50 years 1976-2026

Laboratoire Hubert Curien
UMR ▪ CNRS ▪ 5516 ▪ Saint-Étienne

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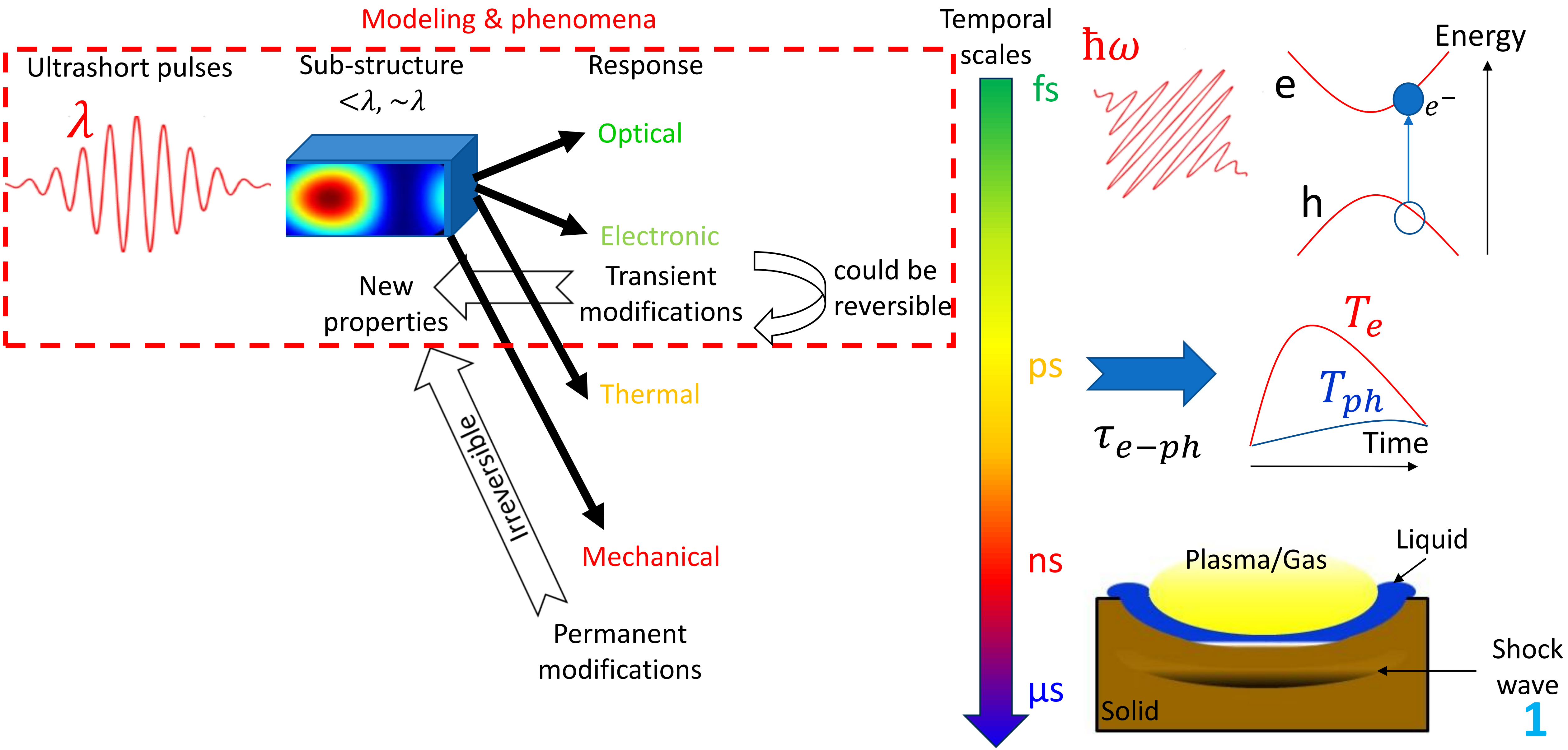
Photo-induced plasma confinement in ultrashort laser-excited nanostructures

