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NATURE OF THE HEAT CAPACITY ANOMALY IN $\underline{Cu}Mn$ at T_{sg}

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Precise measurements near T_{sg} show anomalies in the temperature and field derivatives of the heat capacity of $\underline{Cu}Mn$, but no discontinuities. Thermodynamics fails to predict the observed relationship between the temperature dependence of the susceptibility and the field dependence of the heat capacity.

The discovery[1] of a sharp cusp in the low-field ac susceptibility, χ_{ac}, at a characteristic temperature, T_{sg}, led to a renewal of both theoretical and experimental investigations of spin glasses, and prompted consideration of the possibility of a thermodynamic phase transition at T_{sg}. Mean field solutions of the Edwards and Anderson (EA) model[2] and extensions of it do produce a cusp in χ but they also predict a cusp in the magnetic specific heat, C. The EA theory stimulated a search for a sharp anomaly in C at T_{sg}. Wenger and Keesom[3] measurd both χ_{ac} and C on 1.2 at.% $\underline{Cu}Mn$ alloys but reported no evidence for any discontinuous behavior at T_{sg}. However, Martin[4] noted "knees" in plots of C/T vs T near the expected values of T_{sg} for .083 at.% $\underline{Cu}Mn$ and 1.0 at.% $\underline{Au}Fe$. For more concentrated $\underline{Cu}Mn$ samples the feature is less pronounced but a "similar correlation" was observed.[4]

We report here the results of an investigation of the specific heat of a $\underline{Cu}Mn$ spin glass in the vicinity of T_{sg} that characterizes more precisely the anomaly in C. Near and above T_{sg} essentially all of the data points for C fall within ±0.02% of smooth curves, an improvement of 1 to 2 orders of magnitude over earlier work. Heat capacity and χ_{ac} were measured on a well annealed polycrystalline sample with 0.28 at.% Mn. The χ_{ac} results, shown in Fig. 1, are very similar to those found by Cannela and Mydosh.[1].

A plot of C/T vs T (not shown) does not exhibit a discontinuity in slope of the type reported by Martin.[4] To examine the smoothness of the temperature derivative of C more carefully, we have calculated ΔC/T)/ΔT by taking point-to-point differences between the raw heat capacity data. Δ(C/T)/ΔT varies smoothly and regularly from 0.3 to 25K, the temperature range in which this sample was studied, except in a 4-K interval in the vicinity of T_{sg}. The data in this region are shown in Fig. 2 together with a solid curve that represents an interpolation from data below 2K to data above 7K, and is a possible smooth "background" curve. Figure 2 shows that the zero-field spin ordering in the vicinity of T_{sg} is energetically different from that over the broader range of temperature. However, the anomaly is spread over a temperature interval of the order of T_{sg} itself, in contrast with the predictions of EA-type theories and with qualitative, but unfounded, conjecture based on the sharpness of the susceptibility cusp.

Thermodynamics requires only that an anomaly in χ be reflected in the field dependence of C:

$$\left(\frac{\partial^2 M}{\partial T^2}\right)_H = \frac{1}{T}\left(\frac{\partial C}{\partial H}\right)_T, \tag{1}$$

where M is the magnetization, M=χH. Thus, the strong curvature of χ as a function of temperature that occurs near T_{sg} should be reflected in a strong field dependence of C. Measurements of C in fields from 0 to 1000 Oe were made between 2.5 and 6.5K. The field was always changed at T≈10K and held constant until a complete set of data

in that field was taken. Repeat runs in several fields, in some cases following runs in other fields, showed no evidence of irreproducability. Since M must be an odd function of H, Eq. (1) requires C to be an even function of H. Only the first two terms of a series expansion, $C/T = A + BH^2$, were useful in fitting the data. The coefficient B(T) shows a relatively sharp dip, the minimum value occurring at a temperature that is equal to T_{sg} to within experimental error.

Low-frequency measurements at fields of a few Oe give χ_{ac} as shown in Fig. 1 but dc field-cooled (fc) measurements of M,[5,6] made by cooling from above T_{sg} in fixed fields of a few hundred Oe or less, give values of χ_{fc} corresponding to curve (a) of Fig. 1. No time dependence of M has been reported in fc measurements. When M is measured by changing the applied field at temperatures below T_{sg} time effects can be observed and apparent values of χ are intermediate between χ_{ac} and χ_{fc} depending on the time and sample.[6,7] This picture suggests that the fc data, which are obtained in the same way as the C data are the ones that should be expected to correspond to thermodynamic equilibrium and that should be used in Eq. (1).

