Quadratic magnetooptical reflection effects in ferromagnets

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Low-temperature measurements were made of the equatorial Kerr effect (EKE) and of the orientational magnetooptical effect (OME) in Ni single crystals cut in different crystallographic planes, in the energy region 0.2–1.8 eV. We studied the anisotropy and established the phenomenological law governing the OME anisotropy. An anisotropic fine structure due to spin-orbit splitting of the 3*d* bands, was observed for the anomalies at 0.57 and 0.7 eV. A number of band parameters of Ni are determined within the framework of a model with a reversed sequence of levels.

Krinchik, Gushchin, and Chepurova[1,2] made the first experimental observation of the magnetooptical effect that is quadratic in the magnetization in light reflected from a ferromagnet. Study of quadratic magnetooptical effects is of interest from two points of view. First, this is an additional promising method of investigating the electron energy spectrum of ferromagnets[1,3,4]. Second, joint study of effects that are even in the magnetization with the usual odd magnetooptical effects makes it possible better to understand and describe the crystal-optics features of magnetically ordered crystals. It is also of interest to trace the common features and differences of quadratic magnetooptical effects of reflection and quadratic effects in transmitted light, which were investigated with transparent ferrodielectrics in[5,6].

We report here the results of measurements of the equatorial Kerr effect (EKE) that is linear in the magnetization and the quadratic orientational magnetooptical effect (OME) on Ni single crystals cut along different crystallographic planes, in the energy range 0.2–1.8 eV at T = 295, 80, and 15°K. The singularities observed on these curves are used to determine the band parameters of ferromagnetic Ni. We present also a phenomenological analysis of the theory of quadratic magnetooptical effects in reflected light and establish a law of anisotropy of the OME.

We used for the measurements the magnetooptical setup described by Krinchik, Gan'shina, and Gushchin[3]. The measurements of the EKE were carried out with bipolar magnetization reversal, and the measurements of the OME were made with unipolar magnetization of the sample, such that the magnetization vector I was rotated from a given crystallographic axis to the light axes. To obtain an effect corresponding to rotation of I through 90° we introduced, just as in[3], a normalization factor $P = I_S^2/(I_S^2 - I_r^2)$, where I_S is the saturation magnetization and I_r is the residual magnetization.

PHENOMENOLOGICAL THEORY OF QUADRATIC MAGNETOOPTICAL EFFECTS IN REFLECTION OF LIGHT

The magnetooptical properties of a gyroelectric medium can be described phenomenologically by introducing the dielectric tensor ϵ ($\epsilon_{xx} = \epsilon_{yy} = \epsilon$; $\epsilon_{zz} = \epsilon_0$; $\epsilon_{xy} = -\epsilon_{yx} = -i\epsilon'$; $\epsilon_{xz} = \epsilon_{zx} = \epsilon_{yz} = \epsilon_{zy} = 0$; the magnetization vector I is parallel to z), with two magnetooptical parameters M and f, the values of which determine all the magnetooptical effects[7]. Introduction of the magnetooptical parameter $M = \epsilon'/\epsilon$ is sufficient for the description of effects that are linear in the magnetization (the Faraday effect, the Kerr effect), but when quadratic magnetooptic effects are considered one introduces a second magnetooptical parameter $f = \epsilon_0^{-1}(\partial^2\epsilon/\partial M^2)_{M=0}$, which determines the change of the diagonal components of the tensor ϵ.

Let us consider the magnetooptical effects that are produced when light is reflected from a gyroelectric medium in the case of equatorial magnetization (I lies in the plane of the sample and is perpendicular to the plane of incidence of the light). The Fresnel formula for the reflection coefficient of the p-component of linearly polarized light (E_p lies in the plane of incidence of the light) is written in the form[8]

$$R_p/A_p = (\alpha n^* - \alpha^* + i\beta^* M)/(\alpha n^* + \alpha^* + i\beta^* M), \tag{1}$$

where A_p and R_p are the amplitudes of the incident and reflected waves, respectively; α and α' are the direction cosines of the wave normal in vacuum and in the medium: $\alpha = \cos\varphi$, $\beta = \sin\varphi$, $\alpha^* = \cos\psi$, $\beta^* = (\sin\varphi)/n^*$; φ is the angle of incidence of the light; ψ is the angle of refraction.

