Magneto-optic intensity effects in ferromagnetic metals and insulators

G. S. Krinchik, E. E. Chepurova, and Sh. V. Égamov

Moscow State University

(Submitted 2 August 1977)
Zh. Eksp. Teor. Fiz. **74**, 714–719 (February 1978)

The method of meridional and polar intensity effects was used to investigate the magneto-optic reflection spectra of single-crystal films of iron, nickel, hematite, yttrium orthoferrite, yttrium iron garnet, and bismuth garnet. The observed peaks in the dependences $\delta_m(\omega)$ and $\delta_p(\omega)$ obtained for ferromagnetic dielectrics were identified as the first intense optical transitions in the near ultraviolet ($\hbar\omega > 1.8$ eV). The meridional intensity effect exhibited by single-crystal Ni and Fe films depended weakly on the angle of incidence of light in the range $45° \leq \varphi \leq 70°$ and it reached its maximum value at $\theta = 30$–$40°$. In the case of ferromagnetic insulators an investigation of the dependences of the meridional and polar intensity effects on the angle of polarization of light θ at a fixed wavelength revealed a strong maximum at fairly small angles θ. For example, the meridional intensity effect had its maximum value at $\theta = 10°$ and 8° for α-Fe_2O_3 and $YFeO_3$, and the polar intensity effect had its maxima at $\theta = 8°$, 4°, and 6° for $YFeO_3$, $Y_3Fe_5O_{12}$, and $BiCaVFe_5O_{12}$, respectively. A comparison of the experimental and calculated values of these effects demonstrated a fully satisfactory agreement.

PACS numbers: 78.20.Ls, 78.65.Ez, 78.65.Jd

In an earlier paper[1] we discussed new (linear in the magnetization) effects involving changes in the intensity of the reflected light in the case of polar (with the magnetization vector I in the plane of incidence of light and perpendicular to the surface of the sample) and meridional (I parallel to the plane of incidence of light and to the surface of the sample) magnetization of a ferromagnet. It should be stressed that the usual meridional and polar Kerr effects are due to the circular birefringence, i.e., due to the difference between the refractive indices for left- and right-handed circularly polarized light, which rotates the plane of polarization and changes the polarization of light from linear to elliptic. The intensity of light reflected from a magnetized ferromagnet should remain constant in the first approximation with respect to I, because the influence of the magnetization on the reflection coefficient of the left- and right-handed circularly polarized components is the same but of opposite sign and, consequently, it should cancel out for linearly polarized *p* and *s* waves (the vector **e** is then oriented, respectively, parallel and perpendicular to the plane of incidence of light). However, if the plane of polarization occupies some intermediate position between the *p* and *s* polarizations, the intensity of the reflected light is different for the polar and meridional magnetizations and it is a linear function of I.

Only one effect of this type—the equatorial Kerr effect (with the vector I perpendicular to the plane of incidence of light and parallel to the surface of the sample)—has been investigated so far. The recently discovered new magneto-optic effects have extended the range of the intensity effects and have thus increased the potentialities of the magneto-optic methods. In particular, the task of determination of the permittivity tensor $[\epsilon]$ has become easier (particularly when using cryostats in which it is convenient to have a fixed angle of incidence of light) and there has been simplification in studies of ferromagnetic samples by means of a magneto-optic micromagnetometer,[2] because simple switching of the magnetic field and a change in the state of polarization of the incident light makes it possible to increase the number of linearly independent magneto-optic effects without a change in the experimental conditions. Moreover, these effects are odd not only in respect of the magnetization but also in respect of the angle of deviation of the plane of polarization from the *p* and *s* components. Consequently, we can determine the orientation of the magnetization vector by modulating the plane of polarization of light and this makes it possible to eliminate the contribution due to a change in the ordinary reflection coefficient.

A phenomenological theory yields the following formulas for the meridional and polar intensity effects[1]:

$$\delta_p = \frac{m}{C - D\cos 2\theta}\sin 2\theta, \tag{1}$$

$$\delta_m = \frac{m'}{C - D\cos 2\theta}\sin 2\theta, \tag{2}$$

where

$$\begin{gathered} m = M_2 q \sin 2\varphi \sin\varphi, \\ m' = [(n^2 - k^2 - 1)M_2 \\ -2nkM_1]\sin 2\varphi \cos\varphi, \\ C = (q + \cos^2\varphi)(q\cos^2\varphi + 1) \\ -4n^2\cos^2\varphi, \quad D = (q-1) \\ \times \sin 2\varphi \sin\varphi, \quad q = n^2 + k^2, \end{gathered}$$

φ is the angle of incidence, ϑ is the angle of deviation of the plane of polarization from the *p* component, n and k are the real and imaginary parts of the refractive index $n = n - ik$, M_1 and M_2 are the magneto-optic parameters ($M = M_1 - iM_2$).

