

Nonlinear optics spectroscopy in glasses doped with nanoparticles

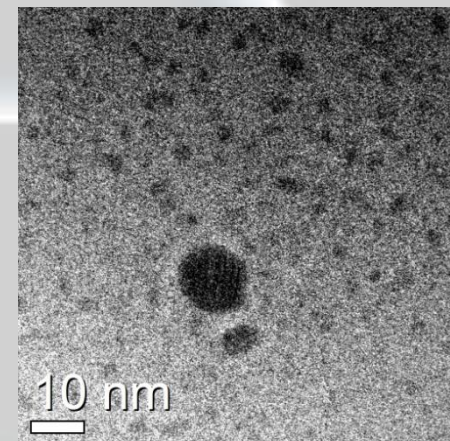
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Outline

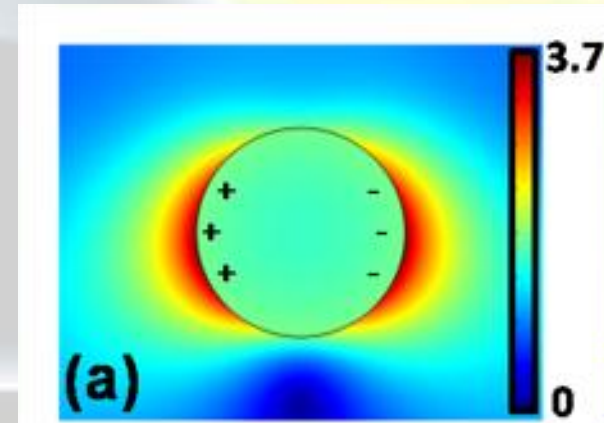
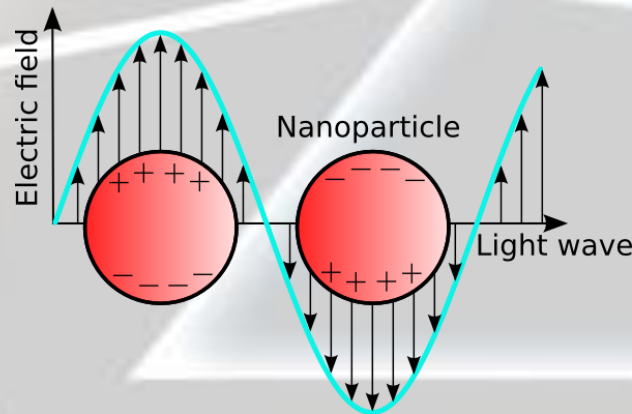
- ✕ Motivation
- ✕ Background (Nonlinear Optics effect)
- ✕ Experimental setup (Z-Scan and WLCZ-Scan and Optical Kerr Gate)
- ✕ Experimental results
- ✕ Conclusion



Motivation

Why study nonlinear effect in glasses with nanoparticles ?

- Local field enhancement effect

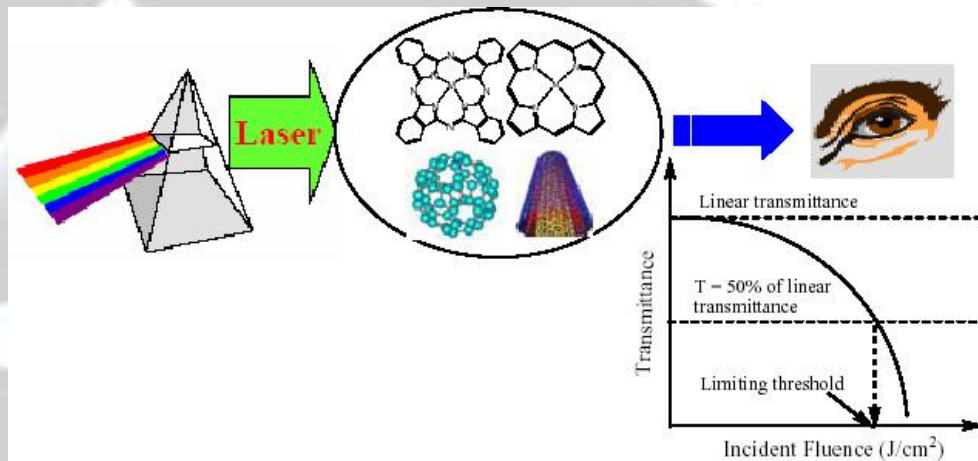
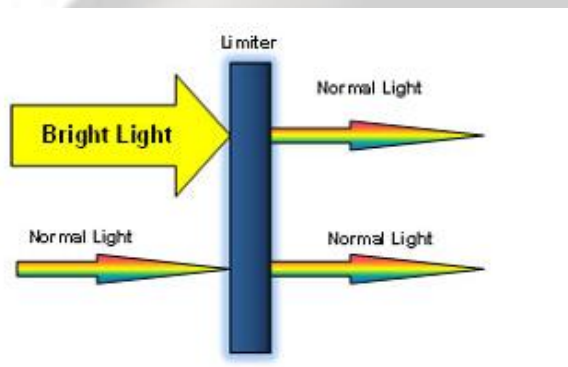


- They are promising materials for photonic applications: ultrafast response times and high third order nonlinearities

Motivation

- ✖ Find new materials with Multi-photon absorption

Optical Limiters

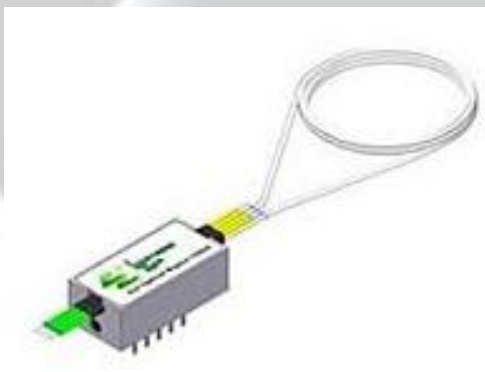


http://www.lightwavelogic.com/applications_optical_filters_limiters.html

<http://spie.org/x14682.xml>

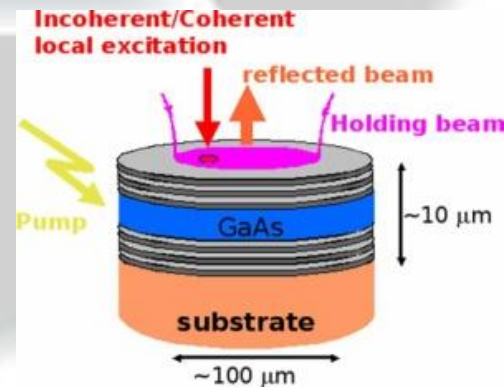
- ✖ Find new materials for telecom

All-Optical Switches



<http://www.directindustry.com/prod/lightwavelink/industrial-fiber-optic-switches-36007-718873.html>