The expression for the refractive indices in the case of transverse propagation of light $k \perp I$ can be expressed in the form

$$n_\perp^{*2} = \varepsilon\mu_0(1-M^2) = n_0^2[1+(1-f)M^2], \qquad n_z^{*2} = \varepsilon_0\mu_0, \tag{2}$$

where $n_0 = f(\epsilon_0\mu_0)^{1/2}$ is the refractive index for a demagnetized medium ($I_S = 0$), while n_I^* and n_Z^* are the refractive indices for light waves with electric-vector oscillations perpendicular and parallel to I, respectively. Using expressions (1) and (2), we obtain a formula for the change of the intensity J of the reflected light upon magnetization of a ferromagnetic medium:

$$\delta = (J - J_0)/J_0 = \delta^{(1)} + \delta^{(2)}; \tag{3}$$

$$\delta^{(1)} = \delta_{eq} = \mathrm{Re}\,(4i\alpha\beta M/(1-\alpha^2 n_0^2)), \tag{4}$$

$$\delta^{(2)} = \mathrm{Re}\left(\frac{4\alpha\Delta n_\perp}{\alpha^2 n_0^2 - 1} + \frac{2\beta^2 M^2}{n_0^2(1-\alpha^2 n_0^2)}\right). \tag{5}$$

Formulas (4) and (5) are written in the approximation $\alpha^* = \cos\psi = 1$, which is valid for metals in the infrared region.

The obtained expressions can be generalized to include the case of cubic crystals, in which the change of the refractive index upon magnetization, for a light wave with a specified direction of oscillations of the vector E, depends on the orientation of the vector I relative to the crystallographic axes. The resultant change Δn in the case of transverse propagation of light can be determined on the basis of the Akulov anisotropy law for even effects in cubic ferromagnets[9]:

$$\Delta n_{\alpha_i\beta_i} = {}^3/_2(\Delta n_{\parallel})_{100}\{\alpha_1^2\beta_1^2+\alpha_2^2\beta_2^2+\alpha_3^2\beta_3^2-{}^1/_3\} + 3(\Delta n_{\parallel})_{111}\{\alpha_1\alpha_2\beta_1\beta_2+\alpha_2\alpha_3\beta_2\beta_3+\alpha_1\alpha_3\beta_1\beta_3\}, \quad (6)$$

where α_i and β_i are respectively the direction cosines of I and E, $(\Delta n_{\parallel})_{100}$ and $(\Delta n_{\parallel})_{111}$ are the changes of n for the light wave with $\mathbf{E} \parallel \mathbf{I}$ for the axes [100] and [111].

A formula for the change in the intensity of the reflected light upon magnetization of a cubic ferromagnetic in an arbitrary direction can be obtained by substituting $\Delta n_{\perp}$ from (6) in (5).

Analogously, the change of the light intensity for the s wave (vector $\mathbf{E}_S$ perpendicular to the plane of incidence of the light) is expressed in the same approximation, cos ψ = 1, in the form

$$\delta_s^{(2)} = \mathrm{Re}\left(\frac{4\alpha\Delta n_{\parallel}}{\alpha^2-n_0^2}\right). \quad (7)$$

ANISOTROPY OF OME

Study of the anisotropy of the magnetooptical effects on Ni single crystals has shown that the EKE is isotropic or, more accurately speaking, weakly anisotropic in the first-order approximation, and that the orientational magnetooptical effect (δ_{or}) depends strongly on the orientation of the vector I relative to the crystallographic axes (see Figs. 1 and 2).

