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Magnetic analog of Pockels' effect in transparent antiferromagnetic crystals

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ABSTRACT

It is shown that light, initially linearly polarized, becomes endowed with elliptical polarisation on traversal of an antiferromagnetic crystal acted on by a static magnetic field (induction $\vec{B}$). The ellipticity can be linear in $\vec{B}$, thus being the Magnetic Analog of Pockels' Effect (MAPE). If the light wave propagates along the z-axis (parallel to the highest-fold symmetry axis of the crystal), the ellipticity will contain only contributions linear in $\vec{B}$ in crystals with the symmetry $\bar{3}m$ ($CoCO_3$) at $\vec{B}=\hat{e}_x B$, in ones with the symmetries $\underline{6}m\underline{m}$ and $\underline{6}/\underline{mmm}$ at $\vec{B}=\hat{e}_y B$, and in ones with the symmetries $\underline{4}/mm\underline{m}$ (CoF_2) and $m3\underline{m}$ ($Dy_3Al_5O_{12}$) at $\vec{B}=\hat{e}_z B$, whereas for $\underline{6}/\underline{m}$, $\underline{6}/\underline{mmm}$ and $\underline{6}m\underline{m}$ at $\vec{B}=\hat{e}_x B$ and for $\bar{3}m$ and $\underline{6}/\underline{m}$ at $\vec{B}=\hat{e}_y B$ the contributions linear in $\vec{B}$ will be accompanied by ones dependent on the square of the magnetic field.

1. INTRODUCTION

In 1956, Tavger and Zaitsev[1] showed that by adjoining time inversion $\underline{1}$ (electric current reversal) as an element of symmetry to the well known 32 point groups G(P) with crystallographically limited fold-ness of their symmetry axes one can construct three kinds of groups G (122 groups in all) as follows: $G(P) \otimes G(\underline{1}) = G(NM)$, G(P) and $G(P') + [G(P) - G)P')] \otimes \underline{1} = G(M)$; where G(P') is a subgroup of the group G(P) [it has to contain one half of the elements of the group G(P)]; whereas G(P) − G(P') denotes the set of those elements of G(P) which did not enter the subgroup G(P').

Magnetic symmetry, once taken into account in different symmetry considerations, has permitted the prediction of numerous new optical effects. Magnetically ordered crystals [the directional symmetry can be described by the groups G(P) and G(M)] with so-called transparency windows[2] in the range of optical frequencies, in the complete absence of perturbing factors (electric or magnetic fields, mechanical stress, etc.) can exhibit, in addition to natural optical birefringence[3,4] and natural optical activity[5-8] [which occur as well for non−magnetic crystals, groups G(NM)], two novel optical effects: natural gyrotropic birefringence (changing its sign on reversal of the light propagation direction),[7-11] and natural gyrotropic rotation.[12-14] Krinchik and Chetkin[15] have shown that in certain magnetically ordered crystals the omission of magnetic susceptibility in the optical frequency range postulated by Landau and Lifshits[16] is unfounded. If taken into account, it gives additional contributions to the above-named optical effects and, moreover, can modify the type of propagation of the light wave.[17,18]

If moreover the crystal possesses spontaneous magnetisation $\vec{M}$, it can exhibit a spontaneous Cotton-Mouton or Voigt, Faraday and optical birefringence effect proportional to the product of $\vec{M}$ and the wave vector of the light wave traversing the crystal.[19,20] These effects resemble the well known effects of Cotton and Mouton, as well as Voigt, and Faraday (the theoretical foundation of which have been expounded in numerous handbooks and monographs)[21-26] as well as optical birefringence proportional to the product of the static magnetic field and the wave vector of the probe beam[5,6,27-30] that occur in non−magnetic media under the action of a static magnetic field. Crystals with spontaneous magnetisation $\vec{M}$ can give rise (in addition to the above) to new effects without counterparts in non-magnetic media. The most important are these:[19] optical birefringence proportional to the first power of the spontaneous magnetisation $\vec{M}$, and rotation proportional to the square of $\vec{M}$ and to the product of $\vec{M}$ and the wave vector of the probe beam. Discussions of the existing theoretical and experimental work in the field of spontaneous magneto-optics are to be found in the respective review articles.[31,32]

The past 10 to 20 years have brought considerable developments in the optical study of magnetically ordered crystals in static magnetic fields.[17,33-39] Here, the magnetic symmetry admits of quite new effects, such as birefringence proportional to the first power of $\vec{B}$ − the Magnetic Analog of Pockels' Effect (MAPE),[17,34-38] as well as rotation proportional to the square of $\vec{B}$[33,38] and proportional to the product of $\vec{B}$ and the wave vector of the light beam.[17] Dillon and co-workers[35] were the first to show that the birefringence linear in $\vec{B}$ can be expected in the longitudinal geometry $\vec{B} \parallel \hat{e}_z \parallel \vec{k}$ (Faraday configuration) in $Dy_3Al_5O_{12}$ crystals ($m3\underline{m}$). The effect has already been measured for the antiferromagnets $DyFeO_3$ (mmm), $Ca_3Mn_2Ge_3O_{12}$ ($\underline{4}/m$), CoF_2 ($\underline{4}/mm\underline{m}$), α-Fe_2O_3 and $CoCO_3$ ($\bar{3}m$).[2] An extensive discussion of the experimental work on the above and other optical effects occurring in antiferromagnetic crystals subjected to external magnetic fields is due to Eremenko and Kharchenko.[39]

