International workshop on

"Laser micro and nano structuring : Fundamentals et applications"

December 10-12, 20012, Ecole Polytechnique, Palaiseau

Theoretical treatments of ultrashort pulse laser processing of transparent materials: Towards explanations of extraordinary phenomena

Nadezhda M. Bulgakova,[1,2] *Vladimir P. Zhukov*,[3]
Yuri P. Meshcheryakov [4]

[1] Optoelectronics Research Centre, University of Southampton, UK

[2] Institute of Thermophysics SB RAS, Novosibirsk, Russia

[3] Institute of Computational Technologies SB RAS, Novosibirsk, Russia

[4] Design and technology Branch of Lavrentyev Institute of Hydrodynamics SB RAS, Novosibirsk, Russia

SEVENTH FRAMEWORK PROGRAMME

MARIE CURIE ACTIONS

Outline

The model of focused laser beam propagation in transparent solids based on the Maxwell equations

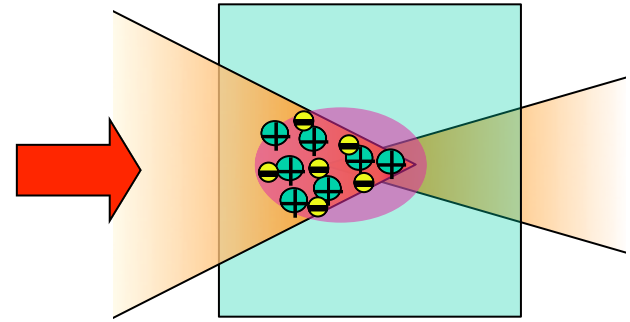
What are the electron plasma densities and energies in the regimes typical for ultrafast laser writing?

Comparing Maxwell and NLSE solutions

On nanograting formation

Laser beam propagation and plasma generation

Non-linear Schrödinger equation (NLSE)



$$\frac{\partial \bar{\mathcal{E}}}{\partial z} = \frac{i}{2k_0} T^{-1}\left(\frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r}\right)\bar{\mathcal{E}} - \frac{ik''}{2}\frac{\partial^2 \bar{\mathcal{E}}}{\partial t^2} + \frac{ik_0 n_2 T}{n_0}\left[(1-f_R)\left|\bar{\mathcal{E}}\right|^2 + f_R \int_{-\infty}^{t} R(t-\tau)\left|\bar{\mathcal{E}}\right|^2 d\tau\right]\bar{\mathcal{E}} -$$

$$-\frac{\sigma}{2}(1+i\omega_0\tau_c)T^{-1}(n_e\bar{\mathcal{E}}) - \frac{1}{2}\frac{W_{\mathrm{PI}}(\left|\bar{\mathcal{E}}\right|)E_g}{\left|\bar{\mathcal{E}}\right|^2}\bar{\mathcal{E}}.$$

coupled with the rate equation for free carriers

$$\frac{\partial n_e}{\partial t} = \left[W_{PI}(\left|\bar{\mathcal{E}}\right|) + \frac{\sigma n_e}{(1+m_r/m_e)E_g}\left|\bar{\mathcal{E}}\right|^2\right]\frac{n_a}{n_l} - \frac{n_e}{t_{tr}}$$

NLSE is obtained from the Maxwell equations in assumption of UNIDIRECTIONAL beam propagation

Time scales of volume modification processes

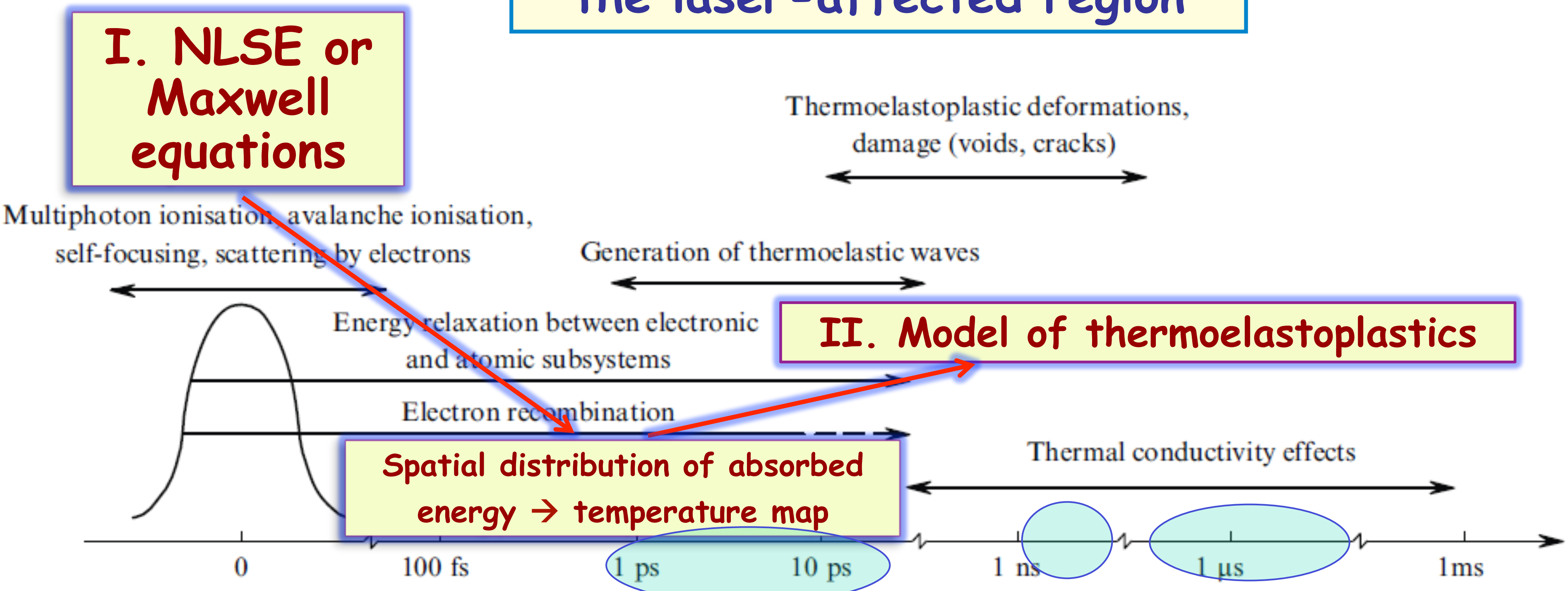
Absorption of the laser energy by electrons

Electron-lattice thermalization

Formation of steep temperature gradients

Generation of thermoelastic waves

Heat conduction → cooling of the laser-affected region

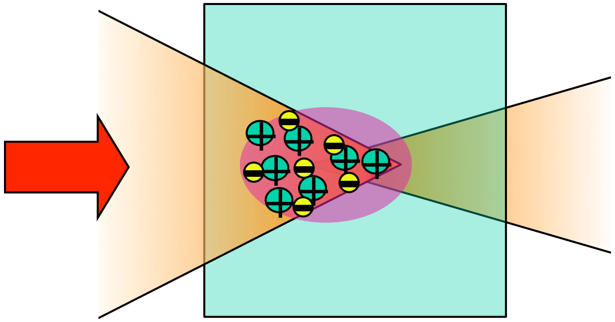


A. Mermillod-Blondin et al. Phys. Rev. B, **77**, 104205 (2008)

Maxwell equations

$$\nabla \times \vec{E} = -\frac{1}{c}\frac{\partial \vec{B}}{\partial t},$$

$$\nabla \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c}\vec{J},$$



Equation for the electric field accounting free carrier generation and associated processes

$$\frac{1}{c}\frac{\partial D}{\partial t} - i\frac{\omega}{c}D = -\frac{4\pi}{c}j + \mathrm{rot}\, H - \frac{8\pi e^2}{mc\omega^2}W_{PI0}\frac{\rho_a}{\rho_0}\left(\frac{|E^2|}{E_*^2}\right)^{\alpha-1}(1+E^2/(4E_*^2))E$$

$$\vec{E} = (\vec{E}_0 e^{-i\omega t} + \vec{E}_0^* e^{i\omega t})/2$$

$$D = E + \sum_m P_m + P_{nl} \qquad P_{nl} = \frac{c}{4\pi}n^2 n_2\left(\left(1-f_r\right)|E^2| + f_r\int_0^\infty R(\tau)|E^2(t-\tau)|\,d\tau\right)E$$

coupled with hydrodynamic model for free carriers

$$j = -\rho e \mathrm{v} \qquad \frac{\partial \rho}{\partial t} = W_{PI} + W_\sigma - \frac{\rho}{\tau_{tr}}$$

