New magneto-optical effect in antiferromagnetic fluorides of transition metals in longitudinal magnetic fields

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We demonstrate the feasibility of inducing linear-in-intensity magneto-optical effects by an external magnetic field that does not violate the collinearity of the spins.

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The influence of the magnetic subsystem of magnets has raised considerable interest in magneto-optical investigations. In particular, detailed investigations were made of effects of circular and linear birefringence in antiferromagnetic fluorides of transition metals[1–3] for different combinations of the directions of the antiferromagnetism vector I, the external magnetic field vector H, and the wave vector of the light Q. To our knowledge, however, no experiment with all three vectors parallel has been performed, although it is precisely this experimental configuration that makes possible the magneto-optical effect that is peculiar to crystals of similar types (space group D_{4h}^{14}) and is due to the presence of single-ion anisotropy. For the analysis of the phenomenon it is convenient to use the exciton model of spin excitations,[4,5] in which it is possible to take exact account of the singularities of single-ion anisotropy by solving the self-consistent problem that determines the spin configuration of the ground state. The latter turns out to be nontrivial even at a temperature $T = 0$ and when the collinearity of the spins and of the external field is preserved.[6] In this case, the self-consistent problem reduces (neglecting the inessential correction necessitated by the zero-point oscillations) to a determination of the field dependences of the quantities

$$\tilde{S}_\alpha = \langle \psi_{n\alpha}^0 | S_{n\alpha}^z | \psi_{n\alpha}^0 \rangle ,$$

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($\psi^0_{n\alpha}$ is a function of the ground state of the ion, n is the site, and α is the number of the sublattice), which are not equal to the spin S because of the quantum-mechanical mixing of the states with different spin projections on the easy (tetragonal) axis OZ, a mixing due to the presence of a "transverse" part in the one-ion anisotropy operator that takes the following form for the crystals of interest to us

$$\mathcal{H}^{aH}_{n\alpha} = -A(S^z_{n\alpha})^2 + (-1)^\alpha B[(S^x_{n\alpha})^2 - (S^y_{n\alpha})^2], \quad A > 0.$$

In strong fields, the solutions for the case of integer and half-integer S are substantially different, but in fields that are weak in comparison with the exchange fields the following expression holds for all S:

$$\tilde{S}_{1,2} = \tilde{S}_0 \pm {}^1/_2 \chi_{\parallel} H,$$

where $\tilde{S}_0$ and $\chi_{\parallel}$ are determined only by the relations between the parameters A and B and the magnitude of the exchange J. (We note that $\chi_{\parallel}$ differs from zero also in the case of biaxial crystals.) It follows directly from the last expression that the sublattices become nonequivalent even at a fixed positions of the nuclei, and consequently, owing to the decompensation of local biaxiality (the local symmetry of the metal ions is rhombic) the optically nonequivalent axes are OX and OY. In addition, a longitudinal angular momentum $m^z = S_1 - S_2$ is produced and induces a gyration vector $\mathbf{G} \| \mathbf{H} \| OZ$. Linearly polarized light $\mathbf{Q} \| OZ$ passing through such a crystal should become elliptically polarized. The nonequivalence of the different light-polarization directions in a plane perpendicular to the tetragonal axis should be substantial also for vectors $\mathbf{Q} \perp OZ$ (in this case there is no circular component of the effect). The resultant biaxiality does not contradict symmetry considerations, inasmuch as at $m^z \neq 0$ the symmetry element $C_4 \times R$ vanishes (C_4 is the rotation through 90° about the tetragonal axis and R is the time-reveral operation).

The change of the refractive index of the crystal is determined by the contributions from the changes of both the magnetic and dielectric susceptibility tensors; in addition, it must also be recognized that part of the change of these tensors at $H \neq 0$ is due to magnetrostriction, which generally speaking is not small for crystals of this type (particularly for CoF_2). The ensuing strains along [110] and [001] should give rise to an additional piezomagnetic effect, which lowers the Zeeman energy.

We consider the magnetic contribution, assuming absence (or cancellation in the experiment) of magnetostriction. The magnetic-susceptibility tensor is determined by the Fourier transforms of the retarded Green's functions $\ll S^i_{n\alpha} | S^j_{m\beta} \gg_\omega (i,j = x,y,z)$, and has a resonant character in the ir region. In the region of optical frequencies it can be represented in the form

$$\chi_{ij}(\omega) = \ll S^i_{n\alpha} | S^j_{m\beta} \gg_\omega = R_{ij}(J/\omega^2).$$

The expressions for R_{ij} are complicated and will be given in a detailed paper for the case $S = \frac{3}{2}$. It is significant that in weak fields we have $R_{xx} - R_{yy} \sim \chi_{\parallel} H$, $R_{xy} \sim \chi_{\parallel} H$ and $R_{zz} \sim H^2$; $R_{xz} = R_{yz} = 0$. One should expect the change in the refractive indices at $H \| OZ$ to be of the same order as in analogous effects in trans-

verse fields for the CoF_2 crystal, where the large single-ion anisotropy gives rise to a value of $\chi_{\parallel}$ which is smaller by only a few times than $\chi_{\perp}$ at $T=0$.

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[1]A. S. Borovik-Romanov, N. M. Kreĭnes, and M. A. Talalaev, Pis'ma Zh. Eksp. Teor. Fiz. **13**, 80 (1971) [JETP Lett. **13**, 56 (1971)].
[2]I. R. Jahn and H. Dachs, Solid State Commun. **18**, 1617 (1971).
[3]A. S. Borovik-Romanov, N. M. Kreĭnes, A. A. Pankov, and M. A. Talalaev, Zh. Eksp. Teor. Fiz. **64**, 1762 (1973) [Sov. Phys. JETP **37**, 890 (1973)].
[4]Yu. B. Gaĭdideĭ and V. M. Loktev, Fiz. Tverd. Tela **16**, 3435 (1974) [Sov. Phys. Solid State **16**, 2224 (1975)].
[5]V. S. Ostrovskii and E. G. Petrov, Phys. Status Solidi B **71**, 369 (1975).
[6]V. S. Ostrovskiĭ, Fiz. Tverd. Tela **18**, 1041 (1976) [Sov. Phys. Solid State **18**, 594 (1976)].