Multiple Reflections in Crystals: Natural and Faraday Optical Activity

Kushnir O.S.

Electronics Department, Lviv National University, Lviv-54, P. O. Box 3154, 79054 Lviv, Ukraine e-mail: o_s_kushnir@rd.wups.lviv.ua

Received: 01.09.2004

Abstract

A previous analysis of the multiple reflections (MR) of light in crystals is extended to description of particular cases of natural and magnetooptic Faraday optical activity. The differences in manifestations of the MR in the latter effects are discussed and compared with the experimental results. In particular, the MR do not affect the optical rotation related to the natural gyration in case of a rigorously normal light incidence. The modifications of the transmittance fit-function of the known polarimetric HAUP technique imposed by the MR are specified. It is shown that the MR in crystals should produce a kind of spurious dichroism.

Key words: crystal optics, multiple reflections, Jones matrices, birefringence, gyration, dichroism, polarimetry, HAUP.

PACS: 78.20.Ci, 78.20.Fm, 42.25.Ja, 07.60.Fs

Introduction

In a recent paper [1], we have studied, by means of the Jones calculus technique, the effect of multiple reflections (MR) on the optical transmittance and polarization characteristics of light in weakly anisotropic absorbing optically uniaxial crystals. However, Herreros-Cedrés et al [2] have considered the particular case of light propagation along the optic axis and on this basis objected to the reflection matrix used in [1] and so our results concerned with the light polarization. Furthermore, some important recent results have gone unnoticed by us, dealing with the measurements and interpretation of the MR effect within the well-known high-accuracy universal polarimeter (HAUP) technique [3-6], the subject that has been also addressed in the study [1]. It has turned out that different authors use for decades the transmittance fit-functions J_{PSA} (see [1] for the detailed explanations of the notation used hereafter) of this widely recognized technique, in which the corrections for MR are sometimes essentially different (see [1,3-8]). All these points clearly need a close consideration.

Derivation of the Jones matrix and analysis of the results

The authors [2] suggest a general relation for the Jones matrix (JM) **M** of crystals in the presence of MR,

$$\mathbf{M} = \mathbf{t}'\mathbf{M}_0 \sum_{m=0}^{\infty} (\mathbf{r}\mathbf{M}_0^{rev}\mathbf{r}\mathbf{M}_0)^m \mathbf{t}, \qquad (1)$$

where $\mathbf{M}_0$ is the single-pass JM, $\mathbf{M}_0^{rev}$ the JM associated with the reversed light propagation direction, **r** the amplitude reflection matrix and **t**, **t'** the amplitude transmission matrices referred to the entrance and exit faces of a crystal plate (the standard geometry is represented in Fig. 1). When combined with the appropriate JM $\mathbf{M}_0^{rev}$, formula (1) generalizes the corresponding relation in [1], where $\mathbf{M}_0^{rev} = \mathbf{M}_0$ is in fact taken. Since we analyse

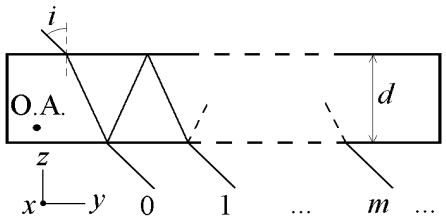

Fig. 1. Schematic representation of MR in an optically uniaxial crystal plate (see [1]). O.A., the optical axis; *x*, *y*, *z*, the principal axes of the optical indicatrix ellipsoid. The incidence angle *i* is taken non-zero only to visualize the multiple beams. The small difference between the refraction angles for ordinary and extraordinary beams is not shown.

the most general case of optical anisotropy for both linearly and circularly polarized waves, the two types of optical gyration effects are to be distinguished further on: a natural optical activity (NA) associated with the dielectric permittivity components ε_{ij} linear in the wave vector **q** and a magnetooptic Faraday optical activity (FA), for which the ε_{ij}'s are dependent on the (external or internal) magnetic field (see, e.g., [9,10]).

