Low-temperature calorimetric investigation of Co-Ga spin-glass

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Low-temperature specific-heat measurements are presented for the spin-glass alloys Co_xGa_{1-x} with x between 0.49 and 0.58. For these compositions the antistructure defects are the principal magnetic entities with concentrations in the range 0.9—8.0%. For the high-concentration samples, nuclear Schottky and spin-wave contributions to the specific heat have been identified. Over the entire composition range a term with a linear temperature dependence has been found and is identified with the spin-glass state. Measurements have been taken in magnetic field from 0 to 2.8 T. These data can be understood in terms of a model with a distribution of two-level systems.

I. INTRODUCTION

The intermetallic compound Co_xGa_{1-x} crystallizes in the cubic CsCl structure when x, the cobalt concentration, has a value between 0.45 and 0.69. In this structure the cobalt ions occupy one simple cubic lattice and the gallium ions occupy another, displaced from the first by one-half the body diagonal. For perfect stoichiometry, $x=0.50$, each ion is surrounded by eight of the opposite type. In this configuration, electrons are transferred from the gallium ions to the d band of the cobalt ions which then become magnetically neutral. Departure from stoichiometry by increasing the cobalt concentration leads to substitutional cobalt ions on the gallium sublattice. These substitutional cobalt ions are surrounded by other cobalt ions and form complexes called antistructure defects (AS defects) which are magnetic with spin $\frac{1}{2}$ and a g value of 5.[1] The AS defects are the essential source of the magnetic properties of this system, and the defect-free alloy is believed to be a nonmagnetic host.[1–4]

Although the concentration of the AS defects, n, is approximately equal to the excess of cobalt ions beyond $x=0.50$, statistical fluctuations and sample preparation will produce additional AS defects. Hence even for cobalt-poor samples down to $x\sim0.49$, AS defects may be present. The importance of sample preparation on the magnetic properties of Co-Ga has been discussed by Meisel *et al.*[4]

Tamminga and de Dood[1] investigated the magnetization and the specific heat of the Co-Ga system down to low temperature. However, their work was performed in the era before spin-glass systems were identified. Meisel *et al.* addressed this problem and measured the low-field ac susceptibility. They discovered a spin-glass regime at low temperature in the composition range $0.50\lesssim x\lesssim0.56$ and identified ferromagnetic transitions for $x>0.56$ with possible antiferromagnetic transitions for $x\lesssim0.49$. The essential magnetic picture is the following:[4,5] The AS defects are randomly distributed in samples prepared by slow, furnace-cooling techniques. At room temperatures, the system is paramagnetic. As the temperature is decreased, the AS defects interact through competing ferromagnetic and antiferromagnetic interactions. The interactions are predominantly ferromagnetic for $x\gtrsim0.51$ and antiferromagnetic for $x<0.51$. For $x>0.56$, ferromagnetic transitions occur; for instance, $T_c=109$ K for $x=0.58$. As the temperature is decreased, the interaction range continues to grow until a spin-glass transition occurs at T_f. This has been discussed previously in terms of cluster growth.[4] T_f is observed to be proportional to the AS defect concentration for $0.50<x<0.54$. Deviations from this behavior for $x\gtrsim0.55$ can be attributed to the dominance of ferromagnetic interactions over competing interactions that are antiferromagnetic. Equilibrium properties of the susceptibility in the spin-glass regime have been studied by Zhou *et al.*[6] in particular, the susceptibility for a wide range of frequencies smoothly approaches zero with decreasing temperature and follows a universal curve independent of AS defect concentration. A universal curve for different magnetic spin concentrations has been reported for other spin-glass systems.[4,7]

In this paper we present measurements of the specific heat of the Co-Ga spin-glasses. Earlier work by Tamminga[1] has shown a substantial effect of sample preparation on the specific heat. Consequently, in order to compare with the magnetic effects reported by Meisel *et al.*,[4] we used their samples.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

Pure (5N) cobalt and gallium were arc-melted in an argon atmosphere, annealed at 950 °C and 48 h under a vacuum of 2×10^{-5} mm Hg and then furnaced-cooled in one

day. The heat-capacity measurements were made between 0.4 and 50 K using a standard heat-pulse technique in a conventional ^{3}He cryostat which has been described elsewhere.[8] Magnetic fields up to 2.8 T were applied at 4.2 K, and measurements started after the samples were cooled in the magnetic field to the lowest temperature. To check for history dependence, one sample, $x=0.51$, was cooled in 2.8 T and then demagnetized at low temperatures. The results were identical to zero-field-cooled measurements. The addenda contribution, as a percentage of the total measured heat capactiy, was of the order of 0.5 at 1 K, 1 at 4 K, and 17 at 30 K.

