Light-induced gyrotropy and anisotropy in a cubic crystal

A. M. Danishevskiĭ, S. F. Kochegarov, and V. K. Subashiev

(Submitted 1 May 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **33**, No. 12, 625–629 (20 June 1981)

We have detected light-induced gyrotropy and anisotropy affects in a cubic InAs crystal. These effects are due to cubic nonlinearity. The components of the nonlinear susceptibility tensor $\chi^{(3)}_{xyxy} \approx 4.6 \times 10^{-8}$ cgs unit and $\chi^{(3)}_{xxyy} = 2.9 \times 10^{-8}$ cgs units at the frequency of the probing radiation $\Omega = 3.92 \times 10^{14}$ sec^{-1} (pump radiation frequency is $\omega = 1.96 \times 10^{14}$ sec^{-1} and $E_g = 0.4$ eV) have been calculated in the approximation of quasispherical crystal symmetry.

PACS numbers: 79.20.Dj

1. Studies of the variation of radiation polarization in a material medium, which is caused by nonlinear optical interactions, make it possible to investigate high-order nonlinear susceptibilities.[1]

In our work we have detected a light-beam-induced gyration and birefringence in a cubic crystal of the $\bar{4}3$m class (InAs). Unlike in Ref. 2, where the self-stimulation-induced variation of the polarization of high-intensity radiation in a GaAs crystal was studied, we have investigated, just as in Ref. 3, the polarization variation of a probing beam of linearly polarized light with a frequency Ω, which is caused by the action on the crystal of the high-intensity, linearly or circularly polarized pump radiation of frequency ω.

The effect was predicted theoretically[4] for circularly polarized pump radiation. This effect is similar to the Faraday effect. The only difference between them is that in our case the circularly polarized light beam plays the role of the magnetic field.

2. Ignoring spatial dispersion and limiting our consideration to cubic nonlinearity, we can write

$$P_i^{NL}(\Omega) = \chi^{(3)}_{ijkl}(\Omega;\ \omega, -\omega, \Omega) E_j(\omega)\, E_k^*(\omega) \mathcal{E}_e(\Omega) \tag{1}$$

for the nonlinear polarization at frequency Ω.

For the linearly polarized pump radiation, which propagates in the z direction that coincides with the [001] axis of the crystal and with the polarization vector along the x axis, and for the probing beam, which propagates opposite to the pumping beam,

$$P_x^{NL}(\Omega) = \chi^{(3)}_{xxxx} E_x^2(\omega) \mathcal{E}_x(\omega)\ ; \quad P_y^{NL}(\Omega) = \chi^{(3)}_{yxxy} E_x^2(\omega) \mathcal{E}_y(\Omega). \tag{2}$$

This polarization results in the appearance of nonlinear increments in the index of refraction; assuming that the nonlinear increments to the index of refraction are small, we obtain

$$\delta n_x \approx \frac{2\pi(\chi^{(3)}_{xxxx} E_x^2(\omega))}{n_0(\Omega)}; \qquad \delta n_y \approx \frac{2\pi\chi^{(3)}_{yxxy} E_x^2(\omega)}{n_0(\Omega)};$$

$$\delta n = \delta n_x - \delta n_y = \frac{2\pi E_x^2(\omega)}{n_0(\Omega)} (\chi^{(3)}_{xxxx} - \chi^{(3)}_{yxxy}). \tag{3}$$

The effect of the variation of E_x^2 with z due to linear absorption$^{1)}$ can be taken into account by averaging over z,

$$\overline{\delta n} = \frac{16\pi^2 j_0 \hbar\omega}{n_0^2(\Omega) c} \; \frac{1 - e^{-\alpha(\omega)d}}{\alpha(\omega)\, d} \; (\chi^{(3)}_{xxxx} - \chi^{(3)}_{yxxy}), \tag{4}$$

where j_0 is the intensity of the pump radiation at the entrance face of the crystal in $\mathrm{cm}^{-2}\ \mathrm{sec}^{-1}$. It follows from Eq. (3) that generally the linearly polarized probing radiation transmitted through the crystal becomes elliptically polarized.

For circularly polarized pump radiation, which propagates along the z axis, $\mathbf{E}(\omega,z) = (E_0/\sqrt{2})(\mathbf{e}_x + i\mathbf{e}_y)$. The rotation of the polarization plane of the probing light, just as the Faraday effect, is attributed to the appearance of the imaginary part of the nondiagonal component of the dielectric constant $\mathrm{Im}\,\epsilon_{ik}$.[5] The rotation angle can be determined from the formula

$$d\theta/dz = (\Omega/2cn_0)\mathrm{Im}\epsilon_{ik}. \tag{5}$$

Thus, using Eq. (1) we can write

$$\theta(\Omega,\ \omega) = \frac{8\pi^2\Omega j_0 \hbar\omega}{n_0^2(\Omega) c} \; \frac{1 - e^{-\alpha(\omega)d}}{\alpha(\omega)} \; (\chi^{(3)}_{xyxy} - \chi^{(3)}_{xxyy}). \tag{6}$$

