Magnetization in Mn-rich γ-$Cu_{100-x}Mn_x$ $(36 \leqslant x \leqslant 83)$ alloys in fields up to 75 kOe

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(Received 28 October 1998)

The field dependence of dc magnetization $[M(H)]$ has been measured in γ-$Cu_{100-x}Mn_x$ ($x=36$, 60, 73, 76, and 83) alloys in magnetic fields up to 75 kOe at different temperatures between 4.2 and 21 K. The $M(H)$ data are found to be almost linear with field beyond 10 kOe. This is expected since the present alloys are antiferromagnetically ordered. But, at lower fields ($H<10$ kOe), the variation is slower than linear. The change of slope in $M(H)$ beyond the critical field of 10 kOe is attributed to the spin-flop transition which marks the collapse of the helical spin structure and the onset of the linear spin-density-wave modulation. Another interesting observation is that the composition dependences of the magnetization and the electrical resistivity exhibit a minimum and a maximum, respectively, around $x=73$. This behavior is described in terms of the transition of magnetic structure from AF3 to AF1. [S0163-1829(99)09125-0]

I. INTRODUCTION

In the last few decades, spin glasses have been a subject of great interest for their unusual magnetic behavior.[1,2] However, in recent times, the intensity of these activities has reduced considerably, although a complete understanding of their basic properties still remains unclear. Most of the earlier studies have dealt with dilute CuMn alloys. In the recent past, very accurate and careful neutron diffraction,[3–6] ac-susceptibility,[7] and dc-magnetization[8] studies have revealed a mixed itinerant antiferromagnetic and spin-glass phase at low temperatures in Mn-rich γ-$Cu_{100-x}Mn_x$ alloys ($x \geqslant 72$ at. %). This has generated a lot of interest since most of the theoretical models, until now, have predicted a mixed ferromagnetic and spin-glass phase. Long back, Overhauser proposed the spin density wave (SDW) model[9,10] to describe antiferromagnetic behavior in dilute CuMn alloys. The SDW is a collective deformed state of electron gas that can be stabilized for particular wave vectors.[9,11] Very recently, a series of neutron-polarization-analysis measurements[11–14] have renewed the interest in CuMn alloys. In detailed studies,[5,12] Cable and co-workers have shown that the spin structure of $Cu_{100-x}Mn_x$ ($x \leqslant 25$ at. %) alloys is fundamentally related to the incommensurate SDW modulation. According to their study, the random atomic short-range order (ASRO) plays an important role in the development of a spin correlation which is ferromagnetic or antiferromagnetic depending on the Mn concentration. Another study by Werner *et al.*[6] had shown that long-range order does not develop in CuMn alloys since it gives SDW instability. Most of the studies reported so far give descriptions of spin structure of Cu-rich CuMn alloys at a fixed temperature. Moreover, until now, there is no experimental evidence of the SDW modulations from dc magnetization, ac susceptibility, and specific heat due to the complicated crystallographic and magnetic structure of the system.[11] Also, as the Mn concentration increases, long-range spin order is expected to appear in Mn-rich alloys. Since most of the recent work is restricted to Cu-rich alloys, it is also not known how the SDW modulation behaves in Mn-rich alloys at lower temperatures well below T_f and with increasing magnetic field. Keeping in mind all the above facts, we have made detailed dc-magnetization measurements on Mn-rich γ-$Cu_{100-x}Mn_x$ alloys ($x=36$, 60, 73, 76, and 83) in magnetic fields up to 75 kOe at nine different temperatures between 4.2 and 21 K which are well below the spin freezing temperatures of all the alloys. To the best of our knowledge, there is as such no report on the field dependence of dc magnetization in Mn-rich CuMn alloys. This is very important in understanding the various magnetic structures of this complicated system. Hence the present investigation attempts to deal with some specific problems: (1) to interpret field and composition dependence of the dc-magnetization data at low temperatures well below T_f and (2) to find whether the dc magnetization gives any supporting evidence for the SDW modulation or not.

II. EXPERIMENTAL DETAILS

The details of alloy preparation and characterization were given earlier.[7] The measurement of magnetization (Lakeshore Model 7229 Extraction Magnetometer/Susceptometer) was performed up to a dc field of 75 kOe in the temperature range of 4.2 to 21 K.

