

Specific heat of spin-glass CuMn below 3 K

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Measurements in the 0.35 to 3 K temperature range on alloys containing 0.083, 0.20, 0.43, 0.88, and 0.92 at. % Mn are reported. The spin-glass contribution to the specific heat does not show a linear dependence on absolute temperature but has an initial positive curvature leading to a linear region which extrapolates back to zero at a positive temperature, dependent on the Mn content. A change in $d(C_p/T)/dT$ at the estimated spin-glass freezing temperature was observed for the 0.083 at. % Mn alloy. The estimated freezing temperatures for the other alloys lie outside the range of the present measurements. The results do not obey the simple scaling law $C_p/c = f(T/c)$, where c is the Mn concentration. However, they are in agreement with earlier specific-heat results below 1 K and recent data above 2 K and also with some recent theory.

I. INTRODUCTION

There is currently considerable interest in the so-called spin-glass systems. These are dilute magnetic systems in which the magnetic moments interact via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism. At low temperatures the moments are frozen into thermal equilibrium orientations but with no long-range order.¹ Moments in positions of low field may be thermally excited. A possible complication is the Kondo or single-impurity effect² which also occurs in such systems and is a correlation of conduction electron and impurity spins so that the effective spin of the impurity becomes zero. In order to investigate pure spin-glass effects it is therefore necessary to choose a system with a very low Kondo characteristic temperature so that Kondo effects do not occur within the measurement range. A suitable system is CuMn for which the Kondo temperature is² about 0.001 K.

The present work was undertaken because of uncertainty regarding the spin-glass contribution to specific heat at low temperatures. This can be calculated from the various theories and the experimental result is an important factor in assessing the validity of these theories. The earliest specific-heat measurements^{3,4} on dilute CuMn alloys, extending down to about 1.5 K, had suggested that, in the low-temperature limit, the spin-glass contribution was approximately linear in absolute temperature and independent of the composition of the sample. The Marshall-Klein-Brout^{5,6} theories of spin-spin interaction via the RKKY mechanism were able to explain this result. When the specific-heat measurements were extended below 1 K a large nuclear contribution to the specific heat was observed.⁷ After this was subtracted, it appeared that the previously observed linear dependence on temperature no longer held

but, in the new low-temperature limit, there was another region of linear dependence with slope about half the magnitude of that previously found. Later measurements below 1 K by Ho⁸ cast some doubt on the accuracy of the earlier measurements⁷ but a similar result was obtained. However, Ho's results were only published indirectly by being summarized in a review paper by Phillips.⁹ More recently, measurements by Wenger and Keesom,¹⁰ extending down to 2 K, suggested a linear region in the spin-glass specific heat which extrapolated to zero at a positive temperature.

The present results, extending down to about 0.35 K, confirm the Wenger and Keesom observation¹⁰ but show that the specific heat deviates from the linear extrapolation to approach the absolute zero with a much lower slope, in general agreement with the Ho result.⁸ A brief report¹¹ of this finding for one of the present alloys was given at a recent conference. At the same conference Fogle, Ho, and Phillips¹² reported some new specific-heat results for the CuMn system which appear to be in substantial agreement with the present work.

II. EXPERIMENTAL

The alloys were prepared from American Smelting and Refining Co. (ASARCO) 99.999% pure copper and Johnson-Matthey spectroscopically pure manganese which were induction melted in an alumina crucible and then chill cast using an apparatus described briefly elsewhere.¹³ (There are no solubility problems in the CuMn system,¹⁴ the chill casting was to preserve homogeneity.) One alloy, finally containing 0.92 at. % Mn, was melted in a helium atmosphere to minimize evaporation. After melting, some residue was found in the crucible suggesting

possible oxidation. The procedure was therefore changed for the other alloys as follows: (a) the apparatus was thoroughly degassed by heating under vacuum before loading the alloy components and (b) melting was done under an atmosphere of flowing hydrogen gas which had been cleaned and dried. It was necessary to pump out the hydrogen before casting, otherwise the resulting sample had a foamlike texture caused by the dissolved hydrogen coming out of solution on solidification. In all cases the alloy was held for about 20 min in the molten state to ensure thorough mixing, stirring being supplied by the induction heating.