III. RESULTS AND DISCUSSION

An overview of the specific heat C measured in zero magnetic field for all samples is presented in Fig. 1 in the form C/T vs T^2. These new results extend to lower and higher temperatures than those of Tamminga and de Dood[1] and agree with theirs when samples with the same AS defect concentration are compared. The result of Tamminga and de Dood for $x=0.48$ is represented by a straight line in Fig. 1. This sample has a negligible concentration of AS defects, and its specific heat can be represented by the sum of electronic and lattice terms,

$$C/T=\gamma+\alpha T^2 , \tag{1}$$

with $\gamma=2.0$ mJ/g-at.K^2 and $\alpha=0.025$ mJ/g-at.K^4. Several features are apparent in Fig. 1. Firstly, at high temperature, the slope of C/T for our samples is not dramatically different from the slope of the $x=0.48$ sample. This indicates that the lattice contribution changes only slightly when AS defects are introduced. Secondly, all the curves appear to converge to the same low-temperature value of C/T. Finally, when attempting to draw straight lines through the higher-temperature data we noted that around $T^2=250$ K^2 all curves show a bump with a magnitude of about 10% of the total specific heat. Using the same equipment and thermometers, we have measured other systems and never observed such an anomaly. The bump is also present in the only sample that Tamminga[1] measured in this temperature range. This anomalous feature is independent of AS defect concentration and magnetic fields. In addition, susceptibility measurements on the same samples do not show any structure near 16 K. Therefore, this anomaly is probably not of magnetic origin and will be ignored in the following discussion.

To infer the effects of AS defects on the specific heat, the electronic and lattice contributions must be subtracted. For the electronic term we use the result of the lowest-cobalt-concentration sample measured by Tamminga and de Dood,[1] $x=0.48$, and assume that γ is independent of the AS defect concentration. Recently, Whittle *et al.*[9] calculated the electronic band structure for the equiatomic $Co_{0.50}Ga_{0.50}$ alloy assuming a perfectly ordered lattice. It is reasonable to neglect changes in the band structure by AS defects and assume that the most important contribution to the electronic structure by these defects is to add electrons to the alloy. We estimate for our range of AS defect concentrations that the coefficient of the electronic contribution will vary, at the most, from 1 to 4 mJ/g-at.K^2. To obtain the lattice contribution we fit our data in Fig. 1 by straight lines for $T^2>200$ K^2. When the anomalous bumps at 16 K are ignored, the resultant lattice heat capacity, cubic in temperature, was found to be

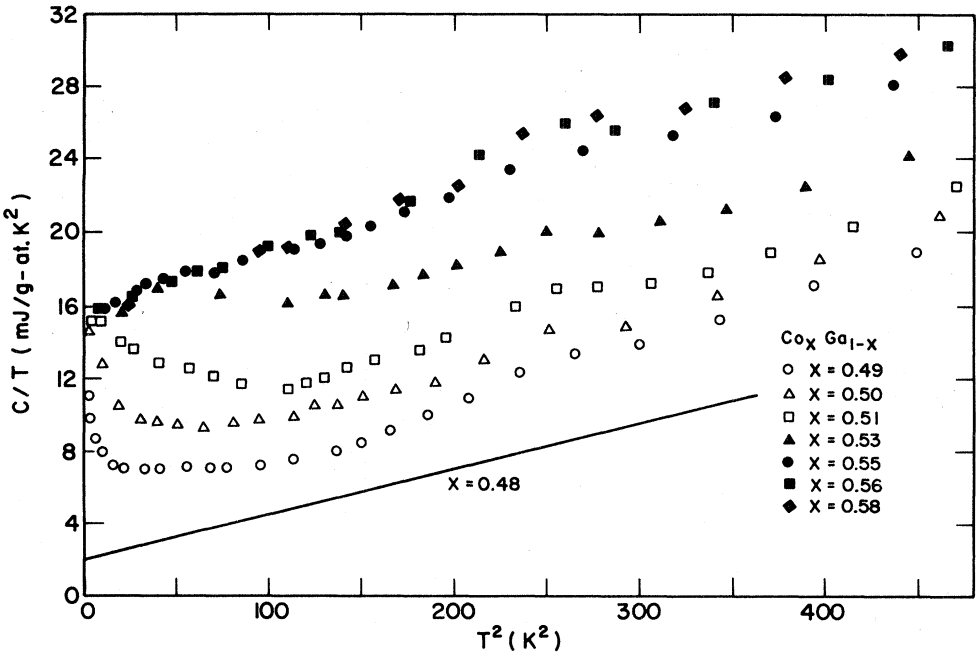

FIG. 1. Overview of the specific heat measured in zero magnetic field is shown as C/T vs T^2 for Co_xGa_{1-x}. The low concentration result of Tamminga and de Dood (Ref. 1) is represented by the straight line.

