Magnetostriction, Thermal Expansion, and Specific Heat of a Nearly Ferromagnetic Compound: Ni_3Ga†

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(Received 1 June 1970)

The properties of Ni_3Ga show considerable variability depending upon annealing conditions. We measured three samples and for the best (selected by the criteria of field-independent and low susceptibility, temperature-independent susceptibility at low temperature, and quadratic magnetostriction) we obtain from the magnetostriction a value, $\partial \ln \bar{I}/\partial \ln V = 1.4$, for the volume dependence of the product, $\bar{I} = IN_0$, of the exchange interaction and the bare density of states. The specific heat, measured in a 100-kOe field to suppress spin-fluctuation and superparamagnetic effects, and the low-temperature susceptibility, give a value $S = 30$ for the exchange-enhancement factor. The thermal expansion has an unusual temperature dependence in zero field and is drastically changed by applying a 90-kOe field. This measurement and the measurement of the specific heat in a large field were unsuccessful in distinguishing spin-fluctuation effects from the superparamagnetic effects which undoubtedly occur even in our best sample of Ni_3Ga.

I. INTRODUCTION

One expects by analogy with the behavior near the ferromagnetic phase transition that a paramagnetic metal having exchange interactions nearly strong enough to produce ferromagnetism at zero temperature will exhibit anomalous properties at low temperatures. The ferromagnetic system retains appreciable short-range order above its Curie temperature and one expects the nearly ferromagnetic system likewise to develop short-range order at low temperatures. In both cases, critical-state spin fluctuations (long-wavelength low-frequency damped spin waves) enhance not only the magnetic susceptibility, but also the specific heat. In a paramagnet, the low-temperature specific heat linear in temperature is enhanced because of virtual scattering of electrons on the Fermi surface by spin fluctuations,[1,2] which corresponds to an enhancement of the effective mass of the electrons. Doniach and Engelsberg[2] showed that, because of the variation of the electron self-energy as one moves away from the Fermi surface with increasing temperature T, the specific heat has an additional term of the form $T^3 \ln(T/T_s)$, where T_s is the characteristic temperature of the spin fluctuations.

The low-temperature thermal expansion should also be enhanced by spin fluctuations because of the volume dependence $\partial \ln \bar{I}/\partial \ln V$ of $\bar{I}$, which is the product IN_0 of the exchange interaction and the bare density of states. When the exchange-enhancement factor S is large (defined as the ratio with respect to N_0 of the density of states calculated from the low-temperature paramagnetic susceptibility), one expects the thermal-expansion enhancement for $\partial \ln \bar{I}/\partial \ln V$ of order unity to be large also. It should be more pronounced than the specific-heat enhancement approximately in the ratio $S\bar{I}(\partial \ln \bar{I}/\partial \ln V)/(\partial \ln N_0/\partial \ln V)$. In Sec. II we derive this expression for a uniform exchange-enhancement model, and also show that $\partial \ln \bar{I}/\partial \ln V$ can be evaluated from S and the magnetostriction. The same ratio expresses the magnitude of the thermal-expansion anomaly relative to the low-temperature specific-heat anomaly, both of which are of the form $T^3 \ln(T/T_s)$.

The predicted enhancement of the specific heat was observed in paramagnetic NiRh[3] and *Pd*Ni[4] alloys. The enhancement increases as the concentration of Ni approaches the critical concentration for ferromagnetism, and the NiRh alloys also develop a low-temperature anomaly similar to the predicted $T^3 \ln(T/T_s)$ term. We described previously measurements of the magnetostriction and thermal expansion of NiRh[5] and *Pd*Ni[6] alloys, which are roughly in accordance with these theoretical predictions, though the thermal expansion of NiRh alloys appears to be very sensitive to annealing.[7]

