Nature of the nonlinearity that leads to self-action of laser radiation in semiconductors of the A_2B_6 group

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The self-action effects of laser radiation in a CdS semiconductor crystal in various crystallographic directions are investigated at temperatures 300 and 77°K. Self-action of a ruby-laser beam is investigated for a broad set of mixed semiconductors CdS_xSe_{1-x} and $Zn_xCd_{1-x}S$. It is shown that at a given frequency the nonlinearity of the crystals increases with decreasing width of the forbidden band. On the basis of an analysis of the experimental data, some conclusions are drawn regarding the nature of the nonlinearity of the refractive index in semiconductors of the A_2B_6 group.

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INTRODUCTION

Our earlier investigations[1–4] of the self-action of a ruby-laser beam in semiconductors of the CdS type have revealed that this is a complicated interaction that manifests itself in the presence of both self-focusing and self-defocusing, depending on the intensity of the beam. This presupposes both a positive and a negative change in the refractive indices of the given crystals in the field of a high-power light wave.

In the case of self-action, when the laser beam, besides exerting a pure field action on the refractive index, produces also changes in certain characteristic parameters of the medium, which in turn influence this refractive index, we can write for the index

$$n(x,E)=n(x)+n_2(x)E^2+\ldots=n(0)+\frac{\partial n}{\partial x}x+n_2(0)E^2+\ldots, \qquad (1)$$

where x is a characteristic of the medium (for example, the free-carrier density, the temperature, etc.) and is altered by the laser radiation. In two-photon excitation of non-equilibrium free carriers, the carrier density is

$$x\sim\int_0^\tau E^4 d\tau,$$

where τ is the lifetime of these carriers. The change in the temperature of the medium in two-photon absorption is also $x\propto E^4$, whereas in the case of linear absorption we have $x\propto E^2$. Thus, expression (1) can be rewritten in the form

$$n(x,E)=n_0+n_2E^2+n_4\int_0^\tau E^4 d\tau+\ldots, \qquad (2)$$

where the coefficients n_2, n_4, etc. are determined respectively by different mechanisms of the nonlinearity of the refractive index.

The purpose of the present paper is to clarify the principal nonlinearity mechanisms that determine the magnitude and sign of the coefficients of the nonlinear change of the refractive index (n_2, n_4) in semiconductors of the CdS type.

ANALYSIS OF THE CONTRIBUTION OF THE PRINCIPAL NONLINEARITY MECHANISMS

Before we proceed to describing the experimental results, let us estimate the principal mechanisms that can lead to a change in the refractive index in these crystals.

We consider first mechanisms that lead to self-focusing of the laser radiation.

1. *Electrostrictive compression.* In the beam of a single-mode laser having an electric field gradient, each volume element of the medium is acted upon by a ponderomotive (striction) force that leads to compression of the medium on the beam axis and to an increase in its density. This leads in turn to an increase of the refractive index. The coefficient of nonlinear variation of the refractive index is determined in this case by the expression[5]

$$n_2=\beta_T\gamma^2/16\pi n_0, \qquad (3)$$

where β_T is the isothermal compressibility $\gamma=\frac{1}{3}(n^2-1)\times(n^2+2)$, and n_0 is the refractive index of the medium at low intensities. Using the parameters of the CdS crystal, we obtain

$$n_2=1.3\cdot10^{-12}\ \text{cgs esu}.$$

It should be noted that expression (3) was obtained for a stationary regime of striction self-focusing, whereas in the field of a Q-switched laser it is necessary to consider the nonstationary solution. Calculations of this kind were carried out in[6] and it was shown that allowance for the non-stationary solution leads to an increase of the threshold power of the striction self-focusing.