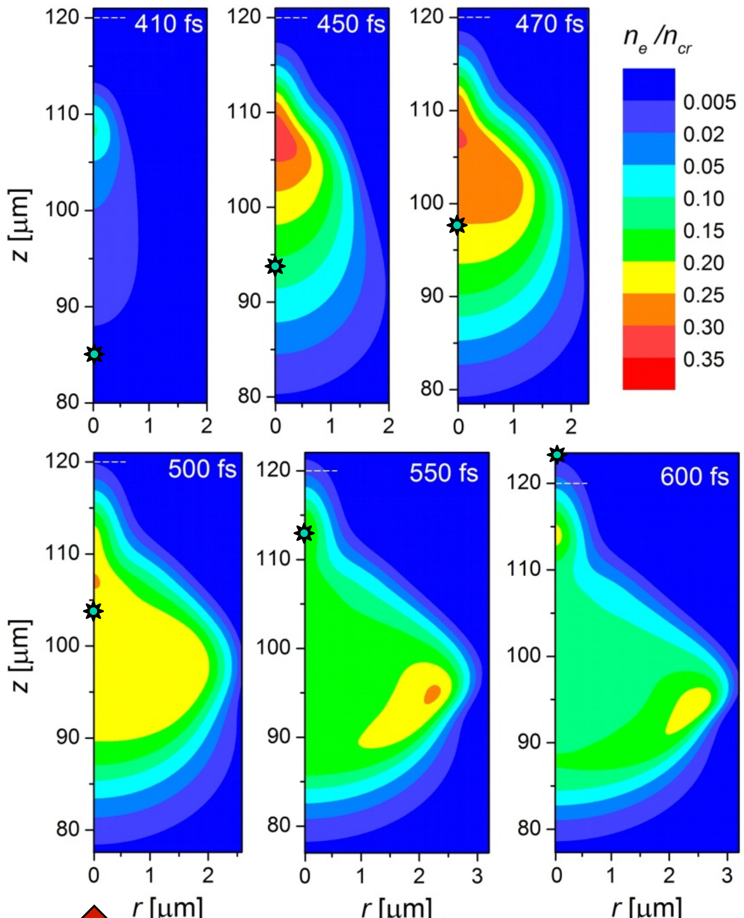
$$\frac{\partial(\rho \mathrm{v})}{\partial t} - i\omega\rho\mathrm{v} = -\rho\frac{e}{m_e}E - \rho\frac{\mathrm{v}}{\tau_c}$$

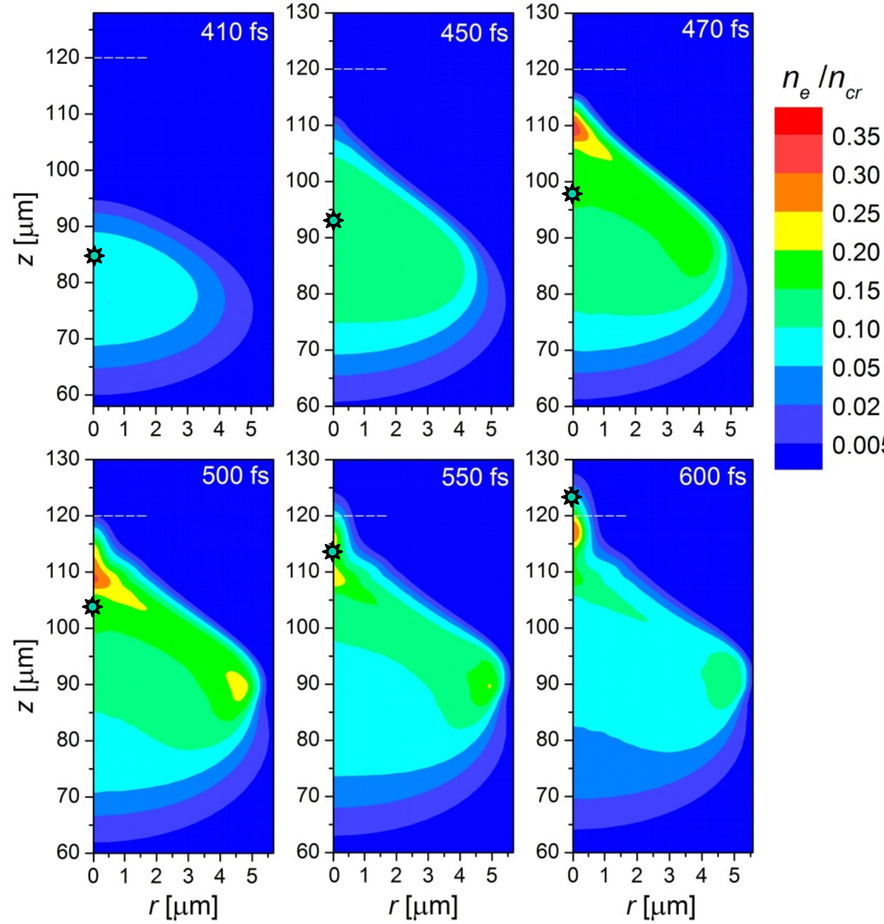
Free electron density inside fused silica

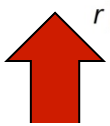
Ti:sappire; 800 nm wavelength; beam waist 1 μm

(Y. Shimotsuma et al. Phys. Rev. Lett. 91, 247405 (2003))

(I.M. Burakov et al. J. Appl. Phys. 101, 043506 (2007))







1 μJ, 150 fs

2.5 μJ, 80 fs

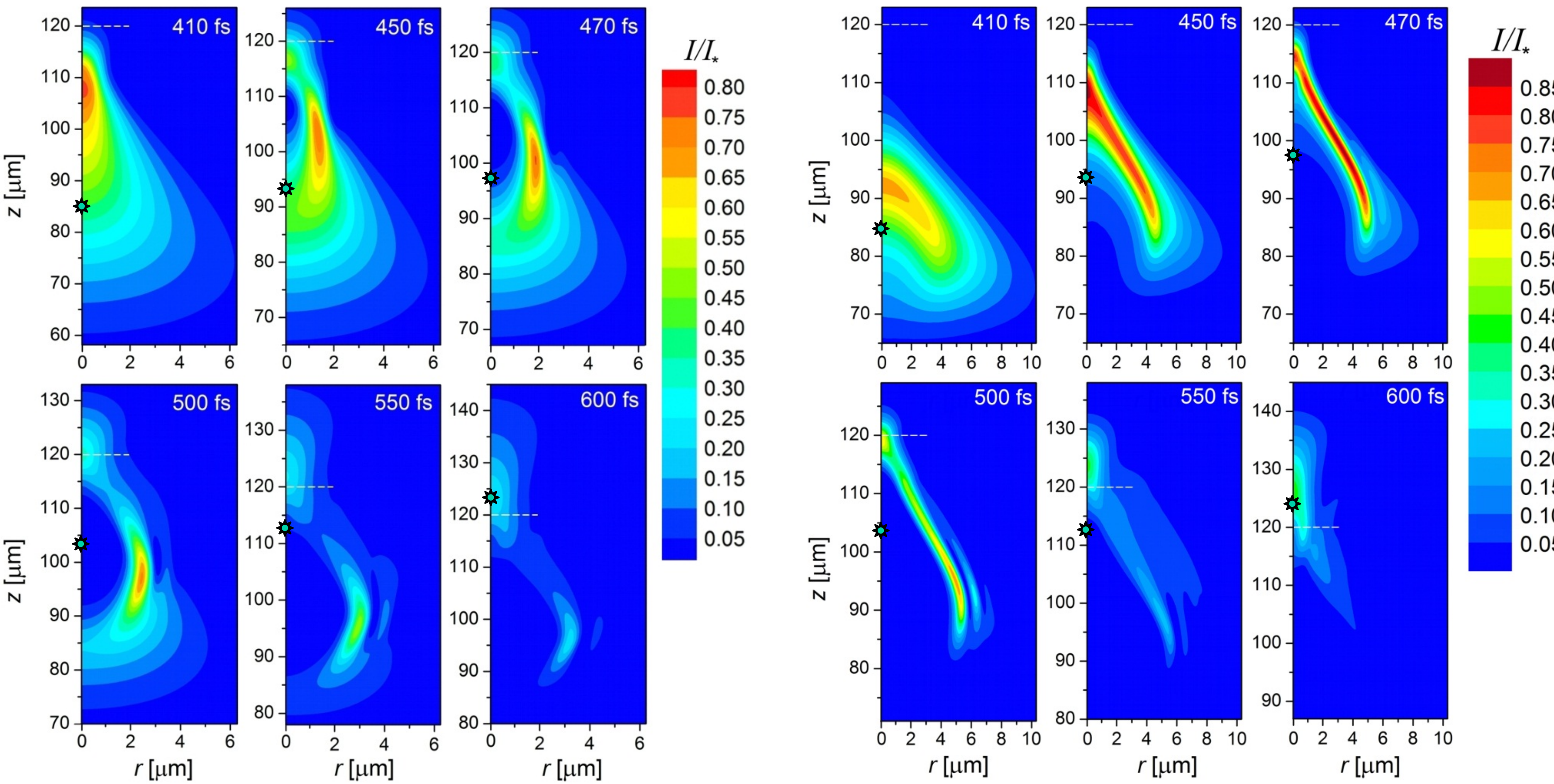
Laser

N.M. Bulgakova, V.P. Zhukov, Yu.P. Meshcheryakov, Applied Physics B, accepted

Snapshots of the laser beam intensity

Ti:sappire; 800 nm wavelength; beam waist 1 μm

I_* is the critical intensity at which $\gamma = 1$



1 μJ, 150 fs

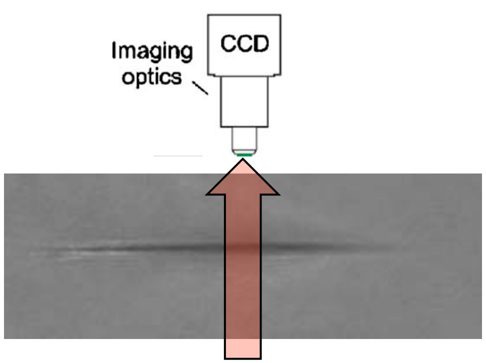
2.5 μJ, 80 fs

N.M. Bulgakova, V.P. Zhukov, Yu.P. Meshcheryakov, Applied Physics B, accepted

What are the densities of free electrons under typical modification conditions?

