Temperature dependence of the magnetization of nickel*

A. T. Aldred
Argonne National Laboratory, Argonne, Illinois 60439
(Received 12 September 1974)

The magnetization of a single-crystal nickel sample has been measured between 4 and 300 K in applied fields up to 13.5 kOe. The results are shown to fit spin-wave theory, but the numerical values of the spin-wave stiffness constants obtained are not in agreement with neutron-determined values. The discrepancy appears to be associated with a contribution to the temperature dependence of the magnetization from Stoner single-particle excitations; this contribution is found to be as large as the spin-wave term.

I. INTRODUCTION

Detailed measurements of the temperature dependence of the magnetization of iron[1,2] and nickel[1,3,4] at low temperature support, in general, the ideas of spin-wave theory,[5] i.e., the magnetization varies initially as $T^{3/2}$. The coefficient of the $T^{3/2}$ term is related to the spin-wave stiffness coefficient D, which can be measured independently by inelastic-neutron-scattering measurements.[6] Results of such experiments have only recently become available. In a previous paper,[2] we showed that quantitative agreement was evident between the values of D obtained from magnetization and neutron-scattering[7] data for iron. In particular, the temperature dependence of D observed in the neutron-scattering experiments was matched by a modification of the basic $T^{3/2}$ dependence of the magnetization. A similar correlation of D values obtained by the two independent techniques was found for Fe-V alloys.[8]

To study this correlation further, magnetization measurements have been made from 4 to 300 K in applied fields up to 13.5 kOe on a high-purity (99.99% pure) single crystal of nickel with the ⟨111⟩ easy axis of magnetization oriented in the direction of the applied field. It is found that the rate of change of magnetization with temperature, which is in good agreement with previous results,[1,3,4] is greater by a factor of ~2 than that calculated from the neutron-determined stiffness coefficient. This discrepancy suggests than an additional contribution is being made to the temperature dependence of the magnetization which arises from Stoner single-particle excitations. The functional form of such a term has been considered by Thompson *et al.*,[9] and the present data are considered in terms of their model.

II. EXPERIMENTAL

The single-crystal specimen, which weighed ~100 mg, was cut in the form of a prolate ellipsoid with an axial ratio of ~2 and a ⟨111⟩ direction oriented along the major axis. Because of the strain-free machining techniques employed, the sample was not annealed.

The current operating procedures for the magnetization equipment have been reported.[10] Magnetizations are determined, sequentially, at one of fourteen applied fields between ~0.5 and ~13.5 kOe as the temperature is slowly increased (~0.5 K/min) from 4 to 300 K. The data points obtained (~1200) are subjected to a cubic spline computer program, which allows evaluation of the data (at each field) at any interpolated temperature. With this procedure, a series of isotherms can be conveniently constructed, although data analysis can also be performed on the original experimental points.

The experiment is a relative one, and the external field gradients were calibrated with the same nickel sample in a separate experiment. The sample was held at 5 K, and, by repetitive measurements, a series of data points were obtained at each field. The results were averaged, and the gradients were calculated from the known magnetization of nickel. Inasmuch as the sample was a single crystal with its easy axis oriented parallel to the applied field, a constant magnetization (= 58.57 emu/g)[11] independent of field was used. This procedure could introduce small errors if the high-field susceptibility term,[12] neglected in the calibration because of the limited field range of the data, were temperature dependent. The general comments in Ref. 2 about experimental errors apply to the present results, although by improvements in experimental methods the uncertainties have now been reduced and vary from ±0.003 emu/g (0.05%) at low fields to ±0.01 emu/g (0.02%) at the highest field. These uncertainties were taken into account, in terms of a weighting function, in the data analysis.

