## VARIATION OF THE SATURATION MAGNETIZATION OF FERROMAGNETIC SUBSTANCES A T HELIUM TEMPERATURES

N. V. ZAVARITSKII and V. A. TSAREV

Institute for Physics Problems, Academy of Sciences, U.S.S.R.

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The variation of the spontaneous moment  $M_s$  of iron and nickel is measured in the temperature range from 1.4 to 5°K. The temperature dependence of the quantity  $dM_s/dT$  is found to be determined by the magnetic field strength. In iron it satisfies the Bloch law only at low fields. The data are compared with the spin wave theory.

According to Bloch's theory, <sup>[1]</sup> the spontaneous magnetic moment  $M_s(T)$  of ferromagnets varies according to the following law in the region of low temperatures:

$$M_{s} = M_{0} \left( 1 - CT^{s/2} \right). \tag{1}$$

This theory has been subjected to numerous refinements (see, e.g., [2]). In particular, it has become clear that in the region of very low temperatures and high magnetic fields deviations from the law (1) should be expected on account of the effect of the magnetic field on the energy spectrum of the spin waves.

A number of papers have been concerned with the experimental investigation of ferromagnets in the region of low temperatures. Fallot, <sup>[3]</sup> Kondorskiĭ and Fedotov, <sup>[4]</sup> Kondorskiĭ, Rode, and Hofman, <sup>[5]</sup> and Foner and Thompson <sup>[6]</sup> have measured the spontaneous magnetization of iron, nickel, and a number of their alloys. According to their data, the dependence  $M_{\rm S}(T)$  in the temperature region 20 to 80°K is better approximated by the  $T^{3/2}$  law than by  $T^2$ . Kouvel <sup>[7]</sup> separated the magnetic part of the heat capacity of a ferromagnetic dielectric at helium temperatures and found that it varied like  $T^{3/2}$ .

We have undertaken the direct measurement of the dependence of the spontaneous magnetization of ferromagnets on temperature in the interval 1.4 to  $5^{\circ}$ K at various magnetic fields.

Since the variation of the magnetic moment  $M_S$ in this temperature interval is not large (0.01%), the quantity  $dM_S/dT$  was determined from the oscillations in the magnetic moment of the sample brought about by oscillations in its temperature. A similar method was first employed by Shal'nikov and Sharin<sup>[8]</sup> to investigate the temperature dependence of the depth of penetration of magnetic



FIG. 1. Basic circuit for the measurements. 1- generator, 2- measuring coil, 3- amplifier, 4- potentiometer.

fields in superconductors. Preliminary measurements by this method carried out by one of the authors earlier showed that the temperature dependence of the saturation magnetization of permalloy in fields of 100-150 Oe did not contradict the Bloch law.

The basic circuit of our apparatus is shown in Fig. 1. The sample S, in the form of a cylinder of diameter 0.18 cm and length 3 cm, was placed in the field of an electromagnet H. A signal of frequency  $\omega$  from a generator is applied to the heater W mounted on the sample. As a result, the temperature of the sample is caused to oscillate at the frequency  $2\omega$ . To measure the amplitude of the oscillations  $\Delta T$  the signal from the thermometer T was applied to the capacitive input of a tuned amplifier 3 with a dc ammeter A at the output. The amplifier was tuned to a frequency of 9.2 cps and had a band width of 0.8 cps. The average temperature of the sample was measured by the thermometer T by means of the potentiometer 4.

The signal from the coil 2, which is proportional to the change in the magnetic moment of the sample  $\Delta M$ , was applied to the transformer input of the same amplifier 3. In order to decrease the effect of fluctuations of the external field on the meter indications, a second coil was mounted in which a sample similar to the one under study was placed. The samples and coils were mounted in a vacuum container placed in a helium dewar.

Nickel and iron were chosen for the investigation. The impurity content of the nickel was 0.1%and of the iron, less than 0.03%. The samples were annealed in vacuum at  $1000^{\circ}$ C for about 3-4hr and cooled in the furnace.

Two means of creating temperature oscillations in the sample were employed. For iron, in which the thermal wavelength is much larger than the dimensions of the sample, the arrangement seen in Fig. 2a was used. One end of the sample was soldered to the copper rod 1, which was in contact with the wall of the container. A film heater W and thermometer T, prepared in the manner described earlier, [9] were arranged on the sample.<sup>1)</sup>



FIG. 2. Distribution of the amplitude of the temperature oscillations  $\Delta T$  and of the mean temperature T of the iron (a) and nickel (b) samples.

