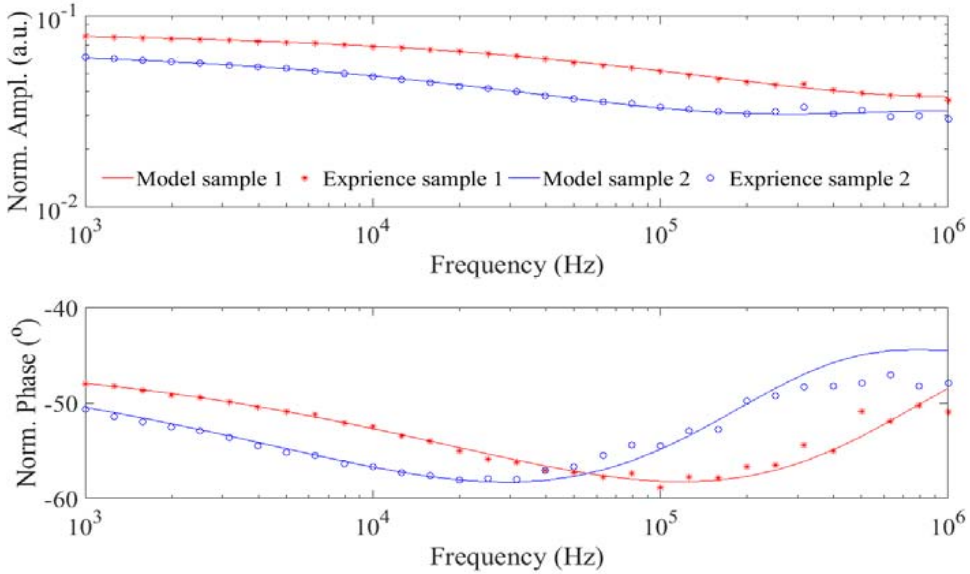
1D model for $T_{ac}(f)$ calculation:

$$T_{ac}(f) = \frac{(1-i)q}{2\sqrt{\pi f k_1 C_1}} \times \frac{1 - \dfrac{\sqrt{k_2C_2/k_1C_1}-(1+i)R_{th}\sqrt{\pi f k_2 C_2}-1}{\sqrt{k_2C_2/k_1C_1}+(1+i)R_{th}\sqrt{\pi f k_2 C_2}+1}\, e^{-2(1+i)\sqrt{\pi f C_1/k_1}d}}{1 + \dfrac{\sqrt{k_2C_2/k_1C_1}-(1+i)R_{th}\sqrt{\pi f k_2 C_2}-1}{\sqrt{k_2C_2/k_1C_1}+(1+i)R_{th}\sqrt{\pi f k_2 C_2}+1}\, e^{-2(1+i)\sqrt{\pi f C_1/k_1}d}},$$



d - film thickness, matter 1

Normalized amplitude and phase of both sample using quadrupole theoretical model blue curves : L = 6.2 µm; red curves : L = 2.8 µm



Using the photothermal signal emitted from the samples it is then possible to find with good uncertainties **$(L / \sqrt{a})_{film}$** and **e_{film}**.

L is the thickness the film, ***a*** and ***e*** the thermal diffusivity and effusivity of the film respectively.

$R_{th} \approx 10^{-8}\, m^2\, K\, W^{-1}$ - the thermal boundary resistance (TBR) between the two materials (Kapitza resistance).

Results

From these results it is possible to fit out the $(L/\sqrt{a})_{film}$ and e_{film} by minimizing the model to fit well the experimental points.

	$(L/\sqrt{a})_{film}$ $[s^{1/2}]$	e_{film} $[J K^{-1} m^{-2} s^{-1/2}]$	e_{film} literature $[J K^{-1} m^{-2} s^{-1/2}]$	a_{film} Literature $[m^2 s^{-1}]$	$L\ [\mu m]$
Sample 1	$6.7\ 10^{-4} \pm 3.9\%$	$1.59\ 10^{4} \pm 9.8\%$	$1.67\ 10^{4}$	$27.4\ 10^{-6}$	$3.5 \pm 12\%$
Sample 2	$1.28\ 10^{-3} \pm 5.3\%$	$1.63\ 10^{4} \pm 9.9\%$			$6.7 \pm 22\%$

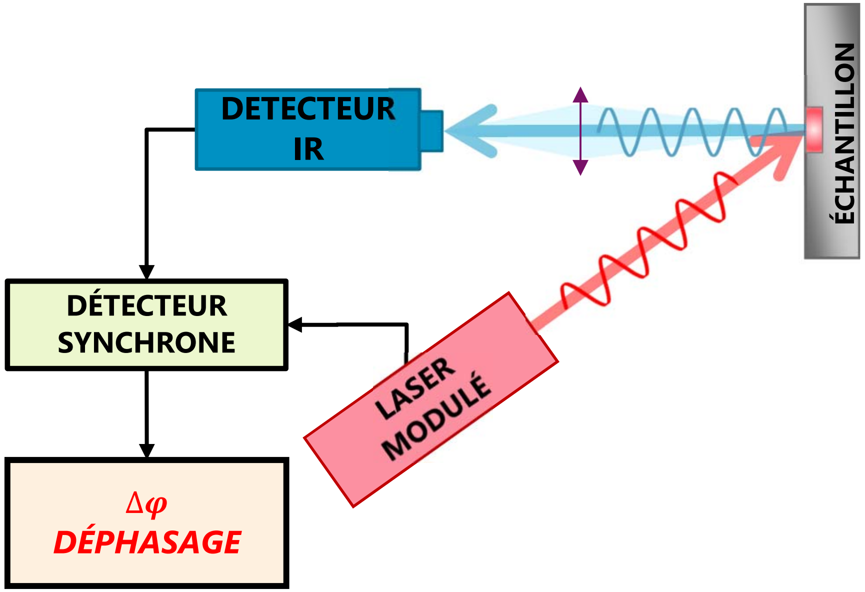
Taking into consideration their respective errors, these values are close to the given ones, **2.8** ***µm*** for sample 1 and **6.2** ***µm*** for sample 2.

