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G. A. Smolenskiĭ, R. V. Pisarev, and I. G. Siniĭ, Investigation of Magnetooptic Phenomena in Ferro- and Antiferromagnets

The first magnetooptic (MO) investigations of ferro- and antiferromagnetic dielectrics were published 10 years ago, when perfect iron garnets of yttrium and rare earths, transparent in the infrared and (in the case of thin layers) in the visible region of the spectrum were synthesized. It was subsequently observed that single-crystal compounds based on divalent europium, chromium halides, and fluorides of the 3d group have good transparency in the infrared, visible, and even ultraviolet regions of the spectrum. This has led to the expansion of optical and MO investigations of magnetically-ordered compounds. MO research was greatly stimulated by the great potential capabilities of using magnetically-ordered materials for practical application in light channels for beam control (for the development of modulators, shutters, circulators, memory elements, and other devices).

The present paper is devoted to MO research carried out during the last few years in the Laboratory of Magnetism and Ferroelectricity of the Semiconductor Institute of the USSR Academy of Sciences. The research deals with the following effects produced in crystals by the passage of light: 1) along the external magnetic field—the Faraday effect (FE), or the rotation of the plane of polarization of the light, and 2) when the light propagates perpendicular to the field—the Cotton-Mouton effect (CME), or magnetic birefringence. The FE effect is linear in the magnetization and the CME is quadratic.

The investigation of the FE and the CME was carried out in a number of magnetically-ordered crystals with different types of magnetic ordering, ferrimagnets and antiferromagnets. An unexpected result was the fact that the FE and the CME in ferrimagnets and the CME in antiferromagnets are quantities of the same order (see the table), whereas in paramagnetic crystals the quadratic effects are weaker by a factor 2—3 than the linear effects. The mechanisms of the MO effects in magnetically ordered compounds were considered within the framework of the concept of the polarizability tensor. It is shown that the FE is determined by the spin-orbit interaction, and the main contribution to the CME is connected with the isotropic exchange interaction between the paramagnetic ions in different sublattices. At the same time, there should exist a contribution to the CME, due to the anisotropy of the single paramagnetic ion, and also due to the anisotropy of the exchange interaction. These two contributions can lead to the CME anisotropy, which can be appreciable in rare-earth iron garnets. The analysis of the mechanisms of these two principal MO effects is in good agreement with the results of the experimental research. Thus, for example, in the ferrimagnet $RbNiF_3$ (Curie point $T_C = 139$°K) at $T < T_C$, the FE and CME differ insignificantly (see the table). On going through T_C, however, the FE decreases by 1—2 orders of magnitude, whereas the decrease of the TME reaches 3—4 orders, i.e., in the paramagnetic region, where the contribution of the exchange mechanism to the CME vanishes, the usual ratio of the values of the linear and quadratic MO effects is observed.

In measurements of the CME in cubic terbium iron garnet $Tb_3Fe_5O_{12}$, a strong anisotropy of the effect was observed, which increased with decreasing temperature. Thus, at a wavelength $\lambda = 1.15\ \mu$ and $T = 295$°K, the result was $\Delta n^{100} = 6 \times 10^{-5}$, and with decreasing temperature the birefringence increased to $\Delta n^{100} = 70 \times 10^{-5}$ at 77°. Similarly, $\Delta n^{111} = 3 \times 10^{-5}$ at 295°K, and with decreasing temperature Δn^{111} reverses sign at 200°, reading $\Delta n^{111} = -72 \times 10^{-5}$ at 77°K. An analysis of the $\Delta n(H, T)$ curves shows that magnetic birefringence in $Tb_3Fe_5O_{12}$ must be connected not only with the CME, but also with birefringence due to magnetostriction, which reaches large values in this crystal. An anomaly of the birefringence was observed in the vicinity of the magnetic compensation point T_{comp}, namely a double change in the sign of the birefringence together with an increase in the depolarization and scattering of light passing through the crystal. These phenomena were observed in sufficiently strong magnetic fields and are the consequence of stimulated phase transitions from one ferromagnetic state into the antiferromagnetic state, and then into another ferromagnetic state with changing temperature. The turning of the magnetic sublattices at the compensation temperature leads to a reversal of the sign of Δn, and the fluctuation of the direction and magnitude of the magnetic moments cause an increase in the depolarization and scattering of the light.

The foregoing experiments were performed in a spectral region where the investigated crystals were

Quadratic CME and linear FE in ferromagnets and antiferromagnets (in a field $H = 20$ kOe)

Crystal	T_{CM}, °K	T_{expt}	λ, μ	Δn_{CM}	β_{CM} deg/cm	Δn_F	α_F deg/cm
$Y_3Fe_5O_{12}$	550	295	1,15	$4{,}5\cdot10^{-5}$	141	$1{,}6\cdot10^{-4}$	260
α-Fe_2O_3	950	295	1,15	$2{,}1\cdot10^{-4}$	657	—	—
$RbNiF_3$	139	77	0,555	$2{,}2\cdot10^{-5}$	142	$3\cdot10^{-5}$	95
$RbFeF_3$	102	77	0,556	$2{,}5\cdot10^{-4}$	1600	$2{,}2\cdot10^{-4}$	680
$Tb_3Fe_5O_{12}$, [100]	568	295	1,15	$6{,}0\cdot10^{-5}$	188	$2{,}9\cdot10^{-4}$	450

transparent. The presence of intrinsic-absorption bands leads to the appearance in these regions of a spectrum of magnetic circular dichroism, accompanying the FE, and magnetic linear dichroism, accompanying the CME. The developed MO research procedure makes it possible to investigate all these effects. The spectral, temperature, and field dependences of these effects were investigated in the ferromagnets $RbNiF_3$ and $Rb(Ni, Co)F_3$ in the weak ferromagnetic $RbFeF_3$, and in the antiferromagnet $KNiF_3$. In the region of the absorption lines, dispersion of the MO effects was observed. The investigations have shown that the observed phenomena are connected with the splitting of the electronic states of the paramagnetic ions below T_C in the exchange field.

The investigation of the MO effects turns out to be fruitful for the interpretation of the magnetic structure of crystals. This is connected with the fact that the magnitude of the linear MO effects is proportional to the magnetization, and oppositely oriented magnetic sublattices make contributions of opposite signs and of different magnitudes to the summary effect. This circumstance has made it possible to confirm the two-sublattice model of the magnetic structure of $RbNiF_3$ and to confirm that this substance is a ferrimagnet of the "easy plane" type, and addition of Co^{2+} up to 30 mol.% leads to its transformation into a uniaxial ferromagnet. It was also established that $RbFeF_3$ is a weak ferromagnet in fields up to 32 kOe. The turning of the magnetic moments of the sublattices in hematite (α-Fe_2O_3) at the Morin point is accompanied by a vanishing of the CME with decreasing temperature at a given observation geometry.

In conclusion, we note that until now most magneto-optic investigations of ferromagnetic and antiferromagnetic dielectrics have been devoted to the FE. However, the large value of the CME in magnetically-ordered crystals and the relative ease of observing the CME uncovers interesting possibilities of investigating exchange interactions in crystals, the temperature dependences of the sublattice magnetizations, the orientations of the magnetic moments relative to the crystallographic axes, and other phenomena. In addition, the CME can be employed with equal success to investigations of both ferromagnets and antiferromagnets, whereas linear MO effects can be used mainly for the investigation of ferromagnets.

Translated by J. G. Adashko