The change in the amplitude of the temperature oscillations along the sample was measured with three thermometers. This distribution agreed with calculations obtained under the assumption that the temperature conductivity of iron is 188 cm<sup>2</sup>/sec (the length of the temperature wave at 9.2 cps is 16 cm). This is indicated by the dashed curve in Fig. 2a. From these data and the amplitude of the temperature oscillations of the thermometer T it was possible to calculate the mean amplitude of the temperature oscillations of the sample.

Another method of creating temperature oscillations was used for nickel, whose temperature conductivity is 9.5 cm<sup>2</sup>/sec (temperature wavelength 3.6 cm) (see Fig. 2b.). The heater was bifilarly wound of constantan wire,  $30 \mu$  in diameter, over the whole length of the sample with a pitch of about 0.17 cm; the pitch was reduced by about 15% at the ends of the sample. Both ends of the sample were in contact with the container walls via the duraluminum rods 2. This material was selected in order that the phase of the temperature wave reflected from the cold surface would coincide with the phase of the incident wave.

The distribution of the amplitude of the temperature oscillations and the mean temperature along the nickel sample are also shown in Fig. 2b. This distribution, as in the case of iron, also was independent of the mean temperature of the sample.

Controlled experiments, in which the ferromagnetic samples were replaced by ones of tin, showed that no spurious effects were caused by this method of creating temperature oscillations in the sample.

The calculation of the quantity dM/dT was carried out according to the relation

$$\frac{dM}{dT} = \frac{pn}{b} \frac{\Delta u^M}{\Delta u^T} \frac{dV(T)}{dT}, \qquad (2)$$

where  $\Delta u^M$  and  $\Delta u^T$  are the signals at the output of the amplifier when measuring the oscillations in magnetic moment and temperature, respectively, p is the coefficient of proportionality between the magnetic moment oscillations of the sample  $\Delta M$ and the signal at the amplifier output  $\Delta u^{M}$ , n is the reading of the amplifier input divider for temperature measurements, b is a corrective factor for the change in amplitude of the temperature oscillations along the sample, and dV/dT was calculated for each measurement according to the thermometer calibration determined during the course of a run (V is the thermometer voltage). In Eq. (2) the characteristics of the amplifier enter through the constant p. Measurements of the magnetic and thermal amplitudes were made successively through one channel; the amplification coefficient came out of their ratio  $\Delta u^{M}/\Delta u^{T}$ . Errors in determining  $\Delta u^{M}$  amounted to 2 and 5% at the temperatures 5 and 2°K, respectively. At temperatures near 1.5°K the noise level reached 20% of the signal. Errors in measurement of the thermal amplitudes  $\Delta u^{T}$  did not exceed 2 and 5% in the intervals 2-5 and 1.5-2°K, respectively. The error in calculating dV/dT did not exceed 5%.

Besides the random errors listed above a systematic error of about 10% also enters into the final result, on account of inaccuracy in the determination of the constants p, n, and b. In the case of nickel a correction of about 10% was introduced owing to the non-uniform distribution of mean temperature over the sample investigated. Errors in determining this correction could have caused some distortion in the temperature dependence of the quantity dM/dT.

The sum of all errors in the determination of the absolute value of dM/dT amounted at 3°K to 15-20% for iron and 20-25% for nickel.

<sup>&</sup>lt;sup>1</sup>)It was established in an auxiliary experiment that the difference in the amplitudes of the temperature oscillations of the thermometer and the sample at the frequency of the measurement did not exceed 2-3%.

## **RESULTS AND DISCUSSION**

The magnetic susceptibility of nickel was measured at 4.2°K in the field interval 1–11 kOe. Its variation agrees with the law  $\kappa = QH^{-3}$ , where  $Q \approx 10^8$ . Because of this, the field dependence of the magnetic moment has the form

$$M = M_s (1 - g H^{-2}), (3)$$

where  $g \cong Q/2M_0 \sim 10^5$  (for nickel  $M_0 \cong 510$  cgs emu<sup>[4]</sup>). Equation (3) is usually used to describe the approach of the magnetic moment to saturation. The factor g is associated with the anisotropy constant, which depends on temperature.

It follows from Eq. (3) that

$$\frac{dM}{dT} = \frac{dM_s}{dT} \left(1 - \frac{g}{H^2}\right) - \frac{M_s}{H^2} \frac{dg}{dT}.$$
 (4)

Figure 3 shows dM/dT vs. magnetic field for nickel, in relative units. As can be seen from the graph there is a strong change in dM/dT in fields from 1 to 5 kOe. In this field interval  $g/H^2 \ll 1$ ; hence the change in dM/dT can be ascribed only to the change in the term dg/dT in Eq. (4). The coincidence of the dependence for different temperatures shows that dg/dT is proportional to  $dM_g/dT$ .



