Breaking the limits in in glass and optical fibres: from quantum interference to femtosecond nanostructuring

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Outline

- Self-organised second-order nonlinearity in glass
- Coherent current, coherent photoconductivity and quantum interference in glass - mechanism of self-organised $\chi^{(2)}$ gratings formation
- Self-organised modifications by femtosecond irradiation in glass: refractive index nano-gratings and form-birefringence
- Mechanism of self-organised nano-structures formation
- Evidence of nano-vortex formation

Nonlinear optics: introduction

$$P = \varepsilon_0 \chi^{(1)} E + \underbrace{\varepsilon_0 \chi^{(2)} E}_{\chi^{(1)}} E + \underbrace{\varepsilon_0 \chi^{(3)} E}_{\chi^{(2)}} E E + \ldots$$

$E/E_{at} \sim 1$, $E_{at} \sim 10^8$ V/cm

$$\frac{P_{2\omega}}{P_\omega} \propto \left(\chi^{(2)}\right)^2 L^2 \left(\frac{\sin \Delta k \mathrm{L}}{\Delta k \mathrm{L}}\right)^2$$

$$\Delta k = k_{2\omega} - 2k_\omega$$

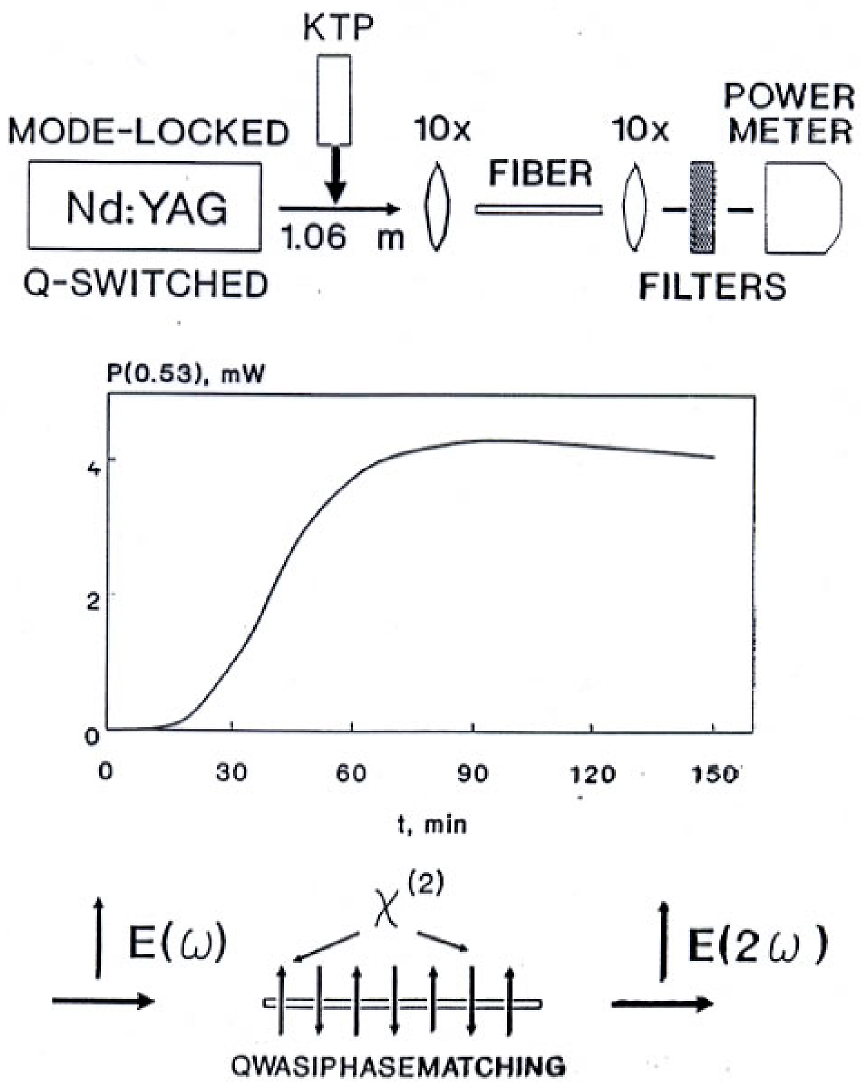
$$l_c = \frac{\pi}{\Delta k} = \frac{\lambda}{4\left(n_{2\omega} - n_\omega\right)}; \Lambda = 2 l_c$$

$$\chi^{(2)} \sim \cos\left(2\pi / \Lambda\right) \sim \cos\left(k_\omega - 2k_{2\omega}\right)$$

$$k_{2\omega} - 2k_\omega + \frac{2\pi}{\Lambda} = 0$$

Photoinduced SHG in Optical Fibres

(Osterberg and Margulis, Opt. Lett. 11, 516 (1986))



Stolen and Tom (1987)

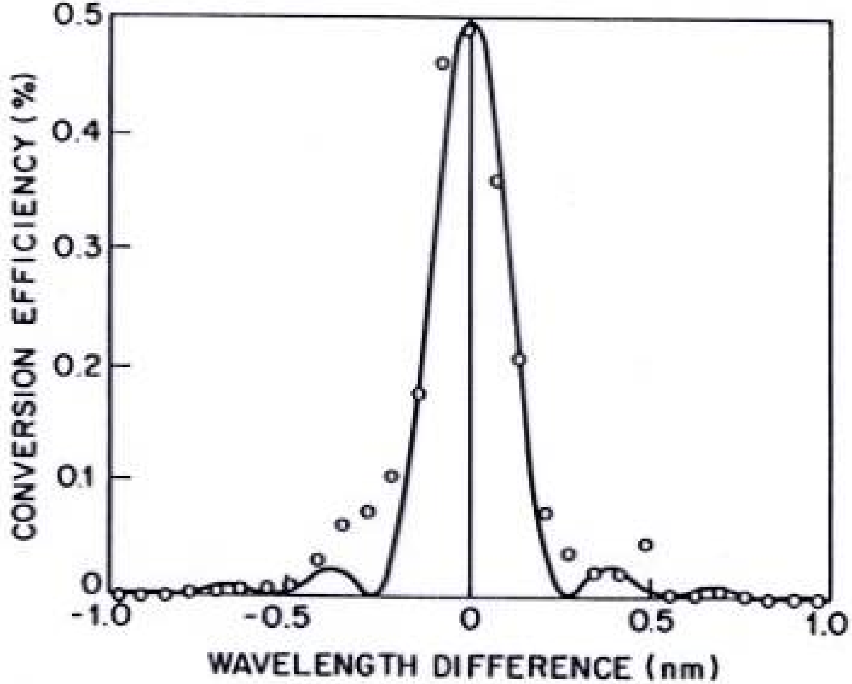
$$\omega + \omega - 2\omega = 0$$

$$P^{(3)} = \varepsilon_0 \chi^{(3)} E_\omega E_\omega E_{2\omega} \sim$$

$$\sim \cos (k_{2\omega} - 2k_\omega)$$

$$\chi^{(2)} = \gamma P^{(3)}$$

- **M.C. Farries, P.St.J. Russell, M.E. Fermann and D.N. Payne (1987)**



$$\frac{P_{2\omega}}{P_{\omega}} \propto d^2 L^2 \left(\frac{\sin \Delta k \mathrm{L}}{\Delta k \mathrm{L}} \right)^2$$

$$\Delta k = k_{2\omega} - 2k_{\omega} + \frac{2\pi}{\Lambda} = \frac{4\pi (n_{2\omega} - n_{\omega})}{\lambda} + \frac{2\pi}{\Lambda}$$

$E_{dc} \sim P^{(3)} \sim 1$ V/cm

is very small !

$\chi^{(2)} = 3\chi^{(3)} E \sim 10^{-3}$ pm/V

$E_{dc} \sim 10^{4}$ V/cm

PHOTOREFRACTIVE EFFECT

Photogalvanic Effect
in Media
Without inversion Symmetry

$$j_i = \beta_{ijk} E_j E_k^*$$

Ferroelectrics: $LiNbO_3$, $LiTaO_3$, $BaTiO_3$, ...

Coherent
Photogalvanic Effect

$$j_i = \beta_{ijkl} E_j^{\omega} E_k^{\omega} E_l^{2\omega *}$$

Any media: glass, crystals, ...

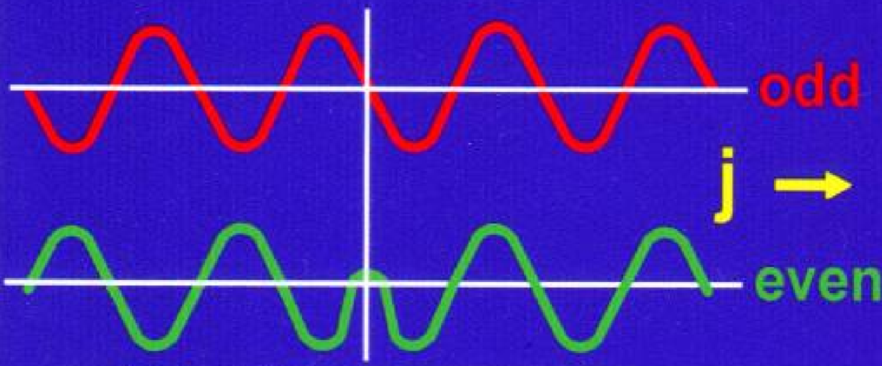
$\sigma E + j = 0$

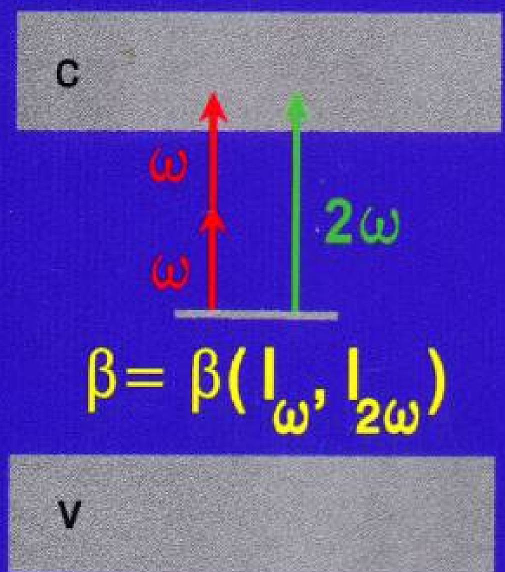
$$E_{dc} = -j/\sigma \sim 10^3 - 10^5 \text{ V/cm}$$

COHERENT PHOTOCURRENT (centrosymmetric media):

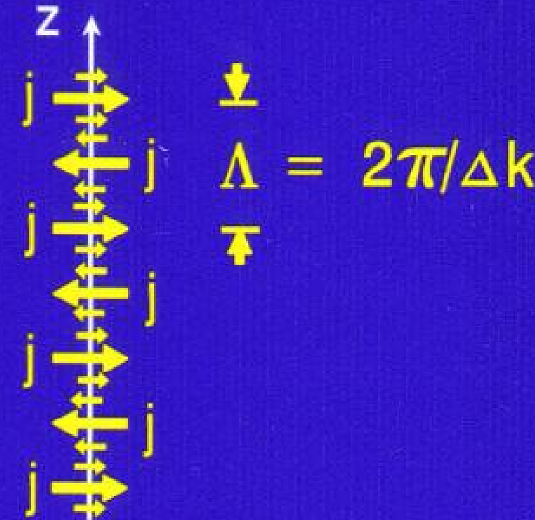
Dianov, Kazansky and Stepanov, 1989

$$j = \beta \, E_\omega E_\omega E^*_{2\omega} \sim \cos \Delta kz$$





$$\beta = \beta(I_\omega, I_{2\omega})$$



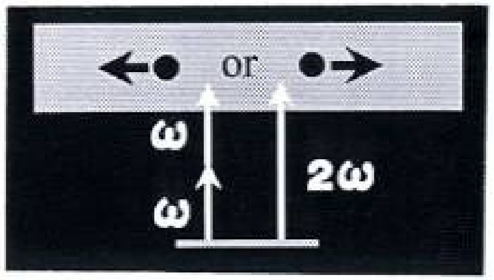
$E = j/\sigma$, $\sigma(z) = \text{const}$

$E \sim 10^5$ V/cm

$\chi^{(2)} = \chi^{(3)} E \sim \cos \Delta kz$

MODULATION OF ANGULAR DISTRIBUTION OF PHOTOELECTRONS

(COHERENT PHOTOCURRENT):



- **rubidium atoms**
  Y.Y. Yin, C. Chen, D.S. Elliot and A.V. Smith
  Phys. Rev. Lett., v.69, 2353 (1992)

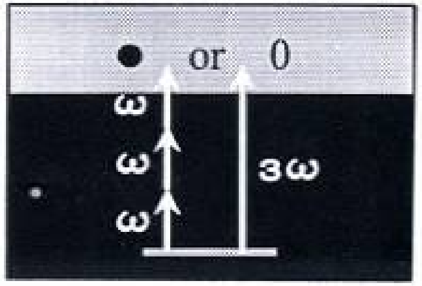
- **Sb-Cs photocathodes**
  N.B. Baranova, A.N. Chudinov, A.A. Shulginov and B.Ya. Zel'dovich
  Opt. Lett., v.16, 1346 (1991)

- **AlGaAs/GaAs superlattices; bulk GaAs**
  E. Dupont, P.B. Corkum, H.C. Liu, M. Buchanan and Z.R. Wasilevski
  Phys. Rev. Lett., v.74, 3596 (1995);
  R. Atanasov, A. Hache, J.L.P. Hughes, H.M. van Dreil and J.E. Sipe
  Phys. Rev. Lett., v.76, 1703 (1996)

