ORIENTATION MAGNETO-OPTIC EFFECT IN NICKEL AND FERROSILICON MONOCRYSTALS

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The recently discovered orientational magneto-optical effect δ_{or} in nickel and ferrosilicon monocrystals is studied. This effect is sharply anisotropic against the background of the isotropic equatorial Kerr effect. The anisotropy is such that δ_{or} cannot be reduced to the Voigt effect. The interpretation of the frequency dependence of δ_{or} is based on the idea that a change in the orientation of the magnetization affects the zone structure of the ferromagnet as a result of spin-orbit interaction.

IN[1] we reported the discovery of a new magneto-optic effect in ferromagnetic metals—a change in the intensity of reflected light that is even with respect to the magnetization. The measurement was made with the geometry of the equatorial Kerr effect (EKE): the magnetization vector I was parallel to the surface of the sample and perpendicular to the plane of incidence of the light. Later, in[2], it was established that this effect, called the orientational magneto-optic effect (OME), is proportional to the square of the component of magnetization that is perpendicular to the plane of incidence of the light. From these first observations it followed that the orientational effect is comparable in order of magnitude with the usual odd EKE, is strongly anisotropic, and is distinguished by an unusual frequency dependence. The hypothesis was put forward that the orientational effect is due to a change in the electronic structure of the ferromagnetic metal during rotation of the magnetization vector on account of the spin-orbit interaction.

In this paper we report on a special study of the anisotropy of OME in monocrystals of nickel and ferrosilicon and of its frequency dependence in the region from 0.17 to 1.5 eV. The study of OME anisotropy has particular value for the elucidation of the question of its physical origin, since, for example, the spin-orbit splitting of degenerate energy bands at a given point in the Brillouin zone changes from zero to a maximum value when the orientation of the vector I in a ferromagnetic crystal changes. Investigation of the frequency dependence of OME is also of great interest. According to the hypothesis mentioned above one should expect for OME a more complex dependence on frequency than for the ordinary Kerr and Faraday magneto-optic effects. This is because electronic transitions with participation of degenerate energy bands are localized in the vicinity of definite lines and points of symmetry of the Brillouin zone; for example, in the face-centered cubic lattice these are the lines Δ, Λ, the points L, X, and a few accidentally degenerate points. This, in turn, opens up the possibility of a reliable identification of specific inter-band transitions and, correspondingly, of a quantitative determination of the band parameters of ferromagnetic metals and alloys.

MEASUREMENT METHOD

Our method consisted of measuring the change in intensity of the p-wave of linearly polarized light ($e \perp I$) reflected from the ferromagnet while it was being magnetized.

The arrangement is shown in Fig. 1. Monochromatization of the light was accomplished by an IKM-1 monochromator (0.15–0.5 eV) and a DMR-4 double monochromator (0.5–1.75 eV) with the appropriate light sources: a Silit resistor and an incandescent lamp. For polarization, a polarizer made up of silver chloride plates with protective layers deposited on them and a Franck-Ritter polarizer were employed. The detectors were a PbS photoresistor (0.5–1.75 eV), liquid-nitrogen cooled InSb and InSn photodiodes (0.23–0.5 eV), and a GeAu photoresistor (0.15–0.3 eV). Under the action of the light flux, two signals are generated in the detection system: a dc current $i_=$ proportional to the intensity of light R_0 reflected by the sample in the absence of a magnetic field, and an ac current $\Delta i_\sim$ proportional to the depth of modulation of light ΔR as a result of the application of the magnetic field. The first signal was registered by a mirror galvanometer G, and the second was taken from the photodetector load and fed into a selective microvoltmeter MV and then a synchronous detector SD. The measured quantities determine the sign and magnitude of the effect, which is characterized by the

