Nonlinear optical rotation in magnetic crystals

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Nonlinear optical rotation in magnetic crystals is theoretically investigated. The various Faraday mechanisms of the interaction of electromagnetic waves which give rise to the effect are considered. The dependence of the rotation angle of the polarization plane of the incident light on the external magnetic field is studied.

1. INTRODUCTION

Nonlinear optical rotation (NOR) consists in the rotation of the polarization plane (equivalently, of the principal axis of the polarization ellipse) of a wave as it passes through a nonlinear crystal. This rotation depends on the intensity of the radiation. In magnetic media the NOR angle contains a contribution associated with the magnetization and is usually identified with the nonlinear Faraday effect. The essence of this latter effect can be understood from simple heuristic considerations. The Verde constant V, defined as the ratio of the optical rotation angle β to the length of the medium z and the magnitude of the external magnetic field H_0, begins to depend on the intensity I of the electromagnetic wave at large values of this intensity. Expanding V in powers of I and stopping at the linear term, we obtain

$$V(I)=V+\alpha I. \tag{1}$$

Then

$$\beta=V(I)H_0z=\beta_{\mathrm{L}}+\beta_{\mathrm{NL}}, \tag{2}$$

where β_L and β_{NL} are the linear and nonlinear Faraday rotation angles, defined by

$$\beta_L=VH_0z, \quad \beta_{NL}=\alpha IH_0z. \tag{3}$$

As follows from formulas (3), the angle β_{NL} is proportional to the length of the medium in the direction of propagation of the wave, the magnitude of the external magnetic field, and the intensity of the light. These basic conclusions were given a theoretical and experimental basis in Refs. 1–3, which investigate the diluted magnetic semiconductor $Cd_{0.75}Mn_{0.25}Te$.

In actual fact, in crystals the situation should be substantially more complicated as a result of the following circumstances:

1. The phenomenon is due to self-interaction of the light, which is described by tensors of fourth and higher rank, whose symmetry is lower than that of the tensor associated with linear optical rotation. This can manifest itself in an orientational dependence of the effect, i.e., the NOR angle becomes a function of the orientation of the azimuth of the polarization of the incident wave relative to the crystallographic axes.[4,5]

2. As is well known, magnetic crystals are gyrotropic. Therefore the orientation of the polarization of the light wave in the crystal depends on the coordinate of the wavefront and the magnitude of the constant magnetic field. A consequence of this should be a nonmonotonic, oscillating dependence of the NOR angle both on the dimensions of the crystal in the direction of propagation and on the magnitude of the magnetic field.

3. In analogy to the self-induced nonlinear rotation of the polarization plane in nonmagnetic, naturally gyrotropic crystals,[6,7] in magnetic media the combination of linear magnetic gyration and nonlinear Kerr refraction should contribute to such rotation.

4. The wave obtained in experiments with linearly polarized light possesses a finite ellipticity.[8] It should therefore be possible to observe self-rotation of the polarization ellipse,[9] which, generally speaking, also depends on the intensity of the magnetic field and which can be erroneously interpreted as rotation of the polarization plane.

In the present paper we theoretically investigate the properties of NOR in magnetic crystals placed in a constant magnetic field in the direction of which propagates an electromagnetic wave. We base our treatment on the phenomenological approach in which the intensity of the electric field of the light wave is found from the wave equation, after which its polarization characteristics—the polarization azimuth and ellipticity are determined in the usual way. We consider the case in which the incident wave has arbitrary polarization and propagates along the symmetry axis of a cubic or highly symmetric single-axis crystal. The contributions of the various interaction mechanisms of the wave to the NOR angle are estimated and its dependence on the external magnetic field is studied for the diluted magnetic semiconductor $Cd_{0.75}Mn_{0.25}Te$.