n_e^{max} values obtained numerically for single-pulse ultrafast laser excitation of silica glasses. Laser wavelength is 800 nm. $N_e^{cr} = 1.74\times10^{21}$ cm^{-3}

Pulse energy, μJ	Pulse duration, fs	NA	Material	Plasma density, cm^{-3}	Ref.
≥ 0.1	50	0.25	BK7	~ 1.7×10^{20}	D.M.Rayner et al. *Opt. Exp.*, **13**, 3208 (2005)
1.3	50	0.25	BK7	~7.5×10^{20}	Ibid
0.25–1.25	160	0.5	fused silica	$(4-6)\times10^{20}$	A.Couairon, et al, *Phys. Rev. B*, **71**, 125435 (2005)
1	120	0.45	fused silica	~3×10^{20}	I.M.Burakov, et al., *J. Appl. Phys.*, 101, 043506 (2007)
0.5	150	0.45	BK7	~5×10^{20}	A.Mermillod-Blondin, et al., *Phys. Rev. B*, **77**, 104205 (2008)
0.4	50	0.65	fused silica	~8×10^{20}	K.I. Popov, et al., Opt. *Express*, **19**, 271 (2010)
1-2.5	50,80,150	0.25-0.36	fused silica	~6×10^{20}	Present study



Probe pulse

$$n_e = -\frac{\ln T\cdot\lambda}{4\pi L}\,\frac{2n_0\varepsilon_0\omega^2 m_e^*}{e^2}\,\frac{1+\omega^2\tau_c^2}{\omega\tau_c}.$$

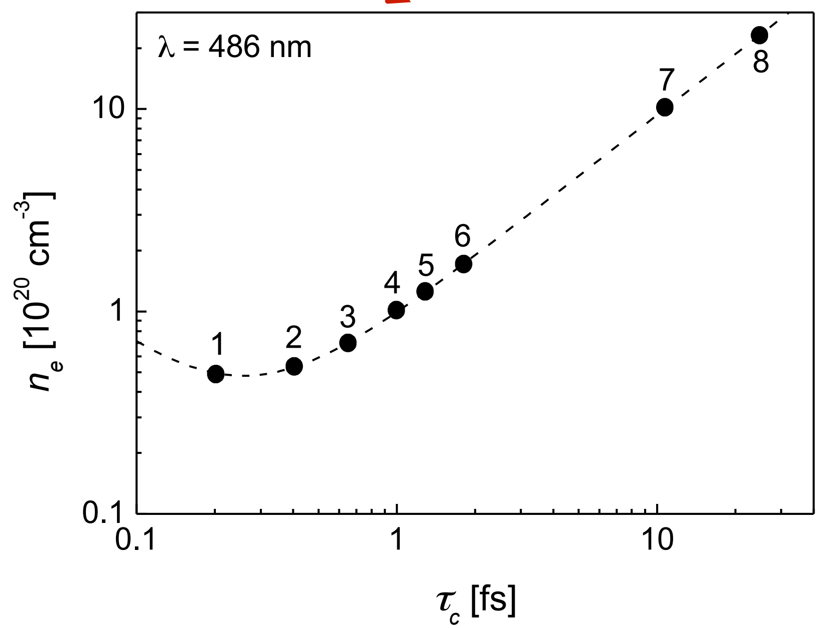
D.G. Papazoglou, et al. *Opt. Lett.*, 32, 2055 (2007)
n_e ~ 4×10^{19} cm^{-3}

M.C. Richardson, CLEO/QELS 2008, San Jose
n_e ~ 10^{17} - 10^{18} cm^{-3}

$$n_e = -\frac{\ln T \cdot \lambda}{4\pi L}\frac{2n_0\varepsilon_0\omega^2 m_e^*}{e^2}\frac{1+\omega^2\tau_c^2}{\omega\tau_c}.$$

n_e is averaged along the probe beam path *L* through the plasma region

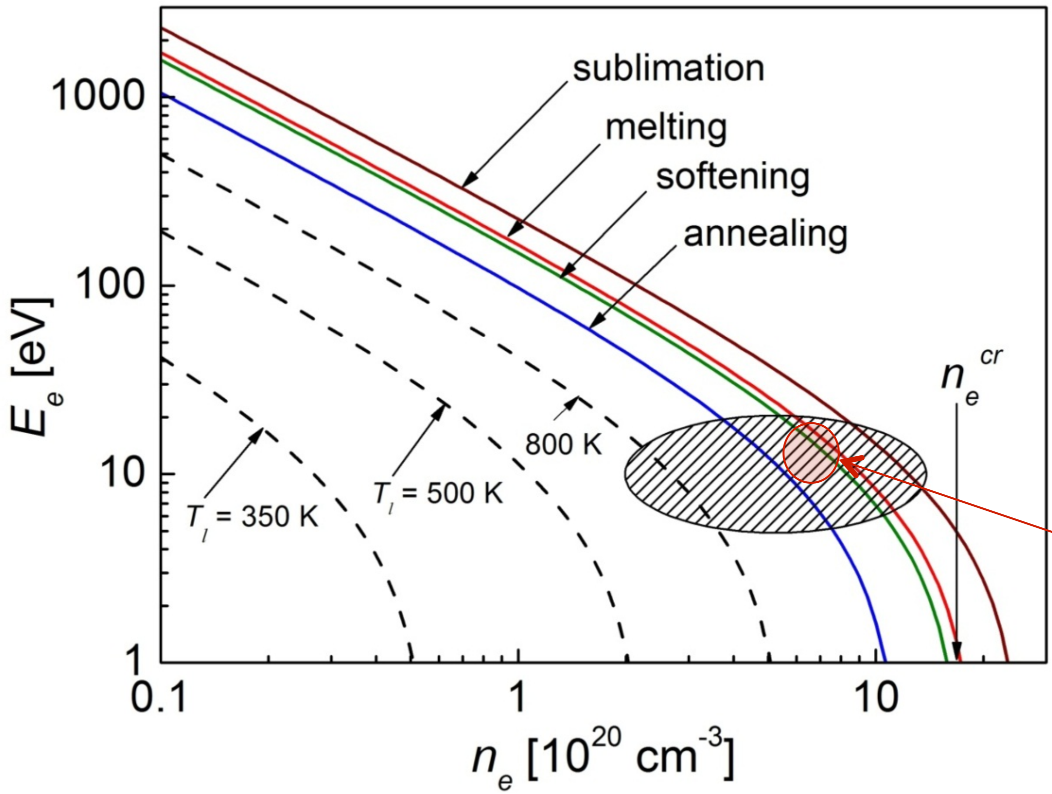
m_e^* is reported in the range from $0.5m_e$ to $2.2m_e$



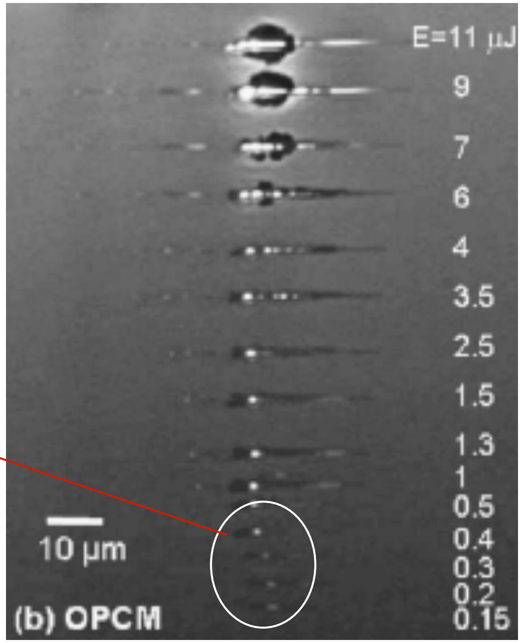
A possible solution can be in applying probe pulses whose frequency is smaller than ω_{pl} expected in the laser-excited region:
For a 1560 nm femtosecond fiber laser (critical plasma density is ~4.6×10^{20} cm^{-3}), assuming that in fused silica τ_c ~ 10 fs, the characteristic absorption length α_{ab}^{-1} is ~1 μm at $n_e = 8\times10^{20}$ cm^{-3} and α_{ab}^{-1} ~ 5 μm at $n_e = 3.3\times10^{20}$ cm^{-3}.

