*The theory of second harmonic generation in media with natural and magnetic gyrotropy. Approach of the coupled waves

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Abstract

In this paper approaching slowly varying amplitudes the investigation of the problem of second harmonic generation in media with natural and magnetic gyrotropy is obtained.

Key words: nonlinear optics, second harmonic generation, natural optical activity, magnetic gyration.

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1. Introduction

A number of theoretical and experimental researches have shown the importance of gyrotropy influence on processes of coherent interaction of electromagnetic waves in nonlinear media [1-9]. The presence of natural and magnetic gyrotropy results in occurrence of new details of nonlinear interaction of electromagnetic waves. Simultaneously the influence of gyrotropy on conditions of phase matching and, as a consequence, on efficiency frequency transformations of radiation is most essential.

2. Nonlinear wave equations in electrodynamics of bi-gyrotropic media

Lets consider the problem of nonlinear interaction of optical fields in anisotropic media with natural optical activity and magnetic gyrotropy. At the solution of this problem we shall use methods developed in [6-12].

We shall proceed from the Maxwell equa-

tions of macroscopic electromagnetic field

$$\nabla \times \mathbf{E} = -\frac{1}{\mathbf{c}} \frac{\partial \mathbf{B}}{\partial t}, \tag{1}$$

$$\nabla \times \mathbf{H} = \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}$$
 (2)

and the phenomenological material equations

$$\mathbf{D} = (\varepsilon + i\mathbf{G} \times)\mathbf{E} + i\alpha\mathbf{H} + 4\pi\mathbf{P}^{nl}, \tag{3}$$

$$\mathbf{B} = (\mu + i\mathbf{O} \times)\mathbf{H} - i\widetilde{\alpha}\mathbf{E}. \tag{4}$$

Here $\widetilde{\alpha}$ means the tensor, transposed α , so $\widetilde{\alpha}_{ij} = \alpha_{ji}$; $\mathbf{G} \times$ and $\mathbf{Q} \times$ are the antisymmetric tensors corresponding to vector product:

$$(\mathbf{G}\times)_{ij}=-e_{ijk}G_k,$$

$$(\mathbf{Q}\times)_{ij} = -e_{ijk}Q_k$$

where e_{ijk} is absolutely antisymmetric unit tensor of third rank..

Excluding the equations (1) - (4) vectors **D**, **B** and **H** we shall obtain the equation for the strength of electrical field

$$\left\{ \nabla \times \left(\mu + i \mathbf{Q} \times \right)^{-1} \nabla \times + \left(\varepsilon + i \mathbf{G} \times -\alpha \widetilde{\alpha} \right) \frac{1}{c^2} \frac{\partial^2}{\partial t^2} + \frac{i}{c} \frac{\partial}{\partial t} \left(\gamma \nabla \right) \times \right\} \mathbf{E} = -\frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}^{nl}}{\partial t^2}. \tag{5}$$

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The equation for a magnetic induction can be similarly constructed:

$$\left\{ \nabla \times \left(\varepsilon - \alpha \widetilde{\alpha} + i \mathbf{G} \times + \frac{i}{c} \frac{\partial}{\partial t} (\gamma \nabla) \times \right)^{-1} \nabla \times + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right\} \mathbf{B} = \frac{4\pi}{c} \nabla \times \frac{\partial \mathbf{P}^{nl}}{\partial t}.$$
 (6)

In these equations the pseudo-tensor of natural gyrotropy γ is coupled with pseudo-tensor of magneto-electrical susceptibility of medium α by means of the following relation:

$$\gamma = \mu^{-1} \widetilde{\alpha} - \text{Tr} \left(\alpha \mu^{-1} \right) \cdot I , \qquad (7)$$

where I is the unit tensor of the second rank with components δ_{ij} , $\operatorname{Tr} \eta = \eta_{ii}$.

Using the monochromatic waves of nonlinear polarization with frequencies ω and 2ω as a source of an electromagnetic field, we shall assume that the medium is transparent on these frequencies. On this basis electrodynamics parameters of medium ε , μ , α , G and Q we shall consider as real parameters.

