Progress on METAMATERIALS development for EO bunch time profile monitor *for* CLIC

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CHALLENGE

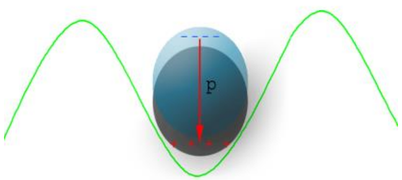
Precision measurement of femtosecond relativistic electron bunches.

- **Currently**: Uses electro-optic (EO) effect in crystal that samples Coulomb field of 1 TeV electron beam.
- **How**: “EO transposition scheme” spectrally shifts THz Coulomb field to optical frequency.
- **Limitation**: Now limited by materials properties – inadequate optical bandwidth.

We are attempting to replace inorganic crystals such as GaP and ZnTe by a “*Metamaterial*” based on *Metal-Glass Nanocomposites (MGNs)*.

Metal-Glass Nano-composites (MGNs)

Glass with metallic nanoparticles



Surface Plasmon Resonance (SPR)

Mie theory for scattering & absorption of light by spheres

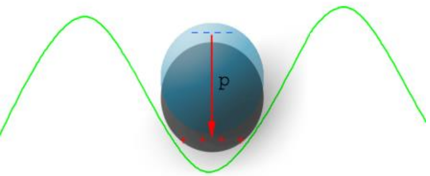
The polarizability α and induced dipole moment p of a metal sphere embedded in dielectric are given by:

$$\alpha = 4\pi R^3 \frac{\varepsilon_i(\omega) - \varepsilon_h}{\varepsilon_i(\omega) + 2\varepsilon_h}$$

$$\vec{p}(\omega) = \alpha\varepsilon_0 \vec{E}_0(\omega) = 4\pi\varepsilon_0 R^3 \frac{\varepsilon_i(\omega) - \varepsilon_h}{\varepsilon_i(\omega) + 2\varepsilon_h} \vec{E}_0(\omega)$$

- R Radius of the nanoparticle
- $\varepsilon_i(\omega)$ Complex electric permittivity of the metal
- ε_h Complex electric permittivity of the host matrix
- E_0 Electric field strength of the incident electromagnetic wave
- ε_0 Electric permittivity of vacuum

- Michael Faraday, Phil. Trans. Royal Society 147, 145 (1857).
- Gustav Mie, Ann. Phys. (Leipzig) 25, 377 (1908).
- Kreibig, U. & Volmer, M. Optical Properties of Metal Clusters, Springer (1995).



The absorption cross-section of a spherical metal inclusion placed in a transparent dielectric matrix (Im[ε_h]→0)

$$\sigma(\omega) = 12\pi R^3 \frac{\omega}{c}\varepsilon_h^{3/2} \frac{\varepsilon_i''(\omega)}{\left[\varepsilon_i'(\omega) + 2\varepsilon_h\right]^2 + \varepsilon_i''(\omega)^2}$$

Real & **Imaginary** parts of the electric permittivity of the metal

$\varepsilon_i(\omega)$ can be described by the ***Drude-Sommerfeld*** relationship

$$\varepsilon_i(\omega) = \varepsilon_b + 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}$$

- ε_b Complex electric permittivity associated with inter-band transitions of the core electrons in the atom
- γ Damping constant of the electron oscillations
- ω_p Free electron plasma frequency
- N Density of the free electrons
- m Effective mass of electron

$$\omega_p = \sqrt{\frac{Ne^2}{m\varepsilon_0}}$$

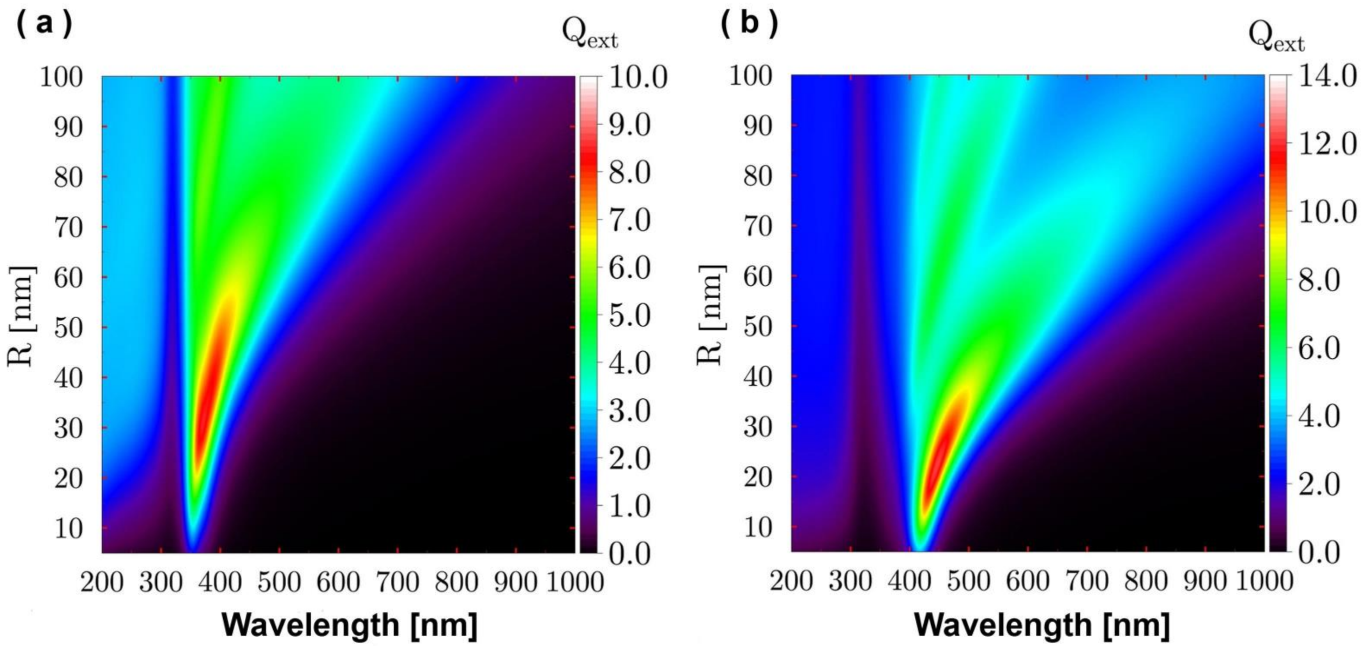
The **Mie resonance** occurs at the SP frequency $\boldsymbol{\omega_{SP}}$ when:

