Faraday effect in antiferromagnetic NiO in a strong magnetic field

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An experiment is described on the Faraday effect in fields reaching 1.6 MOe at a wavelength $\lambda = 6328$ Å at room temperature. A nonlinear dependence and a reversal of the sign of the effect is observed with increasing field. The possible causes of this phenomenon are discussed.

The use of strong magnetic fields that compete with exchange spin-orbit and other interactions in magnetic materials yields information on the magnetic-ordering mechanisms and on the splitting of the electronic states. To our knowledge, optical investigations of magnetic crystals have not been performed in fields stronger than 1 MOe.

We have investigated the Faraday magnetooptical effect in the antiferromagnet NiO ($T_N = 523$ °K). The experiment was performed in the ordered region at the temperature 290 °K, using an explosive-type setup producing a field on the order of 2 MOe.[1] Two identical samples 0.44 cm long of TF-5 glass (Verdet constant 0.046 min/cm-Oe) were placed in the field, and an NiO plate 32 μ thick was placed on one of the samples. A helium-neon laser beam ($\lambda = 6328$ Å) illuminated both samples and the rotation of the polarization plane after passage of the light through the polarizer, sample, and analyzer, was determined from the darkening of two photomultipliers with a two-beam oscilloscope. The field strength was measured by two independent methods, with induction pickups and by rotation of the plane in the TF-5 sample.[2]

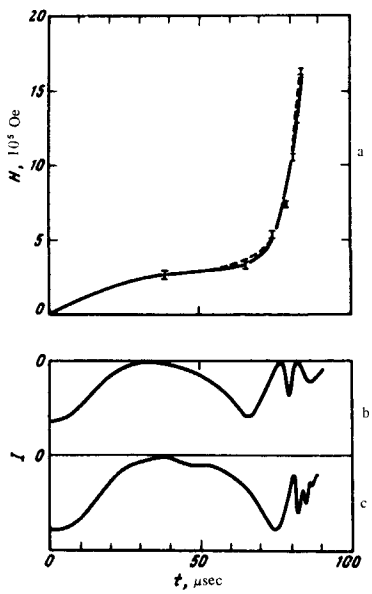

FIG. 1. a) Increase of field with time, measured by induction pickups (dashed) and by the Faraday effect (solid line) in the first experiment; b) oscillogram from TF-5 sample with NiO; c) oscillogram from TF-5 sample.

Figure 1a shows the growth of the field as measured by induction pickups and by rotation in the glass. Figures 1b and 1c show oscillograms of the rotation of the polarization plane in the TF-5 sample with and without the NiO plate, respectively. The distance between the two minima or maxima of the oscillogram corresponds to rotation through 180°, and the accuracy with which the angle is measured is estimated here at $\mp 10°$. Two experiments were performed on the measurement of the Faraday rotation up to a maximum field ~1.6 Ma (Figs. 2a and 2b). Both experiments yielded close results, and the field dependence of the Faraday effect, obtained on their basis, is shown in Fig. 3.

As seen from Fig. 3, in weak fields the rotation of the polarization plane increases with increasing field. At an intensity $H \sim 0.9$ MOe the rotation reaches a maximum, then decreases, and the effect reverses sign at $H \sim 1.5$ MOe. In our opinion, the Faraday effect in the antiferromagnet NiO can be altered either by the action of

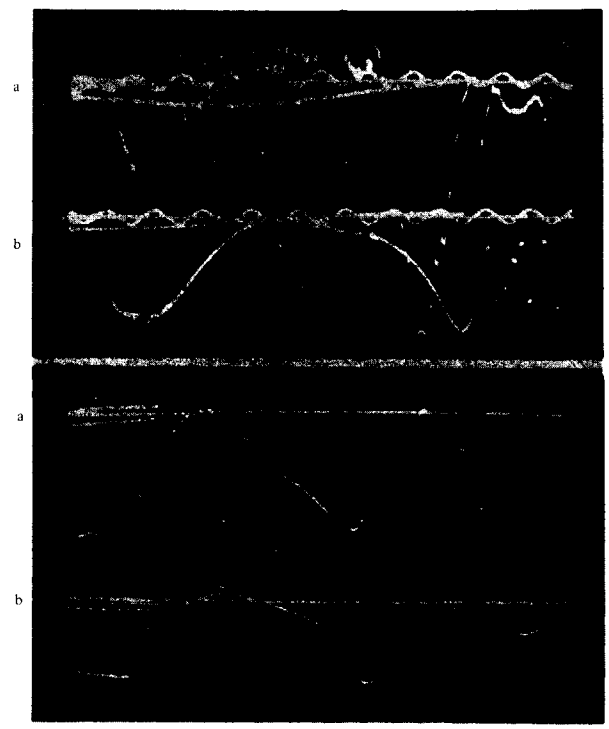

FIG. 2. Oscillograms of polarization-plane rotation in the sample TF-5 plus NiO (top) and TF-5 (bottom) in the first experiment (a) and in the second experiment (b).

