EXPERIMENTAL INVESTIGATION OF THE ELECTRON STRUCTURE OF NICKEL BY THE MAGNETO-OPTICAL METHOD

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The nondiagonal component of the permittivity tensor of nickel was measured in the region 0.7—5.8 μ. The natural frequencies detected at 0.3, 0.8, and 1.4 eV were identified with interband electron transitions in the Fermi surface model of ferromagnetic nickel, proposed by Phillips.[2] The values of the exchange splitting of the d- and s-bands of nickel were determined from the experimental data: $\Delta E_{dd} = 0.7 \pm 0.05$ eV and $\Delta E_{ss} = 0.45 \pm 0.05$ eV. The sharp increase of the intensity of the magneto-optical resonance at $h\nu_0 = 0.75$ eV in an alloy Ni_3Fe was interpreted as being due to the emptying of the $L_{2'}\uparrow$ level and a transition from the open to the closed Fermi surface, due to a shift of the Fermi level.

DEFINITE models of a Fermi surface have been proposed for ferromagnetic nickel in recently published work.[1,2] Both these models start from the general representations of the main features of the Fermi surface of various bands. For example, it is assumed that the Fermi surfaces of all the bands are closed except for one s↑ surface,[1)] which is open and has the same topology as the Fermi surface of copper. However, the positions of the energy levels, with respect to one another and with respect to the Fermi level, differ considerably in these two models: for example, in the first model[1], the exchange splitting of the 3d-band ΔE_{dd} is taken to be ~2 eV and the separation $E(L_{32}\downarrow) - E_F \approx 0.24$ eV, while in the second model[2] $\Delta E_{dd} \approx 0.6$ eV and $E(L_{32}\downarrow) - E_F \lesssim 0.05$ eV, etc.

Experimental measurements of the electron energy spectrum characteristics may be used as the basis for developing a quantitative theory of ferromagnetism in transition d-metals. Therefore, in the present work, we made an attempt to determine accurately the natural frequencies of nickel in the region 0.21—1.8 eV in order to compare the results with the proposed models of the Fermi surface of ferromagnetic nickel.

The natural frequencies of nickel were determined by the magneto-optical method, in the same way as in the earlier work,[3] but the experimental accuracy was much greater due to the improvement of the method of measuring the equatorial Kerr effect. The block diagram of the apparatus is given in Fig. 1. The sample, placed in an electromagnet, was subjected to alternating magnetization by an audio-frequency current and, consequently, the intensity of the reflected light was modulated at the same frequency (f = 79 cps). The depth of modulation determined the equatorial Kerr effect:

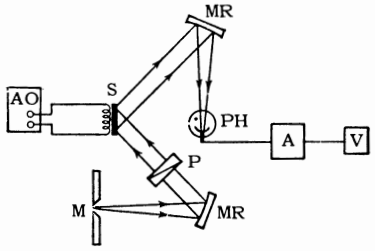

FIG. 1. Block diagram of the apparatus: M – exit slit of the monochromator; MR – mirror; S – sample; P – polarizer; PH – photocell; AO – audio oscillator; A – selective amplifier; V – tube voltmeter.

$$\delta = (J - J_0) / J_0,$$

where J_0 and J are, respectively, the intensities of light reflected by a demagnetized and a magnetized ferromagnet. The signal from a photo-receiver load was fed to an input of a selective amplifier (f_{res} = 79 cps) whose output was applied to an oscillograph or a tube voltmeter. The selective amplifier circuit is given in[4] where similar apparatus was used for a different purpose. Depending on the range of wavelengths used, a germanium photodiode FD-1, a cooled PbS photoreceiver, or a cooled InSb photodiode was used as the radiation detector. The measurements were carried out on a spectrograph (type IKS-12) with an attachment (IPO-12) for measuring the reflection. The monochromator slit width was such that the range of frequencies transmitted did not exceed $\nu/10$ cm^{-1}. Using an auxiliary diaphragm, the aperture for the incident light was reduced to 1°.

