



## افزایش خواص اپتیکی غیر خطی شیشه سیلیکا با استفاده از نانو ذرات فلزی به روش تی- ماتریس

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چکیده- در این مقاله، مدلی برای افزایش خواص اپتیکی غیر خطی در شیشه سیلیکا با استفاده از نانو ذرات فلزی به روش تی-ماتریس ارائه می شود. ابتدا برای به دست آوردن ضریب دی الکتریک موثر و ضریب پذیرفتاری غیر خطی موثر مرتبه سوم برای ترکیب شیشه سیلیکا و نانو ذرات کروی فلزی (طلا، نقره و مس)، تعمیم داده می شود، در این مقاله هم شیشه سیلیکا و هم نانو ذرات فلزی دارای ضریب دی الکتریک غیر خطی هستند. سپس از نظریه ماکسول-گارنت<sup>1</sup> برای جایگزین کردن نانو ذرات استوانه ای و بیضی گون به جای نانو ذرات کروی استفاده می شود و ضریب پذیرفتاری غیر خطی موثر مرتبه سوم در حالت ترکیب چهار موجی تبهگن محاسبه می گردد. طبق محاسبات انجام شده در این مقاله و نمودار های مرتبط با آن مشاهده می شود که ترکیب نانو ذرات کروی شکل نقره در شیشه سیلیکا بیشترین فاکتور کیفیت را از بین سه فلز مورد بحث و سه هندسه به کار رفته برای ترکیب مورد نظر (شیشه و نانو ذرات) ایجاد می کند.

کلید واژه- ضریب پذیرفتاری غیر خطی، فاکتور کیفیت، نانو ذرات فلزی

## Enhancement of Nonlinear Optical Properties of Silica Glass by Using Metallic Nanoparticle Compounds

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Abstract- Current paper is aimed at introducing a method for enhancing the nonlinear optical properties in silica glass by using metallic nanoparticles. First, the T-matrix method is developed in order to calculate the effective dielectric constant as well as the compound of silica glass and metallic nanoparticles, both of which possessing nonlinear dielectric constants. In second step, the Maxwell-Garnett theory is exploited to replace the spherical nanoparticles with cylindrical and ellipsoidal ones, facilitating the calculation of the third-order nonlinear effective susceptibility for a degenerate four-wave mixing case. The results are followed by numerical computations for silver, copper and gold as nanoparticles. It is shown, graphically, that the maximum and minimum of the real part of the reflection coefficient for nanoparticles of silver occurs in smaller wavelengths in comparison to that of copper and gold. Further, it is found that the nanoparticles of spherical shape exhibit greater figure of merit in comparison to those with cylindrical or ellipsoidal geometries.

**Keywords:** Figure of merit; Metallic nanoparticle; Nonlinear susceptibility coefficient

<sup>1</sup> Maxwell-Garnett

### 1 Introduction

The linear optics of heterogeneous media has been extensively studied, e.g., in Refs. [1-5]. By utilizing appropriate approximations, various methods for the calculation of effective dielectric constant can be found [6-10].

In this paper, in the parallel of Refs. [11, 12], we calculate degenerate four-wave mixing in silica glass and nanoparticles compound for a compound heterogeneous medium consisting of silica glass (host) and ellipsoidal gold, silver or copper metallic nanoparticle (guest). Finally, this research investigates the nonlinear properties of spherical, cylindrical and ellipsoidal metallic nanoparticles in silica glass for the case of degenerate four-wave mixing. Moreover, it shows how these parameters can be enhanced.

