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1996 J. Phys.: Condens. Matter 8 3921

(http://iopscience.iop.org/0953-8984/8/21/017)

J. Phys.: Condens. Matter **8** (1996) 3921–3932. Printed in the UK

Character of normal light waves in absorbing crystals with modulated dielectric parameters

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Received 2 January 1996

Abstract. On the basis of the superposition principle for the optical anisotropy parameters and the Jones calculus methods a phenomenological model is developed to interpret the optical properties of absorbing, spatially modulated dielectric materials. The character of the normal electromagnetic waves propagated in a uniaxial, weakly dichroic crystal for which the complex gyration tensor is modulated with the simple square wave form is ascertained. The structure with a perfect modulation wave and the unipolar structure are considered and should describe the multidomain ferroelectric and incommensurately modulated phases. Specific crystal optical effects are found which have no analogues in uniform materials. Proceeding from the normal wave polarization the modulated crystal is shown to be equivalent to a low-symmetry biaxial crystal. The origin of a non-zero eigenwave ellipticity is discussed in connection with the problem of optical activity in macroscopically centrosymmetric incommensurate crystals of A_2BX_4 group.

1. Introduction

In recent years the optical properties of multilayer solid state structures and superlattices of different physical natures have attracted the permanent interest of researchers. When compared with uniform objects, those structures often possess a wider set of elementary excitations. Crystalline media characterized by a spatially modulated dielectric function, i.e. layered dielectric crystals, incommensurately modulated and ordered multidomain ferroelectric phases, may be an important example (see, e.g., Dijkstra *et al* 1992a, Sorge and Hempel 1986, Vlokh *et al* 1992 and Yariv and Yeh 1984). It is known that the optical anisotropy of the systems mentioned has some specific features. So, the modulation of dielectric parameters notably affects the optical birefringence of crystals (Dijkstra 1991, Fousek 1991). Despite the average macroscopic inversion symmetry of the A_2BX_4 group incommensurate crystals, the existence in these materials of the controversial phenomenon of optical activity has been reported by different workers (Dijkstra *et al* 1992b, Kobayashi *et al* 1994, Kushnir *et al* 1993, Ortega *et al* 1992). The latter phenomenon described in terms of a second-rank axial material tensor is forbidden by the point symmetry group associated with the superspace group of the incommensurate phase and must therefore be related to inhomogeneity, owing to the incommensurate modulation of the dielectric medium on a semimacroscopic scale (Dijkstra *et al* 1992a, Kobayashi 1990, Kushnir and Vlokh 1993). Even the simple model developed by Dijkstra (1991) points to an essential difference in the crystal optics of homogeneous and spatially modulated crystals. Indeed, the system of purely birefringent optical platelets which simulates the modulation, with the square wave form, of the off-diagonal components of the real symmetric dielectric tensor

0953-8984/96/213921+12$19.50

turns out to be optically active, for its normal light waves are the elliptically polarized states.

It should be stressed that all the studies referred to above dealt with transparent crystals. At the same time, the presence of anisotropic absorption (dichroism) is known to influence markedly the process of transformation of polarized light by an optical medium and can result in the appearance of some new effects impossible in transparent crystals (see e.g. Pancharatnam 1957). From this point of view it should be interesting to test the crystal optical properties of the modulated dichroic materials, at least in the most simple and practical case of weak absorption.

In this paper a phenomenological approach to light propagation in a medium with a periodic space-dependent dielectric tensor (Kushnir and Vlokh 1993) is applied to absorbing crystals, to reveal the character of normal light waves in the latter. The preliminary results have been published elsewhere (Vlokh and Kushnir 1995). In section 2 the typical patterns for normal wave polarization in dielectric crystals are discussed, and the working methods and approximations are described. Sections 3 and 4 deal with the normal electromagnetic modes in the uniform and the modulated weakly dichroic uniaxial crystals. Conclusions are drawn in section 5.

2. Normal electromagnetic waves in crystals

The effect of the crystalline medium on the plane monochromatic electromagnetic wave can be understood in such a manner that the incident wave is resolved into two normal modes (eigenmodes) characteristic for a given type of optical anisotropy in crystal. These modes, which alone can propagate through the medium without alteration of their polarization and with definite phase velocity and decay, specify well the electromagnetic properties of that medium. The modes are defined completely by the polarization state, together with the refractive indices n and the absorption coefficients κ. If the polarized light passes through isotropic material, its polarization remains unchanged, i.e. each wave in such a material can be treated as normal. In anisotropic materials, several typical patterns for eigenwave polarization have to be distinguished, depending on the crystal optical effects available and the light propagation direction.

