Effect of magnetic field on the heat capacity of Ni_3Al alloys

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(Received 28 July 1988; revised manuscript received 31 October 1988)

The heat capacity C of three alloys of nominal composition $Ni_{74}Al_{26}$, $Ni_{74.25}Al_{25.75}$, and $Ni_{74.5}Al_{25.5}$ (hereafter, alloys I, II, and III, respectively) has been measured in zero and applied fields of 2.5, 5.3, 7.5, and 9.8 T in the range 1.5–20 K. The zero-field C/T-versus-T^2 plots show a mild upturn in all three alloys at temperatures below ~ 10 K. No discernible effect on heat capacity is observed in the 2.5-T field for alloys I and II, and only at higher fields does the upturn get suppressed. In the maximum applied field of 9.8 T, the C/T-versus-T^2 plots are linear. For alloy III (magnetization measurements at 4.2 K show this material to be magnetically ordered), the upturn is completely removed in the 2.5-T field and the heat capacity decreases progressively in increasing applied fields at all temperatures. The origin of the upturn in C/T is discussed as arising from either spin fluctuations or magnetic clusters. While the zero-field data can be interpreted using either of the two models, the data obtained in applied fields strongly suggest that the upturn in C/T plots is caused by spin fluctuations and that spin fluctuations in these materials are depressed in applied magnetic fields.

INTRODUCTION

The magnetic and other physical properties of the intermetallic compound Ni_3Al have been studied extensively over the past two decades. The continuous interest in the study of this system can be attributed to its magnetic properties. Over a relatively narrow composition range, from 72.5 to 77 at. % Ni, the compound crystallizes in the simple $AuCu_3$-structure type with Al atoms occupying the cube corners and Ni atoms at the face-center positions. In the composition range 73.5–74.5 at. % Ni the alloys exhibit strong exchange-enhanced paramagnetism; at higher Ni concentrations, ferromagnetism sets in.[1] The saturation magnetic moment and the transition temperature of the ferromagnetic alloys vary smoothly over the composition range. From the shape of the plots of reciprocal magnetic susceptibility as a function of temperature and the observation of linear σ^2-versus-(H/σ) plots, where σ is the magnetization, De Boer *et al.*[1] concluded that the magnetism of this system is of the itinerant type, qualitatively similar to that discussed within the framework of the Stoner collective-electron model. This initial classification of Ni_3Al as a weak itinerant ferromagnet, which put it in the class of $ZrZn_2$, naturally led to an extensive study of the various physical properties across the whole composition range. For example, volume magnetostriction measurements were carried out and explained on the basis of the Stoner model.[2]

More recently, the self-consistent renormalization theory of spin fluctuations,[3] which takes into account the coupling between different modes of spin fluctuations in a self-consistent way, as opposed to the Stoner model, where only single-particle excitations are considered, has been successfully applied to Ni_3Al to explain the temperature dependence of magnetization, resistivity, magnetoresistance,[4] nuclear-spin-lattice relaxation time,[5] magnetostriction,[6] thermal expansion,[7] and the dependence of the Curie temperature on Ni concentration.[8] This shows that spin fluctuations are significant in this system. A similar conclusion has been derived from band-structure calculations[9] and de Haas–van Alphen (dHvA) effect measurements on stoichiometric Ni_3Al.[10] By performing self-consistent calculations using the augmented-spherical-wave (ASW) method, Buiting *et al.*[9] found their calculated density of states to be far lower than that obtained from existing heat-capacity data,[11,12] and concluded that the renormalization in Ni_3Al due to spin fluctuations is extremely large. They obtained values of renormalization parameter (γ_{expt}/γ_0)-1 ranging from 1.54 to 1.95. dHvA-effect measurements on a sample of Ni_3Al in fields up to 10 T reveal appreciable electronic mass enhancements ranging from 2 to 3.4 depending on the character of the band states.

The enhancement of the effective electronic mass and, therefore, the electronic heat capacity due to spin fluctuations in exchange-enchanced paramagnetic and weakly ferromagnetic materials, was predicted on theoretical grounds within the framework of paramagnon model long ago[13,14] and, more recently, within the self-consistent renormalization theory of spin fluctuations.[15] Qualitatively, the results of both the models are similar. Theory further predicts an additional contribution to the heat capacity of the form $T^3\ln(T/T_{SF})$ for $T < T_{SF}$, where T_{SF} is the spin-fluctuation temperature. The presence of this contribution gives rise to an upturn in (C/T)-versus-T^2 plots at low temperatures. The paramagnon theory also predicts that sufficiently large magnetic fields, of the order of T_{SF}, would quench spin fluctuations and lead to a reduction of the electronic heat capacity.[16–18]

Experiments have shown that in some exchange-enhanced paramagnetic materials, e.g., $CeSn_3$,[19] UAl_2,[20] $TiBe_2$,[21] $CeSi_x$,[22] RCo_2 (R = Y,Lu,Sc),[23] etc., the coefficient of the linear term in the electronic heat capacity, γ,

is relatively large and is lowered in applied magnetic fields compared to its value in the zero field. Except for RCo_2, these materials have a $T^3 \ln T$ term in the heat capacity at low temperatures which gives rise to an upturn in (C/T)-versus-T^2 plots. In $CeSn_3$, for example, the upturn is completely removed in an applied field of 5.3 T, while in $TiBe_2$ the onset of spin fluctuations depression occurs at 5.2 T.

