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Crystal optical properties of incommensurate phases in the plane-wave modulation region

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Abstract. Crystal optical properties of anisotropic optical materials of which the dielectric tensor is spatially modulated with a sinusoidal wave form are studied in the framework of the Jones calculus. Propagation of polarized light along the directions parallel to and far from the optical axes is considered. Polarization of the normal waves of the medium and the Jones matrix of a finite modulated crystal are derived, enabling us to ascertain the parameters of the apparent macroscopic optical activity. The developed model should describe the optical effects in a plane-wave region of incommensurate phases with the average inversion symmetry, occurring in the A_2BX_4 family crystals. The boundary conditions for the phase of the modulation wave, which play a key role in crystal optics of incommensurate phases, are discussed. The model predicts a relatively small optical activity in the birefringent crystal sections and negligible or zero effect in the optical axis directions. The conclusions agree well, at least, with the non-contradictory experimental results on optical rotatory power of the A_2BX_4 crystals. A comparison with the results derived earlier for the square modulation wave proves that the main conclusions of the model do not depend on the exact modulation shape.

1. Introduction

In spite of remarkable success in understanding the nature and properties of incommensurately modulated phases in dielectrics (Cummins 1990) there still remain experimental results that have not yet been satisfactorily explained. One of the most prominent examples is the existence of optical activity (OA) in incommensurate (IC) crystals of the A_2BX_4 family. The effect has been discovered by Uesu and Kobayashi (1985) with a complex experimental technique (the so-called HAUP, high-accuracy universal polarimeter—see Kobayashi *et al* 1986) applied to linearly birefringent crystal sections. In the last decade, an increasing number of experimental studies has been reported by different authors on this controversial problem (see, e.g. Dijkstra *et al* 1992b, Folcia *et al* 1993, Kobayashi *et al* 1993, 1994, Kushnir *et al* 1993, Meekes and Janner 1988, Ortega *et al* 1992, Saito *et al* 1990). From the very recent results we mention here those obtained by Ortega *et al* (1995), Simon *et al* (1996), Kremers and Meekes (1995, 1996) and Kremers *et al* (1996). In the first of the quoted works, an OA value which is almost outside the capacity of the experiment has been found in the case of Rb_2ZnCl_4, and the data of Kobayashi *et al* (1988), when reprocessed, have been shown to give nearly the same result. Kremers *et al* (1996) have reported a detailed study of $(N(CH_3)_4)_2ZnCl_4$, with a conclusion concerning the clearly zero gyration components g_{11}, g_{33} and g_{13} (see also Simon *et al* 1996) that contradicts the observations by Kobayashi *et al* (1993). At the same time, the measurements by Kremers and Meekes (1996) on the related compound

$(N(CH_3)_4)_2ZnCl_{2.8}Br_{1.2}$ have revealed that the OA can be affected by the IC modulation. Finally, a non-zero OA and optical indicatrix rotation have been observed by Kremers and Meekes (1995) in the IC $(N(CH_3)_4)_2CuCl_4$. Although differing in many important details, this agrees with the findings of Uesu and Kobayashi (1985) and Saito *et al* (1990).

Theoretical aspects of the problem attract much attention from researchers (Dijkstra *et al* 1992a, Etxebarria 1994, Kobayashi 1990, Kushnir and Vlokh 1993, Meekes and Janner 1988 etc). Indeed, the characteristic dimension relevant for the optical response increases in the modulated medium up to the spatial period of the IC superstructure, providing the possibility of a strong spatial dispersion that can give rise to an OA effect (Agranovich and Ginzburg 1979). However OA as a third-rank tensorial property should be macroscopically forbidden (see Dvorak *et al* 1983, Folcia *et al* 1993 and several references therein, Nye 1985) because of the inversion centre included in the point symmetry group of the 'average' IC structure (the superspace groups describing exhaustively the symmetry of the modulated crystals are centrosymmetric too).

The most consistent approach interpreting the observed OA seems to be that of a macroscopic electrodynamics, developed by Golovko and Levanyuk (1979), Meekes and Janner (1988) and Dijkstra *et al* (1992a). It proceeds from an order parameter dependent spatial inhomogeneity of the dielectric tensor in the IC phases. To simplify the analytical description, the crystal optical parameters of the IC materials have been mainly evaluated considering a square form for the modulation wave (Dijkstra *et al* 1992a, Kushnir 1996, Kushnir and Vlokh 1993). As a rough simulation of sinusoidal wave, Dijkstra (1991) and Kushnir and Vlokh (1993) analysed rather formally the properties of the medium whose dielectric function was modulated with a triangular shape. They assumed those properties to be independent of the exact form of the modulation wave. Rigorously speaking, all these results are valid directly for the multidomain low-temperature ordered phases in the A_2BX_4 group crystals, as well as the IC phases in the soliton regime of modulation. However, a question of principle is associated with the OA in the plane-wave (sinusoidal) regime occurring not far from the normal-to-IC phase transition temperature T_i, the more so because the work by Kobayashi (1990) points to an essential difference in the behaviour of the OA in the mentioned modulation regimes. As seen from the data of Saito *et al* (1990) for $(N(CH_3)_4)_2CuCl_4$ of which the ordered phase is centrosymmetric too, the OA near T_i can by no means be interpreted as a residual effect of the ordered phase. Unlike the model of 'discontinuously homogeneous' medium applicable for the domain-like region, the analysis of properties of sinusoidally modulated phases represents a cumbersome problem which does not have an exact analytical solution. The influence of the plane wave modulation on the polarization of electromagnetic waves has been dealt with in the study by Stasyuk and Shvaika (1991), but the interpretation of the optical effects has been omitted there.

It is to be noted that the experimental results on the optical rotatory power reported to date for the light propagation directions parallel to the optical axes in the IC crystals (Chern and Phillips 1972, Vlokh *et al* 1985, 1987, Kityk 1994) are much less known, although they seem to be worthwhile owing to the simple and reliable experiment technique used. Preliminary theoretical analysis of these results (Kushnir 1996, Kushnir and Vlokh 1993) has been made only with the assumption of a square modulation wave. It indicates that mechanisms of the OA manifested by the modulated materials in the absence of linear birefringence must be different from those in the case of birefringent crystal sections, a point that cannot be understood within the approach of Kobayashi (1990).

The purpose of this paper is a detailed theoretical discussion of crystal optical properties of incommensurately modulated materials in the plane-wave region, including the light propagation directions parallel to and far from the optical axes. Some preliminary results on this topic have been published elsewhere (Kushnir *et al* 1997). The main assumptions leading to a model developed in this work are explained in section 2. Section 3 is devoted to calculations of optical parameters of the modulated crystals. The results derived are interpreted in section 4. Finally, the conclusions are drawn in section 5.

