

# Components of the third-order nonlinear susceptibility tensors in KDP, DKDP and LiNbO<sub>3</sub> nonlinear optical crystals

I.A. Kulagin, R.A. Ganeev, R.I. Tugushev, A.I. Ryasnyansky, T. Usmanov

**Abstract.** The ratio of the components of the third order nonlinear susceptibility tensors (which are responsible for the self-action effects) in KDP, DKDP and LiNbO<sub>3</sub> crystals is measured at the 1064-nm wavelength of a Nd:YAG laser. Measurements were made on the basis of analysis of polarisation dependences of the focused radiation transmission by the  $z$ -scan method. The obtained polarisation dependences of the effective Kerr nonlinearity correspond to a tensor of the fourth rank that is symmetric in all its indices. The measured values of the Kerr nonlinearities that are responsible for the self-action of o-polarised radiation were found to be  $(1.75 \pm 0.55) \times 10^{-14}$  and  $(2.4 \pm 0.8) \times 10^{-13}$  esu for KDP and LiNbO<sub>3</sub> crystals, respectively.

**Keywords:** nonlinear optical crystals, nonlinear susceptibility, anisotropic media.

## 1. Introduction

The most efficient frequency converters and devices for controlling radiation parameters are based on second-order nonlinear optical processes (second-harmonic generation, parametric oscillation and amplification of light, frequency mixing, electrooptical effect, etc.). The development of the technology of generation and amplification of femtosecond laser pulses and the corresponding enhancement of the radiation intensity necessitate the inclusion of higher-order nonlinear effects influencing the second-order nonlinear optical processes [1, 2]. The most significant among such higher-order effects is the self-action of laser radiation, i.e., processes associated with the variation in the refractive index and absorption of a medium in a strong light field.

Despite the wide use of nonlinear optical crystals, their higher-order nonlinearities have not been studied in detail. Third-order nonlinear susceptibilities were earlier determined only for certain spectral regions and only for certain crystal orientations (see, for example, Refs [3–5]).

The anisotropy of Kerr nonlinearities was analysed much less frequently [6].

In this study, we have analysed and measured the ratios of the components of third-order nonlinear susceptibilities of KDP, DKDP and LiNbO<sub>3</sub> crystals at a wavelength of 1064 nm on the basis of the investigation of polarisation dependences of the transmission of focused radiation by the  $z$ -scan method.

## 2. Basic relations

If radiation of frequency  $\omega$  is present in the medium, the components of the third-order nonlinear polarisation tensor can be written in the form

$$P_i^{(3)}(\omega) = \frac{3}{4} \chi_{ijkl}^{(3)}(\omega = -\omega + \omega + \omega) E_j^* E_k E_l, \quad (1)$$

where  $E_{i,j,k,l}$  are the field intensities of the interacting waves. The tensor  $\chi_{ijkl}^{(3)}$  of the fourth rank possesses the frequency-permutation symmetry properties [7]. In particular, such a tensor is symmetric to the permutation of the last pair of indices. If frequency dispersion is neglected, tensor  $\chi_{ijkl}^{(3)}$  is completely symmetric. Obviously, such an approximation can be used away from the spectral regions of radiation absorption.

To analyse the propagation of linearly polarised radiation in anisotropic media, the field amplitude can be decomposed into orthogonally polarised components, and the interaction of ordinary (o) and extraordinary (e) waves can be analysed using the equation

$$\begin{aligned} \frac{\partial E_i}{\partial z} + \beta_i \frac{\partial E_i}{\partial x} - \frac{i}{2k_i} \Delta_{\perp} E_i \\ = -i \sum_{j,k,l} \gamma_{ijkl} E_j^* E_k E_l \exp(-i\Delta k z) - \alpha_i E_i, \end{aligned} \quad (2)$$

$$i, j, k, l = o, e,$$

where  $k_i = (2\pi/\lambda)n_i$  is the wave number;  $\gamma_{ijkl} = [g\pi^2/(\lambda n_i)]\chi_{ijkl}^{(3)}$  are the nonlinear coupling coefficients;  $\beta_i$  is the birefringence angle;  $\alpha_i$  is the linear absorption coefficient;  $\Delta k = k_i + k_j - k_k - k_l$  is the phase mismatch;  $\Delta_{\perp} = \partial^2/\partial x^2 + \partial^2/\partial y^2$ ;  $g = 1, 3, 6$  is the degree of degeneracy;  $n_i$  is the refractive index; and  $\lambda$  is the radiation wavelength. Note that the tensor  $\chi_{ijkl}^{(3)}$  is given in the

