

## Temperature dependence of the magnetization of nickel using $^{61}\text{Ni}$ NMR

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The temperature dependence of the hyperfine field of  $^{61}\text{Ni}$  in pure nickel has been measured in the range 4.2–292 K using continuous-wave NMR. The hyperfine field is a measure of the magnetization in this temperature range since the hyperfine coupling constant at constant volume is shown to be essentially independent of the temperature. The present measurements confirm the difference, noted by Aldred, between the spin-wave parameters calculated from neutron scattering and magnetization measurements. This discrepancy is discussed in terms of the Stoner theory of single-particle excitations and (because of the improved accuracy attainable using NMR) it is possible to show that the simple expression for the magnetization for a strong ferromagnet,  $M \sim \exp(-\Delta/k_B T)$ , is unsatisfactory, and a large term proportional to  $T^2$  definitely exists. It is concluded that although nickel is a strong ferromagnet, the value of the gap parameter  $\Delta$  is an order of magnitude smaller than has been estimated from magnetization measurements.

### I. INTRODUCTION

The existence of spin waves in the  $3d$  metals has been established by both theory and experiment. The low-temperature magnetization of iron, for example, has been found to have the form

$$(M_0 - M_T)/M_0 = B_0 T^{3/2}, \quad (1)$$

where  $B_0$  is a constant (in agreement with spin-wave theory) and the values of  $B_0$  found from magnetization,<sup>1,2</sup> neutron-scattering<sup>3</sup> and hyperfine-field measurements<sup>4</sup> are identical within experimental error. At higher temperatures the coefficient  $B_0$  in Eq. (1) becomes a function of temperature and will be written  $B_T$ . The value of  $B_T$  for iron for  $T \leq 300$  K ( $0.3T_C$ ) obtained from magnetization measurements is in good agreement with the value ( $B_T^n$ ) calculated from neutron-scattering measurements of the spin-wave stiffness coefficient, and it is justified to conclude that only spin-wave excitations are important for iron.

However, the temperature dependence of the magnetization of nickel appears to be quite different from that of iron. The value of  $B_0$  found from magnetization and hyperfine-field measurements will be shown to be in good agreement, but  $B_0^n$  is some 40% smaller than  $B_0$ . Aldred<sup>5</sup> defined the spin-wave contribution to the magnetization to be

$$[(M_0 - M_T)/M_0]_{\text{sw}} = B_T^n T^{3/2} \quad (2)$$

and examined the difference between his magnetization measurements and Eq. (2) searching for evidence of single-particle (Stoner) excitations. In the Stoner theory,<sup>6</sup> which is discussed more fully in Sec. II, the  $3d$  band of a transition metal at temperatures below the Curie point is split into two subbands each containing electrons of one spin direction. The subbands are shifted in energy by

an amount proportional to the magnetization. A strong ferromagnet is defined as a material in which this energy shift is sufficiently large for the  $d_{\uparrow}$  at 0 K to be completely filled with electrons and only  $d_{\downarrow}$  electrons are present at the Fermi surface. A minimum energy,  $\Delta$ , has to be supplied to a  $d_{\uparrow}$  electron to promote it to the  $d_{\downarrow}$  band. When  $\Delta$  is large the temperature dependence of the magnetization is proportional to  $e^{-\Delta/k_B T}$ . A weak ferromagnet has both spin bands partially full and Stoner showed that, under certain circumstances, the magnetization decreased as  $T^2$ .

Aldred found that a better fit to his measurements was obtained using the  $T^2$  term instead of the exponential term, but neither fit was particularly good. It is difficult to measure the magnetization with sufficient accuracy if the difference from Eq. (2) is to exceed the experimental errors over a wide temperature range below room temperature and, as pointed out by Aldred, it was not possible to decide whether nickel was a strong or weak ferromagnet on the basis of his measurements. Schlosser<sup>7</sup> has indeed suggested that the simple equation

$$(M_T/M_0)^2 = 1 - \alpha(T/T_C)^2 \quad (3)$$

is a satisfactory fit to the magnetization measurements on both iron and nickel up to  $0.45T_C$ , where  $\alpha$  is equal to 0.56. This equation is of the form expected for a weak ferromagnet but  $\alpha$  would then have to be equal to or greater than unity according to Stoner's theory.