about 10—20% higher than that of the $x=0.48$ reference sample. This procedure, however, may not be reliable since the fitting extends to temperatures where higher-order terms of the lattice heat capacity may be significant. Because of these uncertainties we will focus our attention mainly on the region below $T^2=50$ K^2, where the electronic and the lattice contributions play a minor role. In this region the specific heat, C_0, of the $x=0.48$ sample may be taken as representative to these two contributions. Then the excess specific heat above the straight line in Fig. 1 is the contribution from the AS defects. This excess specific heat is shown in Figs. 2 and 3 as $(C-C_0)/T$ vs T. The specific heat has been measured in four different fields, but for clarity only the extreme fields of 0 and 2.8 T are presented. The excess specific heat of the higher-cobalt-concentration samples $x=0.53$, 0.55, and 0.56 in zero fields are the same within 2% between 1 and 6 K. The excess specific heat of the 0.58 sample is 5% lower in this range. The influence of a magnetic field on these high-concentration samples is small, less than 2% for the 0.56 sample and less than 6% for the other three samples.

The Co-Ga samples with AS defect concentrations up to 7% exhibit spin-glass behavior in the temperature dependence of the magnetic susceptibility.[4] Experimentally, most spin-glasses have a linear, or nearly linear, excess contribution to the specific heat below the freeezing temperature T_f, where a sharp peak in the ac susceptibility is observed. In our work on the Co-Ga alloys, this behavior is also observed: $x=0.50$ and 0.51; see Figs. 2 and 3. An exception to this general behavior for spin-glasses appears to be the amorphous Gd-Al alloy for which Coey *et al.*[10] found a $T^{3/2}$ contribution. For samples with $x\geq 0.53$, the magnetic AS defects give rise to two additional contributions to the specific heat. These specimens show a nuclear

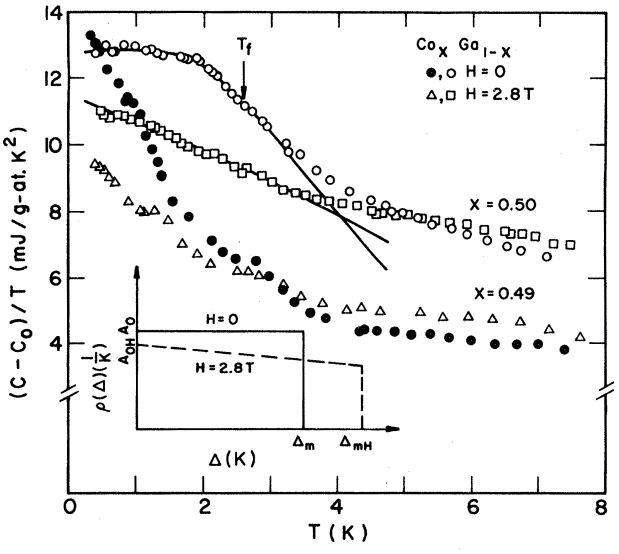

FIG. 2. Excess specific heat above the lattice and electronic contributions is shown as $(C-C_0)/T$ vs T for $x=0.49$ and 0.50 in zero field and in a magnetic field of 2.8 T. The solid lines are calculated using a model discussed in the text. The values of the fitting parameters are given in Table II.

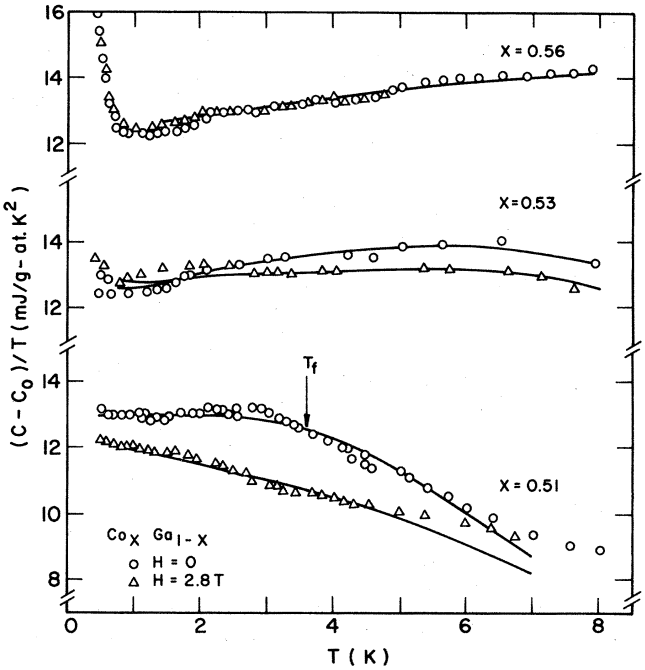

FIG. 3. Excess specific heat above the lattice and electronic contributions is shown as $(C-C_0)/T$ vs T for $x=0.51$, 0.53, and 0.56 in zero field and in a magnetic field of 2.8 T. The solid lines are calculated using a model discussed in the text. The values of the fitting parameters are given in Table II. For $x=0.56$ only the calculated curve for 0 field is shown as the results in the magnetic field are essentially identical.

