Spin-glass-like distribution of interaction fields in Ni-Cu alloys

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Low-temperature heat-capacity measurements were made in magnetic fields up to 42 kOe on several Ni-Cu alloys close to (on both sides of) the critical composition for ferromagnetism. The magnetic component of the specific heat (C_m) is shown to derive from dilute concentrations of magnetic clusters with giant moments and low effective spins. From the field and temperature dependences of C_m, it is also deduced that the interaction (exchange and anisotropy) fields seen by the clusters extend from large positive to large negative values, relative to the applied field. This spin-glass-like distribution of interaction fields, as it pertains to the weakly ferromagnetic alloys, suggests a coexistence of spin-glass and ferromagnetic order. This interaction-field distribution can also account for the anomalously slow approach to saturation of the low-temperature magnetization of these alloys in high fields.

I. INTRODUCTION

The Ni-Cu system has continued to play a prototypal role in various models concerning the magnetic and related properties of disordered transiton-metal alloys. After long service as a principal application of the collective-electron rigid-band theory,[1] the magnetism of Ni-Cu has now come to be regarded as a prime example for which local environmental effects are a governing factor.[2,3] As deduced from neutron scattering[4,5] and magnetic[6-8] measurements, the reduction in the moment of the Ni atoms near a Cu atom in the Ni-rich alloys ultimately culminates with decreasing Ni concentration in a weak ferromagnetic state whose magnetization is distributed inhomogeneously in magnetic clusters with giant moments. For alloys of less than the critical Ni concentration for ferromagnetism (~ 43 at.%), magnetic[8] and heat-capacity[9] measurements have shown that the giant-moment clusters persist as superparamagnetic entities and are situated at statistically Ni-rich regions.

Although the exchange coupling within the clusters in Ni-Cu is strongly ferromagnetic, as evidenced by the high cluster-disordering temperatures (~ 600 K) deduced from resistivity data,[10] very little is known about the interactions between the clusters, especially in the paramagnetic and weakly ferromagnetic regimes. Recently, however, detailed low-field magnetic measurements have revealed that as the Ni concentration in Ni-Cu is effectively decreased through the critical value to the paramagnetic side, the susceptibility versus temperature transition-point anomaly changes from a divergence to a finite peak.[11] This result suggests the appearance of a low-temperature spin-glass state and thus gives support to a recent conjecture about the magnetic states of nearly ferromagnetic Ni-Cu (and similar) alloys.[12]

In this paper, we present the results of low-temperature heat-capacity measurements on several paramagnetic and weakly ferromagnetic Ni-Cu alloys near the critical composition in various externally applied magnetic fields. Our results give clear confirming evidence for the existence of dilute concentrations of giant-moment clusters, and they also reveal that the interaction (exchange and anisotropy) fields experienced by the clusters have a broad continuous distribution that extends from large positive to large negative fields, relative to the applied field. This type of interaction-field distribution has usually been associated with the spin-glass state of disordered alloys such as Cu-Mn, whose local atomic moments are coupled via an oscillatory indirect exchange potential.[13] In the case of Ni-Cu, the entities coupled in this fashion are magnetic clusters, and our results indicate that spin-glass order occurs in both the paramagnetic and weakly ferromagnetic alloys, where in the latter it appears to coexist with the small spontaneous magnetization signifying ferromagnetic order. Our calorimetric results for a single paramagnetic Ni-Cu alloy were recently reported in a brief letter.[14]

II. EXPERIMENTAL PROCEDURES

For the calorimetry, alloy samples of nominal atomic compositions: $Ni_{40}Cu_{60}$, $Ni_{43}Cu_{57}$, and $Ni_{45}Cu_{55}$, each weighing ~ 10 g, were cut from ingots prepared by induction melting and chill casting, followed by cold swaging. Together with smaller samples (~ 0.3 g) cut from the same ingots, they were annealed for three days at 1000 °C and quenched into water. The smaller samples were used for the purpose of magnetic characterization; their magnetizations were measured in a vibrating-sample magnetometer between 2.4 and 50 K in fields up to 40 kOe.