The heat capacity between 1.5 and 15 K in zero field of Ni_3Al alloys for Ni compositions varying from 73 to 76 at. % has been studied earlier[11] to look for effects predicted by the paramagnon theory. A relatively small upturn in C/T was observed in all alloys. An analysis of some of the plots gave nearly the same spin-fluctuation temperature, although the Stoner enhancement varied by a factor of 5, contrary to what one would expect on the basis of paramagnon theory. Therefore, deDood and deChatel[11] concluded that the upturn was not related to spin-fluctuation effects. They[11] also measured the heat capacity of a related system, Ni_3Ga, between 1.5 and 15 K, and for some of these alloys they extended the measurements down to 0.2 K. The upturn in Ni_3Ga alloys turned out to be the high-temperature side of a Schottky-like peak at lower temperatures arising due to magnetic clusters. This observation may also have prompted them to assume a similar physical situation in Ni_3Al alloys. However, an explanation of Ni_3Al heat-capacity data based on magnetic clusters also ran into difficulties (the reader may refer to Ref. 11 for details) and, thus, the origin of the upturn remained unclear.

In the present work we have readdressed this question and have measured the heat capacity of three alloys of nominal composition $Ni_{74}Al_{26}$, $Ni_{74.25}Al_{25.75}$, and $Ni_{74.5}Al_{25.5}$ in zero and applied fields up to 9.8 T. In what follows, we shall refer to these alloys as I, II, and III, respectively. Our field data strongly suggest that the upturn in these alloys is associated with spin fluctuations. We also find that spin fluctuations are depressed in applied fields in these alloys.

EXPERIMENTS AND RESULTS

The samples were prepared by melting appropriate amounts of Ni and Al in an argon atmosphere in an arc furnace. The alloy buttons were repeatedly melted to ensure homogenization. They were given an annealing of about 4 weeks at 1160 °C and furnace cooled. The buttons were then cut on a diamond saw to get a sample of desired shape and mass (about 10 g) for the heat-capacity measurements. The samples were further annealed at 750 °C for 24 h and the furnace temperature decreased at the rate of 50 °C/d. Metallographic examination revealed all the samples to be single-phase materials. Heat capacity was measured in the temperature range 1.5–20 K using a semiadiabatic heat-pulse technique with a germanium-resistance thermometer (GRT) calibrated at 0, 2.5, 5.3, 7.5, and 9.8 T.

Figures 1–3 show the heat capacity in the form of (C/T)-versus-T^2 plots in the temperature range 1.5–6 K. Data taken over the entire temperature range, 1.5–20 K,

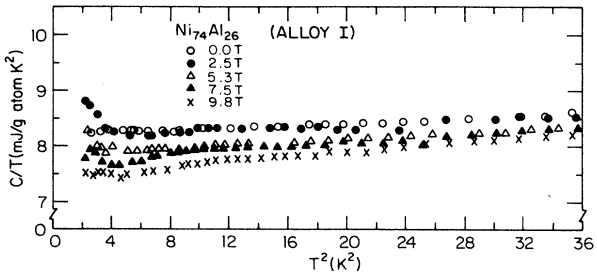

FIG. 1. Heat capacity in the form of C/T vs T^2 of alloy I (nominal composition $Ni_{74}Al_{26}$, see text) in the (1.5–6)-K range for $H=0$, 2.5, 5.3, 7.5, and 9.8 T.