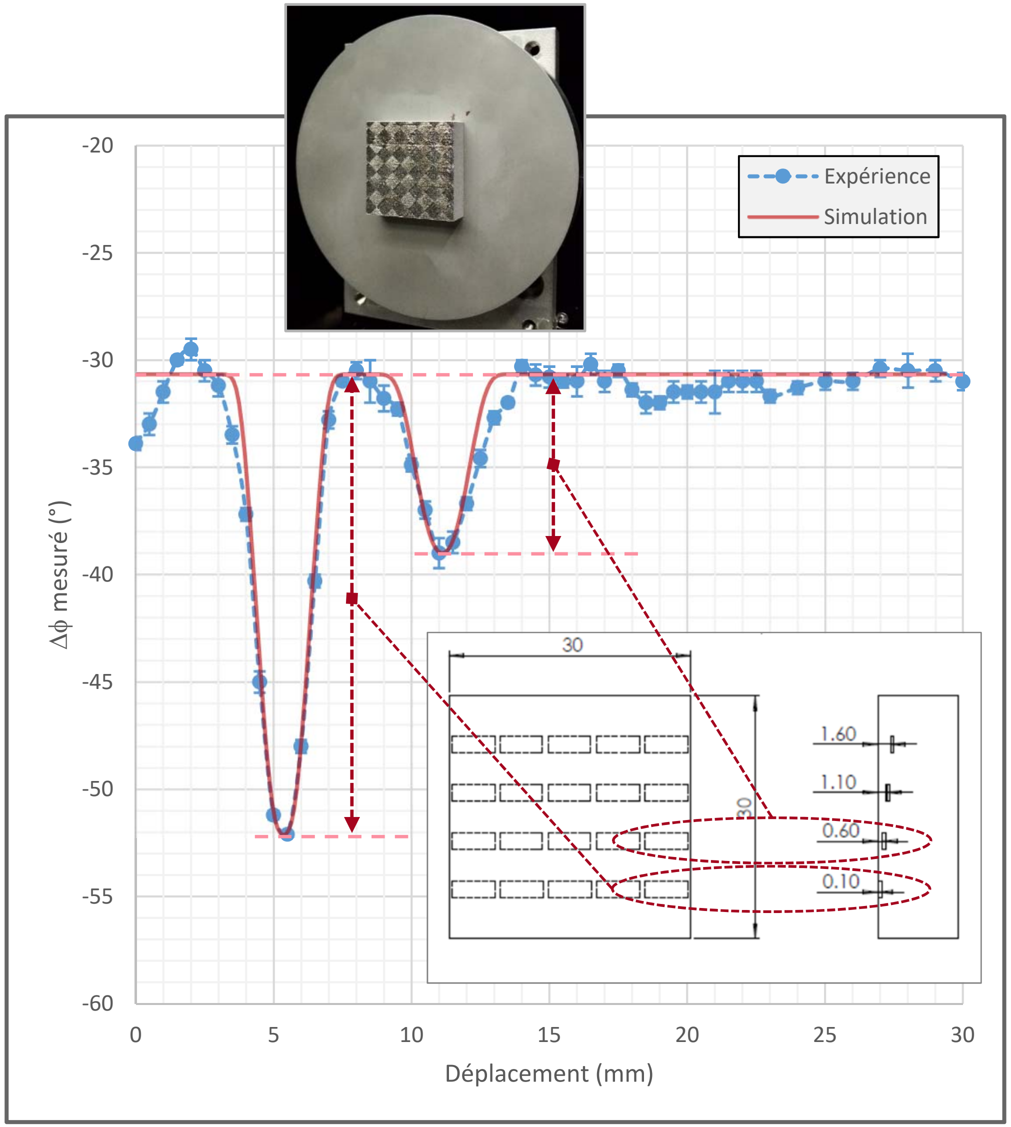
Conclusion: by using the PTR measurements it was possible to find the thickness of a coating of Cr over Zr using the only assumption that heat capacity of the Cr film is equal to the one of the bulk from literature.

Possible improvement: high Cr reflectivity on 532 nm $\rightarrow \lambda$ or carbon nm layer,

Perspective: R_{th} the thermal boundary resistance (TBR) measurement between the two materials;