3. The experiments were performed on n-type InAs with a density of $n = 1.6 \times 10^{16}\ \mathrm{cm}^{-3}$. The 0.15 cm-thick sample was made in the form of a wedge-shaped plate with a 5° angle in order to avoid modulated interference of the probing beam.[6] The crystal was not oriented. The pulsed radiation of a CO_2 laser with a wavelength of 9.5 μm served as the pump, and a beam split off from the main beam or a reflected beam, which was converted to the second harmonic, was used for the probing. The pump and probing-radiation beams, which were directed toward each other, were combined by using a small diaphragm (~300 μm), which was attached to the sample on the side of the probing-beam incidence. The beams passed through the sample at a small angle ζ (in the sample $\zeta < 0.5°$); this made it possible to separate them outside the sample and record the beams. A polarizer was placed in the path of the probing beam in front of the sample, and an analyzer was placed in front of the detector.

Figure 1 shows the dependence of the probing-radiation flux I_p intercepted by the photodetector on the rotation angle ϕ of the analyzer without pumping and with pumping of left-hand and right-hand circular polarization ($\sigma_\pm$). It can be seen that

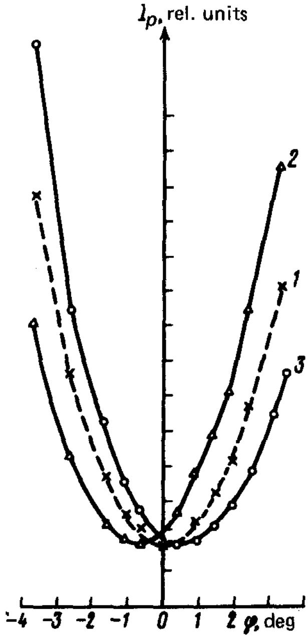

FIG. 1. Dependences of the probing-radiation flux I_p at the photodetector on the analyzer rotation angle ϕ for circular pumping: InAs, $n = 1.6 \times 10^{16}$ cm^{-3}, $T = 90$ K, $d = 0.15$ cm, $\omega = 1.96 \times 10^{14}$ sec^{-1}, and $\Omega = 3.92 \times 10^{14}$ sec.$^{-1}$. 1, x; $j_0 = 0$, and $I_p(\phi = 0)/I_p(\phi = \pi/2) = 7 \times 10^{-4}$; 2 and 3, $j_0 \approx 1.2$ MW/cm^2; σ_+ and σ_- pump-radiation polarizations.

the polarization plane of the probing beam is rotated as a result of the action of the pumping with polarizations σ_+ or σ_- on the crystal. The induced gyration constant is $\xi_{\text{exp}} = (1.46 \pm 0.06) \times 10^{-27}$ cm sec.

For linearly polarized pumping (Fig. 2) the appearance of ellipticity was observed in the probing beam. The degree of introduced ellipticity depends strongly on the polarization direction of the pump radiation. The maximum observed ellipticity corresponded to $\overline{\delta n} = 1.2 \times 10^{-4}$ at $j_0 = 1.76 \times 10^{25}$ cm^{-2} sec^{-1}.

4. The relations (2)-(6) presented in Sec. 2 were obtained for a crystal of cubic symmetry. In InAs the isoenergy surfaces in **k** space for the bands, which give the major contribution to the cubic nonlinearity, are very close to spherical.[7] Therefore, an analysis of the presented experimental data can be made on the basis of a quasispherical (isotropic) model. Thus, by choosing the z axis as the propagation direction of the light beams, it is convenient to write the nonlinear polarization in the form

$$\mathbf{P}^{NL}(\Omega) = \chi^{(3)}_{xyyx}(\mathbf{E}^*(\omega)\mathbf{E}(\omega))\,\vec{\mathcal{E}}(\Omega) + \chi^{(3)}_{xxyy}(\mathbf{E}^*(\omega)\,\vec{\mathcal{E}}(\Omega))\mathbf{E}(\omega) + \chi^{(3)}_{xyxy}(\mathbf{E}(\omega)\,\vec{\mathcal{E}}(\Omega))\mathbf{E}^*(\omega)\,. \tag{7}$$

It can be seen that the first term in Eq. (7) provides only an isotropic contribution to the index of refraction, while the other two terms determine the induced anisotropy and gyrotropy in the crystal. In this case it is possible to use Eqs. (4) and (6) for $\overline{\delta n}$ and θ after supplementing them with the condition that

$$\chi^{(3)}_{xxxx} - \chi^{(3)}_{yxxy} = \chi^{(3)}_{xyxy} + \chi^{(3)}_{xxyy}\,. \tag{8}$$

