

VARIATION OF THE MAGNETIZATION OF NICKEL AND IRON AT LOW TEMPERATURES

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The derivative of magnetization with respect to temperature  $\partial M(T, H)/\partial T$  is measured for nickel and iron at temperatures between 0.7 and 4.2°K and in magnetic fields up to 34 kOe. It is shown that in the expression for the variation of saturation magnetization  $\delta M_g(T, H)$  there is, along with the term involving spin-wave excitation, a term  $AT^2$  (A is a constant). For  $T \lesssim 1^\circ\text{K}$  and  $H \gtrsim 30$  kOe, it is found that  $\delta M_g(T, H) \approx AT^2$ . The values of the spin-wave spectrum gap are obtained for the two metals. The experimental data are compared with existing theories.

THE literature contains a number of papers concerned with the saturation magnetization  $M_g$  of ferromagnetic substances at low temperatures<sup>[1-5]</sup>. According to spin-wave theory (see, for example, the survey<sup>[1]</sup>) the function  $M_g(T, H)$  follows the  $T^{3/2}$  law. Dyson<sup>[5]</sup> shows that when the spin-wave interactions are taken into account terms proportional to  $T^{5/2}$ ,  $T^{7/2}$ ,  $T^4$ , etc., appear in  $M_g(T, H)$ . There are several approximations in the band theory of ferromagnetism. The calculations of Stoner<sup>[2]</sup> lead to

$$M_g = M_0(1 - BT^2), \tag{1}$$

where B is a constant. If in the ground state one of the two possible spin directions corresponds either to a completely filled or a completely empty band, then

$$M_g = M_0[1 - ET^{3/2} \exp(-\Delta^*/kT)], \tag{2}$$

where E is a constant, and  $\Delta^*$  depends on the Fermi energy and the exchange interaction constant. Recent calculations<sup>[3,4]</sup> have shown that in the band model the magnetization  $M_g(T, H)$  can be represented as a combination of expressions containing a dependence of the form  $T^{3/2}$  and (1) or  $T^{3/2}$  and (2). For example, Kondratenko<sup>[3]</sup> has

$$M_g = M_0[1 - CT^{3/2} - AT^2/M_0], \quad H = 0, \tag{3}$$

where C and A are constants. The term  $AT^2$  is due to Fermi excitations of the "magnetic" electrons.

These theoretical models may be checked by measuring the dependence of  $M_g$  on T and H in the low-temperature region ( $T \ll \theta_C$ ,  $\theta_C$  the Curie temperature). Experiments that aim to do this, however, yield ambiguous results. Thus, the recent investigations of Argyle et al.<sup>[6]</sup> and of Rode and Germann<sup>[7]</sup> above 4.2°K showed that for iron the variation of  $M_g$  with temperature follows the spin-wave theory rather well. Argyle et al. arrive at the same conclusion with respect to nickel. On the other hand, Rode and Germann<sup>[7]</sup>, following Thompson, et al.<sup>[4]</sup>, consider that in nickel a contribution like (2) is important. Measurements of the magnetization of iron and nickel in the temperature interval 1.5-5°K and in fields up to 22 kOe by Tsarev and Zavaritskii<sup>[8]</sup>, show that  $M_g(T, H)$  is evidently better described by the band-theory expression (3).

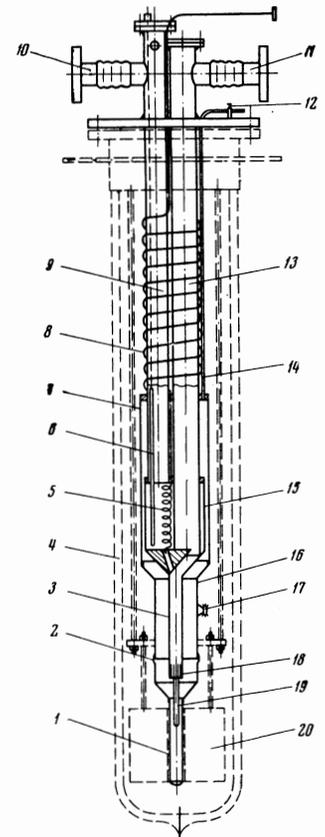


FIG. 1. He<sup>3</sup> apparatus for measurements at  $T \lesssim 1^\circ\text{K}$ : 1, 2, 7, 16—external walls of metal dewar; 3, 18—stainless steel tube with copper base for collecting liquid helium; 4—outer Dewar for He<sup>4</sup>; 5, 8—coils for cooling and condensing He<sup>3</sup>; 6—superconducting level indicator for He<sup>4</sup>; 9, 10—He<sup>4</sup> pumping tubes; 11, 13—He<sup>3</sup> pumping tubes; 12, 14—tube and valve for pumping out the "jacket"; 15—internal He<sup>4</sup> Dewar; 17—leads for the measuring system; 19—copper finger to which samples are soldered; 20—superconducting solenoid.