[1)]The symbol ↑ denotes electrons with uncompensated spin.

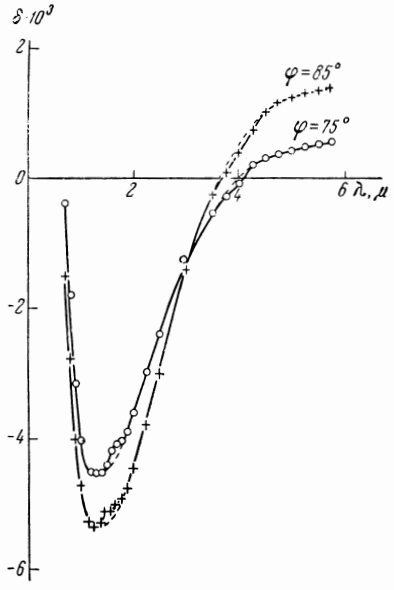

FIG. 2. Equatorial Kerr effect for pure nickel.

Figure 2 gives the values of the equatorial Kerr effect for a pure (99.994%) polycrystalline sample of nickel measured at angles of incidence of light equal to 75 and 85°. The sample was prepared in the form of a plate 16 × 24 × 0.3 mm; it was polished mechanically, annealed and then polished electrolytically.

The nondiagonal component of the permittivity tensor was calculated using the formula

$$\delta = a\varepsilon_1' + b\varepsilon_2',$$

where

$$a = 2\sin 2\varphi \frac{A_1}{A_1^2 + B_1^2}, \qquad b = 2\sin 2\varphi \frac{B_1}{A_1^2 + B_1^2},$$

$$A_1 = \varepsilon_2(2\varepsilon_1\cos^2\varphi - 1),$$

$$B_1 = (\varepsilon_2^2 - \varepsilon_1^2)\cos^2\varphi + \varepsilon_1 - \sin^2\varphi,$$

$$\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon = \varepsilon_1 - i\varepsilon_2,$$

$$\varepsilon_{xy} = -\varepsilon_{yx} = i\varepsilon', \qquad \varepsilon' = \varepsilon_1' - i\varepsilon_2'.$$

The value of δ depends not only on ϵ_1' and ϵ_2', but also on ϵ_1 and ϵ_2.

Since the dispersion curves of ϵ_1 and ϵ_2 are simpler (cf. the table), we may conclude that the nature of the frequency dependence of δ is governed by the frequency dependence of ϵ_1' and ϵ_2'. Indeed, the ϵ' curves (cf. Figs. 3 and 4) obtained for different optical constants are of the same kind and only the absolute values of ϵ_1' and ϵ_2' differ somewhat.

In the given curves of δ and ϵ', we note the following features (in the natural frequency region): a) ϵ_1' changes sign and ϵ_2' passes through a maximum at $h\nu$ = 0.23 eV; b) the δ curves have a characteristic inflection in the region of 0.3 eV (cf. Fig. 2), which leads to pronounced anomalies in ϵ_1' and ϵ_2'; c) a pronounced anomaly is observed in the δ curves in the region of 0.8 eV, but it is less pronounced in the ϵ' curves; d) ϵ_1' changes sign and ϵ_2' has a broad maximum in the region 1—1.8 eV. In further discussion of the results, we shall, to make the case definite, assume that the natural frequencies of these magneto-optical resonances ($h\nu_0$) are 0.23, 0.3, 0.8 and 1.4 eV.

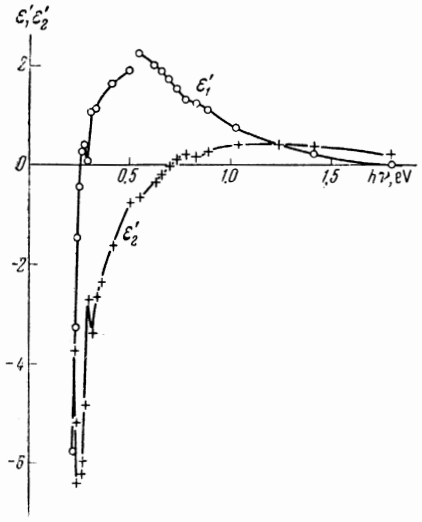

FIG. 3. Values of $\epsilon' = \epsilon_1' - i\epsilon_2'$, calculated from the optical constants given in [5,6] for pure nickel.