III. RESULTS

Some typical magnetic isotherms are shown as a function of internal field H_i in Fig. 1. Because

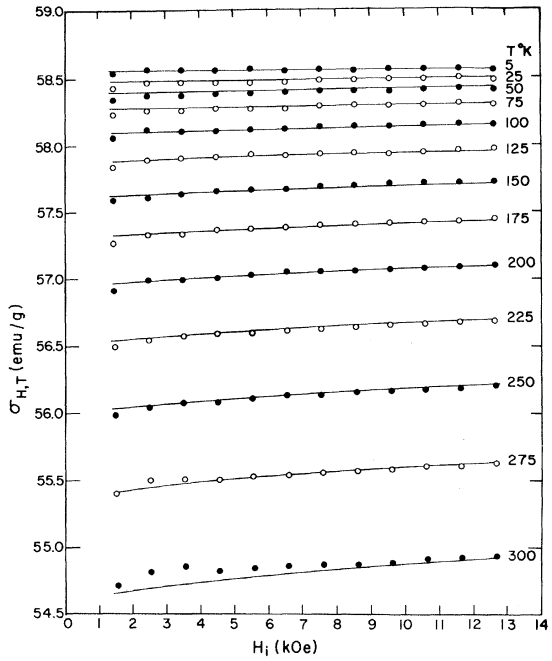

FIG. 1. Magnetic isotherms for a single-crystal nickel sample at the temperatures indicated. Solid lines represent a fit to the spin-wave equation detailed in the text.

of possible uncertainties in the demagnetizing field corrections (~1 kOe with an uncertainty of ± 3%), data below $H_i = 1$ kOe are not shown and were not used in the analyses to be presented. The value of the magnetization (relative to the calibration value) and the lack of field dependence at 5 K (Fig. 1) gives a good measure of the precision of the data. The fact that the values at the lowest field seem to fall systematically below an extrapolation of the other data points at each temperature indicates either an uncertainty in the gradient calibration or a small H^{-1} contribution to the magnetization which arises from possible defects, impurities, etc. in the sample. The increase in field dependence of the magnetization as the temperature increases is apparent.

An analysis of the results in terms of the spin-wave equation was performed on the original data points. According to spin-wave theory, the expression for the specific magnetization $\sigma_{H,T}$ (emu/g) as a function of temperature T(°K) and field H(Oe) is[2,5]

$$\sigma_{H,T} = \sigma_{0,0} - \frac{g\mu_B}{\rho}\left(\frac{k_B T}{4\pi(D_0 - D_1 T^2)}\right)^{3/2} F(\tfrac{3}{2}, t) + \cdots, \quad (1)$$

where g is the Landé splitting factor, μ_B is the Bohr magneton, ρ is the density, k_B is Boltzmann's constant, and $F(\frac{3}{2}, t)$ is a Bose-Einstein integral function ($t = k_B T/g\mu_B H$).[2,5] The parameters D_0 and D_1 represent the temperature dependence of the spin-wave stiffness constant

$$D = D_0 - D_1 T^2, \quad (2)$$

where D is the coefficient of the quadratic term of the spin-wave dispersion relation.[5] The theoretical and experimental justifications for Eq. (2), which is phenomenologically equivalent to a $T^{7/2}$ term in the expansion of Eq. (1) to higher order in T, are noted in Ref. 2. Values of $\rho = 8.9$ g/cm^3 and $g = 2.22$ (independent of temperature[13]) were used in the analyses to be presented. The field H is the sum of the internal and anisotropy fields; the anisotropy field at each temperature was determined from the anisotropy constant as a function of temperature.[13]

The data were fitted, as in the case of iron,[2] to two versions of Eq. (1) one in which Eq. (1) is as written and one in which the next term in the expansion ($T^{5/2}$) is included and a temperature-independent D is substituted for $D_0 - D_1 T^2$. However, three disposable parameters ($\sigma_{0,0}$ and either D_0 and D_1 or D and the coefficient of the $T^{5/2}$ term) are fitted in each version. With the present results for nickel, the second fit gives an rms error ~40% larger than for the first fit, and thus, as in the case of iron,[2] the first fit is deemed more appropriate. Foner and Thompson[3] also note that the deviations from the $T^{3/2}$ law for nickel are better fitted to a $T^{7/2}$ than a $T^{5/2}$ dependence.

