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Characterization of signs change of nonlinear refraction in ZnSe based on a modified double 4*f* imaging system with a phase object

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ABSTRACT

Signs change of nonlinear refractive index in ZnSe is observed by employing a modified double 4*f* imaging system at the wavelength of 800 nm using picosecond pulses with different pulse energies. This process results from the competition of the bound electronic nonlinear refraction and the free carrier refraction. At low intensity, positive nonlinear refraction is obtained, which is attributed to bound electrons. As the increase of laser beam intensity, the nonlinear refractive index become small, and changes to negative. This is ascribed to free carriers generated by two-photon absorption. Additionally, the nonlinear refractive index of bound electron and the refractive index change of free carrier are determined unambiguously by a simple method.

1. Introduction

The optical nonlinearities of semiconducting materials have been investigated extensively because of its potential applications in various optical devices, such as optical limiter, and all-optical switcher [1,2]. Picosecond time-resolved degenerate four-wave mixing experiments at much lower irradiance levels performed on semiconductors in the presence of two-photon absorption (TPA) showed a fast and large third-order nonlinearity that is due to bound electrons (Kerr effect), and a high-order nonlinearity, which is attributed to free charge carriers generated by TPA [3]. This third- and fifth-order nonlinearity of semiconductor-doped glasses is also reported and distinguished by using different pulse durations [4]. That is to say there are two nonlinear effects dominates for semiconducting materials. Therefore, characterization of the two nonlinear mechanisms clearly is necessary for optical devices based on nonlinearities of semiconductors.

As reported in Ref. [4], free carrier nonlinearity of semiconductors depends on the fluence of the laser pulse. So it is hard to observe the contribution of free carrier to the nonlinear refraction using femtosecond pulses at 800 nm. Although the nonlinear refraction of free carrier has been observed by time-resolved *Z*-scan in ZnSe with 200 fs pulses at 790 nm [5], the competition between the two effects was not observed directly. Here, we will study the nonlinear refraction of ZnSe, including third- and fifth-order effects, using picosecond pulses at 800 nm, and distinguish these two effects in quantity using a modified double 4*f* coherent imaging system. This technique improved from the 4*f* coherent imaging system [6] is powerful for poor output beam.

2. Theoretical model

The experimental setup is shown in Fig. 1. The extended beam (the extender system does not shown in Fig. 1) illuminates on an aperture with a phase object (PO). Then, the beam is separated into two beams: an intense beam and a much weaker one. The intense arm of the optical setup is a 4*f* nonlinear imaging system using for measurement of nonlinearities. The other arm is also a 4*f* system, which is used to monitor the spatial distribution of the laser beam. This is the most prominent difference between our experimental setup and that in Ref. [6]. A CCD camera (LAVISION) with 1040×1376 pixels and 4095 gray levels located at the rear focal plane of L_2 is used to record the images. The size of each pixel is $6.4 \times 6.4\ \mu m^2$.

The aperture with PO (shown in Fig. 1b) is illuminated by a linear polarized plane wave field $E = E_0(t)\exp[-i(\omega t - kz)] + c.c.$, where ω is the angular frequency, k is the wave vector in free space, and $E_0(t)$ is the amplitude of the electric field containing the temporal envelop of the laser pulse. The slowly varying envelop approximation [7] is used to describe the propagation of the electric field in the nonlinear medium. Employing aperture with transmittance $t_p(x,y)$, the amplitude of the electric field behind this aperture is $O(x,y,t) = Et_p(x,y)$. And the field amplitude at the focal plane of the L_1 is spatial Fourier transform of $O(x,y,t)$:

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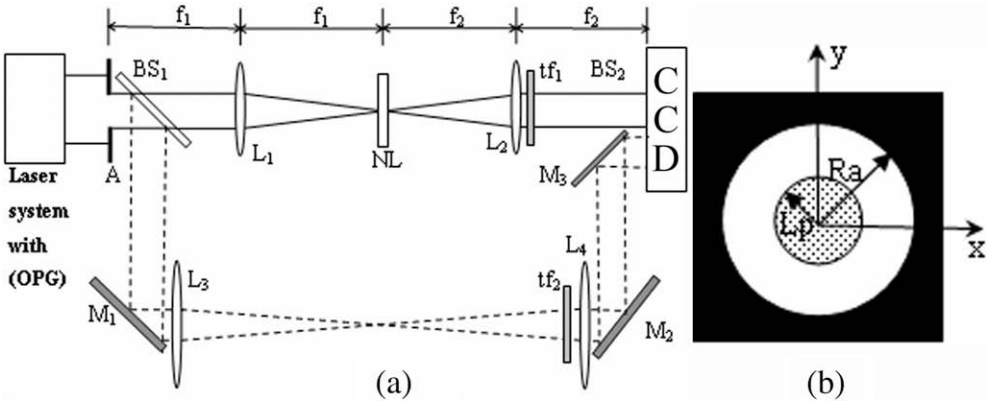

Fig. 1. (a) Schematic of the modified double 4*f* coherent imaging system. The nonlinear material is ocated at the focus plane of L_1. A, an aperture with a phase object; BS_1, BS_2, beam splitters; $L_1 - L_4$, convex lenses; tf_1, tf_2, neutral filters; $M_1 - M_3$, mirrors; CCD, charge-coupled device. (b) Schematic of the circular aperture with a phase object.

$$S(u,v) = \frac{1}{\lambda f_1} FT[O(x,y)] = \frac{1}{\lambda f_1} \iint O(x,y) \exp[-2\pi i(ux+vy)]dxdy, \tag{1}$$

where *FT* denotes the spatial Fourier transform operation, $u = \frac{x}{\lambda f_1}$ and $v = \frac{y}{\lambda f_1}$ are the spatial frequencies in the focal plane, λ is the wavelength of the laser pulse, and f_1 is the focal length of lens L_1.