2. Basic assumptions

Spatially averaged structure of the IC phase of the A_2BX_4 group crystals corresponds to a centrostymmetric point group *mmm* of a high-temperature parent phase. This implies that the dielectric tensor is real, symmetric and diagonal in the principal coordinate system, and the components $\varepsilon_{ii}^{(0)}$ have different values. However, taking into account spatial inhomogeneity of the IC phase, the material equation may be written generally as (see Agranovich and Ginzburg 1979, Meekes and Janner 1988)

$$D_i(\boldsymbol{q}) = \sum_{\boldsymbol{h}} (\varepsilon_{ij}^{(0)}(\boldsymbol{h}) + \mathrm{i} e_{ijk} g_{kl}(\boldsymbol{h}) q_l^{(u)}) E_j(\boldsymbol{q} - \boldsymbol{h}) \tag{1}$$

where $\varepsilon_{ij}^{(0)}$ is a symmetric part of the dielectric tensor, e_{ijk} the unit pseudotensor antisymmetric in all its indices, g_{kl} the gyration pseudotensor, $\boldsymbol{q}$ the wave vector of light, $\boldsymbol{q}^{(u)}$ the unit vector along $\boldsymbol{q}$, and $\boldsymbol{h}$ the vectors of the reciprocal lattice modified by the IC modulation (Meekes and Janner 1988). In other words, besides the homogeneous contributions $\varepsilon_{ij}^{(0)}(\mathbf{0})$ and $g_{kl}(\mathbf{0})$ and ($g_{kl}(\mathbf{0})$ is zero in our case) corresponding to the approximation of an average IC structure, one has to consider also that the components related to the modulation ($\boldsymbol{h} \neq \mathbf{0}$) can appear in the Fourier transform of the dielectric tensor ε_{ij}. In particular, the superspace symmetry of the IC phases does not forbid in general the existence of the off-diagonal components $\varepsilon_{ij}^{(0)}(\boldsymbol{h})$ leading to local monoclinic distortions of the structure, together with the local gyration components $g_{kl}(\boldsymbol{h})$. According to Agranovich and Ginzburg (1979), the contributions to the optical properties originating from structural inhomogeneities are proportional to the ratio of the inhomogeneity dimension and the wavelength λ of light. This is why it is sufficient to retain in (1) only the longest-wavelength vectors from the set of $\boldsymbol{h}$. We recall that the IC soft-mode wave vector $\boldsymbol{q}_{IC}$ may be represented in the form (see, e.g. Cummins 1990)

$$\boldsymbol{q}_{IC} = \gamma \boldsymbol{c}^* = \boldsymbol{q}_C + \boldsymbol{q}_I \tag{2}$$

where $\boldsymbol{q}_C = (r/s)\boldsymbol{c}^*$ is the lock-in commensurate modulation wave vector, $\boldsymbol{q}_I = (\delta(T)/s)\boldsymbol{c}^*$, $\boldsymbol{c}^*$ the reciprocal lattice vector of the parent phase along the modulation axis $\boldsymbol{c}$ (we adopt hereafter the crystallographic orientation of Meekes and Janner (1988)), r and s the integers that differ for different representatives of the A_2BX_4 group, and $\delta(T) \ll 1$ the irrational incomensurability parameter varying with temperature. As a result, the two independent periodicities are superposed in an incommensurately modulated crystal, their values being very close to each other. The small deviation of $\boldsymbol{q}_{IC}$ from $\boldsymbol{q}_C$ just results in a long-wave superlattice periodicity of which the period is an irrational fraction of the underlying lattice parameter. Of course, consideration of the long-wavelength components $\boldsymbol{q}_I$, instead of the $\boldsymbol{h}$ vectors, should mean a transition from a microscopic to a macroscopic description. Relevant discussion related to this point may be found in the work of Dijkstra *et al* (1992a). The modulation associated with the wave vector $\boldsymbol{q}_I$ is expected to contribute notably to the properties of the IC crystal. So, the electric polarization and the mechanical deformation in the IC phase are modulated with the wave vector $s\boldsymbol{q}_I$ (see Hamano *et al* 1980). Dvorak and Esayan (1982) and Esayan (1985) explained the striking effect of asymmetry in the characteristics of transverse ultrasonic waves observed in the IC Ba_2MnF_4 (Fritz 1975) and $RbH_3(SeO_4)_2$ (Esayan *et al* 1981) by the efficient coupling between the acoustic wave and the modulation wave related to $\boldsymbol{q}_I$. It should therefore be reasonable to put simply $\boldsymbol{h} = \boldsymbol{q}_I$ in our further calculations. Simple estimation gives $q_I \ll q_{IC}$ and $q/q_I \approx 10^{-1}$ or somewhat less, thus justifying the correctness of working in terms of macroscopic dielectric parameters. Notice that the procedure proposed by Meekes and Janner (1988) allows us to find shorter Fourier wave vectors $\boldsymbol{h}$. However their structural importance and, probably, the contributions to any physical properties of the crystal decrease progressively on increasing the corresponding indices (see also Etxebarria 1994).

Similarly to Dijkstra *et al* (1992a), we assume the dielectric properties of the IC crystal to be spatially modulated with a uniquely defined (and constant over the entire volume of a sample) period λ_I, being determined by $\lambda_I = 2\pi/q_I$, where $\boldsymbol{q}_I$ has a dominant role at the given temperature in the IC phase. This also means that the interaction of the modulated structure with defects and impurities is neglected completely. Below we shall not account for the modulation of diagonal components $\varepsilon_{ii}^{(0)}$ (see Golovko and Levanyuk 1979), for the corresponding modulated increments are much less than the average values $\varepsilon_{ii}^{(0)}(\mathbf{0})$. Let us restrict ourselves to considering only the local off-diagonal components $\varepsilon_{ij}^{(0)}(\boldsymbol{q}_I)$ and the gyration components $g_{kl}(\boldsymbol{q}_I)$ which are known (Dijkstra *et al* 1992a, Kushnir and Vlokh 1993, Stasyuk and Shvaika 1991) to affect most appreciably the polarization of electromagnetic waves travelling in the crystal.