Hahn and Wohlfarth[8] have proposed that the low-temperature anomaly in the specific heat (and there-

fore also in the thermal expansion) of paramagnetic NiRh alloys is caused by the superparamagnetic behavior of Ni-rich ferromagnetic clusters rather than by spin fluctuations. The sensitivity of the thermal expansion to annealing indicates that inhomogeneity is indeed an important factor in the NiRh alloys, though the magnetization and magnetostriction are quite reproducible after annealing.[7]

The compound Ni_3Ga is a strongly exchange-enhanced paramagnet at the stoichiometric composition.[9] The slight excess of Ni in $Ni_{76}Ga_{24}$, or a trace of Fe impurity, gives rise to ferromagnetic ordering.[10] With suitable annealing Ni_3Ga can be prepared with a high degree of atomic order in the face-centered-cubic Cu_3Au structure. van Laar and Loopstra[11] performed a neutron study on filings of the stoichiometric compound annealed at 700 °C and found the order to be $99.2 \pm 0.6\%$. This encourages the belief that Ni_3Ga is a system in which one might observe spin-fluctuation effects without the complications due to the superparamagnetism associated with inhomogeneities.

We present in Sec. III the experimental technique and in Sec. IV the results of our measurements of the magnetostriction, thermal expansion, and temperature dependence of the magnetic susceptibility of Ni_3Ga. We found that the low-temperature susceptibility of three samples of Ni_3Ga varied about the average by about $\pm 35\%$, and the magnetostriction of the one having the highest susceptibility was much higher than that of the other two samples. The sample having the lowest susceptibility had a magnetization accurately linear in field H up to $H \sim 14$ kOe and independent of temperature T below $T \sim 4$ °K. Its magnetostriction was accurately quadratic in field up to $H \sim 37$ kOe, unlike the higher-susceptibility samples which showed larger magnetostriction at low fields suggestive of superparamagnetism.

This behavior of the lowest-susceptibility sample encouraged the hope that it would not exhibit superparamagnetism in its thermal properties. A very pronounced low-temperature anomaly in the thermal expansion, and a small but clearly evident specific-heat anomaly in the same temperature range, were thought to signal the spin-fluctuation effects predicted for a strongly exchange-enhanced paramagnet.

When the thermal expansion was measured in a large field, $H = 90$ kOe, the low-temperature anomaly disappeared. The specific-heat anomaly was also reduced, but not completely removed, by the application of a large field. This suppression by a field of the low-temperature anomalies in the specific heat, and more clearly in the thermal expansion, suggests that their origin is in the superparamagnetic behavior of locally ferromagnetic Ni-rich clusters. Hahn and Wohlfarth[8] proposed a model which describes the low-temperature specific-heat anomaly of a system containing ferromagnetic particles by assuming them to have uniaxial magnetic anisotropy, and pointed out that for this model a magnetic field would decrease the superparamagnetic contribution to the specific heat. Brinkman and Engelsberg[12] showed that a large field would suppress the contribution of spin fluctuations to the specific heat (and therefore also to the thermal expansion), but one would not expect so large an effect as we observe for the parameters appropriate to Ni_3Ga. It seems therefore that even our best sample of Ni_3Ga did not provide the hoped-for unambiguous evidence for spin-fluctuation effects in the thermal properties.

II. SPIN-FLUCTUATION CONTRIBUTIONS TO THERMAL EXPANSION

We now proceed to derive the effects of spin fluctuations on the thermal expansion for a uniform exchange-enhancement mode, following Brinkman and Engelsberg's[12] discussion of the specific heat. The magnetic susceptibility for this model may be written, as in the Stoner model,

$$\chi = 2\mu_B^2 N_0/(1-\bar{I}) = 2S\mu_B^2 N_0 \tag{1}$$

where $S = 1/(1-\bar{I})$ is the exchange-enhancement factor. The mass enhancement is

$$m/m_0 = 1 + A \ln S, \tag{2}$$

the coefficient A being much less than unity because of the finite range of the interaction,[13] which strongly reduces the large mass enhancement predicted by the zero-range model. The electronic specific heat including the term which arises from variation of the electron self-energy away from the Fermi surface is

$$C_v/C_v^0 = m/m_0 + BS(T/T_s)^2 \ln(T/T_s), \tag{3}$$

where $C_v^0 = \frac{2}{3}\pi^2 N_0 k^2 T$, B is a numerical coefficient, and

$$T_s = 4T_F/\pi S, \tag{4}$$

T_F being the Fermi temperature.