The input electric field before the sample is $S(u,v)$, and I, the intensity of the laser beam, is proportion to $|S(u,v)|^2$. If the sample could be regarded as "thin", the field at the exit surface of the sample is given by [8]:

$$S_L(u,v,t) = S(u,v,t)\exp(-\alpha_0 L/2)[1+q(u,v,t)]^{(ikn_2^{eff}/\beta - 1/2)}, \tag{2}$$

where $q(u,v,t) = \beta I(u,v,t)L_{eff}$, $L_{eff} = [1-\exp(-\alpha_0 L)]/\alpha_0$, β is the nonlinear absorption coefficient, α_0 is the linear absorption coefficient, L is the thickness of the sample, and n_2^{eff} is the effective nonlinear refractive index. Specially, in our present work, both the third- and the fifth-order nonlinearities contribute to this effective nonlinear refractive index.

At the receiving plane of the CCD camera, the image intensity is expressed by:

$$I_{im}(x,y,t) = |U(x,y,t)|^2 = |FT^{-1}[S_L(u,v,t)H(u,v)]|^2, \tag{3}$$

where FT^{-1} denotes the inverse spatial Fourier transform, and $H(u,v) = circ[(u^2+v^2)^{1/2}\lambda G/N_A]$ is the coherent optical transfer function applicable to aberration-free lenses. The function $circ(\rho)$ is defined to be 1 if the radius $\rho(u,v) \leqslant 1$ and 0, otherwise. N_A is the numerical aperture of the lens L_1, and G is the magnification of the 4*f* nonlinear imaging system.

As the image recorded by the CCD camera is the fluence distribution of the laser beam, this image can be expressed by the integration of the intensity:

$$F(x,y) = \int_{-\infty}^{+\infty} I_{im}(x,y,t)dt. \tag{4}$$

Considering a top-hat beam experimental setup used in our present work, the transmittance of the aperture is defined as:

$$t_a(x,y) = circ\left[(x^2+y^2)^{1/2}/R_a\right], \tag{5}$$

where R_a is the radius of this aperture. As a PO of radius L_p ($L_p < R_a$) centered inside the top-hat beam with a uniform ϕ_L is added, the whole transmittance of the aperture can be given by:

$$t_p(x,y) = t_a(x,y)\exp\left\{i\phi_L circ\left[(x^2+y^2)^{1/2}/L_P\right]\right\}. \tag{6}$$

Using equations above, we can simulate experimental images recorded by the CCD camera to obtain optical parameters of materials. More details about the 4*f* coherent imaging system with a phase object have been reported in Refs. [6,9]. In our present work, lenses L_3 and L_4 constitute another 4*f* system. Through this improvement, we can not only monitor the energy fluctuation of the input laser beam, but also acquire the intensity distribution before the sample accurately.

3. Experiment

The excitation pulse is provided by a commercial optical parametric generator (OPG), which is pumped by the third harmonic generation from a *Q*-switched Nd: YAG (EKSPLA) laser delivering pulses at 355 nm. The tunable range of the OPG is from 420 to 680 nm and from 740 to 2300 nm. In our experiment, the laser pulse is set at the wavelength of 800 nm, and the pulse duration is 10 ps (FWHM). Because of the ultrashort laser pulse with a repetition rate of 1 Hz, thermal effect could be neglected in the following discussion. The radius of the aperture is $R_a = 1.7$ mm, $L_p = 0.5$ mm, $\phi_L = 0.266\pi$, the focal lengths of L_1 and L_2 are $f_1 = f_2 = 200$ mm giving magnification of $G = 1$, $f_3 = f_4 = 300$ mm are the focal lengths of L_3 and L_4, respectively, and the sample thickness is $L = 2$ mm. The Airy radius at the focal plane of the lens L_1 is $\omega_0 = 1.22\lambda f_1/(2R_a) = 57.4$ μm giving a Rayleigh range $z_0 = \pi\omega_0^2/\lambda = 12.9$ mm, which is much larger than the sample thickness, so the sample can be considered as 'thin' without doubt. The surface of the ZnSe crystal was polished and front surface reflection losses is considered.

In the experiment three images are needed, including linear image, nonlinear image and without material image. (i) *Linear image:* this image is obtained at low intensity condition by placing high-density neutral filters (tf) before the nonlinear material. (ii) *Nonlinear image:* by placing the same high-density neutral filters after the nonlinear material, the nonlinear image at high intensity is acquired. (iii) *Image without material:* it is acquired by removing the nonlinear material while leaving the neutral filters in the optical layout. Integrating all pixels of the linear image, transmitted energy of the laser pulse under linear condition E_l is obtained. Similarly, the transmitted energy of the laser pulse under nonlinear condition En_l and the input laser pulse energy E_i are acquired respectively. The liner transmittance of the material is determined by $T_l = E_l/E_i$, and the nonlinear transmittance of the material could be given by $T_{nl} = E_{nl}/E_i$. If the nonlinear transmittance is determined, the nonlinear absorption coefficient β can be deduced unambiguously. Then, the effective nonlinear refractive index n_2^{eff}, the only parameter not known in Eq. (2), can be deduced by fitting to the nonlinear image. At last, the third- and fifth-order nonlinearities could be determined in quantity by a simple method.

Since a commercial OPG is used as the beam output in this modified nonlinear-image technique, the energy fluctuation must be