In view of the limited accuracy of the present magnetization measurements it is worthwhile re-examining the possibility of using the hyperfine field of nickel as a measure of the magnetization. Using continuous-wave NMR, the hyperfine field of iron and nickel may be measured an order of

magnitude more accurately than the magnetization. In iron,<sup>4</sup> however, only at low temperatures, where  $B_T$  is equal to the constant  $B_0$ , the two measurements had the same temperature dependence; the hyperfine coupling constant  $A_T$  defined by the equation

$$\nu_T/\nu_0 = A_T M_T / M_0 \quad (4)$$

is a function of temperature. The hyperfine coupling constant of nickel at constant volume is known<sup>8</sup> to be a weak function of temperature, and in Sec. V it is shown that within the accuracy of the magnetization measurements, is constant up to 300 K.

The results of continuous-wave NMR measurements on <sup>61</sup>Ni in nickel are now summarized. In the low-temperature region (4.2–23 K) our value of  $B_0$  is in good agreement with the value obtained from the magnetization.<sup>1</sup> The difference between  $B_0$  and  $B_0^v$  is therefore confirmed. At higher temperatures the measurements are reduced to constant volume and, after subtracting Eq. (2), are examined for evidence of single-particle excitations. The fit to the measurements near room temperature is in good agreement with the values of the Stoner coefficients calculated by Aldred, confirming that the hyperfine coupling constant is independent of temperature, but the greater accuracy of the NMR measurements clearly showed that these values are unsatisfactory at lower temperatures. Nickel appears to be on the border line between weak and strong ferromagnetism. Therefore, the simple equations used by Aldred are no longer applicable and the Stoner equations have to be solved numerically. The best fit to the measurements suggests that nickel is a strong ferromagnet with  $\Delta/k_B \lesssim 18$  K, but it is exactly in this region of small  $\Delta$  that it is most difficult to evaluate the importance of the various approximations implicit in the Stoner theory of ferromagnetism. The best test of the magnetism of nickel, the de Haas–van Alphen effect,<sup>9</sup> seems to definitely establish that nickel is a strong ferromagnet in agreement with our conclusion, but it is unlikely that more definite results can be reached from magnetization studies until the theory of band ferromagnetism at the transition between weak and strong ferromagnetism has been extended.

In Sec. II a brief review of the theory of spin waves and single-particle excitations in ferromagnetic metals will be given; in Sec. III the experimental technique is discussed. The measurements are analyzed purely in terms of spin-wave theory in Sec. IV; a review of the temperature dependence of the hyperfine coupling constant of

nickel is given in Sec. V and the fit to Eqs. (4) and (5) is discussed in Sec. VI.

## II. THEORY OF MAGNETIZATION IN 3d METALS

The theory of spin-wave and single-particle contributions to the magnetization of ferromagnetic metals was begun in the 1930's and at that time the theories were treated as mutually exclusive. Spin waves were associated with the localized (Heisenberg) model of exchange interactions and the single-particle excitations corresponded to a band scheme. However, the clear-cut distinction between the two models breaks down when electron correlations are considered, as emphasized by Herring<sup>10</sup> in his extensive review of exchange interactions between itinerant electrons. It is then possible to obtain spin waves within the band theory and the main support for Heisenberg exchange in nickel is removed. Lowde and Windsor<sup>11</sup> have shown that the neutron-scattering data for nickel is inconsistent with a model of localized exchange interactions and the temperature dependence of the spin-wave stiffness coefficient discussed below is also a band effect.