It is our aim here to determine the polarisation state of light on traversal of a, transparent antiferromagnetic crystal of thickness z (in the direction parallel to its highest-fold symmetry axis) acted on by a static magnetic field $\vec{B}$, at Faraday configuration ($\vec{B}$ parallel to the light propagation direction $\vec{k}$) and at Voigt configuration ($\vec{B}$ perpendicular to $\vec{k}$), for the case when the light incident on the antiferromagnet is polarized linearly. We shall show that in either case the initially linearly polarized light wave changes its state of polarisation from linear to elliptical, with ellipticity Φ proportional to the first power of $\vec{B}$. This effect is governed by a third-rank axial tensor ${}^{(1)}_{e}\alpha^{(1)m}_{e\,(ij)k}(\omega)$ antisymmetric with respect to time inversion and symmetric in its first two indices. The effect, with regard to the permutational symmetry of this tensor and the linear dependence of Φ on $\vec{B}$, represents the analog of the respectively longitudinal and transversal Pockels effect. Accordingly, we refer to it as the magnetic analog of the (longitudinal or transversal) Pockels effect (MALPE or MATPE). It is shown that MALPE and MATPE will not be perturbed by the natural spatial-dispersional effects[5,11] and the dispersional magneto-optical effects[17,19,27-32] as well as the Faraday and/or Voigt effects only in antiferromagnetic crystals with the magnetic symmetry classes $\bar{3}m$ ($CoCO_3$, α-Fe_2O_3) when $\vec{B}=\hat{e}_x B$, $\underline{6}/m\underline{m}m$ and $\underline{6}m\underline{m}$ when $\vec{B}=\hat{e}_x B$, and $\underline{4}/mm\underline{m}$ (CoF_2) and $m3\underline{m}$ ($Dy_3Al_5O_{12}$). when $\vec{B}=\hat{e}_z B$.

2. FOUNDATION OF CLASSICAL MAGNETO-OPTICS

2.1. The material equations

The electric and magnetic properties of a medium acted on by a time-variable electric field $\vec{E}(\vec{r},t)$ and magnetic field $\vec{H}(\vec{r},t)$ is described by the electric induction vector $\vec{D}(\vec{r},t)$ and, respectively, magnetic induction vector $\vec{B}(\vec{r},t)$. In SI units, they take the well known form[16]

$$\vec{D}(\vec{r},t) = \varepsilon_o\, \vec{E}(\vec{r},t) + \vec{P}_e(\vec{r},t)$$

$$\vec{B}(\vec{r},t) = \mu_o[\vec{H}(\vec{r},t) + \vec{P}_m(\vec{r},t)] \qquad (1)$$

with: ε_o and μ_o the electric and, respectively, magnetic permittivity, and $\vec{P}_e(\vec{r},t)$, $\vec{P}_m(\vec{r},t)$ the electric and magnetic polarisation vectors of the medium at the moment of time t and the point $\vec{r}$.
Assume a moderately intense light wave to be incident on the medium. The amplitude $\vec{E}(\omega,\vec{k})$ of the electric field of the wave is assumed to be moderate compared with the intra-atomic field; at the moment of time t and in the point $\vec{r}$ within the medium, $\vec{E}(\vec{r},t)$ as well as the other vectors of Eq.(1) have the form

$$\vec{E}(\vec{r},t) = \vec{E}(\omega,\vec{k})\, \exp[-i(\omega t - \vec{k}\cdot\vec{r})] + cc. \quad (2)$$

the wave vector being given by the well known formula $\vec{k}=(\omega n/c)\vec{s}$, where n is the refractive index of the medium, c the light velocity in vacuum, $\vec{s}$ the unit vector in the direction of light propagation and ω the circular frequency of the wave, whereas cc. stands for complex conjugate.

2.2. Linear electric and magnetic multipole susceptibilities

Information concerning the electric and magnetic properties of the medium in the range of optical frequencies is conveyed by the amplitudes $\vec{P}_e(\omega,\vec{k})$ and $\vec{P}_m(\omega,\vec{k})$ of the electric and, respectively, magnetic polarisations which, in the case of weak excitation[40] for antiferromagnetic crystals in a static magnetic field $\vec{B}$ can be written in the following form:[24]

$$P_{Ai}(\omega,\vec{k}) = {}_A\chi_{e\,ij}(\omega,\vec{k},\vec{B})\, E_j(\omega,\vec{k}) +$$

$$\mu_o\, {}_A\chi_{m\,ij}(\omega,\vec{k},\vec{B})\, H_j(\omega,\vec{k}) \qquad (3)$$

with A=e or A=m. Here we apply the Einstein summation convention. The polar tensors of second rank ${}_e\chi_e(\omega,\vec{k},\vec{B})$ and ${}_m\chi_m(\omega,\vec{k},\vec{B})$ describe the electro-electric and magneto-magnetic susceptibilities of the antiferromagnetic under the action of the static magnetic field of induction $\vec{B}$. The axial tensors ${}_e\chi_m(\omega,\vec{k},\vec{B})$ and ${}_m\chi_e(\omega,\vec{k},\vec{B})$ describe its linear electro-magnetic and magneto-electric susceptibilities. In the case of crystals, where spatial dispersion is not excessively great[6] and in a moderately strong magnetic field $\vec{B}$, the above four linear susceptibility tensors can be written in multipole expansion form[5] as follows:

$${}_e\chi_{e\,ij}(\omega,\vec{k},\vec{B}) = {}^{(1)}_{e}\chi^{(1)}_{e\,ij}(\omega,\vec{B}) + \frac{i\omega n}{3c}\left[{}^{(1)}_{e}\chi^{(2)}_{e\,i(jl)}(\omega,\vec{B}) - {}^{(2)}_{e}\chi^{(1)}_{e\,(il)j}(\omega,\vec{B})\right]s_l + \ldots \qquad (4)$$

$${}_e\chi_{m\,ij}(\omega,\vec{k},\vec{B}) = {}^{(1)}_{e}\chi^{(1)}_{m\,ij}(\omega,\vec{B}) + \ldots$$

$${}_m\chi_{e\,ij}(\omega,\vec{k},\vec{B}) = {}^{(1)}_{m}\chi^{(1)}_{e\,ij}(\omega,\vec{B}) + \ldots \qquad (5)$$

$${}_m\chi_{m\,ij}(\omega,\vec{k},\vec{B}) = {}^{(1)}_{m}\chi^{(1)}_{m\,ij}(\omega,\vec{B}) + \ldots \qquad (6)$$