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coordinate system of the laser radiation beams. It is assumed in Eqn (2) that second-order processes do not have a significant effect on the wave propagation (the phase-matching conditions are not satisfied for the process of second harmonic generation, i.e.,  $\Delta k_2 \gg (8\pi\chi^{(2)})^2/(3n_0 \times \lambda\chi^{(3)})$ ). It follows from Eqn (2) that the measurement of nonlinear susceptibility in anisotropic media is complicated due to birefringence.

In the presence of birefringence, the orthogonally polarised components propagate at an angle to each other because of the lateral shift, and the interaction of various beam regions varies along the longitudinal coordinate. Terms in the sum in Eqn (2) describe the self-action ( $i = j = k = l$ ), cross-action ( $i = l \neq j = k$ ), amplification ( $i = j \neq l = k$ ) and generation ( $i \neq j = l = k$ ) of the  $i$ th component. In addition to nonlinearity, the contribution of each of the terms is determined by the phase-mismatch  $\Delta k$  and the amplitude of the corresponding field components. Thus, the contribution from various nonlinear polarisation terms can be separated by selecting the necessary orientation of the nonlinear crystal and the intensity of radiation.

If the effect of the terms describing the amplification and generation of the corresponding components is insignificant, and if the redistribution of the light beam intensity due to nonlinear refraction (when the length of the medium is smaller than the diffraction length) and nonlinear absorption in a nonlinear medium are disregarded, the self-action and cross-action effects change the space–time distribution of the radiation phase. In this case, the field distribution at the exit from a uniaxial nonlinear medium can be described by a simple expression

$$E_i(z, x, y, t) = (1 - R_i)^{1/2} E_{i0}(z, x, y, t) \times \exp\{-\alpha_i L/2 - i[\gamma_{iiii} L_{\text{eff}} I_i(z, x, y, t) + \gamma_{ijij} \int e^{-\alpha_j z} I_j(z, x + \beta_j z, y, t) dz]\}, \quad (3)$$

where  $L_{\text{eff}} = [1 - \exp(-\alpha_i L)]/\alpha_i$  is the effective length of nonlinear interaction;  $L$  is the length of the medium; and  $R_i$  is its reflectivity. Analysis of this equation leads to the conclusion that the contribution of components with different polarisations to the overall field distribution is determined by the birefringence angles, radiation intensity and the length of the nonlinear medium, as well as the spatial distribution of the incident laser radiation. For a sufficiently long medium, the lateral drift leads to a decrease in the effective interaction length of the orthogonally polarised components.

Expression (3) was used as the boundary condition for reconstructing the distribution of the field  $E_r(z, x, y, t)$  in the radiation-detector region in the  $z$ -scan method with a limiting aperture. The intensity distributions were obtained by using the Gaussian function expansion employed traditionally in the  $z$ -scan method [8].

The normalised transmission coefficient  $T(z)$  at the limited aperture of the detector was determined from the relation

$$T(z) = \int_{-\infty}^{\infty} \int_{x_1}^{x_2} \int_{y_1}^{y_2} \{|E_{\text{or}}(z, x, y, t)|^2 \cos^2 \psi + |E_{\text{er}}(z, x, y, t)|^2 \sin^2 \psi\} dx dy dt \times \left[ \int_{-\infty}^{\infty} \int_{x_1}^{x_2} \int_{y_1}^{y_2} |E_{\text{or}}(z, x, y, t)|^2 dx dy dt \right]^{-1}, \quad (4)$$

where  $E_{\text{or}}(z, x, y, t)$  is the field distribution of the o-polarised radiation in the detector region in the absence of nonlinearities in the crystal, and  $\psi$  is the angle defining the direction of polarisation. Expression (4) describes the dependence of the transmission coefficient on the position of the nonlinear crystal relative to the beam waist.

