Determination of the coefficients of the nonlinear refractive index of a CdS crystal by the nonlinear refraction method

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A method which should yield not only the cubic nonlinearity coefficients but also higher-order nonlinearities (in particular, n_4) was developed for determining the nonlinear refractive index of material media. The method was applied to the determination of the coefficients of the nonlinear change in the refractive index of a CdS semiconductor crystal, which was very much a noncubic medium. The values of the coefficients n_2 and n_4 obtained in this way indicated a strong nonlinearity of the refractive index of such semiconductors.

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INTRODUCTION

Determination of the coefficients of the nonlinear change in the refractive index of a material in the field of a high-power light wave is a task of major practical importance (for example, in the development of new laser systems) and is also of intrinsic physical interest because the nonlinear coefficients are essentially new physical parameters of material media and studies of these coefficients give important information on the structure of matter.

Our investigation [1,2] have shown that II–VI semiconductors are characterized by a strong nonlinearity of the refractive index n and the nature of this nonlinearity is very different from the cubic form. For this reason we decided to determine directly the nonlinearity coefficient n for CdS crystal which, depending on the incident light intensity, can exhibit self-focusing or self-defocusing of laser beams.

Difficulties arise in measurements of this kind: dimensions of an optical inhomogeneity region are small and the duration of its existence is short. There are at present the following main methods for the determination of the coefficients of the nonlinear variation of the refractive index: interference,[3,4] frequency mixing,[5] rotation of the polarization ellipse,[6] and measurement of the self-focusing length[7] or of the focal length under "external" self-focusing conditions.[8] However, for a number of reasons, including the inability to measure nonlinearities of higher order than cubic because the overall value of Δn (without separation into the coefficients n_2, n_4, etc.) is obtained or even because of the limited range of validity, all these methods are found to be inappropriate for the determination of the nonlinearity of cadimium sulfide crystals. Therefore, we shall propose a new method for determining the nonlinearity of the refractive index of material media which can be used to determine not only the cubic nonlinearity coefficients but also the higher coefficients (in particular, n_4). This method is based on the change in the angular parameters of a beam during its self-interaction in a nonlinear beam. Its essence is as follows.

Let us assume that a powerful beam of light with specified input parameters is incident on a nonlinear layer of matter. After passing through this layer the beam now has different parameters. If we know the law governing the changes which occur in the beam in matter and both input and output parameters, we can find the constants which determine the properties of matter. We shall use the law of propagation of a laser beam in a nonlinear medium in the form of an equation derived by Akhmanov *et al.*,[9] which has simple physical interpretation:

$$\left(\frac{\partial f}{\partial z}\right)^2 = n_2E_0^2\left[\frac{1}{a^2f^2n_0}-\frac{1}{a^2n_0}\right]+n_4E_0^4\left[\frac{1}{a^2f^4n_0}-\frac{1}{a^2n_0}\right] + \frac{1}{R_{\text{in}}^2}+\frac{1}{a^2k^2}\left[\frac{1}{a^2}-\frac{1}{a^2f^2}\right], \qquad (1)$$

where k is the wave vector; $f(z)$ is the dimensionless half-width of the beam; a is the input radius of the beam; R_{in} is the initial radius of curvature of the wavefront; n_0 is the nonlinear refractive index of the medium; n_2 and n_4 are the nonlinearity coefficients of the refractive index; E_0 is the field intensity on the beam axis. Equation (1) describes axially symmetric self-focusing of the paraxial part of a Gaussian beam in an infinite medium. It has been found that Eq. (1) cannot explain some the the experimental features of the self-interaction of light. Therefore, it is desirable to determine the range of parameters in which the theory of Akhmanov *et al.*[9] gives the results in good agreement with the experimental data.

Clearly, this is the range of the parameters in which the undesirable features of the self-interaction, such as the collapse of a beam into a filament, formation of rings, etc.[10] are absent or weak. On the basis of the available results,[11,12] we can put forward the following conditions under which Eq. (1) should describe quite satisfactorily the real situation:

1) the input beam power should not be much greater than the critical value ($P_{\text{cr}}=cn_0/2n_2k^2$);

2) the self-focusing length should be greater than the dimensions of the sample (the self-focusing should be external);

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3) the beam entering a medium should be weakly focused.

