Self-defocusing in CdSe induced by charge carriers created by two-photon absorption

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We observe self-defocusing of picosecond, 1.06-μm pulses in CdSe. The effective nonlinear refraction can be 2 orders of magnitude larger than that of CS_2. We obtain good agreement with the theory presented here, which assumes that the self-refraction is caused by charge carriers created by two-photon absorption.

Two-photon absorption of 1.06-μm picosecond pulses in several semiconductors, including CdSe, has been reported.[1] The refractive effects arising in a semiconductor from the charge carriers created by single-photon absorption and two-photon absorption have also been demonstrated in Si and InSb, respectively.[2] Here we report a study of nonlinear refraction arising from charge carriers generated by two-photon absorption of picosecond 1.06-μm radiation in CdSe. Self-defocusing of laser pulses of Gaussian spatial and temporal profiles incident upon a 2-mm-thick sample of CdSe was observed and studied as a function of the input irradiance and pulse width. Such self-defocusing has potential device applications in nonlinear optical energy (irradiance) limiting switches and integrated optics.[3-6]

The experimental configuration used is that of external self-action,[7] in which the thickness of the nonlinear medium is much smaller than the confocal parameter of the incident beam. In this case the Maxwell wave equation describing the propagation of the electric field E can be written as

$$-2ik\frac{\partial E}{\partial z} = i\omega\sigma\mu_0 E - \frac{\omega^2}{c^2}\chi^{(3)}\frac{|E|^2}{2}E, \qquad (1)$$

where $\chi^{(3)}$ denotes the third-order nonlinear susceptibility. By using the Drude formula the conductivity can be written as

$$\sigma = \alpha n_0\left(\frac{\epsilon_0}{\mu_0}\right)^{1/2} + i\frac{Ne^2}{m_{eh}\omega}. \qquad (2)$$

α denotes the linear absorption coefficient of the medium, n_0 is its refractive index, and N is the population density of the electron–hole pairs created in the medium. m_{eh} denotes the reduced effective mass of an electron–hole pair, and ω is the angular frequency of the incident radiation. By writing the electric field as

$$E = Ae^{i\phi}$$

with irradiance $I = (n_0\epsilon_0 c/2)A^2$, Eq. (1) can be reduced to

$$\partial I/\partial z = -\alpha I - \beta I^2, \qquad (3)$$

$$\partial\phi/\partial z = \beta_1 I - \gamma_1 N, \qquad (4)$$

where $\beta_1 = \omega\gamma/c$. γ is related to the nonlinear refractive index n_2 arising from the real part of $\chi^{(3)}$ through the relationship

$$n_2\,(\text{esu}) = \frac{cn_0}{40\pi}\gamma,$$

where the right-hand side of the equation is in MKS units. β is the two-photon absorption coefficient proportional to the imaginary part of $\chi^{(3)}$. Although for the material considered here (CdSe) the contribution of n_2 to the total nonlinear refraction is negligible, we include the term β_1 so that the theory can be applicable to materials for which n_2 is appreciable. Moreover,

$$\gamma_1 = \frac{\mu_0 e^2 c}{2n_0 m_{eh}\omega}p.$$

p is a parameter introduced here to account for the contributions to the nonlinear refraction that are proportional to N but that are not explained by the Drude model. An example of such a contribution is that arising from interband transitions.[8] While the refraction induced by the photogenerated carriers is large, the absorption of the incident radiation by these carriers has been ignored in Eq. (3). This is justified by our experiments, in which up to the maximum incident irradiance used ($\sim$2 GW/cm^2) no appreciable dependence of the total transmission on pulse width was found. For appreciable free-carrier absorption, a dependence on pulse width would be expected and has been observed for longer pulses. Ignoring the diffusion and recombination of the charge carriers during the picosecond pulses, the population density N is given by

$$\frac{\partial N}{\partial t} = \frac{\beta}{2\hbar\omega}I^2. \qquad (5)$$

We assume that the laser beam is Gaussian in spatial and temporal profile and incident upon the sample at $z = 0$ with irradiance

$$I(o, r, t) = I_0\exp[-(r/r_0)^2 - (t/t_0)^2].$$

Ignoring multiple reflections within the sample, the transmitted irradiance for a medium of thickness l and reflectivity R is obtained by solving Eq. (3) to be

$$I(l, r, t) = \frac{(1-R)^2\exp(-\alpha l)I(o, r, t)}{1 + q(r, t)}, \qquad (6)$$

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where

$$q(r,t) = \beta(1-R)\left[\frac{1-\exp(-\alpha l)}{\alpha}\right] I(o,r,t).$$

Also, assuming the input face of the sample to be at the beam waist, Eqs. (3) and (4) are solved for the phase change

$$\phi(l,r,t) = \beta_1(1-R)\ln[1+q(r,t)]\Big/ \beta + (1-R)^2\gamma_1/(2\hbar\omega\beta)\int_{-\infty}^{t} dt' F_1, \quad (7)$$

where

$$F_1 = \alpha\ln[1+q(r,t')] - \frac{q(r,t')\alpha}{1-\exp(-\alpha l)} \times\{1-\exp(-\alpha l)/[1+q(r,t')]\}. \quad (8)$$

