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Z-scan and four-wave mixing characterization of semiconductor cadmium chalcogenide nanomaterials

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Abstract. The possible physical origin of third-order nonlinearity of cadmium chalcogenide (Te, Se, and S) semiconductor nanocrystals were discussed based on the results of both Z-scan and degenerate four-wave mixing spectroscopies at 532, 775, 800, and 1064 nm in nanosecond, picosecond, and femtosecond time scale for nonlinear photonic applications.

1. Introduction

Nanomaterials have drawn great attention because of their distinguishable linear and nonlinear optical properties, which differ remarkably from bulk materials. Nanomaterials with a diameter less than 10 nm are composed of only several hundred atoms. The optical properties are strongly affected because of the quantum confinement of electrical carriers within nanoparticles, the huge local electric field enhancement, and the significantly enhanced surface and interface effects. By controlling the dimensions and the chemistry of their surfaces, both linear and nonlinear optical properties of the nanomaterials can be finely tailored. Design and characterization of nanomaterials have attracted a great deal of attention in several efforts to discover and develop materials possessing large nonlinearities and satisfying various technological requirements for photonic device applications, such as optical switching, optical limiting, and optical communication. For example, ideal nonlinear optical materials for optical limiting applications should have a large nonlinearity, high figure of merit, wide dynamic range, and fast response time.

Many techniques have been developed to characterize the third-order nonlinearities of various materials. Popular spectroscopy techniques to estimate the third-order nonlinearities are Z-scan [1] and degenerate four-wave mixing (DFWM). The Z-scan measurement is completed by detecting the transmittance of a nonlinear medium through a finite aperture in the far field as a function of the sample position (Z) measured with respect to the focal plane. The open aperture transmittance (the aperture size is infinitely large) is used to measure the nonlinear absorption coefficient while the closed Z-scan is used to measure the nonlinear refractive index coefficient. Z-scan method is a simple technique with high accuracy for nonlinearity characterization. The most important advantage of Z-

scan is its indication of the sign and type of nonlinearity immediately after the measurement is finished. However, there are major disadvantages associated with this method. For example, the Z-scan curve will be distorted by the misalignment, linear scattering, sample imperfections, and intensity fluctuation during the measurement [1]. DFWM is also popularly used to measure the third-order nonlinear optical susceptibility of material. In DFWM experiment, the same part of the sample is irradiated by the laser beams. Thus the sample imperfection induced errors will be reduced significantly. Since the linear scattering does not contribute to the signal, DFWM is a better choice to characterize the nonlinearity of sample which possesses linear scattering.

In this work, the third-order nonlinearities of cadmium chalcogenide (Te, Se, and S) semiconductor nanocrystals was investigated with both Z-scan and DFWM methods in nanosecond (ns), picosecond (ps), and femtosecond (fs) time scale. The semiconductor nanocrystals were prepared by either ball milling or colloidal chemical reaction.

2. Experimental Results and Discussion

The three laser systems in our experiments were a fs laser (1 KHz, 150 fs Ti:Sapphire laser operating at 775 nm) for Z-scan, a ps laser (87 MHz, 1 ps Ti:Sapphire oscillator operating at 800 nm) for Z-scan, and a ns laser (10 Hz, 8 ns Nd:YAG laser running at both 532 nm and 1064 nm) for both Z-scan and DFWM spectroscopies.

The synthesis of the nanocrystals can be found in references [2] and [3]. Linear absorption spectra show that the absorption peaks for all of the samples are in the visible range. Thus, the nonlinearities of these samples at 775 nm, 800 nm, and 1064 nm are mainly contributed by two-photon absorption or pure electron cloud distortion. The obtained large nonlinearity as well as the high nonlinear figure of merit indicates our samples are excellent candidates for photonic device applications, such as optical limiting and optical switching.

The typical Z-scan results of the CdTe and CdSe nanocrystals are shown in figures. 1-3. The valley structure of the open Z-scan results shown in all three figures indicate two-photon absorption, while the peak-valley structures shown in figures 2 and 3 imply the negative nonlinear refractive index in both CdTe and CdSe nanocrystals. The two-photon absorption coefficient (β) and nonlinear refractive index coefficient (γ) are given the figures.

In both nanosecond and high repetition picosecond cases, large third-order nonlinearities of the nanomaterials are obtained. It is well known that accumulative effects, such as two-photon absorption induced thermal and density effects will enhance the third-order nonlinearity significantly in long pulse or high repetition rate short pulse cases.

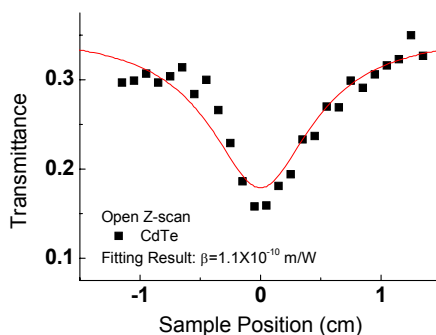


Fig. 1. Open Z-scan results for CdTe nanoparticles in PMMA. The fluctuation is due to the excitation laser power oscillation (Continuum, Surelite II, 1064 nm, 8 ns, and 10 Hz). The solid line is the best fitting to the data.