1. EXPERIMENTAL RESULTS

We measured the derivative of the magnetization with respect to temperature  $\partial M/\partial T$  for nickel and iron at temperatures 0.7-4.3°K in fields up to 34 kOe. For the temperatures below 1°K, the He<sup>3</sup> apparatus shown in Fig. 1 was employed. To reduce vibration, evacuation of the He<sup>3</sup> was accomplished by an adsorption pump. The magnetic field was created by a superconducting solenoid. The magnitude of  $\partial M/\partial T$  was determined from the EMF induced in a coil containing the sample. The temperature was varied periodically at a frequency of 9.6 Hz about some average value, the oscillation amplitude being measured with a carbon-

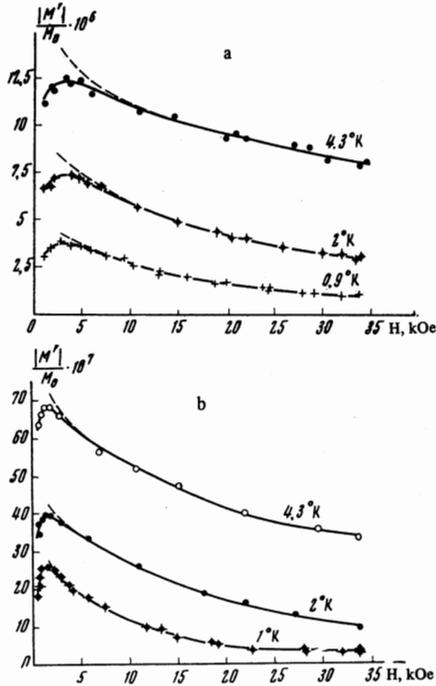


FIG. 2. Dependence of the quantity  $|M'/M_0|_{T=\text{const}}$  on magnetic field for nickel (a) and iron (b) samples ( $M' \equiv \partial M/\partial T$ ) for different temperatures. The averaged dependence of  $|M'/M_0|$  on  $H$  is shown by solid lines. The dependence of  $|M'_g/M_0|$  on  $H$  is given by the dashed lines.

film thermometer. Calibration of the measuring coil was carried out with a single-layer solenoid the same size as the sample. The measuring scheme was similar to the one described in<sup>[9]</sup>. A polycrystalline sample of iron and a monocrystal of nickel with the axis of magnetization along its length served as the specimens. Impurities in the iron amounted to less than 0.03%; the basis of thermal conductivity data we estimate the impurity content of the nickel to be about the same. The specimens were in the form of cylinders, 3-cm long, the iron sample being 0.18 cm in diameter and the nickel 0.28 cm.

The results for the dependence of  $M' \equiv \partial M/\partial T$  (the prime will always indicate the partial derivative with respect to temperature) on magnetic field at constant temperature for the samples of nickel and iron are given in Fig. 2. Figure 3 gives the results for the dependence on temperature at a fixed field. The random error in measurements of  $M'|_{H=\text{const}}$  was not greater than 15%; in measuring  $M'|_{T_{\text{av}}=\text{const}}$ , it was not greater than 5% ( $T_{\text{av}}$  is the average temperature of the sample, called  $T$  from now on). The systematic error is 10% in  $M'|_{H=\text{const}}$  and 20% in  $M'|_{T_{\text{av}}=\text{const}}$ . A detailed error analysis is given in<sup>[9]</sup>. The results of the measurements of the relative changes in  $M'(T, H)$  at  $T > 1.5^\circ\text{K}$  and  $H < 22 \text{ kOe}$  agree with the data of Zavaritskiĭ and Tsarev<sup>[8]</sup>. Our absolute values of  $M'$  are approximately 40% less than those given in<sup>[8]</sup>.

## 2. METHOD OF ANALYSIS OF THE EXPERIMENTAL DATA

According to the theory of spin waves, the dependence of the magnetization of a ferromagnet on tempera-