- ✖ Excited state life time
- ## Saturable absorbers



http://www.lpn.cnrs.fr/en/PEQ/AutoOrg_SC.php

Nonlinear Optics

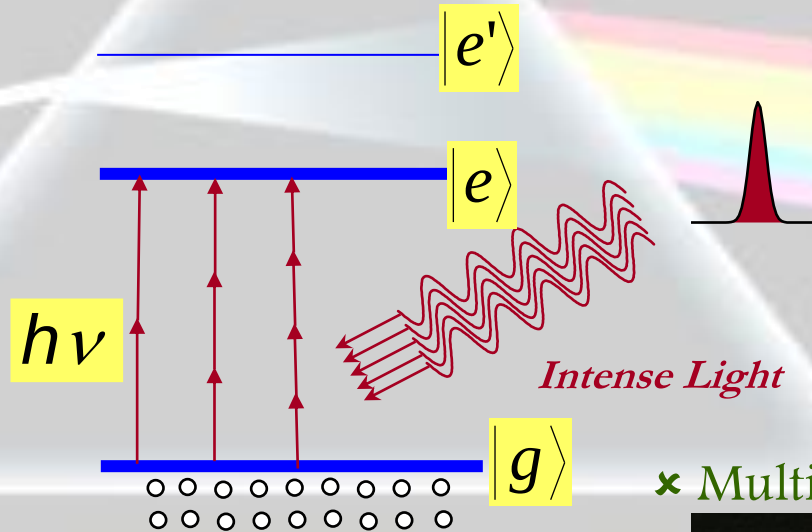
✓ Nonlinear Polarization (transparent medium)

$$P = \chi^{(1)}E + \chi^{(2)}EE + \chi^{(3)}EEE + \chi^{(4)}EEEE + \chi^{(5)}EEEEEE \dots$$

$$\text{Im } \chi^{(3)}$$

$$\text{Im } \chi^{(5)}$$

$$\text{Im } \chi^{(7)}$$



✖ Multi-photon absorption

Nonlinear absorption

$$\alpha(I) = \cancel{\alpha_0} + \alpha_2 I + \alpha_3 I^2 + \alpha_4 I^3$$



Nonlinear Optics introduction

✓ Nonlinear Polarization (transparent medium)

$$P = \chi^{(1)}E + \chi^{(2)}EE + \chi^{(3)}EEE + \chi^{(4)}EEEE + \chi^{(5)}EEEEEE \dots$$

✚ Optical Kerr effect

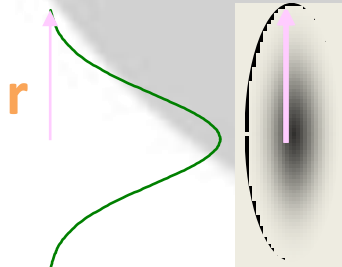
$$\text{Re } \chi^{(3)}$$

$$n(I) = n_0 + \Delta n = n_0 + n_2 I$$

$$n_2 = \frac{3\chi^{(3)}}{4n^2 \epsilon_0^2 c}$$

Nonlinear refraction

Nonlinear medium

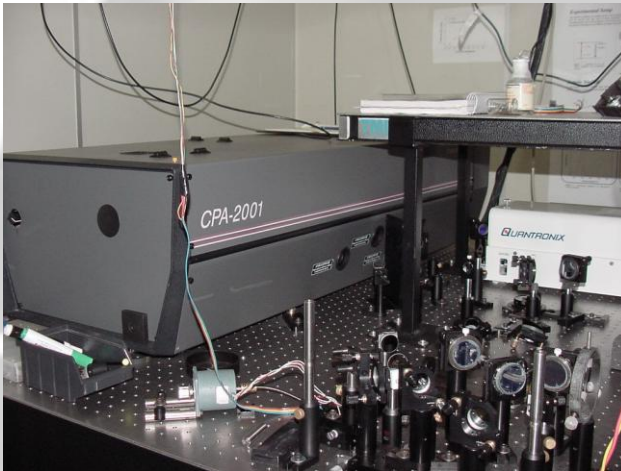


$$\Delta\phi = kn_2 I(r)L$$

Electronic effects

Experimental setup

Laser System



✓ *Ti:sapphire chirped pulse amplified system (CPA-2001)*

✓ 775 nm

✓ 150 fs

✓ 800 μ J



✓ *Optical parametric amplifier (TOPAS)*

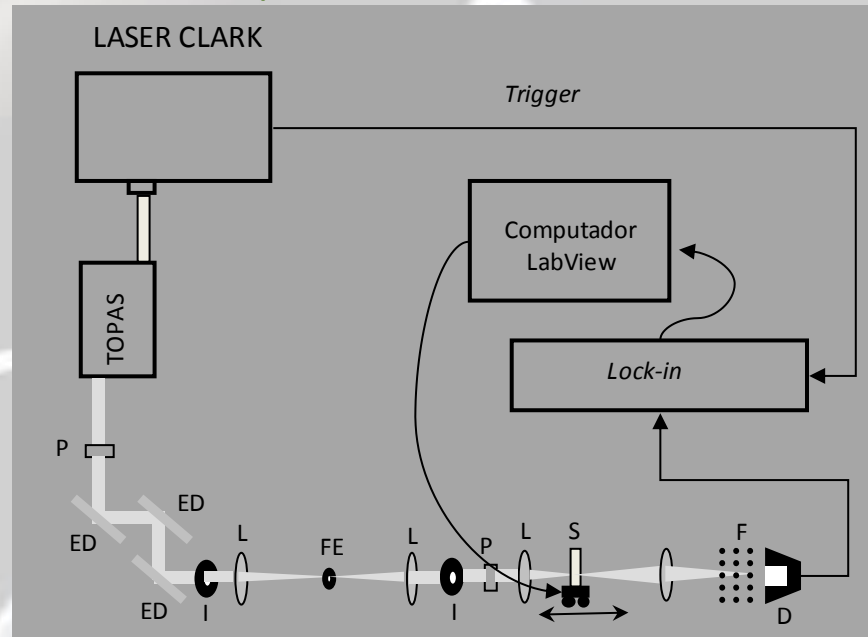
✓ 460 - 2600 nm

✓ $\approx$ 120 fs

✓ 20-60 μ J

Experimental setup

Z-scan technique



To obtain the spectrum of the nonlinear effect

$$T = \frac{1}{\sqrt{\pi} q_0(z,0)} \int_{-\infty}^{\infty} \ln[1 + q_0(z,0)e^{-\tau^2}] d\tau$$

