

# Anisotropy of nonlinear refraction of II-VI semiconductors near fundamental absorption edge

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(Submitted 30 May 1990)

Pis'ma Zh. Eksp. Teor. Fiz. **52**, No. 1, 703–706 (10 July 1990)

The anisotropy of the nonlinear refractive index of  $\text{Cd}_x\text{Se}_{1-x}$  crystals has been measured near the fundamental absorption edge. A comparison of the measured spectra with the spectrum calculated for a quasicubic model makes it possible to link this nonlinearity at room temperature with interband transitions of electron-hole pairs.

The nonlinearity of semiconductors near their fundamental absorption edge has recently been the subject of active research in connection with the possible utilization of this nonlinearity in devices for optical data processing.<sup>1</sup> Several mechanisms for this nonlinearity have been proposed in order to describe the behavior of the refractive index in this region.<sup>1</sup> It has been suggested that the nonlinearity might be due to a mechanism based on a "direct saturation" model or the model of a dynamic Burshstein–Moss shift. There is the further possibility that exciton nonlinearity mechanisms are manifested, particularly at low temperatures. There is also a significant increase in the polarizability component due to interband carrier transitions in semiconductors near the fundamental absorption edge.<sup>2,3</sup>

Studies of the latter nonlinearity mechanism ordinarily use a simple two-band model which describes allowed transitions through the  $C$  and  $V$  bands.<sup>1,2</sup> When the complex structure of the valence band of real semiconductors is taken into account, these results change. In III–V materials, for which the contributions of virtual transitions to the states of light and heavy holes differ only because the effective masses of these states differ, the spectra for the increments in the refractive index due to non-equilibrium electrons ( $\Delta n^{(e)}$ ) and holes ( $\Delta n^{(h)}$ ) differ significantly<sup>4</sup> from the results of the two-band model, although it appears that no quantitative comparison with experimental data on these materials has been made. Sharper deviations from the two-band model occur in II–VI materials. In this case, the three valence bands,  $V_A$ ,  $V_B$ , and  $V_C$ , are degenerate, because of the crystal splitting and the spin-orbit splitting. They are also distinguished by their pronounced anisotropy. Since the effective masses are large (in comparison with that of the  $C$  band), the anisotropy of the energy spectrum of the  $V$  bands is not important. Of importance instead is the difference between the transition matrix elements for different polarizations of the light with respect to the optic axis of the crystal. It is the polarization dependence of the dispersion,  $\Delta n_{\parallel,\perp}(\omega)$ , which makes it possible to distinguish the contributions of virtual transitions to different  $V$  bands. Our purpose in the present letter is to make a quantitative comparison of the spectra  $\Delta n_{\parallel,\perp}(\omega)$  for II–VI semiconductors with theoretical results based on a

quasicubic model of the energy spectrum.<sup>5</sup> This comparison will make it possible to identify the mechanism for the optical nonlinearity of the refractive index.

The contribution of the nonequilibrium free carriers of band  $k$  to the optical characteristics of the semiconductor, for the uniaxial crystal which we are considering here, is determined by the diagonal tensor of the complex electrical conductivity, found from the expressions given in Ref. 4:

$$\sigma_{\parallel, \perp}^{(k)}(\omega) = \frac{ie^2 N_k}{\omega m_{\parallel, \perp}^{(k)}} + \frac{ie^2}{\omega} \sum_{k \neq k'} M_{\parallel, \perp}^{kk'} \int \frac{dp}{(2\pi \hbar)^3} f_{k p} \times [(\epsilon_{k p} - \epsilon_{k' p} + \hbar\omega - i\gamma)^{-1} + (\epsilon_{k p} - \epsilon_{k' p} - \hbar\omega + i\gamma)^{-1} - 2(\epsilon_{k p} - \epsilon_{k' p})^{-1}] \quad (1)$$

Here  $\epsilon_{k p}$  and  $f_{k p}$  are the dispersion law and distribution function of the electrons,  $N_k$  is the density of free carriers in band  $k$ ,  $m_{\parallel, \perp}^k = \partial^2 \epsilon_{k p} / \partial p_{\parallel, \perp}^2$ , and  $\gamma$  is the attenuation which results from the broadening of the extremum by various collision mechanisms. We assume below that this attenuation is small and is the same for all transitions. Since the distribution  $f_{k p}$  is localized at small momenta, we can use the square of the interband matrix element at the point  $p = 0$  and write, in accordance with Ref. 6,

$$m M_{\parallel, \perp}^{CV_i} = \epsilon_g \chi_{\parallel, \perp}^i / m_c; \quad \chi_{\parallel}^A = 0, \quad \chi_{\perp}^A = 1/2, \\ \chi_{\parallel}^B = 2/3, \quad \chi_{\perp}^B = 1/6, \quad \chi_{\parallel, \perp}^C = 1/3,$$

where  $\epsilon_g$  is the band gap, and  $m_c$  is the isotropic effective electron mass in band  $C$ .