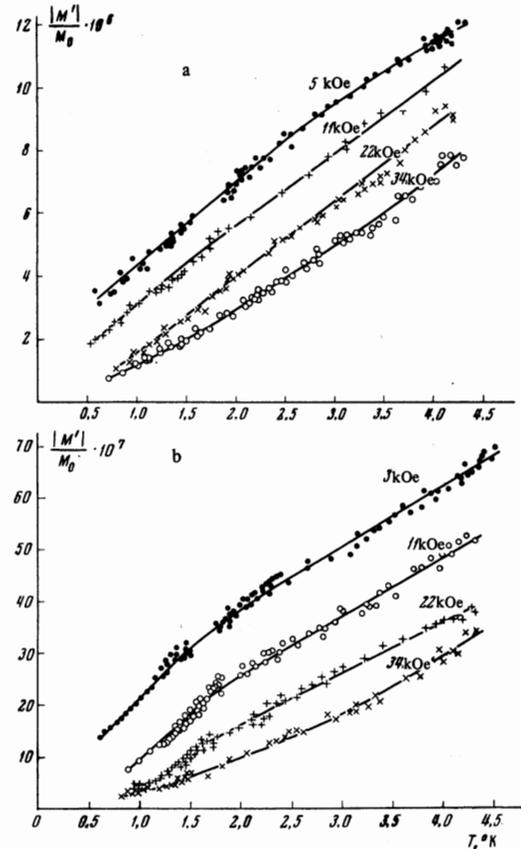


FIG. 3. Dependence of  $M'/M_0|_{H=\text{const}}$  on temperature for nickel (a) and iron (b) samples in different magnetic fields. The solid lines are averaged from several experiments.

ture and magnetic field for a single-domain sample has the form<sup>[10]</sup>

$$\Delta M_\alpha(T, H) = M_0 - M_\alpha(T, H) = CT^{3/2} J(\alpha, \beta), \quad (4)$$

where  $M_0$  is the magnetization at  $T = 0^\circ\text{K}$ ,  $C$  is a constant, and

$$J(\alpha, \beta) = \int_0^{\pi/2} \sin \theta d\theta \int_{\pi(\theta)}^{\infty} \frac{[(z^2 + \alpha^2 \beta^2 \sin^4 \theta)^{1/2} - \alpha(1 + \beta \sin^2 \theta)]^{1/2}}{e^z - 1} dz,$$

$$x(\theta) = \alpha(1 + 2\beta \sin^2 \theta)^{1/2},$$

$$\alpha = \frac{g\mu_0(H_0^{(e)} + H_A)}{kT}, \quad \beta = \frac{2\pi M_0}{H_0^{(e)} + H_A},$$

$$H_A = \frac{2K}{M_0} \text{ for Fe, } H_A = \frac{4|K|}{3M_0} \text{ for Ni,}$$

$$H_0^{(e)} = H_0^{(e)} - 4\pi N_z M_0, \quad H_0^{(e)} \parallel M_0 \parallel n; \quad (5)$$

$H_0^{(e)}$  is the applied magnetic field,  $n$  is the axis of easy magnetization,  $K$  is the anisotropy constant,  $N_z$  is the demagnetization factor,  $\mu_0$  is the Bohr magneton, and  $g$  is the gyromagnetic ratio.

The relations (4) and (5) are valid at sufficiently low temperatures ( $T \ll \Theta_C$ )<sup>[11]</sup>, where the interaction between the spin waves is weak. This condition is very well satisfied in our experiments. In comparing experimental data with theory it is expedient to use the expression  $(M_0 T)^{-1} dM_g(T)/dT$ . The derivative  $dM_g/dT$  can be represented in the form (in Eq. (5) the depend-

ence of the anisotropy constant is introduced)

$$\frac{dM_z}{dT} = M'_z + \frac{\partial M_z}{\partial K} \frac{dK}{dT}, \quad (6)$$

$$M'_z \equiv \partial M_z / \partial T = CT^{1/2} I(\alpha, \beta), \quad (7)$$

where

$$I(\alpha, \beta) = \int_0^1 du \int_0^{\pi} \{y + \alpha[1 + 2\beta(1 - u^2)]^{1/2}\} f(y) \cdot \\ \times \frac{\exp\{y + \alpha[1 + 2\beta(1 - u^2)]^{1/2}\}}{[\exp\{y + \alpha[1 + 2\beta(1 - u^2)]^{1/2}\} - 1]^2} dy, \\ f(y) = \{[(y + \alpha[1 + 2\beta(1 - u^2)]^{1/2})^2 + \alpha^2\beta^2(1 - u^2)^2]^{1/2} - \\ - \alpha[1 + \beta(1 - u^2)]^{1/2}\}^2, \quad (8)$$

and

$$\frac{\partial M_z}{\partial K} \frac{dK}{dT} = CT^{1/2} \frac{\partial J(\alpha, \beta)}{\partial K} \frac{dK}{dT}. \quad (9)$$