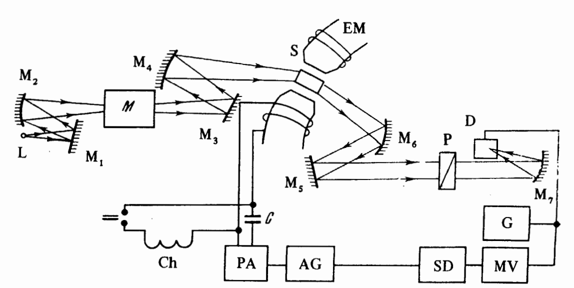

FIG. 1. Schematic of the experimental setup: L–light source, M–monochromator, S–sample, P–polarizer, D–detector, M_1 to M_7–aluminized mirrors, EM–electromagnet, AG–audio generator, PA–power amplifier, C–capacitor, Ch–choke, G–mirror galvanometer, MV–selective microvoltmeter, SD–synchronous detector.

relation $\delta = \Delta R/R_0$. The setup permitted reliable measurements of relative changes in the intensity of the reflected light of the order of 1×10^{-5}.

In this dynamic measurement method, the change in the intensity of the reflected light ΔR occurs upon application to the sample of an ac magnetic field at audio frequency. The periodic magnetic field was created by the electromagnet EM, in whose gap the sample S was placed. The electromagnet is supplied simultaneously with dc and ac currents. The ac component of the field $H_\sim$ was produced by voltage from the audio generator AG and power amplifier PA. To compensate for inductive reactance, a capacitor C is in series with the electromagnet coil. The circuit consisting of the active resistance of the electromagnet coil, its inductance, and the capacitor has a resonant frequency of about 80 Hz. The capacitor also blocks dc from the output transformer of the power amplifier. The choke Ch prevents shorting of the ac component by the dc source. The measurements were made at an angle of incidence $\varphi = 75°$ for FeSi and $\varphi = 80°$ for Ni.

The EKE, which is an odd function of the magnetization, was measured as before,[3,4] by changing the magnetic field from $-H$ to $+H$ ($H = 2700$ Oe), which corresponds to the magnetization of the sample changing from $-I_s$ to $+I_s$ (I_s is the saturation magnetization. However, with such magnetization of the sample, it was not possible to detect the existence of even effects. This was done by "unipolar" magnetization of the sample. By supplying to the coils an ac current $j_\sim = j_0 \sin\omega t$, as well as a dc current $j_= = j_0$ first of one sign and then another, we change the magnetic field from 0 to $+H$ and from $-H$ to 0. The corresponding changes of magnetization from I_r to I_s and from $-I_s$ to $-I_r$, where I_r is the residual magnetization, evokes an effect δ_a in the first case and δ_b in the second. Then,[1] $\delta_{eq} = (\delta_a + \delta_b)/2$ is the EKE effect, and the difference $(\delta_a - \delta_b)/2$ determines the even magneto-optic effect δ_{or} (OME).

The samples were monocrystals of nickel and ferrosilicon cut in the (110) plane and having the shape of a disk 10 mm in diameter and 0.5 mm thick. The Ni sample was cut by the spark method from a rod grown from the melt, and the FeSi (3% Si) sample from a sheet of cold-rolled silicon steel. The samples were polished, first mechanically and then electrolytically. The thickness of the stripped electropolished layer exceeded 100 μ. The EKE and OME measurements were made on the same samples.

EXPERIMENTAL RESULTS

Figure 2 presents the results of measurements of the frequency dependence of EKE, $\delta_{eq}^{[lmn]}$, obtained for monocrystals of nickel and ferrosilicon magnetized along various crystallographic directions. To calculate the quantity $\delta_{s\,eq}^{[lmn]}$ corresponding the magnetization of the sample from a completely demagnetized state to saturation, we found the normalized factors $K^{[lmn]} = I_s/(I_s - I_r)$, which are the coefficients of proportionality between the values $\delta_{eq} = (\delta_a + \delta_b)/2$ and the EKE δ_s values obtained by the usual switching method ($\delta_s = K\delta_{eq}^{[lmn]}$). The average values of K for Ni are 4.0, 6.5, and 1.8 for the [111], [110], and [100] axes,