2. FIELD OF THE WAVE IN A NONLINEAR CRYSTAL

Let us consider the crystal-vacuum interface, which we take to be located in the xy plane. The z axis, along which the external magnetic field $\mathbf{H}_0$ is taken to be oriented, is

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directed into the medium. We investigate the case in which an intense electromagnetic wave is normally incident from vacuum onto the surface of the crystal. Then the electric field of the light wave in the medium $\mathbf{E}$ $(E_x, E_y, 0)$ is found from the equation

$$\frac{d^2\mathbf{E}(z)}{dz^2} = \frac{\omega^2}{c^2}[\mathbf{E}(z) + 4\pi\mathbf{P}^L(z) + 4\pi\mathbf{P}^{NL}(z)], \tag{4}$$

where $\mathbf{P}^L(z)$ and $\mathbf{P}^{NL}(z)$ are the linear and nonlinear polarization vectors of the crystal

$$P_i^L(z) = (\chi_{ij}^{(1)} + \chi_{ijz}^{(1)EH} H_0) E_j, \tag{5}$$

$$P_i^{NL}(z) = (\chi_{ijkl}^{(3)} + \chi_{ijklz}^{(3)EH} H_0) E_j E_k E_l^*, \tag{6}$$

and the subscripts i,j,k,l take the values x,y. In formulas (5) and (6) $\hat{\chi}^{(1)}, \hat{\chi}^{(1)EH}$ and $\hat{\chi}^{(3)}, \hat{\chi}^{(3)EH}$ are the linear and cubic nonlinear optical and magnetooptical susceptibility tensors.

Assuming the nonlinear polarization to be small, we solve Eq. (4) in the prescribed field approximation, in which the vector $\mathbf{P}^{NL}$ is a known function of the field $\mathbf{E}^0(z)$, which is found by solving the corresponding linear problem:

$$\frac{d^2\mathbf{E}^0(z)}{dz^2} = \frac{\omega^2}{c^2}[\mathbf{E}^0(z) + 4\pi\mathbf{P}^L(z)]. \tag{7}$$

After transforming in this equation to the circular variables $E_\pm(z) = E_x(z) \pm iE_y(z)$, taking (5) into account, it splits into two independent equations:

$$\left[\frac{d^2}{dz^2} + (k_0^2 + i\alpha_0 \pm f)\right] E_\pm^0(z) = 0. \tag{8}$$

Here

$$k_0^2 + i\alpha_0 = \omega^2 c^{-2}(1 + 4\pi\chi^{(1)}), \quad \chi^{(1)} = \chi_{xx}^{(1)} = \chi_{yy}^{(1)}$$

$$f = 4\pi\omega^2 c^{-2}\,\mathrm{Im}(\chi_{xyz}^{(1)EH}) H_0.$$

In addition, we have assumed that $\mathrm{Re}(\hat{\chi}_{xyz}^{(1)EH}) = 0$, which corresponds to neglecting linear dichroism.

The solutions of system of equations (8) must satisfy the boundary conditions

$$E_\pm(0) = E_\pm \exp(i\delta_\pm), \tag{9}$$

where $E_\pm$ and $\delta_\pm$ are the amplitudes and phases of the circular components of the field upon entrance to the crystal. The angle $\beta_0 = (\delta_+ - \delta_-)/2$ defines the orientation of the polarization of the incident wave relative to the X axis. In addition, taking the crystal to be semi-infinite, we require that $E_\pm(z) \to 0$ as $z \to \infty$. Fulfillment of this condition is ensured by the finiteness of α_0, which defines the linear absorption of the radiation. Assuming for simplicity the latter to be weak, after solving Eqs. (8) and (4), in what follows we set this parameter equal to zero.

Integrating Eqs. (8) gives

$$E_\pm(z) = E_\pm \exp i(k_\pm z + \delta_\pm), \tag{10}$$

where $k_\pm^2 = k_0^2 \pm f$.

In the current approximation the problem of determining the electric field of the wave in a nonlinear crystal reduces to solving the following system of equations:

$$\left[\frac{d^2}{dz^2} + k_0^2 \pm f\right] E_\pm(z) = 4\pi\omega^2 c^{-2} P_\pm^{NL}(E_\pm^0(z)),$$

and upon integrating these we obtain

$$E_\pm(z) = C_1^{(\pm)} \exp(ik_\pm z) + C_2^{(\pm)} \exp(-ik_\pm z)$$
$$- 4\pi\omega^2(c^2 k_\pm)^{-1} \int_0^z P_\pm^{NL}[E_\pm^0(z')]$$
$$\times \sin(k_\pm(z - z'))]dz'. \tag{11}$$

Here $P_\pm^{NL} = P_x^{NL} + iP_y^{NL}$, and the conditions for determining the constants $C_1^{(\pm)}$ and $C_2^{(\pm)}$ are the same as were used to solve Eq. (8).