$$\left[\varepsilon_i'(\omega) + 2\varepsilon_h\right]^2 + \varepsilon_i''(\omega)^2 \rightarrow Minimum$$

$$\varepsilon_i'(\omega_{SP}) = -2\varepsilon_h$$

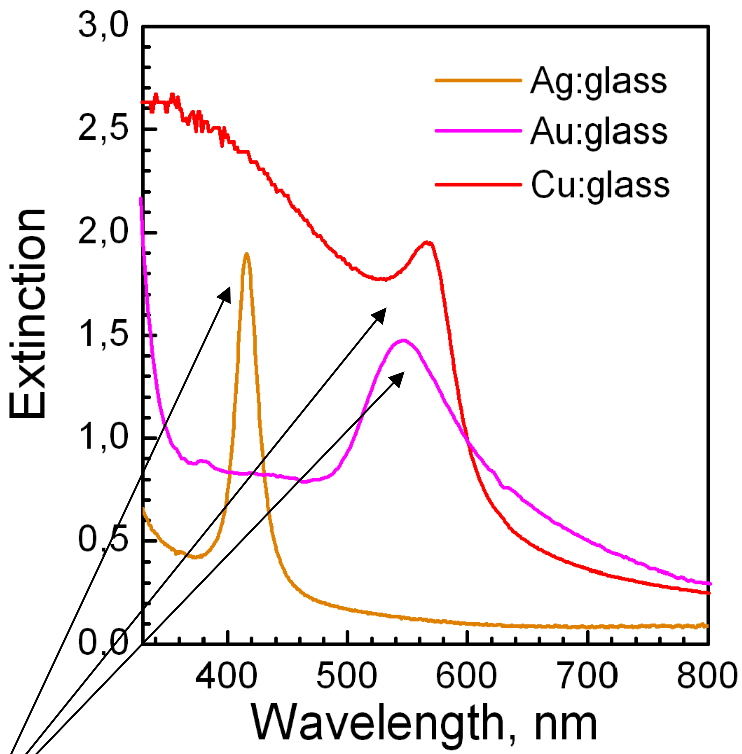
Bohren, C. F.; Huffman, D. R. Absorption & Scattering by Small Particles; Wiley (1983).

Spectral positions of SPRs for silver nanoparticles in a) vacuum (ε_h=1) and b) glass (ε_h=2.25) as a function of particle size.

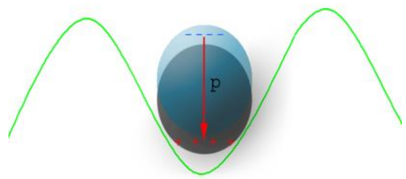


- SPR maxima are red shifted in (b) as compared to (a);
- SPR position depends on the particle size;
- SPR position remains quasi-constant for R<10-15nm, while the bands HW for these clusters differs by a factor of 4 (*intrinsic size effect*);
- For R≤10nm (below the mean free path of electrons in metals), the electron scattering mainly increases the imaginary part of the dielectric function;
- For R>1 nm, the *spill-out* of electrons from the particle surface results in an inhomogeneous dielectric function, leading to very broad SP bands;
- SPR shifts to longer wavelengths with a simultaneous increase in the band HW for R>15nm [*extrinsic size effect*], leading to higher order oscillations of conduction electrons.

Absorption (Extinction) spectra of glass containing **spherical** Silver, Gold & Copper nanoparticles.



SPR peaks

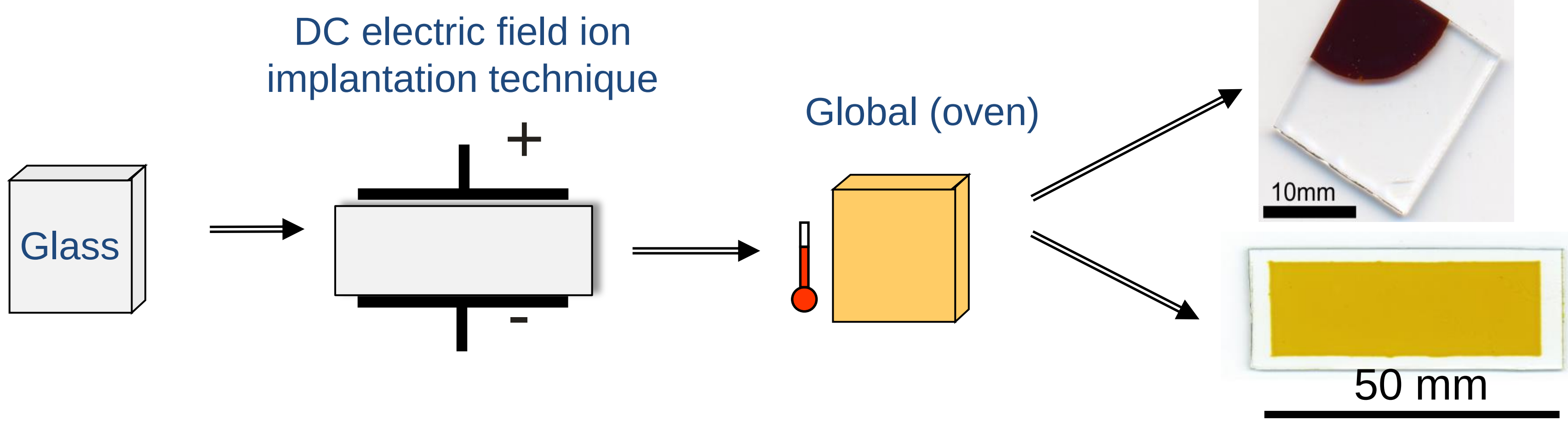


Surface Plasmon Resonance (SPR)

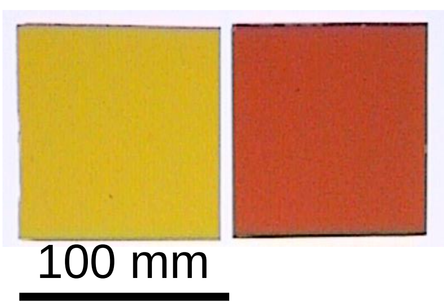
Core electrons define the position of the SPR in the extinction spectra for different noble metals.

