PHYSICAL REVIEW B

SOLID STATE

THIRD SERIES, VOL. 2, NO. 8 15 OCTOBER 1970

Effects of Configurational Order on the Specific-Heat Capacity of Ni_3Fe between 1.2 and 4.4 K*

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(Received 16 March 1970)

Measurements of the specific-heat capacity of configurationally ordered and disordered Ni_3Fe between 1.2 and 4.4 K are presented. Coefficients of the electronic, magnetic, and harmonic phonon contributions decreased significantly on ordering (21.5, 8.5, and 4.8%, respectively). These results are justified theoretically and compared where possible with other measurements in the literature.

I. INTRODUCTION

Within the past decade, a large number of measurements of the specific-heat capacity C_p of solids at liquid-helium temperatures have been reported in the literature. An excellent review of these data for transition metals and their alloys has been prepared by Heiniger, Bucher, and Müller.[1] Of the investigations on transition metal alloys, few have been concerned with the very significant effects of long-range configurational order on this property. Metallurgically speaking, this overwhelming concern with the disordered state is quite unusual because the ordered state is always the stable state near the absolute zero of temperature. The purpose of this work, therefore, is to report the effects of ordering on the C_p of the alloy Ni_3Fe between 1.2 and 4.4 K, and to discuss the theoretical implications of the measured differences in C_p.

The alloy Ni_3Fe (75 at. % Ni + 25 at. % Fe) undergoes both a Curie and a configurational-ordering transformation. Under equilibrium conditions the Curie transformation, which is a change from the ferro- to paramagnetic state, occurs at 871 K and the configurational ordering transformation, which is of the type of Cu_3Au, takes place at 773 K. Due to the sluggish kinetics of the latter transformation, it is possible to quench in the disordered state by rapid cooling from above 773 K. Obviously, the electron and phonon configurations of the metastable-ferromagnetic-disordered state differ from that of the stable-ferromagnetic-ordered state and thereby cause different C_p's near 0 K.

At low temperatures, the dilation contribution to the specific-heat capacity at constant pressure C_p is negligible. Consequently, C_p is equal to C_v, the specific-heat capacity at constant volume. It is common practice[1-4] to assume that C_v of a ferromagnetic metal near 0 K is separable into three independent contributions – a harmonic phonon contribution C_{vh}, an electronic contribution C_{ve}, and a magnetic contribution C_{vm}. With few exceptions, the Debye model is used for C_{vh}, which at these temperatures is adequately represented by βT^3, and the expression γT of the free-electron model is employed for C_{ve}.

Choice of a model for C_{vm} is not as straightforward. However, lacking the desired band-theory description, one is forced to employ localized-electron models based on a Heisenberg-type Hamiltonian. Of course, the validity of applying these models to metals such as Ni_3Fe is questionable because of the itinerant nature of their electrons. Paradoxically, some success has been achieved with these models on magnetic transition metals and, therefore, the spin-wave $\alpha T^{3/2}$ expression for C_{vm} is used herein. The resulting expression for C_p is,

$$C_p = C_v = C_{vh} + C_{ve} + C_{vm} = \beta T^3 + \gamma T + \alpha T^{3/2} . \quad (1)$$

Changes in C_p due to the configurational order-dis-

order transformation are reflected in the coefficients β, γ, and α and these changes are discussed below.

II. EXPERIMENTAL DETAILS

A. Sample Preparation and Characterization

The Ni_3Fe specimen utilized in this research was prepared from stock materials having the chemical compositions shown in Table I. The total of the contaminants present was less than 450 ppm in the nickel stock and less than 1050 ppm in the iron stock. Appropriate proportions of the stock materials were arc-melted several times to ensure homogenization and drop-cast into a 2.5-cm-diam water-cooled copper mold. This casting was machined to remove surface imperfections and then swaged to about 1.9 cm to eliminate casting voids. A 3-cm-long section of this rod was cut off for the specific-heat capacity specimen, and the instrumentation holes requisite to the technique were machined in it. This 64-g specimen was annealed for 24 h at 1200 K in a vacuum of 10^{-8} Torr and then quenched into ice water, resulting in the microstructure shown in Fig. 1. Chemical analyses of the specimen taken after the quench are listed in Table I. From these, the alloy is estimated to contain 0.1 at. % (1000 ppm) impurities, 74.7 ± 0.1 at. % Ni and 25.2 ± 0.1 at. % Fe.

After the C_p measurements were completed on the disordered state, the specimen was heated to 1100 K in a vacuum of 10^{-7} Torr, cooled to 758 K, and annealed according to the schedule in Table II. This heat treatment produced a long-range configurationally ordered structure with antiphase domains less than 0.1 μ wide,[5] but did not affect the microstructure shown in Fig. 1.