After casting, a flat was turned on one end of each alloy. This was for thermal contact with the calorimeter but was done at this time to remove any adhesions with the water-cooled hearth. On two alloys some porous areas were cut or turned off at this time. After acid cleaning and washing each alloy was placed in a cleaned aluminum crucible and sealed up in a quartz tube. For the alloy melted in a helium atmosphere the tube was evacuated while for all the other alloys it contained about one third of an atmosphere of hydrogen. Each alloy was then homogenized for 30 d at 1000 °C by placing the sealed-off tube in a furnace. Finally, each alloy was degassed by heating under vacuum to about 900 °C (when evaporation became apparent), then holding at about 700 °C overnight and then slowly cooling (still under vacuum) to room temperature. Pieces were then cut off the top and bottom of each alloy for analysis. After another cleaning the alloy was sealed under vacuum in a Pyrex glass tube unless measurements were to be made immediately.

The analysis results are summarized in Table I. The two entries for each alloy refer to the two ends of that alloy. The Mn content was determined by a flame atomic absorption method after plating the copper out of a solution of the alloy. The impurities were determined by a dc arc emission spectrographic method and the accuracy is within a factor of 3 of the stated values. The first two alloys were both nominally 1 at. % Mn and the others were 0.5, 0.25, and 0.1 at. % Mn, respectively. The manganese loss, presumably by evaporation, during fabrication of these alloys is similar to that reported elsewhere. (Nominal 1 at. % Mn found¹⁵ to be 0.901 and¹⁶ 0.900.) To check the reported difference in Mn content of the first two alloys in Table I, further samples from one end of each were sent to the analysts. The results are shown in parentheses in Table I and confirm the results of the first analysis.

The specific-heat measurements were made in apparatus described elsewhere.¹⁷

III. RESULTS

Two runs were made on each alloy with an intermediate warm to room temperature. There was some doubt regarding the best analytic expression to represent the specific heat. The electronic plus lattice specific heats can be represented by a polynomial in odd power of temperature (T). For the more concentrated alloys there appeared to be a nuclear term represented by a $(1/T^2)$ term. The form of the spin-glass contribution is less certain. Phillips⁹ (following Ho⁸) suggests that the leading terms are proportional

TABLE I. Chemical analysis of samples. All parts per million wt/wt.

Mn	Fe	Ag	Mg	Si	Average Mn content Atomic %	Average atomic weight	Sample weight g
8000 ± 80 (7900)	1 (0.5)	0.5 (1.5)	0.05 (0.3)	5 (9)	0.92	63.46	69.18
7900 ± 80	0.3	0.2	0.05	1			
7600 ± 76 (7700)	0.5 (0.5)	... (...)	0.05 (0.3)	2 (4.5)	0.88	63.46	74.90
7600 ± 76	0.5	...	0.05	1			
3700 ± 37	0.5	...	0.6	0.9	0.43	63.50	86.23
3700 ± 37	0.5	...	1.0	1.5			
1700 ± 17	0.5	1.5	0.3	0.9	0.20	63.52	84.66
1700 ± 17	1.0	1.5	0.3	1.5			
710 ± 35	0.6	8	0.083	63.53	83.75
720 ± 35	0.6	8			

to T and T^2 . Least-squares fits of various expressions were therefore tried for each alloy with the number of terms being selected to minimize the standard deviation. The coefficients of the selected polynomials are given in Table II and the deviation of the measured points from these fits is shown in Fig. 1.

For the 0.92 at. % Mn alloy it was clear that the nuclear term was necessary. The 0.88 at. % Mn-alloy results have a worse scatter (probably caused by ambient electrical noise during the period of the measurements) and the best fit was obtained with a ten term power series in T . However, the selected poly-

nomial (Table II), which includes the nuclear term, has only a slightly worse (0.02%) standard deviation with four fewer terms. For neither of these alloys do the present results show the need for a term in T^2 . A nuclear term is also clearly needed for the 0.43 at. % Mn alloy and the expression with both even and odd powers of T gave the best fit. The best fit for the 0.20 at. % Mn alloy gave a negative coefficient for the "nuclear" term and was therefore rejected in favor of the expression in Table II which has a slightly worse (0.003%) standard deviation. The 0.083 at. % Mn alloy presumably needs the long power-series ex-

TABLE II. Polynomial coefficients representing specific heat $C_p = \sum a_n T^n$. Units cal/Kg atom (1 cal = 4.186 J). Error limits are 95% confidence limits for each coefficient from the statistical analysis. Each polynomial reproduces the smoothed specific heat to within 0.1%.