- **silica glass**
  E.M. Dianov, P.G. Kazansky and D.Yu. Stepanov
  Sov. J. Quantum. Electron., v.19, 575 (1989)

MODULATION OF CROSS SECTION OF IONIZATION

(COHERENT PHOTOCONDUCTIVITY):



- **xenon gas**
  J.C. Miller, R.N. Compton, M.G. Payne and W.R. Garrett
  Phys. Rev. Lett., v.45, 114 (1980)

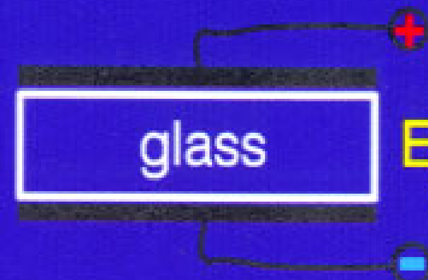
- **HCl molecular beam**
  S.M. Park, S.P. Lu and R.J. Gordon
  J. Chem. Phys., v.94, 8622 (1991)

- **mercury vapor**
  E. Dupont, P.B. Corkum, H.C. Liu, M. Buchanan and Z.R. Wasilevski
  Phys. Rev. Lett., v.74, 3596 (1995)

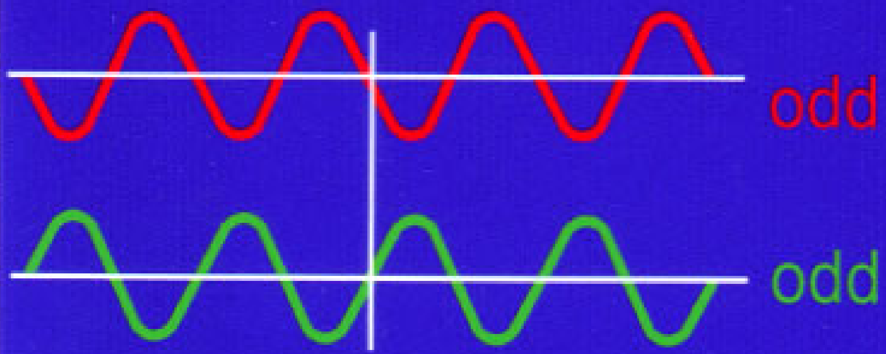
- **media without inversion symmetry, $\omega + \omega - 2\omega$**
  R.J. Glauber
  Quantum Optics (1969)

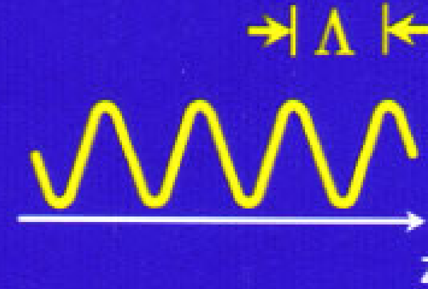
COHERENT PHOTOCONDUCTIVITY (non-centrosymmetric media):

$$\sigma_c = \gamma \; E_\omega E_\omega E^*_{2\omega} \sim \cos \Delta k\, z$$

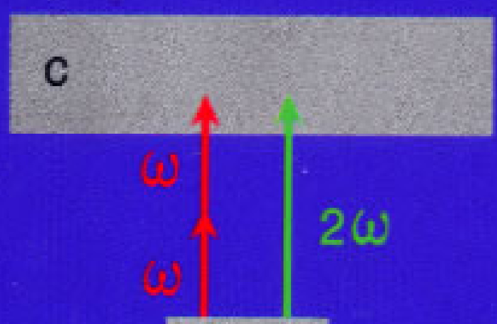


$E_0 \sim 10^7$ V/cm





$$\sigma = \sigma_0 + \sigma_c$$



$$\Delta E \simeq \sigma_c \, \sigma_0^{-1} E_0 (1 - e^{-t/\tau_0}) e^{-t/\tau_0}$$

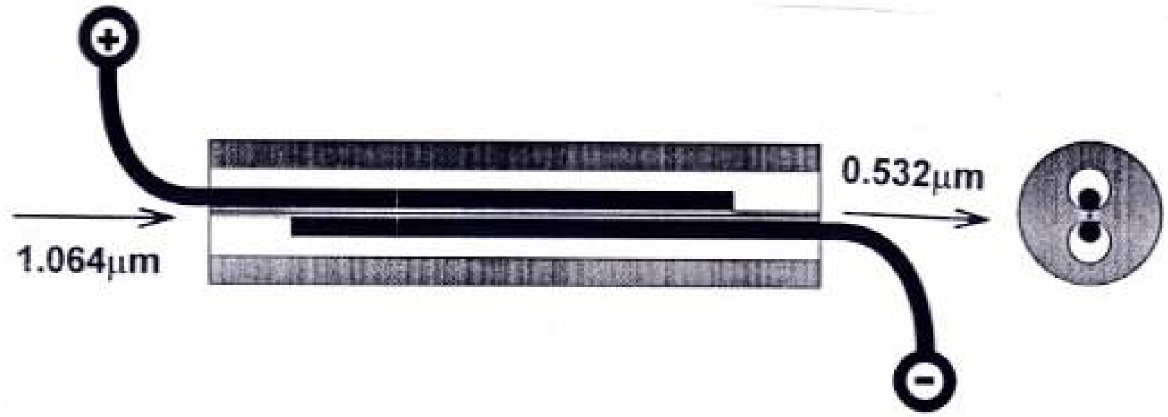
$$\tau_0 = \epsilon / \sigma_0$$

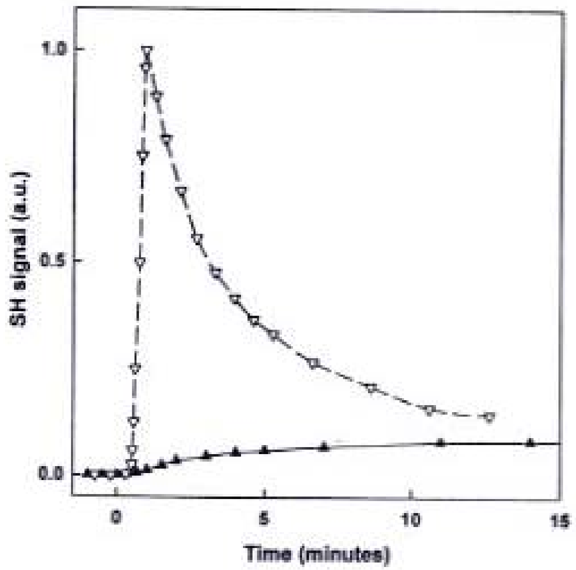
$$\gamma = \gamma \, (I_\omega, I_{2\omega}, E_0)$$

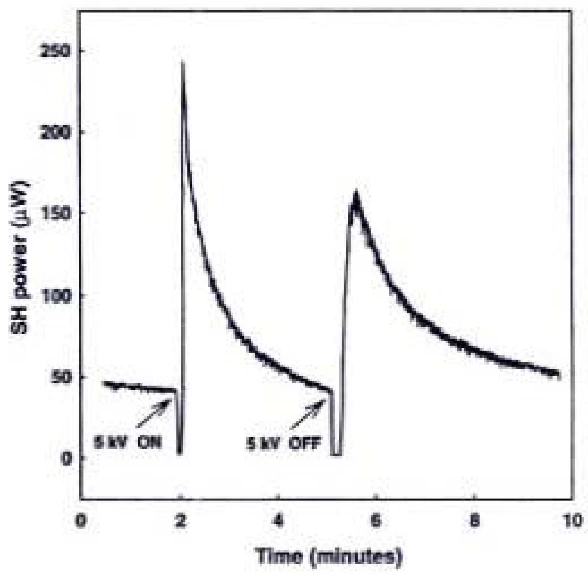
$$\chi^{(2)} = \chi^{(3)} \Delta E \sim \cos \Delta kz$$

Electrically stimulated light-induced SHG in glass

(P.G. Kazansky and V. Pruneri, Phys. Rev. Lett. 78, 2956 (1997))







The observed phenomenon represents the first evidence of *coherent photoconductivity* in solid state materials!

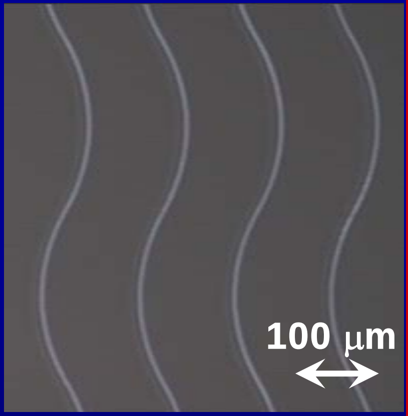
Femtosecond Direct-writing: *The Principle*

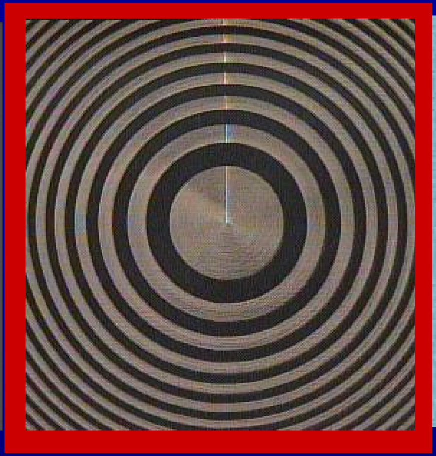
- Tight focusing of laser (λ =850 nm, $\Delta\tau$ =150 fs) into glass
- High intensity leading to multi-photon absorption
- Structural changes in matter confined to focal volume due to short pulse duration – 3-D
- Photosensitivity not required
- -ve or +ve index changes

Intensity ~ 10^{14} W/cm^2
Temperature ~ 10^6 K
Pressure ~ 10^6 bar

Classification of direcly-written structures

- Type 1: smooth positive index change (waveguides, couplers, etc.)
- Type 2: birefringent features, anisotropic reflection and negative index change (embedded microreflectors, Fresnel zone plates, etc.)
- Type 3: voids embedded into glass (photonic crystals, data storage)



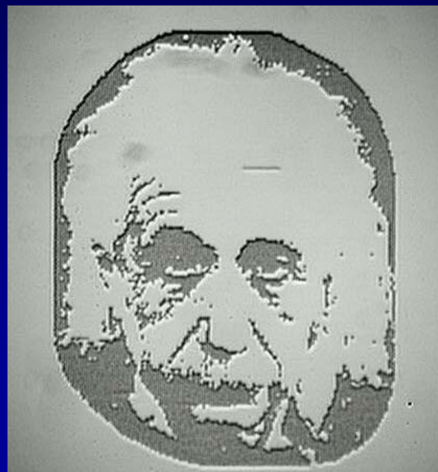


K. M. Davis et al., Opt. Lett., 21, 1729 (1996).

E. N. Glezer & E. Mazur, Appl. Phys. Lett., 71, 882 (1997).

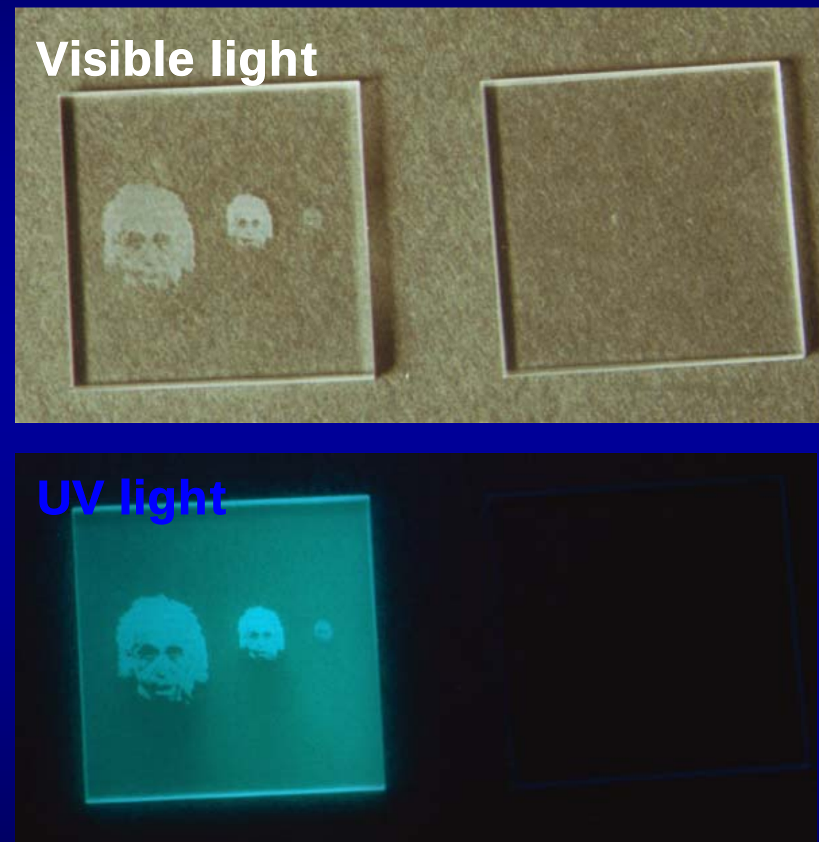
The transition intensity threshold between one kind of structure to the other depends on the processed material, laser's pulse duration and wavelength.

Application of refractive index changes



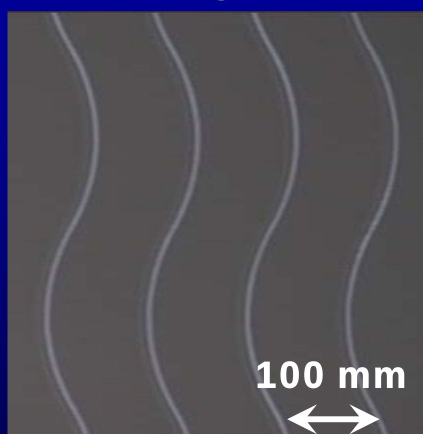
K. Hirao et al., New Glass, 16, 15 (2001).

