Laser : le couteau suisse des procédés haute résolution:

Micro-procédés 3D dans les matériaux transparents par laser femtoseconde

David Grojo
Lasers, Plasmas et Procédés Photoniques (LP3)
Femtosecond laser modification of transparent materials

...3D localization of energy depositior.

4/10/2017 – David GROJO
Femtosecond laser 3D nano/micro-fabrication and surgery

...but only in transparent matter including dielectrics and some polymers?
Non-linear Ionization inside Band gap solids

**Dielectrics**

- Band-Gap: $\Delta_D \approx O(10 \text{ eV})$

- $h\nu_{\text{las}} < \Delta_D$
  - Ti:Saphire fs laser
  - Fund.: $\lambda_{\text{las}} = 800 \text{ nm} \ (1.55\text{eV})$
  - SHG: $\lambda_{\text{las}} = 400\text{nm} \ (3.1\text{eV})$

- Ultrafast optical spectroscopy and imaging with visible light

**Semi-conductors**

- Band Gap: $\Delta_{SC} \approx O(1 \text{ eV})$

- $h\nu_{\text{las}} < \Delta_D$
  - Optical Parametric Amp. (OPA)
    - $\lambda_{\text{las}} > 1.2\mu\text{m} \ (h\nu_{\text{las}} < 1 \text{ eV})$

- **Appropriate Methods?**

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"Similar MP processes must arise in semiconductors with focused SWIR fs pulses"
3D-fs = Complex interplay between NL phenomena

\[ \nabla \times \nabla \times E = -\frac{1}{c^2} \frac{\partial^2 D}{\partial t^2} ; \quad D = \varepsilon E \]

3D Maxwell equations

divD = 0

\[ \varepsilon = \varepsilon_{bond} + \varepsilon_{free}(n_e) \]

Intensity/time/temperature dependent dielectric function

Mainly nonlinear optics in progressively ionized matter before reaching the target (focus) !!

The space-time dependent permittivity can be simply derived using a Drude model but \( n_e(t) \) must account for all the contributions in ionization (NL)

Non-Paraxial case (strong focusing) requires « exact » vectorial Maxwell solvers leading to unrealistic computational resources in most cases

Alternative transformation optics solution: PRL 117 (2016) 043902

“Propagation simulation required because of coupling through the permittivity function”
Micro-procéédés 3D dans les matériaux transparents par laser femtoseconde

Introduction

1. Maitrise des impulsions femtosecondes fortement focalisées dans la matière
2. Propagation non-linéaire et « clamping » des conditions d’interactions
3. Microplasmas de faible densité et applications
4. Claquage optique et applications de structuration 3D
5. Le défi de la microfabrication 3D dans le silicium.

Conclusions et perspectives
Controlling extreme space-time localization of light in matter

NA=0.45

NA=0.65
Ideal focusing with an objective

From textbook *Principle of Optics (7th ed.)* p484-494

1959 Born and Wolf

45° (NA=0.7)

Numerical


« Easier to turn to numerical simulations for non ideal cases (non-ideal plane wave at the entrance, focusing in medium which is not air, etc...) »
Focusing a Gaussian Beam with an objective

Filling parameter: \( \beta_T = R/\omega_0 \)

**Designed case**

\( \beta_T = 0 \)
\( \beta_T = 1 \)
\( \beta_T = 2 \)

**Gaussian optics**

\[ I(r, z) \propto \left( \frac{\omega_0}{\omega(z)} \right)^2 e^{x_p(z)} \frac{-2r^2}{(\omega(z))^2} \]

\[ \omega(z) = \omega_0 \sqrt{1 + \left( \frac{z}{z_R} \right)^2} \]

\[ z_R = \frac{n \pi \omega_0^2}{\lambda} \]

**Parameters:**

- NA
- \( \lambda \)
- \( \beta_T \)

NA=0.7; \( \lambda = 0.8 \mu m \)


“With a decreasing filling factor, the apparent NA decreases and solution tends toward Gaussian beam optics”
Focusing inside a material: spherical aberration

Filling parameter:
\( \beta_T = \frac{R}{\omega_0} \)