The analysis of optical properties of the modulated medium is simplest for the light propagation direction coincident with the modulation axis $c = z$. Then the equiphase planes of the modulation wave are orthogonal to $\boldsymbol{q}$, and the properties of the medium remain constant along the transverse directions x and y but vary along the z axis. It is sufficient to account for only a spatial modulation of the dielectric parameters $\varepsilon_{12}^{(0)}(\boldsymbol{r})$ and $g_{33}(\boldsymbol{r})$. Below, we shall show that, under certain conditions, the obtained results may have a more general character, being applicable for the other propagation directions. With the symmetry conditions $\varepsilon_{ij}^{(0)}(-\boldsymbol{r}) = \varepsilon_{ij}^{(0)}(\boldsymbol{r})$ and $g_{kl}(-\boldsymbol{r}) = -g_{kl}(\boldsymbol{r})$ (Meekes and Janner 1988), we may write for the plane-wave region

$$\begin{aligned} \varepsilon_{12}^{(0)}(\boldsymbol{r}) &= \varepsilon_{a,12}^{(0)} \cos\varphi \\ g_{33}(\boldsymbol{r}) &= g_{a,33} \sin\varphi \end{aligned} \tag{3}$$

where $\varepsilon_{a,12}^{(0)}$ and $g_{a,33}$ are the amplitude factors and

$$\varphi = q_I z + \varphi_0 \tag{4}$$

the phase of the modulation.

It is difficult to deal with the optical phenomena in a crystal for which the parameters $\varepsilon_{12}^{(0)}$ and g_{33}, on one hand, and the difference $\varepsilon_{11}^{(0)} - \varepsilon_{22}^{(0)}$, on the other hand, have arbitrary relative values. A more convenient way is to concentrate separately on the two limiting cases for the optical anisotropy, namely when (i) the light propagation direction z is inclined appreciably compared to the optical axes ($\varepsilon_{12}^{(0)}, g_{33} \ll \varepsilon_{11}^{(0)} - \varepsilon_{22}^{(0)}$) and (ii) the given direction is parallel to the optical axis ($\varepsilon_{11}^{(0)} = \varepsilon_{22}^{(0)}$). The latter corresponds to a hypothetical degenerate case of a uniaxial crystal, while in reality the crystals of the A_2BX_4 group are optically biaxial, with the optical axes lying in the xz plane. The method therefore looks somewhat artificial but enables us to formulate distinctly the boundary conditions for the phase of the modulation (see sections 3 and 4).

3. Calculations of optical parameters of the modulated crystal

3.1. Light propagation directions far from the optical axes

To reveal the character of the light waves travelling in an optically anisotropic, inhomogeneous medium characterized by (3), it is convenient to employ the operating method of differential Jones matrices (Azzam and Bashara 1988, Jones 1948). In the framework of the approach, the spatial evolution of the complex amplitude vector $\boldsymbol{E}$ describing the transverse electric field component of electromagnetic wave (the so-called Jones vector) is determined by the relation (see Jones 1948)

$$\mathrm{i}\frac{\mathrm{d}\boldsymbol{E}}{\mathrm{d}z} = \mathbf{N}\boldsymbol{E} \tag{5}$$

where $\mathbf{N}$ is a differential propagation matrix related in a fundamental manner to the dielectric parameters of the medium and dependent on the propagation direction and, for inhomogeneous media, also on the longitudinal coordinate z. Note that, for transparent spatially homogeneous crystals, solving the equation (5) leads to a well known superposition principle in crystal optics (see Azzam and Bashara 1988, Nye 1985). This is why the equation (5) has the same wide limits of applicability as that principle. It must be stressed that the exact electromagnetic theory of light propagation in dielectrics (Fedorov 1976) is unnecessarily complicated. If the optical anisotropy in solid is weak (i.e. the difference between the refractive indices of the normal waves is much less than the mean refractive index, a condition that is fulfilled for the overwhelming majority of crystals), and we neglect the feeble effects of non-orthogonality of the normal waves in transparent crystals, the generality of the above approach is in fact the same as that of the exact electromagnetic theory (see, e.g. the data of a quantitative analysis by Evdishchenko *et al* (1991)).

Based on (3), one can arrive at the following propagation matrix $\mathbf{N}$ (see the appendix):

$$\mathbf{N} = \frac{1}{2}q_0 \begin{pmatrix} l & -\mathrm{i}c_{33}\sin\varphi - l_{12}\cos\varphi \\ \mathrm{i}c_{33}\sin\varphi - l_{12}\cos\varphi & -l \end{pmatrix} \tag{6}$$

with l and l_{12} the non-modulated and the modulated parts of the linear birefringence, respectively, c_{33} the modulated circular birefringence (or the OA) and q_0 the module of the wave vector of light in vacuum (see the appendix). We should emphasize that the relation (6) is correct unless the light wave normal becomes too close to the optical axes, i.e. just under the conditions of $l_{12},\ c_{33} \ll l$ obeyed in the experimental studies of OA performed with the HAUP technique.

Equation (5) with the $\mathbf{N}$ matrix (6) dependent on z can be solved approximately with a standard perturbation theory. In the coordinate presentation it becomes

$$\left[\mathrm{i}\delta_{ij}\frac{\mathrm{d}}{\mathrm{d}z} + \frac{1}{2}q_0(\mathrm{i}e_{ijk}c_{kk}\sin\varphi + l_{ij}\cos\varphi)\right]E_j = \pm\frac{1}{2}q_0 l E_i \tag{7}$$

where δ_{ij} denotes the Kronecker delta, $i = x,\ y$, $+$ refers to the component x and $-$ to y. According to the periodic character of the perturbation caused by the modulation of $\varepsilon_{12}^{(0)}$ and g_{33}, we suppose the solutions of (7) to take the form of Bloch-type waves (Golovko and Levanyuk 1979). Their Cartesian components are

$$E_i = u_i(\varphi)\exp(\mathrm{i}qz) \tag{8}$$

where the functions $u_i(\varphi)$ which have the period of the long-wave IC superstructure may be represented by

$$u_i(\varphi) = \sum_n U_i^{(n)}\exp(\mathrm{i}n\varphi). \tag{9}$$

Similarly, for the wave vector of light we write the expansion

$$q = q^{(0)} + q^{(1)} + \cdots. \tag{10}$$

In the unperturbed medium ($n = 0$) the two linearly polarized waves abbreviated hereafter as '1' and '2' are the solutions, with

$$\begin{aligned} &q_{1,2}^{(0)} = \mp q_0 l/2 \\ &U_{1x}^{(0)} = 1 \qquad U_{1y}^{(0)} = 0 \qquad U_{2x}^{(0)} = 0 \qquad U_{2y}^{(0)} = 1. \end{aligned} \tag{11}$$

In the first approximation ($n = \pm 1$)

$$\begin{aligned} &U_{1x}^{(\pm 1)} = 0 \qquad U_{1y}^{(\pm 1)} = q_0(\mp c_{33} + l_{12})/[4(q_1 - q_2 \pm q_I)] \\ &U_{2x}^{(\pm 1)} = q_0(\pm c_{33} + l_{12})/[4(q_2 - q_1 \pm q_I)] \qquad U_{2y}^{(\pm 1)} = 0 \end{aligned} \tag{12}$$

where $q_{1,2} = q_{1,2}^{(0)}$ are determined by (10) and (11). Since the next coefficients $U_i^{(\pm n)}$ are proportional to higher powers of the perturbation, we break the Fourier spectrum of the Bloch amplitude off, retaining the same accuracy as that of deriving the matrix (6).