For the process of the second harmonic generation we assume

$$\mathbf{P}^{nl} = \frac{1}{2}\chi : \mathbf{EE}. \tag{8}$$

From the nonlinear wave equation (1) we shall obtain the system of two reduced equations

$$\left\{ \nabla \times \nabla \times -\frac{\omega^2}{c^2} \left(\varepsilon(\omega) - \alpha(\omega) \widetilde{\alpha}(\omega) \right) + i \frac{\omega^2}{c^2} \mathbf{G}(\omega) \times + \frac{\omega}{c} \left(\gamma(\omega) \nabla \right) \times \right\} \mathbf{E}^{(\omega)} = \frac{2\pi \omega^2}{c^2} \chi : \mathbf{E}^{(2\omega)} \mathbf{E}^{(\omega)*}, \tag{9}$$

$$\left\{ \nabla \times \nabla \times - \frac{(2\omega)^2}{c^2} \left(\varepsilon (2\omega) - \alpha (2\omega) \widetilde{\alpha} (2\omega) \right) + i \mathbf{G} (2\omega) \times + \frac{2\omega}{c} \left(\gamma (2\omega) \nabla \right) \times \right\} \mathbf{E}^{(2\omega)} = \frac{4\pi\omega^2}{c^2} \chi : \mathbf{E}^{(\omega)} \mathbf{E}^{(\omega)}. \tag{10}$$

These equations describe the interaction of two electromagnetic modes on frequencies ω and 2ω in a view of mutual energy exchange.

Natural and magnetic gyrotropy in anisotropic media can be observed in pure view in a direction of axes of higher symmetry as effect of circular birefringence. Considering this circumstance, we shall consider the process of nonlinear interaction of electromagnetic waves at their distribution along the third fold axis. The electromagnetic waves in crystal, polarized on a circle, are

$$\mathbf{E}_{\lambda}^{\omega} = E_{\lambda}^{\omega} \mathbf{e}_{\lambda} e^{\left[i\left(k_{\lambda}z - \omega t\right)\right]}, \tag{11}$$

$$\mathbf{E}_{-\lambda}^{2\omega} = E_{-\lambda}^{2\omega} \mathbf{e}_{-\lambda} e^{\left[i\left(K_{-\lambda}z - 2\omega t\right)\right]}.$$
 (12)

Let's us write the equation for the waves of nonlinear polarization, raised by electromagnetic waves

$$\mathbf{P}_{\lambda}^{\omega} = \frac{1}{2} \chi : \mathbf{E}_{\lambda}^{2\omega} \mathbf{E}_{\lambda}^{\omega*}, \qquad (13)$$

$$\mathbf{P}_{-\lambda}^{2\omega} = \frac{1}{2} \chi : \mathbf{E}_{\lambda}^{\omega} \mathbf{E}_{\lambda}^{\omega} \tag{14}$$

in the next form:

$$\mathbf{P}_{\lambda}^{\omega} = \frac{1}{2} \mathbf{e}_{\lambda} d_{\lambda}^* E_{-\lambda}^{2\omega} E_{\lambda}^{\omega*} e^{\left[i\left((K_{-\lambda} - k_{\lambda})z - 2\omega t\right)\right]}, (15)$$

$$\mathbf{P}_{-\lambda}^{2\omega} = \frac{1}{2} \mathbf{e}_{-\lambda} d_{\lambda} \left(E_{\lambda}^{\omega} \right)^{2} e^{\left[i \left(2k_{\lambda} z - 2\omega t \right) \right]}. \tag{16}$$

In these expressions E_{λ}^{ω} and $E_{-\lambda}^{2\omega}$ - are the amplitudes of two cooperating waves depending on coordinate z. Two units polarization vectors \mathbf{e}_{λ} and wave vectors

$$k_{\lambda} = \frac{\omega}{c} \left(\sqrt{\varepsilon_o(\omega) + \lambda G_z(\omega)} + \lambda \alpha_{11}(\omega) \right)$$
 (17)

$$K_{\lambda} = \frac{2\omega}{c} \left(\sqrt{\varepsilon_o(2\omega) + \lambda G_z(2\omega)} + \lambda \alpha_{11}(2\omega) \right)$$
 (18)

correspond to the right $(\lambda = +1)$ and left $(\lambda = -1)$ circular polarization of these waves; $d_{\lambda} = \mathbf{e}_{\lambda} \chi : \mathbf{e}_{\lambda} \mathbf{e}_{\lambda}$ is the imaginary coefficient of

nonlinear interaction. Substituting (11), (12), (15), (16) in (9), (10) and taking into account

