

Linear field dependence of magnetic linear birefringence of rare-earth compounds at low temperatures

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It is predicted theoretically that the magnetic linear birefringence will be a linear function of the field in a paramagnetic rare-earth compound, in which the ground state of the rare-earth ion is an isolated doublet, in a strong magnetic field. The theoretical predictions are confirmed by experiments with erbium aluminate garnet.

In a phenomenological description of a magneto-optical anisotropy, in particular, the magnetic linear birefringence, the dielectric constant tensor ϵ_{ij} is usually expanded in powers of the components of the magnetization \mathbf{M} (Ref. 1–4). It is assumed that the magnitude of the magnetic moment does not depend on its orientation in the crystal. This expansion, like the similar expansion of the thermodynamic potential in the theory of the magnetic anisotropy of orientational phase transitions, is based on the relativistic small parameter⁵ v/c .

From the microscopic standpoint, this interpretation of magnetic linear birefringence should be valid for slightly anisotropic magnetic ions such as Gd^{3+} , Cr^{3+} , and Fe^{3+} , for which the splitting of the energy levels of the ground state in the crystal field is much smaller than the Zeeman splitting in an external field (or the effective exchange field), and the energy spectrum of the ion is nearly equidistant.

There is, however, the possibility of another situation (which generally occurs for rare-earth ions): that in which the splitting of the ground multiplet in the crystal field is much greater than the Zeeman splitting, and the magnitude of the magnetic moment can depend on its orientation. In this case, the choice of the small parameter for the expansion of ϵ_{ij} in \mathbf{M} and \mathbf{H} and the expansion formula itself are not obvious. We have accordingly derived a microscopic theory to determine them.

The rare-earth contribution to the dielectric tensor ϵ_{ij} is

$$\Delta\epsilon_{ij} = 4\pi \left(\frac{n^2 + 2}{3} \right)^2 \sum \alpha_{ij}^{(r)}, \quad (1)$$

where n is the average refractive index, α_{ij} is the polarizability tensor, and the summation is over all the rare-earth ions in a unit volume. The tensor α_{ij} can be written

$$\alpha_{ij} = a_0 \delta_{ij} + ia_1 e_{ijk} m_k + a_2 Q_{ij}, \quad (2)$$

where m_k and Q_{ij} are the components of the average magnetic moment and the quadrupole moment of the rare-earth ion, and the coefficients α_i are proportional to the

oscillator strengths of the allowed optical transitions. The first term in (2) determines the polarizability of a free ion, the second determines the gyrotropy, and the third determines, the optical-anisotropy effects. The quadrupole moment of the rare-earth ion can be expressed as the sum of two terms: $Q_{ij} = Q_{ij}^0 + \Delta Q_{ij}$, where Q_{ij}^0 is the spontaneous quadrupole moment (at $H = 0$), and ΔQ_{ij} is the quadrupole moment induced by the magnetic field. The effects of the magneto-optical anisotropy and, in particular, the dependence of the magnetic linear birefringence on the orientation and magnitude of the field and the temperature are determined completely by the induced quadrupole moment.

As an example, we consider a Kramers rare-earth ion ($\text{Er}^{3+}, \text{Dy}^{3+}, \dots$), whose ground state in a crystal field is a doublet separated from the higher-lying (excited) levels by a rather large energy interval ΔE . If the temperature is low ($\Delta E \ll T$), and we can ignore the population of excited levels, the system can be described by an effective spin $S_{\text{eff}} = 1/2$. An external magnetic field splits the doublet and mixes its wave functions, ψ_1 and ψ_2 . In other words, an external magnetic field rotates the (ψ_1, ψ_2) spinor in spin space. An important point here is that the expectation value of Q_{ij} does not change upon this rotation; i.e., the induced quadrupole moment is zero, so that a magnetic linear birefringence does not arise in this approximation. To find the final value of the magnetic linear birefringence, we need to take into account the excited levels of the rare-earth ion. In other words, in the doublet system the magnetic linear birefringence and other even effects are determined not by the ground state but by the interaction of the ground state with excited levels. The admixture of excited states to the ground state leads, in lowest-order perturbation theory, to

$$\Delta Q_{ij} = \sum_{kl} G_{ijkl} H_l \frac{m_k}{g_k}, \quad (3)$$

where g_k is a component of the g -tensor of the ground doublet of the ion in its local coordinate system, and the G_{ijkl} are real numerical coefficients (for which we will not reproduce here the specific expressions).

We direct the magnetic field along the symmetry axes of the crystal. Summing relation (3) over all the rare-earth ions in the crystal for this case, and using (1) and (2), we find an expression for the magnetic linear birefringence:

$$\Delta n = AMH, \quad (4)$$

where M is the magnetization of the rare-earth subsystem of the crystal for the given orientation of the magnetic field. The coefficient A is a known function of the field orientation and of the light propagation direction.