The change quadratic in I in the intensity of the p wave of the reflected light upon rotation of the magnetization vector is expressed using the first term in formula (5) in the following form:

$$\delta_{or} = [(\Delta n_{\perp}) - (\Delta n_{\parallel})] f(n_0, \varphi), \quad (8)$$

where $\Delta n_{\perp}$ and $\Delta n_{\parallel}$ are the corresponding changes in the refractive index in the cases of equatorial ($\mathbf{E}_p \perp \mathbf{I}$) and meridional ($\mathbf{E}_p \parallel \mathbf{I}$) magnetization, the expression $f(n_0, \varphi)$ does not depend on the magnetization, and we neglect the small isotropic increment due to the second term in formula (5), since estimates have shown that this increment does not exceed 10^{-5} for Ni.

In measurement of the OME for the [001] axis in the plane (100), the vector I becomes reoriented from one cubic axis [001] to another [010], in which case δ_{or} is proportional to the birefringence produced when the magnetization vector is oriented along the cubic axis. The presence of δ_{or} in this case proves that the observed OME anisotropy cannot be understood on the basis of the Argyres theory when account is taken of second-order terms in the spin-orbit interaction in the wave functions, inasmuch as, according to Donovan and Medcalf[10], there is no linear birefringence when a cubic crystal is magnetized along the cubic axis.

We have also estimated the isotropic contribution made to the quadratic magnetooptical effect by the existence of off-diagonal dielectric tensor components ϵ' linear in I. As follows from (2), at $\epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz} = \epsilon_0$, i.e., at f = 0, we have

$$\Delta n = n_{\perp} - n_{\parallel} = -M^2/2n_0. \quad (9)$$

The results of the calculation of $\delta^{(2)}$ by formula (5) with Δn from (9) are given in Fig. 3. The values of n, k, and ϵ_1', ϵ_2' were taken the same as in[11]. The obtained isotropic contribution is insufficient to explain the observed effect for the [001] axis in the (100) plane.

To obtain a phenomenological law of OME anisotropy, we specify the magnetization direction in the crystal relative to the crystallographic axes in terms of the direction cosines α_i and γ_i for the equatorial and meridional cases, respectively, and rewrite (8) in the form

$$\delta_{or(l,m,n)}^{[\alpha_i]} = [\Delta n_{\alpha_i\gamma_i} - \Delta n_{\gamma_i\gamma_i}] f(n_0, \varphi), \quad (10)$$

where $[\alpha_i]$ and $[\gamma_i] \in (l, m, n)$.

Substituting now Δn from (6) in (10), we obtain an expression for the change δ_{or}, quadratic in the magnetization, of the intensity of the reflected light that is valid when the OME is measured in any crystallographic direction, namely when the vector I is reoriented from the direction α_i to the direction γ_i:

$$\delta_{or(l,m,n)}^{[\alpha_i]} = -\delta_{or(100)}^{[001]}\{\gamma_1^2(\alpha_1^2-\gamma_1^2)+\gamma_2^2(\alpha_2^2-\gamma_2)+\gamma_3^2(\alpha_3^2-\gamma_3^2)\} - 2\delta_{or(100)}^{[110]}\{\gamma_1\gamma_2(\alpha_1\alpha_2-\gamma_1\gamma_2)+\gamma_2\gamma_3(\alpha_2\alpha_3-\gamma_2\gamma_3)+\gamma_1\gamma_3(\alpha_1\alpha_3-\gamma_1\gamma_3)\}, \quad (11)$$

where the coefficients

$$\delta_{or(100)}^{[001]} = -{}^3/_2(\Delta n_{\parallel})_{(100)} f(n_0, \varphi), \qquad \delta_{or(100)}^{[110]} = -{}^3/_2(\Delta n_{\parallel})_{(111)} f(n_0, \varphi)$$

correspond to the OME measured for the axes [001] and [110] in the (100) plane.