are depicted in Figs. 4–6. C/T curves in zero field show a gentle upturn beginning at temperatures below ~10 K in all three alloys (see Figs. 4–6) and the upturn is gradually suppressed in applied fields. For alloys I and II the upturn is almost removed in a 7.5-T field, while for alloy III the upturn is completely removed in 2.5 T. In the maximum applied field of 9.8 T, the C/T plots for all three alloys are linear. It is noteworthy that the heat capacity is independent of field above ~12 K in alloys I and II, but for alloy III the heat capacity is reduced over the entire temperature range in applied fields. It is also observed that the applied field of 2.5 T has no effect on the upturn in alloys I and II. The different heat-capacity response of alloy III in applied fields compared to that of alloys I and II was rather unexpected. We therefore took magnetization data of our samples at a few selected temperatures and fields, using the Faraday method, and compared it with the results reported in literature. We find all three alloys are slightly richer in Ni content than given by their nominal composition. The susceptibility of alloy II is slightly smaller than that reported for the $Ni_{74.5}Al_{25.5}$ composition, which is paramagnetic.[7] Alloy I is less magnetic than II, but has a susceptibility greater than that reported for this nominal composition in the literature.[7] The susceptibility of alloy III is of the same magnitude as reported for $Ni_{75}Al_{25}$, which orders magnetically at 40 K. Indeed, a plot of σ^2 versus H/σ at 4.2 K for alloy III gave a positive intercept on the σ^2 axis, corresponding to a spontaneous moment of $0.076\mu_B$/Ni. We draw the important conclusion from magnetization measurements that only alloys I and II are paramagnetic,

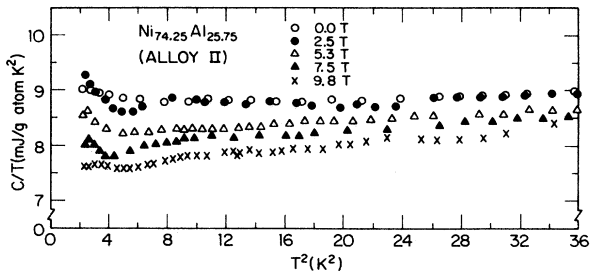

FIG. 2. Heat capacity in the form of C/T vs T^2 of alloy II (nominal composition $Ni_{74.25}Al_{25.75}$, see text) in the (1.5–6)-K range for $H=0$, 2.5, 5.3, 7.5 and 9.8 T.

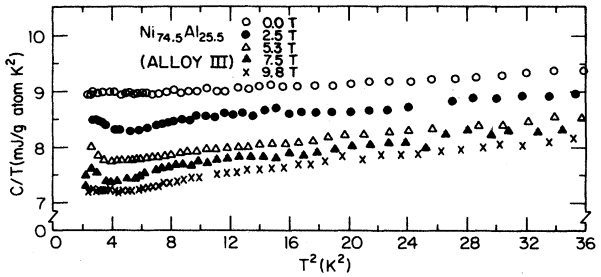

FIG. 3. Heat capacity in the form of C/T vs T^2 of alloy III (nominal composition $Ni_{74.5}Al_{25.5}$, see text) in the (1.5–6)-K range for $H=0$, 2.5, 5.3, 7.5, and 9.8 T.

while alloy III magnetically orders with a Curie temperature near 40 K.

DISCUSSION

Heat capacity in zero field

For a normal metal the heat capacity at low temperatures is usually represented by the following expression:

$$C/T=\gamma+\beta T^2 , \quad (1)$$

where γ and β represent, respectively, the electronic and lattice contributions to heat capacity. Equation (1) normally holds for $T\leq\Theta_D/(50\text{ K})$, Θ_D being the Debye temperature. Θ_D can be obtained from β by using the relation $\Theta_D^3=1.9437\times10^6/\beta$, where β has the units of mJ/g-at. K^4. Elastic constants[24] and inelastic-neutron-scattering measurements[25] on Ni_3Al give a Debye temperature of 470 K. Between 0 and 15 K, the Debye temperature decreases slightly.[25] For the present analysis, we take Θ_D as 465 K, which corresponds to a β value of 0.01927 mJ/g-at. K^4. The zero-field C/T plots depicted in Figs. 1–3 are nearly linear, except for a slight upward curvature in alloy II (see Fig. 2). However, a fit of the zero-field data from 1.5 to 6 K for alloys I and III to Eq. (1) gives Debye temperatures of 653 and 548 K, respectively, which are grossly incompatible with the known value. Robbins and Claus, who proposed that magnetic ordering in Ni_3Al alloys evolved as a result of interactions between magnetic clusters, had analyzed their heat-capacity data[26] in the temperature range 1.5–5 K by including an extra term (varying as $1/T$) in Eq. (1) to account for the cluster contribution. A value of 524 K is obtained for the Debye temperature from their analysis which is, again, not in accordance with the inelastic-neutron-scattering results. Therefore, in obtaining proper values of Θ_D and γ from the heat capacity of Ni_3Al, a more extended temperature range must be considered. This is not surprising considering the upturn involved in C/T. deDood and deChatel[11] fitted their data to Eq. (1) in the range 10–15 K above the upturn, where C/T plots are linear to obtain γ values for various compositions.

