Susceptibility of the *Cu*Mn spin-glass: Frequency and field dependences

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Alternating current susceptibility measurements on powdered samples of the spin-glass *Cu*Mn (Mn concentrations: $0.23 \leq c \leq 6.3$ at.%) showed relatively broad maxima as well as sharp cusps in χ' as a function of temperature depending on the method of preparing the sample. In contrast to the broadly peaked $\chi'(T)$ curves, the sharply cusped ones were more affected by small, external, static fields H. This field dependence was measured at various temperatures above and below T_f, the freezing temperature. At T_f the behavior of $\chi'(H)$ was analyzed and the critical exponent δ determined. The susceptibility data for the lower concentration alloys were independent of the measurement frequency ($1 \text{ Hz} < \nu < 10 \text{ kHz}$) within the absolute experimental accuracy of about 1%. However, in the concentration regime of ≈ 1 at.% Mn and above, the sharply cusped, so-called "quenched" samples exhibited a small frequency dependence in $\chi'(T)$ near T_f. The relative shift in freezing temperature $\Delta T_f/T_f$ per decade of frequency was found to be 0.0050 independent of concentration from 1 to 6.3 at.% Mn. Below T_f the various $\chi'(\nu)$ curves seemed to converge towards a single, nonzero χ' value as $T \rightarrow 0$ K. Above T_f and below about 50 K, the susceptibility obeyed a simple Curie-like law, whereas in the higher temperature region from 100 to 150 K a Curie-Weiss-like behavior was observed with a small, positive paramagnetic Curie-Weiss temperature. To analyze the behavior of $\chi'(T)$ near T_f and at the lowest temperatures of measurement 0.4 K, the superparamagnetic blocking model of Wohlfarth was used. A distribution of blocking temperatures was thus determined from the experimental χ' data. This distribution function shows a rather sharp step at T_f, indicating that there are cooperative effects in the freezing of a spin-glass.

I. INTRODUCTION

The term spin-glass has been broadly applied to the freezing behavior and low-temperature magnetic properties of many different classes of random magnetic systems. At present the spin-glass problem is the subject of intensive study,[1] from the experimental side in order to characterize systematically the exact nature of the observed behavior, and theoretically to determine the best and simplest model with which to describe these properties. The experimental results encompassing a large variety of different measurements are somewhat muddled by the complexity of the spin-glass phenomena and by inconsistencies in the sample preparations. While among the existing theories, there remain many controversies over the question of a phase transition and how best to describe the low-temperature properties.[1]

One of the simplest to observe and most striking experimental features of a spin-glass is the "sharp cusp" in the low-field ac susceptibility.[2] While many different measurement techniques have been employed to study spin-glasses, the susceptibility as a function of temperature not only gives the clearest determination of the freezing temperature T_f, but also is of intrinsic importance in investigating the nature of this "freezing" and presents a distinct challenge to the theoretical description of the spin-glasses. Recently there has been a large amount of experimental effort devoted to the frequency and field dependences of $\chi(T)$.[3-16]

The question of a frequency or time dependence is fundamentally related to the description of the transition which occurs at the freezing temperature. A strong "time of measurement" behavior in the various experimental properties suggests a gradual freezing over a wide temperature range and is thus commensurate with a glasslike or thermally activated change of phase. On the other hand, freezing behavior which is independent of time, occurring at the same T_f for various frequency windows indicates a usual type of static phase transition with the associated critical phenomena. Somewhere between these two extremes lies the not fully explored regime of a dynamical phase transition which is cooperative yet time dependent.

Spin-glasses are sensitively affected in the temperature region around T_f by an applied magnetic field. At T_f the susceptibility "cusp" is severely rounded in rather small fields of order 100 Oe.[2] The characteristics of the magnetization, M, are also drastically modified as T_f is traversed. For $T > T_f$ a Brillouin-function-like $M(H)$ curve is found, while below T_f an "S" shaped $M(H)$ contour appears at T_f which permits a determination of T_f.[17,18] Here ($T < T_f$) above a certain rather low threshold field, irreversible and time-dependent behavior becomes visible in the magnetization. Such metastable effects are characteristic to the low-temperature spin-glass state over a wide range of fields. Yet even in fields around 400 kOe (40 T) the spin-glass cannot be fully saturated.[19] This wide range response to an external field represents a peculiar and complicating property. So it is of particular interest to study the field dependence of χ at T_f and at temperatures below T_f. Not only will additional information be gained concerning the type of phase transition, but also about the low-temperature excitations and activation processes.

For many years now there have been a large number of measurements of the spin-glass susceptibility. This quantity is most difficult to define and to measure for these nonlinear $M(H)$, irreversible, metastable, time-dependent systems. We have decided to reconsider the *Cu*Mn archetypal spin-glass by systematically measuring the differential susceptibility, both its in-phase and out-of-phase components with calibrated magnitudes. The available temperature region for these measurements was 0.3 to 150 K which nicely spanned the freezing temperatures ($3 \lesssim T_f \lesssim 30$ K) by a factor of at least 5 for our Mn concentrations of from 0.23 to 6.3 at.%. Two distinctly different sample preparations and subsequent heat treatments were employed in order to illustrate the effects of a metallurgical nature on the spin-glass state. The magnitude of the ac driving field was varied to test for possible alterations in the susceptibility. In addition its frequency was changed from 1 Hz to 10 kHz, and the susceptibility was carefully measured as a function of the frequency through the freezing temperature. Finally, dc external fields up to 25 kOe (2.5 T), parallel to the ac driving field, were applied to determine the field dependence of the susceptibility.

In the next section we describe our sample preparations and experimental apparatus for the susceptibility. Section III, on experimental results, is divided into three parts: the temperature and frequency dependence of χ; its field dependence; and finally the low-temperature behavior and the shape of $\chi(T)$. In Sec. IV we discuss our results and offer some conclusions.

II. EXPERIMENTAL DESCRIPTION

We have studied *Cu*Mn samples with various Mn concentrations, c, in the range from 0.23 to 6.3 at.% Mn. The samples were prepared in two different ways. Alloys with Mn concentrations of 0.23, 0.46, and 1.48 at.% Mn (samples Ia, Ib, and Ic, respectively) were made by induction melting at 1100° C in a 0.75-bar argon atmosphere. In order to avoid the effects of eddy currents and to reduce the skin effect as much as possible, the experiments were performed on finely powdered samples, with grain size of 100 μm or less. These powders were then annealed at 500° C for several hours, and slowly furnace cooled to room temperature. Alloys with 0.57, 0.70, 0.94, 2.0, and 6.3 at.% Mn (IIa to IIe, respectively) were fabricated by arc melting, followed by 24 h of homogenization annealing at 900° C, and then rapidly quenched in ice-water. These samples did not receive any further heat treatment after filing into powder (grain size also $\lesssim 100$ μm). The Mn concentrations were determined by chemical analysis, and were found to agree to within 5% of the initial, nominal concentrations. A listing of the samples studied, their concentration, heat treatment, and freezing temperature T_f is given in Table I.

The differential susceptibility was measured by means of a mutual inductance technique in the frequency range from 1 Hz to 10 kHz. Both the in-phase component χ' and the out-of-phase component

TABLE I. Samples of *Cu*Mn alloys (powders < 100 μm).