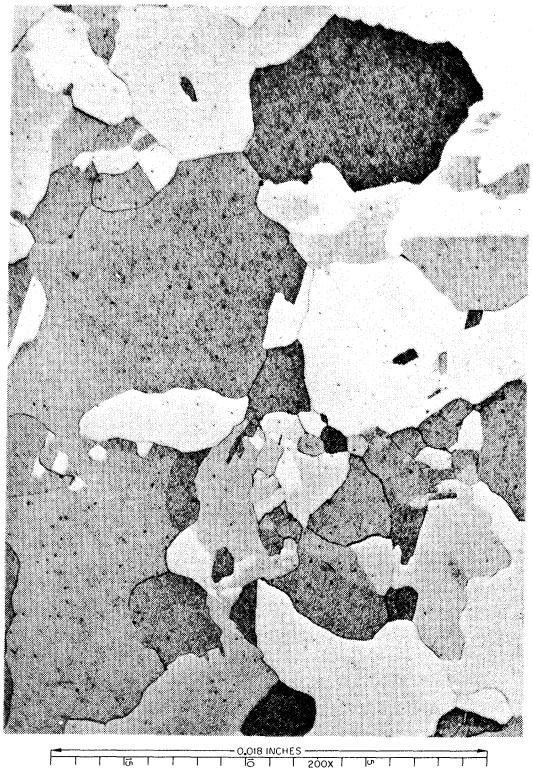

FIG. 1. Tranverse section of quenched Ni_3Fe specimen etched with ammonium persulfate (×200 original magnification). Difference in brightness of grains is due to orientation differences, including twinning.

TABLE I. Chemical composition of specimen stock and specimen.

Element	Contents in ppm by weight: Nickel stock	Iron stock	Ni_3Fe specimen
Ni			75.4×10^4
Fe	40		24.2×10^4
Al	6	< 200	< 500
C	70	50	20
Co	30	10	< 200
Cr	1.5	100	< 200
Cu	5	500	200
H_2	3		2
Mn	1	10	< 200
N_2	5	9	23
O_2	130	120	88
P	5	5	
S	< 0.4	40	
Si	<150	2	< 100
Ti	0.3	0.3	< 100
V	0.3		< 200
Total	<450	<1046	<1000[a]

[a]Estimate based on above limits of detection.

Due to the similarity in the scattering factors of nickel and iron, standard x-ray techniques of determining the Bragg-Williams parameter S of the ordered alloy were not attempted. Instead, S was estimated by electrical resistivity ρ measurements using the relationship[6]

$$\Delta\rho = A(1 - S^2) \ , \qquad (2)$$

TABLE II. Annealing schedule to produce long-range order in Ni_3Fe specimen.

Soak temperature (° K)	Days
758	4
733	10
713	7
693	8

where $\Delta\rho$ was the change in electrical resistivity due to ordering and A was approximately a constant. Measurements of ρ of the quenched and ordered C_p specimen were performed at 4.2 K using a technique described elsewhere,[7] and these data are listed in Table III. Also included therein is the result of another investigation[7] on a ρ specimen from the same stock which had been cooled from 1362 to 767 K in 34 days and from 767 to 602 K in 180 days. It was assumed that S for the latter specimen was 1.0. Since water quenching produces only a small amount of local order[8] in Ni_3Fe, S of the quenched C_p specimen was 0.0, yielding S for the ordered specimen of 0.69. For comparison, neutron-diffraction measurements[9] on a single crystal of Ni_3Fe annealed 25 days at 720 K gave an S of 0.75.

B. Measurement of Specific-Heat Capacity

The specific-heat capacity of the ordered and disordered specimen was measured using a previously described technique.[10] Briefly, a manganin heater was noninductively wound upon the specimen and a graphite resistance thermometer was inserted into a hole drilled in the specimen. Varnish was employed to secure the heater and thermometer in place and to ensure good thermal contact with the specimen. Nylon threads were used to suspend the specimen in an adiabatic vacuum chamber. After submerging the vacuum chamber in a liquid-helium bath, the pressure over the bath was changed to produce the desired specimen temperature. When thermal equilibrium was obtained between bath, chamber, and specimen, a direct current I was passed through the manganin heater and the time Δt required to change the specimen temperature by an amount ΔT was measured. Normally, ΔT was about 0.03 K. The C_p of a specimen of mass M at the mean temperature of the pulse was

$$C_p = (1/M)\ (EI\Delta t/\Delta T - C_a)\ , \tag{3}$$

where E was the potential across the manganin heater. The quantity C_a was the heat capacity of the thermometer, heater, and varnish at the mean temperature of the pulse and was determined by auxiliary measurements with a copper standard specimen.[10] The magnitude of C_a was less than 0.4% of C_p at 4 K and less than 0.3% of C_p at 1 K. For increased accuracy, the carbon-resistance thermometer was calibrated against the vapor pressure of the liquid-helium bath. Errors of $\pm 0.5\%$ in C_p and ± 0.002 K in temperature were ascribed to measurement uncertainties, and the repeatability of the technique was $\pm 0.25\%$.