Optical memory



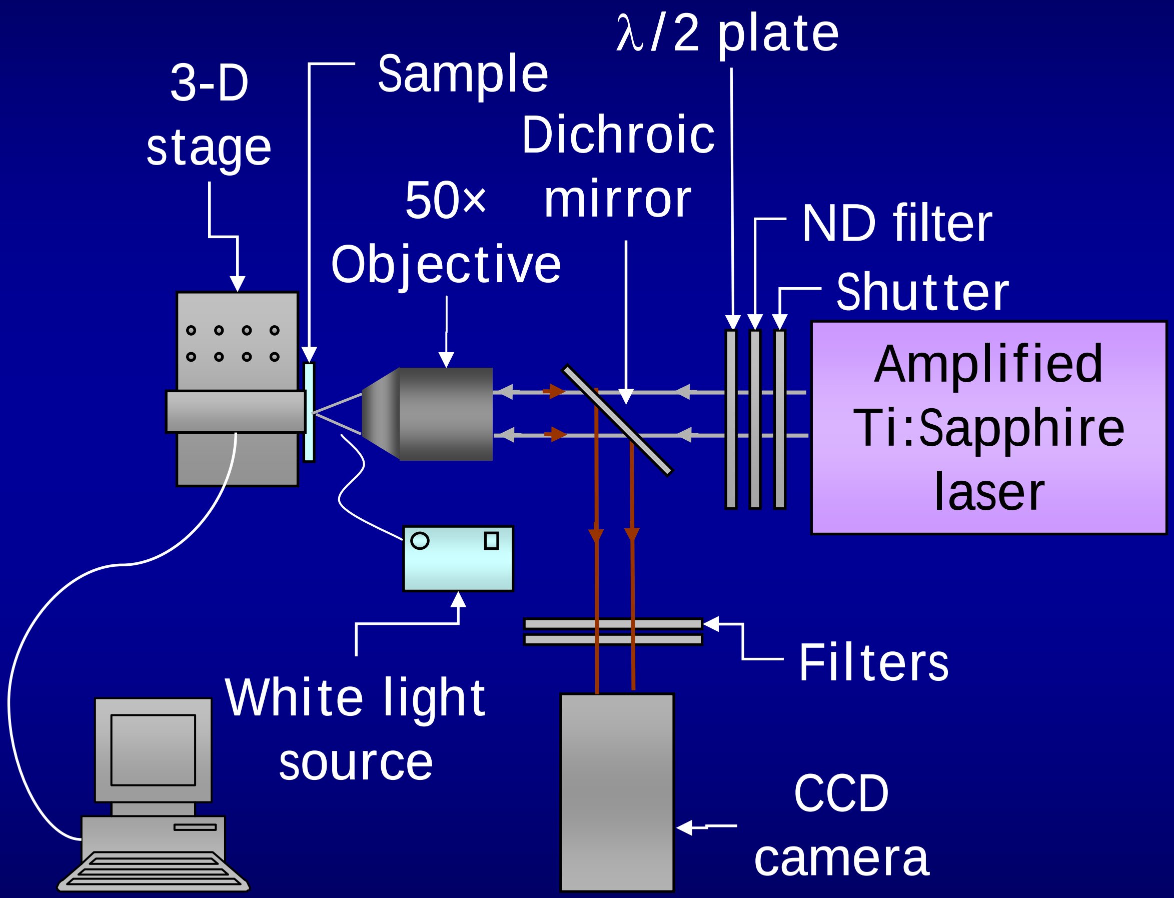
J. Qiu et al., O Plus E, 74, 10 (2001).

Waveguides



K. M. Davis et al., Opt. Lett., 21, 1729 (1996).
K. Miura et al., Appl. Phys. Lett., 74, 10 (1999).

Experimental set up



<u>Laser parameters</u>:

λ = 850 nm

f = 250 kHz

T_{FWHM} = 150 fs

E_{pulse} = 1.1 to 1.3 μJ

<u>Objective</u>: 50×

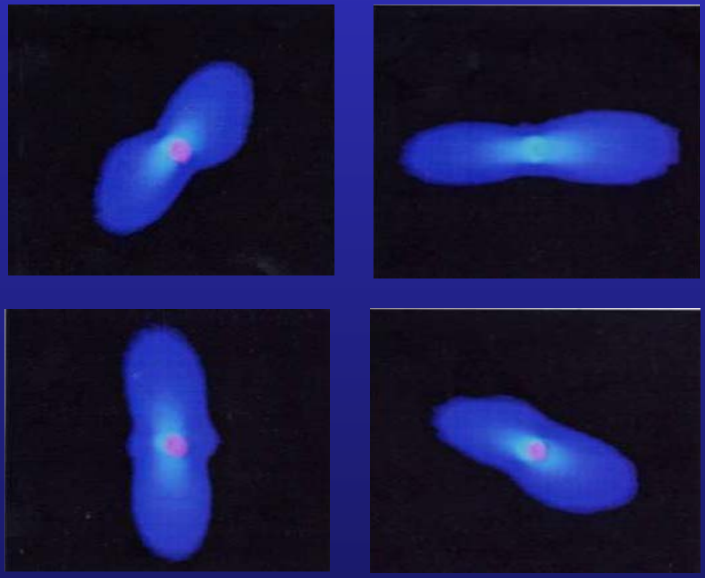
NA=0.55

<u>3-D stage</u>:

100 nm position accuracy

Anisotropic phenomena during femtosecond direct writing

Anisotropic blue luminescence



Blue luminescence pattern
is elongated along laser polarization

Anisotropic 3d harmonic generation

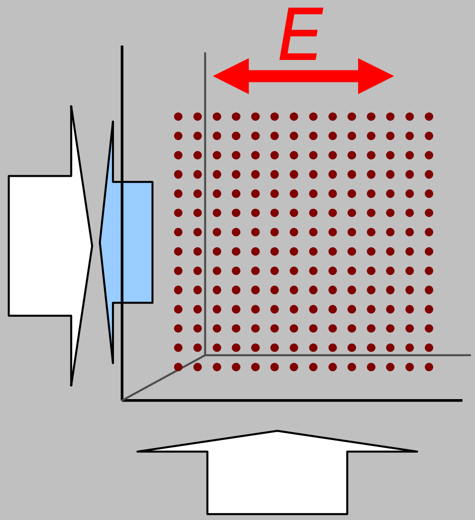
Silica: Microscope images of anisotropic reflection

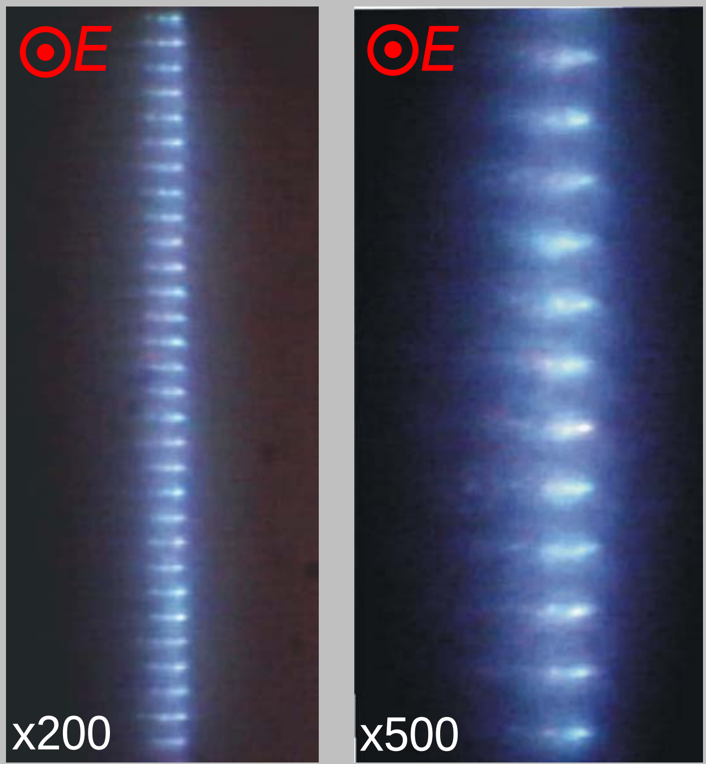
Unpolarized white light

Reflected blue light

E Direct-write polarization

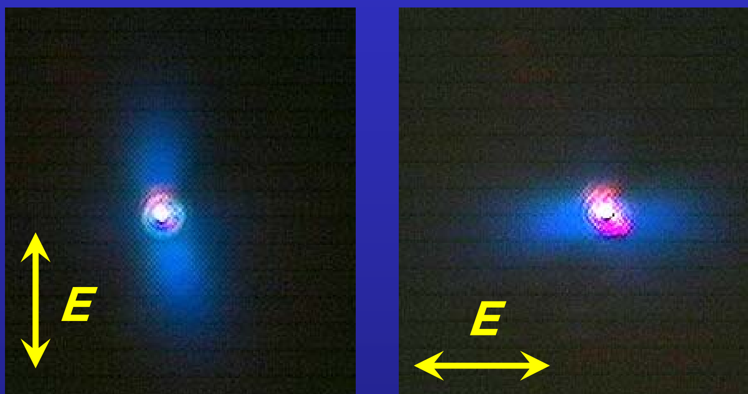
period = 7 µm
pulse energy = 0.9 μJ





Anisotropic phenomena

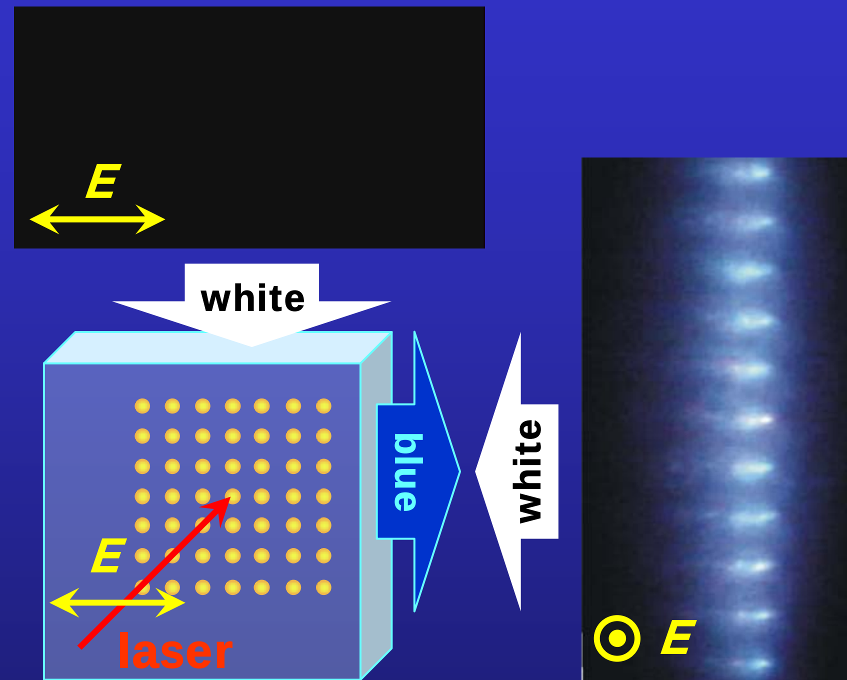
Anisotropic light scattering



P. G. Kazansky et al.,
Phys. Rev. Lett., 82, 2199 (1999).

Blue luminescence pattern is elongated along laser polarization.

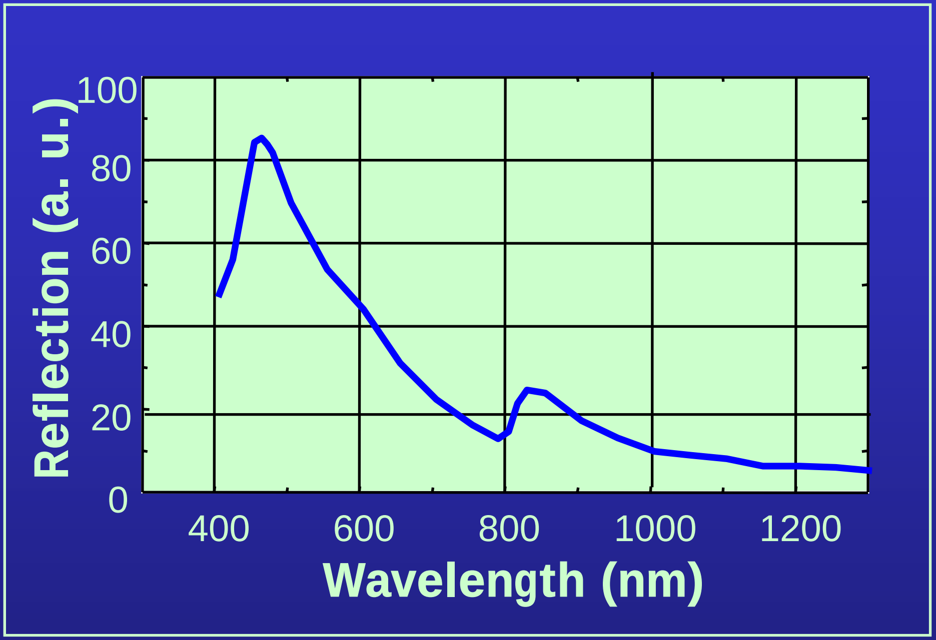
Anisotropic reflection

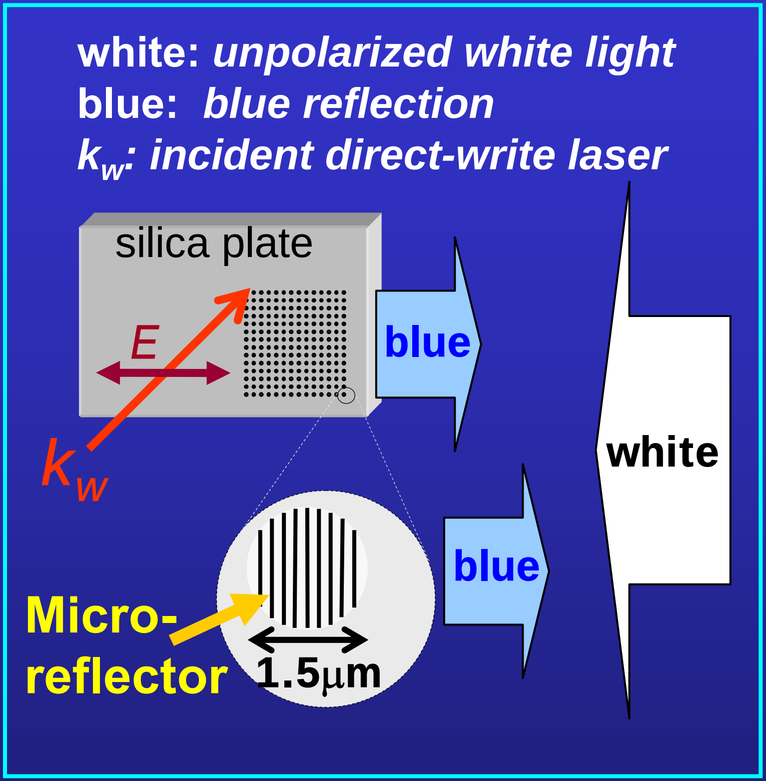


J. D. Mills, et al.,
Appl. Phys. Lett., 81, 196 (2002).

Reflection occurs only in direction parallel to laser polarization.

