Volume : 4 | Issue : 10 | Special Issue Oct- 2015 • ISSN No 2277 - 8179

NONLINEAR OPTICAL PROPERTIES OF SIZE DEPENDENT ZnSe NANOPARTICLES USING Z-SCAN TECHNIQUE

Physics

KEYWORDS: Nonlinear refraction, Three photon absorption, Four photon absorption

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ABSTRACT

Nonlinear optical properties of different sizes of ZnSe nanoparticles are investigated. The sizes were smaller as well as larger than the Bohr radius. Nonlinear absorption and refraction coefficients were measured with 1064 nm wavelength and 100 fs laser pulses at 10 KHz repetition rate with Nd: YAG laser pulses using an open aperture and closed aperture Z-scan set up. Nonlinear refraction (n2) coefficients were estimated for different sized particles. All the particles show positive nonlinearity. The investigated samples with 5 nm and 10 nm sizes show four photon absorption (4PA) and 30 nm and 400 nm show three photon absorption (3PA).

INTRODUCTION

In recent years the study of nonlinear optical properties of semiconductor quantum dots or nanoparticles with dimension close to the Bohr radius shows eminent opportunities due to their direct band gap and control on the band gap over wide spectral range. The nanoparticles of ZnSe as semiconductor are important materials for use in nonlinear optics. These nanoparticles of ZnSe are synthesized by injecting diethylzinc and trioctylphosphine (TOP) selenide heating at different temperature in hexadecylamine (HDA) [1]. ZnSe nanoparticles smaller than the Bohr radius demonstrate unique optical properties which are due to the effect of three dimensional quantum confinements. Large confinement potential for the smaller crystal sizes leads to broadening in the band gap which is inversely proportional to the particle size. The two factors mainly affect the nonlinear properties of nanoparticles namely, a very high surface to volume ratio and the quantum mechanical effects. Three photon absorption (3PA) and four photon absorption (4PA) are particularly interesting as IR photons get converted to a blue or UV region. There is a lack of direct measurement on the 3PA and 4PA spectra of wide gap semiconductor. In the present study a systematic investigation of optical properties of ZnSe nanoparticles those are smaller as well as larger than the Bohr radius have been measured using open and closed aperture Z-scan technique with Nd:YAG laser at 1064 nm. The four different sizes of ZnSe nanoparticles investigated are 5nm, 10nm, 30 nm and 400 nm [1-5].

SYNTESIS AND CHARACTERIZATION

In this paper we report a systematic investigation into both 3PA and 4PA in ZnSe nanoparticles with change in size of the particles. The ZnSe nanoparticles are prepared through injecting diethylzinc and trioctylphosphine (TOP) selenide when heated at different temperature in hexadecylamine (HDA) at 110 o C for 60 minute then 5 nm size is obtained (at zero reflux times). When it is taken over different reflux times of 30 minutes, 60 minutes and 120 minutes, we obtained particles of 10 nm, 30 nm and 400 nm respectively. These particles are then isolated by standard procedure. The nonlinear absorption and nonlinear refraction properties of synthesized particles are studied at 1064 nm wavelengths, 100 femtosecond pulse width Nd: YAG laser at a repetition rate of 10 KHz using the standard Z-scan technique with maximum pulses energy of 30 mJ [4]. The sample is moved along the axial direction of the focused beam in such a way that it moves away from the lens passing through the focal point. At the focal point the sample experiences maximum pump intensity, which decreases gradually in either direction from the focus. Now, the aperture is introduced in front of the detector to record closed aperture Z-scan curve and obtain the nonlinear refraction coefficients.

RESULTS AND DISCUSSION

The nonlinear absorption measurements were performed using a hybrid mode locked Nd:YAG laser. Multiphoton absorption coefficients are calculated by the relations given by Sutherland et al. [6].

$$\frac{dI}{dz} = -\alpha_n I^n$$

$$T_{OA(nPA)} = \frac{1}{\left[1+(n-1)\alpha_n L\left(I_{oo}/\left(1+(Z/Z_o)^2\right)\right)^{n-1}\right]^{1/n-1}} \quad (1)$$

$$T_{OA(2PA)} = \frac{1}{\left[1+\alpha_2 L_{eff}\left(I_{oo}/\left(1+(Z/Z_o)^2\right)\right)\right]} \quad (2)$$

$$T_{OA(3PA)} = \frac{1}{\left[1+2\alpha_3 L'_{eff}\left(I_{oo}/\left(1+(Z/Z_o)^2\right)\right)^2\right]^{1/2}} \quad (3)$$

$$T_{OA(4PA)} = \frac{1}{\left[1+3\alpha_4 L''_{eff}\left(I_{oo}/\left(1+(Z/Z_o)^2\right)\right)^3\right]^{1/3}} \quad (4)$$

Where, Ioo is the peak intensity, λ **is the laser wavelength, z is the sa**mple position, zo = $\pi\omega$o2 / λ is Rayleigh range; ωo is the beam waist at the focal point (Z = 0),

$$L_{eff} = \left[1-\exp(-\alpha_o L)\right]/\alpha_o$$

$$L'_{eff} = \left[1-\exp(-2\alpha_o L)\right]/2\alpha_o \text{ and}$$

$$L''_{eff} = \left[1-\exp(-3\alpha_o L)\right]/3\alpha_o \text{ are the effective path}$$

lengths in the sample of length L for 2PA, 3PA and 4PA respectively [7].