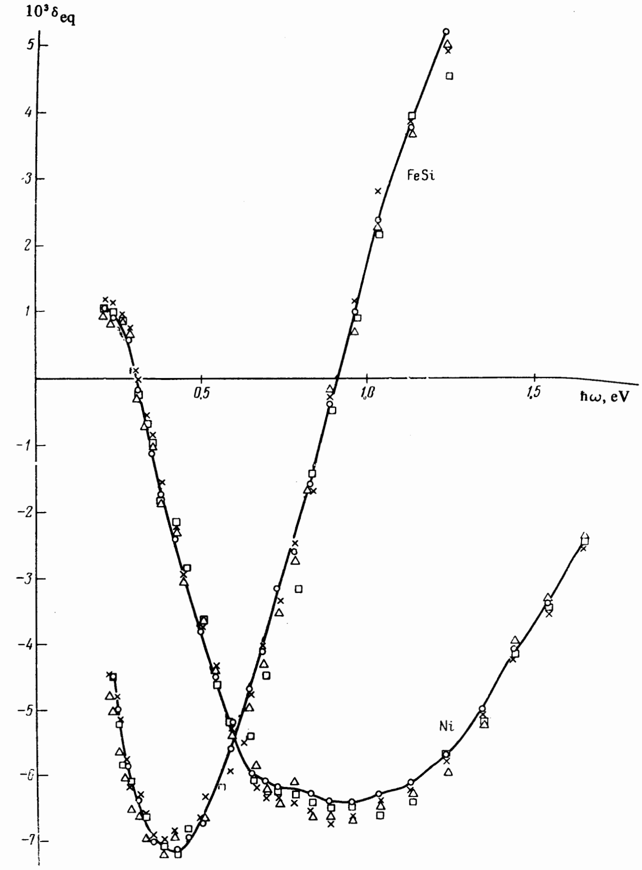

FIG. 2. Equatorial Kerr effect in monocrystals of Ni and FeSi: (X–**H** ∥ [111], △–**H** ∥ [110], □–**H** ∥ [100], ○–δ_s).

respectively. In the last two cases the numerical values of K correspond approximately to rotation of I_s from the [110] and [100] axes to the nearest easy axes [111]. The values of K for FeSi are 1.5, 1.3, and 1.15 for the [111], [110], and [100] axes, respectively. From the values of $\delta_{s\,eq}^{[lmn]}$ in Fig. 2 obtained as described above, it is seen that EKE is isotropic to within 5%. The same result was obtained in measurement of δ_s by the switching method for the different crystallographic directions.

The curves in Fig. 3 represent the frequency dependence of the orientational effect for Ni. Here also the experimental values of $\delta_{or}^{[lmn]}$ were multiplied by $p = I_s^2/(I_s^2 - I_r^2)$, although this ignores the fact that when the magnetization changes from I_s to I_r the vector I passes through different crystallographic axes. From the values of K presented above we find the following values of p: 2.2 for [111], 3.5 for [110], and 1.2 for [100]. In this figure is also given the function $\delta_{or}(\omega)$ for a polycrystalline sample of Ni ($p = 1.1$). Regions where abrupt changes in OME occur can be seen on the curves. There is such a change, with sign reversal, in all the curves, both for mono- and polycrystals, in the region 0.17 to 0.25. In the region near 0.3 eV there is a maximum on the curves for [100] and [110]. The maximum in the [111] curve is shifted somewhat towards the shortwave end and is found at about 0.35 eV. In the interval 0.4 to 0.9 eV, the [100] and [110] curves show a broad maxi-