In uniaxial non-absorbing crystals the character of the eigenmode polarization is the simplest. When the ordinary (linear) birefringence is present, the eigenmodes are represented by the orthogonal states linearly polarized along the principal axes of the optical indicatrix. In optically active (circularly birefringent) material the modes become circularly polarized with the opposite signs of rotation. That is why, according to the usual approximation, the waves propagating in the elliptically birefringent crystal with coexisting birefringence and optical activity can be regarded as orthogonal elliptical states, their numerical ellipticities being equal and the signs opposite (figure 1(a)):

$$\tan(2\varepsilon_{e1,2}) = \pm\frac{2k}{1-k^2} = \pm\frac{\Delta n_c}{\Delta n_l} \tag{1}$$

where ε_{ei} denotes the ellipticity angle for the eigenwave (the ratio of minor to major semiaxes of the polarization ellipsis), and Δn_c and Δn_l are the circular and the linear birefringences, respectively. Except for very close to the optical axis direction, the eigenwave ellipticity is small because the optical activity is overwhelmed by a dominating birefringence effect:

$$\varepsilon_{e1,2} = \pm k = \pm\frac{\Delta n_c}{2\Delta n_l}. \tag{2}$$

Qualitatively the same pattern occurs in a crystal where pure absorption anisotropy (the elliptical dichroism) is present (Azzam and Bashara 1988, Vlokh and Kushnir 1996). The only peculiarity is that the normal waves suffer different absorptions but have the same velocities. In particular, we have linear or circular polarization of the waves in linearly or circularly dichroic crystals.

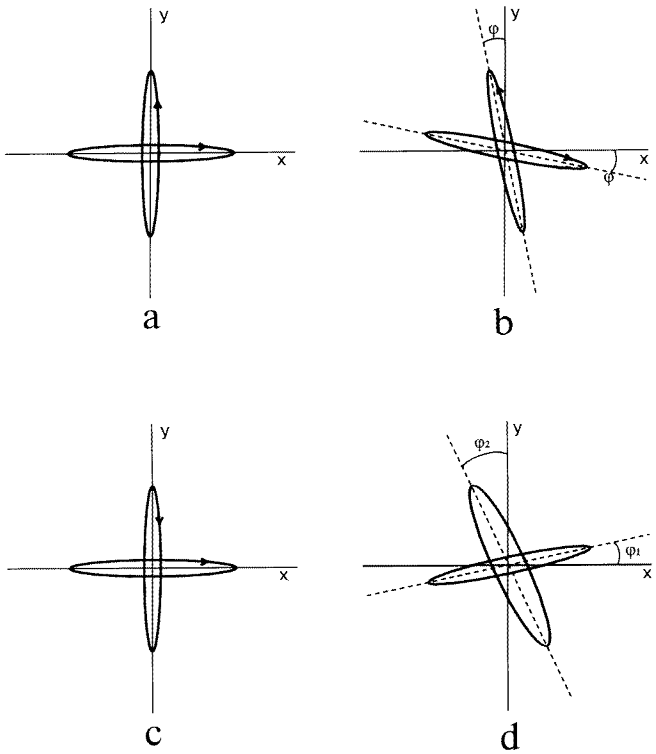

Figure 1. Typical patterns for the normal wave polarization in (a) transparent, (b) dichroic uniaxial, (c) birefringent linearly dichroic biaxial and (d) dichroic biaxial crystals. x, y are the principal axes of the optical indicatrix. Details are discussed in the text.

The situation becomes more complicated if the crystal possesses anisotropy of both the refractive and the absorption properties. In the most general case of an elliptically birefringent, elliptically dichroic biaxial crystal in which the principal directions of birefringence and dichroism do not coincide, the geometrical forms of the eigenwave polarization ellipses do not have a simple relation to one another as shown by Pancharatnam (1957). Namely, their major axes are not crossed, being inclined at different angles to the principal axes of refraction; the ellipticity moduli are not equal whereas their handedness can be either the same or the opposite (figure 1(d)). In some approximations, results have been obtained for a number of particular simpler cases (see, e.g., Konstantinova *et al* 1983, Pancharatnam 1955, 1957). For example, the normal waves in an absorbing uniaxial crystal have the same numerical ellipticities described by opposite signs (Pancharatnam 1957). The (usually weak) effect of dichroism is known to manifest itself in the non-orthogonality of the waves as displayed in figure 1(b).

