



MATERIAL STUDIES

into

EO CRYSTALS

Amin Abdolvand

a.abdolvand@dundee.ac.uk

School of Engineering, Physics & Mathematics
University of Dundee
United Kingdom

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Materials & Photonics Systems (MAPS) Group

- ① **Fabrication & processing of nanomaterials;**
- ② **Applied Laser Technology;**
- ③ **Complex photonics.**



IMPACT & POTENTIAL APPLICATIONS:

- Storage of information;
- Sensing, Circuitry & Security;
- Energy sector;
- Particle accelerators;
- Healthcare;
- Creative industries;
- Fundamental optical studies;
- Beam shaping; Laser technology.

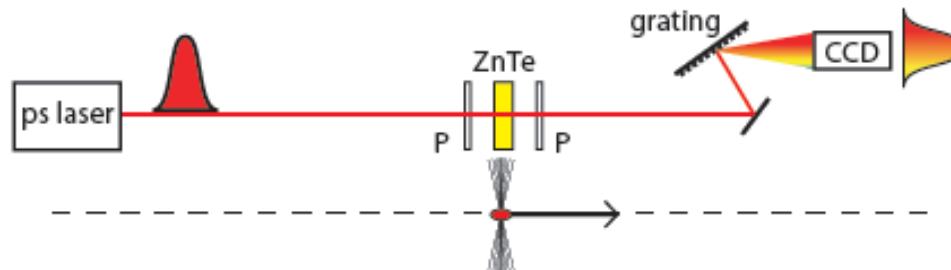


Stating the problem ...

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.



New materials

We are attempting to replace inorganic crystals such as GaP and ZnTe by a

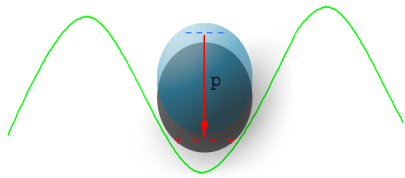
“*Metamaterial*” based on *Metal-Glass Nanocomposites (MGNs)*.

This talk is about the

fabrication, design & functionalisation of

MGNs

& MAPS efforts to address the challenge



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 \vec{p} of a metal sphere embedded in dielectric are given by:

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

$$\vec{p}(\omega) = \alpha \epsilon_0 \vec{E}_0(\omega) = 4\pi \epsilon_0 R^3 \frac{\epsilon_i(\omega) - \epsilon_h}{\epsilon_i(\omega) + 2\epsilon_h} \vec{E}_0(\omega)$$

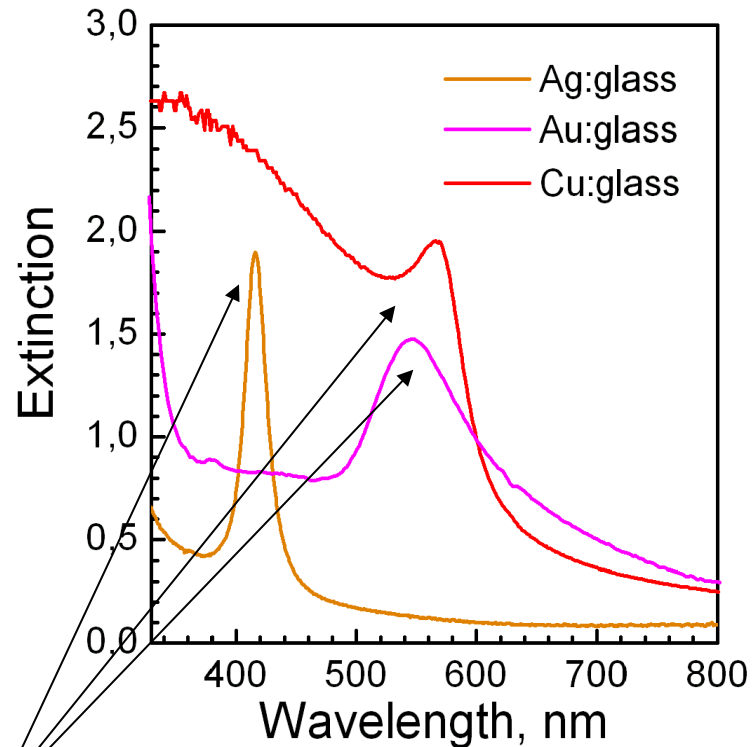


Lycurgus cup (4th century AD) Roman art in the British Museum.

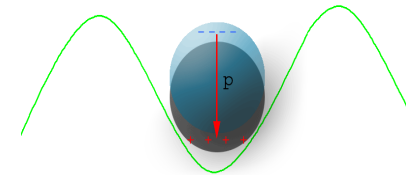
- R Radius of the nanoparticle
- $\epsilon_i(\omega)$ Complex electric permittivity of the metal
- ϵ_h Complex electric permittivity of the host matrix
- E_0 Electric field strength of the incident electromagnetic wave
- ϵ_0 Electric permittivity of vacuum

- 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).

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



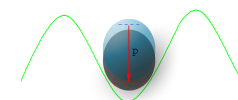
SPR peaks



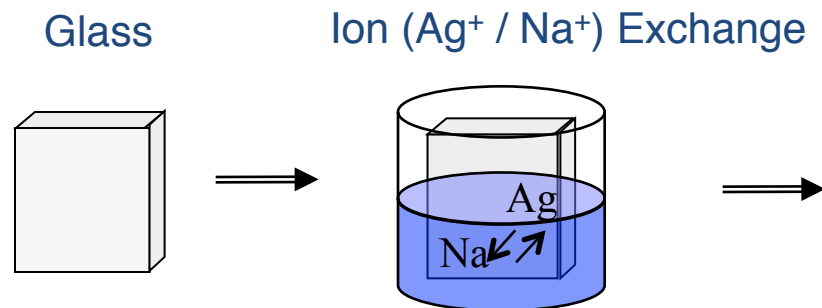
Surface Plasmon Resonance (SPR)

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).



Traditional ion-exchange technique & “modern” laser annealing in air



Case in point

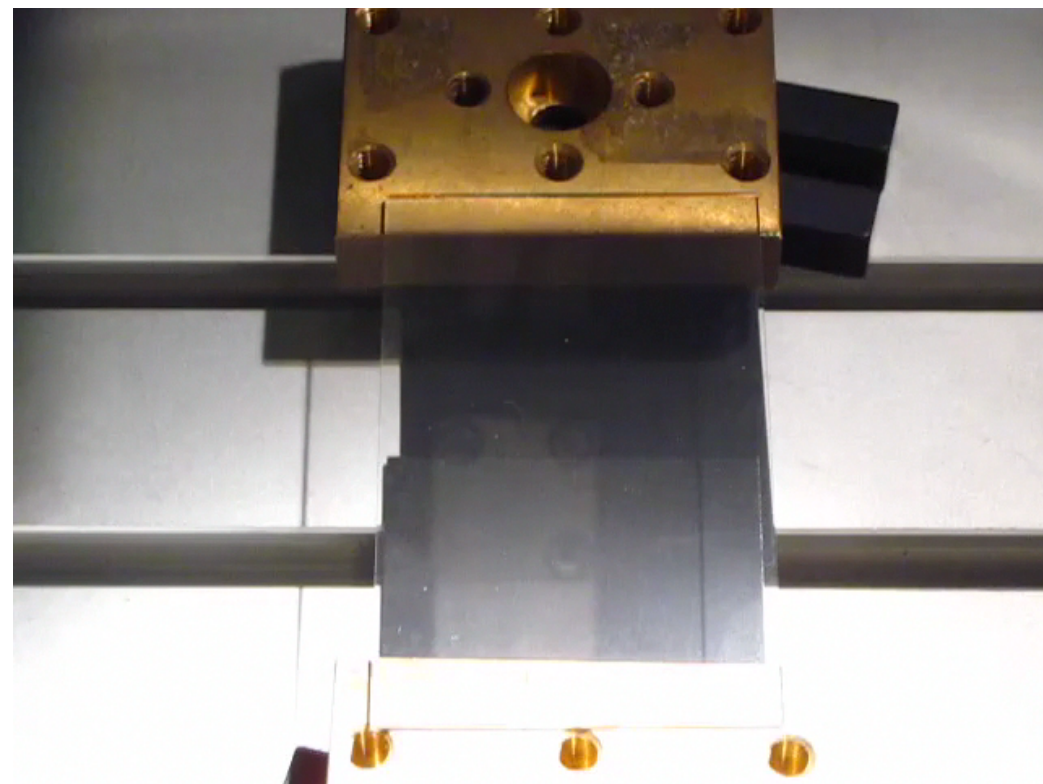
Material: window glass after ion-exchange