The general nature of the ϵ_1' and ϵ_2' curves and, in particular, their behavior in the regions $h\nu_0$ = 0.23 eV and 1—1.8 eV is in agreement with the magneto-optical results,[3,7] discussed earlier[1,2,7] and with the optically determined values of the natural frequencies for nickel.[1] The following may be said about the small anomalies detected

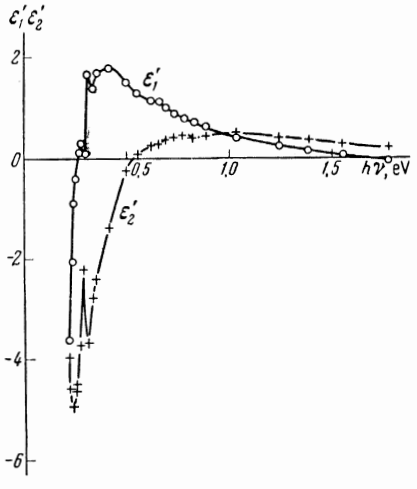

FIG. 4. Values of $\epsilon' = \epsilon_1' - i\epsilon_2'$, calculated from the optical constants of pure nickel measured by Noskov.

λ		Data of Ingersoll, [5] Beattie and Conn [6]		Noskov's data		λ		Data of Ingersoll, [5] Beattie and Conn [6]		Noskov's data	
μ	eV	$-\varepsilon_1$	ε_2	$-\varepsilon_1$	ε_2	μ	eV	$-\varepsilon_1$	ε_2	$-\varepsilon_1$	ε_2
0.69	1.8	11.80	16.40	11.56	14.81	2.5	0.496	70.1	75.4	64.4	63.5
0.88	1.41	18.01	24.01	14.70	19.01	3.0	0.414	88.8	91.5	92.9	81.1
1.0	1.24	21.33	27,58	16.81	21.62	3.5	0.355	117.1	111.1	122.0	97.9
1.2	1.03	27.59	34,80	20.79	26.00	3.75	0.330	136,9	120,2	145,6	105.6
1.4	0.886	35.03	42,10	25.43	31.72	4.0	0.310	156,6	128,7	152.5	114,2
1.5	0.827	38.44	45,50	27.82	34.31	4.25	0.290	187.2	139.2	168,2	122.2
1.6	0.775	42,13	48,75	30.94	37,30	4.5	0.276	217.2	148.8	189.6	131.6
1.7	0.730	46.82	52,20	34.32	40,11	4.75	0.261	250,7	159.6	212,8	141,0
1.8	0.690	50,16	55,30	37.04	43.00	5.0	0.248	285.8	172.5	234,1	149.8
1.9	0.653	54,28	59.04	40,01	45,58	5,25	0.236	313.6	184,0	258.0	159.1
2.0	0.620	58.71	62,55	42.41	48,17	5.5	0.226	338.8	194,8	286,4	169.7
2.25	0.551	69.66	71.25	52,04	55.35	5.75	0,216	364,4	208.0	314,6	180.3

again for the natural frequencies 0.3 and 0.8 eV: the existence of an anomaly of δ in the region of 0.8 eV is not in doubt because it has been observed in numerous measurements on various samples of nickel using different monochromator slit widths and two receivers (a germanium photodiode and a PbS photoresistor). The anomaly of δ in the region of 0.3 eV ($\approx 4\,\mu$) was subjected to a very careful check because here the effect changed sign and at $\lambda = 4.26\,\mu$ there was a strong absorption band due to the atmosphere. However, a thorough analysis of these factors (allowing for the monochromator slit width, the spectral density of the radiation from the source, and the sensitivity of the InSb receiver) showed that the systematic measurement error could not be greater than 25% of the deviation of the continuous line from the dashed line in Fig. 2.

