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A new analysis of spin-glass magnetic isotherms

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Abstract. It is shown that the empirical expression

$$\partial M/\partial H = \chi(0) - (\beta/2)\log(1 + H^2/K^2)$$

provides a very close fit to experimental spin-glass magnetic isotherms both at and above the freezing temperature T_g. Above T_g the parameter K is shown to vary in a very similar manner to $X_{nl}(T)$ as reported by previous researchers. At T_g, however, K is found to be finite. It is argued that this result demonstrates that there are no divergent quantities in the spin-glass magnetic isotherm.

1. Introduction

In spite of the very large body of experimental and theoretical work on spin glasses published in the past 30 years the central question of the existence of a phase transition in real dilute, magnetically disordered systems is still largely unresolved (Binder and Young 1986). In recent years, following the seminal work of Chikazawa *et al* (1981) and Monod and Bouchiat (1982) a great deal of experimental activity has centred around the behaviour of the non-linear susceptibility $\chi_{nl} = M_{nl}/H$, defined by the relationship

$$\chi_{nl} = [M(H) - \chi(0)H]/H \simeq -a_3H^2 + a_5H^4 + \ldots \tag{1}$$

at temperatures above the spin-glass freezing temperature T_G. χ_{nl} is thought to be particularly important in the context of spin glasses because it is associated with the order parameter $[\langle S^i\rangle^2]_{disorder}$ of Edwards and Anderson (EA) (1975). It is now thought that above T_g the Sherrington–Kirkpatrick equations provide an exact mean-field solution. In this solution, all the terms in equation (1) are thought to diverge as $T \rightarrow T_g$. Thus it is thought that measurements of χ_{nl} shed light on the experimental analogue of the EA order parameter. Attention has been focused on the low-field expansion of equation (1) since in this limit the predicted behaviour is particularly simple and it has been thought possible to separate the first two terms in equation (1) by suitable choice of field and temperature range. According to Chalupa (1977), one would expect the exponent γ of a_3 to diverge near T_g with an exponent of unity and the magnetisation on the critical isotherm to be a non-analytic function of the field $H^{2/\delta}$ with $\delta \simeq 2$.

In their original work, Monod and Bouchait (1982) studied the DC magnetisation of a Ag–10 at.% Mn sample and extracted χ_{nl} from plots of M/H against H^2 taken over a restricted field range at temperatures above T_g. They concluded that $\chi_{nl} \propto (1 - T/T_g)^{-\gamma}$ and estimated γ to be in the range $1 < \gamma < 2$. Subsequently other workers making similar

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analyses on other systems have produced results consistent with γ in the range $3 < \gamma < 4$ for Cu–Mn (Berton *et al* 1982, Omari *et al* 1983, Barbara *et al* 1981) and for amorphous $Al_2Mn_3Si_3O_{12}$. (Beauvillain *et al* 1984). All workers concede that, close to T_g, $M(H)$ becomes highly curved so that it is very difficult to obtain a finite region of field in which $M_{nl} \propto H^2$ and thus reliable estimates of γ. Some workers have tried to solve this problem by progressively limiting the field range used as T_g is approached (Monod and Bouchiat 1982, Bouchiat 1986). Others (Barbara *et al* 1981, Beauvillain *et al* 1984) have resorted to the unphysical procedure of assuming a form $\chi_{nl} \propto H^{a(T)}$ allowing $a(T)$ to vary with temperature. This approach is entirely wrong in the case of a paramagnet for which symmetry under field inversion must be observed. Omari *et al* (1983) attempted to find values for the higher terms in the expansion by an iterative procedure in which the successive terms in equation (1) were estimated from plots of χ_{nl} against powers of H. The severity of this problem is demonstrated by the manner in which all researchers analyse the critical isotherm. All attempt to extract the critical exponent δ from log–log plots of χ_{nl} against H. Bouchiat (1986) provides apparently strong evidence for $\delta = 0.64$ for Ag–Mn. de Courtenay *et al* (1986) show that, if a wider field range is employed, then the data appear curved on a log–log plot so that high- and low-field exponents are required. Even though T_g is not known in any system to better than about 2%, de Courtenay *et al* use radically different analyses on the assumed critical isotherm itself and on isotherms taken at temperatures much less than 2% above T_g. As we shall demonstrate in this paper, it is not necessary to invoke different hypotheses for T_g and temperatures above T_g. In our analysis, all the isotherms can be described by equation (1) provided that a sufficient number of terms are considered. This was an impossible task using the methods of others described above.