Energy balance

$$c\rho(T^{*}-T_0)=n_e\left(E_e+E_g\right)$$



I.M.Burakov, et al., *J. Appl. Phys.*, 101, 043506 (2007)

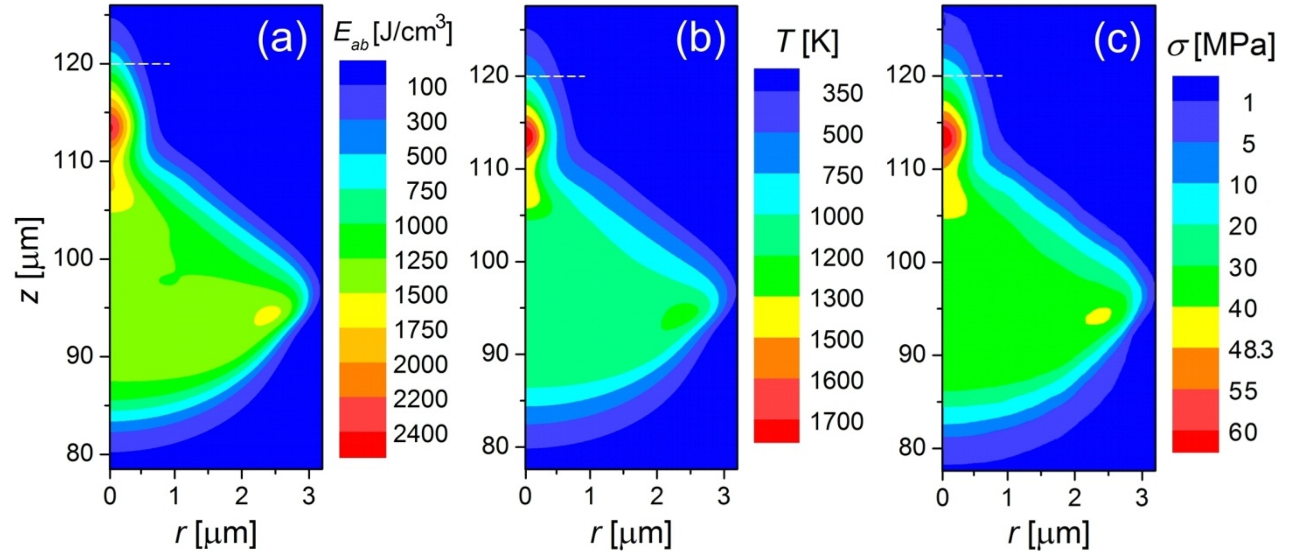


Pulse energy, μJ	Pulse duration, fs	NA	Material	Plasma density, cm^{-3}	Ref.
≥ 0.1	50	0.25	BK7	~ 1.7×10^{20}	D.M.Rayner et al. *Opt. Exp*., **13**, 3208 (2005)
1.3	50	0.25	BK7	~7.5×10^{20}	Ibid
0.25–1.25	160	0.5	fused silica	$(4-6)\times10^{20}$	A.Couairon, et al, *Phys. Rev. B*, **71**, 125435 (2005)
1	120	0.45	fused silica	~3×10^{20}	I.M.Burakov, et al., *J. Appl. Phys*., 101, 043506 (2007)
0.5	150	0.45	BK7	~5×10^{20}	A.Mermillod-Blondin, et al., *Phys. Rev. B*, **77**, 104205 (2008)
0.4	50	0.65	fused silica	~8×10^{20}	K.I. Popov, et al., Opt. *Express*, **19**, 271 (2010)
1-2.5	50,80,150	0.25-0.36	fused silica	~6×10^{20}	Present study