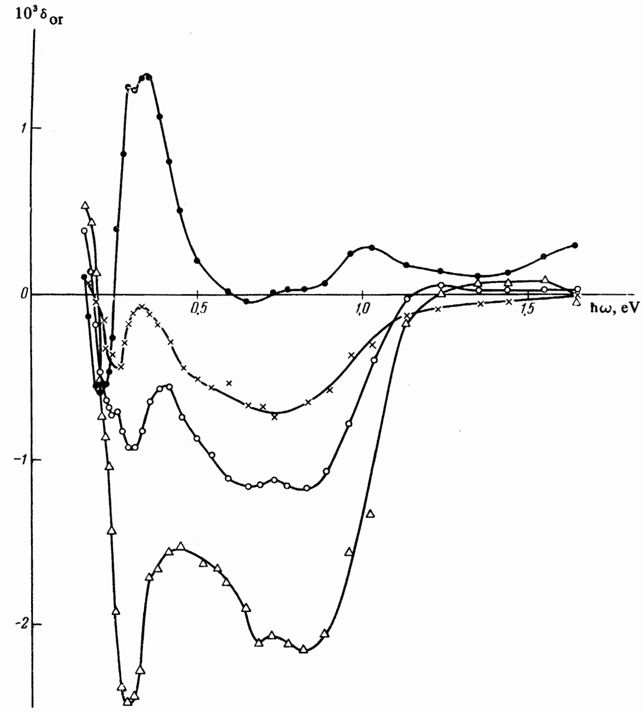

FIG. 3. Orientational magneto-optic effect in Ni: (●—**H** ‖ [111], Δ—**H** ‖ [110], O—**H** ‖ [100], X—polycrystal Ni).

mum with a characteristic break corresponding to the anomaly at 0.8 eV found earlier in EKE measurements.[4] The region 0.9 to 1.2 eV is characterized by a drop in δ_{or} for the [100], [110] axes and for the Ni polycrystal.

In Fig. 4 are the curves for the dependence of OME on the angle α between the direction of the magnetizing field and the crystallographic axis [100] in the (110) plane; these demonstrate the strong anisotropy of the orientational effect with complete isotropy of EKE (within the limits of experimental error). These curves, too, have been normalized by means of the coefficient p.

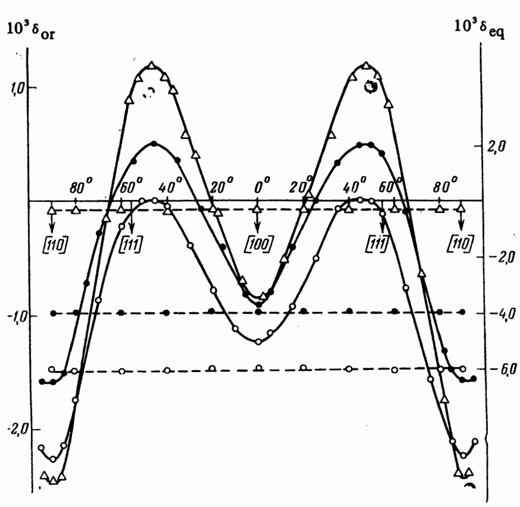

FIG. 4. Dependence of OME (solid curves) and EKE (dashed) for Ni on the direction of the magnetization vector in the crystal for three values of wavelength: (Δ—0.31 eV, ●—0.5 eV, O—0.7 eV).

The function $\delta_{or}(\omega)$ for FeSi with magnetization of the sample along [100] is in Fig. 5. Note, that even for the FeSi monocrystal the orientational magneto-optic effect is comparable to the EKE in magnitude (see Fig. 2).

ANISOTROPY OF THE ORIENTATIONAL MAGNETO-OPTIC EFFECT

Anisotropy of magneto-optic effects in ferromagnetic metals was examined by Donovan and Medcalf.[5] They carried out a calculation according to the Argyres scheme,[6] taking into terms of the second order in the spin-orbit interaction, and showed that in cubic ferromagnetic metals, just as in cubic semiconductors with anisotropic Fermi surfaces, the anisotropy of magneto-optic effects arises as a consequence of second-order effects. The observation of OME corresponds to the Voigt configuration when $\mathbf{e} \perp \mathbf{I}$.