When the light passes through the crystal in the opposite direction ($\mathbf{q} \to -\mathbf{q}$), the mentioned terms in ε_{ij} (and so the corresponding complex circular birefringence $\Delta n_C + i\Delta\kappa_C$) change their signs in case of the NA and remain invariant in case of the FA (see, e.g., [11]). On the basis of the relations between the dielectric tensor and the JM, one infers that the same holds true of those parts of components of the JM that include the odd functions of $\Delta n_C + i\Delta\kappa_C$. As a result of such the symmetry difference, the NA and FA would behave quite differently with respect to the MR. The two examples cited in [2] for the first-order ($m = 1$) MR beam correspond respectively to the FA and the NA. In other words, it is necessary to put $\mathbf{M}^{rev}_{0,FA} = \mathbf{M}_{0,FA}$ and $\mathbf{M}^{rev}_{0,NA}(\Delta n_C + i\Delta\kappa_C, k) = \mathbf{M}_{0,NA}(-\Delta n_C - i\Delta\kappa_C, -k)$, where k is the normal wave ellipticity angle. Still more intricate situations may happen when finding the JM $\mathbf{M}^{rev}_0$, e.g., for the crystals possessing a non-reciprocal gyrotropic linear birefringence Δn^{nr}_L [11–13] added to the usual linear birefringence Δn_L. Again, we should put $\mathbf{M}^{rev}_0(\Delta n^{nr}_L) = \mathbf{M}_0(-\Delta n^{nr}_L)$.

As follows from the said above, the method $\mathbf{M}^{rev}_0 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \mathbf{M}_0 \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ [2] for calculating the reversed JM represents an artificial attempt to solve the problem using purely geometrical means within the Jones formalism, rather than real physical arguments. Nevertheless, it succeeds by chance in a particular case of NA when being combined with the incorrect reflection matrix (see below). However, the method fails whenever the JM $\mathbf{M}_0$ from [1] is written in reference systems that do not coincide with the principal (crystallophysical) one ($\mathbf{M}_0' = \mathbf{R}(-\theta)\mathbf{M}_0\mathbf{R}(\theta)$, with $\mathbf{R}(\theta)$ being the rotation matrix) [1]. Then the terms $\Delta n_L + i\Delta\kappa_L$ and $\Delta n_C + i\Delta\kappa_C$ are "mixed" in all of the $M'_{0,ij}$ components, while the method [2] prescribes simply to change signs of the off-diagonal components ($M'^{rev}_{0,ij} = -M'_{0,ij}$, $i \neq j$) [2]. It fails also in cases of the FA or the non-reciprocal birefringence.

Let us now touch upon the reflection matrix used in the calculations. Its meaning consists in nothing but defining the amplitude reflectivities and the phase relations of the incident and reflected waves. The analysis of the Fresnel formulae under the conditions specified in [1]

[1] By the way, one must work in such the reference systems, e.g., in case of the light propagation along the optic axes in biaxial crystals.

[2] Contrary to the view [2], the relation $M_{0,21} = -M_{0,12}$ does not represent a general property of JMs describing anisotropic crystals. It gets broken in an arbitrary reference system ($\theta \neq 0, 90^\circ$), even in the simplest case of transparent purely linearly birefringent crystal (see formulae (2)–(4) in [1]).

(first of all, the normal light incidence ($i = 0$) and the weak optical anisotropy) leads to the JM $\mathbf{r} \approx r\mathbf{I}$ ($\mathbf{I}$ being the identity matrix) and the authors [2] themselves agree with the arguments [1]. It is quite another matter that the JM $\mathbf{M}$ [1] does not describe the NA (see [2]). This originates from the relation $\mathbf{M}_0^{rev} = \mathbf{M}_0$ [1] describing the FA and has nothing to do with the matrix $\mathbf{r}$. It is easy to prove (see [2] and subsection 2.1 in [1]) that the relation (1) suggested in [2], combined with our $\mathbf{r}$ and the general technique for calculating $\mathbf{M}_0^{rev}$ presented here, give the correct results for both the NA and FA cases.

As mentioned above (see also [2]), the simultaneous use of the algorithms $\mathbf{r} \approx r\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ and $\mathbf{M}_0^{rev} = \mathbf{M}_0$ for the case of NA in the work [14] (further reproduced in [4-6]) has also led to the correct JM, although both algorithms separately should be considered as inadequate. To our opinion, the method [14] has represented merely a practical means for deriving the JM, without being involved into lengthy discussions of the reversibility properties and physical justification of the $\mathbf{r}$ form.

