



## اشباع شدگی جذب توسط نانو-ذرات نقره نشانده شده درون شیشه تحت تابش پالسهای فمتولیزری در طول موج 400 نانومتر

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چکیده - پدیده اشباع شدگی جذب یک رفتار وابسته به شدت است که به علت کاهش شدید جمعیت تراز پایه به هنگام تابش پالسهای پراثری رخ می دهد. این پدیده برای نانو ذرات نقره نشانده شده درون شیشه تحت تابش پالسهای فمتوثانیه با طول موج 400 نانومتر مشاهده شد. این طول موج بیشینه ی طیف جذب نانو ذرات نقره به علت پدیده پلاسمون سطحی است. از طرفی این طول موج هارمونیک دوم تابش لیزر تانیم-سفایر تولید کننده پالسهای فمتوثانیه است. اشباع شدگی شدید جذب در نمونه تولید شده روی طول موج 400 نانومتر برای استفاده در ابزاری همچون سویچ های تمام اپتیکی بسیار نویدبخش است.

کلید واژه- اپتیک غیر خطی، اشباع شدگی جذب، پالسهای فمتولیزر، شیوه انتقال و تعویض یون، نانو ذرات نقره

## Saturation of Absorption for Silver Nanoparticles in Glass irradiated with Femtosecond Laser pulses at 400 nm

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Abstract- Saturation of absorption (SA) is an intensity dependent behavior occurs at high intensity regime due to depletion of the ground state population during the laser pulse irradiation. In this report the observation of SA for silver nanoparticles (NPs) embedded in glass prepared by ion exchange method is presented. The nonlinear transmission and the Z-scan methods both were employed to verify the SA as well as to determine the saturation intensity and linear absorption coefficient for silver NPs. The light source was the second harmonic of the Ti-sapphire laser corresponding to 400 nm wavelength which is close to the surface Plasmon resonance (SPR) of the silver NPs. The obtained results show that the saturation pulse energy for the examined sample is 330 nJ corresponding to the saturation intensity of 845 GW/cm<sup>2</sup>. Observation of strong SA at low saturation intensity ( $I_s$ ) for the prepared sample make it as a promising candidate for applications exploiting SA phenomenon such as producing short laser pulses and all optical switching devices.

Keywords: Femtosecond laser pulses, Ion exchange method, Nonlinear optics, Saturation of absorption, Silver nanoparticles

## 1 Introduction

Metal-glass composites containing nanoparticles (NPs) such as copper, gold and silver have attracted great interest due to their versatile applications in optical and photonic devices. Interaction between electric field of the irradiation light and the free electrons confined in the metal NPs results in the collective oscillation of the electrons and thus absorption of light as a consequence. This phenomenon known as surface plasmon resonance (SPR) [1] underlies the narrow band absorption spectrum of transparent solid materials like glass or polymer with embedded metal NPs.

When a bulk glass containing metal NPs is irradiated with high intense light at the frequency around the SPR, the saturation of absorption (SA) may be observed due to the depletion of the ground state population of the oscillating electrons of the conduction band of the metal NPs. SA has been exploited extensively to produce short pulses via passive mode-locking technique [2] and has recently been examined to design all-optical switching devices [3]. This optical nonlinear (i.e. intensity dependent) behavior has been observed for different kind of metal nanoparticles such as copper [4], gold [5] and silver [6-8] around their surface plasmon absorption wavelengths [9]. The SPR of the silver NPs occurs around 400 nm matching the second harmonic generation (SHG) of the Ti-Sapphire laser; a solid state laser enables to produce femtosecond pulses. To our knowledge, it is the first report on the SA of silver NPs embedded in glass using femtosecond laser pulses at 400 nm.

In this research, silver NPs were embedded in BK7 glass by the ion exchange method in order to obtain silicate glass having well-defined nonlinear properties. The details can be found in [10].

## 2 Theory and simulation results

SA is observed as an increase in the transmittance of a medium when the incident light intensity is increased. The dependence of absorption coefficient on intensity is given by

$$\alpha(I) = \frac{\alpha_0}{1 + I/I_s} \quad (1)$$

where  $\alpha_0$  is the linear absorption coefficient (low intensity regime) and  $I_s$  the saturation intensity [11]. The relation between the exit and incident intensity is governed by

$$\frac{I_{ex}}{I_s} + \ln\left(\frac{I_{ex}}{I_s}\right) = \frac{I_{in}}{I_s} + \ln\left(\frac{I_{in}}{I_s}\right) - \alpha_0 L \quad (2)$$

where  $L$  is the thickness of the sample.

In the Z-scan technique the incident intensity is varied via translating the sample along the propagation direction of a focused laser beam. The exit intensity is measured as a function of the sample position. Then, the transmittance (as the quotient of the exit intensity to incident intensity) is obtained as the function of the sample position.

Figure 1 shows the simulated Z-scan normalized transmittance. In the Z-scan method, as the sample is moved towards the focus, the medium is irradiated with higher light intensity thus the SA appears leading to higher transmittance. As the sample passes through the focus and moves away from the focal point, the inverse process takes place leading to a decrease in the transmittance due to reduction of intensity.