We present some particular cases of the application of formula (11):

$$\delta_{or(110)}^{[001]} = {}^1/_2[\delta_{or(100)}^{[001]} + \delta_{or(100)}^{[110]}],$$

$$\delta_{or(110)}^{[1\bar{1}0]} = \delta_{or(100)}^{[001]}, \qquad \delta_{or(110)}^{[112]} = \delta_{or(100)}^{[110]},$$

$$\delta_{or(111)}^{[\alpha_i]} = {}^1/_3\delta_{or(100)}^{[001]} + {}^2/_3\delta_{or(100)}^{[110]}.$$

Figures 1 and 2 show results of measurements of the OME anisotropy in three crystallographic planes. The dashed curves calculated from formula (11) show satisfactory agreement between the phenomenological theory of the OME anisotropy and experiment.

For quadratic magnetooptical effects with transmitted light, a formula analogous to (6) was obtained in[5] on the basis of allowance for the influence of the magnetization on the optical indicatrix of the material. However, the main difference between quadratic transmission effects and reflection effects is that for the

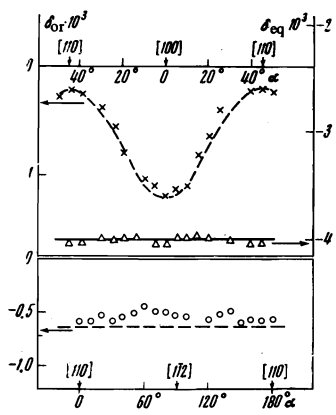

FIG. 1

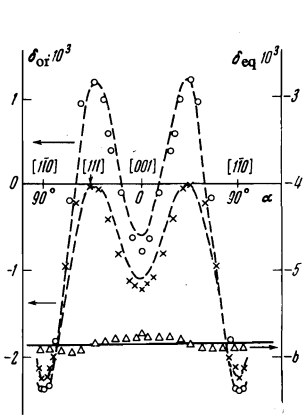

FIG. 2

FIG. 1. Magnetooptical effects vs. the direction of the magnetization vector in the crystal for the crystallographic planes: X–OME for (100), $\hbar\omega$ = 0.7 eV, φ = 70°; ○–OME for (111), $\hbar\omega$ = 0.5 eV, φ = 80°; △–EKE. Dashed curves–values of the OME calculated from formula (11).

FIG. 2. Dependence of the magnetooptical effect on the direction of the magnetization vector in the (110) plane for two wavelengths: ○–0.31 eV, X–0.7 eV, dashed–value of OME calculated from formula (11); △–EKE, $\hbar\omega$ = 0.7 eV, φ = 80°.

former no dependence of the effect on the wavelength of light has as yet been observed[5,6], whereas a strong frequency dependence is observed for the latter[3].

Parker[12] has proposed a magnetostriction mechanism to explain the anisotropy of quadratic magnetooptical effects in Ni. To estimate the contribution of this mechanism to the OME, it is necessary to measure the piezooptical coefficients for ferromagnetic metals directly. An analogous magnetoelastic contribution to quadratic effects in ferrodielectrics turned out to be smaller by two orders of magnitude than the observed effects[6,13]. We propose that the OME for nickel is due mainly to the influence of the spin-orbit interaction on the band structure when the orientation of the vector I is changed[3,4].

FREQUENCY DEPENDENCE OF THE OME AND THE EKE AND DETERMINATION OF THE BAND PARAMETERS OF Ni

Figures 3—5 show the results of measurements of the OME and EKE frequency dependences for different crystallographic directions at T = 295, 80, and 15°K. It is possible to distinguish on these curves a number of anomalies (0.3, 0.57, 0.77, 1.1 eV), the structure of which becomes more pronounced when the sample temperature is lowered. Simultaneous study of the singlarities in the spectra of the OME and the EKE at low temperatures allows us to propose a variant for identifying the interband transitions in the band-structure model for ferromagnetic nickel with inverted order of the levels[14], based only on magnetooptical measurements on single-crystal Ni. In[14] we used the 0.4 eV maximum obtained in the thermal reflection spectrum of a polycrystalline-film Ni sample[15] for identification of the transition B (see Fig. 6).