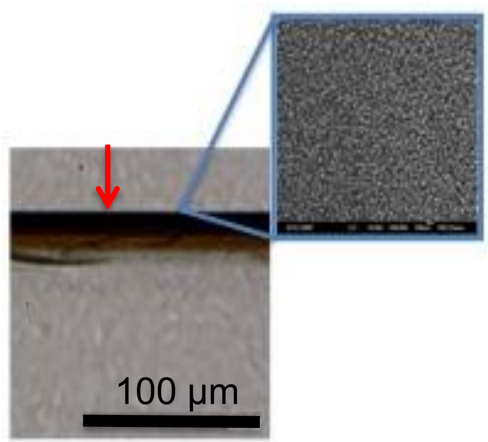
• Kreibig, U & Volmer, M. Optical Properties of Metal Clusters. Springer (1995).
• Bohren, C. F.; Huffman, D. R. Absorption & Scattering by Small Particles. Wiley (1983).

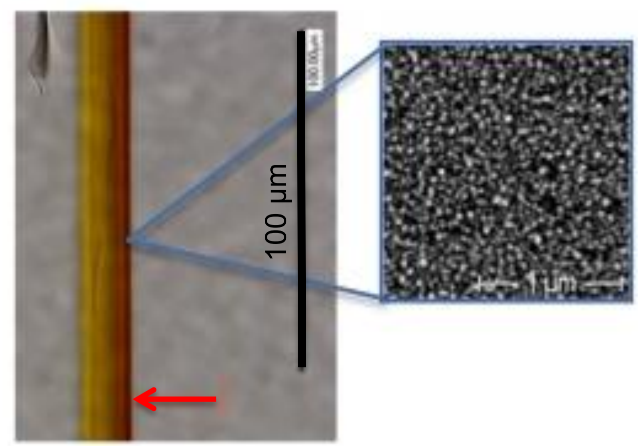
DC electric field-assisted ion exchange & reduction in air



- Control over the size of the embedded nanoparticles (up to 50nm in diameter);
- Control over the thickness of the nanoparticle-containing layer in glass (500nm – 250µm);
- Control over the spatial distribution (filling factor) of the nanoparticle-containing layer.







MAPS

- Optical Materials Express 1, 1224 (2011).
- Optics Express 20, 23227 (2012).
- Patent.

Effective dielectric constant of a composite material with spherical metal inclusions having a filling factor *f*

$$\varepsilon_{eff}(\omega)=\varepsilon_h\frac{(\varepsilon_i(\omega)+2\varepsilon_h)+2f(\varepsilon_i(\omega)-\varepsilon_h)}{(\varepsilon_i(\omega)+2\varepsilon_h)-f(\varepsilon_i(\omega)-\varepsilon_h)}$$

Complex index of refraction of a composite medium: $n(\omega)=n'+in''=\sqrt{\varepsilon_{eff}(\omega)}$

Absorption coefficient of a composite medium: $\alpha=\frac{2\omega}{c}\mathrm{Im}\sqrt{\varepsilon_{eff}(\omega)}$

Refractive index of a composite medium: $n'(\omega)=\mathrm{Re}\sqrt{\varepsilon_{eff}(\omega)}$

Reflectivity of a composite medium for normal incidence: $R(\omega)=\left|\frac{n(\omega)-1}{n(\omega)+1}\right|^2$

$\varepsilon_i(\omega)$ *Complex electric permittivity of the metal*
ε_h *Complex electric permittivity of the host matrix*
c *Speed of light in vacuum*
f *Volume of the silver inclusions per unit volume of the composite material* $f = V_{Ag} / V_{total}$

Absorption cross-section, Dispersion & Reflectivity Spectra of glass containing spherical silver nanoparticles as a function of the volume filling factor of the metal clusters

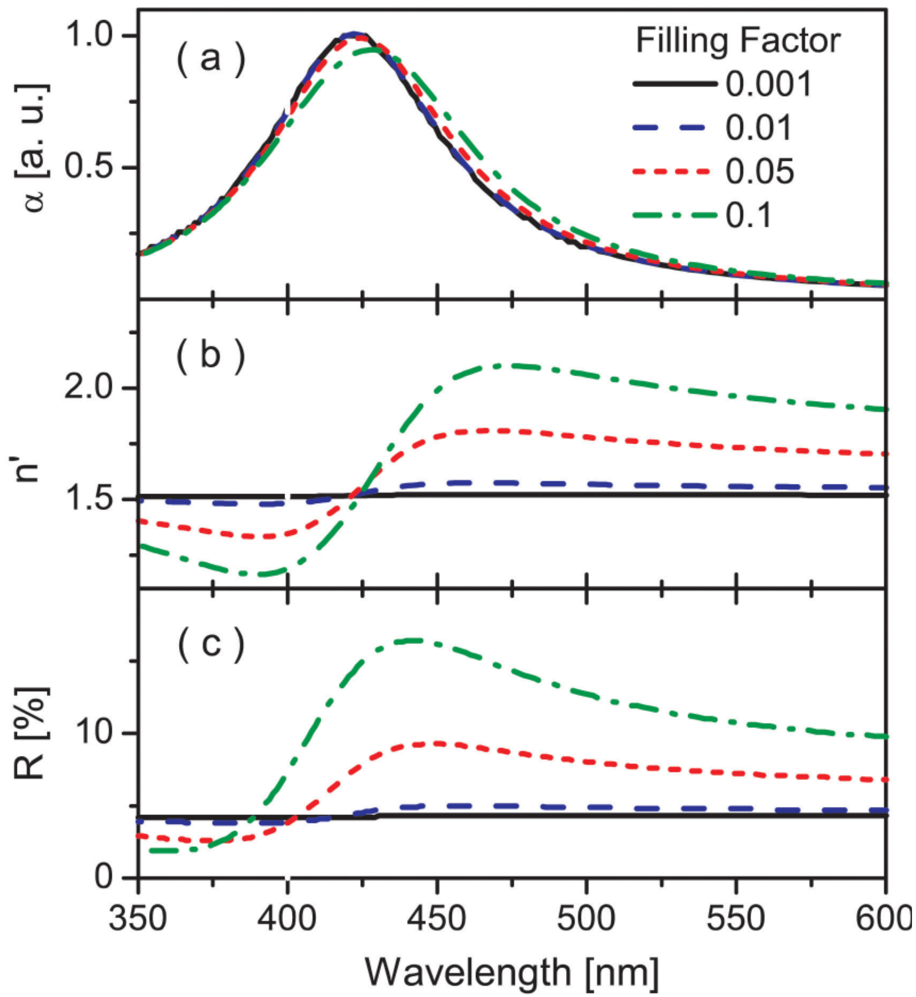
$$n(\omega) = n' + i n'' = \sqrt{\varepsilon_{eff}(\omega)}$$