2. *Thermal heating of the material medium.* Absorption of light (linear and nonlinear) leads to heating of the medium in the light-beam channel, and consequently to a change of its refractive index. The total change of the refractive index due to heating can be represented in the form[7]

$$\delta n_{\text{therm}}=\left(\frac{\partial n}{\partial \rho}\right)_T\delta\rho+\left(\frac{\partial n}{\partial T}\right)_\rho\delta T. \qquad (4)$$

The first term in (4) is connected with the expansion of the medium and is usually negative. The second term is connected with the change in the width of the forbidden band of the crystal E_g as the crystal becomes heated, and is positive for crystals such as CdS. The two terms have different rise times and, particularly in the case of Q-switched lasers, the principal effect on the refractive index is that of the second term, which has a very short relaxation time ($\tau \sim 10^{-11}$–10^{-12} sec). In addition, for short laser pulses ($\tau_p \sim 10^{-8}$ sec), the heating of crystals such as CdS is determined mainly by the thermalization of the "hot" electrons in the band, which are excited as a result of two-photon transitions, whereas the time of the nonradiative recombination of the "cold" electrons in these crystals is of the order of 10^{-6}–10^{-3} sec.[8] Furthermore, under our experimental conditions the two-photon absorption was much larger than the linear absorption. Consequently the nonlinearity of the refractive index and of crystals of the CdS type, due to the thermal heating, is determined mainly by the temperature variation of the width of the forbidden band E_g as a result of two-photon absorption of part of the laser radiation. The coefficient of the nonlinear change of n then takes the form

$$n_4 = \frac{1}{16\pi^2} \frac{\beta c^2 \tau_p n_0^2}{\rho c_\rho} \left(\frac{2\hbar\omega - E_g}{2\hbar\omega} \right) \frac{dn}{dT},$$

where β is the two-phonon absorption coefficient, c_ρ and ρ are the specific heat and the density of the medium, c is the speed of light, n_0 is the linear refractive index, $\hbar\omega$ is the energy of the laser photons, and E_g is the width of the forbidden band.

At the present time there are no measurements of dn/dT in the dynamic regime, and we have therefore used the value measured under stationary conditions.[9] As a result of these estimates we have obtained for the CdS crystal at a laser intensity $I \approx 1$ MW/cm^2

$$n_4 E^2 \approx 8 \cdot 10^{-13} \text{ cgs esu}$$

3. *Nonlinear polarizability of bound electrons.* The change due to the nonlinear polarizability of the bound electrons is determined by the nonlinear third-order susceptibility $\chi^{(3)}$. Within the framework of the two-band model, $\chi^{(3)}$ can be represented in terms of the linear susceptibility $\chi^{(1)}$ and the second-order nonlinear susceptibility $\chi^{(2)}$.[10] The coefficient of the nonlinear change of n can then be written in the form

$$n_2 = \frac{4\pi}{\chi^{(1)} n_0} \left\{ \left[\frac{2}{3} \chi^{(2)} \right]^2 - \frac{2[\chi^{(1)}]^3}{E_g N_v} \right\},$$

where N_v is the number of valence electrons per unit volume. The experimental values of $\chi^{(2)}$ and $\chi^{(1)}$ for CdS crystals were measured sufficiently accurately in[10]. By using these values we obtain $n_2 = 6 \times 10^{-12}$ cgs esu.

The contribution of the other mechanisms is certainly less. For example, the nonlinear polarizability of excitons, in contrast to their appreciable resonant contribution in the exciton region,[1] yields for CdS at the ruby-laser emission frequency the value $n_2 \sim 10^{-14}$ cgs esu.

The following are the most probable nonlinearity mechanisms that lead to self-defocusing of laser radiation in semiconductors of the CdS type:

a) *Thermoelastic stresses.* That part of the crystal in which the laser beam propagates, will be heated somewhat as a result of absorption (predominantly two-photon), and the heating will not be uniform, i.e., a temperature gradient will be produced. In this case, however, in accordance with the results of V'yun, Lokhov, and Fiveĭskiĭ,[12] radial compressive stresses will act in this part of the crystal during the time of the laser monopulse. Investigations of CdS crystals under hydrostatic pressure have shown that the width of their forbidden band increases in this case, i.e., the refractive index of the crystal on the long-wave side of the absorption edge should decrease with increasing pressure. When two-photon absorption is taken into account, the coefficient of the nonlinear variation of n is in this case

$$n_4 = \frac{k^2 \alpha_T c^2 n_0 \tau_p \beta}{16\pi^2 c_\rho \rho (\xi + 2\mu)} \left(\frac{2\hbar\omega - E_g}{2\hbar\omega} \right) \left[1 - \frac{\xi + 2\mu}{k} - \cos\left(\frac{\alpha_T n_0 v_\rho \tau_p}{r_0^2} \right) \right] \frac{dn}{dp},$$

where k is the isothermal modulus of hydrostatic compression, $\xi = k = 3\mu/2$, and α_T is a coefficient of temperature expansion.