As seen from (10)–(12), the wave vectors of the light waves propagating in the modulated medium, as well as the dispersion equation, are unaltered compared to a homogeneous (non-modulated) medium, making impossible the optical effects such as the existence of forbidden gaps for $\boldsymbol{q}$ (see Yariv and Yeh 1984). This is a consequence of the fact that, for the propagation directions under investigation, the main contribution to the normal wave refractive indices originates from a non-modulated fraction of the linear birefringence. Incidentally, according to Fousek (1991), the IC modulation influences the linear birefringence associated with the difference between diagonal components of the dielectric tensor, resulting in corrections proportional to the square of the modulated parameters. It should be stressed (see formula (11)) that $\boldsymbol{q}_{1,2}$ contain only their anisotropic parts. This corresponds to the fact that the basic propagation matrix (6) is normalized (Azzam and Bashara 1988), i.e. its isotropic part related to the mean refractive index is omitted as being unable to affect the polarization of light.

One can find from (8)–(12) the expressions for the electric fields $\boldsymbol{E}_1$ and $\boldsymbol{E}_2$ of electromagnetic waves characteristic of the modulated medium:

$$\begin{aligned} \boldsymbol{E}_1 &= \left\{ \boldsymbol{e}_x + (q_0/4)\left[\frac{(-c_{33} + l_{12})\exp(\mathrm{i}\varphi)}{q_1 - q_2 + q_I} + \frac{(c_{33} + l_{12})\exp(-\mathrm{i}\varphi)}{q_1 - q_2 - q_I}\right]\boldsymbol{e}_y \right\} \exp(\mathrm{i}q_1 z) \\ \boldsymbol{E}_2 &= \left\{ (q_0/4)\left[\frac{(c_{33} + l_{12})\exp(\mathrm{i}\varphi)}{q_2 - q_1 + q_I} + \frac{(-c_{33} + l_{12})\exp(-\mathrm{i}\varphi)}{q_2 - q_1 - q_I}\right]\boldsymbol{e}_x + \boldsymbol{e}_y \right\} \exp(\mathrm{i}q_2 z) \end{aligned} \tag{13}$$

where $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$ are the unit vectors along the x and y axes. Equations (13) prove that the structural modulation manifests itself mainly in the polarization state of these waves. It may be ascertained on the basis of the complex parameter $\kappa_{e1,2} = E_{1,2y}/E_{1,2x}$ (see Azzam and Bashara 1988). As with the lossless media, the waves appear to be orthogonal ($\kappa_{e1}\kappa_{e2}^* = -1$, * denoting a complex conjugation). Their polarization is in general ellipical and evolves with passing on through the medium, depending on the exact coordinate z. In other words, because of their spatial inhomogeneity, these waves cannot be regarded as normal waves in the usual sense (see Agranovich and Ginzburg 1979, Azzam and Bashara 1988).

It is well known that the character of the normal light waves is determined by the crystal optical effects manifested by the anisotropic medium. For an optically inhomogeneous medium, unambiguous solving of the inverse problem (identification of crystal optical effects on the basis of the known polarization of the waves) is difficult, particularly because of a spatial dependence of polarization state of the normal waves mentioned before (see also Kushnir 1996). In order to avoid ambiguity, one has to consider a crystal plate of a finite thickness (d). Our next step corresponds to the method proposed by Kushnir (1996). Let us calculate the integral Jones matrix M of the plate defined by the relation $\boldsymbol{E}_{out} = \mathsf{M}\boldsymbol{E}_{in}$ ($\boldsymbol{E}_{in}$ and $\boldsymbol{E}_{out}$ being respectively the Jones vectors of the light incident on ($z = 0$) and emergent ($z = d$) from the plate), using decomposition of $\boldsymbol{E}_{in}$ and $\boldsymbol{E}_{out}$ on the Jones vectors of the normal waves. Then it is possible to pass in our analysis from the spatially inhomogeneous normal waves to the effective normal waves of the entire crystal (the eigenvectors of the M matrix). The polarization state of the latter does not formally depend on coordinate but only the crystal thickness and the values of the phase of the modulation wave at the boundaries ($\varphi_0 = \varphi(0)$ and $\varphi_1 = \varphi(d)$). We shall just characterize the modulated crystal in terms of the effective parameters referred to its effective normal waves.

Following the procedure described above, we obtain

$$\mathsf{M} = \begin{pmatrix} \exp(-\mathrm{i}\,\Delta/2) & -2(k + \mathrm{i}\,\Delta\theta)\sin(\Delta/2) \\ 2(k - \mathrm{i}\,\Delta\theta)\sin(\Delta/2) & \exp(\mathrm{i}\Delta/2) \end{pmatrix} \tag{14}$$

where

$$\begin{aligned} k &= \alpha_+(\cos\varphi_1 - \cos\varphi_0)\cot(\Delta/2) + \alpha_-(\sin\varphi_1 + \sin\varphi_0) \\ \Delta\theta &= -\alpha_-(\sin\varphi_1 - \sin\varphi_0)\cot(\Delta/2) + \alpha_+(\cos\varphi_1 + \cos\varphi_0). \end{aligned} \tag{15}$$

The coefficients $\alpha_\pm$ are expressed via the sums of and the differences between the wave amplitudes $U_{2x}^{(+1)} \pm U_{2x}^{(-1)}$ (or $U_{1y}^{(+1)} \pm U_{1y}^{(-1)}$):

$$\alpha_+ = \frac{c_{33}(q_I/q_0) - l_{12}l}{2(l^2 - (q_I/q_0)^2)} \qquad \alpha_- = \frac{c_{33}l - l_{12}(q_I/q_0)}{2(l^2 - (q_I/q_0)^2)} \tag{16}$$

and $\Delta = (q_2 - q_1)d = q_0 l d$ represents the total phase retardation for the two normal waves attributed to the linear birefringence. The optical parameters k and $\Delta\theta$ in (14) and (15) determine respectively the ellipticity and the polarization azimuth of one of the (orthogonal) effective normal waves (see Kushnir and Vlokh 1993). Note that the relations (15) cannot be applied when Δ approaches zero, owing to the assumptions made above. The analysis shows (see also the next subsection) that the terms proportional to $\cot(\Delta/2)$, and responsible for the behaviour of k and $\Delta\theta$ when the wave normal becomes close to the optical axis, should remain finite in reality ($k \to \pm 1$ or 0, and $\Delta\theta \to \pm\pi/4$ or 0).

