Laboratoire Lasers, Plasmas et Procédés Photoniques
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Laser damage and ablation in ultrashort pulsed regime: recent measurements and applications

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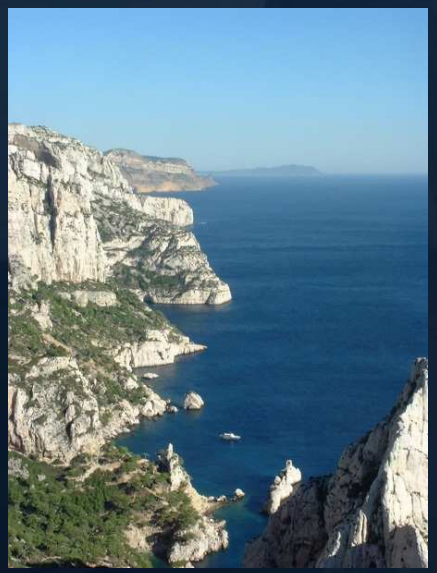
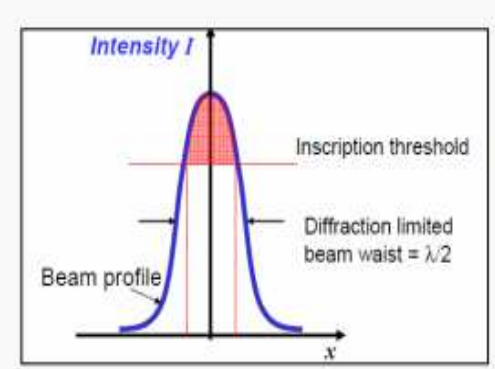
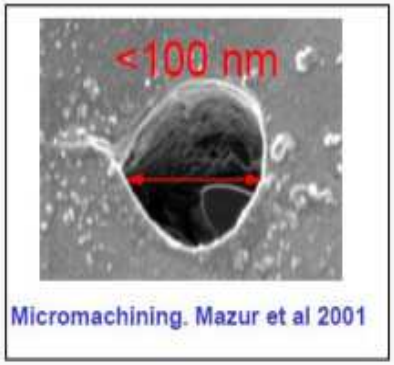
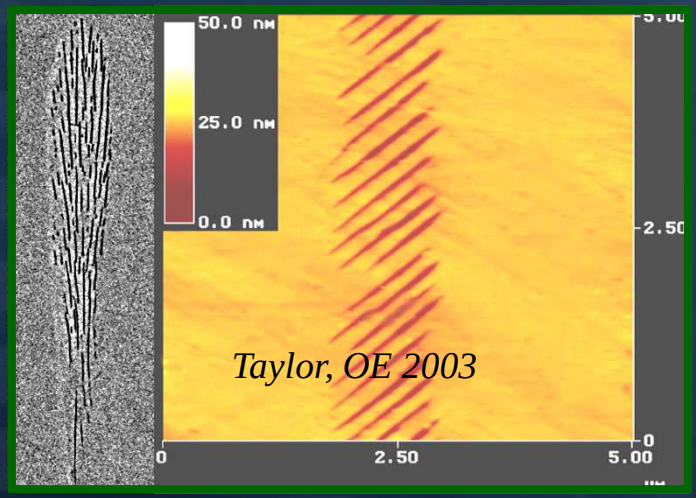
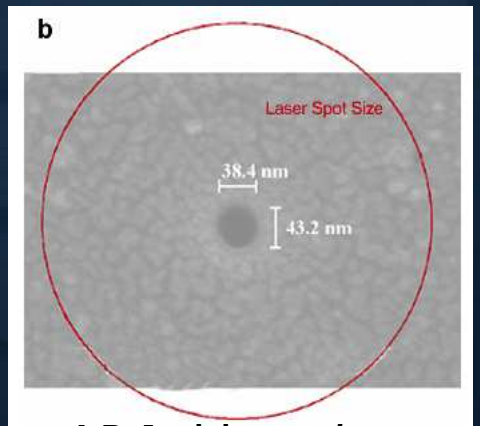


Due to NL interaction, a fs pulse provides resolution and localization of induced effects

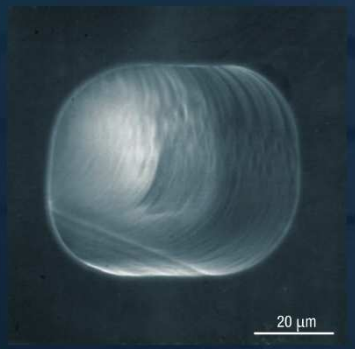




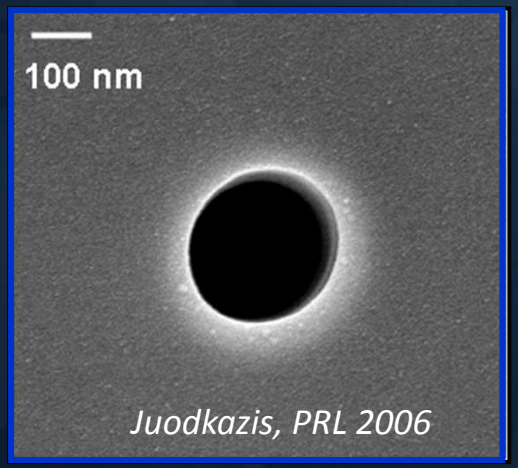




A.P. Joglekar *et al.* PNAS, 101, 5856-5861 (2004)



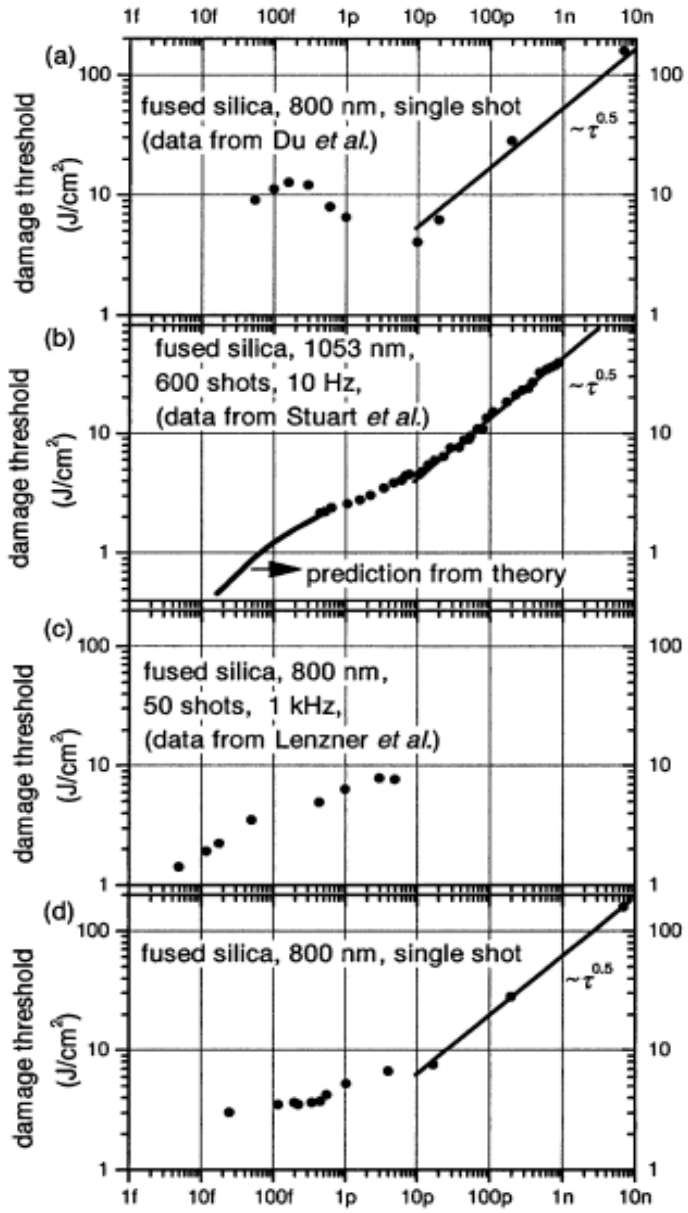
Bellouard. Y. and al (2004).



- Find out the optimum laser conditions for 2D and 3D machining with high resolution, high efficiency, quality and selectivity,….
- Progress in understanding of fs laser – matter interaction mechanims

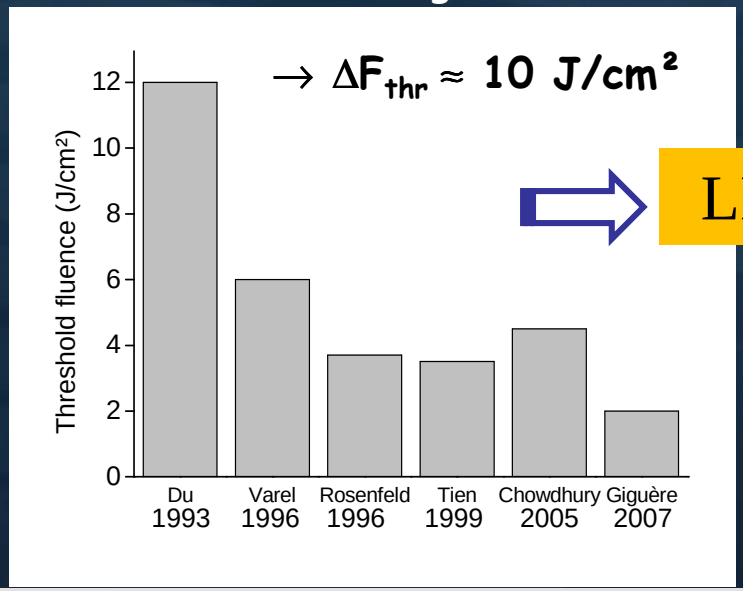
A- Laser Induced Damage Threshold (LIDT): the "Big" problem !

For instance, SiO_2: <u>huge LIDT data **dispersion** (different motivations, etc.)</u>



Tien et al., PRL, 1999

<u>Same experimental conditions</u> : SiO_2, ~ 800 nm, ~100 fs, single shot



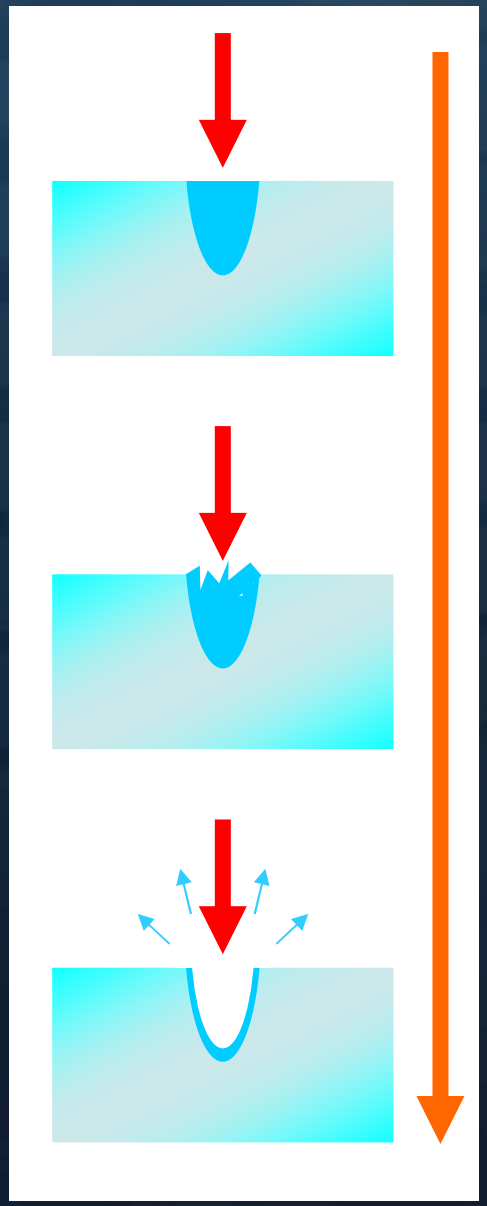
<u>Data dispersion comes from:</u>

- Different threshold criteria and diagnostics (ex-situ/in-situ; TOF, light scattering, plasma formation, time-resolved interference, transient reflectivity, microscopy, etc.)

- laser source (beam distribution, NA, contrast ratio, energy measurements, etc.)

- different kind of sample (polishing, cleaning, surface state, exact chemical composition and defects, etc.)

B- Lets' clarify: Definitions and Methodology*



Structural modification

($I < 10^{13}$ W/cm^2)

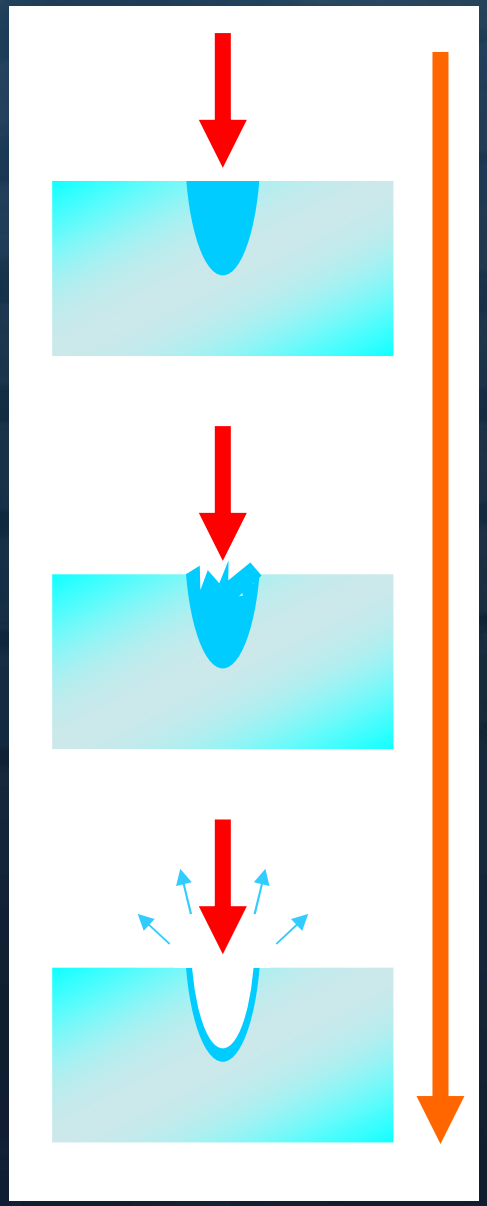
- linear and/or nonlinear refractive index change,…)
- No morphology change on the surface

Structural modification

Not studied. No tools available for this measure → development of specific analysis tools (THG microscopy)

* F. Korte et al., OE 2000; N. Sanner et al., APA 2009

Lets' clarify: Definitions and Methodology*



Structural modification

- linear and/or nonlinear refractive index change,...)
- No morphology change on the surface

Damage (I ~ 10^{13} W/cm^2)

- Morphology change on the surface
- No matter ejection

Structural modification

Not studied. No tools available for this measure → development of specific analysis tools (THG microscopy)