contribution below 1 K and a small $T^{3/2}$ contribution above 1 K, Fig. 4, in addition to the linear spin-glass term. Consequently, we can express the measured heat capacity in the form

$$C=C_0+C_{sw}+C_N+C_{sg}\ ,$$

where $C_0=\gamma T+\alpha T^3$ is attributable to the electrons and the lattice $C_{sw}=\beta T^{3/2}$ is from spin waves, $\beta\neq 0$ for $x\geq 0.53$, $C_N=AT^{-2}$ is the nuclear Schottky contribution, $A\neq 0$ for $x\geq 0.53$, and C_{sg} is the spin-glass specific heat which we find to be of the form a_0T at very low temperature in zero magnetic field for the samples with $x=0.50$ and 0.51, Figs. 2 and 3. It will be assumed that this proportionality with T also holds for the higher concentration samples. We will discuss these contributions separately and indicate how they can be independently determined.

The specific heat below 1 K of samples with strong ferromagnetic interactions, $x\geq 0.53$, shows the tail of a nuclear Schottky contribution, Fig. 3, $x=0.53$ and 0.56. This contribution, C_N, can be fit to a T^{-2} temperature dependence allowing a determination of the coefficient A. For these higher Co concentration samples, the separation of contributions with a T^{-2} and $T^{3/2}$ temperature dependence is easily performed since the former is only important below 1 K. Examples of such analysis are shown in Fig. 4. In Fig. 4(a) the nuclear Schottky term is displayed through the slope of a graph of $(C-C_0-C_{sw})/T$ vs T^{-3} for the sample $x=0.58$. From the slope we determine the coefficient A and deduce the effective hyperfine field. In

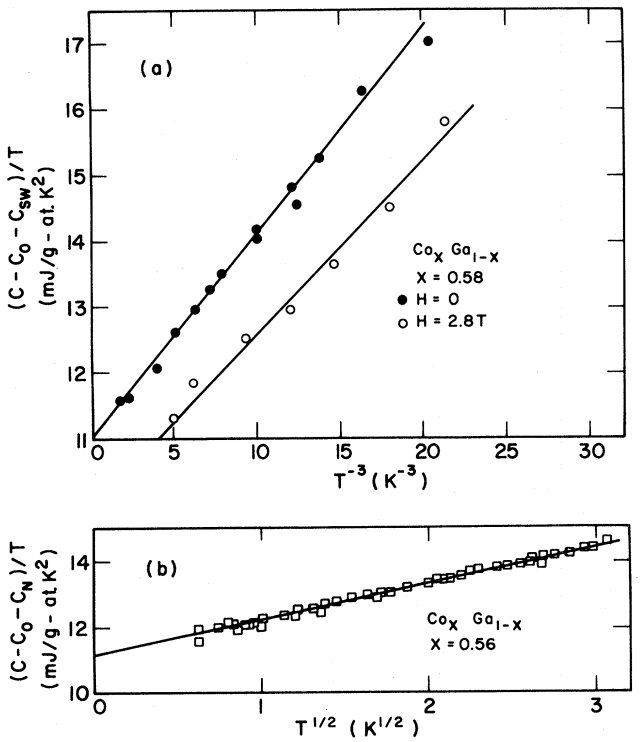

FIG. 4. (a) Excess specific heat above the lattice, electronic, and spin-wave contribution $(C-C_0-C_{sw})$ is shown for $x=0.58$ in $H=0$ and 2.8 T. The slope of the straight line gives the coefficient of the nuclear contribution A. (b) Excess specific heat above the lattice, electronic, and nuclear contribution $(C-C_0-C_N)$ is shown for $x=0.56$. The slope of the straight line gives the coefficient of the spin-wave contribution β.

Fig. 4(b) a plot of $(C-C_0-C_N)/T$ vs $T^{1/2}$ for the case of $x=0.56$ is shown and demonstrates the existence of a $T^{3/2}$ contribution to the heat capacity which we have determined from the slope β of this graph. In the following we will discuss these two contributions in more detail.