(17), (18) we shall receive the following system of nonlinear coupled equations:

$$\left(\frac{d^2}{dz^2} + 2i\frac{\omega}{c}\sqrt{\varepsilon_o(\omega) + \lambda G_z(\omega)}\frac{d}{dz}\right)E_{\lambda}^{\omega} = -\frac{2\pi\omega^2 d_{\lambda}^*}{c^2}E_{-\lambda}^{2\omega}E_{\lambda}^{\omega*}e^{i\Delta k_{\lambda}z}$$
(19)

$$\left(\frac{d^{2}}{dz^{2}} + 4i\frac{\omega}{c}\sqrt{\varepsilon_{o}(2\omega) + \lambda G_{z}(2\omega)}\frac{d}{dz}\right)E_{-\lambda}^{2\omega} = -\frac{4\pi\omega^{2}d_{\lambda}}{c^{2}}\left(E_{\lambda}^{\omega}\right)^{2}e^{i\Delta k_{\lambda}z} \tag{20}$$

In the equations (19), (20) is introduced

$$\Delta k_{\lambda} = K_{-\lambda} - k_{\lambda}$$

for the value of phase mismatch at nonlinear interaction of the circular polarized waves. In approximation of amplitudes E_{λ}^{ω} and $E_{-\lambda}^{2\omega}$, slowly varied in space, we neglect in (19), (20) by second derivative on coordinate z:

$$\frac{d}{dz}E_{\lambda}^{\omega} = i\sigma_{\lambda}^{\omega}E_{-\lambda}^{2\omega}E_{\lambda}^{\omega*}e^{i(\Delta k_{\lambda} - \psi_{\lambda})z}, \quad (21)$$

$$\frac{d}{dz}E_{-\lambda}^{2\omega} = i\sigma_{-\lambda}^{2\omega} \left(E_{\lambda}^{\omega}\right)^{2} e^{-i\left(\Delta k_{\lambda} - \psi_{\lambda}\right)z} . \tag{22}$$

In these equations the following designations are used:

$$\psi_{\lambda} = \operatorname{Arg} d_{\lambda},$$

$$\sigma_{\lambda}^{\omega} = \frac{\pi \omega \left| d_{\lambda} \right|}{c_{\lambda} \left(\varepsilon_{\lambda} \right) + \lambda G_{\lambda}(\omega)}, \qquad (23)$$

$$\sigma_{-\lambda}^{2\omega} = \frac{\pi\omega \left| d_{\lambda} \right|}{c\sqrt{\varepsilon_{o}(2\omega) - \lambda G_{z}(2\omega)}}.$$
 (24)

From the equations (21), (22) the ratio of Manly – Rou can be obtained

$$\frac{d}{dz} \left(\sqrt{\varepsilon_o(\omega) + \lambda G_z(\omega)} \cdot E_{\lambda}^{\omega} E_{\lambda}^{\omega*} + \right) \\
+ \sqrt{\varepsilon_o(2\omega) - \lambda G_z(2\omega)} \cdot E_{-\lambda}^{2\omega} E_{-\lambda}^{2\omega*} = 0.$$
(25)

This relation means, that the sum of energy

flows

$$S_{\lambda} = \frac{c\sqrt{\varepsilon_o + \lambda G_z}}{8\pi} \cdot E_{\lambda} E_{\lambda}^*$$
 (26)

of cooperating waves on both frequencies remains constant on all thickness of gyrotropic sample.

Representing now the complex amplitudes of cooperating waves in the form

$$E_{\lambda}^{\omega} = A_{\lambda}^{\omega} e^{i\phi_{\lambda}}, \ E_{-\lambda}^{2\omega} = A_{-\lambda}^{2\omega} e^{i\Phi_{\lambda}}$$
 (27)

and separating (21), (22) real and imaginary parts, we come to the following system of the equations:

$$\frac{d}{dz}A_{\lambda}^{\omega} = -\sigma_{\lambda}^{\omega}A_{-\lambda}^{2\omega}A_{\lambda}^{\omega}\sin\theta_{\lambda}, \qquad (28)$$

$$\frac{d}{dz}A_{-\lambda}^{2\omega} = \sigma_{-\lambda}^{2\omega} \left(A_{\lambda}^{\omega}\right)^2 \sin\theta_{\lambda}, \quad (29)$$

$$\frac{d\theta_{\lambda}}{dz} = \Delta k_{\lambda} + \left(\sigma_{-\lambda}^{2\omega} \frac{\left(A_{\lambda}^{\omega}\right)^{2}}{A_{-\lambda}^{2\omega}} - 2\sigma_{\lambda}^{\omega} A_{-\lambda}^{2\omega}\right) \cos\theta_{\lambda}, (30)$$

where

$$\theta_{\lambda} = \Delta k_{\lambda} z + \Phi_{-\lambda} - 2\phi_{\lambda} - \psi_{\lambda}.$$