### 2 Modeling

The electric field components in an ellipsoidal along x, y and z axes are as follow:

$$E_{ik} = \frac{E_{0k}}{1 + L_k(\epsilon_{2k}^{non} - \epsilon_{1k}^{non}) / \epsilon_{1k}^{non}}, k = x, y \text{ and } z, \quad (1)$$

where *i* represents interior of the guest medium and the term  $\epsilon_{1k}^{non}$  ( $k = x, y, z$ ) is the nonlinear effective dielectric constant of the host and  $\epsilon_{2k}^{non}$  ( $k = x, y, z$ ) for guest. The nonlinear effective dielectric constant of ellipsoidal nanoparticles of the guest is:

$$\epsilon_{2k}^{non}(\omega) = \epsilon_1'(\omega) + \epsilon_2' |E_{0k}|^2, \quad k = x, y, z. \quad (2)$$

where  $\epsilon_1'$  and  $\epsilon_2'$  are linear and nonlinear parts of guest nanoparticles effective dielectric constant respectively. Then, the nonlinear effective dielectric constant of host medium will be:

$$\epsilon_{1k}^{non}(\omega) = \epsilon_1(\omega) + \epsilon_2 |E_{0k}|^2, \quad k = x, y, z, \quad (3)$$

where  $\epsilon_1$  and  $\epsilon_2$  are linear and nonlinear parts of host nanoparticles effective dielectric constant respectively.  $E_{ik}$  is the component of guest's electric field and  $E_{0k}$  is the same one for the exerting field. Calculations and simplifications in the *x* direction show that

$$\bar{\chi}_{112}^{(3)} = \chi_{112}^{(3)} \cdot f \cdot \frac{|x_1'|^2 x_2'^2}{[1 + f(x_1' - 1)]^2 [1 + f(x_2' - 1)]^2}. \quad (4)$$

Experiments and calculations show that the nonlinear refractive index will increase after the addition of metallic nanoparticles and in conclusion the 2-photon absorption coefficient will increase too [14]. Thus, the figure of merit will not change due to the following equation.

$$FOM = \frac{|\chi^{(3)}|}{4\pi \text{Im}(\chi^{(3)})}. \quad (5)$$

In tables 1 and 2, the influence of the addition of different nanoparticles with different shapes is shown. They are for the case of small volume fractions (0.06 and 0.08) in a degenerate four-wave mixing at  $\lambda = 532nm$  [15- 17]

Table 1. Variations of figure of merit in silica glass after addition of different cylindrical metallic nanoparticles for a degenerate four-wave mixing at  $\lambda = 532nm$

Composite	<i>f</i>	Re $\bar{\chi}_{211}^{(3)}$ (esu)	Im $\bar{\chi}_{211}^{(3)}$ (esu)	$ \bar{\chi}_{211}^{(3)} $ (esu)	figure of merit
Silica & Au	0.08	-2.49×10 <sup>-7</sup>	4.10×10 <sup>-7</sup>	4.80×10 <sup>-7</sup>	0.93
Silica & Cu	0.08	-1.09×10 <sup>-8</sup>	1.33×10 <sup>-8</sup>	1.72×10 <sup>-8</sup>	0.03
Silica & Ag	0.08	4.93×10 <sup>-12</sup>	1.11×10 <sup>-13</sup>	4.93×10 <sup>-12</sup>	3.25
Silica & Au	0.06	-1.45×10 <sup>-7</sup>	2.72×10 <sup>-7</sup>	3.09×10 <sup>-7</sup>	0.90
Silica & Cu	0.06	-7.07×10 <sup>-9</sup>	9.09×10 <sup>-9</sup>	1.15×10 <sup>-8</sup>	1.08
Silica & Ag	0.06	3.37×10 <sup>-12</sup>	7.59×10 <sup>-14</sup>	3.37×10 <sup>-12</sup>	3.34

Table 2. Variations of figure of merit in silica glass after addition of different spherical metallic nanoparticles for a degenerate four-wave mixing at 532 nm wavelength