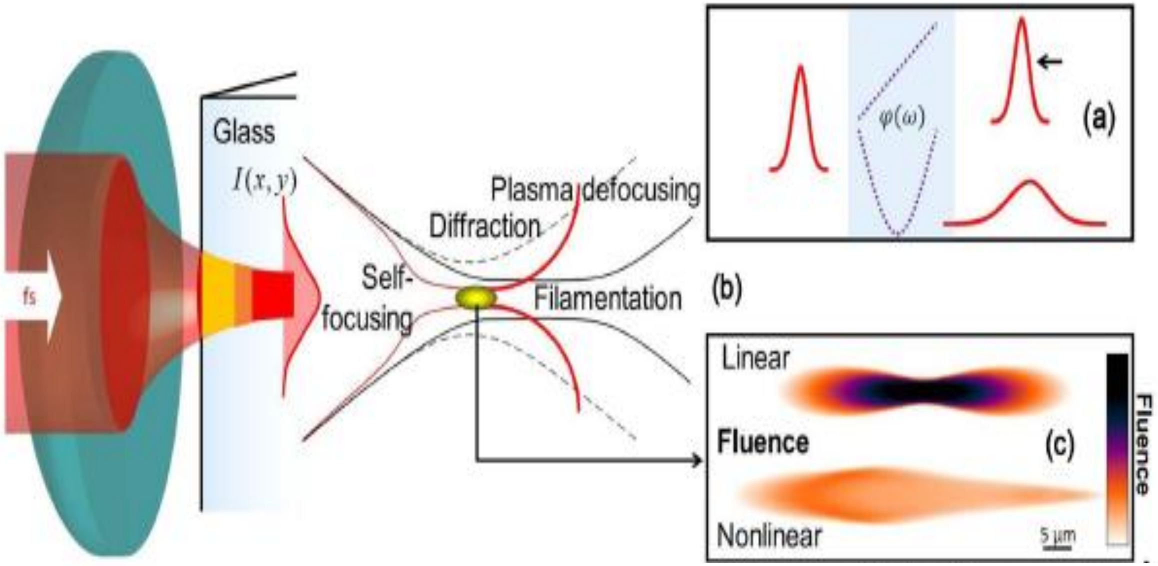
Anton Rudenko

Ultrashort laser-matter interaction: scales & response

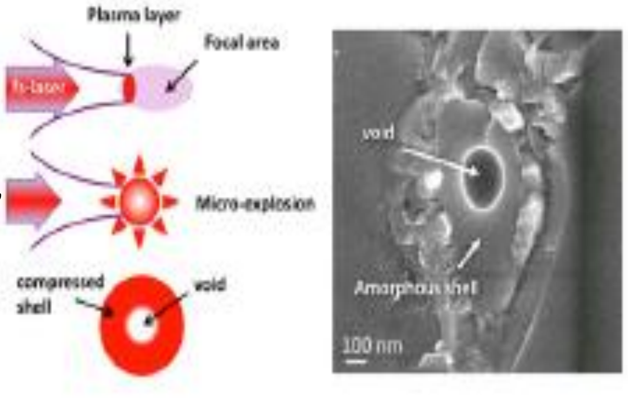


Tight focusing: Context/Problem formulation

Ultrashort laser pulse focused inside sapphire/glass



Microexplosions
S. Juodkazis, Phys. Rev. Lett., Vol. 96, 166101 (2006).



E = 100 nJ, $\lambda = 800$ nm, $\tau = 200$ fs, NA $= 1.35$

Modelling challenges

Coupled

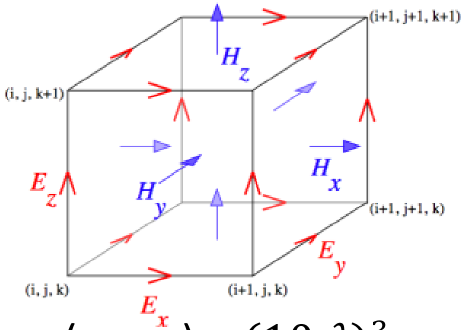
- 3D Maxwell solver (few µm-size)[3]
- Tight non-paraxial focusing conditions (NA=1.35)
- Model for electronic response: ionization mechanisms (w_{PI}, w_{AI}) & cold plasma fluid dynamics ($\overrightarrow{v_e}, \overrightarrow{J_e}, N_e, T_e, W$)
- Model for nonlinear optical response ($\overrightarrow{J_{NL}}, \overrightarrow{P_{NL}}$)

Maxwell equations

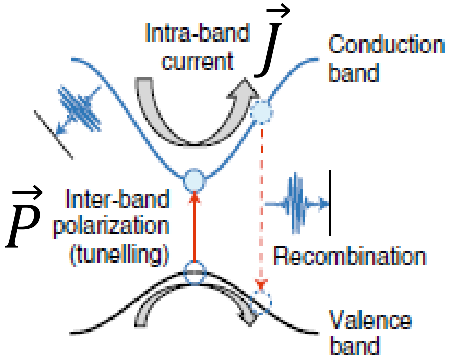
(optical response)

$$\frac{\partial \vec{E}}{\partial t} = \frac{\nabla \times \vec{H} - \overrightarrow{J_{NL}} - \frac{\partial \overrightarrow{P_{NL}}}{\partial t}}{\varepsilon_0}$$

$$\frac{\partial \vec{H}}{\partial t} = -\frac{\nabla \times \vec{E}}{\mu_0}$$



(x, y, z) $\sim(10\ \lambda)^3$



System of equations & beyond

Maxwell equations coupled to electron plasma fluid equations

Nonlinear Maxwell equations

$$\frac{\partial(\varepsilon\vec{E})}{\partial t}=\frac{\nabla\times\vec{H}-\vec{J}_e-\vec{J}_{Kerr}-\vec{J}_{PI}}{\varepsilon_0}$$

$$\frac{\partial\vec{H}}{\partial t}=-\frac{\nabla\times\vec{E}}{\mu_0}$$

Momentum equation

$$\frac{\partial\vec{J}_e}{\partial t}=-\vec{J}_e\nu_e+\frac{N_e e^2}{m_e^*}(\vec{E}+\vec{v}_e\times\vec{H})-\vec{\nabla}\cdot(\vec{J}_e\otimes\vec{v}_e)-\frac{e}{m_e^*}\vec{\nabla}P_e$$