In this paper, we adopt a different approach which allows us to obtain estimates of all the terms in equation (1) at temperatures at and above T_g. Our method arose out of a desire to make quantitative comparisons between magnetic isotherms taken for ternary spin glasses based on Cu–Mn containing Pt and Al as impurities (Cayless and Guy 1988). In the absence of any quantitative theoretical description which could reliably be used over a wide range of field, we sought an empirical expression that closely described the experimental data with as few adjustable parameters as possible. We found a very simple expression, equation (4) below, that needed only one adjustable parameter. The temperature variation in this empirical quantity was so obviously closely linked to the question of the possible divergence of χ_{nl} that we have extended the use of our method to this central issue. It is not our intention to suggest that our new expression has any particular physical significance although it is remotely possible that it or something like it may ultimately arise from a microscopic theory. Rather we use this expression as a template which has the right characteristics dictated by equation (1) to lay over the data and to allow us to determine all the terms without recourse to artificial limitations on field or temperature range. Our expression also allows us to make a detailed appraisal of the validity of previous analyses.

We used our expression to fit magnetic isotherms over as wide a field range as was available. For each fit, we obtain a value for the adjustable parameter K. From a power series expansion of our original expression, equation (6) below, we can relate all the terms in equation (1) to powers of K. It should be noted that fitting a wide field range in our method is not equivalent to that employed by Omari *et al* (1983) who attempted to fit just two terms in equation (1) to data taken at high fields. This procedure has rightly been criticised for this reason and naturally produces results which differ from those of researchers who use lower field ranges. The temperature variation of K thus provides