DESCRIPTION OF THE METHOD

Let us assume that the monochromatic axially symmetric light beam characterized by a set of input parameters (R_{in}, a, and average input intensity I_{av}) is incident perpendicularly to the plane of a sample of thickness l. The diameter and radius of curvature of the beam wavefront change during the passage through the sample. Having determined experimentally the input and output parameters [including the output parameter b and $G=(\partial f/\partial z)^2$ which is governed by the angle of incidence of the beam on the exit plane], we obtain a relationship between the constants n_2 and n_4 if we substitute these parameters in Eq. (1). If a similar operation is carried out for two other input parameters of the beam, an additional relationship of the same kind is obtained. The two of them form a system of two algebraic equations, whose solution is of the form:

$$n_2=\frac{B_1A_2-B_2A_1}{A_2-A_1},\qquad n_4=\frac{B_2-B_1}{A_2-A_1},\tag{2}$$

where

$$A_i=\frac{a_i^2+b_i^2}{b_i^2}E_{0i}^2,\qquad R_i=\frac{1}{R_{in\,i}^2},$$

$$B_i=\frac{n_0}{E_{0i}}\left\{\frac{b_i^2a_i^2}{a_i^2-b_i^2}(G-R_i)+\frac{1}{k^2a_i^2}\right\}.$$

It is convenient to relate R_{in} and G to set of the input and output diameters of the beam, respectively, and E_0 to the average input intensity I_{av}. The measured parameters are shown in Fig. 1. In the case of paraxial beams the value of G is related to the angle of emergence of the beam β by

$$G=\frac{1}{a^2n_0^2}\tan^2\beta.$$

The tangent of β can be expressed in terms of the radii b and g and allowance for the diffraction of light can be made by substituting $n_2=n_4=0$ in Eq. (1); integration of this equation and certain transformations finally give

$$\tan\beta=\frac{b}{c}-\left(\frac{g^2}{c^2}-\frac{1}{k^2b^2}\right)^{1/2}.$$

Similarly, the quantity $R_{in\,i}$ can similarly be expressed in terms of the radii m and a:

$$R_{in\,i}=\frac{1}{n_0^2R_0^2},\qquad R_0=\left[\frac{1}{s}-\left(\frac{a_i^2}{s^2m_i^2}-\frac{1}{k^2m_i^4}\right)\right]^{-1}.$$

The average intensity at the entry to the nonlinear layer is given by

$$I_{av}=\frac{cn_0}{4\pi}E_0^2\alpha,\qquad \alpha=\int_0^{+\infty}f(x)x\,dx,$$

where $f(x)$ is the distribution function of the laser wave amplitude over the transverse cross section at the entry and x is the dimensionless radius. The exact form of the function $f(x)$ was determined by us experimentally (Fig. 2) and with a high degree of accuracy could be regarded as Gaussian; the calculated value of α was $\frac{1}{2}$.

Bearing in mind that at an interface between two media there is partial reflection of the beam energy, we obtain the following expression between the field intensity on the beam axis and the average input intensity:

$$E_0^2=8\pi I_{av}(1-R_{refl})/cn_0,$$

where R_{refl} is the reflection coefficient of the entry face. In this case we can assume

$$R_{refl}=\left(\frac{n_0-1}{n_0+1}\right)^2.$$

The method in the form proposed above can be used under steady-state self-focusing conditions. In reality the intensity depends on time in accordance with a near-Gaussian law. This dependence results in scanning of the focal length of the nonlinear lens formed in the investigated layer. Therefore, a time integrated beam diameter $d=2b_m$ is obtained in the measurement plane and this diameter is the solution of the equation

$$I(r=b_m)=I(0)/e,$$

$$I(r)=\frac{cn_0}{8\pi^{1/2}}\frac{1}{\tau}\int_{-\infty}^{+\infty}\frac{E_0^2\exp(-t^2/\tau^2)}{f^2(z,t)}\exp\left\{-\frac{r^2}{a^2f^2(z,t)}\right\}dt,$$

where $I(0)=I(r)|_{r=0}$, τ is the pulse duration, and $f(z,t)$ is the solution of Eq. 1, where E_0^2 should be replaced with $E_0^2\exp(-t^2/\tau^2)$. Clearly, $b_m>b$ in the self-focusing case and $b_m<b$ in the case of self-defocusing. Num-

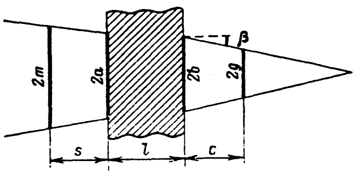

FIG. 1. Schematic diagram showing the measured diameters.