The anisotropy of quadratic magneto-optic effects for the Voigt configuration in the (100) plane was considered in[5]. We shall do the same for the (110) plane of our case. The components of the dielectric permeability tensor, which are quadratic in magnetization,

$$\hat{\varepsilon}^{(2)} = 4\pi\hat{\alpha}^{(2)} + 4\pi\hat{\sigma}^{(2)}/i\omega,$$

where $\hat{\alpha}^{(2)}$ and $\hat{\sigma}^{(2)}$ are the polarizability and conductivity tensors, can be written with respect to arbitrary axes ξ_i in accordance with Eq. (34) of[5] in the following fashion:

$$\varepsilon_{ij}^{(2)} = {}_{i}\beta_k\beta_l(\delta_{ij}\delta_{kl} + \delta_{il}\delta_{jk} + \delta_{ik}\delta_{jl} - 2c_{im}c_{jm}c_{lm}c_{km})\,\varepsilon^{(2)},$$

where β_k are the direction cosines of the magnetization vector in the crystal, c_{ij} is the matrix of transformation from cubic axes x_j to axes $\xi_i = c_{ij}x_j$. The index 2 indicates terms of second order with respect to the spin-orbit interaction.

We introduce the axes ξ_i such that ξ_1 is parallel to **H**, which makes an angle ψ with 001; the axis ξ_3 is parallel to **k** and makes an angle φ with [110]. Then $\beta_1 = 1$, $\beta_2 = \beta_3 = 0$, and the transformation matrix c_{ij} has the form

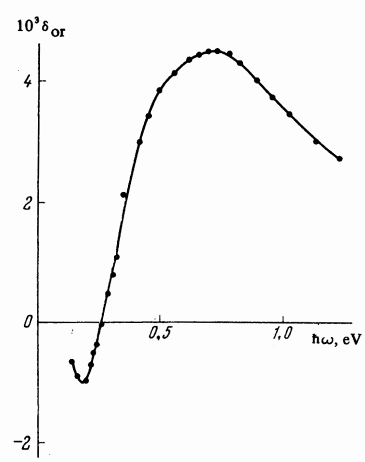

FIG. 5. Dependence of OME on frequency of light for FeSi (**H** ‖ [100]).

$$c_{ij}=\begin{pmatrix} 2^{-1/2}\sin\psi & 2^{-1/2}\sin\psi & \cos\psi \\ 2^{-1/2}(\cos\varphi+\cos\psi\sin\varphi) & -2^{-1/2}(\cos\varphi-\cos\psi\sin\varphi) & -\sin\psi\sin\varphi \\ -2^{-1/2}(\sin\varphi-\cos\psi\cos\varphi) & 2^{-1/2}(\sin\varphi+\cos\psi\cos\varphi) & -\sin\psi\cos\varphi \end{pmatrix}$$

From this we find the angular dependence of the tensor components:

$$\varepsilon_{12}^{(2)}=\varepsilon_{21}^{(2)}={}^1/_2\sin 2\psi\sin\varphi(\cos 2\psi+\cos^2\psi),$$

$$\varepsilon_{11}^{(2)}=\sin^2\psi(1+\cos^2\psi),$$

$$\varepsilon_{22}^{(2)}=\sin^2\psi(\cos^2\varphi+3\cos^2\psi\sin^2\varphi).$$

From these expressions it follows that when $\psi = 0$, i.e., for the case of magnetization of the crystal along [100], all the components $\epsilon_{ij}^{(2)}$ vanish and there is no linear double refraction in the Voigt configuration. This conclusion is completely contradicted by the experimental data we obtained on both Ni and FeSi.