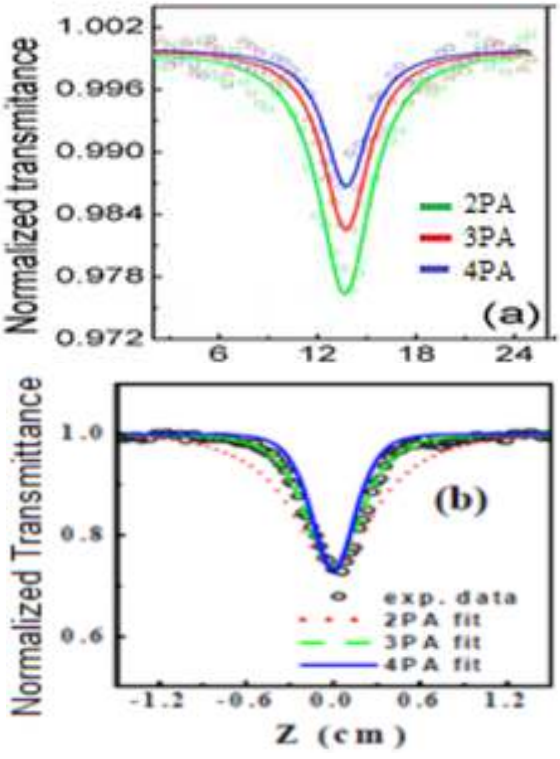

Figure 1(a): Open aperture Z-scan curve for 5 nm ZnSe nanoparticles (b): Open aperture Z-scan curve for 400 nm ZnSe nanoparticles.

By fitting these curves, with equations (2), (3) and (4), 2PA, 3PA and 4PA coefficients were estimated. From the graph shown in figure 2 it is found that the nonlinearity is negative which is estimated by closed aperture arrangement. The closed aperture normalized transmittance (TCA) is obtained by equation.

$$T_{CA} = 1 - \frac{4\Delta\varphi_o (z/z_o)}{[1+(z/z_o)^2][9+(z/z_o)^2]} \qquad (5)$$

Where, $\Delta\varphi$o is the phase change of the laser beam due to nonlinear refraction. The value $\Delta\varphi$o is estimated for all the sizes of the ZnSe nanoparticles by fitting the experimental data.

$$n_2(cm^2W^{-1}) = \frac{|\Delta\varphi_o|\lambda}{2\pi I_{oo} L_{eff}} \qquad (6)$$

$$n_2(esu) = 10^{-4}\frac{cn_o}{40\pi} n_2(cm^2W^{-1}) \qquad (7)$$

$$n_o = \sqrt{1+\frac{\varepsilon_{bulk}-1}{1+(0.75/D)^{1.2}}} \qquad (8)$$

Where, D is the diameter of nanoparticles, ε is the dielectric constant of bulk ZnSe and C is the velocity of light [8]. The nonlinear refractive index (n2) and linear refractive index (no) are estimated from equation 7 and 8 and the phase change of the laser beam due to nonlinear refraction ($\Delta\varphi$o) is less than one for all the closed aperture Z-scan curves.

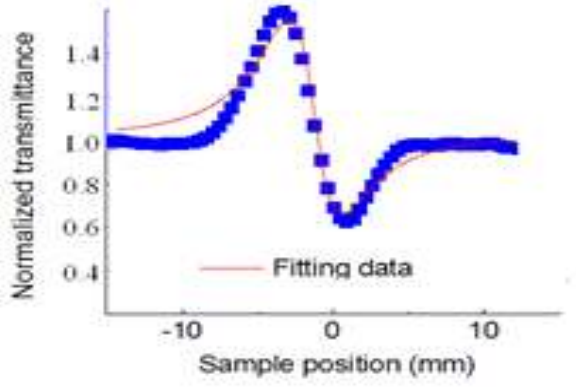

Figure 2: Closed aperture Z- scan curves for 5 nm ZnSe nanoparticles.

Table1: Estimation of 3PA, 4PA, linear and nonlinear refractive index coefficients of the ZnS nanoparticles.

Size of ZnSe nanoparticles (nm)	Three photon absorption coefficient α 3 × 105 (cm3/GW2)	Four photon absorption coefficient α 4 × 105 (cm5/GW3)	Linear refractive index (no)	Non-Linear refractive index n2 ×1013 (esu)
5	-	2.1	2.36	3.5
10	-	1.1	2.41	3.3
30	6.7	-	2.44	2.7
400	8.7	-	2.46	1.1

CONCLUSIONS

The temperature dependent ZnSe nanoparticles of four different sizes have been synthesized through chemical route. Nonlinear absorption and refraction coefficient have been calculated using Z-scan technique. The Observed result indicate 4PA for 5 nm and 10 nm and 3PA for the 30 nm and 400 nm particles. Nonlinear refraction coefficients are estimated for different sized particles. This study is important in identifying the best nonlinear material that can be used for the development of optoelectronics and photonics devices.

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