TABLE III. Estimated S and measured ρ of various states of Ni_3Fe at 4.2 K.

State	ρ ($\mu\Omega$ cm)	S
Quenched from 1200 K[a]	4.25	0.00
Ordered (Table II)[a]	3.56	0.69
Ordered (see text)[b]	2.80	1.00

[a] C_p specimen. [b] ρ specimen.

III. RESULTS

If the magnetic contribution to C_p is negligible, Eq. (1) may be rewritten as

$$C_p/T = \beta T^2 + \gamma\ , \tag{4}$$

indicating that C_p/T is a linear function of T^2. Figure 2 depicts the data obtained on the ordered and disordered states of Ni_3Fe plotted as C_p/T versus T^2. (These data are tabulated in Ref. 7.) The straight lines in this figure were obtained by fitting the data to Eq. (4) by the method of least squares. Note that a negative deviation of the data from the straight lines exists below 2.2 K^2; that is, a smooth curve drawn through the data would be concave toward the T^2 axis. Dixon, Hoare, and Holden[4] predict this type behavior in the presence of a spin-wave contribution. They showed this by differentiating C_p/T obtained from Eq. (1) twice with respect to T^2, obtaining

$$\frac{d^2(C_p/T)}{d(T^2)^2} = -\frac{3\alpha}{16T^{7/2}} \approx \kappa\ . \tag{5}$$

Equation (5) is the first term in the Taylor-series expansion for the curvature κ of the function C_p/T of T^2. Since α is small and positive, κ is small and negative. In addition, κ is more than 2 orders of magnitude smaller at 4 K than at 1 K, indicating that C_p/T should be slightly concave toward the T^2 axis at 1 K^2 but almost linear in T^2 near 16 K^2. From this analysis, it was concluded that a small spin-wave contribution was probably present in the C_p of ordered and disordered Ni_3Fe. Consequently, the method of least squares was employed also to fit the data to Eq. (1).

Figures 3 and 4 depict the absolute differences between the data and the least-square fits to Eqs. (1) and (4) for disordered and ordered Ni_3Fe, respectively. Several observations may be made regarding these figures. First, the deviations are periodic rather than random and have the same temperature dependence for both the two-parameter [Eq. (4)] and three-parameter [Eq. (1)] fits of each alloy state. Second, below 3 K, where the curvature caused by the spin-wave contribution is important, the deviations of the three-parameter fits are much smaller. Third, few of the deviations of either fit are outside the $\pm 0.25\%$ repeatability of the experimental technique. Last, as noted in Table IV, the standard deviations of β and γ are higher for the

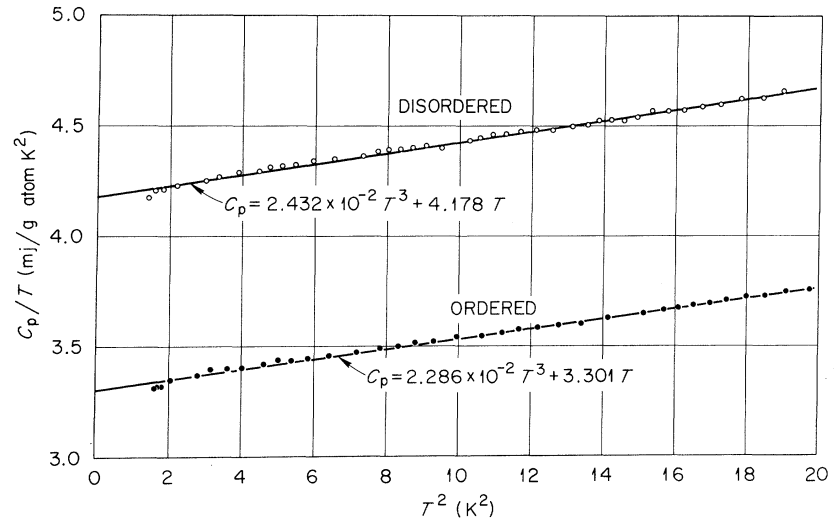

FIG. 2. Low temperature C_p of ordered and disordered Ni_3Fe.

three-parameter fits for both alloy states.

The question now arises, "Which fit is the more exact?" To assist in answering this question, one further investigation was conducted. This consisted of systematically omitting points from the low- or high-temperature end of the data and refitting the remaining points to the three-parameter equation.

As shown in Table IV, α decreased and β and γ increased as data points were excluded from the low-temperature end. After four ordered-state and five disordered-state points were dropped, α became negative, which is physically unreal, and β and γ were about the same as the two-parameter fits. Obviously, the value of α is highly dependent on data near 1 K, as predicted by Eq. (5).

