Mesures pompe-sonde pour le diagnostic des plasmas laser créés lors de la nanostructuration des matériaux

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-Laser technologies
-Carrier plasmas
-Pump-probe review
-3D structuring
Laser processing

Micro and Nano fabrication

- Key technological issue in manufacturing
2D surface engineering

Fabrication

- Drilling
- Functional Microstructures
- Tribology
- Mechanics
- Electronics
- etc.....
3D optical functions

Fabrication

• Embedded optical elements
• Embedded lasers
• Photonic crystals
• Quantum information
• Optofluidics
• Astrophotonics
• etc.....

Watanabe OE 2002
Marshall OL 2008
Szameit OE 2006
Sansoni PRL 2010
Cheng & Sugioka 2006
Thomson OE 2011
laser irradiation: enabling tool

Master the process via fundamental knowledge

Probing laser processes - a review of ultrafast probing
Material functions

Fabrication

• Involve lasers

But which lasers?
Long versus short pulses

Long pulse

\[ P = \frac{E}{\tau} = \frac{1\text{mJ}}{100\text{ fs}} = 10GW \]

Short pulse

femtosecond

\[ P = \frac{E}{\tau} = \frac{1J}{10\text{ fs}} = PW \]

Clark-MXR web site
Long versus short pulses

- **1 watt** = $10^3$ W
- **1 kilowatt** = 1 W
- **1 megawatt** = $10^6$ W
- **1 gigawatt** = $10^9$ W
- **1 terawatt** = $10^{12}$ W
- **1 petawatt** = $10^{15}$ W

**High intensities** (comparable with atomic fields)

- **Electron Motion**
- **Molecular Rotation /Vibration**

- **10^{-15} sec** (femto)
- **10^{-18} sec** (atto)

- **Electric bulb**
- **Electric cooker**
- **Car**
- **Flashlight**
- **Bomb**
- **Laser**
Laser action

Laser structuring = consequence of laser ablation

Energy coupling
Heating
Transformation
Ablation

CAN THIS BE CONTROLLED?
(achieve upgrade beyond the material limits?)

What is happening to the material: optically, structurally
Process dynamics

Laser ablation

Initial state

Excitation

Change

Probe

E. Mazur, Harvard University
Dynamics

in 1878

Optical stroboscopy
$10^{-1}$ to $10^{-3}$ seconds.

Ref. Eadweard Muybridge 1878
Dynamics

Optical stroboscopy
10^{-1} to 10^{-3} seconds.

Ultrafast spectroscopy by pulsed laser pump probe 10^{-10} to 10^{-15} s

Ref. Eadweard Muybridge 1878

Molecular motion in real time
Laser-matter interaction

• How is the laser energy deposited in materials?
  
  • ELECTRONS (CARRIER PLASMAS)
  • (confined in condensed matter)
3D nonlinear excitation: ultrafast laser pulses

- Inv. Bremsstrahlung
- Multiphotonic PHOTOIONIZATION
- COLLISIONAL Avalanche

$E_{fs}$
Electron on a spring with position $x_e(t)$, and driven by a light wave, $E_0 \exp(-i\omega t)$:

$$m_e \frac{d^2 x_e}{dt^2} + m_e \omega_0^2 x_e = eE_0 \exp(-i\omega t)$$

The solution is:

$$x_e(t) = \left[ \frac{e / m_e}{\left( \omega_0^2 - \omega^2 \right)} \right] E_0 \exp(-i\omega t)$$

Infinite amplitude at $\omega = \omega_0$
Free electrons

\[ x_e(t) = \left[ \frac{-e/m_e}{\omega^2 + i\omega\gamma} \right] E(t) \]

Energy stays constant \( E_{\text{kin}} = E_{\text{pot}} \)
no absorption on free electrons
(no energy and momentum conservation)

Energy varies because of collisions
ABSORPTION

R. Trebino Georgia Tech
3D nonlinear excitation: ultrafast laser pulses

Drude model

\[ \epsilon^*_\omega = \epsilon_0 \left( \frac{N_e}{N_{cr}} \right) \frac{1}{1 + i \frac{1}{\omega \tau}} = \epsilon_r + i \epsilon_i \]

\[ N_{cr} = \epsilon_0 m^*_e \omega^2 / e^2 \]

\[ \text{Re} \left[ \epsilon^*_\omega \right] = 0 \]

DEPOSIT ENERGY

ELECTRON DENSITY

RFLECTION

No coll

Coll=fs
3D nonlinear excitation: ultrafast laser pulses

Result: refractive index change
Nonlinear excitation: ultrafast laser pulses

Microexplosions: New matter states

Self-organization

Index changes: waveguides
Excitation of carrier plasmas

Q1: Ionization/Excitation
Optical dynamics:

Changes in optical properties
- Reflectivity
- Absorbivity

Example 1: Coherent vibrations

Energy coupling

Lattice: Vibration to heat

E. Mazur, Harvard University
Digital holography

\[ n^* \approx n_0 - \frac{N_e}{2N_{cr}} \frac{1}{1 + \left( \frac{1}{\omega \tau} \right)^2} \]

\[ k^* \approx \frac{N_e}{2N_{cr}} \frac{1}{1 + \left( \frac{1}{\omega \tau} \right)^2} \]

Interferogram

Phase: \( \phi \)  
Amplitude: \( T \)
Ionization of carrier plasmas in dielectrics

\[ \frac{dN_e}{dt} \sim I^n \]

\[ \frac{N_e}{m_e} \]
Excitation of carrier plasmas

Q2: Relaxation
Spectral interferometry

\[ n^* \approx n_0 - \frac{N_e}{2N_{cr}} \frac{1}{1 + (1/\omega \tau)^2} \]

Spectral fringes

\[ \Delta \Phi(t) = \frac{2\pi}{\lambda} \int_0^L \Delta n(l, t) \cdot \, dl \]
Optical dynamics: time-resolved ellipsometry

Changes in optical properties
- dielectric function
  \( \text{Re}(\varepsilon) \) via Fresnel eq.
  \( \text{Im}(\varepsilon) \)

Phase transitions

SOLID TO LIQUID

1 ps

GaAs

1.7 kJ/m²
Time-domain techniques

THz waves
THz time-domain spectroscopy

Razvan Stoian

M. Bonn, T. Heinz, MIT
Transient gratings: ultrafast laser pulses

Similarly:
- plasma waves (conserve k)

Diffraction depends on modulation contrast

Temnov OE 2009
Light conversion

- Generation of frequencies and phase matching

\( \chi^2 \) sensitive to crystalline anisotropy

In glasses? Electronic gradients

SOLID TO LIQUID

Beresna et al APL 2009

\[ \bar{P}(2\omega) = \chi_{\text{fe}}(2\omega) \left[ 2 \nabla E^2 / 2 + 2 \bar{E} (\nabla \cdot \nabla \ln n_e) / \bar{\varepsilon} \right] \]

Tom et al PRL 1998

SH generation
Excitation of carrier plasmas

Q3: Material ejection
Material removal: ultrafast laser pulses

Laser

Photoemission

Electron depleted region

Electron excited region

drift-diffusion transport

Photoemission

Coulomb explosion

R. Stoian PRL 2002
Material removal: ultrafast laser pulses

decay of excitation

$\Delta \tau$
Material removal: ultrafast laser pulses

DIELECTRICS
- electrostatic ion emission

METALS
- thermal

COULOMB EXPLOSION (CE)

\[ \frac{p(O^+)}{p(Al^+)} \]

NUMBER OF PULSES

NORMALIZED YIELD

TIME [ps]

Razvan Stoian

R. Stoian PRL 2002
Excitation of carrier plasmas

Q4: Internal energy; Scattering, plasma radiation
Energy of plasmas: ultrafast laser pulses

Simulation codes

Electron heating  Lattice heating  Hydrodynamic expansion

Plasma surdense
$10^{21} < N_e < 10^{23} \text{ cm}^3$

Chauffage électronique

Gaz

Liquide

Fusion

Solide

Densité ($\text{g cm}^{-3}$)

Comobier PhD 2005 CEA
Energy of plasmas: ultrafast laser pulses

Properties:
- density
- structure
- temperature

Proton scattering

Front evolution

Various labs: SLAC, DESSY, Los Alamos, CEA, Berkeley
Ablation pulses: ultrafast laser pulses

Nanoparticles

Shock (schlieren & shadowgraphy)
\( \frac{dn}{dx} \) \( \frac{d^2n}{dx^2} \)

Properties:
- acoustic
- optical
- luminescence
- electrical
- structural
- waves
- ions (core level)

Luminescence and excitation temperature
\[ I_\lambda \sim e^{-E/kT} \]

Amoruso APL 2009
Hermann JAP
Lippert JAP 2010
Car GE 2005
Challenge: dynamics of the surface movement?
Optical methods:
- visualizing surface modulation

Time-resolved diffraction?
The interest?