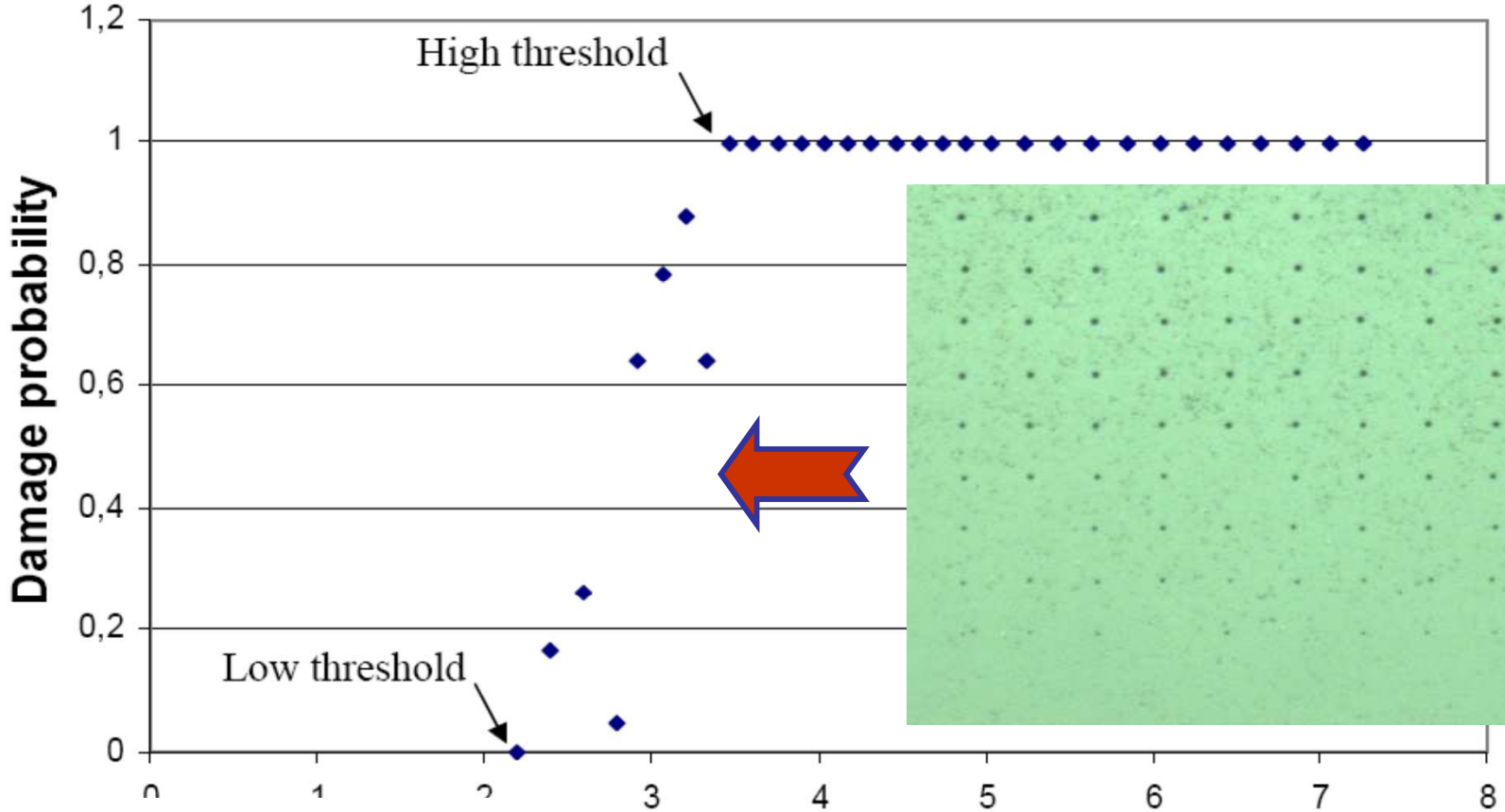
Damage ↔ LIDT

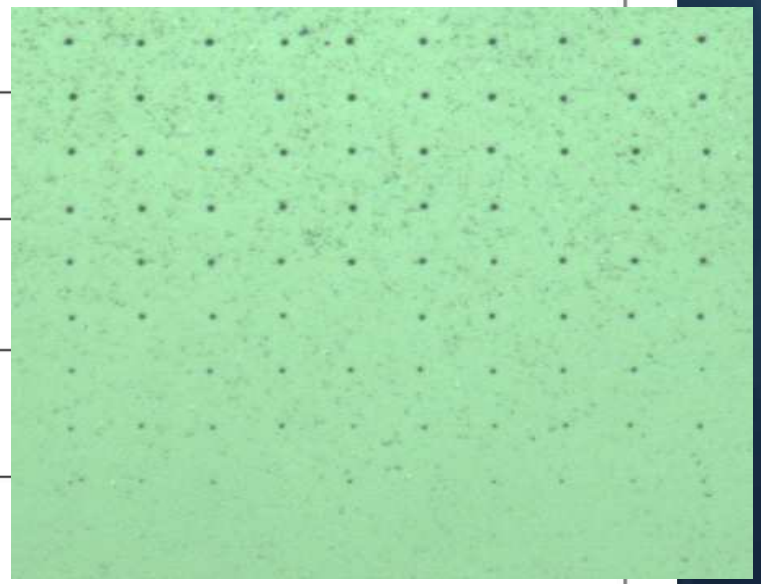
(Laser-induced Damage Threshold)

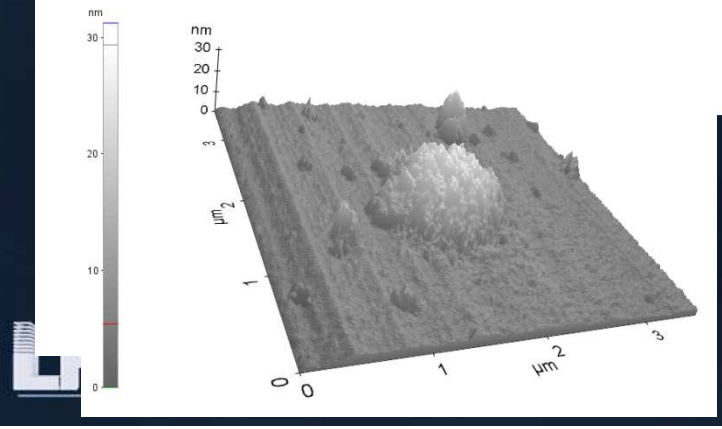
→ qualitative analysis (visible damage): **optical microscopy and statistical approach**

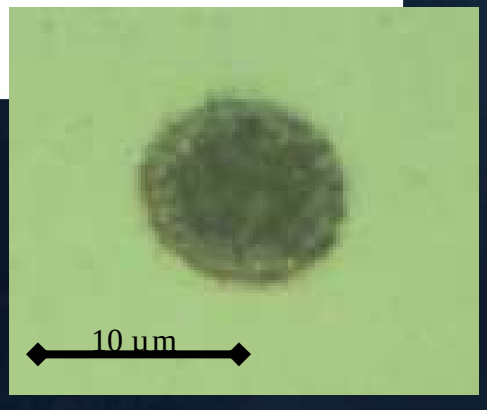
* F. Korte et al., OE 2000; N. Sanner et al., APA 2009

Laser damage – LIDT (best evidenced with optical microscope, « Yes/No » damage diagnostic, statistical approach)









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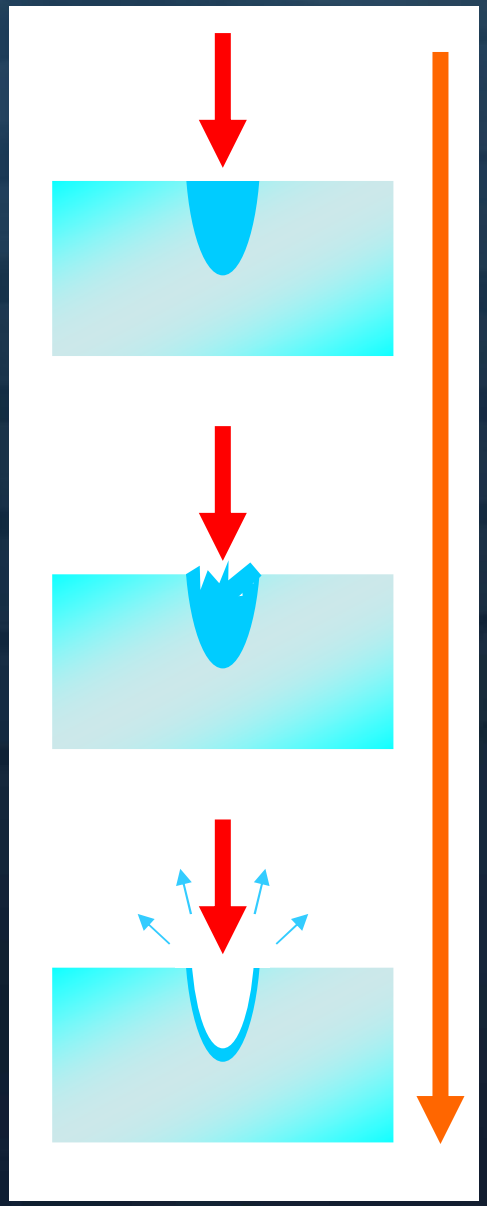


Structural modification

- linear and/or nonlinear refractive index change,...)
- No morphology change on the surface

Damage

- Morphology change on the surface
- No matter ejection

Ablation

($I > 10^{13}$ W/cm^2)

- ejection of matter

Structural modification

Not studied. No tools available for this measure → development of specific analysis tools (THG microscopy)

Damage ↔ LIDT

(Laser-induced Damage Threshold)

→ qualitative analysis (visible damage): **optical microscopy and statistical approach**

Ablation ↔ LIAT

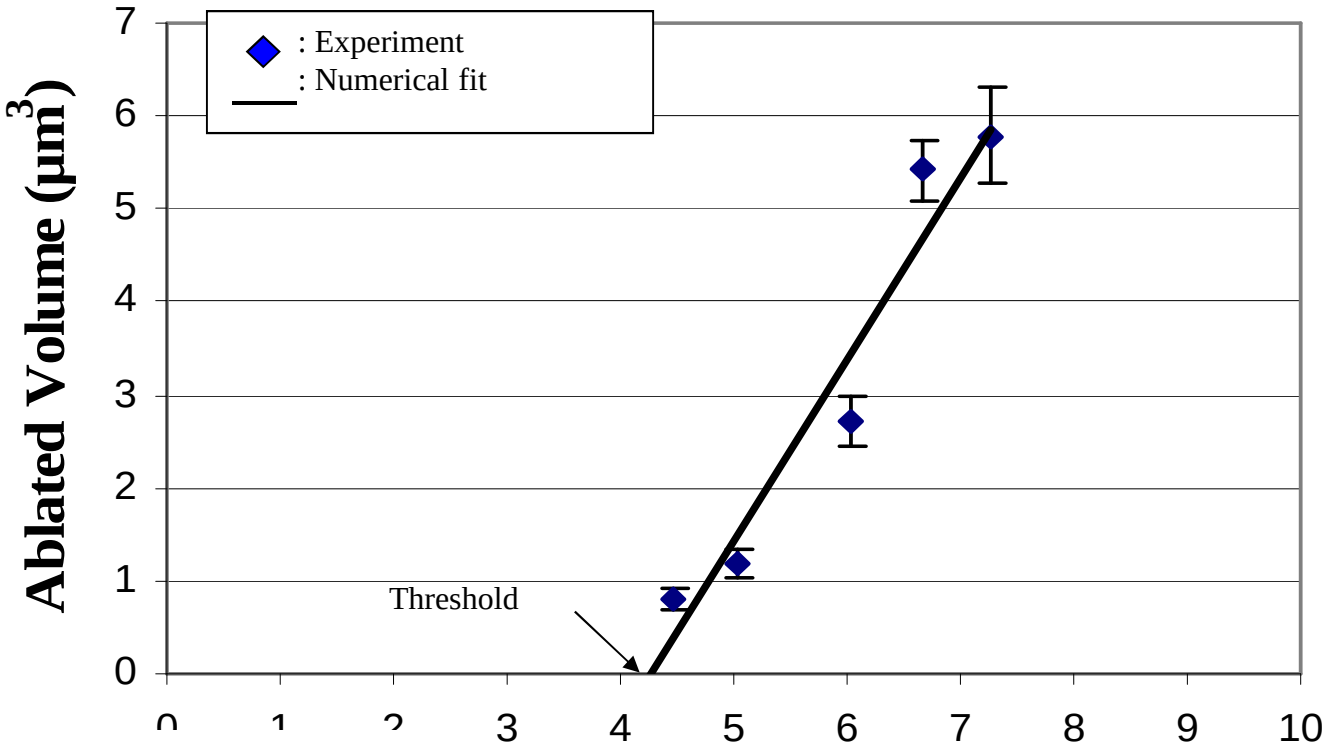
(Laser-induced Ablation Threshold)

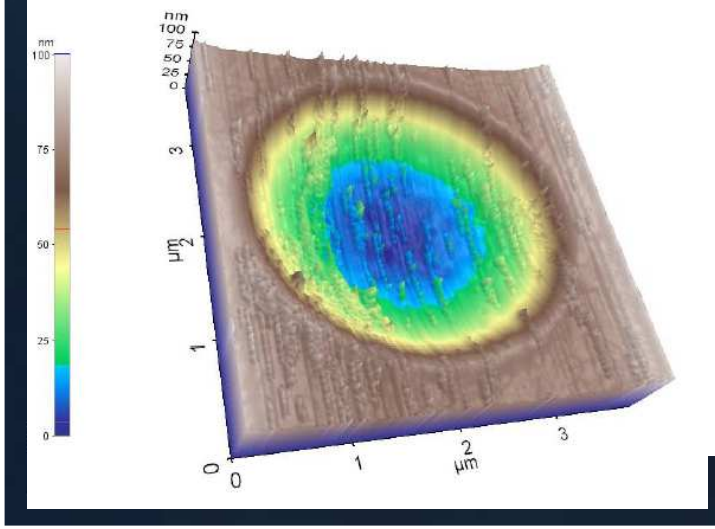
Measure of ablated volume

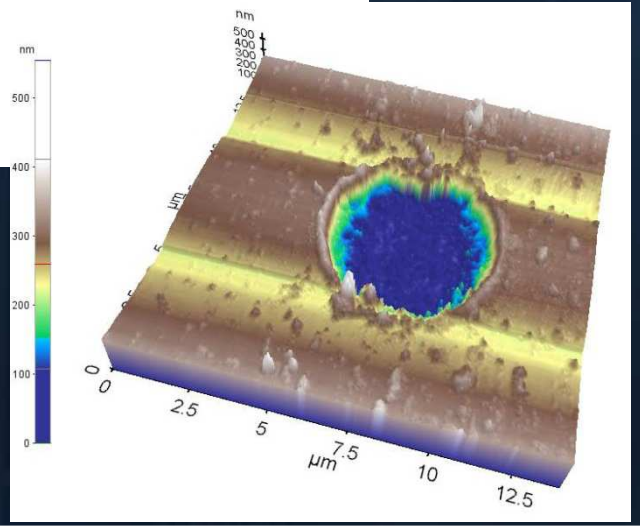
→ **AFM and regression approach**

* F. Korte et al., OE 2000; N. Sanner et al., APA 2009

Laser Ablation – LIAT (best evidenced with AFM)