When data points were excluded from the high-temperature end, the changes in the parameters were far less drastic, yielding an increase in α and a decrease in β and γ. These changes were affected by the dependence of β on these high-temperature data. This can be seen from Fig. 5 in which the contributions of $\alpha T^{3/2}$ and βT^3 to C_p are plotted as a percent of the total C_p versus temperature. In addition, the value of β is more important in determining the slope of C_p/T versus T^2 at high temperatures because

$$\frac{d(C_p/T)}{d(T^2)} = \frac{1}{4}\alpha \frac{1}{T^{3/2}} + \beta \,. \tag{6}$$

Near 4 K, the second term in Eq. (6) is 1 order

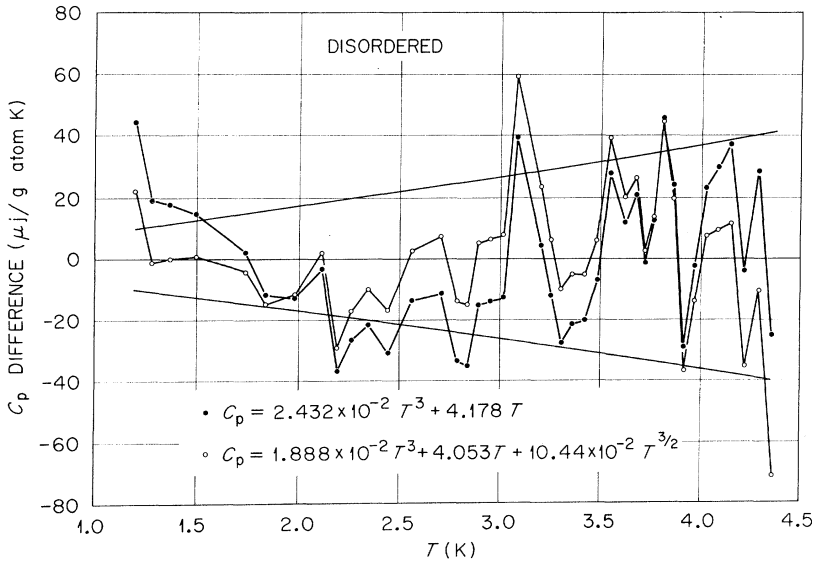

FIG. 3. C_p calculated from two-parameter [Eq. (4)] and three-parameter [Eq. (1)] least-squares fits minus the experimental values for disordered Ni_3Fe. The smooth solid curves represent ±0.2% deviations from the experimental values.

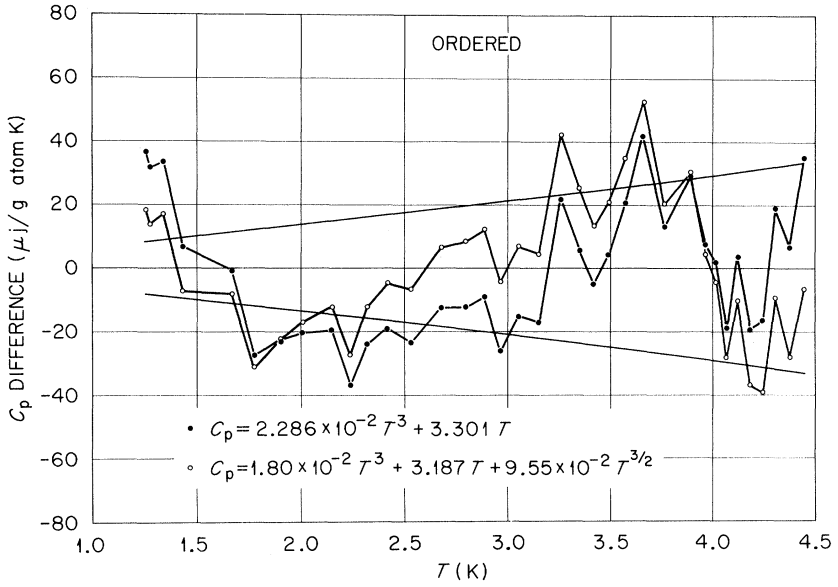

FIG. 4. C_p calculated from two-parameter [Eq. (4)] and three-parameter [Eq. (1)] least-squares fits minus the experimental values for ordered Ni_3Fe. The smooth solid curves represent ±0.2% deviations from the experimental values.

of magnitude greater than the first, whereas at 1 K they are about equal.

This analysis indicates that the values of α and β derived from a least-squares fit of C_p data of a ferromagnet such as Ni_3Fe are susceptible to the temperature interval of the fit because both contributions are small, totaling less than 15% of C_p below 4.4 K. In the absence of a nuclear contribution as found by Martin[11] in Cu_3Au, data below 1.2 K would probably establish the existence of a spin-wave term in the C_p of Ni_3Fe and would yield a better value of α. Unfortunately, the technique employed

TABLE IV. Least-squares parameters from fits of C_p data on ordered and disordered Ni_3Fe.

