4P074 **Third-order nonlinear properties of CdTe nanoparticles by femtosecond Z-scan and four-wave mixing spectroscopy**

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Introduction: The determination of optical nonlinearities and their response time in semiconductor nanoparticals are of great important for possible applications of optical limiting devices and all-optical switching elements. It is known that third-order nonlinearities arising from bound-electronic effects and two-photon absorption (TPA) can be analyzed by Z-scan technique and / or four-wave mixing experiment. In the present study, we have examined newly synthesized CdTe nanoparticles by single beam Z-scan technique with femtosecond Ti:sapphire laser to measure the magnitude and the sign of the third-order nonlinear refractive index and two-photon absorption (TPA) cross section. Femtosecond time-resolved four-wave mixing (FWM) spectroscopy is also applied for CdTe nanoparticles to measure the third-order nonlinear properties. Crystal size and wavelength dependence on nonlinear properties will be discussed on the basis of these results.

Experiments: For Z-scan measurements, experimental set up is described in ref. [1]. The excitation wavelength is 800nm of amplified femtosecond Ti:Sapphire laser (100fs, 1kHz). For FWM experiments, sample was excited by fundamental wavelength (720 nm, 250fs FWHM) from a hybridly mode-locked, dispersion compensated femtosecond dye laser (Coherent Satori 774) and a dye amplifier (Continuum, RGA 60-10 and PTA 60) at an operating repetition rate of 10Hz. The excitation pulse was slit into two beams, which were crossed in the sample at an appropriate angle to produce an optical interference pattern. The induced transient grating was probed by a femtosecond supercontinuum obtained by focusing a portion of fundamental laser beam into a 1-cm H_2O cell. A computer-controlled translation stage was used to change the time delay between pump and probe pulse. By setting a $\lambda/2$ plate in one of the pump beam, the grating condition was adjusted. The polarization of the probe beam was changed for both intensity and polarization grating. The intensity fluctuation of the supercontinuum was compensated for splitting off a portion of the supercontinuum to form the reference beam.

Results and Discussion: Typical time resolved FWM signals with 720 nm as pump and 550 nm as probe show sharp rise as well as sharp decay without any slow components as shown in Fig. 1, the signal shapes are very similar even for the different size of CdTe nanoparticles and different pumping power. This is suggesting that CdTe nanoparticles are highly polarizable materials, and the signal is mainly due to the electronic polarization. $\chi^{(3)}$ value was determined by using the function given in the textbook [2]. According to our experimental results, $\chi^{(3)}$ values are almost independent on the size of the nanoparticles (diameter: 2.8 nm-4.9 nm). The power dependence on $\chi^{(3)}$ for CdTe nanoparticles with a size of 2.8 nm and 4.9 nm was shown in Fig. 2. $\chi^{(3)}$ was normalized with the signal of standard sample, CCl_4, measured in the same condition. $\chi^{(3)}$ is almost constant irrespective of excitation intensity within an experimental uncertainty. That means imaginary part of $\chi^{(3)}$, which is related with the two-photon absorption, is negligibly small as compared with the real part of $\chi^{(3)}$ in FWM experiments; even though the TPA induced fluorescence can be detected. As the same reason, the slow component related with the population grating induced by two-photon absorption cannot be detected in FWM experiments of intensity grating condition. As to observe the imaginary part of $\chi^{(3)}$, Z-scan experiments for determining the two-photon absorption cross section were carried at the excitation wavelength of 800 nm. Typical experimental data is shown in Fig. 3. The third order nonlinear parameters, γ, of CdTe nanocrystals are positive as well as the absorption is anti-saturated. Two-photon absorption increases linearly with increasing the excitation intensity, which means two-photon absorption corresponding to imaginary part of $\chi^{(3)}$ give great contributions in Z-scan measurement. Further increase of the excitation intensity shows nonlinear behavior of two-photon absorption, suggesting the contribution of excited-state absorption.

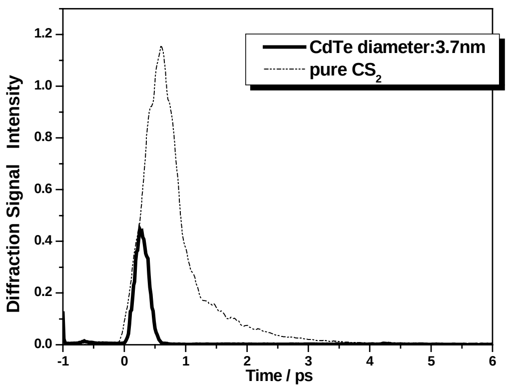

Fig. 1 Typical time resolved FWM signal of CdTe nanoparticles in solution and standard sample CS_2 detected with polarization grating condition at 550nm.

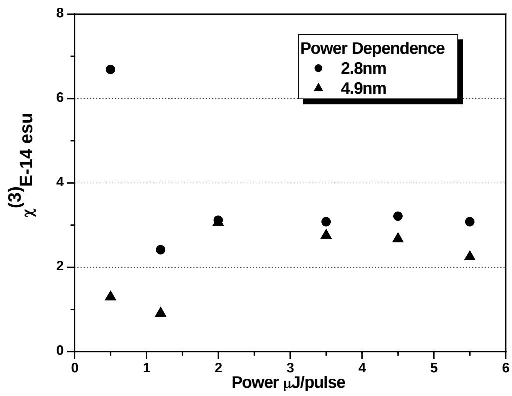

Fig. 2 Time resolved FWM signal of CdTe nanoparticles in solution at 550nm as a function of excitation intensity.

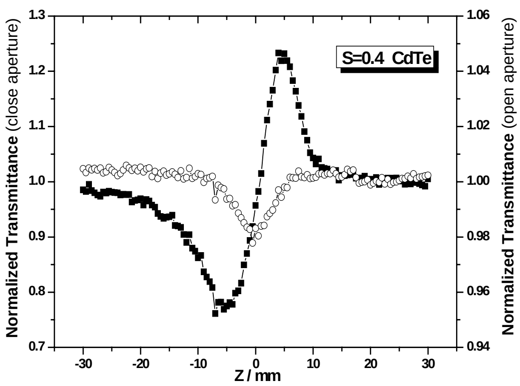

Fig. 3 Typical Z-scan signal at 800nm. Open circles are corresponding to open aperture related to nonlinear absorption; close squares corresponding to close aperture related to both index change and nonlinear absorption.

Reference:

[1] P. Audebert, K. Kamada, K. Matsunaga, K. Ohta, *Chem. Phys. Lett.* 367(2003)62-71

[2] Paras N. Prasad, David J. Williams, *Introduction to nonlinear optical effects in molecules & polymers*. Wiley-Interscience (1990)