where

$${}^{(a)}_{A}\chi^{(q)}_{Q}(\omega,\vec{B}) = {}^{(a)}_{A}\chi^{(q)}_{Q}(\omega) + {}^{(a)}_{A}\chi^{(q)m}_{Q}(\omega)\cdot\vec{B} + {}^{(a)}_{A}\chi^{(q)mm}_{Q}(\omega)\cdot\cdot\vec{B}\vec{B} + \ldots \qquad (7)$$

with[16] ${}_m\chi_{m\,ij}(\omega,\vec{k},\vec{B}) = 0$ in the optical region. Above, two dots denote the double scalar product. The tensor component indices i, j and l refer to the laboratory coordinates and take the values x,y,z. Above ${}^{(a)}_{A}\chi^{(q)}_{Q}(\omega)$ describes the linear electric-multipole (A=e) or magnetic-multipole (A=m) susceptibility of order a (for a=1,2,...we have respectively the dipole and quadrupole moment) related with electric multipole (Q=e) and magnetic multipole (Q=m) transitions of order q[5,7] (thus, for Q=e and q=1 or 2 we have a transition E1 or E2 whereas for Q=m and q=1 we have a transition M1). In formula (4) the subscripts in semicircular parentheses (....) label the components of the electric quadrupole moment; these parentheses at the same time serve to denote the invariancy (symmetricity) of the respective components with respect to transposition of the subscripts. The tensors with one or two superscripts m express the variations induced in the multipole susceptibilities by a static magnetic field; respectively in a linear (first order of stationary perturbation calculus) and quadratic approximation (second order of stationary perturbation calculus).

<u>2.2.1 Permutational symmetry for linear electric and magnetic multipole susceptibilities.</u> In a transparent (loss-less) medium, the time-averaged divergence of the Poynting vector has to vanish.[16,41] From the above theorem and the formulae (4) - (7) the multipole susceptibilities in a loss-less medium in the absence as well as in the presence of a dc field $\vec{B}$ can be shown to fulfil[11,13] the relation ${}^{(a)}_{A}\chi^{(q)}_{Q}(\omega)=[{}^{(q)}_{Q}\chi^{(a)}_{A}(\omega)]^{*}$ (their linear and quadratic magnetic variations too fulfil a similar relation) signifying hermiticity of the tensor. Moreover, in magnetic materials, the multipole susceptibilities as well as their magnetic variations are conjugate[11,13,24] so that each of them can be expressed in the form

$$ {}^{(a)}_{A}\chi^{(q)}_{Q}(\omega) = {}^{(a)}_{A}\alpha^{(q)}_{Q}(\omega) + i\,{}^{(a)}_{A}\gamma^{(q)}_{Q}(\omega). \quad (8) $$

The hermiticity and the conjugate nature of the multipole susceptibilities lead to the following transposition (permutation) relations:

$$ {}^{(a)}_{A}\alpha^{(q)}_{Q} = {}^{(q)}_{Q}\alpha^{(a)}_{A}, \qquad {}^{(a)}_{A}\gamma^{(q)}_{Q} = -{}^{(q)}_{Q}\gamma^{(a)}_{A} \quad (9) $$

and similarly for the linear and quadratic magnetic variations of the preceding multipole susceptibilities (for the sake of brevity we have omitted ω).

<u>2.2.2 Time-reversal symmetry for linear electric and magnetic multipole susceptibilities.</u> Making use of the transposition relations we can express the electric polarization vector $\vec{P}_e(\vec{r},t)$ and magnetic polarization vector $\vec{P}^e_m(\vec{r},t)$ in a form involving the electric and magnetic field strength $\vec{E}(\vec{r},t)$ and $\vec{H}(\vec{r},t)$ as well as their time-derivatives $\vec{E}(\vec{r},t)_t$ and $\vec{H}(\vec{r},t)_t$. With the respective expressions, and keeping in mind that $\vec{E}(\vec{r},t)$, $\vec{H}(\vec{r},t)_t$ and $\vec{P}_e(\vec{r},t)$ are invariant with respect to time inversion whereas $\vec{E}(\vec{r},t)_t$, $\vec{H}(\vec{r},t)$, $\vec{B}(\vec{r},t)$, $\vec{P}_m(\vec{r},t)$, $\vec{B}$ and $\vec{k}$ undergo a change in sign if $t \longrightarrow -t$, we are immediately in a position to determine how the linear multipole susceptibilities transform on time inversion. In this way we find that components α with an even number of lower and upper indices m (subscripts and superscripts jointly) and components γ with an odd number of indices m are invariant under time inversion (after Birss,[42] we shall be referring to them as i-tensors), whereas the others (α with an odd number of indices m and γ with an even number of indices m) undergo a change in sign (c-tensors) for arbitrary m and q.

<u>2.3. Neumann's principle as selection rule for magneto-optics.</u>

It follows from Neumann's principle[23,42] that i-tensors can exist both in magnetic and non-magnetic crystals whereas c-tensors can exist in magnetic crystals only. Hence, the expressions (4) - (7) determining the linear electro-electric and respectively electro-magnetic susceptibilities of non-magnetic crystals will involve only i-tensors. For this case these susceptibilities will fulfil the relations

$$ {}_{A}\chi_{Q\,ij}(\omega,\vec{k},\vec{B}) = w_A w_Q\, {}_{Q}\chi_{A\,ji}(\omega,-\vec{k},-\vec{B}) \quad (10) $$

where w_A=1 (likewise w_Q) for A=e and w_A = -1 for A=m in complete agreement with the result of Onsager's symmetry principle for kinetic coefficients[16] which is considered in the literature as the selection rule for the existence of linear magneto-optical effects in crystals. Kleiner[43] has shown that the relations (10) are valid only for non-magnetic crystals. For the magnetic crystals whose directional symmetry is described by 32 magnetic symmetry classes G(P) no Onsager principle can be enounced, whereas for the remaining 58 magnetic symmetry classes G(M) a generalized Onsager principle can be formulated. From what has been said we draw the conclusion that with regard to magnetic crystals one has to drop the Onsager principle as a selection rule for the existance of linear magneto-optical effects. Here, the Neumann principle imposes itself as more adequate selection rules: it is applicable to non-magnetic as well as magnetic crystals provided that the transformation properties of the tensors under time inversion are known. In other words, the expansions (4) - (7) in the case of magnetic crystals will involve i-tensors as well as c-tensors, the latter leading to new magneto-optical effects, forbidden by Onsager's principle with regard to non-magnetic crystals, Eqs. (10), one of which is MAPE.