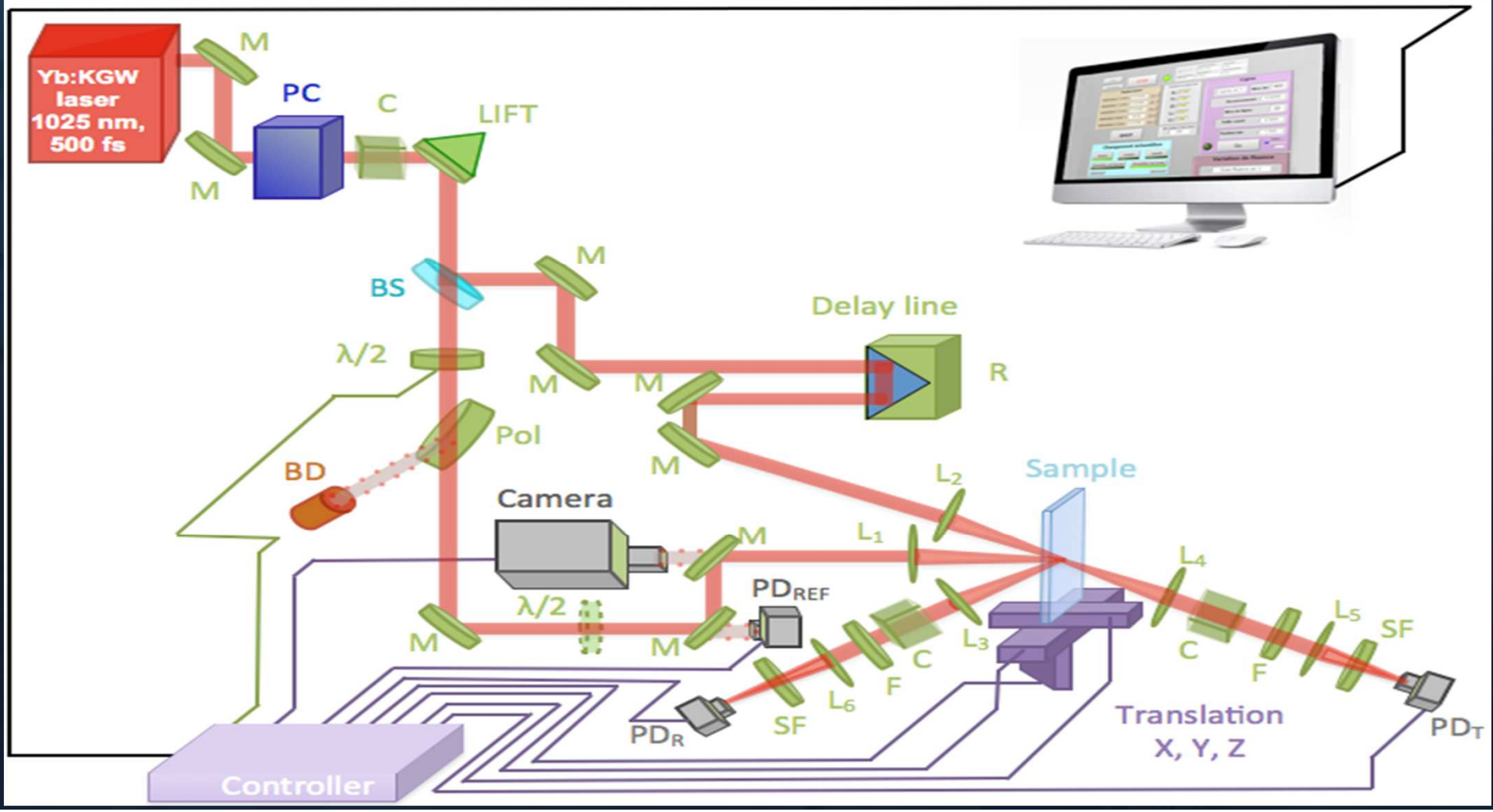




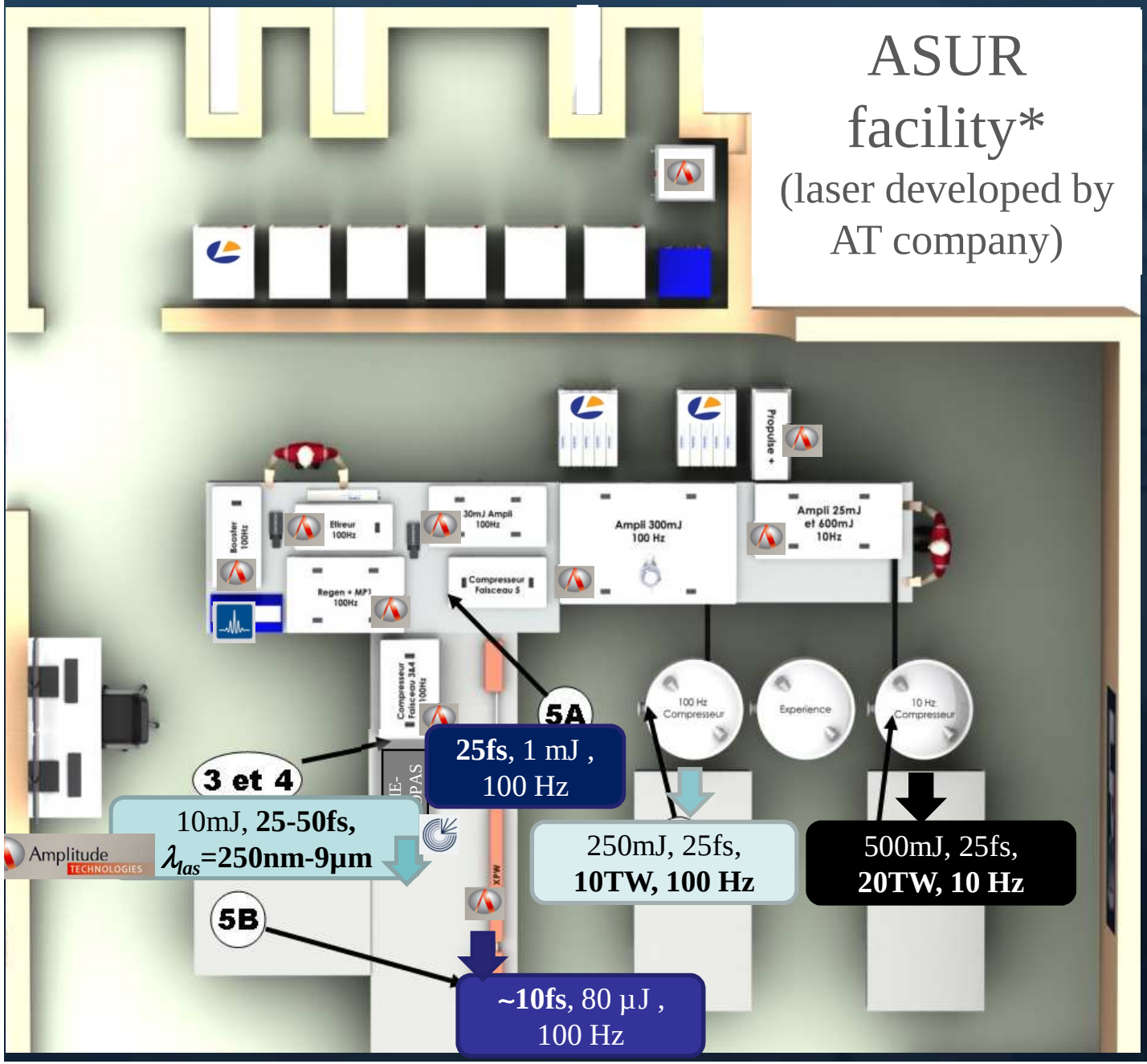


C- Experiments: Damage/ablation test-bench in LP3

- Laser (pulse duration, fluence): τ_{pulse} : **500 fs (operational, AS laser @ 1025nm) and 10 fs → 400 fs + OPA (ASUR)**; F : 1 – 20 J/cm², linear pol.
- Single-shot/Nshots regime (1on1, Non1), air/vacuum ambiance
- Diagnostics: ex-situ: AFM, optical microscope; in-situ**:** pump-pump, **pump-probe**
- **Materials (SiO_2, Al_2O_3, Ti^{3+}:Al_2O_3) and components (gratings, mirrors, etc.)**



ASUR laser system (10 TW@100 Hz) in LP3

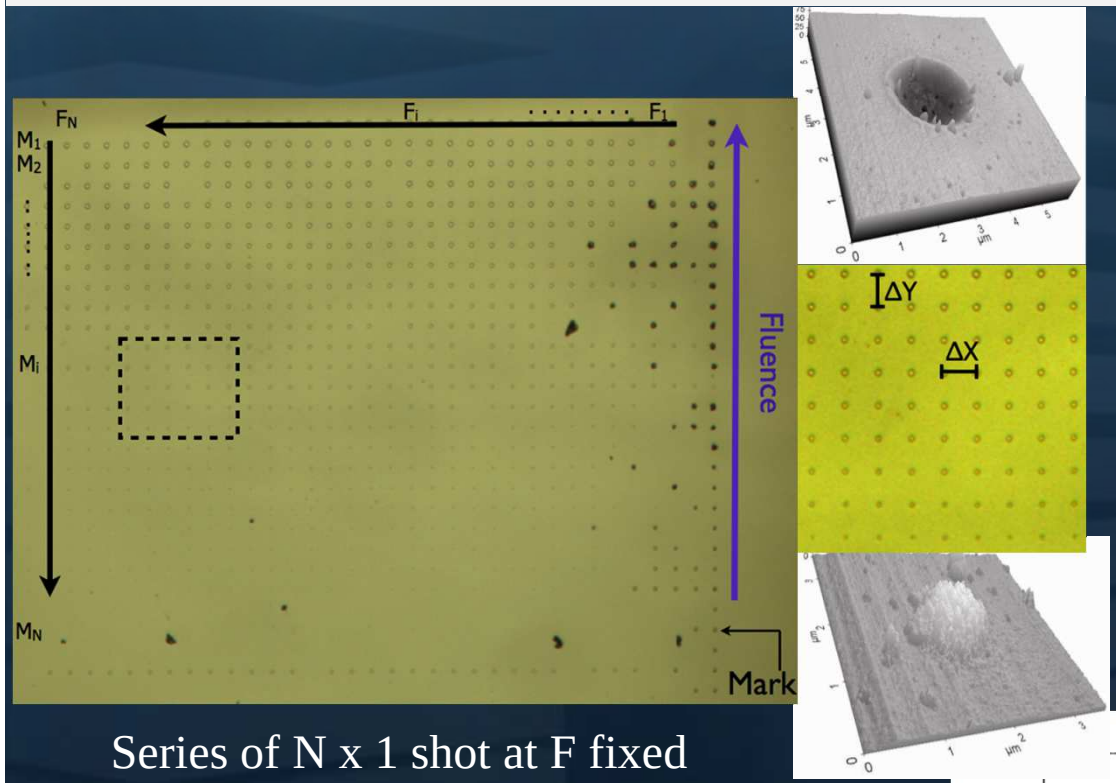


- 5 laser lines
- possible to combine low and high energy beamlines
- Ultrashort pulses (~10 fs, XPW): available test-bench (air and soon vaccuum)
- Adaptative optics
- radioprotection

Secondary Sources :

- OPA : < 35 fs, 100 Hz – 0.24 µm à 9 µm - > 30 µJ to 1 mJ
- X Ray (Kα) 10 Hz et 100 Hz (2013)
- X-Ray probes (2014)

D- let's do first a simple damage experiment…



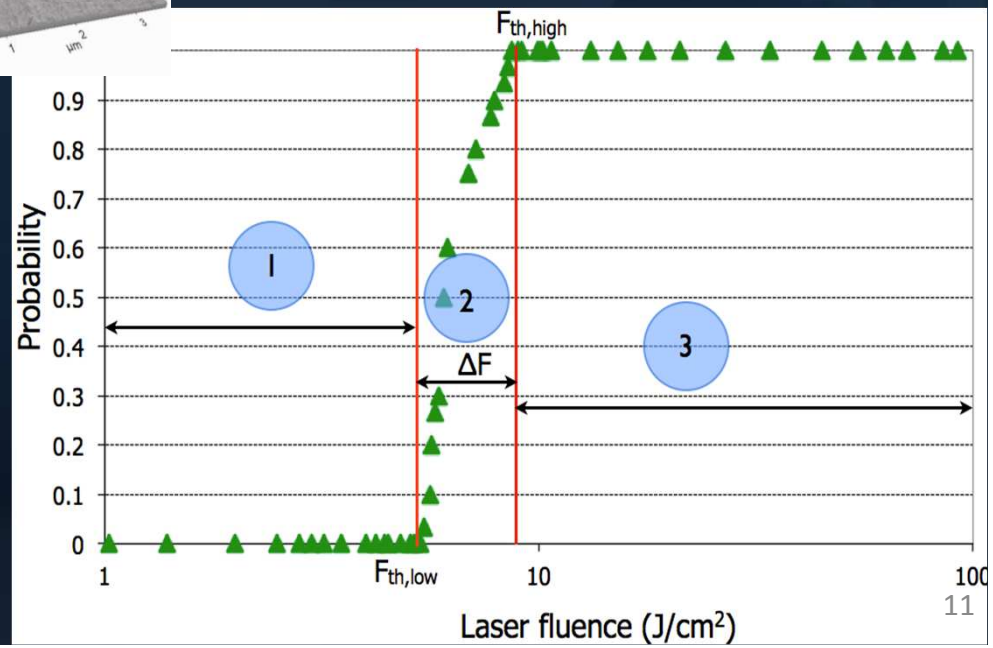
Series of N x 1 shot at F fixed

- Minimal distance not to have material surface contamination (>> ω_0)
- Space consuming experiment (minimization of the experiment area)
- **High level of metrology** (beam size and focusing geometry, energy, material,…)
- Reference and calibration samples

<u>Statistical approach to define damage threshold</u>

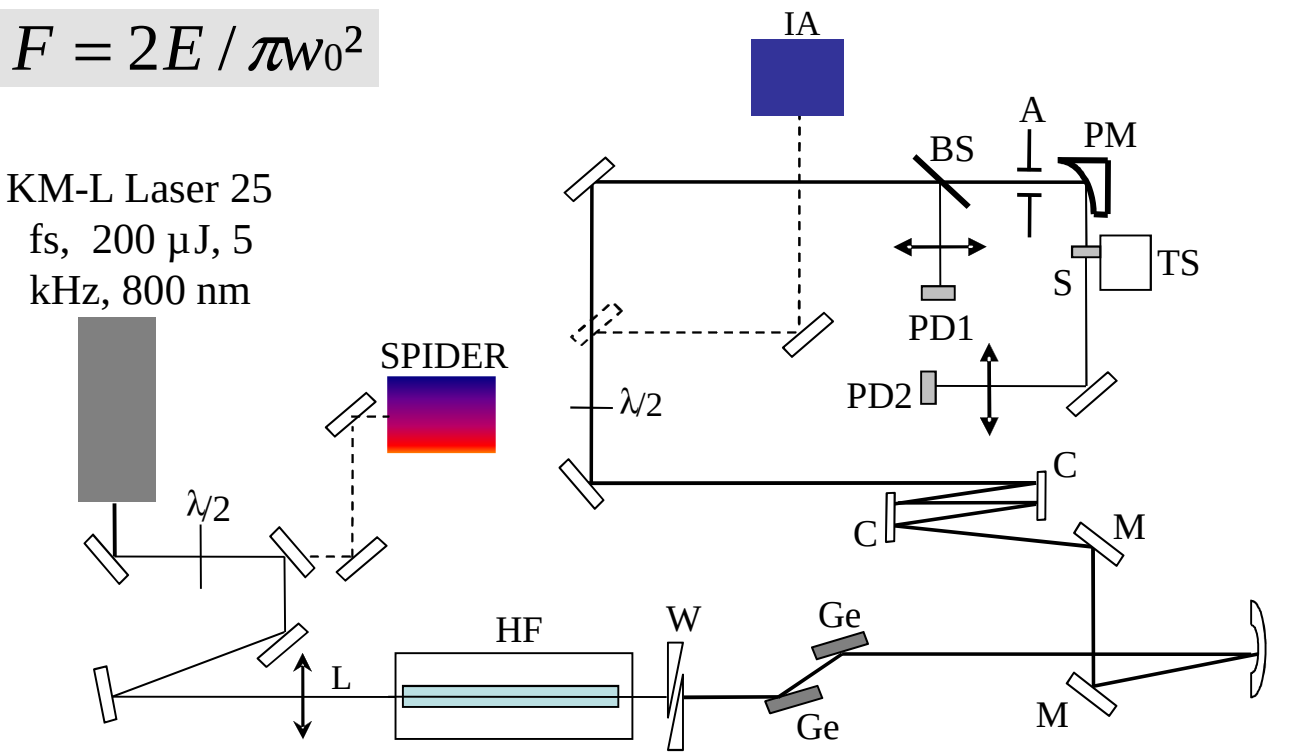
$$F_{th,\ damage} = F_{th,\ low}$$

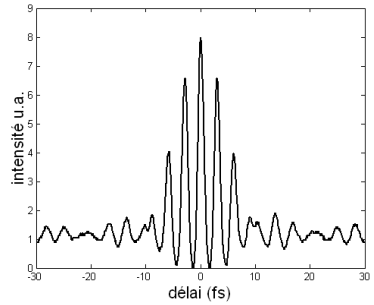
$\Delta F \leftrightarrow$ **determinism strength**