	Type I (slow cooled) Concentration (at.% Mn)	T_f (K)		Type II (quenched) Concentration (at.% Mn)	T_f (K)
Ia	0.23	2.85	IIa	0.57[a]	6.00
Ib	0.46	5.00	IIb	0.70[b]	7.65
Ic	1.48	12.40	IIc	0.94	9.40[c]
			IId	2.0	15.50[c]
			IIe	6.3	32.3[c]

[a]A portion of this sample was additionally annealed at 500 °C and then slowly cooled in a similar fashion to the type I samples.

[b]On this sample bulk measurements were also performed.

[c]These samples exhibit a small frequency dependency of the susceptibility maximum. The above values for the freezing temperatures are determined at a frequency of 234 Hz.

χ'' of the complex susceptibility are measured simultaneously. The ac driving field, h, usually employed was ≈ 1 Oe. In certain measurements h was varied between 0.1 and 10 Oe without any noticeable effect in the susceptibility. So the magnitude of the ac driving field does not seem to be an important parameter in our study of the *Cu*Mn susceptibility.

The absolute value of the susceptibility is obtained from a calibration of the apparatus with the standard paramagnet $Mn(NH_4)_2(SO_4)_2 \cdot 6H_2O$. The accuracy of this calibration is better than 1%. However, at a fixed frequency the relative accuracy of χ' is of the order of 0.1%. Measurements are usually performed in the temperature range from 1 to 150 K.[20] In the range 1.2 to 20 K, where the spin-glass freezing temperatures occur for most of these *Cu*Mn alloys, the temperature could be kept constant to within 0.1%, while the absolute temperature accuracy is better than 0.5%. At 100 K the absolute error in the thermometer calibration is about 1%. For certain susceptibility experiments temperatures down to 0.3 K were obtained using a ^{3}He cryostat.[21] An external static field, H, could be applied parallel to the ac driving field via two different magnets. A water cooled, copper wire solenoid was used to generate fields up to 4.5 kOe (0.45 T) and a superconducting coil was available to produce magnetic fields with a maximum value of 36 kOe (3.6 T).[22]

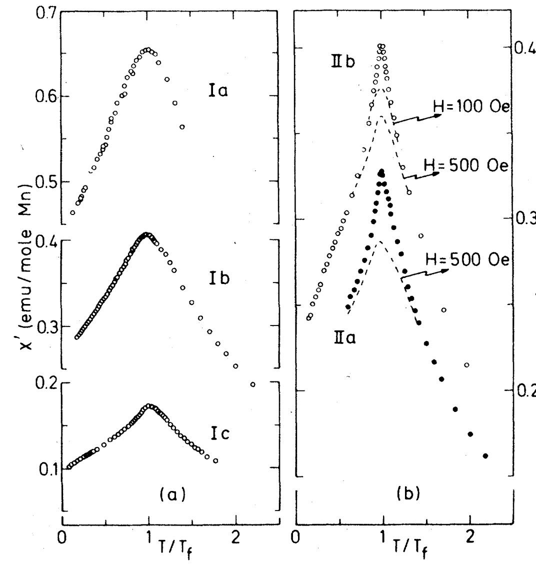

FIG. 1. Susceptibility χ' as a function of reduced temperature T/T_f for various *Cu*Mn spin-glass (powder) alloys: (a) Ia(0.23 at.% Mn) $T_f = 2.85$ K, Ib(0.46 at.% Mn) $T_f = 5.00$ K, and Ic(1.48 at.% Mn) $T_f = 12.40$ K; (b) IIa(0.57 at.% Mn) $T_f = 6.00$ K and IIb(0.70 at.% Mn) $T_f = 7.65$ K. The dashed lines represent data in an external magnetic field as labeled. The samples Ia, Ib, Ic and IIa, IIb, respectively, have been prepared and annealed using different methods, as described in text.

III. EXPERIMENTAL RESULTS

In this section we first limit ourselves to measurements concerning the shape of the maximum in the susceptibility versus temperature. Then for a number of different Mn concentrations we present data on the frequency dependence of the susceptibility cusp and on the susceptibility at high temperatures. The field dependence of the susceptibility $\chi(h,H)$ is given in the next part (Sec. III B). Finally in Sec. III C we analyze the low-temperature behavior of the susceptibility and its shape in terms of a distribution of blocking temperatures employing the model of Wohlfarth.[23]

A. Temperature and frequency dependence of χ

Some typical temperature dependences of the susceptibility $\chi'(T)$ are shown in Fig. 1 for five different *Cu*Mn alloys.[24] The temperature axis is normalized to the freezing temperature T_f (see Table I). All these measurements were performed with the ac driving field $h \leq 1$ Oe at a frequency $\nu = 332$ Hz. The data were taken in "zero" field—no compensation was made for the earth magnetic field of approximately 0.5 Oe. The dashed lines in Fig. 1(b) represent measurements performed in external fields of $H = 100$ and 500 Oe. The samples Ia (0.23 at.% Mn), Ib (0.46 at.% Mn), and Ic (1.48 at.% Mn) show relatively broad maxima [Fig. 1(a)] at the freezing temperatures 2.85 K, 5.00 K, and 12.40 K, respectively. The freezing temperatures are frequency independent between 1 Hz and 10 kHz within our experimental accuracy. Thus, the frequency dependence of T_f in these metallic spin-glasses is much weaker than those reported for the so-called "insulating spin-glasses."[25] The susceptibilities of the three slow cooled alloys shown in Fig. 1(a) are less dependent on the external dc field ($\vec{H} \parallel \vec{h}$) than for the quenched alloys. For example, an overall reduction of $\simeq 15\%$ is achieved for $H = 2$ kOe, independent of the temperature for T below 2 T_f.

In Fig. 1(b) the susceptibility $\chi'(T)$ is given for two quenched *Cu*Mn alloys. These samples exhibit a sharp cusp in $\chi'(T)$ at T_f which occurs at 6.00 K (IIa—0.57 at.% Mn) and 7.65 K (IIb—0.70 at.% Mn). It should be noted that samples of type I and samples of type II represent two different extremes in the preparation and heat treatment procedures. Thus, the effect of various heat treatments plays an important role not only in the magnitude of χ' near T_f,[26] but also in the sharpness and form of $\chi'(T)$

around T_f. Nevertheless, the temperature T_f, at which the peak in $\chi'(T)$ occurs is relatively independent of the heat treatment. Again, for these type II quenched samples, the freezing temperatures are independent of frequency (1 Hz $< \nu <$ 10 kHz) within the experimental accuracy. An important difference between the susceptibility measurements for the two types of *Cu*Mn samples is the sensitivity to the external field, *H*. For quenched samples the sharpness of the peak is strongly influenced by small external fields—see the dashed lines in Fig. 1(b). A similarly sensitive field dependence was found in the early susceptibility experiments on quenched *Au*Fe alloys by Cannella and Mydosh.[2] A detailed discussion of the external field dependence for the *Cu*Mn alloys will be given later on in this paper.

In our experiments much attention was devoted to the possibility of thermal-magnetic history effects in the ac susceptibility. All measured $\chi'(T,H)$ curves did reproduce within the experimental accuracy, independent of whether the sample was cooled in zero applied field or not. Thus, in contrast to the dc susceptibility,[27,28] the ac susceptibility does not depend on when ($T >> T_f$ or $T << T_f$) the H field is applied. In addition, no changes in the susceptibilities were found by repeating the measurements after storing the samples at room temperature for a period of one month.