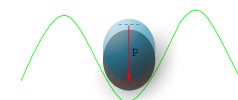
Wavelength: 355 nm

Pulse length: 10 ns at 80 kHz

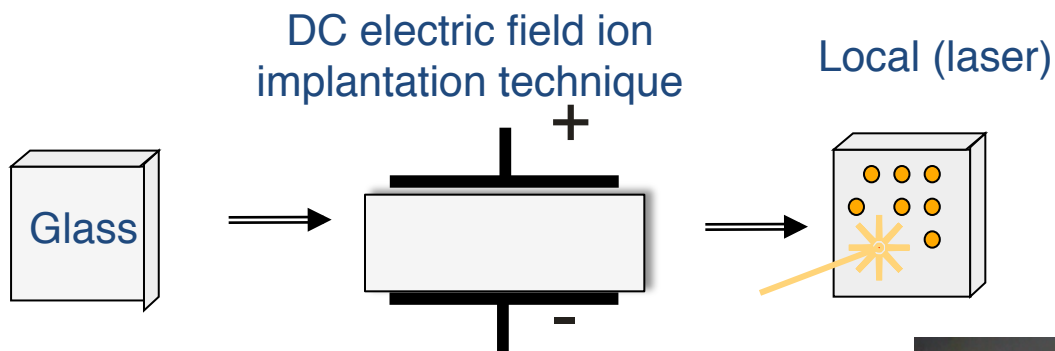
Focal spot diameter: 30 μm

Processing speed: 20 mm/s





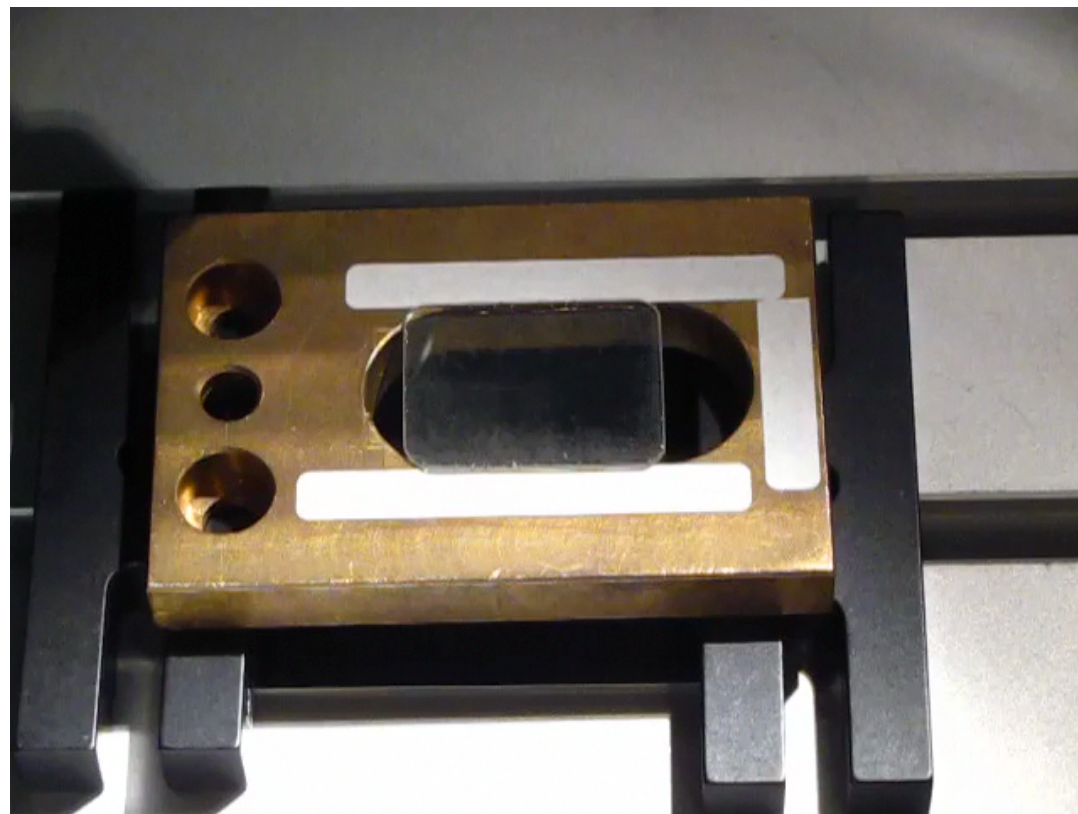
DC electric field-assisted ion exchange & laser reduction in air



Case in point
Wavelength: 355 nm
Pulse length: ~ 6 ps at 80 kHz
Focal spot diameter: 30 μm
Processing speed: 20 mm/s

