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

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 mechanisms
A- Laser Induced Damage Threshold (LIDT): the “Big” problem!

For instance, SiO$_2$: huge LIDT data dispersion (different motivations, etc.)

Data dispersion comes from:
- 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- Let’s clarify: Definitions and Methodology*

Structural modification

(I < 10^{13} \text{ 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 \( \rightarrow \) 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** ($I \sim 10^{13} \text{ W/cm}^2$)
- Morphology change on the surface
- No matter ejection

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

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**Damage ↔ LIDT**
(Laser-induced Damage Threshold)
→ qualitative analysis (visible damage): optical microscopy and statistical approach

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* 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)

![Graph showing damage probability vs. fluence on a log-log scale](image)

- High threshold
- Low threshold

Fluence (J/cm²)

![Image of laser damage site](image)

10 µm
**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**

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

- Ejection of matter

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**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

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* F. Korte et al., OE 2000; N. Sanner et al., APA 2009
Laser Ablation - LIAT (best evidenced with AFM)

Fluence (J/cm²)

Ablated Volume (µm³)

- Experiment
- Numerical fit

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

- Laser (pulse duration, fluence): $\tau_{\text{pulse}} : 500 \text{ fs (operational, AS laser @ 1025nm)}$
  and $10 \text{ fs } \rightarrow 400 \text{ fs + OPA (ASUR)}$; $F : 1 - 20 \text{ J/cm}^2$, linear pol.
- Single-shot/Nshots regime (1on1, Non1), air/vacuum ambiance
- Diagnostics: ex-situ: AFM, optical microscope; in-situ: pump-pump, pump-probe
- Materials ($\text{SiO}_2$, $\text{Al}_2\text{O}_3$, $\text{Ti}^{3+}:\text{Al}_2\text{O}_3$) and components (gratings, mirrors, etc.)
ASUR laser system (10 TW@100 Hz) in LP3

ASUR facility*
(laser developed by AT company)

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

- Minimal distance not to have material surface contamination ($\gg \omega_0$)
- Space consuming experiment (minimization of the experiment area)
- High level of metrology (beam size and focusing geometry, energy, material,...)
- Reference and calibration samples

Series of $N \times 1$ shot at $F$ fixed

Statistical approach to define damage threshold

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

$\Delta F \leftrightarrow$ determinism strength
* Laser (pulse duration, fluence): $\tau_{\text{pulse}}: \text{7 fs} \rightarrow \text{300 fs}$; $F : 1 - 20 \text{ J/cm}^2$, linear pol.
* Focusing: parabolic mirror EFL =50 mm ($w_0 = 4.65 \mu\text{m}$, $2z_R = 140 \mu\text{m}$)
* Single-shot regime; Surface experiments
* Sample: superpolished (Ra< 0.2 nm) high-purity suprasil ($\alpha$-SiO$_2$) (< 0.065 ppm)
* Diagnostics: AFM, optical microscope
Strong LIDT reduction at ultrashort pulse duration < 25 fs (importance of tunnel ionization and electronic effects)

Avalanche dominates laser absorption at « long » timescale ($\tau_{\text{pulse}} > 50$ 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 \((\sigma_6 = 2 \times 10^{25} \text{ cm}^{-3}\text{s}^{-1} \text{(cm}^2/\text{TW})^6, C. Mézel, POP, 2008)\)
- 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 \((\tau_{\text{pulse}} > 50 \text{ fs})\)

\textbf{but} 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

Strong deterministic character for « long » pulses $300 \text{ fs} > \tau > 30 \text{ fs}$

$\text{Rq: } >>$ transition slope ($\Delta F > \sim 10 \text{ J/cm}^2$) observed for nanosecond pulses ($\text{SiO}_2$, similar operating conditions)

Very strong determinism for pulses $< 30 \text{ fs}$
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.

Multiphoton ionization is more sensitive to laser and matter fluctuations.

* 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*

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

Precious indications for material processing*

* Utéza et al., JLMN 5, 2010; APA, 2011
* Hoffart et al., JFO 33, 2010; OE 19, 2011
For illustration...

450 fs @ 1025 nm

(a) Epithelium
(b) Bowman's layer

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

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

7 fs – F = 1.2 J/cm²
7 fs – F = 1.5 J/cm²

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: $A = 1 - R - T$
• 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/cm²

Ablation threshold: 5.9 J/cm²

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* 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)

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

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

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

* 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

- 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 ($F < F_{\text{opt}}$) to top-hat shape ($F > F_{\text{opt}}$)

- **saturated absorption window**
  - reproducibility
  - high control
  - top-hat shaped crater

- **selective processing window**
  - resolution ~10 nm with fluence (for a given pulse length)

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$7 \text{ fs, } 1.1 \times F_{\text{th}} (1.5 \text{ J/cm}^2)$

$7 \text{ fs, } 5.4 \times F_{\text{th}} (7.1 \text{ J/cm}^2)$
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.

\[ D/W_0 = 1.4 \times F_{th} \]

\[ W_0 \approx 4.65 \mu m \]
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

![Graph showing fluence threshold vs. pulse duration](image1)

- **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)

![Image of mixed gratings «LaHC»](image2)

- **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 ($\sigma_{\text{cross section}}, m^*, \nu_{\text{coll}}, \text{etc.}$)
  - Consider nonlinear effects (NLES)
  - 3D models to link « energy absorbed » and « ablated volume »
Thank you for your attention

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Damage criteria: \( T_i = T_{\text{melting}} \) (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)

\[ P = P_i(T_i, n_e) + P_e(T_e, n_e) \]

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