A prominent effect can exist in biaxial crystals with the superposed birefringence and linear dichroism (Pancharatnam 1955). The eigenmodes turn out to be similarly rotating elliptical vibrations with equal ellipticities which may be written as (Okorochkov *et al* 1984, Pancharatnam 1957)

$$\tan(2\varepsilon_{e1,2}) = \frac{2p}{1-p^2} = \frac{\Delta\kappa_l}{\Delta n_l}\sin(2\varphi) \tag{3}$$

where $\Delta\kappa_l$ is the linear dichroism and φ the acute angle between the principal planes of birefringence and dichroism. Under the condition of a relatively small amount of dichroism, e.g. in the directions not too near the optical axes, the major axes of the ellipses are almost parallel to the principal planes of optical indicatrix (figure 1(c)) and

$$\varepsilon_{e1,2} = p = \frac{\Delta\kappa_l}{2\Delta n_l}\sin(2\varphi). \tag{4}$$

If, additionally, optical activity is present, it also contributes to the normal wave ellipticities, modifying them to

$$\varepsilon_{e1,2} = \pm k + p. \tag{5}$$

Now we shall concentrate on clarifying the main features of the normal waves imposed by a combined effect of absorption anisotropy and a periodic variation in the dielectric parameters in the crystal. It should be difficult to solve the problem of light propagation in a medium characterized by several coexisting crystal optical effects from the standpoint of exact electromagnetic theory. In view of the complexity of the subject, one has to make some reasonable simplifying assumptions. First, we shall restrict ourselves to considering the uniaxial crystals. Second, in the most practical cases the optical anisotropy is small, i.e. mathematically speaking the differences between the refractive and absorption coefficients are much less than the mean values of the latter. Then we can neglect the mutual influence of the elementary optical effects (birefringence, optical activity and dichroism) and use a superposition principle (Nye 1985). Note that the majority of the results discussed above have been derived within the framework of such an approach. We adopt the principle in its most general formulation as postulating that the anisotropy components for linearly and circularly polarized light waves are composed together according to a vectorial law (see also Jones 1948):

$$\Delta_0 = \sqrt{(\Delta_l + i\delta_l)^2 + (\Delta_c + i\delta_c)^2} \tag{6}$$

where Δ_0 is the (complex) amplitude-and-phase difference for the normal waves, Δ_l, Δ_c, δ_l and δ_c are the partial contributions to Δ_0 of the birefringence, optical activity, linear and circular dichroism, respectively:

$$\Delta_l = \gamma\Delta n_l \qquad \Delta_c = \gamma\Delta n_c \qquad \delta_l = \gamma\Delta\kappa_l \qquad \delta_c = \gamma\Delta\kappa_c \qquad \gamma = 2\pi d/\lambda \tag{7}$$

with d the thickness of crystal and λ the wavelength in vacuum.

From the two convenient calculation methods based on employing the superposition principle, the Poincaré sphere and the Jones calculus (Azzam and Bashara 1988), we choose hereafter the algebraic matrix method as more powerful when applied to inhomogeneous media. With this approach the characteristics of the normal waves can be easily determined from the eigenvectors and eigenvalues of the appropriate Jones matrices.

3. Homogeneous, weakly dichroic crystal

As a first step, let us consider the optical properties of a homogeneous (non-modulated) dichroic crystal. In the coordinate system associated with the principal axes, the general normalized Jones matrix of a uniaxial absorbing crystal may be written as follows (Jones 1948):

$$\mathbf{T} = \begin{bmatrix} \cos\left(\frac{\Delta_0}{2}\right) - \mathrm{i}\cos(2\beta)\sin\left(\frac{\Delta_0}{2}\right) & -\sin(2\beta)\sin\left(\frac{\Delta_0}{2}\right) \\ \sin(2\beta)\sin\left(\frac{\Delta_0}{2}\right) & \cos\left(\frac{\Delta_0}{2}\right) + \mathrm{i}\cos(2\beta)\sin\left(\frac{\Delta_0}{2}\right) \end{bmatrix} \tag{8}$$

where

$$\sin(2\beta) = \frac{\Delta_c + \mathrm{i}\delta_c}{\Delta_0} \qquad \cos(2\beta) = \frac{\Delta_l + \mathrm{i}\delta_l}{\Delta_0}. \tag{9}$$

A common situation is that the anisotropy for the linearly polarized waves exceeds appreciably that for the circularly polarized waves. This is valid unless the wave normal is very close to the optical axis direction, in which case a special analysis should be necessary. Otherwise we may disregard the contributions of Δ_c and δ_c to Δ_0. Using further the condition of weakness of the dichroism fulfilled for a large number of materials, we have from (9)

$$\sin(2\beta) \approx \frac{\Delta_c + \mathrm{i}\delta_c}{\Delta_l} \qquad \cos(2\beta) \approx 1. \tag{10}$$