The type of the tensor  $\chi_{ijkl}^{(3)}$  is determined by the crystal symmetry. In the dispersionless approximation, the third-order Kerr nonlinearities in KDP crystals (symmetry class  $\bar{4}2m$ ) and LiNbO<sub>3</sub> crystals (symmetry class  $3m$ ) have four independent components. For radiation directed at angles  $\theta$  and  $\varphi$  to the crystal axes, the Kerr nonlinearities for a crystal of the symmetry class  $\bar{4}2m$  are expressed in terms of these components as:

$$\chi_{\text{oooo}}^{(3)} = \chi_{11} - \frac{1}{2}(\chi_{11} - 3\chi_{12}) \sin^2 2\varphi, \quad (5)$$

$$\chi_{\text{oooe}}^{(3)} = \chi_{\text{eooo}}^{(3)} = \frac{1}{2}(\chi_{11} - 3\chi_{12}) \sin^2 2\varphi \cos^2 \theta + \chi_{12} \cos^2 \theta + \chi_{23} \sin^2 \theta, \quad (6)$$

$$\chi_{\text{eeee}}^{(3)} = \chi_{\text{oooo}}^{(3)} \cos^4 \theta + \frac{3}{2}\chi_{23} \sin^2 2\theta + \chi_{33} \sin^4 \theta, \quad (7)$$

while the Kerr nonlinearities for a crystal of the symmetry class  $3m$  have the form

$$\chi_{\text{oooo}}^{(3)} = 3\chi_{12}, \quad (8)$$

$$\chi_{\text{oooe}}^{(3)} = \chi_{\text{eooo}}^{(3)} = \chi_{12} \cos^2 \theta + \chi_{14} \sin 3\varphi \sin 2\theta + \chi_{23} \sin^2 \theta, \quad (9)$$

$$\chi_{\text{eeee}}^{(3)} = 3\chi_{12} \cos^4 \theta - 4\chi_{14} \sin 3\varphi \sin \theta \cos^3 \theta + \frac{3}{2}\chi_{23} \sin^2 2\theta + \chi_{33} \sin^4 \theta. \quad (10)$$

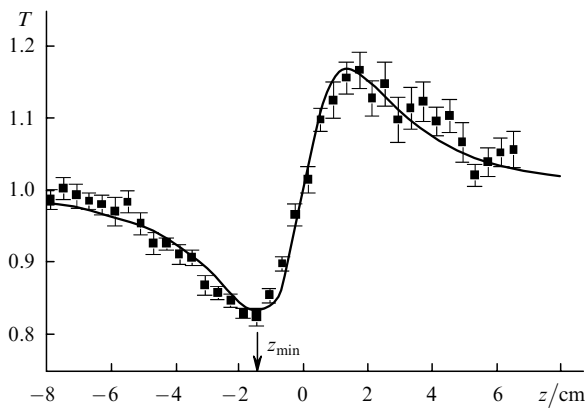
Expressions (5) and (6) have been written using the concise form of notation of tensors of the fourth rank with the help of two indices [7]. One can see from (5)–(7) and (8)–(10) that the anisotropy of the Kerr nonlinearity is determined by the relation between the tensor components and may be quite significant. To analyse this relation, we studied experimentally the nonlinear susceptibilities of crystals cut at various angles for different values of the ratio of the o- to e-components of linearly polarised laser radiation in the  $z$ -scan method. The  $z$ -scan method is a comparatively simple single-beam method of measuring nonlinear optical parameters and is based on an analysis of the variation in transmission in a sample placed in the beam waist using a detector whose field of vision is limited by the aperture [8].

### 3. Experimental

We used in the experiments a picosecond Nd : YAG laser ( $\lambda = 1064$  nm,  $\tau = 55$  ps,  $W = 2$  mJ) with a pulse repetition rate of 2 Hz [9]. The measuring scheme differed from the one used in Ref. [9] in that a polarisation rotator was mounted in front of the focusing lens of focal length 25 cm.