Sample	Coefficients	Symbol in figures
Cu-0.92 at. % Mn	$a_{-2} = +(0.123\ 63 \pm 0.004\ 4) \times 10^{-4}$ $a_1 = +(0.674\ 08 \pm 0.007\ 4) \times 10^{-3}$ $a_3 = +(0.316\ 55 \pm 0.016) \times 10^{-3}$ $a_5 = -(0.931\ 72 \pm 0.12) \times 10^{-4}$ $a_7 = +(0.172\ 15 \pm 0.035) \times 10^{-4}$ $a_9 = -(0.161\ 65 \pm 0.045) \times 10^{-5}$ $a_{11} = +(0.594\ 26 \pm 0.21) \times 10^{-7}$	■ ◆
Cu-0.88 at. % Mn	$a_{-2} = +(0.112\ 5 \pm 0.004\ 4) \times 10^{-4}$ $a_1 = +(0.688\ 4 \pm 0.006\ 8) \times 10^{-3}$ $a_3 = +(0.280\ 2 \pm 0.012) \times 10^{-3}$ $a_5 = -(0.646\ 3 \pm 0.061) \times 10^{-4}$ $a_7 = +(0.811\ 5 \pm 0.11) \times 10^{-5}$ $a_9 = -(0.382\ 9 \pm 0.065) \times 10^{-6}$	□ ◇
Cu-0.43 at. % Mn	$a_{-2} = +(0.424\ 3 \pm 0.10) \times 10^{-5}$ $a_1 = +(0.699\ 6 \pm 0.085) \times 10^{-3}$ $a_2 = -(0.130\ 4 \pm 0.30) \times 10^{-3}$ $a_3 = +(0.810\ 2 \pm 0.40) \times 10^{-3}$ $a_4 = -(0.600\ 9 \pm 0.26) \times 10^{-3}$ $a_5 = +(0.185\ 8 \pm 0.079) \times 10^{-3}$ $a_6 = -(0.210\ 1 \pm 0.091) \times 10^{-4}$	▷ ◁
Cu-0.20 at. % Mn	$a_1 = +(0.768\ 91 \pm 0.003\ 0) \times 10^{-3}$ $a_3 = +(0.348\ 64 \pm 0.013) \times 10^{-3}$ $a_5 = -(0.181\ 49 \pm 0.015) \times 10^{-3}$ $a_7 = +(0.538\ 56 \pm 0.074) \times 10^{-4}$ $a_9 = -(0.888\ 42 \pm 0.17) \times 10^{-5}$ $a_{11} = +(0.761\ 68 \pm 0.18) \times 10^{-6}$ $a_{13} = -(0.264\ 38 \pm 0.074) \times 10^{-7}$	▽ △
Cu-0.083 at. % Mn	$a_1 = +(0.503\ 074 \pm 0.24) \times 10^{-3}$ $a_2 = +(0.173\ 950 \pm 0.18) \times 10^{-2}$ $a_3 = -(0.239\ 057 \pm 0.52) \times 10^{-2}$ $a_4 = +(0.057\ 697\ 7 \pm 0.81) \times 10^{-2}$ $a_5 = +(0.188\ 043 \pm 0.73) \times 10^{-2}$ $a_6 = -(0.217\ 434 \pm 0.40) \times 10^{-2}$ $a_7 = +(0.101\ 562 \pm 0.13) \times 10^{-2}$ $a_8 = -(0.223\ 604 \pm 0.23) \times 10^{-3}$ $a_9 = +(0.191\ 355 \pm 0.17) \times 10^{-4}$	+ ×