$$\alpha = \frac{2\omega}{c} \mathrm{Im} \sqrt{\varepsilon_{eff}(\omega)}$$

$$n'(\omega) = \mathrm{Re} \sqrt{\varepsilon_{eff}(\omega)}$$

$$R(\omega) = \left| \frac{n(\omega) - 1}{n(\omega) + 1} \right|^2$$

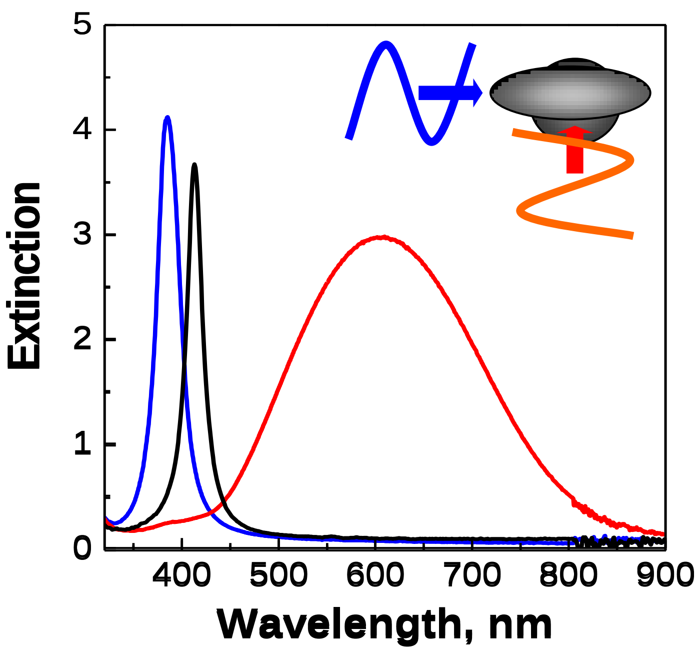
$\varepsilon_h = 2.3$
$\omega_p = 9.2$ eV
$\gamma = 0.5$ eV
$\varepsilon_b = 4.2$



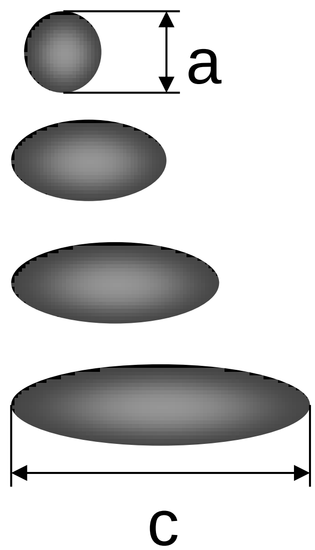
- Opt. Materials Express 4, 969 (2014).
- Appl. Opt. 48, F38 (2009).
- Appl. Phys. A 80, 1647 (2005).
- Opt. Exp. 13, 1266 (2005).

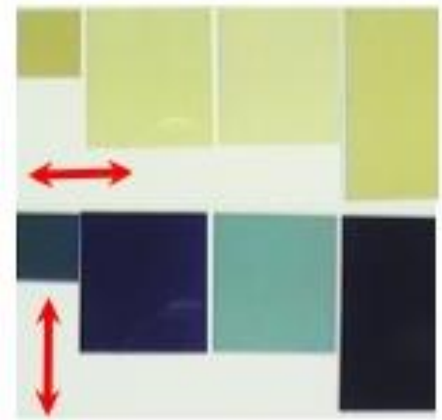
Non-spherical nanoparticles

SPR position is size & **shape dependent**. Therefore metal nanoparticles with non-spherical shape should demonstrate several SPR in their spectra.