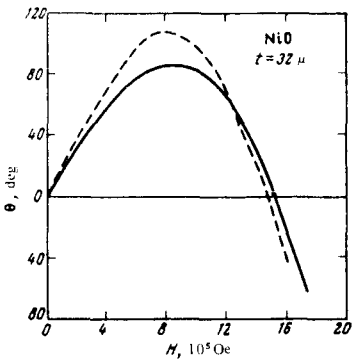

Fig. 3. Experimental (dashed) and calculated (solid) dependence of the polarization-plane rotation angle θ on the field H.

the external field on the magnetic structure, or by the action of the field on the splitting of the electronic states and on the intensities of the transitions that determine the effect. We consider initially the first mechanism.

In weak fields on the order of several kOe, the moments of the antiferromagnetic sublattices are oriented parallel to the field. Further increase of the field causes "flipping" of the sublattices, i.e., a transition from the antiferromagnetic to the ferromagnetic structure. The sublattice "flipping" field is determined only by the exchange interaction[3-4]

$$H_E = \frac{3\sigma T_N}{g\mu_B (S+1)} \approx 4{,}8\ \text{MOe}, \qquad (1)$$

where $\sigma = M(T)/M(0)$ is the relative magnetization of each sublattice and μ_B is the Bohr magneton. Thus, in fields ~1.5 MOe the sublattice "flipping" is appreciable. An increase of the magnetic field component in the field direction leads to an increase of the so-called paramagnetic rotation $\theta_p = -ch$, where $h = H/H_E$ and c is a constant. Thus, the sublattice "flipping" itself cannot explain the observed nonlinear field dependence of the Faraday rotation.

The magnetic field can exert a strong influence on the electronic structure of NiO, i.e., on the position of the absorption band and on the transition intensity. In fact, a field ~1 MOe leads to a splitting of ~100 cm^{-1} of the electronic states (at $g = 2$), which may be comparable with the widths of the absorption bands and with their spin-orbit splitting. This may distort the linear dependence of the rotation on the field, but we know of no calculations that take these factors into account. It is possible that a similar influence obtains in other magnetic crystals, where violation of the proportionality of the Faraday effect to the magnetization was observed.[5,6]

It appears that an effective role can be played in the case of antiferromagnetic crystals by the diamagnetic contribution θ_d to the Faraday effect, which is proportional to h and to the sum of the squares of the matrix elements of the transitions from the ground to the excited state.[2] We consider only the contribution due to the polarizability that depends on the ferromagnetic exchange interaction. This polarizability is connected with transitions described by the operator $(\mathbf{e}\cdot\pi)(\mathbf{S}_1\cdot\mathbf{S}_2)$, where $\mathbf{S}_1$ and $\mathbf{S}_2$ are the spin operators of ions of two antiferromagnetic sublattices and are nondiagonal, $\mathbf{e}$ is the electric vector of the light wave, and π is the dipole moment.[7] This operator describes many optical phenomena in magnetic crystals, such as two-magnon, exciton-magnon, or two-exciton absorption, etc.

If the antiferromagnetic ordering is destroyed by an external magnetic field, then the contribution made to the polarizability by such transitions decreases like $(1-h^2)^2$.[8] A similar influence of the field on the intensities of individual lines was observed in the absorption spectrum of $FeCO_3$.[9] In the case of the Faraday effect, we can describe the observed field dependence by the formula

$$\theta = a(1-h^2)^2 h - c^* h, \qquad (2)$$

where c^* takes into account the paramagnetic and diamagnetic contributions that depend linearly on the field. This formula makes it possible to describe satisfactorily the experimental curve (Fig. 3) at $a = 3670$ deg and $c^* = 2940$ deg. An estimate of c^* in accordance with formula (15) of[2] yields a close value, but the value of a turns out to be larger than expected. It is possible that so large a value of a is due to the proximity of the observation frequency to the electronic transition in the divalent-nickel ion, particularly to the transition to the ^1E level.

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[1] A. D. Sakharov, R. Z. Lyudaev, E. N. Smirnov, Yu. I. Plyushchev, A. I. Pavlovskiĭ, V. K. Chernyshev, E. A. Feoktistova, E. I. Zharinov, and Yu. A. Zysin, Dokl. Akad. Nauk SSSR 165, 65 (1965) [Sov. Phys.-Dokl. 10, 1045 (1966)].
[2] V. V. Druzhinin and O. M. Tatsenko, Opt. spektr. 36, 733 (1974).
[3] R. E. Dietz, G. I. Parisot, and A. E. Meixner, Phys. Rev. B4, 2302 (1971).
[4] A. Tsuchida, J. Phys. Soc. Jap. 21, 2497 (1966).
[5] N. F. Kharchenko, L. I. Belyi, and O. P. Tutakina, Fiz. Tverd. Tela 10, 2819 (1968) [Sov. Phys.-Solid State 10, 2221 (1969)].
[6] G. S. Krinchik and S. A. Gushchina, ZhETF Pis. Red. 4, 244 (1966) [JETP Lett. 4, 164 (1966)].
[7] T. Moriya, J. Appl. Phys. 39, 1042 (1968).
[8] V. V. Druzhinin and V. V. Teĭmurazyan, Fiz. Tverd. Tela 14, 1600 (1972) [Sov. Phys.-Solid State 14, 1381 (1972)].
[9] V. V. Eremenko, Yu. G. Litvinenko, and V. P. Myatlik, ZhETF Pis. Red. 12, 66 (1970) [JETP Lett. 12, 47 (1970)].