As was mentioned above, in the dynamic method of OME measurements the results pertaining to anisotropy of the effect can evoke some doubt. For example, in measuring OME for Ni along [100] during one period of magnetization, the vector I moves in the (110) plane from the [100] axis to the easy [111] axes and back again, i.e., we are measuring some sort of integral effect. The formation of a 180-degree boundary when the sample is demagnetized can also lead to underestimation of the OME, which is quadratic in magnetization. At the same time, the nonzero value and even the attainment of a maximum value of δ_{or} during magnetization of the sample along [100] have basic significance. Therefore, for a qualitative check of our results, we undertook direct static measurements of the OME anisotropy, using the compensation method. Reorientation of I from a given axis perpendicular to the plane of incidence of the light to a transverse direction was effected by turning the magnet 90°; the change in intensity of the reflected light was then recorded from the mirror galvanometer. This could only be done for $\varphi = 60°$ and with the widest slit on the monochromator, leading to a decrease in the absolute values of δ_{eq} and δ_{or}. The following values were obtained for Ni: $\delta_{eq} = -1.4 \times 10^{-3}$,

$$\delta_{or}^{[100]}=-0.5\cdot 10^{-3},\ \delta_{or}^{[110]}=-0.8\cdot 10^{-3},\ \delta_{or}^{[111]}=$$

$$-0.1\cdot 10^{-3}\ \text{for}\ \hbar\omega=0.73\,\text{eV}$$

and $\delta_{eq}=1.8\cdot 10^{-3},\ \delta_{or}^{[100]}=\delta_{or}^{[110]}=\delta_{or}^{[111]}=0$ for $\hbar\omega=1.1$ eV[1)].

In this the experimental error was $\pm 0.1 \times 10^{-3}$. It is seen that the anisotropy of δ_{or} at $\varphi = 80°$ by the dynamic method agrees qualitatively with that of the static method at $\varphi = 60°$. Analogous measurements for FeSi gave: $\delta_{eq} = -0.7 \times 10^{-3}$,

$$\delta_{or}^{[100]}=1.8\cdot 10^{-3}\quad \delta_{or}^{[110]}=1.5\cdot 10^{-3},$$

$$\delta_{or}^{[111]}=1.6\cdot 10^{-3}\ \text{for}\ \hbar\omega=0.73\,\text{eV}$$

and $\delta_{eq}=1.3\cdot 10^{-3},\ \delta_{or}^{[100]}=2.6\cdot 10^{-3},\ \delta_{or}^{[110]}=1.7\cdot 10^{-3},$

$$\delta_{or}^{[110]}=2.8\cdot 10^{-3}\ \text{for}\ \hbar\omega=1.1\,\text{eV}.$$

We said that our dynamic method for FeSi yields values of δ_{or} for the [111] and [110] axes that are too low compared to those for [100]; this is evidently due to the formation of 180-degree domain boundaries when FeSi is demagnetized. Hence in Fig. 5 we give the curve only for [100], in order to show the dependence of δ_{or} on frequency.

Thus, the experimentally observed even change of the intensity of reflected light is comparable in order of magnitude to the usual EKE. And, the OME does not reduce to the Voigt effect in cubic crystals as is obtained in theory with account taken of terms to the second order with respect to the spin-orbit interaction,[5] since the change in the intensity of reflected light takes place with I along [100]. We suggest that OME is associated with the effect of the spin-orbit interaction on the band structure of the ferromagnetic metal when the orientation of the magnetization vector changes, and that the observed OME anisotropy can be explained by taking into account the local symmetry of interband transitions in the vicinity of those points and lines of symmetry where the degeneracy of the energy bands is lifted by the spin-orbit interaction.

FREQUENCY DEPENDENCE OF THE ORIENTATIONAL MAGNETO-OPTIC EFFECT

In this section we shall attempt to correlate the experimentally observed peculiarities on the $\delta_{or}(\omega)$ curves with the interband transitions with participation of spin-orbitally degenerate bands on the basis of existing models for the electronic structure of ferromagnetic nickel.