The nonlinear susceptibility tensors were measured for nonlinear KDP crystals ( $L = 1$  and 2 cm,  $\theta = 90^\circ$  and  $59^\circ$ ), a DKDP crystal ( $L = 1$ ,  $\theta = 0$ ), and a LiNbO<sub>3</sub> crystal ( $L = 0.8$  cm,  $\theta = 90^\circ$ ). The polarisation dependences of nonlinear susceptibilities were measured in the following way. When a sample length is smaller than the diffraction length and the nonlinear absorption is absent, the difference between the maximum and minimum values of the normalised transmission coefficient as a function of the sample position is proportional to the Kerr nonlinearity. Analysis of the variation in the maximum or minimum value of transmission for different radiation polarisations makes it possible to determine the relation between effective Kerr nonlinearities because the transmission dependence on the sample position in the absence of nonlinear absorption is symmetric with respect to the beam waist ( $z = 0$ ). The effective nonlinear susceptibility here means the nonlinearity determined for arbitrary crystal orientations and radiation polarisation directions.

The point  $z_{\min}$  at which the transmission coefficient has its smallest value (Fig. 1) was determined in the experiments by scanning the sample along the  $z$  coordinate. Figure 1 shows typical experimental and theoretical dependences of the normalised transmission coefficient on the sample position (a 1-cm-long KDP crystal cut at angles  $\theta = 90^\circ$  and  $\varphi = 45^\circ$ ). The sample was placed at a given point and the transmission was measured as a function of the laser radiation polarisation direction.



**Figure 1.** Typical experimental (squares) and theoretical (curve) dependences of the normalised transmission coefficient  $T$  on the position of a KDP crystal ( $\theta = 90^\circ$  and  $\varphi = 45^\circ$ ).

The processing of results involved averaging of the measured values over a certain number of points ( $N = 20$ ), as well as rejection of individual measurements obtained in the case of strong intensity fluctuations of the incident radiation. This enabled us to lower considerably the overall measuring errors in sample transmission at each point. A PC was used to control the operation of the measuring system.

### 4. Results and discussion

Crystals cut at various angles were used for determining the relation between nonlinear susceptibility components. Figure 2 shows the dependence of transmission on the polarisation angle  $\psi$  in a KDP crystal ( $L = 1$  cm), cut at angles  $\theta = 90^\circ$  and  $\varphi = 45^\circ$ , for various radiation intensities. One can see that an increase in the intensity leads to a decrease in transmission, while the amplitude of oscillations of these dependences remains almost unchanged. In this crystal, the birefringence angle for the e-polarised component is zero. The o- and e-polarised components have different refractive indices, and the phase mismatch is significant for the amplification and generation of new components. Estimates of the contribution from various terms in the sum in Eqn (2) show that the effect of generation and amplification of radiation components in the field of the orthogonal component can be neglected for the radiation intensities ( $I < 10^{11}$  W cm<sup>-2</sup>) used in measurements. In this case, if the condition of a thin medium ( $L < R_d$ , where  $R_d$  is the diffraction length of radiation) is satisfied [8], the minimum transmission coefficient  $T_{\min}$  as a function of angle  $\psi$  can be described, taking Eqns (2)–(4) into account, by the expression:

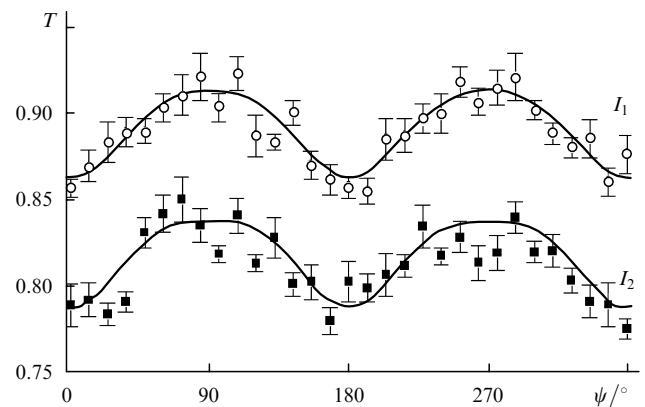
$$T_{\min}(\psi) = C(\psi) + \mu\rho^2 S(\psi) - \Gamma [\chi_{oooo}^{(3)} C^2(\psi) + 2(l + \mu) \times \rho\chi_{oeoe}^{(3)} C(\psi)S(\psi) + l\mu\rho^2\chi_{eeee}^{(3)} S^2(\psi)], \quad (11)$$