Thus, the results [1] for the circular birefringence Δn_C and dichroism $\Delta\kappa_C$ correspond to the FA and the magnetic circular dichroism, which have been dealt with in the studies [15-17] appealed to in [1]. Nevertheless, most of those results are universal and valid also for the case of NA. Below, we shall specially mention all the exceptions that are "sensitive" to the reversal behaviour of Δn_C and $\Delta\kappa_C$.

First, the JM of crystals with the NA affected by the MR, written in the principal reference system, is as follows (cf. with [1]):

$$\mathbf{M}_{NA} = F_{NA}\mathbf{m} = F_{NA}\begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix}, \qquad (2)$$

where

$$\begin{aligned} F_{NA} &= \frac{(1-R)e^{i\varphi-\alpha d/2}}{1-2Re^{2i\varphi-\alpha d}[\cos^2(\Delta/2)-\cos 4k\sin^2(\Delta/2)]+R^2e^{4i\varphi-2\alpha d}} \\ m_{11} &= (1-Re^{2i\varphi-\alpha d})\cos(\Delta/2) - i(1+Re^{2i\varphi-\alpha d})\cos 2k\sin(\Delta/2), \\ m_{12} &= -m_{21} = -(1-Re^{2i\varphi-\alpha d})\sin 2k\sin(\Delta/2), \\ m_{22} &= (1-Re^{2i\varphi-\alpha d})\cos(\Delta/2) + i(1+Re^{2i\varphi-\alpha d})\cos 2k\sin(\Delta/2) \end{aligned} \qquad (3)$$

Here d denotes the thickness of the crystal plate, φ the "isotropic" phase shift upon a single pass through the plate, α the mean absorption coefficient, Δ the total (complex) "phase retardation" defined by the superposition principle in crystal optics ($\Delta = (2\pi d/\lambda)\sqrt{(\Delta n_L + \Delta\kappa_L)^2 + (\Delta n_C + \Delta\kappa_C)^2}$, with λ being the light wavelength in vacuum), and $R = r^2$. Comparing with formulae (8) in [1], the sign of the R term in m_{12} and m_{21} is opposite, while for the "isotropic" pre-factor we have $F_{NA} = F_{FA}$ only when the circular anisotropy is absent ($k = 0$).

Analysis of the results

Unlike the results [1], the JM (2) reduces to the matrix obtained by Melle [14] for transparent crystals with the NA (see formulae (13) from [14]) and, when ignoring the pre-factor F, to the matrix [6] for the propagation direction along the optic axis ($k = \pm\pi/4$). On the other hand, the JM $\mathbf{M}_{NA}$ derived in [4] for the experimental HAUP geometry ($\alpha = \Delta\kappa_L = \Delta\kappa_C = 0$, $k << 1$; the factor F dropped, i.e., $F = 1$) differs from (3) by replacement of $R\exp(2i\varphi)$ with the "scalar reflection parameter" $R\cos 2\varphi$ [7]. Inspection of the calculations [4] shows that all the terms $iR\sin 2\varphi$ are discarded without any reasoning (cf. formulae (27)–(30) and (31)–(34) from [4]).

There is a sharp physical distinction between our $\mathbf{M}_{NA}$, $\mathbf{M}_{FA}$ matrices and the JM [4]. It is well known that the MR in transparent isotropic material (for much familiarity, placed into Fabri-Perot resonator) produce an "apparent

absorption" $\alpha_{eff} \neq 0$ (in reality, a destructive interference at certain wavelengths, thicknesses, refractive indices n and R). For anisotropic crystal, the transmittances of the Fabri-Perot etalon would be obviously different for the ordinary and extraordinary rays, due to the inequality $n_o \neq n_e$. We arrive naturally at an idea of "spurious dichroism" originated from the MR (see the parameter δ_{eff} in [1]). Though without coming to terms, the effect has been in fact described by Holmes [18] in frame of the exact electromagnetic theory for transparent linearly birefringent crystals (see also the conclusions [7] about similar actions of the MR and the linear dichroism on the light transmittance within the HAUP). The fact of availability of apparent absorption and dichroism due to the MR must be reflected in violation of a set of sufficient conditions $m_{22}^* = m_{11}$, $m_{21}^* = -m_{12}$ and $\det \mathbf{m} = 1$ for unitarity of the corresponding JM. Both the results [14] and the R-dependent terms in formulae (3) testify that. Surprisingly, the JM [4] is unitary and so disagrees with the possibility for differential transmittances of the normal waves.