As the temperature approaches zero, the second term in Eq. (3) vanishes, and to determine the expected behavior of the thermal expansion we differentiate the first term to obtain the electronic Grüneisen parameter

$$\gamma \equiv \frac{\partial \ln C_v}{\partial \ln V} = \frac{\partial \ln N_0}{\partial \ln V} + \frac{m_0}{m} A \frac{\partial \ln S}{\partial \ln V}. \tag{5}$$

We write Eq. (5) in the form

$$\Delta\gamma = \gamma - \gamma_0 = A \frac{m_0}{m} \frac{\partial \ln S}{\partial \ln V} = A \frac{m_0}{m} S\bar{I} \frac{\partial \ln \bar{I}}{\partial \ln V}, \tag{6}$$

where γ_0 is the Grüneisen parameter without spin

fluctuations. Thus for a nonzero value of $\partial \ln \bar{I}/\partial \ln V$, the zero-temperature Grüneisen parameter is enhanced by spin fluctuations. From the definition,

$$\gamma = \beta/\kappa C_v, \tag{7}$$

where β is the volume coefficient of thermal expansion and κ the compressibility, we obtain the ratio of the shift in the low-temperature value of β to the shift in the low-temperature value of C_v,

$$\frac{\Delta\beta/\beta_0}{\Delta C_v/C_v^0} = 1 + \frac{1}{\gamma_0 \ln S}\frac{\partial \ln S}{\partial \ln V}. \tag{8}$$

We can determine $\partial \ln S/\partial \ln V [= S\bar{I}(\partial \ln \bar{I}/\partial \ln V) \equiv (S-1) \times (\partial \ln \bar{I}/\partial \ln V)]$ from the magnetostriction, using a thermodynamic relation corresponding to the isothermal, isobaric conditions,

$$\frac{1}{V}\frac{\partial V}{\partial H} = \kappa\chi\frac{\partial \ln\chi}{\partial \ln V}H, \tag{9}$$

which with Eq. (1) gives,

$$\frac{\partial \ln\chi}{\partial \ln V} = \gamma_0 + \frac{\partial \ln S}{\partial \ln V}. \tag{10}$$

As temperature increases, the second term in Eq. (3) causes the specific heat to decrease rapidly. The corresponding low-temperature anomaly $\delta\beta$ in the thermal-expansion coefficient is obtained by taking the volume derivative of the entropy. Neglecting the derivative of the slowly varying logarithmic term, we obtain

$$\frac{\delta\beta/\beta_0}{\delta C_v/C_v^0} = \frac{1}{\gamma_0}\frac{\partial \ln S}{\partial \ln V}, \tag{11}$$

which is rather similar to the expression for the relative enhancements of the Grüneisen γ and the specific heat, apart from the relatively small $\ln S$ factor in the denominator for Eq. (8).

III. EXPERIMENTAL TECHNIQUE

The thermal expansion and longitudinal magnetostriction of the Ni_3Ga samples were measured by a capacitance method. The capacitance dilatometer is described in Ref. 14. Its thermal expansion was calibrated against intrinsic Si, whose thermal expansion was measured absolutely by Sparks and Swenson.[15] The thermal expansion coefficient at each temperature was obtained by computing a least-squares fit of the function $(a_0 + a_1T^2 + a_2T^4)$ to the set of five data points centered on that temperature, and then differentiating this function.