It is deduced from figure 1 that, lower saturation intensity along with higher linear absorption coefficient result in stronger saturation of absorption. The surface Plasmon absorption of silver nanoparticles arises from the transition of the free electrons of the conduction band during the laser pulse excitation. The larger linear coefficient implies that more ground-state electrons are pumped to the excited-state. Once these electrons are excited by a laser pulse, they do not oscillate at the same frequency as that of the unexcited electrons. The larger linear coefficient in silver nanoparticles the more ground-state plasmon band to bleach and hence stronger SA will be observed [8].

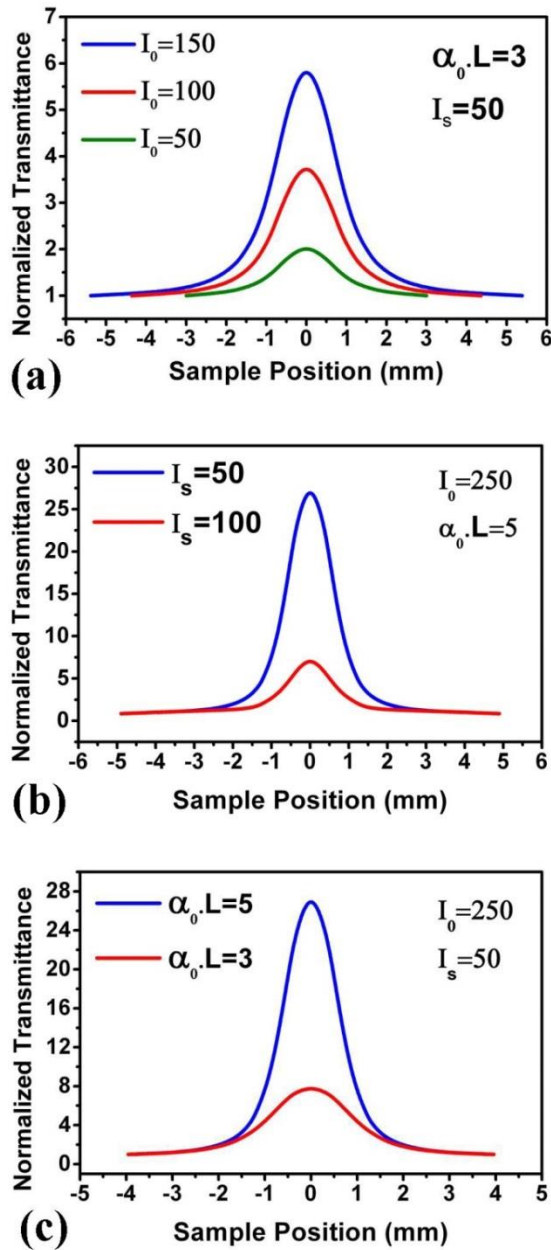


Figure 1. Simulation of Z-scan curves for a saturable absorber material. (a) For different on-axis peak intensity  $I_0$ , (b) for different saturation intensity  $I_s$  and (c) for different values of  $\alpha_0 L$ . ( $I_0$  and  $I_s$  have the same arbitrary unit)

### 3 Optical nonlinear properties

The saturation of absorption of the silver NPs were investigated using the open aperture Z-scan technique as well as nonlinear transmission method (NLT). A Ti-sapphire amplifier producing 25 fs pulses at the central wavelength of around 800 nm with the repetition rate of 1 kHz was employed.

A BBO crystal was used to generate 400 nm femtosecond pulses from the original laser pulses at 800 nm. The crystal was followed by a blue filter to block the 800 nm pulses in order to measure only the degenerate process at 400 nm. It is an excellent natural happening that the SHG of the Ti-sapphire laser matches the SPR of the silver NPs [10] where saturation of absorption is expected.

In NLT method the sample was placed and fixed at the focal point of the focused laser beam. The input intensity was varied using neutral density (ND) filters. The laser beam power and then the pulse energy were measured before and after the sample. Figure 2 shows the output pulse energy versus input pulse energy measured over a range of pulse energies which were practically producible in our setup. The circle points show the measured data and the solid curve represents the best fit to the data using Equation (2) from which the linear absorption coefficient was extracted to be 57.4 1/cm and the saturation pulse energy was extracted as 330 nJ corresponding to the saturation intensity of 845 GW/cm<sup>2</sup>.

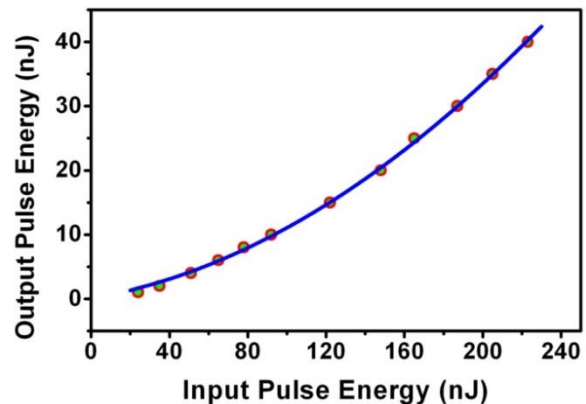


Figure 2. Exit pulse energy versus input pulse energy. Circle points show the experimental data and the solid curve is the best fit to the data using Equation (2).

