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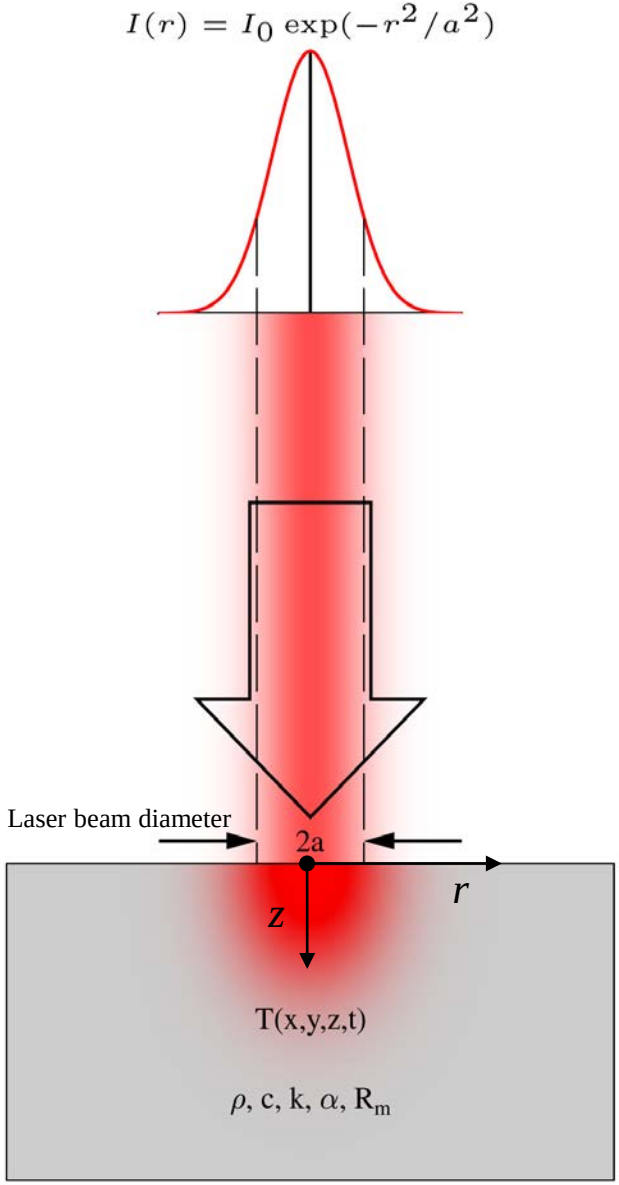
MULTI-PARAMETRIC MODELING OF SOLID SAMPLE HEATING BY NANOSECOND LASER PULSES FOR NANO-ABLATION

A. Semerok[1], C. Jabbour[1], S.V. Fomichev[2,3], J.-L. Lacour[1], M. Tabarant[1], F. Chartier[1]

[1] DEN-Service d'Etudes Analytiques et de Réactivité des Surfaces, 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



BASIC SCHEME AND EQUATIONS FOR ns-LASER HEATING



$$c(T)\frac{\partial T}{\partial t} = div\left(k(T)\nabla T\right) + Q(t,r,z)$$

$$Q(t,r,z) = I(t,r)A\alpha\exp(-\alpha z)$$

$$T(t=0,r,z) = T_o \quad \text{and} \quad \left.\frac{\partial T(t,r,z)}{\partial z}\right|_{z=0} = 0$$

c (J m^{-3} K^{-1}) – thermal capacity;
k (W m^{-1} K^{-1}) – thermal conductivity;
α (m^{-1}) – absorption coefficient of media;
A – surface absorptivity at laser wavelength;

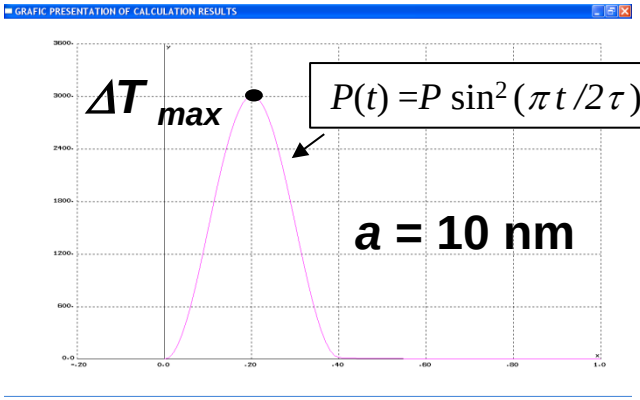
I (W/cm^2) $\rightarrow$ T (K) = f (t, r, z)
for $a \in$ [100 µm $\rightarrow$ 10 nm]

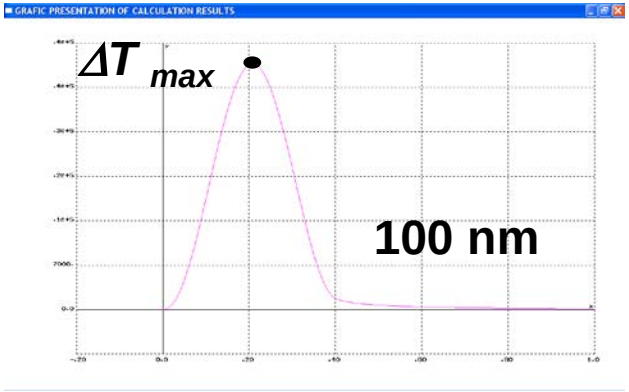
[*] A. Semerok, S. V. Fomichev, J.-M. Weulersse, F. Brygo, and P.-Y. Thro, Heating and ablation of tokamak graphite by pulsed nanosecond Nd-YAG lasers, JOURNAL OF APPLIED PHYSICS **101**, 084916, 2007.