3. The solution of coupled equations

The solution of this system in the case of phase matching condition

$$\sqrt{\varepsilon_o(2\omega) - \lambda G_z(2\omega)} - \sqrt{\varepsilon_o(\omega) + \lambda G_z(\omega)} = (31)$$

$$= \lambda(\alpha_o(2\omega) + \alpha_o(\omega))$$

can be found similarly [11]. In linear approximation on parameters of gyrotropy α_o and G_z this solution looks like:

$$A_{\lambda}^{\omega} = A_{0} \operatorname{sch} \left(\frac{\pi \omega \left| d_{\lambda} \right| A_{0}}{c \sqrt{\varepsilon_{o}(\omega)}} \left(1 - \frac{\alpha_{o}(\omega) + \alpha_{o}(2\omega) + G_{z}(\omega)}{2\lambda \sqrt{\varepsilon_{o}(\omega)}} \right) (z + z_{0}) \right), \tag{32}$$

$$A_{-\lambda}^{2\omega} = A_0 \left(1 - \frac{\alpha_o(\omega) + \alpha_o(2\omega)}{2\lambda \sqrt{\varepsilon_o(\omega)}} \right) \text{th} \left(\frac{\pi\omega |d_{\lambda}| A_0}{c \sqrt{\varepsilon_o(\omega)}} \left(1 - \frac{\alpha_o(\omega) + \alpha_o(2\omega) + G_z(\omega)}{2\lambda \sqrt{\varepsilon_o(\omega)}} \right) (z + z_0) \right)$$
(33)

The constants of integration should be determined from boundary conditions. Taking into account that a field of the second harmonic on an input of nonlinear medium equal to zero, we finally find the strength vectors of an electrical field of the interacted waves by phase matching

$$\mathbf{E}_{\lambda}^{\omega} = \mathbf{e}_{\lambda} A_{0} \operatorname{sch} \left(\frac{\pi \omega |d_{\lambda}| A_{0}}{c \sqrt{\varepsilon_{o}(\omega)}} \left(1 - \frac{\alpha_{o}(\omega) + \alpha_{o}(2\omega) + G_{z}(\omega)}{2\lambda \sqrt{\varepsilon_{o}(\omega)}} \right) z \right) \exp[i(k_{\lambda}z - \omega t)], \tag{34}$$

$$\mathbf{E}_{-\lambda}^{2\omega} = i\mathbf{e}_{-\lambda} A_{0} \left(1 - \frac{\alpha_{o}(\omega) + \alpha_{o}(2\omega)}{2\lambda \sqrt{\varepsilon_{o}(\omega)}} \right) \times$$

$$\operatorname{th} \left(\frac{\pi\omega |d_{\lambda}| A_{0}}{c\sqrt{\varepsilon_{o}(\omega)}} \left(1 - \frac{\alpha_{o}(\omega) + \alpha_{o}(2\omega) + G_{z}(\omega)}{2\lambda \sqrt{\varepsilon_{o}(\omega)}} \right) z \right) \exp \left[i \left(K_{\lambda} z - 2\omega t + \psi_{\lambda} \right) \right].$$
(35)

The obtained equations generalize the decision of a task of coherent second harmonic generation in the anisotropic media in the presence of natural and magnetic gyrotropy.

The limiting value of wave amplitude of the second harmonic follows from (34), (35). This value

$$E_{-\lambda}^{2\omega}(z \to \infty) = \left(1 - \frac{\alpha_o(\omega) + \alpha_o(2\omega)}{2\lambda\sqrt{\varepsilon_o(\omega)}}\right) A_0 \qquad (36)$$

differs from radiation amplitude on the fundamental frequency on an input in a nonlinear crystal. This difference has value about dimensionless parameter natural gyrotropy in (36) and is caused by difference of factors (23), (24) in the equations (28), (29) for natural gyrotropic medium at phase matching.

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