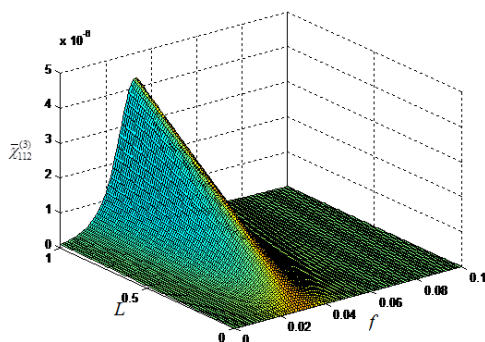
Composite	<i>f</i>	Re $\bar{\chi}_{211}^{(3)}$ (esu)	Im $\bar{\chi}_{211}^{(3)}$ (esu)	$ \bar{\chi}_{211}^{(3)} $ (esu)	figure of merit
Silica & Au	0.08	-2.71×10 <sup>-6</sup>	-2.05×10 <sup>-6</sup>	3.39×10 <sup>-6</sup>	0.32
Silica & Cu	0.08	-1.25×10 <sup>-7</sup>	2.45×10 <sup>-8</sup>	1.28×10 <sup>-7</sup>	0.16

Silica & Ag	0.08	$2.99 \times 10^{-11}$	$7.07 \times 10^{-13}$	$2.99 \times 10^{-11}$	3.69
Silica & Au	0.06	$-2.13 \times 10^{-6}$	$-1.29 \times 10^{-6}$	$2.49 \times 10^{-6}$	0.54
Silica & Cu	0.06	$-8.45 \times 10^{-8}$	$2.07 \times 10^{-8}$	$8.70 \times 10^{-8}$	0.35
Silica & Ag	0.06	$2.03 \times 10^{-11}$	$4.79 \times 10^{-13}$	$2.03 \times 10^{-11}$	3.8

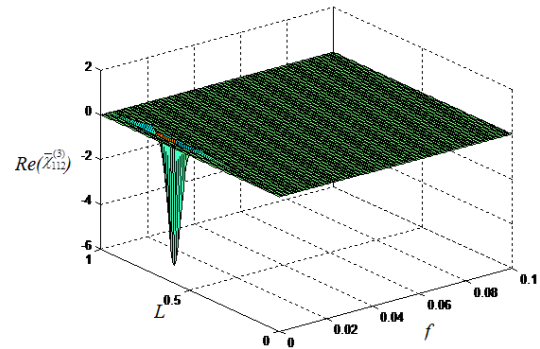
Figures 1 and 2 respectively show the variations of the magnitude of effective third-order nonlinear susceptibility coefficient  $\bar{\chi}_{112}^{(3)}$  after adding ellipsoidal nanoparticles of gold and silver for a degenerate four-wave mixing at  $\lambda = 532\text{nm}$  on the basis of  $L$  &  $f$ . Here, the values of effective third-order nonlinear susceptibility coefficient are expressed in terms of  $esu$ .

### Conclusions

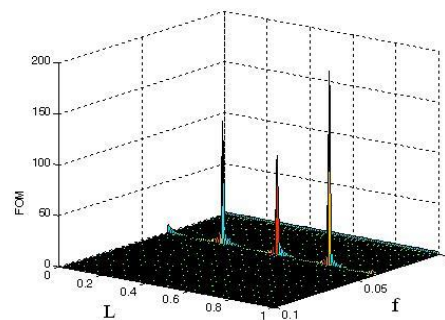
In this research, the physics of enhancement of nonlinear properties of silica glass MNPs has been investigated. Increasing the intensity of the external field causes an increase in the medium. The models presented in this paper describe the effects of the nanoparticles investigated for silica-gold, silica-silver and silica-copper, and it is shown that the ETNSC and FOM for spherical nanoparticles is larger than those of cylindrical ones. Considering the tables 1 and 2 and the information gathered, because of high values of nonlinear properties



**Fig. 1.** Variations of the effective third-order nonlinear susceptibility coefficient ( $\bar{\chi}_{112}^{(3)}$ ) after addition of gold ellipsoidal nanoparticles for a degenerate four-wave mixing at 532 nm wavelength on the basis of  $L$  and  $f$



**Fig. 2.** The Same as Fig. 1, but for the silver ellipsoidal nanoparticles



**Fig. 3.** Variations of the figure of merit after addition of silver ellipsoidal nanoparticles for a degenerate four-wave mixing at 532 nm wavelength on the basis of  $L$  and  $f$