Rate equation

$$\frac{\partial N_e}{\partial t}=\frac{N_a-N_e}{N_a}(w_{PI}+w_{AI})-\vec{\nabla}\cdot(N_e\vec{v}_e)+D_a\Delta N_e$$

Energy equation

$$\frac{\partial E_e}{\partial t}=(\vec{J}_e+\vec{J}_{PI})\cdot\vec{E}-\vec{\nabla}\cdot([E_e+P_e]\vec{v}_e)+\vec{\nabla}\cdot(k_e\vec{\nabla}T_e)$$

Intraband current $\vec{J}_e=eN_e\vec{v}_e$

Kerr effect

$$\vec{J}_{Kerr}=\varepsilon_0\chi^{(3)}\frac{\partial(\frac{N_a-N_e}{N_a}\vec{E}|\vec{E}|^2)}{\partial t}$$

Photo-ionization depletion

$$\vec{J}_{PI}=w_{PI}I_p\frac{\vec{E}}{|\vec{E}|^2}\frac{N_a-N_e}{N_a}$$

(1) Beyond single-band representation?
(2) Interband polarization? Beyond dipolar?
(3) Non-perturbative nonlinearities of higher order?

DFT Band structure → SBE Dynamics

System of equations & beyond

Maxwell equations coupled to electron plasma fluid equations

Nonlinear Maxwell equations

$$\frac{\partial(\varepsilon\vec{E})}{\partial t} = \frac{\nabla\times\vec{H} - \vec{J}_e - \vec{J}_{Kerr} - \vec{J}_{PI}}{\varepsilon_0}$$

$$\frac{\partial\vec{H}}{\partial t} = -\frac{\nabla\times\vec{E}}{\mu_0}$$

Momentum equation

$$\frac{\partial\vec{J}_e}{\partial t} = -\vec{J}_e\nu_e + \frac{N_e e^2}{m_e^*}(\vec{E} + \vec{v}_e\times\vec{H} + \cdots) - \vec{\nabla}\cdot(\vec{J}_e\otimes\vec{v}_e) - \frac{e}{m_e^*}\vec{\nabla}P_e$$

Rate equation

$$\frac{\partial N_e}{\partial t} = \frac{N_a - N_e}{N_a}(w_{PI}+w_{AI}) - \vec{\nabla}\cdot(N_e\vec{v}_e) + D_a\Delta N_e$$

Energy equation

$$\frac{\partial E_e}{\partial t} = (\vec{J}_e + \vec{J}_{PI})\cdot\vec{E} - \vec{\nabla}\cdot([E_e + P_e]\vec{v}_e) + \vec{\nabla}\cdot(k_e\vec{\nabla}T_e)$$

Collision frequency

$$\nu_e = \nu_{e-ph} + \nu_{e-h}(T_e) + \nu_{ei}(N_e, T_e), \nu_e \le \nu_{max}(\overrightarrow{v_e}, T_e)$$

Reduced electron mass (m_e^*)

Electron pressure $P_e = N_e k_B T_e$

Electron energy density (thermal+kinetic+bandgap)

$$E_e = (\frac{3}{2}k_B T_e + \frac{m_e^* v_e^2}{2} + I_p)N_e$$

(1) Validity of continuum representation?
(2) How to evaluate collisions ?

Kinetic Boltzmann
Electron/phonon sub-systems

(1) Beyond single-band ?
(2) Electron pressure beyond the ideal gas ?

DFT
Band structure

System of equations & beyond

Maxwell equations coupled to electron plasma fluid equations

Nonlinear Maxwell equations

$$\frac{\partial(\varepsilon\vec{E})}{\partial t} = \frac{\nabla\times\vec{H} - \vec{J}_e - \vec{J}_{Kerr} - \vec{J}_{PI}}{\varepsilon_0}$$

$$\frac{\partial\vec{H}}{\partial t} = -\frac{\nabla\times\vec{E}}{\mu_0}$$

Momentum equation

$$\frac{\partial\vec{J}_e}{\partial t} = -\vec{J}_e\nu_e + \frac{N_e e^2}{m_e^*}\left(\vec{E} + \vec{v}_e\times\vec{H}\right) - \vec{\nabla}\cdot\left(\vec{J}_e\otimes\vec{v}_e\right) - \frac{e}{m_e^*}\vec{\nabla}P_e$$

Rate equation

$$\frac{\partial N_e}{\partial t} = \frac{N_a - N_e}{N_a}(w_{PI}+w_{AI}) - \vec{\nabla}\cdot(N_e\vec{v}_e) + D_a\Delta N_e$$

Energy equation

$$\frac{\partial E_e}{\partial t} = \left(\vec{J}_e + \vec{J}_{PI}\right)\cdot\vec{E} - \vec{\nabla}\cdot([E_e + P_e]\vec{v}_e) + \vec{\nabla}\cdot(k_e\vec{\nabla}T_e)$$

Photo-ionization rate

$$w_{PI}(|\vec{E}|, m_e^*, I_p, \omega_0, \varepsilon)$$

Avalanche ionization rate

$w_{AI}(\vec{v}_e, T_e)$ from Maxwellian distribution

(1) Beyond static e-field single-wavelength single-parabolic band ionization?
(2) Towards dynamic band structure?
(3) Beyond Maxwellian velocity distribution?