III. RESULTS AND DISCUSSION

The present γ-$Cu_{100-x}Mn_x$ ($x=36$, 60, 73, 76, and 83) alloys have exotic magnetic structures at low temperatures below T_f. The earlier ac-susceptibility,[7] dc-magnetization,

0163-1829/99/60(1)/440(5)/$15.00 PRB 60

TABLE I. Alloy compositions and the values of T_f, T_N, electrical resistivity ($\rho_{4.2\ K}$), magnetization at 75 kOe and 4.2 K, and fitting parameters to Eq. (1).

$Cu_{100-x}Mn_x$ x (at. %)	T_f (K)	T_N (K)	$\rho_{4.2\ K}$ ($\mu\Omega$ cm)	M (emu/g)	n	K (10^{-2}) (emu/g(kOe)^n)
36	135		93	1.91	1.06	1.94
60	149		176	0.99	0.96	1.44
73	172		184	0.80	0.97	1.21
76	145	275	196	0.83	0.95	1.32
83	45	484	120	0.91	0.91	1.79

and neutron diffraction[3] studies have revealed that the alloys with $x \leqslant 73$ have short-range order. But, as the Mn concentration increases, bigger and bigger clusters are formed and finally long-range antiferromagnetic order appears. According to the magnetic phase diagram,[7] the alloys with $x=36$, 60, and 73 are cluster glasses whereas $x=76$ and 83 are in the mixed cluster-glass and long-range antiferromagnetic phase. The values of the spin freezing (T_f) and Neel (T_N) temperatures are given in Table I. Recently, a very precise and detailed electrical resistivity $[\rho(T)]$ study[15] has shown that the present γ-$Cu_{100-x}Mn_x$ ($x=36$, 60, 73, 76, and 83) alloys are substantially disordered with large residual resistivity ($\rho_0 \sim 100$–200 $\mu\Omega$ cm) (see Table I). At low temperatures below 30 K, a distinct resistivity minimum is found in all the alloys. This is interpreted convincingly in terms of electron-electron interaction (EEI) effects in the weak-localization limit. Also the positive magnetoresistance[16] ($\Delta\rho/\rho$) in both longitudinal and transverse directions below 20 K in all the Mn-rich alloys is found to be very much consistent with the prediction of the EEI effects.

The field dependence of dc magnetization $[M(H)]$ in magnetic fields up to 75 kOe is shown in Figs. 1–4 for nine different temperatures between 4.2 and 21 K in the alloys with $x=36$, 60, 73, 76, and 83. The measurements were done in the zero-field-cooled (ZFC) state. Here, one can see that the dispersion in the data is much less than the width of the symbols. The values of M at 75 kOe are found in the range of (0.8–1.9) emu/g at 4.2 K which are in good agreement with the values reported earlier for concentrated CuMn alloys.[7,8] Interestingly, the $M(H)$ data exhibit an almost temperature-independent behavior between 4.2 and 21 K in $x=60$, 73, and 76 whereas only a small variation is observed in $x=36$ and 83. In $x=36$ (see Fig. 1), the magnetization at

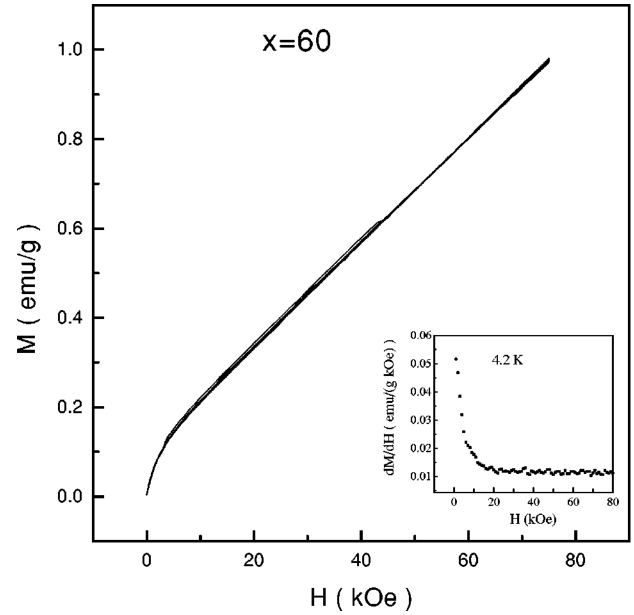

FIG. 2. External field dependence of dc magnetization for the alloy with $x=60$ at 4.2, 5.9, 8.0, 10.1, 12.1, 14.1, 16.1, 18.2, and 21.3 K. In the inset, a first derivative plot of dc magnetization (dM/dH) at 4.2 K is shown.