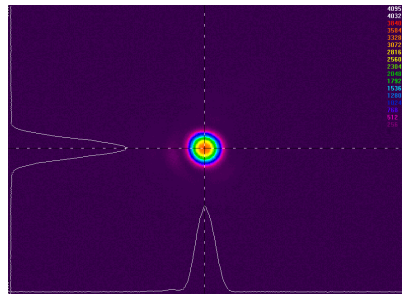


LIDT, LIAT vs τ_{pulse} (in collab. with INRS, Celia, LaHC)

$$F = 2E / \pi w_0^2$$

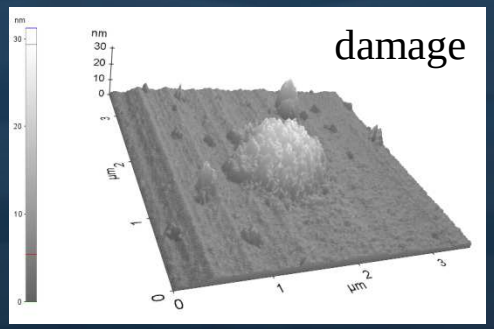


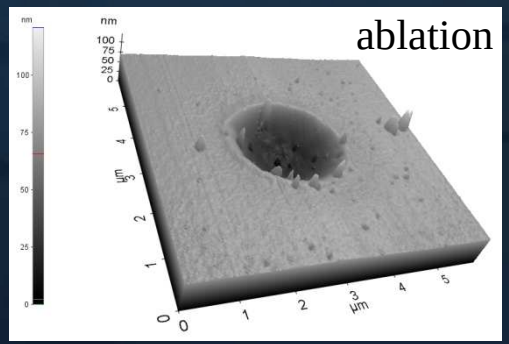


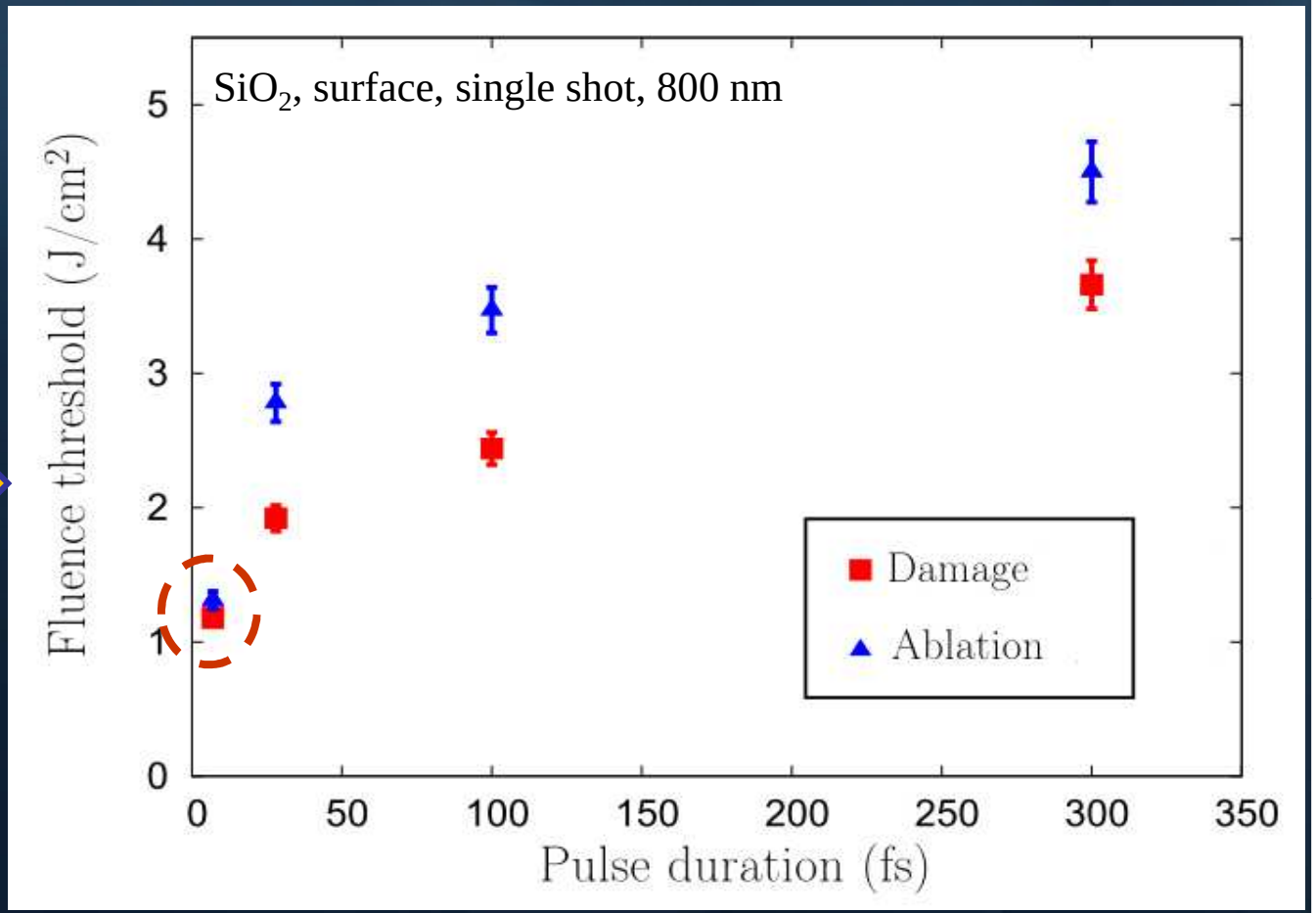


* Laser (pulse duration, fluence): τ_{pulse} : **7 fs → 300 fs** ; F : 1 – 20 J/cm², linear pol.
* Focusing : parabolic mirror EFL =50 mm (w_0 = 4.65 µm, $2z_R$ = 140 µm)
* Single-shot regime, Surface experiments
* Sample : superpolished (Ra< 0.2 nm) high-purity suprasil (α-SiO_2) (< 0.065 ppm)
* Diagnostics: AFM, optical microscope

Ablation and damage thresholds vs pulse duration (< 10 fs – 300 fs)



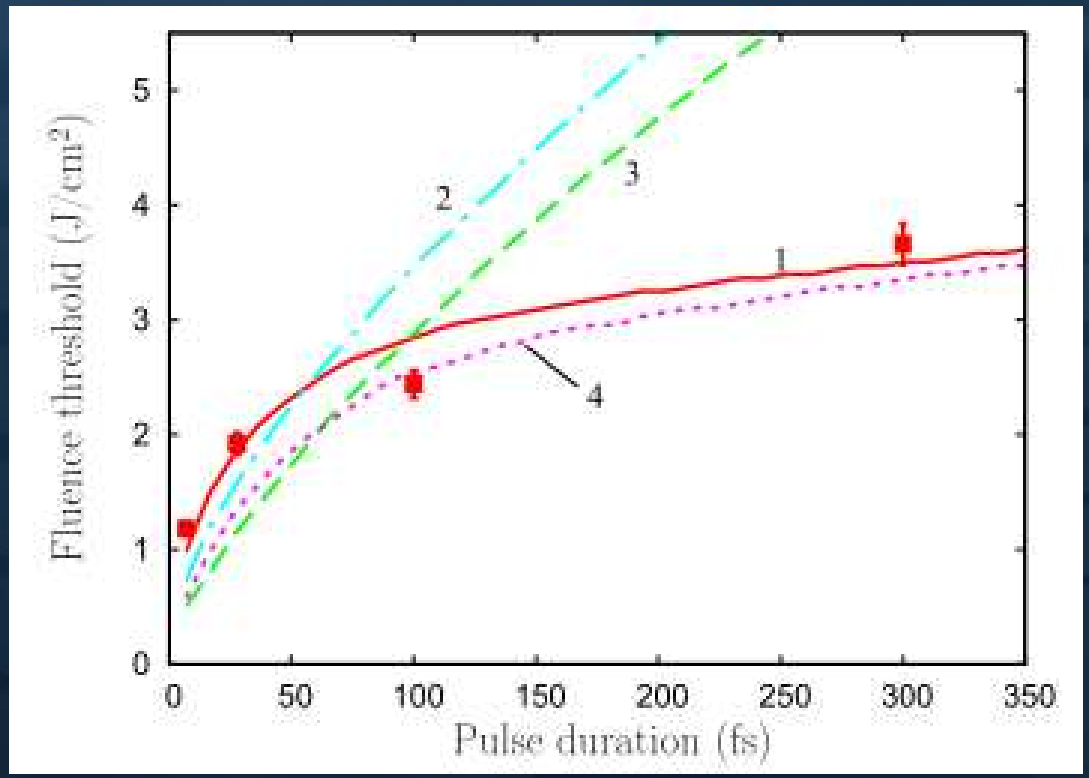




- Strong LIDT reduction at ultrashort pulse duration < 25 fs (importance of tunnel ionization and electronic effects)
- Avalanche dominates laser absorption at « long » timescale (τ_{pulse}> 50 fs)

See B. Chimier et al, PRB 84, 2011

Damage threshold versus pulse duration (< 10 fs – 300 fs): Ionization **mechanisms***

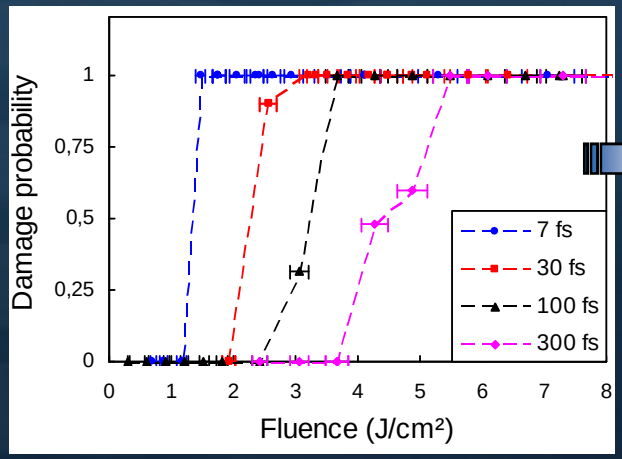


- 1 = Keldysh + avalanche (II)
- 2 = Keldysh (MPI/TI)
- 3 = MPI (σ_6 = 2 10^{25} $cm^{-3}s^{-1}$ $(cm^2/TW)^6$, C. Mézel, POP, 2008)
- 4 = MPI + avalanche
- ■ = experiment (damaging)

- Photoionization cannot only be described by MPI at short pulse durations (< 250 fs)
- Progressive significance of Tunnel Ionization (TI) at short timescale (< 250 fs)
- Avalanche (II) dominates laser absorption at « long » timescale (τ_{pulse}> 50 fs) **<u>but</u>** contributes to matter ionization whatever the pulse duration, even at ultrashort timescale (< 10 fs)

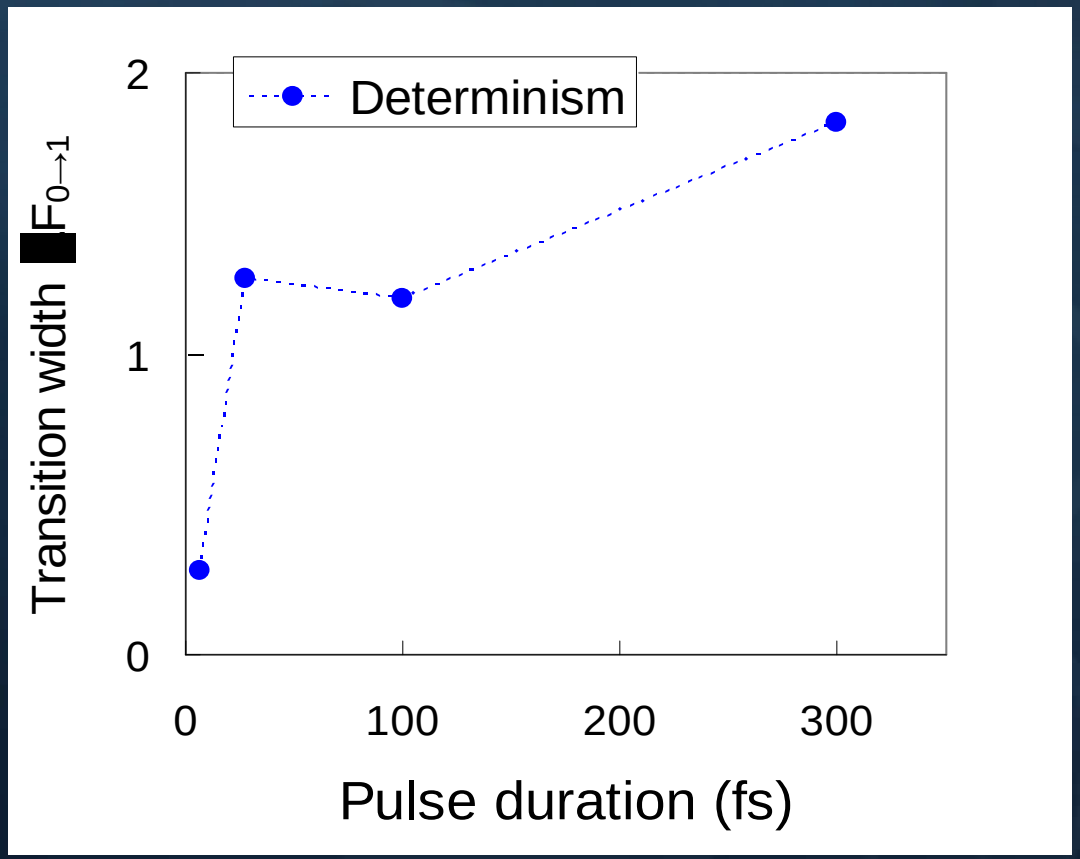
** Chimier et al, PRB 84, 2011*

A simple experience … and a lot of information: Determinism



Determinism parameter

$$\Delta F_{0\to1}^{th}$$



➡ **Strong deterministic character for « long » pulses 300 fs > τ > 30 fs**

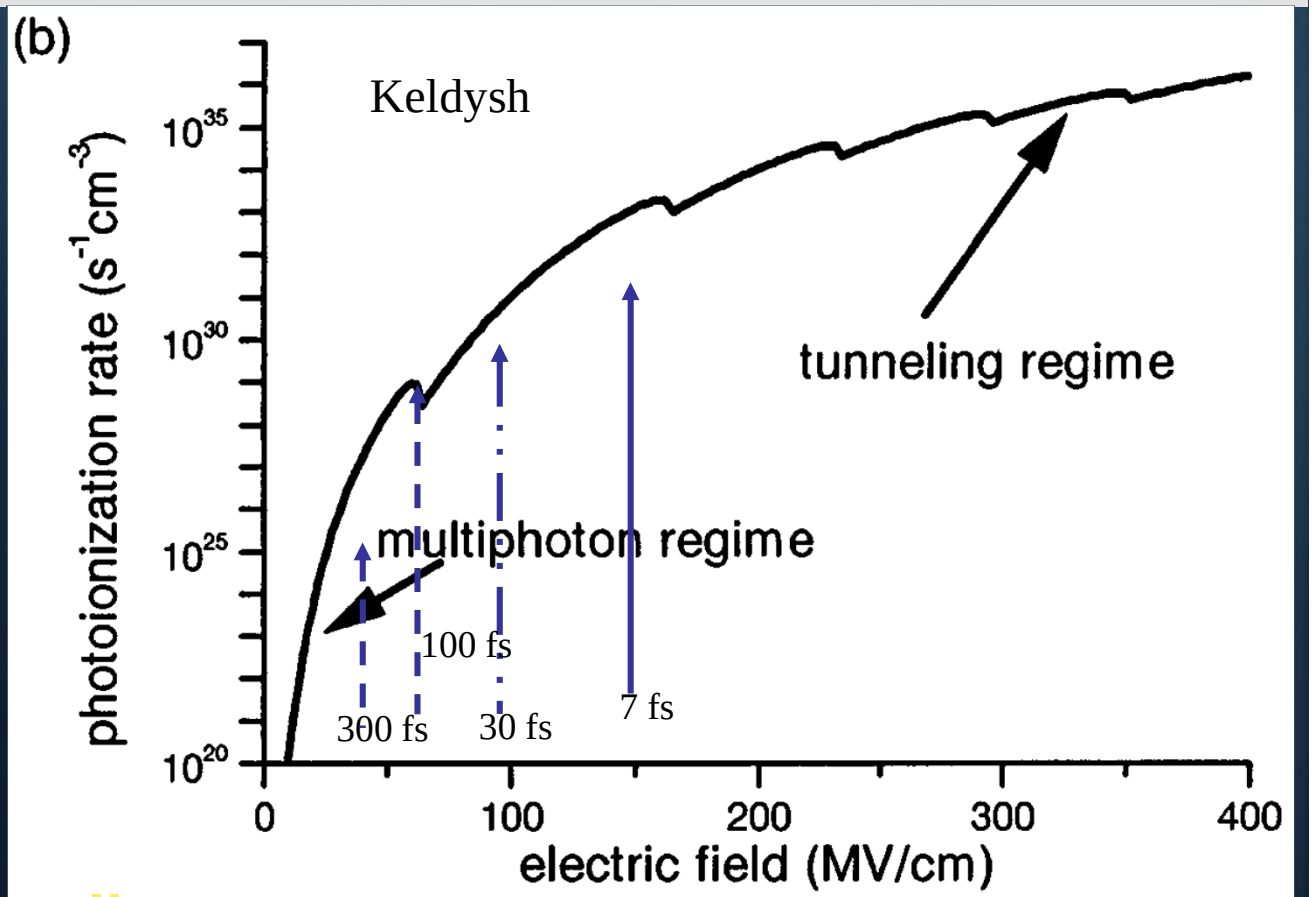
Rq : >> transition slope (ΔF > ~10 J/cm²) observed for nanosecond pulses (SiO_2, similar operating conditions)

