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Influence of free carrier refraction due to linear absorption on Z-scan study of porous Si

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Abstract

We analyze the limitations imposed by free carrier refraction induced by the sample linear absorption in determination of nonlinear refractive index in porous silicon by the Z-scan technique. By simulation we show that for picosecond laser pulses there is a strong contribution to the Z-scan signal due to free carriers generated by linear absorption even in highly transmitting sample. By simulating various experimental conditions we show that for femtosecond pulses the contribution due to bound electronic nonlinearity is significant for samples with small two-photon absorption coefficient.

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The characterization of nonlinear optical properties of materials is important both from fundamental physics and device point of view. In particular, great effort has been devoted to measure the intrinsic third order nonlinearity (nonlinear refractive index and nonlinear absorption coefficients). Large intrinsic third order nonlinearity is important for optical switching device due to its ultrafast response. A wide range of techniques have been used to measure third order nonlinearity: e.g. Z-scan [1], degenerate four wave mixing [2], nonlinear interferometry [3], optical third harmonic generation (THG) [4], and frequency resolved optical gating [5].

The Z-scan technique is widely used due to its simplicity both in the experimental set up and data analysis. The technique provides sensitive and straightforward determination of the sign and the values of nonlinear refractive index (γ), two-photon absorption (TPA) coefficient (β), and the free carrier refraction coefficient (σ). The free carrier refraction is due to the electron–hole pairs

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generated by combination of linear or nonlinear absorption process. The decay time of free carrier nonlinear refraction is limited by the lifetime of the carriers. In general, for the transparency region of semiconductor the carriers are generated by TPA. The Z-scan technique has been applied in the transparency region to measure the nonlinear optical coefficients of bulk semiconductors [6] and quantum confined structures such as semiconductor-doped glasses (SDGs) containing nanocrystals of CdS_xSe_{1-x} [7–9]. The technique has been demonstrated to work successfully for bulk semiconductors for determination of γ, β and σ [6]. However, for dilute systems such as SDGs as well as any other material with very low TPA coefficient the Z-scan technique poses serious limitation [8] in the interpretation of the experimental results in distinguishing the bound electronic nonlinearity and the free carrier nonlinearity due to carriers generated by TPA particularly in case of excitation with picosecond pulses. For systems with low TPA coefficient as shown in [8] the refractive index change per unit intensity saturates in Z-scan experiment at low value ~0.2 of peak-valley transmission difference and hence an erroneous interpretation about the lack of free carrier nonlinearity is drawn.

The nonlinear optical properties of bulk Si has been extensively studied using pico-second and nanosecond pulses of Nd:YAG laser [10,11]. Bulk Si crystal with an indirect band gap of 1.11 eV absorbs photons of Nd:YAG laser of energy 1.17 eV with the absorption coefficient ranging from 10 to 100 cm^{-1} depending on the doping concentration [12,13]. The carriers are generated by the linear absorption. The mechanism of nonlinearity in bulk Si is thus attributed to carriers generated by linear absorption [11,14,15]. Recently, Z-scan technique has been applied to measure nonlinear optical coefficients of self-suspended p-type porous Si of 75% porosity and 20 μm thickness at the laser wavelength of 1064 nm using pico-second pulses [16]. The band gap of the porous Si shifts to about 2 eV due to quantum confinement [16]. The laser excitation wavelength 1064 nm is far from the peak luminescence wavelength at 630 nm and the sample is also highly transmitting at the laser excitation wavelength. Thus the nonlinearity in the porous Si was claimed to be of bound electronic type. However it may be noted that in this porous Si sample the linear absorption coefficient is quite large of ~10 cm^{-1} [17] and the sample appears transparent only because of small thickness. The calculated transmission for sample of thickness 20 μm with $\alpha = 10\ cm^{-1}$ is 98%. Although the sample is highly transparent, due to significant value of linear absorption coefficient substantial number of free carriers are generated and hence large free carrier nonlinearity is possible. However, Lettieri et al. [16] neglected the effect of nonlinear refraction due to free carriers generated by linear absorption. In this paper, we show that the Z-scan data in these sample of porous Si at similar intensities have a very strong contribution from the free carrier nonlinearity due to linear absorption and the experimental Z-scan data reported in [16] can be explained on the basis of this process alone rather than due to bound electronic nonlinearity. By simulating various experimental conditions, we also examined the contribution of free carrier nonlinearity and the bound electronic nonlinearity to the total nonlinear refraction signal for picosecond and femtosecond laser pulses.

To simulate the Z-scan results, we follow the procedure as described in [8]. However the intensity dependent change of refractive index (Δn) would be modified to include the contribution from the carriers generated by linear absorption. $\Delta n = \gamma I + \sigma N$, with the two terms representing the contribution due to bound electrons and the electron hole pairs generated by absorption of the laser pulse. N is the carrier density generated by the linear absorption and by the TPA. Thus the contribution to free carrier nonlinearity would be due to both the carriers generated by linear as well as TPA. For simulation, the linear transmission through the aperture, S, was kept at 2%. The spot size of the beam at the focus is taken to be 52 μm at the laser wavelength of 1064 nm with 40 ps (FWHM) laser pulses. The sample thickness and the linear absorption are taken as 20 μm and 10 cm^{-1}, respectively.

The details of calculations for estimation of nonlinear coefficients for the porous Si are given in the later part of the paper. The theoretically estimated nonlinear optical coefficients for porous Si