The low-temperature OME curves (Figs. 3 and 4) reveal a sharply pronounced maximum in the region of 0.3 eV, which was identified by us with the transition $G(L_{32\beta}^{\uparrow} \rightarrow L_{32\alpha}^{\uparrow})$ in the region of intersection of the bands $Q_+^{\uparrow}$ and $Q_-^{\uparrow}$ (Fig. 6). The position of this maximum determines the exchange splitting of the 3d bands of ferromagnetic Ni. The positive maximum of the OME at $\hbar\omega = (1.1 \pm 0.05)$ eV will be identified with the transition E, which is of similar character.

An investigation of the EKE spectra at T = 15°K with better spectral resolution $\Delta\hbar\omega = 0.005$ eV has revealed an anisotropic fine structure of the singularities at 0.55 —0.59 and 0.67—0.9 eV.

On the EKE curve at I ∥ [111] (Fig. 5) in the region 0.57 eV there are observed two maxima: at $\hbar\omega(\beta_1) = (0.55 \pm 0.005)$ eV and $\hbar\omega(\beta_2) = (0.59 \pm 0.005)$ eV. The splitting of this transition by an amount $\Delta = (0.04 \pm 0.01)$ eV can be related to the spin-orbit splitting of the level $L_{32}^{\uparrow}$, and the observed singularity can be identified with the transition $B(L_{2'}^{\uparrow} \rightarrow L_{32}^{\uparrow})$. The aniso-

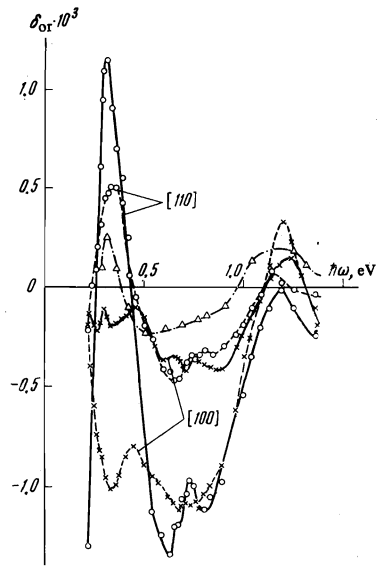

FIG. 3. Orientational magnetooptical effect for Ni single crystal in the (001) plane. Solid curves—T = 80°K, dashed—T = 295°K, ○—[110], X—[100], dash-dot—OME calculation by formula (5); light-incidence angle $\varphi = 70°$.

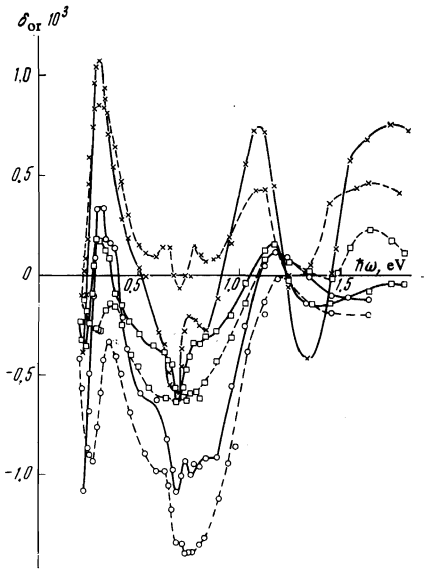

FIG. 4. Orientational magnetooptical effect for Ni in (100) plane. Solid curves—T = 80°K, dashed—T = 295°K; X—H∥[111]; □—H∥[001]; ○—H∥[1$\bar{1}$0]; $\varphi = 70°$.