For a nucleus with a nuclear spin I ($I=\frac{7}{2}$ for cobalt) the coefficient A is given by

$$A=nN_0(\mu_N H_N)^2\frac{I(I+1)}{3k_B}=n(1.016\times10^{-2})H_N^2 \quad (2)$$

(measured in mJ K/g-at. %), where μ_N is the nuclear Bohr magneton, H_N is the effective magnetic field (in tesla) acting on the nucleus, N_0 is Avogadro's number, and k_B is the Boltzmann constant. The effects have been studied in several magnetic fields, but only the $H_a=0$ and 2.8 T results are shown. The values of A and H_N that have been extracted from the data using Eq. (2) are given in Table I. It is interesting to note that the decrease in the effective field with increasing applied magnetic field indicates that the hyperfine field and the magnetization are in opposite directions. The hyperfine fields inferred from our specific-heat measurements are in good agreement with the values obtained for pure cobalt from specific-heat[11] and from resonance and nuclear orientation experiments.[12]

The dispersion relation for spin waves in zero applied field can be written $\hbar\omega=Dk^2$ in which $\hbar\omega$ is the energy of the spin wave, k is the wave number, and D is the magnetic stiffness parameter. From this relation follows the spin-wave specific heat:[13] $C_{sw}=\beta T^{3/2}=0.113V_m k_B(k_BT/D)^{3/2}$; V_m is the molar volume. The value of β and D for different concentrations are given in Table I. Thomson and Thompson[14] derived that β should scale as $1/\sqrt{n}$. For the three samples with the highest cobalt concentration, β obeys this relation. The estimate of β for the $x=0.53$ sample is less accurate since we were constrained to stay well below the freezing temperature 7.2 K. The specific-heat analysis shown in Figs. 4(a) and 4(b) clearly indicates the existence of a linear term in temperature as well. This term is associated with the spin-glass behavior, so our data indicate the coexistence of spin-glass and ferromagnetic ordering. Evidence for ferromagnetic clustering for $T\lesssim 2T_f$ was found by Meisel *et al.*[4] from susceptibility measurements in these cobalt-concentrated Co-Ga systems. We believe that the determination of a $T^{3/2}$ term in the heat capacity of these alloys indicates the existence of short-range ferromagnetic ordering inside clusters and is an independent demonstration of cluster formation. Very recently Murani[15] investigated Au—19 at. % Fe by neutron scattering and also concluded that there was coexistence of spin-glass and ferromagnetic ordering. The value of the magnetic stiffness parameter for his sample is 40% lower than what we find for Co-Ga. Suggestions for the coexistence of spin-glass and ferromagnetic behavior have also been advanced by Gabay and Toulouse[16] and by van Hemmen.[17]

TABLE I. Cobalt concentration is x; the AS defect concentrations n and n_S are obtained from susceptibility, Ref. 4, and entropy considerations. Terms in the specific heat proportional to T^{-2} and $T^{3/2}$ lead to A and β, respectively, and from these are obtained H_N, the effective magnetic fields acting on the AS defect nuclei and D, the magnetic stiffness parameter.

x	n[a]	n_S	A (mJ K/g-at.) $H_a=0$	A (mJ K/g-at.) $H_a=2.8$ T	$H_N(T)$ $H_a=0$	$H_N(T)$ $H_a=2.8$ T	β(mJ/g-at. $K^{5/2}$)	D (10^{-40} J m²)
0.49	0.009	0.0096						
0.50	0.015	0.0180						
0.51	0.020	0.0230						
0.53	0.035	0.0370	0.17	0.14	21.9	19.8	1.1	0.64
0.55	0.052		0.25	0.21	21.7	19.9	1.1	0.64
0.56	0.064		0.33	0.31	22.5	21.8	1.0	0.69
0.58	0.080		0.31	0.27	19.5	18.2	0.9	0.74

[a]Reference 4.

Experimentally, no clear evidence for the coexistence of a linear and a $T^{3/2}$ term in the specific heat has previously been found although Thomson and Thompson[14] reanalyzed data on the archetypal spin-glasses CuMn, AgMn, and AuMn and found the specific heat at very low temperatures, below $T_f/3$ to contain a $T^{3/2}$ term. For the insulating spin-glass $Eu_xSr_{1-x}S$, Scherzberg *et al.*[18] found in the concentration range where neutron scattering shows ferromagnetism, $0.51<x<0.70$, that the specific heat contains a linear term, presumably a spin-glass contribution. On the other hand, the metallic spin-glass GdAl, investigated by Coey *et al.*,[10] has a ferromagnetic $T^{3/2}$ contribution. It should then not be surprising that the Co-Ga system close to the crossover between spin-glass and ferromagnetism shows both contributions: one proportional to T, the other proportional to $T^{3/2}$.

Finally, this brings us to a discussion of the excess contribution to the heat capacity, C_{sg}, that we ascribe to the spin-glass state. This contribution is summarized in Fig. 5. For the samples with $x=0.49$, 0.50, and 0.51, no evidence for nuclear or spin-wave contributions was found. Consequently, in these three cases, we attribute $C-C_0$ as the spin-glass specific heat C_{sg}. In Figs. 2 and 3 it can be seen that for the sample with $x=0.51$ in zero field C_{sg}/T is constant up to 3 K; its freezing temperature is 3.6 K; for $x=0.50$, C_{sg}/T is constant up to 1.3 K which is 50% of T_f. The lowest concentration sample, $x=0.49$, has a freezing temperature below 1.3 K and C_{sg}/T is never constant; it is already decreasing with increasing temperature at 0.3 K. For the higher-Co-concentration samples, $x=0.53$, 0.55, and 0.56, the linear contribution to the heat capacity is reduced from that at lower concentrations.