Polarised extinction spectra of spherical & spheroidal silver particles in glass

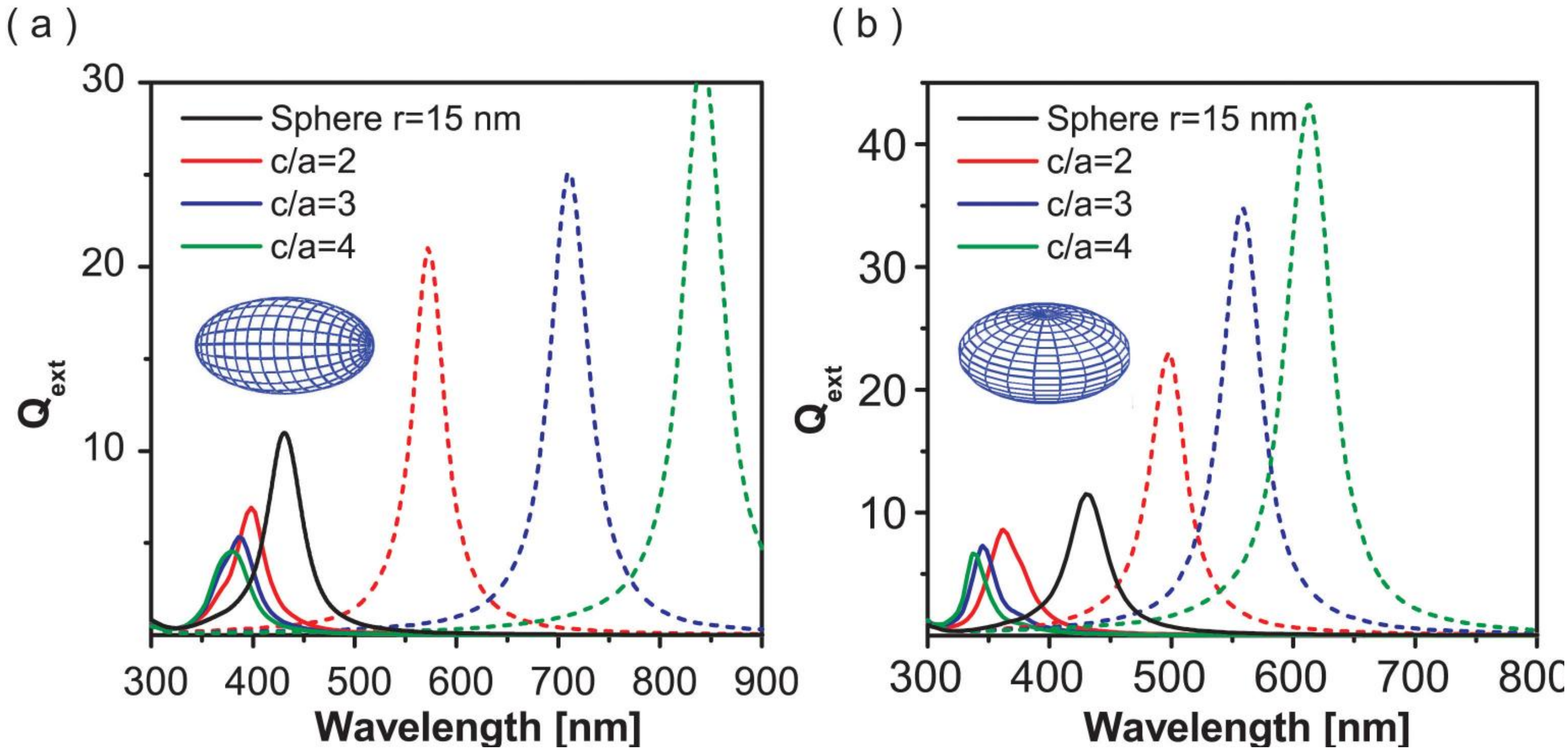




Non-spherical nanoparticles

Mie theory for silver spheroids embedded in glass - with different aspect ratios

Polarization extinction spectra for (a) **prolate** and (b) **oblate** silver spheroids

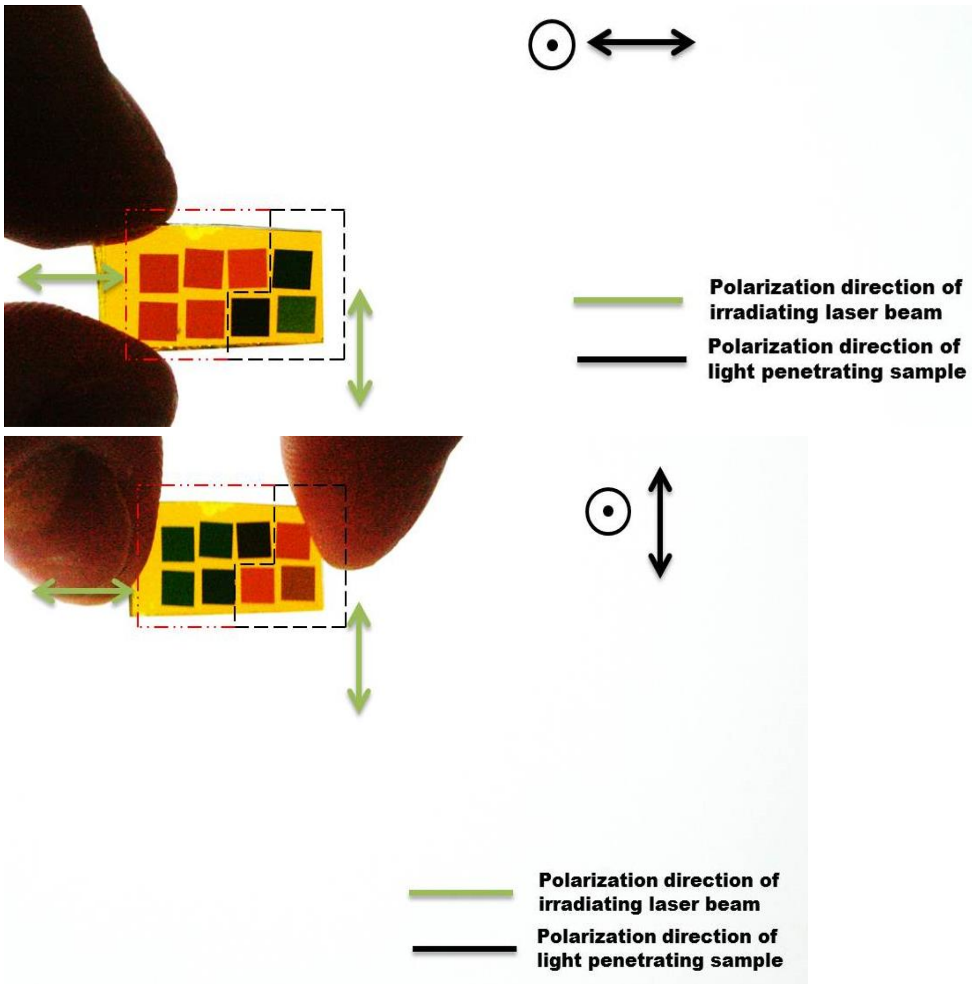


- The volume of the spheroids is equal to the volume of a nanosphere with radius of 15 nm;
- **Dashed curves:** polarization of the light is **parallel** to the **long axis**;
- **Solid curves:** polarization of the light is **parallel** to the **short axis**.

Astrophys. Space Sci. **204**, 19 (1993).

