Nonlinear optical activity in crystals

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We report on the first investigation of the effect of nonlinear optical activity (NOA) in crystals (dependence of the angle of rotation of the polarization plane on light intensity) which is dependent on the electronic nonlinearity. Measurements are made in $LiIO_3$ crystals. Electronic NOA is separated from the thermal effect background and is $(1\pm0.6)\times10^{-11}$ deg·cm·w^{-1}.

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1. The object of this article is to present results of an experimental investigation involving the first study of nonlinear optical activity (NOA) in crystals (dependence of the angle of rotation of the polarization plane on light intensity) which is dependent on "fast" electronic nonlinearity.

Electronic NOA as identified in lithium iodate crystals in the background of slower thermal effects; the nonlinear rotation constant, measured at $\lambda=0.532$ μm, was $(1\pm0.6)\times10^{-11}$ deg·cm·W^{-1}.

Measurement of NOA dispersion may, in our opinion, be a new effective method in nonlinear polarization spectroscopy of condensed media which yields unique information on the combined effects of anharmonicity and spatial dispersion.

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2. The NOA effect was predicted in Ref. 1; phenomenologically, it is based on taking into account third-order terms of the optical field intensity in an expression for the polarization. The following holds for a medium with spatial dispersion, to within third-order field terms:

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \gamma_{ijk}^{(1)} \frac{\partial E_j}{\partial x_k} - \gamma_{ijkl}^{(2)} E_j \frac{\partial E_k}{\partial x_l} + \gamma_{ijklm}^{(3)} E_j E_k \frac{\partial E_l}{\partial x_m}, \quad (1)$$

where the terms containing $\gamma^{(1)}$, $\gamma^{(2)}$, and $\gamma^{(3)}$ correspond, clearly, to the terms $\chi^{(1)}$, $\chi^{(2)}$, and $\chi^{(3)}$.

NOA is associated with nonlinear susceptibility $\gamma^{(3)}$; in the case of a gyrotropic cubic crystal the following obvious formula—using standard designations—may be written:

$$\mathbf{D} = \epsilon_0 \mathbf{E} - if^L(\omega)[\mathbf{k}\mathbf{E}] + if^{NL}|\mathbf{E}|^2[\mathbf{k}\mathbf{E}],$$

where

$$f^L(\omega) = \mathrm{Re}\{\gamma^{(1)}(\omega)\}, \qquad f^{NL}(\omega) = \mathrm{Re}\{\gamma^{(3)}(\omega)\}. \quad (2)$$

Here the term responsible for NOA has the same structure as that responsible for linear activity (LOA).

A detailed analysis of the tensor $\gamma^{(3)}$ is given in Ref. 2. The angles of linear and nonlinear rotation may be expressed as follows:

$$\phi^L = \frac{k^2 l}{2n_0} f^L(\omega), \qquad \phi^{NL} = \frac{k^2 l}{2n_0} f^{NL}(\omega)|\mathbf{E}|^2. \quad (3)$$

3. The same physical mechanisms contributing to self-interactions, described by the cubic nonlinear susceptibility $\chi^{(3)}$, contribute to NOA in the general case. In this connection, NOA can be called polarization self-interaction, the simplest manifestation of which is the heating of a medium by the laser beam. This mechanism was studied in Ref. 3, yielding results that are similar to those obtained for the temperature measurements of LOA.[1] Coarse estimates of $\gamma^{(3)}$ (and, consequently, f^{NL}) which depend on electronic nonlinearity may be made for the following simple reasons. Away from the absorption bands, $\gamma^{(3)} \sim \chi^{(3)} a$, where a is the characteristic lattice dimension.[2] In the case of typical nonlinear crystals in the optical region $\gamma^{(3)} \approx 10^{-20}$–$10^{-22}$ CGSE may be expected. Substituting this value into Eq. (3) yields a value of intensity $I = 10^8$ W/cm^2 and $l = 1$ cm, $\phi^{NL} \approx 3 \times 10^{-15}$–$3 \times 10^{-7}$ rad.

Thus, isolation of the intrinsic electronic NOA from the background of thermal effects and parasitic signals caused by frequency instability and LOA dispersion, is a difficult task in itself.[3] To solve this problem, we developed a two-channel pulsed polarimeter capable of measuring a change in the angle of rotation of the polarization plane during a single laser pulse, $\Delta\phi = 5\times10^{-5}$ rad ($\tau = 10^{-8}$ sec).

As the test object we used the $LiIO_3$, crystal which is characterized by a relatively high susceptibility $\chi^{(3)} = 3\times10^{-12}$ CGSE.

4. The NOA effect was studied in a 62-mm long $LiIO_3$ crystal at the second-harmonic frequency ($\lambda = 0.532\ \mu$m) of a monochromatic YAG : Nd laser. The experimental set-up is shown in Fig. 1.