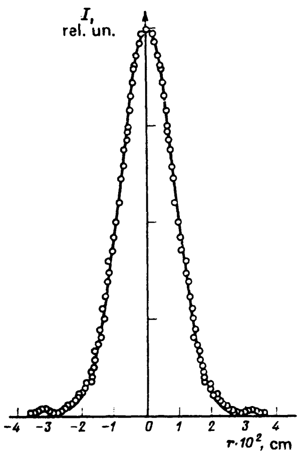

FIG. 2. Distribution of the intensity in a transverse cross section through a laser beam at the entry to a nonlinear medium: the points are the experimental results and the continuous curve is a Gaussian distribution.

erical calculations carried out for the values of the parameters satisfied by the conditions (1)–(3) indicate that the difference between the true radius b and the measured one b_m is slight, amounting to ~ 2% of b_m. Therefore, we can write $b = b_m \pm 0.02 b_m$, where the plus sign corresponds to self-defocusing and the minus to self-focusing of the beam.

EXPERIMENTS AND RESULTS OBTAINED

The proposed method for determining the nonlinearity coefficients was implemented using the apparatus shown schematically in Fig. 3. A Q-switched single mode ruby laser *1* produced pulses of $\tau = 20$ nsec duration. Frequency control, ensuring the emission of a single transverse mode, was based on the distribution of the intensity in the far field zone and on laser radiation interferograms. The instability of the laser operation was within the limits $\Delta P/P < 7\%$, $\Delta d/d < 3\%$, where P was the laser beam power and d was the beam diameter. The laser radiation was focused by a lens *3* ($F = 182$mm) on a sample *4* (crystal or a cell with a liquid) and the focus of the lens *3* was located behind the investigated substance so that all the singularities appearing in a nonlinear medium in the region of the focus were avoided. A system comprising a microscope and a photographic camera (*5* and *6*) made it possible to photograph the laser beam cross sections in various measurement planes. A set of calibrated filters *2* was used to vary the intensity of the laser beam reaching the sample but the intensity of the beam incident on a photographic film was kept constant irrespective of the size of the photographed cross section (this intensity was within the normal sensitivity range of the film). A calorimeter *7* was used to determine the laser radiation energy. The absolute error in the power determination was 15%.

The suitability of the method was checked by determining the nonlinearity coefficients n_2 of the refractive index of benzene and nitrobenzene, because these values had been determined earlier by many authors.[13,14] In these measurements we used a cell 10–15 mm long with benzene or nitrobenzene. The range of the parameters used to determine n_2 was $I_{av} = 100–600$ MW/cm^2 and $R_{in} = 30–20$ cm. The results of experimental determinations and machine calculation of n_2, by means of the expressions in Eq. (2), are presented in Table I. We can see that the average scatter of the experimental values of n_2 is 20% for benzene and 10% for nitrobenzene. It is also clear from Table I that the nonlinearity coefficients n_2 found by us are in good agreement with the data for benzene and nitrobenzene found in the literature. This confirms the correctness of our method for determining the nonlinearity of the refractive index in the restricted range under consideration.

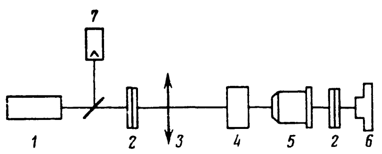

FIG. 3. Experimental setup: *1*) laser; *2*) neutral filters; *3*) lens; *4*) sample; *5*) microscope; *6*) photographic camera; *7*) calorimeter.

TABLE I.

Benzene		Nitrobenzene	
n_2(exp.) × 10^{12}, cgs esu	n_2(publ.) × 10^{12}, cgs esu	n_2(exp.) × 10^{12}, cgs esu	n_2(publ.) × 10^{12}, cgs esu
1.6	1.8[13] 0.7[14]	6.1	9.2[13] 4.9[14]

Next, we used the same method to determine the nonlinearity coefficients of the refractive index of a CdS semiconductor crystal. The input parameters of the beam were found to be as follows: $I_{av} = 1–30$ MW/cm^2, $R_{in} = 7–15$ cm. Machine analysis of the experimental data gave the following values of n_2 and n_4 for CdS crystal: $n_2 = 3.9 \times 10^{-11}$ cgs esu and $n_4 = -2.7 \times 10^{-15}$ cgs esu.

A theoretical estimate of the error in the determination of the values of n_2 for benzene and nitrobenzene gave 40% and for the CdS crystal in n_2 and n_4 did not exceed 50 and 60%, respectively. This was due to the fact that the greatest contribution to the error was the inaccuracy of the average input intensity and output diameter of the beam. The experimental scatter of the values of n_2 for a CdS crystal did not exceed 40% and the corresponding scatter of n_4 was ~ 50%.

Our values of the nonlinearity coefficients demonstrate a strong nonlinearity of the refractive index of the semiconducting CdS, which arises from the nonlinear polarizability of the valence (bound) electrons, represented by n_2, and from the contribution of nonequilibrium free electrons to the refractive index, represented by n_4.[1,2] These mechanisms can, in principle, give rise to values of the nonlinearity coefficients n of the kind found by us for the CdS crystal.