3. THE REFRACTIVE INDICES

On taking into account the expressions (4)-(6) in the Maxwell equations[6] for insulators we obtain the following equation for the light refractive index n of the medium:

$$ \{n^2(\delta_{ij}s_k^2 - s_i s_j) + n\sqrt{\varepsilon_0/\mu_0}\;\chi_{iju}(\omega,\vec{B})\,s_u - $$

$$\delta_{ij} - (1/\varepsilon_o)\ {}_e\chi_{e\ ij}(\omega,\vec{B}) + \dots \}E_j(\omega,\vec{k}) = 0 \qquad (11)$$

with

$$\chi_{iju}(\omega,\vec{B}) = \frac{i\omega}{3}\Big[{}^{(1)}_{e}\chi^{(2)}_{e\ i(ju)}(\omega,\vec{B}) - {}^{(2)}_{e}\chi^{(1)}_{e\ (iu)j}(\omega,\vec{B})\Big] + {}_e\chi_{m\ ip}(\omega,\vec{B})\ \delta_{pju} + \delta_{iuw}\ {}_m\chi_{e\ wj}(\omega,\vec{B}). \qquad (12)$$

Here, δ_{ij} and δ_{ijk} denote respectively the Kronecker and Levi-Civita unit tensors.

Let the light wave propagate in the crystal along the z-axis taken as parallel to the highest of its axes of symmetry. We accordingly have

$$s_x = s_y = 0,\quad s_z = 1 \text{ and } E_z(\omega,\vec{k}) = 0. \qquad (13)$$

On equating to zero the determinant of the coefficients at $E_x(\omega,\vec{k})$ and $E_y(\omega,\vec{k})$ in Eqs (11) and with regard to (13) we arrive at an equation of the fourth degree for n. Since it involves terms in n^3 and n, it is not easily solvable. One immediately notes that these terms vanish if one puts

$$\chi_{xxz}(\omega,\vec{B}) = \chi_{yyz}(\omega,\vec{B}) = \chi_{xyz}(\omega,\vec{B}) = \chi_{yxz}(\omega,\vec{B}) = 0. \qquad (14)$$

By having recourse to Tables[42] giving the form of i - and c - tensors of the second, third, fourth and fifth rank one can determine those crystal symmetries that ensure fulfilment of the condition (14). Obviously, if the static magnetic field is applied along the x- or y- axis, the condition (14) is fulfilled for crystals with the following symmetries: $(4/\underline{m})$, $\underline{4}/m$, 4/mmm, $\underline{4}/mm\underline{m}$, $(4/m\underline{mm})$, $(\bar{3})$, $\bar{3}m$, $(\bar{3}\underline{m})$, (6/m), $\underline{6}/m$, $\underline{6}/\underline{m}$, $\underline{6}m\underline{m}$, 6/mmm, $\underline{6}/\underline{m}m\underline{m}$, $(6/m\underline{mm})$, $\underline{6}/m\underline{m}m$, m3, m3m and $m3\underline{m}$, whereas if the field is applied strictly along the z-axis it is fulfilled moreover for the classes: $(4\underline{mm})$, $4/\underline{mmm}$, $(3\underline{m})$, $\underline{\bar{3}m}$, $(\bar{6}, 6\underline{mm})$, $\bar{6}m2$, $\underline{\bar{6}}2\underline{m}$, $(\bar{6}\underline{m2})$, $6/\underline{mmm}$ and $\underline{m}3\underline{m}$. Thus, the condition (14) restricts our solution to uniaxial crystals ($n_x = n_y = n_o$) and isotropic ones ($n_x = n_y = n_z = n_o$) with the above stated symmetries. The magnetic symmetry classes in parentheses do not admit of antiferromagnetic ordering. Formula (11) with the condition (14) taken into account gives the following solution for n:

$$n_\pm = n_o + n(B) \pm \delta(B) \qquad (15)$$

where we have introduced the notations

$$n_o^2 = 1 + (1/\varepsilon_o)\ {}^{(1)}_e\alpha^{(1)}_{e\ xx}(\omega),$$

$${}^{(1)}_e\alpha^{(1)}_{e\ xx}(\omega) = {}^{(1)}_e\alpha^{(1)}_{e\ yy}(\omega) \qquad (16)$$

$$n(B) = (1/4\varepsilon_o\mu_o)\Big\{{}^{(1)}_e\alpha^{(1)m}_{e\ xxu}(\omega) + {}^{(1)}_e\alpha^{(1)m}_{e\ yyu}(\omega) + \Big[{}^{(1)}_e\alpha^{(1)mm}_{e\ xx(uw)}(\omega) + {}^{(1)}_e\alpha^{(1)mm}_{e\ yy(uw)}(\omega)\Big] B_w + \dots\Big\}B_u \qquad (17)$$

$$\delta(B) = (1/4n_o)\sqrt{h(B)^2 + 4[g(B)^2 + f(B)^2]} \qquad (18)$$

$$h(B) = (1/\varepsilon_o)\Big\{{}^{(1)}_e\alpha^{(1)m}_{e\ yyu}(\omega) - {}^{(1)}_e\alpha^{(1)m}_{e\ xxu}(\omega) + \Big[{}^{(1)}_e\alpha^{(1)mm}_{e\ yy(uw)}(\omega) - {}^{(1)}_e\alpha^{(1)mm}_{e\ xx(uw)}(\omega)\Big]B_w + .\Big\}B_u \qquad (19)$$

$$g(B) = (1/\varepsilon_o)\Big[{}^{(1)}_e\alpha^{(1)}_{e\ (xy)}(\omega) + {}^{(1)}_e\alpha^{(1)m}_{e\ (xy)u}(\omega)B_u + {}^{(1)}_e\alpha^{(1)mm}_{e\ (xy)(uw)}(\omega)B_uB_w + \dots\Big] \qquad (20)$$

$$f(B) = (1/\varepsilon_o)\Big[{}^{(1)}_e\gamma^{(1)}_{e\ [xy]}(\omega) + {}^{(1)}_e\gamma^{(1)m}_{e\ [xy]u}(\omega)B_u + {}^{(1)}_e\gamma^{(1)mm}_{e\ [xy](uw)}(\omega)B_uB_w + \dots\Big]. \qquad (21)$$