What is happening with the material

- changes in the dielectric function: OPTICAL
- changes in the structure: THERMODYNAMIC
- changes in the shape: FUNCTION
Monitoring and control of carrier plasmas

Q5: Carrier dynamics

Ex: Application to 3D structuring
3D nonlinear excitation: ultrafast laser pulses

Result: refractive index change
3D material modifications

refractive index $\Delta n$

- Building block of embedded optical functions

Light guiding
Davis et al OL 1996
Energy density regimes: a-SiO$_2$ Phase contrast

NA: 0.45 - 150 fs

Black $\Delta n > 0$ enables guiding - core
White $\Delta n < 0$ defines guiding - cladding

Razvan Stoian
Type I regime: a-SiO$_2$

Phase contrast

- NO STRONG POLARIZATION SENSITIVITY
- ISOTROPIC OPTICAL GUIDING
- LOW LOSSES <0.5dB/cm

Optical functions

Type I
$\Delta n (10^{-4} - 10^{-3})$

V-inj

H-inj

100 nJ

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3D photonic structure

Mauclair et al. OE 2009
Role of polarization: a-SiO$_2$

Energy

PCM WAVEGUIDE IMAGE

Type I - Isotropic guiding

Type II - WG Polarization sensitive

Guiding only here
Anisotropic regimes: a-SiO$_2$

- Birefringent regions
- Type II
- Core $\Delta n > 0$
- $\lambda/2n$ - controllable

Guiding when $E$ is parallel to the planes
Cladding: form birefringence

Polarization function

Shimotsuma PRL 2003
Bhardwaj PRL 2006

Cheng et al. OE 2009, 2010

Razvan Stoian
Optical functions: a-SiO$_2$

- Polarization maintaining waveguides
- Birefringent phase retardation properties

Quarter Wave Plate

Razvan Stoian

Mishchik et al. OE 2010
Refractive index changes

Local modification

Can this be improved (controlled)?
Refractive index changes a-SiO$_2$

SINGLE PULSE EFFECT (N=1): a-SiO$_2$

Q1: How is the energy distributed?
Q2: How does the material react?
Refractive index changes

Q: How is the energy deposited?
Nonlinear pulse propagation

NLSE-Schrödinger
E-field propagation
- self-focusing and self-phase modulation
- plasma generation
- filamentation
Pulse evolution: sequential energy deposition

INTENSITY

TIME [ps]

DEPOSITED ENERGY DENSITY [J/cm³]

FOCUS

z [µm]

Thermo-elasto plastic model (dynamic elasticity)

Material deformation

Accumulation of energy (expansion & rarefaction)

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Refractive index changes a-SiO$_2$

Q2: how does the material react?
Pump-probe apparatus: time-resolved microscopy

Excitation and relaxation dynamics

MODES:
- Phase contrast (PCM)
- Optical transmission (OTM)
Refractive index changes a-SiO$_2$: time-sequence

- Fast plasma decay (low excitation)
- Persistent absorptive zone (ns)
  - electrons?
  - liquid phase?
  - defects?
- But not permanent

Matrix response
Thermo-mechanics

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Refractive index changes a-SiO$_2$: electrons

- Residual absorption can be linked to defects

- But also to free electrons in liquid phase where trapping is not efficient (indication of a «phase transition»)
Comparison short & long pulses 6.2µJ, NA=0.45

160fs pulse

3ps pulse

Focal region
Refractive index changes a-SiO₂: electrons

150 fs

3 ps

Delayed plasma
-less defocusing

Better absorption and confinement
Comparison fs/ps, single pulse irradiation

- Pressure wave
- ps pulse – Stronger amplitude of the PW
Electronic transitions a-SiO$_2$: spectra

N=1

a) Electron plasma at peak

b) Permanent damage

c) Oxygen deficiency centers
Slow el. decay

d) Non-bridging oxygen
(Fast el. decay)
Refractive index changes a-SiO$_2$: stress

Slow relaxation

Slow $\Delta n$ Dynamics

-stress relaxation

Mermillod-Blondin et al. RSI 2011
Refractive index changes

Q5: the energy density is important

Energy density is important!!!
- regulates the physical excitation
- determines relaxation paths
Modulated refractive index changes

Q: How controllable is the nanoscale pattern?
Nano-control via diffraction feedback: \(\text{a-SiO}_2\)

Control via electronic excitation

Mauclair et al. OE 2012
Nano-control via diffraction feedback: a-SiO$_2$

Controllable Periodicity

Controllable arrangement

Linear chirp, symmetric stretching

Mauclair et al. OE 2012
Nano-control via diffraction feedback: a-SiO$_2$

Electron density

Non monotoneous $N_e$ -max 0.6ps

Mauclair et al. OE 2012
Nanostructuring hypothesis: a-SiO$_2$

Plasmonic hypothesis?

$$\Delta = \frac{\lambda}{\sqrt{\frac{\varepsilon_r + 1}{\varepsilon_r}}}$$

$$\varepsilon_r = \varepsilon_{r0} - \frac{N_e}{N_{cr}}$$

Shimotsuma et al. MPL 2005
Nanoscale control

Transition Order-Disorder on polymers

150 fs

2 ps

Forster JPPC 2011
Possible applications

What do we want to achieve?

- Flexibility in designing laser-interaction
Its all about the electrons

Process control
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