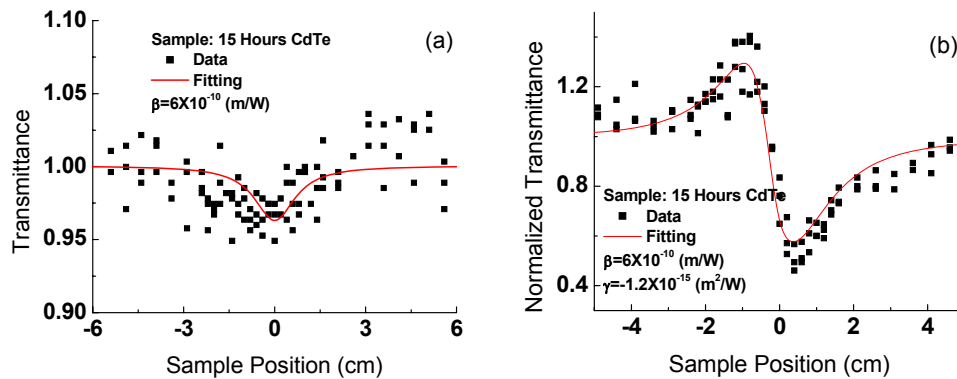


Fig. 2. Z-scan results of capped CdTe nanoparticles ball milling for 15 h suspended in hexane with a 1-ps laser excitation at 800 nm. The solid line is the result of the best fitting to the experimental data.

In the femtosecond scale, accumulative effects such as thermal and density effects are negligible for the 1 KHz repeat case. Therefore, the third-order nonlinearities of the nanoparticles in the femtosecond scale are 4-5 orders smaller than those in the nanosecond and high repetition picosecond cases, as shown in Fig. 4.

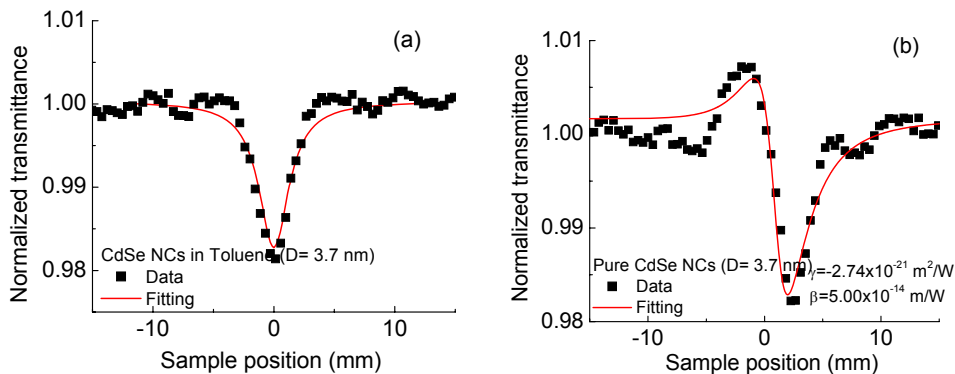


Fig. 3. Z-scan results of CdSe nanoparticles with a 150 fs, 1 KHz Ti:Sapphire laser running at 775 nm. The solid line is the result of the best fitting to the experimental data.

The resonant third-order nonlinearities of these nanomaterials were also investigated by degenerate four-wave mixing as shown in Fig. 5. The cubic dependence of the DFWM signal on the total input intensity indicates the third-order nonlinear optical process dominates the contribution to the DFWM signal [4]. Many different processes may contribute to the third-order nonlinearity of the material, such as saturable absorption, excited-state absorption, thermal effect, etc.

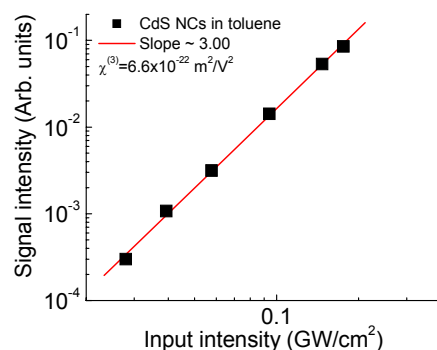


Fig. 4. Logarithmic plot of the DFWM signal in CdS nanocrystals at 532 nm as a function of the total pump intensity. The cubic dependence of the signal on the input intensity indicates the third-order nonlinearity.

In conclusion, the third-order nonlinearities of CdTe, CdSe, and CdS were measured at 532 nm, 775 nm, and 1064 nm by using ns, ps, and fs laser systems. It is found that the nonlinearity at the femtosecond scale is much smaller than those at the picosecond and nanosecond scale. This is attributed to the accumulative effects. In the femtosecond measurement, only fast processes, such as pure electron distortion, contribute to the nonlinearity. However, in the nanosecond and high repetition rate picosecond experiments, much slower processes, such as thermal effect, density effect, will also contribute or even dominate the nonlinearity. In order to further identify the physical origin of the third-order nonlinearities of these nanomaterials, especially in order to distinguish the contributions from fast electronic process and those from slow accumulative processes, the time-resolved four-wave mixing, pump-probe, and polarization-resolved four-wave mixing are being carried out in our laboratories. Other evidence, such as the excited-state lifetime and the ratio of different components of the third-order susceptibility are also being measured.

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