where

$$C(\psi) = \cos^2 \psi; \quad S(\psi) = \sin^2 \psi; \quad \Gamma = \frac{3\pi^2 b s}{\lambda n_o} |E_0|^2 L_{\text{oeff}};$$

$$\rho = \frac{1 - R_e}{1 - R_o}; \quad \mu = \exp[(\alpha_o - \alpha_e)L]; \quad l = \frac{n_o L_{\text{oeff}}}{n_e L_{\text{oeff}}};$$

$b$  and  $s$  are coefficients determined by using the standard  $z$ -scan method ( $b = 0.203$  and  $s = 1/\sqrt{2}$  for a Gaussian time intensity distribution [8]). The solid curves in Fig. 2 are obtained from expression (11) by the method of least squares. We used the refractive indices  $n_o = 1.4954$  and  $n_e = 1.4604$  in the calculations, and the reflection coefficients calculated from expressions presented in Ref. [10]. In addition, the absorption coefficients at 1054 nm [10] were



**Figure 2.** Dependences of the normalised transmission coefficient  $T$  of a KDP crystal ( $\theta = 90^\circ$  and  $\varphi = 45^\circ$ ) on the polarisation angle  $\psi$  for the intensity ratio  $I_1 : I_2 = 0.65 : 1$  and  $I_1 = 54$  GW cm<sup>-2</sup>.

also used ( $\alpha_o = 0.058 \text{ cm}^{-1}$  and  $\alpha_e = 0.02 \text{ cm}^{-1}$ ). The above dependences and expressions (5)–(7) lead to the relation

$$0.5\chi_{11} + 1.5\chi_{12} = 2.34\chi_{23} = 1.11\chi_{33} \quad (12)$$

between the components of the nonlinear susceptibility tensor.

The error in this relation was determined by the error of measurement and the error of processing of the results of measurements. The error amounted to about 2% in most measurements.

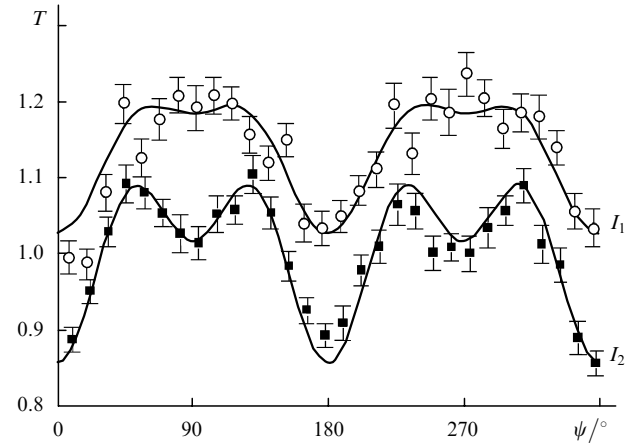
The Kerr nonlinearity  $\chi_{oooo}^{(3)}$  measured with the help of expression (11) was  $(1.75 \pm 0.55) \times 10^{-14}$  esu. It should be recalled that  $\chi^{(3)}$  (in esu) =  $(9/4\pi) \times 10^8 \chi^{(3)}$  (in SI units). The maximum error in the absolute measurement of the Kerr nonlinearities ( $\pm 30\%$ ) was introduced by the error in determining the radiation intensity, which depends mainly on the error in measuring the size of the focal spot (or diffraction length) and the duration of laser radiation pulses. In order to reduce the error, the focal spot size was estimated more precisely by using the known relation between the diffraction length (equal to 1.6 cm in our case) and the separation between the transmission maximum and minimum, obtained by using the traditional  $z$ -scan method for sample lengths smaller than the diffraction length (i.e., for a thin medium [8]).