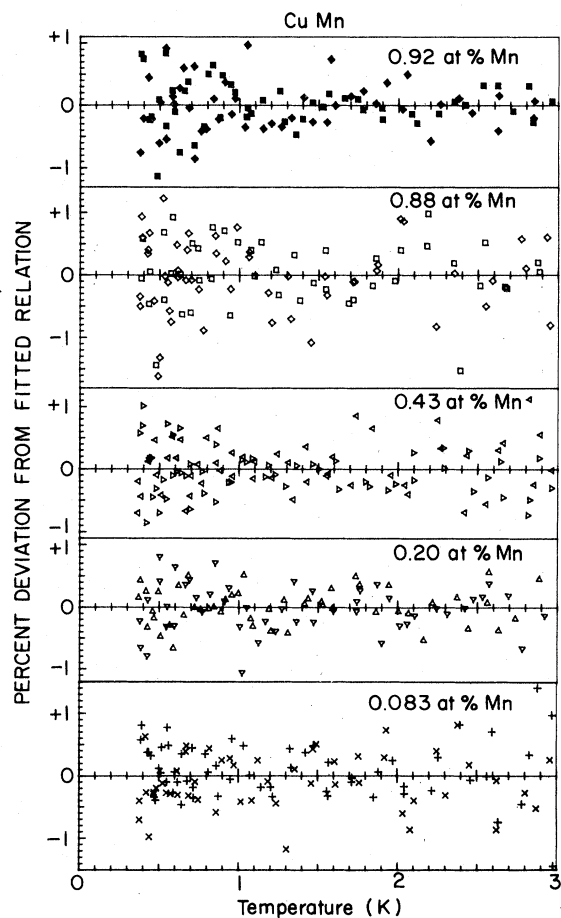


FIG. 1. Percentage deviation of the raw specific-heat data for each sample from the fitted relation for that sample given in Table II.

pression (Table II) because the result indicates much more curvature at higher temperatures than for the other alloys. The inclusion of a nuclear term gives a slightly worse fit.

IV. DISCUSSION

First the results for the two nominally 1 at. % Mn alloys, melted in helium and hydrogen respectively, will be compared. Figure 2 is a plot of the deviations of the raw data for the 0.92 at. % Mn (melted in helium) alloy from the fit (Table II) for the 0.88 at. % Mn (melted in hydrogen) alloy. Any difference is clearly very small. (The large deviation for the highest temperature point suggests an "end-effect" error in the fit of the 0.88 at. % Mn alloy.) The differ-

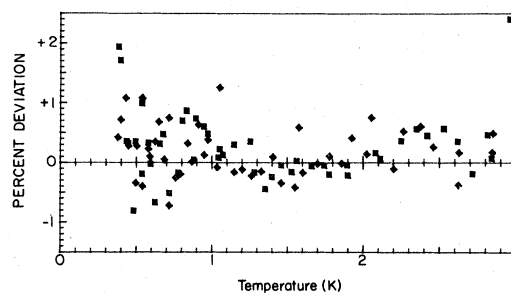


FIG. 2. Percentage deviation of the raw specific-heat data for the 0.92 at. % Mn sample from the fit for the 0.88 at. % Mn sample (Table II).

ence in Mn content (Table I), confirmed by a second analysis (value in parentheses, Table I), is about 0.04 at. %. Referring to Fig. 3, it may be deduced that such a composition difference would result in a specific-heat change of a few tenths of a percent at 3 K. This is as seen in Fig. 2 and it may therefore be concluded that there is no significant difference in alloys made by the two methods. Since the composition of these alloys is so similar, only the results for the 0.88 at. % Mn alloy will be considered in later discussion.

In order to discuss the spin-glass (or magnetic) contribution to specific heat it is necessary to subtract the lattice, electronic, and nuclear contributions. It will be assumed that the first two contributions are exactly the same as those for pure copper for which the results¹⁷ obtained previously with the present apparatus are used. (Alloys of copper with nonmagnetic elements at the 1 at. % level generally show a very small increase of electronic specific heat.¹⁸ The elastic constants of dilute alloys of Mn in Cu have been measured at low temperatures.¹⁹ The change of lattice specific heat on alloying may be calculated and the largest effect, for the most concentrated alloy of the present work, is less than 0.2% of the total specific heat of copper or less than 0.05% of the spin-glass contribution to the specific heat of that alloy.) The nuclear specific heat of dilute alloys of Mn in Cu has been determined elsewhere from specific-heat measurements extending to much lower temperatures than the present work.^{12,15} The value obtained (directly proportional to the Mn content) is consistent with the nuclear orientation measurements of Campbell, Compton, Williams, and Wilson²⁰ and within 2% of the value found in the present work for the 0.88 at. % Mn alloy. The latter value has therefore been taken as the basis for calculating the nuclear specific-heat term for the more dilute alloys.