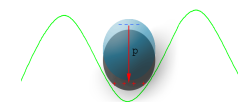


10 mm

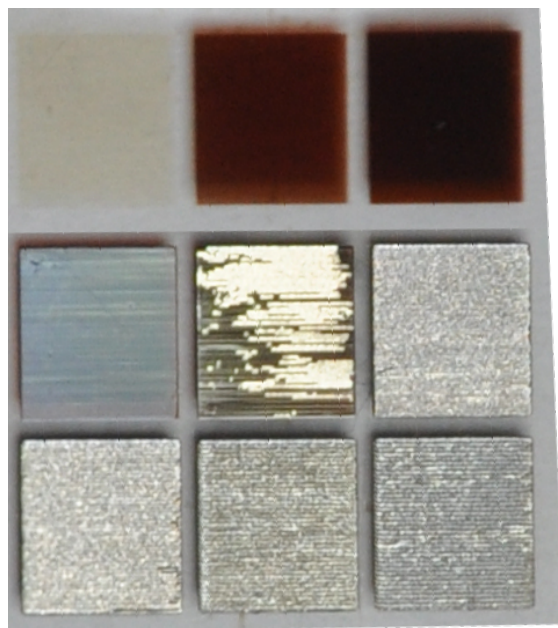


MAPS

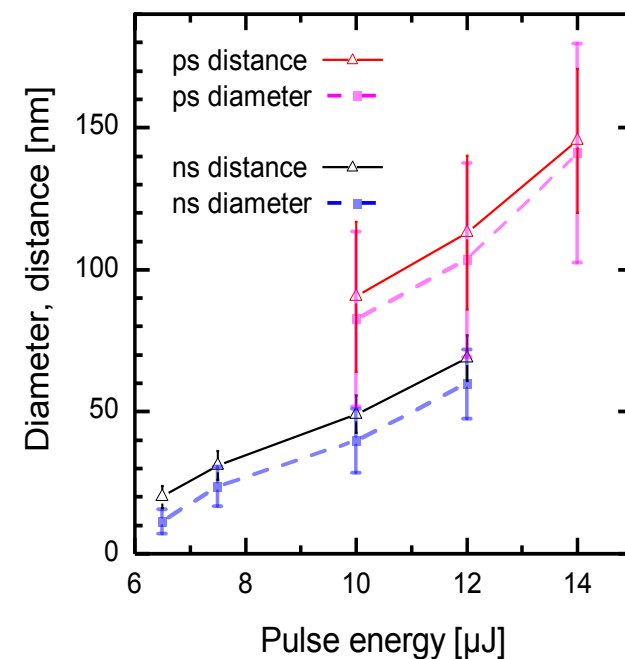
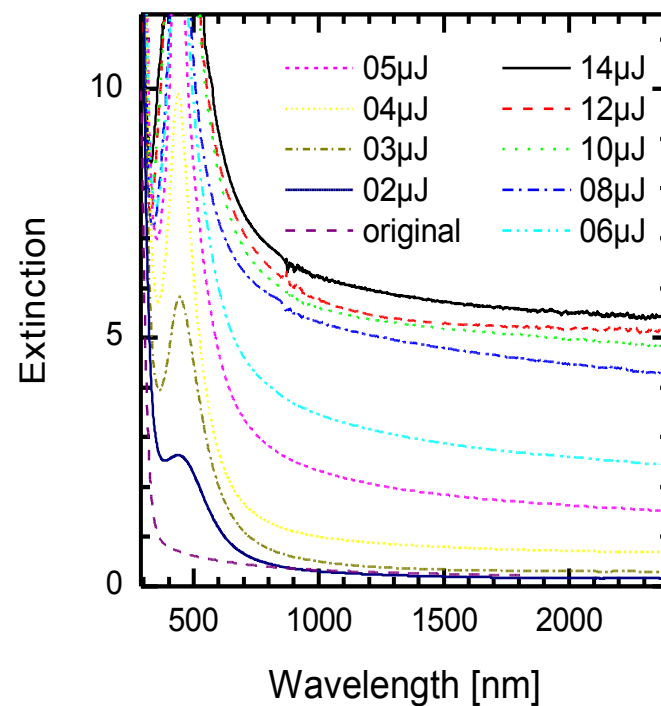
- Appl. Phys. A (Rapid Commun.) 109, 45 (2012).
- Optics Express 22, 5076 (2014). + Patent



Picosecond pulsed laser irradiation of silver-ion doped glass

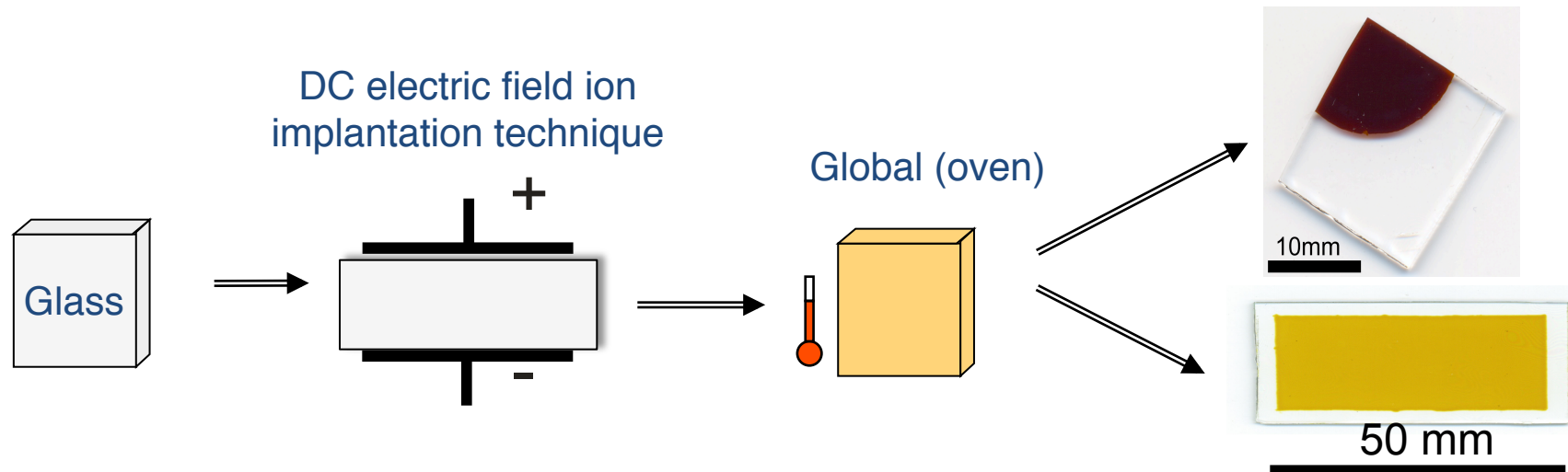
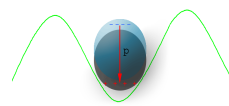


10 mm

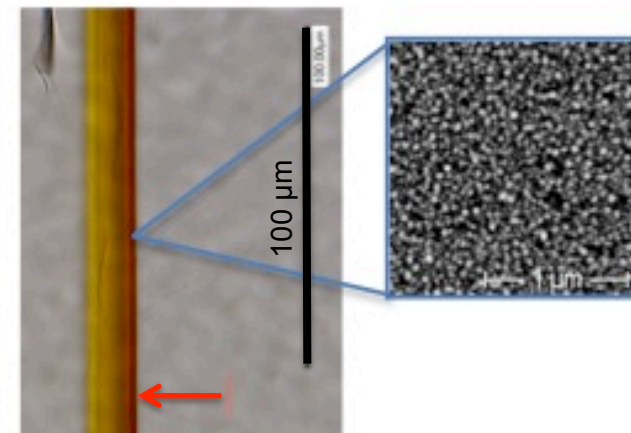
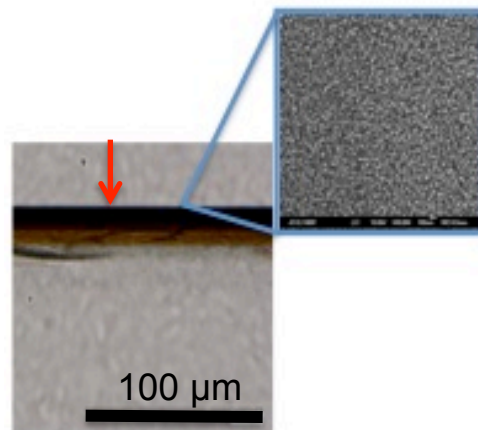
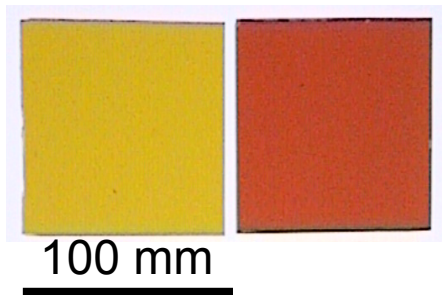


- Optics Express 22, 5076 (2014).
- Appl. Phys. A (Rapid Commun.) 109, 45 (2012).
- Patent – 2012.

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 – 250 μ m);
- 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.