Energy – temperature – stress

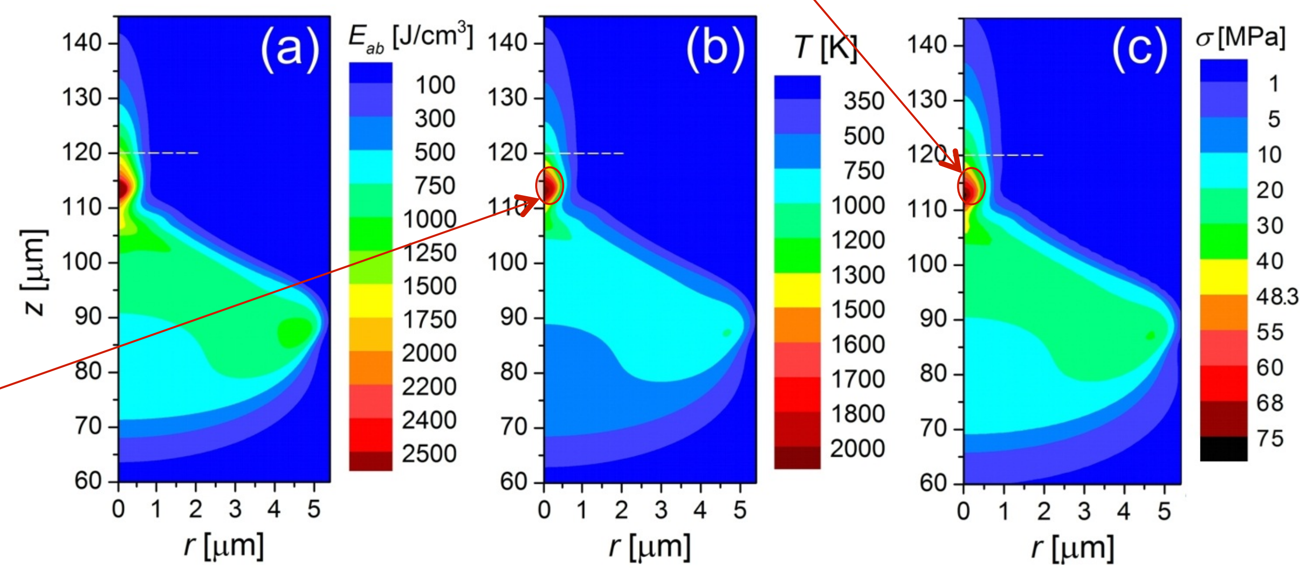
1 μJ, 150 fs

$$c\rho(T^{*} - T_{0}) = E_{\text{abs}}$$



Possible homogeneous nucleation of the vapor phase

2.5 μJ, 80 fs



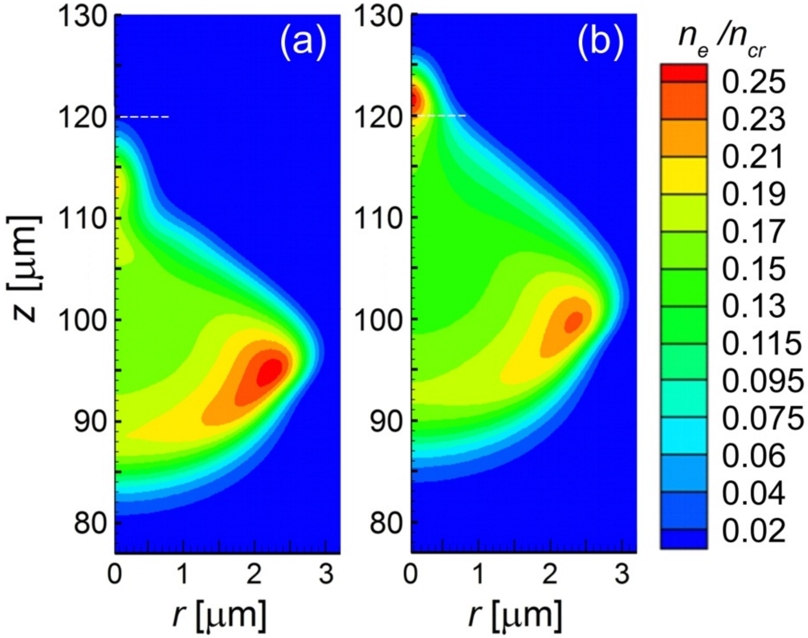
melting

N.M. Bulgakova, V.P. Zhukov, Yu.P. Meshcheryakov, Appl. Phys. B, accepted

Comparing Maxwell and NLSE solutions

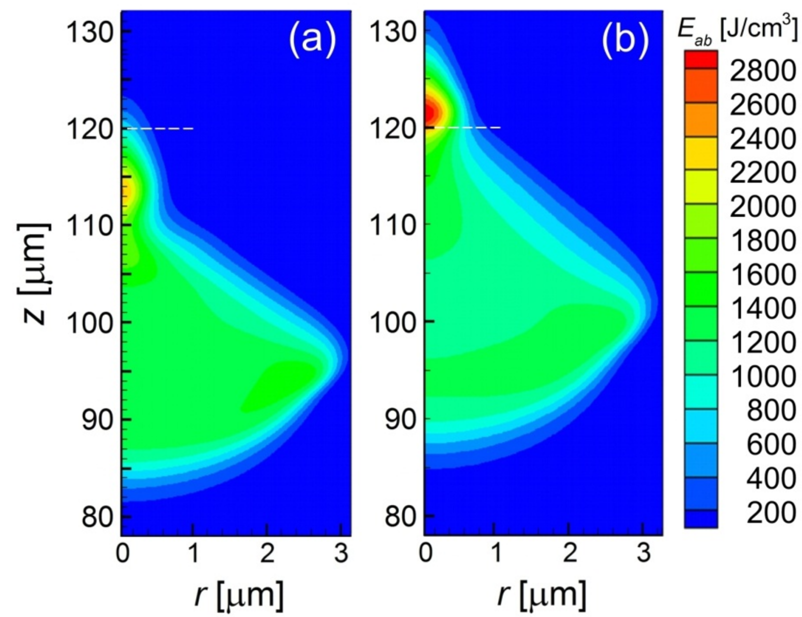
1 μJ, 150 fs, NA = 0.25

Free electron density
at t = 500 fs



Maxwell NLSE

Absorbed laser energy



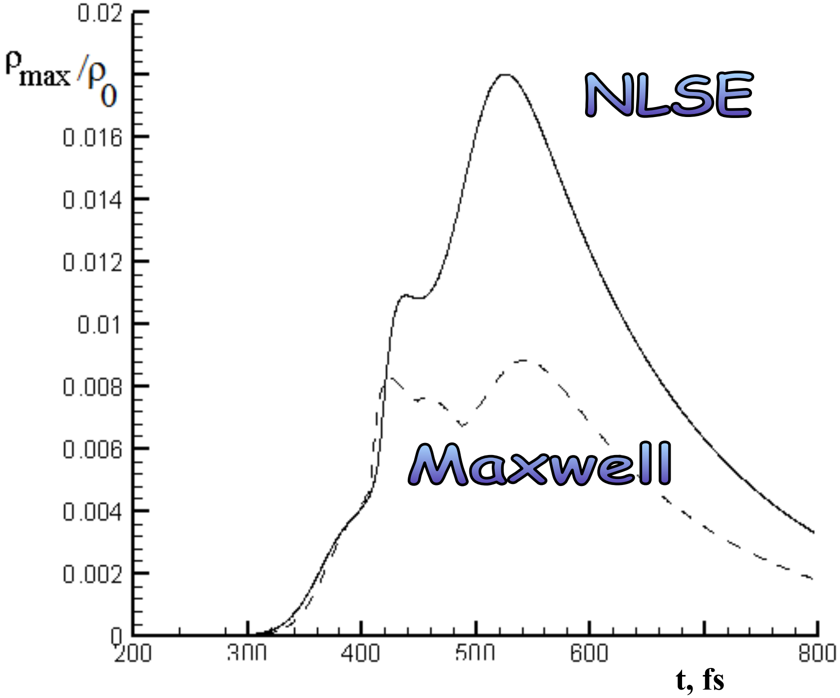
Maxwell NLSE

N.M. Bulgakova, V.P. Zhukov, M.P. Fedoruk, to be published

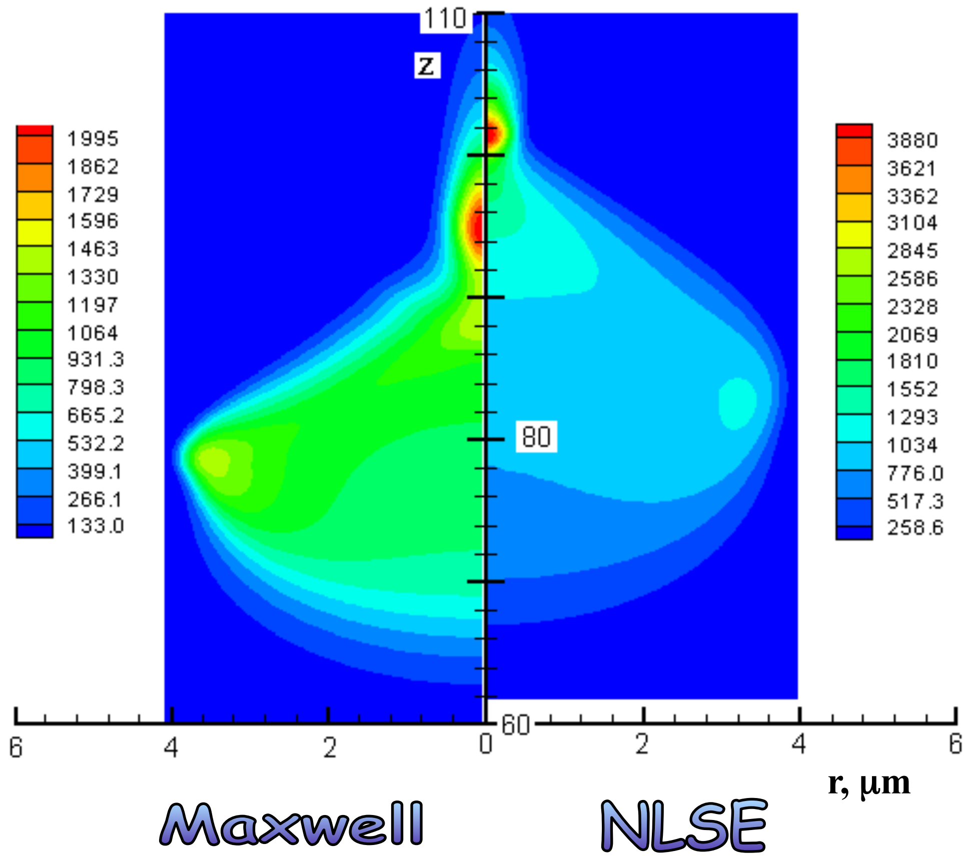
Comparing Maxwell and NLSE solutions

1 μJ, 50 fs, NA = 0.36

Maximum free electron density
as a function of time

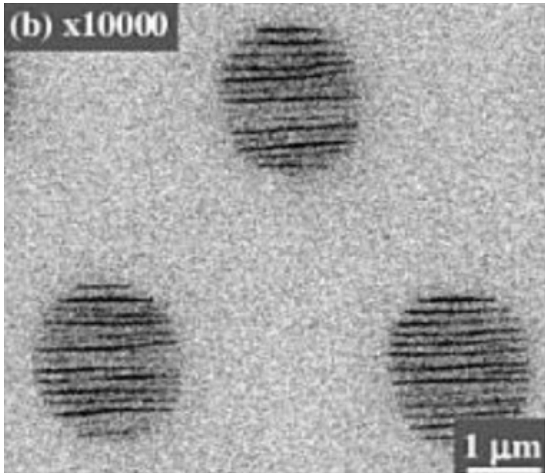


Absorbed laser energy



N.M. Bulgakova, V.P. Zhukov, M.P. Fedoruk, to be published

Nanogratings in transparent materials



Existing explanations:

1. **Plasma wave interfering with optical beam**

Y. Shimotsuma et al. PRL, 91, 247405 (2003)

2. **Nanoplasma self-ordering**

V. R. Bhardwaj et al. PRL, 96, 057404 (2006)

3. **Exciton "self-organization"**

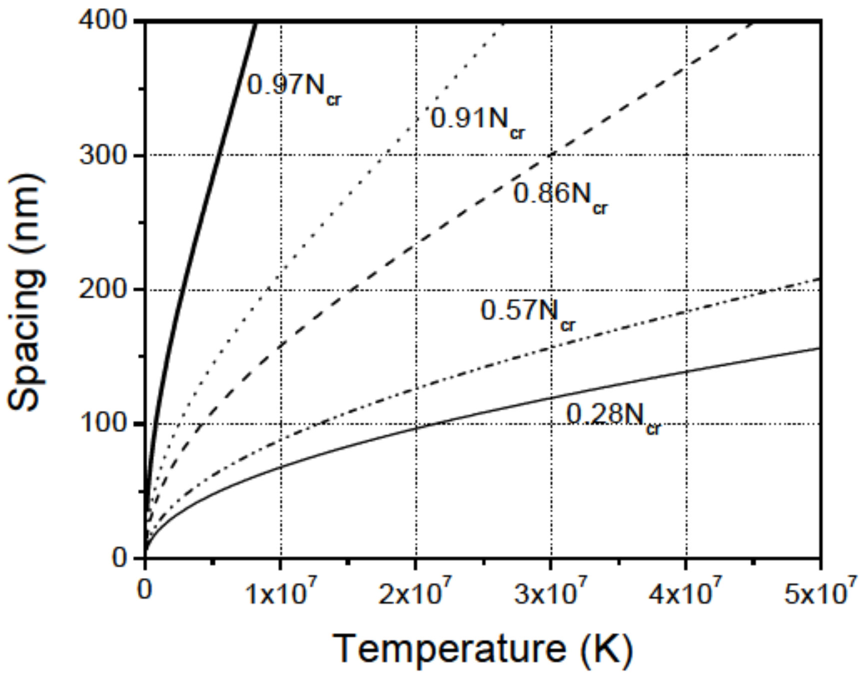
M. Beresna et al., Appl. Phys. Lett., 101, 053120 (2012)

Key questions:

1. What is the actual mechanism?
2. Why do the nanogratings form only in several materials?
3. What are the main material properties for nanograting formation?