The magnetic susceptibility as a function of field and the magnetization in fixed field as a function of temperature were measured with a pendulum magnetometer. The specific heat was measured by the heat pulse method.[16]

The Ni_3Ga samples were made by melting together the stoichiometric quantities of high-purity Ni and Ga in an alumina container *in vacuo* and cooling slowly in a Bridgman furnace. The samples were annealed *in vacuo* for several days with the results shown in Table I. The high value of the residual resistivity ratio for sample $Ni_3Ga\,C$ indicates that this is the most well-ordered sample, but the lower value and smaller temperature dependence of the low-temperature susceptibility for $Ni_3Ga\,B$ suggests this sample to be the least affected by superparamagnetism. The field dependence of the magnetostriction supports the conclusion that $Ni_3Ga\,B$ is the best sample for exploring spin-fluctuation effects, with the least likelihood of their being masked by superparamagnetic effects. We describe in Sec. IV the thermal expansion and specific-heat data for this sample in zero field and in a large field.

TABLE I. Annealing and properties of Ni_3Ga samples.

Sample	Anneal	RRR[a]	χ_g[b] (10^{-6} emu)
$Ni_3Ga\,A$	3 days at 1000 °C	11	110
$Ni_3Ga\,B$	3 days at 1000 °C		
	4 days at 950 °C	28	57
$Ni_3Ga\,C$	3 days at 1000 °C		
	4 days at 950 °C	44	
	7 days at 1000 °C	42	73

[a]RRR is the residual resistivity ratio between room temperature and 4.2 °K.

[b]χ_g is the susceptibility per g as temperature tends to zero.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

The susceptibility of the three Ni_3Ga samples as a function of temperature is shown in Fig. 1. The susceptibility of $Ni_3Ga\,B$ is considerably lower than that of the other two samples, especially at low temperature where for this sample alone the susceptibility is independent of temperature below $T \sim 4$ °K. The value $\chi_g(0) = 56.7 \times 10^{-6}$ emu of the susceptibility extrapolated to zero temperature is considerably lower than the value $\chi_g(0) = 79.4 \times 10^{-6}$ emu reported by de Boer.[10,17] This may indicate that our sample $Ni_3Ga\,B$ is nonstoichiometric, since $\chi_g(0)$ drops rapidly as the Ni concentration decreases, to the value $\chi_g(0) = 35 \times 10^{-6}$ emu for $Ni_{74.5}Ga_{25.5}$.[17] However, the high value of the residual resistivity ratio for both $Ni_3Ga\,B$ and $Ni_3Ga\,C$ given in Table I suggests this not to be the case, and the low value of $\chi_g(0)$ may be due to better ordering than de Boer's sample.[18] The temperature dependence of the susceptibility for $Ni_3Ga\,B$ is qualitatively similar to that reported by de Boer.[17]

The susceptibility of samples $Ni_3Ga\,A$ and $Ni_3Ga\,C$ is even higher at low field, but decreases with increasing field and becomes field independent above

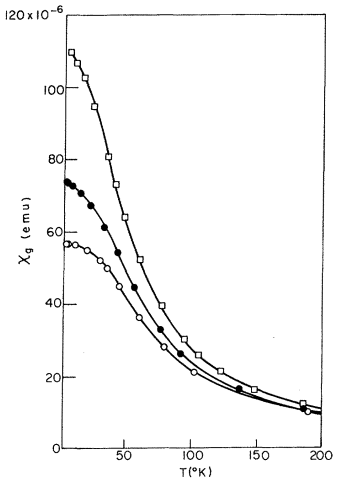

FIG. 1. Susceptibility of Ni_3Ga as a function of temperature T in a field $H = 14.24$ kOe. □ $Ni_3Ga\,A$; ○ $Ni_3Ga\,B$; ● $Ni_3Ga\,C$.