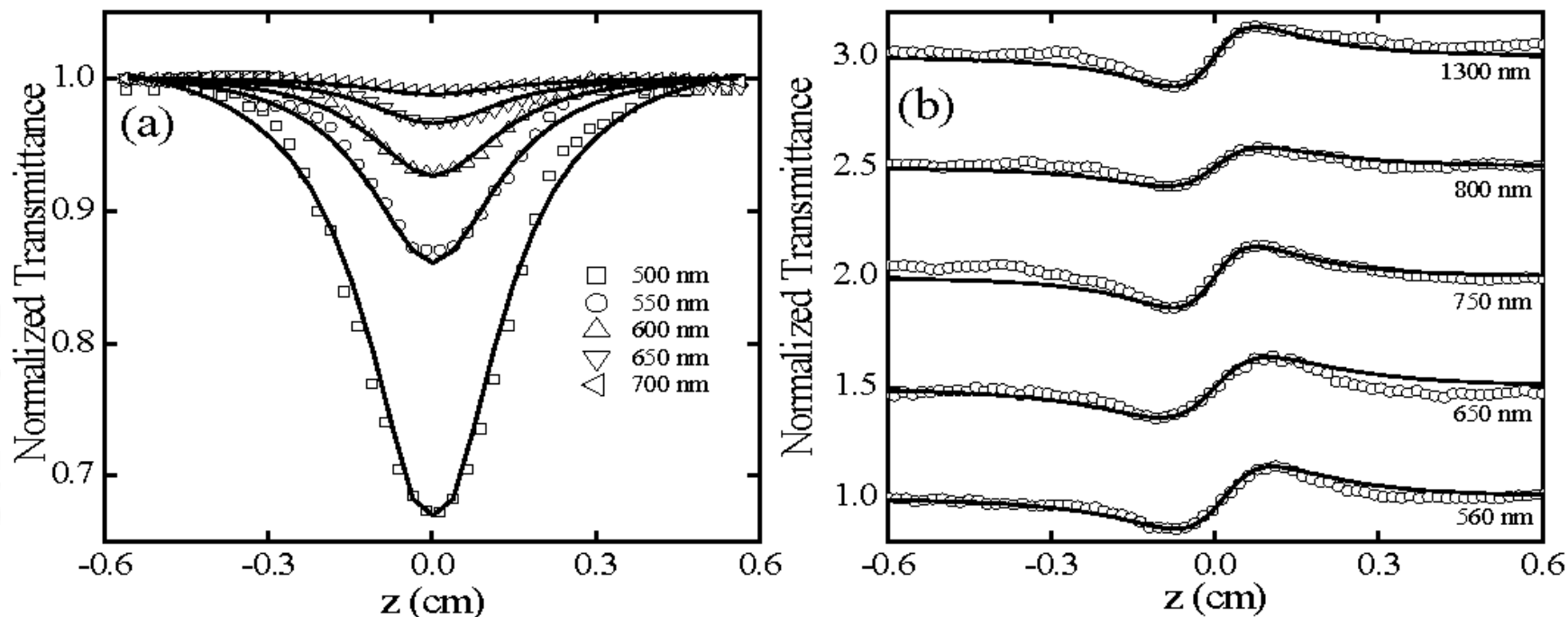
$$T(z, \Delta\phi_0) = 1 + \frac{4\Delta\phi_0 x}{(x^2 + 9)(x^2 + 1)}$$

$$q_0(z,t) = \frac{\beta I_0(t)L}{1 + z^2 / z_0^2}$$

$$\Delta\phi_0 = kn_2 I_0 L.$$

Z-scan Results

Traditional Z-scan



WLC Z-scan

Typical (a) open- and (b) closed-aperture Z-scan curves obtained at several wavelengths. The solid lines represent fittings.

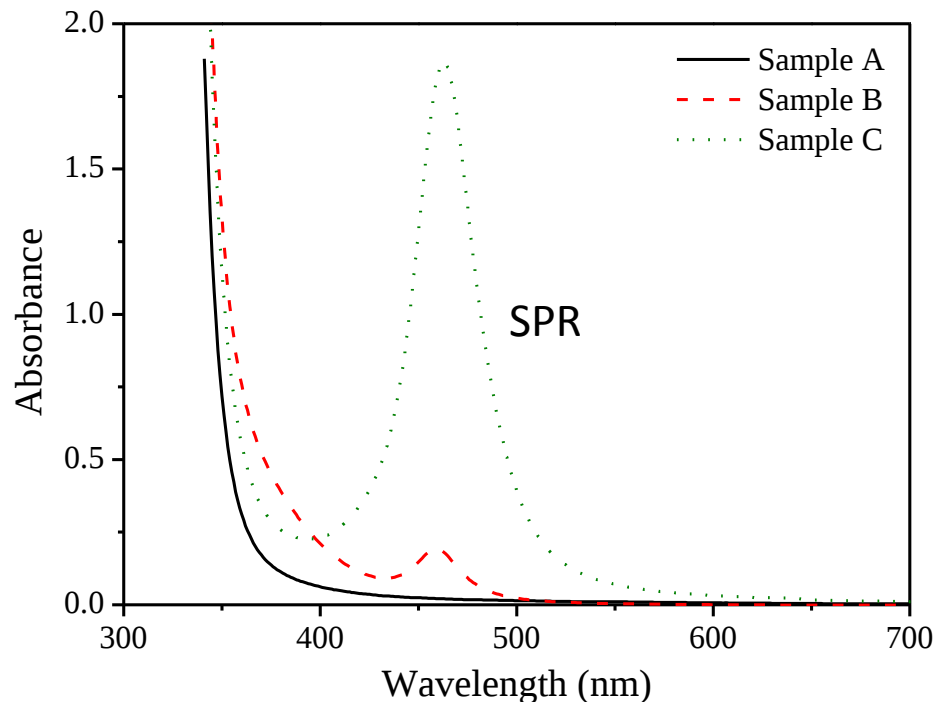
The pulse energy and the beam waist used in the open aperture Z-scan are 34 nJ and 16 μm for 500 nm, 29 nJ and 17 μm for 550 nm, 27 nJ and 18 μm for 600 nm, 23 nJ and 19 μm for 650 nm, 19 nJ and 20 μm for 700. The pulse energy and the beam waist used in the closed aperture Z-scan are 8 nJ and 16 μm for 560 nm, 16 nJ and 17 μm for 650 nm, 16 nJ and 18 μm for 750, 17 nJ and 18.5 μm for 800 nm and 62 nJ and 20 μm for 1300 nm.

