

CRYSTAL OPTICS TAKING INTO ACCOUNT THE MAGNETOELECTRIC EFFECT

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Abstract

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CRYSTALLOGRAPHY

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CRYSTAL OPTICS TAKING INTO ACCOUNT THE MAGNETOELECTRIC EFFECT

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The theoretically predicted ⁽¹⁾ magnetoelectric effect (m.e.) was experimentally discovered in the radio-frequency range ^(2,3). The m.e. consists in the fact that, for substances of a definite magnetic symmetry, when an electric field is applied a magnetization proportional to the field appears, and when a magnetic field is applied an electric polarization proportional to this field appears. It may be supposed that the m.e. should also occur in the optical frequency range. Below we consider the generalization to which taking the m.e. into account leads in crystal optics.

Taking the m.e. into account, the media in which electromagnetic waves propagate must, in the most general case, be characterized by the following dependences of the inductions **D** and **B** on the fields **E** and **H**:

$$\mathbf{D} = \varepsilon \mathbf{E} + \nu \mathbf{H}, \quad \mathbf{B} = \mu \mathbf{H} + \nu' \mathbf{E}. \quad (1)$$

The requirement that the law of conservation of energy be satisfied imposes restrictions on the properties of the tensors of dielectric (ε) and magnetic (μ) permeabilities, as well as on the properties of the magnetoelectric tensors ν and ν' introduced here. In this case, from the condition

$$\frac{\partial w}{\partial t} = \frac{1}{4\pi} (\mathbf{E} \dot{\mathbf{D}} + \mathbf{H} \dot{\mathbf{B}}),$$

where w is the field-energy density, it follows that $\nu' = \tilde{\nu}$ (along with the usual requirement $\varepsilon = \tilde{\varepsilon}$ and $\mu = \tilde{\mu}$). Here the sign $\sim$ denotes transposition. This conclusion is analogous to the result ⁽⁴⁾ obtained from other considerations for static processes. The requirement that there be no absorption of energy leads to new restrictions. If the time dependence of the fields and inductions is determined by the expression $e^{i\omega t}$, then the condition $\dot{\bar{w}} = 0$ (the bar denotes averaging over time) leads to the requirement $\nu^* = \nu$ (together with the usual requirement $\varepsilon^* = \varepsilon$ and $\mu^* = \mu$). The asterisk here denotes complex conjugation.

Fig. 1. Optical indicatrix of uniaxial crystals in the case $\mu = 1$ and $\nu = pe^\times$

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The specific form of the tensors ν for various classes of magnetic symmetry can be taken from ⁽⁵⁻⁷⁾. In these works ν are written in a crystallophysical basis. For our purposes, however, it is more convenient to use an invariant treatment, without resorting to any chosen coordinate system. Like the tensors of optical activity in the generalized sense ^(8,9), the tensors ν are nonsymmetric with respect to permutations of their indices. An invariant notation for the activity tensors for groups of ordinary symmetry is available in ^(8,9). Using below the results of ⁽¹⁰⁾, we note that the tensors of optical activity are tensors of electric type. According to ⁽¹⁰⁾, the form of the tensors ν (tensors of magnetoelectric type) is exactly the same as that of the tensors of optical activity. Knowing the form of the latter in classical symmetry, one can indicate the magnetic classes for which the tensors ν will have exactly the same form. Of all 143 crystallographic and limiting groups of magnetic symmetry, the m.e. is forbidden by all 64 classes containing a center of symmetry $\bar{1}$ or the anti-identity operation $1'$

(or both), and, in addition, by 11 more classes ^[11]. Hence it is clear that a necessary condition for the m.e. is the presence of magnetic ordering (ferro- or antiferromagnetic).

Like any nonsymmetric tensor of the second rank, ν may be represented as the sum of a symmetric part γ and an antisymmetric part $\mathbf{p}^\times$ (we regard the antisymmetric part as an antisymmetric tensor, the dual vector $\mathbf{p}$, the cross here denoting duality). In this case Maxwell's equations for plane waves may be written in the form

$$\varepsilon \mathbf{E} + \gamma \mathbf{H} = -[\mathbf{m}' \mathbf{H}], \quad \mu \mathbf{H} + \gamma \mathbf{E} = [\mathbf{m}' \mathbf{E}], \quad (2)$$

where the wave refraction vector $\mathbf{m}$ is related to $\mathbf{m}'$ and $\mathbf{p}$ by the relation $\mathbf{m}' = \mathbf{m} + \mathbf{p}$. A similar approach was also used in ^[12]. Let us note that in (2) the equations transform into one another under the replacement $\mathbf{E} \rightleftharpoons \mathbf{H}$, $\varepsilon \rightleftharpoons \mu$, $\gamma \rightleftharpoons \gamma$ and $\mathbf{m}' \rightleftharpoons -\mathbf{m}'$. From the last two relations it also follows that, under such a replacement, $\mathbf{p} \rightleftharpoons -\mathbf{p}$, $\mathbf{p}^\times \rightleftharpoons -\mathbf{p}^\times = \tilde{\mathbf{p}}^\times$ and $\nu \rightleftharpoons \tilde{\nu}$.

