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# Modeling of Nonlinear Optical Activity Characteristics of Layered-Periodic Crystal Structures

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## Abstract

Analytical and numerical modeling of tensor characteristics of nonlinear optical activity for layered-periodic crystal structures (superlattices) is carried out. Calculations are executed in frames of the long wavelength approximation for electromagnetic field at neglecting of the harmonics generation and effect of the probe wave on the effective medium optical characteristics. The relations determining nonlinear optical activity, electrogyration effective tensors for the structures from bianisotropic layers are derived. The detailed analysis is made for the superlattices formed by cubic crystals of GaAs-type. Domains of parameters values at which the effective nonlinear gyrotropy characteristics exceed ones for the monocrystal components of the superlattices are ascertained.

## 1. General Relations

Effects of parametric crystal optics of layered-periodic structures or superlattices (SL) are intensively investigated last time. Even in frames of the long wavelength approximation for electromagnetic field (the case of short-period SL) these structures can exhibit electrooptical, gyrotropic, magneto-optical properties which are distinguished from ones for the monocrystals forming SL [1-3]. The aim of the paper is investigation of nonlinear optical activity (NLOA) and electrogyration in short-period SL. For the description of electromagnetic properties of SL with account of NLOA one can use material equations [4]

$$D_q = [\varepsilon_{qj}]E_j + i[\alpha_{qj}]H_j, \quad B_q = \mu_{qj}H_j - i[\alpha_{jq}]E_j, \quad (1)$$

where  $i^2 = -1$ , and the quantities

$$[\varepsilon_{qj}] = \varepsilon_{qj} + \chi_{qjk}E_k^0 + \theta_{qjkl}E_k^0E_l^0, \quad [\alpha_{qj}] = \alpha_{qj} + \nu_{qjk}E_k^0 + \tau_{qjkl}E_k^0E_l^0 \quad (2)$$

are the tensors of dielectric permittivity and optical activity depending on the field of the monochromatic controlling wave  $E^0$ ,  $q, j, k, l = 1, 2, 3$ . In Eqs. (2) tensors  $\chi$  and  $\theta$  describing nonlinear dielectric properties are real and symmetric relatively to any permutation of the indexes that corresponds to non-absorbing media. The forms of characterizing NLOA real pseudotensors  $\nu$  and  $\tau$  are determined by the crystallographic symmetry of the medium [4]. We will assume satisfying Eqs. (1) and (2) for the monocrystal layers and for the short-period SL at substitution of the corresponding

material tensors for the effective tensors. According to Eqs. (1), components of magnetic permeability tensor  $\mu$  are supposed to be not depending on the controlling electric field  $\mathbf{E}^0$ .

The general forms of effective tensors  $\varepsilon^{(e)}$ ,  $\chi^{(e)}$ ,  $\theta^{(e)}$  and pseudotensor  $\alpha^{(e)}$  of linear optical activity were determined (see Refs. in [1,2]). Analogically to [4], let us consider NLOA in the given field approximation, at neglecting: 1) generation of harmonics in the effective medium, 2) effect of the probe wave with a low intensity on the material characteristics of SL. Moreover, the following assumptions will be used: 1) tensors  $\varepsilon$  (non-disturbed by field  $\mathbf{E}^0$ ) and  $\mu$  are diagonal, 2)  $\mu \gg \alpha\chi$ ,  $\tau \gg \alpha\theta$ , that takes place in a wide range of parameters values [5,6].

Then the methods described in [2,3] and Eqs. (1) and (2) lead to the expressions

$$\begin{aligned} (\mu_{33}\varepsilon_{33})^{-1}v_{33j}, \quad (\mu_{33}\varepsilon_{33}\varepsilon_{33}^0)^{-1}v_{333}, \quad \varepsilon_{33}^{-1}v_{3ij}, \quad (\varepsilon_{33}\varepsilon_{33}^0)^{-1}v_{3i3}, \quad \mu_{33}^{-1}v_{i3j}, \\ (\mu_{33}\varepsilon_{33}^0)^{-1}v_{i33}, \quad v_{ijn}, \quad (\varepsilon_{33}^0)^{-1}v_{ij3}, \end{aligned} \quad (3)$$