DÉFAUTS: CAVITÉS SOUS LA SURFACE

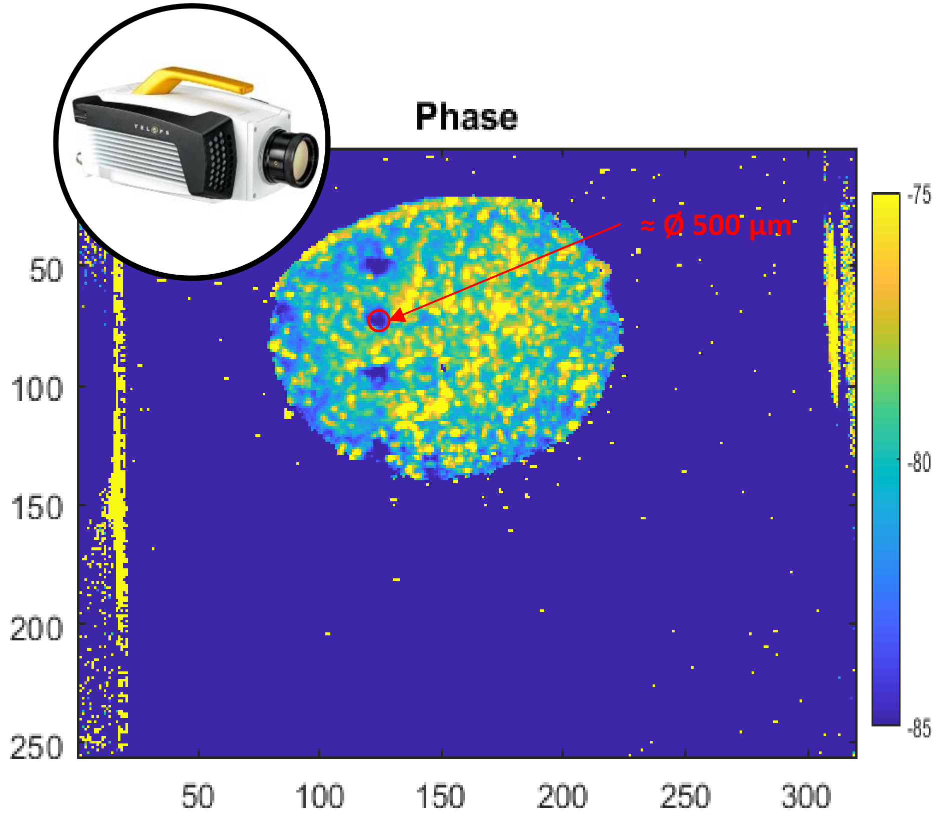


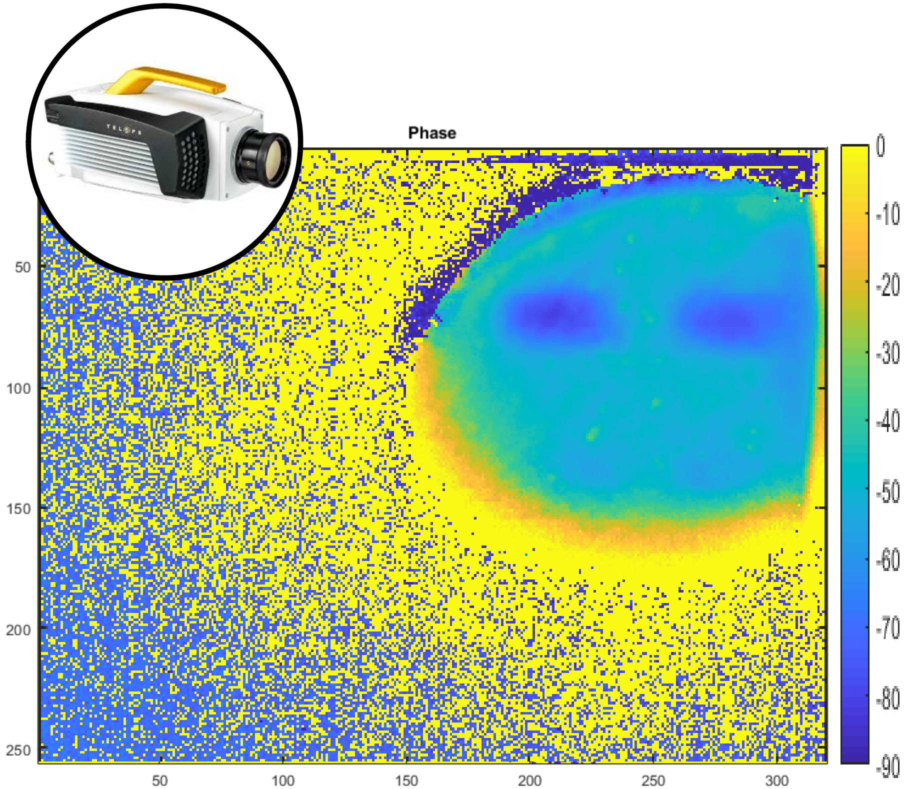


- La valeur du déphasage mesurée dépend de l'épaisseur du matériau
- En déplaçant le faisceau sur l'échantillon, on observe un changement de valeur du déphasage

► Détection d'une cavité sous la surface

CAMÉRAS FLIR & TELOPS: RÉSULTATS PROMETTEURS





Conclusions

Results:

- **New laser methods for materials characterization (h, L and D) with advantages:**
  - o **contactless**
  - o **remote**
  - o **fully-optical**
  - o **non-destructive**