The samples with concentration $x<0.52$ show also a distinct difference from the samples with $x>0.52$ when magnetic fields are applied. For higher concentration samples a magnetic field of 2.8 T does not change the specific heat by more than a few percent. The specific heat for the three lower concentration samples drops by 10 to 20% at the lowest temperatures; at higher temperatures, around $2T_f$, the specific heats in zero and 2.8 T become equal and at still higher temperatures the specific heat in a magnetic field has increased above the zero-field results by 20% for $x=0.49$ and somewhat less for the two other samples.

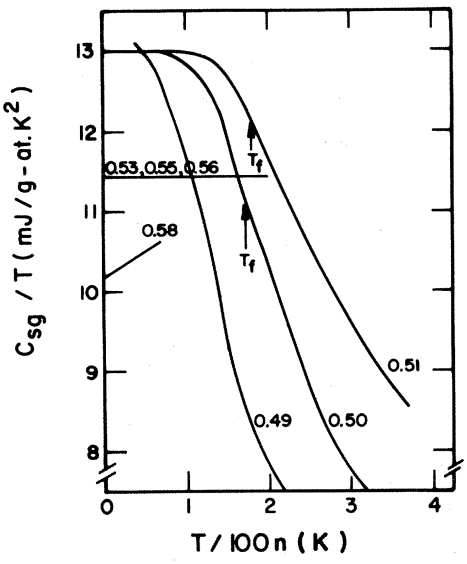

FIG. 5. C_{sg}/T vs $T/100n$ for various cobalt concentrations is shown. C_{sg} is the spin-glass contribution to the specific heat and n is the concentration of AS defects.

From our determination of C_{sg} in zero field we estimated the high-temperature magnetic entropy S_m and calculated the concentration of magnetic entities n_S using

$$S_m = n_S R \ln(2s+1) \tag{3}$$

and $s=\frac{1}{2}$.[1] The results are shown in Table I. The concentration of magnetic constituents obtained by this method is in excellent agreement with the AS defect concentrations found by Meisel *et al.*[4] from their magnetic susceptibility measurements. This agreement indicates that individual AS defects are the principal magnetic entities in Co-Ga in accordance with several workers[1-4] and contrary to proposed models in which single AS defects are nonmagnetic and only clusters are magnetic.[19-21] The magnetic entropy at the spin-glass transition temperature, T_f, is 30%, 35%, and 40% of the high-temperature magnetic entropy for the samples with $x=0.50$, 0.51, and 0.53, respectively. These percentages are similar to the results for the typical spin-glass *Cu*Mn (Ref. 22) and indicate that the majority of the ordering occurs above T_f. The entropy change above T_f occurs in the same temperature region where the paramagnetic Curie temperature, positive at higher temperature, is dropping rapidly to zero on approaching T_f.

For dilute magnetic alloy systems, the interaction is of the type proposed by Ruderman, Kittel, Kasuya, and Yosida (RKKY) (Ref. 23) and is proportional to $1/r^3$; r is the distance between the magnetic ions. With the use of the relation that $nr^3\cong 1$, Souletie and Tournier[24] deduced scaling laws for several properties of these magnetic systems. For sufficient low concentration they derived, for example, that the specific heat divided by the concentration n should follow a universal function of T/n. From these considerations it follows also that C_{sg}/T vs T/n should be a unique function for all concentrations. This function is shown in Fig. 5; for clarity individual points are omitted. For the samples with $x=0.50$ and 0.51 scaling occurs only at the very lowest temperatures. The spin-glass contribution of the 0.53, 0.55, and 0.56 samples are identical so that they mutually scale. It is interesting to note that the AS defect concentration of the measured samples increases by a factor of 8, but that the low-temperature limit of C_{sg}/T differs only by 30%.

In developing a model we will use the following experimental results.

(a) From magnetization and also from entropy considerations it follows that the single AS defects are the magnetic entities and they have a concentration n (or n_S). These defects have spin $\frac{1}{2}$ and effective magnetic moment $(5\pm0.5)\mu_B$.

(b) At the lowest temperatures the spin-glass specific heat in zero magnetic field is proportional to the temperature with a coefficient a_0. With increasing temperatures,

$T \cong T_f$, C_{sg}/T drops below a_0 for $x < 0.52$. For the samples with concentration above 0.52, C_{sg} is linear up to temperatures for which it can be determined with accuracy.