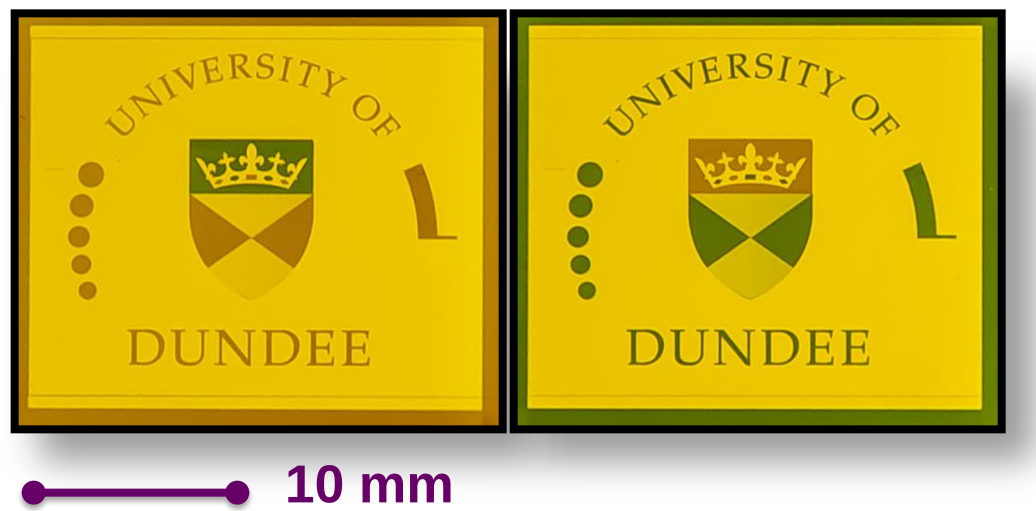
Non-spherical nanoparticles

Picosecond laser irradiation of glass with embedded silver nanoparticles



Case in point

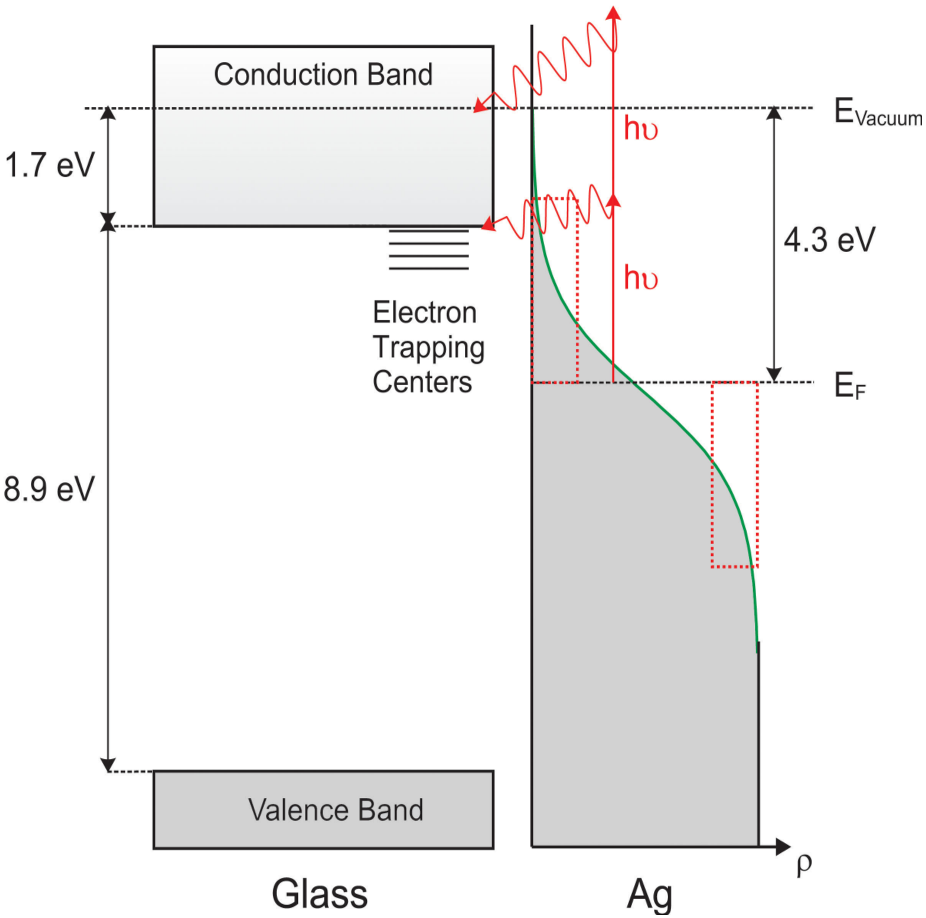
Wavelength: 532 nm
Pulse length: ~ 6 ps at 200 kHz
Laser fluence: ~ 0.3 J/cm^2
Focal spot diameter: 15 µm

