



# Nanosecond high-order nonlinear optical effects in wide band gap silver sulfide nanoparticles colloids



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## ABSTRACT

In this paper, we report the magnitude and the sign of optical nonlinearities of silver sulphide nanoparticles ( $\text{Ag}_2\text{S}$  NPs) dispersed in the dimethyl sulfoxide (DMSO) under exposure to 532 nm, 15 nanosecond (ns) pulsed laser irradiation. Large enhancement of two-photon absorption effect is observed with increase in concentration of the  $\text{Ag}_2\text{S}$  NPs in the DMSO. The closed-aperture Z-scan data with ns pulsed laser shows a positive nonlinear refraction which is in agreement with the two-band model of Sheik-Bahae. This reveals that the bound electronic nonlinearity (third-order nonlinearity) is the dominant mechanism causing nonlinear refraction in the colloids. The presence of a fifth-order nonlinear refraction is also observed and analyzed. The concentration dependence of the optical limiting is also investigated. The optical limiting threshold and the clamping energy decrease with increasing the  $\text{Ag}_2\text{S}$  NPs in the DMSO. The nonlinear optical performance of the  $\text{Ag}_2\text{S}$  NPs colloids in ns time regime in comparison with the reported continuous-wave (CW) nonlinear optical results is discussed. The results show that  $\text{Ag}_2\text{S}$  NPs colloid could be a very promising medium for nonlinear photonics devices in both time regimes.

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## 1. Introduction

With the wide-scale use of CW lasers and short laser pulses in the nano-, pico- and femto-second time domains for their prospective applications in many different areas, there arises a greater need for materials offer effective protection from lasers of known wavelengths in different power regimes [1–8]. Some material devices can make a response to long laser pulses and CW but are not effective against short laser pulses and vice versa. So it is of great interest to find a material presenting optical nonlinearities for applications in both long and short laser pulses. Among the materials, semiconductor nanoparticles have excellent and interesting applications in electronics, photonics and nonlinear optics [1–10]. The widening of the band gap energy for semiconductors can dramatically change their electronic structure and optoelectronic properties when their size is reduced to the nanoscale [11,12]. With increasing the band gap of semiconductor nanoparticles, higher order nonlinearities can be strongly enhanced in comparison to that of the bulk [9,13,14]. For this reason, any new semiconductor nanoparticles fabricated are examined under different conditions with laser pulses of various energies, wavelengths and durations

to understand the nonlinear physical mechanisms and distinguish their contribution for finding potential materials for applications such as optical limiting [1–8], all-optical switching [15,16] and optical imaging [17]. Semiconductor nanoparticles have presented good optical limiting performances due to the nonlinear absorption, nonlinear refraction, nonlinear scattering and thermal nonlinear refraction [1–8,18–21].

Wide band gap (WBG) semiconductor nanostructures are often used in optoelectronics, blue-ultraviolet light emitter, detectors and devices in which high-switching speed, high-voltage and high-temperature operation are needed and allow them to be more energy efficient, more powerful, faster and smaller [22,23]. Several wide band gap semiconductors such as CdS, ZnO,  $\text{Ag}_2\text{S}$  and GaN nanostructures were reported as good materials for nonlinear optical and photonic devices [22,24–27]. Wide band gap silver sulfide nanostructures have been studied as applicable to the development of different devices duo to their interesting properties [27–30]. As shown by Karimzadeh et al. the silver sulfide nanoparticles colloids can be used as a low power optical limiter because they have exhibited large third-order nonlinearity using CW lasers at 532 nm [27].

In this work, WBG  $\text{Ag}_2\text{S}$  NPs colloids at different concentrations of silver sulfide nanoparticles are synthesized by ns pulsed laser ablation of a silver plate in the DMSO. We use Z-scan method to measure the nonlinear optical coefficients of the  $\text{Ag}_2\text{S}$  NPs colloids

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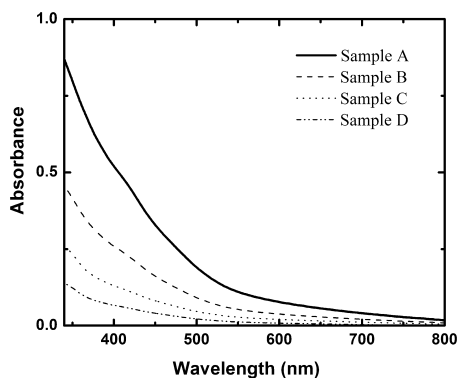


Fig. 1. Absorption spectra of the  $\text{Ag}_2\text{S}$  NPs colloids at different concentrations.

using ns pulsed laser irradiation at 532 nm. The open- and closed-aperture Z-scan behaviors of the samples are investigated based on the local nonlinear responses including two-photon absorption, third- and fifth-order nonlinear refraction. The results obtained from the experimental Z-scan measurements are compared with the theoretical models. The concentration dependence of the optical limiting is also reported. Finally, the origin of the nonlinear optical properties of the  $\text{Ag}_2\text{S}$  NPs colloids in CW and ns time regimes is briefly discussed.