$$\begin{aligned} (\mu_{33}\varepsilon_{33})^{-1}\tau_{33ij}, \quad (\mu_{33}\varepsilon_{33}\varepsilon_{33}^0)^{-1}\tau_{333i}, \quad (\mu_{33}\varepsilon_{33}\varepsilon_{33}^0)^{-1}\tau_{33i3}, \quad (\mu_{33}\varepsilon_{33})^{-1}(\varepsilon_{33}^0)^{-2}\tau_{3333}, \quad \varepsilon_{33}^{-1}\tau_{3mij}, \\ (\varepsilon_{33}\varepsilon_{33}^0)^{-1}\tau_{3m3i}, \quad (\varepsilon_{33}\varepsilon_{33}^0)^{-1}\tau_{3mi3}, \quad \varepsilon_{33}^{-1}(\varepsilon_{33}^0)^{-2}\tau_{3m33}, \quad \mu_{33}^{-1}\tau_{m3ij}, \quad (\mu_{33}\varepsilon_{33}^0)^{-1}\tau_{m33i}, \\ (\mu_{33}\varepsilon_{33}^0)^{-1}\tau_{m3i3}, \quad \mu_{33}^{-1}(\varepsilon_{33}^0)^{-2}\tau_{m333}, \quad \tau_{ijmn}, \quad (\varepsilon_{33}^0)^{-1}\tau_{ij3m}, \quad (\varepsilon_{33}^0)^{-1}\tau_{ijm3}, \quad (\varepsilon_{33}^0)^{-2}\tau_{ij33}, \end{aligned} \quad (4)$$

where  $i, j, m, n=1, 2$ . Here  $\varepsilon$  and  $\varepsilon^0$  are the permittivity tensors corresponding to the frequencies of the probe and controlling electric fields. At averaging of the form

$$A^{(e)} = xA^{(1)} + (1-x)A^{(2)}, \quad (5)$$

where  $x=d^{(1)}/D$  is the relative thickness of the first layer in the SL period  $D$ , upper indexes  $e, 1, 2$  here and below denote the quantities characterizing the effective medium, the first and the second crystal layers in the SL period, expressions (3,4) determine all the components of effective tensors  $v^{(e)}$ ,  $\tau^{(e)}$  describing NLOA. Expressions (3,4) are also true in the case of the constant controlling electric field  $\mathbf{E}^0$ . Then tensors  $v^{(e)}$  and  $\tau^{(e)}$  describe linear and quadratic electrogyration [5] in the considered media.

## 2. The Structures from GaAs-Type Crystals

Let us determine the form of tensor  $\tau^{(e)}$  in the practically important case of SL formed by cubic crystals of class  $\bar{4}3m$  (GaAs-type). According to [7,8], these crystals are characterized by the following nonzero components of tensor  $\tau$  (the axes of the orthogonal coordinate system are parallel to the axes  $\bar{4}$ ):

$$\begin{aligned} \tau_{3311} = \tau_{1122} = \tau_{2233} = -\tau_{2211} = -\tau_{3322} = -\tau_{1133} = \lambda, \quad \tau_{2323} = \tau_{3131} = \tau_{1212} = -\tau_{3232} = -\tau_{1313} = -\tau_{2121} = \lambda', \\ \tau_{3223} = \tau_{1331} = \tau_{2112} = -\tau_{2332} = -\tau_{3113} = -\tau_{1221} = \lambda'', \end{aligned} \quad (6)$$

where  $\lambda$ ,  $\lambda'$ ,  $\lambda''$  are real independent parameters. From expressions (4)-(6) we have (axis  $Z$  is perpendicular to the boundaries of the layers)

$$\tau_{3311}^{(e)} = -\tau_{3322}^{(e)} = \frac{\langle \lambda/(\mu\varepsilon) \rangle}{\langle 1/\mu \rangle \langle 1/\varepsilon \rangle}, \quad \tau_{3131}^{(e)} = -\tau_{3232}^{(e)} = \frac{\langle \lambda'/(\varepsilon\varepsilon^0) \rangle}{\langle 1/\varepsilon \rangle \langle 1/\varepsilon^0 \rangle}, \quad \tau_{3223}^{(e)} = -\tau_{3113}^{(e)} = \frac{\langle \lambda''/(\varepsilon\varepsilon^0) \rangle}{\langle 1/\varepsilon \rangle \langle 1/\varepsilon^0 \rangle},$$