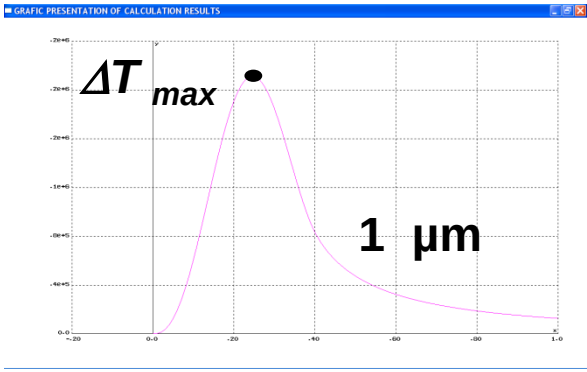
“3D+t” HEATING CODE, Si -bulk

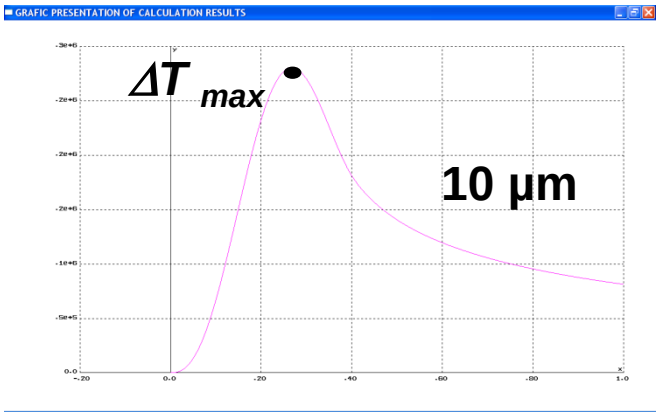
$$\Delta T\,(t, r = 0, z = 0) = \mathrm{f}\,(t)$$

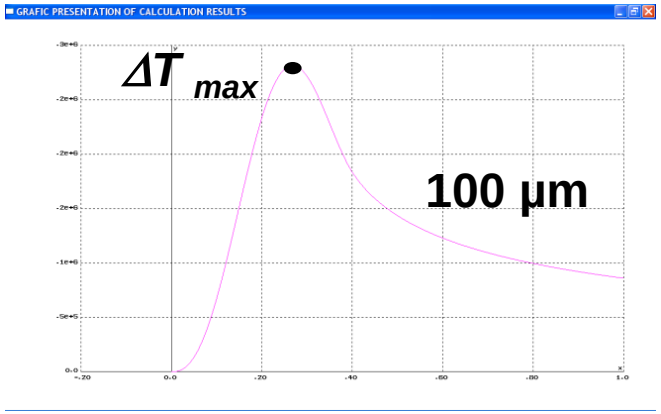
Si-bulk: c = 700 J/kg K, k = 149 W/m K, ρ = 2330 kg/m^3, α = 200 μm^{-1}, A = 0.3.
Laser: Gaussian F(r)=F_o exp -(r^2/a^2), $P(t)$ =P sin^2 ($\pi t/2\tau$) - pulse of τ = 4 ns, λ = 266 nm, F = 100 J/cm^2;