Lead-germanium oxide glasses + Ag

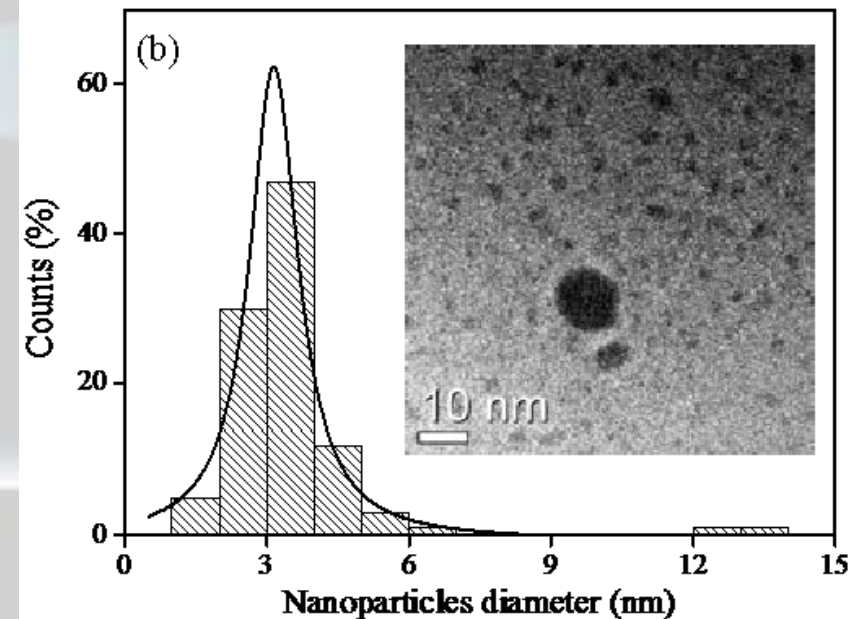
59PbO-41GeO₂ (in wt. %) for glass host (sample A)

+
AgNO₃ (5.0 wt. %)

Annealed { 420 °C for 1 h (sample B)
480 °C for 3 h (sample C)

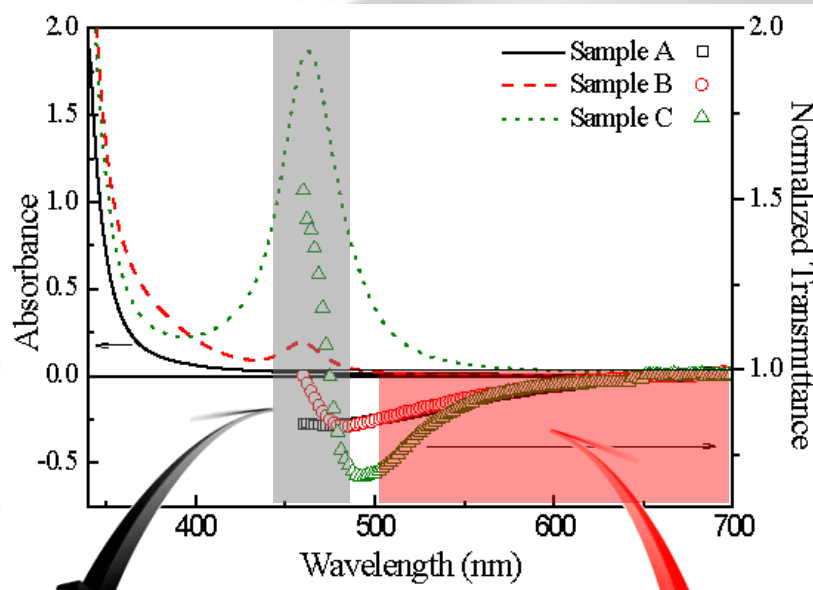


Filling factor of about **2%** for sample C



The inset shows the image of Ag NPs investigated with a high resolution TEM

Two-photon absorption spectra



Saturable absorption

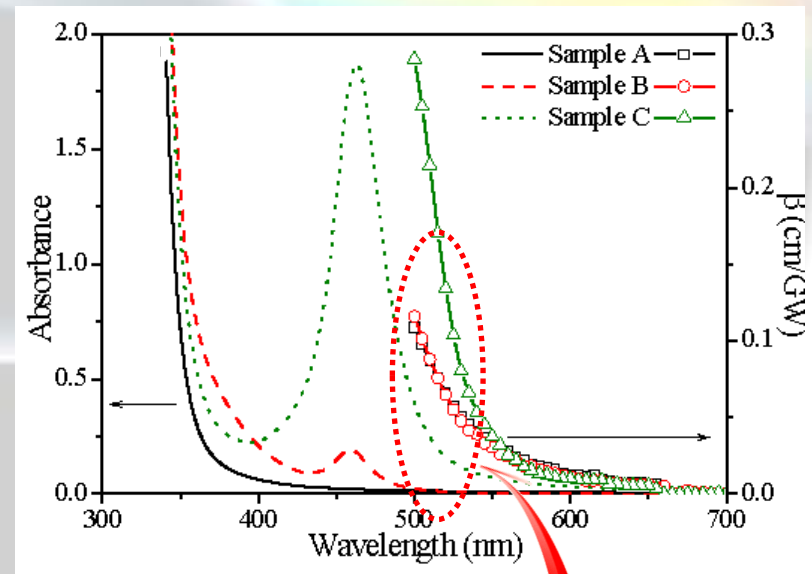
Two-photon absorption

This increment is attributed to an interband transition in the Ag nanoparticles, also mediated by a 2PA excitation.

Qu S. *et al* Opt. Mater. **28**(3), 259–265 (2006).

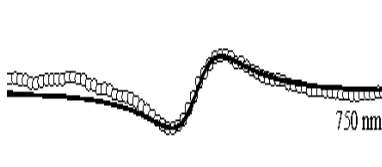
$$T = \frac{1}{\sqrt{\pi} q_0(z, 0)} \int_{-\infty}^{\infty} \ln[1 + q_0(z, 0) e^{-\tau^2}] d\tau$$