In the experiments the imaginary part of the susceptibility signal χ'' was typically about 1% of χ' for frequencies less than 1 kHz. The $\chi''(T)$ curves showed *no* measurable anomaly at T_f and are insensitive to applying an external field and for varying the frequency from 1 Hz to 1 kHz. However, even for our grain size ($\lesssim 100$ μm), the onset of skin effect and eddy current absorptions causes a steady increase of the out-of-phase signal for measuring frequencies above 1 kHz. Such effects reduce the absolute value of the measured in-phase signal. The physical quantities χ' and χ'' due to the magnetic ions in the spin-glass are, of course, not affected by eddy current losses and the effect of the excluded volume of the sample. So the observed increase in out-of-phase signal and decrease in the in-phase signal are an artifact of the measurement, and if too large, it is then no longer possible to identify the observed signals as due to the magnetic behavior of *Cu*Mn. In Fig. 2 for sample IIb, *Cu*Mn 0.46 at.% (powder) both χ' and χ'' are plotted as a function of frequency at T_f. Note that the magnitude of the out-of-phase signal approaches the in-phase signal as $\nu \rightarrow 10$ kHz. Additionally, we performed measurements on bulk samples of the *Cu*Mn spin-glass. Using rods with a typical bulk volume of ≈ 2 mm^3, the χ' signal at 10 Hz was already the same order of magnitude as the χ'' signal (see Fig. 3). At higher frequencies an enormous increase in χ'' was observed up to, for example, $\chi'' > 100\chi'$ at 1 kHz as is illustrated for bulk IIb

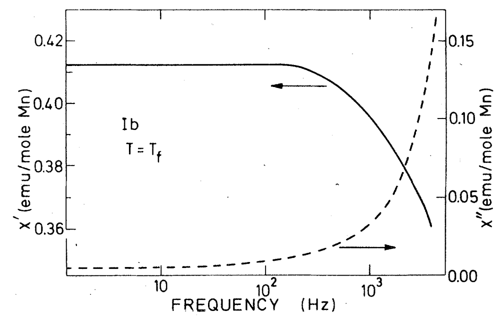

FIG. 2. The in-phase component χ' and the out-of-phase component χ'' of the complex susceptibility as a function of the log of the frequency at $T_f = 5.00$ K for sample Ib (Cu-0.46 at.% Mn, powder).

Cu-0.70 at.% Mn in Fig. 3. This elucidates the extreme caution which must be employed in determining the true χ' for bulk, dilute spin-glasses. It is noted, that the frequency-dependent behavior of $\chi'(T)$ due to the above effects introduces a pseudo frequency dependence of the susceptibility maxima because they vary with temperature. This (additional) variation of the "freezing temperature" with frequency is of course not an intrinsic spin-glass effect.

The essential difference between the measurements on samples I (slow-cooled) and samples II (quenched) is the shape of the χ' maximum around the freezing temperature. It was previously suggested by a number of authors[29,30] that this effect is related to the preparation and heat treatment of the particular sample. In order to confirm these ideas and to verify the difference in χ' shape between our two

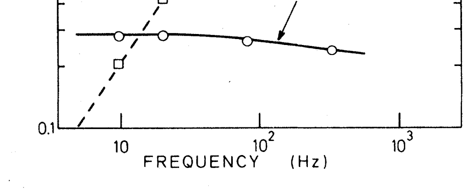

FIG. 3. The in-phase component χ' and the out-of-phase component χ'' as a function of the frequency at $T_f = 7.65$ K for sample IIb (Cu-0.70 at.% Mn, bulk). The volume of the rod is about 2 mm^3.

types of $CuMn$ samples, we annealed the Cu-0.57 at. % Mn powder (IIa) for 3 h at 500° C and allowed it to cool slowly back to room temperature in a few hours. This is the heat treatment used for the type I $CuMn$ alloys. From Fig. 4 it can be seen that the previously, sharply peaked, $\chi'(T)$ now exhibits a relatively broad maximum at approximately the same T_f. The difference in amplitude between the two $\chi'(T)$ curves can thus clearly be attributed to the effect of the heat treatment. The broad maximum shaped $\chi'(T)$ is found to be typical for type I samples. Also after this slow cooling, effects of applying a small H field were somewhat weaker, again a characteristic of the type I samples.

Obviously much care should be taken in preparing the samples. Possible changes due to the ac frequency and the external magnetic field near the susceptibility maximum are expected to be observed more clearly on type II (quenched) samples. In addition, a nonrandom distribution in the manganese concentration over the sample can be avoided by rapidly quenching the samples after the homogenizing anneal.

According to Tholence[11] it should be possible to observe a frequency dependence of the freezing temperature in the higher concentration regime of the $CuMn$ spin-glass. Correspondingly we performed susceptibility measurements on $CuMn$ alloys for concentrations up to 6.3 at. %. Figure 5 shows the results for Cu-0.94 at. % Mn (IIc) over a wide temperature range. The open circles represent data taken at a frequency of 234 Hz with an ac driving field $h \approx 1$ Oe. In the temperature region immediately around the peak, susceptibility data with maximum accuracy were taken as a function of frequency from

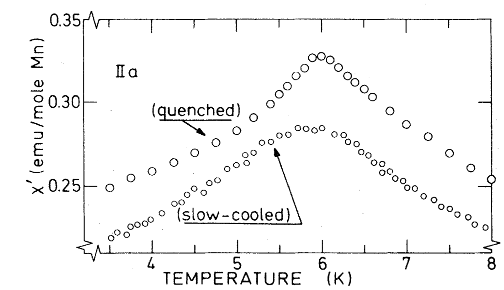

FIG. 4. The susceptibility χ' as a function of temperature for two samples of $CuMn$ IIa (0.57 at. %, powder) which were differently heat treated as labeled.

1 Hz to 10 kHz. Some of the $\chi'(T,\nu)$ curves are depicted in the inset of Fig. 5. The frequency dependence is extremely small for this alloy ($\Delta T_f = 0.14$ K for $\Delta\nu = 10^3$ Hz). It should be noted here that the relative accuracy of the measurements is $\approx 0.1\%$, decreasing to 0.3% at the lowest frequency, whereas the absolute calibration of the susceptibility apparatus is of the order of 1%. Similar measurements on higher concentration samples IId and IIe Cu-2.0 and 6.3 at. % Mn showed an enhanced frequency dependence of the freezing temperature, i.e., $\Delta T_f = 0.23$ and 0.49 K for $\Delta\nu = 10^3$ Hz. This also explains why frequency effects in $\chi'(T)$ are hardly observable for concentrations less than ≈ 1 at. % Mn.[24] Nevertheless, the normalized frequency shift $\Delta T_f / T_f$ is equal for all three $CuMn$ samples. This constant