Variant I.—On the basis of the model for the electronic structure of ferromagnetic nickel with inverted order of levels proposed and discussed in[4,7], the broad maximum in the $\delta_{or}^{[110]}$ and $\delta_{or}^{[100]}$ curves in the region 0.4 to 1.1 eV can be identified with a transition from $\Lambda_1\downarrow$ to $\Lambda_3\uparrow$, a band split by spin-orbit interaction. In Fig. 6 this region corresponds to transitions from B to A. The maxima on the $\delta_{or}^{[100]}$, $\delta_{or}^{[110]}$, and $\delta_{or}^{[111]}$ curves in the region 0.3 to 0.35 eV, as well as the complex behavior of $\delta_{or}(\omega)$ in the longer wavelength region are due to transitions C($\hbar\omega \approx 0.25$ eV) and D($\hbar\omega \approx 0.4$ eV).

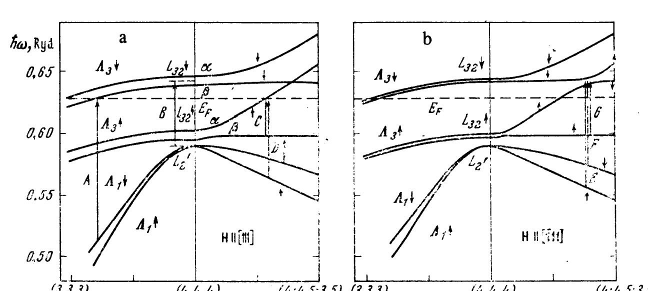

FIG. 6. Model of the band structure of Ni in the vicinity of the point L (4, 4, 4) (in units of $2\pi/a$) for different orientations of the magnetic field H. [9] a—H parallel to [111]; b—H parallel to [$\bar{1}$11]. Values of the energy $\hbar\omega$ are given in Rydbergs.

[1)]The static measurements permitted direct establishment of the sign of the magneto-optic effects. In all the curves presented in this paper, a positive sign corresponds to an increase, and negative to a decrease, in the intensity of the reflected light in going from a demagnetized to a magnetized state, i.e., when the vector I switches from the plane of incidence of the light to a perpendicular direction.

Variant II.—Falicov and Ruvalds,[8] and Zornberg[9] considered in detail the question of spin-orbit lifting of degeneracy arising in the crossing of bands with opposite spin directions. Using the figure from[9], we show in Fig. 6 a diagram for such a lifting of degeneracy of the bands $L_{32\alpha}\uparrow$ and $L_{32\beta}\downarrow$ with a change in orientation of the magnetization vector for the model with inverted order of levels.[4,7,9] Taking into account this new possibility of an effect of the change of orientation of the magnetization vector on the band structure of a ferromagnetic metal and, correspondingly, the new possibility for the origin of OME, let us consider the following identification of features on the δ_{or} curves with the interband transitions B, E, F, and G:

The maximum on the $\delta_{or}^{[100]}$ and $\delta_{or}^{[110]}$ curves in the region 0.7 to 0.9 eV can be connected with the transition F, and the maximum on $\delta_{or}^{[111]}$ in the region of 1.0 eV with the transition E. This identification is also based on additional low-temperature measurements. In Fig. 7 are the EKE curves obtained with an orientation of the magnetic field along the three principal crystallographic directions, as well as the $\delta_{or}^{[100]}$ curve for $T = 80°K$. Significance should not be attached to the absolute values of the effect, since the measurements were made in a cryostat on a nickel platelet, as a consequence of which the demagnetization factor for the different crystallographic directions was not the same. Normalization of $\delta_{or}^{[100]}$ by the coefficient p was not done, either.

In Fig. 7 it is seen that the maximum in $\delta_{or}^{[100]}$ at 0.7 to 0.85 eV divides more sharply at the lower temperature. The center of this anomaly, a dip at 0.76 eV, coincides exactly with a corresponding dip in the δ_{eq} curves, leading to the characteristic two-component structure of the feature.[4,7] For the transition F this dip corresponds to a minimum of the interband density of states arising on account of the spin-orbital splitting of the band $L_{32\alpha}\uparrow$ at the place where it intersects the band $L_{32\beta}\downarrow$. If Variant I is correct, we would