The heat-capacity measurements were made between 1.2 and 20 K by standard adiabatic methods. The sample was clamped to a copper holder, to which carbon and germanium resistor thermometers and a Manganin wire resistance heater were thermally attached. The sample unit was suspended adiabatically inside a copper heat shield connected to a vapor-pressure chamber and a He-gas thermometer bulb, used in calibrating the germanium thermometer in zero field and the carbon thermometer in various fields. Calibration reference points were established with the vapor-pressure chamber filled with liquid He or liquid Ne. An adjacent chamber filled with liquid He was pumped down to different pressures to obtain calibrations below 4.2 K. A mechanical heat switch allowed the sample to be brought to the temperature of the heat shield. The small temperature and field dependences of the heater resistance were measured and their effects on the input power were taken into account. The vacuum-sealed calorimeter assembly was immersed in a liquid-He bath containing a Nb-Ti superconducting solenoid, which was operated in the persistent mode to give a steady uniform field of up to 42 kOe at the sample. The thermometer resistances before and after each heating interval were monitored potentiometrically, the off-balance signal being fed to a sensitive strip-chart recorder, from which the data were taken, computer reduced, and tabulated. More complete experimental details are available in a fuller report.[15]

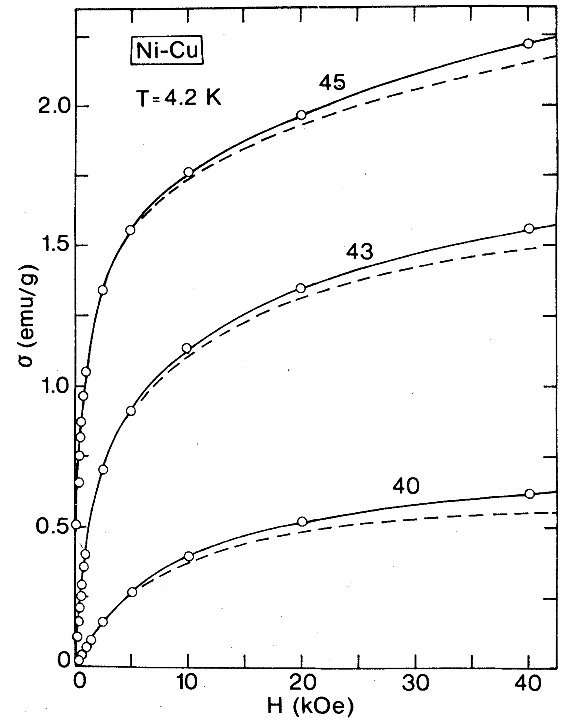

FIG. 1. Magnetization (σ) vs field (H) for Ni-Cu alloys (designated by at.% Ni) at 4.2 K. Dashed curves show field dependence of $\sigma - \chi' H$, where χ' is the Pauli susceptibility.

III. RESULTS AND DISCUSSION

The magnetization versus field (σ-vs-H) curves for our three alloy samples at 4.2 K are displayed in Fig. 1. Rapid changes with composition in this critical region are clearly evident, especially in the steepness of the curves at low fields. However, none of these alloys are ferromagnetic at this temperature, for which the Arrott plots (σ^2 vs H/σ) of the same data give smooth curves with positive H/σ intercepts. (Moreover, the Arrott curves have normal concave-downward shapes down to fields of ~ 25 Oe, which testifies to the macroscopic homogeneity of the alloy samples.) Our results for the initial susceptibility (χ_0) as a function of temperature show that all these alloys follow a Curie–Weiss-like behavior. For $Ni_{45}Cu_{55}$ and $Ni_{43}Cu_{57}$ χ_0^{-1} is seen to head into zero at 3.5 and ~ 1.2 K, respectively, thus marking the ferromagnetic Curie points; for $Ni_{40}Cu_{60}$ χ_0^{-1} appears to have a zero-temperature intercept, indicating the absence of long-range ferromagnetic order.

Another feature of interest in Fig. 1 is the fact that the low-temperature magnetization curves, after rising rapidly at low fields, approach high-field saturation extremely slowly. Even if allowance is made for the Pauli susceptibility[8] ($\chi' \approx 1.7 \times 10^{-6}$ emu/Oe g) and $\chi' H$ is subtracted from the measured σ, the remaining magnetization associated with the clusters[8] (represented in Fig. 1 by dashed curves) shows a much slower approach to saturation than would be predicted from any Brillouin-function fit of the low-field data. A possible cause of this apparent anomaly, as proposed previously,[16] is a broad distribution in the magnitude of the cluster moments, such that the clusters with the smaller moments require stronger fields to become substantially magnetized. However, theoretical considerations of local magnetic environment effects in Ni-Cu indicate that, since these effects are cooperative, the magnetic clusters that form at local Ni-rich regions will have moments that exceed a fairly large critical size.[2,17] It therefore appears that the anomalously slow high-field saturation of the low-temperature magnetization of Ni-Cu must have a different origin. Our calorimetric results presented below indicate a plausible alternative explanation.