The temperature dependence of the magnetization of nickel must, therefore, be due to both spin-wave and single-particle excitations. It seems to be generally held at present that these two terms contribute independently to the magnetization of a strong ferromagnet, as it is assumed by Aldred and in the present paper, but there is no direct experimental evidence for this view.

The spin-wave theory is reviewed in an article by Keffer<sup>12</sup> and in Ref. 4 on NMR in iron. The coefficient  $B_T$  in Eq. (1) is given by

$$B_T = (2.612 g \mu_B / M_0) (k_B / 4\pi D)^{3/2}, \quad (5)$$

where  $g$  is the Landé splitting factor,  $\mu_B$  the Bohr magneton,  $k_B$  Boltzmann's constant, and  $D$  the spin-wave stiffness coefficient.  $D$  may be measured by neutron-scattering experiments and according to Aldred's analysis<sup>5</sup> the current best values for nickel are 555 meV Å<sup>2</sup> at 4.2 K and 455 meV Å<sup>2</sup> at 295 K with an accuracy of (1–2)%. The temperature dependence of  $D$  should be of the form

$$D = D_0 - D_1 T^{5/2} \quad (6)$$

(e.g., Marshall and Lovesey<sup>13</sup>), therefore using the above experimental values  $D_0 = 555$  meV Å<sup>2</sup> and  $D_1 = 6.69 \times 10^{-5}$  meV Å<sup>2</sup> K<sup>-5/2</sup>. Aldred uses the form  $D_0 - D_1 T^2$ , but the difference is not very important up to 300 K. The equation for the spin-wave magnetization of nickel is therefore

$$\left(\frac{M_0 - M_T}{M_0}\right)_{\text{sw}} = \frac{4.48 \times 10^{-6} T^{3/2}}{(1 - 1.20 \times 10^{-7} T^{5/2})^{3/2}} = B_T^n T^{3/2} \quad (7)$$

and  $B_0^n$  is equal to  $4.48 \times 10^{-6} \text{ K}^{-3/2}$ . The coefficient  $B_T$  must be modified if a gap exists in the spin-wave spectrum. When the gap temperature  $T_g$  is much less than the lowest experimental temperature the form

$$B_T [1 - 1.355(T_g/T)^{1/2} \dots] \quad (8)$$

is sufficient, so the leading correction term in the magnetization is linear in the temperature. The analysis of the NMR measurements of iron<sup>4</sup> showed that the temperature dependence of the hyperfine field of the atoms within domain walls, which contribute to the NMR signal, was consistent with a gap equal to the anisotropy field

$$k_B T_g = g \mu_B H_A \quad (9)$$

and it will be seen in Sec. IV that this is also the case for nickel.

The contribution of single-particle excitations to the magnetization of a ferromagnetic metal was first studied by Stoner.<sup>6</sup> Stoner considered the case of a nearly full  $3d$  parabolic band. The exchange energy was represented by a term proportional to the square of magnetization, as in the Weiss molecular-field theory, leading to a splitting of the  $d_{\uparrow}$  and  $d_{\downarrow}$  subbands proportional to the magnetization. The application of Fermi-Dirac statistics to the subbands then lead to an expression for the spontaneous magnetization as a function of temperature which can be evaluated numerically.

The basic variable of the Stoner theory is the ratio  $k_B \Theta' / \epsilon_0$ , where  $2k_B \Theta' \zeta$  is the energy splitting of the two subbands for a relative magnetization  $\zeta$ , and  $\epsilon_0$  is the Fermi energy in the absence of exchange interactions. The criteria for ferromagnetism in the Stoner theory is that  $k_B \Theta' / \epsilon_0 > \frac{2}{3}$ , but for the range  $2^{-1/3} > k_B \Theta' / \epsilon_0 > \frac{2}{3}$  both spin bands are only partially full and the system is said to be a weak ferromagnet. It has not always been appreciated that a simple  $T^2$  law for the temperature dependence of the magnetization occurs in fact for only a limited range of weak ferromagnetism, roughly speaking for  $0.7 > k_B \Theta' / \epsilon_0 > \frac{2}{3}$  and that for  $2^{-1/3} > k_B \Theta' / \epsilon_0 > 0.7$  the coefficient of the  $T^2$  term also has a complicated temperature dependence. Stoner gave the  $T^2$  law in the form of Eq. (3), but it is more usually written in the low-temperature form