Parameters:
- NA
- \( \lambda \)
- \( \beta_T \)
- \( n, \text{ depth} \)

\[ \lambda = 0.8 \mu m; \quad n = 1.5 \text{(Glass)}; \quad \beta_T = 1 \]

\[ \lambda = 1.3 \mu m; \quad n = 3.5 \text{(Si)}; \quad \beta_T = 1 \]

« The focus shifts and spreads along the optical axis by about a factor \( n \) and spherical aberrations must be a concern »

Main geometrical characteristics

Parameters:
- NA, $\lambda$, $\beta_T$, $n_{mat}$, depth
- FWHM $\approx \lambda / 2NA$
- $b \propto n.\lambda^2$

Shape affected by aberrations

- $\omega^\text{beam} >> R$ ($\beta_T << 1$)
- $\omega^\text{beam} << R$ ($\beta_T >> 1$)

- $d = 1.22 \frac{\lambda}{NA}$
- $l/d = \left( \frac{3 - 2 \cos(\theta) - \cos(2\theta)}{1 - \cos(\theta)} \right)^{\frac{1}{2}}$
- [JOSA A 16, 2658(1999)]

- $2\omega_0 = \frac{2\lambda}{\pi \theta}$
- $b/(2\omega_0) = \frac{\pi}{\lambda} \omega_0$

- $NA = \sin(\theta)$

«In practice, not only on the spatial characteristics but also the temporal characteristics and non-linear effects acting on the beam used for writing»
Group velocity dispersion

\[ E_0(t) = \int E(\omega)e^{-i\omega t} \]

\[ E_0(t) = \int E(\omega)e^{-i\omega t + \phi(\omega)} \]

\[ \varphi(\omega) = \varphi(\omega_0) + \varphi'(\omega_0)(\omega - \omega_0) + \frac{1}{2}\varphi''(\omega_0)(\omega - \omega_0)^2 + \frac{1}{6}\varphi'''(\omega_0)(\omega - \omega_0)^3 + \ldots \]

Linear Phase shift  
Pulse is delayed but undistorted

Quadratic phase shift  
Pulse is stretched/linear chirp

Higher order dispersive effects

«Not the shortest possible pulse duration at the focus unless GVD is precompensated»
Pulse stretching estimations

For unchirped transform limited Gaussian pulses:

\[
\tau = \tau_0 \sqrt{1 + \left(4 \ln(2) \cdot \Phi'' / \tau_0^2 \right)^2}
\]

GVD in \( \text{fs}^2 \), Initial pulse duration FWHM

For instance
- 10 fs pulses at 800nm will increase its duration by 40% after only \( \sim 2.5 \text{m} \) in air (GVD=0.2fs\(^2\)/cm) or \( 1.4 \text{mm} \) in glass (GVD=360 fs\(^2\)/cm).
- 100 fs pulses are not stretched with <1000 fs\(^2\) (about 2-3cm in glass)
Dispersion compensations strategies

Materials

\[ \phi'' > 0 \]

Added

\[ \phi'' < 0 \]

Gratings

Prisms

Chirped mirrors

« One must be also careful with higher order dispersion for the shortest pulses »
Practical solution with CPA systems

Adjust compressor gratings (CPA systems) to look for maximum NL absorption

Source: PRL 102, 083001 (2009)

NA=0.25
λ=800 nm
SiO₂

«Adjust compressor gratings (CPA systems) to look for maximum NL absorption »
Space-time confinement of laser light inside matter

"Technical solutions exist for compensations for linear interactions... nonlinear space-time-spectrum distortions are expected"

Sources: www.microscopyu.com and CVI Technical notes
Nonlinear propagation and intensity clamping
Intensity dependent index = positive and negative lensing on Gaussian beams

Third order nonlinearity

\[ P^{(3)}(t) = \varepsilon_0 \chi^{(3)} E(t)^3 \]

\[ n(r, t) = n_0 + n_2 I(r, t) \]

Near critical power: \( P_{cr} = \frac{\pi (0.61)^2 \lambda^2}{8n_0 n_2} \)

Above ionization threshold near critical plasma density: \( N_{cr} = \frac{\varepsilon_0 m_e \omega^2}{e^2} \)

“Self-trapping of intense light at equilibrium conditions”

Kerr effect

Filamentation

Self-focusing

Plasma-defocusing

Courtesy S. Tzortzakis
Multiple Filamentation $P \gg P_{cr}$

"Built from modulational instabilities or growing up from small fluctuations in the beam intensity profile."