We note, however, that the separation of two natural frequency regions at 0.23 and 0.3 eV in the ϵ_1' and ϵ_2' curves is somewhat arbitrary and is bound up with the explanation, proposed below, [2)] of the obtained experimental data. We are assuming that the general nature of the curves of ϵ_1' and ϵ_2' in the range 0.21—1.0 eV and, in particular, the change of sign of ϵ_1' at $h\nu_0 = 0.23$ eV, are due to the motion of free carriers—the ferromagnetic Hall effect at optical frequencies—in agreement with the theory proposed by one of the present authors.[7] The singularities observed at $h\nu_0 = 0.3$, 0.8, and 1.4 eV are interpreted as the natural frequencies of the interband electron transitions in nickel.

According to the Phillips model,[2] the transitions ($L_{32}\downarrow$, $L_{2'}\uparrow$) have the frequency $h\nu_0 = 0.3$ eV. Since the level $L_{2'}\uparrow$ lies 0.15 eV below the Fermi level,[2] we find that the exchange splitting of the s-p band is $\Delta E_{ss} = E(L_{2'}\downarrow) - E(L_{2'}\uparrow) = 0.45 \pm 0.05$ eV. On the other hand, the level $L_{32}\downarrow$ lies near the Fermi level [$E(L_{32}\downarrow) - E_F \lesssim 0.05$ eV[2]], and therefore the natural frequency of the transition ($L_{32}\uparrow$, $L_{2'}\uparrow$) gives directly the value of the splitting of the d-band $\Delta E_{dd} = E(L_{32}\downarrow) - E(L_{32}\uparrow)$. Phillips[2] identified the anomaly in the ϵ_1' curve at $h\nu_0 = 0.6$ eV[7] with the transition ($L_{32}\uparrow$, $L_{2'}\uparrow$), and hence obtained the value $\Delta E_{dd} = 0.6$ eV. However, the ϵ_1' curve which Phillips took from[7] was calculated from values of δ which were not very accurate. Moreover, he joined the experimental values of the optical constants taken from[5,6] in the region of 0.55 eV and, therefore, his value for $\Delta E_{dd} = 0.6$ eV cannot be regarded as reliable. The results of our measurements allow us to identify the natural frequency 0.8 eV with the transition ($L_{32}\uparrow$, $L_{2'}\uparrow$). Thus we obtain $\Delta E_{dd} = 0.7 \pm 0.05$ eV, because it is natural to determine the exchange splitting of the d-band from the long-wavelength absorption band edge, which is equal to 0.7 eV (cf. Fig. 2). This value does not contradict Phillips' model since his estimates of the value of ΔE_{dd}, obtained by various methods, lie within the limits 0.6—0.8 eV.[2]

Finally, the natural frequency in the region of 1.4 eV can be identified, in accordance with[2], with the transition ($W_1\downarrow$, $W_1'\downarrow$).

An additional confirmation of the validity of the proposed classification of the natural frequencies of nickel on the basis of the Phillips model is provided by the results of measurements of δ of the ordered alloy Ni_3Fe.

A sharp magneto-optical resonance was observed in the alloy Ni_3Fe in the region $h\nu_0 = 0.75$ eV.[8] Since this frequency coincided with one of the natural frequencies of nickel found in the present work, we carried out more careful measurements of the frequency dependence of δ for this

[2)] It is worth noting that the splitting of the maximum of ϵ_2' in the far infrared region is 0.07 eV, i.e., it coincides in magnitude with the spin–orbital splitting of the L_3 level predicted by Phillips.[2]

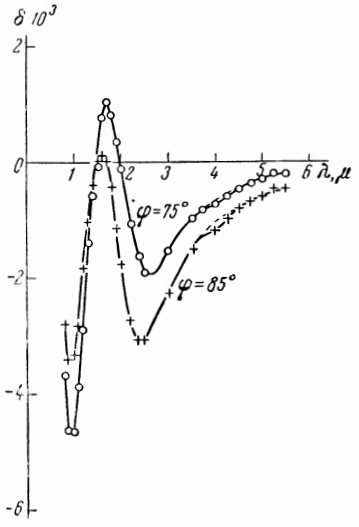

FIG. 5. Equatorial Kerr effect for the ordered alloy Ni_3Fe.