$$[(M_0 - M_T)/M_0]_{\text{sp}} = A' T^2. \quad (10)$$

When  $k_B \Theta' / \epsilon_0 > 2^{-1/3}$  the  $d_{\uparrow}$  band is completely full at 0 K and the system is said to be a strong ferro-

magnet. It is again possible to write a simple expression for the magnetization, at low temperature provided that the system is not too close to the boundary with a weak ferromagnet

$$[(M_0 - M_T)/M_0]_{\text{sp}} = B' T^l e^{-\Delta/k_B T}, \quad (11)$$

where  $B'$  is a constant,  $l$  depends upon the form of the density of states curve, and  $\Delta$  is the energy required to promote an electron at the top of the  $d_{\uparrow}$  band into the  $d_{\downarrow}$  band. The constant  $l$  is found to be equal to  $\frac{3}{2}$  for a parabolic band and seems to be little changed for a more realistic band structure for nickel.<sup>14</sup>

In the region where  $k_B \Theta' / \epsilon_0 \approx 2^{-1/3}$  it is not possible to write simple expressions of the form of Eqs. (10) and (11), but it is clear from Stoner's calculations that as  $\Delta$  goes to zero a transition must take place between a  $T^{3/2}$  term and the  $T^2$  form for a weak ferromagnet and it will be seen later that a sum of such terms is in fact adequate to describe the present measurements on nickel.

The Stoner theory has been discussed in some detail because the limited validity of Eqs. (10) and (11) has not always been appreciated. A further complication is that in a weak ferromagnet it has been shown by Herring<sup>10</sup> that Eq. (10) is modified in the presence of spin waves to read

$$[(M_0 - M_T)/M_0]_{\text{sp}} = A' T^2 - C' T^{5/2}, \quad (12)$$

where  $C'$  is probably positive for nickel.

The attractive feature of the Stoner theory is its simplicity, but the calculated values of parameters such as  $A'$  are found to be too large to give quantitative agreement with experiment. The problem therefore is to study the difference between the measured magnetization of nickel as a function of temperature and that predicted by Eq. (7) in terms of powers of the temperature which even in the simplest cases have been seen to contain  $T^{3/2}$ ,  $T^2$ ,  $T^{5/2}$ , and an exponential. Since at 300 K the magnetization of nickel has decreased by about 7% of its value at 0 K, and since roughly half of this decrease is attributed to spin waves, it is not surprising that extremely high accuracy is required before a choice can be made between the various possibilities. A further complication is that many-body effects<sup>15</sup> seem to act to reduce  $\Delta$ , i.e., to move a strong ferromagnet towards the regime of weak ferromagnetism, and therefore real ferromagnets may not exist in which  $\Delta$  is sufficiently large for Eq. (11) to be valid.

### III. EXPERIMENTAL DETAILS

The nickel sample (Johnson Matthey sponge, 99.9985% nickel) was annealed under hydrogen and cooled slowly to room temperature. The NMR

measurements were made in a continuous-flow helium cryostat that has already been described.<sup>16</sup> The temperature was measured using a germanium resistance thermometer over the range 4.2–77 K, and between 77 K and room temperature a Au(Fe)-chromel thermocouple was used. The thermocouple was calibrated against a platinum resistance thermometer in a separate experiment.