Source: PRL 92, 225002 (2004)
Self-Limited Intensity of a Filament

\[ \epsilon_{NL} + Re(\epsilon_{\text{Plasma}}) = 0 \equiv n_2I - \frac{N_e}{2Ncr(1 + \nu^2/\omega^2)} = 0 \]

Taking a simple MPI description and \( \nu \approx \omega \)

\[ N_e \approx \sigma_K I^K t_P \]

We find an equilibrium intensity

\[ I \approx \left( \frac{2n_2N_{cr}}{\sigma_K t_P} \right)^{1/(K-1)} \]

Plasma density (free carrier)

Electron-ion collision rate

Example

**Si at 1300 nm**

- K=2, \( \sigma_2 = 3.3 \times 10^9 \text{ cm/W/s} \)
- \( N_{cr} = 6 \times 10^{20} \text{ cm}^3 \)

Example

<table>
<thead>
<tr>
<th>Material</th>
<th>Intensity (W/cm²)</th>
<th>Wavelength (nm)</th>
<th>Pulse duration (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>( 1.2 \times 10^{12} )</td>
<td>1300</td>
<td>( \approx 100 )</td>
</tr>
<tr>
<td>SiO₂</td>
<td>( 10^{13} )</td>
<td>800</td>
<td>800</td>
</tr>
<tr>
<td>Air</td>
<td>( 10^{13} )</td>
<td>800</td>
<td>800</td>
</tr>
</tbody>
</table>

"There is a limitation on the intensity that can be delivered by simply increasing the incoming pulse energy because of this intensity clamping."
Intensity clamping by high-order NL absorption

“lawn mower” modeling

$I_{th}=1.5 \times 10^{13} \text{ W/cm}^2$

Experiment

“lawn mower” modeling

Fused SiO$_2$
43fs, NA = 0.25, $I_{max} \approx 5\times10^{13} \text{ W/cm}^2$ (vacuum intensity)

Energy distributions (J/cm$^3$)

1000 nJ
250 nJ
100 nJ

Source: OE 13, 3208 (2005)

“Tends to maintain the power below the self-focusing threshold ($\approx 1.5 \text{ MW for SiO}_2$) and lead to nearly uniform excitation in the pre-focal region”
It is hard to exceed significantly the plasma critical density. Plasma effects completely prevent modification in silicon with IR fs pulses in standard configurations.
**Important:** Attempts to deliver more intensity than the clamping level in the bulk degrade the spatial resolution rather than enhance the deposited energy density.
Effects of fs laser induced low-density plasma and applications
Types of applications with low density plasmas

Ultrafast signal processing

Bio-degradation

3D nano/microfabrication

« ... which are not direct micromachining or surgery above breakdown threshold... »
Two-photon lithography (positive)

Near IR fs-pulses

Resin

Sub-100nm resolution

Radicals + weakly cross-linked polymers or monomer (liquid state) → Highly cross-linked polymers (solid state)

Applications. Complex nano-electromechanical systems, active and passive integrated photonic devices, complex architecture photonic bandgap crystals, remotely controllable micromachines, microfluidic and medical devices.

3D Optical Functionalization (Fluorescence writing)

Application:
High capacity optical recording

Figure 4. Recorded images viewed by confocal fluorescence microscopy ($\lambda_{exc} = 405 \text{ nm}$) of three French Nobel laureates in physics: Gabriel Lippmann (1908), Alfred Kastler (1966), and Claude Cohen-Tannoudji (1997). a) The images are 100 × 100 pixels. The pixel spacing is 3 µm, the pixel diameter is 2 µm, the layer spacing is 20 µm, and the encoding is 256 gray levels. The image size is 300 µm × 300 µm. The fluence was kept constant (5 J cm⁻²) and the number of pulses was varied from $10^2$ to $10^3$. b) Side view of the recorded images. No cross-talk between the layers is observed.