3.2. Optical axis directions

In the case of light propagation directions parallel to the optical axes ($l = 0$) we cannot apply directly the perturbation theory when solving the equation (5) with the propagation matrix (6). An elegant alternative way suggested by Azzam and Bashara (1972) consists in employing the equation for the evolution of light polarization with the distance z:

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}\kappa(z) = -N_{12}\kappa^2(z) + (N_{22} - N_{11})\kappa(z) + N_{21} \tag{17}$$

where

$$\kappa(z) = E_y(z)/E_x(z) \tag{18}$$

is the polarization parameter (see above) written in terms of Cartesian components of the Jones vector of light. Equation (17) for inhomogeneous media ($N_{ij} = N_{ij}(z)$) represents the general Ricatti equation which does not have analytical solution for arbitrary dependence $N_{ij}(z)$. One of the exceptions described by Azzam and Bashara (1972) refers to the problem of propagation of light along the helical axis in a cholesteric liquid crystal.

Equation (17) may be solved by separation of variables for the fractions $\mathbf{N}_\varepsilon$ and $\mathbf{N}_g$ of the $\mathbf{N}$ matrix (see the appendix) which describe media with modulated local indicatrix rotation and the gyration, respectively. For example, substituting the elements $N_{g,ij}$ in (17) yields

$$\kappa(z) = \tan(C - \gamma_g \cos\varphi) \tag{19}$$

with

$$\gamma_g = (c_{33} q_0)/(2q_I) \tag{20}$$

and C a constant of integration. It may be found from the initial condition $\kappa(0) = \kappa_0$, where κ_0 refers to the polarization state of the light incident at the crystal:

$$\tan C = \frac{\kappa_0 + \tan(\gamma_g \cos\varphi_0)}{1 - \kappa_0 \tan(\gamma_g \cos\varphi_0)}. \tag{21}$$

From (19) and (21) we have

$$\kappa(d) = \frac{\tan[\gamma_g(\cos\varphi_0 - \cos\varphi_1)] + \kappa_0}{1 - \kappa_0 \tan[\gamma_g(\cos\varphi_0 - \cos\varphi_1)]}. \tag{22}$$

When comparing (22) with the well known relation (Azzam and Bashara 1988)

$$\kappa(d) = \frac{M_{21} + M_{22}\kappa_0}{M_{11} + M_{12}\kappa_0} \tag{23}$$

one can see that the coefficients of a bilinear transformation $\kappa(d, \kappa_0)$ given by (22) determine, up to a complex factor, the integral Jones matrix $\mathbf{M}$ of the modulated crystal. This factor may be found from the condition of unitarity of $\mathbf{M}$, since the latter matrix describes a lossless medium. We obtain the normalized Jones matrix that specifies completely the influence of the crystal on the polarization of light:

$$\mathbf{M}_g = \mathbf{R}[-\gamma_g(\cos\varphi_1 - \cos\varphi_0)]. \tag{24}$$

The matrix of the crystal with the modulated component $\varepsilon_{12}^{(0)}$ may be derived in a similar manner:

$$\mathbf{M}_\varepsilon = \begin{pmatrix} \cos[\gamma_\varepsilon(\sin\varphi_1 - \sin\varphi_0)] & \mathrm{i}\sin[\gamma_\varepsilon(\sin\varphi_1 - \sin\varphi_0)] \\ \mathrm{i}\sin[\gamma_\varepsilon(\sin\varphi_1 - \sin\varphi_0)] & \cos[\gamma_\varepsilon(\sin\varphi_1 - \sin\varphi_0)] \end{pmatrix} \tag{25}$$

where

$$\gamma_\varepsilon = (l_{12} q_0)/(2q_I). \tag{26}$$

From (24) and (25) we are now able to reveal the character of optical phenomena taking place in the modulated crystal. Following Kushnir and Vlokh (1993), we shall consider separately the influence on these phenomena of the modulations of $\varepsilon_{12}^{(0)}$ and g_{33}.

4. Discussion of the model

We begin from discussing the optical properties of the IC crystals for propagation directions other than the optical axes. As in the studies by Kushnir (1996) and Kushnir and Vlokh (1993), it should be natural to refer to the nonzero parameters k and $\Delta\theta$ as the OA and the optical indicatrix rotation which appear owing to the modulation, irrespective of the different origin of those effects compared to spatially homogeneous crystals. One can see from (15) that the OA is still present in the modulated crystal, despite the fact that the symmetry group of the 'average structure' must include an inversion centre. We note that little attention has been paid to the indicatrix rotation effect, although its order of magnitude is expected to be the same as that of the OA (see formula (15)), and the effect has been detected in experimental studies (see Kushnir *et al* 1993, Ortega *et al* 1992, and references therein).

To evaluate the size of the optical effects, it is necessary to account for the smallness of the ratio $q_0/q_I = \lambda_I/\lambda_0$ characterizing the inhomogeneity of the modulated medium, and the parameter l ($l \cong 10^4$–10^{-2}) in (16)†. Then the expressions for the amplitude coefficient may be written as

$$\alpha_+ \approx -k_0(l\lambda_I/\lambda_0) + \Delta\theta_0(l\lambda_I/\lambda_0)^2, \alpha_- \approx -k_0(l\lambda_I/\lambda_0)^2 + \Delta\theta_0(l\lambda_I/\lambda_0) \tag{27}$$

where we put $k_0 = c_{33}/(2l)$ and $\Delta\theta_0 = l_{12}/(2l)$ to be equal to the ellipticity of the normal wave (i.e. the OA) and the indicatrix rotation occurring in a corresponding acentric, low-symmetry non-modulated crystal. With the results for the square-wave modulation (Kushnir and Vlokh 1993) one can derive

$$k/k_0 \cong \Delta_{HP} = \pi(l\lambda_I/\lambda_0) \tag{28}$$

where Δ_{HP} is the phase retardation per half-period of the modulation. Notice that less significant terms of order $(l\lambda_I/\lambda_0)^2$ are dropped from (28). A comparison of formulae (15) and (27) with (28) substantiates that the results obtained for the two modulation regimes agree almost quantitatively. Further inspection shows that the conclusion remains valid also if the model is complicated by analysing local distortions of the modulation wave owing to interactions between the IC structure and defects, as well as the unipolarity effect (see Kushnir and Vlokh 1993).

It is interesting to compare the results of the present study with that of Stasyuk and Shvaika (1991). Proceeding from their relations for the normal light waves, it is easy to derive formulae similar to (15) in which, however, the coefficients α_+ and α_- turn out to be proportional to $(\lambda_I/\lambda_0)^2$ and $(\lambda_I/\lambda_0)^3$, contrary to (27). This should in fact imply the absence of any observable OA. Moreover, the relative values of the contributions to α_+ and α_- originating from k_0 and $\Delta\theta_0$ are different from those obtained by Kushnir and Vlokh (1993) and in the present work. The reasons for such a discrepancy are still not quite clear.