On insertion of n_+ and n_- respectively into the first and second equation of the set (11) and with regard to (14) we get the following relation:

$$E_x(\omega,k_+) = a_+E_y(\omega,k_+),\qquad a_\pm = \frac{2[g(B) \pm if(B)]}{h(B) + 4n_o\delta(B)}$$
$$a_-E_x(\omega,k_-) = -E_y(\omega,k_-), \qquad (22)$$

where obviously $a_- = a_+^*$. Hence we see that in the direction z two light waves propagate in the medium. We have assumed the electric field of either of them at a point $\vec{r} = \hat{e}_z z$ and a moment of time t within the medium in the form of a plane wave (2). The one propagates with the velocity $V_+ = c/n_+$ and the amplitude $\vec{E}(\omega,k_+) = E_x(\omega,k_+)[\hat{e}_x + (1/a_+)\hat{e}_y]$, whereas for the other we have, respectively, $V_- = c/n_-$ and $\vec{E}(\omega,k_-) = E_x(\omega,k_-)[\hat{e}_x - a_-\hat{e}_y]$, where $\hat{e}_x$ and $\hat{e}_y$ are unit vectors along x - and y. The superposition of the electric field vectors of the two waves in an arbitrary point z and at an arbitrary moment t in the medium can be expressed in the form $\vec{E}(z,t) = \hat{e}_xE_x(z,t) + \hat{e}_yE_y(z,t) + $ cc., where

$$\begin{pmatrix} E_x(z,t) \\ E_y(z,t) \end{pmatrix} = \begin{pmatrix} P_- & Q_+ \\ Q_- & P_+ \end{pmatrix} \begin{pmatrix} E_x(0,t) \\ E_y(0,t) \end{pmatrix} \exp\Big\{\frac{i\omega[n_o + n(B)]z}{c}\Big\} \qquad (23)$$

$$P_\mp = \cos\Big(\frac{\omega z\delta(B)}{c}\Big) \mp i\,\frac{1 - a_+a_-}{1 + a_+a_-}\sin\Big(\frac{\omega z\delta(B)}{c}\Big),$$

$$Q_{\pm} = \frac{i2a_{\pm}}{1 + a_{+}a_{-}} \sin\left(\frac{\omega z \delta(B)}{c}\right) \qquad (24)$$

whereas $E_x(0,t)$ and $E_y(0,t)$ are the components of the electric field vector oscillating along the x- and y- axis in the point z=0, i.e., at the input to the crystal: $E_j(o,t)=E_j(\omega)\exp(-i\omega t)$ for j= x and y.

4. APPLICATION AND DISCUSION

For a totally polarized light wave, the azimuth Ψ (the angle between the major axis of the ellipse and the x-axis) and the ellipticity Φ (the ratio of the minor and major axes of the ellipse) are equal to:[4]

$$\Psi = \tfrac{1}{2}\arctan(S_2/S_1), \quad \Phi = \tfrac{1}{2}\arcsin(S_3/S_o) \qquad (25)$$

where S_0, S_1, S_2 and S_3 are the well known Stokes parameters.

We shall consider two cases, the one for f(B) = 0 and the other for h(B) = 0.

4.1. The case f(B) = 0

Using the definition of Stokes' parameters[4] and putting $E_y(0,t) = 0$ in (23) as well as writing f(B) = 0 in $a_{\pm}$ and expanding the function $\sin[\omega z\delta(B)/c]$ of Eq (24) we get

$$\Psi = \tfrac{1}{2}\arctan(A_3/A_1), \quad \Phi = \tfrac{1}{2}\arcsin(A_5) \qquad (26)$$

where

$$A_5 = \frac{\omega z g(B)}{n_o c}\left\{ 1 + \sum_{k=1}^{\infty} \frac{(-1)^k [2U(h,g)]^{2k}}{(2k+1)!} \right\} \qquad (27)$$

$$\frac{A_3}{A_1} = \frac{g(B)h(B)\left(\frac{\omega z}{2n_o c}\right)^2 \left\{1+\sum_{k=1}^{\infty} \frac{(-1)^k U(h,g)^{2k+1}}{(2k+1)!}\right\}^2}{1 - 2\left(\frac{\omega z g(B)}{2n_o c}\right)^2 \left\{1+\sum_{k=1}^{\infty} \frac{(-1)^k U(h,g)^{2k}}{(2k+1)!}\right\}^2} \qquad (28)$$

with

$$U(h,g) = (\omega z/4n_o c)\sqrt{h(B)^2 + 4g(B)^2} \qquad (29)$$

If f(B) = 0, then $a_+ = a_- = a$. In accordance with (22) this signifies that two waves propagate along the z-axis; they are linearly polarized in the directions $\hat{e}_+ = \hat{e}_x + (1/a)\hat{e}_y$ and, respectively, $\hat{e}_- = \hat{e}_x - a\hat{e}_y$ so that with regard to $\hat{e}_+ \cdot \hat{e}_- = 0$ their polarisation directions are mutually perpendicular. The formulae (26) and (27) show that if $g(B) \neq 0$ the superposition of the two waves in a point **z** (on traversal of the path **z** in the medium) is an elliptically polarized wave. Here, we deal with linear birefringence (of linearly polarized waves). In particular, if a = 1 (this takes place if h(B) = 0) the parameters of the ellipse are equal to

$$\Phi = \omega z g(B)/(2n_o c), \qquad \Psi = 0 \qquad (30)$$

its major axis lying along the x-axis. Obviously, the light wave will propagate in the medium with its state of polarisation unchanged if a = 0; this occurs if g(B) = 0.