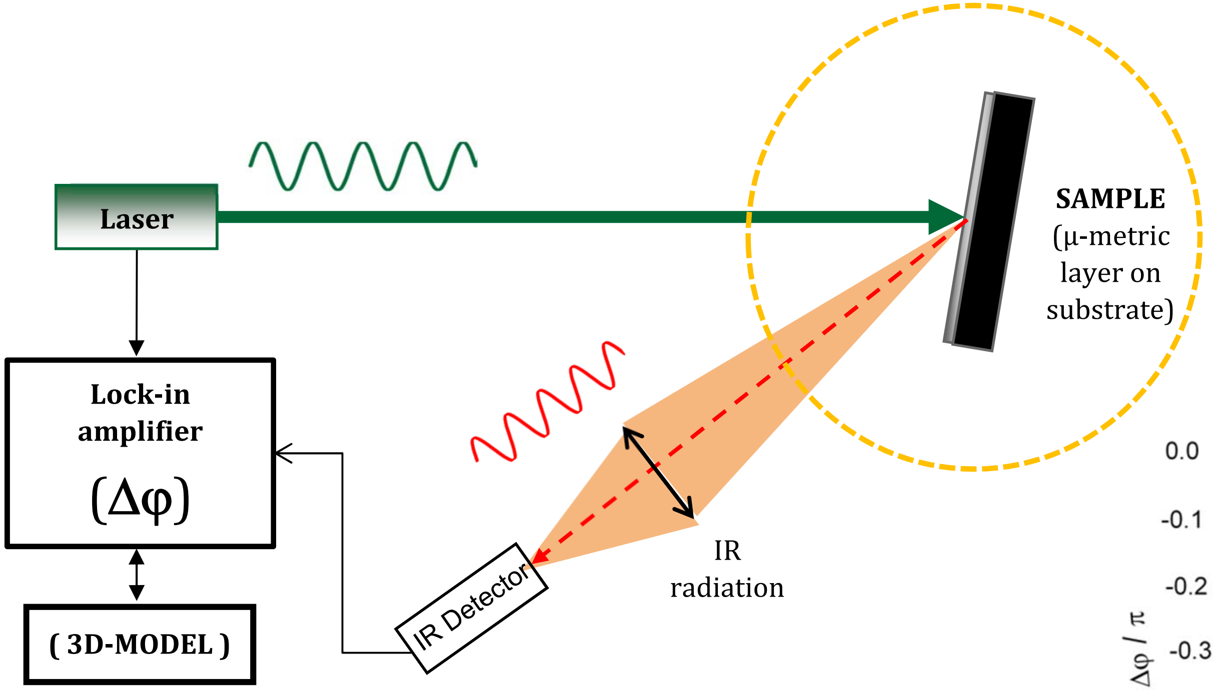
- **Methods validation with a set of samples (metal plates and Zy4 cladding);**

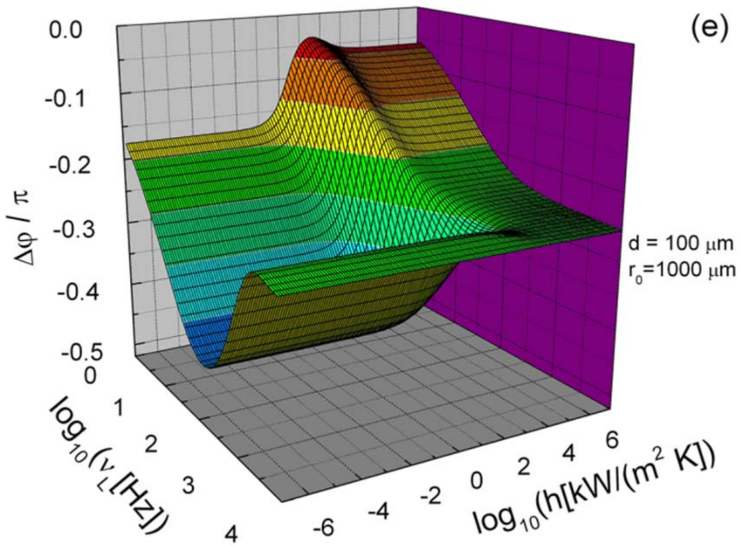
Perspectives:

- **Experimental setup and method simplification (PC code, portable device);**
- **Method application extension for thin layer (<1µm) and multilayer surfaces (oxides on metals);**
- **Method validation on nuclear sites;**
- **In situ control of nuclear construction elements for safety improvement of nuclear installations.**

Lock-in thermography

Heating at different laser repetition rate frequencies: ν_L = (2 – 1000) Hz





thickness, thermal resistance, conductivity and capacity

Lock-in thermography advantages:

- Fully-optical, contactless method
- non-destructive remote measurements;
- no effect of surface reflectivity;
- independence on laser power instability;

Publications, reports and patents

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Thank you for your attention!

Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 65 57

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ABSTRACT

In the search for new materials for nuclear industry and reliable nuclear safe technologies, much attention is given to methods for non-destructive remote testing and control of materials in severe environment of nuclear installations. Photothermal radiometry, as one of these possible methods, is under development in the CEA (French Atomic Energy Commission). In our studies, it was used with repetition rate laser heating of a sample by either a pulsed (100 ns-1 ms, 1-10000 Hz) [1, 2] or a sinusoidal modulated laser power (1 Hz - 1 MHz) [2,3] followed by measurements of the thermal radiation emitted by the sample under study. Thermal radiation time-dependence or phase shifts between the laser power and the thermal radiation measured at different modulated frequencies (lock-in radiometry) were then compared with those obtained with a model of laser heating to characterize some sample properties (thermal diffusivity, thickness, layer adhesion, under-surface defects and their changes in time).

Samples from European TOKAMAKS (graphite or carbon fibre composite protective tiles with 1 - 100 µm deposits) and nuclear installations (metal pieces, Zircaloy cladding, micrometric oxide layers or PVD thin films, 3D-printed metal objects) were tested and characterized with the home-made experimental setups and the developed analytical (3D+t) models of laser heating [4-6]. The numerical simulation of laser heating was used to fit the calculated thermal radiation or phase shifts with the experimental ones by adjusting the material properties. This method of characterization was validated with a set of etalon samples. The applied methods will be presented along with the obtained results. The possible ways to improve the method to widen the scope of its application in the nuclear industry will be discussed as well.