Similarly to [1], let us analyse the matrix (2). The complex amplitude ratios for the normal waves $\xi_{e1,2}$ and the eigenvalues $V_{e1,2}$ may be written as

$$\xi_{e1,2}^{NA} = \frac{i(\mp\sqrt{1+2Re^{2i\varphi-\alpha d}\cos 4k + R^2 e^{4i\varphi-2\alpha d}} - \cos 4k)}{(1-2Re^{2i\varphi-\alpha d})\sin 4k},$$
$$V_{e1,2}^{NA} = F[(1-Re^{2i\varphi-\alpha d})\cos(\Delta/2) \mp i\sqrt{1+2Re^{2i\varphi-\alpha d}\cos 4k + R^2 e^{4i\varphi-2\alpha d}}\sin(\Delta/2)] \quad (4)$$

Unlike the FA [1], the MR affect the elliptical polarization of normal waves in the general case of $k \neq 0, \pm\pi/4$, and the waves become slightly non-orthogonal ($\xi_{e1}\xi_{e2}^* \neq -1$) due to the R terms. The maximal difference between $V_{e1,2}^{NA}$ and $V_{e1,2}^{FA}$ [1] occurs for the optic axis direction, when $V_{e1,2}^{NA} = F[1 - R\exp(2i\varphi - \alpha d)]\exp(\mp i\Delta/2)$. Here we do not write out a cumbersome relation for the apparent dichroism parameter δ_{eff}^{NA} (in general, it is defined as $\delta = (2\pi d/\lambda)\Delta\kappa_L$), which is zero for the optic axis, exactly equal to δ_{eff}^{FA} [1] if $k = 0$ and very close to the latter in a practical case of $k << 1$. Simplifying δ_{eff} under the conditions $k, R << 1$ (see also formula (10) from [1]), we get

$$\delta_{eff} \approx 2R\sin 2\varphi \sin\Delta. \quad (5)$$

On the other hand, the parameter $f = (E_{out,y}/E_{in,y})/(E_{out,x}/E_{in,x})$ included in formulae (14) and (16) in [18], which defines the ratio of amplitude transmittances for the normal waves linearly polarized along the axes x and y, means in fact the same "apparent dichroism" ($f \approx \delta_{eff}$). In the weak optical anisotropy approximation, the relations [18] for f agree excellently with (5), as well as the formulae (30) and (34) from [1] for the experimental parameters used to detect the dichroic consequences of the MR.

It is interesting to compare manifestations of the MR in the conditions of maximal difference between the NA and FA, i.e. for the light propagation direction exactly along the optic axis ($k = \pm\pi/4$). Taking $\alpha = 0$ for simplicity, we obtain

$$\mathbf{M}_{FA} = \frac{(1-R)(1-Re^{2i\varphi})e^{i\varphi}}{1-2Re^{2i\varphi}\cos\Delta + R^2 e^{4i\varphi d}} \begin{pmatrix} \cos(\Delta/2) & -\dfrac{1+Re^{2i\varphi}}{1-Re^{2i\varphi}}\sin(\Delta/2) \\ \dfrac{1+Re^{2i\varphi}}{1-Re^{2i\varphi}}\sin(\Delta/2) & \cos(\Delta/2) \end{pmatrix}, \quad (6)$$

$$\mathbf{M}_{NA} = \frac{(1-R)e^{i\varphi}}{1-Re^{2i\varphi}} \begin{pmatrix} \cos(\Delta/2) & -\sin(\Delta/2) \\ \sin(\Delta/2) & \cos(\Delta/2) \end{pmatrix}. \quad (7)$$