To show the temperature dependence of the data more clearly, the original experimental values at $H_i = 10.6$ kOe are plotted as a function of $T^{3/2} \times F(\frac{3}{2}, t)$ in Fig. 2 together with a line representing the least-squares fit to all data (as a function of temperature and field) up to 300 K. The first fit [Eq. (1) as written] provides a good representation of the experimental results. The parameters obtained are given in the first line of Table I, and the values were used to calculate the isotherms given by the solid lines in Fig. 1; the quality of the fit to the interpolated data lends confidence to the interpolation procedures. A further feature of the data in Fig. 2 is the small but systematic discrepancy between the experimental and fitted points at low temperatures [25–75 K; $T^{3/2}F(\frac{3}{2}, t) < 1000$ K$^{3/2}$]. A similar effect was observed in the earlier measurements on iron, and it is, most likely, an artifact of the experimental setup (see Ref. 2). This apparent uncertainty in the low-temperature values has a substantial effect (in terms of the magnitude of the rms error) when only the data up to 150 K are fitted to the same equation. The results of such an analysis are given in the second line of Table I. The two sets of spin-wave parameters are reasonably consistent. However, they do not agree with values obtained by neutron-

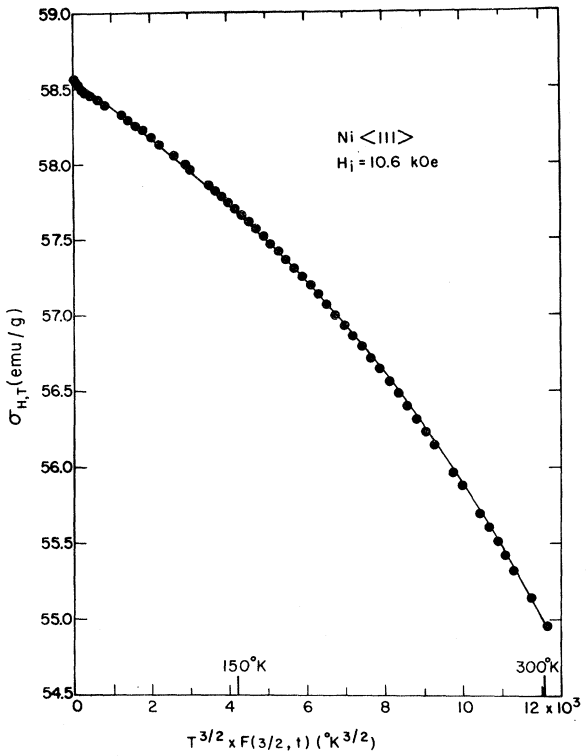

FIG. 2. Temperature dependence of the magnetization of a single-crystal nickel sample at $H_i = 10.6$ kOe. Solid line represents the same fit as in Fig. 1.

scattering techniques[7,14–17]; the source of this discrepancy is considered in detail in the next section.

IV. DISCUSSION

The present results for the temperature dependence of the magnetization of nickel up to room temperature appear to be in good agreement with previous determinations.[1,3,4] (Exact comparisons are difficult because of differences in data presentation.) As noted by the previous authors, the deviations from $T^{3/2}$ behavior are pronounced (see also Fig. 2). The comment by Foner and Thompson[3] that these deviations may be fit by a $T^{7/2}$ term are supported by the present work, and it is proposed that this term arises from the T^2 dependence of the spin-wave stiffness coefficient. Although this fit provides a good representation of the data, the numerical values obtained for the stiffness coefficient are not in agreement with those determined directly by neutron-scattering experiments.