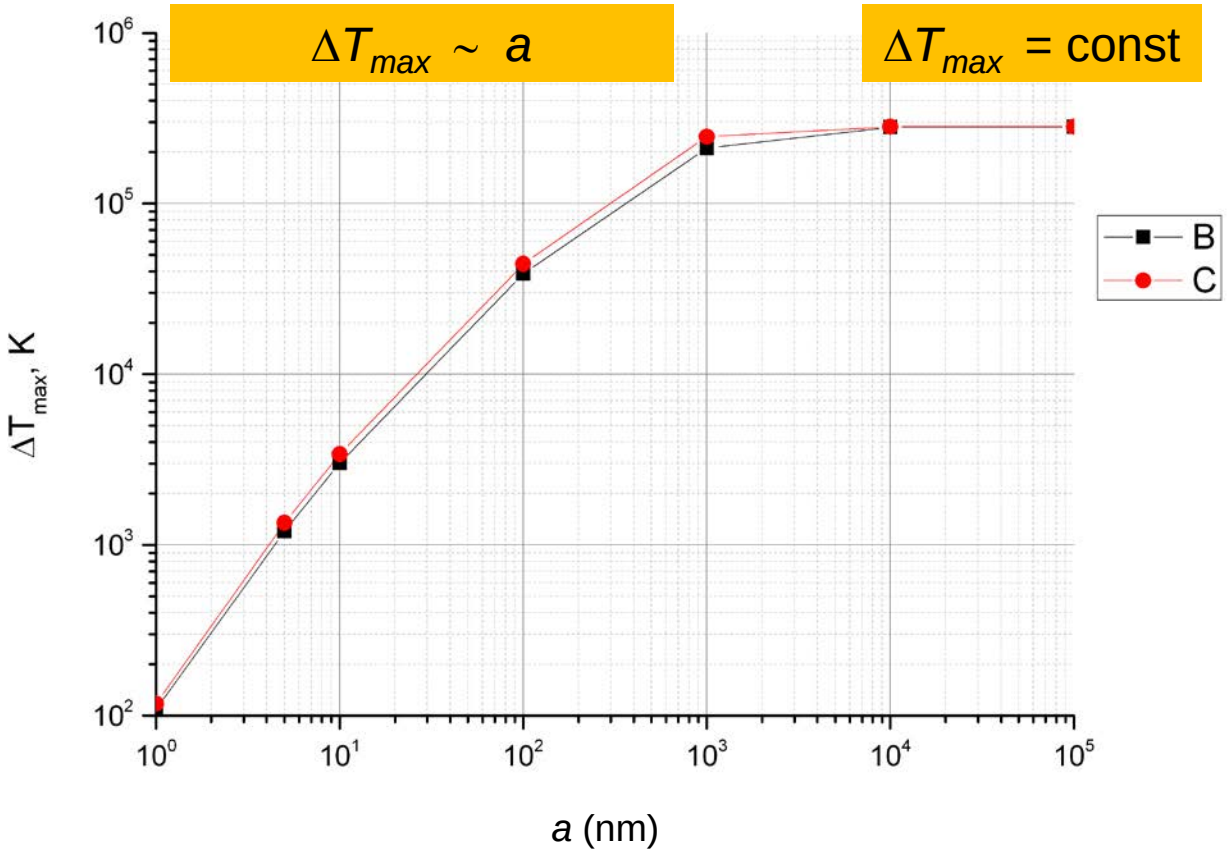






Total duration of calculation with time following laser pulse corresponding to 1 on x-axis is 20 ns.

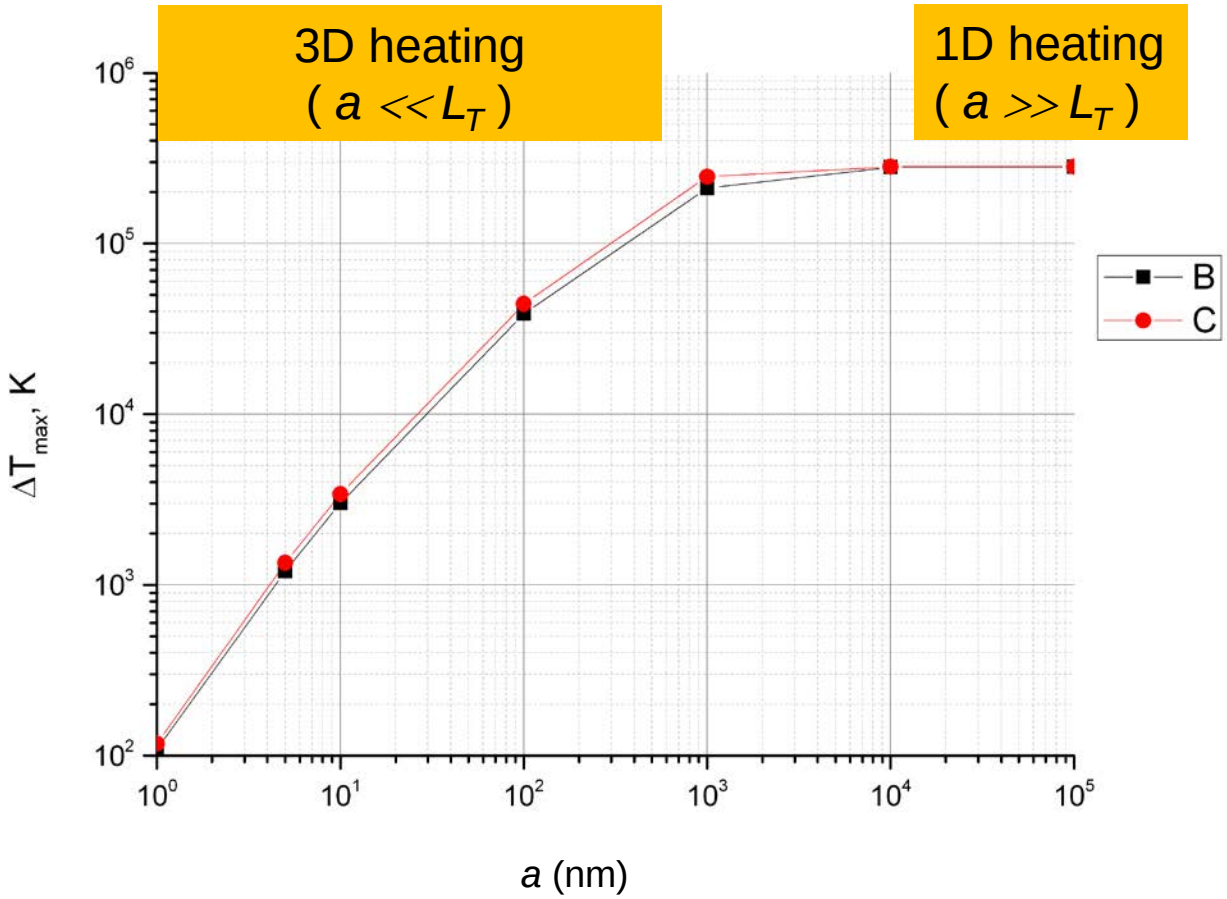
Heating temperature ΔT_{max} = f(a) for given F =100 J/cm^2



B - Gaussian
C - Top-Hat

Heating temperature ΔT_{max} = f(a) for given F =100 J/cm^2

$$Heating\ length:\ \boldsymbol{L_T} = [4\times \boldsymbol{D}(\mathrm{m}^2/\mathrm{s})\times \boldsymbol{\tau}(\mathrm{s})]^{0.5}$$