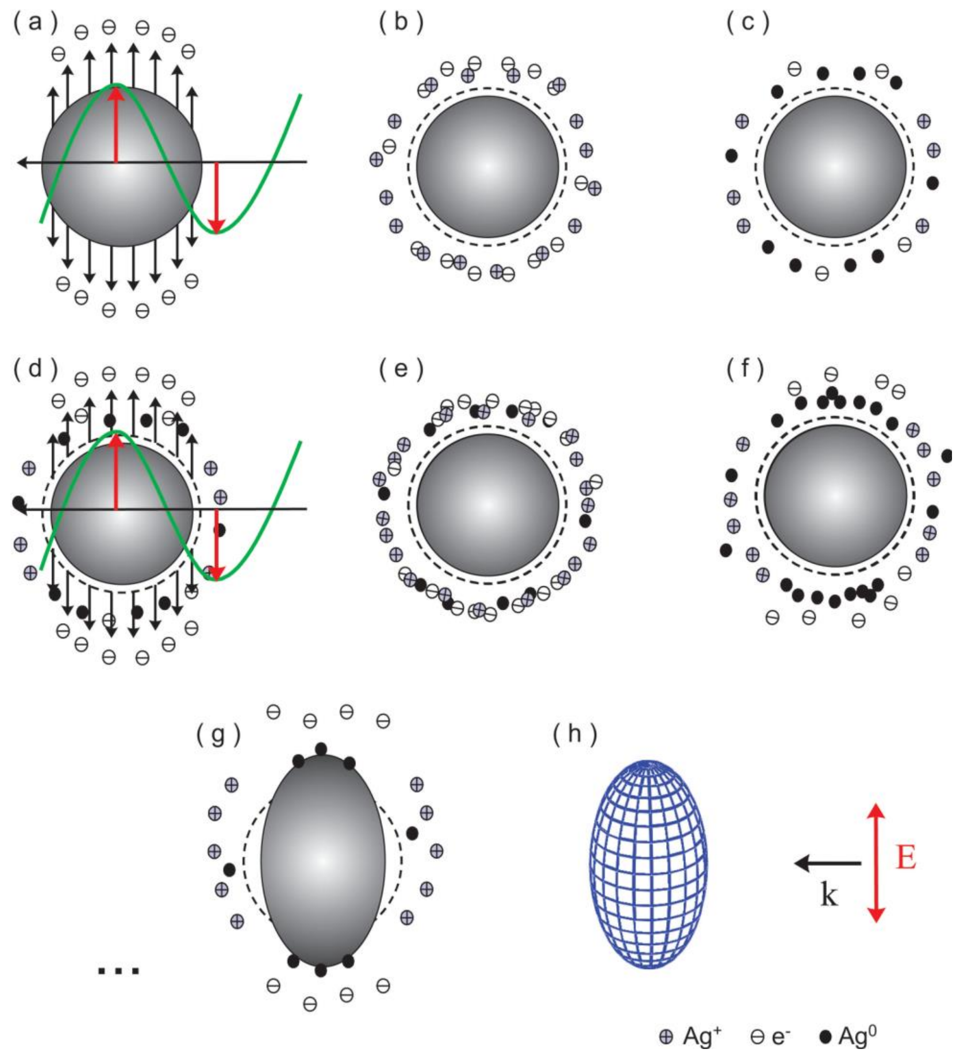


MAPS

- Optics Express 21, 21823 (2013).

Energy level scheme of the electrons in a composite glass containing silver inclusions. The red dotted line indicates a non-thermal distribution of the electrons in the Ag nanoparticle caused by excitation of SPR. Green line –distribution of the electrons after thermalisation.

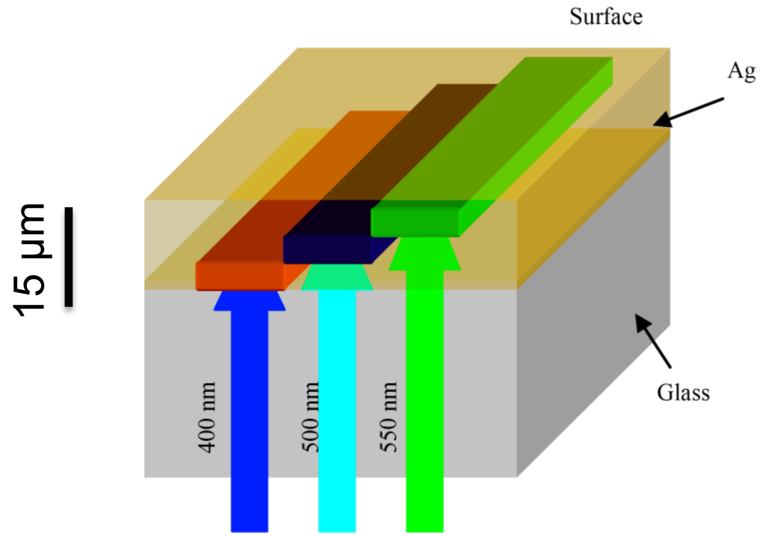




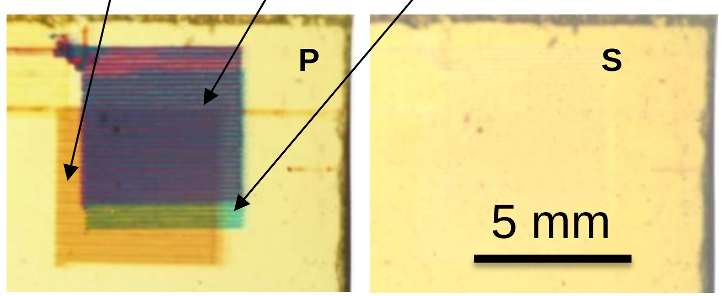
A.Stalmashonak, G.Seifert and A. Abdolvand

Ultra-Short Pulsed Laser Engineered Metal-Glass Nanocomposites in **SpringerBriefs in Physics** (June 2013).

MULTI-LAYER laser irradiation AND CONTROLED MODIFICATION



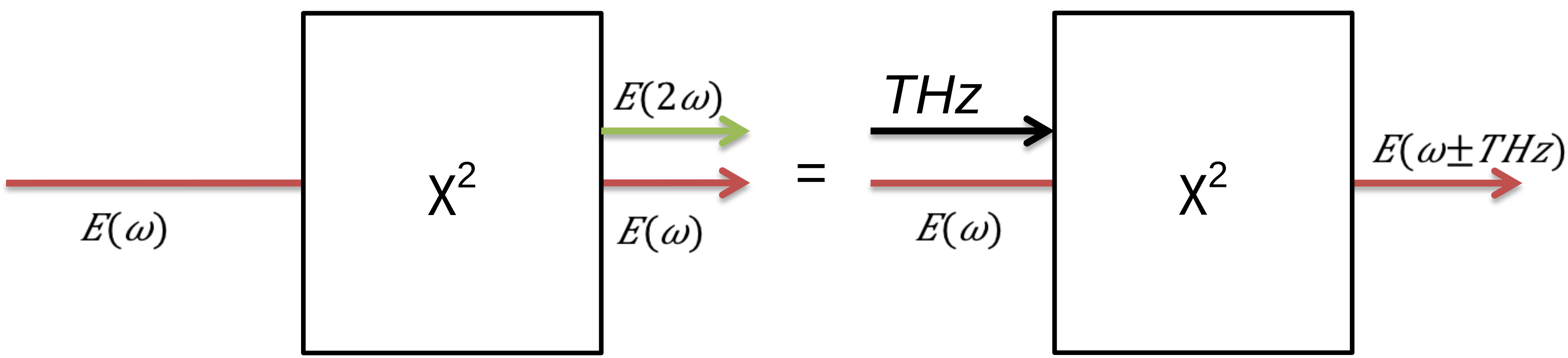
The exposed areas in polarized light
@ 400 nm, 500 nm, 550 nm