Following Moxon and Renshaw (1990) it is convenient now to introduce a new set of anisotropy parameters Δ, E, k and k' describing the elementary optical effects (see also equation (2)):

$$\Delta = \gamma\Delta n_l \qquad E = \gamma\Delta\kappa_l \qquad k' = \frac{\Delta\kappa_c}{2\Delta n_l}. \tag{11}$$

Of these four parameters, E, k and k' are small with respect to unity, and we shall omit the terms of order E^2, k^2 and k'^2 in all further calculations. Inserting (11) in (8) gives the resulting Jones matrix of the homogeneous, weakly dichroic uniaxial crystal:

$$\mathbf{T}_H = \begin{bmatrix} \left(1 + \frac{E}{2}\right)\exp\left(-\mathrm{i}\frac{\Delta}{2}\right) & -2(k + \mathrm{i}k')\left[\sin\left(\frac{\Delta}{2}\right) + \mathrm{i}\frac{E}{2}\cos\left(\frac{\Delta}{2}\right)\right] \\ 2(k + \mathrm{i}k')\left[\sin\left(\frac{\Delta}{2}\right) + \mathrm{i}\frac{E}{2}\cos\left(\frac{\Delta}{2}\right)\right] & \left(1 - \frac{E}{2}\right)\exp\left(\mathrm{i}\frac{\Delta}{2}\right) \end{bmatrix}. \tag{12}$$

Note that (12) may be derived from the matrix of a transparent crystal (Vlokh *et al* 1990) with the formal replacement $\Delta \to \Delta + \mathrm{i}E$ and $k \to k + \mathrm{i}k'$.

Complex characteristic values of Jones matrix (12) testify that the crystal is to be classified as a mixed amplitude-and-phase retardation plate, for which the phase difference Δ is defined by the birefringence alone and the amplitude difference E by the linear dichroism alone. The eigenvectors of (12) specifying the normal wave polarization are characterized by the polarization parameters (Azzam and Bashara 1988) given by

$$\xi^H_{e1} = \mathrm{i}(k + \mathrm{i}k') \qquad \xi^H_{e2} = [\mathrm{i}(k + \mathrm{i}k')]^{-1}. \tag{13}$$

This proves that the normal waves are not orthogonal:

$$\xi^H_{e1}\left(\xi^H_{e2}\right)^* = -1 - \frac{2\mathrm{i}k'}{k}. \tag{14}$$

The effect is defined by a small ratio $\boldsymbol{k}/\boldsymbol{k}'$.

A noteworthy fact is the absence of any contribution from E to the polarization state of the eigenwaves, contrary to the views of Moxon and Renshaw (1990). If, in particular, the crystal manifests only linear dichroism the eigenwaves are orthogonal when the approximation linear in $\Delta_c\delta_l \ll 1$ is adopted (Kushnir and Vlokh 1995).

Checking the eigenvectors of (12) yields

$$\chi^H_{e1} = -k' \qquad \chi^H_{e2} = \tfrac{1}{2}\pi + k' \qquad \varepsilon^H_{e1} = k \qquad \varepsilon^H_{e2} = -k \tag{15}$$

for the azimuths (χ_e) and the ellipticities (ε_e) of the normal waves. The major axes of the polarization ellipses appear to be separated by the angles $\frac{1}{2}\pi + 2|k'|$ or $\frac{1}{2}\pi - 2|k'|$, depending on the sign of k'; each of these axes is rotated by $\mp k'$ compared with those of the transparent crystal, while the ellipticities are equal but opposite in sign. This fits completely into the pattern displayed in figure 1(b), in accordance with the general results obtained by Forsterling (quoted by Szivessy 1928) from the exact electromagnetic theory of light propagation in the uniaxial crystals. Note that, when circular dichroism is absent, the azimuths χ_{ei} define the angular positions of the principal axes of optical indicatrix.

Finally, we examine the normal wave polarization in a weakly anisotropic direction of optical axis. Then symmetry demands that both the birefringence and the linear dichorism disappear, and equation (8) transforms to

$$\mathbf{T}_H = \begin{bmatrix} \cos\left(\frac{\Delta_c}{2}\right) - \mathrm{i}\frac{\delta_c}{2}\sin\left(\frac{\Delta_c}{2}\right) & -\sin\left(\frac{\Delta_c}{2}\right) - \mathrm{i}\frac{\delta_c}{2}\cos\left(\frac{\Delta_c}{2}\right) \\ \sin\left(\frac{\Delta_c}{2}\right) + \mathrm{i}\frac{\delta_c}{2}\cos\left(\frac{\Delta_c}{2}\right) & \cos\left(\frac{\Delta_c}{2}\right) - \mathrm{i}\frac{\delta_c}{2}\sin\left(\frac{\Delta_c}{2}\right) \end{bmatrix}. \tag{16}$$

On the basis of (16), one can arrive at the conclusion that the normal waves are represented by the two orthogonal circularly polarized states, a partial case of the situation illustrated in figure 1(a).