Choosing the optimum absolute single-pass phase shift φ of the wave ($\varphi = \pi p$, p being integer), one can attain "amplification" of the single-pass optical rotation $\phi^{(0)}$ ($\phi^{(0)} = \Delta/2$) for the FA [16], as a result of the corresponding reversal behaviour. According to (6), the "amplification factor" $Q = \phi/\phi^{(0)}$ is equal to $Q \approx (1+R)/(1-R)$, when $\phi^{(0)} << 1$. On the contrary, the anisotropic part **m** of the JM (7) for the NA does not contain any MR terms. Since it is well-known (see, e.g., [19]) that the polarization characteristics of the light emergent from crystal are affected by the JM **m** only, one comes to the conclusion about a complete absence of polarization influence of the MR. Then the formulae (18) and (19) of subsection 2.2.3 in [1] should be replaced with

$$\begin{aligned} &\phi = \chi - \theta = \phi^{(0)} = \chi^{(0)} - \theta = \Delta/2, \\ &\sin 2\varepsilon = \sin 2\varepsilon^{(0)} = \tanh\delta \end{aligned}, \quad (8)$$

where $\Delta = (2\pi d/\lambda)\Delta n_C$, $\delta = (2\pi d/\lambda)\Delta\kappa_C$, θ is the incident polarization azimuth, $\chi^{(0)}$, χ and $\varepsilon^{(0)}$, ε the emergent polarization azimuths and ellipticities, respectively, for the single and multiple pass through the crystal [1]. The result (8) agrees with both the theoretical and experimental findings by Melle [14] for the case of normal light beam incidence. In other words, under the condition of $i = 0$ the MR *do not influence* the light polarization for the light propagating along the optic axis and the corresponding NA characteristics. In this respect, formula (33) for the ϕ parameter in [6], including the MR effect for $i = 0$, represents a clear inaccuracy, though the MR does affect the light transmittance under the given conditions.

MR in the HAUP-type experiments

Now we proceed to analysis of crystal optical parameters measured with the HAUP-type techniques for transparent materials with the NA (see subsection 2.2.4 in [1]). The azimuth χ and the ellipticity ε of the emergent light under the HAUP conditions are expressed in the form

$$\begin{aligned} \chi &= A\theta - Bp + \delta Y + \\ &\quad k[\sin\Delta - 2R(1-\cos\Delta)\sin(2\varphi+\Delta), \\ \varepsilon &= B\theta + + Ap + \delta\varepsilon + \\ &\quad + k(1-\cos\Delta)[1+2R\cos(2\varphi+\Delta)]] \end{aligned}, \quad (9)$$

where $\Delta = (2\pi d/\lambda)\sqrt{\Delta n_L^2 + \Delta n_C^2}$, p, δY and $\delta\varepsilon$ are the instrumental imperfection parameters (see, e.g., [1,7]) and the standard functions A and B are unaltered when compare to the case of FA ($A = \cos\Delta - 2R\sin(2\varphi+\Delta)\sin\Delta$ and $B = [1+2R\cos(2\varphi+\Delta)]\sin\Delta$ [1]). Then formula (36) from [1] for $\theta = 0$ changes to the form $\chi \approx k[\sin\Delta - 2R(1-\cos\Delta)\sin(2\varphi+\Delta)]$, although the "theoretical" $\chi(\lambda)$ oscillation amplitude A_{th} ($A_{th} \approx 2kR$) and the conclusions of subsection 3.5 [1] remain valid. The k-dependent terms in the characteristic azimuth θ_i and ellipticity ε_0 (see formulae (22) in [1]) would modify accordingly.

The most important, the linear birefringence dominates in the experimental HAUP geometry and so the differences between the manifestations of the MR in the NA and FA become insignificant. As a result, the coefficients C_{13}, C_{22} and C_{31} of the HAUP fit-function $J_{PSA} = \hat{Y}\hat{C}\hat{\theta}'$ [1,7] remain the same for both the NA and FA, i.e., we have again (see [1])

$$\begin{aligned} c_{13} &= \cos 2\varphi(1+2\cos\Delta) - 1, \\ c_{22} &= \sin(2\varphi+\Delta)\cot(\Delta/2) - \\ &\quad - 2\sin^2(\varphi - \Delta/2), \\ c_{31} &= -2\sin^2(\varphi - \Delta/2) \end{aligned} \quad (10)$$

if the representation $C_{ij} = C_{ij}^{(0)}(1+2c_{ij}R)$ is used, with $C_{ij}^{(0)}$ being the coefficients in the absence of MR. Moreover, it follows from analysis of the data [3] that the assumption [1] about the re-definition of the quantity C_{31}