$$q_0(z, t) = \frac{\beta I_0(t) L}{1 + z^2 / z_0^2}$$



Enhancement effect

Nonlinear refraction spectra

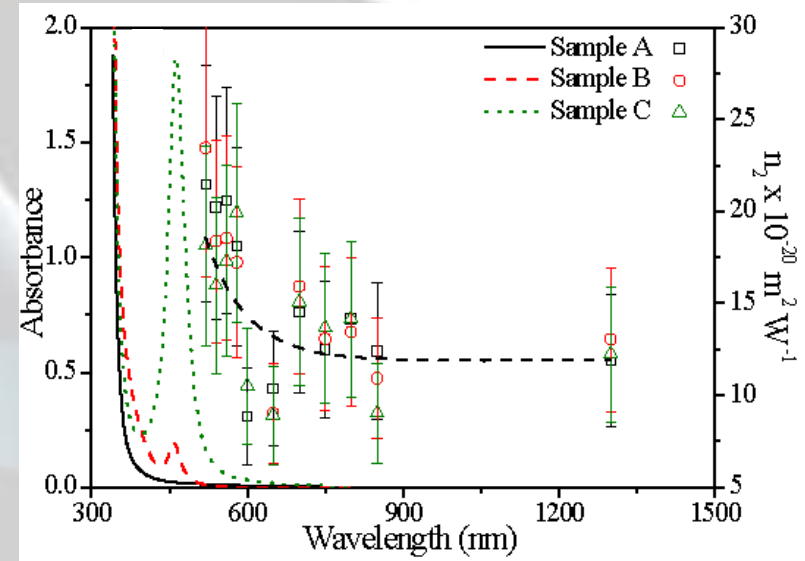


$$T(z, \Delta\phi_0) = 1 + \frac{4\Delta\phi_0 x}{(x^2 + 9)(x^2 + 1)}$$



$$\Delta\phi_0 = kn_2 I_0 L.$$

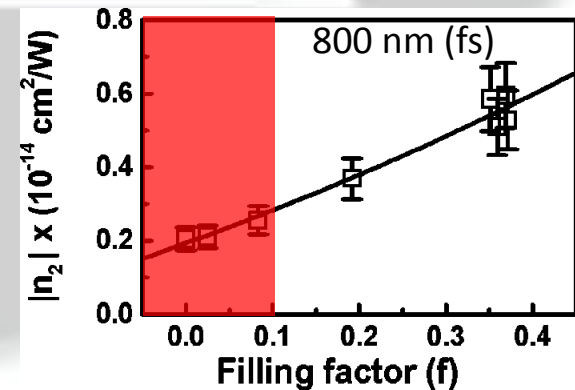
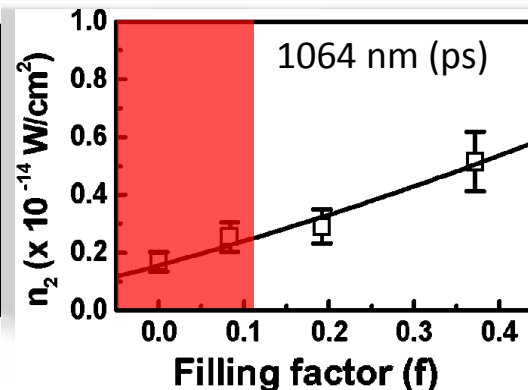
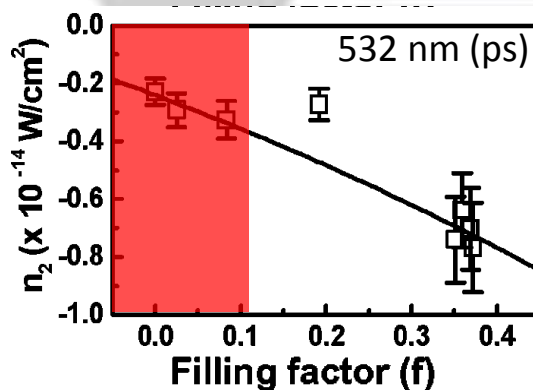
De Boni L *et al* Optics Express, 20, 6844- 6850 (2012)



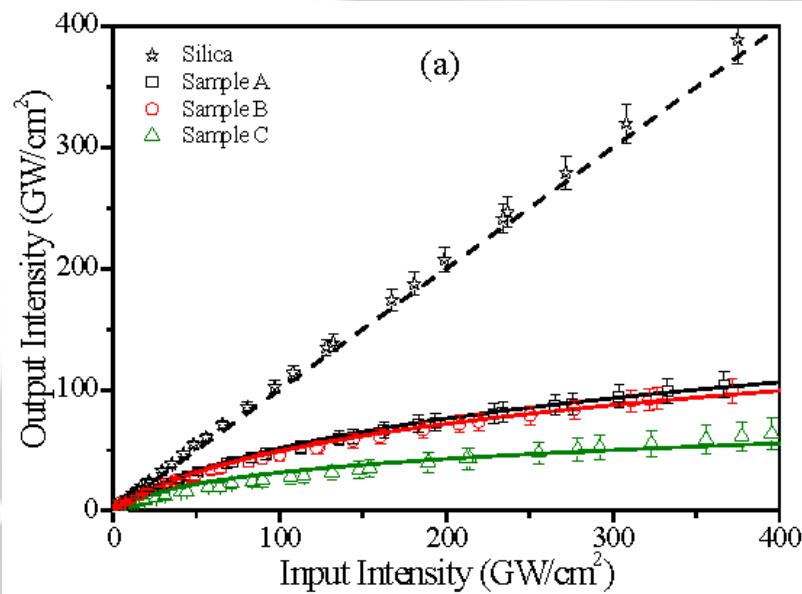
Same value → glass host

Transparent glass ceramic containing sodium niobate nanocrystals.

Falcão-Filho E *et al* Phys. Rev. B **69**(13), 134204 (2004).



Optical limiting effect



$$W = \Delta n_{\max} / \lambda \alpha_0$$

$$FOM = 2\beta\lambda/n_2$$

α_0 is very small

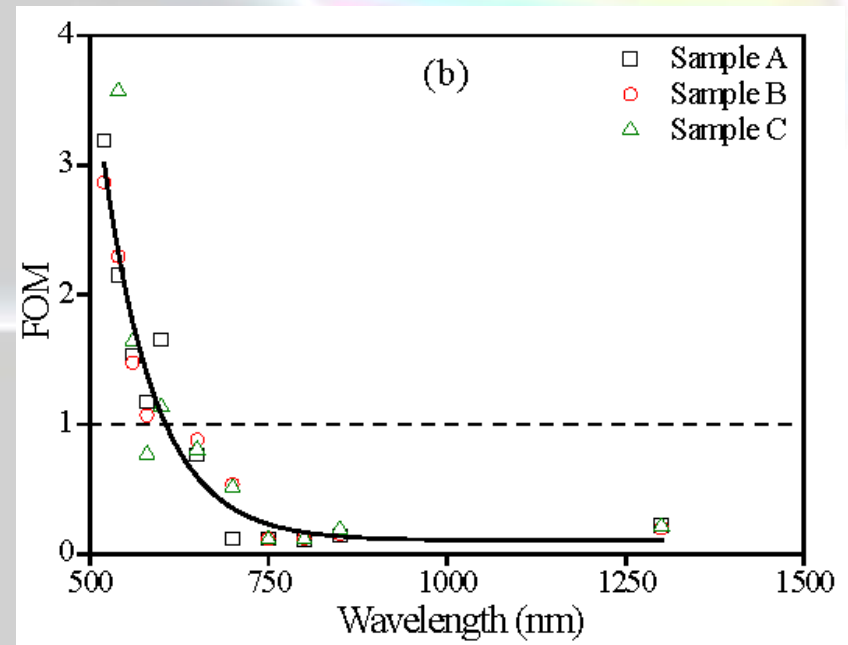
$$W > 2$$



All-optical switch

GOOD

$$FOM < 1$$



Tungsten Lead-Pyrophosphate + Cu

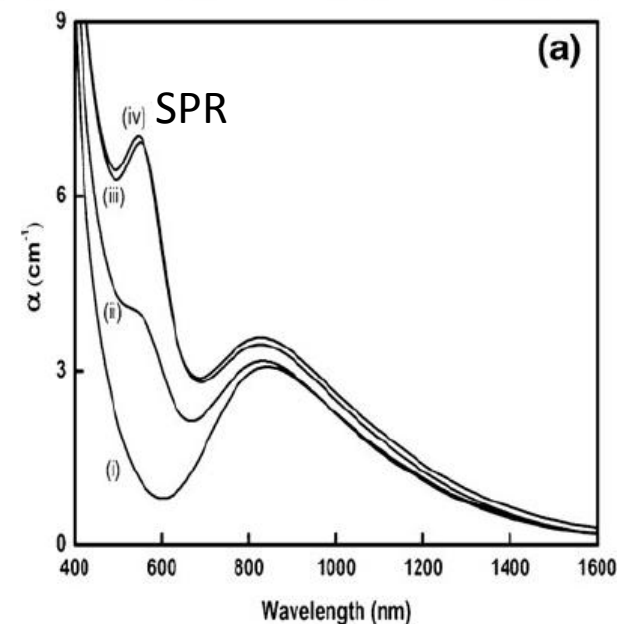
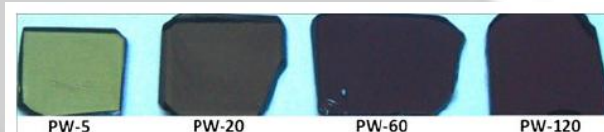
70Pb₂P₂O₇-30WO₃ (in wt. %) for glass host