Application Example

Plasmonics-assisted rapid laser joining of glass

Case in point

Wavelength: 532 nm

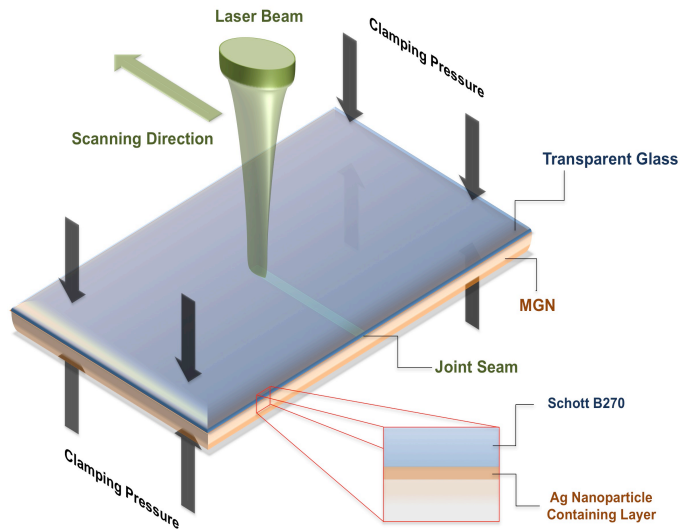
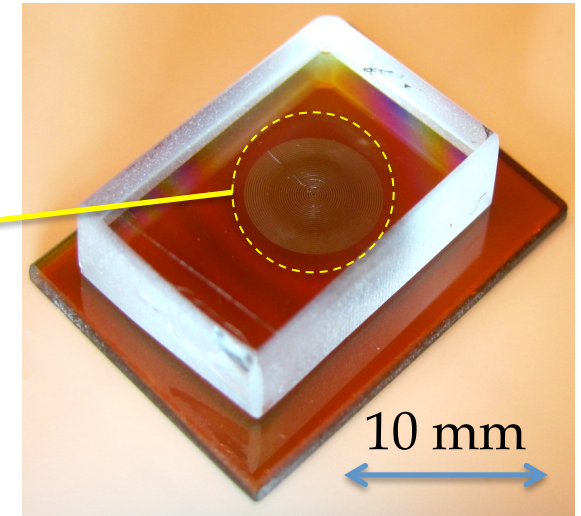
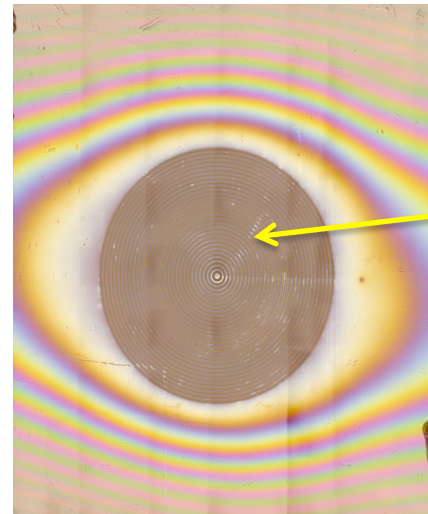
Pulse length: ~ 40 ns at 100 kHz

Laser fluence: ~ 0.2 J/cm²

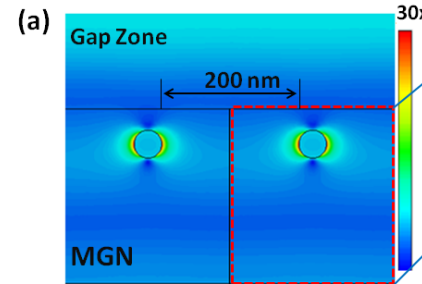
Focal spot diameter: 60 μm

Processing speed: 10 mm/s

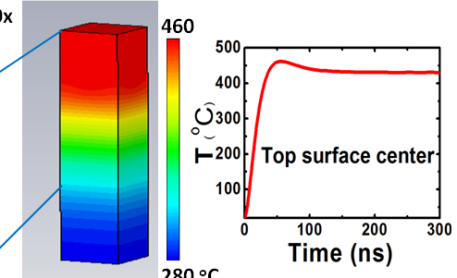
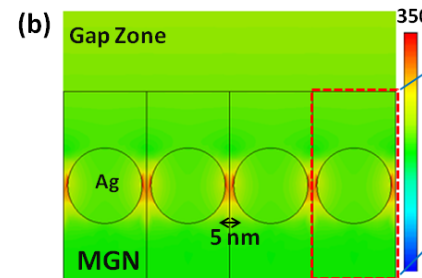
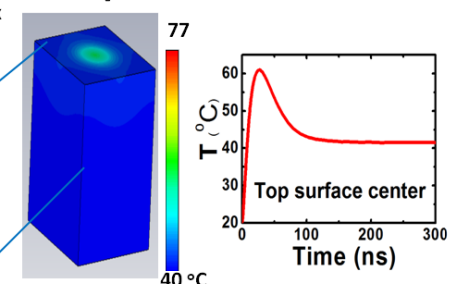
Weld strength: ~ 13 MPa



EM Simulation

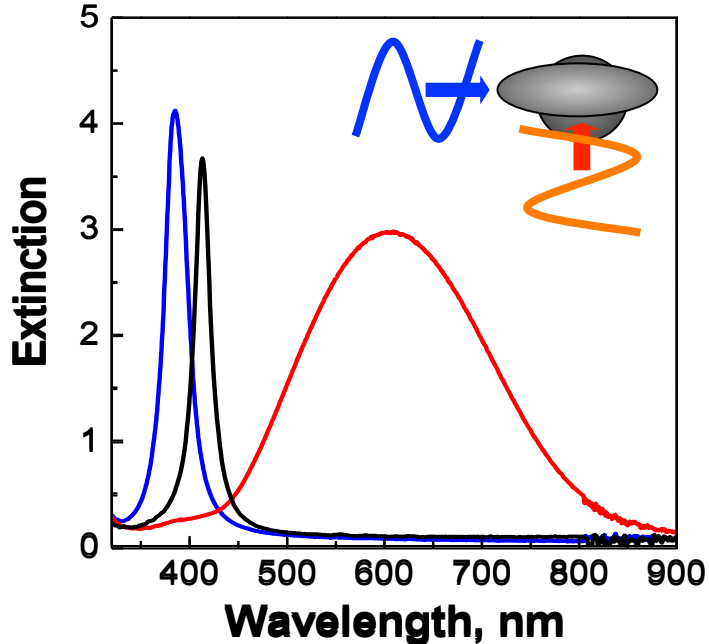


Temperature Simulation

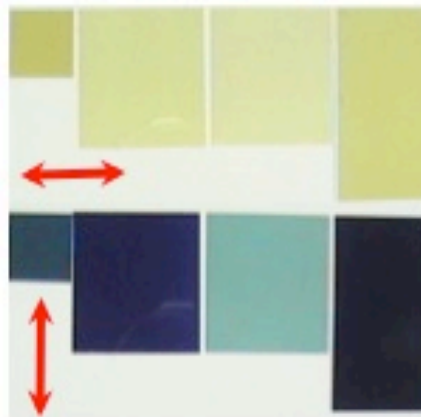
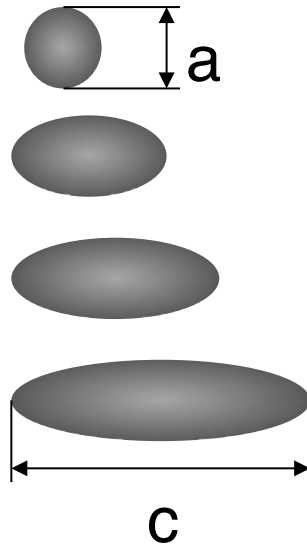