Number of parameters	α $\left(\frac{mj}{\text{g atom } K^{3/2}} \times 10^{+2}\right)$	γ $\left(\frac{mj}{\text{g atom } K^2}\right)$	β $\left(\frac{mj}{\text{g atom } K^4} \times 10^{+2}\right)$	Θ_d[a] (° K)	Number of points
		Ordered[b]			
2		3.301(±0.010)	2.286(±0.084)	439.7(± 5.1)	36
3	9.55(±2.01)	3.187(±0.024)	1.80 (±0.11)	476.6(± 9.4)	36
3	7.86(±2.09)	3.209(±0.025)	1.87 (±0.11)	470.2(± 8.9)	35[c]
3	5.52(±2.19)	3.240(±0.027)	1.97 (±0.11)	462.2(± 8.9)	34[c]
3	− 2.54(±2.10)	3.346(±0.027)	2.31 (±0.10)	438.3(± 6.3)	32[c]
3	9.66(±2.11)	3.186(±0.025)	1.79 (±0.11)	477.2(± 9.4)	35[d]
3	10.16(±2.19)	3.180(±0.026)	1.76 (±0.12)	480.0(±10)	34[d]
3	11.35(±2.32)	3.167(±0.027)	1.67 (±0.13)	488.0(±12)	32[d]
		Disordered[b]			
2		4.178(±0.009)	2.432(±0.083)	430.8(± 4.9)	41
3	10.44(±1.81)	4.053(±0.022)	1.888(±0.097)	468.8(± 7.8)	41
3	7.44(±1.82)	4.088(±0.022)	2.008(±0.094)	459.3(± 7.0)	40[c]
3	6.74(±2.08)	4.101(±0.026)	2.05 (±0.10)	456.1(± 7.5)	39[c]
3	− 1.10(±3.09)	4.206(±0.040)	2.38 (±0.14)	433.9(± 8.1)	36[c]
3	11.61(±1.75)	4.041(±0.021)	1.810(±0.095)	475.4(± 8.0)	40[d]
3	12.02(±1.82)	4.036(±0.022)	1.78 (±0.10)	478.1(± 8.7)	39[d]
3	13.41(±1.98)	4.021(±0.023)	1.690(±0.12)	487.0(±11)	36[d]

[a] $\Theta_d = 124.83(\beta)^{-1/3}$, the Debye temperature.
[b] Terms in parentheses are standard deviation of parameters.
[c] Data dropped from low-temperature end.
[d] Data dropped from high-temperature end.

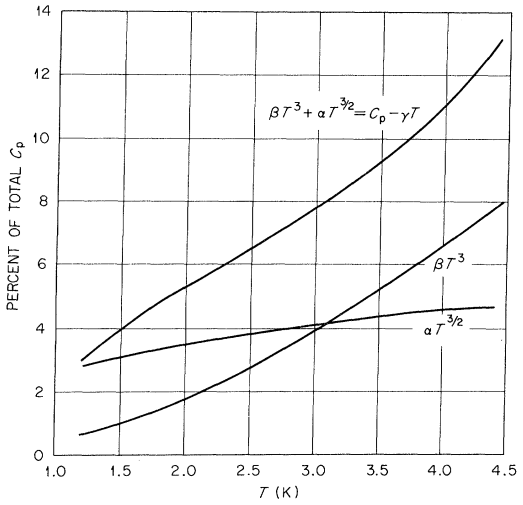

FIG. 5. Contribution in percent of the $\alpha T^{3/2}$ and βT^3 terms to total C_p of disordered Ni_3Fe. ($\alpha = 0.1044 \times 10^{-3}$ j/g atom $K^{5/2}$, $\beta = 0.01888 \times 10^{-3}$ j/g atom K^4.)

herein had a low-temperature limit of 1.2 K.

In recapitulation, neither Eq. (1) nor Eq. (4) appears to be the true functional form of the C_p: T relationship because of the periodicity noted in Figs. 3 and 4. However, to within the experimental uncertainties, both equations are adequate representations of the data. Statistically speaking, the standard deviations of the fits are substantially improved below 3 K by use of the $\alpha T^{3/2}$ term, but inclusion of this term has a detrimental effect on the standard deviations of β and γ. Data below 1.2 K would help establish whether Eq. (1) or (4) is more exact.

IV. DISCUSSION OF RESULTS

A. Comparison with Other Investigations

To the authors' knowledge, no other low-temperature measurements of C_p of ordered and disordered Ni_3Fe exist in the literature. However, Dixon, Hoare, and Holden,[4] Gupta, Cheng, and Beck [12] and Keesom and Kurrelmeyer[13] performed measurements between 1 and 20 K on a series of disordered nickel-iron alloys. The latter two groups of authors employed the two-parameter expression to interpret their data, whereas Dixon *et al.* used the three-parameter expression. The values of γ and Θ_d, the Debye temperature, of these authors are shown in Figs. 6 and 7, respectively. (Note: Θ_d is proportioned to $\beta^{-1/3}$ and it is felt that the results are more meaningful if discussed in term of Θ_d rather than β.)