The failure of Eq. (1) to predict correctly the relation between C and χ is evident in several ways: (1) Since C is a parabolic function of H, $\partial^2\chi/\partial T^2$ should be independent of field. In fact, the curvature of both χ_{fc} and χ_{ac} is changed markedly by fields of a few hundred Oe in the vicinity of T_{sg}. (2) the measured change in slope of χ_{fc} in the vicinity of T_{sg} is twice as great as that calculated from the calorimetric data and Eq. (1). This is illustrated in Fig. 1 by curve (b) which represents $\chi(T)$ calculated from the calorimetric data by a double integration of Eq. (1) from 4.25K.

Recent suggestions[8] that certain disordered systems including spin glasses are inherently "non-ergodic" in nature offer a possible explanation of the failure of the thermodynamic relation Eq. (1). According to this suggestion the configuration space of a spin glass contains many potential valleys separated by barriers, with distributions of minimum energies of the valleys and of barrier heights. When a sample is cooled sufficiently far below T_{sg} it is frozen into a particular potential valley and does not sample microscopic states that are energetically allowed but rendered inaccessible by the intervening barriers. It is reasonable to assume further that cooling in different fixed fields leaves a sample trapped in different regions of configuration space. If the number of possible regions is sufficiently high, the properties of the microscopic states associated with them could still vary essentially continuously as a function of the external field that selects them, but not in the way determined by the local properties of the phase space eg., at the bottoms of the valleys. Under these conditions thermodynamic equalities such as Eq. (1) could break down even though M and C are measured under identical conditions and are "equilibrium" values in the sense of being independent of time. The failure of thermodynamic relations would then arise as a fundamental consequence of the disorder of the system.

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FIGURE CAPTIONS

Figure 1: χ vs T for CuMn. Data points are the χ_{ac} measurements. Curve (a) represents typical χ_{fc} data and curve (b) is derived from C/T vs H data. (XBL 8012-13569)