(c) C_{sg} is strongly magnetic field dependent for the lower concentration samples, and nearly field independent for the higher concentrations.

We will use the two-level model introduced by Marshall.[25] This model assumes that the RKKY interaction produces a local field H_i at the magnetic ions which is random in value and removes the twofold degeneracy of the AS defects. In addition, this model also assumes that the density of levels with energy splitting Δ is independent of Δ up to a cutoff Δ_m, so that $\rho(\Delta)d\Delta = A_0 d\Delta$ for $\Delta \leq \Delta_m$ and 0 otherwise. A_0 is the number of magnetic entities per g-at. of the host material per kelvin. The high-temperature cutoff Δ_m is then determined by the total number of magnetic entities in a g-at.:

$$nN_0 = A_0 \Delta_m \ . \tag{4}$$

Each magnetic ion contributes its Schottky specific heat $C_S(T/\Delta)$, so that the spin-glass contribution is $C_{sg} = \int d\Delta \rho(\Delta) C_S(T/\Delta)$. For $T < \frac{1}{7}\Delta_m$, C_{sg} is linear in temperature and $C_{sg}/T = (\pi^2/6)A_0 k$. When T increases above $\frac{1}{7}\Delta_m$, C_{sg}/T decreases below the constant value.

All samples, except the one with the lowest Co concentration, have a linear spin-glass specific heat at low temperatures. Therefore A_0 and Δ_m can be determined and C_{sg} can be calculated. The results of the calculations are shown in Figs. 2 and 3 and the values of the fitting parameters are given in Table II. The agreement with the experimental values is excellent, including the deviation from linearity which starts below T_f for $x = 0.50$ and 0.51. This indicates that only individual AS defects contribute to the spin-glass specific heat. At somewhat higher temperatures the calculated specific heat for these samples is lower than the experimental one, e.g., 10% lower at 4 K for $x = 0.50$ and at 7 K for $x = 0.51$. Therefore, the assumption that $\rho(\Delta)$ is constant up to the cutoff Δ_m, as shown in the inset of Fig. 2, is an oversimplification. Probably the sharp cutoff should have been smoothed out.

The specific heat of the low-concentration samples, $x \leq 0.53$, are magnetic field dependent. We tried to extend the Marshall model used for zero applied field by adding the applied field, H_a, vectorially, or algebraically to the internal fields H_i. The calculated specific heats, however, did not agree with those that are experimental.

In a field of 2.8 T the spin-glass specific heat for the 0.51 sample can be written: $C_{sg}/T = a_{0H} - a_{1H}T$. This relation can be used to deduce the density function $\rho(\Delta)$. As the Schottky specific heat is a function of T/Δ, it follows that if $C_{sg}/T = \Sigma a_{iH} T^i$, then $\rho(\Delta)$ is also a power series, but in powers of Δ: $\rho(\Delta) = \Sigma A_{iH} \Delta^i$; the coefficients a_{iH} and A_{iH} are directly related. Marshall's model corresponds to all coefficients zero except a_{0H} and the corresponding A_{0H}. The size of the next pair of coefficients is indicated by the behavior of the sample with $x = 0.51$ in 2.8 T. Still higher powers of T may be necessary to characterize C_{sg}/T, but they will not be distinguishable from lattice contributions. We have used, therefore, in this slightly modified Marshall model only the first two terms, and for the high-energy cutoff of the density function in a magnetic field: $\Delta_{mH} = \Delta_m + \mu H_a$. The value of a_{0H} was estimated from the extrapolation of C_{sg}/T to 0 K, and with this we calculated A_{0H}. We drew then, in the inset of Fig. 2, a straight line to Δ_{mH} to obtain the best fit with the experimental results. With this model the values of C_{sg}/T in magnetic fields were calculated, not only for the sample with $x = 0.51$, but also for the $x = 0.50$, 0.53, and 0.56. As shown in Figs. 2 and 3, the agreement with the measured values is rather reasonable. This modified model has similarities with the Ising system in the Marshall model, but with a smoothed out density of energy-level splittings, which is physically more appealing.

To be able to understand the hysteresis effects observed in spin-glasses below the freezing temperature Prejean and Souletie[26] have introduced the concept of objects. Because of the mutual magnetic interactions, a set of magnetic ions forms an object containing several thousands of magnetic ions, and which possesses an asymmetric two-level system. The energy difference between the two levels of the object is 2ϵ, but the transition between the two levels is inhibited by an energy barrier $W \pm \epsilon$. It is assumed that the probability to find an object with a given W or a given ϵ is constant up to energy cutoffs W_m or ϵ_m. They show that with this simple model many of the remanence and hysteresis effects can be understood.

To understand the spin-glass specific heat we have also introduced a two-level system but assumed that the objects are individual AS defects and that the defects contributing to the specific heat are in thermal equilibrium. To obtain a linear term in the specific heat with a two-level system it is not necessary that the objects be single ions, but the successful use of Eq. (4) argues strongly for this assumption.