At a laser intensity $I \sim 1$ MW/cm^2 we obtain for the CdS crystal

$$n_4 E^2 \approx 5 \cdot 10^{-14} \text{ cgs esu}.$$

b) *Contribution of nonequilibrium free carriers.* At the place where the laser beam acts on a crystal, two-photon absorption causes excitation of free carriers that make an appreciable contribution to the negative change of the dielectric constant of the crystal. The coefficient of the nonlinear change of n is then

$$n_4 = \frac{e^2 c^2 n_0 \beta}{64\pi m^* \hbar\omega^3} \tau$$

where e and m^* are the charge and effective mass of the carriers in the band and τ_0 is their lifetime. Since the effective mass of the holes in the CdS crystal is much larger than the effective mass of the electrons, and their lifetime in the band is much shorter, the change of the refractive index will be determined mainly by the concentration of the non-equilibrium electrons. For a laser intensity $I \sim 1$ MW/cm^2 we then have

$$n_4 E^2 \approx 5 \cdot 10^{-13} \text{ cgs esu}.$$

Thus, our estimates show that all the mechanisms noted above can make an appreciable contribution to the nonlinearity of the refractive index of the crystals of the CdS type and lead to self-action of laser radiation in them. However, to reveal those mechanisms that determine the nature of the nonlinearity in these semiconductors, it is necessary to perform experiments that permit a qualitative separation of the contribution of each of them.

EXPERIMENTAL PROCEDURE

We investigated the "external" self-focusing[2] of ruby-laser radiation in bulky single crystals of CdS_xSe_{1-x}

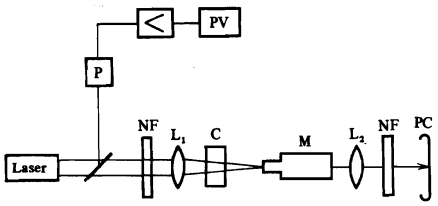

FIG. 1. Diagram of experiment. NF—neutral filters, L_1 and L_2—lenses, C—crystal, M—microscope, PC—photographic camera, PV—pulse voltmeter, P—photocell, <—amplifier.

and $Zn_xCd_{1-x}S$ of good optical quality, having equal thickness ~ 5 mm. The experimental setup is illustrated in Fig. 1. The radiation source was a single-mode Q-switched laser generating ~ 15-msec pulses. The beam from this laser, having a Gaussian intensity distribution over the end face, was attenuated with neutral filters NF to the desired intensity and focused with the aid of lens L_1 and directed to the investigated crystal C. The geometric focus of the lens L_1 was located behind the crystal. The image of the beam cross sections, both in front of the geometric focus of the lens L_1 and behind it, was projected with the aid of a microscope and lens combination (M–L_2) system onto a photographic camera (PC) and was photographed. Comparison of the dimensions of these cross sections makes it possible to assess the changes of the positions of the focus itself at different beam intensities. To obtain a normal photographic density, neutral filters NF were placed in front of the camera. The photographs of the spatial distribution of the laser-beam intensity, obtained in this manner, were then analyzed with an MF-2 microphotometer. The characteristic diameter of the beam was assumed to be its dimension at a level at which the intensity decreases to one half the value at the center. To measure the laser-beam power, part of the beam was diverted to a previously graduated pulsed-photometer system consisting of a photocell P, an amplifier, and a pulsed voltmeter PV. The absolute power measurement error was 15%.

The relative accuracy of our measurements was determined by the instability of the laser operation, and measures were therefore taken to stabilize its operating regime. Careful adjustment, the use of a small-diameter diaphragm, high stability of the voltage on the pump lamps, and good cooling of the active element made it possible to attain high lasing stability at a practically constant half-width of the contour of the section in the far zone ($\Delta I/I \sim 5\%$; $\Delta d/d \sim 3\%$).