$I(l,r,t)$ and $\phi(l,r,t)$ completely characterize the electric field $E(l,r,t)$ at the exit plane of the sample, from which the electric field at any point $(l+z,r,t)$ can be determined by using the Huygens–Fresnel propagation formalism[9] as

$$E(l+z,r,t) = \frac{2\pi}{i\lambda z}\exp(i\pi r^2/\lambda z)\int_0^{\infty} E(l,r',t-z/c) \times \exp(i\pi r'^2/\lambda z) J_0(2\pi r r'/\lambda z) r' dr'. \quad (9)$$

The experimentally measured quantity is the fluence at the observation plane at a distance z given by

$$F(l+z,r) = \frac{c\epsilon_0}{2}\int_{-\infty}^{\infty} |E(l+z,r,t)|^2 dt. \quad (10)$$

We have numerically evaluated $F(l+z,r)$ for different parameters and have compared the results with those obtained experimentally.

The apparatus used is described in greater detail elsewhere.[10] Single pulses of measured Gaussian spatial and temporal profiles were obtained from a microprocessor-controlled, passively mode-locked Nd:YAG system operated at 0.5 Hz. The single pulses were switched out of the mode-locked pulse train and amplified. Attenuation of the energy in the pulse was achieved by using a half-wave-plate–polarizer combination, which produced no measurable aberrations on the spatial profile of the beam. The shot-to-shot variations in both the energy and the pulse width were monitored as described in Ref. 10. The pulse width was varied by using different-thickness étalons as the laser output coupler. The beam out of the laser was collimated with a spot size of 1.02 mm ($HWe^{-1}M$). This corresponds to a confocal parameter of 6.17 m for the 1.06-μm radiation. This beam was incident upon the sample without any focusing, and the transmitted signal was monitored at distances of 0.5 and 2 m from the sample, which are both near-field regions. The change in the spatial profile of the beam with increasing input irradiance was determined using a vidicon tube interfaced with a microprocessor-controlled optical multichannel analyzer (PARC 1215). A 2-mm-thick sample of CdSe (II-VI, Incorporated) with polished surfaces was used in this experiment. CdSe has a hexagonal wurtzite structure, and the incident laser radiation was directed along the c axis of the crystal. Curve (a) of Fig. 1 shows the change in the beam profile with irradiance for the 55-psec ($HWe^{-1}M$) pulse width used. By using a pinhole (25-μm diameter) on a photodetector, the on-axis fluence of the transmitted signal was also measured as a function of irradiance for the different pulse widths at the two distances. The results are shown in Figs. 2 and 3. To verify the validity of the theory further, the irradiance dependence of the transmitted fluence at an off-axis point was also measured. This is shown in Fig. 4.

The theoretical fits in Figs. 2–4 were obtained from the numerical evaluation of F given in Eq. (10). The value of β for CdSe has been measured to be 18 cm/GW.[1] Other parameters used in the calculation are $\alpha = 0.2\ \mathrm{cm}^{-1}$,[1] $n_0 = 2.54$,[1] and $m_{eh} = 0.104 m_e$.[11] The

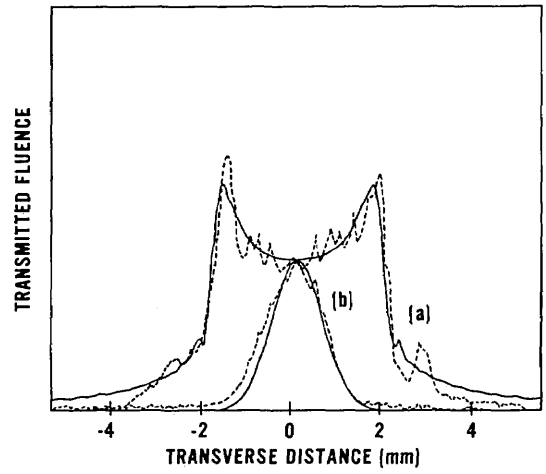

Fig. 1. (a) Defocused beam profile at a distance of 0.5 m from the sample. Incident irradiance is 1 GW/cm^2. (b) The beam profile at the same distance at low irradiance (0.03 GW/cm^2). The pulse width is 55 psec ($HWe^{-1}M$). The beam profiles are normalized to have the same on-axis fluence. The dashed lines are the experimental results and the solid lines are the theoretical fits with $p = 3.5$.