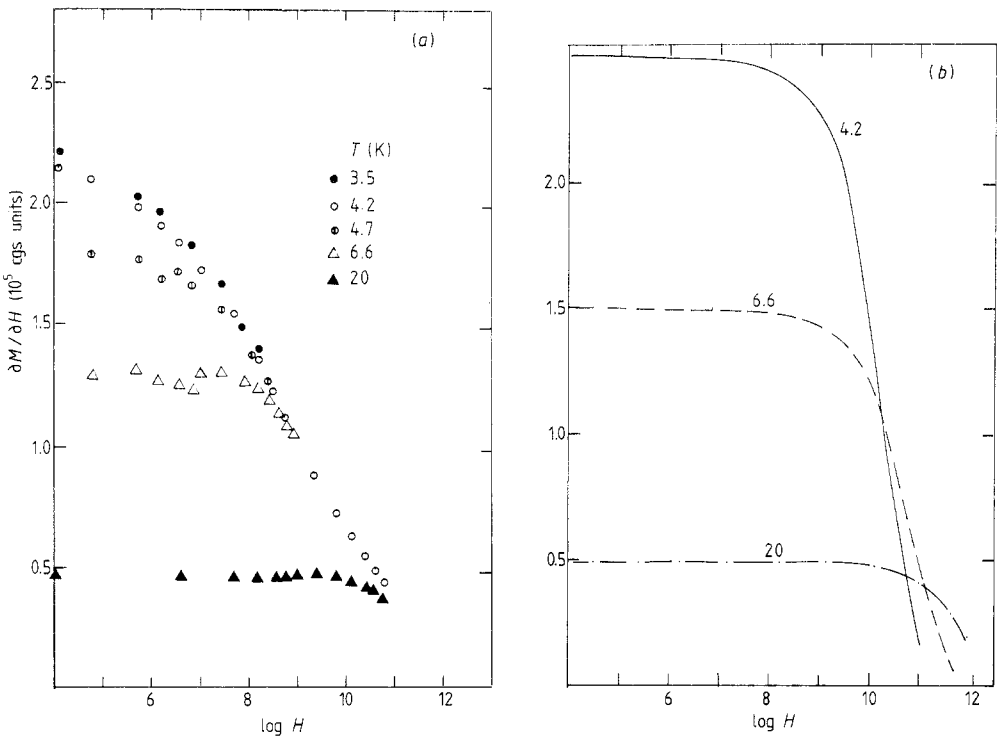

Figure 1. Plots of $\partial M/\partial H$ against log H obtained (a) for a Cu–0.25 at.% Mn alloy in this work, for which T_g was measured to be 3.55 K, and (b) from a Brillouin function with $g = 2$, $J = 1.8$. Temperatures are indicated on curves (in K).

us with a direct test of the divergence hypothesis that is entirely free from the defects in the numerical techniques used by previous workers.

The form of our expression is described in § 2. We justify its form through the use of plots of $\partial M/\partial H$ against log H and show that the same form with different parameters provides a tolerable fit to a Brillouin function. We present an analysis in § 3 of three experiments, two of which were reported by previous workers. In § 4, we discuss and compare our results with those of others.

The new experimental data presented in § 3 were obtained on a home-built vibrating sample magnetometer (VSM) whose resolution is 9×10^{-9} A m^2 with an accuracy of 2%. The Cu–Mn alloy used was prepared in a standard fashion by arc melting, heat treating and quenching in water. T_g for this sample was obtained from the low-field (0.003 T) maximum in $M(T)$.

2. The fitting function

Figures 1(a) and 1(b) show plots of $\partial M/\partial H$ against log H taken from Cu–0.25 at.% Mn ($T_g = 3.55$ K) and a calculated Brillouin function with $g = 2, J = 1.8$, chosen so that the initial values of $\partial M/\partial H$ agreed with the apparent high-temperature Curie law of this sample ($p_{eff} = 4.5\ \mu_B$ per Mn atom, $\theta \simeq 0$).

These figures illustrate the main points of this paper and suggest a simple analytic form for the fitting function. It is clear that both sets of curves become approximately linear in log H at high fields. It is also clear that the spin-glass isotherms are anomalously curved with respect to the Brillouin function over a very wide range of fields and not just at very low values. Finally, as the temperature approaches T_g, the curves appear to be approximately parallel over the entire range. Figure 1(a) includes the critical isotherm and this does not appear to be markedly different in form from its neighbours taken at higher temperatures. As far as we can tell from the published literature, these qualitative observations are true for all dilute spin-glass isotherms.

By a process of trial and error, we have found two expressions which follow the experimental data very closely. These are

$$\partial M/\partial H = \chi(0) - \beta' \sinh^{-1}(H/K') \tag{2}$$

$$\partial M/\partial H = \chi(0) - (\beta/2)\log(1 + H^2/K^2) \tag{3}$$

where in both formulae $\chi(0) = \partial M/\partial H_{\lim H\to 0}$ and β is the high-field slope of $\partial M/\partial H$ against log H. K is an adjustable parameter which is chosen by trial and error to provide a good fit to each isotherm.

Curiously both expressions provide very good fits to the data using different values of K with (2) giving slightly better results than (3). We cannot use (2), however, since it does not, without further adjustment, describe both positive and negative quadrants of a complete isotherm. Furthermore it has a low-field expansion which differs from equation (1). Integrating equation (3), we obtain

$$M(H) = [\chi(0) + \beta]H - (\beta/2)[H\log(1 + H^2/K^2) + 2K\tan^{-1}(H/K)]. \tag{4}$$

In § 4, we compare our analysis with those based on equation (1); thus we require the low-field expansion of (4) valid for $H \ll K$. We obtain

$$M(H) \simeq \chi(0)H - \beta H^3/6K^2 + \beta H^5/20K^4 - \beta H^7/42K^6 + \ldots. \tag{5}$$

It is clear that, if equation (4) fits the data well, then one immediately has estimates for all coefficients in equation (1) in terms of β and K. Our fitting procedure is very simple. We use plots of $\partial M/\partial H$ against log H to estimate $\chi(0)$ and β; then we vary K in equation (4) until it is judged by eye that the fit is uniformly good over the entire range of field considered. We estimate that we can determine K to about 5% by this procedure. It is possible that this could be improved by using a more objective numerical method for assessing the goodness of fit.