➡ **Very strong determinism for pulses < 30 fs**

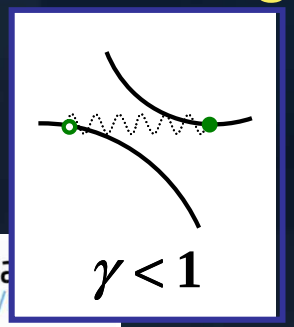
Enhancement of determinism at ultrashort time is correlated with the significance of tunnel ionization *

Tunnel Ionization is less sensitive to laser and matter fluctuations

Radical determinism arises from tunneling ionization

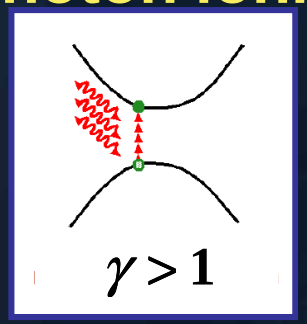


Tunneling



$\gamma = 1$

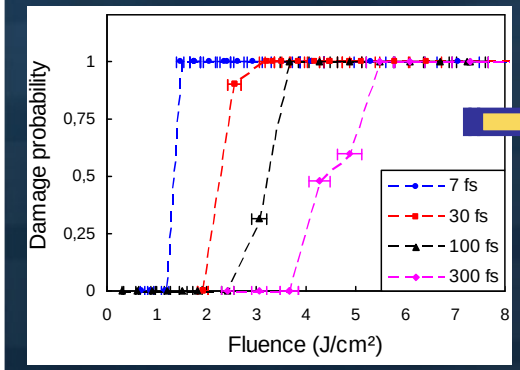
Multiphoton ionization

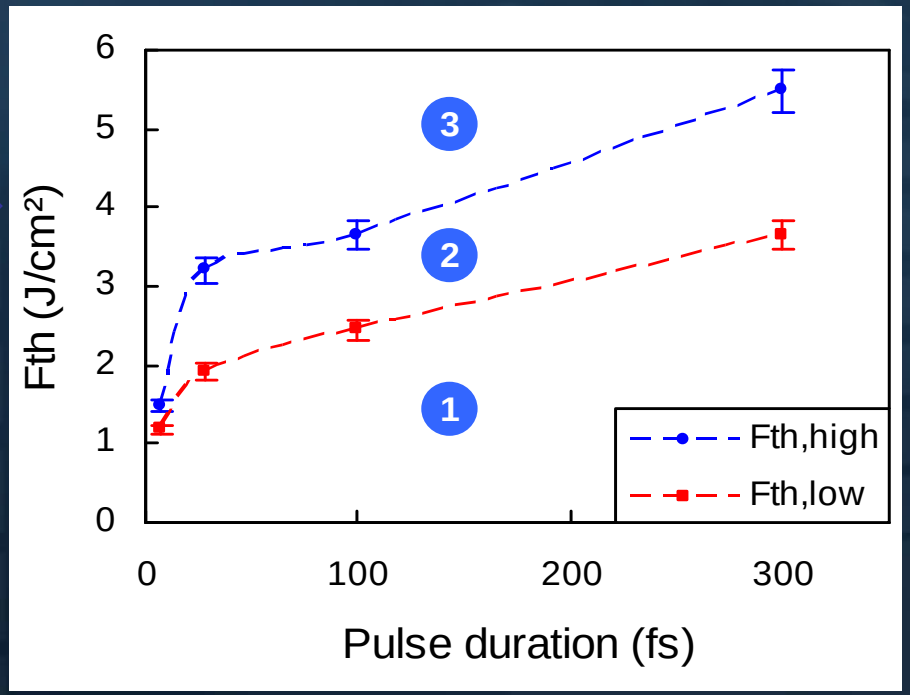


** Sanner et al., APL 96, 2010*

Chimier et al, PRB 84, 2011

A simple experience … and a wealth of information for micromachining and laser damage community*





Precious indications for material processing*

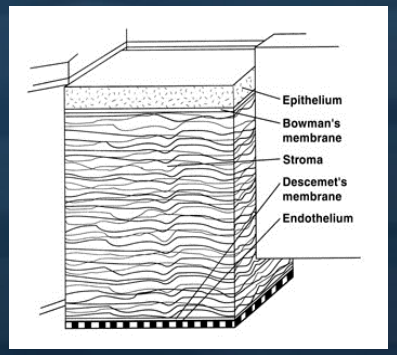
- *Zone 1 :* no damage (P=0) ↔ **safety zone**
- *Zone 2 :* erratic (statistical) damage (0<P<1) ↔ **zone to avoid for a controlled and reproducible process**
- *Zone 3 :* systematic damage (P=1) ↔ **minimum operating fluence level**

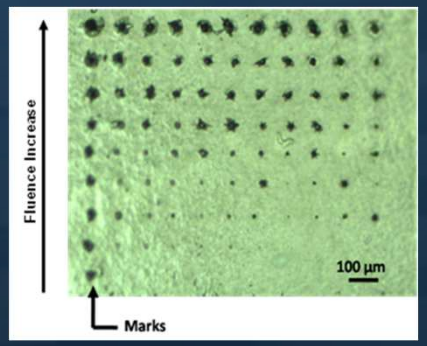
* Utéza et al., JLMN 5, 2010; APA, 2011

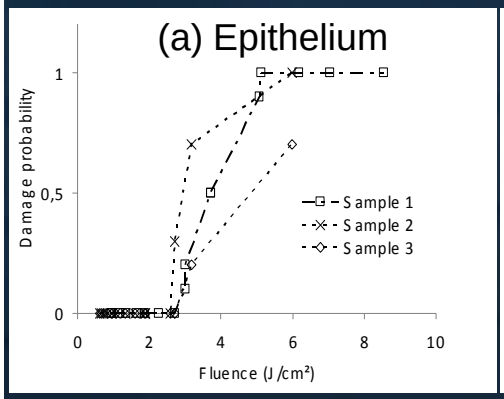
* Hoffart et al., JFO 33, 2010; OE 19, 2011

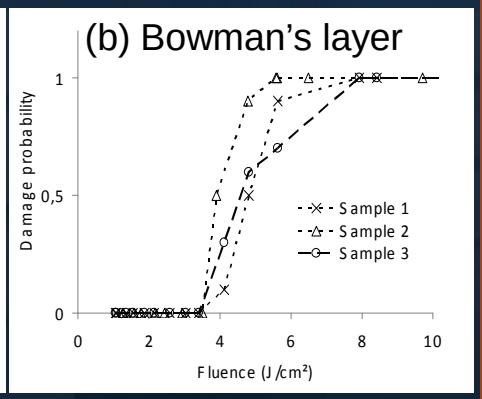
For illustration…

450 fs @ 1025 nm









Cornea Surgery
(Hôpital La Timone – LP3 - INRS)*

* Hoffart et al., JFO 33, 2010; OE 19, 2011

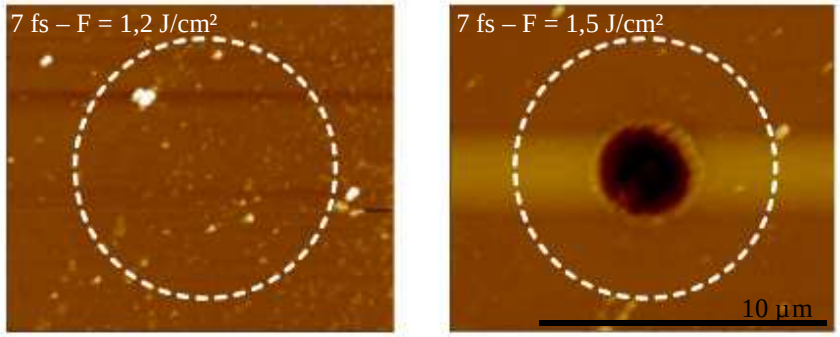
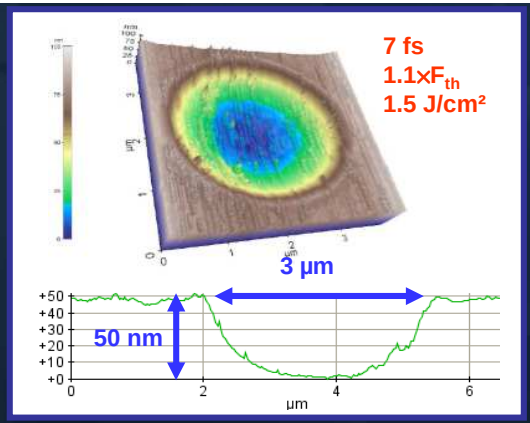


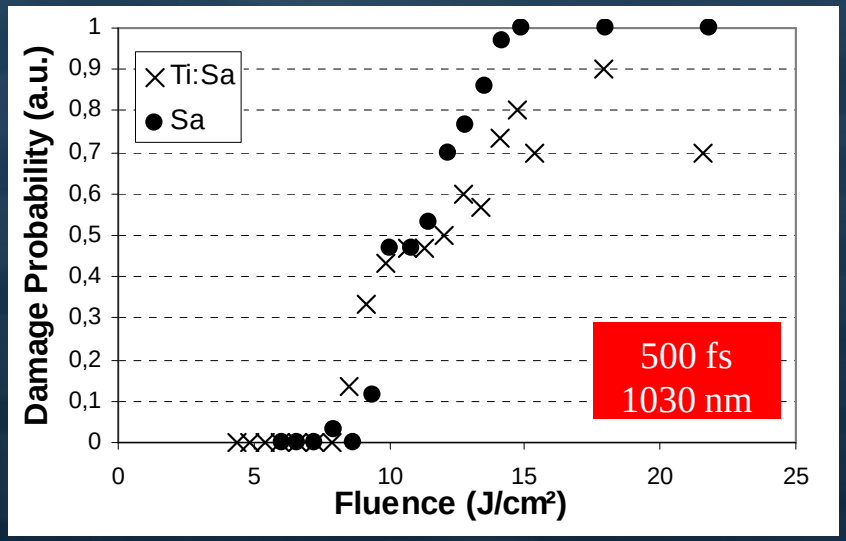
FIG. 4. (Color online) AFM images of the material after irradiation by a single 7 fs pulse, illustrating the sharpness of the transition $\Delta F_{0\to1}$. (Left) irradiation at $F_{th,low}$: no surface change is detected. (right) irradiation at $F_{th,high}$: a crater is clearly observed. The dashed circles, representing the laser beam diameter $2w_0$=9.3 μm, give the scale.

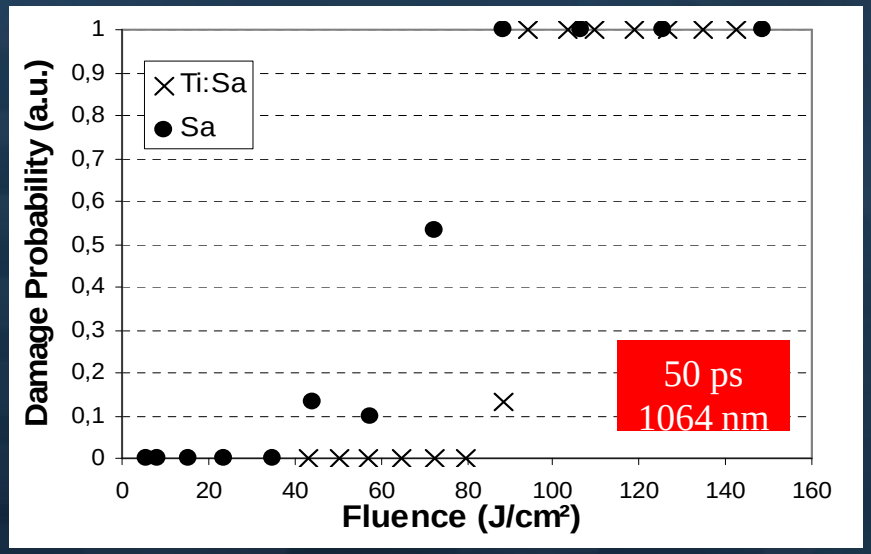


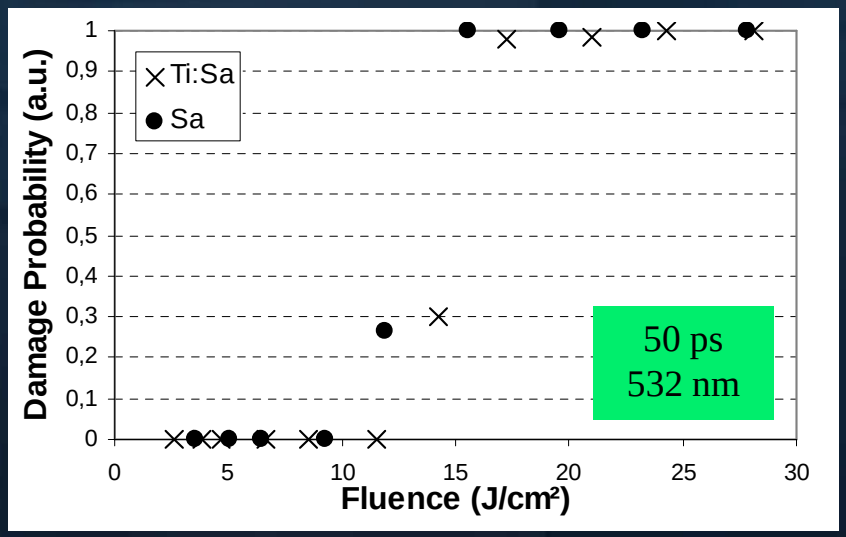
Drilling/Engraving without side effects*

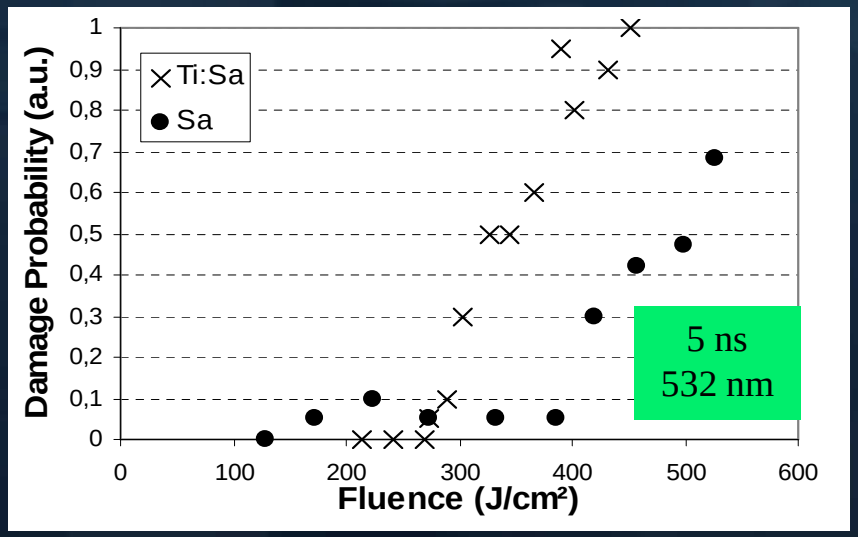
* Sanner et al., APL 96, 2010; Utéza et al., APA 105, 2011