The molar specific heat (C) versus temperature (T) data for our three alloy samples in different fields are plotted as C/T vs T^2 in Fig. 2. The data obtained above ~ 11 K are excluded in order that the low-temperature portion of this figure remain reasonably uncompressed; even so, for clarity, only about half of the low-temperature points are plotted. Our results for zero field, in their pronounced low-temperature departure from the nearly linear behavior at higher temperatures, agree with those of previous zero-field measurements on Ni-Cu of similar compositions.[9,18–20] Since a straight line in this type of plot, expressed as

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

gives the electronic and lattice heat coefficients (γ and β, respectively), the anomalous low-temperature behavior indicates the appearance of an additional contribution, which has been attributed to the existence of magnetic clusters.[9,21] That this contribution to the specific heat is magnetic in origin is confirmed by our results in Fig. 2, which show that it is strongly affected by a magnetic field. In particular, the magnetic specific heat at the lowest temperatures is seen to diminish with increasing field, and our data at other intermediate fields document this effect in detail. We have exploited this effect by extrapolating C/T to infinite field (i.e., for decreasing $1/H$) in order to locate the low-temperature end of the base line representing the sum of the electronic and lattice heats, as expressed in Eq. (1). The base line for each alloy was then adjusted in slope until it was seen to merge tangentially with the zero-field C/T-vs-T^2 data at temperatures of ~ 15 K, which showed that the magnetic specific heat in zero field is vanishing rapidly with increasing temperature. The base lines thus determined for all the alloys are shown dashed in Fig. 2. The values for γ and β and for the Debye temperature derived from β are listed in Table I. The values agree quite well with earlier calorimetric results, considering that the latter were all obtained from zero-field data and involved approximate allowances for the magnetic specific heat.[9,18–20,22]

In our work, the magnetic specific heat (C_m) was determined from the difference between each measured value of C and the base-line value at the same temperature values (T). The value of C_m for our three alloys are plotted against T in Figs. 3(a), 3(b), and 3(c). The scatter of the points, though always less than 1% of the total measured specific heat, corresponds to a much larger relative error in C_m, particularly at the higher temperatures where the base-line subtraction becomes very severe. The figures were therefore truncated at 12 K. Despite the scatter that remains, the plotted points for each alloy at each field are seen to describe a smooth curve with a fairly well-defined maximum. Moreover, with increasing field, the maximum in each case moves to higher temperatures. This suggests that C_m depends in a Schottky-like fashion on the ratio T/H_T, where H_T is the total effective field consisting of the applied field plus the interaction (exchange and anisotropy) fields in the alloy. Presumably, the presence of the interaction fields is what gives rise to the observed C_m in