Embedded micro-reflectors

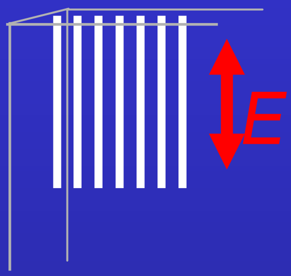




- **Peak at λ=460 nm**
- **Period Λ~150 nm assuming Λ=λ/2n**
- **Birefringent?**

Anisotropic reflection vs birefringence

Polished down to structure

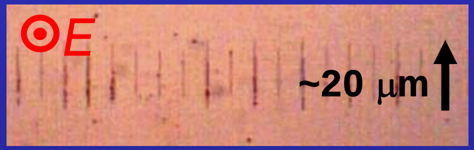


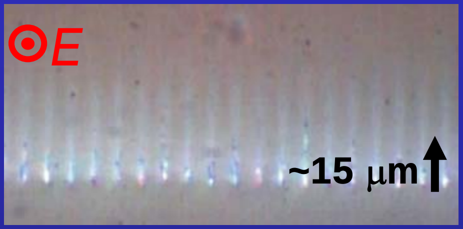
0.4 μJ/pulse

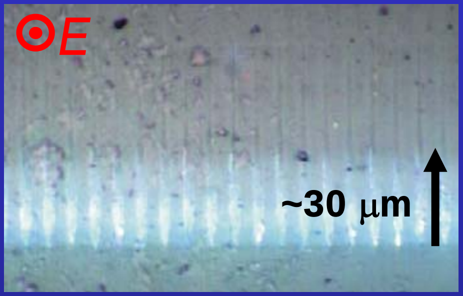
0.6 μJ/pulse

1.1 μJ/pulse

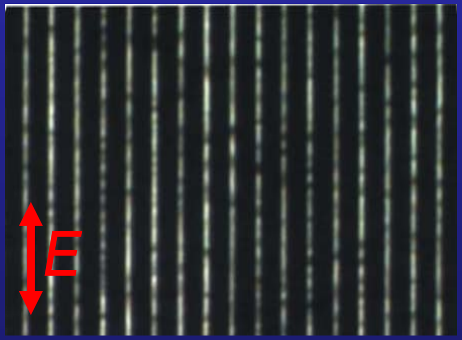
Illuminated and viewed from above

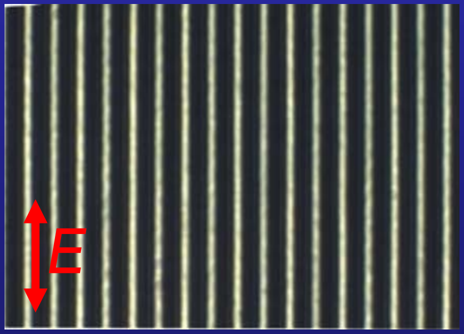






Illuminated from behind, viewed from the front. Sample between cross-polarizers



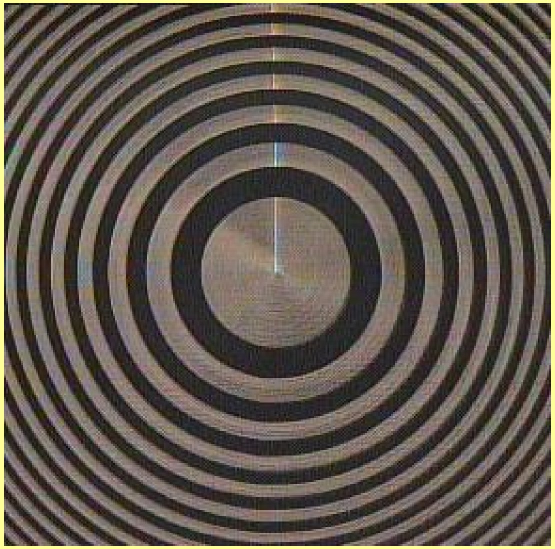


Threshold for onset of reflection AND birefringence ~0.5 μJ/pulse

Birefringence & efficiency

Structures directly-written in silica above a certain threshold (E_{pulse} > 0.5 μJ) show uniaxial birefringence

FZP B



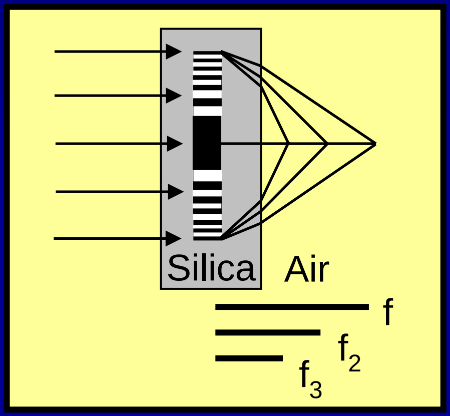
between
Cross polarizers

R = 1 mm
f = 2.4 cm
λ= 632.8 nm
N = 70
E_{pulse} = 1.3 μJ

Fresnel zone plates (FZP)

Focusing element consisting
of a series of alternate concentric zones

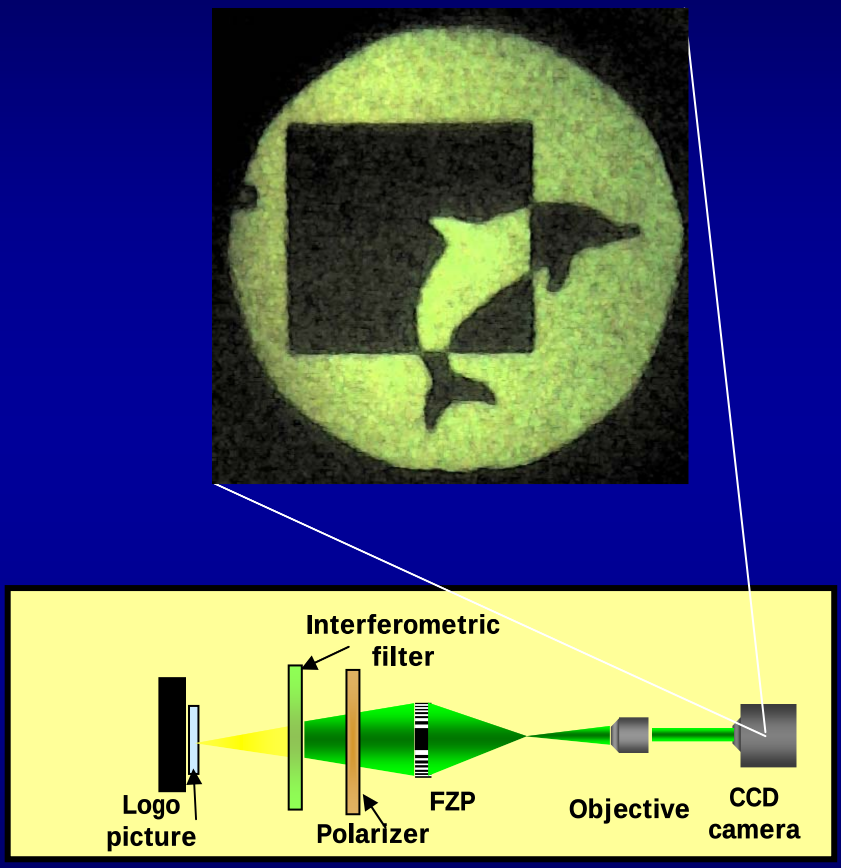
$$R_m = \sqrt{mf\lambda}$$

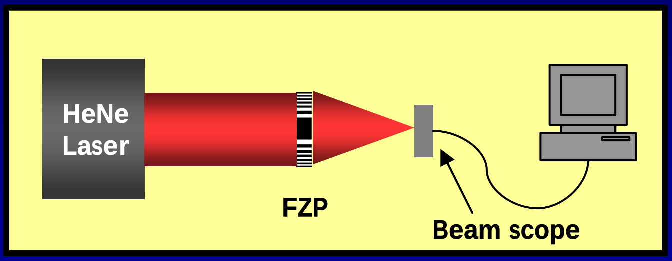


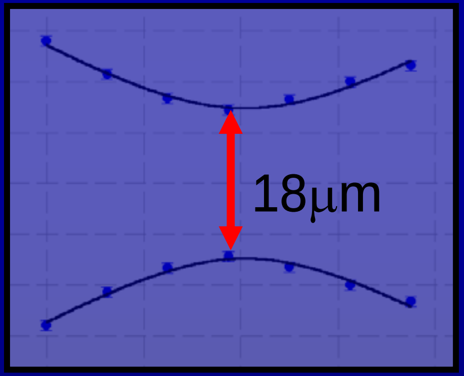
m: integer number
of m^{th} zone
f: focal length
λ: wavelength