$$\tau_{1331}^{(e)} = -\tau_{2332}^{(e)} = \frac{\langle \lambda''/(\mu\varepsilon^0) \rangle}{\langle 1/\mu \rangle \langle 1/\varepsilon^0 \rangle}, \quad \tau_{1212}^{(e)} = -\tau_{2121}^{(e)} = \langle \lambda' \rangle, \quad \tau_{2112}^{(e)} = -\tau_{1221}^{(e)} = \langle \lambda'' \rangle, \quad (7)$$

$$\tau_{2323}^{(e)} = -\tau_{1313}^{(e)} = \frac{\langle \lambda'/(\mu\varepsilon^0) \rangle}{\langle 1/\mu \rangle \langle 1/\varepsilon^0 \rangle}, \quad \tau_{1122}^{(e)} = -\tau_{2211}^{(e)} = \langle \lambda \rangle, \quad \tau_{2233}^{(e)} = -\tau_{1133}^{(e)} = \frac{\langle \lambda/(\varepsilon^0)^2 \rangle}{\langle 1/\varepsilon^0 \rangle^2}.$$

In Eqs. (7) and below the angular brackets denote averaging according to Eq. (5). It is seen from Eqs. (6) and (7) that the transition from monocrystals to SL is accompanied by increasing threefold the number of independent components of tensor  $\tau$ . In this case the indexes of the nonzero components of  $\tau$  do not change. According to [7,8], the general form of this tensor in Eq. (7) has no analogues among monocrystal media. That determines the main characteristic properties of exhibition of NLOA and electrogyration in the considered SL and points to wide opportunities of creating new NLOA materials on the basis of SL.

Values of some components of  $\tau^{(e)}$  in Eqs. (7) can exceed the values of the analogical quantities of the monocrystals originating the SL. These components, besides ones which are equal to quantities  $\pm\langle\lambda\rangle$ ,  $\pm\langle\lambda'\rangle$ ,  $\pm\langle\lambda''\rangle$ , can be written in the form

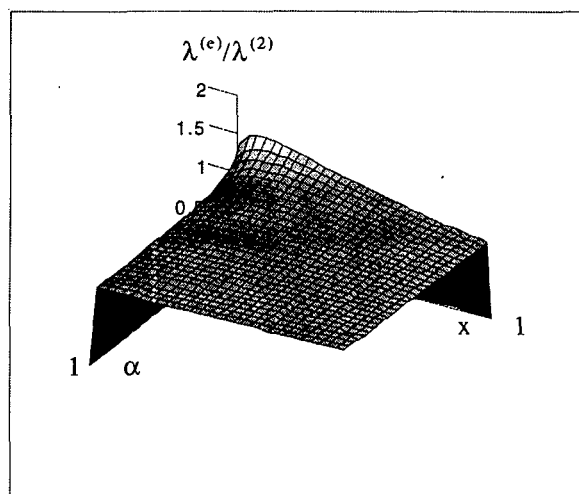
$$\lambda^{(e)} = \frac{\langle \lambda/(ab) \rangle}{\langle 1/a \rangle \langle 1/b \rangle}, \quad (8)$$

where  $\lambda$  and  $a, b$  are scalar parameters taking on values  $\lambda, \lambda', \lambda''$  and  $\mu, \varepsilon, \varepsilon^0$  correspondingly. Then a domain of the parameters values, satisfying the condition  $\lambda^{(e)} > \lambda^{(2)} > \lambda^{(1)}$ , is determined by the system of inequalities