Non-spherical nanoparticles

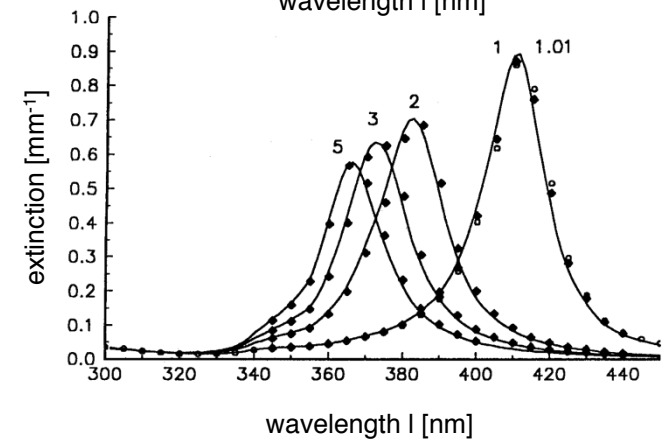
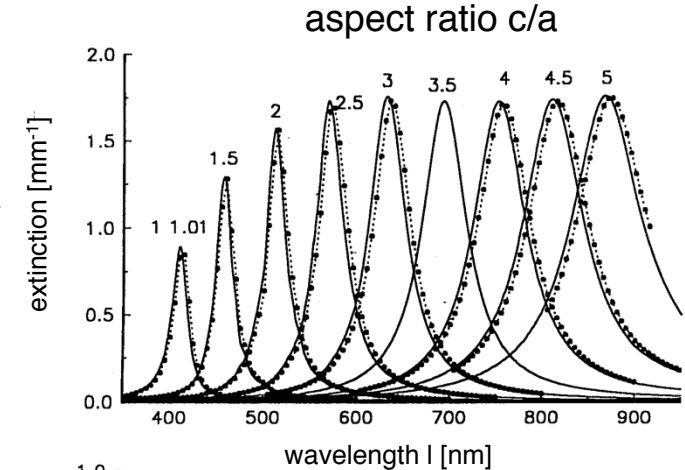
SPR position is size & **shape** dependent. Therefore metal nanoparticles with non-spherical shape should demonstrate several SPR in their spectra.



Polarised extinction spectra of spherical & spheroidal silver particles in glass



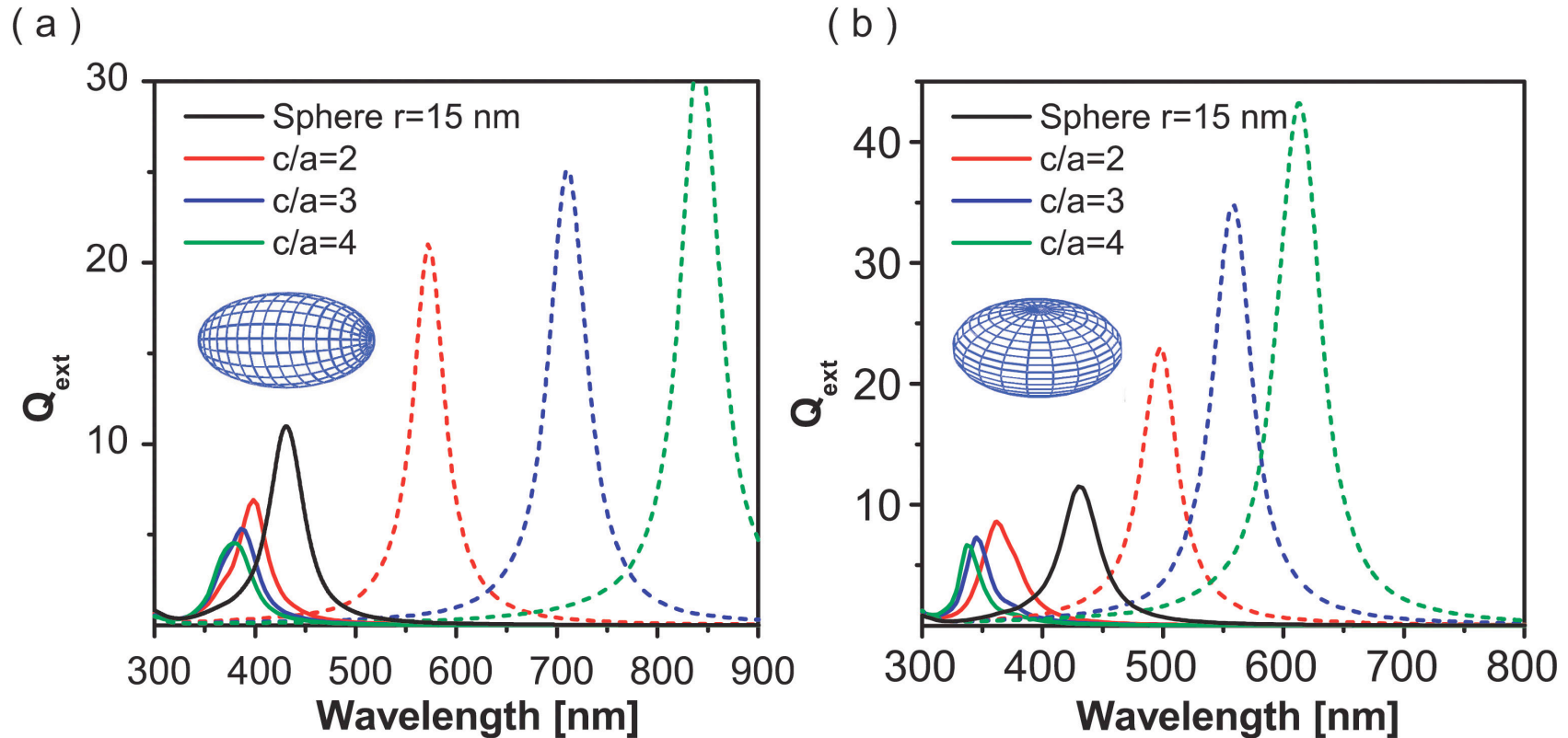
Increase of the aspect ratio leads to the rise of spectral separation between SPRs in polarised spectra



Non-spherical nanoparticles

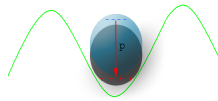
Mie theory for silver spheroids embedded in glass - with different aspect ratios

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**.