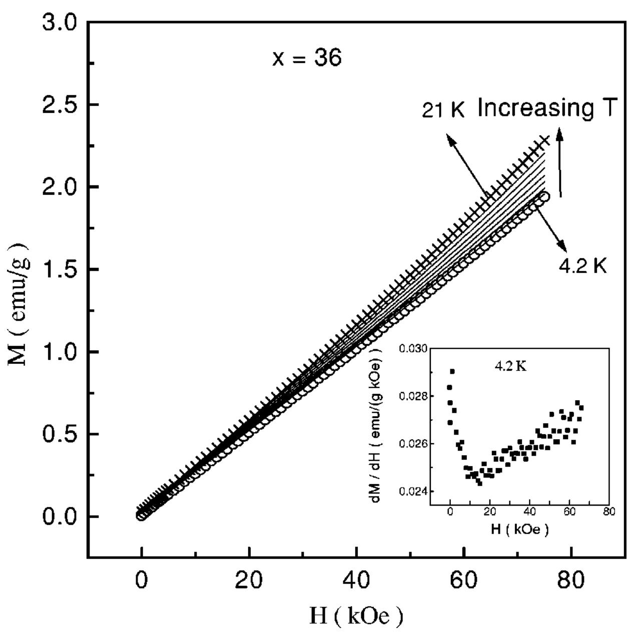

FIG. 1. External field dependence of dc magnetization for the alloy with $x=36$ at 4.2, 5.8, 7.8, 10.0, 12.1, 14.1, 16.1, 18.2, and 21.3 K. In the inset, a first derivative plot of dc magnetization (dM/dH) at 4.2 K is shown.

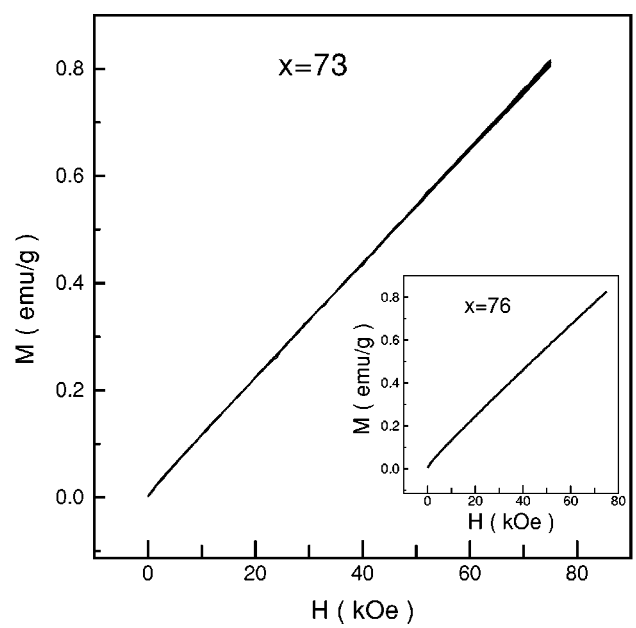

FIG. 3. External field dependence of dc magnetization for the alloys with $x=73$ and 76 (inset) at 4.2, 5.9, 8.1, 10.0, 12.1, 14.1, 16.1, 18.2, and 21.3 K.