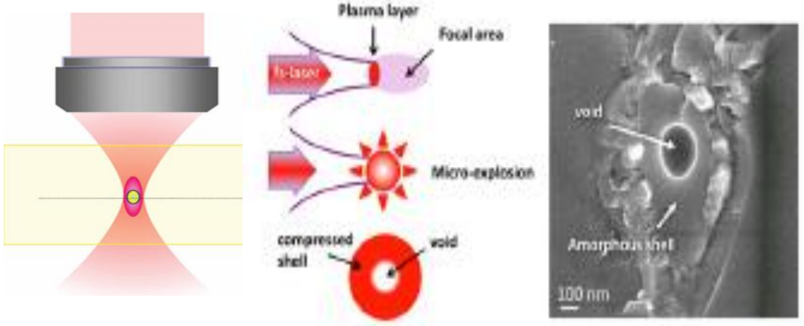
TD-DFT
Band structure

Kinetic Boltzmann
Electron sub-system

Extreme tight focusing in transparent solids

Microexplosions in bulk sapphire

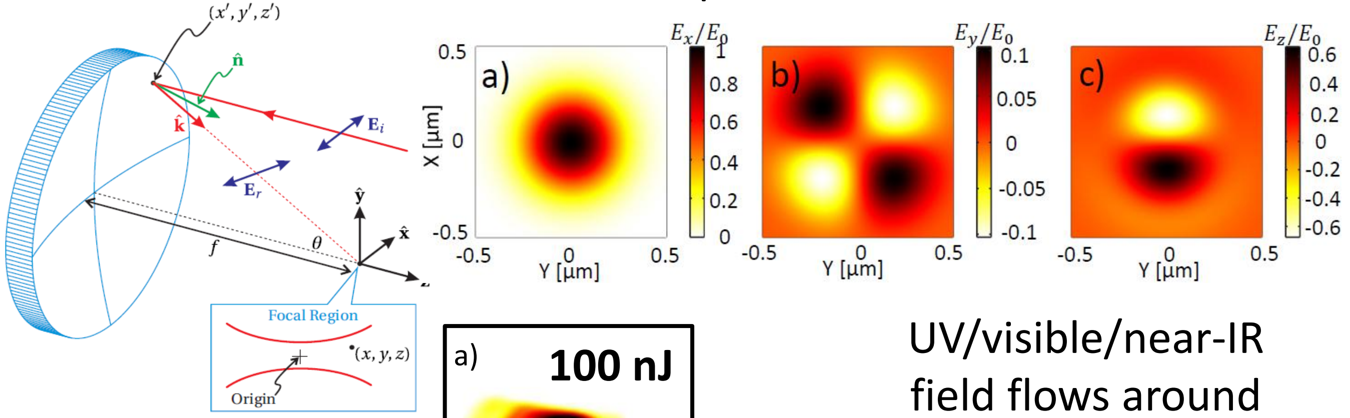
S. Juodkazis, Phys. Rev. Lett., Vol. 96, 166101 (2006)

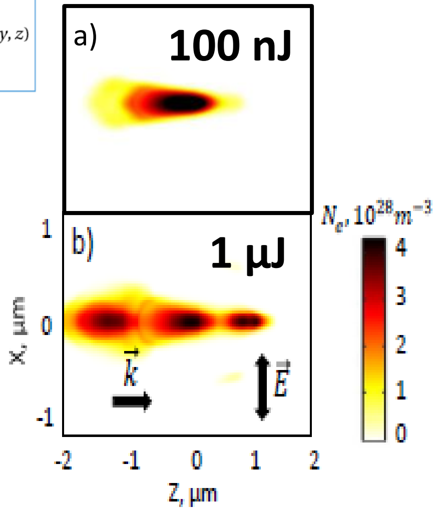


E. Gamaly et al., Phys. Rev. B (2007) Point explosion static estimate	A. Rudenko et al., Phys. Rev. Appl. (2023) Dynamic modeling
$F_p \sim 150 \frac{J}{cm^2}$	Clamping $F_{nl} \sim 15\, J/cm^2$
$N_a \sim 5 \cdot 10^{29} m^{-3}$	No full ionization $N_e \sim N_a/10$
$P_e \sim 13.4\, TPa \gg Y$	$P_e \sim 150\, GPa < Y$

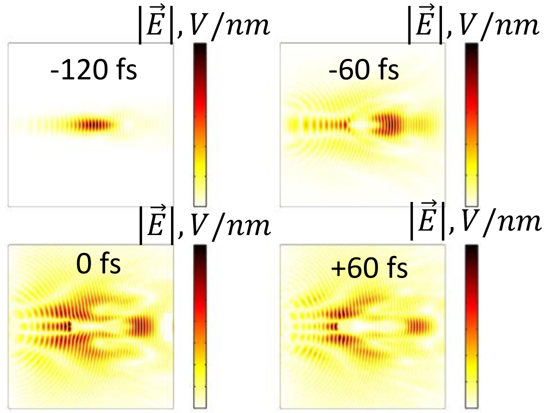
E = 100 nJ, $\lambda = 800$ nm, $\tau = 200$ fs, NA $= 1.35$