Other measurements (collab. with AT, I. Fresnel and LOA): Sa, Ti:Sa LIDT*





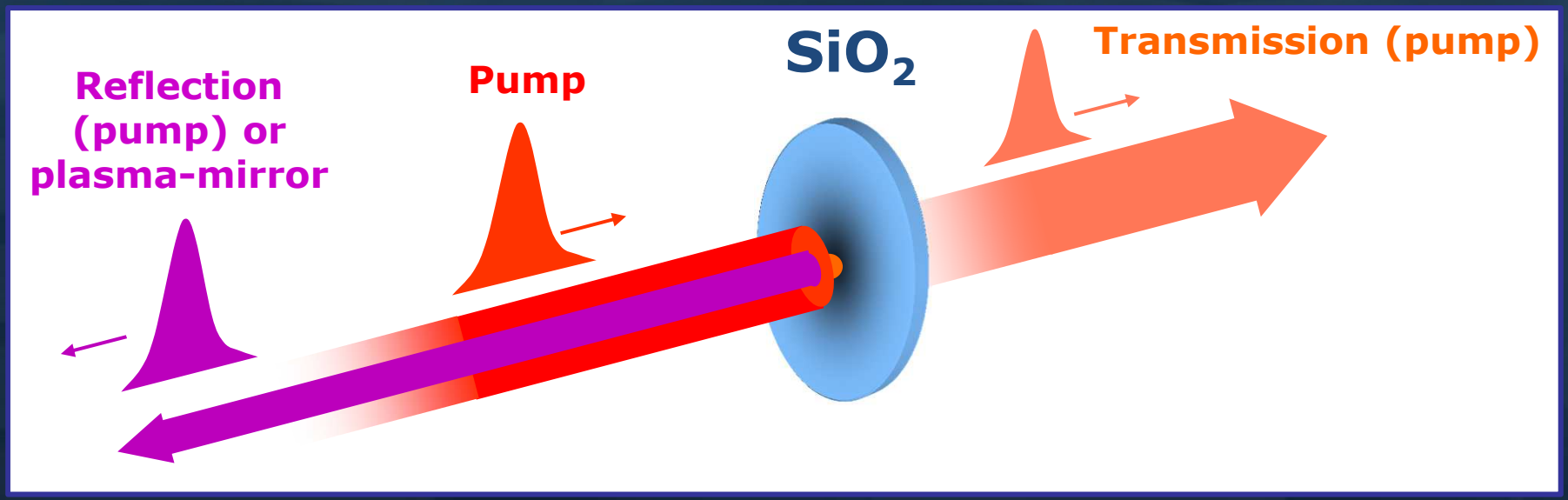




Same batch of samples, same polishing, constant and highly controlled methodology **to recover the LIDT behaviour (ns to fs) of Sa and Ti:Sa crystals**

* Bussière et al., AO 51, 2012

F- Energy balance (plasma mirror effect): self-pump and pump – probe experiments (see also M. Lebugle poster)

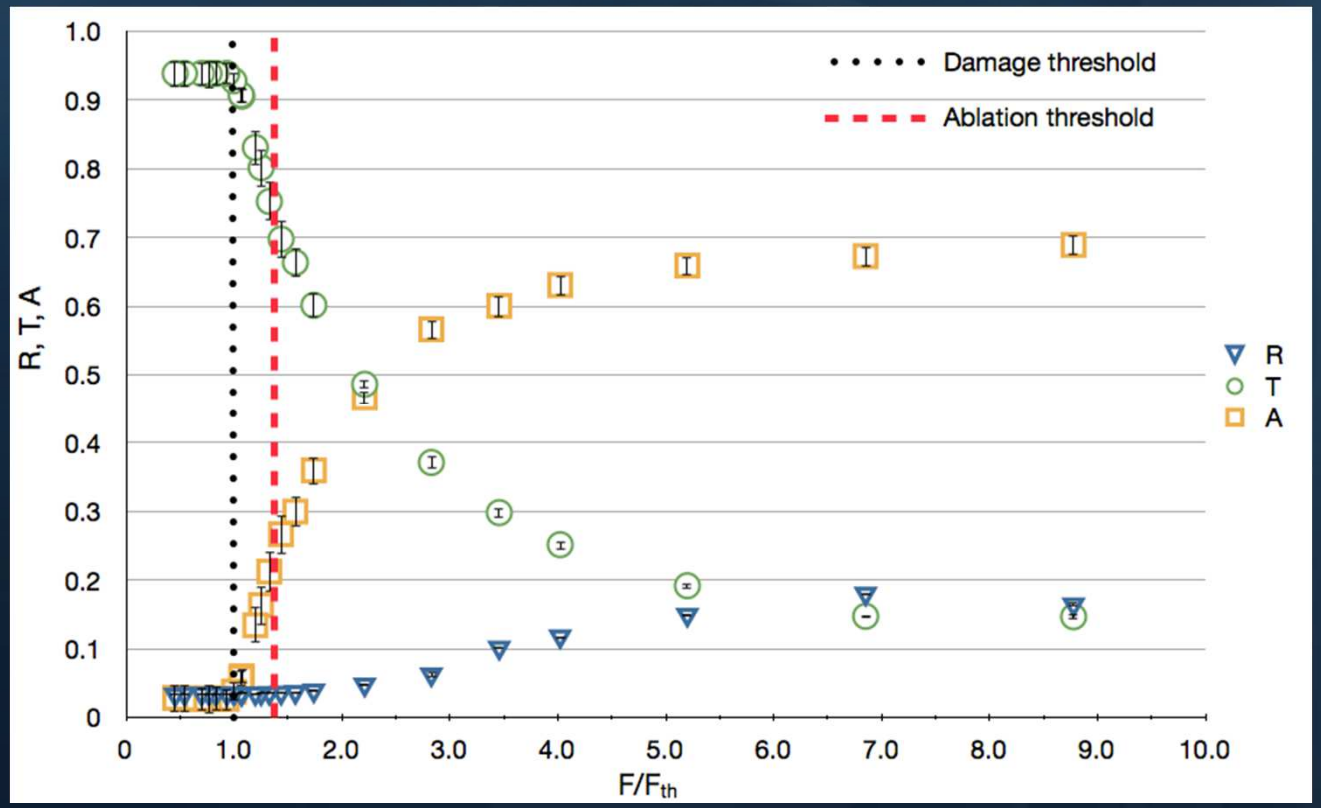


Precise energy balance

Absorption: $A = 1 - R - T$

F- Energy balance: Evolution of R, T and A vs F (500 fs)

- Dynamics of reflection and transmission is different*
- Significant change of T from F_{th} and R for high fluence
- Reflection saturation below 20%**
- Transmission saturation at 15%
- Absorption saturation at 60%
- Plasma shielding effect is delayed with respect to absorption



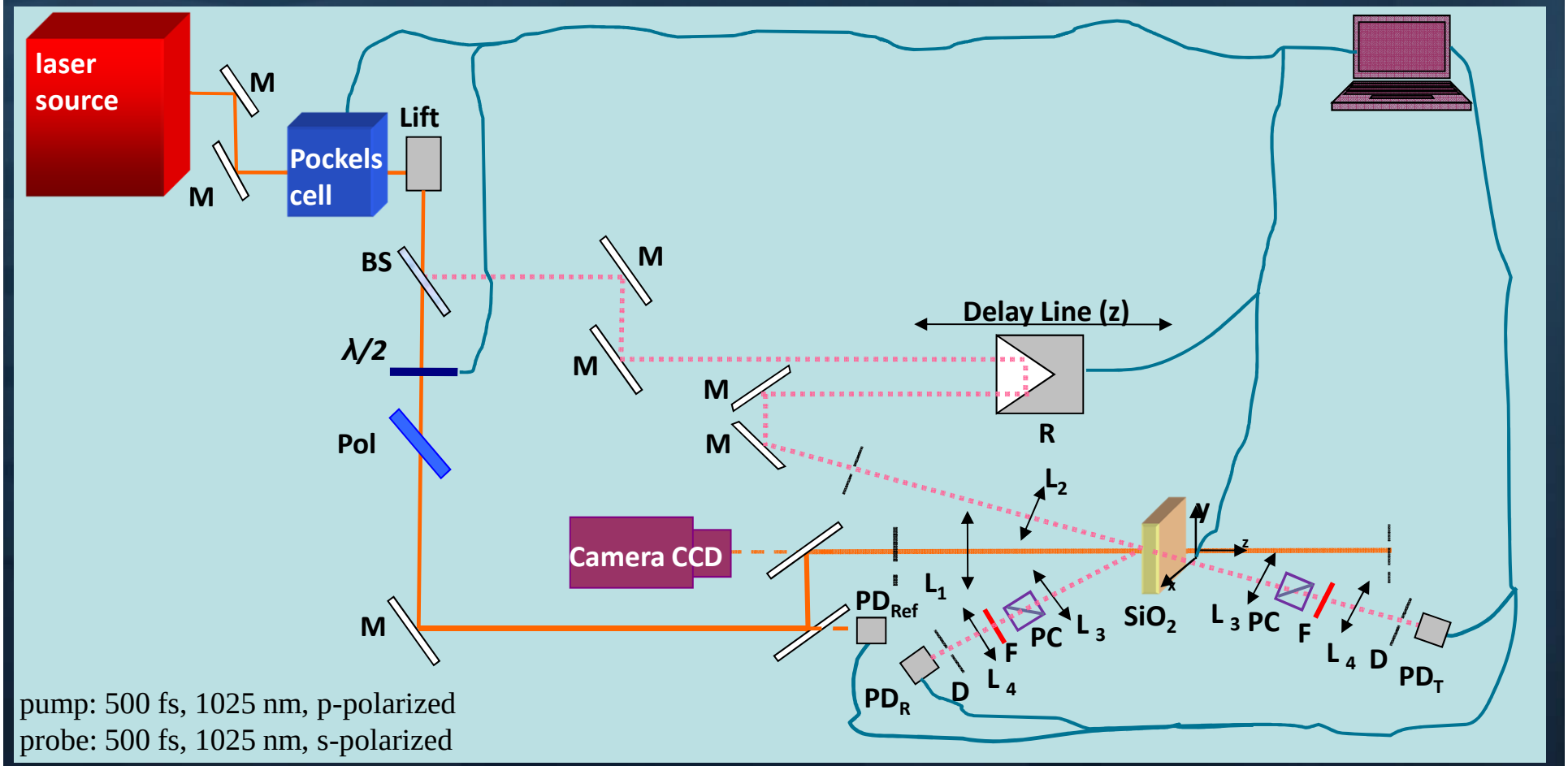
Damage threshold: 4.4 J/cm^2

Ablation threshold: 5.9 J/cm^2

*A. Q. Wu, I. H. Chowdhury, and X. Xu, Phys. Rev. B 72, 085128 (2005)

**Ch. Ziener, P.S. Foster, E. J. Divall, C.J. Hooker, M. H. R. Hutchinson, A. J. Langley and D. Neely, J. Appl. Phys. 93(11), 768 (2003)

F- Energy balance (plasma mirror effect): self-pump and pump – probe experiments (see also M. Lebugle poster)

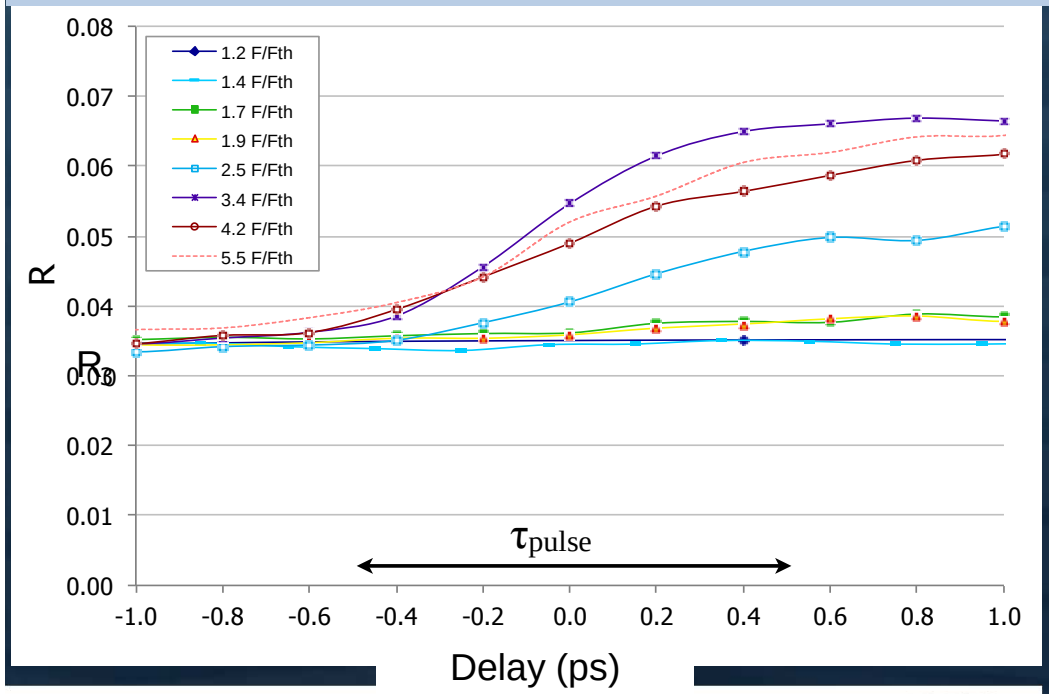


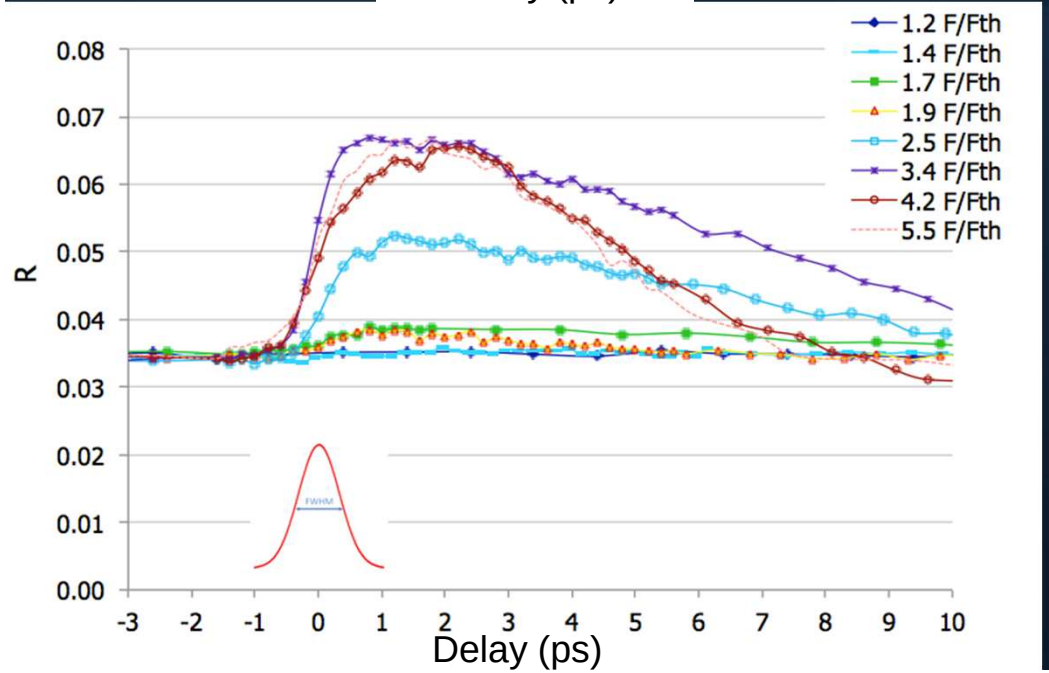
* Measurement of R and T as a function of F

⇨ **Absorption: A = 1 – R – T**

* **Keypoint:** spectral filter, spatial filter and polarization filter for ensuring the precise measurement of the reflected and transmitted probe beams

Dynamics of plasma mirror (experiment @ 500 fs)