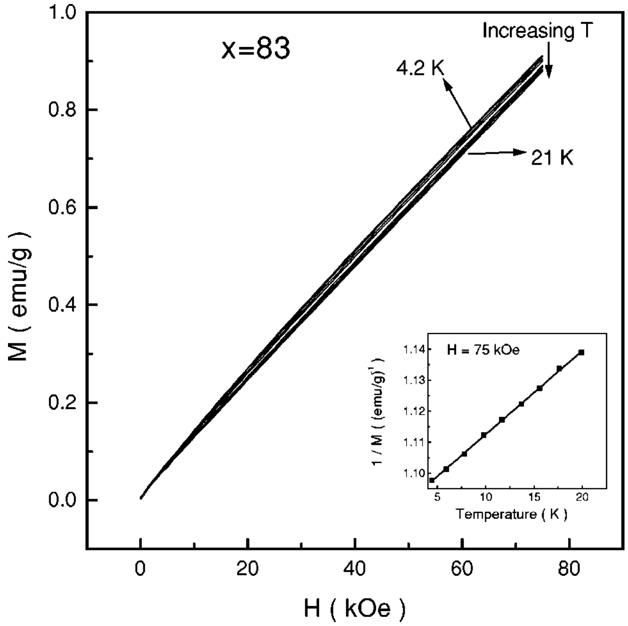

FIG. 4. External field dependence of dc magnetization for the alloy with $x=83$ at 4.2, 5.9, 7.8, 9.8, 13.7, 15.6, 17.6, 19.5, and 21.0 K. In the inset, the temperature dependence of $1/M$ at the magnetic field of 75 kOe and the best-fitted line are shown.

a given field is found to decrease as temperature decreases. The above behavior is generally expected in ZFC states of spin–cluster-glass alloys.[1,7,8] At temperatures well below T_f, more and more spin-cluster moments get frozen. As a consequence, the alignment of moments along a given applied magnetic field becomes less resulting in a decrease in the net moment. The present alloy with $x=36$ is in the critical concentration region of the magnetic phase diagram of CuMn where the system exhibits both the spin and cluster-glass behaviors.[3,7] Moreover, the present temperature range between 4.2 and 21 K are well below the spin-freezing temperature ($T_f=135$ K) of the alloy $x=36$. In addition, the recent magnetoresistance study[16] has clearly shown the dominant presence of spin–cluster-glass phase in $x=36$. Hence, the decrease in magnetization with decreasing temperatures is quite expected in the alloy $x=36$. On the other hand, in the Mn-rich alloys with $x=60$, 73, and 76, the $M(H)$ data exhibit a temperature-independent behavior below 21 K (see Figs. 2 and 3). This is quite puzzling in the sense that the early dc-magnetization study[7] had clearly shown a decrease in $M_{\mathrm{dc}}(T)$ with decreasing temperatures. However, the study was restricted to the range of (30–300) K. It is well known that as Mn concentration increases, clusters in CuMn alloys grow in size and, as a result, the magnetic correlation within the clusters becomes stronger in the Mn-rich alloys. The present temperature range of 4.2 to 21 K is too small for any significant thermal relaxation of these frozen cluster moments for $x=60$, 73, and 76, and, hence, a temperature-independent behavior shows up. On the contrary, in the alloy $x=83$, $M(H)$ at a given field is found to decrease with increasing temperatures (see Fig. 4). This behavior is exactly opposite to what is found in $x=36$. The temperature variation of M at $H=75$ kOe is shown in the inset of Fig. 4 where the plot of 1/M vs T is found to be linear with a positive intercept on the $1/M$ axis. This certainly gives a clear indication of the increase in long-range antiferromagnetic order in the alloy $x=83$ where the magnetization is expected to vary as $1/M=T/\alpha+T_N/\alpha$, where α is a constant. From the fitting parameters (α and T_N/α), the Neel temperature is calculated and is found to be around 470 K. This is in very good agreement with the earlier reported value for $x=83$ (see Table I). However, no such dependence could be found in the present data of the alloy with $x=76$. Moreover, the earlier studies of magnetization[7] and neutron diffraction[3] have revealed a mixed long-range antiferromagnetic order and cluster-glass phase at low temperatures in $x=83$. The behavior of the cluster-glass phase in this highly Mn-rich alloy might be an indication of the presence of SDW modulation at low temperatures. In addition, the neutron-polarization-analysis study by Cable *et al.*[5] had provided a rough estimate of the temperature dependence of the magnetic peak-height intensity and the spin-correlation length for the SDW modulation in concentrated $Cu_{100-x}Mn_x$ ($x=25$ and 15) alloys. The central magnetic peak-height intensity and the correlation length for the alloy $x=25$ are found to be almost temperature-independent below 100 K. Interestingly, the present findings of almost temperature-independent behavior of $M(H)$ in the alloys $x=60$, 73, 76, and 83 are more or less in agreement with those of the neutron-polarization study whereas the variation of $M(H)$ in $x=36$ could not be supported from those observations. It is important to mention here that the neutron-polarization study is restricted only to the low-concentration alloys ($x\leqslant 25$ at. %) and for very few temperatures between 4.2 and 300 K, while the present study deals with the Mn-rich alloys at temperatures well below T_f. Hence, to find a proper correlation, a detailed neutron-polarization study at low temperatures in the Mn-rich alloys is needed. Nevertheless, the present report gives detailed information about the temperature dependence of the magnetic moment of this complicated SDW CuMn system.