“... with specially designed and synthesized materials.”
3D waveguide writing
Repeated bond breaking causing a molecular rearrangement and a local uniform change of the refractive index

\[ \Delta n \approx +10^{-3} \]

Densification, \( \Delta n \approx +10^{-3} \)

\( \frac{5}{6} \) member rings

\( \frac{3}{4} \) member rings (visible in Raman spectra)

Source: Optics Letters 26 (2001) 1726

- Quantum Information Science (QIS)
- Ultra-stable optical devices (e.g. spatial)
- Specific telecommunication systems
- Lab-on-chip hybrid systems

« Low loss waveguides in various glassy materials and no need of material preparation. Flexibility meeting the requirements of challenging applications »
3D waveguide writing for QIS

Quantum circuits requirements

- Low transmission losses 0.1dB/cm
- Embedded – highly stable nested interferometers
- Complex designs requiring true 3D capabilities
- « Cheap » flexible prototyping (growing research)

“A rapidly growing field of research with the most advanced demonstration based on fs-laser writing but also other field of application as: specific telecommunication devices and spatial ultra-sable integrated optics devices applications”
Optical Breakdown and microfabrication applications
Femtosecond Laser writing inside Fused SiO$_2$

- **Uniform index change, $\Delta n \sim +10^{-3}$**
- **Nanoscale cracking, $\Delta n \sim -10^{-2}$**

- $\tau_{\text{laser}} = 50$ fs, < 200 nJ
- $\tau_{\text{laser}} = 130$ fs, 350 nJ

- Fused Quartz
- NA 0.65
- $\sim \lambda/2n$
- $E_{\text{field}}$

- C.B.
- V.B.

- $1.5$ eV
- $9$ eV
- $3$ eV

- $N_{\text{pulses}} = 10^5$
- $N_{\text{pulses}} = 10^4$
- $N_{\text{pulses}} = 10^3$

- 10 um
High refractive index decrease by nanostructuring

$R \ll \lambda$ = critical assumption

$\varepsilon_{\text{avg}} < \varepsilon$

“The apparent reactive index is below the average index of the structure”
High refractive index decrease by nanostructuring

Light energy preferentially resides in low index regions

\[ \Delta n \leq \frac{V}{V_0} (1 - n) \]

Scattering is kept to its minimum with small features

\[ \nu_{scat} \propto R^6 \]

(Rayleigh approximation)

\[ \frac{\nu_{scat}}{\Delta n} \xrightarrow{R \to 0} 0 \]

\( R << \lambda \) = critical assumption
Micro-Lensing

\[ n = 1.45 \]

Measured HWHM

\[ \Delta n/n = -1.8\% \]

After 10^7 shots

\[ \Delta n/n = -1.8 \pm 0.2\% \]

References:

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Form Birefringence

Behaves like an uniaxial crystal

$n_p = n \sqrt{\frac{R+1}{Rn^2+1}}$

$n_s = n \sqrt{\frac{R/n^2+1}{R+1}}$

$\Delta n_{\text{max}} / n = (n-n_p) / n \sim 2\%$

$(n_s-n_p) / n \sim 1\%$

Details in: Born and Wolf, *Principle of Optics*
In-situ Birefringence Diagnostic

\[ \Delta \Phi = 2\pi (n_s - n_p)d/\lambda_{las} \]

**Writing**

**Probing**

- Half-wave plate
- 45deg.
- ASE
- PD
- Polarization Analyzer

Optical data storage

1996 Mazur group, Harvard

Binary

Source: Optics Letters 21 (1996) 2023

2014 Kazansky group, Univ. Southampton

Parallel writing

Position 3D
+ Retardance (32 states=5bits)
+ Slow axis (8 states=3 bits)

5D

High capacity
360TB per disc

Long lifetime

T = 973 K
τ = 208 days

T = 673 K
τ = 8800 years

T = 462 K
τ = 1.3 x 10^9 years

T = 373 K
τ = 7 x 10^14 years

T = 303 K
τ = 3 x 10^20 years

Source: Physical Review Letters 112 (2014) 033901
Confined ultrafast microexplosion