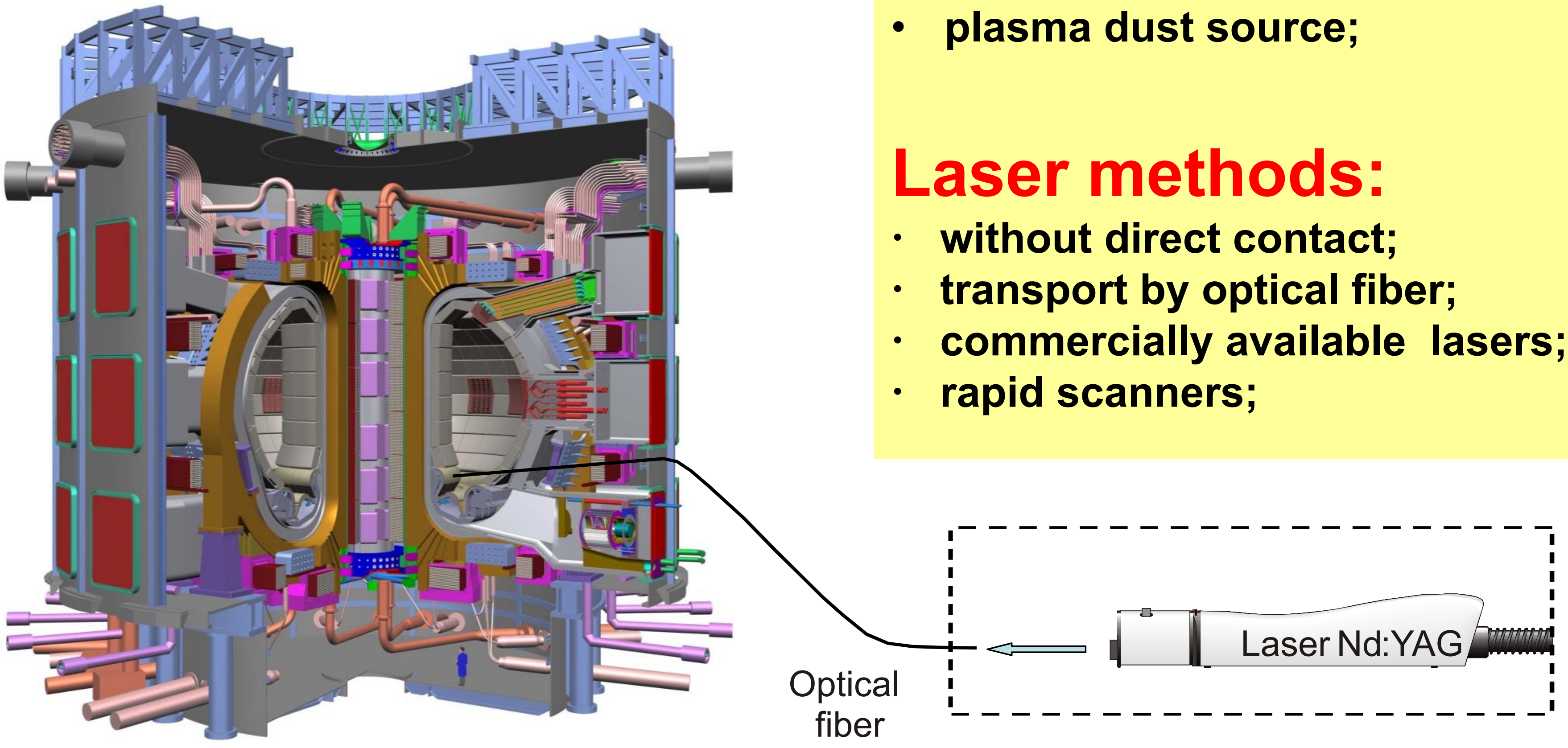
INTRODUCTION

Problems:

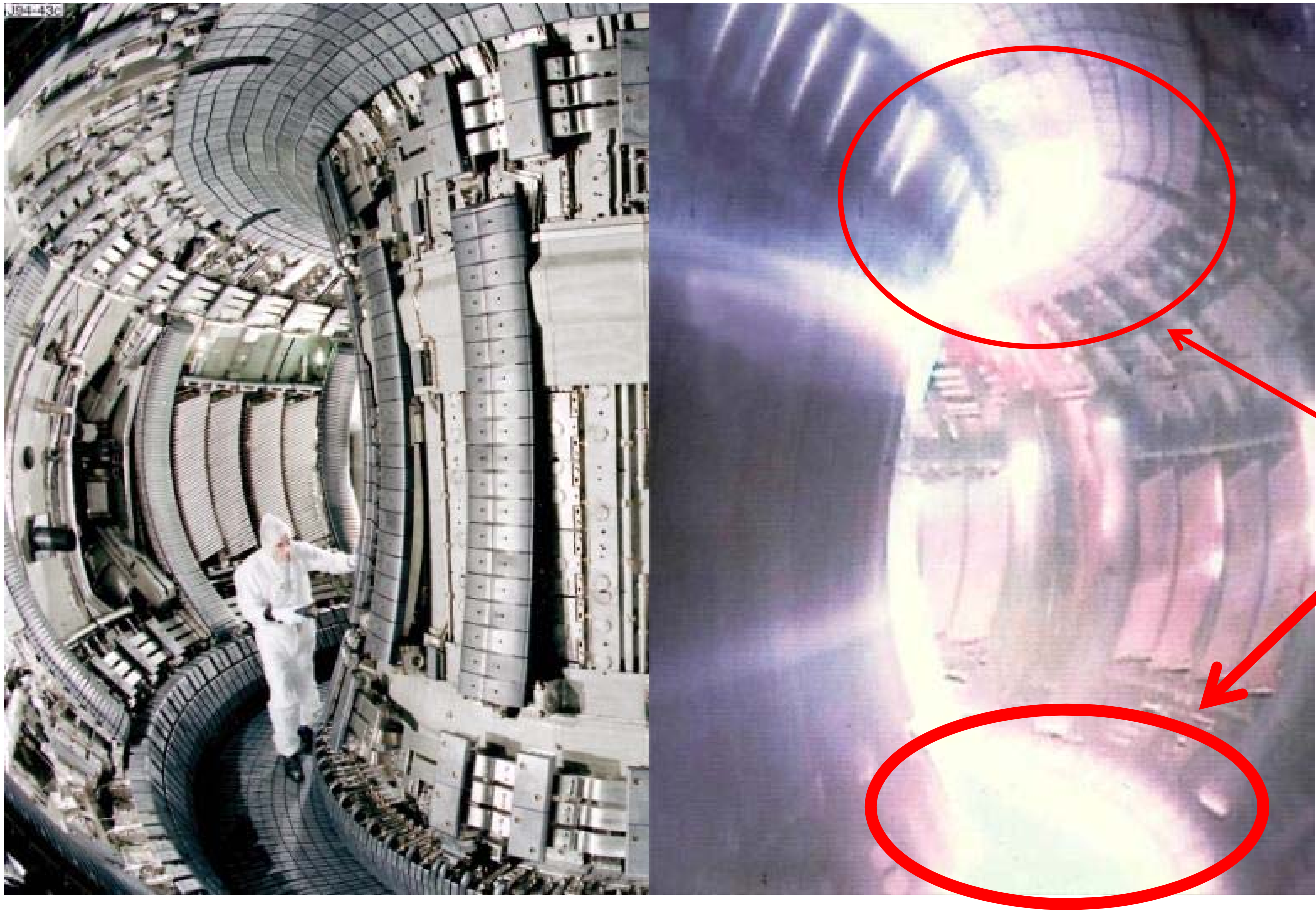
- deposited layers with tritium;
- plasma dust source;