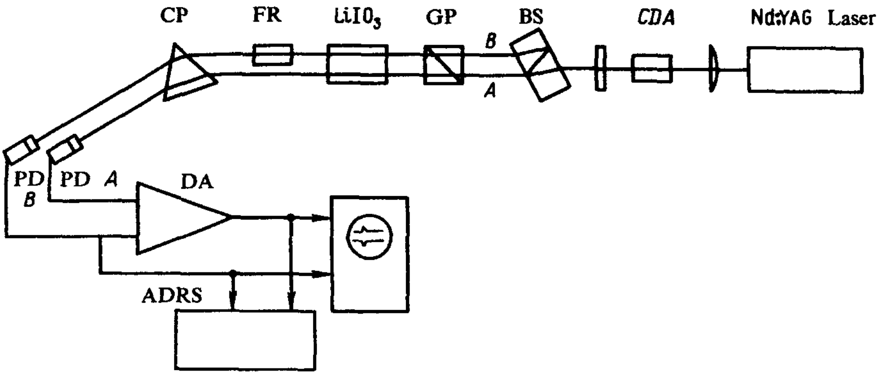

FIG. 1. Bloc diagram of the experimental setup.

The laser output was converted to the second harmonic (SH) by means of a CDA crystal, resulting in SH pulse width of 10 nsec and having the maximum energy $W_2 = 10$ mJ. The SH output was divided by the beam splitter (BS) into two parallel beams A and B with the intensity ratio $I_A/I_B = 30$. After being polarized by the Glann prism (GP), both beams were transmitted through the $LiIO_3$ crystal along the optical axis and, subsequently, through the Cotton prism (CP), a polarization analyzer. In addition to these, channel A contained a Faraday rotator (FR) that compensated for the difference of the linear rotation of the polarization planes of beams A and B which arises due to a difference in the optical path length in the two channels. The electrical part of the recording system consists of a differential amplifier (DA), two-beam oscilloscope and a two-channel pulsed analog-digital recording system (ADRS) with an output printer. Silicon photodiodes (PD) were used as photoreceivers. The nonlinear rotation effect was measured as a function of variations in the ratio of optical intensities in channels A and B produced by increasing the power of the beam propagating in the crystal. The sensitivity of the apparatus used in the NOA observations using the described method is characterized by a level of parasitic signals, associated with LOA, which did not exceed 8×10^{-5} rad.

5. Figure 2 shows the characteristic shape of signals generated at the polarimeter output (maximum beam intensity $I = 300$ MW/cm^2). The first pulses are constrained by electronic NOA; the effect was accompanied by the laser radiation depolarization effect (distinguishable from the NOA signal by changing the position of the working point of the polarimeter) which accumulated with each new laser burst and was,

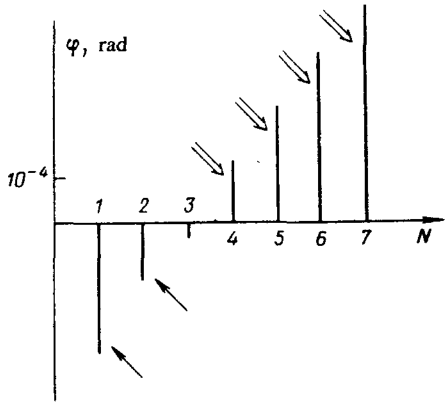

FIG. 2. Signals at the output of two-channel pulsed polarimeter after successive firing of YAG : Nd laser (→NOA signals, ⇒ change in signal sign, associated with optically-induced depolarization in a crystal).

clearly, associated with inhomogeneous heating of the crystal by the laser radiation. As the intensity decreases, so does the depolarization with the characteristic time $\tau_2 = 1$ sec which confirms its thermal origin. The effect of thermal rotation during a single laser pulse did not exceed 10% of the observed NOA effect in our experiments and was characterized by an opposite sign.

The experimental value of the optical rotation constant is $(1 \pm 0.6)\times 10^{-11}$ deg·cm·W^{-1}, and the effective value of the $\gamma^{(3)}$ tensor component (the beam propagating along the optical axis) is

$$\gamma^{(3)} = (6 \pm 4)\cdot 10^{-21} \text{ CGSE}.$$

Although as the NOA effect in $LiIO_3$ is relatively small and measuring it presents some difficulty, the generation of considerably greater effects may be expected. It suffices to point out that the susceptibility in crystals in the vicinity of biexciton resonances is $\chi^{(3)} \approx 10^{-8}$ CGSE (see Ref. 9).

The study of NOA in liquid crystals is of considerable interest; the use of tunable lasers will permit the development of NOA as a new method of nonlinear polarization spectroscopy which simultaneously yields information concerning anharmonicity and spatial dispersion.

Finally, the relation between NOA and the quadratic electrogyration effect is of considerable interest from the standpoint of crystal spectroscopy. A comparison of this kind is not possible at present since the measurement of NOA and quadratic electrogyration were made using different crystals.[10]

1) An interesting manifestation of the thermal effect in the dependence of the Faraday rotation on laser light intensity was first shown in Ref. 4.

2) This relation is actually used for a coarse evaluation of constants of linear gyration. Naturally, this is not a sufficient reason in support of this method of evaluation of the nonlinear constants; in this connection, it should be pointed out that such an evaluation is indirectly supported in Refs. 5–7 where the susceptibility $\gamma^{(2)}$ which describes the quadrupole generation of the second optical harmonic was determined.

3) In our opinion incomplete exclusion of the foregoing effects has affected the accuracy of results described in Ref. 8.

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