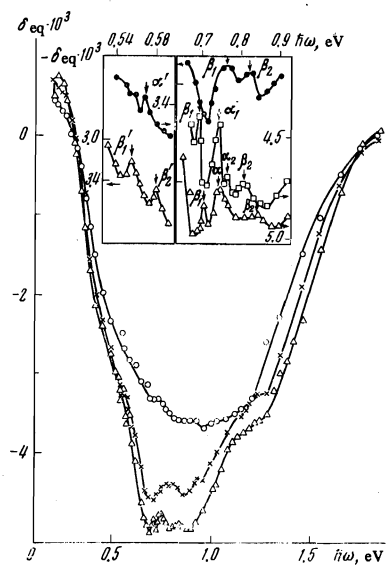

FIG. 5

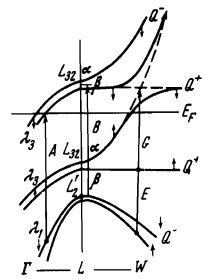

FIG. 6

FIG. 5. Equatorial Kerr effect at different temperatures: ○—295°K, X—80°K, △—15°K at H∥[111]. The insert shows the EKE for different crystallographic directions: △—H∥[111]; □—H∥[1$\bar{1}$0]; ●—H∥[001]; T = 15°K, $\varphi = 70°$.

FIG. 6. Energy level scheme in the vicinity of the point L.

tropic fine structure of the 0.57-eV anomaly was also observed by Stoll[16], who identified this anomaly with the transition G.

The complex structure of the EKE in the region 0.8 eV is interpreted by us as the result of spin-orbit splitting of the edge of the interband transition $\lambda_1^{\downarrow} \rightarrow E_F$ (transition A), the splitting of which must be increased by a factor $\mathfrak{M}$ in comparison with the splitting of the transition B, where $M = m_{L_{32}^{\downarrow}}/m_{L_{2'}^{\downarrow}}$.

In the presence of a magnetic field and with spin-orbit interaction taken into account, the eight L points of the Brillouin zone become nonequivalent. In the discussion of the fine structure we shall use the following symbols: the fine-structure components corresponding to the maximum splitting of the levels will be designated by β, and those corresponding to the minimum splitting by α. With I directed along the [111] axis, for the two L points located along this axis we shall have a maximum splitting of the levels L_{32} and λ_3, and for the remaining six L points the splitting is one-third as large. With I directed along the [100] axis, all L points will be equivalent, and the splitting of the L_{32} and λ_3 levels amounts to $1/\sqrt{3}$ of the maximal splitting[17]. The change of the fine structure of the anomalies at different orientations of I qualitatively confirms the proposed identification. At the orientation $I \parallel [111]$, with transitions to the spin-orbit-split level $\lambda_3^{\downarrow}$, we set the following maxima in correspondence with the nonequivalent L points: $\hbar\omega(\beta_1) = (0.71 \pm 0.005)$ eV and $\hbar\omega(\beta_2) = (0.84 \pm 0.005)$ eV with the energy interval $\Delta_{[111]} = \hbar\omega(\beta_2) - \hbar\omega(\beta_1) = (0.13 \pm 0.01)$ eV and $\hbar\omega(\alpha) = (0.75 \pm 0.005)$ eV. At the orientation $I \parallel [100]$, these transitions correspond to the maxima at $\hbar\omega(\beta_1) = (0.76 \pm 0.005)$ eV and $\hbar\omega(\beta_2) = (0.83 \pm 0.005)$ eV with energy interval $\Delta_{[100]} = (0.07 \pm 0.01)$ eV, and at $I \parallel [110]$ to the maxima $\hbar\omega(\beta_1) = (0.69 \pm 0.005)$ eV and $\hbar\omega(\beta_2) = (0.80 \pm 0.005)$ eV with the energy interval $\Delta_{[110]} = (0.11 \pm 0.01)$ eV and $\hbar\omega(\alpha_1) = (0.74 \pm 0.005)$ eV, $\hbar\omega(\alpha_2) = (0.76 \pm 0.005)$ eV. The ratio of the energy intervals between the β components for the [111] axis and the axes [100] and [110] corresponds to the ratio of the intervals for the maximally split levels at different orientations of I in the single crystal:

$$\Delta_{[100]}/\Delta_{[111]} \sim 1/\sqrt{3}, \quad \Delta_{[110]}/\Delta_{[111]} \sim \sqrt{2}/\sqrt{3}.$$