Fig. 1. Optical indicatrix of uniaxial crystals in the case $\mu = 1$ and $\nu = pe^\times$

Without going here into the details of light propagation in uniaxial crystals, we note only one interesting case. In the following 14 symmetry classes:

$$\infty/m'mm, \quad \infty mm, \quad \infty 2'2', \quad 6/m'mm, \quad 6mm, \quad 62'2', \quad \bar{6}'m2',$$

$$4/m'mm, \quad 4mm, \quad 42'2', \quad \bar{4}'m2', \quad \bar{3}'m, \quad 3m, \quad 32'$$

ν has a purely antisymmetric form: $\nu = pe^\times$ (e is the unit vector of the symmetry axis of highest order). In this case (2) take the form usual for crystals of the middle crystal systems:

$$\varepsilon \mathbf{E} = [\mathbf{m}' \mathbf{H}], \quad \mu \mathbf{H} = [\mathbf{m}' \mathbf{E}], \quad (3)$$

where ε and μ are uniaxial tensors. Taking into account the tensor character of μ , a similar problem was investigated in [13]. For $\mu = 1$ we obtain the case of uniaxial crystals well known in ordinary crystal optics. The optical indicatrix of such crystals will have the usual form (see Fig. 1), but only $\mathbf{m}$ should be laid off not from the center, but from a point displaced from the center by pe .¹ It is then clear that, in the cases under consideration, there will no longer be an ordinary wave in the usual sense, since the phase velocity of both waves will depend on the direction of $\mathbf{m}$. The indicatrix under study lacks a center of symmetry and, in contrast to ordinary crystal optics, the directions "forward" and "backward" become nonequivalent (with the exception of propagation of waves perpendicular to the optical axis). The possibility of such nonreciprocity was pointed out in [12, 14, 15], where, in particular, it is noted that, with allowance for the m.e., the equation of the normals is not invariant with respect to the replacement of $\mathbf{m}$ by $-\mathbf{m}$. If one takes into account that $\mathbf{m}$ is a vector of the magnetoelectric type (it changes sign under inversion and anti-identity and does not change sign under anti-inversion), then it is not difficult to verify that the symmetry of the optical indicatrix (the geometrical locus of the ends of the vector $\mathbf{m}$) coincides with the symmetry of the magnetoelectric vector $\propto/m'mm$.

If in (2) $\mathbf{H}$ is eliminated, then after calculation one can obtain the following equation for $\mathbf{E}$:

$$\{1 + \varepsilon^{-1}(\mathbf{m}' \times + \gamma)\mu^{-1}(\mathbf{m}' \times - \gamma)\}\mathbf{E} = 0. \quad (4)$$

Using the matrices $\rho = \varepsilon^{-1}\mathbf{m}'^\times$, $\sigma = \mu^{-1}\mathbf{m}'^\times$, $\varkappa = \varepsilon^{-1}\gamma$, and $\chi = \mu^{-1}\gamma$, (4) can be represented in the form:

$$\{1 + (\rho + \varkappa)(\sigma - \chi)\}\mathbf{E} = 0. \quad (5)$$

Making the substitution $\mathbf{E} \rightleftharpoons \mathbf{H}$, etc., under which $\rho \rightleftharpoons -\sigma$ and $\varkappa \rightleftharpoons \chi$, instead of (4) and (5) we obtain the following equations for $\mathbf{H}$:

¹Approximately (if only terms linear in the components of ν are retained), exactly the same form will be possessed by the indicatrix also of the following 10 symmetry classes: ∞/m' , ∞ , $6/m'$, 6 , $6'$, $4/m'$, 4 , $4'$, $3'$ and 3 .

$$\{1 + \mu^{-1}(\mathbf{m}'^{\times} - \gamma)\varepsilon^{-1}(\mathbf{m}'^{\times} + \gamma)\}\mathbf{H} = 0, \quad (6)$$

$$\{1 + (\sigma - \chi)(\rho + \varkappa)\}\mathbf{H} = 0. \quad (7)$$

Equating to zero any of the determinants of the systems (4)–(7) (these determinants are equal), we obtain the condition for the existence of nonzero solutions for $\mathbf{E}$ and $\mathbf{H}$, and consequently (see (1)) for $\mathbf{D}$ and $\mathbf{B}$ —the equation of the normals of crystal optics taking into account the m.e. Omitting the calculations, we give the result of expanding the determinant in invariant form:

$$\begin{aligned} & \mathbf{m}'\varepsilon\mathbf{m}' \cdot \mathbf{m}'\mu\mathbf{m}' - (\mathbf{m}'\gamma\mathbf{m}' + |\gamma|)^2 + \mathbf{m}' \left\{ \mu(\bar{\varepsilon}\mu - (\varepsilon\mu)_c) + 2\varepsilon(\gamma\mathbf{m}')^{\times}\mu \right. \\ & \left. - \frac{1}{|\mu|}\gamma\bar{\mu}(\varepsilon\bar{\mu} - (\varepsilon\mu)_c)\gamma + 2\varepsilon\gamma\mu \right\} \mathbf{m}' + 2(\bar{\varepsilon}\gamma\bar{\mu}\mathbf{m}'^{\times})_c \\ & + 2((\gamma\mathbf{m}')^{\times}\mu\gamma\varepsilon)_c - (\bar{\varepsilon}\gamma\bar{\mu}\gamma)_c + (\varepsilon\bar{\gamma}\mu\bar{\gamma})_c + |\varepsilon\mu| = 0. \end{aligned} \quad (8)$$