4. The Jones model for the modulated weakly dichroic crystal

4.1. Polarization of normal waves

Let the basic structure of the crystal be neither optically active nor circularly dichroic. However, the latter effects can exist locally owing to a periodic perturbation of the dielectric tensor by the modulation, provided that the antisymmetric part of this tensor becomes non-zero. In other words, we suppose now that the optical activity k and circular dichroism k' are spatially modulated in a manner shown in figure 2. From symmetry considerations this should take place when, for example, the crystal has a phase transition from a centrosymmetric parent phase to a ferroelectric phase. Another example may be the transitions to incommensurately modulated phases in the A_2BX_4 group crystals (Cummins 1990). Following Dijkstra (1991) and Kushnir and Vlokh (1993) we shall consider analytically the simplest square shape of the modulation wave. Then the crystal can be regarded as a layered optical structure in which layers have alternating k and k' signs (see figure 2). The two neighbouring layers together correspond to a spatial period of the modulation wave. We assume this period to be sufficiently large to allow for the interpretation in terms of macroscopic dielectric parameters but much less than the crystal dimensions. The above conditions are satisfied for both the multidomain ferroelectric and the incommensurate phases.

The layered structure depicted in figure 2 is unipolar as is often with the real multidomain phases. If the layers are identical ($\delta_1 = \delta_2 = \delta$), we have a perfectly periodic modulated structure. Under the conditions discussed in section 3, each of the (optically uniform) constituent layers can be described by the basic Jones matrix (12) in which the parameters Δ and E should be replaced by the phase retardations δ_i and the linear dichroisms e_i in separate layers.

The Jones matrix $\mathbf{T}_M$ of the entire structure is given by

$$\mathbf{T}_M = [\mathbf{T}_H(\delta_2, e_2, -k, -k')\mathbf{T}_H(\delta_1, e_1, k, k')]^N \tag{17}$$

with N the number of the modulation periods fitted in the crystal length. It should be stressed that the Jones matrices of all the phase retardation plates and the partial polarizers

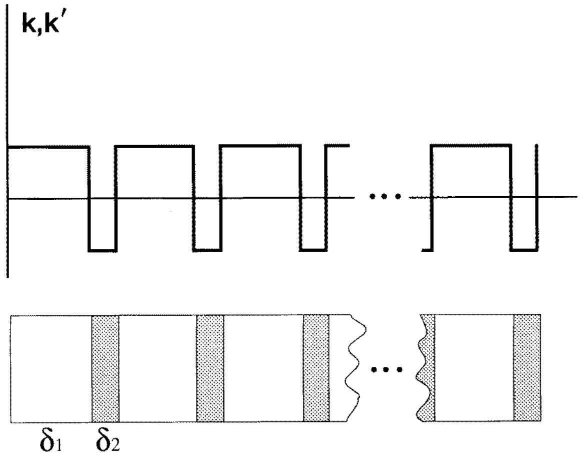

Figure 2. Schematic representation of the modulated crystal with a square wave shape (see text). The modulated optical activity k and circular dichroism k' have opposite signs in the shaded and unshaded optical layers. δ_1 and δ_2 are the phase retardations in the layers.

are unimodular, including those employed in the present work. This permits us to apply the Chebyshev formula for the Nth power of a matrix (Yariv and Yeh 1984) in calculations of (17) which results in

$$\mathbf{T}_M = \begin{bmatrix} \left(1+\frac{E}{2}\right)\exp\left(-\mathrm{i}\frac{\Delta}{2}\right) & -2\frac{k+\mathrm{i}k'}{s_+}(s_- - 2\mathrm{i}s_1s_2)\left[\sin\left(\frac{\Delta}{2}\right)+\mathrm{i}\frac{E}{2}\cos\left(\frac{\Delta}{2}\right)\right] \\ 2\frac{k+\mathrm{i}k'}{s_+}(s_- + 2\mathrm{i}s_1s_2)\left[\sin\left(\frac{\Delta}{2}\right)+\mathrm{i}\frac{E}{2}\cos\left(\frac{\Delta}{2}\right)\right] & \left(1-\frac{E}{2}\right)\exp\left(\mathrm{i}\frac{\Delta}{2}\right) \end{bmatrix}. \tag{18}$$

Here the following notation is introduced:

$$\Delta = N(\delta_1+\delta_2) \qquad E = N(e_1+e_2)$$
$$s_i = \sin\left(\frac{\delta_i}{2}\right) \qquad s_+ = \sin\left(\frac{\delta_1+\delta_2}{2}\right) \qquad s_- = \sin\left(\frac{\delta_1-\delta_2}{2}\right). \tag{19}$$