The smoothed values of the spin-glass specific heat

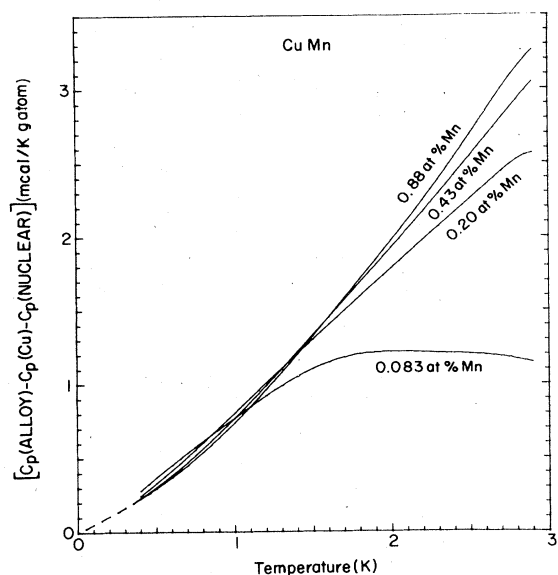


FIG. 3. Smoothed results for the "spin-glass" contribution to specific heat.

thus derived from the present measurements are shown in Fig. 3. (Smoothed values are plotted because the raw data obscures the result on this plot.) The dashed line shows the extrapolation to zero calculated from the coefficients (Table II) for the 0.88 at. % Mn alloy. From this plot it is clear that the spin-glass specific heat is not linearly proportional to temperature over the whole range of the present measurements, nor is it concentration independent. The form for the two more concentrated alloys is made clearer in Fig. 4. There is a linear region at "high" temperatures which extrapolates to zero at a positive temperature (~ 0.6 K for the 0.88 at. % Mn alloy and ~ 0.4 K for the 0.43 at. % Mn alloy). The specific-heat data diverges from this extrapolation to approach zero temperature with a smaller slope (after subtraction of the nuclear specific-heat contribution). The observed linear regions and extrapolated intercepts are consistent with the above 2 K data on more concentrated alloys by Wenger and Keesom.¹⁰ The intercept temperature varies roughly as $c^{1/2}$ where c is the Mn concentration of the alloy. Thus the more dilute the alloy the more the specific heat appears to vary linearly with absolute temperature.