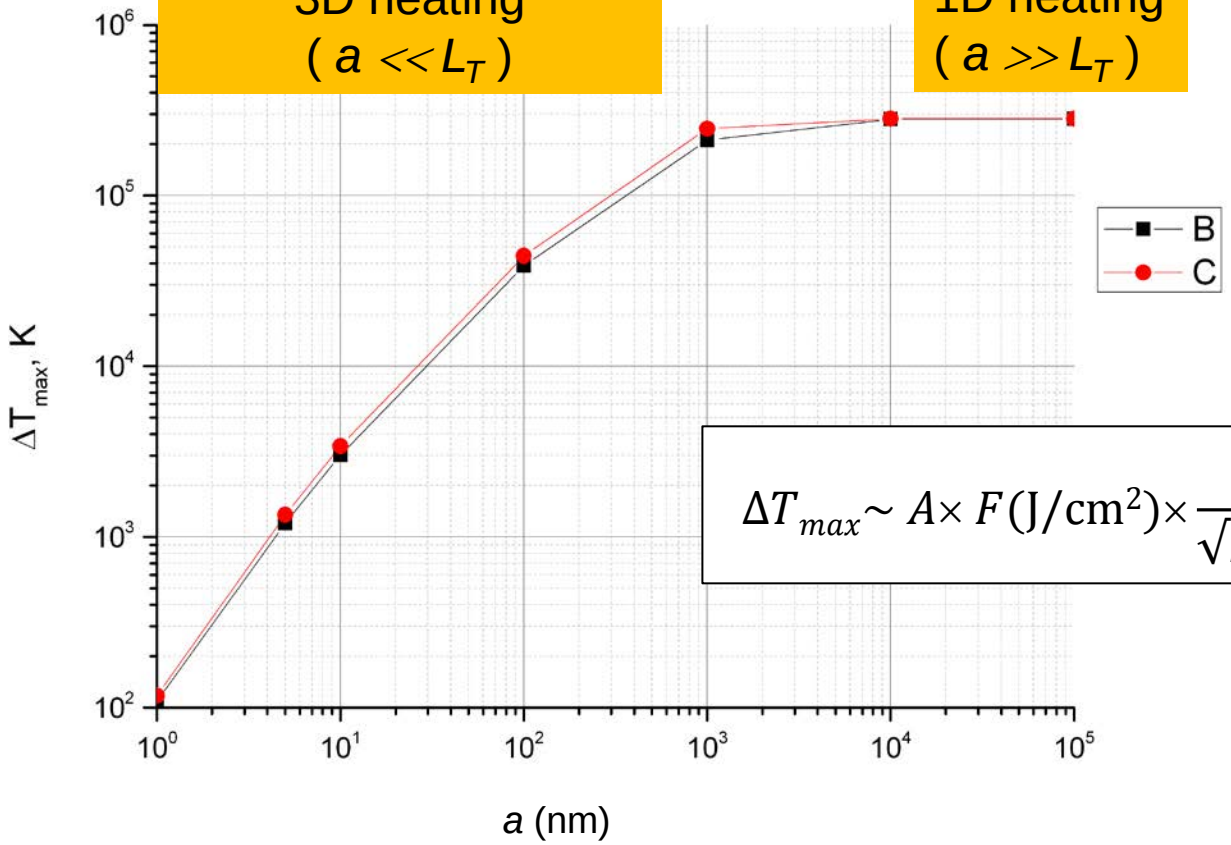
B - Gaussian
C - Top-Hat

Analytical solutions in linear approximation

$$\Delta T_{max} \sim A \times F(\mathrm{J/cm^2}) \times \frac{a(\mathrm{nm})}{k(\mathrm{W/mK}) \times \tau(\mathrm{s})}$$

3D heating
($a << L_T$)

1D heating
($a >> L_T$)