Figure 2: Derivative of C/T with respect to T in zero field and 1000 Oe. The error bar represents a shift of 0.10% in one heat capacity point. The smooth curve is explained in the text. (XBL 8012-13572A)
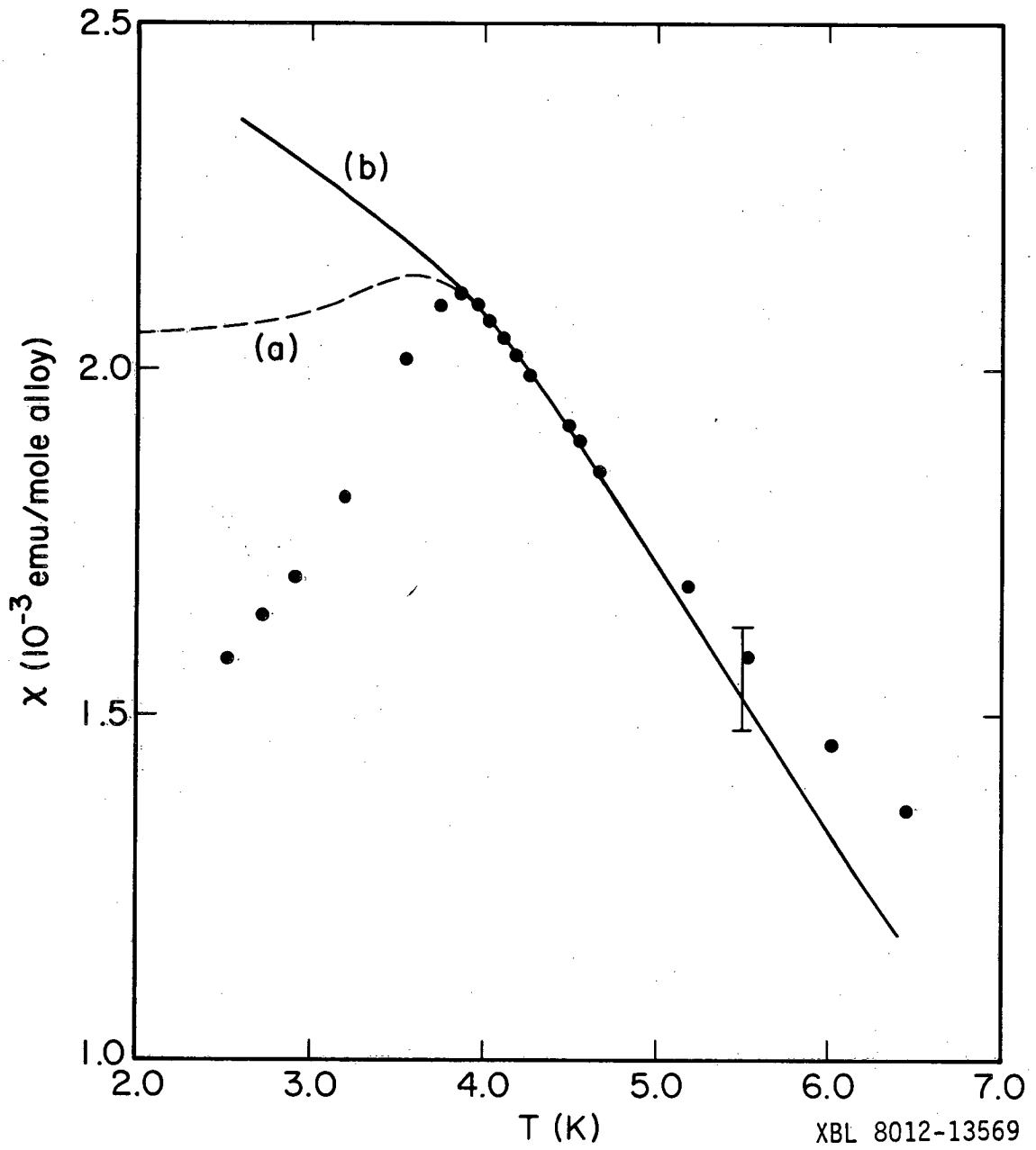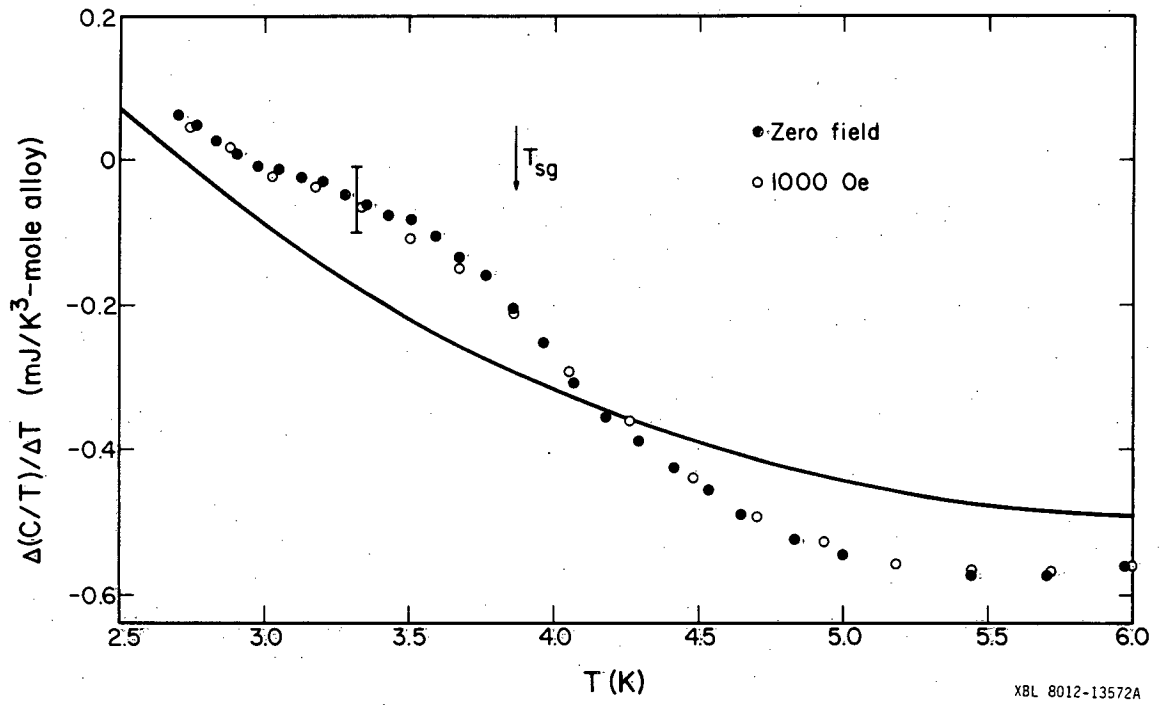
Figure 1

XBL 8012-13569

Figure 2

XBL 8012-13572A