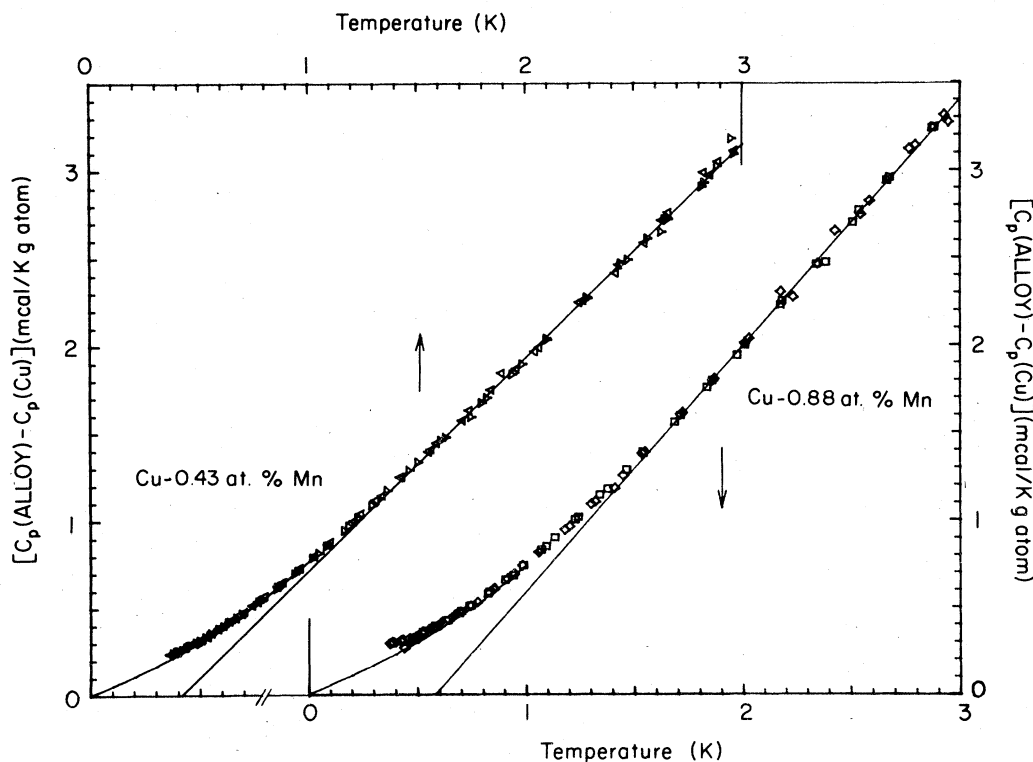


FIG. 4. Specific-heat plots for the two more concentrated alloys. For clarity, the temperature scales are staggered by 1 K. Note the marked linear region of the specific heat and the extrapolation to zero temperature after subtraction of the nuclear specific heat.

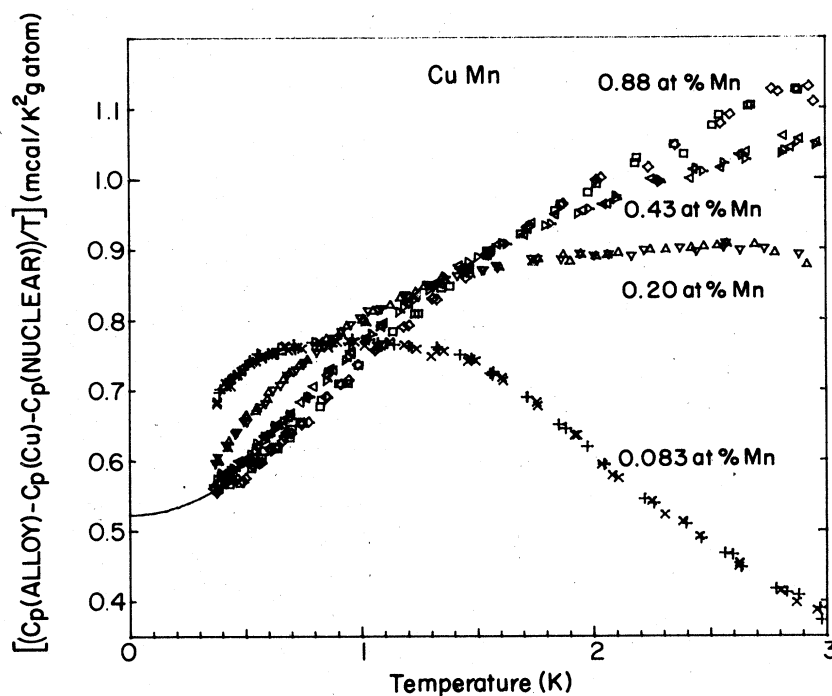


FIG. 5. Plot of (spin-glass specific heat divided by temperature) against temperature. The extrapolation to zero temperature is for the most concentrated alloy using the curve fit coefficients from Table II.

The most dilute alloy shows a broad maximum in the spin-glass specific heat at about 1.8 K (Fig. 3). (A maximum in the electrical resistance occurs at about 2.8 K for an alloy of this composition.¹⁶) In agreement with previous specific-heat results¹⁰ there is no sign of a cusp at the maximum. A cusp is found in the low-field magnetic susceptibility²¹ at a slightly lower temperature^{10,21} and has been taken as the magnetic freezing or ordering temperature (T_f). (One of the recent preoccupations of theoreticians has been to reconcile the cusp in susceptibility with the noncusp in specific heat.¹ The magnetic cluster model²² is a possibility but arguments against this approach have been advanced and an alternative model proposed involving short-range antiferromagnetic coupling above T_f gradually decreasing to zero at a temperature T_{ESR} which is of the order $2T_f$.²³) Taking the present result for the 0.083 at. % Mn alloy with those of Wenger and Keesom¹⁰ for more concentrated alloys, it is seen that the temperature of the maximum of the spin-glass specific heat is roughly proportional to the Mn content of the alloy.