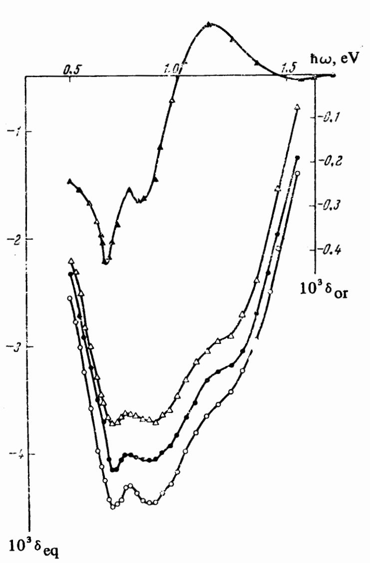

FIG. 7. EKE and OME in a monocrystal of Ni at 80 K ($\varphi = 70°$): O–$\delta_{eq}^{[111]}$, ●–$\delta_{eq}^{[100]}$, △–$\delta_{eq}^{[110]}$, ▲–$\delta_{or}^{[100]}$).

have to expect an anisotropy in the magnitude of the splitting of this two-component structure in the δ_{eq} curve, i.e., a shift of the components A_1 and A_2[4,7] Experimentally, however, we observe only a change in shape of the line, which is evidence for Variant II. In addition, according to Zornberg's calculations,[9] transition A is shifted toward shorter waves. On the basis of all this, we take $\hbar\omega_F = 0.76$ eV. On the $\delta_{or}^{[100]}$ (Fig. 7) a maximum appears also at 1.1 eV, which gives $\hbar\omega_E = 1.1$ eV. The low-frequency maxima on the δ_{or} curves in the region of 0.3 eV can be identified with the transitions B and G. For definiteness, we take $\hbar\omega_B = 0.35$ eV (the maximum on the [111] curve), and $\hbar\omega_G = 0.3$ eV (the maximum on the [100] and [110] curves). The spin-orbit interaction affects the transitions C and D more weakly, although it is possible that the low-energy maximum on the [111] curve at 0.2 eV may be due to C transitions.

The Variant II identification of interband transitions does not contradict the models of the electronic structure of Ni with inverted level order,[4,7] if the calculations of Zornberg[9] are also taken into account. For example, for set VII band parameters,[9] the energies of the B, C, E, F, and G transitions are respectively 0.4, 0.25, ~1, ~0.6, and 0.38 eV.

From what we have said, it is obvious that to carry out a reliable identification of the interband transitions it is necessary to construct a theory of OME (especially of its anisotropy), relating it to the local symmetry of specific interband transitions. In this case, taking into account the enhancement of a given feature when measuring along specific crystallographic directions and correlating the intensities and signs of the features would permit a reliable check of the validity of any particular identification variant. The above considerations are presented in order to show some of the possibilities offered by the study of the frequency dependence of OME for deciphering the band structure of ferromagnetic metals and alloys. We point out only the following interesting circumstance. If identification of any feature on the $\delta_{or}(\omega)$ curves with the frequency $\hbar\omega_G$ turns out to be possible, then we obtain a direct spectral method for determining the magnitude of the exchange splitting of the d band of a ferromagnetic metal ΔE_d. It is clear from Fig. 6, even though we have to do in this case with transitions between subbands with the same sign of spin $L_{32\beta}\uparrow$ and $L_{32\alpha}\uparrow$, the frequency

$$\hbar\omega_G = E(L_{32\beta}\downarrow) - E(L_{32\beta}\uparrow) = \Delta E_d,$$

since the transition goes in the region of crossing of the bands $L_{32\alpha}\uparrow$ and $L_{32\beta}\downarrow$.

In conclusion, we remark that some preliminary experiments we have done show up some characteristic features on the $\delta_{or}(\omega)$ curves also in the region of 2.5 eV, where, according to magneto-optic observations,[4] there is an anomaly associated with the transitions $X_5\uparrow \rightarrow X_4'\uparrow$, i.e., with transitions from the spin-orbitally degenerate band X_5.

We thank Dr. Zornberg for a preprint of his paper[9].

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26