Reflected fields from parabolic mirror





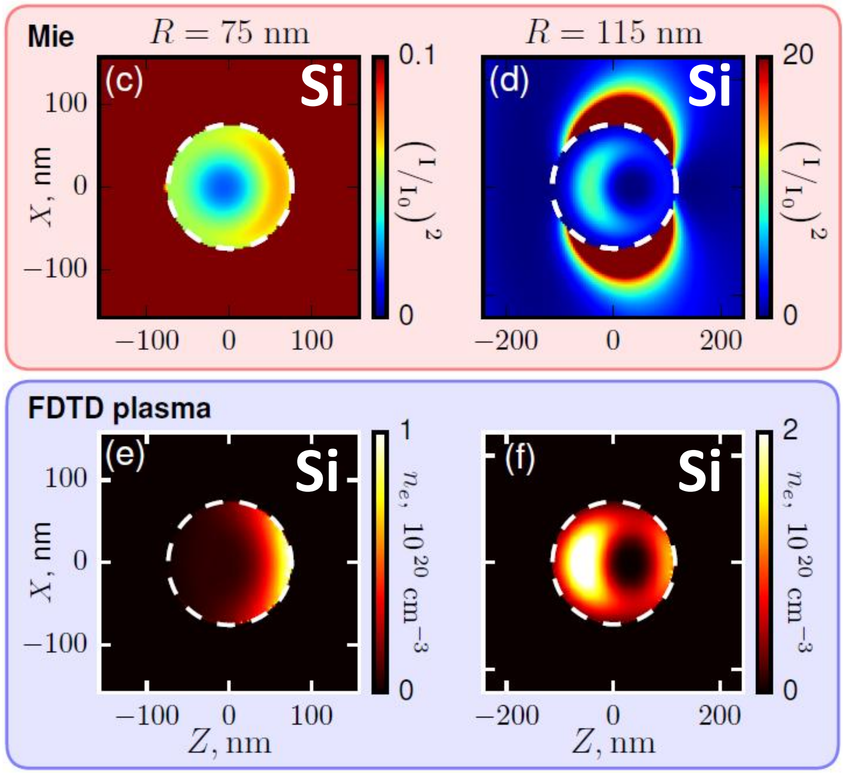
UV/visible/near-IR field flows around plasma ball



1 μJ

Universal phenomenon: ionization clamping prevails within (0.1-10)·($E_p, w_{PI}, w_{AI}, \nu_e$) and in UV/visible/near-IR

Photo-induced plasma inside Si NP



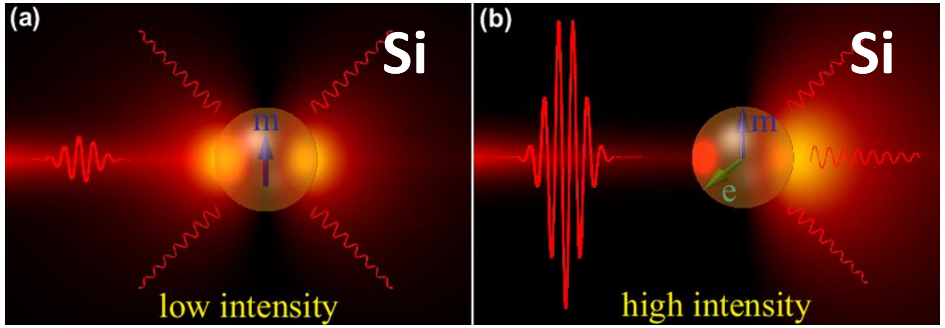
A. Rudenko et al., Adv. Opt. Mater., 2018

How the photo-induced plasma affects:

- Directional scattering
- Resonant mode excitation
- Nonlocal optical response [SHG]
- Non-perturbative harmonic generation [HHG]
- Irreversible reshaping at high intensity

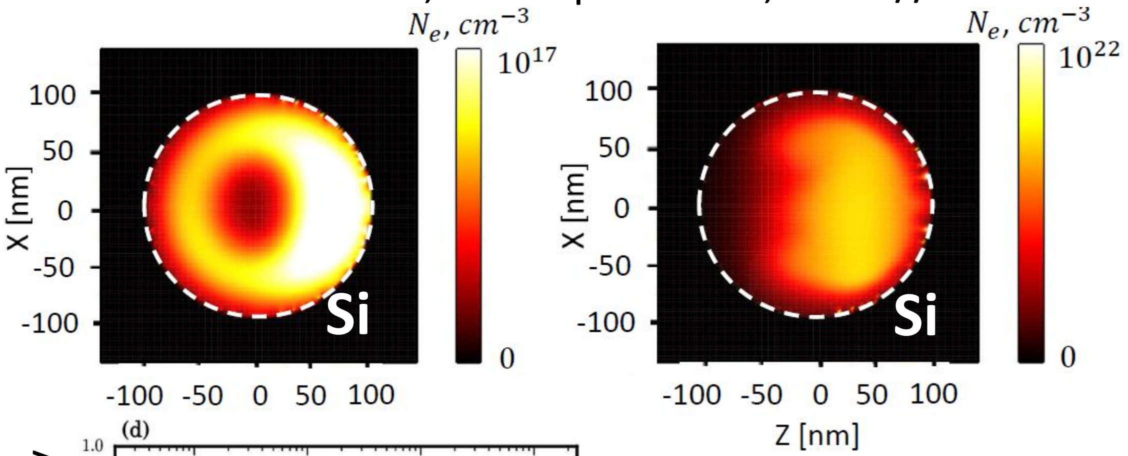
Far-field: backward-to-forward optical switching