For the transitions of types A and B, no clearly pronounced separated maxima were observed on the OME curves. We note, however, that in[18], where an improved procedure was used to measure the OME and at normal incidence of the light, two negative OME maxima, at $\hbar\omega = (0.56 \pm 0.01)$ eV and $\hbar\omega = (0.78 \pm 0.01)$ eV, corresponding to transitions B and A of the first type[4], were observed in single-crystal Ni films grown in the (100) plane even at room temperature.

The identification of the observed structure with definite components from spin-orbit-split interband transitions enables us to determine a number of band parameters in a self-consistent manner.

For example, the mass ratio $\mathfrak{M}$ in the direction ΓL, assuming validity of a quadratic E(k) dependence, can be determined by two methods.

a) From the ratio of the intervals between the β components for the transitions B and A, since

$$\mathfrak{M} = [\hbar\omega(\beta_2) - \hbar\omega(\beta_1)]/[\hbar\omega(\beta_2') - \hbar\omega(\beta_1')] = 3.25.$$

b) From the ratios of the frequencies $\hbar\omega(\beta_1')$, $\hbar\omega(\beta_2')$ and $\hbar\omega(\beta_1)$, $\hbar\omega(\beta_2)$ at $H \parallel [111]$

$$\hbar\omega(\beta_1) = [E_F - E(L_{2'}\downarrow)] + \mathfrak{M}[\hbar\omega(\beta_1') - (E_F - E(L_{2'}\downarrow))],$$
$$\hbar\omega(\beta_2) = [E_F - E(L_{2'}\downarrow)] + \mathfrak{M}[\hbar\omega(\beta_2') - (E_F - E(L_{2'}\downarrow))],$$
$$\mathfrak{M} = \frac{\hbar\omega(\beta_1) - x}{\hbar\omega(\beta_1') - x} = \frac{\hbar\omega(\beta_2) - x}{\hbar\omega(\beta_2') - x} \qquad x = E_F - E(L_{2'}\downarrow).$$

Using the obtained relations, we get

$$x = 0.48, \ \mathfrak{M} = 3.3.$$

The values of $\mathfrak{M}$ obtained by the two methods are practically equal.

Simultaneous knowledge of the quantities $\hbar\omega(\beta')$ and $\hbar\omega(\beta)$ and of the exchange splitting of the 3d band at the point L leads to the following values of the interband intervals:

$$\langle E(L_{32}\downarrow)\rangle - E_F = 0.09 \text{ eV}, \ E_F - \langle E(L_{32}\uparrow)\rangle = 0.21 \text{ eV},$$

$\langle E(L_{32}^{\downarrow})\rangle$ is the mean value of the energy of two spin-orbit-split levels L_{32}.

The obtained position of the Fermi level relative to the L_{32} level in the subband with spins ↑ agrees with the experiments on the de Haas-van Alphen effect[19] and with optical experiments[20]. The positions of the energy levels at the L and X points are not independent. The relative width of the d band, determined by the distance $E(x_S^{\downarrow}) - E(L_{32}^{\downarrow})$, as shown in[21], depends little on the potential used in the calculation of the band structure. According to the estimate[17], this is a quantity on the order of 0.12–0.15 eV. Using this quantity and our estimates of the interband intervals at the point L, we obtain the distance between the vertex of the 3d band and the Fermi level $E_F - \langle E(x_S^{\downarrow})\rangle \approx 0.06$–$0.09$, which agrees with the experiments on the temperature dependence of the magnetization[22] and the spin polarization of the photoelectron[23].

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