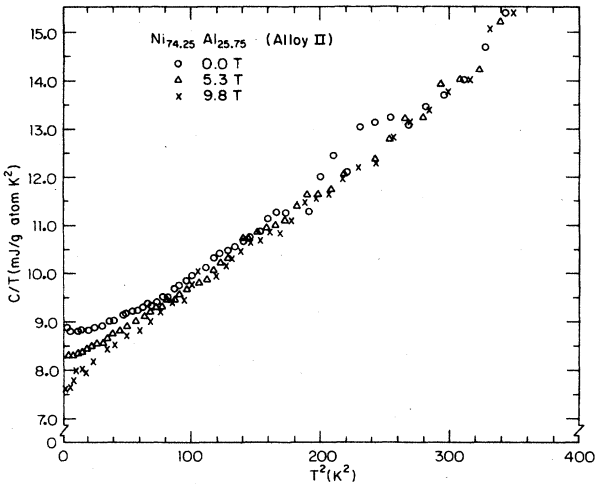

FIG. 5. C/T vs T^2 of alloy II (see text) in the (1.5–20)-K range for $H=0$, 5.3, and 9.8 T.

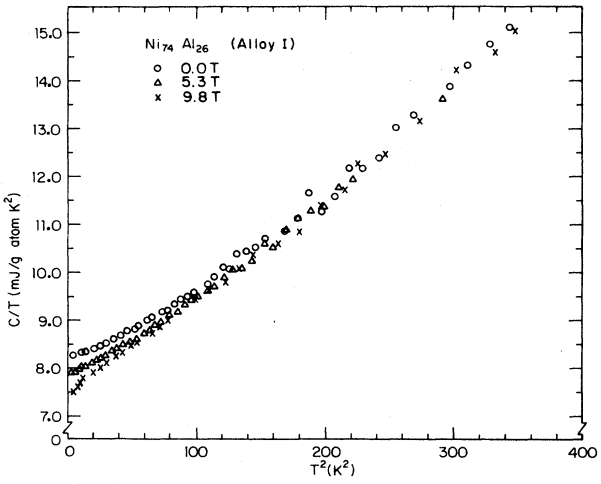

FIG. 4. C/T vs T^2 of alloy I (see text) in the (1.5–20)-K range for $H=0$, 5.3, and 9.8 T.

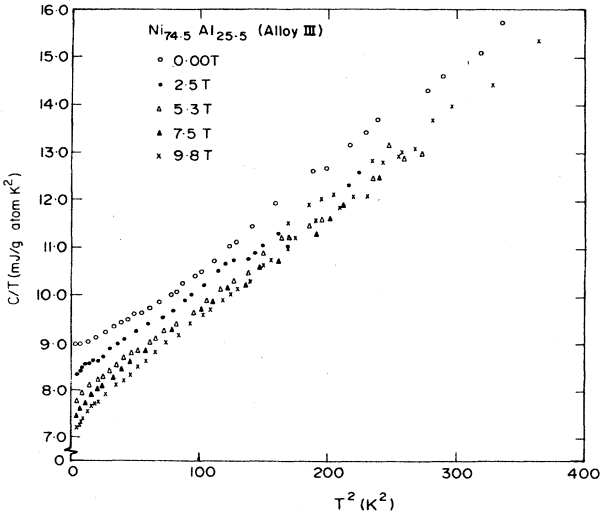

FIG. 6. C/T vs T^2 of alloy III (see text) in the (1.5–20)-K range for $H=0$, 2.5, 5.3, 7.5, and 9.8 T.

They find $\beta=0.0325$ mJ/g-at. K^4, which gives a Θ_D of 390 K. The large discrepancy in the β value of deDood and deChatel is surprising. The Debye temperature of Ni_3Al is rather high and one might expect Eq. (1) to be applicable for the (10–15)-K regime, which is above the upturn. To check this, we have fitted the present data to Eq. (1) in the same temperature regime, and the results are listed in Table I. The coefficient of lattice heat capacity, β, thus obtained for alloys I and II is in excellent agreement with the estimates based on elastic constants and inelastic-neutron-scattering measurements. For alloy III, the β value is slightly larger. Since this alloy orders magnetically, additional magnetic contribution to the heat capacity may result in a larger β from the fitting procedure. We infer from the results of our high-temperature fit that the sample quality of our alloys is excellent. Because of the curvature in C/T at low temperatures the γ_{HT} values listed in Table I may not represent the actual value of the coefficient of electronic heat capacity in these materials. However, they can definitely be taken as the "lower-bound" estimates of this quantity.