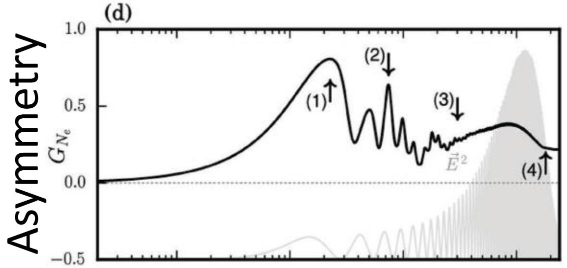
S. Makarov et al., Nano Letters, 2015



Near-field: symmetry breaking

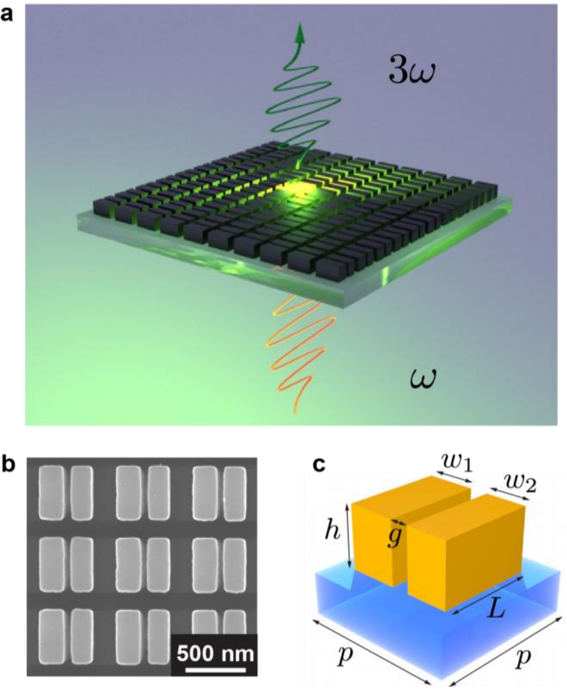
A. Rudenko et al., Adv. Opt. Mater., 2018 // 2023



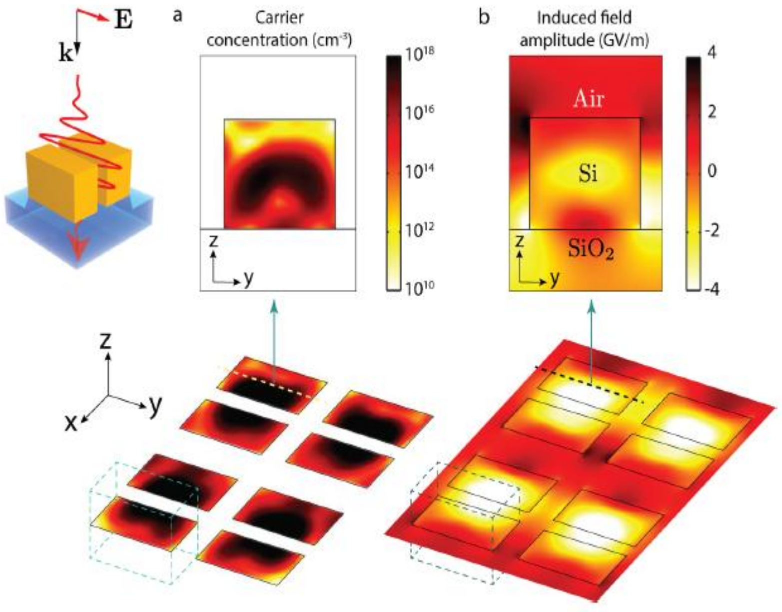


(1) low-Q ED excitation;
(2) non-stationary;
(3) high-Q Mie pattern;
(4) homogenization

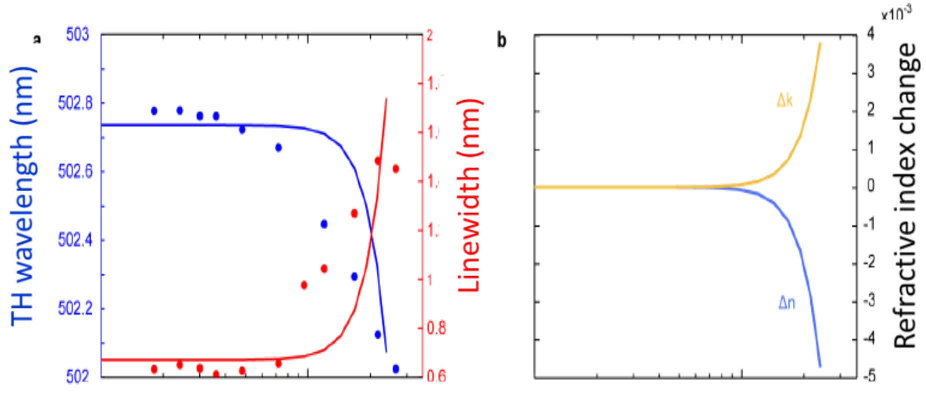
Plasma-induced nonlinear effects in metasurfaces