We checked the theory and measured the intensity effects in new materials with a high magneto-optic figure of merit by a detailed investigation of these effects in the wavelength range from 0.5 to 3.5 eV. We used magneto-optic apparatus based on a DMR-4 monochromator. Natural light from a highly stabilized source passed

0038-5646/78/4702-0375$02.40

through the entry slit of the DMR-4 double monochromator with interchangeable quartz and glass prisms. The resultant monochromatic beam was then focused by a lens on the surface of a ferromagnetic sample. A polarizer was placed between the lens and the sample. The light reflected from the sample was focused on a radiation detector by a plane aluminized mirror and a lens. The detector was a PbS photoresistor in an evacuated cell, which was used in the 0.5 – 1.4 eV range, and an FÉU-79 photomultiplier, employed in the 1.4 – 3.5 eV range. The sample was subjected to an alternating magnetic field of 80 Hz frequency. An electromagnet was supplied from a GZ-34 af oscillator and a UM-50 power amplifier. The modulated signal was measured by means of a U2-6 selective amplifier and a V9-2 synchronous detector. The static component of the current passing through the photodetector was recorded with a V2-11 microvoltmeter.

The frequency and angular dependences of the meridional intensity effect were measured for single-crystal iron and nickel films in the (100) plane, for a $YFeO_3$ plate cut parallel to the c axis, and for hematite in the basal plane. These materials were selected because they could be easily magnetized to saturation parallel to the surface.

The components of the permittivity tensor, needed in the calculation of the effect, were known only for the Ni and Fe single crystals.

Figure 1 shows the frequency dependence of the meridional intensity effect δ_m obtained for a single crystal Ni film at two angles of incidence: $\varphi = 45°$ and $\varphi = 70°$; the angle of deviation of the plane of polarization from the p component was $\theta = 45°$. The same figure includes, for comparison, the dependences of the equatorial Kerr effect obtained in the same wavelength range (0.5 – 3.5 eV) and for the same angles of incidence of light. The upper part of the figure shows (see the inset) the dependence $\delta_m(\theta)$ obtained at a fixed wavelength ($\hbar\omega = 3.1$ eV). The results of a similar investigation carried out on a single-crystal Fe film are plotted in Fig. 2. The dependences $\delta_m(\omega)$ and $\delta_m(\theta)$ (curves denoted by 1) obtained for hematite (α-Fe_2O_3) for $\varphi = 70°$ are plotted in Figs. 3 and 4. Curves denoted by 2 in these figures represent the dependences $\delta_m(\omega)$ and $\delta_m(\theta)$ obtained at $\varphi = 65°$ for yttrium orthoferrite ($YFeO_3$).

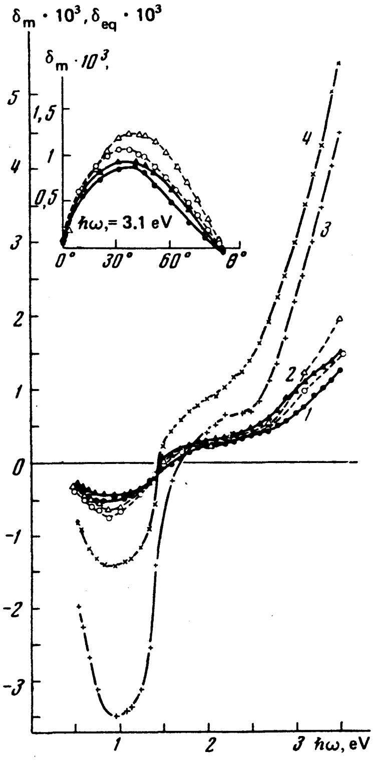

FIG. 1. Meridional intensity (1, 2) and equatorial (3, 4) effects in a single-crystal film of Ni: 1) $\varphi = 70°$, $\theta = 45°$; 2) $\varphi = 45°$, $\theta = 45°$; 3) $\varphi = 70°$; 4) $\varphi = 45°$. Here and later, the dashed curves are calculated.