The form of the present results at the lowest temperatures is more clearly seen in the plot of spin-glass specific heat divided by temperature (Fig. 5) where the extrapolation to zero temperature is from the Table II coefficients for the 0.88 at. % Mn alloy.

It is not clear from the plot whether all compositions would extrapolate to the same point. Obviously, measurements to lower temperatures are necessary to determine this. Such measurements have been reported by Fogle *et al.*¹² who find that four of their samples extrapolate to a point about 20% below that shown in Fig. 5 while the fifth sample is only 5% below. Fogle *et al.*¹² find that in the low-temperature limit the spin-glass specific heat is of the form $AT + BT^2$. This is a straight line on Fig. 5. Extrapolation of the present measured data in this way does lead to better agreement with the result of Fogle *et al.*¹² However, such a result cannot be obtained from the present data considered in isolation. (Showing that extrapolation is no substitute for measurement.)

Another interesting feature seen in Fig. 5 is the "knee" in the curve for the 0.083 at. % Mn alloy at about 1.5 K. This is close to the spin-glass freezing temperature for this composition.²³ Results for other Mn concentrations would be required to see whether this is a real effect or just a fortunate coincidence. (The spin-glass freezing temperatures for the other alloys studied here are above the upper temperature range of the present measurements.) The results of Fogle *et al.*¹² for a 0.0231 at. % Mn alloy show a similar "knee" at about 0.28 K. However this is signifi-

cantly lower than the spin-glass freezing temperature²³ for this composition (~ 0.50 K). The possibility of an error in composition or spin-glass freezing temperature estimate must be considered. Following a reanalysis of early specific-heat data, Mydosh²⁴ reports that "at best" there is "some" correlation between a knee or maximum in C_m/T and the spin-glass freezing temperature.

The general form of the results found here also appears to occur in at least some other spin-glass systems. Thus recent data on dilute PtMn alloys²⁵ shows similar results in the 1 to 2.6 at.% Mn range while the present author's data²⁶ for a 0.2 at.% Mn in Zn alloy fits into the same picture. It is therefore probable that the form of the results of the present work are quite general for spin glasses and that more accurate and extensive measurements on other systems will lead to similar results.

Finally, a comparison of the present results with theory will be made. As mentioned earlier, the Marshall-Klein-Brout theories^{5,6} predicted a spin-

glass specific heat which was linear in temperature over a temperature range of several degrees from the absolute zero and was also independent of the Mn content for dilute alloys. Klein and his collaborators have made many refinements to this theory. Most recently, Klein²⁷ has shown that the coefficient of the linear term in the spin-glass specific heat should increase slightly with increasing Mn content. Clearly, this is not in accord with the present results. However, a computer model calculation by Walker and Walstedt,²⁸ avoiding some of the assumptions implicit in the above theory^{6,27} but using the same RKKY spin-coupling model, appears to be in excellent agreement with the more concentrated alloy results of the present work. (The effective field distributions obtained in Refs. 6 and 28 are quite different.) Walker and Walstedt²⁸ compared their result with the Wenger and Keesom data,¹⁰ in which the spin-glass specific heat appeared to extrapolate to zero at a positive temperature, and concluded that there must be an error in their excitation frequency spectrum at zero fre-