Using the matrix (18) we are able to find the changes in the character of normal light waves induced by the modulation. First of all, note that in the approximation linear in k and k' the characteristic values of (18) coincide with those of (12), i.e. the observed phase retardation Δ^{ob} and the observed linear dichroism E^{ob} of the modulated crystal are equal to the corresponding values of the homogeneous sample with the same geometry:

$$\Delta^{ob} = \Delta \qquad E^{ob} = E. \tag{20}$$

The influence of spatial inhomogeneity of the medium on the birefringence and linear dichroism can be shown to be quadratic in the modulated parameters, in agreement with the conclusions for incommensurate crystals (Fousek 1991).

The polarization parameters of the normal waves are

$$\xi_{e1}^M = \mathrm{i}s_+^{-1}(k+\mathrm{i}k')(s_- + 2\mathrm{i}s_1s_2) \qquad \xi_{e2}^M = [\mathrm{i}s_+^{-1}(k+\mathrm{i}k')(s_- - 2\mathrm{i}s_1s_2)]^{-1}. \tag{21}$$

From (21) it follows that the waves are not orthogonal and, moreover, the relation (14) formally holds. However, we have, instead of (15),

$$\begin{aligned} \chi_{e1}^{M} &= -s_{+}^{-1}(2ks_1s_2 + k's_{-}) \\ \chi_{e2}^{M} &= \tfrac{1}{2}\pi - s_{+}^{-1}(2ks_1s_2 - k's_{-}) \end{aligned} \qquad (22)$$

and

$$\begin{aligned} \varepsilon_{e1}^{M} &= s_{+}^{-1}(ks_{-} - 2k's_1s_2) \\ \varepsilon_{e2}^{M} &= s_{+}^{-1}(-ks_{-} - 2k's_1s_2). \end{aligned} \qquad (23)$$

Taking $\delta_1 = \delta_2 = \delta$ in (22) and (23), one can obtain the relations for a limiting case of a perfect non-unipolar structure. Finally, we pass to (15) for the enantiomorphous uniform material by putting either $s_1 = 0$ or $s_2 = 0$, together with $s_{-}/s_{+} = 1$.

Essential difficulties arise in the analysis of the eigenwave polarization in a general case when all the anisotropy parameters are comparable in magnitude. For simplicity, we shall therefore consider only the light propagation direction parallel to the optical axis, similarly to section 3. Performing the calculations with the basic Jones matrix $\mathbf{T}_H$ (equation (16)) in the manner described above yields the resulting matrix for the modulated material, where the values Δ_c and δ_c are replaced by the differences in the corresponding parameters characteristic for the neighbouring layers: $\Delta_c^{l1} - \Delta_c^{l2}$ and $\delta_c^{l1} - \delta_c^{l2}$. As a result, the perfect modulated structure ($\Delta_c^{l1} = \Delta_c^{l2}$; $\delta_c^{l1} = \delta_c^{l2}$) appears to be optically isotropic, while the unipolar structure has a right-circular vibration and a left-circular vibration as the normal modes. This can be illustrated again by figure 1(a).

4.2. Discussion of results

As seen from (22) and (23), polarization of the normal waves in the weakly absorbing modulated crystal cannot in general be interpreted within any of the simple patterns in figures 1(a)–1(c). In the absence of circular dichroism the waves become orthogonal. However, even in this case the optical properties of the medium differ essentially from those of the uniform crystal where $\chi_{e1}^{H} = 0$ and $\chi_{e2}^{H} = \frac{1}{2}\pi$. Namely, the modulation causes a rotation of the principal axes of optical indicatrix associated with the term $2s_{+}^{-1}ks_1s_2$ in (22). A noteworthy fact is that the basic structure belongs to a high-symmetry group corresponding to the uniaxial crystal, for which any changes in the optical indicatrix orientation should be forbidden. Thus the given symmetry breaking has to be ascribed to structural inhomogeneity of the modulated crystal. The optical indicatrix rotation is dependent on the temperature, wavelength, etc, and may therefore be detected experimentally, e.g. in the polarimetric studies on incommensurate materials (Kushnir *et al* 1993). The magnitude of the effect is equal to $k\tan(\delta/2)$ in the perfectly modulated crystal (Kushnir and Vlokh 1993).

An additional effect arises when the structure acquires unipolarity: non-orthogonality of the major axes of the eigenwave polarization ellipses. It is determined by the term $\pm 2|s_{+}^{-1}k's_{-}|$ (cf the homogeneous dichroic crystal (section 3)).