FIG. 3. Dependence of dM/dT on magnetic field for nickel: 0 - at 2°K,  $\Delta$  - at 4.2°K.

The decrease in the magnitude of dM/dT in fields greater than 6 kOe is apparently explained by the change with magnetic field in the quantity  $dM_S/dT$  itself, i.e., by the para-process effect.

The field dependence of dM/dT for iron is presented in Fig. 4. The value of the constant g in Eq. (3) obtained from magnetic susceptibility measurements equals in this case  $2 \times 10^4$ , if one takes  $M_0 = 1750$  cgs emu.<sup>[4]</sup> From this it follows that in fields greater than 500 Oe, gH<sup>-2</sup>  $\ll 1$ . It is obvious that the variation of dM/dT in the interval 500-2000 Oe can be attributed principally to the second term in Eq. (4). As in the case of nickel, dg/dT is proportional to dM<sub>s</sub>/dT. A numerical comparison shows that for both metals

$$\frac{1}{g}\frac{dg}{dT} = B\frac{1}{M_s}\frac{dM_s}{dT}$$
, where  $B \cong 10$ .

FIG. 4. Dependence of dM/dT on magnetic field for iron:  $O - at 2^{\circ}$ K,  $\Delta - at 4.2^{\circ}$  K.



In fields less than 500 Oe the dependence of dM/dT on H has a more complicated character, similar to that observed earlier in permalloy. This agrees qualitatively with the ideas about the temperature dependence of the magnetization in the region of reversal and incipient domain rotation.

In fields greater than about 2-3 kOe, the second term on the right-hand side of Eq. (4) becomes negligibly small, and the variation of dM/dT is obviously attributable to the variation of dM<sub>s</sub>/dT, i.e., the para-process effect. As can be seen from Fig. 4, in this case the relative change with field of the quantity dM/dT depends on temperature, in contrast with the low-field region. The mean susceptibility of the para-process in the interval 2–10 kOe is about  $10^{-6}$  for iron, which is less than the higher limit of the magnitude obtained by Kapitza<sup>[10]</sup> in experiments in very high fields at room temperature.

Figures 5 and 6 show the temperature dependence of the quantity  $dM/M_0 dT$  for nickel and iron

FIG. 5. Temperature dependence of  $dM/M_0dT$  for nickel: • – experimental values in a field H = 10.8 kOe; curves – theoretical dependences: broken according to Eq. (5), solid according to Eq. (8a). (C = 10<sup>-5</sup>;  $\mu = 0.22 \times 10^{-20}$ erg/G.)



FIG. 6. Temperature dependence of dM/M<sub>o</sub>dT for iron. Experimental values in fields of 2 kOe (0) and 10.8 kOe ( $\bullet$ ). Theoretical dependences represented by curves: broken-according to Eq. (5), solid-according to Eq. (8a). (C = 3.7 × 10<sup>-6</sup>;  $\mu$  = 1.1 × 10<sup>-20</sup> erg/G.)



in different magnetic fields. It follows from the Bloch law that

$$dM/M_0 dT = \frac{3}{2} CT^{1/2}.$$
 (5)

We shall compare our results with this equation. For nickel the dependence of  $dM/M_0 dT$  does not differ from that derived from the Bloch law in the interval 3–5°K; with iron a similar agreement takes place only in a field of 2 kOe; at lower temperatures or higher magnetic fields deviations from this law are observed, and  $dM/M_0 dT$  decreases with temperature faster than  $T^{1/2}$ .

Whereas for nickel and iron in a field of 2 kOe this deviation lies within the limits of maximum experimental error, for iron in a field of about 10 kOe it goes outside the limits of all possible errors.

We can attempt to correlate our results with the theory of spin waves in ferromagnets. In an external field H the energy of a spin wave has the form [2]

$$\boldsymbol{\varepsilon}_{K} = AK^{2} + \boldsymbol{\mu}H, \qquad (6)$$

where K is the wave vector of the spin wave, and A is a quantity proportional to the exchange integral. Terms proportional to  $M_0$  have been omitted from Eq. (6) since according to experiment the dependence of  $dM/M_0 dT$  in small fields is close to the Bloch law, which follows from Eq. (6) at H = 0.