“...multi-TPa pressure, ultrahigh temperature (Warm Dense Matter) and ultrafast cooling”

Source: PRL 96 (2006) 166101
Applications of ultrafast microexplosion

Discovery of new superdense materials

Sapphire ($Y = 400$ GPa) -> Discovery of bcc-Al
See: Nature Communications 2 (2011) 445

Si -> Discovery of new tretagonal polymorphs
Source: Nature Communications 6 (2015) 7555

Nano/microvoid generation for bulk machining: glass cutting, Microfluidic devices

Stealthy cut and/or controls of crack extension (dash line cutting)
Source: http://wophotonics.com

Source: Applied Physics Letters 97(8) 081102
The challenge of 3D processing inside silicon
Challenges with narrow gap semiconductors

Challenge #1: Producing and manipulating infrared ultrafast pulses and novel interaction diagnostics (λ > 1.2 µm for Si)

Challenge #2: Breaking the strong limit to energy space-time confinements inside Si

“Bulk modification impossible in the fs regime for all even the highest intensity tested... in conventional machining configuration [Appl. Phys. Lett. 105, 191103 (2014)] including Bessel [J. Appl. Phys. 117 (2015) 153105]”
Observing and controlling microplasmas inside Si

Time-resolved IR microscopy (fs pump and probe)

The conductivity can be turned on and off by ultrafast plasma formation

From local free-carrier injection to applications in microelectronics...

Novel experimental and numerical tools for studying the complex physical mechanisms taking place in the nonlinear interactions (propagation, ionization, plasma, etc.).

Nature Communications (2017) Accepté DOI: 10.1038/s41467-017-00907-8

Physical Review Letters 117 (2016) 043902
A strict clamping of the delivered intensities

“The minimal target!”

Surface damage threshold (meas.)

“An increase of the maximum fluence with NA but also a quenching below the threshold for modification for all focusing configurations*”

*Simulations reveal a strong depletion prior to the focus region because of 2-photon absorption and plasma defocusing/absorption play essential roles at very modest densities ($N_0 \propto 1/\lambda$) for in the long wavelength regime.

Nature Communications (2017) Accepté DOI: 10.1038/s41467-017-00907-8
Ultrafast optical breakdown in Si with hyper-focusing

NA=2.97, 20nJ/pulse

"Toward solid-immersion lens solutions for femtosecond laser 3D processing..."

Nature Communications (2017) Accepté DOI: 10.1038/s41467-017-00907-8
Femtosecond Laser Index Modification

NA=2.97, 20nJ/pulse

“The perspective of 3D Si Photonics but also an opportunity for extreme interaction experiments and potentially new types of material modifications”

Nature Communications (2017) Accepté DOI: 10.1038/s41467-017-00907-8
Toward Solid-Immersion Laser Processing?

SIL technology:
Succefully demonstrated in microscopy and lithography for resolution enhancement…
But rarely adopted due its practical complexity and the availability of other methods.

“Interestingly, SIL could find in laser processing an application where it provides super-resolution but also high-nonparaxiality which is a requirement for 3D femtosecond laser writing inside narrow gap materials. This may fully justify its complexity”

Note that at \( NA \approx 3 \), the spot size shrinks down to about \( \lambda/6 \approx 220 \) nm for \( \lambda=1.3\mu \text{m} \)...
Phase-imaging-controlled nanosecond laser writing

Positive index change waveguides demonstrated but improved control required to decrease losses !!!

Patent Application FR1655979 (27/06/2016); Optics Letters 41 (2016) 4875
Summary and Conclusions

- Main advantages:
  - Direct – Contact free/Near single step
  - Stability of embedded devices
  - Flexibility – Prototyping tool
  - Precision – sub-wavelength and high aspect ratio

- 3D nonlinear propagation in progressively ionized materials adding complexity in comparison to surface machining studies

- There are natural limits to the spatial resolution and the deposited energy densities (and so type of modifications) that can be achieved in femtosecond bulk interactions.

Microscopy concepts can be used for improved confinement and resolution: this has recently open the field to silicon

A unique opportunity for the development 3D photonics
Merci !

Questions: grojo@lp3.univ-mrs.fr