Let us consider the case when the incident wave is polarized linearly at an angle of $\Pi/4$ to the x-axis in the xy-plane. At the input, we now have $E_x(0,t) = E_y(0,t)$. If moreover the parameter **a** vanishes (this occurs if f(B) = 0 and g(B) = 0), two linearly polarized waves will again be propagating in the medium, in the directions $\hat{e}_x$ and $\hat{e}_y$ respectively, their superposition giving an elliptically polarized wave:

$$\Phi = \omega z h(B)/(4n_o c), \qquad \Psi = \Pi/4 \qquad (31)$$

the major axis of the ellipse coinciding with the polarisation direction of the wave at the input to the crystal.

4.2. The case h(B) = 0

Putting h(B) = 0 we have $|a_+|^2 = 1$. If $E_x(o,t)$ vanishes, Ψ and Φ are still given by the formulae (26), though now

$$A_5 = \frac{g(B)}{\sqrt{g(B)^2 + f(B)^2}} \sin\left(\frac{\omega z}{n_o c}\sqrt{g(B)^2 + f(B)^2}\right) \qquad (32)$$

$$\frac{A_3}{A_1} = \frac{f(B)}{\sqrt{g(B)^2 + f(B)^2}} \tan\left(\frac{\omega z}{n_o c}\sqrt{g(B)^2 + f(B)^2}\right). \qquad (33)$$

Here, two elliptically polarized waves propagate in the medium; their superposition in a point **z** is also an elliptically polarized wave. Thus, we are dealing here with optical birefringence of elliptically polarized waves. In particular, if g(B) = 0, two circularly polarized waves with opposite senses will be propagating in the medium, their superposition giving a linearly polarized wave: $\Phi = 0$ with its polarisation plane at an angle $\Psi = \omega z f(B)/(2n_o c)$ to the xz-plane. Here, we deal with circular birefringence (of circularly polarized waves). The first term of $(21)_1$ determining the parameter f(B), i.e., $e^{(1)}_{\gamma} e^{(1)}_{xy}(\omega)$,[12-14] describes natural gyrotropic rotation; the second term at u = z (magnetic field acting along the z-axis) describes the well known Faraday effect; and the third term at arbitrary orientation of the field $\vec{B}$ describes quadratic magnetic variation in natural gyrotropic rotation (quadratic magnetic rotation of the light polarization plane).[17,38]

5. MAPE

The formulae derived by us for Ψ and Φ are valid for antiferromagnetic crystals which fulfil the condition (14) and, additionally, if f(B) = 0 or h(B) = 0. In other words, the above conditions define a set of antiferromagnetic crystals where the experimenter is invited to measure MAPE (linear optical birefringence proportional to the first power of the field $\vec{B}$, i.e., ellipticity proportional to the first power of $\vec{B}$). By making use of Tables[42] giving the axial and polar i- and c- tensors of the second, third and fourth ranks for the 90 magnetic symmetry classes, we have found the parameters h(B), g(B) and f(B) for all the antiferromagnetics fulfilling the condition (14) for $\vec{B}$ applied along x, or y, or z. The results thus obtained are assembled in Table 1 where, moreover, we give our expressions for the ellipticity Φ and the rotation angle Ψ of the major axis of the ellipse with respect to the x-axis (where the fourth column specifies the configuration of the electric field of the light wave at the input to the crystal). When determining Ψ and Φ we have restricted ourselves to the first term of the series expansion of $\arctan(A_3/A_1)$ and $\arcsin(A_5)$ -an apparently satisfactory approximation[5] since the terms omitted in the two expansions are proportional to the third power of the argument of the respective function, thus giving a contribution to Ψ and Φ whose dependence on $\vec{B}$ has an exponent higher than that assumed in the expressions (4) - (7). For the same reasons, in the expressions defining A_3/A_1 and A_5 ,we have neglected the terms containing the parameters U(h,g) and U(g,f). Obviously, these considerations are valid for weak magnetic fields, as long as $\arctan(A_3/A_1)$ and $\arcsin A_5$ can be expanded in series, i.e., if $|A_3/A_1| < 1$ and $|A_5| < 1$. Table 1 shows that MAPE is described by the constants g_1, g_2 and h_1, and that the ellipticity Φ will be strictly linear in $\vec{B}$ only in crystals having the symmetries $\bar{3}\underline{m}$ ($\vec{B}=\hat{e}_x B$); $\underline{6}/\underline{m}mm$ and $\underline{6}mm$ ($\vec{B}=\hat{e}_y B$); as well as $\underline{4}/mm\underline{m}$ and $m3\underline{m}$ ($\vec{B}=\hat{e}_z B$); with the major axis of the ellipse subtending angles of Ψ_1, Ψ_5 and $\omega z f_1 B/(2n_0 c)$ with the x-axis. In the first two cases we have MATPE —— the magnetic analog of the transversal Pockels effect, whereas in the last case we deal with MALPE — the magnetic analog of the longitudinal Pockels effect. Thus, if a linearly polarized light wave traverses a crystal having the symmetry $\underline{4}/mm\underline{m}$ or $m3\underline{m}$ along the highest-fold symmetry axis with $\vec{B}$ applied parallel to the light propagation direction, the wave becomes endowed with elliptical polarisation, the major axis of the ellipse subtending the angle Ψ with the x-axis. In both these magnetic symmetry classes MALPE is accompanied by Faraday's effect. Luckily for the experimenter, the two effects do not obscure each other since MALPE is apparent in Φ, whereas Faraday's effect reveals itself in Ψ, so they are accessible to measurement separately.