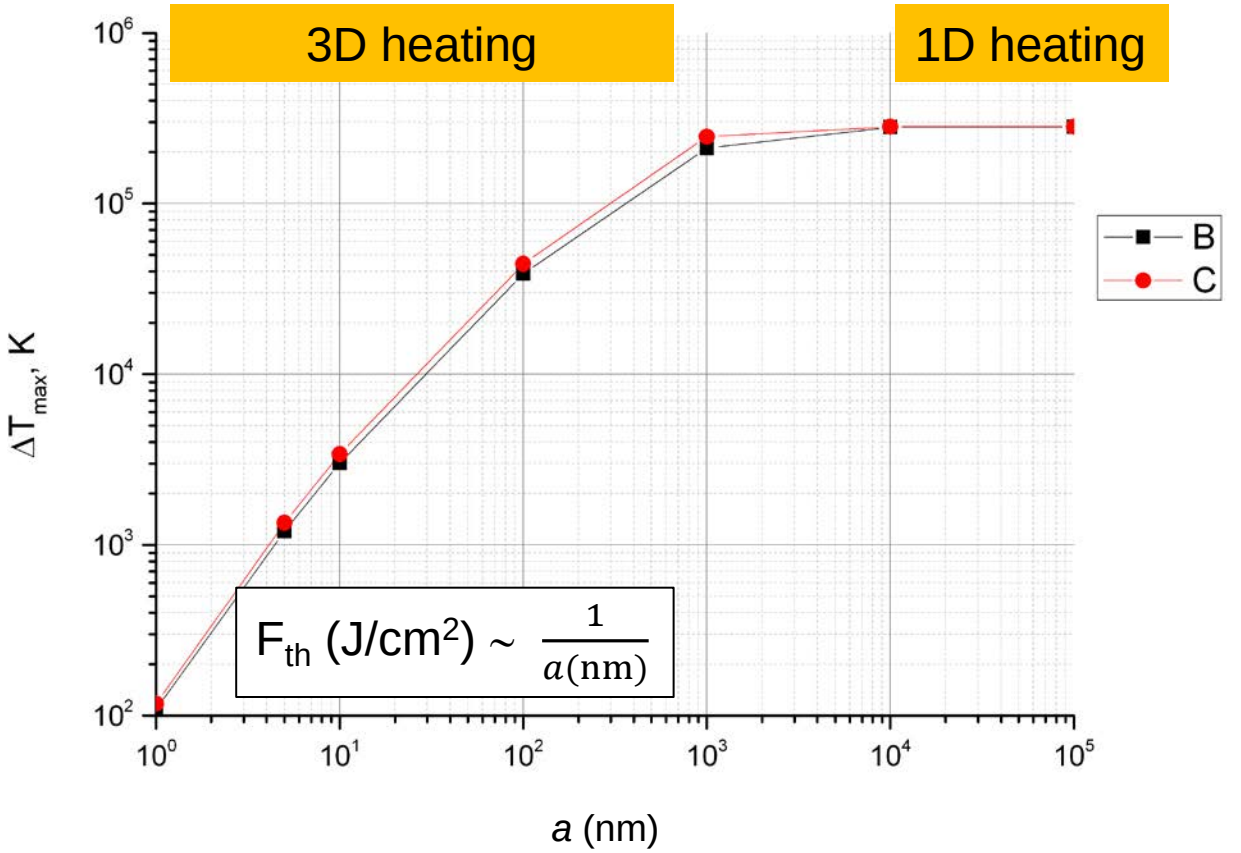


B - Gaussian
C - Top-Hat

$$\Delta T_{max} \sim A \times F(\mathrm{J/cm^2}) \times \frac{\sqrt{\tau(\mathrm{s})}}{\sqrt{k(\mathrm{W/mK})} \times \sqrt{C(\mathrm{J/m^3})}}$$

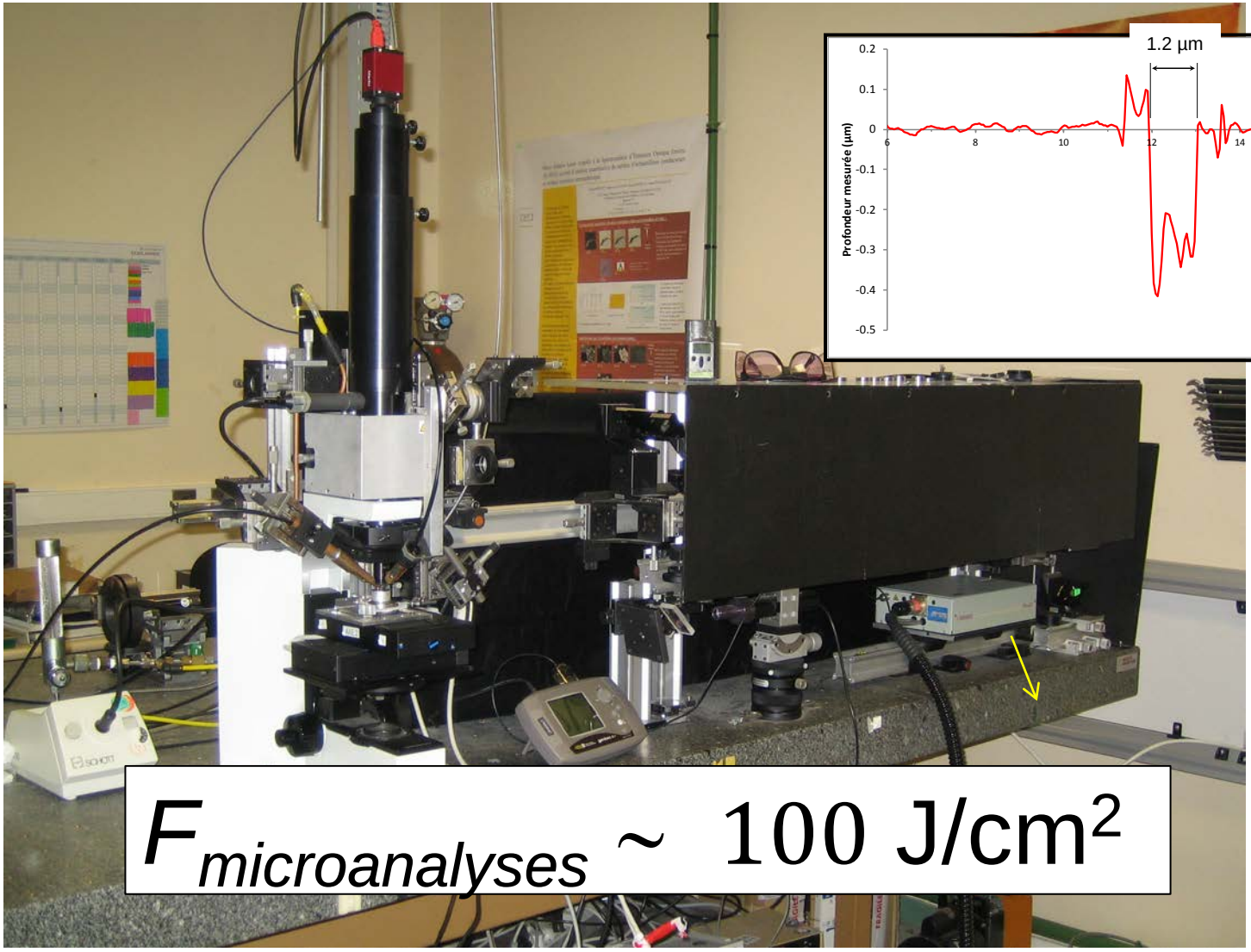
«Ablation» threshold F_{th} (J/cm²)

F_{th} (J/cm²) ≠ f(*a*)