The spin-wave dispersion relationship for nickel has been studied with inelastic neutron-scattering techniques by a number of authors.[7,14–17] The results appear to fit a quadratic dispersion law, and no significant evidence of higher-order terms has been found, except, possibly, in the data of Minkiewicz *et al.*[16] The stiffness constant D has been determined mainly at room temperature, and the values are given in Table II. Inasmuch as the room-temperature data of Ref. 17 are given only graphically, D was determined by least-squares analysis of values read from the graph. To minimize uncertainties due to possible higher-order terms in q, only data below $q = 0.38$ Å^{-1} were used. This same procedure, applied to the data at 4.2 K, gave good agreement with the value quoted by Mook *et al.*[17] (Table II). When the room temperature data of Minkiewicz *et al.*[16] are analyzed in the same way ($q \leq 0.38$ Å^{-1}) the value of D(295 K) is in excellent agreement with that obtained from

TABLE II. Results of neutron-scattering determinations of the spin-wave stiffness constant D of nickel.

Technique[a]	Applied field (kOe)	D(295 K) (meV Å^2)	D(4.2 K) (meV Å^2)	Reference
DM	3	374 ± 20[b]		14
SAS	1.8	410[c]		7
TA	•••	433[b]		15
TA	•••	400[b] (454 ± 9)[d]		16
TA	•••	458 ± 9[d]	555[b] (544 ± 6)[d]	17

[a]DM: diffraction method; SAS: small-angle scattering; TA: triple-axis spectrometry.
[b]Value given by authors.
[c]Value estimated from graph.
[d]Value obtained by least-squares analysis of data ($q < 0.38$ Å^{-1}) read from graph.

TABLE I. Results of various least-squares fits to the temperature and field dependence of the magnetization of nickel. Details of the fits are given in the text; the numbers in parentheses represent statistical uncertainties in the final significant figure(s).

Max. Temp. (°K)	$\sigma_{0,0}$ (emu/g)	D_0 (meV Å^2)	D_1 (10^{-3} meV Å^2 °K^{-2})	B (10^{-5} °K$^{-3/2}$)	Δ/k_B (°K)	A (10^{-7} °K^{-2})	rms error (emu/g)
300	58.564(3)	362(1)	1.05(1)				0.0215
150	58.537(3)	395(5)	2.02(21)				0.0136
300	58.540(3)	384(4)	0.86(5)	0.95(9)	453(44)		0.0124
300	58.545(3)	472(16)	1.52(8)			1.95(20)	0.0173

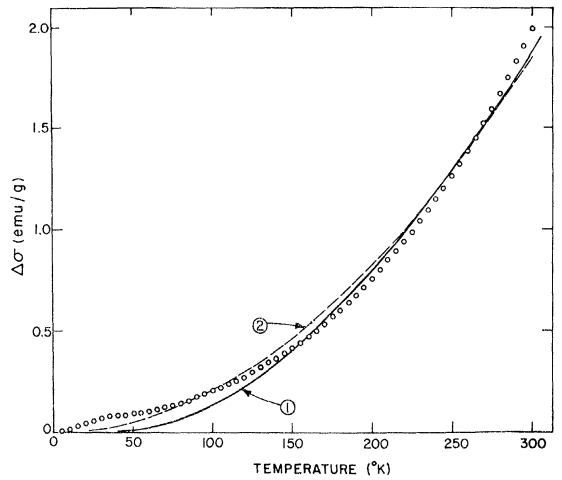

FIG. 3. Non-spin-wave magnetization of nickel $\Delta\sigma$ (see text) plotted as a function of temperature. Solid line labeled 1 represents a fit to Eq. (3), and the dashed line labeled 2 represents a fit to Eq. (4).

the results of Mook *et al.*,[17] and therefore a value of $D(295\ \mathrm{K}) = 455$ meV Å^2 has been used in the subsequent analysis. When the values of $D(4.2\ \mathrm{K})$ and $D(295\ \mathrm{K})$ are inserted in Eq. (2), the following results are obtained: $D_0 = 555$ meV Å^2; $D_1 = 1.15 \times 10^{-3}$ meV Å^2 °K^{-2}. These neutron-determined parameters can then be substituted in Eq. (1) to calculate the spin-wave contribution to the temperature dependence of the magnetization. On this basis, a value of $\sigma_{0,300} = 56.56$ emu/g is obtained, in contrast to the experimental value of 54.57 emu/g (Fig. 1); i.e., the total change in magnetization between 0 and 300 K is a factor of 2 larger than the spin-wave contribution.