The magnetic field dependence of the $M(H)$ data in all the alloys is found to be almost linear in the range of 20–75 kOe (see Figs. 1–4). This is expected since the present alloys are antiferromagnetically ordered. However, one can find that the data vary slightly faster than linear in the alloy with $x=36$ whereas they are slower in the Mn-rich ($x\geqslant 60$ at. %) alloys. Hence to give a complete description, we have fitted the data to the relation

$$M=KH^n, \qquad (1)$$

where K is the proportionality constant. The values of n and K at 4.2 K are found to be in the range of (0.91–1.06) and (1.2–1.9) $\times 10^{-2}$ (emu/g)(kOe)$^{-n}$, respectively for all the alloys. The fitting parameters at 4.2 K are given in Table I. For other temperatures, the values of n and K are found to be almost the same and hence are not included in Table I. It is interesting to note that the temperature dependence of $M(H)$ in the alloy $x=36$ comes mostly from the variation of n [$\simeq$(1.06–1.12)], while the values of K [$\simeq 0.02$ (emu/gm)(kOe)$^{-n}$] are found to be almost the same. The most important finding in the present data is that $M(H)$ deviates sharply from linearity below 10 kOe in all the alloys. This is found to be more pronounced in the alloy $x=60$ (see Fig. 2). To find the deviation more clearly, we

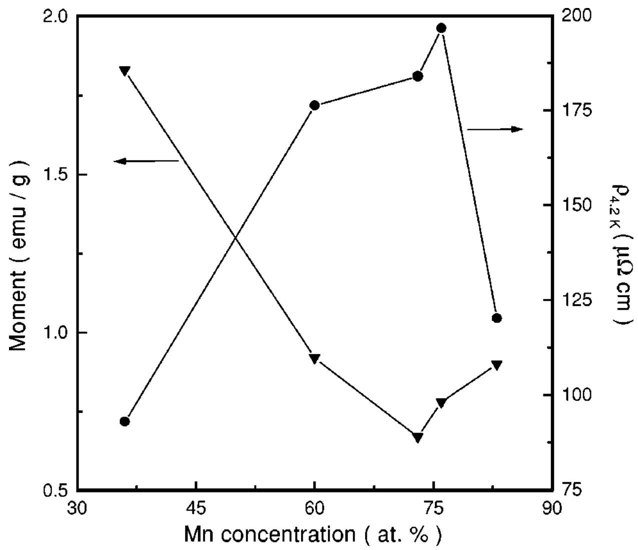

FIG. 5. Concentration dependence of dc magnetization at 75 kOe and 4.2 K, and electrical resistivity at 4.2 K ($\rho_{4.2\ K}$).

have plotted the first derivative of M (i.e., dM/dH) with respect to the magnetic field H (see insets of Figs. 1 and 2). The data have shown a sharp drop around 10 kOe beyond which it becomes almost field independent. This sudden increase in magnetization beyond a critical field of 10 kOe is attributed to the spin-flop transition which marks the collapse of the helical spin structure and the onset of a linear SDW. A similar behavior was observed earlier in the Mn-rich alloys.[7] But the interpretation could not be made since $M(H)$ was restricted up to 16 kOe only. In addition, a neutron diffraction study[17] of Mn-rich CuMn alloys had shown that the average canting of spins with respect to the crystallographic ordering direction [001] is around 5°. This, as a result, gives rise to a helical spin structure. Also, the recent study on single crystal $Cu_{10}Mn_{90}$ alloy[8] has found that spin freezing occurs in both parallel and perpendicular to crystallographic ordering direction [001]. But, the freezing is found to be more in the parallel direction compared to the perpendicular one. The present study of the field dependence of dc magnetization gives a strong evidence of the presence of a helical spin structure in the Mn-rich CuMn alloys, which is nothing but a manifestation of the SDW modulation.