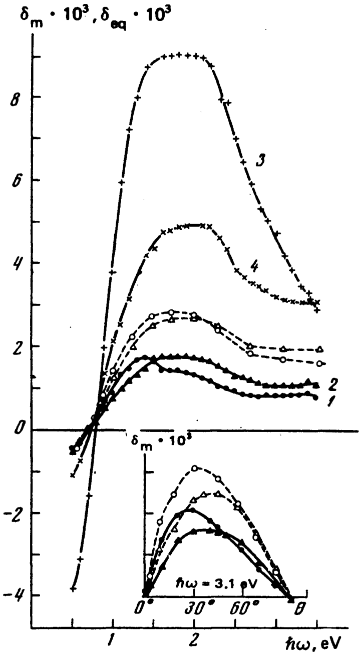

FIG. 2. Meridional intensity (1, 2) and equatorial (3, 4) effects in a single-crystal film of Fe: 1) $\varphi = 70°$, $\theta = 45°$; 2) $\varphi = 45°$, $\theta = 45°$; 3) $\varphi = 70°$; 4) $\varphi = 45°$.

It is clear from the experimental data that, in contrast to the equatorial Kerr effect, the meridional intensity effect in single-crystal Ni and Fe films depends weakly on the angle of incidence of light in the range $45° \leq \varphi \leq 70°$. Moreover, the dependences $\delta_m(\theta)$ obtained at a fixed wavelength show that in the case of metals the maximum value of δ_m is reached at θ of the order of 30–40°, whereas in the case of ferromagnetic insu-

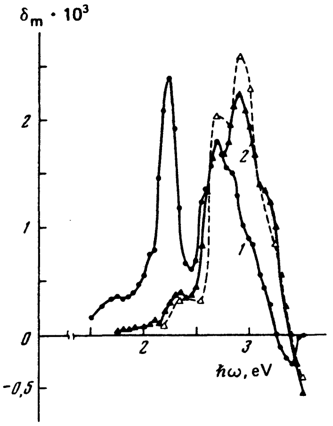

FIG. 3. Meridional intensity effect: 1) α-Fe_2O_3 ($\varphi = 70°$, $\theta = 10°$); 2) $YFeO_3$ ($\varphi = 65°$, $\theta = 8°$).

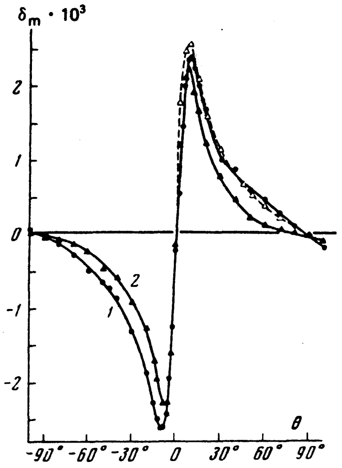

FIG. 4. Meridional intensity effect: 1) α-Fe_2O_3 ($\varphi = 70°$, $\hbar\omega = 2.25$ eV); 2) $YFeO_3$ ($\varphi = 65°$, $\hbar\omega = 2.9$ eV).

lators such as α-Fe_2O_3 and $YFeO_3$ the corresponding angles are $\vartheta = 10°$ and 8°.

The dashed curves in Figs. 1 and 2 are calculated on the basis of Eq. (2) using the components of the tensor $[\epsilon]$ taken from the published work.[3] The experimental and theoretical curves seem to be in good agreement. Some quantitative discrepancy may be explained by the fact that the calculations are based on the data for bulk Ni and Fe single crystals, whereas our results apply to single-crystal films.

Measurements of the meridional intensity effect in spinel ferrite $CoCr_{0.75}Fe_{1.25}O_4$ with the maximum Kerr effect at the He–Ne laser wavelength[4] have revealed also the maximum value of δ_m, which is $\delta_m = 2\%$ for $\varphi = 65°$, $\theta = 4°$, and $\hbar\omega = 1.8$ eV.

Our measurements of the polar intensity effect δ_p were carried out on yttrium orthoferrite, yttrium iron garnet, and bismuth garnet $BiCaVFe_5O_{12}$. Figure 5 shows the dependences $\delta_p(\omega)$ obtained in the wavelength range from 1.8 to 3.5 eV for light incident at the angle of $\varphi = 65°$ on $YFeO_3$ ($\theta = 8°$, curve 1), $Y_3Fe_5O_{12}$ ($\theta = 4°$, curve 2), and $BiCaVFe_5O_{12}$ ($\theta = 6°$, curve 3). The peaks of these curves represent the first intense optical transitions in the near ultraviolet. Spin-forbidden and frequently also parity-forbidden transitions between the levels of the 3*d* ion in an internal crystal field lie in the visible and infrared range. Consequently, the reflection magneto-optic effects in these crystals are negligible in the $\hbar\omega < 1.8$ eV range. The dependences $\delta_p(\theta)$ for light of fixed wavelength incident at the angle of $\varphi = 65°$ on $YFeO_3$ ($\hbar\omega = 2.8$ eV, curve 1), $Y_3Fe_5O_{12}$ ($\hbar\omega = 2.9$ eV, curve 2), and $BiCaVFe_5O_{12}$ ($\hbar\omega = 2.55$ eV, curve 3) are plotted in Fig. 6. We can see that in the case of these ferromagnetic insulators the polar intensity effect depends strongly on the angle of polarization and is fairly large. For example, in the case of the bismuth garnet the value of δ_p reaches 3% at $\hbar\omega = 2.55$ eV for $\theta = 6°$. The dashed curves in Figs. 5 and 6 are the dependences calculated using the published data.[5] We can see that the agreement between the experimental and theoretical values of δ_p is fully satisfactory.