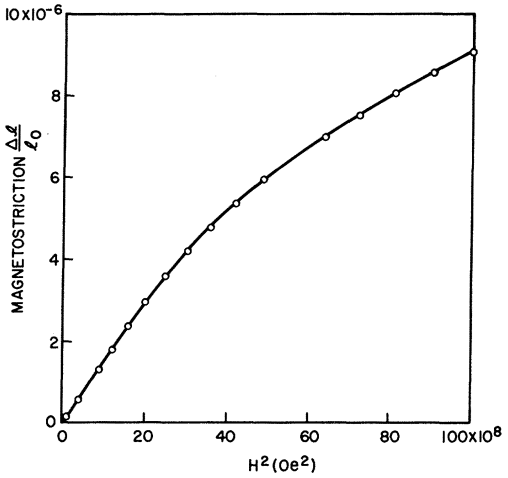

FIG. 3. Magnetostriction of sample $Ni_3Ga\,B$ up to a field $H = 100$ kOe at 4.2 °K.

about 4 kOe. For a field-independent susceptibility the magnetostriction should be quadratic in field, as may be seen by integrating Eq. (9). We see in Fig. 2 that for $Ni_3Ga\,A$ and $Ni_3Ga\,C$ the magnetostriction becomes quadratic in the field only above about 20 kOe. Furthermore, $Ni_3Ga\,A$ has a magnetostriction about six times greater than that of the other two samples, which we attribute to ferromagnetism or superparamagnetism associated with Ni-rich regions in this sample. By contrast, $Ni_3Ga\,B$ has a field-independent susceptibility up to $H \sim 14$ kOe, and as shown in Fig. 2 its magnetostriction is quadratic in field up to $H \sim 37$ kOe. These results indicate that $Ni_3Ga\,B$ is the best candidate for an experimental study of the thermal properties to look for spin-fluctuation effects, and the remainder of the paper will be devoted to measurements on this sample.

The magnetostriction of $Ni_3Ga\,B$ deviates from a quadratic field dependence at higher fields, as shown in Fig. 3. This behavior is consistent with the field dependence of the susceptibility at higher fields found by de Boer *et al.*,[9,10] and is qualitatively in accordance with the Stoner-Wohlfarth theory for a nearly ferromagnetic metal.[19,20] When the Belov equation[21] is modified to describe the relation between the magnetostriction ω and the magnetization M of a strong paramagnet,[22] one obtains a quadratic relationship $\omega \sim M^2$ similar to that predicted for a weak itinerant ferromagnet.[20] We were unable to check this relationship since our magnetization measurements only extend to a low field, $H \sim 14$ kOe, and it is clearly unsatisfactory to use this magnetization data of de Boer *et al.*[9,10] obtained for another sample.

The specific heat of $Ni_3Ga\,B$ shown in Fig. 4 has

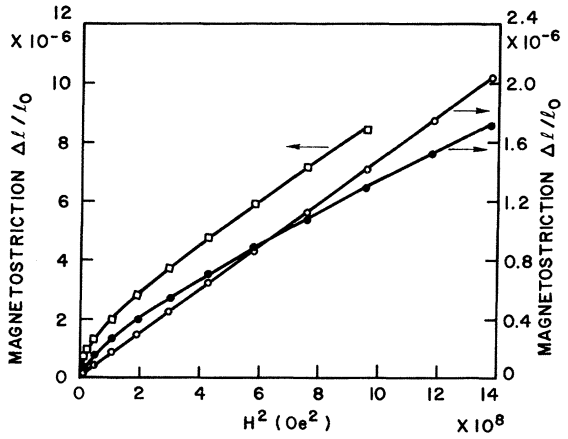

FIG. 2. Magnetostriction of Ni_3Ga up to a field $H = 37$ kOe at 4.2 °K. □ $Ni_3Ga\,A$; ○ $Ni_3Ga\,B$; ● $Ni_3Ga\,C$.