$$\frac{\Delta T_f}{T_f(\Delta \log_{10}\nu)} = 0.0050 \ ,$$

is a significant feature of our measurements and is in

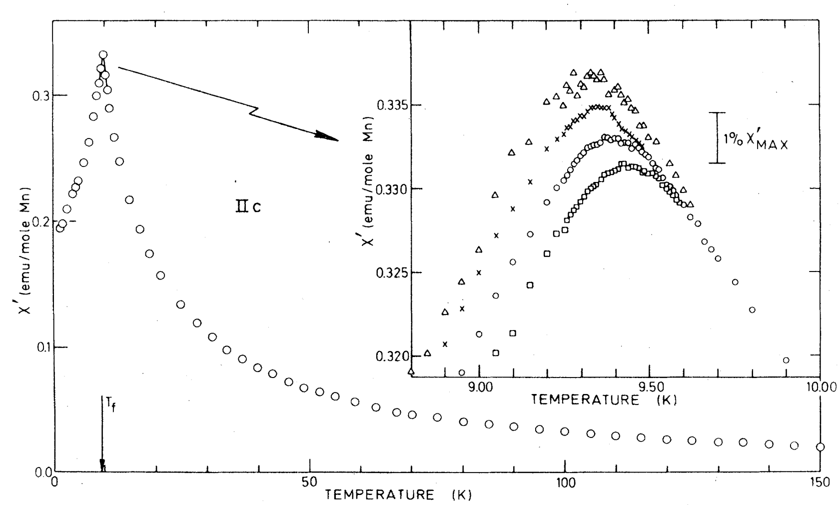

FIG. 5. Zero-field susceptibility χ' as a function of temperature for sample IIc (Cu-0.94 at. % Mn, powder). Measuring frequencies: □, 1.33 kHz, O, 234 Hz; ×, 10.4 Hz; and Δ, 2.6 Hz.

contrast to the suggestion of Tholence[11] that $\Delta T_f/\Delta \log_{10}\nu$ is proportional to the concentration. It should be noted here that $T_f \propto c^{0.63}$ (Ref. 29). The constant, $\Delta T_f/(T_f \Delta \log_{10}\nu)$, can be obtained with the superparamagnetic blocking model with the additional assumptions of Tholence only by scaling temperatures with respect to T_f instead of c.

It can further be seen from the inset of Fig. 5 that for temperatures greater than 9.6 K there is no effect of varying the frequency. All curves merge together in a "Curie-Weiss"-like behavior which persists to very high temperatures (150 K maximum measurement temperature). As the maximum in $\chi'(T)$ is approached from the high-T side, deviations from the single line behavior occur first for the highest frequency. For lower frequencies the observed $\chi'(T_f)$ maxima are shifted to lower T values and the absolute values of $\chi'(T_f)$ are increased. Thus, the "breakaway" on the high-temperature side is frequency dependent along with the shape of the $\chi'(T)$ curve. It would seem to us that the exact behavior of the $\chi'(T)$ near T_f is of more fundamental physical interest than the frequency dependence of T_f, as defined by the maximum in $\chi'(T)$. None the less in keeping with so many other investigations,[5,8,11,16,25] we give the frequency dependence of the freezing temperature in Fig. 6, employing a T_f^{-1} vs $\log_{10}\nu$ plot. If one assumes that the frequency dependence of T_f is describable in terms of a double well potential with activation energy, E_a, then the freezing process which is a simple blocking of cluster spin may be characterized by an Arrhenius law $\nu = \nu_0 \exp(-E_a/kT_f)$. The slope of the line in Fig. 6 gives an activation energy E_a of 4400 K, with even larger values for the higher concentration alloys. In addition the corresponding ν_0 values are $\approx 10^{200}$. Such E_a and ν_0 values are completely without physical meaning and thus cast doubt upon the simple thermally activated blocking model. In an attempt to remove these unphysical magnitudes and yet retain the superparamagnetic blocking model for the spin-glass freezing, Tholence[12] has used a Fulcher law, $\nu = \nu_0 \exp[-E_a/k(T_f - T_0)]$, to describe the frequency dependence of T_f. By using the low temperature, remanent magnetization estimate of $\nu_0 \approx 10^{+13}$ s^{-1}, a value of the parameter T_0 is obtained which is $\approx 0.9T_f$, and E_a is of order 50 K for *Cu*Mn, for 3.3 to 8 at.% Mn.[12] While the Fulcher law is useful in analyzing the freezing properties of real glasses, it remains to be seen whether this law has a physical interpretation with respect to the nature of the spin-glass freezing. At present, we see little to be gained by analyzing our data in terms of a Fulcher law.

Another interesting result from the frequency dependent χ' measurements is that the various $\chi'(\nu)$ curves remain separated for $T < T_f$ (see Fig. 5). This low-T-susceptibility separation for the different frequencies is still observed down to 4 K and is reversible with temperature. In Fig. 7 we illustrate this situation by plotting the normalized susceptibility as a function of frequency for several different temperatures. Note that there is no frequency dependence for $T > 9.60$ K [$T_f(\nu = 234$ Hz$) = 9.40$ K] and that the frequency dependence of χ' decreases for decreasing temperature below T_f.

We shall not discuss in any detail the susceptibility measurements on samples IId (2.0 at.% Mn) and IIe (6.3 at.% Mn) because the results are similar to those of sample IIc (0.94 at.% Mn) fully considered above. However, we must again mention that sample IId and IIe exhibit a more pronounced, but still rela-

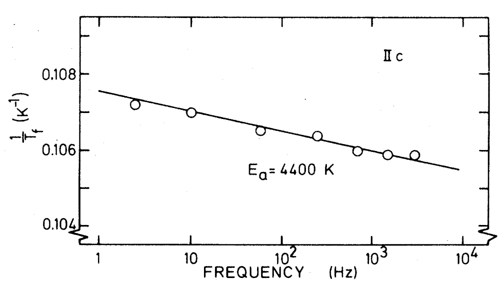

FIG. 6. The inverse freezing temperature T_f^{-1} as a function of the log of the frequency for sample IIc (Cu-0.94 at.% Mn). Drawn line is fitted to an Arrhenius law with $E_a = 4400$ K ($\nu_0 = 10^{200}$ Hz).

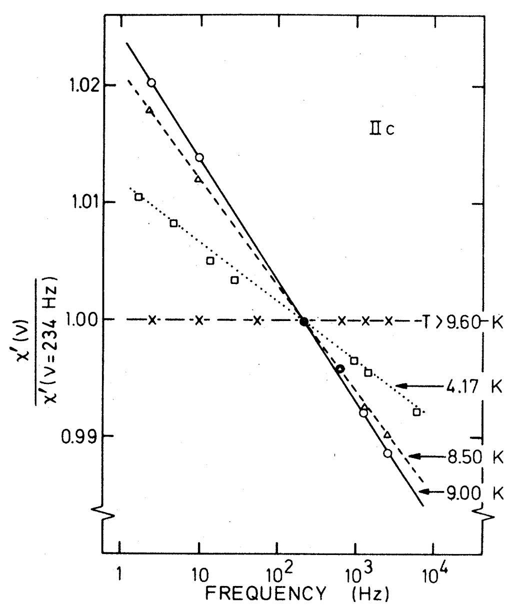

FIG. 7. The normalized susceptibility $\chi'(\nu)/\chi'(\nu = 234$ Hz) as a function of frequency at several temperatures for sample IIc (Cu-0.94 at.% Mn) $T_f = 9.40$ K. The drawn lines are only a guide to the eye.

tively small frequency dependence of the freezing temperature. For $\nu = 234$ Hz T_f (IId) = 15.50 K and T_f (IIe) = 32.3 K. In Fig. 8 for sample IId the normalized susceptibility is plotted as a function of frequency for several temperatures, again indicating that the various $\chi'(\nu)$ curves remain separated for temperatures below T_f. Essentially, the separation in frequency is reversible with temperature and frequency. If plotted as T_f^{-1} vs $\log_{10}\nu$, the activation energies E_a are 7350 K (sample IId) and 14 700 K (sample IIe), respectively. As with all quenched samples the sharpness of $\chi'(T)$ is strongly influenced by small applied H fields.