Near the fundamental absorption edge (with  $|\hbar\omega - \epsilon_g|$  comparable to the electron temperature  $T_e$  and to the splitting energies of the  $V$  bands), we have the following expression for the electron components of the refractive index:<sup>4</sup>

$$\Delta n^{(e)}_{\parallel, \perp}(\omega) = - \frac{2\pi e^2 N_C}{\kappa_{\parallel, \perp}^{1/2} \omega^2 m_e} \left[ 1 + \frac{2(\hbar\omega)^2}{n^{1/2} T_e (\epsilon_g + \hbar\omega)} \right. \\ \left. \times \sum_i \chi_{\parallel, \perp}^i J\left(\frac{\hbar\omega - \epsilon_{V_i}}{T_e}; \frac{\gamma}{T_e}\right) \right], \\ J(x; \gamma) = \int_0^{\infty} dy \frac{y^{1/2}(y-x)\exp(-y)}{(y-x)^2 + \tilde{\gamma}^2}, \quad (2)$$

where  $\epsilon_{V_i}$  is the energy gap between the  $C$  and  $V_i$  bands,  $\kappa_{\parallel, \perp}$  is the high-frequency dielectric constant, and the function  $J(x; \tilde{\gamma})$  is shown in Fig. 1. An estimate of the

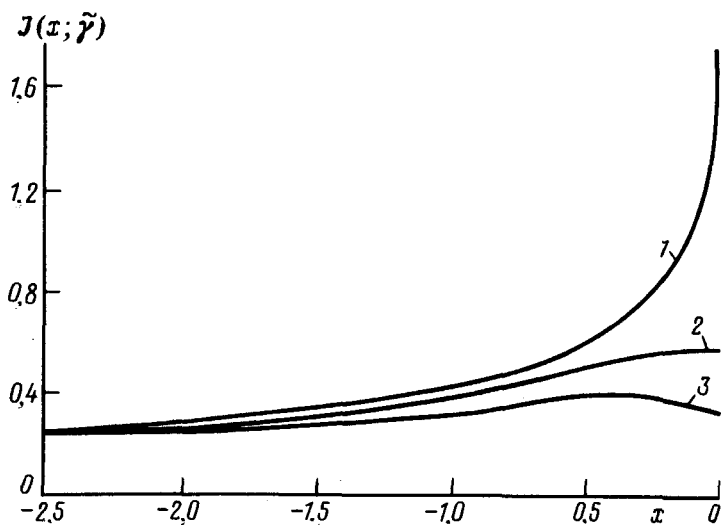


FIG. 1. The dimensionless function  $J(x; \tilde{\gamma})$  versus the parameter  $x = (\hbar\omega - \epsilon_g)/T_c$ . 1— $\tilde{\gamma} = 0$ ; 2— $\tilde{\gamma} = 0.5$ ; 3— $\tilde{\gamma} = 1$ .

contribution from nonequilibrium holes<sup>4</sup> yields an increment for this spectral region which is small, in proportion to the parameter  $m_c/m_{v_i}$ .

To measure the nonlinearity coefficients of the refractive index,  $n_2$ , and their anisotropy, we used a technique which we had developed previously.<sup>7</sup> It is based on the change in the angular characteristics of a beam in the course of a self-effect in a nonlinear medium. The errors in the measurements of  $n_2$  for the various polarizations of the light with respect to the optic axis of the crystal were no worse than 5%. The experiments were carried out on mixed  $\text{CdS}_x\text{Se}_{1-x}$  ( $x \approx 0.9-0.6$ ) single crystals of good optical quality. The crystal faces were oriented with respect to the optic axis and were highly polished. To measure the frequency dependence near the absorption edge, we used a beam from a tunable Kolorit laser with quasilongitudinal pumping by the second harmonic from a single-mode Nd:YAG laser with passive  $Q$  switching. The active medium was a polymer medium (polyurethaneacrylate) activated with the dye rhodamine 6G or 6-aminophenalinon (the tuning range was 560–600 nm or 560–670 nm, respectively; the spectral width was less than 1 nm).

Figure 2 shows the frequency dependence of the negative nonlinear increment in the refractive index of a  $\text{CdS}_{0.84}\text{Se}_{0.16}$  crystal for various polarizations of the light with respect to the optic ( $c$ ) axis. These results were found at room temperature from the experimental values of  $n_2$ , the absorption coefficient, and the carrier lifetime at an excited-carrier density  $N_c \approx 10^{17} \text{ cm}^{-3}$ . Also shown here are corresponding theoretical results calculated from (2).

The good agreement observed between the theoretical and experimental values at  $\tilde{\gamma} = 1$  (this value corresponds to interband relaxation times  $\sim 2.5 \times 10^{-14}$  s and agrees

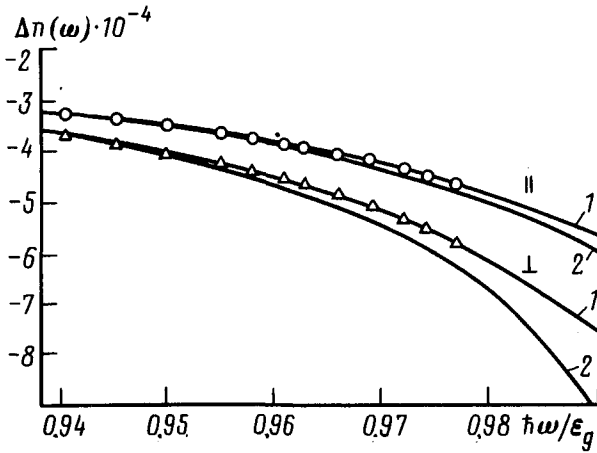


FIG. 2. Frequency dependence of the nonlinear increment in the refractive index,  $\Delta n(\omega)$ , of a  $\text{CdS}_{0.84}\text{Se}_{0.16}$  crystal. 1—Theoretical,  $\tilde{\gamma} = 1$ ; 2—theoretical,  $\tilde{\gamma} = 0$ ;  $\circ$ —experimental,  $E \parallel c$ ;  $\triangle$ —experimental,  $E \perp c$ .

well with the measured value<sup>8</sup>) is consistent with the conclusion that interband transitions of nonequilibrium carriers dominate the nonlinear polarizability of II–VI semiconductors near the fundamental absorption edge at room temperature.

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Translated by D. Parsons