1. Plasma wave interfering with optical beam

Y. Shimotsuma et al. PRL, 91, 247405 (2003)



From: R.P. Pattathil et al. Proc. SPIE 5971, 5971D (2005)

$$\Lambda = \frac{2\pi}{\sqrt{\frac{1}{T_e}\left(\frac{m_e\omega^2}{3k_B} - \frac{e^2 N_e}{3\varepsilon_0 k_B}\right) - k_{\text{ph}}^2}}.$$

Additionally, energy balance estimate

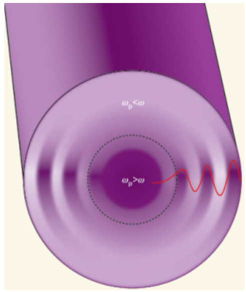
$$c\rho(T^* - T_0) = n_e\left(\frac{3}{2}kT_e + eE_g\right)$$

with near-critical plasma density and temperatures of order of 10^6 K and higher gives huge lattice temperatures which would definitely result in microexplosion

Alternative could be the Tonk-Dattner resonances

L. Tonks, Phys. Rev. 37, 1458 (1931);

A. Dattner, Ericsson Technics No.2, Stockholm, 310 (1957)



$$\omega_j^2 = \omega_p^2 + (3kT/m_e)k_j^2, \quad j = 1, 2, 3, \dots$$

The multiple absorption and reflection peaks when probing a confined gas discharge in a discharge tube (confined plasma) with variable frequency

T.C. Killian, Nature 441, 297 (2006)

$$\int_{-r_f}^{r_f} k(r)dr = \pi j, \quad j = 1,2,3,\dots$$

If to assume uniform distribution of electron density, then the dispersion relation reads as

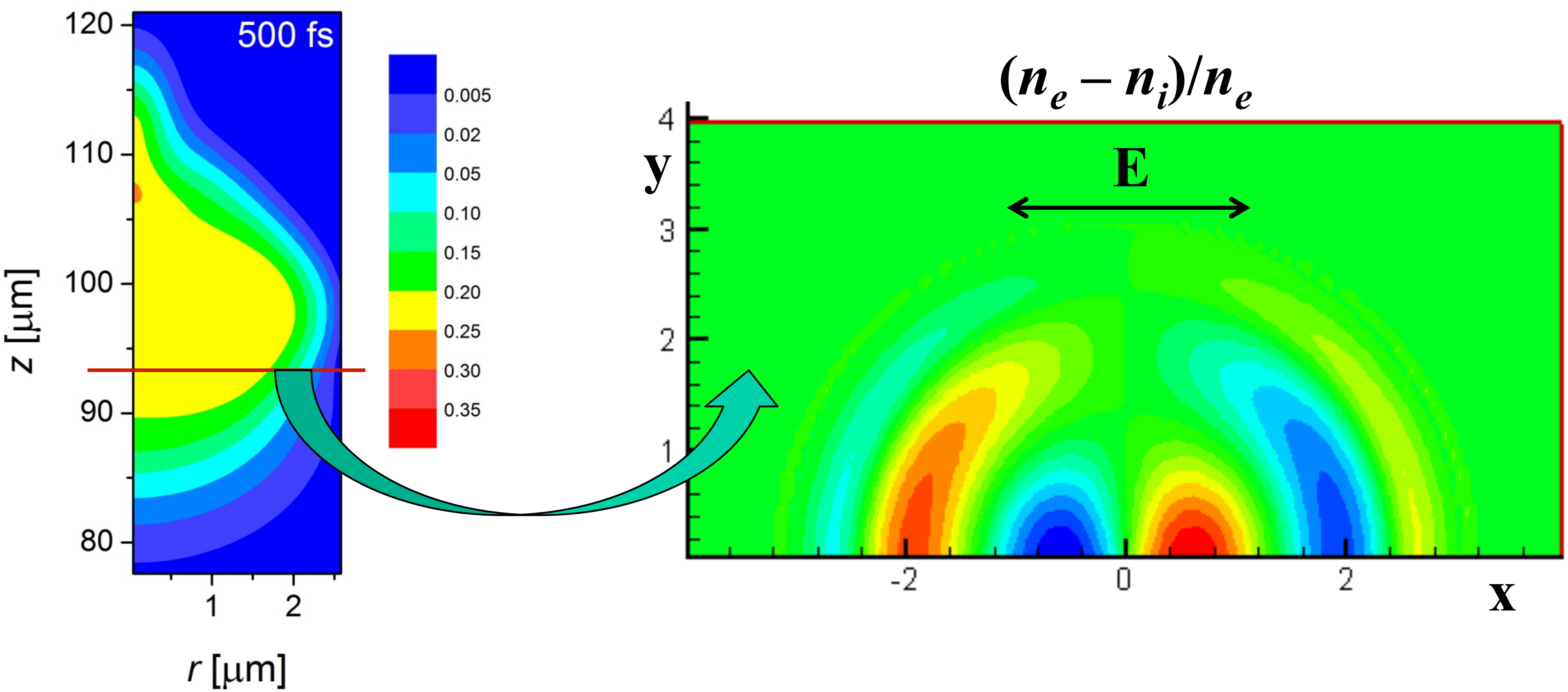
$$\omega_j^2 = \omega_p^2 \left[1 + \left(\pi j \lambda_D / 2r_f\right)^2\right]$$

At typical free electron density of $(2 - 6)\cdot 10^{20}$ cm^{-3} and electron energy of ~10 eV, $j \sim 10^3$. Taking into account actual plasma profile from simulations leads to $j \approx 120$. To obtain the grating periodicity of 200 nm c $E_e \sim 10$ eV, $j = 54$ but n_e must be close to n_{cr}:

$$\omega_j^2 - \omega_p^2 \approx 1.3 \times 10^{25}\ (\mathrm{rad/s})^2$$

Plasma wave geometry in focusing region cross-section

1 μJ, 150 fs, NA = 0.25

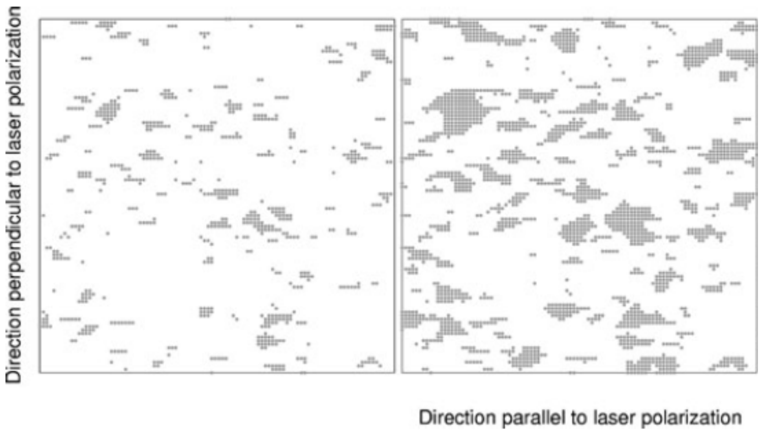


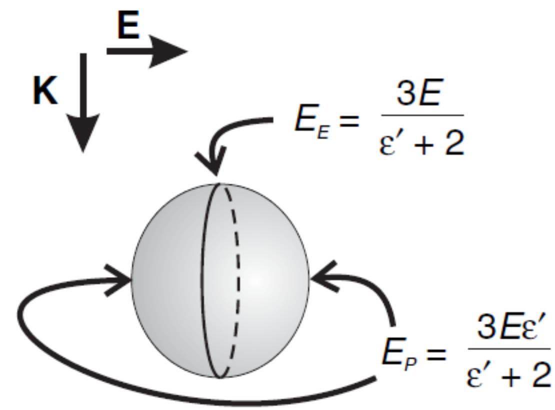
Plasma waves:
No tendency to form plane periodic structures

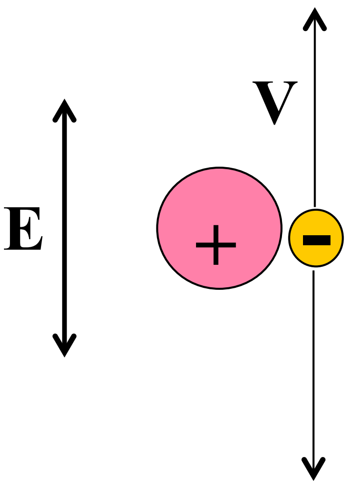
2. Nanoplasma self-ordering

V. R. Bhardwaj et al. PRL, 96, 057404 (2006)

Forest-fire model







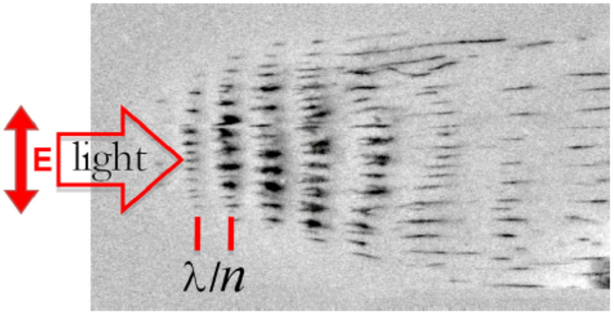
L.N. Gaier et al.
J. Phys. B 37,
L57 (2004)