+
CuO (0.5 wt. %)

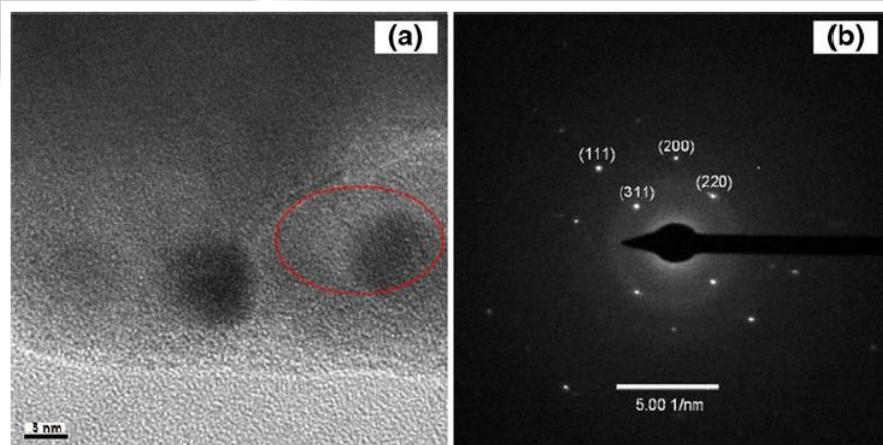
Table 1 Sample labels, synthesis conditions, and characteristic temperatures of PW glasses doped with CuO

Sample labels	Annealing conditions		Characteristic temperatures	
	T_{ht} (°C)	t_{ht} (min)	T_g (°C)	T_x (°C)
(i) PW-0	410	0	410	575
(ii) PW-5		5		
(iii) PW-20		20		
PW-60		60		
(iv) PW-120		120		

Changes in color

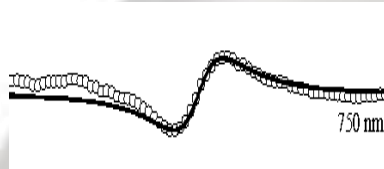


(a) Cu NPs investigated with a high resolution TEM



(b) Crystallographic planes of cubic Cu structure NPs

Nonlinear refraction and Absorption



$$T(z, \Delta\phi_0) = 1 + \frac{4\Delta\phi_0 x}{(x^2 + 9)(x^2 + 1)}$$



$$\Delta\phi_0 = kn_2 I_0 L.$$

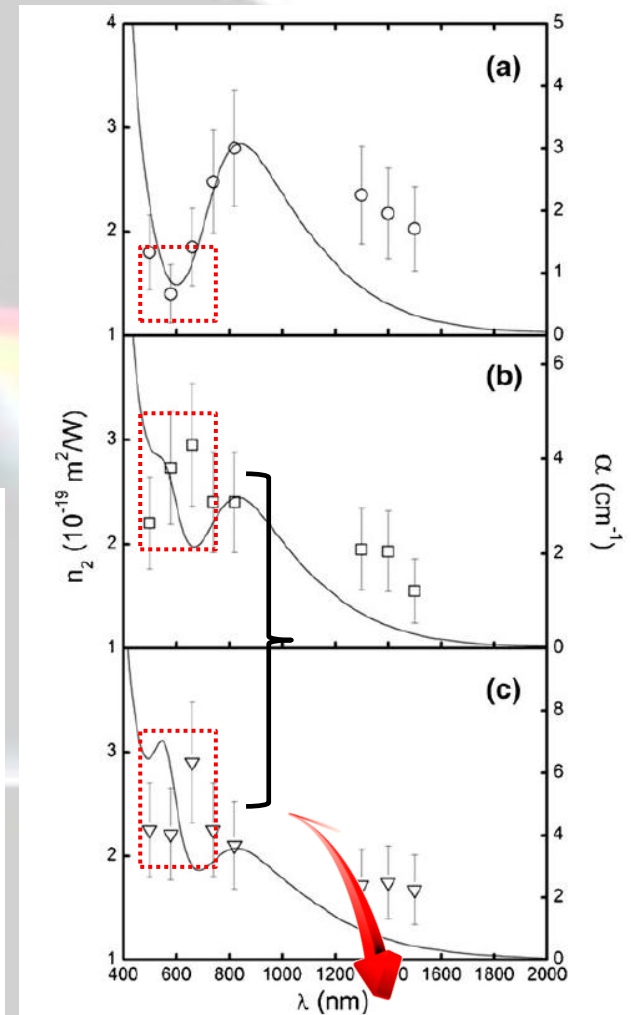
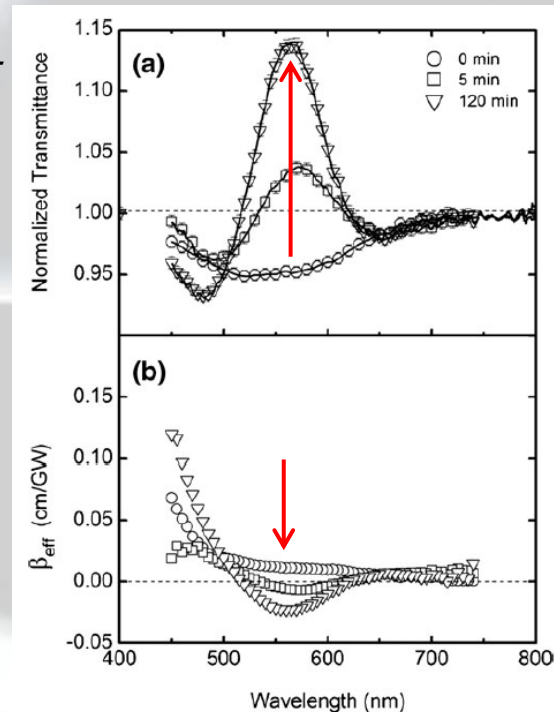
Nonlinear Absorption spectra

$$T = \frac{1}{\sqrt{\pi} q_0(z, 0)} \int_{-\infty}^{\infty} \ln[1 + q_0(z, 0) e^{-\tau^2}] d\tau$$