- Amplitude Fresnel zone plates
- Phase Fresnel zone plates

Focusing Properties

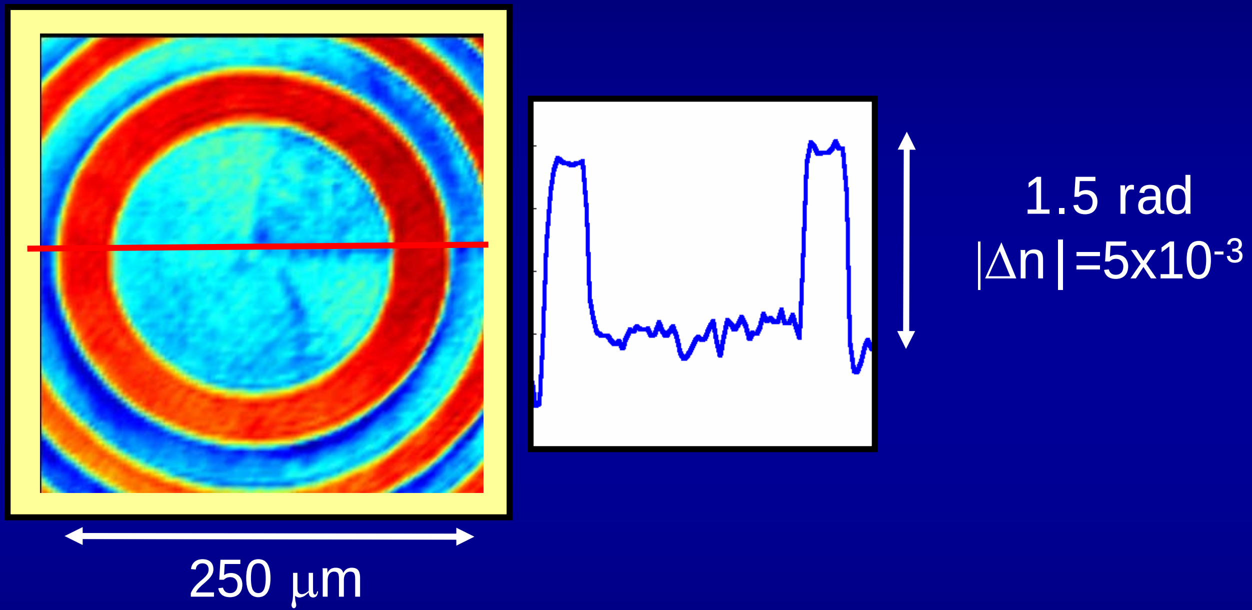






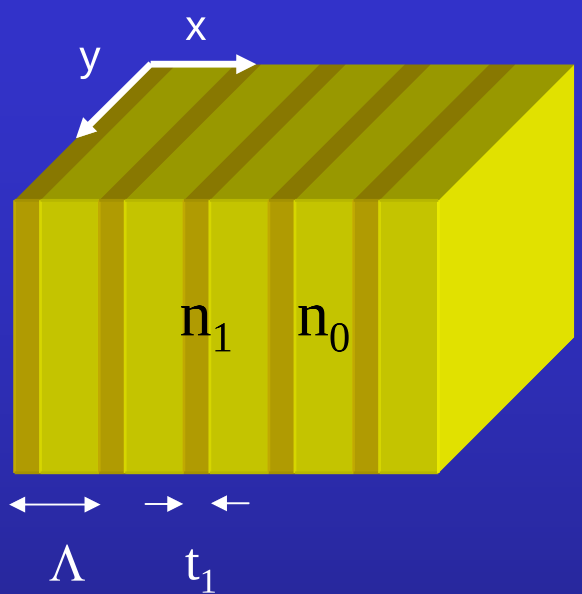
$w_{1/e^2}=18\ \mu m$

Change of refractive index



Index change is negative

Self-organized form birefringence

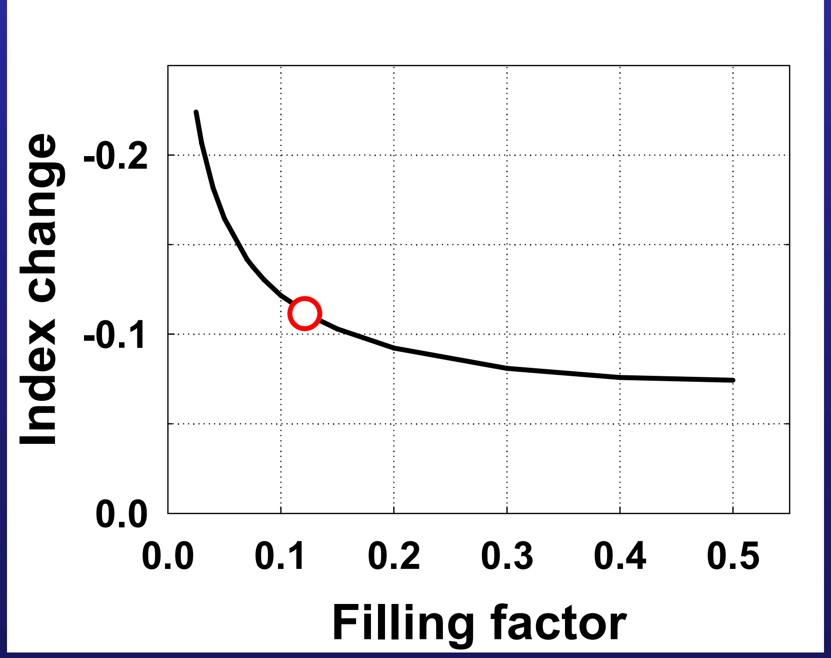


$$n_{xy(//)} = n_0 - \Delta n_{xy} = \sqrt{n_1^2 q + n_0^2 (1-q)}$$

$$n_{xx(\perp)} = n_0 - \Delta n_{xx} = \frac{1}{\sqrt{\frac{1}{n_1^2} q + \frac{1}{n_0^2}(1-q)}}$$

index change : $\Delta n = n_1 - n_0$

filling factor : $q = \frac{t_1}{\Lambda}$



$\Delta n_{xx} = -5 \times 10^{-3}$

$\Delta n_{xy} = -2 \times 10^{-3}$

$n_0 = 1.45$

$t_1 = 20 nm, \Lambda = 150 nm \Rightarrow q = 0.13$

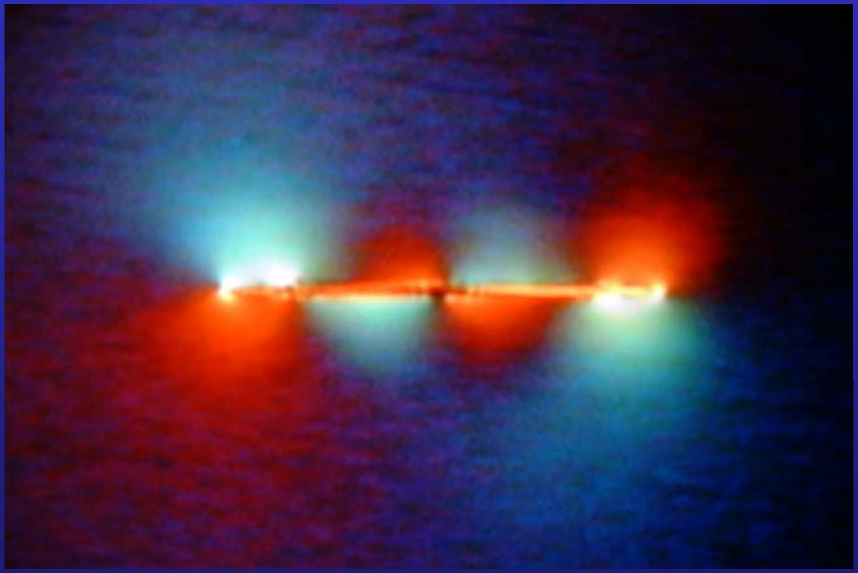
Other materials

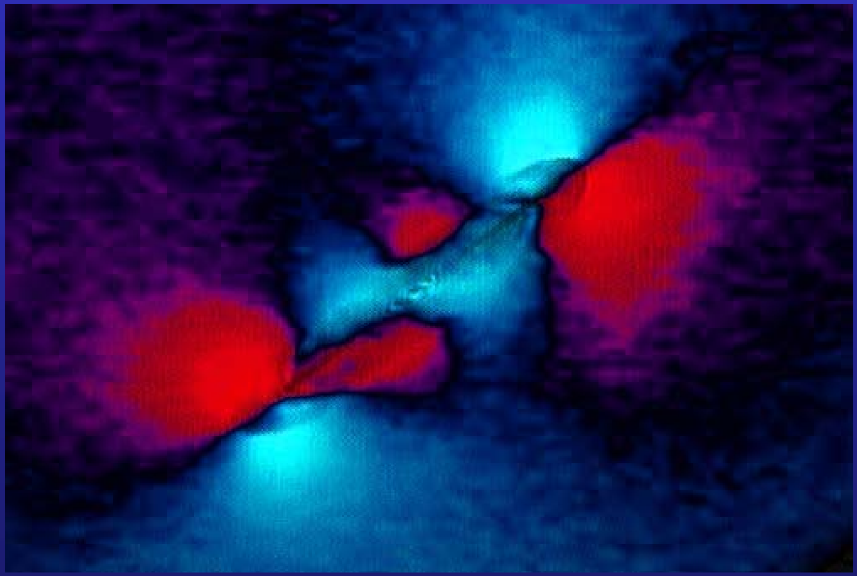
Samples in approximate order of write-uniformity	Birefringent?		Reflection?	
* Soda-lime glass	No		No	
Nanocrystal	No		No	
BK7	No		No	
Sapphire	Yes	One write-direction only	Yes	Small
Ge-doped silica	Yes		Yes	Reddish/Bluish
* Fused silica	Yes		Yes	Blue
Sol-gel silica	Yes		Yes	Reddish

Direct-write performed under identical conditions 1.1 μJ/pulse

Fresnel zone plate in cross polarizers:
Evidence of stress-induced birefringence

Fresnel Galaxy (K2B)

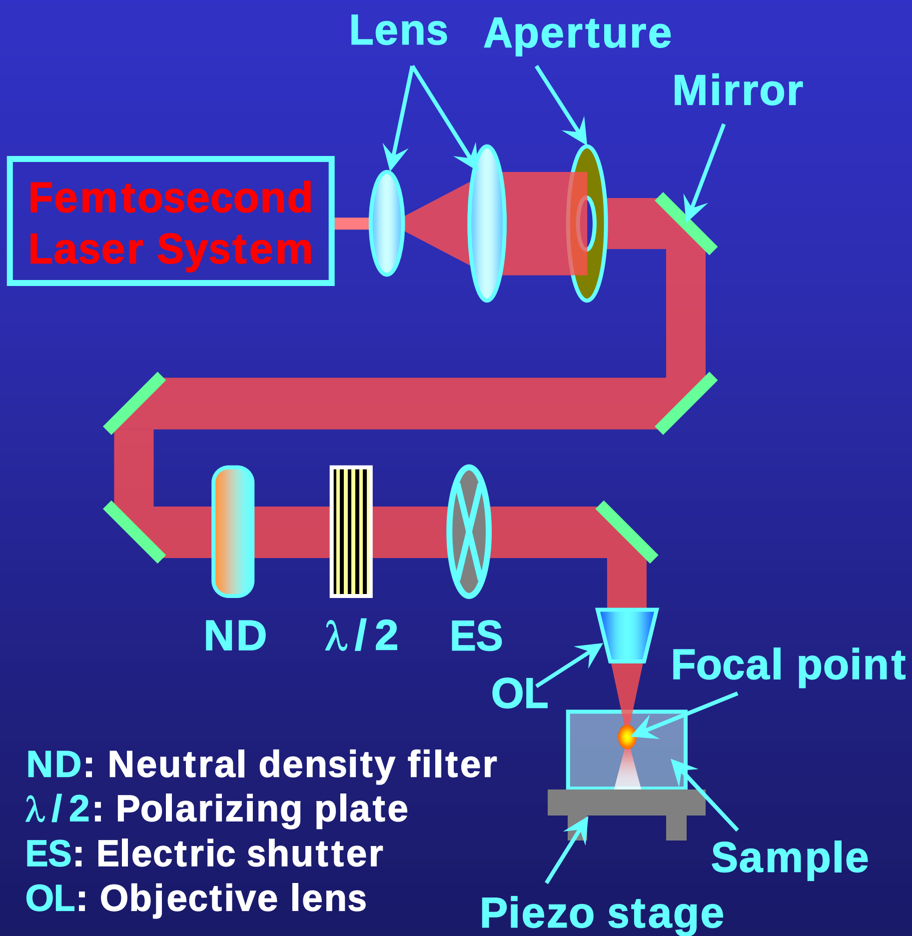




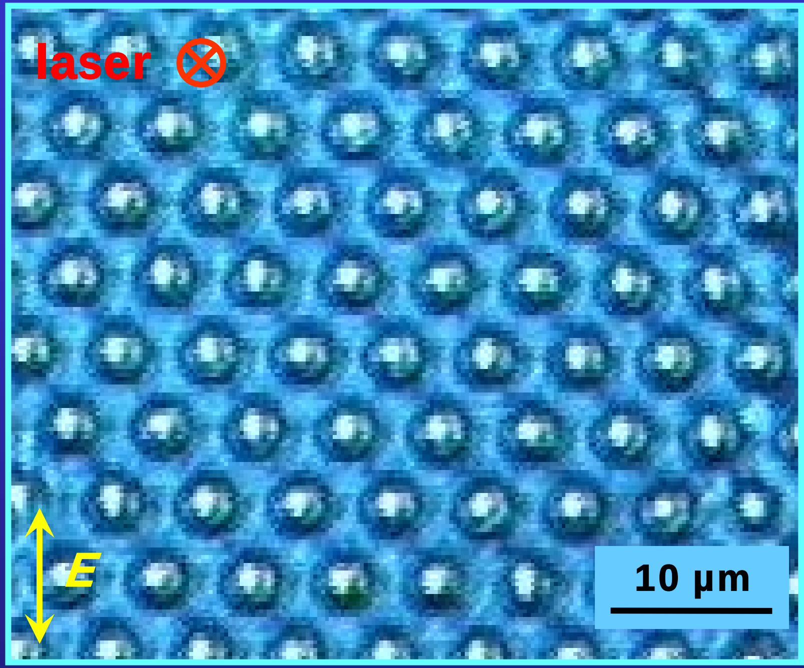
Side-view: stress birefringence in surrounding glass