## 2. Materials and methods

Silver sulfide nanoparticles have been prepared by nanosecond pulsed laser ablation of highly pure silver target in the DMSO. The detailed of experimental procedure has been given in references [31–33]. The  $\text{Ag}_2\text{S}$  NPs colloids were denoted by A, B, C and D with the silver sulfide nanoparticle concentration of  $0.45 \times 10^{-4}$ ,  $0.87 \times 10^{-4}$ ,  $1.70 \times 10^{-4}$  and  $3.51 \times 10^{-4}$  mol/L, respectively. The prepared  $\text{Ag}_2\text{S}$  NPs colloids were studied using transmission electron microscopy (TEM), X-ray diffraction (XRD) and a UV–vis optical absorption spectrophotometer [27,33]. The average  $\text{Ag}_2\text{S}$  nanoparticle radius was found to be about 4.5 nm. The X-ray diffraction pattern of the  $\text{Ag}_2\text{S}$  NPs showed highly crystalline structures with well-resolved diffraction peaks which were indexed as an acanthite phase of  $\text{Ag}_2\text{S}$  (JCPDS 140072)[33].

A CW low power (100 mW) diode-pumped Nd:YVO<sub>4</sub> laser operating at a wavelength of 532 nm was also used to measure the linear absorption coefficient of the  $\text{Ag}_2\text{S}$  NPs colloids. The nonlinear optical properties of the  $\text{Ag}_2\text{S}$  NPs colloids were studied by the Z-scan measurements using a 15-ns (FWHM) laser pulses irradiation at a wavelength of 532 nm. A similar optical geometry as given in Ref. [32] was used for nonlinear optical measurement. The beam was focused onto the samples (10 mm cell) by using a lens with 50 cm focal length. The spot size in the focal region was  $70 \mu\text{m}$  ( $\text{HW}1/e^2\text{M}$ ).

## 3. Results and discussion

The UV–vis absorption spectra of the  $\text{Ag}_2\text{S}$  NPs synthesized in the DMSO at different concentrations of silver sulfide nanocrystals are shown in Fig. 1. The energy and the type of transition for the band gap energy of the colloid were determined by analyzing the UV–vis absorption spectra of the  $\text{Ag}_2\text{S}$  NPs using the band theory of semiconductors. The absorption coefficient for direct transition is written as [34]

$$\alpha = \frac{A(h\nu - E_g)^{1/2}}{h\nu} \quad (1)$$

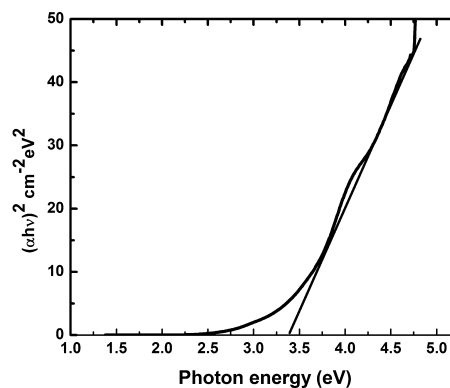


Fig. 2. The solid curve shows the plot of the  $(\alpha h\nu)^2$  of the  $\text{Ag}_2\text{S}$  NPs and the solid line is the extrapolating of the linear part of the plot.

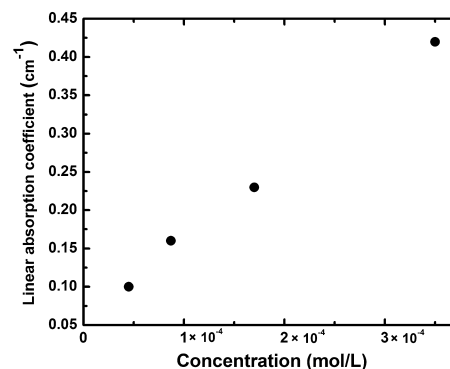


Fig. 3. The figure shows the behavior of the linear absorption coefficient as a function of the  $\text{Ag}_2\text{S}$  NPs concentration in the DMSO.

where  $A$  is the absorption constant,  $h$  is the Planck's constant,  $\nu$  is the frequency and  $E_g$  is band gap energy. The direct band gap energy is determined by extrapolating the linear part of the diagram of  $(\alpha h\nu)^2$  vs photon energy. We obtained the direct band gap energy to be about 3.38 eV for  $\text{Ag}_2\text{S}$  NPs as shown in Fig. 2.

Based on Beer–Lambert law, the linear absorption coefficients were measured using low power laser at 532 nm [32,33]. In Fig. 3, the extracted values of the linear absorption coefficients for the colloids are presented versus concentration. As shown in this figure, linear absorption coefficient increases linearly with the concentration of  $\text{Ag}_2\text{S}$  NPs.

The open- and closed-aperture Z-scan experiments were performed with the nanosecond laser at wavelength of 532 nm. Fig. 4 shows the ns open-aperture Z-scan measurement for the  $\text{Ag}_2\text{S}$

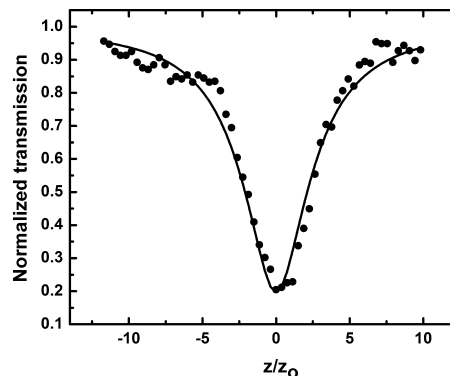


Fig. 4. The figure shows open-aperture Z-scan measurement of the sample A, using ns pulsed laser at 532 nm. The solid curve is the theoretical fit for the two-photon absorption process.

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