It is to be regretted that the angles Ψ_1 and Ψ_5 as well as the others are highly complicated functions; already in a first approximation they are dependent on the third power of the magnetic field strength and thus exceed the approximation assumed in Eq. (7), so that (in the case of fields for which $|\omega z \delta(B)/c| < 1$) one can write $\Psi_1 \cong 0$.

For crystals possessing the symmetries $\underline{6}/\underline{m}$, $\underline{6}/\underline{m}mm$ and $\underline{6}mm$ ($\vec{B}=\hat{e}_x B$) as well $\bar{3}m$ and $\underline{6}/\underline{m}$ ($\vec{B}=\hat{e}_y B$), the ellipticity contains (in addition to the linear term) a quadratic term typical of the Voigt effect and the Cotton-Mouton effect. The linear term is easily separable from the quadratic term since it changes its sign on reversal of the magnetic field.

The birefringence proportional to the first power of the static magnetic field which is at the core of our interest will occur as well as in non-centrosymmetric magnetic crystals, possessing the symmetries 3, $\bar{3}$, 32, $3\underline{2}$, $3\underline{m}$, $\bar{3}\underline{m}$, $\underline{6}$, $\bar{\underline{6}}$, $\underline{6}2\underline{2}$, $\bar{\underline{6}}2\underline{m}$ and $\bar{\underline{6}}m\underline{2}$. In these magnetic symmetry classes, however, the condition (14) ceases to hold so that the birefringence in question will be accompanied by other optical effects such as natural optical activity,[5-8] natural gyrotropic rotation[12-14] and birefringence,[7-11] and their variations are proportional to the first and second powers of the magnetic field strength[5,8,27-30] rendering difficult the observation of birefringence proportional to the first power of the static magnetic field.

5.1. The physics of MAPE

A static magnetic field $\vec{B}$ acting on a magnetic crystal modifies the well known magnetic vectors $\mathcal{L}^{(\lambda)}$ which characterize the magnetic ordering (the vector $\mathcal{L}^{(\lambda)}$ represents the resultant magnetisation of the λ-th sublattice of the crystal) to the following form:

$$\mathcal{L}_i^{(\lambda)}(\vec{B}) = \mathcal{L}_i^{(\lambda)} + \chi_{ij}^{\mathcal{L}^{(\lambda)}}(0)\, B_j + \ldots \qquad (34)$$

where $\chi^{\mathcal{L}^{(\lambda)}}(0)$ is an polar tensor of the second rank symmetric with respect to time inversion and accounting for the change in magnetisation of the sublattice λ under the action of $\vec{B}$. For a two-sublattice antiferromagnet the vectors $\mathcal{L}^{(\lambda)}$ are the well known ferromagnetism vector $\vec{M}$ and the antiferromagnetism vector $\vec{L}$. The multipolar susceptibility ${}^{(a)}_{A}\alpha^{(q)}_{Q}(\omega,\vec{B})$ for a two-sublattice antiferromagnet acted on by a static magnetic field $\vec{B}$ can be expanded in a series in the vectors $\vec{M}(\vec{B})$ and $\vec{L}(\vec{B})$. This leads to expansions similar to the expressions obtained by Pisarev.[31] On comparing his expansion and our expansion in $\vec{B}$, we can write the linear multipolar susceptibility ${}^{(a)}_{A}\alpha^{(q)m}_{Q}(\omega)$ as follows:

$${}^{(a)}_{A}\alpha^{(q)m}_{Q\,iju}(\omega) = {}^{(a)}_{A}\alpha^{(q)ML}_{Q\,ijpr}(\omega)\chi^{M}_{pu}(0)L_r +$$

$${}^{(a)}_{A}\alpha^{(q)MM}_{Q\,ijpr}(\omega)\chi^{M}_{pu}(0)M_r + {}^{(a)}_{A}\alpha^{(q)LL}_{Q\,ijpr}(w)\chi^{L}_{pu}(0)L_r \qquad (35)$$

Table 1. Explicit form of the parameters h(B), g(B), f(B), Ψ and Φ for magnetic symmetry classes ensuring fulfilment of the condition (14) and three selected directions of $\vec{B}$.

$\vec{B} = \hat{e}_x B$

Magnetic symmetry classes	h(B)	g(B)	$\vec{E}(0,t)$	Ψ	Φ
$4/mmm$, $\underline{4}/mm\underline{m}$, $6/mmm$, $\underline{6}/m\underline{m}m$, $m3$, $m3m$, $\underline{m}3\underline{m}$, $m3\underline{m}$	H_1B^2	0	$(\hat{e}_x+\hat{e}_y)E(0,t)$	Π/4	$(\omega z/4n_oc)H_1B^2$
$\bar{3}m$	H_1B^2	g_1B	$\hat{e}_xE(o,t)$	Ψ_1	$(\omega z/2n_oc)g_1B$
$\underline{6}/m$	H_1B^2	G_1B^2	$\hat{e}_xE(0,t)$	Ψ_3	$(\omega z/2n_oc)G_1B^2$
$\underline{6}/\underline{m}$	$2h_1B+H_1B^2$	$g_1B+G_1B^2$	$\hat{e}_xE(o,t)$	Ψ_2	$(\omega z/2n_oc)(g_1B+G_1B^2)$
$\underline{6}/\underline{m}m\underline{m}$, $\underline{6}m\underline{m}$	$2h_1B+H_1B^2$	0	$(\hat{e}_x+\hat{e}_y)E(o,t)$	Π/4	$(\omega z/4n_oc)(2h_1B+H_1B^2)$