($C_{31}=1$), which depends upon the pre-factor F and is common for the all C_{ij} components, has indeed been right. This gives the quite equivalently expressed coefficients $c_{13}=\cos 2\varphi\cos(2\varphi+\Delta)$, $c_{31}=0$ and $c_{22}=\sin(2\varphi+\Delta)\cot(\Delta/2)$, in a perfect consonance with the results [3]. Since the authors [5] have earlier checked out a practical identity of their C_{ij}'s found in the approximation of the first-order MR ($m=1$) and those of the work [3], we infer that formulae (10) are well coordinated with the fit-function [3,5,8] and represent the correct solution of the problem for the MR effect [3]. In particular, we obtain the quantity that characterises the MR (or the relevant "effective dichroism") the simplest way (see (5) and formula (30) from [1]):

$$(C_{22}-C_{13})/(2C_{31})\approx(C_{22}-C_{13})/2=\\ =2R\sin 2\varphi\sin\Delta \quad . \qquad (11)$$

In consonance with the results [3] (section 5) and [5] (subsection 4.3) and similar to the linear dichroism effect [7], the MR "decouple" the basic functions $Y\theta'$ and θ'^2 of the decomposition J_{PSA} (i.e., $C_{22}\neq C_{13}$ [1]). However, the coefficients $c_{13}=c_{22}=2\cos 2\varphi\cos^2(\Delta/2)$ from [4,6] correlate with neither the results [1,3,5,8] nor the earliest data $c_{13}=c_{22}=\cos 2\varphi$ [7]. Basing on the said above, they should be most probably regarded as invalid. It is also worth noticing that appealing to agreement with the results [18] for the pure birefringence (see [4]) does not seem to be justification enough, since the effect of the other optical properties (NA, dichroism, etc) has been ignored in [4].

In order to decide definitely between the two alternative sets of the C_{ij} components, experimental measurements of the well resolved (large-period) oscillation temperature dependences $C_{13}(T)$ and $C_{22}(T)$ may be recommended, with focusing at the relative oscillation phase shifts. The same refers to catching the difference in behaviours of the output light polarization (the χ and ε parameters) for the NA and FA. The most detailed data for the MR oscillation dependences under the rotation of sample by 90° around the beam direction ($\Delta\to-\Delta$ and $k\to-k$) reported so far (see Fig. 6a [3], Fig. 15 [5] and Fig. 1 [8]) strongly indicate to validity of the relations [1,3,5,8]. They show a clear oscillation phase shift after that rotation, consistent with the arguments $(2\varphi+\Delta)$ or $(2\varphi+\Delta/2)$ of the functions C_{13} and C_{22}, rather than the arguments 2φ and Δ separately (the phase shift in the latter case would have been zero).

Analysis of experiments for the pure natural optical activity

Finally, let us concentrate on the interpretation of experimental NA data for the optic axis directions. The relevant oscillating temperature dependences $\phi(T)$ measured with a conventional polarimeter may be found, e.g., in [1] (Fig. 5), [20] (Fig. 1) and [6] (Fig. 12a) respectively for α-ZnP_2 and quartz crystals (see also the data [21] for $LiIO_3$), whereas the $\varepsilon(T)$ dependence for α-ZnP_2 has been represented in [20] (Fig. 3). The example for α-ZnP_2 is cited in Figure 2. Unlike the study [14], the authors [1,6,20] have analysed those data using the invalid relations [1,6] that predict the MR influence (the theoretical oscillation amplitude $A_{th}\approx 2R\phi^{(0)}$, or $A_{th}/\phi^{(0)}\sim 8\%$ for the refractive indices ~ 1.5) at the normal incidence. Nevertheless, they have succeeded in explaining the observed $\phi(T)$ and $\varepsilon(T)$ periodicity (see Figure 2). What has been really established in [1,6,20] is the fact that the above oscillations are indeed to be prescribed to the MR, following

[3] When employing those quantities in practice, one should remember a possibility for opposite signs in the definitions of Δ, φ and Y parameters used by different authors (see [1]).