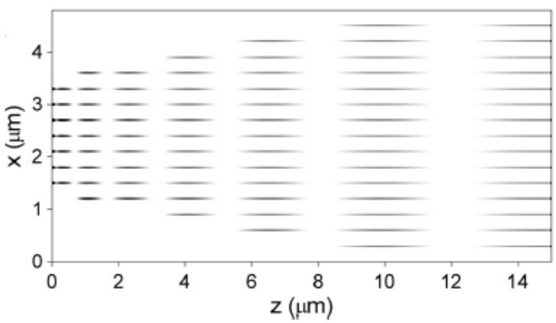
$$\frac{\partial n_e}{\partial t} \sim \sigma_n I^n \to \sigma_n \left(E_L + E_{local}\right)^{2n}$$

The problems:
1. Idea of self-organization of nanoplasmas into nanoplanes is vague
2. Periodicity cannot not be explained

3. Exciton-mediated self-organization

M. Beresna et al. APL, 101, 053120 (2012)





The Gross–Pitaevskii equation describes the ground state of a quantum system of identical bosons using the Hartree–Fock approximation and the pseudopotential interaction model

$$i\hbar\frac{\partial\Psi(\mathbf{r},t)}{\partial t} = \left(-\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r}) + g|\Psi(\mathbf{r},t)|^2\right)\Psi(\mathbf{r},t).$$

$$\Psi(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N) = \psi(\mathbf{r}_1)\psi(\mathbf{r}_2)\ldots\psi(\mathbf{r}_N)$$

Potential

$$H = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2} + \alpha|\psi|^2$$

Trial function

$$\psi = \sqrt{\frac{N}{a}}\left(\frac{2}{\pi}\right)^{\frac{1}{4}} e^{-x^2/a^2}$$

Polarization along z to describe the longitudinal grating

$$\mathbf{P}_{exc}(z,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dz' g(z-z') \int_{-\infty}^{\infty} d\omega[\chi_e(\omega)\mathbf{E}(z',\omega)e^{-i\omega t}]$$

The problems:

1. Developed plasma screens weak dipole interaction between excitons
2. What is the mean free path of excitons?

4. Ionization instability

$$\nabla \times \vec{E} = -\frac{1}{c}\frac{\partial \vec{B}}{\partial t},$$

$$\nabla \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c}\vec{J},$$

$$\frac{\partial n_e}{\partial t} = \sigma_6 E^{12}$$

$$\frac{\partial (n_e \vec{\mathrm{v}})}{\partial t} - i\omega n_e \vec{\mathrm{v}} = -n_e \frac{e}{m_e}\vec{E}$$

Introducing perturbations:

$$\vec{E} = e_x[E_0 + u_1(r,t) + iv_1(r,t)]\exp[i\varphi_0(t) - ikz]$$

$$n_e = n_{e0} + n_{e1}$$

Searching solution in the form

$$u_1, v_1, n_1 \sim \exp(\Gamma t - i\kappa r)$$

The dispersion relation

$$\Gamma[4\Gamma^2 - (\kappa^2 c^2 - 4\omega^2 \varepsilon_0 \varepsilon \cos^2\alpha) - 8i\Gamma\omega\sqrt{\varepsilon_0\varepsilon}\cos\alpha)] =$$

$$= AW_{\mathrm{MPI}}\omega_{pm}^2(1 - \sin^2\alpha\cos^2\phi/(\varepsilon_0\varepsilon))$$

$$A = d\ln W_{\mathrm{MPI}} / d\ln E$$

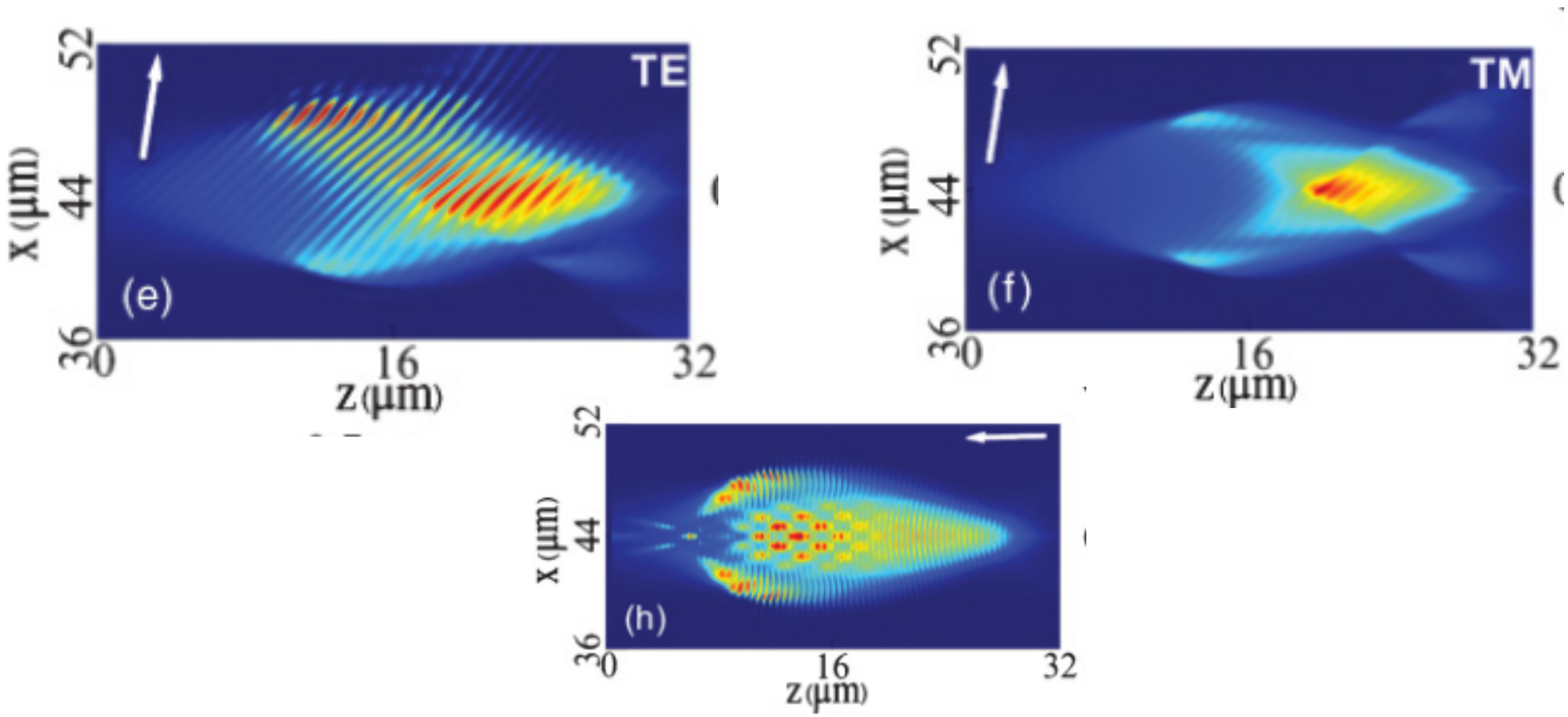
E.S. Efimenko and A.V. Kim, Phys. Rev. E **84**, 036408 (2011)

Instability increment

$$\Gamma_{1,2} = \pm(1-i)\sqrt{\frac{AW_{\mathrm{MPI}}\omega_{pm}^{2}}{16\omega(\varepsilon_0\varepsilon)^{1/2}\cos\alpha}}$$

Instability criterion ($L > c/\mathrm{Re}(\Gamma)$)

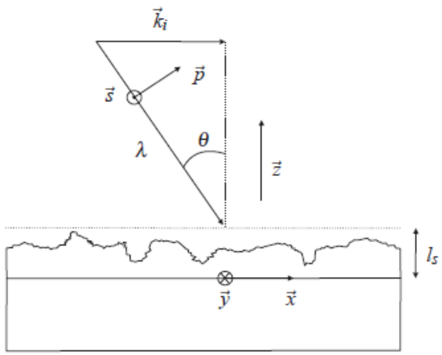
$$\boxed{n_e^{\max}/n_e^{cr} > (16\omega/AW_{\mathrm{MPI}})(\omega^2L^2/c^2)}$$



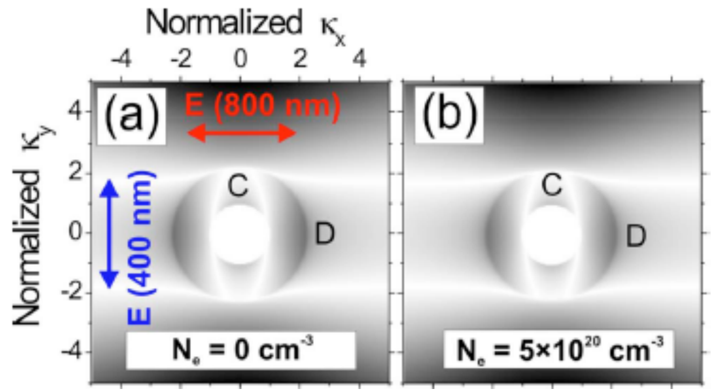
Our case, fused silica:

$W_{\mathrm{MPI}} = 9.6\times10^{-70}I^6\ \mathrm{s}^{-1}$ ⇨ $\mathrm{I} > 2.6\times10^{13}\ \mathrm{W/cm^2}$ or $0.74I^*$

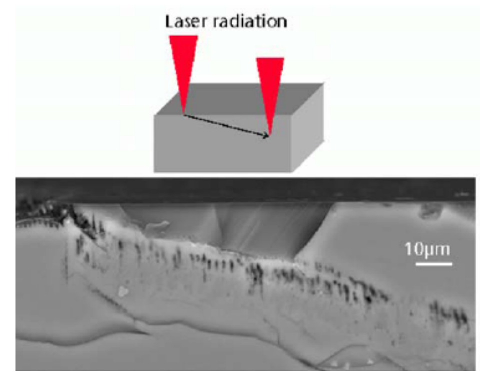
Self-organization of nanoplasma via volumetric Sipe's mechanism?