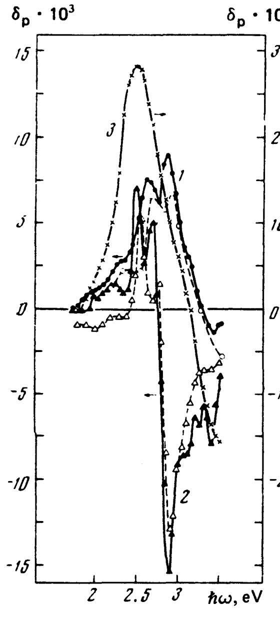

FIG. 5. Polar intensity effect ($\varphi = 65°$): 1) $YFeO_3$ ($\theta = 8°$); 2) $Y_3Fe_5O_{12}$ ($\theta = 4°$); 3) $BiCaVFe_5O_{12}$ ($\theta = 6°$).

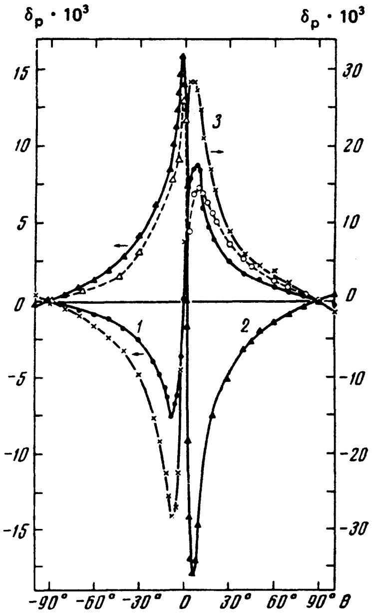

FIG. 6. Polar intensity effect ($\varphi = 65°$): 1) $YFeO_3$ ($\hbar\omega = 2.85$ eV); 2) $Y_3Fe_5O_{12}$ ($\hbar\omega = 2.9$ eV); 3) $BiCaVFe_5O_{12}$ ($\hbar\omega = 2.5$ eV).

The investigated ferromagnetic insulators form a class of new materials used in devices based on magnetic bubble domains. In view of this, the new magnetooptic effects with their unusual dependence on the angle of polarization of the incident of light may be useful in some applications. Modulation of the plane of polarization by just a few degrees (for example, by ±4° in the case of $Y_3Fe_5O_{12}$) makes it possible to investigate in detail the domain structure, magnetization processes, and magnetic anisotropy of ferromagnetic insulators without destroying the magnetic state of the sample, and also to study the nature and structure of bubble domains.

[1] G. S. Krinchik and E. E. Chepurova, Tr. Mezhd. konf. po magnetizmu, M., 1973 (Proc. Intern. Conf. on Magnetism), Nauka, M., 1974, p. 134.

[2] G. S. Krinchik and O. M. Benidze, Zh. Eksp. Teor. Fiz. **67**, 2180 (1974) [Sov. Phys. JETP **40**, 1081 (1975)].

[3] I. I. Sasovskaya and M. M. Noskov, Fiz. Met. Metalloved. **32**,

723 (1971); G. S. Krinchik and V. A. Artem'ev, Zh. Eksp. Teor. Fiz. **53**, 1901 (1967) [Sov. Phys. JETP **26**, 1080 (1968)].

[4] G. S. Krinchik, A. P. Khrebtov, A. A. Askochenskiĭ, E. M. Speranskaya, and S. A. Belyaev, Zh. Eksp. Teor. Fiz. **72**, 699 (1977) [Sov. Phys. JETP **45**, 366 (1977)].

[5] F. J. Kahn, P. S. Pershan, and J. P. Remeika, Phys. Rev. **186**, 891 (1969); S. Wittekoek, T.J. A. Popma, J. M. Robertson, and P. F. Bongers, Phys. Rev. B **12**, 2777 (1975).