The distribution of hyperfine fields was measured using the technique of adiabatic fast passage that has been described in detail previously.<sup>4,17</sup> The width of the <sup>61</sup>Ni distribution of hyperfine fields at low temperature<sup>17</sup> ( $\approx 120$  kHz) limited the accuracy of the measurements, but by averaging four passes through the line it is thought the results shown in Table I are accurate to about 1.5 kHz. The third column in Table I shows the estimated

TABLE I. NMR measurements of the hyperfine field in nickel. The second column shows the measurements at atmospheric pressure and the third column the estimated correction ( $\delta\nu_\nu$ ) to constant volume. The fourth and fifth columns show the correction for a gap in the spin-wave spectrum ( $\delta\nu_A$ ) as discussed in the text.  $\delta\nu_\nu$  must be added, and  $\delta\nu_A$  subtracted, from the second column to find the hyperfine field for constant volume and zero-spin-wave gap.

$T$ (K)	$\nu_T$ (MHz)	$\delta\nu_\nu$ (kHz)	$\delta\nu_A^a$ (kHz)	$\delta\nu_A^b$ (kHz)
4.2	28.455	...	1	
6.4	28.454	...	1	...
7.9	28.454	...	1	...
9.9	28.453	...	2	...
13.1	28.450	...	2	1
14.8	28.448	...	3	2
16.8	28.446	...	3	2
17.1	28.445	...	3	2
18.9	28.443	...	3	2
20.7	28.441	...	4	2
23.2	28.437	...	4	3
26.8	28.431	...	5	3
29.5	28.426	...	5	3
31.7	28.422	...	5	3
35.1	28.415	...	6	3.5
40.6	28.401	1	7	4
45.4	28.388	2	7	4.5
52.7	28.366	4	8	5
69.1	28.306	10	11	7
77.5	28.267	14	12	7
106.8	28.112	35	15	9
126.7	27.988	56	16	9.5
168.6	27.671	107	17	11
209.8	27.279	161	16	10
250.7	26.805	217	14	8
292.4	26.216	275	13	8

<sup>a</sup> Assuming only spin-wave contribution to decrease of magnetization.

<sup>b</sup> Assuming spin-wave contribution to be calculated from neutron data.

correction ( $\delta\nu_\nu$ ) that has to be added to the measurements to reduce them to constant volume.<sup>18</sup> The fourth column shows the correction to be subtracted from the measurements for a gap in the spin-wave spectrum equal to the anisotropy field if only spin-wave terms are considered, and the last column shows the correction if the spin-wave term is defined by  $B_T^n$ .

#### IV. SPIN-WAVE ANALYSIS

The measured values of the hyperfine field in the range 4.2–23 K are shown as a function of  $T^{3/2}$  in Fig. 1. It is clear that a gap exists in the spin-wave spectrum and a least-squares analysis shows that the value of the gap (0.46 K) is consistent with that calculated from Eq. (11) using the anisotropy measurements of Franse and de Vries.<sup>19</sup> The corrected measurements then follow the equation

$$(\nu_0 - \nu_T)/\nu_0 = (7.38 \pm 0.11)10^{-6}T^{3/2},$$

$$\nu_0 = 28.457 \text{ MHz.} \quad (13)$$

The coefficient of  $T^{3/2}$  is in excellent agreement with the value  $7.5 \times 10^{-6} \text{ K}^{-3/2}$  deduced from the low-temperature magnetization measurements of Argyle *et al.*,<sup>1</sup> but it is clearly inconsistent with the neutron result given in Eq. (7).