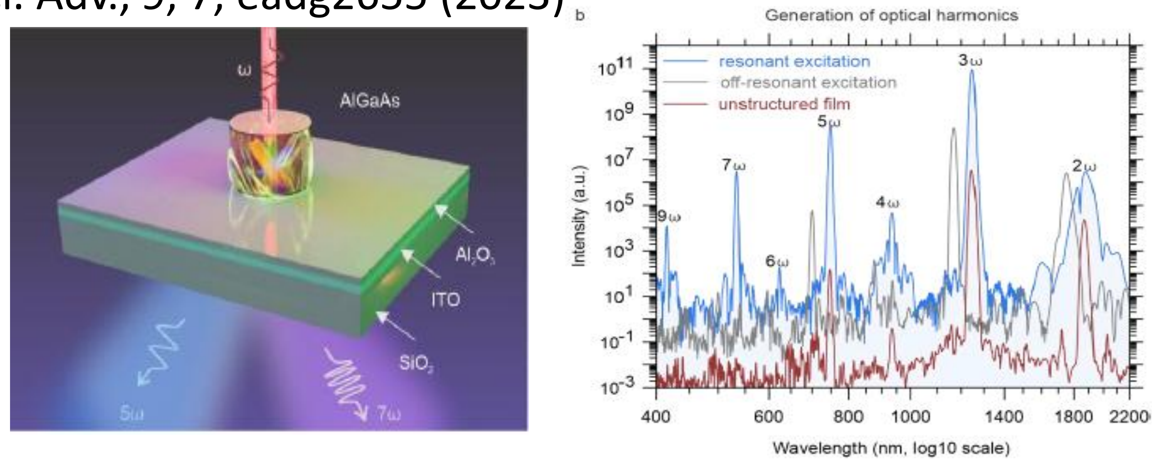
Nano Lett., 21, 20, 8848 (2021)



Plasma-induced blueshift THG in resonant BIC metasurface



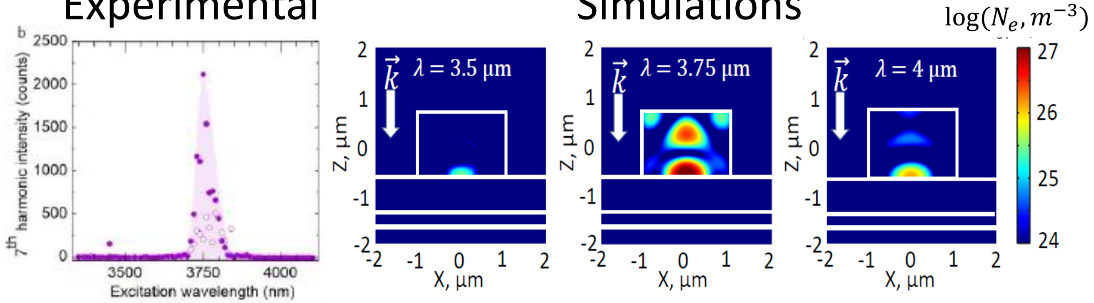
Sci. Adv., 9, 7, eadg2655 (2023)



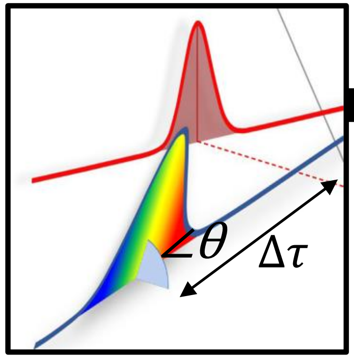
Non-perturbative odd and even HHG in resonant BIC cylinder

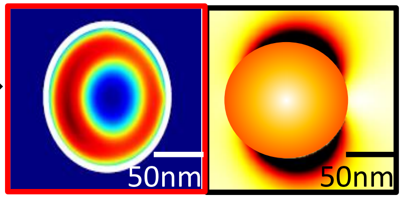
Experimental

Simulations



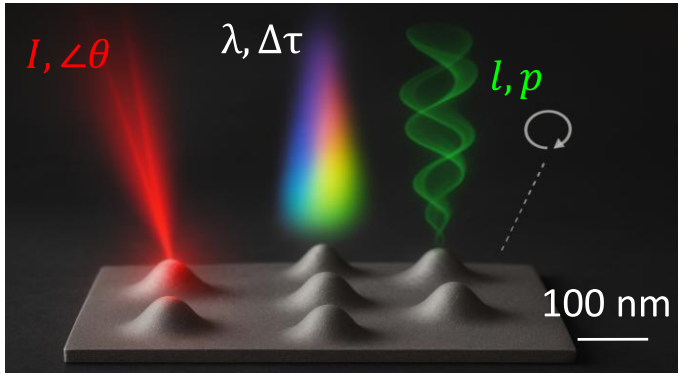
Strategies for laser-induced nanostructuring