$\vec{B} = \hat{e}_y B$

Magnetic symmetry classes	h(B)	g(B)	$\vec{E}(0,t)$	Ψ	Φ
$4/mmm$, $\underline{4}/mm\underline{m}$, $6/mmm$, $\underline{6}/m\underline{m}m$, $m3m$, $m3\underline{m}$, $\underline{m}3\underline{m}$	$-H_1B^2$	0	$(\hat{e}_x+\hat{e}_y)E(0,t)$	Π/4	$-(\omega z/4n_oc)H_1B^2$
$m3$	H_2B^2	0	$(\hat{e}_x+\hat{e}_y)E(0,t)$	Π/4	$(\omega z/4n_oc)H_2B^2$
$\bar{3}m$	$-2g_1B-H_1B^2$	0	$(\hat{e}_x+\hat{e}_y)E(0,t)$	Π/4	$-(\omega z/4n_oc)(2g_1B+H_1B^2)$
$\underline{6}/m$	$-H_1B^2$	$-G_1B^2$	$\hat{e}_xE(0,t)$	Ψ_6	$-(\omega z/2n_oc)G_1B^2$
$\underline{6}/\underline{m}$	$-2g_1B-H_1B^2$	$h_1B-G_1B^2$	$\hat{e}_xE(0,t)$	Ψ_4	$(\omega z/2n_oc)(h_1B-G_1B^2)$
$\underline{6}/\underline{m}m\underline{m}$, $\underline{6}m\underline{m}$	$-H_1B^2$	h_1B	$\hat{e}_xE(0,t)$	Ψ_5	$(\omega z/2n_oc)h_1B$

$\vec{B} = \hat{e}_z B$

Magnetic symmetry classes	g(B)	f(B)	$\vec{E}(0,t)$	Ψ	Φ
$4/mmm$, $4/\underline{mmm}$, $6/mmm$, $\underline{6}/\underline{m}m\underline{m}$, $\underline{6}/m\underline{m}m$, $6/\underline{mmm}$, $\bar{\underline{3}}\underline{m}$, $\bar{3}m$, $\underline{6}/m$, $\underline{6}/\underline{m}$, $\underline{6}m\underline{m}$, $\bar{6}m2$, $\bar{\underline{6}}2\underline{m}$, $m3$, $m3m$, $\underline{m}3\underline{m}$	0	f_1B	$\hat{e}_xE(0,t)$	$(\omega z/2n_oc)f_1B$	0
$\underline{4}/mm\underline{m}$, $m3\underline{m}$	g_2B	f_1B	$\hat{e}_xE(0,t)$	$(\omega z/2n_oc)f_1B$	$(\omega z/2n_oc)g_2B$

where:

$$H_1 = \frac{1}{\varepsilon_o}\left[{}^{(1)}_{e}\alpha^{(1)mm}_{e\,yyxx}(\omega) - {}^{(1)}_{e}\alpha^{(1)mm}_{e\,xxxx}(\omega)\right],\qquad G_1 = \frac{1}{\varepsilon_o}\,{}^{(1)}_{e}\alpha^{(1)mm}_{e\,(xy)xx}(\omega),$$

$$H_2 = \frac{1}{\varepsilon_o}\left[{}^{(1)}_{e}\alpha^{(1)mm}_{e\,xxxx}(\omega) - {}^{(1)}_{e}\alpha^{(1)mm}_{e\,xxyy}(\omega)\right],\qquad g_1 = \frac{1}{\varepsilon_o}\,{}^{(1)}_{e}\alpha^{(1)m}_{e\,(xy)x}(\omega),$$

$$f_1 = \frac{1}{\varepsilon_o}\,{}^{(1)}_{e}\gamma^{(1)m}_{e\,(xy)z}(\omega),\qquad g_2 = \frac{1}{\varepsilon_o}\,{}^{(1)}_{e}\alpha^{(1)m}_{e\,(xy)z}(\omega),$$

$$h_1 = \frac{1}{\varepsilon_o}\,{}^{(1)}_{e}\alpha^{(1)m}_{e\,yyx}(\omega),$$

Table 1 (Contd.)

$$\Psi_2 = -\frac{(1/2)(\omega zB/2n_o c)^2 (g_1 + G_1 B)(2h_1 + H_1 B)}{1 - 2[\omega zB(g_1 + G_1 B)/(2n_o c)]^2} \qquad \Psi_4 = \frac{(1/2)(\omega zB/2n_o c)^2 (2g_1 + H_1 B)(h_1 - G_1 B)}{1 - 2[\omega zB(h_1 - G_1 B)/(2n_o c)]^2},$$

$$\Psi_1 = \Psi_2(h_1=0,\ G_1=0), \quad \Psi_3 = \Psi_2(g_1=0,\ h_1=0) \qquad \Psi_5 = \Psi_4(g_1=0,\ G_1=0), \quad \Psi_6 = \Psi_4(g_1=0,\ h_1=0)$$

where ${}^{(a)}_{A}\alpha^{(q)m}_{Q}(\omega)$ as well as the two others are tensors of the rank a+q+2, axial if A=e and Q=m (or inversely) but polar if A=e and Q=e (or if A=Q=m); their permutational symmetry and transformation properties with respect to time inversion are the same as those of ${}^{(a)}_{A}\alpha^{(q)}_{Q}(\omega)$. In particular, putting A=Q=e and a=q=1 in (35) we get the expression obtained by Eremenko and Kharchenko.[39]

5.2. Applications of MAPE in science and industry

The MAPE is sensitive to the magnetic crystal symmetry and to reorientation of the antiferromagnetic vector. Moreover, it undergoes a change in sign when the direction of the magnetic moments of a sublattice are inverted. Owing to this, MAPE can be used to study the time-reversed domain structure of antiferromagnets, to determine the symmetry of magnetic ordering, and to study the magnetic crystal energy spectra by spectroscopic methods.[39]

Since the experimentally determined susceptibility ${}^{(1)}_{e}\alpha^{(1)}_{e}(\omega)$ of antiferromagnets is relatively great [of order (10^{-7}—10^{-8})Oe^{-1}],[39] a magnetic field of several kOe will suffice to endow linearly polarized light with ellipticity amounting to several tenths of a radian on traversal of a crystal some tenths of a cm thick, thus making MAPE an effective tool for the modulation of laser light.

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