Experiment

Fabrication system



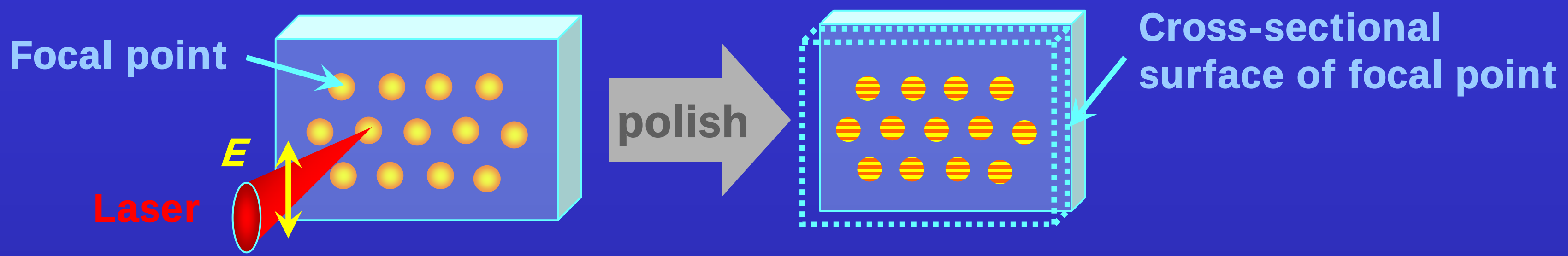
Microscope image

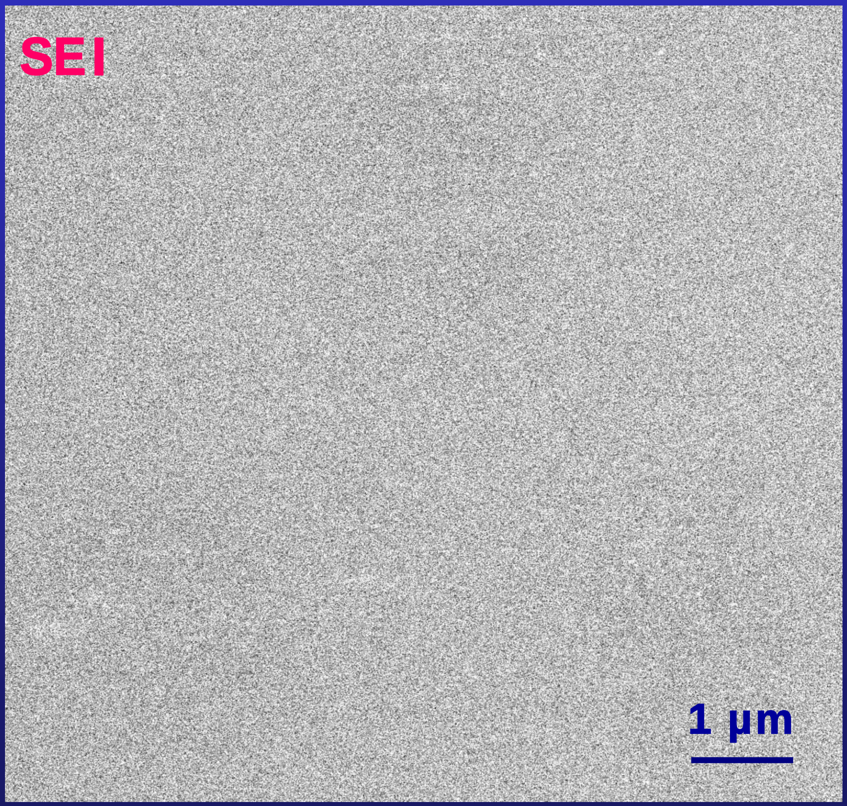


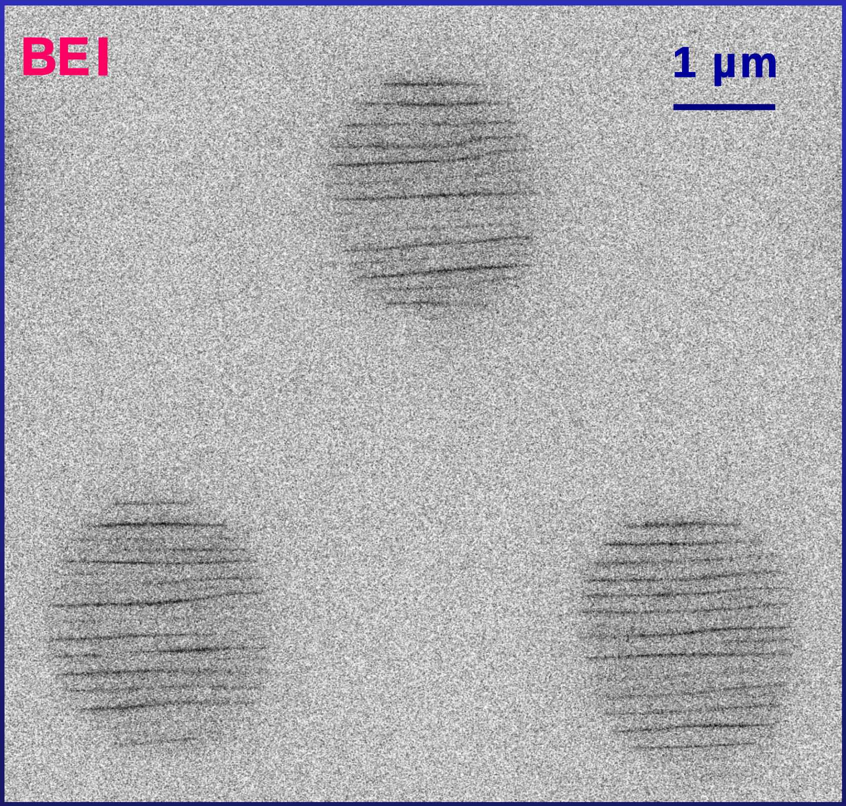
Conditions

wavelength	: 800 nm
pulse duration	: 150 fs
repetition rate	: 200 kHz
pulse energy	: □1.0 μJ
objective	: ×100 (NA=0.95)
polarization	: vertical direction

Induced structural change





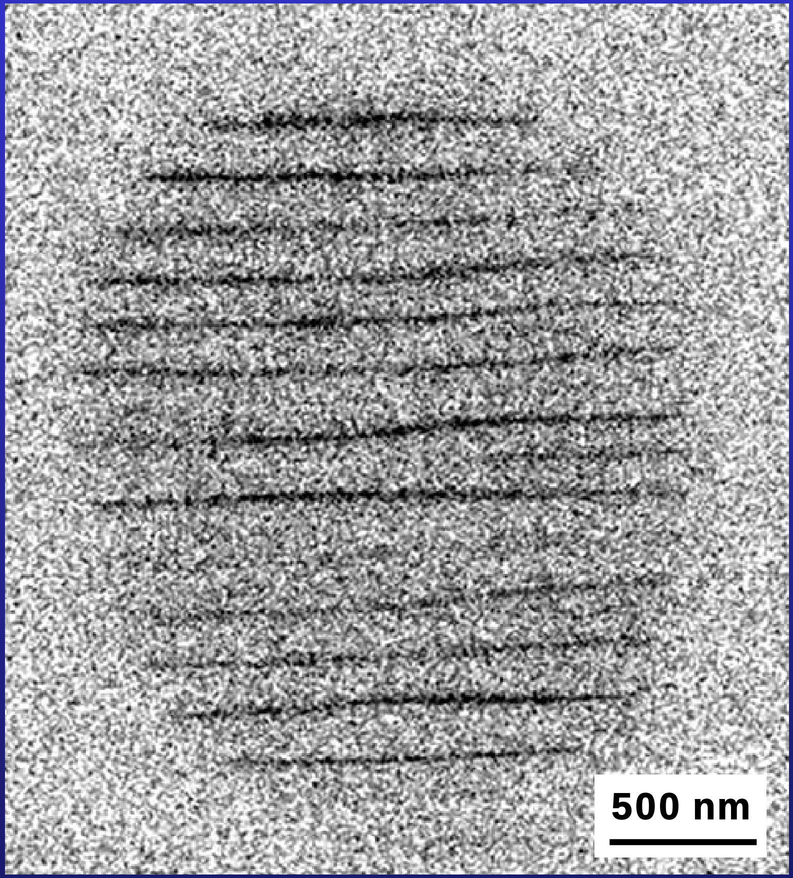


Magnification: x10000
accelerate voltage: 30 kV

Magnification: x10000
accelerate voltage: 30 kV

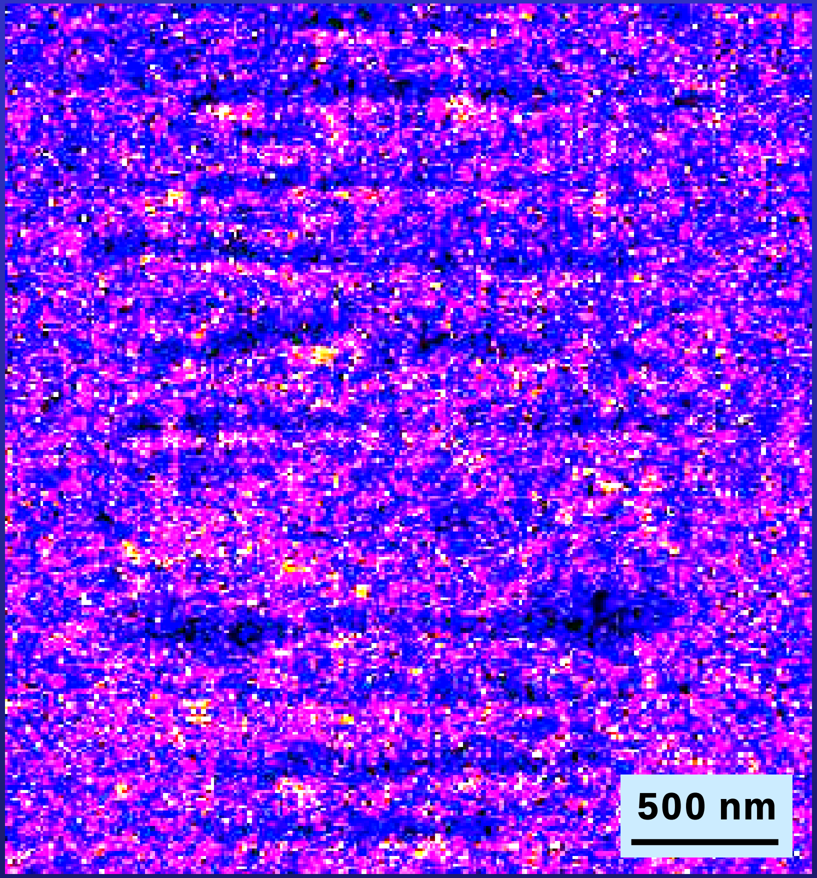
Self-organized nanostructure

BEI



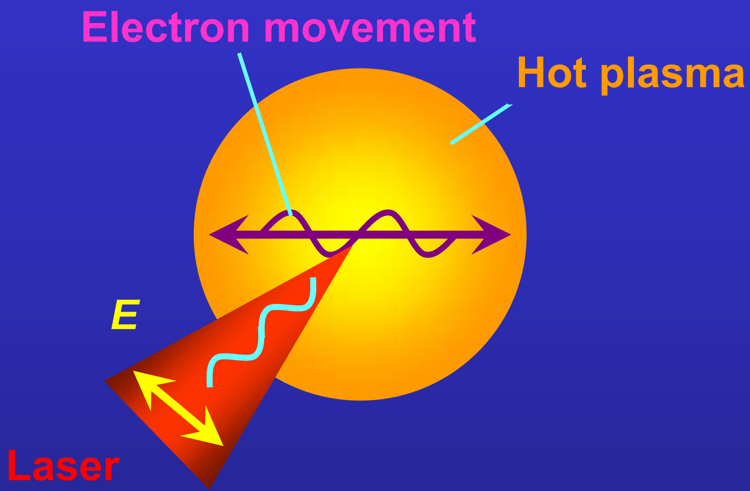
Magnification: x30000
accelerate voltage: 30 kV

Oxygen mapping by AES



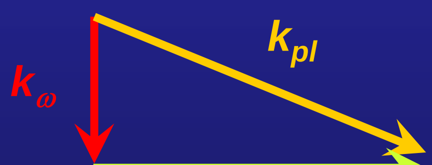
Magnification: x30000
accelerate voltage: 20 kV

Interference between light and electron plasma wave



momentum conservation

$$k_g = k_{pl} - k_\omega$$

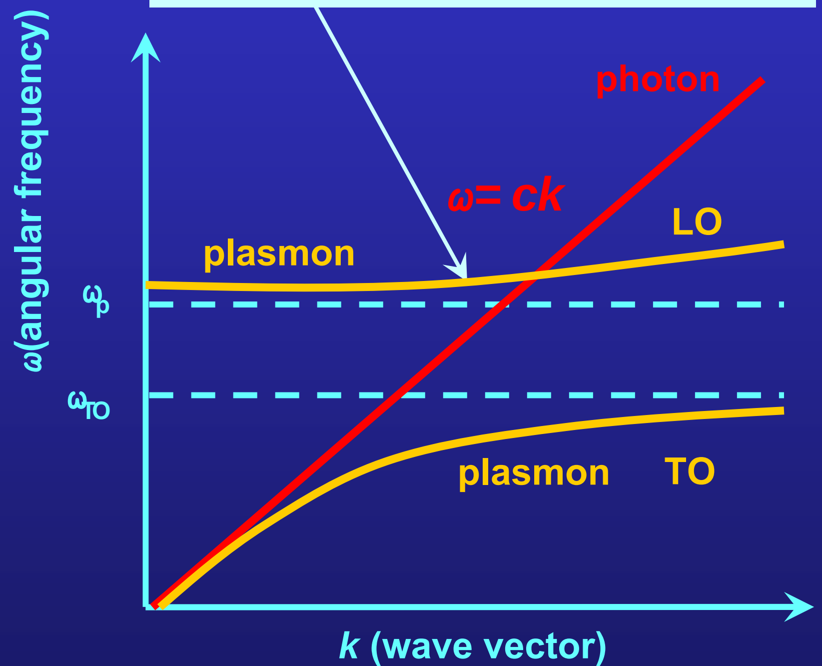


$$k_g = \frac{2\pi}{\Lambda}$$

- k_ω: light wave vector
- k_{pl}: plasmon wave vector
- k_g: grating vector
- Λ: period of nanostructure