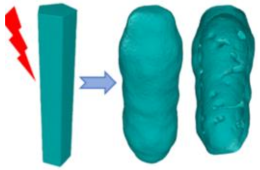


Near-field
(Mie-, plasmon, …)

Spatio-temporal
control of laser beam

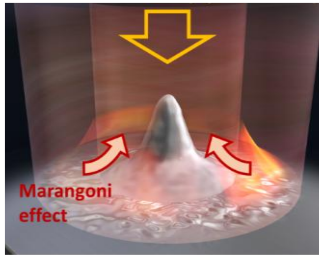


Laser reshaping of nanostructures

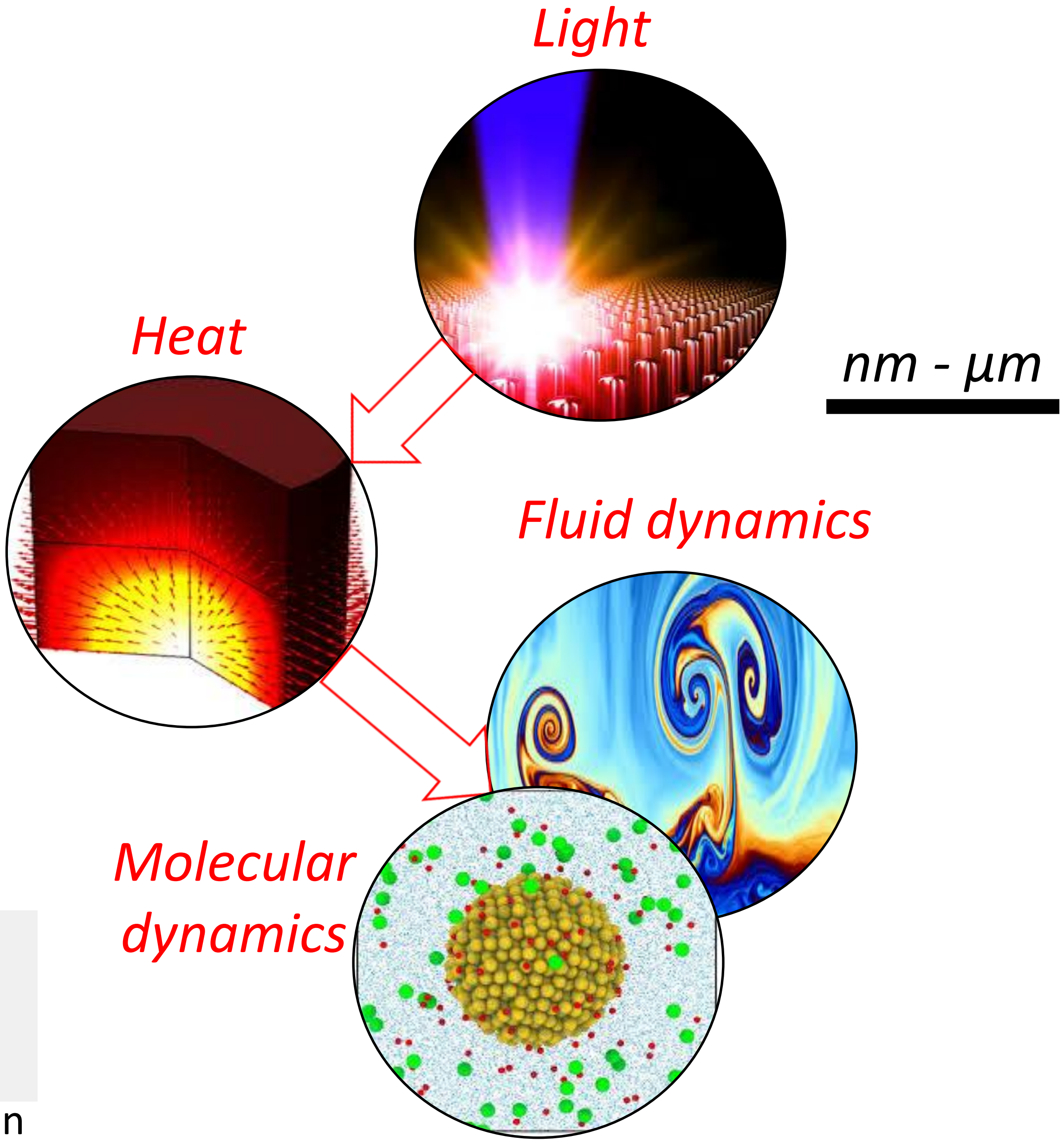


Melting

Nanocavitation



Nanoablation



Summary

- Plasma confinement by tight focusing: challenges due to ionization clamping, spatial nonlinearity, laser damage
- Revisiting the models for ultrashort laser-induced electron plasma dynamics: beyond classical approaches
- Plasma confinement in nanostructures: tunable control of (non)linear, (non)local near(far)-field optical properties avoiding irreversible damage
- Could this strategy help to nanostructure with ultimate precision?

Merci pour votre attention !