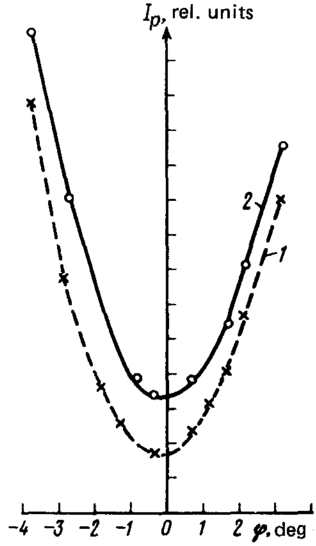

FIG. 2. Dependences of the probing-radiation flux I_p at the photodetector on the analyzer rotation angle ϕ for linear pump polarization: InAs, $n = 1.6 \times 10^{16}$ cm^{-3}, $T = 90$ K, $d = 0.15$ cm, $\omega = 1.96 \times 10^{14}$ sec^{-1}, and $\Omega = 3.92 \times 10^{14}$ sec^{-1}. 1, $j_0 = 0$ and $I_p(\phi = 0)/I_p(\phi = \pi/2) = 7 \times 10^{-4}$; 2, $j_0 = 0.37$ MW/cm^2; the polarization direction of the pump radiation is at an angle of $\pi/4$ to the polarization direction of the probing radiation.

Using the experimental value of $\overline{\delta n}_{\max}$, we obtain $\chi^{(3)}_{xyxy} + \chi^{(3)}_{xxyy} = (7.4 \pm 0.4) \times 10^{-8}$ cgs units, and starting from the given value of $\xi_{\exp}$, we can calculate $\chi^{(3)}_{xyxy} - \chi^{(3)}_{xxyy} = (1.7 \pm 0.06) \times 10^{-8}$ cgs units. Therefore, $\chi^{(3)}_{xyxy} \approx 4.6 \times 10^{-8}$ cgs units and $\chi^{(3)}_{xxyy} \approx 2.9 \times 10^{-8}$ cgs units.

5. It was shown in Ref. 8, where an interaction of the type $\omega_3 = 2\omega_1 - \omega_2$ between two CO$_2$-laser emission lines in InAs was investigated, that the nonlinear susceptibility $\chi^{(3)}_{xxxx}$ is caused primarily by the conduction electrons because of the nonparabolic shape of the band, i.e., $\chi^{(3)} \approx \chi^{(3)}_n$. On the other hand, only the contribution of valence electrons was taken into account in Ref. 4, where the induced gyration constant (IGC) was calculated. In this case the IGC increases resonantly as $\kappa = (\omega + \Omega)/E_g \to 1$. There should be no resonance for $\chi^{(3)}_n$.[8]

We performed experiments to measure the IGC—$\xi_{\exp}$—at $\kappa = 0.58$ and $\kappa = 0.97$. In the second case $\xi_{\exp}$ was 2.6 times larger than in the first. According to Perlin's calculations,[4] a much stronger dependence can be expected. This discrepancy apparently indicates that the contributions of the free and valence electrons to the nonlinear susceptibility $\chi^{(3)}$ are of the same order of magnitude for the studied crystal. For a more detailed investigation of this problem, we plan to perform experiments on samples with different concentrations of free charge carriers.

The authors thank E. L. Ivchenko and I. P. Areshev for a useful discussion of the questions raised in this paper.

1)The small polarization variation of pump radiation caused by self-stimulation is ignored.

---

1. S. A. Akhmanov and V. I. Zharikov, Pis'ma Zh. Eksp. Teor. Fiz. 6, 644 (1967) [JETP Lett. 6, 137,(1967)].

2. A. I. Kovrigin, D. V. Yakovlev, B. V. Zhdanov, and N. I. Zheludev, Opt. Commun. **35**, 92 (1980).
3. K. Kubota, J. Phys. Soc. Jpn. **29**, 986 (1970); **29**, 999 (1970).
4. E. Yu. Perlin, Fiz. Tverd. Tela **22**, 66 (1980) [Sov. Phys. Solid State **22**, 38 (1980)].
5. L. D. Landau and E. N. Lifshitz, Elektrodinamika sploshnykh sred (Electro-dynamics of Continuous Media), GIFML, Moscow, 1959.
6. I. P. Areshev, A. M. Danishevskii, S. F. Kochegarov, and V. K. Subashiev, Opt. Spektrosk. **48**, 975 (1980) [Opt. Spectrosc. 48, 534 (1980)].
7. F. Stern, Bull. Am. Phys. Soc. **2**, 347 (1957).
8. C. K. N. Patel, R. F. Slusher, and P. A. Fleury, Phys. Rev. Lett. **17**, 1011 (1966); S. Iha and N. Bloembergen, Phys. Rev. **171**, 891 (1968).

Translated by Eugene R. Heath
Edited by S. J. Amoretty