According to a common belief, a non-zero eigenwave ellipticity is to be ascribed to the optical activity effect. Then the analysis of (23) proves the optical activity in the dichroic modulated materials to have some mutual features with that in transparent materials (Kushnir and Vlokh 1993). Specifically, a non-zero eigenwave ellipticity results from unipolarity of the structure. However, another contribution is also present in (23) originating from the circular dichroism k' in the optical layers. This is the only effect characteristic of the perfect structure, although its size is relatively small with respect to the effect arising from optical activity in the layers. We recall that the spatial average of the perfect structure has inversion symmetry. The effect mentioned should therefore be related to a combined influence of the modulation of dielectric parameters and the absorption anisotropy.

Of particular interest is comparison of the ellipticities of the two normal waves. In both the uniaxial non-dichroic homogeneous crystal (equation (1)) and the modulated crystal (Kushnir and Vlokh 1993) the condition $\varepsilon_{e2} = -\varepsilon_{e1}$ holds, as it also does in a weakly dichroic homogeneous crystal (equation (15)). For the transparent modulated crystal the conclusion may be arrived at, for example, by checking the behaviour of ε_e (Kushnir and Vlokh 1993) when the crystal is rotated by the angle $\frac{1}{2}\pi$ around the light propagation direction. Since the principal axes are then replaced, we have $\varepsilon_{e1} \to \varepsilon_{e2}$, $\delta_i \to -\delta_i$, $\Delta \to -\Delta$ and $k \to -k$. In the absorbing modulated crystal (see (23)),

$$\begin{aligned} \varepsilon_{e1}^M &= \varepsilon_{OA} + \varepsilon_{CD} \\ \varepsilon_{e2}^M &= -\varepsilon_{OA} + \varepsilon_{CD} \end{aligned} \tag{24}$$

where ε_{OA} and ε_{CD} denote the terms with k and k', respectively. This contrasts drastically with the usual situation in uniaxial materials but resembles that described by (5). Specifically, $\varepsilon_{e2}^M = \varepsilon_{e1}^M$ in the perfectly modulated structure, as in a biaxial birefringent, linearly dichroic crystal (figure 1(c)). Although having their major axes crossed, the normal elliptical vibrations are rendered non-orthogonal by the fact that they are of the same handedness. Note that here the effect is not concerned with different orientations of the principal axes of dielectric and conductivity tensors (Pancharatnam 1955) but is evoked by the modulation.

It must be stressed that the given effect of circular dichroism is quite new. In all other cases known, the circular dichroism gave either opposite signs of the eigenwave ellipticities (optically active crystal (figure 1(a))) or non-orthogonality of their major axes (birefringent crystal (see, e.g. Konstantinova *et al* 1976, Moxon and Renshaw 1990)).

The nature of the crystal optical effects in the modulated medium can be elucidated more by considering their behaviour under the symmetry operation of time inversion (Kushnir and Vlokh 1993). Then $k \to -k$ and $\delta_1 \leftrightarrow \delta_2$ should be put in (22) and (23). As a result, ε_{OA} remains invariant under the operation, unlike the term ε_{CD}. In a hypothetical crystal with no birefringence and no linear dichroism, the behaviour of ε_{CD} should lead to an alteration in the sign of the measured optical rotatory power when the light passes through the sample in the opposite direction, similarly to the magneto-optical Faraday effect. Qualitatively the same may be said of the birefringent, linearly dichroic crystal sections, although there is no pure optical rotation in the latter case. However, it is known (Konstantinova *et al* 1969) that, when birefringence is present in the crystal, the sign of the optical activity may be determined from the thickness-dependent variations in the emergent light polarization azimuth χ. If the incident light is linearly polarized in the principal plane, we have for a transparent crystal (Konstantinova *et al* 1969)

$$\chi = k \sin\Delta \tag{25}$$

with k the normal wave ellipticity taken from (2). A similar relation for the weakly dichroic modulated crystal may be derived using (18):

$$\begin{aligned} \chi = s_+^{-1}(1-E)\{&k[s_- \sin\Delta - 2s_1 s_2(1+E-\cos\Delta)] \\ &-k'[s_-(1+E-\cos\Delta) + 2s_1 s_2 \sin\Delta]\}. \end{aligned} \tag{26}$$

As seen from (26), all the terms available in (22) and (23) contribute to the emergent light polarization. Under the time inversion operation the contribution to χ originating from ε_{CD} changes its sign, thus testifying that the normal wave ellipticity ε_{CD} in (23) in no case can be explained as the optical activity effect. Moreover the above-mentioned methods of

Konstantinova *et al* (1969) for determining the optical activity sign should not be in general applicable to the crystals under consideration.