Figure 2 shows some examples of our fits to the Brillouin function shown in figure 1(b). Here we used a constant value for the high-field slope $\beta = 1.6 \times 10^{-5}$ cgs chosen to be a rough mean of the high-field slopes shown in figure 1(b). In § 3 where we fit spin-glass data, there is no ambiguity in the choice of β since in these cases it appears from figure 1(a) that all isotherms have a common high-field slope.

In the case of the Brillouin function it is clear how K must vary with temperature; as T increases, K must increase very rapidly in order that the fitting function should mimic the increasing range of field over which $\partial M/\partial H$ remains approximately constant. The inset in figure 2 confirms this. In § 3 where we apply equation (4) to real data, we find a similar temperature variation for K.

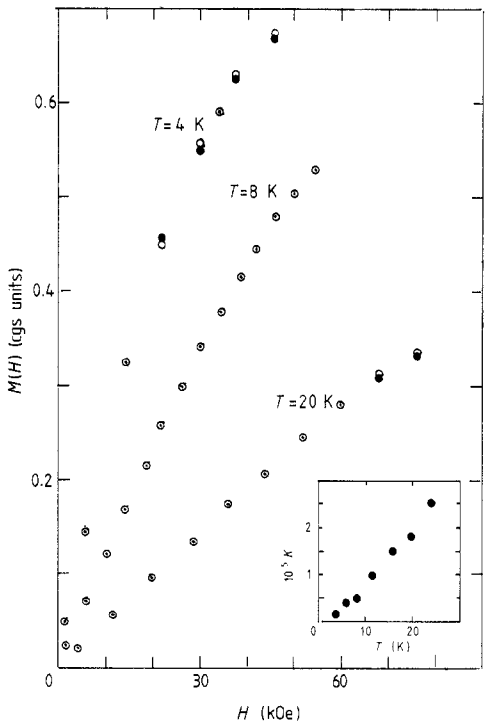

Figure 2. Comparison of calculated values (●) of $M(H)$ for the same Brillouin function as in figure 1(*b*) and the fits (○) using equation (4): ⊙, overlap. The inset shows the variation in K with temperature. All the first were obtained using $\beta = 1.66 \times 10^{-5}$ cgs.

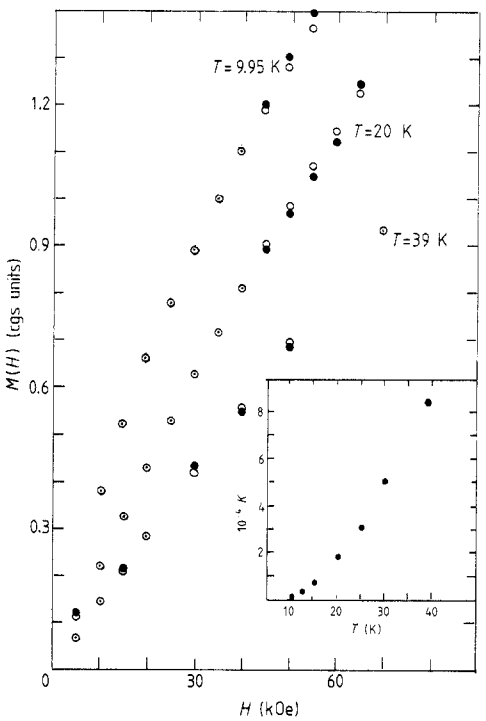

Figure 3. Comparison of experimental values (●) of $M(H)$ taken from Omari (1982) for a Cu–1 at.% Mn alloy and fits (○) using equation (4): ⊙, overlap. All the fits were obtained using $\beta = 1 \times 10^{-5}$ cgs. The inset shows the variation in K with temperature. T_g for this sample was given as 10.05 K.