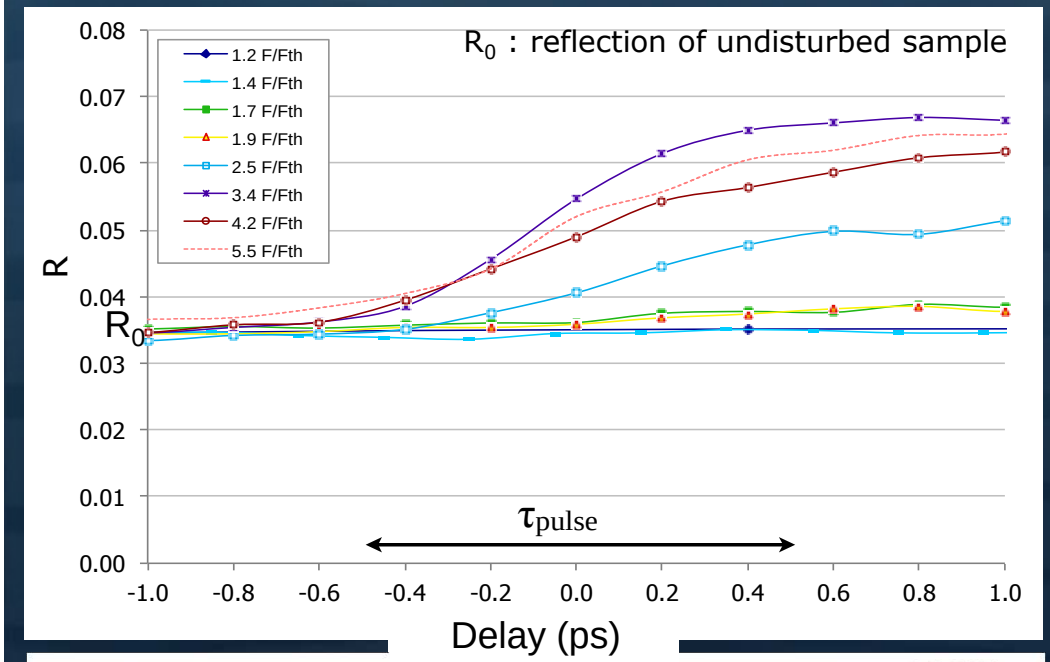


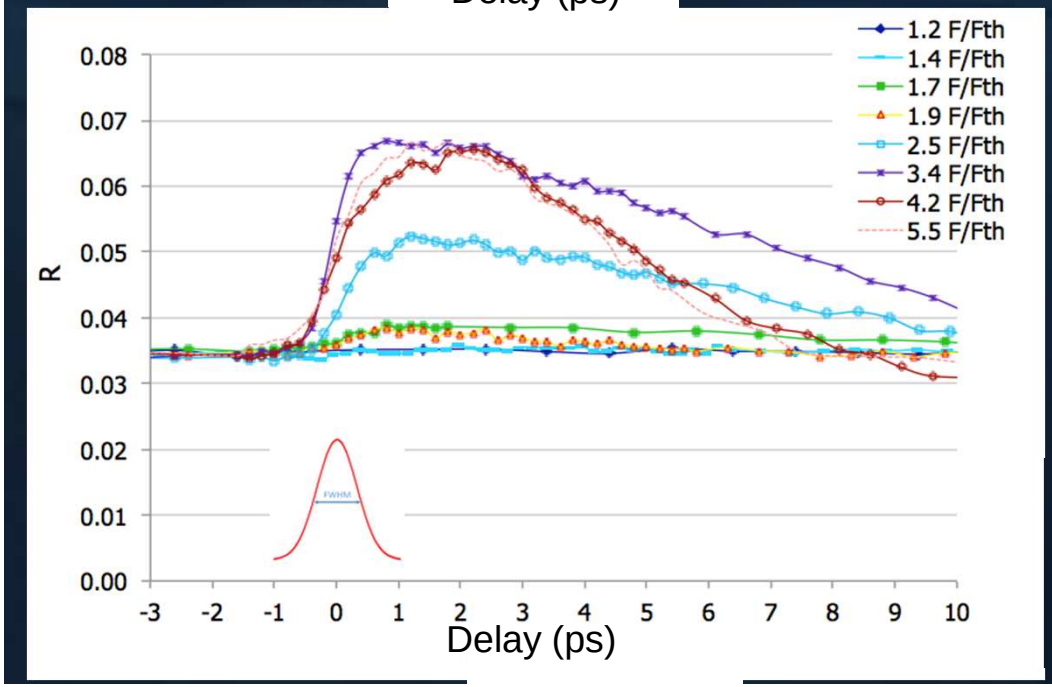
* Convolution of pump with probe in experiment (560 fs FWHM)
Time increment of 200 fs

* R_0 : reflection of undisturbed sample

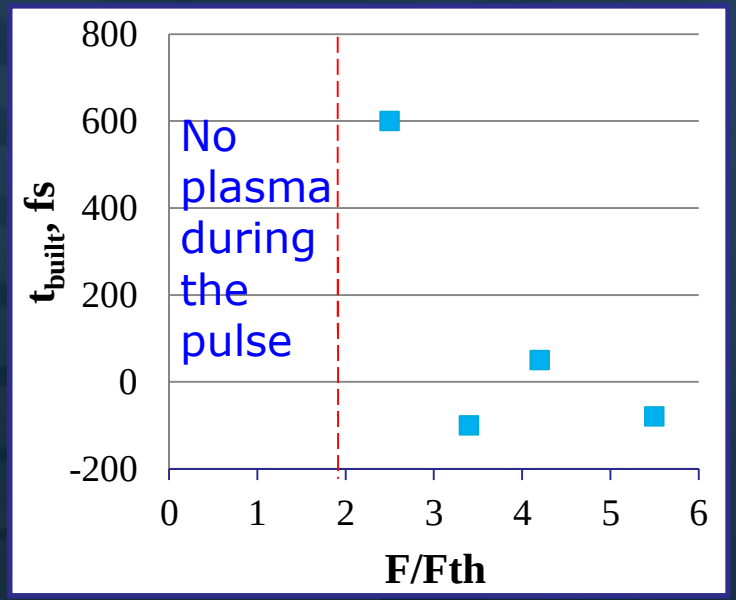
- Increase of R is fluence sensitive
- Increase of R up to 2 times its initial value during the pulse
- Relaxation to initial values (R_0) of R up to $1.9F_{th}$
- Slow relaxation of R
- Change of slope for different F
- Saturation of R below R_0. The dynamics of this decrease is faster at high than at low and medium fluence.

Dynamics of plasma mirror (experiment @ 500 fs)





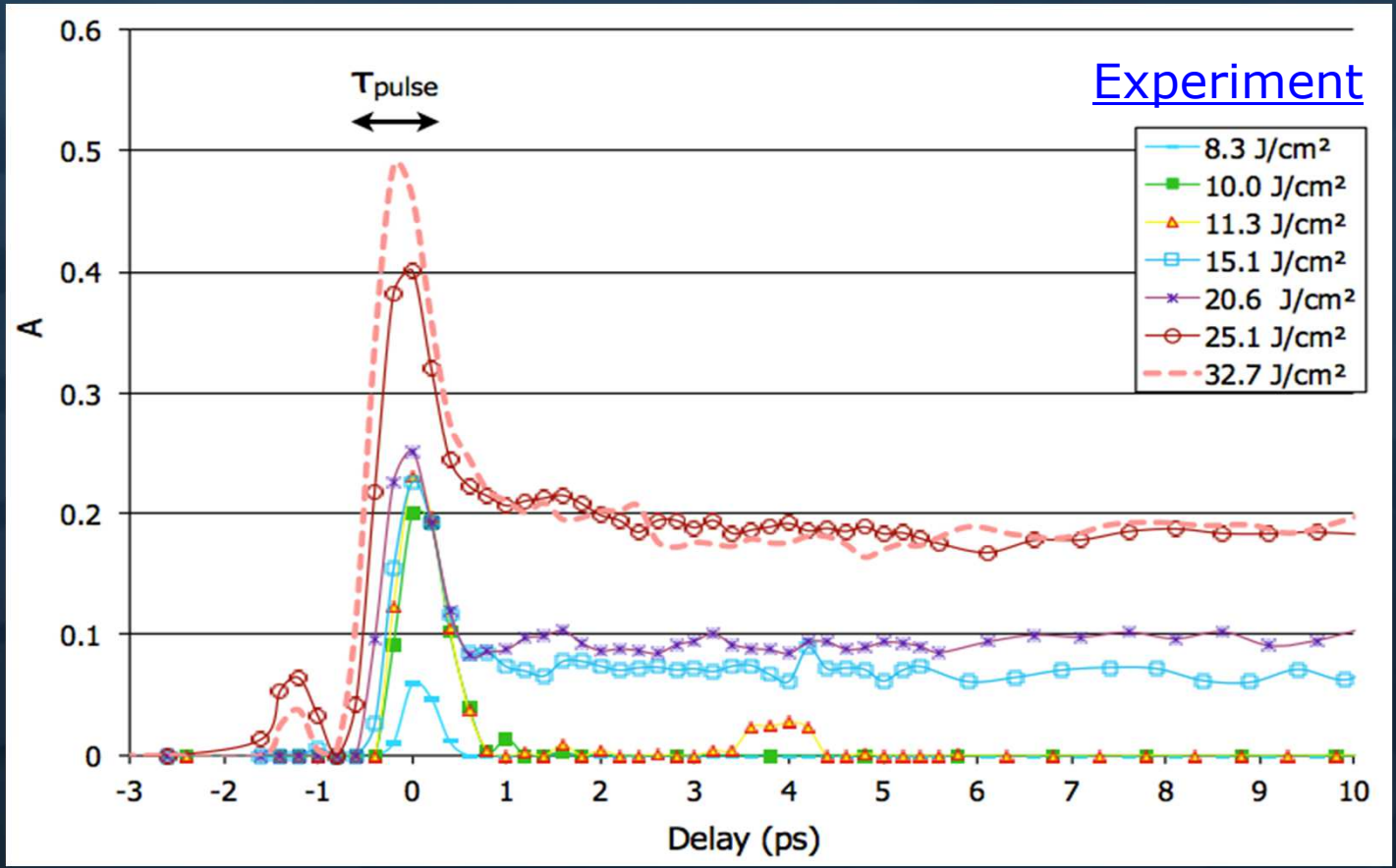
t_{built} : increase of 50 % from the max of R



- The onset of reflection change starts earlier at high fluence
- Plasma mirror formation during the pump pulse from 2 F_{th}

Study with smaller pulse duration currently in progress (see M. Lebugle poster)

Dynamics of absorption A = 1- R – T, absorption duration

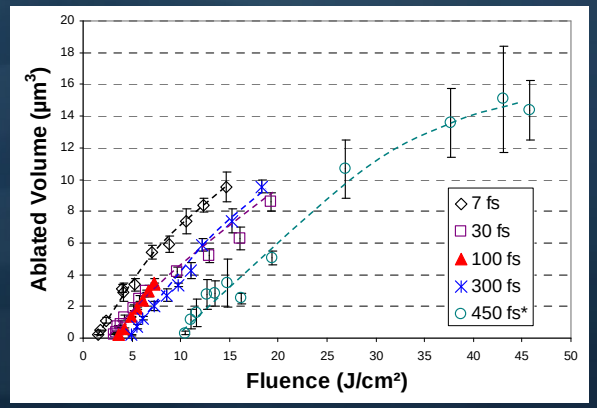


- Low fluences: $\tau_{absorption} < \tau_{pulse}$ (strong absorption starts at the second part of the pulse)
- High fluences: $\tau_{absorption} \approx \tau_{pulse}$

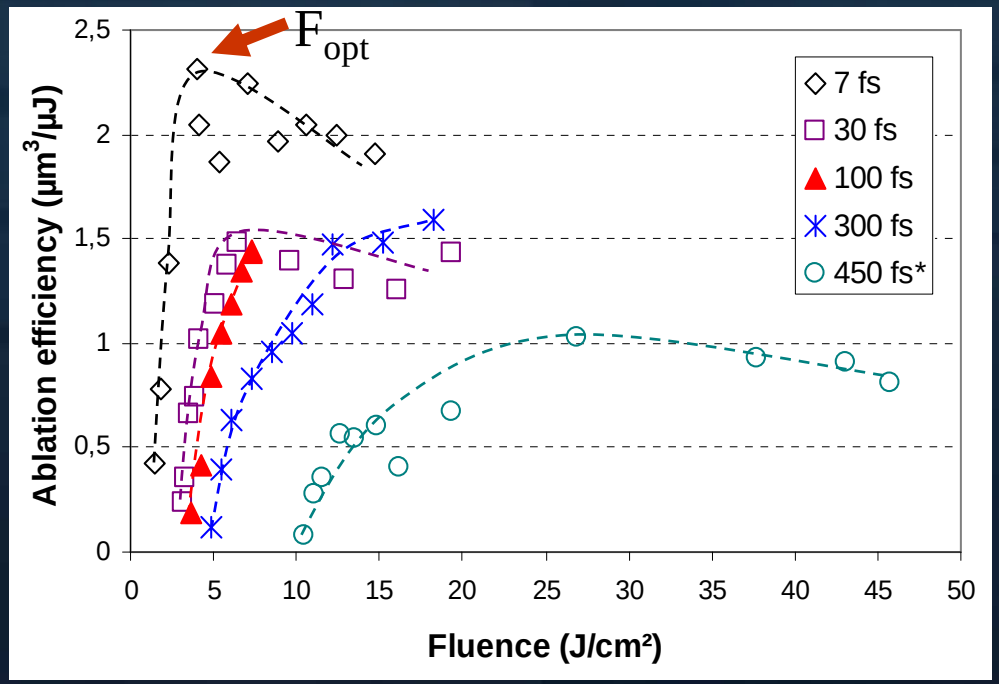
* Absorption saturation after the pump pulse is a result of both plasma (electrons, ions) effects and structural modification

G- *Ablation* vs pulse duration and applied fluence ($F \leq 10\ F_{th}$): efficiency

Remind: SiO_2 - suprasil, single shot, surface experiment



$$\textit{ablation efficiency} = \frac{\textit{ablated volume}}{\textit{incident pulse energy}}$$

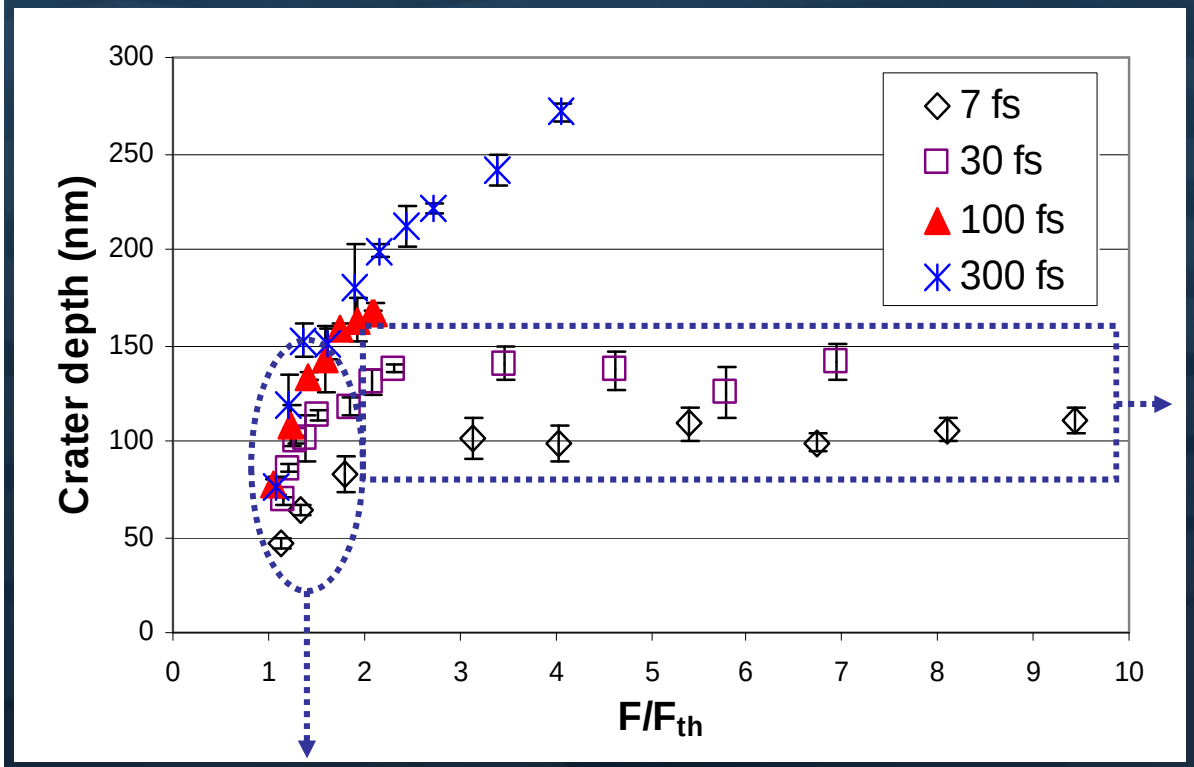


- Identification of an **optimum fluence F_{opt}** for each pulse duration
- This originates from the development of a free-electrons plasma becoming overcritical for increasing fluences and reflecting the late part of the beam ("**ultrafast optical shutter**" UOS, B. Stuart et al., PRB, 1996, etc.)
- The UOS effect is quicker and more efficient (closure) for ultrashort pulses (see also crater depth evolution)?
- Other possibility: NL propagation effects (in discussion, see M. Lebugle poster)