To determine the relation between the components  $\chi_{11}$  and  $\chi_{12}$  of the nonlinear susceptibility tensor, we used a DKDP crystal cut along the crystal axis. In this case, the refractive indices of the orthogonally polarised components are equal, which may lead to the generation of an orthogonally polarised component (or, in other words, to the rotation of radiation polarisation in a nonlinear medium). For the nonlinear susceptibility responsible for self-action of the linearly polarised radiation, expression (5), in which  $\varphi$  is the angle between the direction of polarisation and the crystal axis  $X$ , remains valid. The expression for the nonlinear susceptibility responsible for the generation of the orthogonal component, will have the form

$$\chi_{\perp}^{(3)} = \frac{1}{4}(\chi_{11} - 3\chi_{12}) \sin 4\varphi. \quad (13)$$

It follows from this relation that the nonlinearity is equal to zero for  $\varphi = m\pi/4$ . According to Eqn (5), measurement of the relation between Kerr nonlinearities for  $\varphi = 0$  and  $45^\circ$  leads to a relation between  $\chi_{11}$  and  $\chi_{12}$ . The Kerr nonlinearities were measured by analysing the dependence of transmission coefficient on the sample position relative to the beam waist. The ratio of nonlinearities measured by turning the polarisation angle by  $45^\circ$  was 0.71, and hence the relation between the nonlinear susceptibility components is  $\chi_{12} = 0.14\chi_{11}$ .

Figure 3 shows the dependences of the normalised transmission on the polarisation angle for a KDP crystal of length 2 cm cut at angles  $\theta = 59^\circ$  and  $\varphi = 0$  for various radiation intensities. Under these conditions, the effect of birefringence leads to a spatial ‘breakdown’ of the ordinary and extraordinary transverse components of the beam. In the case under study, the characteristic size of the region of interaction of orthogonally polarised beams  $a/\beta_i \sim 0.2$  cm (where  $a$  is the beam radius), which is much smaller than the length of the crystal. Thus, the middle term in the brackets in (11) can be neglected for intensities  $I < 10^{11} \text{ W cm}^{-2}$ .



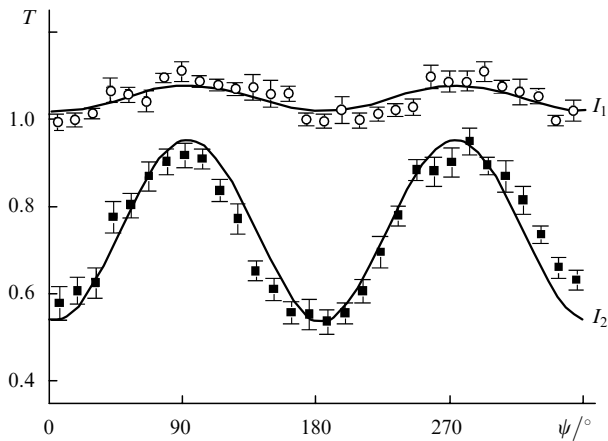
**Figure 3.** Dependences of the normalised transmission coefficient  $T$  of a KDP crystal ( $\theta = 59^\circ$  and  $\varphi = 0$ ) on the polarisation angle  $\psi$  for the intensity ratio  $I_1 : I_2 = 0.45 : 1$  and  $I_1 = 16 \text{ GW cm}^{-2}$ .

The relative displacement of beams leads to different positions of the highest intensity regions of the orthogonally polarised components at the detector. For a limited aperture of the detector, this leads to a variation in the oscillation amplitude. Hence, in the presence of birefringence, the coefficient  $\rho$  also depends on the geometry of the experiment. One can see from Fig. 3 that the variation in the coefficient  $\rho$  due to birefringence under these conditions increases the amplitude of oscillations. The solid curves show the dependences obtained for  $\mu\rho^2 = 1.13$  and for the above relation between the nonlinear susceptibility tensor components. It also follows from Fig. 3 that the theoretical and experimental dependences are in good agreement with each other for various intensities.