The behavior of the spin-glass susceptibility for $T >> T_f$ is an interesting question. This is especially so when a comparison is drawn between the predictions of the Edwards-Anderson model[31] and those which take into account the existence of short-range magnetic order far above T_f. As is indicated in Fig. 5, there is an apparent Curie-Weiss-like behavior. So let us now examine the inverse susceptibility χ'^{-1} over a wide temperature interval. Such a plot is made in Fig. 9 for samples IIc (0.94 at. % Mn) and IId (2.0 at. % Mn). Above T_f and below 50 K, the susceptibility obeys a simple Curie law with C (IIc) = 3.3 emu K/mole Mn and C (IId) = 3.6 emu K/mole Mn. The effective moments per Mn atom, $p_{\text{eff}} = (3kC/N\mu_B^2)^{1/2}$, are $5.2\mu_B$ and $5.4\mu_B$, respectively. However, in our experiments the data were taken up to $T = 150$ K. Above 50 K considerable deviations from the "simple" Curie law were observed. In the higher-temperature region from 100 to 150 K, χ'^{-1} was again linear in temperature, but now with a positive Curie-Weiss temperature Θ (IIc) = 5 ± 1 K and Θ (IId) = 10 ± 3 K, and C(IIc) = 3.1 emu K/mole Mn [p_{eff}(IIc) = $5.0\mu_B$] and C(IId) = 2.9 emu K/mole Mn [p_{eff}(IId) = 4.8 μ_B]. Our results at extended temperatures ($T_f \lesssim T \lesssim 150$ K) demonstrate that the simple Curie law found in Ref. 27 is an artifact of the limited low-temperature range of measurement. It should be mentioned here that for concentrations below about 1 at. % Mn the Curie-Weiss Θ determined by susceptibility measurements above ~50 K becomes negative.[32] The same change in the sign of Θ was reported earlier at similar Mn concentrations in *Ag*Mn and *Ag*SnMn.[33] We must conclude, in contrast with the Edwards-Anderson model, that short-range magnetic order far above T_f must be taken into account in the theoretical description.

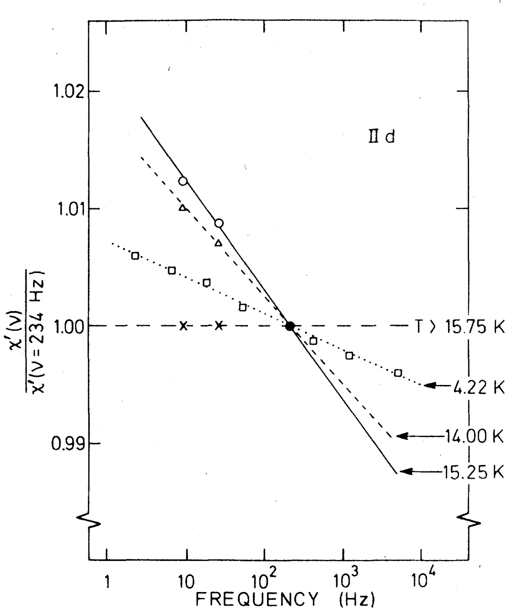

FIG. 8. The same as Fig. 7, but now for sample IId (Cu-2.0 at. % Mn) $T_f = 15.50$ K.

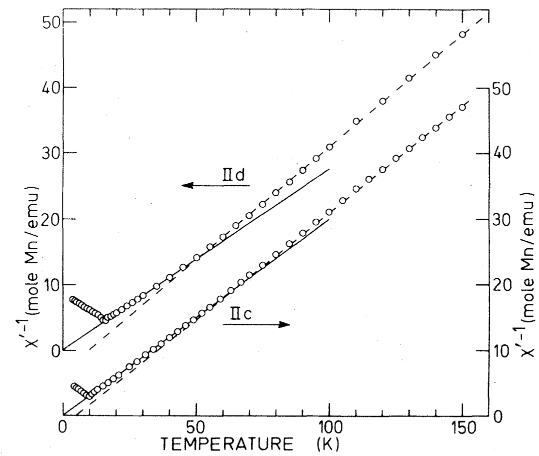

FIG. 9. The inverse susceptibility $1/\chi'$ as a function of temperature for samples IIc (Cu-0.94 at. % Mn) and IId (Cu-2.0 at. % Mn). Drawn lines: Curie-law fit for temperatures $T_f \lesssim T \lesssim 50$ K. Dashed lines: Curie-Weiss fit for temperatures $100 \lesssim T < 150$ K.

B. Field dependence of the ac susceptibility

A preliminary account of the field dependent susceptibility has already been given in the previous subsection. We now wish to consider in detail the effects of a homogeneous, external field H ($\vec{H} \parallel \vec{h}$) on the magnetic susceptibility.

As was mentioned earlier the two types of *Cu*Mn samples exhibit a different behavior with respect to small applied magnetic fields. The sharp susceptibility peaks of the type II (quenched) samples are rounded off in fields of only a few hundred oersteds (see also Fig. 1), while weaker effects were seen for the broad maxima in the susceptibility of type I (slow-cooled) samples. A comparison between the field dependences of the susceptibility at T_f for these two different types of CuMn alloys is presented in Fig. 10. Here $\chi'(T_f)/\chi'_0(T_f)$, for a frequency of 332 Hz, is plotted against the static, applied field where χ'_0 is the zero-field susceptibility. The initial decrease of $\chi'(T_f)$ is stronger for the type II samples. As the accuracy of determining the temperature of the maximum in χ' is smaller for rounded peaks, we never observed an H-field shift of the freezing temperature for both types of samples.

In Fig. 11 a comparison is made for the field dependence at T_f between the CuMn (0.57 at.% Mn) sample in the quenched and slow-cooled states. Now a $\log_{10}$-$\log_{10}$ plot is used with the normalized change in susceptibility $(\chi'_0-\chi')/\chi'_0$ versus the field up to 25 kOe (2.5 T). Once again the susceptibility of the sharp-cusp, quenched sample (squares) is more sensitive to the applied field than the broad peak, slow-cooled sample (circles). However, at field strengths approaching 25 kOe both curves merge together and here the zero-field susceptibility χ'_0 is suppressed by about 60%. The reduction at strong fields is in accordance with the results of Smit *et al.*[19] who measured magnetization curves of CuMn alloys with Mn concentrations between 1.5 and 10 at.% Mn at 4.2 K in fields up to 400 kOe (40 T), using a pulsed-field technique. In preparing the samples, Smit *et al.* employed a type I heat treatment (500 °C anneal, slow cooled). However, in high-field measurements there is little difference in magnetic behavior between the two types of sample (see Fig. 11).