$$q_0(z, t) = \frac{\beta I_0(t) L}{1 + z^2 / z_0^2}$$

Inversion of the effect



Enhancement effect

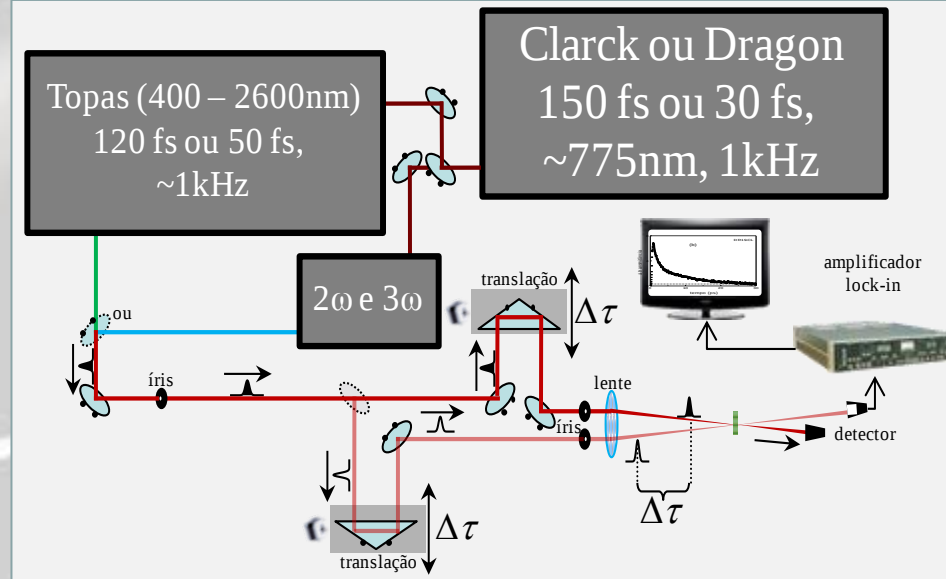
Optical Kerr Gate effect

Outside of the
Plasmon band

780 nm

200 fs

FAST

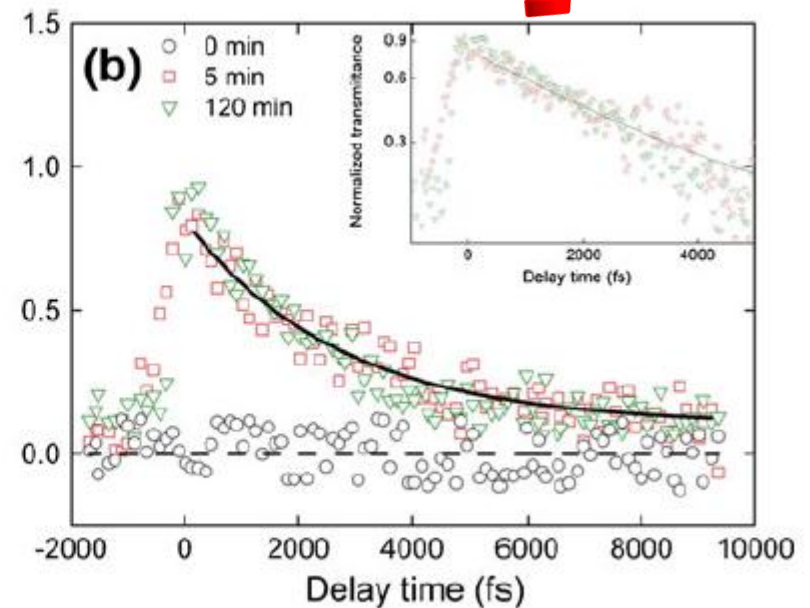
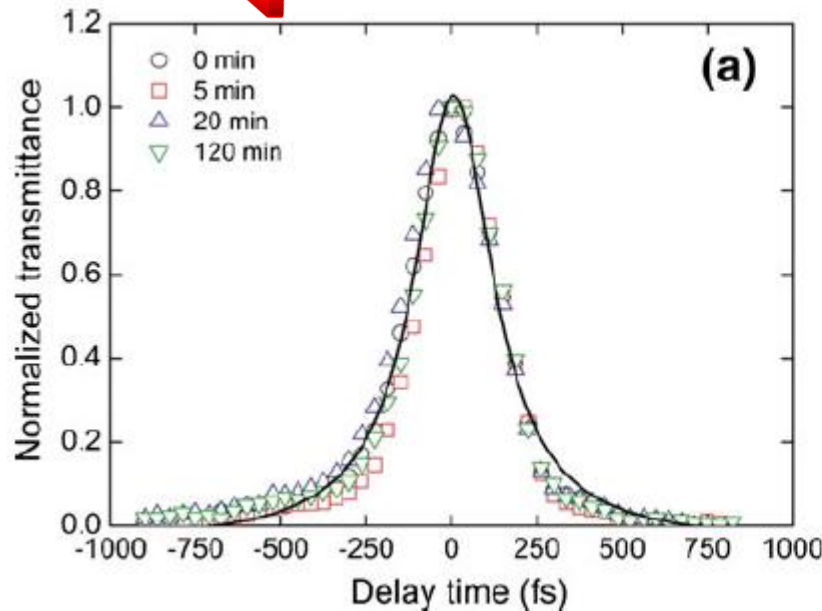


Inside of the
Plasmon band

560 nm

2.3 ps

SLOW



Heavy metal oxide glasses + Au

58.4 GeO₂–41.6 Bi₂O₃ (in wt. %) for glass host

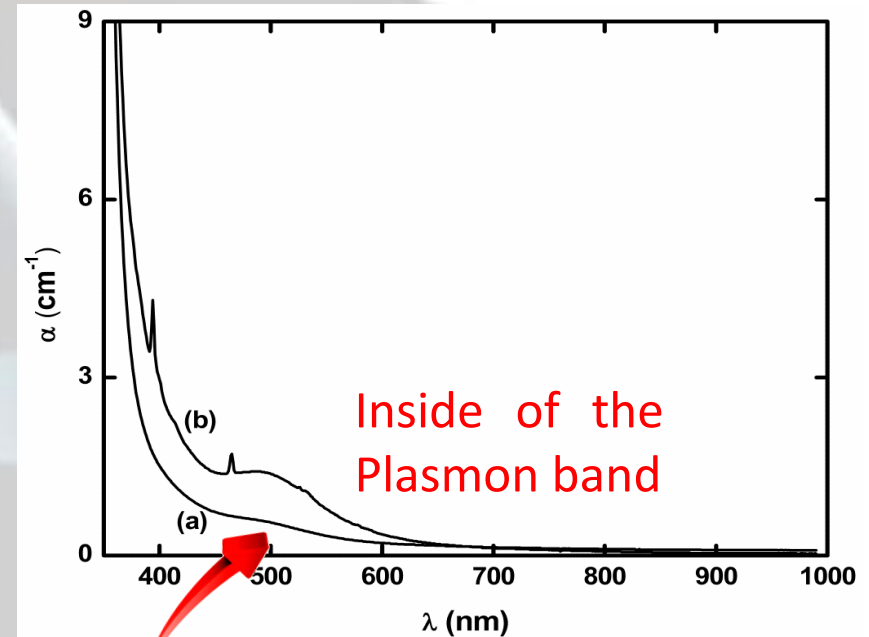
+

3Au₂O₃ – 0.5Eu₂O₃ (wt%)