We shall show that under the conditions of our experiment ( $T \lesssim 4.2^\circ\text{K}$ )

$$dM_z/dT \approx M'_z. \quad (10)$$

In the low-temperature region ( $T \ll \Theta_C$ ) we have<sup>[11,12]</sup>

$$\delta K = 10 \frac{K_0}{M_0} \delta M_z,$$

where  $K$  is the anisotropy constant and  $K_0$  is the anisotropy constant at  $T = 0$ . From this we have

$$\frac{dK}{dT} = 10 \frac{K_0}{M_0} \frac{dM_z}{dT}. \quad (11)$$

Using (11) and (6), we obtain

$$\frac{dM_z}{dT} \approx M'_z \left(1 - 10 \frac{K_0}{M_0} \frac{\partial M_z}{\partial K}\right)^{-1}. \quad (12)$$

It is not hard to see that  $10(K_0/M_0)\partial M_z/\partial K \ll 1$ , from which (10) follows:

The very same result can be obtained using the temperature dependence  $K(T)$ . For nickel<sup>[13,14]</sup>

$$K = K_0 e^{-\alpha T}, \quad K_0 = 8 \cdot 10^5, \quad \alpha = 34 \cdot 10^{-6}. \quad (13)$$

Using (7)–(9) and (13), it can be shown that

$$M'_z \gg \frac{\partial M_z}{\partial K} \frac{dK}{dT}. \quad (14)$$

In the case of iron (14) is obviously fulfilled with high accuracy.

It follows directly from (10) that

$$\frac{1}{M_0 T} |M'_z| = CQ(T, H), \quad (15)$$

where  $Q(T, H) = I(\alpha, \beta)/T^{1/2} M_0$ . Following<sup>[3]</sup>, it is easy to see that in the band model

$$\frac{1}{M_0 T} |M'_z| \equiv \mathcal{A} = CQ(T, H) + \frac{2A}{M_0}. \quad (16)$$

For an examination of the peculiarities of the dependence  $M'_z(T, H)$  in the coordinates  $\mathcal{A}$ ,  $Q$ , we expand the integral  $I(\alpha, \beta)$  appearing in the expression for  $Q$ . We represent the function  $f(y)$  as

$$f(y) = y^{1/2} \left[ \frac{y+d}{y} \left(1 + \frac{2y}{(y+d)^2} (c-d)\right)^{1/2} - \frac{d}{y} \right]^{1/2},$$

where

$$\gamma = \alpha[1 + \beta(1 - u^2)], \quad c = \alpha[1 + 2\beta(1 - u^2)]^{1/2}.$$

It is easily seen that

$$2y(c-d)/(y+d)^2 < 1$$

for any values of  $T$  and  $H$ . Expanding  $f$  in series for the region of values of  $\alpha$  and  $\beta$  we used in the experiment, we obtain

$$f \approx y^{1/2} [1 + (c-d)/2(y+d)]. \quad (17)$$

Using (17) we have

$$I(\alpha, \beta) \approx \frac{\sqrt{\pi}}{2} \int_0^1 du \sum_{k=1}^{\infty} \frac{\exp\{-\alpha[1 + 2\beta(1 - u^2)]^{1/2} k\}}{k^{1/2}} \cdot \\ \times \left\{ \alpha[1 + 2\beta(1 - u^2)]^{1/2} + \frac{3}{2k} \right\} \text{ for } \frac{c-d}{2d} \ll 1, \quad (18a)$$

$$I(\alpha, \beta) \approx \frac{\sqrt{\pi}}{2} \sum_{k=1}^{\infty} \frac{e^{-\alpha k}}{k^{1/2}} \left( \alpha + \frac{3}{2k} \right) \text{ for } \beta \ll 1. \quad (18b)$$

Keeping these expansions in mind, it is easy to see that the function  $Q(T)_{H=\text{const}}$  has a maximum  $Q_{\text{max}}$  at the temperature

$$T_{\text{max}} \sim 2g\mu_0(H_0^{(0)} + H_A)/\sqrt{3},$$

where  $g\mu_0(H_0^{(1)} + H_A)$  is the energy gap  $\Delta$  in the spin-wave spectrum. The magnitude of  $Q_{\text{max}}$  depends on  $H_0^{(e)}$ . Thus, the function  $M'_z(T)_{H=\text{const}}$  in coordinates  $\mathcal{A}$ ,  $Q$  (see (16)) should have a "turnaround point" at  $Q_{\text{max}}$ , which is directly associated with the existence of the gap. In case  $\Delta = 0$ , the function  $\mathcal{A}(Q)$  is single-valued and there is no "turnaround point."

Consider a multi-domain sample. Near saturation, the magnetic moment can be expanded in series

$$M(T, H) = M_z(T, H) (1 - q/H^2), \quad (19)$$

where  $q \sim c(K/M_0)^2$ ,  $c$  is a constant, and  $K$  is the anisotropy constant. Taking the derivative with respect to  $T$  and using (11), we obtain for  $T \ll \Theta_C$

$$M_0 \frac{dq}{dT} \approx D \frac{dM_z}{dT},$$

where  $D \approx 20cK_0^2/M_0^2$  and

$$M'(T, H) = M'_z(T, H) (1 - D/H^2). \quad (20)$$

At temperatures  $T \lesssim 4^\circ\text{K}$ , these relations are fulfilled very accurately.