At a given composition, the maximum difference in the γ values of Fig. 6 is about 8%, implying that the C_p measurements are essentially in agreement and that the value of γ is relatively insensitive to the mathematical form of second-order terms. Results of this investigation on disordered Ni_3Fe are plotted for comparison and illustrate the excellent agreement (0.2%) in γ with that of Dixon *et al.*

Figure 7 illustrates the extreme sensitivity of Θ_d to the functional form used to extract it from C_p measurements. Note, however, that the Θ_d values of this investigation for disordered Ni_3Fe agree within the combined experimental errors (about 18°) with that of Gupta *et al.* for the two-parameter fit and with that of Dixon *et al.* for the three-parameter fit. Again, the unresolved question of the last section appears: Which is the correct function, the two- or three-parameter one?

Using a wide variety of solids, Alers[3] has demonstrated that Debye temperatures calculated from calorimetric measurements are within 2% of those calculated from elastic constant measurements if all measurements are made below $\frac{1}{150}\Theta_d$. In the case of the ferromagnetic transition metals such as nickel, this excellent agreement was only achieved when the spin-wave term was included in the expression for C_p. For comparison, the elastic constant measurements of Θ_d of disordered Ni_3Fe obtained by Bower, Claridge, and Tsong[14]

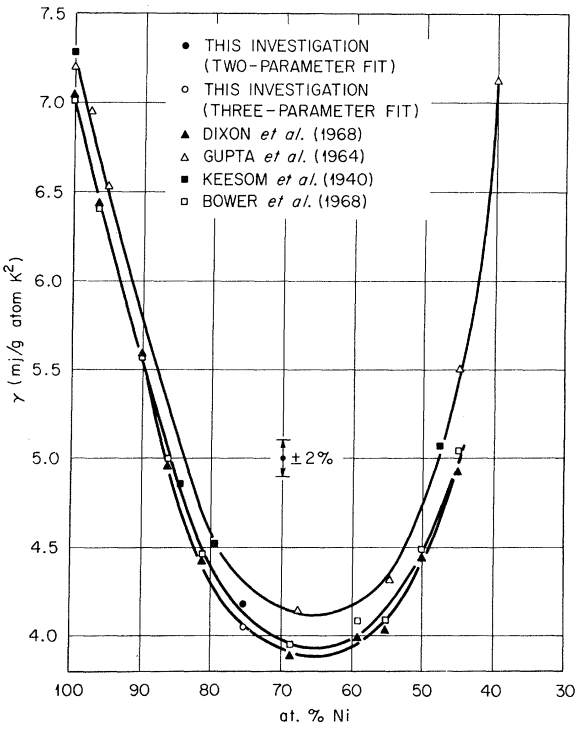

FIG. 6. Temperature coefficient γ of the electronic contribution to C_p of disordered nickel-iron alloys.

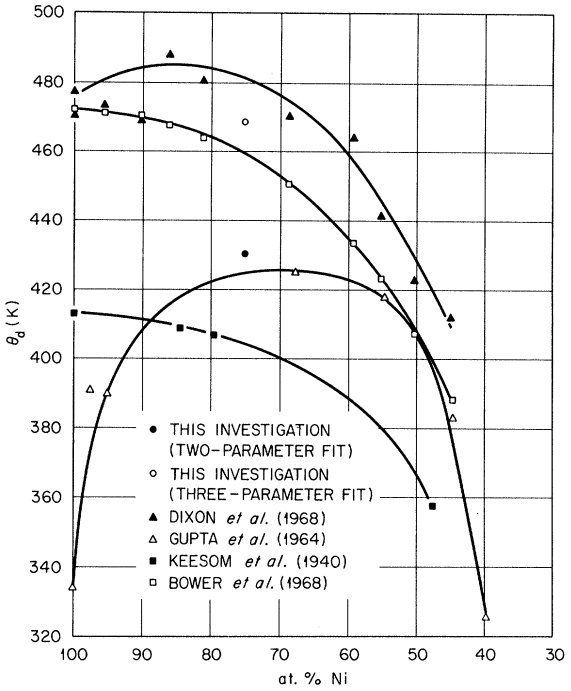

FIG. 7. Debye temperature Θ_d of disordered nickel-iron alloys.

are also shown in Fig. 7. If the criterion of agreement of calorimetric and elastic constant determinations of Θ_d is employed, the conclusion is reached that exculsion of the spin-wave term from the C_p results leads to erroneous Θ_d's for nickel-iron alloys.

If the spin-wave term is truly present, how then does the value of the spin-wave coefficient α compare with those calculated from other measurements? Figure 8 attempts to answer this question and includes α's derived from specific-heat capacity measurements,[4] specific-heat capacity measurements in conjunction with elastic-constant measurements,[14] neutron-scattering measurements,[15] spontaneous-magnetization measurements,[16] and Curie-temperature measurements.[17] Obviously, the specific-heat capacity α's are of the correct order of magnitude but are too high for Ni_3Fe. However, since spin-wave theory is only a first approximation for the magnetic contribution to C_p, this agreement tends to substantiate the presence of the spin-wave $\alpha T^{3/2}$ contribution.