At low field the slopes of the $\log_{10}$-$\log_{10}$ plots in Fig. 11 are 1.0 ± 0.2. For similar measurements on sample IId (2.0 at.% Mn), presented in Fig. 12, we find for the low-field slope at the freezing temperature 1.6 ± 0.3. Chalupa[34] has argued that the critical exponent δ for an Edwards-Anderson spin-glass can be extracted from the magnetic susceptibility in a weak uniform magnetic field. His result is $\chi(T_f,H) \propto H^{2/\delta}$. Since the susceptibility is usually depressed in an external field, we assume our normalized change in susceptibility $(\chi'_0-\chi')/\chi'_0$ to be proportional to $H^{2/\delta}$ at T_f. The average of the above quoted slopes is 1.3, which directly yields $\delta = 1.5$ (± 0.3). This value is considerably smaller than that found by Simpson,[15] $\delta = 2.9 \pm 0.4$ for Cu-10 at.% Al–1 at.% Mn in fields up to 150 Oe. The pos-

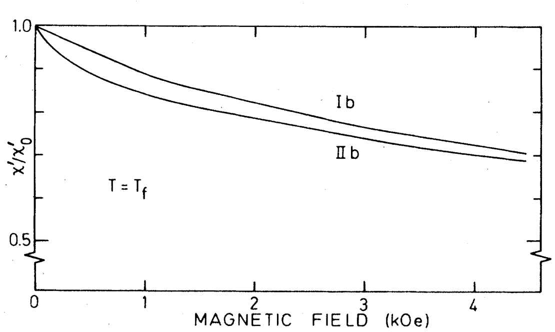

FIG. 10. The normalized susceptibility χ'/χ'_0 (χ'_0 is the zero field susceptibility) as a function of external, static magnetic field at $T = T_f$ for samples Ib (Cu-0.46 at.% Mn) $T_f = 5.00$ K and IIb (Cu-0.70 at.% Mn) $T_f = 7.65$ K.

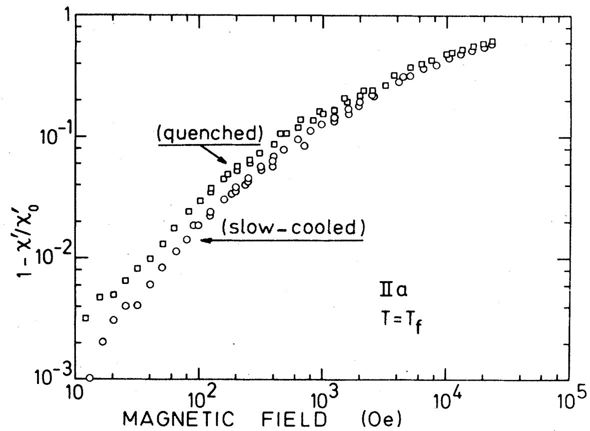

FIG. 11. The normalized change in susceptibility $(\chi'_0-\chi')/\chi'_0$ as a function of magnetic field at $T_f = 6.00$ K for a differently heat treated CuMn IIa sample (0.57 at.%): □ quenched, sharp cusp; and ○ slow-cooled, broad peak.

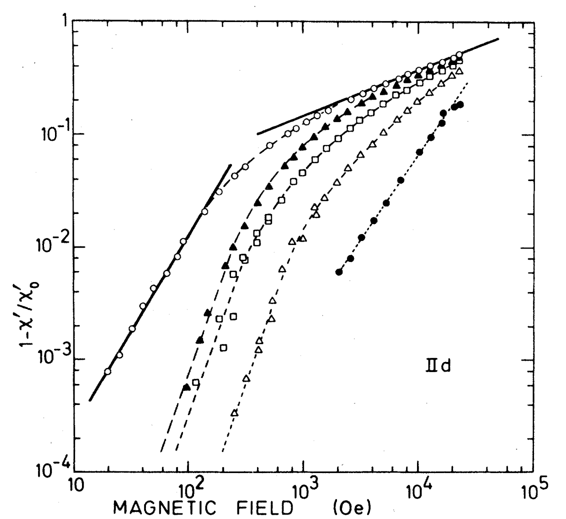

FIG. 12. The normalized change in susceptibility $(\chi'_0-\chi')/\chi'_0$ as a function of magnetic field at several temperatures for sample IId (Cu-2.0 at.% Mn). Temperatures: ● 31 K$=2\,T_f$, ▲ 17 K, ○ 15.5 K$=T_f$, □ 14 K, Δ 12 K. Drawn lines: fits to $\chi'(T_f,H) \propto H^{2/\delta}$ as described in the text. Dashed and dotted lines are for visual aid only.

sible cause for this discrepancy may be associated with the different sample preparation techniques, especially the inclusion of 10 at.% Al in the sample of Simpson in order to sharpen the $\chi(T)$ cusp.[15] If we follow the suggestion of Chalupa and make use of the scaling law $\alpha = 2 - \beta(1+\delta)$, where α and β are the specific heat and order parameter critical exponents, a value of $\alpha = -0.5$ is obtained in the low-field limit (β had been previously found to be ≈ 1). This -0.5 value for α is larger than those suggested theoretically ($\alpha < -1$) (Ref. 35) and those indicated by indirect experiment ($\alpha = -2$).[36] We believe that the above results support the conclusion that the application of critical exponents and scaling laws to the spin-glass freezing has little validity.

Recently Parisi has developed a new mean-field theory of spin-glasses.[37] At the critical temperature he predicts the behavior of the magnetic susceptibility to be initially proportional to $H^{1.33}$, with the power of H *increasing* at larger fields. Our value for the low-field exponent is in agreement with this result. However, the experiments show a clear *decrease* in the power of H at larger fields.

The field dependence of the susceptibility for sample IId is given in Fig. 12 not only for $T_f(=15.5\text{ K})$ but also for other temperatures. For this *Cu*Mn (2 at.% Mn) sample there is a dramatic change of behavior as T_f is reached from above. At high temperatures ($T = 31\text{ K} = 2T_f$; closed circles) χ' is roughly independent of field below fields of about 500 Oe; this is followed by a strongly decreased susceptibility in larger fields. As the freezing temperature is approached ($T = 17$ K; closed triangles) there is always a field dependent susceptibility which is strongest for $T = T_f$ (open circles). For $T < T_f$ (14 K, open squares; 12 K, open triangles) the suppression of χ' becomes weaker, i.e., much larger fields are required to reduce the susceptibility by a given amount. Once the field dependence at low fields sets in, there is at the highest fields a crossover to a weaker field dependence and all $\chi'(H)$ curves ($T = 17$ to 12 K) seem to merge into one curve with decreased dependence on the field. The data in Fig. 12 show that maximum sensitivity to external field is found at temperatures around the freezing temperature. In addition $\chi'(H)$ at T_f does seem to possess a special character (or symmetry point) with its strongest sensitivity to the applied field as compared to $\chi'(H)$ curves just above and just below T_f, the latter $\chi'(H, T = 17\text{ K})$ and $\chi'(H, T = 14\text{ K})$, respectively, showing rather similar field dependences with each other.

C. Low temperature susceptibility and distribution of blocking temperatures model

In order to further analyze the behavior of the magnetic susceptibility we shall use the superparamagnetic blocking model as proposed by Wohlfarth.[23] This model enables one to calculate the temperature dependence of χ for a system of clusters or superparamagnetic particles. The susceptibility of each cluster obeys a simple Curie law above the blocking temperature, T_B, and is assumed to be zero below T_B. Using this approach the susceptibility can be calculated when the distribution of blocking temperatures is known

$$\chi'(T) = \frac{C}{T}\int_0^T f(T_B)\,dT_B \quad ,$$

where $C = \lim_{T\to\infty}(\chi' T)$ is the Curie constant, obtained from the high-temperature susceptibility and $f(T_B)$ represents the distribution of blocking temperatures. We may invert this equation in order to obtain $f(T_B)$ from our experimental χ' data

$$f(T_B) = \frac{1}{C}\frac{d}{dT}(\chi' T) \quad .$$

In the model the shape of the distribution function around the freezing temperature is directly related to the sharpness of the peak in the susceptibility.