Taking relations (10) into account, we can represent the circular components of the nonlinear polarization in the form

$$P_\pm^{NL} = \sum_{m=1}^{4} A_m^{(\pm)} \exp(iq_m^\pm z), \tag{12}$$

where $q_1^\pm = k_\pm$, $q_2^\pm = 2k_\pm - k_\mp$, $q_3^\pm = k_\mp$, $q_4^\pm = 2k_\mp - k_\pm$, and the coefficients $A_m^{(\pm)}$ do not depend on z and are combinations of the nonlinear susceptibility and the components of the field strength $E_\pm(0)$ at the front face of the crystal. Substituting (12) into (11) and determining the integration constants $C_1^{(\pm)}$ and $C_2^{(\pm)}$, we find

$$E_\pm(z) = E_\pm \exp[i(k_\pm z + \delta_\pm)] + 4\pi\omega^2 c^{-2} \sum_{m=1}^{4} A_m^{(\pm)}$$
$$\times [(q_m^\pm)^2 - k_\pm^2)]^{-1} [\exp(iq_m^\pm z) - \exp(ik_\pm z)]$$
$$= E_\pm^0(z)[1 + i\Delta_\pm(z)]. \tag{13}$$

Formulas (13) thus solve for the wave field in a nonlinear magnetic crystal. Their explicit form requires that we specify the values of the coefficients $A_m^{(\pm)}$, which depend critically on the symmetry properties of the medium. For simplicity, let us restrict the discussion to cubic crystals or those uniaxial crystals in which only the following "transverse" components of the nonlinear optical and magnetooptical susceptibilities are nonvanishing[10]:

$$\chi_{xxxx}^{(3)} = \chi_{yyyy}^{(3)} = \chi_1^{(3)}, \quad \chi_{xyxy}^{(3)} = \chi_{xxyy}^{(3)} = \chi_2^{(3)},$$

$$\chi_{xyyx}^{(3)} = \chi_3^{(3)}, \quad \chi_{xxxyz}^{(3)EH} = \Gamma_1,$$

$$\chi_{yxxxz}^{(3)EH} = \Gamma_2, \quad \chi_{xxyxz}^{(3)EH} = \Gamma_3.$$

In this case the coefficients $A_m^{(\pm)}$ are given by

$$A_1^{(\pm)} = \tfrac{1}{2} E_\pm^3 \exp(i\delta_\pm)[(\chi_1^{(3)} + 2\chi_2^{(3)} - \chi_3^{(3)})$$
$$\pm iH_0(\Gamma_1 + \Gamma_2 - 2\Gamma_3)] + \tfrac{1}{4} E_\mp^2 E_\pm$$
$$\times \exp(i\delta_\pm)[\chi_1^{(3)} + \chi_3^{(3)} \pm iH_0(\Gamma_2 - \Gamma_1)],$$

$$A_4^{(\pm)}=\tfrac{1}{2}E_\mp^2E_\pm \exp(i(2\delta_\mp-\delta_\pm))$$
$$\times[(\chi_1^{(3)}-2\chi_2^{(3)}-\chi_3^{(3)})\pm iH_0(\Gamma_1+\Gamma_2+2\Gamma_3)], \tag{14}$$
$$A_2^{(\pm)}=A_3^{(\pm)}=0.$$

Substituting these $A_m^{(\pm)}$ into (13), we obtain expressions for the nonlinearity parameters $\Delta_\pm(z)$:

$$\Delta_\pm(z)=\pi\omega^2(2c^2)^{-1}\{E_\pm k_\pm^{-1}[(\chi_1^{(3)}+2\chi_2^{(3)}-\chi_3^{(3)})$$
$$\pm iH_0(\Gamma_1+\Gamma_2-2\Gamma_3)]z+2E_\mp k_\pm^{-1}$$
$$\times[\chi_1^{(3)}+\chi_3^{(3)}\pm iH_0(\Gamma_2-\Gamma_1)]z\pm iE_\mp k_\pm^{-1}$$
$$\times\exp(i(2\delta_\mp-\delta_\pm))[(\chi_1^{(3)}-2\chi_2^{(3)}-\chi_3^{(3)})$$
$$\pm iH_0(\Gamma_1+\Gamma_2+2\Gamma_3)]$$
$$\times[\exp(\mp i2\Delta kz)-1](2\Delta k)^{-1}, \tag{15}$$

where $\Delta k=k_+-k_-$.