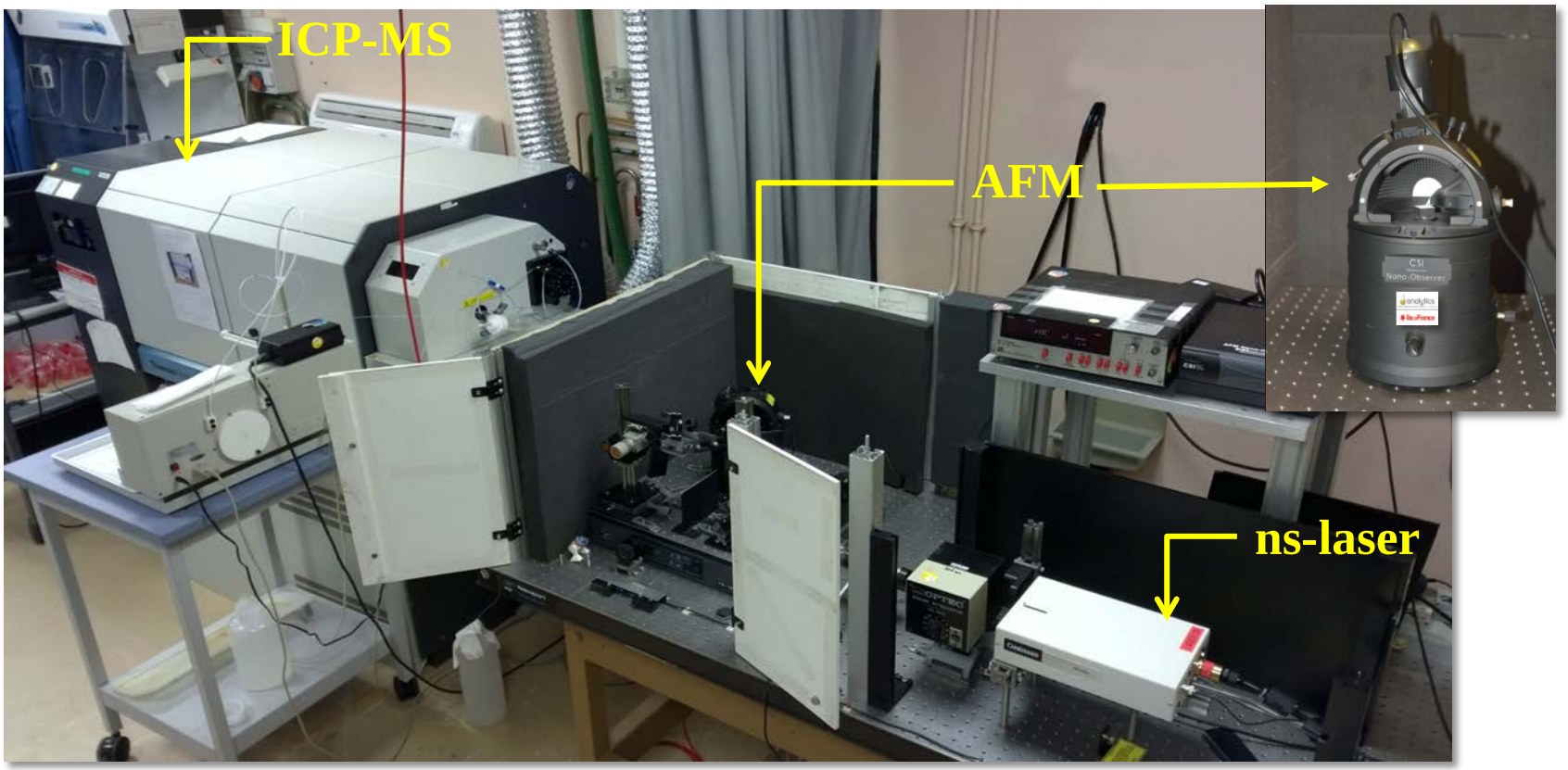
B - Gaussian
C - Top-Hat

CEA Saclay micro-LIBS instrument



[**] D. Menut, P. Fichet, J.-L. Lacour, A. Rivoallan, and P. Mauchien, Micro-laser-induced breakdown spectroscopy technique: a powerful method for performing quantitative surface mapping on conductive and nonconductive samples, APPLIED OPTICS **42**, 2003, p. 6063

CEA Saclay near-field LA-ICP-MS installation

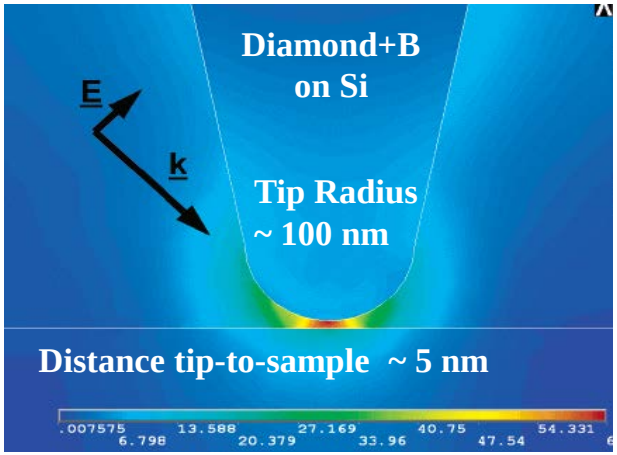


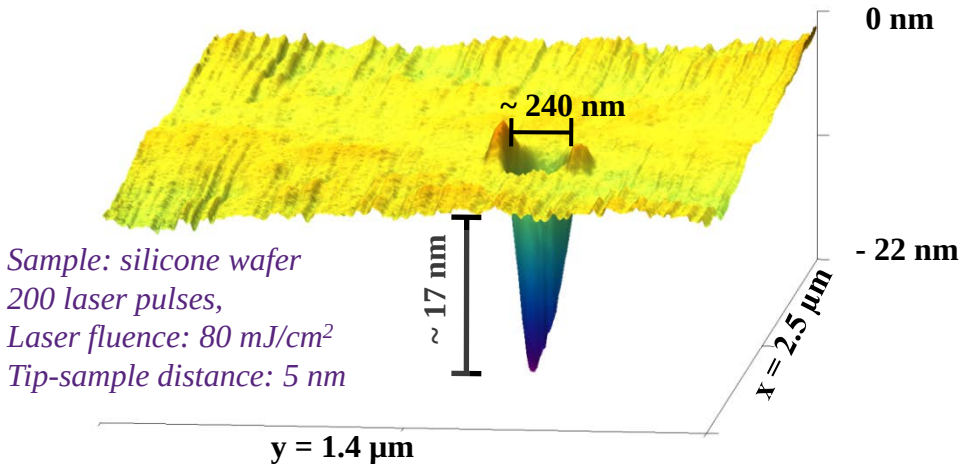
Nd-YAG laser *(266 nm) , pulse duration 4 ns.*