Thus, for an appropriate orientation of the crystal, the effect of the Kerr nonlinearities leads to the additional modulation of the polarisation dependences of high-intensity radiation transmission. The agreement between the experimental and theoretical dependences obtained for various angles of crystal orientation and for various intensities of laser radiation suggests that the dispersionless approximation can be used to analyse the tensor properties of Kerr nonlinearities in the investigated spectral range. Note that the photon energy in this range is one-sixth the band gap in the KDP crystal.

We also analysed the anisotropy of the Kerr nonlinearity in  $\text{LiNbO}_3$  crystals. Figure 4 shows the dependences of transmission on the radiation polarisation angle in a  $\text{LiNbO}_3$  crystal of length 0.8 cm cut at an angle  $\theta = 90^\circ$ . The relatively small linear absorption coefficients for the  $\text{LiNbO}_3$  crystal ( $\alpha_o = 0.0019 - 0.0023 \text{ cm}^{-1}$  and  $\alpha_e = 0.0014 - 0.0019 \text{ cm}^{-1}$  [10]) must not lead to significant oscillations of such dependences at low intensities. However, the large difference in the reflectivities of the orthogonally polarised components leads to an increase in the amplitude of such oscillations. Calculations were made using the refractive indices obtained with the help of the dispersion formulas [10] ( $n_o = 2.2337$  and  $n_e = 2.1495$ ). The oscillation amplitude increases with intensity. An analysis of these dependences using expression (11) leads to the following relation between the Kerr nonlinearity components:

$$\chi_{12} = 1.2\chi_{23} = 1.4\chi_{33}. \quad (14)$$



**Figure 4.** Dependences of the normalised transmission coefficient  $T$  of a  $\text{LiNbO}_3$  crystal ( $\theta = 90^\circ$ ) on the polarisation angle  $\psi$  for the intensity ratio  $I_1 : I_2 = 0.042 : 1$  and  $I_1 = 1.5 \text{ GW cm}^{-2}$ .

The Kerr nonlinearity obtained from (11) for the o-polarised radiation component  $\chi_{oooo}^{(3)}$  as  $(2.4 \pm 0.7) \times 10^{-13}$  esu.

The above parameters and relations between nonlinearity components can be used to determine various types of third-order nonlinearities, including the nonlinearities responsible for the self-action of radiation, cross-action of radiation at different frequencies, as also for frequency conversion.

In addition to the nonlinearities, values of the nonlinear index  $n_2 = (3\pi/n_o)\chi_k^{(3)}$  are also given in the literature [11]. It must be noted that various interpretations are given for nonlinearities and the nonlinear index. In this work, we have used the concepts developed in Refs [8, 11]. The nonlinear index  $n_2$  or  $\gamma$  are introduced with the help of the following relations [8]:

$$n = n_o + n_2 \frac{|E_0|^2}{2} = n_o + \gamma I, \quad (15)$$

where  $E_0$  is the field amplitude. As a rule, the quantity  $n_2$  is measured in the esu system, while  $\gamma$  is measured in  $\text{m}^2 \text{ W}^{-1}$  (or  $\text{cm}^2 \text{ W}^{-1}$ ). These two coefficients are connected through the relation  $n_2 = cn_o\gamma/(40\pi)$  (here, the velocity of light  $c$  and  $\gamma$  are taken in the SI system) [8]. It should also be noted that the coefficient  $\gamma$  is denoted as  $n_2$  in some publications.