The low-temperature upturn below 10 K in the heat capacity could have its origin most likely either in spin fluctuations or magnetic clusters. In the paramagnon model, the heat capacity at temperatures $T<T_{SF}$ is given by

$$C=\gamma_0 T[m^*/m+\alpha(T/T_{SF})^2\ln T/T_{SF}]+\beta T^3 . \quad (2)$$

m^*/m is the zero-temperature many-body mass enhancement, γ_0 is given by the band-structure density of states, α is proportional to $S(1-S^{-1})^2$, where S is the Stoner-enhancement factor, and βT^3 is the usual lattice contribution. A fit of the zero-field data of the three alloys in the temperature range 1.5–12 K to the equation

$$C/T=A+BT^2+DT^2\ln T \quad (3)$$

represents the data quite well, where $A=(m^*/m)\gamma_0=\gamma$, $B=\beta-D\ln T_{SF}$, and $D=\alpha\gamma_0/T_{SF}^2$. (We may mention here that a fit based on cluster contribution appears to be almost equally good; see below.) The coefficients A, B, and D are listed in Table I. $A(\equiv\gamma)$ increases with increasing enhanced paramagnetism (i.e., increasing S) in alloys I and II. Theoretically, the mass enhancement is given by $m^*/m=1+\delta\ln S/3$. In the zero-range paramagnon model, which overestimates the mass enhancement, $\delta=\frac{9}{2}$. When the finite range of interaction[27] is taken into account, mass enhancement is found to be lower, which is more in accord with the experiment.

The coefficient of the $\ln T$ term is small, but its relative magnitude for the three compositions appears to follow the expected trend. As one approaches the critical composition of the onset of ferromagnetism, one expects the spin fluctuations and, therefore, the upturn—if it is associated with spin fluctuations—to become stronger. For alloy II, which is (paramagnetic but) very close to magnetic instability, D is about 50% larger than in alloy I. Once magnetic order sets in, D should decrease. That is exactly what is observed for alloy III. It is plausible to argue that D would become vanishingly small as Ni concentration is increased further and as one moves away

TABLE I. Least-squares-fit parameters of the zero-field heat-capacity data of the three Ni_3Al alloys. γ_{HT} and β are the results of the fit of the zero-field heat-capacity data in the range 10–15 K to $C/T=\gamma+\beta T^2$. A, B, and D are the fit coefficients for the zero-field data in the (1.5–12)-K range to $C/T=A+BT^2+DT^2\ln T$. A', B', and D' are the fit parameters obtained by fitting the (1.5–12)-K data to cluster model $C=A'+B'T+D'T^3$. The error limits are the standard deviations of the fit parameters.

Alloy	γ_{HT} (mJ/g-at. K^2)	β (mJ/g-at. K^4)	Θ_D (K)
I	7.63±0.15	0.0197±0.0010	462
II	8.03±0.10	0.0191±0.0007	467
III	8.17±0.15	0.0225±0.0009	442

Alloy	A (mJ/g-at. K^2)	B (mJ/g-at. K^4)	D (mJ/g-at. $K^4\ln K$)	T_{SF} (K)
I	8.32±0.01	−0.0114±0.0009	0.0107±0.0004	17.7
II	8.93±0.01	−0.0274±0.0011	0.0161±0.0004	18.1
III	8.95±0.01	−0.0035±0.0006	0.0083±0.0002	

Alloy	A' (mJ/g-at. K)	B' (mJ/g-at. K^2)	D' (mJ/g-at. K^4)
I	0.716±0.002	7.92±0.01	0.0159±0.0001
II	1.211±0.021	8.23±0.01	0.0159±0.0001
III	0.468±0.021	8.67±0.01	0.0177±0.0001

from the regime of weak ferromagnetism to relatively strong ferromagnetism. The (C/T)-versus-T^2 curve would, in that case, show a linear behavior far below the Curie temperature.

These remarks may be quite pertinent in considering the heat-capacity results on single crystals of Ni_3Al by Ho *et al.*[28] and polycrystalline Ni_3Al by Collocott *et al.*[29] The former report measurements between 2 and 14 K in zero field on two single-crystal samples and find linear (C/T)-versus-T^2 behavior. However, the γ values of the two samples differ appreciably, i.e., 8.48 and 7.72 mJ/g-at. K^2, respectively. They give the following composition for their two specimens: Ni, 87.27%; Al, 11.3%; and Ni, 87.68%; Al, 11.47%, by weight, which corresponds to the formulas $Ni_{78}Al_{22}$ and $Ni_{77.84}Al_{22.15}$, respectively, surprisingly higher than the limiting single-phase stoichiometry $Ni_{77}Al_{23}$. Collocott *et al.* have measured the heat capacity of nominally stoichiometric Ni_3Al between 0.5 and 15 K. They find some evidence of an upturn in C/T below 2 K where the data show appreciable error bars. They fit the data to Eq. (1) and obtain a γ value of 8.95 mJ/g-at. K^2 and a Θ_D of 475 K. We feel that Eq. (1) is not a good representation of their data, particularly at low temperatures, i.e., below about 5 K.