Let us examine more closely the case of light propagation along the optical axes. Rigorously speaking, the Jones matrices (24) and (25) describe, respectively, an optical rotator (purely optically active crystal) and a linear phase retardation plate (linearly birefringent crystal) with the fast axis tilted by the angle $\pm\pi/4$ with respect to x and y axes. However both the optical rotation

$$\phi = -\gamma_g(\cos\varphi_1 - \cos\varphi_0) \tag{29}$$

and the phase retardation Δ_ε attributed to the birefringence l_{12},

$$\Delta_\varepsilon = 2\gamma_\varepsilon(\sin\varphi_1 - \sin\varphi_0) \tag{30}$$

appear to be feeble effects. Indeed, apart from a small ratio q_0/q_I in (20) and (26) (see above), the modulation amplitude $g_{a,33}$ cannot exceed the typical values for the gyration tensors in acentric non-modulated crystals (10^{-6}–10^{-4}—see e.g., Nye 1985). The parameter $\varepsilon^{(0)}_{a,12}$ has to be of the same order of magnitude. This is why the matrices (24) and (25) only slightly differ from a unit matrix which describes an optically isotropic medium.

† Under the matching condition for the linearly polarized normal waves of the unperturbed system ($\boldsymbol{q}_2 - \boldsymbol{q}_1 = \boldsymbol{q}_I$ or $\lambda_I/\lambda_0 = 1/l$), equation (16) displays a possibility for resonance increase of k and $\Delta\theta$. This should mean that the linear birefringence concerned with the difference between the diagonal components $\varepsilon^{(0)}_{ii}$ is overwhelmed, and the crystal becomes either optically active only ($k \to \pm 1$) or linearly birefringent only, but with the corresponding fast axis inclined by $\Delta\theta \to \pi/4$ to the principal axes (cf the conclusions drawn by Zapasskiy and Kozlov (1995)). In incommensurately modulated crystals the phase matching condition may be surely regarded as impossible.

Furthermore, the optical rotation effect does not accumulate with increasing thickness of sample, unlike the situation in homogeneous crystals. Finally, as seen from (29) and (30), ϕ and Δ_ε are rigorously zero whenever the phase of the modulation remains the same at both sample surfaces ($\varphi_1 = \varphi_0$).

To compare the results for the optical axis directions with those of the study by Kushnir and Vlokh (1993), we consider now the most general case of $\varphi_1 \neq \varphi_0$. It may conventionally be termed as a local boundary unipolarity† of the modulated structure, using the analogy with the domain-like layered structures considered in the mentioned work. It must be emphasized that the model of 'extended' (or 'regular') unipolarity in which each neighbouring half-period of the modulation has different lengths (see Kushnir and Vlokh 1993) looks more realistic but cumbersome to describe analytically in the plane-wave region. Here we do not display the relevant results, which lead to conclusions qualitatively the same as those presented below. One can expect the unipolarity $\Delta\varphi = \varphi_1 - \varphi_0$ to be small. Expressing $\Delta\varphi$ in terms of the modulation wavelength,

$$\Delta\varphi = 2\pi(\Delta\lambda_I/\lambda_I) \tag{31}$$

and introducing the unipolarity parameter

$$\eta = \Delta\lambda_I/d \tag{32}$$

one derives from (20), (29), (31) and (32) that the optical rotation may be written as

$$\phi = \phi_0\eta \text{ or } \phi = \phi_0\eta^2\pi(d/\lambda_I) \tag{33}$$

for the structures with the initial phase values $\varphi_0 = \pi/2$ and $\varphi_0 = 0$, respectively. In (33), ϕ_0 has the meaning of the optical rotation in a homogeneous crystal sample of the same thickness d:

$$\phi_0 = \pi c_{33} d/\lambda_0. \tag{34}$$

Equations (33) agree with the corresponding relations obtained for the soliton region (Kushnir and Vlokh 1993), thus proving that the main conclusions of the model based on treating the IC phase as a periodic spatial distribution of the dielectric tensor do not depend on the exact shape of the modulation wave.

Of particular interest is a comparison of the results presented for the alternative light wave normal directions, at least for the reason of verifying the self-consistency of the developed model. A correspondence between these results is concerned with the polarization states of the normal waves. It is provided by the first terms on the right-hand side of (15) which should result in optical behaviours of rotator or a linear phase retardation plate types when $l \to 0$, in agreement with (24) and (25). In contrast, the values of the retardation parameters ϕ and Δ_ε predicted by (29) and (30) cannot be deduced from (14), since the contributions to the total phase retardation Δ arising from the modulation have been neglected (see subsection 3.1). On the whole, the optical parameters determined by (15), (29) and (30) may be divided into the amplitude and phase factors dependent, respectively, on the modulation amplitude and the phase of the modulation wave. As to the amplitude factor, it is proportional to $(q_2 - q_1)/q_I$ in both cases. This term is much smaller for the optical axis directions, being referred exclusively to a modulated fraction of the total birefringence ($q_2 - q_1 = q_0 l_{12}$ or $q_0 c_{33}$). Another drastic difference between the propagation directions parallel to and far from the optical axes lies in the phase factors. In the former case the optical anisotropy arises only when a periodicity condition $\varphi_1 = \varphi_0$ is broken, i.e. in a unipolar modulated structure (see the above discussion).

† The term appears because the neighbouring half-periods of the modulation, having opposite signs of the modulated parameters, may also include spontaneous polarizations (or strains) of the opposite signs.

It is well known that the IC phase can acquire a small unipolarity, due to the interactions with structural defects. However the effect is possible perhaps not far from the IC-to-lock-in phase transition temperature but not in the plane-wave region. The model hence predicts a zero or negligible OA along the optical axes in sinusoidally modulated IC phase. All of the relevant experimental studies performed on the A_2BX_4 family representatives and the $NaNO_2$ crystals (Chern and Phillips 1972, Vlokh *et al* 1985, 1987, Kityk 1994) have revealed an insignificant optical rotatory power observed only in the vicinity of the low-temperature lock-in phase. Furthermore, a correlation of the effect size and the unipolarity of samples has been demonstrated. This confirms the validity of the presented model. At the same time, in linearly birefringent crystal sections the phase factor may, in principle, be of the order of unity (see equations (15)), and the magnitude of the OA requires a further examination.