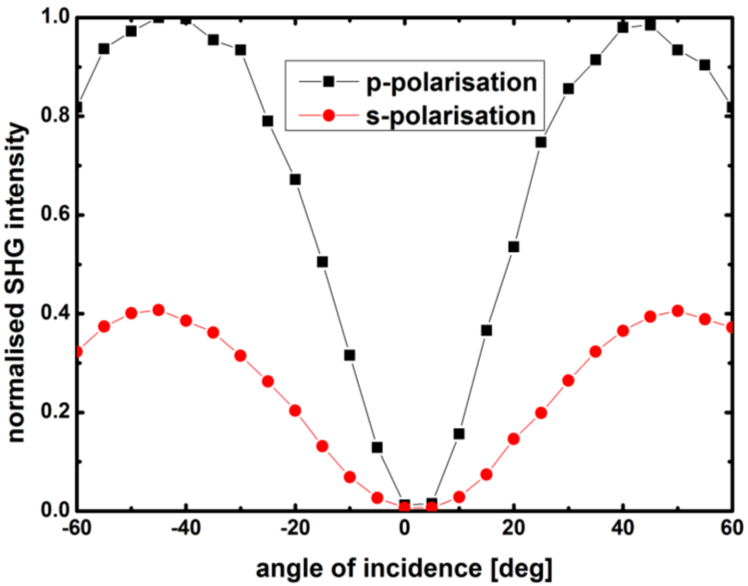
MAPS: Appl. Phys. Lett. 99, 201904 (2011).

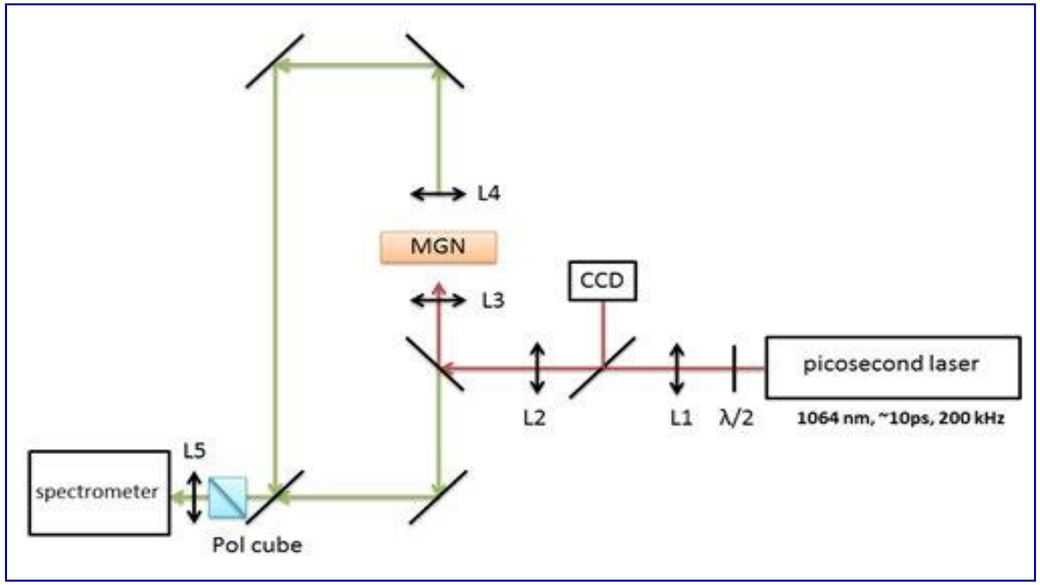
Second Harmonic Generation (SHG) observation

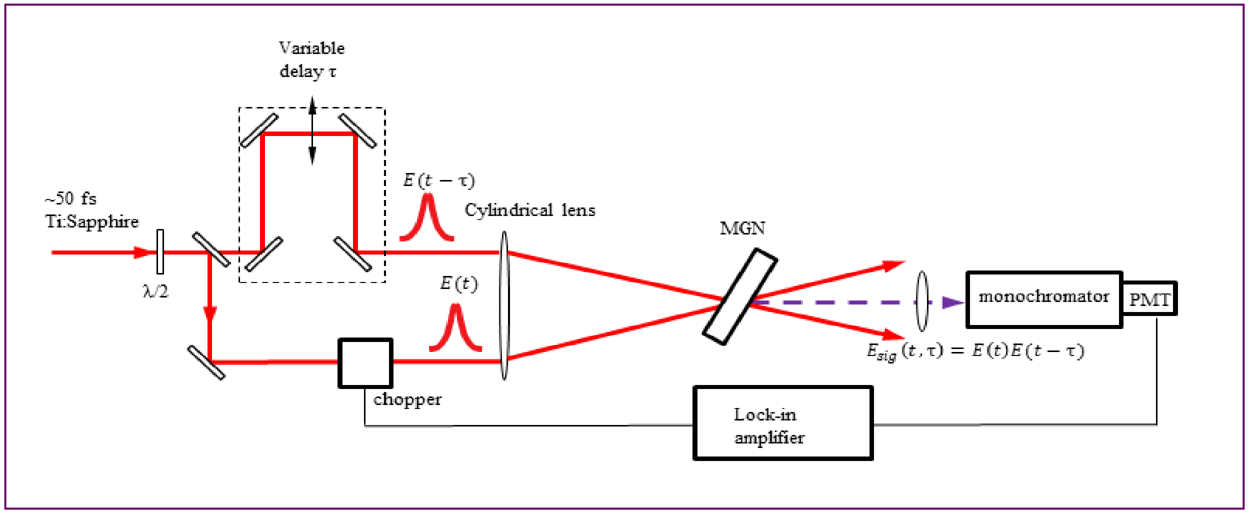
Due to the identical matrix elements in the susceptibility and EO tensors ε_{xx} and r_{52} , a good test of a useful EO effect is to measure SHG from these samples…..



Second Harmonic Generation (SHG) from laser-reshaped silver nanoparticles embedded in glass







Final stage

Further tests at **Daresbury Laboratory** with a THz antenna will verify the usefulness of our metamaterial (MGNs) for overcoming the challenge with the current EO materials (the bandwidth limitations).

THANK YOU FOR YOUR ATTENTION!