It follows from this investigation that the proposed method can be used to determine the nonlinearity coefficients of various material media used in practice and that the theory of Akhmanov *et al.*[9] describes satisfactorily the processes of propagation of laser beams in nonlinear media in a limited range of parameters. Moreover, our measurements demonstrate that semiconducting crystals of the CdS type are characterized by a strong nonlinearity of the refractive index and this may be of considerable practical importance.

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Čerenkov and transition radiations in the γ-resonance frequency region

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Coherent (Čerenkov and transition) radiation of a charged particle moving through a bounded medium with resonantly scattering nuclei is considered. It is shown that an essential role in the formation of radiation in the γ-resonance frequency region is played by allowance for the smallness of the radiation-absorption length in the medium, and that the interference between the Čerenkov and transition radiations in the vacuum must be taken into account without fail. Attention is called to the existence of a region of emission frequencies and angles in which the waves from the sections of particle motion in the medium and in the vacuum are in phase. The possible quantum yields of coherent emission in the γ band are presented for media containing the Mössbauer isotopes ^{57}Fe, ^{119}Sn, and ^{73}Ge.

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1. It is known that resonant scattering of γ quanta by nuclei exerts an appreciable influence on the character of the frequency dispersion and on the refractive index of the medium at frequencies close to the resonant frequency of the nucleus, since the coherent amplitude of forward nuclear scattering can be comparable and may even exceed the electronic scattering amplitude. It therefore becomes possible to obtain Čerenkov radiation[2,4] and to increase the transition radiation[1,3] (in an inhomogeneous medium) in the γ band in the case of a fast charged particles moving in a medium containing resonantly scattering nuclei.

The estimates of the intensity of the Čerenkov radiation in Refs. 2 and 4 pertain to an unbounded medium and describe the additional contribution made to the energy losses of the fast charged particle in the resonantly scattering medium, whereas practical interest attaches to coherent radiation emitted into vacuum (or into another weakly absorbing medium).

For such an analysis it becomes important to consider the following singularities of the case in question: the short absorption length in the γ-resonance frequency region, and the fact that the coherence lengths in vacuum and in the medium are comparable. Account must therefore be taken of the limited region in which the Čerenkov radiation is formed in the medium, and the interference between this radiation and the transition radiation in the vacuum.

Allowance for these circumstances, and the consequences ensuing from such an analysis, are in fact the subject of the present paper.

2. The refractive index of a medium at frequencies close to the γ-transition frequency in the nucleus can be expressed in the form[5,6]

$$n=1+\delta n_{el}+\delta n_{nuc}$$
$$\delta n_{el}=-Z_0 r_e N\lambda^2/2\pi,\quad \delta n_{nuc}=-4a\Delta/(\Delta^2+1)+i\cdot 4a/(\Delta^2+1);\tag{1}$$
$$a=N(2j'+1)f^2\lambda^3/32\pi^2(2j+1)(1+\alpha_0),$$

where δn_{el} is the characteristic part of the refractive index,[1)] δn_{nuc} is the nuclear component of the refractive index, r_e is the classical radius of the electron, N is the number of atoms per unit volume, $\lambda(\omega)$ is the wavelength (frequency) of the γ quantum, Z_0 is the atomic number, j' and j are respectively the spins of the excited and ground states of the nucleus, f is the Lamb-Mössbauer factor (assumed hereafter equal to unity), α_0 is the internal conversion coefficient, ω_0 is the frequency of the resonant level of the nucleus, Γ is the total width of the nuclear level, and $\Delta=2(\omega-\omega_0)/\Gamma$.

According to (1), for example, for a medium of ^{57}Fe (nuclear-transition energy $E_\gamma=14.4$ keV, $\Gamma=4.67\times10^{-9}$ eV) we have

$$\delta n_{el}=-7.5\cdot10^{-6},\quad a=3.37\cdot10^{-5},$$

for ^{119}Sn ($E_\gamma=23.9$ keV, $\Gamma=2.47\times10^{-8}$ eV)

$$\delta n_{el}=-1.71\cdot10^{-6},\quad a=4.61\cdot10^{-6}$$

and for ^{73}Ge ($E_\gamma=67.03$ keV, $\Gamma=2.45\times10^{-7}$ eV)

$$\delta n_{el}=-2.16\cdot10^{-7},\quad a=9.2\cdot10^{-7}.$$

To calculate a we used the data of Ref. 7 on the Mössbauer isotopes.

3. Let a particle with charge e move uniformly with

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