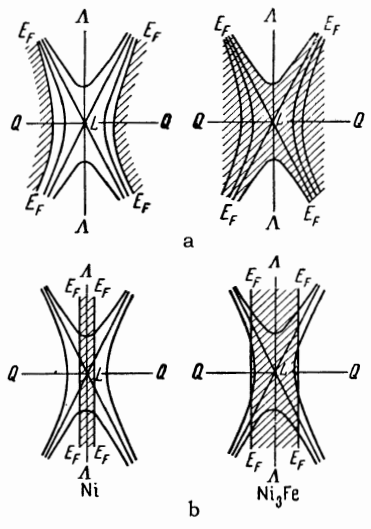

FIG. 6. Scheme of the interband transitions near the L point in nickel and in the alloy Ni_3Fe: a) transition ($L_{32}\uparrow$, $L_{2'}\uparrow$), the Fermi level position in the s-band is shown, transitions possible only to the hatched region; b) transitions ($L_{32}\downarrow$, $L_{2'}\downarrow$), the Fermi level position in the d-band is shown, transitions possible only from the unhatched region.

alloy. The results of these measurements are given in Fig. 5. It is evident that the δ curves exhibit, apart from the cited resonance at $h\nu_0 = 0.75$ eV, an additional small anomaly in the region of $h\nu_0 = 0.3$ eV. From the point of view of the proposed classification of the transitions in nickel, this result may be interpreted as follows: with the transition from nickel to the alloy Ni_3Fe, the number of 4s + 3d electrons decreases from 10 to 9.5. In the rigid band approximation, the Fermi level should drop by about 0.17 eV for a density of states near E_F given by $dN/dE \approx 3$ states/eV. atom.[9]

According to the Phillips model, this will empty the level $L_{2'}\uparrow$ [in nickel $E_F - E(L_{2'}\uparrow) = 0.15$ eV] and consequently increase the intensity of the transition ($L_{32}\uparrow$, $L_{2'}\uparrow$). Conversely, the population of the level $L_{32}\downarrow$ and the intensity of the transition ($L_{32}\downarrow$, $L_{2'}\downarrow$) should decrease.

Figure 6 shows the cross sections of the constant energy surfaces for the interband transitions near the point L, which may be denoted by ($L_{32}\uparrow$, $L_{2'}\uparrow$) (Fig. 6a) and ($L_{32}\downarrow$, $L_{2'}\downarrow$) (Fig. 6b). In the former case, transitions are possible to the hatched region (to the states lying above the Fermi level in the s-band), while in the latter case transitions are possible from the unhatched region (from the states lying below the Fermi level in the d-band). The expected sharp increase of the intensity of the transitions ($L_{32}\uparrow$, $L_{2'}\uparrow$) in the alloy Ni_3Fe follows from the fact that the lowering of the Fermi level leads to the emptying of a conical surface, with its vertex at the point L, having a high density of states.[2] We note that the validity of this assumption about the change in the electron structure of Ni_3Fe compared with pure Ni may be checked by independent experiments: optical measurements on the ordered alloy Ni_3Fe should show a sharp absorption line in the region of 0.75 eV, and galvanomagnetic measurements should reveal a transition from a Fermi surface open at the points L to a closed Fermi surface of the s$\uparrow$-band. Some confirmation of the transition to the closed Fermi surface in the alloy Ni_3Fe may be seen in the fact that the δ curve in the magneto-optical resonance region $h\nu_0 = 0.75$ eV is practically the same also for the ordered alloy Ni_3Mn,[8] i.e., it remains the same with the further lowering of the Fermi level in a rigid band.

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