We mention here that in the theory of Moriya and Kawabata[30] the square of the spontaneous magnetization behaves as $T^{4/3}$ in some temperature range below T_c and this is considered to arise due to spin fluctuations. Experimentally, it is observed that the temperature range in which this behavior is valid increases as one approaches the critical composition from the ferromagnetic side.[8] This means that spin-fluctuation effects become weaker as one moves away from the critical composition. Makoshi and Moriya[15] have considered the effect of spin fluctuations on the heat capacity and have given a graphic depiction of C/T (here, C denotes only the electronic contribution) at low temperatures for various values of α close to the ferromagnetic instability, where α is defined as the product $I\chi_0/2$; I is the interaction constant and χ_0 the susceptibility without electron-electron interaction.[15] It is found that C/T increases and the temperature range, where C/T remains nearly constant, decreases as the system approaches the ferromagnetic instability from either end. We suggest that in the Ni_3Al system the spin-fluctuation-induced upturn in the heat capacity may not exist across the whole composition range and, therefore, stoichiometry is an important consideration.

It is possible to estimate the spin-fluctuation temperature T_{SF} by using the relation $B=\beta-D\ln T_{SF}$. Using $\beta=0.019\,27$, we get $T_{SF}=17.7$ and 18.1 K, respectively, for alloys I and II. These are listed in Table I. The spin-fluctuation temperature is nearly the same for both of the compositions, which is rather contrary to one's expectation of observing a decreasing T_{SF} with increasing susceptibility. As mentioned earlier, deDood and deChatel[11] also observed the same behavior for their alloys and took it as an indication that the upturn in their C/T plots was not caused by spin fluctuations. However, this apparently anomalous relationship between T_{SF}, as obtained from the application of the simple paramagnon model [Eqs. (2) and (3)] to the heat-capacity data, and the magnetic susceptibility (or Stoner enhancement) is also observed in other systems that have an appreciable $T^3\ln T$ contribution to their heat capacity, e.g., $CeSi_x$ (Ref. 22) and the pseudoternaries $U_xLa_{1-x}Al_2$.[31] More realistic models of spin fluctuations are needed.

While the above discussion shows that the present zero-field heat-capacity results are compatible with the predictions of the spin-fluctuation theory, there remains the alternative of explaining the upturn as arising due to magnetic clusters. A cluster-based fit,

$$C=A'+B'T+D'T^3\ , \tag{4}$$

describes the zero-field data almost equally well over most of the temperature range, except from $\sim$9 to 12 K, where it is marginally poorer compared to the $\ln T$ fit. The results of the clusters based fit are summarized in Table I. The Debye temperature deduced from D' for alloys I and II is larger than the known value by about 5%. However, this does not necessarily imply that the cluster-based fit is unphysical. The magnetic cluster contribution to heat capacity, A', is a constant only when $T\gg t_A$, where $t_A=T_A/S$ and $kT_A=E_A$,[32] where E_A is the average anisotropy energy of the cluster and S its spin. Normally, t_A is some tenths of a degree kelvin, but if it happens to be within the experimental range of measurement of a few degrees kelvin, then the cluster specific-heat contribution is no longer a constant, but follows an Einstein specific-heat function in the simplest approximation, t_A being the Einstein characteristic temperature.[33,34] In such a situation, fitting the data to Eq. (4) would enhance B' and lower D' (increase Θ_D).[32] We note that a polarized-neutron-diffraction study in a field of 10 kOe on a single crystal of ferromagnetic $Ni_{75.9}Al_{24.1}$ (Ref. 35) did not reveal the presence of any giant-moment clusters. It was found that the excess Ni atoms possess a moment which is nearly the same as the other Ni atoms. For substoichiometric Ni_3Al (i.e., Ni-deficient) alloys, it is therefore unlikely that any antistructure Ni atom sitting at Al site would form giant-moment clusters. The Ni_3Al system is known to have a highly ordered crystal structure and it does not favor preferential atomic clustering.

Field dependence of the heat capacity of paramagnetic alloys I and II

Although the zero-field data can be interpreted using either the spin-fluctuation or cluster-based models, we feel the data in applied fields favor the spin-fluctuation model. An inspection of Figs. 1 and 2 shows that an applied field of 2.5 T has practically no effect on the heat capacity of paramagnetic alloys I and II. If the upturn were caused by clusters, one would have observed at least a partial suppression of the upturn in a field of 2.5 T. On the other hand, if the upturn is due to $T^3\ln T$ term, then one would observe the diminishment of heat capacity and the associated upturn only at fields at which spin fluctuations begin to get quenched. For example, in $CeSi_{1.90}$, where the electronic heat capacity has a $T^3\ln T$ form at low temperatures, spin fluctuations are not quenched in

fields up to 7.5 T.[22] In what follows we shall base our conclusions on the spin-fluctuation model.