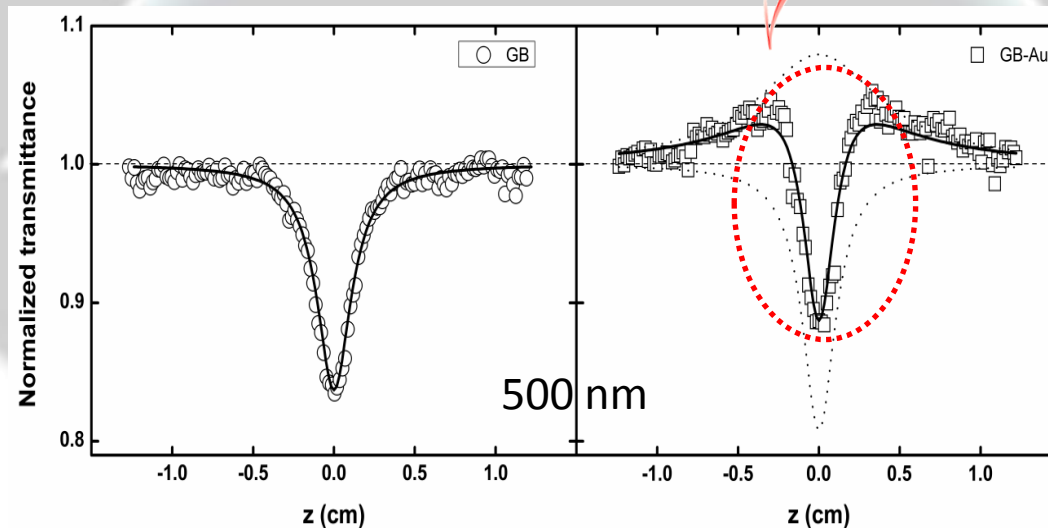
Nonlinear Absorption spectra

$$T = \frac{1}{\sqrt{\pi}q_0(z,0)} \int_{-\infty}^{\infty} \ln[1 + q_0(z,0)e^{-\tau^2}] d\tau$$

$$q_0(z,t) = \frac{\beta I_0(t)L}{1 + z^2 / z_0^2}$$



Two-photon
Absorption



Two-photon
Absorption +
Saturable
absorption

Heavy metal oxide glasses + Au

58.4 GeO₂–41.6 Bi₂O₃ (in wt. %) for glass host

+

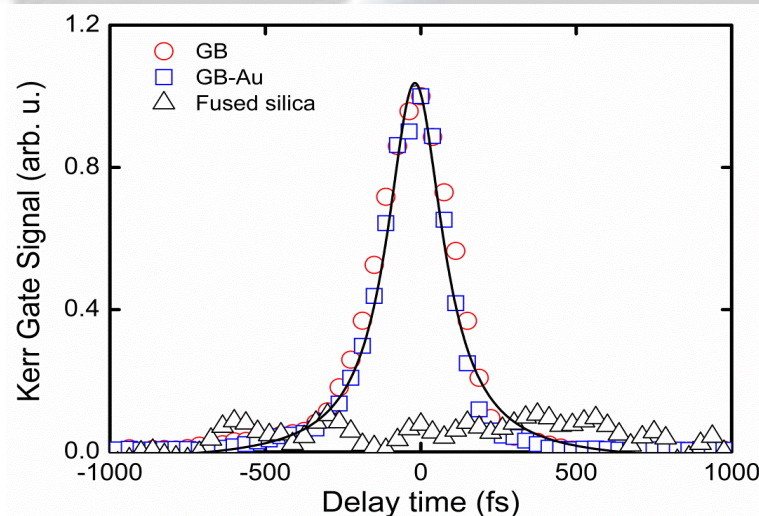
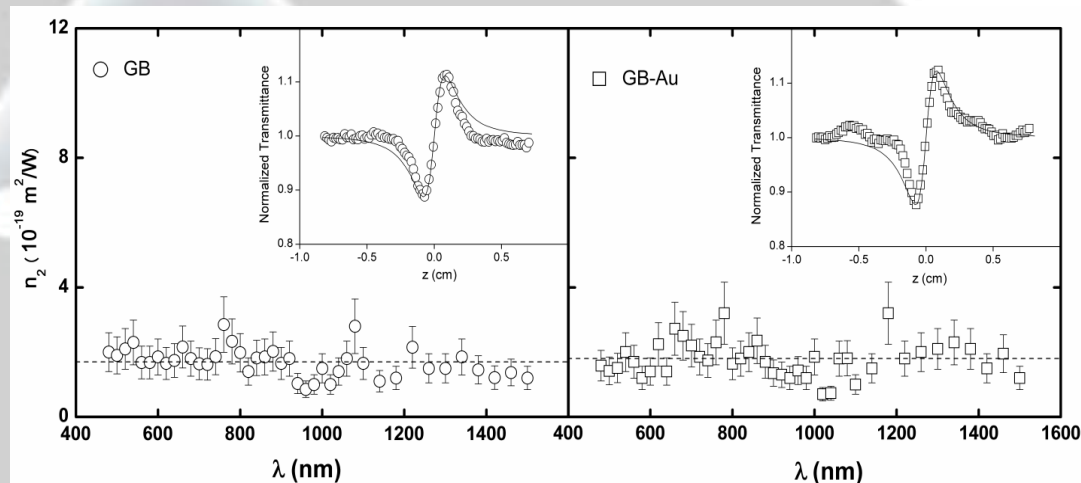
3Au₂O₃ – 0.5Eu₂O₃ (wt%)

Nonlinear refraction spectra

$$T(z, \Delta\phi_0) = 1 + \frac{4\Delta\phi_0 x}{(x^2 + 9)(x^2 + 1)}$$



$$\Delta\phi_0 = kn_2 I_0 L.$$



Outside of the Plasmon band

780 nm

~200 fs

FAST

Conclusion

- ✓ **Saturable** and **two-photon** absorption were observed

plasmon band

Increased because of an *interband transition* in the case of Ag nanoparticles

- ✓ **Nonlinear refraction** was observed to be the same

10 times higher than fused silica

low filling factor Ag and Au

- ✓ **Optical limiting effect** was observed to increase a factor of **2** due to the AgNP

- ✓ Samples can be used as **optical limiters**, **all-optical switches** and **saturable absorbers** because of the distinct response times

FAST

SLOW

Acknowledgements



Thank you