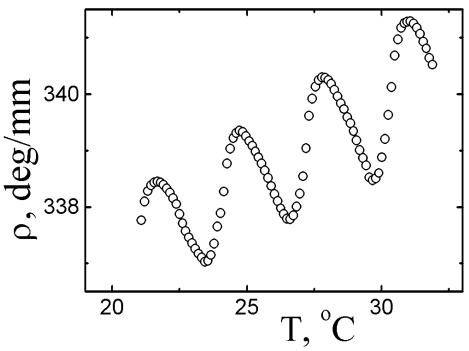

Fig. 2. Temperature dependence of specific optical rotatory power $\rho = \phi / d$ due to NA for α-ZnP_2 crystals (d = 0.34 mm, heating run, temperature scan rate 6 °C/h – see the data [1]).

from the temperature period values. However, in order to decide among the specific theoretical relations for $\phi(T)$, one should again analyse the oscillation amplitude and phase. Comparison of A_{th} with the experimentally observed amplitude A_{exp} for $\phi(T)$ and $\varepsilon(T)$ in α-ZnP_2 (Figure 2) has given unexpectedly small parameters $\beta = A_{exp} / A_{th}$ ($\beta \sim 0.02 - 0.04$ – see also [1,20]). It is an order of magnitude smaller than that found for the *x*-cut sample tested for the ordinary birefringence [1], in spite of that the latter sample has larger dimensions (d = 1.41 mm) and it is its β that should be possibly smaller, due to a stronger reduction of the parameters β_V and β_{SR} by the light scattering ($\beta = \beta_V \beta_{SR} \beta_p$, with β_p being the surfaces' non-parallelity contribution – see [1]). According to our calculations, the results [6] for quartz also demonstrate very small oscillation amplitudes ($\beta \sim 0.03$ at the most – see Table), again unlike the anisotropic birefringent propagation directions. The only remained assumption has been a notable non-parallelity of the entrance and exit faces of the crystal plate [1,6,20].

Let us now use the order-of-magnitude estimation $\beta_p \approx \sin(a\gamma\varphi/d)/(a\gamma\varphi/d)$ (a being the diameter of the probing light beam, γ the angle between the sample faces and $a\varphi/d \sim 10^4$) [3] and the results for the MR in the NA along the optic axis reported in a number of studies [1,6,14,20–22]. As seen from Table, the sample wedge angles necessary for efficient reduction of the MR are $\sim 10'$. As a matter of fact, the value $\gamma \sim 1 - 5''$ represents high enough commercial standards required only if the laser resonators or special-purpose plane-parallel plates are dealt with, while the values $\gamma \sim 10 - 50''$ are quite bearable and cannot be the reason for the fact of $\beta \sim 0.03$. It is therefore impossible to explain the extreme weakness of the MR oscillations observed in [6] on the basis of formulae like $\phi = \phi^{(0)}(1 - 2\beta_p R \cos 2\varphi)$ (see [6]).

The correct interpretation should be based on the fact of rising of the MR effect with a deviation of light propagation direction from the optic axis (cf. the JMs (2) and (7)). In practice, this should mean a nonzero incidence angle $i \neq 0$ [14] which, when using no special means, may be as large as $i \sim 0.5 - 2^\circ$. Derivation of explicit functions $\phi(T)$ and $\varepsilon(T)$ in case of $i \neq 0$ and the presence of the MR is enormously complicated task, because the parameter i, determining the refractive index values, affects all the intermediate quantities, including the one-surface reflectances and transmittances (see subsection 2.1 in [1]). We shall confine ourselves to rough estimations of $\phi(T)$ and $\varepsilon(T)$ oscillation amplitudes, using the experimental and numerical-solution $\phi(i)$ and $\varepsilon(i)$ data by Melle [14] for the quartz crystals. Following from the results [14] and the proportionality $A_{exp} \propto 2\phi^{(0)} \beta R \exp(-\alpha d)$ ($\beta \sim 1$), the values $A_{exp} \approx 0.04$ deg ($\phi^{(0)} \approx 20$ deg, $d \approx 1$ mm) for the quartz [6] and $A_{exp} \approx 0.4$ deg ($\phi^{(0)} \approx 120$ deg – see Figure 2) for α-ZnP_2 should correspond respectively to $i \approx 0.9^\circ$ and

Table. Some experimental and calculated parameters concerned with the MR manifestations in the NA measured nearly along the optic axis.