For the  $\text{LiNbO}_3$  crystal ( $\theta = 90^\circ$ ), we obtained the following values:  $n_2 = 10.1 \times 10^{-13}$  esu ( $\gamma = 1.9 \times 10^{-15} \text{ cm}^2 \text{ W}^{-1}$ ) and  $n_2 = 2.5 \times 10^{-13}$  esu ( $\gamma = 0.49 \times 10^{-15} \text{ cm}^2 \text{ W}^{-1}$ ) for o- and e-polarised radiation, respectively. The value  $n_2 = 4.8 \times 10^{-13}$  esu measured in Ref. [12] is close to the values obtained by us. At the same time, the value  $\gamma = 8.5 \times 10^{-15} \text{ cm}^2 \text{ W}^{-1}$  [13] estimated in the periodic structure of  $\text{LiNbO}_3$  from an analysis of frequency conversion upon optical parametric oscillation using pumping at 800 nm is much higher than the value measured by us in  $\text{LiNbO}_3$  single crystals. Apart from structural variations, this may be due to a comparatively large dispersion in  $\text{LiNbO}_3$ . The nonlinearity of this crystal in the 532-nm region is about an order of magnitude higher than its nonlinearity at 1064 nm [12].

The values of  $n_2$  calculated from the measured values of nonlinearities and relations between the nonlinearity com-

ponents, as well as under the assumption that these relations are identical in KDP and DKDP crystals, were found to be  $1.1 \times 10^{-13}$  and  $1.0 \times 10^{-13}$  esu for o- and e-polarised radiation, respectively. The corresponding values of  $\gamma$  were  $3.1 \times 10^{-16}$  and  $2.9 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$ , respectively. Within the limits of the measuring error, these values are close to the values of  $n_2$  measured in Ref. [3] for a KDP crystal ( $0.78 \times 10^{-13}$  and  $0.72 \times 10^{-13}$  esu for o- and e-polarised radiation, respectively) and to the values of  $\gamma$  obtained in Ref. [14] ( $3.2 \times 10^{-16}$  and  $2.84 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$ ) for a DKDP crystal. The cubic nonlinearity determined by us for a KDP crystal ( $\theta = 0$ ) for radiation polarised along the crystal axis  $X$  was found to be equal to  $2.5 \times 10^{-14}$  esu. The nonlinearity determined in Ref. [15] for a KDP crystal with the same orientation was  $1.43 \times 10^{-14}$  esu. Taking into account the error ( $\pm 50\%$ ) in such measurements made while determining the critical self-focusing power, the agreement between the results can be considered satisfactory in this case also.

Using Miller's dispersion rule [ $\chi_{ijkl}^{(3)}/(\chi_{ii}^{(1)}\chi_{jj}^{(1)}\chi_{kk}^{(1)}\chi_{ll}^{(1)}) = \delta_{ijkl}$ , where  $\delta_{ijkl}$  is a constant independent of frequency], we can estimate the nonlinear parameters of transparent materials in various spectral ranges. For  $\theta = 90^\circ$  and  $\varphi = 45^\circ$ , the coefficients  $\gamma$  calculated from our measurements and responsible for cross-action of the 800-nm radiation with the 400-nm radiation were found to be  $3.5 \times 10^{-16}$  and  $3.1 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$  for o- and e-polarised radiation, respectively. The values of  $\gamma$  measured in [16] for DKDP crystals were  $(4.35 \pm 0.97) \times 10^{-16}$  and  $(4.61 \pm 0.72) \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$  for o- and e-polarised radiation, respectively. Within the limits of the measuring error, the values of  $\gamma$  for o- and e-polarised radiation are close to each other.

It can be concluded from the above comparison that the results obtained by us explain the difference in the absolute values of the nonlinearities measured in Refs [3] and [16] for various orientations of nonlinear crystals.

## 5. Conclusions

We have analysed the polarisation dependences of transmission of focused radiation in nonlinear crystals at 1064 nm. The observed dependences can be attributed to the effect of Kerr nonlinearities. An analysis of these dependences leads to the relations between the Kerr nonlinearity tensor components for KDP, DKDP and  $\text{LiNbO}_3$  crystals. It is shown that in the given spectral range, Kerr nonlinearities in KDP crystals can be described in terms of the dispersionless model. The components of the nonlinear susceptibility tensor and the nonlinearities themselves can be determined for various angles of crystal orientation from the measured Kerr nonlinearities. The obtained results were compared with the results of measurements made by other authors.

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