Expressions (13)–(15) can serve as a basis for determining the polarization of a wave that has passed through the crystal.

3. NONLINEAR OPTICAL ROTATION

The azimuth of the polarization ellipse β of the wave in the crystal is given by the relation

$$\mathrm{tg}(2\beta)=\frac{\mathrm{Im}(E_+E_-^*)}{\mathrm{Re}(E_+E_-^*)} \tag{16}$$

and in accordance with formulas (13)–(15) can be represented in the form

$$\beta=\beta_0+\beta_L+\beta_{NL}. \tag{17}$$

Here $\beta_L=\Delta kz/2$ is the linear Faraday rotation angle, and the nonlinear contribution to the optical rotation angle is given by

$$\beta_{NL}=\tfrac{1}{2}\,\mathrm{Re}(\Delta_+(z)-\Delta_-(z)). \tag{18}$$

Substituting the values of $\Delta_\pm(z)$ from (15) into formula (18), we find that the nonlinear rotation angle β_{NL} can be represented as a sum of six terms:

$$\beta_{NL}=KI_0z[\phi_1(z)+\phi_2(z)+\phi_3(z)+\theta_1(z)$$
$$+\theta_2(z)+\theta_3(z)]. \tag{19}$$

Here $K=4\pi^2\omega(n_0c^2)^{-1}$, I_0 is the intensity of the incident radiation, n_0 is the index of refraction, and the quantities $\phi_i(z)$, $\theta_i(z)$, $i=1,2,3$, characterize the contributions to NOR from the various interaction mechanisms of the waves.

The first term $\phi_1(z)$ takes account of rotation due to anisotropy of two-photon absorption in a crystal with a linear Faraday effect, and is given by

$$\phi_1(z)=\mathrm{Im}(\Delta\chi^{(3)})\mathrm{sinc}(\Delta kz)\sin[4\alpha(z)], \tag{20}$$

where the nonlinear anisotropy parameter $\Delta\chi^{(3)}=\chi_1^{(3)}-2\chi_2^{(3)}-\chi_3^{(3)}$, $\mathrm{sinc}(x)=x^{-1}\sin(x)$, and $\alpha(z)=\beta_0+(\Delta kz)/4$.

Since in what follows we will investigate the dependence of the NOR angle on the magnetic field, we separate out from Eq. (20) that part of $\phi_1(z)$ that depends on H_0. The corresponding contribution to the effect is given by

$$\tilde{\phi}_1(z)=\mathrm{Im}(\Delta\chi^{(3)})\{\mathrm{sinc}(\Delta kz)\sin[4\alpha(z)]$$
$$-\sin(4\beta_0)\}. \tag{21}$$

The term

$$\phi_2(z)=-H_0\{\mathrm{Im}(3\Gamma_2-2\Gamma_3-\Gamma_1)-\mathrm{Im}(\Gamma_1+\Gamma_2$$
$$+2\Gamma_3)$$
$$\times\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)]\} \tag{22}$$

gives the inherently nonlinear Faraday effect, which by analogy with the corresponding linear phenomenon we define as the optical rotation proportional to the magnetooptical susceptibility. In the case of weak linear Faraday rotation, when $\Delta kz\ll 1$, formula (22) coincides with the corresponding expression from Ref. 2.

The contribution

$$\phi_3(z)=\{-\mathrm{Re}(3\chi_1^{(3)}+2\chi_2^{(3)}+\chi_3^{(3)})+\mathrm{Re}(\Delta\chi^{(3)})$$
$$\times\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)]\}\Delta k(2k_0)^{-1} \tag{23}$$

is due to a combination of the two effects: nonlinear refraction and linear gyrotropy, the latter of which is produced in the crystal by the static magnetic field.