Zavaritskii and Tsarev<sup>[15]</sup> have shown that  $dq/dT \sim M'_z$ . Keeping in mind the form of the function  $M'_z(T, H)$  (see (18)), we see easily that the curve  $M'(H)|_{T=\text{const}}$  has a maximum at a definite value of the field  $H_{\text{max}}$ . From this maximum condition we obtain

$$D = H_{\text{max}}^2 \left\{ H_{\text{max}} - 2 \frac{\partial M_z}{\partial T} \Big|_{H=H_{\text{max}}} / \frac{\partial^2 M_z}{\partial H \partial T} \Big|_{H=H_{\text{max}}} \right\}^{-1}. \quad (21)$$

It can be shown that

$$\frac{\partial M_z}{\partial T} \Big|_{H=H_{\text{max}}} / \frac{\partial^2 M_z}{\partial H \partial T} \Big|_{H=H_{\text{max}}} \approx \frac{kT}{g\mu_0}.$$

The comparison of experimental data with theory was carried out with Eq. (15) graphically in coordinates  $\mathcal{A}$  and  $Q$ . The value of  $\mathcal{A}$  (see (16)) was calculated from experimental data using relations (20) and (21). Calculation of  $Q(T, H)$  was by electronic computer using formulas (15) and (3). The following values were used in this calculation:  $M_0 = 510$  CGSM units<sup>[16]</sup>,  $K_0 = 8 \times 10^5$  erg/cm<sup>3</sup><sup>[17]</sup>,  $N_z = 0$  in the case of nickel, and  $M_0 = 1750$  CGSM units<sup>[16]</sup>,  $K_0 = 5.2 \times 10^5$

erg/cm<sup>3</sup><sup>[17]</sup>,  $N_Z = 0$  in the case of iron.

Neglect of demagnetization ( $N_Z = 0$ ) leads to an error in determining the effective field  $H_{\text{eff}} = H + H_A - 4\pi N_Z M_0$  the maximum value of which is less than 1% under our experimental conditions. The energy gap is taken equal to  $\Delta = p(H + H_A)$ . In the Heisenberg model, as in the band theory,  $p = 2\mu_0$ . In the phenomenological spin-wave theory  $p$  is an experimentally determined quantity. This parameter was determined from the functions  $|M'(H)|/M_0|_{T=\text{const}}$ .

### 3. COMPARISON WITH THEORY

The experimental function  $|M'(H)|/M_0|_{T=\text{const}}$  (see Fig. 2) has a maximum at  $H = H_{\text{max}}$ . It is obvious that the variation of the magnitude of  $M'$  with field  $H$  when  $H < H_{\text{max}}$  is due to the domain structure of the sample, whereas in fields  $H > H_{\text{max}}$  it is due to the change in the energy gap in the spin-wave spectrum with magnetic field. The parameter  $p$  entering in  $\Delta$  was determined by comparison of the experimental function  $|M'(H)|/M_0|_{T=\text{const}}$  with the theoretical one by the method indicated above for  $H > H_{\text{max}}$ . The correction to  $p$  for the multi-domain structure of the sample (see relations (20) and (21)) was found to be insignificant. The constant  $D$  given by formula (21) and the data of Fig. 2 amounts to  $\sim 10^6$  for nickel and  $\sim 0.2 \times 10^6$  for iron. The function  $|M'_g(H)|/M_0$  is shown by the dashed line in Fig. 2. In the calculations we used values of  $M'_g$  in which the correction for multi-domain structure was not greater than 10%. The criterion for the correct determination of  $p$  was linearity of the dependence of  $\mathcal{M}$  on the calculated quantity  $QT$ . As is seen in Fig. 4, this condition is satisfied by  $p = 1.2 \pm 0.2 \mu_0$  for nickel and  $p = 1.8 \pm 0.3 \mu_0$  for iron. The departure of  $p_{\text{exp}}$  from  $2\mu_0$  in the case of nickel was found to be outside the limits of experimental error.