A crucial problem is represented by the boundary conditions for the phase of the modulation wave, which seems to be worth considering in more detail. In the early study by Golovko and Levanyuk (1979) is was suggested that the crystal boundaries could occupy arbitrary positions, irrespective of the IC superstructure period, and these positions could change not only by an integer number of spatial periods of the modulation. This has to lead to a random distribution of φ_0 and φ_1 values over the crystal surfaces. When accounting for the 'averaging' processes for the measured optical parameters owing to extreme smallness of λ_I, a finite cross-section of the probing light beam and inevitable non-parallelity of the sample surfaces, one can conclude within the framework of the model (see formulae (15)) that the OA observed in any practical experiment should be cancelled out to zero (see also Etxebarria 1994). To explain the OA measured in the HAUP experiments, Dijkstra *et al* (1992a) supposed that, in accordance with some energy considerations and periodicity requirements, the modulation phase was to be the same at both surfaces of crystal plate. This implies that integer number m of the modulation wavelengths must fit into the crystal dimension along the modulation axis, i.e. the condition $m\lambda_I = d$ is fulfilled similarly to some extent to the situation with the normal vibration modes of a finite optical string.

Comparing to the natural relation $pc = d$ (with c the cell parameter in the modulation direction and p an integer), the assumption $\varphi_1 = \varphi_0$ (equivalent to $m\lambda_I = d$) does not look evident, the more so because the properties of crystals in the vicinity of their surfaces are less investigated so far, and they can differ from those of the crystal bulk (Chen *et al* 1981). It should be reasonable, although somewhat oversimplified, to suppose that a pinning of the modulation wave, due to the requirement of energy stability of the modulated system, takes place at the surface which represents in fact a defect-like formation (see e.g. Hamano *et al* 1980, Janovec 1983). For the soliton modulation regime, the condition $m\lambda_I = d$ correlates with the conclusion that the phase solitons nucleate and annihilate in pairs (Nattermann 1986). Specific mechanisms ensuring a constancy of the modulation phase over the surfaces under changing sample thickness may be variations of the number m and (or) local variations of the IC modulation wavelengths λ_I occurring in the surface layer (Chen *et al* 1981). In all other respects, the presented model should not be critical to the local changes in λ_I, as expected from the analysis by Kushnir and Vlokh (1993) for the square modulation wave.

One can see that the exact equality $m\lambda_I = d$ contradicts an incommensurate character of the modulation, since the fraction λ_I/c cannot be irrational with m and p simultaneously integer. Besides the mentioned local λ_I variations, a way out can be offered by the hypothesis of a 'devil's staircase' behaviour of the modulation wave vector (Bak and Bachm 1980). With $\boldsymbol{q}_{IC} = (r'/s')\boldsymbol{c}^*$ (r' and s' being integers), it becomes clear that the condition

$m\lambda_I = d$ reduces to a general requirement that the spatial period of (any) superstructure must fit into the corresponding crystal dimension. If it is really so, then even the approximate equality $\boldsymbol{q}_{IC} \approx (r'/s')\boldsymbol{c}^*$, taking place for each temperature inside the incommensurately modulated phase, should provide an approximate fulfilment of the condition $\varphi_1 = \varphi_0$. As a result, the boundary conditions for the phase of the IC modulation should affect different physical properties of the crystal. An appropriate example is the studies of influence of sample shape on the low-frequency dielectric parameters of $(N(CH_3)_4)_2ZnCl_4$ compound (Sveleba *et al* 1995), irrespective of the exact interpretation given by the authors.

Under the condition $\varphi_1 = \varphi_0$ the optical parameters of the modulated crystal have to be described by the second terms on the right-hand side of (15), and the OA is mainly associated with the modulation of the $\varepsilon_{12}^{(0)}$ component (see equation (27)). Furthermore, the OA in the optical axis directions will be zero, in accordance with the above discussion. It is still not clear enough which of the values φ_0 ($=\varphi_1$) are preferable and realized in practice. A sequence of complete modulation cells (the half-periods of a square modulation wave) considered by Dijkstra (1991) and Dijkstra *et al* (1992a) corresponds to a zero value of the modulated parameter at the crystal boundaries and should mean that $\varphi_0 = 0$ (or π) in the case of modulation of the gyration tensor, as seen from (3) and (4). Besides such a 'perfect' (in terms of Kushnir and Vlokh 1993) structure, the modulated structure with $\varphi_0 = \pi/2$ (or $-\pi/2$) for which the g_{33} module has a maximum value at the boundaries is also physically distinguishable and may be suspected to be stable. We conclude from (15) that only the values $\varphi_0 = \pm\pi/2$ provide a situation observed in most of the HAUP experiments (the OA $k \neq 0$ and the indicatrix rotation $\Delta\theta = 0$). Then the 'node' of the $\varepsilon_{12}^{(0)}$ modulation wave and the 'hump' of the g_{33} wave will be at the crystal boundaries. In the framework of the present approach, it is hardly believable that there exist some grounds for one of the enantiomorphous structures, characterized by $\varphi_0 = 0$ or π (and $\pi/2$ or $-\pi/2$), to be privileged. If the selection happens due to accidental effects such as the interactions with defects, then the OA observed on different samples may be of the opposite signs (see also discussion by Dijkstra (1991), Dijkstra *et al* (1992a) and Kushnir and Vlokh (1993)).

Since a macroscopic gyration tensor component g_{13} has been measured in most of the HAUP experiments and the actual optical axis directions lie in the xz plane, we now turn to the problem of how to generalize our model for light wave normals different from the z axis. The problem is not trivial, for we cannot simply use the model of a 'transversely homogeneous' optical medium as above. The most important fact is that the dielectric properties of the medium should vary from one point of the surface to another, and it becomes impossible to formulate the boundary conditions for the phase of modulation in the case of a plane electromagnetic wave travelling along a direction not parallel to the modulation axis (see Stasyuk and Shvaika 1991). The assumption of Golovko and Levanyuk (1979) that, for the propagation directions perpendicular to z axis, the electric field of the light wave can be averaged over z, and structural modulation should not manifest itself in the optical properties, seems reasonable. For the 'tilted' directions, particularly in the xz plane, we obviously have a case intermediate between that discussed throughout the paper and that just mentioned. The main conclusions of the present work are then expected to be valid. Only when assuming that the dielectric properties do not vary evenly along the crystal boundaries (i.e. the surfaces have a tendency to keep definite values of the phase of modulation, being 'quasi-equiphase' surfaces) can we avoid cancellation of the measured macroscopic OA. Insignificant extra peculiarities of the model will be longer modulation period ($\lambda_I^{eff} = \lambda_I/\cos\alpha$, with α the angle between the light wave normal and the modulation axis), together with the fact that the modulation of the other $\varepsilon_{ij}^{(0)}$ and g_{kl} components must be additionally accounted for.