The Z-scan results using femtosecond pulses at 400 nm for three different incident pulse energies are presented in figure 3. The transmittance is normalized with respect to the low irradiance transmittance when the sample is far enough from the focus and the saturation effect is very weak. As seen in figure 3, the transmittance increases as the sample is moved towards the focus which is an indication of saturation of absorption. Repeating the Z-scan with higher pulse energy gives higher

maximum transmittance supporting the results obtained from NLT experiment.

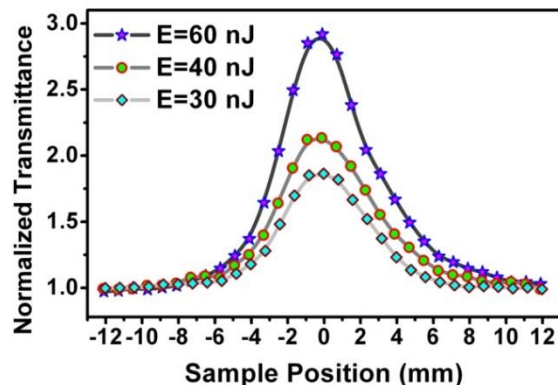


Figure 3. Z-scan results with different pulse energies. The points are the measured data and the solid lines are just displayed to guide the eye.

#### 4 Conclusion

Silver nanoparticle embedded in BK7 glass showed strong saturation of absorption at 400 nm wavelength matching the SHG of the Ti-sapphire laser. This behavior makes the glass doped with silver NPs a promising candidate for designing all optical devices and producing shorter laser pulses.

#### References

- [1] V. Amendola, O. M. Bakr and F. Stellacci, "A study of the surface plasmon resonance of silver nanoparticles by the discrete dipole approximation method: Effect of shape, size, structure, and assembly" *Plasmonics*. 5, 1, 85-97, 2010.
- [2] T. K. Lim and M. M. Denariez-Roberge, "Ultrashort-pulse generation by lasers with saturable absorbers" *Applied physics*. 15, 4, 439-450, 1978.
- [3] N. Venkatram, R. Sai Santosh Kumar, D. Narayana Rao, S. K. Medda, S. De and G. De, "Nonlinear optical absorption and switching properties of gold nanoparticle doped SiO<sub>2</sub>-TiO<sub>2</sub> sol-gel films" *Journal of*

*Nanoscience and Nanotechnology*. 6, 7, 1990-1994, 2006.

- [4] B. Ghosh, P. Chakraborty, S. Mohapatra, P. A. Kurian, C. Vijayan, P. C. Deshmukh and P. Mazzoldi, "Linear and nonlinear optical absorption in copper nanocluster-glass composites" *Materials Letters*. 61, 23-24, 4512-4515, 2007.
- [5] H. I. Elim, J. Yang, J. Y. Lee, J. Mi and W. Ji, "Observation of saturable and reverse-saturable absorption at longitudinal surface plasmon resonance in gold nanorods" *Applied Physics Letters*. 88, 8, 2006.
- [6] M. Hari, S. Mathew, B. Nithyaja, S. A. Joseph, V. P. N. Nampoori and P. Radhakrishnan, "Saturable and reverse saturable absorption in aqueous silver nanoparticles at off-resonant wavelength" *Optical and Quantum Electronics*. 43, 1-5, 49-58, 2012.
- [7] C. Zheng, X. Y. Ye, S. G. Cai, M. J. Wang and X. Q. Xiao, "Observation of nonlinear saturable and reverse-saturable absorption in silver nanowires and their silica gel glass composite" *Applied Physics B: Lasers and Optics*. 101, 4, 835-840, 2010.
- [8] C. Zheng, W. Li, W. Chen and X. Ye, "Nonlinear optical behavior of silver nanopentagons" *Materials Letters*. 116, 0, 1-4, 2014.
- [9] O. Plaksin, Y. Takeda, H. Amekura, N. Kishimoto and S. Plaksin, "Saturation of nonlinear optical absorption of metal-nanoparticle composites" *Journal of Applied Physics*. 103, 11, 2008.
- [10] L. Salavcová, B. Švecová, S. Janáková, O. Kolek, M. Míka, J. Špírková and A. Langrová, "Planar optical waveguides in newly developed Er:silicate glasses: A comparative study of K<sup>+</sup> and Ag<sup>+</sup> ion exchange" *Ceramics - Silikaty*. 49, 1, 53-57, 2005.
- [11] M. Hercher, "An Analysis of Saturable Absorbers" *Applied Optics*. 6, 5, 947-954, 1967.