With this parameter  $p$  it is possible to juxtapose the experimental data with the theoretical relations over the entire interval of temperature and field. Figures 5 and 6 show the results of this comparison. The "turnaround point" is clearly seen in the temperature curves. The straight dashed lines take into account primarily the data obtained in high magnetic fields, where the influence of  $M_0$ ,  $H_A$ ,  $D$ , and  $N_Z$  on the results for  $Q$  is a minimum. It is obvious that the magnetization of these metals follows the relation (16)

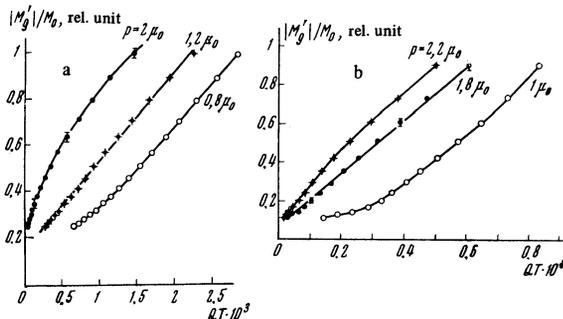


FIG. 4. Dependence of  $|M'_g|/M_0|_{T=\text{const}}$  on  $QT$  for samples of nickel and iron for different values of the parameter  $p$ : a—nickel,  $T = 0.9^\circ\text{K}$ ; b—iron,  $T = 1^\circ\text{K}$ .

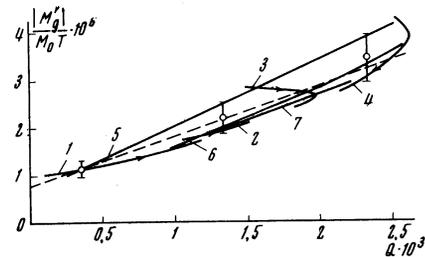


FIG. 5. Dependence of the quantities  $\mathcal{M}|_{H=\text{const}}$  and  $\mathcal{M}|_{T=\text{const}}$  on  $Q$  for nickel ( $\mathcal{M} \equiv |M'_g|/M_0 T$ ): curve 1— $H = 34$  kOe; 2—22 kOe; 3—11 kOe; 4—5 kOe (the arrows indicate direction of increasing temperature curve 5— $T = 0.9^\circ\text{K}$ , 6— $2^\circ\text{K}$ , 7— $4.3^\circ\text{K}$ ). The dashed line is drawn for calculation of the constants  $C$  and  $2A/M_0$ . The maximum possible random error is indicated.

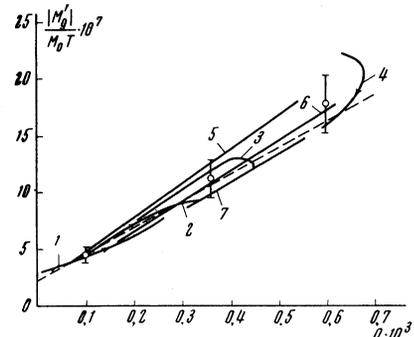


FIG. 6. Dependence of the quantities  $\mathcal{M}|_{H=\text{const}}$  and  $\mathcal{M}|_{T=\text{const}}$  on  $Q$  for iron: 1— $H = 34$  kOe, 2—22 kOe, 3—11 kOe, 4—3 kOe (the arrows show the direction of increasing temperature); 5— $T = 1^\circ\text{K}$ , 6— $2^\circ\text{K}$ , 7— $4.3^\circ\text{K}$ . The dashed line is drawn for calculation of the constants  $C$  and  $2A/M_0$ . The maximum possible random error is indicated.

for the corresponding values of the gap presented above.

The  $y$ -intercept of the line  $\mathcal{M}(Q)$  equals  $2A/M_0$ , and its slope is  $C$ . The constants determined this way are

$$\begin{aligned} \text{Ni: } C &= (1.1 \pm 0.3) \cdot 10^{-3}, \\ 2A/M_0 &= (0.8 \pm 0.2) \cdot 10^{-6}, \\ \text{Fe: } C &= (2.3 \pm 0.7) \cdot 10^{-3}, \\ 2A/M_0 &= (2.3 \pm 0.7) \cdot 10^{-7}. \end{aligned}$$

It should be emphasized that the quantity  $2A/M_0$  depends weakly on  $p$ . Thus in the case of nickel a change in  $p$  from 1.2 to  $2\mu_0$  brings about a 20% change in  $2A/M_0$ . However, the observed departures of  $\mathcal{M}(Q)$  from theory exceed the experimental error by more than three times.

It is easy to see that at  $T < 1^\circ\text{K}$  in fields greater than 30 kOe, the change in saturation magnetization is  $\delta M_g \approx AT^2$ . Using (16) and the above parameters  $A$ ,  $C$ ,  $p$ , it is possible to calculate  $\delta M_g(T, H)$  for  $T > 4.2^\circ\text{K}$  and compare it with the experiments in this temperature region. A similar comparison was made with Argyle's data<sup>[16]</sup>. The calculation was made for the sample configurations investigated in<sup>[6]</sup> with the contribution to  $M_g(T, H)$  resulting from spin-wave interactions taken into account. The relative changes of both the experimental and the theoretical values of  $\delta M_g T$  with temperature agree to within about 10%. The maximum departures of the absolute values of  $\delta M_g(T)$  are 30% for iron and 15% for nickel.