Using Eq. (6), we obtain the following expressions for the spontaneous moment  $M_s$ :

$$M_{s} = M_{0} \left[ 1 - \frac{C}{\zeta^{(3/2)}} \frac{4}{\sqrt{\pi}} \left(\frac{A}{K}\right)^{s/2} \int_{0}^{\infty} \frac{K^{2} dK}{\exp\left[(AK^{2} + \mu H)/kT\right] - 1} \right]$$
$$= M_{0} \left[ 1 - \frac{C}{\zeta^{(3/2)}} T^{s/2} \sum_{n=1}^{\infty} n^{-s/2} \exp\left(-n\frac{\mu H}{kT}\right) \right]$$
(7)

or (see the appendix to Schafroth's paper<sup>[11]</sup>)

$$M_{s} = M_{0} \left\{ 1 - \frac{CT^{3/2}}{\zeta \left(\frac{3}{2}\right)} \left[ \zeta \left(\frac{3}{2}\right) - 2\Gamma \left(\frac{1}{2}\right) \left(\frac{\mu H}{kT}\right)^{1/2} - \zeta \left(\frac{1}{2}\right) \frac{\mu H}{kT} \dots \right] \right\}$$
(8)

$$\left|\frac{dM_{s}}{M_{0}dT}\right| = \frac{3}{2}CT^{1/2}\left[1 - \frac{4}{3}\frac{\Gamma(1/2)}{\zeta(3/2)}\left(\frac{\mu H}{kT}\right)^{1/2} - \frac{1}{3}\frac{\zeta(1/2)}{\zeta(3/2)}\frac{\mu H}{kT}\dots\right]$$
(8a)

 $[\zeta(x) \text{ is the Riemann zeta-function}].$ 

Our experimental data were compared with Eq. (8a). The parameters C and  $\mu$  were regarded as quantities subject to determination. In the case of iron both the field dependence of dM/dT and the temperature dependence of dM/M<sub>0</sub> dT were used. In the latter case the results obtained at temperatures higher than 2°K were mainly used; the accidental errors of measurement are not yet very large in this region. It was found that both these

dependences are satisfactorily described by Eq. (8a) with  $C = 3.7 \times 10^{-6}$  and  $\mu = 1.1 \times 10^{-20} \text{ erg/G} \cong 1.2 \,\mu_0$ , where  $\mu_0$  is the Bohr magneton. For nickel a much better agreement of Eq. (8a) with the experimental data is found with  $C = 10^{-5}$  and  $\mu = 0.22 \times 10^{-20} \text{ erg/G} \cong 0.25 \,\mu_0$ .

Obviously, the accuracy of determining the constants C and  $\mu$  depends on various factors. The quantity C is related to the absolute value of  $dM/M_0 dT$ , and its accuracy is limited mainly by the systematic errors, the magnitude of which in the above cases can reach 10–15%. The magnitude of  $\mu$  is determined from the relative change in  $dM/M_0 dT$ , and its accuracy is limited mainly by the accidental errors.<sup>2)</sup> In the case of iron the error in the determination of  $\mu$  reaches 25%. For nickel the possible error is still larger. In this case it can be asserted that  $\mu$  is positively less than 0.4  $\mu_0$ .

The values we obtained for C in the Bloch law agree with the results of determinations of this quantity at temperatures higher than  $20^{\circ}$ K (see the table).

Authors	Constant 10 <sup>6</sup> C		Measure- ment
	Iron	Nickel	interval, °K
This work Fallot <sup>[3]</sup> Kondorskif and Fedotov <sup>[4]</sup> Foner and Thompson <sup>[6]</sup>	3.70 3,5 4.18	$     \begin{array}{c}       10 \\       8.6 \\       9 \\       6.0     \end{array} $	$ \begin{array}{r} 1,5-5\\20-80\\20-80\\40-120\end{array} $

Apparently, there is a lack of investigations of the variation of the energy spectrum of spin waves in a magnetic field and a lack of values for  $\mu$  derived thereby. According to the microscopic theory of spin waves <sup>[2,12]</sup> values of the constant  $\mu$ in Eq. (6) equal to  $2\mu_0$  can be expected. The values we obtained are considerably less than this. They are different for nickel and iron.

Thus, the variation of the saturation magnetization of the ferromagnets investigated here at helium temperatures can be satisfactorily explained by the spin wave theory. However, in order to get this agreement it is necessary to assume an anomalously small value for the constant  $\mu$  which determines the field dependence of the spin-wave energy spectrum.

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<sup>&</sup>lt;sup>2)</sup>Of course, the magnitude of  $\mu$  is obtained in the approximation that terms involving M<sub>0</sub> are omitted from Eq. (6).

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