On the whole, despite the fact that a non-zero ellipticity of the waves propagating through the dichroic modulated medium can be easily revealed within the model developed, it is much more difficult to interpret unambiguously the effects found there. All of them are a distinct result of the modulation being produced by the local optical activity k or circular dichroism k' in the optical layers. Nevertheless the effects cannot be simply identified with the 'optical activity' or 'circular dichroism' for that reason only. A good illustration may be the term in equation (22) proportional to ks_1s_2 which, surprisingly, gives rise to a rotation of the optical indicatrix. The most consistent definition of the optical activity proceeds from a material equation describing the light propagation in the dielectric medium (Agranovich and Ginzburg 1979). This way, however, seems to be clear enough only in the simple case of uniform non-absorbing media and gives little when applied to inhomogeneous materials. As shown above, the spatially averaged complex gyration tensor in the dichroic modulated crystal may be zero, but the normal waves become elliptical owing to 'spatial dispersion' of this tensor, unlike the crystal optics of the uniform transparent crystals. Another apt example is reported by Dijkstra (1991): the 'optical activity' resulting from modulation of the purely symmetric dielectric tensor. This can be understood in terms of local deviations of the optical indicatrix axes. Regarding the material equations, in both cases quoted, the nature of the 'optical activity' is quite different from that of the uniform crystals. Hence the effects described by (23) may be referred to as the optical activity only conventionally, in view of the fact that they cause elliptical polarization of the normal waves.

5. Concluding remarks

In this work the crystal optical effects in the inhomogeneous absorbing medium are discussed on the basis of the well known superposition principle for the optical anisotropy parameters and the Jones calculus methods. A visual Jones model is developed for the dichroic crystal with a modulated dielectric function. We considered a relatively simple and practical case of a uniaxial, weakly dichroic material, assuming that the antisymmetric part of its dielectric tensor was modulated with a square wave shape. Although the basic structure is neither optically active nor circularly dichroic, the modulated structure formed by local periodic perturbations of the complex gyration tensor reveals a number of unexpected effects which manifest themselves in the polarization of the normal waves propagating in the crystal. Those are the optical indicatrix rotation, the non-orthogonality of polarization ellipses of the normal waves and their elliptical polarization caused by the modulation, i.e. both local optical activity and circular dichroism. In relation to their origin, at least the last two effects have to be qualified as quite new and cannot be interpreted in terms of the usual crystal optical phenomena such as the optical activity and circular dichroism occurring in the uniform crystals. One can thus see that the optics of inhomogeneous crystals have essential differences from that of uniform materials. The dichroic crystals represent the most prominent example. In fact, the symmetry of the uniaxial, weakly dichroic crystal is broken due to the modulation and, in accordance with the character of the normal waves, it becomes optically equivalent to an absorbing biaxial crystal.

A key point is that the effects imposed by the modulation in fact disappear when light propagates along the optical axis. So, the structure with the perfect modulation wave has similarly rotating elliptically polarized normal modes when birefringence and linear dichroism are present. However, the eigenmodes do not transform into circularly polarized states with the same handedness on approaching the optical axis direction, but the structure suddenly becomes optically isotropic. This discontinuity shows the important role of anisotropy for the linearly polarized waves in the existence of the effects described by the eigenmode ellipticity ε_{CD}. The origins of the optical activity in transparent modulated crystals are also different in the two alternative cases considered, as found by Kushnir and Vlokh (1993). Furthermore, no optical activity that can be related to structural modulation has been reported experimentally up to now for the weakly anisotropic directions of optical axes in the incommensurate crystals of the A_2BX_4 group. In our view, the latter point must be taken into account for proper understanding of the problem of optical activity in incommensurate phases. We have therefore to emphasize that the reason for the peculiar crystal optical phenomena in the modulated medium is a combined effect of the modulation and 'mixing' anisotropy for linearly and circularly polarized light waves taking place for the wave normal far from the optical axis directions.

The results of the present paper should explain, first of all, the optical properties of the multidomain ferroelectric and the incommensurate phases in the soliton regime. However, using the conclusions of study by Kushnir and Vlokh (1993), we may assume the main results to be correct also in the plane wave modulation regime occurring immediately below the parent-to-incommensurate phase transition. The important questions remaining within the model and disregarded here are its sensitivity to the boundary conditions for the modulation wave and the size of the optical effects, the points considered in more detail by Dijkstra (1991), Dijkstra *et al* (1992a) and Kushnir and Vlokh (1993). To compare the theory with experiments, further experimental investigations of the optical anisotropy in absorbing dielectrics are necessary.

Acknowledgment

This work was supported by a Scholarship from the Ukrainian Cabinet.

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