A further indication that the simple spin-wave analysis is incorrect is found when the measurements up to room temperature are examined. In the case of iron a single term  $T^{5/2}$  is sufficient to fit the NMR measurements and produced a value for the coefficient of  $T^{3/2}$  consistent with the direct

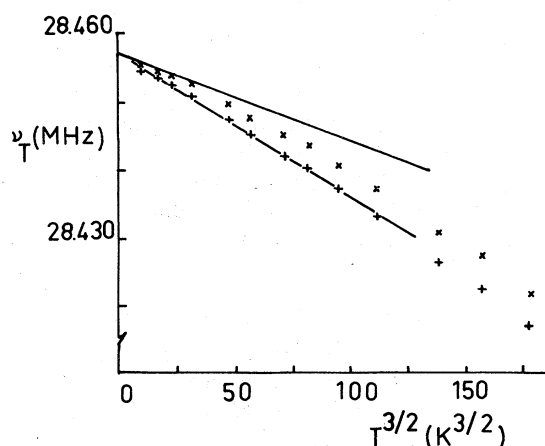


FIG. 1. Hyperfine field of <sup>61</sup>Ni in natural nickel as a function of  $T^{3/2}$ . The original measurements are shown (x) and after correction for a gap equal to the anisotropy field (+). The lower line is given by Eq. (13) and the upper line shows the temperature dependence predicted by neutron scattering measurements, Eq. (7).

measurement at low temperature; for nickel however no equation of the form

$$(\nu_0 - \nu_T)/\nu_0 = B_0 T^{3/2} + C T^{n/2}, \quad (14)$$

where  $n$  was 4, 5, 6, or 7 would fit both the measurements and give a value of  $B_0$  consistent with Eq. (13). The smallest change in  $B_0$  was found for  $T^{5/2}$ , but the best fit to the high-temperature measurements occurred for  $T^{7/2}$  with an increased value for  $B_0$ . The increase in the value of  $B_0$  when the range of temperature is extended may also be seen in Table I of Aldred's paper.<sup>5</sup> A two-term spin-wave analysis is therefore inadequate to describe the present measurements and in Sec. VI the effect of single-particle excitations will be discussed.

#### V. HYPERFINE COUPLING CONSTANT

The hyperfine-field measurements can be substituted for the magnetization at high temperature only if the hyperfine coupling constant of nickel is independent of temperature and equal to unity. Streever and Bennett<sup>8</sup> first showed that after the reduction of both hyperfine-field and magnetization measurements to constant volume, the value of  $1 - A_T$  was less than  $5 \times 10^{-3}$  up to 450 K ( $0.7T_C$ ). The magnetization measurements of Kaul and Thompson<sup>20</sup> gave a larger value for  $1 - A_T$ , but using the latest magnetization measurements, due to Aldred,<sup>5</sup> and the present hyperfine-field measurements, the discrepancy is found to be  $2 \times 10^{-3}$  at 300 K. The equation given by Aldred, line 1 of Table I, as the best fit to his measurements gives a value of less than  $1 \times 10^{-3}$  for  $1 - A_T$  at 300 K, but increases to almost  $3 \times 10^{-3}$  at 250 K before reducing to less than  $1 \times 10^{-3}$  below 100 K. In summary the change in  $A_T$  up to 300 K ( $\approx 0.5T_C$ ) is certainly small and quite probably zero within experimental error. A more extensive set of measurements on the pressure dependence of the hyperfine field and the magnetization would however be very desirable since, as may be seen from Table I, the correction to constant volume at room temperature is quite large.

#### VI. SINGLE-PARTICLE EXCITATIONS

In this section the difference between the reduced hyperfine field of nickel at temperature  $T$  and the spin-wave term given by Eq. (7) will be considered. At low temperature ( $T < 69$  K) it was found that a small term linear in  $T$  occurred that was attributed to a gap in the spin-wave spectrum of the nickel atoms within the domain walls that contribute to the NMR signal (Sec. IV). At higher temperatures, as may be seen from Table I, this term is not important because the anisotropy field

of nickel is nearly zero at room temperature. The hyperfine-field measurements were then corrected to constant volume and  $B_T^n$  was also corrected to constant volume<sup>18</sup> assuming that the spin wave and total magnetization had the same dependence on pressure.