Non-spherical nanoparticles



Femtosecond laser irradiation of glass with embedded silver nanoparticles

Case in point

Wavelength: 515 nm

Pulse length: ~ 300 fs

Laser fluence: ~ 30 mJ/cm²

Dichroic spots: ~ 500 nm

left to right: 200, 300, 400, 500 pulses per spot

\longleftrightarrow : Laser polarization

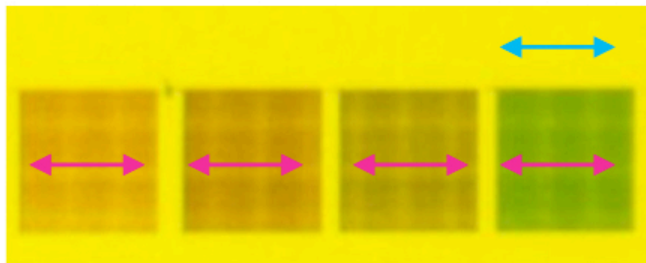
\longleftrightarrow : Readout polarizer

(a) Horiz. Pol. 0°

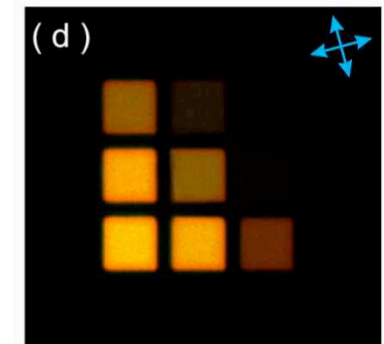
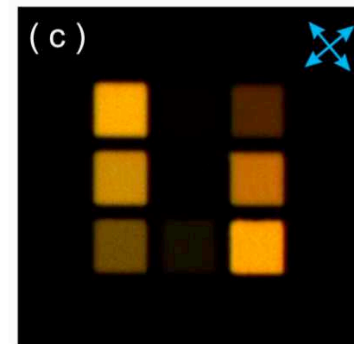
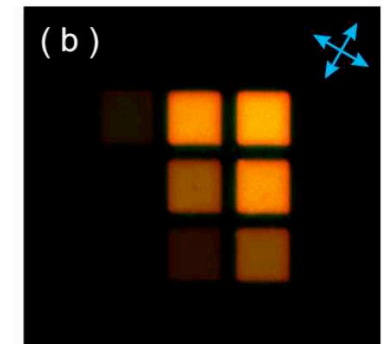
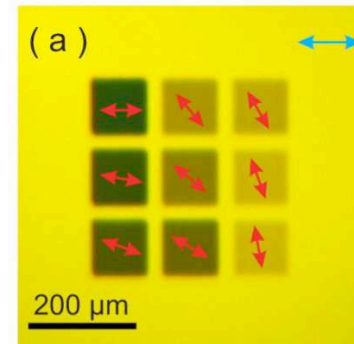
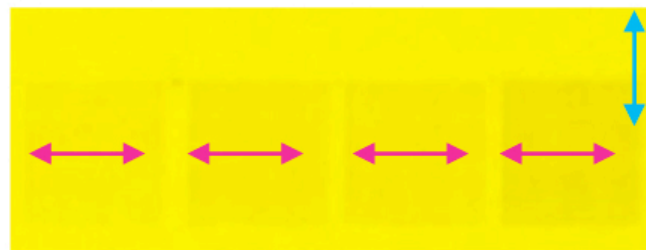
(b) Cross Pol. 20°

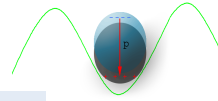
(c) Cross Pol. 45°

(d) Cross Pol. 70°

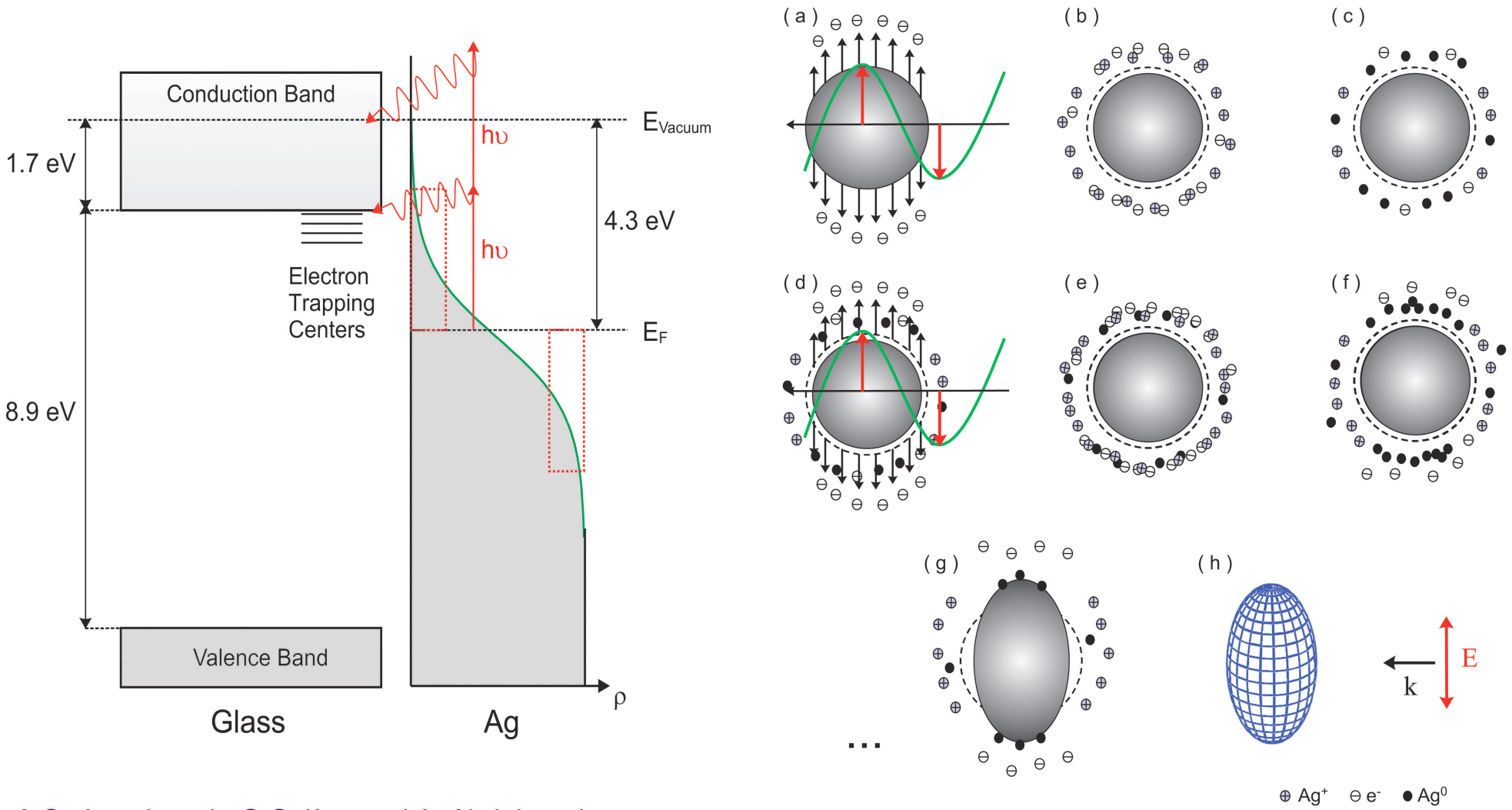


30 μ m



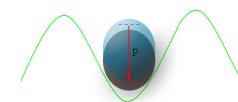


Energy level scheme of the electrons in a composite glass containing silver inclusions. The red dotted line indicates a non-thermal distribution of the electrons in the Ag nanoparticle caused by excitation of SPR. Green line –distribution of the electrons after thermalisation.