From our experiments on *Cu*Mn (e.g., Fig. 9) it is found that at high temperatures a deviation from the apparent Curie law occurs. Therefore we use the C values obtained from our measurements in the temperature range $T_f \lesssim T \lesssim 3\,T_f$. The distribution functions for the two types of samples Ib and IIb are shown in Fig. 13. A reduced temperature scale T/T_f is used in this figure. These examples of $f(T_B)$ are representative for the different types (I or II) of samples and their characteristic χ' peaks. In comparing these samples, the quenched ones (II) exhibit a rather sharp, cooperative like transition at T_f. Thus while the method of Wohlfarth[23] is useful in comparing different samples, the resulting $f(T_B)$ is incom-

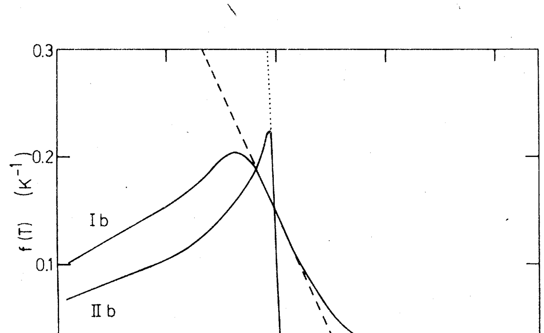

FIG. 13. The calculated distribution of blocking temperatures $f(T_B)$ as a function of reduced temperature T/T_f for samples Ib (Cu-0.46 at.% Mn) $T_f = 5.00$ K and IIb (Cu-0.70 at.% Mn) $T_f = 7.65$ K. The dashed and dotted lines represent the respective slopes $(T_f/C)(d^2\chi'/dT^2)_{T=T_f}$.

patible with the basic assumptions underlying his model. For, Wohlfarth[38] bases his model and defines his blocking temperature according to the Néel theory of superparamagnetism.[39] In this theory the blocking temperature is proportional to an energy barrier, and thermal activation (Arrhenius law) is responsible for the unfreezing (or unblocking) of the noninteracting superparamagnetic clusters. A random distribution of spins, such as is found in quenched *Cu*Mn alloys, would produce a random distribution of energy barriers and thus the $f(T_B)$ should be a smooth Gaussian-like function. A sharp steplike $f(T_B)$ requires interactions between the clusters and these cooperative effects have not been taken into account in the Néel theory.

As the peak in the measured susceptibility is not infinitely sharp, we can perform a simple check as to the internal reliability and self-consistency of the *analysis*. At T_f the slope of the distribution function should be proportional to the second derivative of $\chi'(T_f)$, or

$$\left.\frac{df(T)}{dT}\right|_{T=T_f}=\left.\frac{T_f}{C}\frac{d^2\chi'}{dT^2}\right|_{T=T_f} .$$

In Fig. 14 $d\chi'/dT$ is plotted as a function of reduced temperature. For both samples the straight (dashed or dotted) lines represent the second derivative of χ' at T_f. By means of the above formula these results for $d^2\chi'/dT^2|_{T=T_f}$ are used to calculate the slopes of the distribution function at T_f. The results of this calculation are consistent with the slopes of the distribution functions around T_f (see Fig. 13).

When observable, the effects of the frequency dependence of $\chi'(T)$ are also reflected in the distribution function. For sample IIc (0.94 at. % Mn) a part of the distribution function at temperatures around the susceptibility peak temperature, i.e., T_f, is given in Fig. 15 for three different frequencies. As one can see, the distribution function is shifted to higher temperatures without any essential change of shape.

The behavior of the low-temperature $T\rightarrow 0$ susceptibility is also predicted by the Wohlfarth model. In the low T limit the susceptibility may be expanded in a Taylor series

$$\chi'(T)=C[f(0)+\tfrac{1}{2}Tf^{(1)}(0)+\tfrac{1}{6}T^2f^{(2)}(0)+\cdots] ,$$

where $f^{(1)}(0)=df/dT|_{T=0}$ and $f^{(2)}(0)=d^2f/dT^2|_{T=0}$. In order to check this formula we extended our susceptibility measurements down to 0.4 K. Such measurements, presented for samples Ia and Ib in Fig. 16, clearly show a nonzero value for $\chi'(0)$, in agreement with the value of $\chi'(0)=Cf(0)$. A nonzero $\chi'(0)$ seems a rather strange result from the Wohlfarth model.[23] Since the susceptibility of all clusters is assumed to be zero below their blocking temperature, the model susceptibility should also be zero at $T=0$ K, in contrast with our measurements. The experimental variation of the susceptibility with temperature is essentially linear at low temperatures. This dependence is reflected in the experimentally derived distribution function (Fig. 13) which is typi-

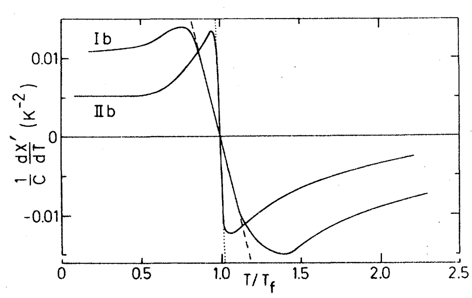

FIG. 14. The normalized first derivative $d\chi'/dT$ as a function of reduced temperature T/T_f for samples Ib (Cu-0.46 at. % Mn) $T_f=5.00$ K and IIb (Cu-0.70 at. % Mn) $T_f=7.65$ K. The dashed and dotted lines represent the second derivative of χ' at T_f: ---, $(T_f/C)\times(d^2\chi'/dT^2)_{T=T_f}=-0.088$ K^{-2}; and $\cdots$, $(T_f/C)\times(d^2\chi'/dT^2)_{T=T_f}=-0.73$ K^{-2}.

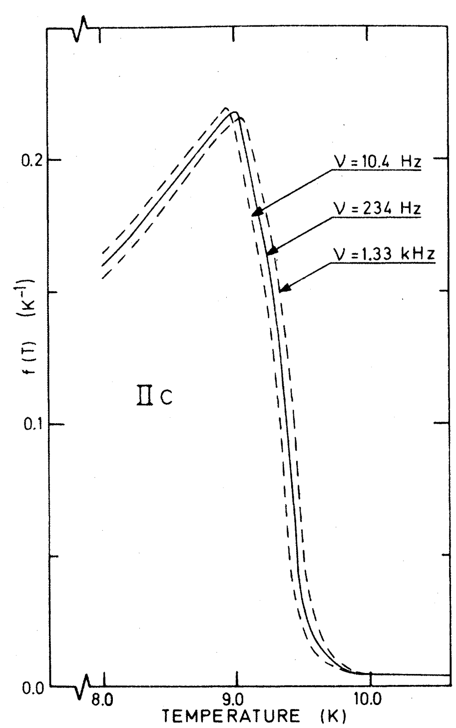

FIG. 15. The distribution function $f(T_B)$ as a function of temperature at several frequencies around the freezing temperature $T_f(\nu=234$ Hz$)=9.40$ K for sample IIc (Cu-0.94 at. % Mn).