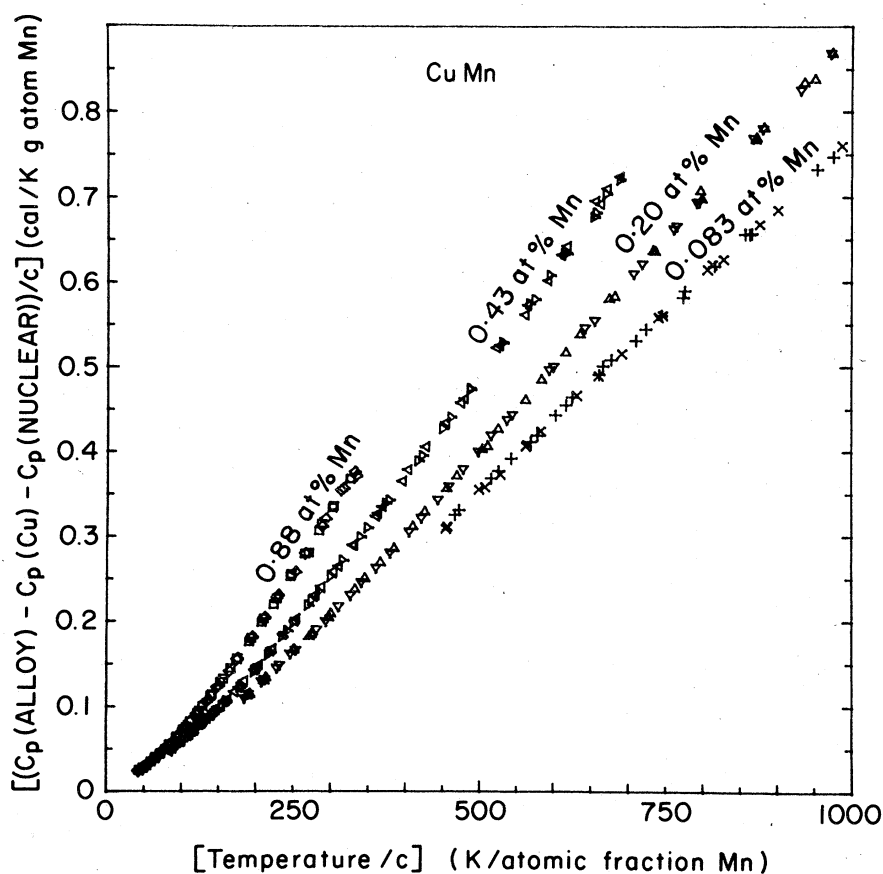


FIG. 6. Plot of scaled spin-glass specific heat against scaled temperature. The concentration c is expressed as an atomic fraction. Higher temperature points for the two most dilute alloys are off scale on this plot.

quency. The present results show that the calculation²⁸ is much better than comparison with earlier data suggested. Other recent theoretical results, for example some^{29,30} based on the Sherrington-Kirkpatrick model,³¹ also show that the spin-glass specific heat has the same general form as that predicted by the computer model calculation mentioned above²⁸ and as found in the present experimental work. For a recent summary of numerical simulation work see Binder.³² Some workers³³ have emphasized the existing of scaling laws [i.e., for the present case $C_p/c = f(T/c)$ where c is the Mn content] and some specific-heat data^{33,34} have been shown to obey this law. (One consequence is the concentration independence of the low-temperature limiting linear specific heat.) The present results (Fig. 6) quite clearly do not fall into this pattern and therefore cast doubt on the general applicability of scaling laws to spin-glass phenomena. (Unless, of course, the view, taken in some papers,³⁵ that systems not obeying scaling laws are not spin glasses, is adopted. This view has been criticized elsewhere.^{23,36}) For a recent survey of the theory see Blandin.¹

V. CONCLUSION

The present measurements show that the spin-glass specific heat of CuMn at low temperatures has a more complex form than was suggested by the earliest measurements. However, the results presented here are in agreement with more recent experimental data above 2 K and below 1 K and also with some recent theoretical work.

ACKNOWLEDGMENTS

Professor M. W. Klein suggested that further specific-heat measurements on the CuMn spin glass were necessary in order to quantify the low-temperature behavior and to facilitate comparison with theory. Professor P. A. Beck kindly supplied estimates of the spin-glass freezing temperatures for the present alloys. I am indebted to W. Fisher for his care and interest in the alloy preparation, to members of the Analytical Chemistry Section (V. P. Clancy, R. Ironside, H. B. MacPherson, and A. Mykytiuk) for the chemical analysis and to R. L. Snowdon for help with several other aspects of the experiments.

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