It is convenient to define the parameter  $Y$  by the expression

$$Y = (\nu_0 - \nu_T)/\nu_0 T^{3/2} - B_T^n, \quad (15)$$

where both the hyperfine field and  $B_T^n$  have been reduced to constant volume. A preliminary attempt to fit all the measurements up to room temperature with a simple power law showed that  $Y$  was roughly proportional to  $T^{0.36}$ .

The agreement between the measurements and the form of the two simple expressions for single-particle excitations, Eqs. (10) and (11), is shown in Fig. 2. The fit to a  $T^2$  law at low temperature is obvious, but although the value for  $\Delta/k_B$  of 162 K found by Aldred is in good agreement with the present measurements in the range 210–292 K it is evident that the greater accuracy of the NMR measurements has established that this value is not in agreement with the low-temperature results. The apparent value of  $\Delta/k_B$  decreases from 162 K near room temperature to 18 K at temperatures below 50 K.

The choice between weak and strong ferromagnetism for nickel is not however as clear-cut as might appear from Fig. 2. A strong ferromagnet with a small energy gap ( $\approx 18$  K) would not be ex-

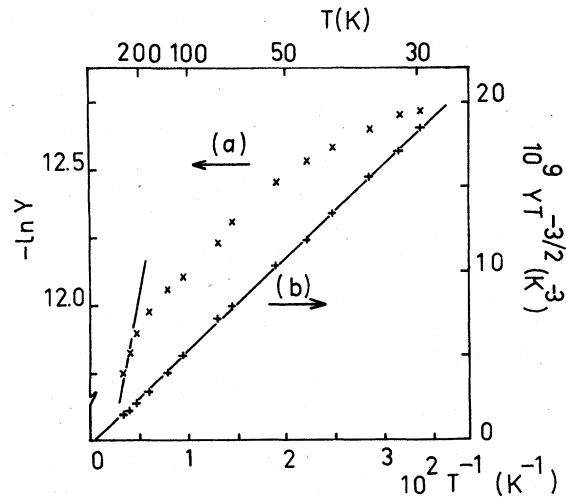


FIG. 2. (a)  $\ln Y$  as a function of  $T^{-1}$ . The slope estimated from magnetization measurements ( $\Delta/k_B = 162$  K) is shown. (b)  $Y T^{-3/2}$  as a function of  $T^{-1}$ . The straight line shows that the temperature dependence of single-particle excitations in nickel contains a large term in  $T^2$ . [ $Y$  is defined in Eq. (15)].

pected to obey Eq. (11) as was remarked in Sec. II, but would follow a polynomial containing a term in  $T^{3/2}$ . In Fig. 3 the dependence of  $Y$  on  $T^{1/2}$  is shown. The equation

$$(\nu_0 - \nu_T)/\nu_0 = (B_T^n + 1.68 \times 10^{-6})T^{3/2} + 3.22 \times 10^{-7}T^2 \quad (16)$$

provides a fit to the measurements with a rms error of less than 3 kHz (0.01% accuracy) and may therefore indicate that nickel is indeed a strong ferromagnet, but with a much smaller value of  $\Delta/k_B$  than had been calculated from the magnetization measurements. This conclusion would be in agreement with de Haas-van Alphen measurements that show only one type of spin at the Fermi surface,<sup>9</sup> and with the theoretical prediction<sup>15</sup> that the effect of many-body interactions is to reduce  $\Delta$ .

The value of the coefficient of  $T^{3/2}$  in Eq. (16) is an order of magnitude greater than the estimated random error in  $B_T^n$ . Using the early neutron measurements of Stringfellow<sup>3</sup> to calculate  $B_T^n$  it is found that there is little change in the coefficient of  $T^2$  in Eq. (16), but the term in  $T^{3/2}$  reduces to  $B_T^n$ . The  $T^2$  contribution to the single-particle term in the magnetization is therefore well established, but the term in  $T^{3/2}$  depends more sensitively upon the accuracy of the neutron-scattering measurements. A reasonable fit to the measurements can also be made using the theoretical expression for a weak ferromagnet given in Eq. (12):

$$(\nu_0 - \nu_T)/\nu_0 = B_T^n T^{3/2} + 5.64 \times 10^{-7} T^2 - 8.6 \times 10^{-9} T^{5/2}, \quad (17)$$

but the fit is not as good as Eq. (16) and in addition the magnitude of the  $T^{5/2}$  term is probably too large to be physically reasonable since it arises from a second-order effect.