Laser methods:

- without direct contact;
- transport by optical fiber;
- commercially available lasers;
- rapid scanners;

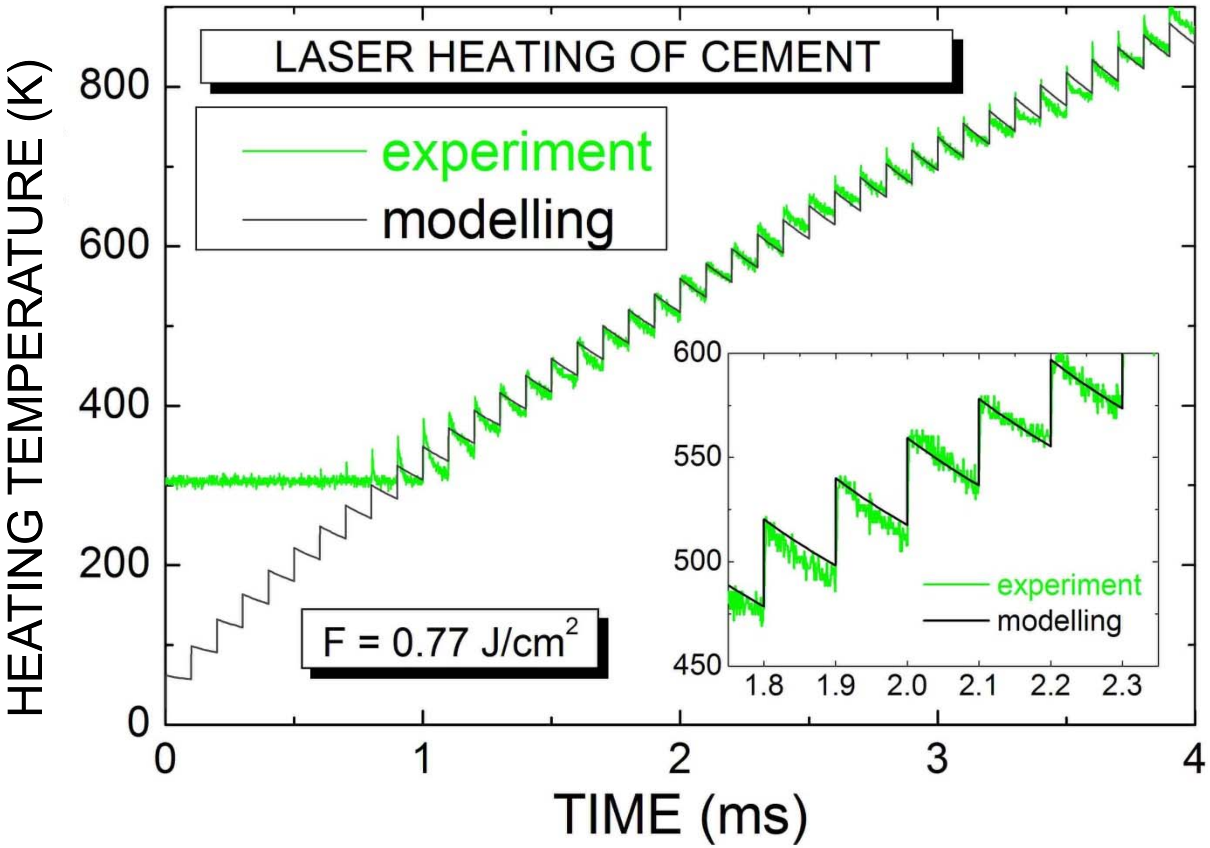


European tokamak JET, Culham, GB



Places of the plasma wall interactions

VOLUME HEATING OF CONCRETE



Simulation parameters: R = 0.38, D = 0.0123 cm^2/s, k = 2.7 W/m·K, α = 2.2$\times$10^4 m^{-1}