AFM*: **A**tomic **F**orce **M**icroscope.*

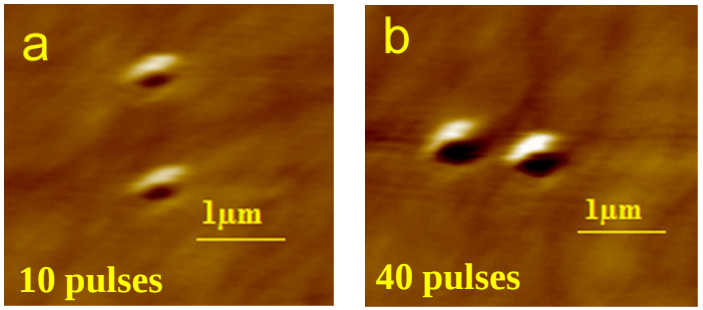
ICP-MS*: **I**nductively **C**oupled **P**lasma - **M**ass **S**pectrometry : double focusing sector field*

Nanometric laser ablation





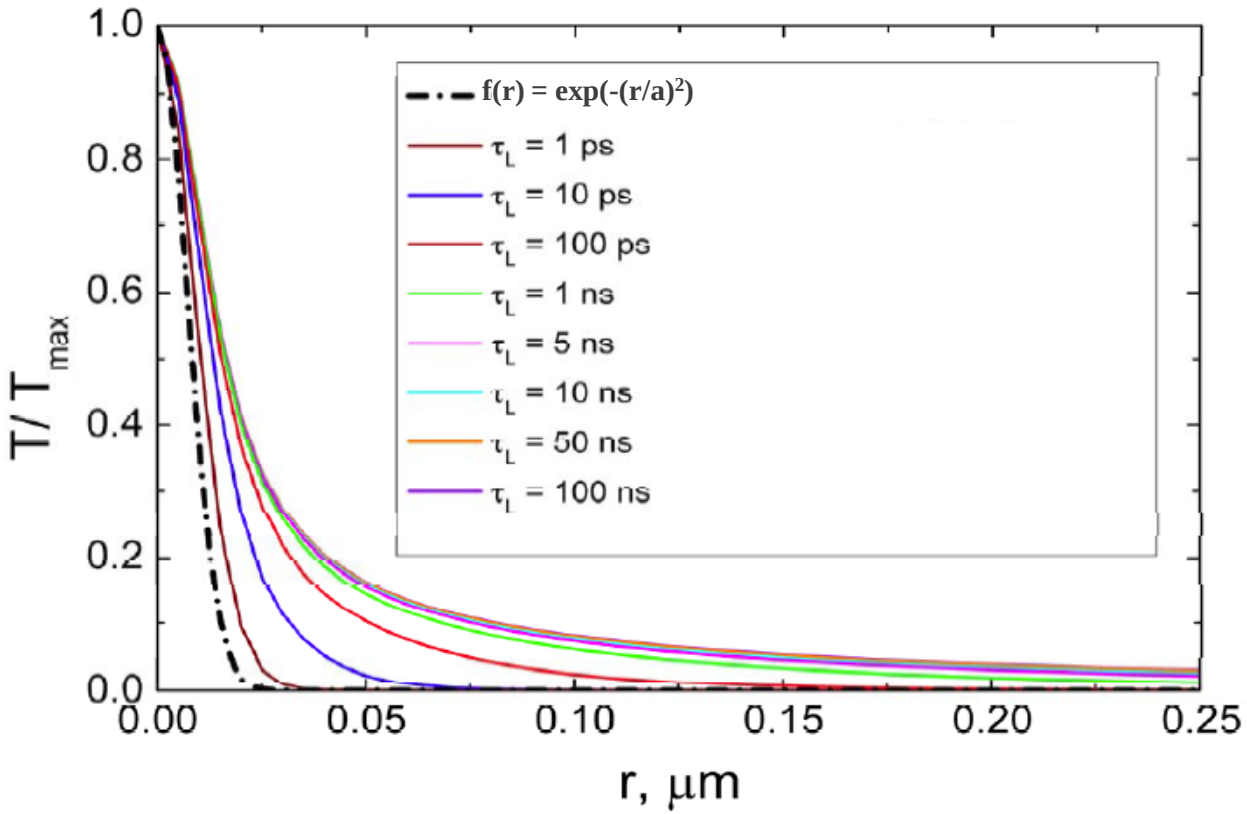
Sample: silicone wafer
200 laser pulses,
Laser fluence: 80 mJ/cm²
Tip-sample distance: 5 nm



[***] C. Jabbour, J.-L. Lacour, M. Tabarant, A. Semerok, F. Chartierc, Development of a tip enhanced near-field laser ablation system for sub-micrometric analysis of solid samples, J. Anal. At. Spectrom., 2016, **31**, 1534–1541