Translated by A. Tybulewicz

Direct observation of nuclear magnetic resonance in a rotating coordinate system

A. E. Mefed and V. A. Atsarkin

Institute of Radio Engineering and Electronics, USSR Academy of Sciences

(Submitted 25 August 1977)
Zh. Eksp. Teor. Fiz. **74**, 720–733 (February 1978)

A method for the direct observation of nuclear magnetic resonance in a rotating coordinate system is proposed and realized. The method is based on registering the longitudinal (relative to the constant magnetic field) component of the nuclear magnetization at the Larmor frequency of the spin precession in an effective magnetic field. It is shown that this method has sufficiently high sensitivity and can be used to improve the resolution and to measure NMR chemical shifts in solids, without the use of coherent pulse sequences or Fourier transformation of the output signal. The spectra of the nuclear magnetic resonance in a rotating coordinate system are obtained from the ^{19}F nuclei in CaF_2 crystals, from the protons in water, and others. In CaF_2, under the conditions of the "magic" angle, a narrowing of the nuclear magnetic resonance line by a factor of 50 has been obtained, and the spin-temperature saturation theory has been confirmed. The chemical shifts were resolved for CaF_2, BaF_2, MgF_2, and CdF_2. Further possibilities of the method for the investigation of spin dynamics and structure analysis in solids are analyzed.

PACS numbers: 07.58.+g, 76.60.Cq

1. PRINCIPLE OF DIRECT OBSERVATION OF NUCLEAR MAGNETIC RESONANCE IN A ROTATING COORDINATE SYSTEM

Considerable progress has been made recently in spectroscopy of nuclear magnetic resonance (NMR) in solids, as a result of investigations of the dynamics of nuclear spin systems in strong high-frequency magnetic fields. Thus, the sensitivity of NMR signal registration has been increased by several orders of magnitude; exceedingly slow atomic and molecular motions have been detected; methods have been developed for the suppression of nuclear dipole interactions, and high-resolution spectra were obtained from solids (see, e.g., [1-3]). In all these cases, the motion of the nuclear magnetic moments must be considered in the so-called rotating coordinate system (RCS).[4-5]

Let a solid sample containing nuclear spins I be placed in a constant magnetic field H_0 directed along the z axis and in a high-frequency field $2H_1\cos\omega t$ perpendicular to it, with $\omega\approx\omega_0\equiv\gamma H_0$ where γ is the nuclear gyromagnetic ratio. In a coordinate system (RCS) that rotates about the z axis with frequency ω, the spins are acted upon by a static effective magnetic field $H_e=(\Delta^2+H_1^2)^{1/2}$, where $\Delta=H_0-\omega/\gamma$, which is directed at an angle $\theta=\tan^{-1}(H_1/\Delta)$ to the z axis (Fig. 1). At $H_1\gg H_L$, where H_L is the local magnetic field produced by the spin-spin interactions in the laboratory frame (l.s.), the quantization axis Z in the RCS is directed along $\mathbf{H}_e$, i.e., it makes an angle θ with the z axis.[1] The effective spin Hamiltonian in this coordinate system is[4]

$$\mathscr{H}=-\hbar\gamma H_e I_Z+\hat{\mathscr{H}}_{dp}{}^0, \tag{1}$$

where I_Z denotes the Z component of the total spin of the sample, while $\hat{\mathscr{H}}_{dp}^0$ denotes the secular part of the nuclear dipole-dipole ineractions in the RCS, and its value in first-order perturbation theory is

$$\hat{\mathscr{H}}_{dp}{}^0={}^1/_2(3\cos^2\theta-1)\sum_{i<j}B_{ij}(I_{Zi}I_{Zj}-{}^1/_3\mathbf{I}_i\mathbf{I}_j), \tag{2}$$

with

$$B_{ij}=\frac{3\gamma^2\hbar^2}{2r_{ij}^3}(1-3\cos^2\theta_{ij}),$$

$\mathbf{r}_{ij}$ is the radius vector joining the spins I_i and I_j; θ_{ij} is the angle between $\mathbf{r}_{ij}$ and $\mathbf{H}_0$.

The Hamiltonian (1) is analogous to the ordinary spin

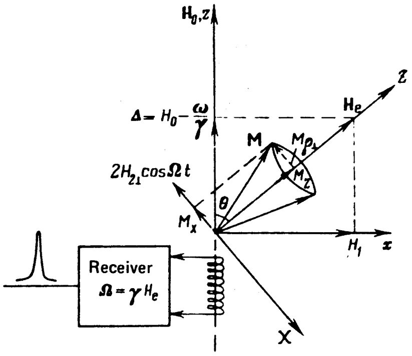

FIG. 1. Geometry of magnetic fields and principle of direct observation of NMR in a rotating coordinate system.

0038-5646/78/4702-0378$02.40