## VII. CONCLUSION

The discrepancy between the neutron-scattering and magnetization values for the spin-wave coefficient of nickel has been confirmed by hyperfine-field measurements. Assuming that the true spin-wave term is given by the neutron value  $B_T^n$  it has been shown that the energy difference ( $\Delta$ ) between the top of the  $d_{\uparrow}$  band and the Fermi level must be at least an order of magnitude smaller than the value of  $\Delta/k_B = 162$  K estimated from magnetization measurements. This result is consistent with the theoretical prediction that in a real ferromagnet the many-body interaction effect is to reduce  $\Delta$  from the value expected on the simple Stoner theory to a value close to zero.

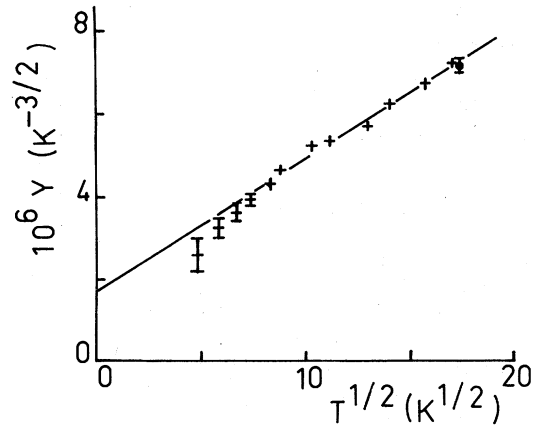


FIG. 3.  $Y$  as a function of  $T^{1/2}$ . The straight line is given by Eq. (16). The error bars show the effect of an error of  $\pm 1.5$  kHz in the NMR measurements, i.e., no account is taken of a possible error in the neutron measurements used to derive Eq. (7). The value of  $Y$  at 300 K calculated from the magnetization is shown (●).

There seems little doubt that nickel is a strong ferromagnet, particularly in view of the evidence from de Haas-van Alphen measurements, but it must be noted that, even though the NMR measurements are an order of magnitude more accurate than the magnetization measurements, it has still not proved possible to distinguish with certainty between the single-particle equations appropriate to strong and weak ferromagnetization.

The contrast between the magnetic properties of iron and nickel is extremely striking. The spin-wave theory is sufficient to explain the properties of iron, but single-particle excitations appear to be as important as spin waves in the case of nickel. A more direct separation of the two contributions than is possible using magnetization measurements may be achievable using the de Haas-van Alphen effect. Edwards<sup>21</sup> has shown that the temperature dependence of de Haas-van Alphen frequencies can give information about the  $T^2$  term in ferromagnetic metals without interference from spin waves. Lonzarich and Gold<sup>22</sup> found that one such frequency in iron was independent of temperature between 1 and 4 K, as would be expected since single-particle terms are unimportant in iron, but in view of the present measurements it would be most interesting to repeat this experiment for nickel.

*Note added in proof.* D. M. Edwards (private communication) has recently proposed that the ferromagnetism of face-centered-cubic transition metals has a quasi-two-dimensional character. The discrepancy between  $B_T$  and  $B_T^n$  is then attributed to low-lying flat modes of the spin-wave

spectrum that were not observed in the neutron-scattering measurements from which  $B_T^n$  was deduced. The importance of the contribution of independent particle excitations to the magnetization of nickel is therefore still open.

## ACKNOWLEDGMENTS

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