Normalized curves $T(r)$ for different laser pulse duration τ

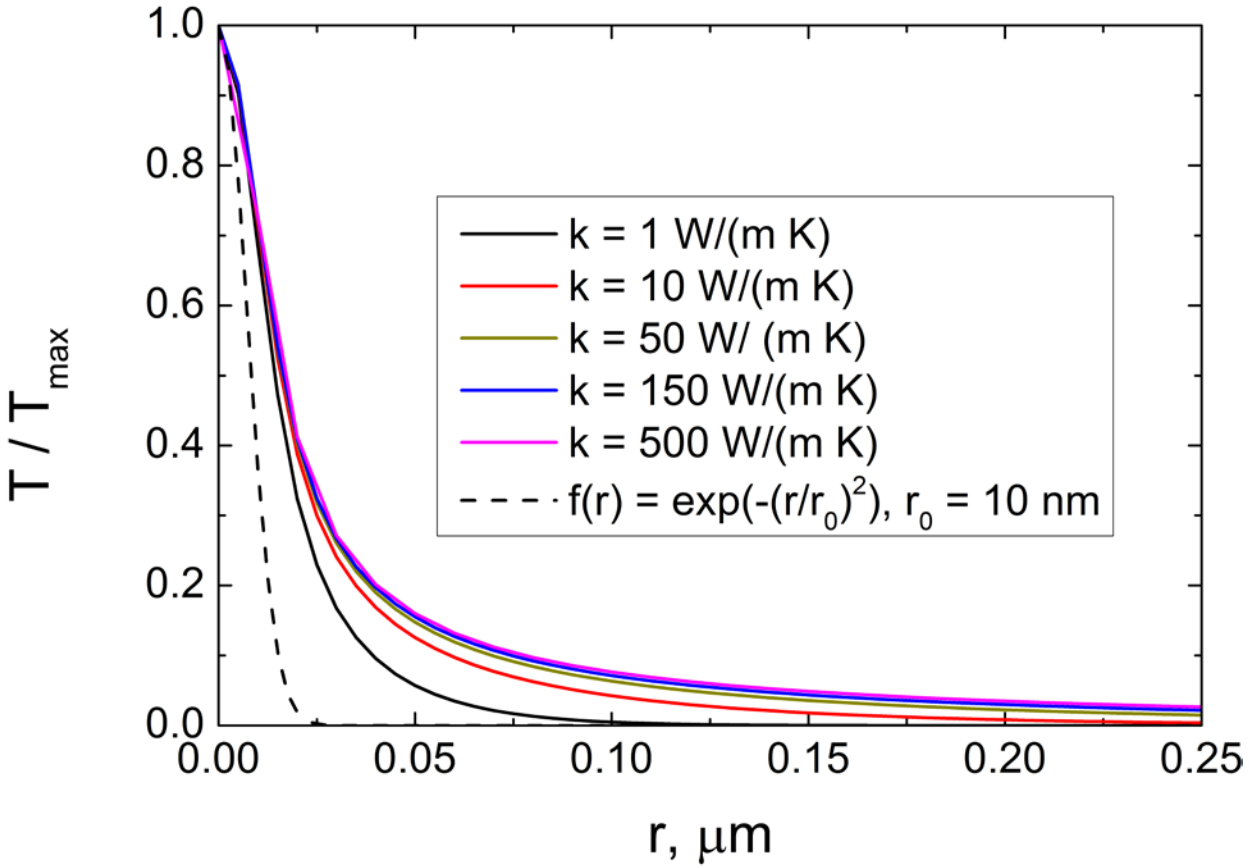
Si, λ=266 nm, α^{-1} = 5 nm, D_T = 0.8 cm^2/s, L_T = 20 nm ÷ 6.0 µm, $2a$ = 20 nm



T= f (r) for z = 0 and t = τ_{laser}

Normalized curves $T(r)$ for different k (W/m K)
(thermoconductivity coefficient)

Si, λ=266 nm, α^{-1} = 5 nm, L_T = 100 nm ÷ 2.0 µm, $2a$ = 20 nm



T= f (r) for z = 0 and t = τ_{laser} = 4 ns

CONCLUSIONS

1) **"Ablation threshold"** depends on laser spot diameter for $L_T >> a$;

2) **Low dependence** of $T(r, z)/T_{max}$ on τ (s) for 3D heating regime;

3) **Low dependence** of $T(r, z)/T_{max}$ on k (W/m K) for 3D heating regime;

4) Possibility to make **nanometric** laser ablation with ns lasers, when $L_T >> a$;

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Thank you for your attention

ABSTRACT

In LIBS microanalysis, spatial resolution is determined by laser beam diffraction limits (of the order of a laser wavelength) and thermal diffusion of a deposited heating energy during laser pulse duration (proportional to the square root of the pulse duration and matter diffusivity). Being limited by these features of a laser beam and heating energy, the best LIBS spatial resolution (the crater diameter) of 1 μm was obtained by ablation with the laser pulses of 266 nm wavelength and 4 ns duration. One of the ways to improve spatial resolution of microanalysis may be suggested as the use of laser pulses of a lower wavelength ($<$ 266 nm) and shorter pulse durations (ps and fs).

In the search for the other ways to improve spatial resolution as an important feature of microanalysis, we made laser ablation experiments with a highly localized electromagnetic field obtained by a tip near-field enhancement. In our experiments, a tip of an atomic force microscope was illuminated by the laser pulses of 266 nm wavelength and 4 ns duration. The experiments were followed by extensive multi-parametric theoretical studies to analyze the effect of both matter properties (absorption coefficient, thermal conductivity and capacity) and electromagnetic field parameters (spatial distribution and pulse duration) on the resulted temperature field distribution T(r, t). The simulations were made with a home-developed “3D+t” heating model. The simulation results have demonstrated a good agreement with the craters of 100 nm diameters which have been experimentally obtained with Si-, Au- and Ta-samples.

Discussion on the possibilities of application of the ablation with tip near-field enhancement for consecutive chemical analysis with nanometric resolution will be presented.