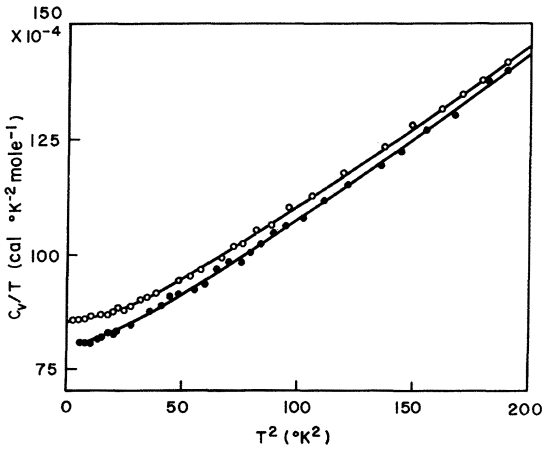

FIG. 4. Specific heat C_v of sample $Ni_3Ga\,B$ up to a temperature $T \sim 14$ °K. ○ – field $H = 0$; ● – field $H = 100$ kOe.

a low-temperature anomaly beginning between 7 and 10 °K. The application of a field of 100 kOe decreases the specific heat over the whole temperature range and diminishes but does not suppress the low-temperature anomaly. Putting aside for the present the question of the origin of this field-dependent low-temperature anomaly, we might assume that extrapolation of the high-field curve to zero temperature would give a more reliable value, $(C_v T)_0 = 80 \times 10^{-4}$ cal °K^{-2} mole^{-1} for the specific heat in the absence of spin-fluctuation or superparamagnetic effects, than extrapolation of the zero-field curve. This value gives a density of states 4.8(eV)$^{-1}$mole^{-1}, and if we take this to be the value of the bare density of states N_0, we can evaluate from the exchange-enhanced susceptibility 57×10^{-6} emu g^{-1} (see Table I) an exchange-enhancement factor, $S = 30$.

The thermal expansion coefficient of Ni_3Ga *B* divided by the temperature, which is shown in Fig. 5, has a low-temperature anomaly starting at $T \simeq 25$ °K, with a sharp upturn with decreasing temperature at $T \simeq 6$ °K. The application of a field of 90 kOe drastically changes the thermal expansion. The measurements were taken only to 20 °K since it was difficult to calibrate the germanium resistance thermometer in a field above the boiling point of liquid hydrogen.[23] The curve for the thermal expansion in a field appears to extrapolate roughly as shown by the dashed curve in Fig. 5 to approach the zero-field curve at about 25 °K. At low temperature the thermal expansion decreases rapidly in contrast to the behavior in zero field.

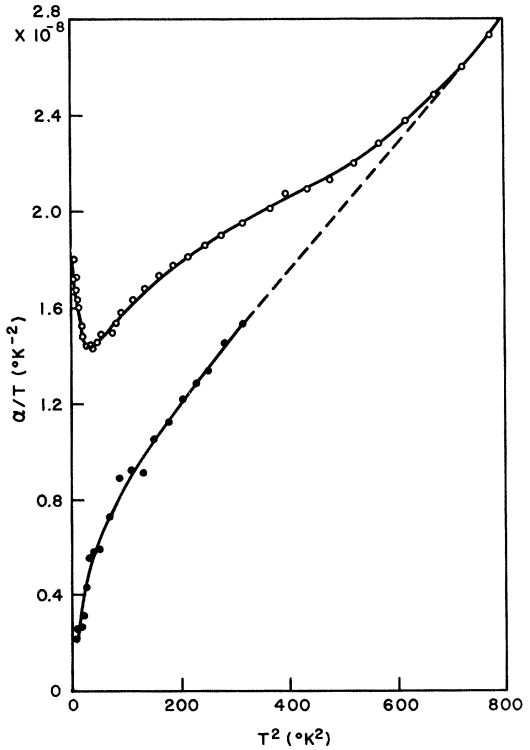

FIG. 5. Linear thermal expansion coefficient α of sample Ni_3Ga *B* up to a temperature $T \sim 28$ °K. ○ – field $H = 0$; ● – field $H = 90$ kOe.