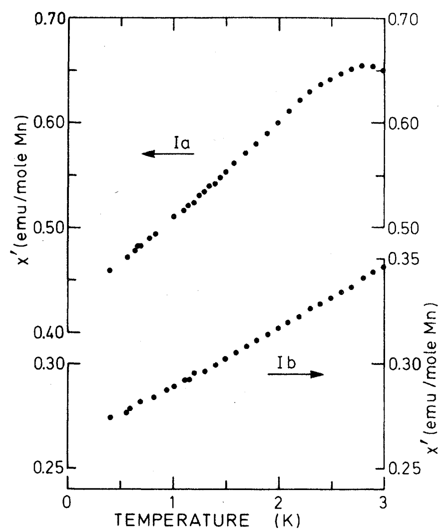

FIG. 16. The low-temperature susceptibility $\chi'(T)$ for samples Ia (Cu-0.23 at.% Mn) $T_f = 2.85$ K and Ib (Cu-0.46 at.% Mn) $T_f = 5.00$ K.

cally linear with temperature up to $\approx \frac{1}{2} T_f$, indicating a finite value for $f^{(1)}(0)$. However, this result has caused controversy over the third law of thermodynamics,[40] which would require [because of $(\partial\chi/\partial T)_{H,T\to 0} = 0$] a zero slope of the susceptibility versus temperature curve as $T \to 0$ K. In the model of Wohlfarth this "violation" of the third law is prevented only if the initial variation of $\chi'(T)$ at the lowest temperatures is parabolic. We observe a slight curvature at low temperatures which requires the T^2 term in the Taylor series. Still, within the limits of our experimental accuracy and of the 0.4 K minimum temperature, the low-temperature χ behavior can be reasonably well described by the first two terms of the Taylor expansion.

IV. DISCUSSION AND CONCLUSIONS

It has been shown in the experimental section that the sample preparation has a large influence on the shape and sharpness of $\chi'(T)$ near T_f. However, different sample preparations do not seem to affect the value of the freezing temperature T_f. For homogeneous, quenched *Cu*Mn alloys $\chi'(T)$ was found to exhibit a rather sharp peak thus giving a well-defined T_f (the uncertainty is $\delta T_f / T_f \approx 5 \times 10^{-3}$).

Regarding the form of the sample it is important to use fine powder specimens for these metallic spin-glasses, if the frequency dependence of $\chi(T)$ is to be determined. On bulk *Cu*Mn alloys, already at 100 Hz, the out-of-phase signal significantly exceeds the in-phase signal and causes a distortion in the $\chi(T)$ behavior. Consequently, frequency dependences of T_f measured on bulk alloys must be regarded with skepticism unless it can be shown that $\chi'' << \chi'$.

A small frequency dependence of $\chi'(T)$ is clearly seen for Mn-concentrations $c \gtrsim 1$ at.% with our measurement accuracy in the shape of these curves near T_f (see Fig. 5). The breakaway from pseudo-Curie behavior occurs at slightly higher temperatures as the frequency is increased. This is a very small effect and results in a frequency dependence of T_f which is only 0.5% per decade of frequency and is essentially independent of the Mn concentration.

The change in slope of the $\chi'(\omega)$ curves with temperature (see Figs. 7 and 8) is more important and appears to have a definite, characteristic form. At different frequencies the various $\chi'(T)$ curves converge at low temperatures. The behavior around T_f is reflected in the displacements of the distribution of blocking temperatures as is shown in Fig. 15. The attempt to describe such frequency shifts via an Arrhenius law of thermally activated diffusion produces unphysical parameters. This suggests that effects of a cooperative nature are present and that the freezing is more than a simple blocking process of individual clusters. Any theory which treats the spin-glass freezing must explain not only the sharpness of the susceptibility behavior, but also these subtle frequency effects. The present theories are insufficient and new efforts in the dynamical theory of critical phenomena are required.

Our measurements in Fig. 9 have revealed the nonapplicability of the Curie-Weiss law to spin-glasses just above the freezing temperature. It is only far above T_f (at ≈ 150 K or $T > 10T_f$) that a Curie-Weiss fit is appropriate with reasonable, paramagnetic effective moments and positive Curie-Weiss temperatures. This would indicate that a net ferromagnetic exchange is present in *Cu*Mn. Also we must point out that our results are in disagreement with the Edwards-Anderson theory of a simple $\chi(T) = (C/T)[1 - q(T)]$ where the order parameter $q(T) = 0$ for $T > T_f$. The wide temperature region deviation from such a $\chi(T) = C/T$ behavior demonstrates the evolution of clusters or groups of magnetically correlated spins which grow as the temperature is reduced and which cooperatively freeze at T_f. Here again the experimental results would require in the theory a dynamical process of cluster growth over a large temperature interval above T_f.

The field dependence of the differential susceptibility is stronger for the quenched samples than for the slow-cooled ones. So the sharper the peak in $\chi'(T)$ the more it is smeared in an external field. We have found no indication of shifts in the temperature where the maximum of χ' occurs, i.e., in T_f, with applying an external field. An attempt to treat $\chi'(H)$ at $T = T_f$ as a "critical isotherm" and to evaluate the critical exponent δ via the Edwards-Anderson model has given a rather small (≈ 1.5) value of δ. This, via a scaling law, results in a too large value of the specific-heat exponent α. Furthermore, there is a cross-over behavior at larger fields to a much larger value of δ. It is very difficult to interpret these field dependences within the standard critical phenomenon theory using the Edwards-Anderson model. Yet as can be seen in Fig. 12, there is something special about the $T = T_f$ behavior, for here the susceptibility is most sensitive to the external field.

The distribution of blocking temperatures model allows one to normalize the susceptibility and to compare many different samples and systems. This analysis has been proven self-consistent by comparing the slope in $f(T)$ at T_f with that calculated from the high-temperature experimental behavior. The resulting forms of the distribution function are a very sharp, step-like function for the quenched samples and a more rounded tail behavior for the slow-cooled ones. A sharp peak in $\chi'(T)$ requires from the analysis a step in $f(T)$ at T_f, see Fig. 13. This distinctive form of the distribution function indicates that the freezing is cooperative and is in contradiction with the assumption of superparamagnetic cluster blocking. For, $f(T)$ possesses a near vertical slope and peaks at T_f meaning that there is a sudden onset of blocked clusters. Independent, noninteracting clusters could produce no such distribution function as that determined from the measured susceptibility. A cooperative phase transition model based upon the percolation of clusters has been suggested by Mydosh[41] to describe the freezing process of a spin-glass. Figure 15 illustrates how the distribution function scales with increasing frequency. Only a very small shift to higher temperatures and depression at lower temperatures is needed to account for the frequency dependence of $\chi'(T)$. Further using the Wohlfarth analysis at the lowest temperatures ($T \gtrsim 0.4$ K) via a Taylor expansion has shown that the $\chi'(T) - \chi'(0)$ is mainly proportional to T with a small additional T^2 term giving a somewhat better fit. More accurate and still lower temperature measurements are needed to unambiguously determine the initial variation of $\chi'(T)$.

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