The terms $\theta_1(z)$ are proportional to the ellipticity ψ_0 of the incident wave, and are absent for linearly polarized light. The value of $\theta_1(z)$ defines the rotation of the polarization ellipse due to the anisotropy of two-photon absorption and is given by

$$\theta_1(z)=-\mathrm{Im}(\Delta\chi^{(3)})\mathrm{sinc}(\Delta kz)$$
$$\times\sin[4\alpha(z)]\Delta k(2k_0)^{-1}\psi_0. \tag{24}$$

The term

$$\theta_2(z)=\{\mathrm{Im}(\Gamma_2+2\Gamma_3-3\Gamma_1)-\mathrm{Im}(\Gamma_1+\Gamma_2+2\Gamma_3)$$
$$\times\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)]\}H_0\psi_0\Delta k(2k_0)^{-1} \tag{25}$$

describes the rotation of the polarization ellipse due to the magnetooptical nonlinearity of the crystal, and the term

$$\theta_3(z)=-\{\mathrm{Re}(\chi_1^{(3)}-2\chi_2^{(3)}+3\chi_3^{(3)})$$
$$+\mathrm{Re}(\Delta\chi)\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)\}\psi_0 \tag{26}$$

defines the ordinary Maker–Terhune–Savage effect[9] in a gyrotropic crystal. In contrast to $\theta_{1,2}(z)$, at $H_0=0$ the quantity $\theta_3(z)$ is different from zero. Therefore, as was done for the term $\phi_1(z)$, in formula (26) we separate out the part that depends on H_0:

$$\tilde{\theta}_3(z)=-\mathrm{Re}(\Delta\chi^{(3)})\psi_0\{\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)]$$
$$-\cos(4\beta_0)\}. \tag{27}$$

It should be pointed out that in crystals of the symmetry classes $3m$, 32, $\bar{3}m$, $6m2$, $\bar{6}mm$, 622, $6/mmm$, the nonlinear anisotropy parameter $\Delta\chi^{(3)}=0$ and, consequently, the quantities $\phi_1(z)$, $\theta_1(z)$, $\tilde{\theta}_3(z)$, and the corresponding NOR angles are identically zero. For crystals of the symmetry classes $4mm$, $\bar{4}2m$. 422, $4/mmm$, $\bar{4}3m$, 432, $m3m$, 23, $m3$ the nonlinear anisotropy parameter is nonzero, and all six of the indicated mechanisms normally contribute to the effect.

4. SOME ESTIMATES. DEPENDENCE OF THE NOR ANGLE ON THE MAGNETIC FIELD

We now compare the relative contributions to the NOR angle associated with the various interaction mechanisms of the waves and on this basis establish the dependence of the effect on the magnitude of the magnetic field.

We will obtain estimates for a crystal of $Cd_{0.75}Mn_{0.25}Te$, which has the zinc blende structure (symmetry $\bar{4}3m$),[11] in which the Verde constant $V=1.4\cdot10^{-4}$ cm^{-1} G^{-1} and $k_0=2\cdot10^5$ cm^{-1} at the frequency of the neodymium laser $\omega=1.7\cdot10^{15}$ s^{-1} (Ref. 2). For optical cubic nonlinear susceptibilities we can set[12]

$$|\mathrm{Im}(\Delta\chi^{(3)})/\mathrm{Re}(\Delta\chi^{(3)})|\approx0.5,$$

$$|\mathrm{Re}(\Delta\chi^{(3)})/\mathrm{Re}(\chi^{(3)})|\approx0.1,$$

and for the nonlinear magnetooptical susceptibility it is natural to adopt the following estimate:

$$H_0\,\mathrm{Im}(\Gamma)/\mathrm{Re}(\chi^{(3)})\sim H_0\,\mathrm{Im}(\chi^{(1)EH})/\mathrm{Re}(\chi^{(1)})\sim\Delta k k_0^{-1}. \tag{28}$$

In addition, we will evaluate the most interesting case, namely when $\Delta kz=2VH_0z\sim1$ and the linear Faraday rotation is of the order of tens of degrees.