The upturn in C/T is distinctly reduced for $H=5.3$ T and higher fields and in a 9.8-T field C/T versus T^2 is linear. The γ ($\equiv A$) values obtained by fitting the data from 1.5 to 12 K in $H=0$, 2.5, 5.3, and 7.5 T to Eq. (3) and $H=9.8$ T data to Eq. (1) for alloys I and II are plotted in Fig. 7, and the fit coefficients are listed in Table II. For the data obtained in 7.5 T, where C/T versus T^2 is almost linear, there is little difference in the γ values obtained by fits to Eq. (1) or Eq. (3). In the maximum applied field of 9.8 T, we obtain $\beta=0.019\,62$ ($\Theta_D=463$ K) and $0.020\,84$ ($\Theta_D=454$ K) mJ/g-at. K^4 for alloys I and II, respectively. For alloy II, β is slightly larger than $0.019\,27$ mJ/g-at. K^4 (Refs. 24 and 25). It is possible that the applied field induces a moment in this sample which is almost magnetic. The induced moment gives rise to a magnetic contribution to the heat capacity which has a T^3 temperature dependence and, therefore, increases β. Such induced moment effects have been observed in $CeSn_3$,[19] $CeSi_x$,[22] Sc_3In,[36] and other materials. The magnitude of the induced moment in these systems was found to be quite small. It may be noted that, for alloy II, γ in a 9.8-T field is smaller than γ_{HT}, the "lower-bound" estimate of γ in zero field. Also, γ ($H=7.5$ and 9.8 T, see Table II) of alloys I and II is lower than B' [$B'\equiv\gamma(H=0)$ in the cluster model]. These observations strongly support the conclusion that spin fluctuations are partially quenched in applied fields (≥ 5.3 T) in the paramagnetic alloys I and II. In the maximum applied field of 9.8 T, $\Delta\gamma/\gamma(H=0)=-10.2\%$ and -15.2% in the two alloys, respectively.

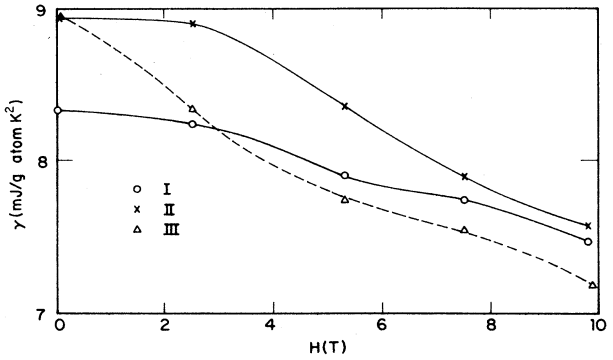

FIG. 7. Variation of the electronic specific-heat constant with the applied magnetic field for alloys I, II, and III. The lines drawn through the data points are guides to the eye. The errors associated with the least-squares fitting of the γ values are less than the size of the symbols.

It is interesting to compare the experimentally observed depression of γ with theoretical predictions. Béal-Monod *et al.*[17] give the following expression for $\Delta\gamma$:

$$\frac{\Delta\gamma}{\gamma(H=0)}=0.1\frac{S}{\ln S}\left[\frac{\mu_B H}{k_B T_{SF}}\right]^2. \tag{5}$$

In order to calculate $\Delta\gamma$, we need to know the values of the Stoner-enhancement factor S for alloys I and II,

TABLE II. Least-squares-fit parameters of the heat-capacity data in applied fields of the three Ni_3Al alloys. A, B, and D are the fit coefficients for the data in the (1.5–12)-K range to $C/T=A+BT^2+DT^2\ln T$; γ and β are the fit coefficients to $C/T=\gamma+\beta T^2$. The error limits are the standard deviations of the fit parameters.

Alloy	H (T)	A (mJ/g-at. K^2)	B (mJ/g-at. K^4)	D (mJ/g-at. K$^4\ln K$)
I	0	8.32±0.01	−0.0114±0.0009	0.0107±0.0004
	2.5	8.24±0.01	−0.0065±0.0012	0.0085±0.0005
	5.3	7.89±0.00	0.0045±0.0005	0.0048±0.0002
	7.5	7.74±0.01	0.0141±0.0014	0.0013±0.0005
II	0	8.93±0.01	−0.0274±0.0011	0.0161±0.0004
	2.5	8.89±0.03	−0.0286±0.0028	0.0166±0.0010
	5.3	8.35±0.02	−0.0092±0.0022	0.0101±0.0009
	7.5	7.89±0.01	0.0210±0.0015	0.0016±0.0006
III	0	8.95±0.01	−0.0035±0.0006	0.0083±0.0002