5. Conclusions

In this work a phenomenological model is developed for interpreting the crystal optical properties of the IC phases characterized by a spatially average inversion symmetry. It should be valid for description of the OA of the IC crystals in a plane-wave region and in the case of light propagating along the modulation axis, although the methods of generalizing the model to arbitrary propagation directions are outlined. Unlike the study by Kushnir and Vlokh (1993), we worked mainly in terms of an ideal modulated structure which was not unipolar and distorted by interactions with defects. It is revealed that incommensurability of the modulation, leading to appearance of long-wave Fourier components of the dielectric tensor, acts notably on the normal wave polarization and can cause an apparent OA effect. The model is not in general tied to a certain IC crystal, but involves the dielectric tensor allowed by the superspace symmetry, which depend on the value and the temperature behaviour of the modulation wave vector (Meekes and Janner 1988). Because of the insignificant role of external defects in the properties of the IC phases in the plane-wave region, consideration of the related effects was rather schematic. It is evident that defects and structural unipolarity can also affect the optical parameters. However the latter contributions are sample dependent and cannot result in reproducible OA.

Taking into account the difficulties in analysing the most general light wave normal direction, we considered the two limiting cases of practical importance, the directions parallel to and far from the optical axes. Essentially different conclusions are drawn in these cases. For the optical axis directions, the ideal incommensurately modulated structure shows no OA. The effect can be imposed by the unipolarity and defect influences only, in excellent agreement with all of the experimental data reported on the A_2BX_4 family compounds. In our belief, comprehending the mentioned difference must be a necessary feature of the true theory that explains the OA observed in the IC crystals (see Kushnir 1996).

As seen from a comparison with the results presented by Dijkstra (1991), Dijkstra *et al* (1992a) and Kushnir and Vlokh (1993), the crystal optical properties of the IC phases are not sensitive to the exact modulation shape (cf Kobayashi 1990). However, specific physical mechanisms for the modulation of local dielectric parameters in the domain-like and the plane-wave modulation regimes are different. They are concerned respectively with a spontaneous electrogyration effect occurring due to the spontaneous electric polarization included in the nearly commensurate domains, and a direct coupling of dielectric susceptibility with the order parameter of the IC phase transition.

On the whole, the presented model predicts a small value of the macroscopic OA in the modulated crystals, when compared with acentric macroscopically homogeneous crystals. Furthermore, the model is very critically dependent on the boundary conditions for the phase of the modulation wave, and, in order to avoid cancelling out the observed OA, one has to assume this phase to be the same at both crystal surfaces (see also Dijkstra 1991). This means in general that the spatial period of any (one-dimensional) superlattice in a solid should fit into the length of the crystal along the modulation direction.

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Appendix

The $\mathbf{N}$ matrix of the modulated crystal may be derived from the integral Jones matrix $\mathbf{M}(z, \Delta z)$ of a thin slice of the medium of thickness Δz located at the coordinate z. Let the propagation direction z be parallel to the optical axis. Consider first the modulation of $\varepsilon_{12}^{(0)}$ parameter only. It is easy to show that the real symmetric part of the dielectric tensor with the components $\varepsilon_{11}^{(0)} = \varepsilon_{22}^{(0)}$, $\varepsilon_{33}^{(0)}$ and $\varepsilon_{12}^{(0)}$ can be reduced to diagonal form in the coordinate system $x'y'z'$ with the axes x' and y' rotated by $\pm 45^\circ$ compared to the principal axes x and y. Then using a standard Jones matrix of a linear phase retardation plate with the rotated principal axes (Azzam and Bashara 1988) and the relation (3) yields

$$\mathbf{M}_\varepsilon(z, \Delta z) = \begin{pmatrix} \cos[(l_{12}q_0\,\Delta z/2)\cos\varphi] & \mathrm{i}\sin[(l_{12}q_0\,\Delta z/2)\cos\varphi] \\ \mathrm{i}\sin[l_{12}q_0\,\Delta z/2)\cos\varphi] & \cos[(l_{12}q_0\,\Delta z/2)\cos\varphi] \end{pmatrix} \tag{A1}$$

where $l_{12} = \varepsilon_{a,12}^{(0)}/\bar{n}$ is the amplitude of the modulated contribution to the linear birefringence associated with $\varepsilon_{12}^{(0)}$, $\bar{n}$ the mean refractive index, $q_0 = 2\pi/\lambda_0$ and λ_0 the wavelength of light in vacuum. Taking into account the modulation of g_{33} only, we have

$$\mathbf{M}_g(z, \Delta z) = \mathbf{R}[(c_{33}q_0\,\Delta z/2)\sin\varphi] \tag{A2}$$

with

$$\mathbf{R}(\beta) = \begin{pmatrix} \cos\beta & -\sin\beta \\ \sin\beta & \cos\beta \end{pmatrix} \tag{A3}$$

the matrix of rotation by the angle β, and $c_{33} = g_{a,33}/\bar{n}$ the amplitude of the modulated circular birefringence (see Nye 1985). From the relation between $\mathbf{N}$ and $\mathbf{M}(z, \Delta z)$ matrices (Jones 1948)

$$\mathbf{N} = \lim_{\Delta z \to 0} \frac{\mathbf{M}(z, \Delta z) - \mathbf{I}}{\Delta z} \tag{A4}$$

where $\mathbf{I}$ is the identity matrix, and (A1) and (A2) we obtain

$$\mathbf{N}_\varepsilon = -\frac{1}{2} q_0 l_{12} \cos\varphi \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \tag{A5}$$

$$\mathbf{N}_g = \frac{\mathrm{i}}{2} q_0 c_{33} \sin\varphi \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}. \tag{A6}$$

Since the propagation matrices characterizing different elementary types of optical anisotropy may be put together algebraically (Jones 1948), the overall matrix of the medium with the modulated parameters $\varepsilon_{12}^{(0)}$ and g_{33} takes the form

$$\mathbf{N} = \mathbf{N}_\varepsilon + \mathbf{N}_g. \tag{A7}$$

Finally, the $\mathbf{N}$ matrix for the propagation directions inclined appreciably to the optical axes may be derived in a quite similar manner. The only difference is that we have to proceed from a more general integral Jones matrix (Azzam and Bashara 1988) describing a crystal possessing a large non-modulated linear birefringence $l = (\varepsilon_{11}^{(0)} - \varepsilon_{22}^{(0)})/(2\bar{n})$ referred to the diagonal components of the dielectric tensor. Taking into account the relative smallness of the modulation amplitudes $\varepsilon_{a,12}^{(0)}$ and $g_{a,33}$ and performing the calculations give equation (6) of section 2 which generalizes the $\mathbf{N}$ matrix defined by (A5)–(A7).

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