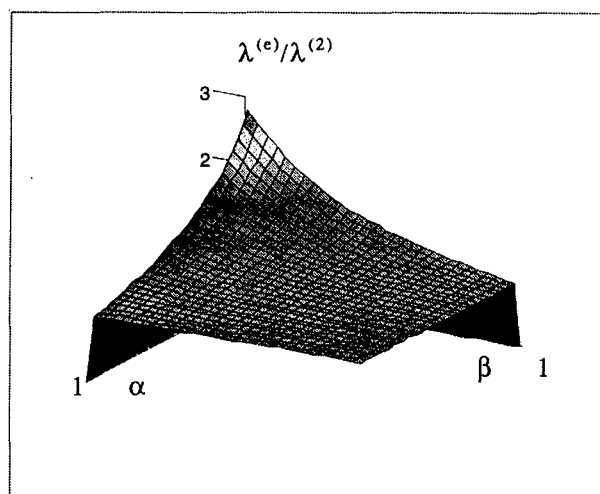
$$l < 1, \quad 0 < x < 1, \quad \alpha > 0, \quad \beta > 0, \quad (1 - \alpha)(1 - \beta)(1 - x) > 1 - l, \quad (9)$$

where  $l = \lambda^{(1)}/\lambda^{(2)}$ ,  $\alpha = a^{(1)}/a^{(2)}$ ,  $\beta = b^{(1)}/b^{(2)}$ .

According to system (9) an amplification of the considered induced gyrotropy properties at forming SL can take place at a small difference between the NLOA constants ( $l \approx 1$ ) and a strong difference between the dielectric ( $\varepsilon, \varepsilon^{(0)}$ ) and magnetic ( $\mu$ ) constants ( $\alpha \neq 1, \beta \neq 1$ ) of the SL components. In particular, at nonmagnetic layers satisfying the condition  $\lambda^{(e)} > \lambda^{(2)} > \lambda^{(1)}$  is not possible for all the components of tensor  $\tau^{(e)}$  in Eqs. (7), besides  $\tau_{2233}^{(e)} = -\tau_{1133}^{(e)}$ .



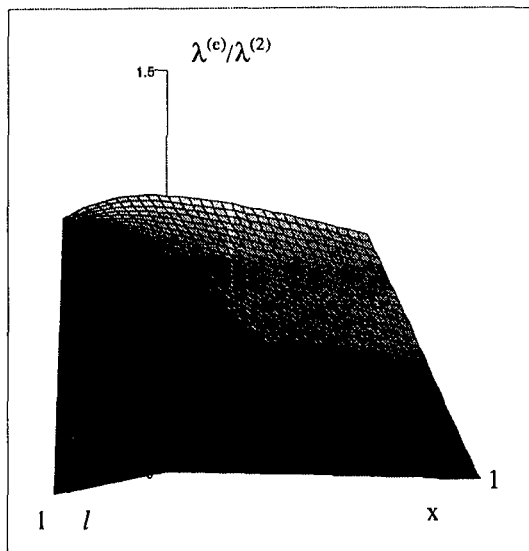
$\beta = 0.5, l = 0.99$   
Fig. 1



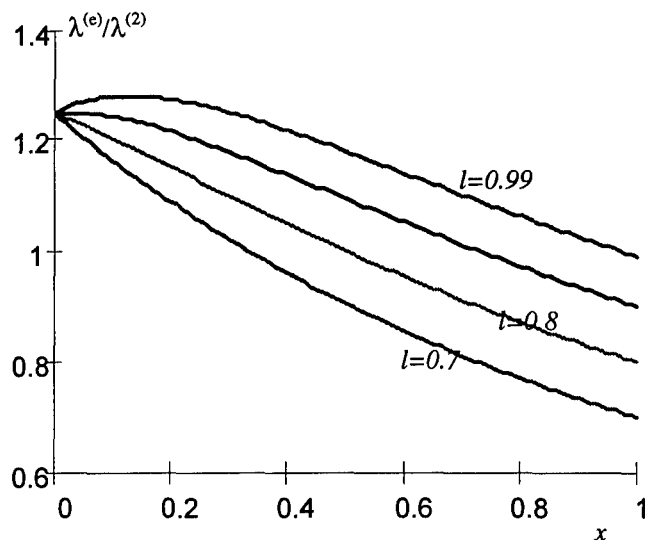
$x = 0.3, l = 0.99$   
Fig. 2

Relations (8,9) determine also the analogical domains of parameters values for the considered SL at quadratic electrooptical and induced magneto-optical effects [2,3].

Graphs calculated on Eqs. (8) and (9) illustrate possibilities of realization of the condition  $\lambda^{(e)} > \lambda^{(2)} > \lambda^{(1)}$  at various values of the parameters.



$\alpha = \beta = 0.5$   
Fig. 3



$\alpha = \beta = 0.5$   
Fig. 4

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