To extract the non-spin-wave term, the spin-wave contribution to the temperature dependence of the magnetization calculated from the neutron-determined parameters given above was subtracted point by point from the data of Fig. 1, and the remainder $\Delta\sigma$ was averaged over the six highest fields (to minimize random errors). The values of $\Delta\sigma$ are plotted as a function of temperature in Fig. 3; the systematic errors associated with the experimental data in the range from 25 to 75 K, noted earlier, are clearly reflected in the values of $\Delta\sigma$ in the same temperature range. In addition, the procedure given above yields the difference between two large numbers and this increases the chance of systematic errors in the values of $\Delta\sigma$. Nevertheless, the general magnitude of $\Delta\sigma$ and its temperature dependence are established.

The data of Fig. 3 have been analyzed in terms of the equations given by Thompson *et al.*[9] for the contributions of single-particle Stoner excitations to the temperature dependence of the magnetization. In Thompson *et al.*'s terminology, one would expect nickel to be a strong ferromagnet; i.e., holes are present only in the spin-down band. In this case,

$$\Delta\sigma = \sigma_{0,0}[2I(T)/n]\, e^{-\Delta/k_B T}, \tag{3}$$

where n is the number of holes in the spin-down band and Δ is a measure of the band splitting. According to Thompson *et al.*, $I(T)$ is a function related to the density of states which, for nickel, varies as $T^{3/2}$. Therefore $2I(T)/n$ in Eq. (3) was replaced by $BT^{3/2}$, and the data of Fig. (3) were least-squares fit to Eq. (3). The results are presented as the solid line in Fig. (3); the parameters obtained were $B = (1.06 \pm 0.08) \times 10^{-5}$ K$^{-3/2}$ and $\Delta/k_B = 162 \pm 10$ K, with an rms error of 0.056 emu/g. These values are in reasonable agreement with values obtained by Thompson *et al.* from an analysis of the literature data. When the temperature range of the data to be fitted was gradually decreased, the values of B and Δk_B decreased rapidly; this effect presumably reflects the spurious bump in the data of Fig. 3 near 50 K.

An alternative approach in fitting the data of Fig. 3 is to consider nickel as a weak ferromagnet (holes in both spin bands). In this case, according to Thompson *et al.*,[9] $\Delta\sigma$ should vary primarily as

$$\Delta\sigma = \sigma_{0,0} A T^2, \tag{4}$$

where A is related to various band parameters. The least-squares fit to Eq. (4) is shown as the dashed line in Fig. 3; the rms error is 0.051 emu/g (smaller than for the two-parameter exponential fit), and the value of A is $(3.50 \pm 0.03) \times 10^{-7}$ K^{-2}. When the temperature range of data fitted was decreased to 5–150 K, a value of $A = (3.19 \pm 0.04) \times 10^{-7}$ K^{-2} was obtained with an rms error of 0.025 emu/g.

An analysis of the fits to Eqs. (3) and (4) in terms of the rms errors, and the systematics of the parameters obtained over different temperature ranges would marginally favor the T^2 fit. However, an examination of Fig. 3 suggests that neither Eq. (3) nor Eq. (4) provides a good representation of the data. This may well reflect systematic errors accumulated in the data reduction. A further point is that Eqs. (3) and (4), with a suitable choice of parameters, can produce quite similar magnetization-versus-temperature curves at higher temperatures. It will therefore only be possible to determine which equation is applicable by precise measurements at low temperatures, where the changes in magnetization with temperature are small.