As mentioned above,  $Q(T, H)$  was calculated in the

spin-wave approximation. Note that at small values of the wave vectors  $k$  the continuous spin-wave spectrum turns into a discrete spectrum of magnetostatic modes and other vibrations<sup>[18]</sup>. Since when  $T \lesssim \Delta$  vibrations with minimum energy are excited, it is necessary in calculating  $Q(T, H)$ , generally speaking, to consider the detailed form of the excitation spectrum. However, in this temperature region it is true that  $AT^2 \gg CT^{3/2} I(\alpha, \beta)$ , and accounting for the magnetostatic modes and other vibrations obviously cannot affect the results very much.

Thus the dependence of  $M_g$  on  $T$  and  $H$  for  $T \lesssim 10^\circ\text{K}$  leads to the relations

$$\begin{aligned} \Delta M_g &\approx CT^{3/2} + AT^2 \quad \text{for } \Delta \ll kT, \\ \Delta M_g &\approx AT^2 \quad \text{for } \Delta \gg kT. \end{aligned} \quad (22)$$

In the region of higher temperature ( $T \sim 100^\circ\text{K}$ ) at realistic fields ( $H \sim 100$  kOe) the condition  $\Delta \ll kT$  is satisfied. Along with the terms of (22), Dyson terms become important in the temperature dependence of the magnetization. At these temperatures the term  $AT^2$  is comparable in magnitude with  $CT^{3/2}$ .

#### 4. DISCUSSION

Experiments to measure the magnetization of ferromagnetic metals show that the magnetic moment per atom  $\langle \mu \rangle$  does not equal an integral number of Bohr magnetons. For nickel and iron it is respectively  $0.65 \mu_0$  and  $2.22 \mu_0$ . Neutron diffraction studies of these metals, which confirm the magnetization data, show that  $\langle \mu \rangle$  is almost completely localized on the atom<sup>[19]</sup>. Since in these metals the magnetic moment at a lattice site does not equal a whole number of Bohr magnetons, the theory of localized spins is strictly speaking inapplicable. But for a description of spin-wave excitations it is possible to make use of a Heisenberg-type model with an effective magnetic moment  $\langle \mu \rangle$  at the site.

Studies of neutron diffraction<sup>[18]</sup>, x-ray scattering<sup>[20]</sup>, and nickel alloys<sup>[21]</sup> suggest that in metallic nickel the configuration of 3d electrons is intermediate between  $3d^{10}$  and  $3d^9$ . The ground state of the nickel atom in the metal can be considered approximately to be a superposition of states with configurations  $3d^{10}$  and  $3d^9$  (these  $3d^{10}$  and  $3d^9$  states are not exact states of the free atom). The atomic spin in state  $3d^{10}$  is 0 and in  $3d^9$  it is  $1/2$ . In this case, in examining the contribution of spin waves to the temperature dependence of magnetization, the Hamiltonian is approximately the Heisenberg Hamiltonian

$$H_0 = -\frac{1}{2} \sum_{l \neq m} J_{lm} s_l s_m,$$

where  $s_l$  and  $s_m$  are spin  $1/2$  operators for the  $l$ -th and  $m$ -th sites. It is easy to see that when the magnetic field is turned on an additional term appears

$$H_1 = 2\langle \mu \rangle \sum_i s_i^z.$$

The gap in the spin-wave spectrum is  $\Delta = 2\langle \mu \rangle(H + H_A) = 1.3 \mu_0(H + H_A)$ . This agrees with our measurements.

In metallic iron the equilibrium configuration of 3d electrons is between magnetic states with  $s$  equal to

1 and  $3/2$ . In this case the least separation between the levels of the system in a magnetic field (Zeeman splitting) is

$$\Delta E = 2\mu_0(H + H_A).$$

The effective gap width is determined by transitions between levels and is  $\Delta \approx \Delta E$ , at least for  $kT \sim 2\mu_0(H + H_A)$ .

Spin waves may also be excited by an electromagnetic field, as in ferromagnetic resonance. The data of ferromagnetic resonance shows that the gap must be  $\Delta \approx 2\mu_0(H + H_A)$  no matter what the metal<sup>[22]</sup>. If there is a localized magnetic moment in nickel<sup>[19]</sup> this would yield a value  $\langle \mu \rangle = \mu_0$ . Thus, magnetic diffraction of neutrons and ferromagnetic resonance lead to different results in the present conceptual framework. Our data agrees with neutron diffraction. In the localized spin and band models<sup>[1-3]</sup> we have  $\Delta = 2\mu_0(H + H_A)$ , which agrees with the data of ferromagnetic resonance, but it is difficult to understand the aforementioned results of neutron diffraction investigations in terms of these models.