Crystal and reference	Sample wedge angle γ	Sample-wedge MR reducing factor β_p	Total MR reducing factor β*	Angle of incidence i
SiO_2 [14]	5 – 20'' **	0.9–1	≈ 0	≈ 0
SiO_2 [22]	18'	0.02	≈ 0	?
SiO_2 [6]	2' 20''	0.1 0.9	<0.01 0.03	? 0.9^o
α-ZnP_2 [1,20]	40''	0.5	0.02–0.04	1.3^o $\approx 1^o$ ***

* The values calculated as if the MR were present at $i = 0$;

** The values calculated on the basis of the accuracy of 1 μm (see [14]) for the sample thickness control;

*** The value calculated with the $\varepsilon(T)$ ellipticity data [20].

$i \approx 1.3^\circ$ (see Table). If we compare the result $A_{exp} \approx 4 \cdot 10^{-3}$ [20] for the $\varepsilon(T)$ amplitude in α-ZnP_2 ($A_{exp} \propto \beta R \exp(-\alpha d)$, with $\beta \sim 1$) to the average ellipticity $\sim 4 \cdot 10^{-2}$ predicted for $i = 3^\circ$ [14], the estimation $i \sim 1^\circ$ follows again.

Thus, small values of the total MR-reducing factor β presented in Table reflect a gradual disappearance of the MR with approaching to the $i = 0$ condition, rather than a scattering of light or large sample wedge angles [6]. The true β values, which can be derived after specifying the exact analytic form of the function $A_{th}(i)$, should be most likely $\beta \sim 1$ for the high-quality samples and the wedge angles $\gamma \sim 20''$. It is also evident from the above discussion that there is no need in making wedge-like crystal samples in order to reduce the interference MR effects, which may impose difficulties in the Landau-type thermodynamic analysis or, when unresolved, produce experimental noises for the NA (see [22,23]). The simplest way consists in accurate orientation of crystal in the optical system that ensures the absence of the MR under the exact condition of normal ($i = 0$) light beam incidence.

References

1. Kushnir O.S. J. Opt. A: Pure & Appl. Opt. **5** (2003) p. 478.
2. Herreros-Cedrés J., Hernández-Rodríguez C., Guerrero-Lemus R. J. Opt. A: Pure Appl. & Opt. (submitted).
3. Simon J., Weber J., Unruh H.-G. J. Phys. D: Appl. Phys. **30** (1997) p.676.
4. Gomes P., Hernandez C. J. Opt. Soc. Amer. B **15** (1998) 1147.
5. Folcia C.L., Ortega J., Etxebarria J. J. Phys. D: Appl. Phys. **32** (1999) p.2266.
6. Hernández-Rodríguez C., Gómez-Garrido P. J. Phys. D: Appl. Phys. **33** (2000) p.2985.
7. Moxon J.R.L., Renshaw A.M. J. Phys.: Condens. Matter **2** (1990) p.6807
8. Simon J., Weber J., Unruh H.-G. Ferroelectrics **183** (1996) p.161.
9. Landau L.D., Lifshitz E.M. Electrodynamics of continuous media (New York, Pergamon Press) 1960.
10. Yariv A., Yeh P. Optical waves in crystals (New York, Wiley) 1984.
11. Vlokh O.G. Spatial dispersion phenomena in parametric crystal optics (Lviv, Vyshcha Shkola) 1984 (in Russian).
12. Hornreich R.M., Shtrikman S. Phys. Rev. **171** (1968) p.1065.
13. Pisarev R.V. Ferroelectrics **183** (1996) 39.

14. Melle H. Optik **72** (1986) p.157.
15. de Lang H. Philips Res. Rep. **8** (1967) p.1.
16. Rosenberg R., Rubinstein C.B., Herriott D.R. Appl. Opt. **3** (1964) p.1079.
17. Tronko V.D., Dovgalenko G.E. Opt. Spektrosk. **34** (1973) p.1157.
18. Holmes D.A. J. Opt. Soc. Amer. **54** (1964) p.1115.
19. Azzam R.M.A., Bashara N.M. Ellipsometry and Polarized Light (Amsterdam, North-Holland) 1988.
20. Kushnir O.S., Bevz O.A., Polovinko I.I., Sveleba S.A. Phys. Status Solidi b **238** (2003) p.92.
21. Herreros-Cedrés J., Hernández-Rodríguez C., Guerrero-Lemus R. J. Appl. Cryst. **35** (2002) p.228.
22. Bachheimer J.P. J. Phys. C: Solid State Phys. **19** (1986) p.5509.
23. Bosch G., Jahn I.R., Prandl W., Verhein M. Physica B **142** (1986) p.320.