In an effort to separate the two contributions (from spin-wave and Stoner excitations) to the total temperature dependence of the magnetization of nickel, the terms given by Eqs. (3) and (4) were

incorporated, in turn, into Eq. (1), and the original experimental data were reanalyzed. The parameters obtained in the fit that included the exponential term are given in line 3 of Table I. Although the rms error is considerably improved, the spin-wave parameters (D_0 and D_1) are not significantly different from those obtained in the spin-wave-only analysis presented in Sec. III (line 1 of Table I). Thus this analysis is inconsistent with the neutron-scattering results. When a T^2 term (in the magnetization) was added to Eq. (1), the parameters given in line 4 of Table I were obtained. The spin-wave parameters are in better agreement with the neutron-determined values than any of the earlier fits, and the T^2 coefficient is not inconsistent with that obtained above.

V. CONCLUSIONS

The temperature dependence of the magnetization of nickel up to 300 K can be well represented in terms of the spin-wave equation. However, the parameters obtained are not in good agreement with the values determined directly by inelastic neutron-scattering measurements. If it is assumed that a contribution to the temperature dependence of the magnetization arises from Stoner single-particle excitations, the data can be analyzed to yield this term, which appears to be of the same magnitude as the spin-wave contribution. The temperature dependence of the Stoner excitation term may be analyzed satisfactorily in terms of two alternative forms proposed by Thompson *et al.*[9] The results do not show, conclusively, whether nickel has holes in either one or both spin subbands, and additional extremely precise measurements at low temperatures (< 50 K) are required.

ACKNOWLEDGMENTS

The author would like to thank P. H. Froehle for experimental and computational assistance. Helpful discussions with E. D. Thompson, S. Doniach, J. S. Kouvel, and J. Hertz are gratefully acknowledged.

---

*Work supported by the U.S. Atomic Energy Commission.

[1]B. E. Argyle, S. H. Charap, and E. W. Pugh, Phys. Rev. 132, 2051 (1963).
[2]A. T. Aldred and P. H. Froehle, Int. J. Magn. 2, 195 (1972).
[3]S. Foner and E. D. Thompson, J. Appl. Phys. 30, 229S (1959).
[4]R. Kaul and E. D. Thompson, J. Appl. Phys. 40, 1383 (1969).
[5]F. Keffer, in *Handbuch der Physik* (Springer, Berlin, 1966), Vol. 18, pp. 21–31.
[6]G. Shirane, V. J. Minkiewicz, and R. Nathans, J. Appl. Phys. 39, 383 (1968).
[7]M. W. Stringfellow, J. Phys. C 1, 950 (1968).
[8]A. T. Aldred, Int. J. Magn. 2, 223 (1972).
[9]E. D. Thompson, E. P. Wohlfarth, and A. C. Bryan, Proc. Phys. Soc. Lond. 83, 59 (1964).
[10]A. T. Aldred, B. D. Dunlap, D. J. Lam, and I. Nowik, Phys. Rev. B 10, 1011 (1974).
[11]H. Danan, A. Herr, and A. J. P. Meyer, J. Appl. Phys. 39, 669 (1968).
[12]S. Foner, A. J. Freeman, N. A. Blum, R. B. Frankel, E. J. McNiff, Jr., and H. C. Praddaude, Phys. Rev. 181, 863 (1969).
[13]D. S. Rodbell, *Proceedings of the International Conference on Magnetism*, Nottingham, 1964 (Institute of Physics and The Physical Society, London, 1965), p. 420.
[14]S. J. Pickart, H. A. Alperin, V. J. Minkiewicz, R. Nathans, G. Shirane, and O. Steinsvoll, Phys. Rev. 156, 623 (1967).
[15]H. A. Mook, R. M. Nicklow, E. D. Thompson, and M. K. Wilkinson, J. Appl. Phys. 40, 1450 (1969).
[16]V. J. Minkiewicz, M. F. Collins, R. Nathans, and G. Shirane, Phys. Rev. 182, 624 (1969).
[17]H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. 30, 556 (1973).