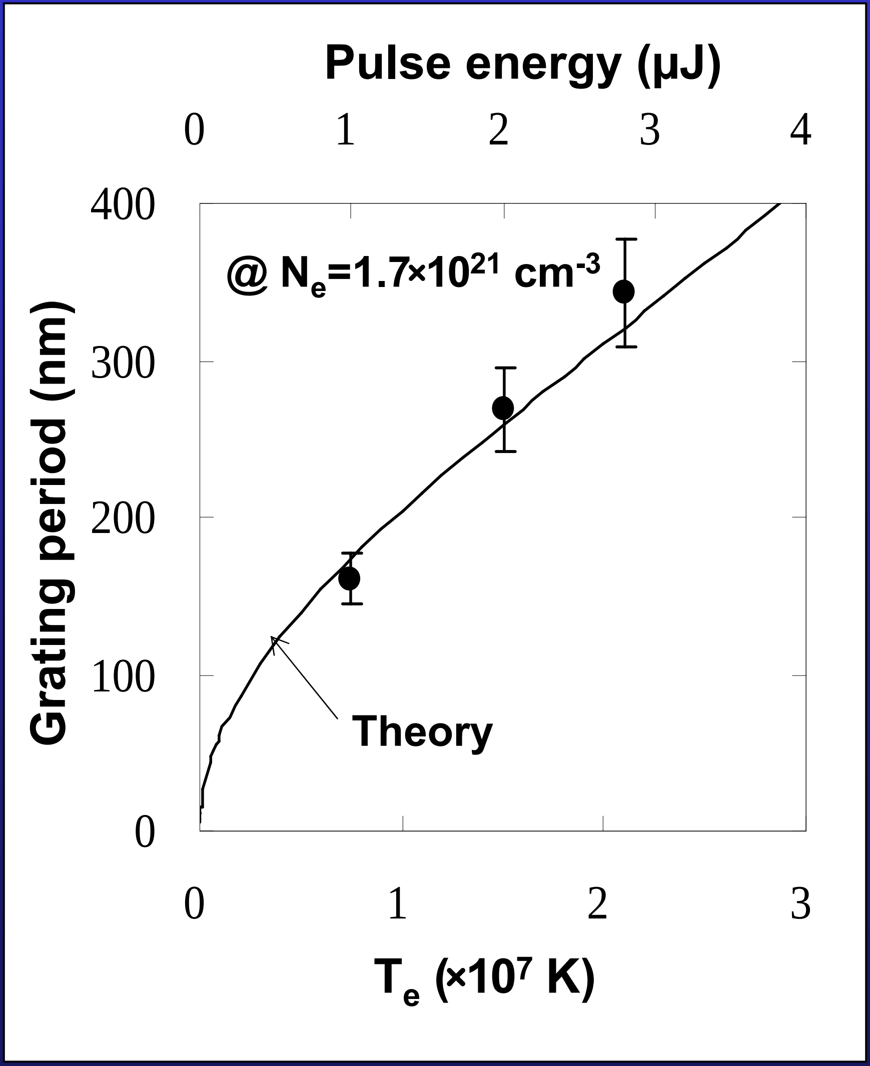
Dispersion relations and energy conservation

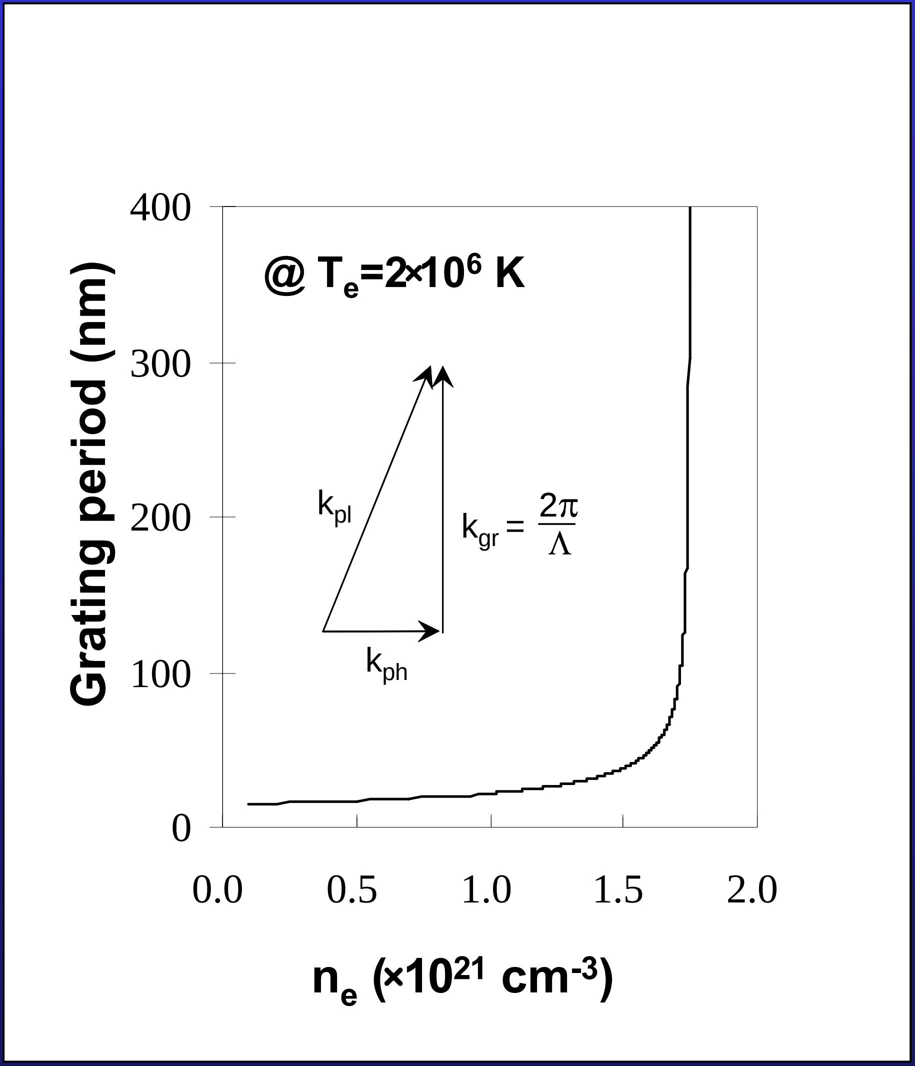
$$\omega_{pl}{}^2 = \omega_p{}^2 + \frac{3k_B T_e}{m_e} k^2, \omega_p = \sqrt{\frac{N_e e^2}{\varepsilon_0 m_e}}$$



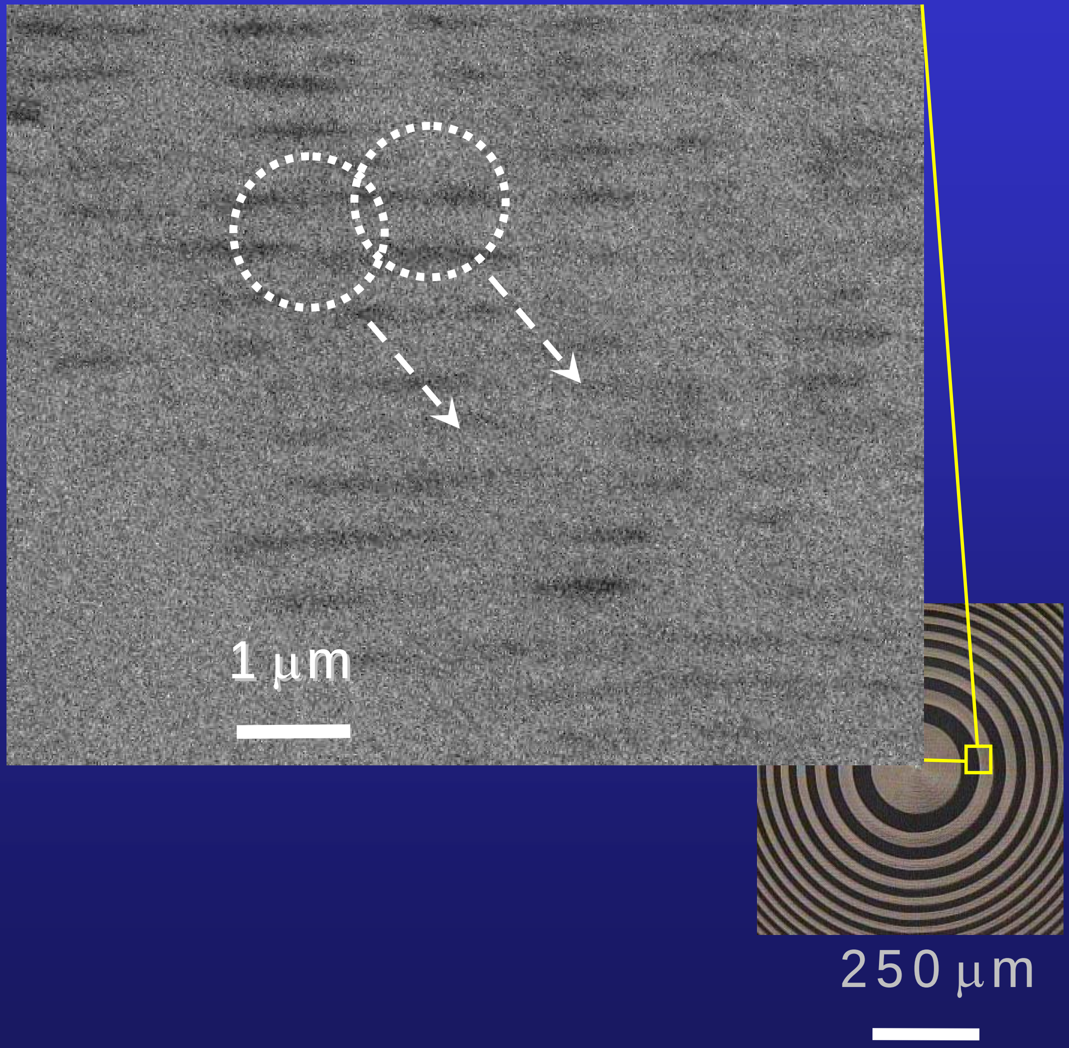
$$\omega_{pl} = \omega$$

Theoretical dependence of grating period on electron temperature and concentration