It should be noted that the procedure of "external" self-focusing offers undisputed advantages from the point of view of the study of the nonlinear refraction itself, for in this case the focus lies outside the nonlinear medium, thereby eliminating all the phenomena and singularities that appear in the focal region of the nonlinear medium and make observation of its behavior difficult.

Figure 2 shows photographs and microphotographs of the beam of a ruby laser passing through a CdS crystal placed in front of the geometric focus of the lens, at different intensities. It is seen that slight self-focusing of the beam is observed (the cross section decreases) at an intensity on the order of several MW/cm^2, and the beam becomes self-defocused (the cross section increases) at an intensity on the order of several dozen MW/cm^2. With appreciable increase of the intensity (70–90 MW/cm^2), the self-defocusing of the beam is accompanied by a change in the shape of the beam and by its stratification, as a result of which first one ring and then a number of less distinct rings appear in the cross section (Fig. 2c). The total dimension of the cross section continues to increase. This behavior of the beam passing through a CdS crystal is obviously governed by aberrations of the defocusing nonlinear lens produced in the crystal.

In the subsequent report of the experimental results, so as not to clutter the figures, they show only the change of the cross section in front of the focus of lens L_1, although we obtained also the cross sections behind the focus, and were thus able to monitor the behavior of the nonlinear focus with changing laser intensity. In addition, we measured the diameter of the cross section only at those intensities at which the beam behaved as a unit and revealed no stratification.

DISCUSSION OF RESULTS

According to our experiments on self-focusing of the ruby-laser beam in cadmium sulfide[1] the coefficient n_2 amounts to 1.4×10^{-12} cgs esu. The quantitative estimates presented above yield coefficients that are close to the experimental ones, and do not make it possible to identify the main mechanism of the nonlinearity that leads to self-focusing of the laser radiation in this crystal. To permit this identification, we performed experiments on the temperature dependence of the self-focusing in cadmium sulfide.

It is known[5] that electrostriction has a strong positive temperature dependence $(dn_2/dT)_{str} > 0$. The contribution of this mechanism of nonlinearity should therefore increase with increasing temperature. The contribution of the temperature variation of the width E_g of the forbidden band also depends essentially on the temperature interval in which it is observed. Thus, the temperature coefficient (dE_g/dT) for the CdS crystal near room temperature is 6 cm^{-1}/deg,[9] as against 2 cm^{-1}/deg at a temperature 77 °K.[13] On the

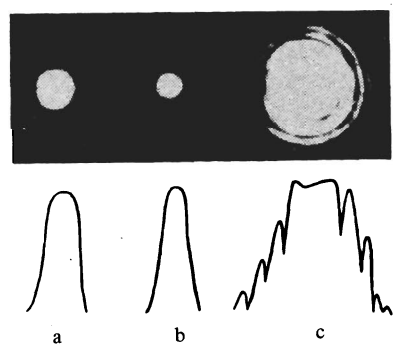

FIG. 2. Photographs and microphotograms of the cross section of a laser beam passing through a CdS crystal at intensities 0.1 (a), 5 (b), and 80 (c) MW/cm^2.

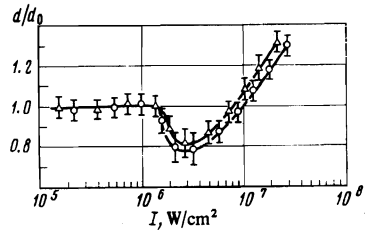

FIG. 3. Ratio of the diameter of a ruby-laser beam passing through a CdS crystal at a definite intensity to the diameter of the same beam at the initial intensity vs the laser intensity in a section located ahead of the geometric focus of the lens. $\Delta - T = 300$ K, $k \perp C$, $E \perp C$; $o - T = 77$ K, $k \perp C$, $E \perp C$.

other hand, the contribution of the nonlinear polarizability of the bound electrons is practically independent of the temperature.

Figure 3 shows plots of the ratio d/d_0 of the diameter of the beam passing through the CdS crystal at a definite intensity to the diameter of the same beam at the initial minimal intensity, against the intensity of the laser beam at temperatures 300 °K (triangles) and 77 °K (circles). It is seen from the figure that the self-focusing threshold remains practically unchanged when the temperature is changed from 77 to 300 °K, so that the main contribution to the nonlinear polarizability of the crystals is made by a mechanism that does not change its contribution with changing temperature. This mechanism is apparently the nonlinear polarizability of the bound electrons.