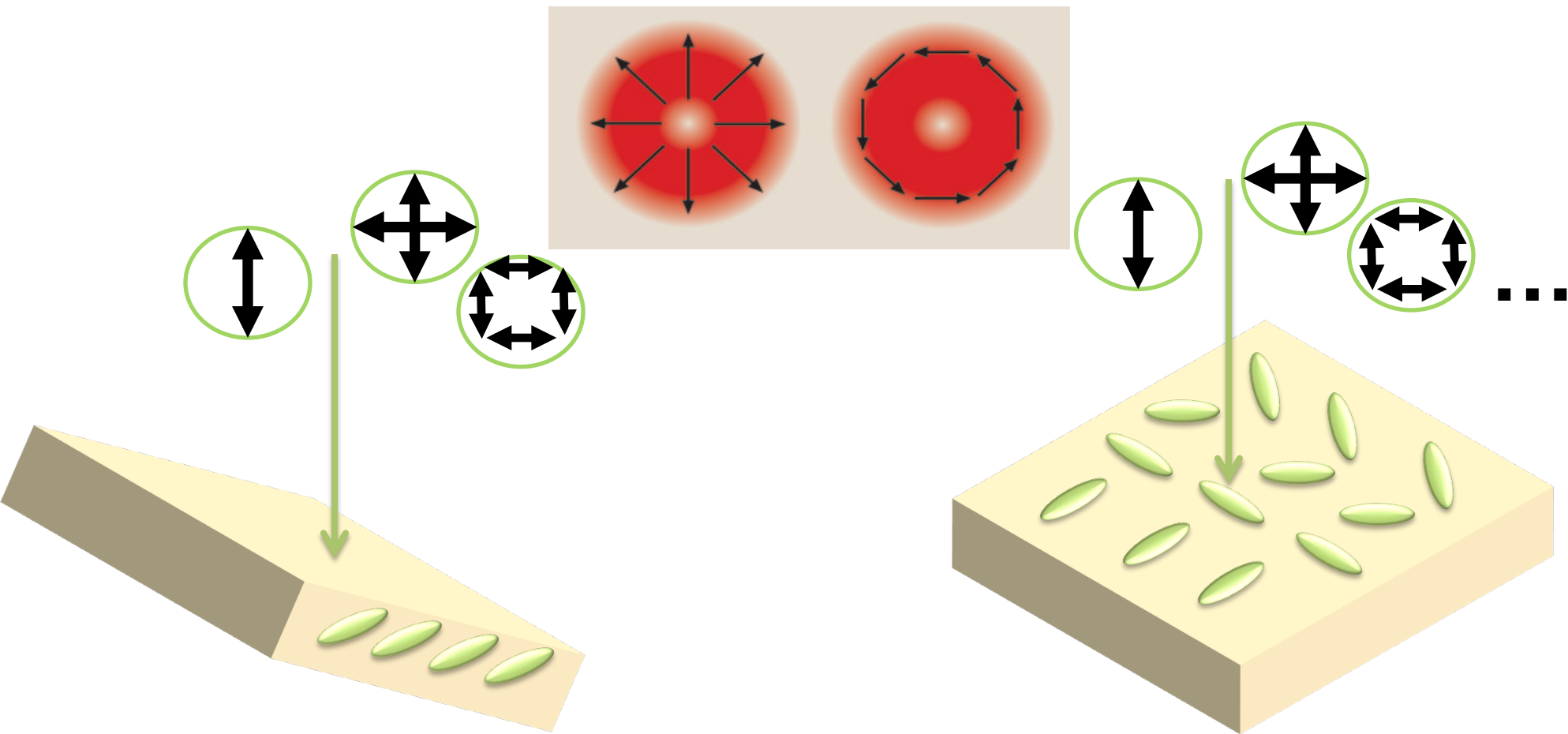
A. Stalmashonak, G. Seifert and A. Abdolvand

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



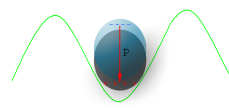
More advanced nanoparticle re-shaping ...

We are looking at more esoteric polarisation states for a range of applications:
radial and azimuthal polarisation



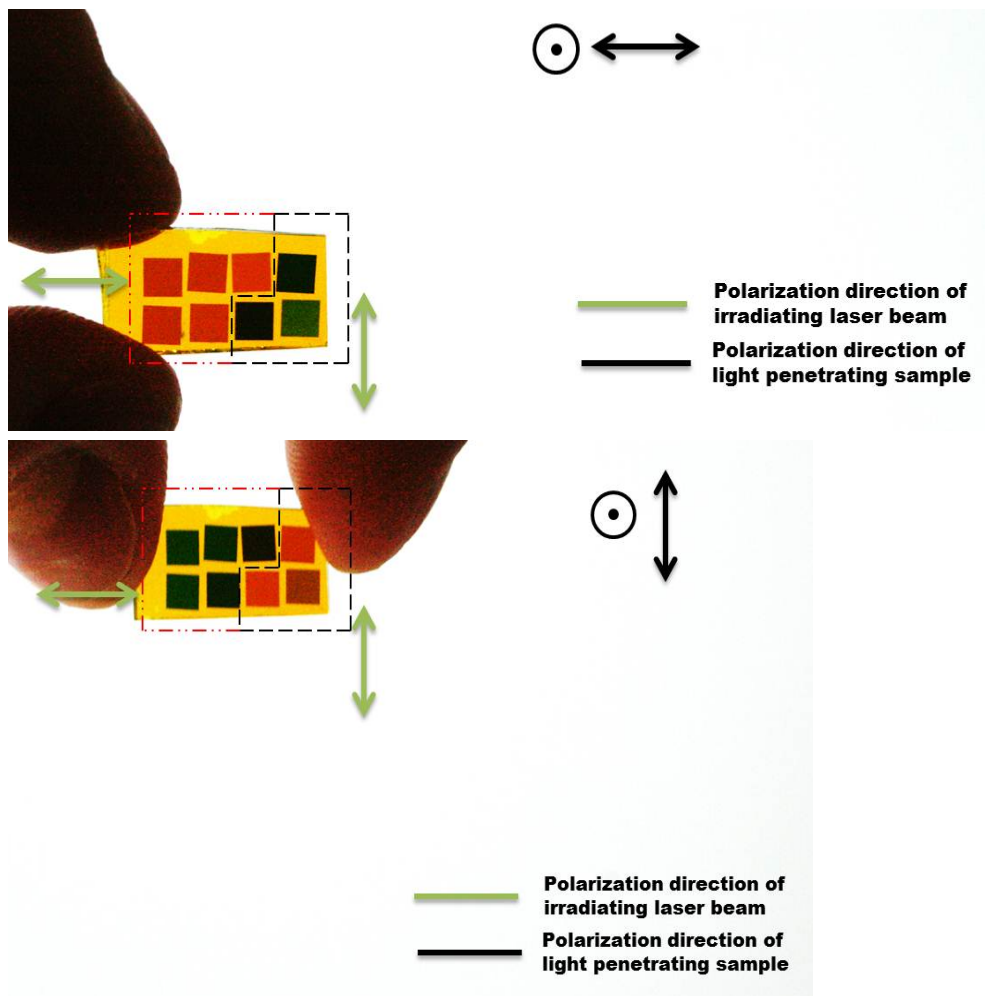
Mateusz A. Tyrk's work at Dundee

ESR Marie Curie Fellow

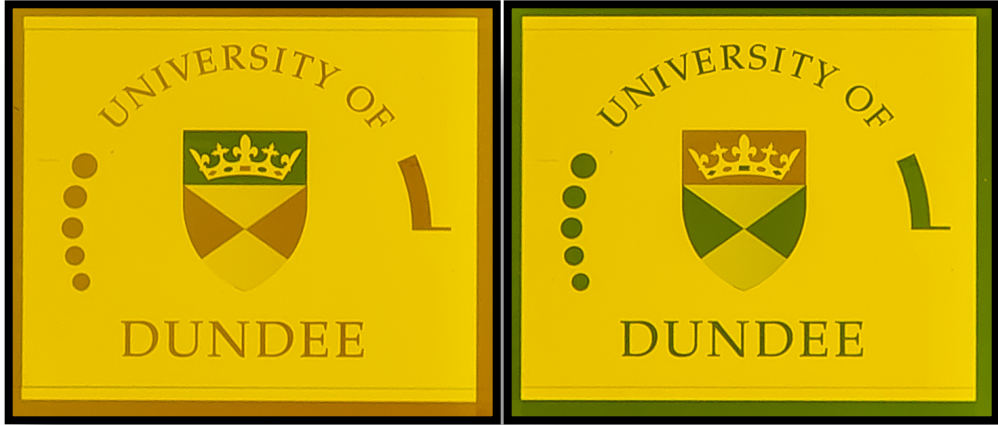


Non-spherical nanoparticles

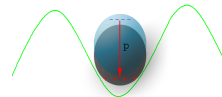
Picosecond laser irradiation of glass with embedded silver nanoparticles



Case in point
Wavelength: 532 nm
Pulse length: ~ 6 ps at 200 kHz
Laser fluence: ~ 0.3 J/cm²
Focal spot diameter: 15 μm