TABLE II. First two coefficients of the polynomial $C_{sg}/T = \Sigma a_i T^i$ representing the spin-glass specific heat in zero field: a_i, and in 2.8 T a_{iH}, $i = 0$ and 1. These coefficients lead to the coefficients A_i and A_{iH} of the polynomial representing the density of systems with energy-level splitting Δ: $\rho(\Delta) = \Sigma A_i \Delta^i$; finally Δ_m and Δ_{mH} are the high-energy cutoff of the systems density in 0 and 2.8 T.

	$H_a = 0$			$H_a = 2.8$ T				
x	a_0 $\left[\frac{\text{mJ}}{\text{g-at. K}^2}\right]$	A_0 $\left[\frac{10^{20}}{\text{g-at. K}}\right]$	Δ_m (K)	a_{0H} $\left[\frac{\text{mJ}}{\text{g-at. K}^2}\right]$	A_{0H} $\left[\frac{10^{20}}{\text{g-at. K}}\right]$	a_{1H} $\left[\frac{\text{mJ}}{\text{g-at. K}^3}\right]$	A_{1H} $\left[\frac{10^{20}}{\text{g-at. K}^2}\right]$	Δ_{mH} (K)
0.50	12.8	5.6	14	11.4	5.0	0.85	0.114	25
0.51	13.0	5.7	26	12.4	5.5	0.45	0.060	34
0.53	11.5	5.1	44	11.8	5.2	0.23	0.031	55
0.56	11.4	5.1	76	11.4	5.1	0.00	0.00	87

From the experimental value of a_0 and the total number of AS defects the high-energy cutoff (in our notation Δ_m) follows directly. With this cutoff the break in the calculated spin-glass specific heat falls at the experimental value, while if the objects contained more than one AS defect the cutoff would be at much lower energy and consequently leads to a break in the spin-glass specific heat at much lower temperatures than observed.

This analysis, in terms of a distributed ensemble of two-level systems, is quite consistent with the ideas presented by Prejean and Souletie.[26] However, in the Co_xGa_{1-x} alloys, it appears that the magnetic objects are single AS defects, at least for $x \leq 0.52$. The picture that forms, then, is that on warming a sample toward T_f increasing numbers of AS defects become unlocked and are free to contribute to the specific heat. Furthermore, it could be expected that upon unfreezing these AS defects, they would also contribute to the reversible susceptibility, and indeed that is what has been observed.[4] However, close to T_f there remains the dilemma, universally found in spin-glasses, that there is an abrupt change (or cusp) in the susceptibility, yet continuous behavior in the temperature dependence of the heat capacity.

IV. CONCLUSIONS

Meisel *et al.*[4] from their susceptibility measurements concluded that they observed spin-glass behavior for the Co-Ga alloys which contained less than 8% AS defects. These AS defects lead to an extra specific heat above the electronic and lattice contributions. There is a distinct difference in the extra specific heat for Co concentration less than 0.52 (AS defect concentration less than 3%) as compared with cobalt concentration more than 0.52. This correlates with the sign of the dominant magnetic interaction which is antiferromagnetic below 0.52, and predominantly ferromagnetic above this concentration. The following observations can be made.

(a) Samples with $x < 0.52$ have an extra specific heat which is attributable to their spin-glass nature. When $x > 0.52$ the extra specific heat contains in addition contributions from the Co nuclei of the AS defects and a small ferromagnetic spin-wave contribution proportional to $T^{3/2}$.

(b) The spin-glass contribution is also different between the two composition regimes. For the samples with $x = 0.53$, 0.55, and 0.56, the spin-glass contributions are identical. They are proportional to T at least up to 7 K, the highest temperature for which this contribution can be deduced with reasonable accuracy. In addition C_{sg} for these samples is nearly magnetic field independent. For $x = 0.50$ and 0.51 the spin-glass specific heat is initially linear, but this behavior weakens when the freezing temperature is approached. These samples show a strong magnetic field dependence.

(c) Entropy considerations show that the single AS defects are magnetic entities and not just clusters containing more than one single AS defect, as has been proposed.

(d) The model proposed by Marshall to explain the extra linear specific heat originally found for dilute magnetic alloys must be modified slightly and can then represent the experimental spin-glass contribution reasonably well.

It follows from this model that individual AS defects are the magnetic entities responsible for the specific heat and entropy. Prejean and Souletie[26] suggested a model which explains hysteresis and remanence by assuming objects or clusters which contain large numbers of magnetic ions. These objects are frozen in an equilibrium state at absolute zero, and unfreeze when the temperature increases. We speculate that on warming, increasing numbers of single AS defects become free to contribute to the thermal properties as well as to the ac susceptibility.

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