To verify this it is important to obtain the frequency dependence of this nonlinearity, since the nonlinear polarizability of the bound electrons[14] presupposes a characteristic frequency dependence that increases resonantly when the edge of the intrinsic absorption is approached. Unfortunately, for the lack of a sufficient set of laser wavelengths, this dependence cannot be obtained with one CdS crystal. However, data of this kind can be obtained with a single laser frequency and a definite set of crystals. Indeed, it is well known that mixed crystals of the A_2B_6 group form a series of substitutional solid solutions, which have a band structure similar to the band structure in CdS, but have a smooth variation of the forbidden-band widths with changing percentage composition. Consequently, by using crystals of this type and by performing the measurements at a fixed frequency, we obtain in effect information on the frequency dependence of the nonlinearity of the CdS crystal.

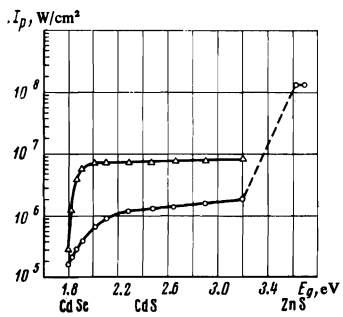

FIG. 4. Dependence of the threshold intensity of self-focusing (circles) and self-defocusing (triangles) on the width of the forbidden band of the crystals CdS_xSe_{1-x} and $Zn_xCs_{1-x}S$.

To perform such an experiment we used the mixed crystals, CdS_xSe_{1-x} and $Zn_xCd_{1-x}S$, in which the width of the forbidden band changes, with changing composition, from 1.8 to 2.5 eV for the CdS_xSe_{1-x} and from 2.5 to 3.7 eV for the $Zn_xCd_{1-x}S$ system. We used "external" self-focusing of the radiation of Q-switched ruby laser. Figure 4 shows the dependence of the threshold intensity of the self-focusing (circles) on the width of the forbidden band of the investigated crystals. It shows clearly the resonant increase (i.e., the corresponding decrease of the threshold intensity of the self-focusing) of the positive contribution to the nonlinear change of the refractive index as the laser frequency ($\hbar\omega = 1.78$ eV) approaches the intrinsic absorption edge of the crystals. This behavior of the nonlinearity is in qualitative agreement with the frequency dependence of the nonlinear polarizability of bound electrons. We assume therefore that the principal mechanism leading to self-focusing of laser radiation in crystals of the CdS type is nonlinear polarizability of bound electrons.

We note that at values of E_g between 3.4 and 3.6 eV a jump is observed in the threshold characteristics of the self-focusing; this jump can obviously be attributed to the contribution made to the nonlinear polarizability by two-photon absorption, since it is precisely in this region that the coefficient of the two-photon absorption of ruby-laser radiation vanishes.

We consider now the nature of the negative change of the refractive index in crystals of the CdS type. As shown by estimates carried out above, the most probable mechanisms can be thermoelastic stresses which are produced by local heating of the crystal by the laser beam, and also the contribution of the nonequilibrium free carriers that are produced in the path of the beam as a result of its two-photon absorption.

To separate the contributions of these two mechanisms in the CdS crystal, we have investigated the self-action effects in different crystallographic directions. A study of the piezo-optical properties of CdS crystals[15] has shown that the short-wave shift of the intrinsic absorption edge is brought about by compression stresses directed only along the optical axis of the crystal C. On the other hand, compression of the crystal in all other directions either leads to no shift of the absorption edge at all, or leads to a slight long-wave shift in the opposite direction. On the other hand, the contribution of the non-equilibrium free carriers is practically isotropic.