The constant  $C$  in the temperature dependence of magnetization can be evaluated from the relation

$$C \approx M_0 / 2\pi^2 \Theta_c^{3/2}.$$

From this we have  $C = 1.6 \times 10^{-3}$  for nickel and  $C = 2.6 \times 10^{-3}$  for iron. These values are close to the ones we find experimentally.

It is easy to see that the treatment above is actually a modified localized spin model. In band models<sup>[2,3]</sup> it is difficult to explain so strong a localization of the magnetic moment. On the other hand, the term  $AT^2$  in the temperature dependence of magnetization does not arise in localized spin models but can be described in the band theory. An estimation of this term gives  $\sim \langle \mu \rangle a^{-3} (T/\epsilon_d)^2$ . According to Kondratenko<sup>[3]</sup>,  $\epsilon_d \approx \epsilon_F$ , where  $\epsilon_F$  is the Fermi level in the d band. In the Stoner model<sup>[2]</sup>,  $\epsilon_d \approx \Theta_C$  for  $\zeta_0 < 1$ , where  $\zeta_0$  is the relative magnetization at  $T = 0$ . Estimates from our data show that  $\epsilon_d \approx \Theta_C$ . In the temperature region  $T \lesssim 10^\circ\text{K}$  the heat capacity of a ferromagnetic metal in the band model is

$$c \sim \frac{\pi^2 kT}{3a^3 \epsilon_F} + \frac{1}{a^3} \frac{kT}{\epsilon_d} + \frac{1}{2\pi^{3/2} a^3} \left( \frac{T}{\Theta_c} \right)^{3/2}.$$

The first term in this expression represents the contribution of conduction electrons with Fermi energy  $\epsilon_F$ , the second, the contribution of d-band electrons, the third, the contribution of spin waves. If  $\epsilon_d \approx \Theta_C$ , then

$$c \approx a^{-3} T / \Theta_c.$$

This leads to the correct magnitude of the heat capacity of nickel and iron.

It follows that features from both the theory of localized spins and the band theory are required to describe the function  $\Delta M_g(T, H)$ . As is well known, it is difficult to describe the results of measurements of various physical parameters of ferromagnetic metals using only one theory. Obviously, the model of a ferromagnetic metal must contain the characteristics of both theories.

It seems to us that the above experimental results on the distribution of the magnetic moment in the cell

and its magnitude are important for the description of ferromagnetism. Starting from these data, it is natural to suppose that some of the d electrons complete transitions between d states of the atom and a state in the conduction band, where the magnetization is small compared to the lattice magnetization. This is described by a single-particle s-d exchange. It is easy to see that if a ferromagnetic consists of atoms of only one kind, other types of exchange cannot lead to an effective moment per atom that is not an integral number of Bohr magnetons. This type of s-d exchange creates the possibility of a single-particle d-d exchange, on the basis of which a ferromagnetic correlation of the spins of neighboring atoms becomes conceivable. It is clear that s-d and d-d exchange describe the transitions considered above between states with different configurations of d electrons.

This exchange concept is like the Anderson model<sup>[23]</sup>. On the basis of a certain generalization, Liu considered this model in relation to the ferromagnetism of iron and nickel<sup>[24]</sup>. Let us examine our results from the point of view of this model. Following Liu, it is easy to show that in the simplified Anderson model for  $\mu_0 H \ll U$  ( $U$  the Coulomb repulsion of the d electrons) the Zeeman splitting is  $\Delta E \approx 2\langle\mu\rangle H$ . Since  $U > 1$  eV, the condition  $\mu_0 H \ll U$  is practically always fulfilled. This model is directly applicable to nickel and the gap is

$$\Delta = \Delta E \approx 2\langle\mu\rangle H.$$

In the case of iron two electrons with parallel spins are evidently strongly coupled, their magnetic moment  $2\mu_0$  is described by Hund's rules, and only a portion of  $\langle\mu\rangle$  equal to  $0.2\mu_0$  is described by this model. The separation between the Zeeman levels in this case is  $2\mu_0 H$ . For the quasiclassical case Liu showed that the Heisenberg Hamiltonian is derivable from the Anderson Hamiltonian. It follows that the Anderson model agrees with our results in describing spin wave excitations. On the other hand, since "magnetic" d electrons participating in the exchange are described by wave functions that are a strong hybridization of d wave functions of the atom and wave functions of the conduction band, excitations of the Stoner type<sup>[2]</sup> are possible in this model. This can lead to an additional term  $AT^2$  in  $\Delta M_g(T, H)$ . However, the proof of existence of a term  $AT^2$  in this model, as well as of the general applicability of this model to ferromagnetism, requires further investigation.

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