We consider first the zero-field curve. The characteristic temperature T_s of spin fluctuations in Ni_3Ga can be evaluated by the use of Eq. (4). We use de Boer's[17] estimate of the degeneracy temperature $T_F = 375$ °K, and our estimate of the exchange-enhancement factor $S = 30$, and obtain a value $T_s = 16$ °K. The sharp upturn in the zero-field curve of Fig. 5 occurs at a temperature much higher than one would expect for spin fluctuations having this characteristic temperature. For comparison, the low-temperature anomaly in the specific heat of $Ni_{0.63}Rh_{0.37}$, which was attributed by Bucher *et al.*[24] to spin fluctuations, occurs at about 5 °K though their estimate of ~ 250 °K for T_s is much higher than in Ni_3Ga. The anomalously high thermal expansion extending to about 25 °K is clearly not due to spin fluctuations. Hahn and Wohlfarth[8] showed that superparamagnetic behavior resulting from local ferromagnetic regions with magnetic anisotropy E_A gives a roughly constant contribution to the specific heat up to temperature $T_A \simeq E_A/k_B$. If we make the reasonable assumption that the volume dependence of E_A is negative, this would result also in a positive contribution to the thermal expansion. Hahn and Wohlfarth showed also that the application of a field should remove this anomalous specific-heat contribution, and therefore also the thermal expansion anomaly, up to a temperature of about $2\mu_B H/k$. A field of 90 kOe corresponds to an equivalent temperature of only 12 °K, so that it is surprising that the thermal expansion is strongly affected to as high a temperature as 25 °K.

Brinkman and Engelsberg[12] showed that a field having an equivalent temperature of the order of T_s should suppress the mass enhancement associated with spin fluctuations. In Fig. 5 the low-temperature thermal expansion anomaly starting at about 6 °K in zero field appears to be reversed in sign in a high field. This behavior is difficult to understand if we attribute the anomaly either to spin fluctuations or to superparamagnetism resulting from inhomogeneities.

Finally, we can estimate $\partial \ln \bar{I}/\partial \ln V$ from the magnetostriction. We estimate the value $\partial \ln \chi/\partial \ln V = 42$ by substituting in Eq. (9) the volume magnetostriction of Ni_3Ga *B*, $(1/V)\partial V/\partial H = 1.00 \times 10^{-14} H$ Oe^{-1}, obtained from Fig. 2 with the assumption that the magnetostriction is isotropic, the magnetic susceptibility at zero temperature, $\chi = 5.1 \times 10^{-4}$ emu cm^{-3}, obtained from Table I, and the compressibility[25] $\kappa = 4.65 \times 10^{-13}$ cgs. A very rough estimate of γ_0 for substitution in Eq. (10) is obtained by taking the low-

est-temperature value of α/T from the high-field curve in Fig. 5, $(\alpha/T)_0 = 0.2\times10^{-8}$ (°K)$^{-2}$, and the high-field low-temperature value $(C_v/T)_0 = 85\times10^{-4}$ (cal °K^2 mole^{-1}) from Fig. 4 which gives $\gamma_0 = 1.0$, and therefore $\partial \ln S/\partial \ln V = 41$. The resultant value $\partial \ln T/\partial \ln V = 1.4$ is of the same sign and approximately equal to the value 1.0 observed in NiRh,[7] but opposite in sign to the value -2.0 for Pd.[6]

We are unable to draw any definite conclusions from the behavior of the thermal properties of Ni_3Ga. The thermal expansion in zero field suggests that both superparamagnetic and spin-fluctuation effects are present, but the application of a large field does not appear to help to distinguish between them. We expect from the theory of spin fluctuations that the thermal expansion will show their effects much more clearly than the specific heat for reasonable values of the volume dependence of the exchange interaction, and our results indicate that the same is true for the superparamagnetic effects.

ACKNOWLEDGMENT

We are indebted to T. R. Kyle for technical assistance.

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†Some parts of this paper will be published also in the Proceedings of the 1968 Symposium on Thermal Expansion of Solids.

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[25]We are indebted to L. R. Testardi for the measurement of the compressibility.