Therefore, although purely mathematical reasoning did not demonstrate the superiority of the two- or three-parameter fits, the physical interpretation of the resulting parameters Θ_d and α reveals the three-parameter fit to be more theoretically meaningful for disordered Ni_3Fe.

B. Comparison of Results for Ordered and Disordered States

The measurements described herein demonstrate that Θ_d increases 1.7% (7.8 K), that α decreases 8.5%, and that γ decreases 21.5% when Ni_3Fe is configurationally ordered. These results are discussed below and the theoretical implications of them them are given where possible.

1. *Debye Temperature* Θ_d

To the authors' knowledge, no measurements of the Debye temperature of ordered Ni_3Fe exist in the literature. However, measurements[11,18,19] on structurally similar Cu_3Au show increases in Θ_d upon ordering of between 1 and 6%. Therefore, the change in Θ_d noted herein is reasonable in comparison with existing knowledge.

2. *Temperature Coefficient* α *of the Spin-Wave Contribution*

No measurements of the spin-wave coefficient α have been made, to our knowledge, on ordered Ni_3Fe. However, the ratio of the ordered to dis-

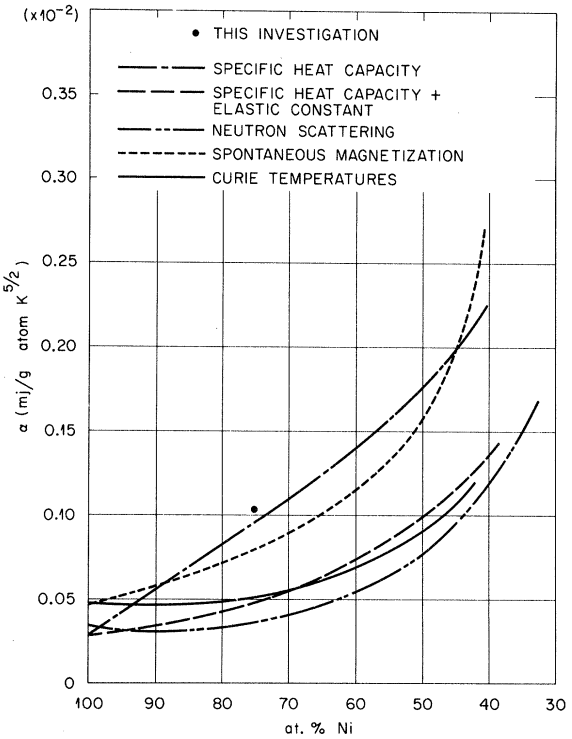

FIG. 8. Temperature coefficient α of spin-wave contribution to C_p of disordered nickel-iron alloys derived from this investigation, specific-heat capacity (Ref. 4), specific-heat capacity plus elastic-constant (Ref. 14), neutron scattering (Ref. 15), spontaneous-magnetization (Ref. 16), and Curie-temperature (Ref. 17) measurements.

ordered α can be shown to be

$$\frac{\alpha_0}{\alpha_d} = \left(\frac{4\bar{S}_0+1}{4\bar{S}_d+1}\right)^{3/2}\left(\frac{\Theta_C^d}{\Theta_C^0}\right)^{3/2}, \quad (7)$$

where $\bar{S}$ is the average spin and Θ_C is the Curie temperature. Crangle and Hallam[16] measurements yield a $\bar{S}_d$ of 1.1 and this coupled with the saturation magnetization data of Wakelin and Yates[17] gives $\bar{S}_0$ of 1.15. Kollie[7] has shown that Θ_C^d is 871 K and that Θ_C^0 is 940 K; hence, α_0/α_d is 0.94. The experimental data listed in Table IV yield an α_0/α_d of 0.915, which is in good agreement with the theoretical calculation.

3. *Temperature Coefficient γ of Electronic Specific-Heat Capacity*

There has been considerable discussion in the literature as to the phenomena contributing to the γT term of the low-temperature specific-heat capacity of metals. (See, for example, Refs. 11, 12, and 18–27.) For a noninteracting degenerate electron gas, the contribution to C_p at low temperatures is[1]

$$C_{ve} = \gamma T = \tfrac{2}{3}(\pi k)^2 N(E_F) T, \quad (8)$$

where $N(E_F)$ is the density of electronic states at the Fermi energy E_F, and k is Boltzmann's constant. Equation (8) is changed little if electron-electron interactions are considered.[1,20] However, if electron-phonon interactions occur, the expression for C_{ve} becomes[21]

$$C_{ve} = \gamma T = \tfrac{2}{3}(\pi k)^2 N(E_F)\, T\left[1+(\tfrac{1}{2}m)^{1/3}F\right], \quad (9)$$

where F is the interaction constant and m the number of free electrons per atom. For transition metals, the second or electron-phonon interaction term in Eq. (9) is thought to be as large as the first term, which is a free-electron-type contribution.