### References

- [1] I.D. Rukhlenko, W. Zhu, M. Premaratne, G. P. Agrawal, Effective third-order susceptibility of silicon-nanocrystal-doped silica, *Opt. Express*, 20(24) (2012) 26275-26284
- [2] C.Z. Fan, J.P. Huang, K.W. Yu, Third-Order Nonlinear Optical Susceptibilities of Colloidal Crystals, *J. Phys. Chem. C*, 113 (2009) 18504-18514.
- [3] I. Webman, J. Jortner, M.H. Cohen, Optical properties of amorphous semiconductors, *Phys. Rev. B* 15 (1977) 5712-5717.
- [4] W. Lamb, D.M. Wood, N.W. Ashcroft, DNA-linked metal nanosphere materials: Fourier-transform solutions for the optical response, *Phys. Rev. B* 21 (1980) 2248- 2266.
- [5] G.S. Agarwal, R. Inguva, Effective-medium theory of a heterogeneous medium with individual grains having a nonlocal dielectric function, *Phys. Rev. B* 30 (1984) 6108 -6117.
- [6] S. Sharma, D. Mohan, S. Ghoshal, Measurement of nonlinear properties and optical limiting ability of Rhodamine6G doped

- silica and polymeric samples, *Optics Communications*, 28 (2008) 2923-2929.
- [7] D.A.G. Bruggeman, Berechnung verschiedener physikalischer Konstanten von heterogenen Substanzen, *Ann. Phys.* 24 (1935) 636-679.
- [8] A. Zakery, H. Shahmirzaee, The effect of graded nonmetal particles and their shape on the enhancement of nonlinear optical properties of oxide glasses, *J. Phys. D. Appl. Phys.* 41(22) (2008) 225106(1-7).
- [9] A. Zakery, H. Shahmirzaee Modeling of enhancement of nonlinearity in oxide and chalcogenide glasses by introduction of nanometals, *Phys. Lett. A* 361 (2007) 484-487.
- [10] N. Boris, A. Vitaly, G. Nikolia, Determination of the size, concentration and refractive index of silica nanoparticles from turbidity spectra, *Langmuir* 24(16) (2008) 8964-8970.
- [11] S. Jafari, H. Shahmirzaee, A. Gharaati, Modeling of Enhancement of FIGURE OF MERIT of silica glass by introducing core-shell nanometals, *J. Nonlinear Optic. Phys. Mat.* 21 (2012) 1250020(1-23).
- [12] A. Zakery, H. Shahmirzaee, Modeling of enhancement of nonlinearity in oxide and chalcogenide glasses by introduction of nanometals, *Phys. Lett. A*, 361(6) (2007) 484-487.
- [13] G.S. Agarwal, S.D. Gupta, T-matrix method to the nonlinear susceptibilities of heterogeneous media, *Phys. Rev. A.*, 38(11) (1988) 5678-5687.
- [14] A. Zakery, H. Shahmirzaee, Modeling of enhancement of nonlinearity in oxide and chalcogenide glasses by introduction of nanometals, *Phys. Lett. A*, 361(6) (2007) 484-487.
- [15] Y. Takeda, V.T. Gritsyna, N. Umeda, C.G. Lee, N. Kishimoto, Linear and nonlinear optical properties of Cu nanoparticles fabricated by high-current Cu-implantation in silica glass, *Nucl. Instr. Meth. B* 148 (1999) 1029-1033.
- [16] D.D. Smith, Y. Yoon, R.W. Boyd, J.K. Campbell, L.A. Baker, R.M. Crooks, M. George, Z-scan measurement of the nonlinear absorption of a thin gold film, *J. Appl. Phys.* 86(1999) 6200-6205.
- [17] G.S. Agarwal, S.D. Gupta, T-matrix method to the nonlinear susceptibilities of heterogeneous media, *Phys. Rev. A.*, 38(11) (1988) 5678-5687.