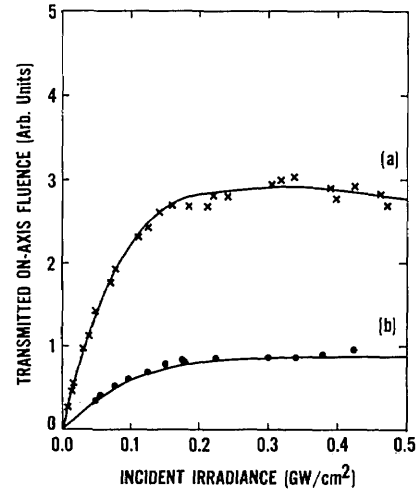

Fig. 2. Transmitted on-axis fluence as a function of incident irradiance at a distance of 0.5 m from the sample. (a) 55-psec ($HWe^{-1}M$) pulse width, (b) 26-psec ($HWe^{-1}M$) pulse width. The solid lines are numerically calculated from Eq. (10) with $p = 3.3$.

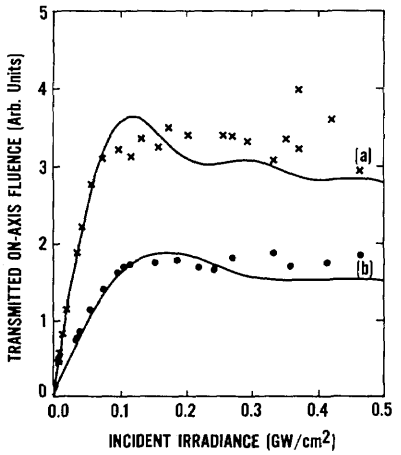

Fig. 3. Transmitted on-axis fluence as a function of incident irradiance at a distance of 2 m from the sample. (a) 55-psec (HWe^{-1} M) pulse width, (b) 26-psec (HWe^{-1} M) pulse width. The solid lines are numerically calculated from Eq. (10) with $p = 3$ for case (a) and $p = 3.3$ for case (b).

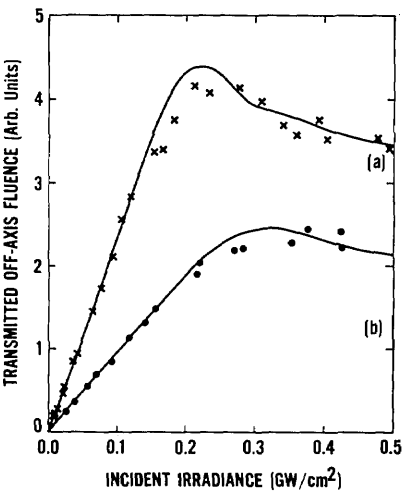

Fig. 4. Transmitted fluence measured at a point 1.1 mm off axis as a function of incident irradiance at a distance of 0.5 m from the sample. (a) 55-psec (HWe^{-1} M) pulse width, (b) 26-psec (HWe^{-1} M) pulse width. The solid lines are numerically calculated from Eq. 10 with $p = 3.3$ for case (a) and $p = 4.2$ for case (b).

total number N of charge carriers generated by two-photon absorption can be calculated by integration of Eq. (5). We obtain

$$N = \frac{(1-R)^2\beta e^{-2\alpha l}}{2\hbar\omega}\int_{-\infty}^{\infty}\frac{I^2(0,0,t')\mathrm{d}t'}{[1+q(0,t')]^2}.$$

At peak input irradiance of 1 GW/cm^2, we obtain a peak value of N evaluated at the input plane of the sample to be 2×10^{18} cm^{-3}. We find that the best agreement between the theory and the experiments is obtained for $p \sim 3.5$. The fits shown in Figs. 2–4 are for values of $p = 3.6 \pm 0.6$, with the exact value of p adjusted between 4.2 and 3.0 to obtain the best fit. Using a two-level model, p has been calculated to be $E_g{}^2/(E_g{}^2 - \hbar^2\omega^2)$,[12] where E_g is the band-gap energy. For CdSe, $E_g = 1.75$ eV at room temperature so that, at 1.06 μm, $p = 2$.

The peak phase change undergone by the beam calculated by integration of Eq. (4) is (for $I_0 = 1$ GW/cm^2) $\Delta\phi = -8.1$, which is a 1.3-wavelengths distortion. We have ignored the n_2 that is due to bound electronic effects in these calculations. Even if this nonlinearity for CdSe were as high as the n_2 of CS_2, i.e., 10^{-11} esu, the maximum contribution to the phase change would be $40\pi lI\omega n_2/n_0$, which is 0.2 for $I_0 = 1$ GW/cm^2. This index change also would be a self-focusing effect and not a defocusing effect, as observed. Thus the nonlinear refraction observed in CdSe is approximately 40 times larger than in CS_2 at this irradiance. Higher irradiances give rapidly increasing values of defocusing since the nonlinearity is induced by two-photon absorption. The abrupt and dramatic optical limiting behavior previously reported in GaAs at 1.06 μm (Ref. 6) also occurs through two-photon absorption and can be explained by this theory.

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