Spatial dispersion and new magneto-optical effects in magnetically ordered crystals

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It is shown that magnetic-field-induced optical activity can occur in crystals belonging to the group D_{2h}^{16} in the presence of magnetic ordering.

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It is well known that the terms in the dielectric-constant tensor ϵ_{ij}, which are linear with respect to the components of the wave vector K (optical activity), can exist only in crystals lacking an inversion center.[1] An interesting situation arises in magnetic crystals, whose crystal structure has an inversion center and whose magnetic structure lacks one. In a paramagnetic state, they do not exhibit optical activity, but the transition to a magnetically ordered state (without a change in crystallographic symmetry) can induce it. In this paper we will not make a complete symmetry analysis of the phenomena, but we shall only examine, as a particular example, the possibility for the existence of optical activity in crystals with an inversion center in the presence of an external magnetic field and we shall mention some new magneto-optical effects, which are due to the presence of terms in the dielectric constant of the magnetic crystals that are linear in K.

Specifically, we shall examine crystals belonging to the spatial group D_{2h}^{16}. Such crystals include a wide class of materials of the type RMO_3, where R is a rare-earth ion, and M is a magnetic d ion (Fe, Cr,...) or nonmagnetic ion (Al,Ga,...). Let $\mathscr{M}^0$ be the magnetic modes existing in these crystals, whose transformation properties are presented, for example, in Ref. 2. We use a system of coordinates whose axes coincide with the crystalline axes a, b, and c. We represent the tensor ϵ_{ij}in the form

$$\epsilon_{ij} = \epsilon_{ij}^0 + ie_{ijk} J_k + \text{terms quadratic in } \mathscr{M}_i^\rho\ H_i,\ k_i.$$

The effects related to the quadratic terms are studied in Refs. 3 and 4. The vector J is $J = G + g$, where the vector G determines the usual Faraday effect and g is the magnetic-field-induced optical activity. It is the vector g that is of greatest interest here $(g \sim k)$:

$$g_i = \delta_{ijkm} \mathfrak{M}_j^0 H_k k_m .$$

Let us determine the form of the tensor δ_{ijkm}. This can be done with the help of a multiplication table of the inrreducible representations of the group D_{2h}^{16}. The components g_i and the product $\mathcal{M}_j^0 H_k k_m$ must transform according to the same represention, i.e., $\Gamma(g_i) = \Gamma(\mathcal{M}_j^0)\Gamma(H_k)\Gamma(k_m)$. We shall present the results of such an analysis for effects induced by an external magnetic field, which are realized in the simplest magnetic structures $\Gamma_5 - \Gamma_8$ (see Ref. 2). We note that the gyration vector g vanishes in magnetic structures described by the representations $\Gamma_1 - \Gamma_4$. In addition, the coefficients in front of terms of the type $\mathcal{M}_j \mathcal{M}_k^0 k_m$ vanish in magnetic structures $\Gamma_5 - \Gamma_8$. For the X component of the gyration vector, we find

$$g_x = (a_{xyz}^{(1)} H_y k_z + a_{xzy}^{(1)} H_z k_y)\Gamma_5^m + (a_{xzx}^{(2)} H_z k_x + a_{xxz}^{(2)} H_x k_z)\Gamma_6^m$$

$$+ (a_{xxy}^{(3)} H_x k_y + a_{xyx}^{(3)} H_y k_x)\Gamma_7^m + (a_{xzx}^{(4)} H_z k_x + a_{xxz}^{(4)} H_x k_z)\Gamma_8^m ,$$

where $\Gamma_n^m (n = 5,...8)$ denote linear combinations of the modes $\mathcal{M}^0$, which transform according to the nth representation, for example $\Gamma_5^m = \alpha A_y + \beta G_x$, etc.[2] The two other components of the vector g can be written out in an analogous manner.

Let us examine, for example, the magnetic structure determined by the representation Γ_5 (magnetic group $\overline{m}\overline{m}\overline{m}$). In this group, the components of the gyration vector are given by

$$g_x = B_{xyz}^{(1)} H_y k_z + B_{xzy}^{(1)} H_z k_y ,$$

$$g_y = B_{yzx}^{(1)} H_z k_x + B_{yxz}^{(1)} H_x k_z , \qquad (1)$$

$$g_z = B_{zxy}^{(1)} H_x k_y + B_{zyx}^{(1)} H_y k_x ,$$

where $b_{ijk}^{(1)} = a_{ijk}^{(1)} \Gamma_5^m$, etc. We shall examine below only the simplest cases, in which the vectors H and k are oriented along the coordinate axes. If $H \| k$, then, as is evidence from (1), $g = 0$. If $K \perp H$, then the optical activity will be manifested in the birefringence, although there is no rotation of the polarization plane in the direction of the principal axes, so that a linearly polarized ordinary wave, which is polarized perpendicular to the direction of the magnetic field, and an elliptically polarized extraordinary wave, whose polarization plane coincides with the plane of the vectors H and k, will propagate in the crystal. This effect is analogous to the natural optical activity predicted in Ref. 5. Since the rotation of the plane of polarization as such does not occur, the effect can be observed only with reflection of light from the crystal. For natural optical activity, such observations in the resonant part of the spectrum were recently performed in Ref. 6. For magnetic structure determined by the representation Γ_6, we find

$$g_x = B^{(2)}_{xxx} H_x k_x + B^{(2)}_{xyy} H_y k_y + B^{(2)}_{xzz} H_z k_z ,$$

$$g_y = B^{(2)}_{yyx} H_y k_x + B^{(2)}_{yxy} H_x k_y ,$$

$$g_z = B^{(2)}_{zzx} H_z k_x + B^{(2)}_{zxz} H_x k_z .$$

For this structure, the X axis is the distinguished direction. If the magnetic field is oriented along the X axis, the magnetic circular birefringence exists with propagation of light along any axis and, in particular, perpendicular to the magnetic field, in contrast to the usual Faraday effect, which is possible only for $k \parallel H$. It is also easy to analyze other mutual orientations of the vectors k and H. We note, in addition, the possibility of the appearance of "new waves" (polaritons) near resonances in the presence of a magnetic field; further, the magnetic field makes it possible in this case to control the index of refraction of the "new wave." For magnetic structures determined by representations Γ_7 and Γ_8, the same effects will occur as for crystals with representation Γ_6, but with the difference that the y and z axes, respectively, will be the distinguished direction in this case. The phenomenon examined above concerns a class of effects linear in H (piezomagnetism, linear magnetostriction, linear magnetoelectrical effect, linear birefringence), i.e., these effects change sign under the transformation $H \to -H$.

The crystals examine above also exhibit nonlinear optical effects induced by an external magnetic field. These effects include second-harmonic generation, three-frequency parametric conversion, etc., all of which are determined by a nonlinear susceptibility tensor of rank three, which depends linearly on the magnetic field.

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