A magnetic contribution γ_m to γ has been proposed by Overhauser,[26] Marshall,[27] and Gupta, Cheng, and Beck.[12] Overhauser's theory for γ_m concerns the formation of long-range antiferromagnetic order in a paramagnetic solute by a static spin-density wave in the conduction electrons of the solvent (e.g., manganese in copper). Conversely, Marshall's theory, which employs the Ruderman-Kittel-Yosida model of spin-spin coupling of the paramagnetic solute via the conduction electrons, requires only a small fraction of the solute spins to be unaligned in a small effective field. Both of these theories were derived for *dilute* solutes. However, Marshall suggested that the anomalously large γ values obtained by Wei, Cheng, and Beck[28] in vanadium-iron and chromium-iron alloys could be explained by his model. Subsequently, Gupta, Cheng, and Beck[12] extended Marshall's model to explain the large γ's observed in nickel-manganese and nickel-iron alloys by assuming that compensating ferromagnetic and antiferromagnetic exchange interactions in small volume regions produce the required small effective fields in these alloys.

For the three contributions discussed above, C_{ve} for an alloy such as Ni_3Fe becomes

$$C_{ve} = \gamma T = \left\{\tfrac{2}{3}(\pi k)^2 N(E_F)\left[1+(\tfrac{1}{2}m)^{1/3}F\right]+\gamma_m\right\}T. \quad (10)$$

Consequently, changes in the band structure, the electron-phonon interaction, or the effective field would account for the 21% change in γ noted when Ni_3Fe was ordered configurationally. Unfortunately, the theories employed to derive Eq. (10) cannot be used to predict quantitative effects caused by configurational ordering. Only plausibility arguments can be given for each. For example, Goldman[24] found that γ of Ni_3Mn decreased by a factor of 2 upon ordering. Using Slater's[23] band model, Goldman asserted that the superposed double periodicity of the lattice potential of the ordered alloy splits the d band and alters the density-of-states energy curve such that the half-band of one spin is full or nearly full; hence, only d electrons of the other spin contribute to γ of the ordered alloy. On the other hand, Gupta *et al.*[12] attributed the decrease in γ on ordering to the disappearance of a large γ_m contribution. They found that γ of disordered Ni_3Mn decreased 10.4% when measured in an applied field of 14.5 kOe, whereas γ of ordered Ni_3Mn did not. Gupta *et al.* postulate that the applied field increased the effective fields in the disordered alloy, reducing γ_m. They also state that Marshall's theory does not predict a γ_m contribution in ordered Ni_3Mn since Mn-Mn antiferromagnetic exchange interactions are not possible because the ordered manganese atoms have 12 nearest-neighbor nickel atoms. Thus, application of an external field should not change γ of ordered Ni_3Mn.

Both Goldman and Gupta *et al.* have suggested that their results were applicable to nickel-iron alloys. However, Dixon and Stravely[29] (see Ref. 14) were able to explain the in-field decreases in C_p of disordered nickel-iron alloys using spin-wave theory; that is, the external magnetic field increases the energy gap inthe spin-wave spectrum and in effect reduced α.[30] Thus, it is the opinion of the authors that near-zero fields do not exist in disordered Ni_3Fe and consequently the $\gamma_m T$ contribution is not operable in this alloy, whether ordered or disordered.

In view of the above, it is our opinion that the change in γ noted upon ordering Ni_3Fe is due to a decrease in $N(E_F)$ and/or to a decrease in the electron-phonon interaction brought about by the signifi-

cant increase in the periodicity of the lattice potential.

ACKNOWLEDGMENTS

Dr. C. R. Brooks, the University of Tennessee, is thanked for his suggestions and discussion on this work. Helpful comments on the paper were received from Dr. H. L. Davis, J. P. Moore, Dr. O. M. Stocks, and Dr. W. Fulkerson. R. E. McDonald is thanked for fabrication of the specimen.

*Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corp. tion.

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[30]K. G. Gupta *et al*. (Ref. 12) did not employ the $\alpha T^{3/2}$ spin-wave contribution in analyzing their C_p measurements on Ni_3Mn. A ratio α_d/α_0 of about 3 was calculated for Ni_3Mn using Curie temperatures given by M. Hansen [*Constition of Binary Alloys* (McGraw-Hill, New York, 1958)], the saturation magnetization data of G. R. Piercy and E. R. Morgan [Can. J. Phys. 31, 529 (1953)], and Eq. (7). Because the spin-wave contribution in disordered Ni_3Mn is about 5% of C_p near 4 K, decreases in C_p of several percent would be expected in fields of 14.5 kOe. However, the effect of this field on the spin-wave contribution of ordered Ni_3Mn would be too small to be discernible. Thus, the results of Gupta *et al*. can be explained qualitatively by spin-wave theory.

FIG. 1. Tranverse section of quenched Ni_3Fe specimen etched with ammonium persulfate (×200 original magnification). Difference in brightness of grains is due to orientation differences, including twinning.