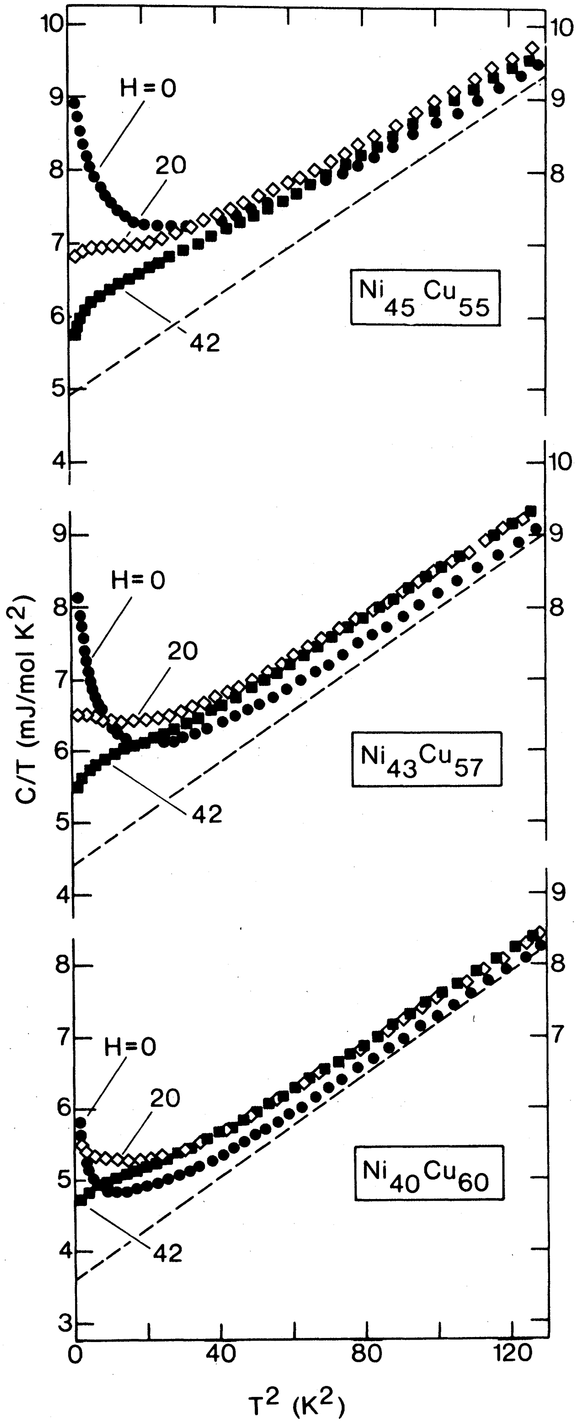

FIG. 2. C/T vs T^2, where C is the total specific heat measured at temperature T, for Ni-Cu alloys in various fields (in kOe). The base lines, shown dashed, represent estimated sums of electronic and lattice heats.

TABLE I. Electronic and lattice heat coefficients and Debye temperatures.

Alloy	γ (mJ/mol deg^2)	β (mJ/mol deg^4)	Θ_D (K)
$Ni_{40}Cu_{60}$	3.6	0.0360	378
$Ni_{43}Cu_{57}$	4.4	0.0355	380
$Ni_{45}Cu_{55}$	4.9	0.0340	385

zero applied field. However, a simple Schottky function associated with a single discrete value of H_T behaves exponentially at low temperatures, whereas our results for $C_m(T)$ do not appear to follow such a behavior. Especially at our highest field (42 kOe), where the $C_m(T)$ points for each alloy reach down to very small values at our lowest temperatures, they are seen to be heading into the origin linearly or almost linearly. An initial linear temperature dependence of C_m was also recently observed by Lanchester *et al.*[23] from similar measurements on nearly ferromagnetic Ni-Cu at comparable fields but lower temperatures (0.3 to 4.0 K). Their temperatures did not extend high enough to reach and delineate the C_m maxima, and they fitted their data with Einstein functions appropriate for very large spin. The distinctly peaked behavior of C_m shown by our results (and those of others in zero field[22]) indicates that the effective spin (per magnetic cluster) is quite small.

An initial linear variation of C_m with temperature is observed characteristically in spin-glass alloys such as Cu-Mn,[24,25] and has been attributed to an exchange-field distribution $P(H_{ex})$ which, as derived from an oscillatory indirect-exchange coupling in a disordered Ising system,[13] extends from positive to negative H_{ex} symmetrically, with a substantial magnitude at zero H_{ex}. In applying this rationale to our results for Ni-Cu, we can ignore the effects that may arise according to more recent observations that $P(H_{ex})$ vanishes at and very close to zero H_{ex} (Refs. 26 and 27); these effects would occur well below our temperatures of measurement. Since our Ni-Cu results were obtained at various applied fields (H), we will be concerned with the total-field distribution $P(H_T)$ where, within an Ising model, H_T is the scalar sum of H and the net exchange field at a cluster, H_{ex}. For simplicity, it will be assumed that $P(H_T)$ is rectangular in shape, extending from $\bar{H}_T - \Delta H_T$ to $\bar{H}_T + \Delta H_T$ at a constant value of $(2\Delta H_T)^{-1}$, that changes in H cause $P(H_T)$ to shift rigidly along the H_T axis without changing in shape, and that the shape of $P(H_T)$ is also unchanged by variations in temperature over the range of our measurements. Furthermore, from the form of the $C_m(T)$ curves in