3. Comparison with experiment

3.1. Cu–1 at.% Mn (Omari et al 1983)

Figure 3 shows a comparison between the experimental values of $M(H)$ and fitted values. The inset shows the temperature variation in K. The experimental curves were taken from a large graph in Omari's *PhD Thesis* (1982) with an estimated accuracy of 5%. Here β was estimated from the 9.95 K isotherm to be 1×10^{-5} cgs and was held constant for all the data. We used the values of $\chi(0)$ given by Omari (1982) for the sample that he used. It should be noted that these are larger by about 20% than the values given by Nagata *et al* (1979) for the same concentration. If we use the values of Nagata *et al* in our fits but keep β at the same value, then the values of K that we have to use to obtain a good fit are systematically smaller than those shown in figure 3. The average difference between fit and data was 2%; the maximum difference was 4.6% at low-field values on the higher-temperature isotherms. Figure 3 includes the critical isotherm 9.95 K. It is clear that this curve is very similar in form to that at 12.5 K and that our fits are equally good both at and above the critical temperature. In § 4, we demonstrate that the fractional power fit ($\chi_{nl} \propto H^{0.35}$) used by Omari *et al* for the critical isotherm is equivalent to our analysis using equation (4).

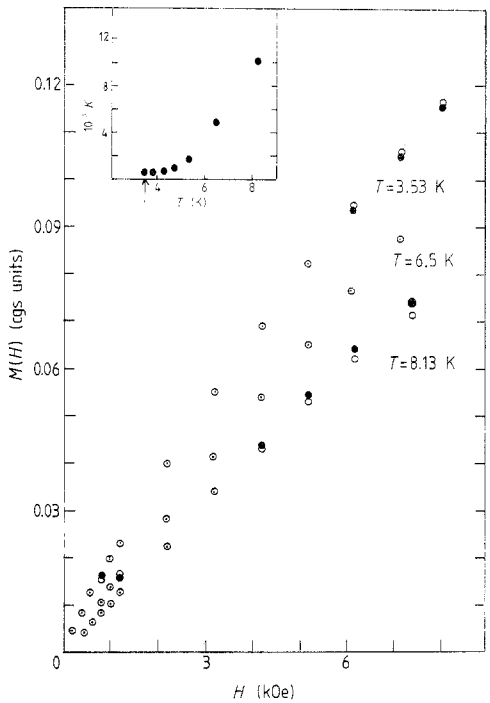

Figure 4. Comparison of experimental data of $M(H)$ and fits using equation (4) for the Cu–0.25 at.% Mn alloy. $\beta = 3.6 \times 10^{-6}$ cgs for all the curves. The inset shows the variation in K with temperature. T_g for this sample was measured to be 3.55 K.

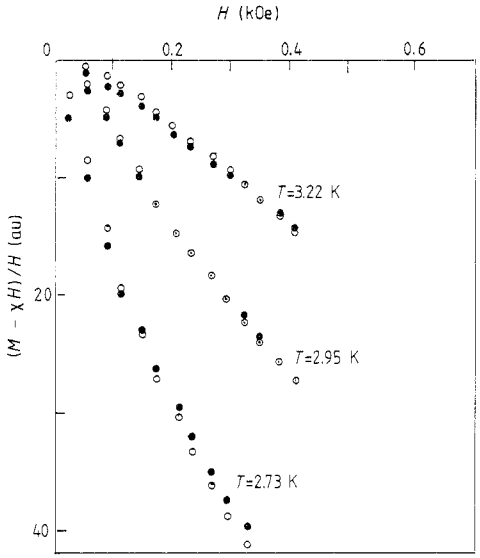

Figure 5. Comparison of experimental data of $[M(H) - \chi(0)H]/H$ taken from Bouchiat (1986) for a Ag–0.5 at.% Mn alloy. Here β was found by trial and error to be 30 au. T_g for this sample was given as 2.70 K.