We have investigated the self-action of a ruby-laser beam in a CdS crystal in the two following experimental geometries:

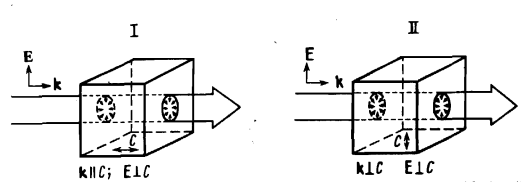

FIG. 5. Geometry of experiments (I, II) on the self-action of a ruby-laser beam in a CdS crystal in different crystallographic directions.

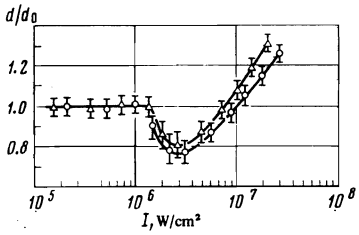

FIG. 6. Dependence of the dimensionless diameter of a ruby-laser beam passing through a CdS crystal on the laser intensity. o—experimental geometry *I*, $\mathbf{k} \parallel C$, $\mathbf{E} \perp C$; Δ—experimental geometry II, $\mathbf{k} \perp C$, $\mathbf{E} \perp C$.

I. The laser beam is directed along the optical axis of the crystal. The electric vector of the light-wave field is perpendicular to the *C* axis ($\mathbf{E} \perp C$) (Fig. 5.I).

II. The laser beam is directed perpendicular to the optical axis of the crystal, and the electric vector of the light-wave field is also perpendicular to the *C* axis (Fig. 5.II).

In the first experimental geometry, inasmuch as the thermoelastic compression stresses in a Gaussian beam are radially directed towards the center of the beam,[12] there is no compression along the optical axis. In the second geometry the compression stress along the optical axis is maximal. Consequently the contribution of the thermoelastic stresses to the negative change of the refractive index of the CdS crystal should be practically nonexistent in the first geometry and maximal in the second.

Figure 6 shows the results of this experiment. We see that in both the first and the second geometry an appreciable self-defocusing of the laser beam was observed, whereas if a contribution of the thermoelastic stresses is assumed, there should be no self-defocusing in the first geometry. Consequently this experiment indicates that the principal mechanism of the negative change of the refractive index of CdS crystals, leading to self-defocusing of the laser radiation in these crystals, is the contribution of the non-equilibrium free carriers produced in the light-beam channel as a result of two-photon absorption.

This explains a certain increase of the self-focusing threshold[1)] with decreasing temperature (see Fig. 3). Indeed, on going from room temperature to liquid nitrogen temperature, in spite of the fact that the coefficient of the two-photon absorption in the CdS crystal remains practically unchanged, the fraction of the electrons bound into excitons increases (the thermal disintegration of the excitons decreases abruptly). This leads to a certain decrease of the total density of the free carriers generated by the laser at nitrogen temperature and to a corresponding increase of the self-defocusing threshold. In addition, the self-defocusing threshold in the second geometry (see Fig. 6) turned out to be somewhat lower than in the first. This decrease can be attributed to an increase of the coefficient of two-photon absorption and to a corresponding increase of the density of the excited carriers, which is due to the appearance of compression stresses along the optical axis in the second geometry. This increase of the two-photon absorption following compression of the CdS crystal along the *C* axis was observed by Lisitsa and Sidorko.[16]

Using the mixed crystals CdS_xSe_{1-x} and $Zn_xCd_{1-x}S$, we also investigated the frequency dependence of the nonlinearity that leads to self-defocusing of the laser radiation in these crystals. The results of these measurements are shown in Fig. 4, where the triangles denote the dependence of the threshold intensity of the self-defocusing on the width of the forbidden band. Such a course of this nonlinearity agrees well with the theoretically predicted frequency dependence of the contribution of the free carriers to the dispersion of crystals of the A_2B_6 group.[17]

Thus, from the analysis of all our experiments we can draw the conclusion that the main mechanism of the nonlinearity of semiconductors of the A_2B_6 group, which leads to self-focusing of the laser radiation, is the nonlinear polarizability of the bound electrons, and the nonlinearity mechanism that leads to self-defocusing of the laser radiation in the crystals is due to the contribution of the nonequilibrium free carriers.

[1)]The threshold self-defocusing intensity was assumed to be that minimal value of the laser-beam intensity at the entrance to the crystal at which the divergence of the beam past the crystal becomes larger than the diffraction divergence.

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