Simplified rapid measurements of sample thickness L and diffusivity D

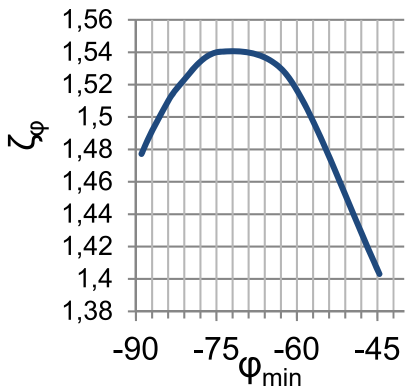
$$\mathbf{L}\,[\mu m] \cong (3/2) \times \mathbf{r_o}\,[\mu m] \times \ln(90°/\boldsymbol{\varphi}_{\mathbf{min}})$$

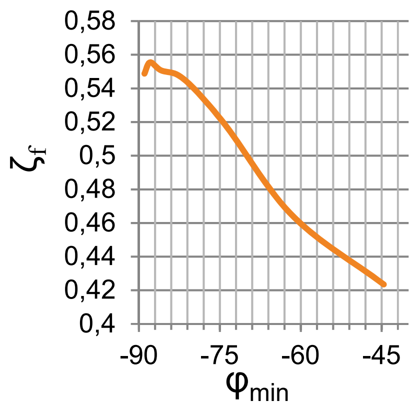
$$\mathbf{D}[\mu m^2/s] \cong 2 \times \mathbf{r_o}\,[\mu m] \times \mathbf{L}[\mu m] \times \mathbf{f_{min}}\,[Hz]$$

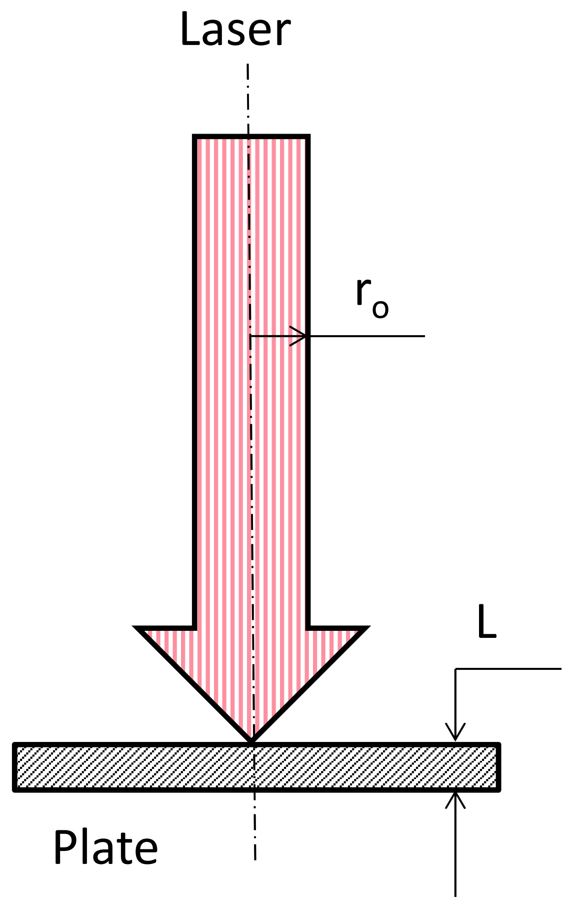
$$2 \le \frac{r_0}{L} \le 20$$

$$\mathbf{L}\,[\mu m] = (\mathbf{r_o}\,[\mu m]/\zeta_\varphi) \times \ln(90°/\boldsymbol{\varphi}_{\mathbf{min}})$$

$$\mathbf{D}[\mu m^2/s] = (1/\zeta_f) \times \mathbf{r_o}\,[\mu m] \times \mathbf{L}[\mu m] \times \mathbf{f_{min}}\,[Hz]$$