3.2. Cu–0.25 at.% Mn: present work

Figure 4 shows similar fits to our own data taken over the field range 0–0.8 T. β was estimated to be 3.6×10^{-6} from a separate experiment taken at 4.2 K over the field range 0–6 T. This value of β was held constant for the other fits in this series. The insert shows that K varies in qualitatively the same manner as for the Cu–1 at.% Mn sample and the Brillouin function. In both the Cu–Mn samples we find that K is non-zero at T_g.

3.3. Ag–0.5 at.% Mn (Bouchiat 1986)

Figure 5 shows a comparison of experimental values of $(M - \chi(0)H)/H$ and our fits. The experimental data were taken from a small published graph with an estimated accuracy of 7%. Bouchiat obtained these data in an ingenious fashion; a measure of M_{nl} was obtained directly from the apparatus by bucking out (backing off), using an additional coil, the contribution linear in H. This method does of course require very accurate knowledge of $\chi(0)$. In our fits to these data, we had to find a value of β by trial and error since high-field data for this sample were not available. It should be noted that in this case the values of the field are very much lower than the previous two cases but the fits are reasonably good both above and near the critical temperature of 2.7 K. In this case

our values of K may be in error by as juch as 30% because we have guessed values of β and $\chi(0)$. Bouchiat (1986) shows data for 2.70 K following the form $\chi_{nl} \propto H^{0.64}$.

4. Discussion

In the three cases that we have considered, the fits, although not perfect, are sufficiently good for us to assert that K remains finite and thus χ_{nl} does not diverge at T_g. In addition, we have shown that the critical isotherm can be described in the same way as isotherms taken at higher temperatures. This result is so much out of step with current thinking on spin glasses that is necesary for us to demonstrate in detail just how this major difference arises. We can do this in three ways. First, we can examine the exact conditions under which equiation (5) (and hence equation (1)) provides a faithful description of the data when the series is truncated at H^4 or below. Secondly, we can compare our values of K with the values of the quantity $a_3^{-1/2}$ where a_3 are the coefficients of the H^2 term in equation (1) reported by Bouchiat for Ag–Mn and by Omari *et al* for Cu–Mn. Finally, we can compare our estimates of χ_{nl} on the critical isotherm with those reported by these workers. Both Omari *et al* and Bouchiat show convincing evidence that their critical isotherms are described by power laws as described in § 3. Here there is a very simple direct comparison between the two methods since our equation (4) when plotted in the same fashion should yield the same apparent power law over the same ranges of field.

It is clear that a truncated version of the equation

$$M(H)/H = \chi(0) - \beta H^2/6K^2 + \beta H^4/20K^4 \tag{6}$$

can only describe the data when $H \ll K$. Near T_g, for Bouchiat's Ag–Mn sample, we find a value of 35 G for K and, for the Cu–Mn samples of Omari *et al*, a value of 1000 G. Bouchiat found that, near T_g, she had to limit the range of field used to extract the coefficient of H^2 to 50 G so that higher-order terms did not affect her estimate. Omari *et al* used a field range of 400 G at 11.15 K (1.1 T_g) in their analysis of this isotherm. In both cases these values are to be expected given our values of K in equation (4). At fields of the order of K the series is only slowly convergent and hence equation (6) does not describe the data faithfully. It should be noted, however, that even in fields below K the effects of the higher terms are still important.

Figure 6 shows our values of K plotted against T together with the values of $a_3^{-1/2}$ reported by Bouchiat and Omari *et al* on a log–log plot. We have multiplied the published values by constants so that, for each case, K and $a_3^{-1/2}$ lay close together on the graph. It is clear from this figure that there is good agreement between our analysis and those of Bouchiat and of Omari *et al*.

Figure 6 also demonstrates our central point that there is no experimental evidence for a divergence in χ_{nl} at T_g. As far as log–log plots are reliable, figure 6 suggests that all three spin glasses are characterised by $K(T) \propto T^a$ with a in the range $5 < a < 7$ in a limited range of temperatures.