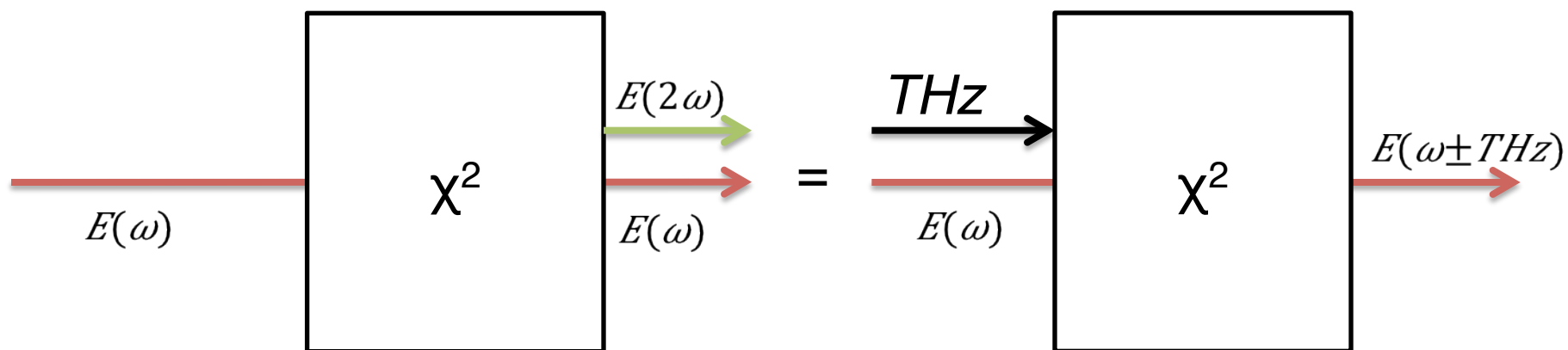


● — ● **10 mm**

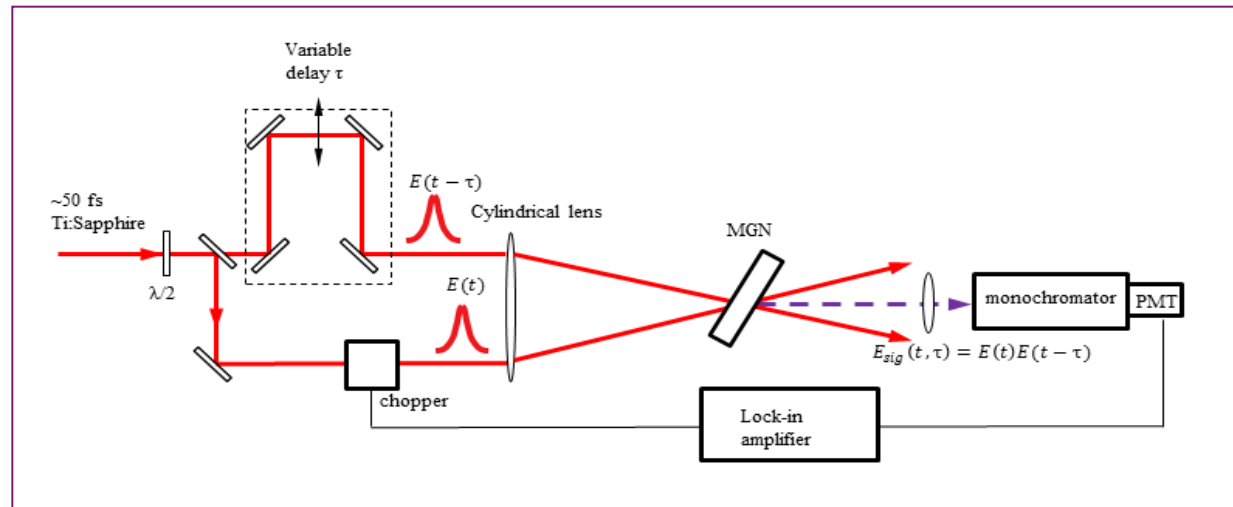
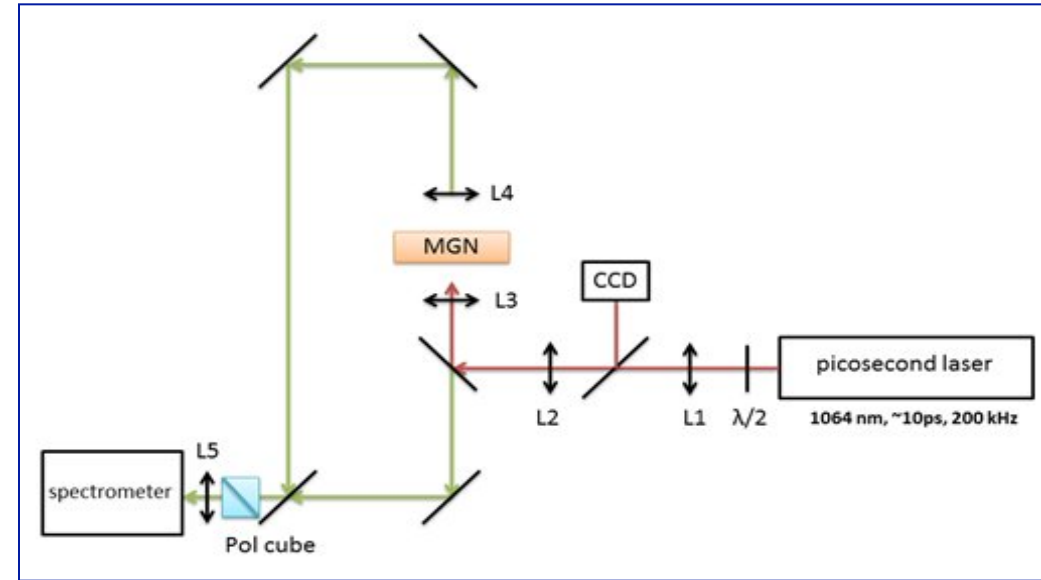
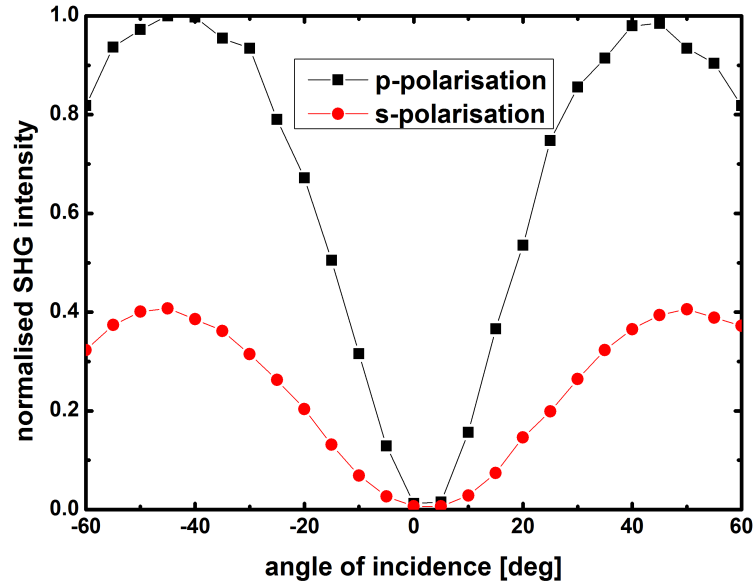


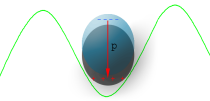
Second Harmonic Generation (SHG) observation

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

Results to be reported in June (Liverpool)

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!