Given the indicated assumptions, from formulas (21)–(23) we have

$$|\tilde{\phi}_1(z)|/|\phi_2(z)|\sim|\mathrm{Im}(\Delta\chi^{(3)})|/|H_0\,\mathrm{Im}(\Gamma)|\sim5\times10^{-2}(\Delta k)^{-1}k_0, \tag{29}$$

$$|\phi_2(z)|/|\phi_3(z)|\sim1. \tag{30}$$

In fields $H_0\approx5\cdot10^3$ G, the ratio (29) is of the order of 10^4 and, consequently, in crystals in which $\Delta\chi^{(3)}\neq0$ the main contribution to the nonlinear rotation angle is associated with the anisotropy of two-photon absorption.

Similarly, with the help of formulas (24)–(27) we have

$$|\tilde{\theta}_1(z)|/|\theta_2(z)|\sim|\mathrm{Im}(\Delta\chi^{(3)})|/|H_0\,\mathrm{Im}(\Gamma)|\sim10^4, \tag{31}$$

$$|\tilde{\theta}_1(z)|/|\theta_3(z)|\sim|\mathrm{Im}(\Delta\chi^{(3)})/\mathrm{Re}(\Delta\chi^{(3)})|\sim0.5. \tag{32}$$

Thus, as in nonmagnetic media, nonlinear rotation of the polarization ellipse is due mainly to nonlinear refraction.

Finally, let us estimate the contribution to the nonlinear rotation angle associated with the finite ellipticity of the incident wave. Comparison of the largest of the quantities $\phi_i(z)$ and $\theta_i(z)$ gives

$$|\tilde{\phi}_1(z)|/|\tilde{\theta}_3(z)|\sim(\psi_0)^{-1}|\mathrm{Im}(\Delta\chi^{(3)})|/|\mathrm{Re}(\chi^{(3)})|\sim5\times10^{-2}(\psi_0)^{-1}. \tag{33}$$

In actual experiments $\psi_0\geqslant10^{-2}$ (Ref. 8), from which we can assume that the quantities $\tilde{\phi}_1(z)$ and $\tilde{\theta}_3(z)$ are of comparable magnitude.

In the expression for the total NOR angle (19) we will retain the largest terms. Thus, the part that depends on the magnetic field is given by

$$\begin{aligned}\Delta\beta_{NL}(H_0)&=\beta_{NL}(H_0)-\beta_{NL}(0)\\&=KI_0z\{\mathrm{Im}(\Delta\chi^{(3)})\\&\quad\times[\mathrm{sinc}(\Delta kz)\sin[4\alpha(z)]-\sin(4\beta_0)]-H_0\\&\quad\times[\mathrm{Im}(3\Gamma_2-2\Gamma_3-\Gamma_1)-\mathrm{Im}(\Gamma_1+\Gamma_2+\Gamma_3)\\&\quad\times\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)]]+[-\mathrm{Re}(3\chi_1^{(3)}\\&\quad+2\chi_2^{(3)}+\chi_3^{(3)})+\mathrm{Re}(\Delta\chi^{(3)})\mathrm{sinc}(\Delta kz)\\&\quad\times\cos[4\alpha(z)]](2k_0)^{-1}\Delta k\\&\quad-\mathrm{Re}(\Delta\chi^{(3)})\psi_0\\&\quad\times[\mathrm{sinc}(\Delta kz)\cos[4\alpha(z)]-\cos(4\beta_0)]\}.\end{aligned} \tag{34}$$

As follows from (34), under conditions in which the linear Faraday effect is small (e.g., weak magnetic fields or thin samples) and the parameter $\Delta kz\ll1$, $\beta_{NL}(H_0)\sim H_0$. If $\Delta kz\sim1$, then the dependence of β_{NL} on the magnetic field, $\beta_{NL}(H_0)$, begins to oscillate, and for $\Delta kz\gg1$ it again becomes linear. In a crystal of $Cd_{0.75}Mn_{0.25}Te$ with thickness $z=0.1$ cm in the magnetic field range $0\leqslant H_0\leqslant6\cdot10^3$ G, the value of $\Delta kz=2\beta_L$ varies within the limits 0 to 0.16 (Refs. 1–3) and, consequently, the dependence of β_{NL} on H_0 should be quasilinear, which is found to be in agreement with the results of experiment.[1–3] This dependence is illustrated in Fig. 1, from which, in particular, it can be seen that its character is determined largely by the orientation of the azimuth of the polarization of the incident wave relative to the crystallographic axes.