There is a close analogy between the mechanism for the appearance of a magnetic linear birefringence for a rare-earth ion with a doublet ground state and the mechanism for the Van Vleck magnetization of ions in a singlet ground state. In the latter case, when we take into account only the ground state, the magnetic moment of the ion is zero, and the admixture of excited states to the ground state exclusively determines the magnetic moment and the susceptibility of the singlet state. For a doublet, the expectation value of the induced quadrupole moment is zero, so that in calculating

it we need to take into account the admixture of excited states to the ground doublet, i.e., the effect that determines the Van Vleck magnetization.

Calculations show that expression (4) describes the magnetic linear birefringence of non-Kramers rare-earth ions (Tb^{3+}, Ho^{3+}, \dots) if their ground state is a well-isolated quasideoublet. For ions with $S_{\text{eff}} > 1/2$ in the ground state (a triplet, a quartet, etc.) and, in particular, for ions in the S state, the magnetic linear birefringence is given by

$$\Delta n = \Delta n_0 \hat{I}_{5/2} [\mathcal{L}^{-1}(m)], \quad (5)$$

where $\hat{I}_{5/2}$ is the reduced Bessel function, \mathcal{L}^{-1} is the reciprocal of the Langevin function, and m is the relative magnetization. Expression (5) is analogous to that which has been derived for the magnetostriction in the theory of even Akulov effects.⁶

These arguments can easily be tested by studying the magnetic linear birefringence of paramagnets with a doublet ground state in strong magnetic fields at low temperatures. If these arguments are correct, then in the region of paramagnetic saturation, where the magnetization stops changing, the magnetic linear birefringence should be linear function of the field, while a saturation of the magnetic linear birefringence should be observed according to the "classical" model in the field region corresponding to paramagnetic saturation.

Convenient systems for an experimental test of the field dependence of the magnetic linear birefringence are the rare-earth gallates and aluminates with the garnet structure, in which the ground state of the rare-earth ion is often a doublet separated by a considerable distance from the higher-lying levels.⁷ In particular, in erbium aluminate garnet, $Er_3Al_5O_{12}$, the ground doublet lies 27 cm^{-1} below the first excited level.⁸

Figure 1 shows the results we found on the dependence of the magnetic linear

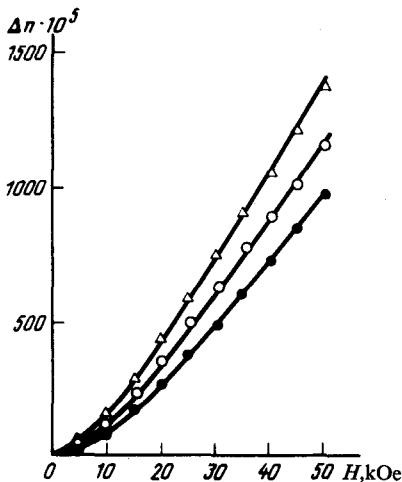


FIG. 1. Magnetic linear birefringence of $Er_3Al_5O_{12}$ versus the magnetic field at 4.2 K. The light propagation vector is perpendicular to the (110) plane of the crystal. The points are experimental. \bullet — $H \parallel [100]$; \circ — $H \parallel [110]$; \triangle — $H \parallel [111]$. The lines are theoretical [expression (4)].

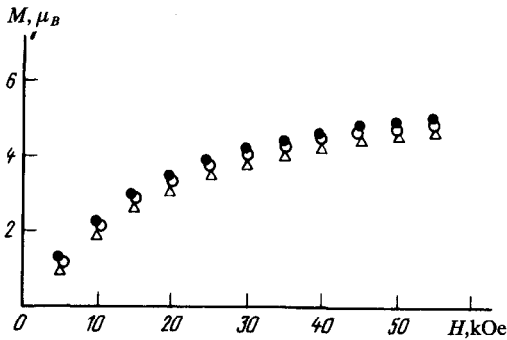


FIG. 2. Magnetization per rare-earth ion of $\text{Er}_3\text{Al}_5\text{O}_{12}$ versus the magnetic field at 4.2 K. ●— $H \parallel [100]$; ○— $H \parallel [110]$; △— $H \parallel [111]$.

birefringence of the $\text{Er}_3\text{Al}_5\text{O}_{12}$ single crystal on the magnetic field at 4.2 K; Fig. 2 shows the field dependence of the magnetization of the crystal. We see that in weak fields, where the magnetization is proportional to the field, the magnetic linear birefringence is a quadratic function of the field, while in strong fields, where the magnetization tends toward paramagnetic saturation, the magnetic linear birefringence does not reach saturation and instead increases linearly with the field. Consequently, in erbium aluminate garnet, which is a highly anisotropic paramagnet with a doublet ground state, relation (4) for the magnetic linear birefringence does hold. In other words, this magneto-optical effect cannot be expressed in terms of the magnetization alone, as in the "classical" case.

Finally, we note that preliminary experimental data indicate that a linear dependence of the magnetic linear birefringence on the field is observed in many rare-earth gallates and aluminates: garnets with a doublet or quasidoublet ground state for the rare-earth ion.

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