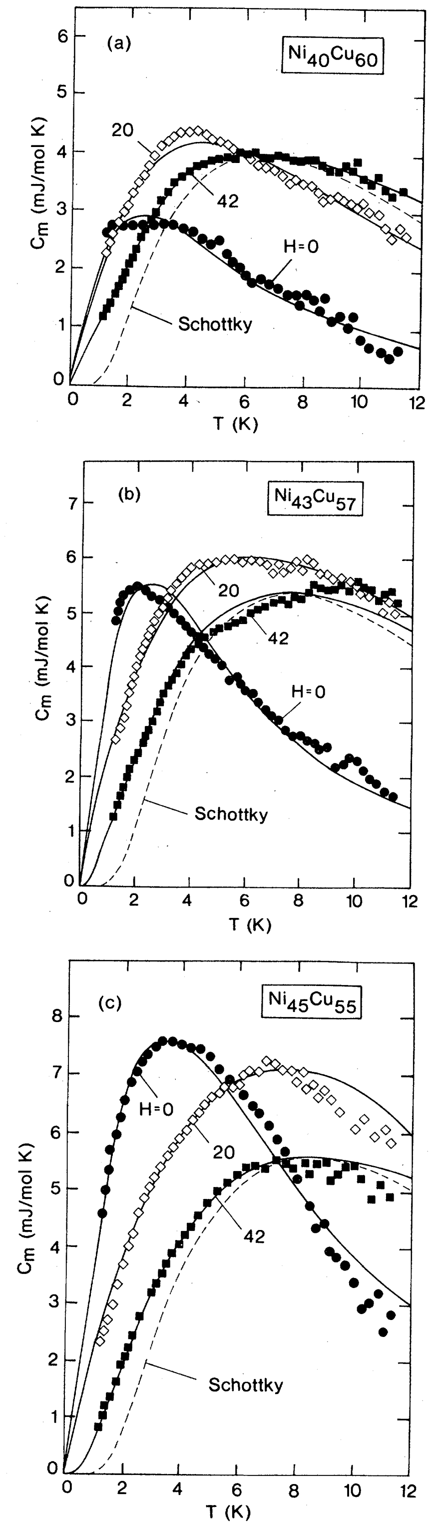

FIG. 3. Magnetic specific heat (C_m) vs temperature for (a) $Ni_{40}Cu_{60}$, (b) $Ni_{43}Cu_{57}$, and (c) $Ni_{45}Cu_{55}$, in various fields (in kOe). Solid curves were fitted to experimental points as described in text. Schottky curves (for $S=2$) are shown dashed.

Figs. 3(a), 3(b), and 3(c), allowing for some broadening due to the $P(H_T)$ distribution, we estimate that the effective spin, $S=2$, so that the effective moment per cluster, $\mu^*=2g\mu_B$, g being the effective Landé factor and μ_B the Bohr magneton. For reasons given earlier, μ^* is assumed to have a negligible distribution and is represented here by its average value. In terms of the reduced variable, $x=g\mu_B H_T/kT=\mu^* H_T/2kT$, the magnetic specific heat per mole may then be expressed as

$$C_m(T)=c^*N_A k(2\Delta x)^{-1}\int_{\bar{x}-\Delta x}^{\bar{x}+\Delta x} C_S(x)\,dx \ , \quad (2)$$

where c^*N_A is the number of magnetic clusters per mole (N_A being Avogadro's number), k is the Boltzmann constant, and

$$C_S(x)=(x/2)^2\,\mathrm{csch}^2(x/2)-(5x/2)^2\,\mathrm{csch}^2(5x/2) \ , \quad (3)$$

the Schottky function for $S=2$.

Within our simplified model, it follows from Eq. (2) that the shape of the $C_m(T)$ curve is determined uniquely by the value of the parameter, $\alpha\equiv\Delta x/\bar{x}=\Delta H_T/\bar{H}_T$, where $\Delta H_T(=\Delta H_{ex})$ is the half-width of the total-field (and exchange-field) distribution and $\bar{H}_T(=H+\bar{H}_{ex})$ is the average total field. In Fig. 4, $C_m(T)$ curves calculated for various representative values of α are shown plotted universally as $C_m/c^*N_A k$ vs $2kT/\mu^*\bar{H}_T$. As expected, all the curves