The concentration dependence of the magnetization at 75 kOe is shown in Fig. 5 for the present alloys with $x=36$, 60, 73, 76, and 83. The plot shows a decrease until $x=73$ beyond which it starts increasing. This is quite interesting in the sense that the minimum gives a clear indication of a critical concentration of $x=72$ where the antiferromagnetic structure of the alloys goes from AF3 to AF1.[3] In the AF3 structure, both the nearest (J_1) and the next-nearest neighbor (J_2) interactions of Mn are antiferromagnetic while, in AF1, J_1 and J_2 are antiferromagnetic and ferromagnetic, respectively. In addition, J_2 is found to be almost twice of J_1.[3] In addition to this, the neutron-polarization-analysis study[12] has shown that the average number of Mn next-nearest neighbors is large compared to those of the nearest neighbors. Hence, the decrease in magnetization until $x=73$ (below which it is AF3 type) can be attributed to the increase in antiferromagnetic ordering. But, beyond $x=73$, the magnetic structure becomes AF1 type where the next-nearest ferromagnetic interactions (J_2) aligned some of the random moments. Thus, the magnetization is enhanced. Here, one can also expect that the above behavior of increase in the magnetization should be reflected in the composition dependence of electrical resistivity [$\rho(T)$]. In Fig. 5, we have plotted the concentration dependence of the electrical resistivity at 4.2 K where the values of $\rho_{4.2\ K}$ are taken from our earlier report.[15] It is interesting to see that the data exhibit a peak around $x=76$. The increase in resistivity until $x=76$ is an indication of enhanced disorder of the magnetic moments (i.e., spin) in AF3 structure. But, as soon as the AF1 structure sets in, the next-nearest neighbor ferromagnetic interactions align some of the randomly oriented moments in a preferred direction resulting in a decrease in the disorder and hence the resistivity. The above findings certainly show that random atomic short-range order plays an important role in the present alloys. However, no such correlation was observed between the EEI effects and the alloy composition.[15] In our earlier study of $\rho(T)$,[15] it was clearly shown that the temperatures of the resistivity minima had a roughly linear dependence on the values of resistivity of the present alloys and not on their composition (see Fig. 2 of Ref. 15). Moreover, a correlation between the EEI effects and the magnetization is not really expected here since magnetic sates of any three-dimensional disordered alloys do not have any major effect on the EEI in the weak-localization limit.[18,19] Nevertheless, this is a study on Mn-rich CuMn alloys where the change from AF3 to AF1 magnetic structure is observed from the concentration dependence of the dc magnetization as well as the electrical resistivity.

IV. CONCLUSIONS

The studies of the field and the composition dependence of dc magnetization were presented in γ-$Cu_{100-x}Mn_x$ ($x=36$, 60, 73, 76, and 83) alloys in magnetic fields of up to 75 kOe at different temperatures between 4.2 and 21 K. The $M(H)$ data have been found to be almost temperature independent for the alloys with $x=60$, 73, and 76. However, small variations are observed in the alloys $x=36$ and 83 which are interpreted in terms of spin–cluster-glass and long-range antiferromagnetic orders, respectively. The field dependence of M is found to be almost linear beyond 10 kOe. This is expected since the present alloys are antiferromagnetically ordered. But, in the low-field limit ($H<10$ kOe), there is a deviation where $M(H)$ goes slower than linear. The faster increase in magnetization beyond 10 kOe is attributed to the spin-flop transition which marks the collapse of the helical spin-structure and the onset of a linear SDW modulation. Another important observation in the present study is that the composition dependences of the magnetization and the electrical resistivity data exhibit a minimum and a maximum, respectively, almost around $x=(73–76)$. This is explained in terms of a crossover of antiferromagnetic structure from AF3 to AF1 at $x=73$.

The work at Texas A & M University was supported by the National Science Foundation (INT-9603137) and the Robert A. Welch Foundation, Houston, Texas (Grant No. A-0514). We are also grateful to the Department of Science and Technology, Government of India, for providing support through Project No. SP/S2/M-24/93.

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