The shorter is the pulse, the higher is the ablation efficiency

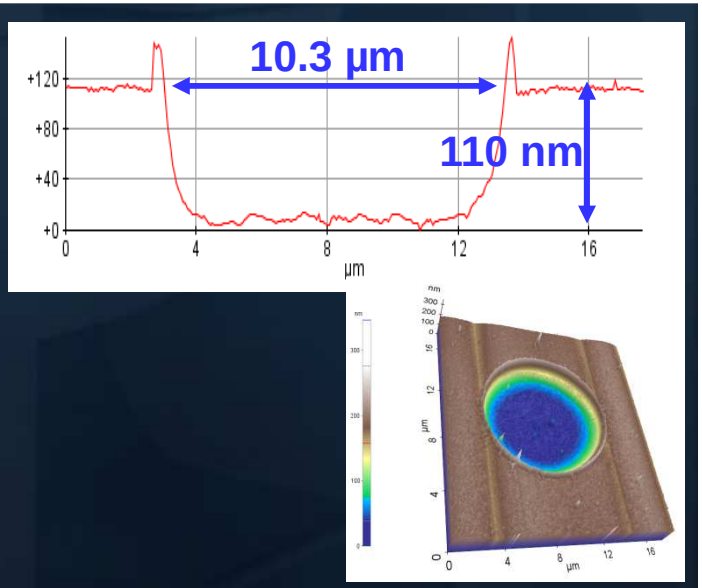
* Utéza et al., JLMN 5, 2010; APA, 2011

G- Crater depth vs pulse duration and applied fluence: Axial selectivity depends on intensity (balance pulse duration / fluence)



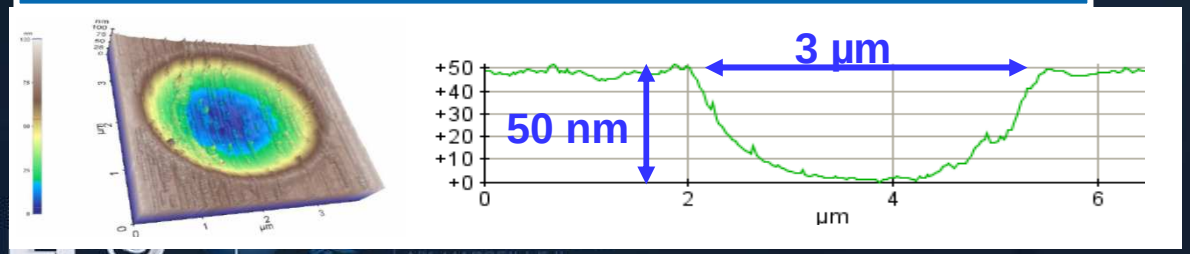
The UOS also drives a transition concerning the **crater morphology**, from Gaussian-like ($F < F_{opt}$) to top-hat shape ($F > F_{opt}$)

« saturated absorption window »
→ reproducibility
→ high control
→ top-hat shaped crater



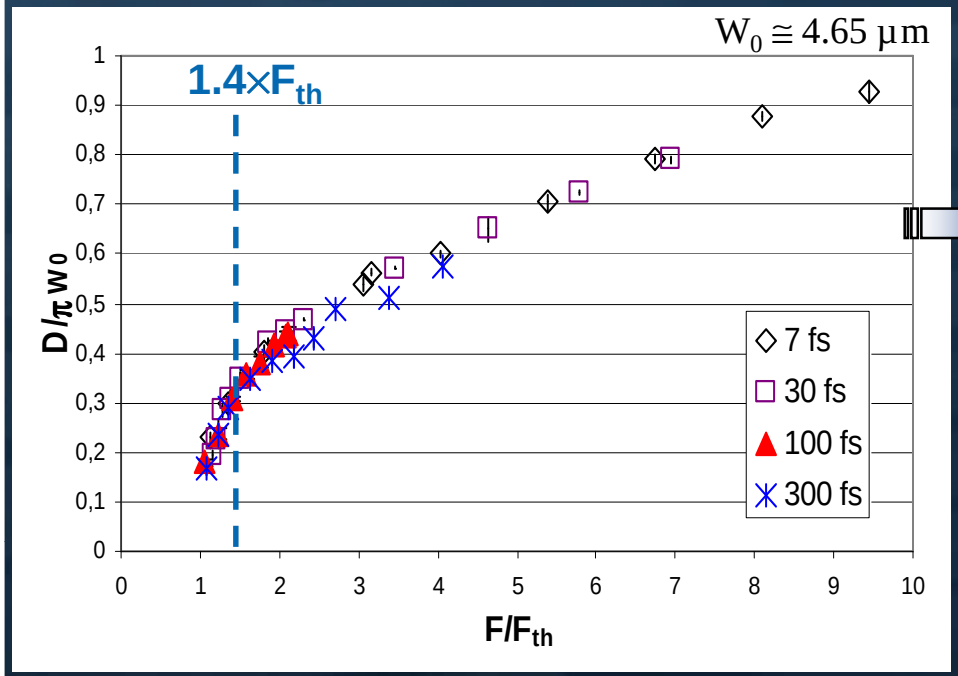
7 fs, 5.4×F_{th} (7.1 J/cm²)

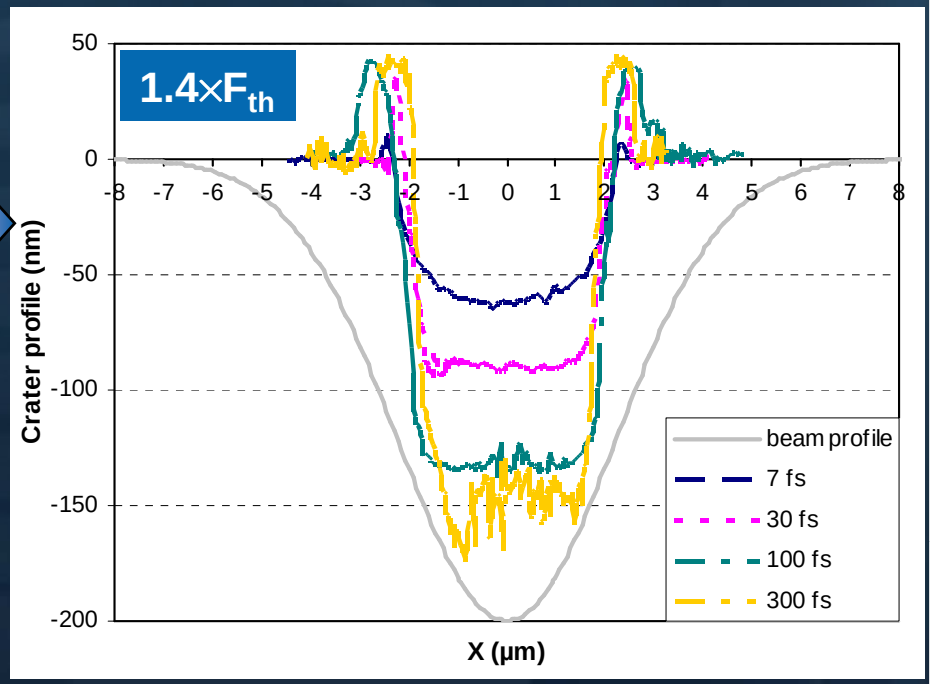
« selective processing window »
→ resolution ~10 nm with fluence (for a given pulse length)



7 fs, 1.1×F_{th} (1.5 J/cm²)

G- Crater diameter vs pulse duration and applied fluence: Transverse selectivity depends on fluence





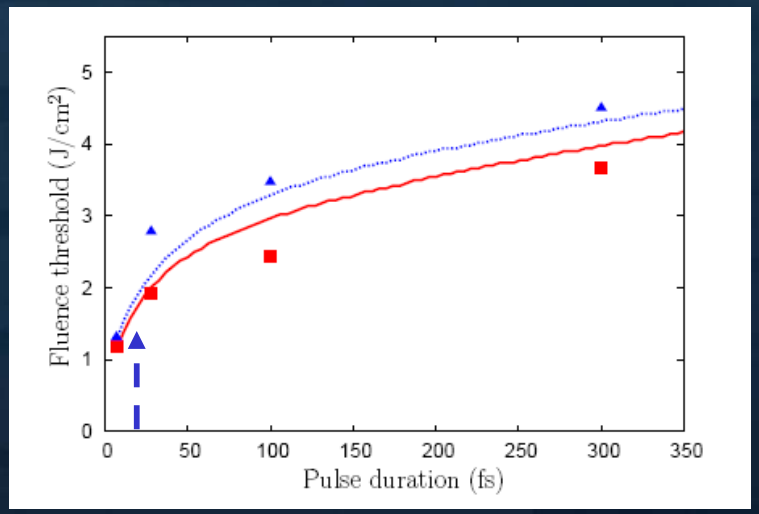
The crater diameter (transverse selectivity) depends only on normalized fluence, not on pulse length

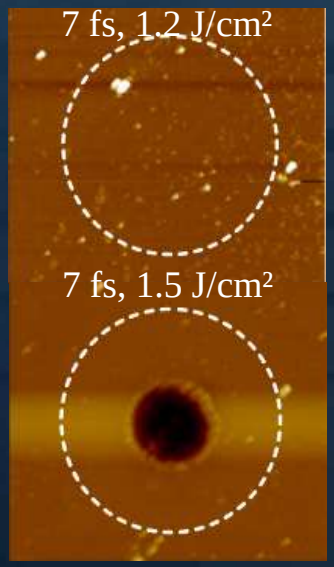
Tuning of pulse duration enable to control the crater depth while keeping its diameter fixed

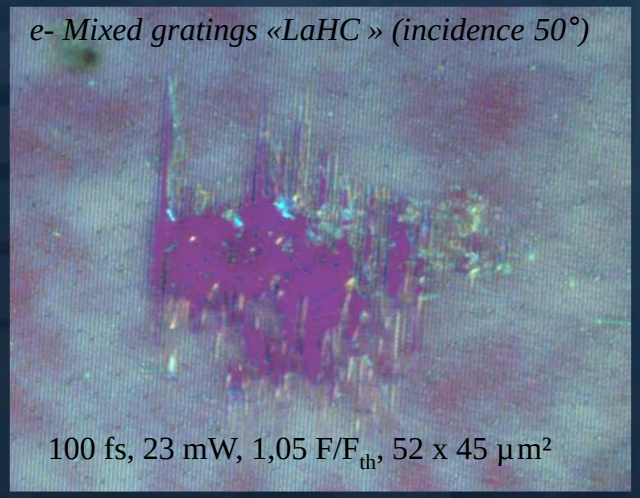
Conclusions: Laser-damage test bench in operation in LP3

➢ LIDT test-bench (and also ablation): ~ 10 fs – 500 fs

* LIDT of materials and optical components (ILE/Eli, CNRS, SESO, HJY, AT, etc.)
* Safety zone and fluence working window for high quality micro-machining process







➢ Fundamentals of fs Laser – matter Interaction:

* Ionization mechanisms and energy relaxation
* Energy balance, importance of electronic effects at ultrashort pulse duration
* laser – plasma Interaction (dynamics of plasma mirror)

A wealth of applications: laser damage, micro-processing, ophtalmology, etc.

Note: LP3 is in Laserlab

To progress, we should increase the number of observable information…!

We also should better measure and modelize and combine both information!

- Use diagnostics able to follow the band structure (lattice) of matter (Time-resolved X-ray, acoustics)

- … and in the same time the **transient (ps)** optical properties (dielectric function – optical probes) and plasma characteristics (interferometry, spectroscopy)

- Hire « atto people »: with attosecond pulse, we have the shorter event for optimal investigation !

- Measure also carefully the post-mortem characteristics (thresholds, ablation and morphology details)

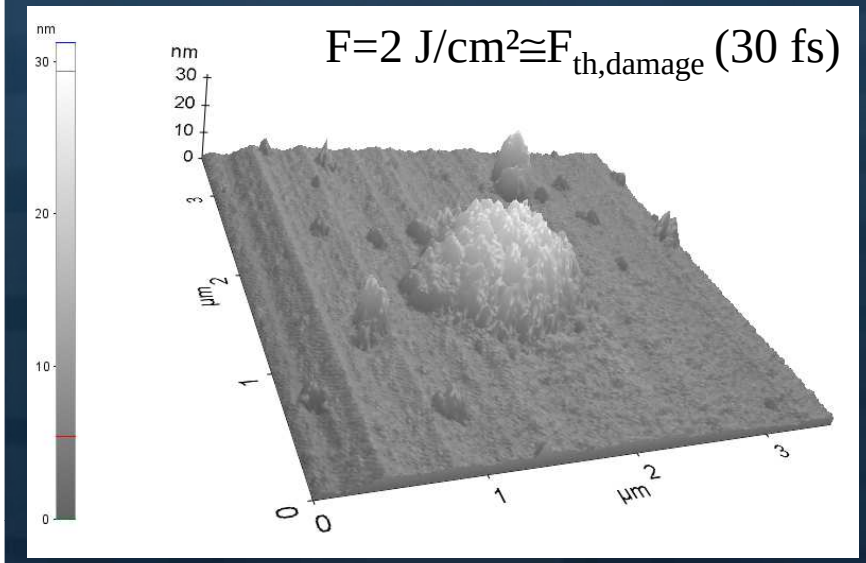
-**Towards predictive models** (threshold, ablation depth, etc.)
- Consider band structure evolution ($\sigma_{cross\ section}$, m^*, υ_{coll}, etc.)
- Consider nonlinear effects (NLES)
- 3D models to link « energy absorbed » and « ablated volume »

Thank you for your attention

*Acknowledgments and funding supports:

ANR funding « Festic » and « Nanomorphing » contracts, CNRS, Europe, Ministry of Research, PACA region, BDR Department Council, City of Marseille

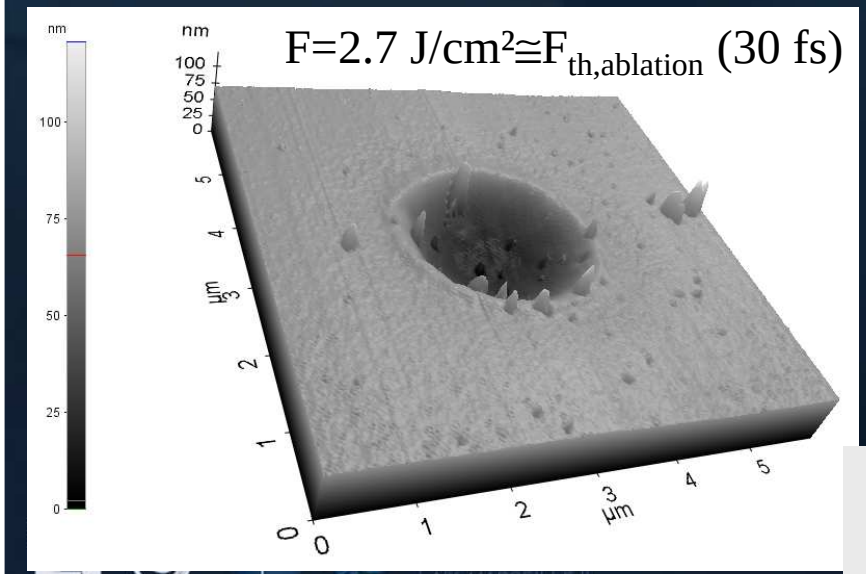
4- Ablation and damage thresholds vs pulse duration (< 10 fs – 300 fs): **criteria are chosen in accordance with experiments**



Damage criteria:

$T_i = T_{melting}$

(SiO_2: 1800 K)



Ablation criteria: $T_e \geq T_{ec}$

Cohesion Temperature : $T_{ec} \leftrightarrow T_e$
For which isotherm (T_e,T_i) passes by non-equilibrium critical point ($\partial P/\partial \rho = \partial^2 P/\partial \rho^2 = 0$):
$T_{ec} = f(T_i, n_e)$ (e^- and lattice effects)

$$P = P_i(T_i, n_e) + P_e(T_e, n_e)$$

$P_i(T_i, n_e) = P_{SES}(T_i, \rho_s) - P_e(T_i, n_e)$ (Table 7387 Sesame, LANL)