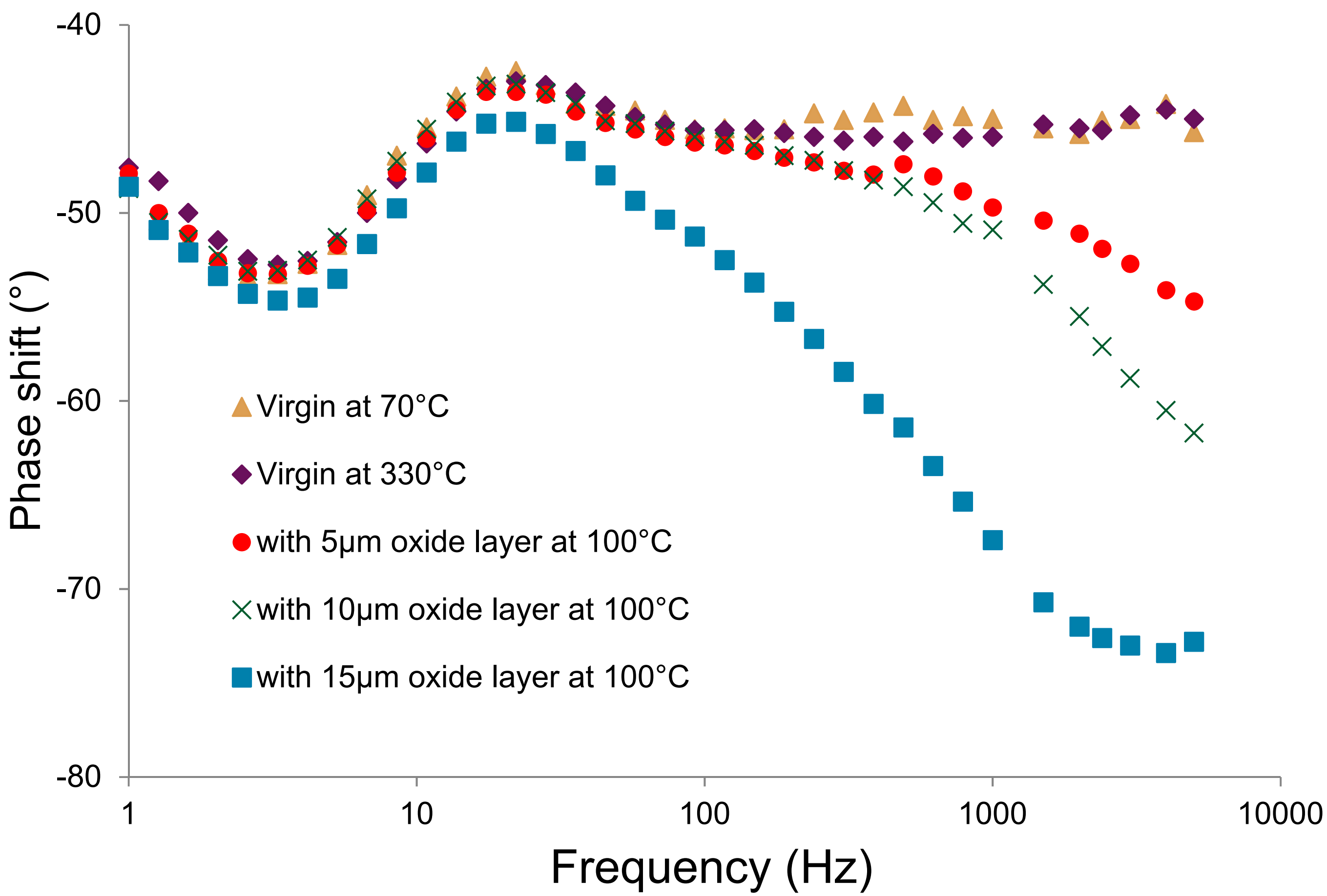






Two patents of the methods

Zy4 cladding

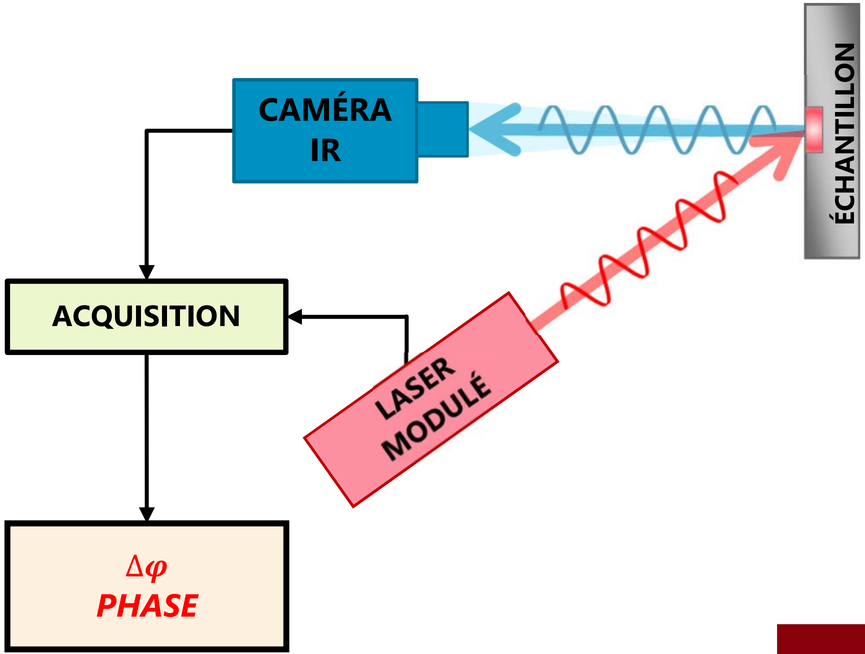


Zircaloy-4 claddings with oxide layers

Oxide thickness	µm	5	10	15
Laser mean power	W	2.7		
Temperature	°C	100±5		
D_{ref} reference	cm^2/s	**0.073 ±0.003**		
L_{ref} reference	µm	**570 ± 2.5**		
r_0 measured	µm	1740±30		
φ_{min} measured	°	-53.3 ±0.5	-53.1±0.5	-54.7±0.5
f_{min} measured	Hz	3.1± 0.3	3.1±0.3	3.4±0.3
L_m measured	µm	**594±15**	**598±15**	**564±15**
D_{m1} measured with L_m	cm^2/s	**0.068±0.006**	**0.069±0.006**	**0.071±0.006**
D_{m2} measured with L_{ref}	cm^2/s	**0.066 ± 0.006**	**0.066 ± 0.006**	**0.071 ±.006**

GRESPI team, 05 February 2018, Reims

TEST AVEC 2 SOCIÉTÉS: FLIR & TELOPS



Spécifications	FLIR X6900sc	TELOPS M2k
Gamme spectrale	1,5 à 5,0 µm	1,5 à 5,5 µm
Résolution	640 x 512	320 x 256
Pas du détecteur	25 µm	30 µm
Sensibilité thermique/NETD	< 20 mK	25 mK
Fréquence d'acquisition (max)	1004 Hz	1910 Hz
Ouverture de l'objectif (f)	F/2.5 ou F/4.1	F/2.5