Here the index c denotes the trace of a matrix, and the bar above denotes the reciprocal tensor. Using the Hamilton–Cayley theorem, one can show that (8) does not change under the simultaneous substitution $\varepsilon \rightleftharpoons \mu$ and $\mathbf{m}' \rightleftharpoons -\mathbf{m}'$. At the same time, (8), generally speaking, is not invariant with respect to the substitution $\mathbf{m} \rightarrow -\mathbf{m}$. For symmetry classes in which $\rho \neq 0$, this is obvious. The classes in which this can occur are easily determined by Curie's principle (subgroups $\infty/m'mm$) or from the data of ¹⁰. These include 31 crystallographic classes and 5 limiting groups (these 36 classes are isomorphic to the ferroelectric and ferromagnetic classes). Among these 36 classes are the 14 groups listed above, as well as the following 22 groups, in which γ has both symmetric and antisymmetric parts:

$$\begin{aligned} & \infty/m', \quad \infty, \quad 6/m', \quad 6, \quad \bar{6}', \quad 4/m', \quad 4, \quad \bar{4}', \quad \bar{3}', \quad 3, \quad mmm', \\ & mm2, \quad 2'2'2, \quad 2'm'm, \quad 2/m', \quad 2, \quad m', \quad 2'/m, \quad m, \quad 2', \quad \bar{1}', \quad 1. \end{aligned}$$

Terms containing odd powers of the components of $\mathbf{m}'$ (there are 3 such terms) can lead to the noninvariance of (8) with respect to the substitution $\mathbf{m} \rightarrow -\mathbf{m}$ also for a number of symmetry groups in which $\rho \equiv 0$, $\mathbf{m}' \equiv \mathbf{m}$, and $\gamma = \bar{\gamma}$ (there are 32 such groups in all). Let us single out these cases. The terms

$$2(\bar{\varepsilon}\gamma\bar{\mu}\mathbf{m}'^{\times})_c \equiv 2((\gamma\mathbf{m})^{\times}\mu\gamma\varepsilon)_c \equiv 0,$$

since the matrices $\bar{\varepsilon}\gamma\bar{\mu}$ and $\mu\gamma\varepsilon$ are symmetric for these groups ($\varepsilon, \bar{\varepsilon}, \mu, \bar{\mu}, \gamma$, and $\bar{\gamma}$ have common systems of principal axes and therefore commute).

The term

$$2m\varepsilon(\gamma m)^\times \mu m = 2m\varepsilon[\gamma m, \mu m]$$

can differ from zero only if the vectors εm , μm , and γm are noncoplanar. This term vanishes in groups where ε , μ , and $\gamma \equiv \bar{\gamma}$ are scalars and where, consequently, εm , μm , and γm are collinear (7 symmetry groups). In groups where ε , μ , and $\gamma \equiv \bar{\gamma}$ are uniaxial tensors of the form $\varepsilon = \varepsilon_1 + (\varepsilon_3 - \varepsilon_1)\mathbf{e} \cdot \mathbf{e}$, etc. (with common axis $\mathbf{e}$), all the vectors εm , μm , and γm lie in the same plane ($m, \mathbf{e}$) and therefore are coplanar (14 symmetry groups). In this case it turns out that the term under consideration can differ from zero in the following 11 groups:

$$4'/m'm'm, \quad \bar{4}2m, \quad \bar{4}2'm', \quad 4'mm', \quad 4'22', \quad m'm'm, \quad 222, \quad m'm'2, \quad 4'/m', \quad \bar{4}, \quad 4.$$

As a result, it turns out that among all 68 crystallographic and limiting groups of magnetic symmetry that allow the m.e., nonreciprocity of electromagnetic-wave propagation (propagation with different phase velocity for the “forward” and “backward” directions) can occur

in 47 symmetry classes. For all these classes there appears the possibility of detecting, by optical methods, 180° domains (for example, ferro- or antiferromagnetic), which are indistinguishable according to the notions of ordinary crystalloptics.

For a sufficiently small m.e. in (8) one may discard all terms containing components of γ to powers higher than the first. Then (8) is simplified and takes the form:

$$\mathbf{m}'\varepsilon\mathbf{m}' \cdot \mathbf{m}'\mu\mathbf{m}' + \mathbf{m}'\{\mu(\bar{\varepsilon}\mu - (\varepsilon\mu)_c) + 2\varepsilon(\gamma\mathbf{m}')^\times\mu\}\mathbf{m}' + 2(\bar{\varepsilon}\gamma\bar{\mu}\mathbf{m}'^\times)_c + |\varepsilon\mu| = 0. \quad (9)$$

In the particular case $\gamma = 0$, (8) goes over into the equation of the normals of the optics of magnetic crystals (see ⁽¹³⁾).

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