5. CONCLUSION

The above treatment grounds the qualitative ideas about the peculiarities of the NOR effect in magnetic crystals set forth in the Introduction. As has been shown, it is not a trivial generalization of the natural Faraday effect to the nonlinear case, but is rather a fairly complicated and multifaceted phenomenon associated with various physical mechanisms of the polarization interaction of waves.

On the one hand, this circumstance substantially complicates the interpretation of the corresponding experimental data, which can occasionally lead to fundamentally erroneous conclusions. In particular, as has been shown, experimental measurement of the linear (or quasilinear)

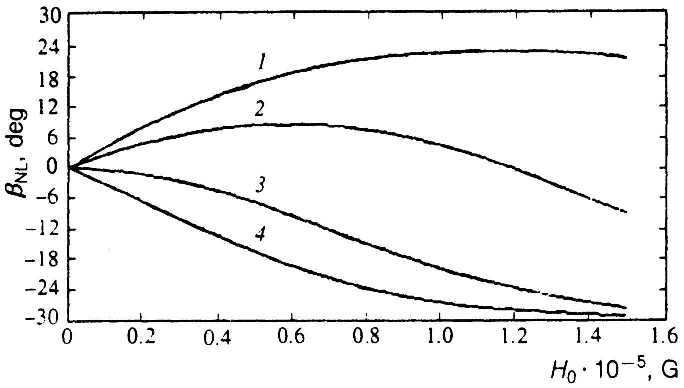

FIG. 1. Dependence of the nonlinear optical rotation angle β_{NL} on the external magnetic field H_0 for the following parameter values: wavelength $\lambda = 1.06$ μm, refractive index $n = 3.5$, intensity of the incident light $I_0 = 700$ MW/cm^2, $\mathrm{Im}(\Delta\chi^{(3)}) = 10^{-11}$ cgsOe units, $\mathrm{Re}(\Delta\chi^{(3)}) = 5 \cdot 10^{-11}$ cgsOe units, $\psi_0 = 10^{-1}$. Curves *1*, *2*, *3*, *4* correspond to the following values of β_0: 0, $\pi/16$, $\pi/8$, $\pi/4$.

dependence of the NOR angle on the magnitude of the magnetic field cannot serve as a basis for direct determination of the values of the nonlinear magnetooptic susceptibilities—rather a more detailed treatment is required. In this regard, we point to Refs. 1 and 2, in which the values of the nonlinear optical and magnetooptic susceptibilities were determined from independent measurements:

$$\mathrm{Re}(\chi^{(3)}) \approx 10^{-10} \text{ cgsOe units},$$

$$\mathrm{Im}(\Gamma) \approx 10^{-14} \text{ cgsOe units} \cdot \mathrm{G}^{-1}$$

and, consequently,

$$|\mathrm{Re}(\chi^{(3)})/\mathrm{Im}(\Gamma)|_{\mathrm{exp}} \approx 10^4 \text{ G}.$$

At the same time, according to (28) this ratio is

$$|\mathrm{Re}(\chi^{(3)})/\mathrm{Im}(\Gamma)|_{\mathrm{est}} = k_0/2V = 7 \times 10^8 \text{ G},$$

which exceeds by more than four orders of magnitude the value obtained from the experimental data. Such a discrepancy can be explained if we assume that in reality what was being investigated was the effect associated with the anisotropy of two-photon absorption or with rotation of the polarization ellipse or both of these phenomena together, since according to estimates (29) and (33), their combined contribution to NOR is also four orders of magnitude larger than that of the nonlinear Faraday effect. In particular, the authors of Refs. 1 and 2 determined from their measurements apparently not the susceptibility $\mathrm{Im}(\Gamma)$ but some other parameter, proportional to $\Delta\chi^{(3)}$, the anisotropic part of the cubic nonlinear-optical susceptibility.

On the other hand, the multifacetedness of the effect makes it attractive and promising from the point of view of experimental study of the subtle features of the polarizational interaction of light in magnetic crystals.

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