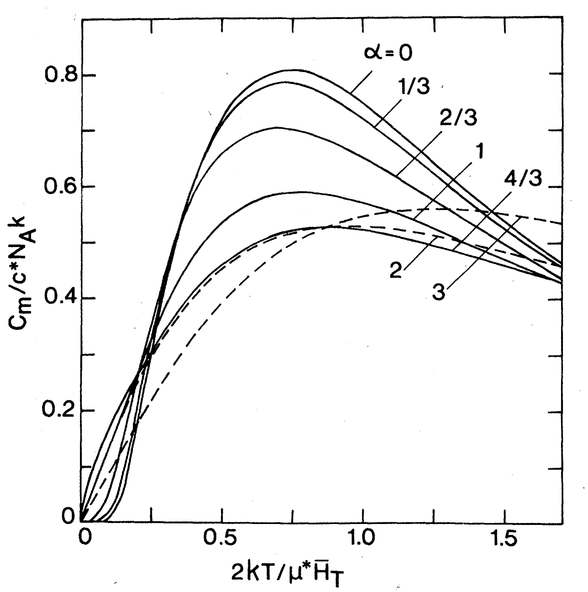

FIG. 4. $C_m/c^*N_A k$ vs $2kT/\mu^*\bar{H}_T$ curves for various values of $\alpha(=\Delta H_T/\bar{H}_T)$ calculated from Eqs. (2) and (3) for rectangular distribution of total effective field H_T. Note that as $\alpha\rightarrow\infty$ ($\bar{H}_T\rightarrow 0$) the shape of the curve approaches that for $\alpha=1$.

for $\alpha>0$ have broader maxima than the curve for zero α, the latter being the simple Schottky function given by Eq. (3). Moreover, the initial rise of the curves continues to be exponential, though less pronouncedly, as α increases from zero to unity, where it becomes linear and remains so for all values of α greater than unity, corresponding to the condition that $P(H_T)\neq 0$ for $H_T=0$.

In applying this model to our experimental results, we start with the $C_m(T)$ points for $Ni_{40}Cu_{60}$ plotted in Fig. 3(a). The fact that this alloy is not ferromagnetic suggests that at zero H the rectangular distribution in $H_T(=H_{ex})$ is symmetrical about $H_T=0$ (i.e., $\bar{H}_{ex}=0$) and thus, that at all fields, $\bar{H}_T=H$. Noting that the $C_m(T)$ points for $H=42$ kOe are approaching the origin linearly, we conclude that $P(H_T)$ must be nonzero down to $H_T=0$ (or to some negative H_T value); we therefore set $\Delta H_T=42$ kOe as the smallest distribution half-width consistent with experiment. The values of $\alpha(=\Delta H_T/\bar{H}_T)$ for the three fields of measurement are thus determined (and are listed in Table II) and they, in turn, determine the pertinent curves from among the set represented in Fig. 4. In order to fit our $C_m(T)$ results, the curves must be scaled horizontally and vertically by a suitable choice of values for the cluster moment μ^* and the cluster concentration c^*, respectively. An optimum fit was made individually for each field of measurement. The $C_m(T)$ curves corresponding to these fits are shown in Fig. 3(a), and the values used for μ^* and c^* are listed in Table II.

Proceeding to our $C_m(T)$ results for the other two alloys, we note in Figs. 3(b) and 3(c) that the plotted points for $H=42$ kOe are heading slightly to the right of the origin, thus appearing to give small positive temperature intercepts. This feature, which is clearly larger for $Ni_{45}Cu_{55}$ than for $Ni_{43}Cu_{57}$, suggests that $C_m(T)$ is mildly exponential rather than linear in its initial variation, and it follows from our model that for $H=42$ kOe the rectangular distribution $P(H_T)$ extends down to small positive values of H_T but is zero at $H_T=0$. Since both these alloys are weakly ferromagnetic, according to our magnetic measurements [though in neither case does $C_m(T)$ at zero field display a sharp anomaly at the Curie temperature], it seems reasonable to assume that $P(H_T)$ is slightly asymmetrical about $H_T=H$, such that $\bar{H}_T=H+H_d$, where H_d $(=\bar{H}_{ex})$ represents a small positive displacement. If we further assume that the distribution half-width (ΔH_T) for these two alloys is the same (namely, 42 kOe) as for $Ni_{40}Cu_{60}$, then $P(H_T)$ for $H=42$ kOe will extend only down to (and be zero below) $H_T=H_d$, thus giving rise to an initial exponential variation of $C_m(T)$. Hence, the $C_m(T)$ points for the two alloys at 42 kOe were fitted with curves from the set in Fig. 4 for $\alpha=\Delta H_T/(H+H_d)=(42\text{ kOe})/(42\text{ kOe}+H_d)$, again scaled individually by appropriate choice of values for μ^* and c^*. The

TABLE II. Fitting parameters for the $C_m(T)$ curves in Fig. 3.