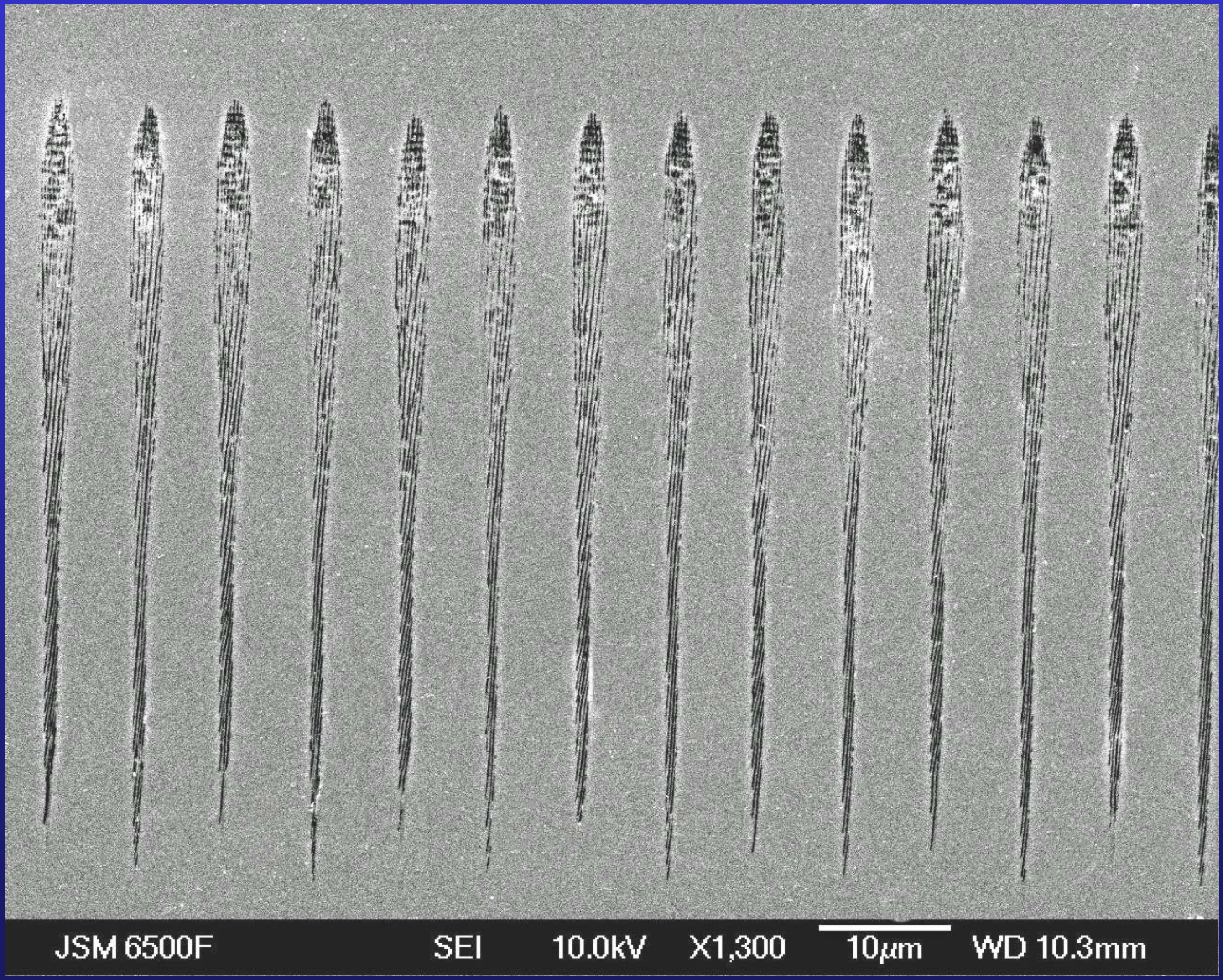




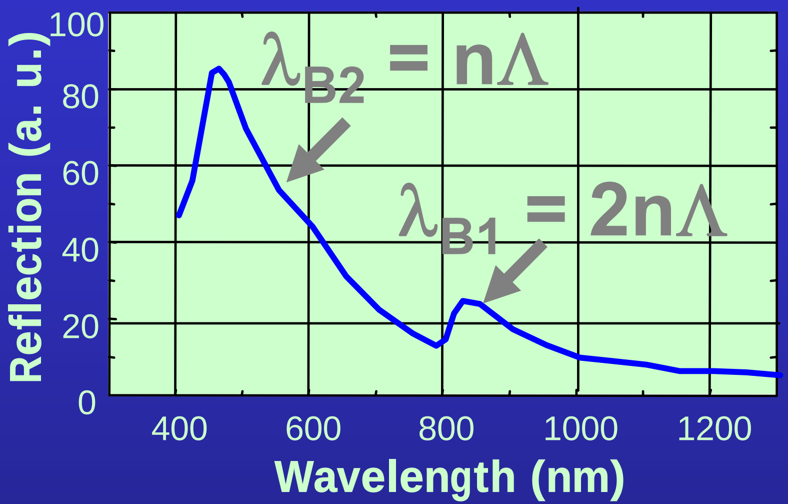
Evidence of self-organization

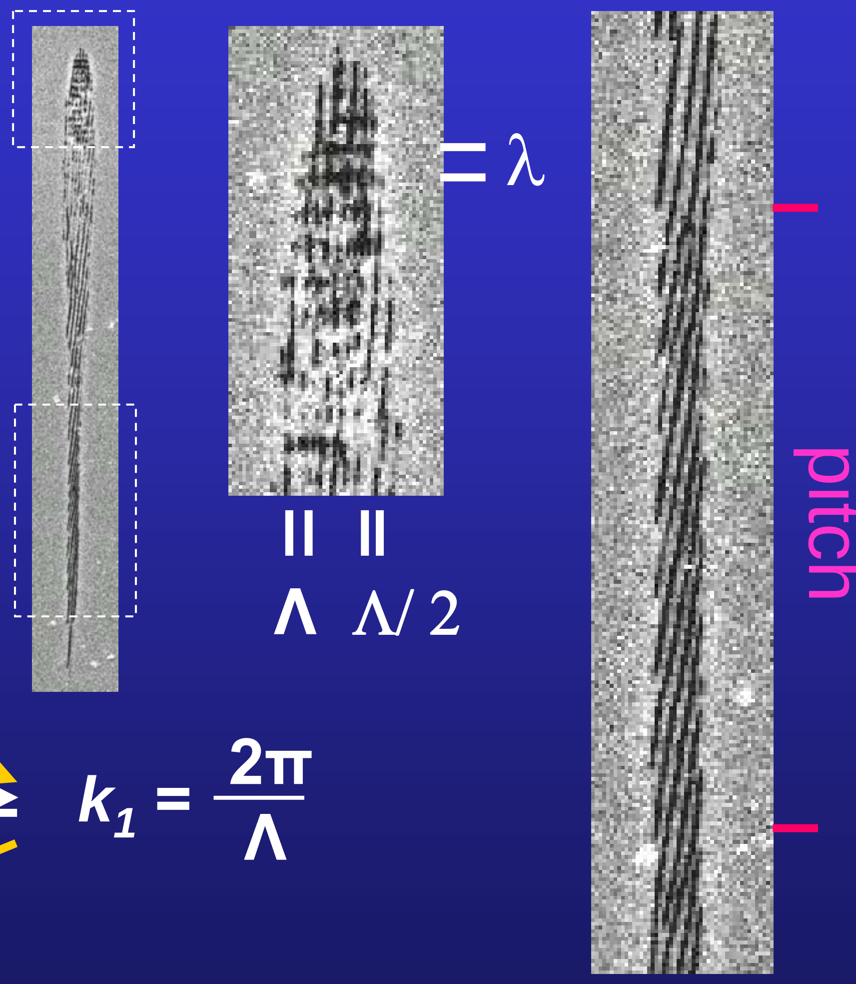


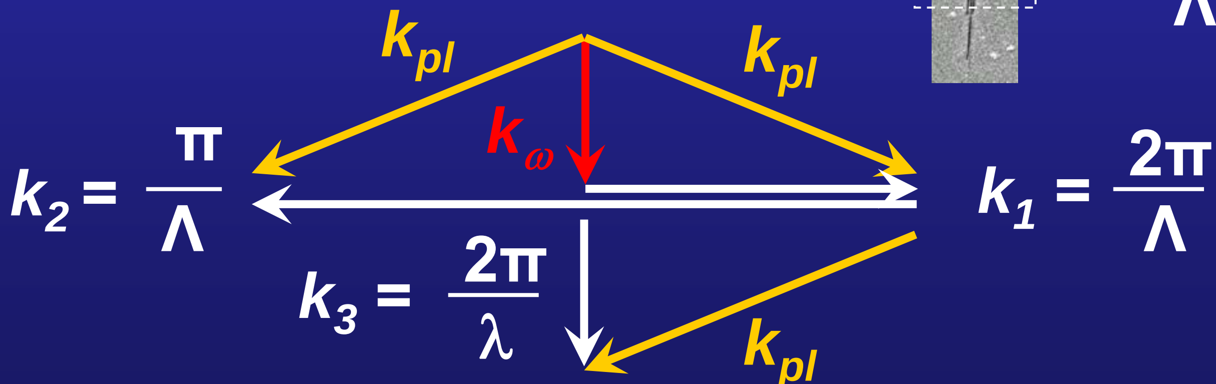
SEM images of tracks written in silica



Gratings and …"nano-tornado" in silica

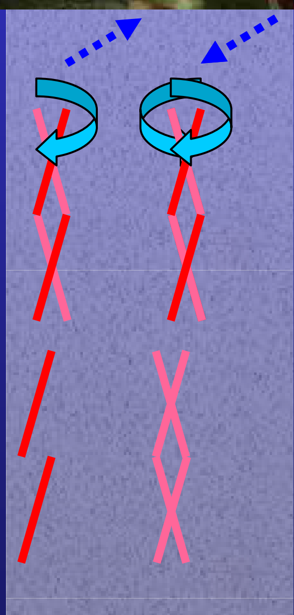


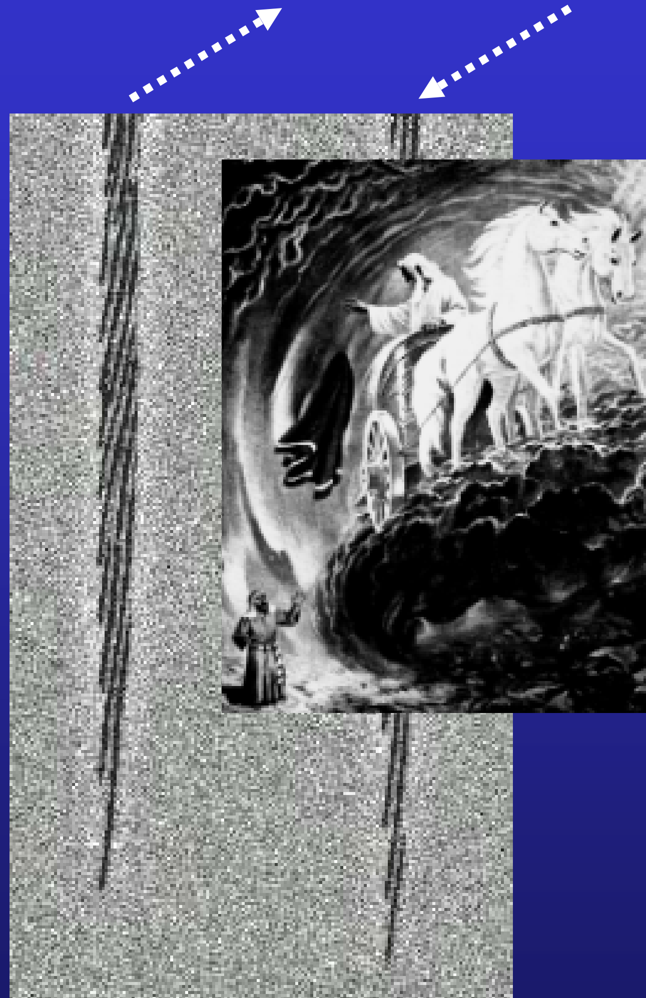




Λ= 330 nm, Λ/2 =165 nm, $\lambda = \lambda_0/n$ = 600 nm , pitch = 10 μm

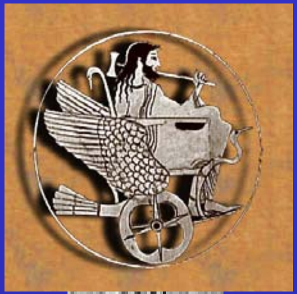
Photonic "nano-tornado" and …"chariot of fire"

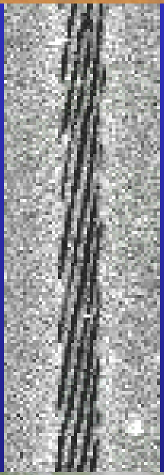




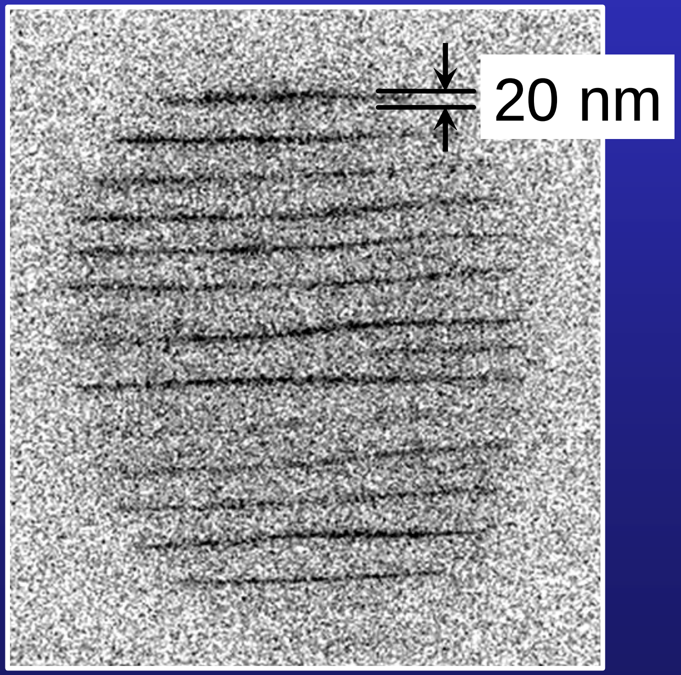
"...And it came to pass, as they still went on, and talked, that, behold, there appeared a **chariot of fire**, and horses of fire, and parted them both asunder; and Elijah went up by a **whirlwind** into heaven." II Kings 2:11

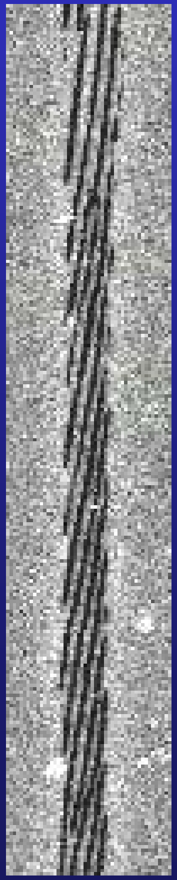
Vortices: from macro to nano-scale

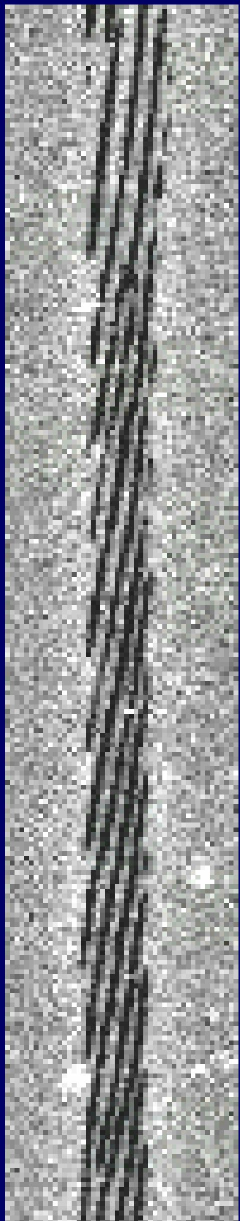


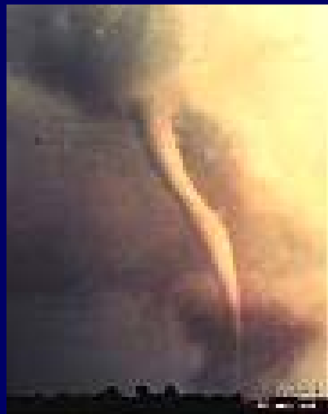


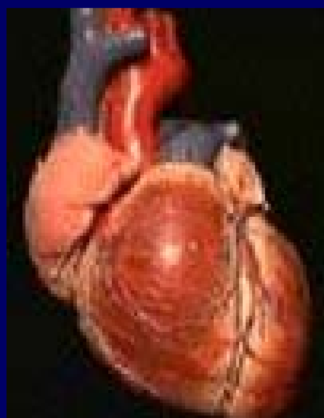
Light “fingerprint” and “nano-tornado”: The smallest embedded structures ever created by light











Spirals, Spirals, everywhere....

There are many spiral forms in nature, both on Earth and in space. Spirals occur in physical forms such as DNA and the shell formation of mollusks such as the conch. They also occur in wind patterns, including hurricanes and tornadoes. They are present in air and flame forms known as vortexes and whorls. And they occur in the way things fall in the atmosphere, from leaves to aircraft. In the human body, the spiral pattern of the heart's bioelectric impulses causes the chambers to beat with a spiral pulsing rhythm. Brain waves, comprised of neuron impulses, seem to flow along the neurons and down the spinal cord in a spiral pattern. We see spiral forms omnipresent throughout the visible and invisible universe, in galaxies, accretion disks around black holes, coalescing interstellar clouds and many other forms of matter and energy. Finally, spiral patterns in glass irradiated by ultrashort light pulses are some of the smallest ever created in nature.

Conclusions

- The smallest *embedded* structures ever created by light are observed in the experiments on femtosecond direct writing
- The phenomenon is interpreted in terms of interference between light and electron plasma wave, resulting in periodic structural changes in glass