Alloy	H (T)	γ (mJ/g-at. K^2)	β (mJ/g-at. K^4)
I	9.8	7.47±0.00	0.0196±0.0001
II	9.8	7.57±0.00	0.0208±0.0001
III	2.5	8.33±0.01	0.0177±0.0002
	5.3	7.73±0.00	0.0207±0.0002
	7.5	7.54±0.00	0.0214±0.0002
	9.8	7.18±0.00	0.0240±0.0002

which, as has already been mentioned, are slightly richer in Ni content than given by their nominal compositions of $Ni_{74}Al_{26}$ and $Ni_{74.25}Al_{25.75}$, respectively. deDood and deChatel[11] quote $S=21$ for $Ni_{74}Al_{26}$ and 116 for $Ni_{74.5}Al_{25.5}$. We, therfore, calculate $\Delta\gamma$ for $S=30$ and 100. While these numbers are arbitrarily chosen, hopefully they are close to the actual values of S for alloys I and II. We obtain (for $T_{SF}=18$ K) $\delta\gamma/\gamma(0)=-11.8\%$ and -29% in the maximum applied field of 9.8 T for alloys I and II, respectively. Thus the theoretically predicted magnitude of quenching is in reasonably good agreement with experiment.

Field dependence of the heat capacity of ferromagnetic alloy III

The heat-capacity response of alloy III in applied fields is significantly different from that of alloys I and II. The zero-field upturn is completely removed in 2.5 T and, also, the heat capacity is reduced in the entire temperature range 1.5–20 K. The heat capacity is further reduced in higher applied fields. If the complete removal of the upturn in alloy III in 2.5 T was due to the effect of an applied field on magnetic clusters, one is at a loss to understand why similar behavior is not observed in alloy I and II (the coefficient A' is of the same order of magnitude for all three alloys). We feel that this dissimilarity in the response to applied field reinforces the view that the upturn in these alloys is most likely due to spin fluctuations.

The magnitude of the spin-fluctuation temperature determines the field at which spin fluctuations (and the upturn) begin to get suppressed. Table II lists the values of fit parameters obtained by fitting the linear (C/T)-versus-T^2 plots in applied fields in the range 1.5–12 K to Eq. (1). We find that γ decreases by 19.8% relative to its zero-field value in a field of 9.8 T. The γ values of alloy III are plotted in Fig. 7.

It was mentioned in the beginning that alloy III orders magnetically and its spontaneous magnetization obtained from Arrott plots at 4.2 K is the same as reported for $Ni_{75}Al_{25}$ ($0.076\mu_B$/Ni), which is known to be a weak itinerant ferromagnet with a Curie temperature of 40 K. Earlier studies on another weak itinerant ferromagnet, Sc_3In ($T_c\approx 6$ K, $\mu_s=0.066\mu_B$/Sc), have shown that an applied field affects both the spin-fluctuation and magnetic contributions to the heat capacity.[36,37] The magnetic entropy of Sc_3In in a 10-T field, which completely quenches the spin fluctuations in this material with a low T_{SF} of 5 K, is reduced to 11–19 % of its zero-field value. The lowering of the heat capacity in applied fields of alloy III in the whole temperature range 1.5–20 K is thus consistent with earlier observations on Sc_3In.

The coefficient of the electronic heat capacity, γ, for weak itinerant ferromagnets with low exchange-splitting energies, is shown to decrease with applied fields even within the framework of Stoner model which does not take into account the spin fluctuations. The depression is calculated by Wohlfarth to be proportional to the applied field[38] H, i.e., $\Delta\gamma_H=-[\sigma(0,0)]^2H/\sigma(H,0)T_c^2$, where σ is the magnetization and T_c the Curie temperature. Using the magnetization values of Ni_3Al as reported in Ref. 4, where the magnetization was measured up to 7 T, Wohlfarth's expression gives, for $T_c=40$ K, $\Delta\gamma_H\sim -4\%$ in an applied field of 7.5 T, which is much smaller than the experimentally observed change of -15.6%. The much larger change seen experimentally is certainly due to the partial quenching of spin fluctuations. A comparison with band-structure calculations,[9] which give a γ of 2.56 mJ/g-at. K^2, shows that an appreciable enhancement of electronic heat capacity of the three alloys still persists at the maximum field used in the present investigation. It is likely that the application of fields exceeding 10 T would result in a further decrease of the electronic heat capacity due to quenching of spin fluctuations.

ACKNOWLEDGMENTS

The authors wish to thank F. X. Kayser for preparing the $Ni_{3-x}Al_{1+x}$ alloys used in this study. The Ames Laboratory is operated for the U.S. Department of Energy (D.O.E.) by Iowa State University under Contract No. W-7405-ENG-82. This research was supported by the Director of Energy Research, Office of Basic Energy Sciences, of the U.S. DOE.

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