J.E. Sipe et al.
Phys. Rev. B 27, 1141 (1983)



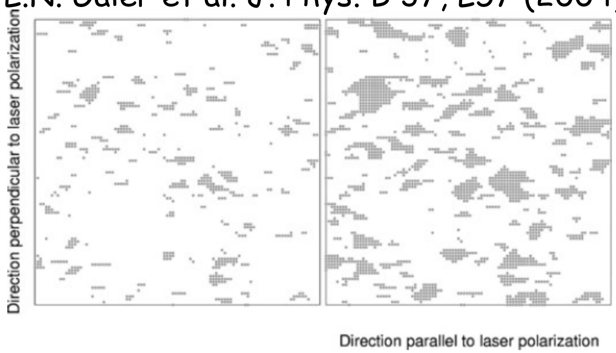
D. Duft et al. J. Appl. Phys. 105, 034908 (2009)



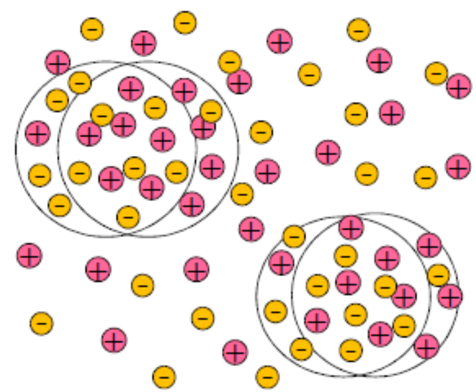
M. Hörstmann-Jungemann et al.
J. Laser Micro/Nanoeng. 4, 135 (2009)

Forest-fire model

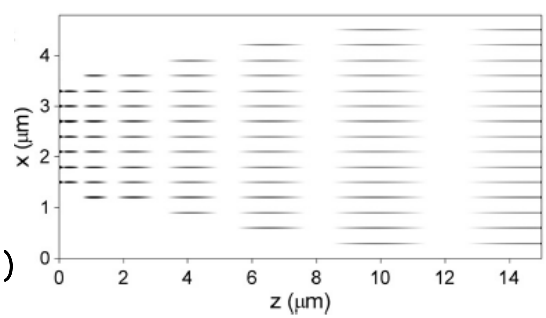
L.N. Gaier et al. J. Phys. B 37, L57 (2004)



Interacting dipoles



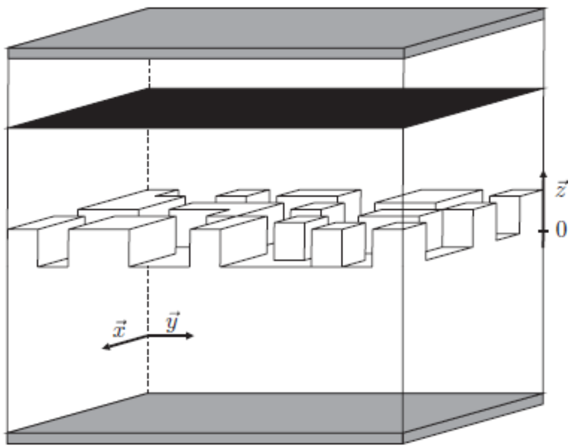
Periodic absorption + ionization instability



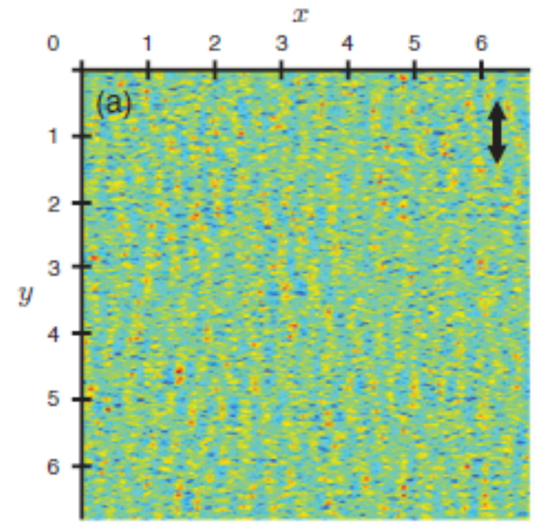
M. Beresna et al.
APL, 101, 053120 (2012)

Volumetric FDTD simulations?

J.Z.P. Skolski et al. Phys. Rev. B 85, 075320 (2012)



D. Duft et al. J. Appl. Phys. 105, 034908 (2009)



In volumetric case refractive index change is governed by several processes that complicates theoretical consideration:

$$\Delta n = \Delta n_{Drude} + \Delta n_{Kerr} + \Delta n_{trap} + \Delta n_{th} + \Delta n_{P} + \Delta n_{\rho}$$

Further details:

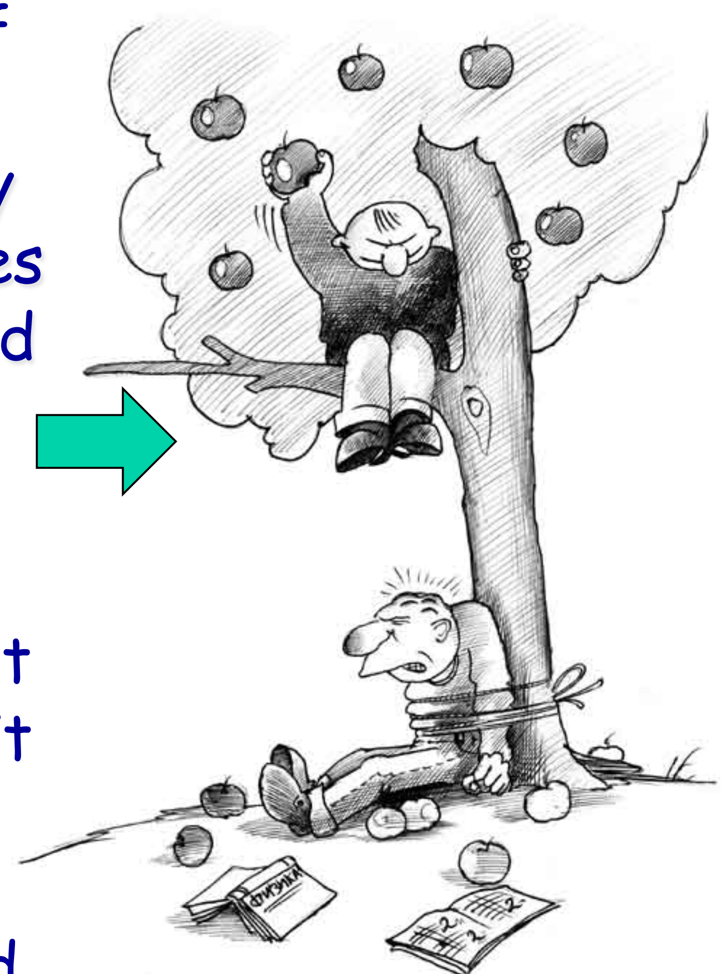
N.M. Bulgakova, V.P. Zhukov, Yu.P. Meshcheryakov, Applied Physics B, accepted

Plausible explanation of pulse front tilt effect;
Material properties favorable for nanograting imprinting

Conclusion

Deep understanding of interaction of ultrashort laser pulses with bulk transparent materials is an extremely complicated phenomenon which requires consolidating knowledge of optics, solid state physics and chemistry, plasma physics, thermodynamics, theory of elasticity and plasticity.

The numerical modeling is an important supplement of experimental studies. It may be used as a very helpful tool to predict and foresee the underlying physics of phenomenon. Many detailed aspects which can be overlooked in experiments may be revealed by numerical modeling.



"Physics lesson"
by Sergei Korsun

Acknowledgements

Co-authors

Dr. V.P. Zhukov,
Institute of Computational Technologies SB RAS,
Novosibirsk, Russia

Dr. Yu. P. Meshcheryakov
Institute of Hydrodynamics SB RAS, Novosibirsk, Russia

Prof. Peter G. Kazansky and Dr. Jörn Bonse for valuable discussions

SEVENTH FRAMEWORK PROGRAMME

Marie Curie International Incoming Fellowship grant, No. 272919

MARIE CURIE ACTIONS

Thank you for attention!