Alloy	H (kOe)	α	μ^* (μ_B)	c^* (10^{-3})
$Ni_{40}Cu_{60}$	0	∞	4.5	0.59
	20	2.10	7.4	0.95
	42	1.00	5.8	0.82
$Ni_{43}Cu_{57}$	0	10.50	5.5	1.12
	20	1.75	8.3	1.37
	42	0.91	6.2	1.04
$Ni_{45}Cu_{55}$	0	3.50	6.1	1.63
	20	1.31	8.0	1.61
	42	0.78	6.7	1.02

optimum fits, represented by the $C_m(T)$ curves in Figs. 3(b) and 3(c) for $H=42$ kOe, were achieved with $H_d=4$ and 12 kOe for $Ni_{43}Cu_{57}$ and $Ni_{45}Cu_{55}$, respectively, and with the values of μ^* and c^* (and α) listed in Table II. Using the same values for H_d and ΔH_T, we then fitted the $C_m(T)$ points for the two alloys at 20 kOe and zero field; our optimum results obtained with the μ^* and c^* (and α) values listed in Table II are again represented by $C_m(T)$ curves in Figs. 3(b) and 3(c).

On the whole, the curves in Fig. 3 are very reasonable fits to the $C_m(T)$ points for the three alloy samples measured in different fields. The curves are particularly illuminating in their initial low-temperature behavior, which is linear in all cases except for $Ni_{43}Cu_{57}$ and $Ni_{45}Cu_{55}$ at 42 kOe, where the behavior is mildly exponential. But even the latter is in contrast with the strongly exponential initial behavior of a simple Schottky function, and this is seen in Fig. 3 where such a function has been fitted to the peak of each of the $C_m(T)$ curves for $H=42$ kOe. This contrast is a direct consequence of the broad exchange-field distribution assumed within our model for each of the alloys. Moreover, the values of μ^* and c^* in Table II show that this distribution, which extends from large positive to large negative exchange fields, pertains to small concentrations of magnetic clusters with very sizable moments. Although the μ^* and c^* values deduced for each alloy are not the same for different fields, probably in large part due to the oversimplicity of our model assumptions, the averages of these values and their increase (especially of c^*) with increasing Ni concentration are in fair agreement with the cluster moments and concentrations derived from magnetic measurements on Ni-Cu alloys of similar composition.[8]

A more exact analysis of our $C_m(T)$ results would assume a more plausibly shaped exchange-field distribution, taking also into account the changes in this distribution with temperature and applied field,[28] and would explicitly include the effects of local anisotropy.[29] These and other model improvements are currently being explored. Nevertheless, our present model assumptions appear to capture a very essential feature of the magnetic states of Ni-Cu near the critical composition for ferromagnetism—namely, that there are giant-moment clusters which experience a broad spin-glass-like distribution of exchange fields. Furthermore, the conclusion drawn from our heat-capacity results that the exchange-field distribution $P(H_{ex})$ extends to large negative values of H_{ex}, also provides an explanation for the fact that the low-temperature magnetization of these alloys shows an anomalously slow approach to saturation at high fields, as described earlier. According to this explanation, an increasing applied field will reverse the moments of an increasing number of magnetic clusters until its magnitude ultimately exceeds that of the strongest negative H_{ex}. Thus, our exchange-field model for Ni-Cu gives a fairly consistent rationale for several of the basic thermodynamic properties of the alloys.

Finally, our calorimetric results for Ni-Cu suggest a comparison with those of recent measurements on the insulating spin-glass system (Eu, Sr)S near the critical composition for ferromagnetism.[30] The magnetic specific heat of the latter also was found to contain a large temperature-linear component which decreases substantially (after a small initial rise) with increasing field. Thus, it appears that the competition between the short-range direct interactions in these insulating pseudobinary compounds gives rise to an exchange-field distribution similar to that produced by the long-range indirect interactions between the magnetic clusters in Ni-Cu or between the magnetic atoms in more typical spin-glass alloys such as dilute *Cu*Mn.

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