The more severe test is that of the critical isotherm itself. Bouchiat shows an exponent of 0.64 for the Ag–Mn sample; Omari *et al* show a value of 0.35 for the same quantity. Using our values of K of 35–50 for Ag–Mn and 1000 for Cu–Mn, we have plotted, in figure 7, log–log plots of χ_{nl} generated using equation (4) for the two cases on the respective critical isotherms. There it can be seen that our data mimic the supposed fractional exponent over the field range used as long as $H \gg K$. At lower field values the fractional exponent description breaks down. In their papers, both Omari *et al* and

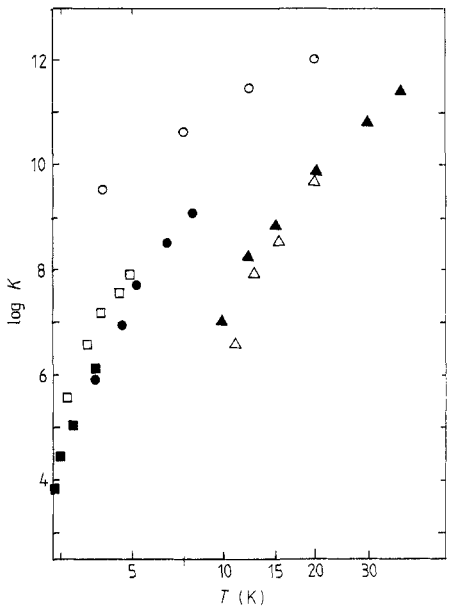

Figure 6. log–log– plots of K against T (○, ▲, ●, ■) and $a_3^{-1/2}$ against T (△, □) for the three spin-glass samples (▲, △, ●, ■, □) and the Brillouin function (○). The values of $a_3^{-1/2}$ for Cu–1 at.% Mn obtained by Omari *et al* and for Ag–0.5 at.% Mn by Bouchiat were multiplied by constants so that they lay close to the values of K at T_g for each sample.

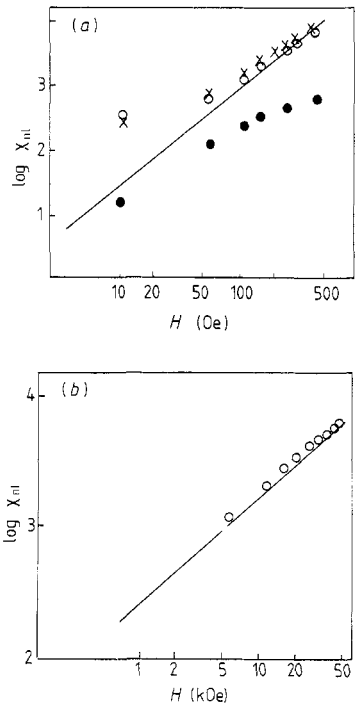

Figure 7. (*a*) log–log plots of χ_{nl} against H very near the critical isotherm for Ag–0.5 at.% Mn obtained in this work via equation (4) (●, ○, ×) and the $H^{0.64}$ form reported by Bouchiat (——). Our values of χ_{nl} have been multiplied by a constant to bring them close to the straight line. The three curves derived in this work correspond to the different values of K: ●, 10; ×, 35; ○, 50. (*b*) The same plot as in (*a*) for the critical isotherm of for Cu–1 at.% Mn obtained in this work (○) and the $H^{0.35}$ form reported by Omari *et al*. Our results have been multiplied by a constant so that they lay close to the straight line and are for $K = 1000$.

Bouchiat describe similar low-field departures from their chosen exponent. This shows that there is no real disagreement between the two methods of analysis in terms of the empirical description of the data.

We have shown, however, that there is no functional difference between the critical isotherm and those taken at higher temperatures. With hindsight this result could have been anticipated from the fact that some researchers found that they obtained better descriptions of $\chi_{nl}(H)$ by using a non-physical temperature-dependent fractional exponent at all temperatures (Beauvillian *et al* 1984). By accident such an analysis mimics the analytic expression over limited ranges of field. As we have already mentioned, de Courtenay *et al* (1986) found that over a wide range of fields a single value of the exponent was not sufficient; rather different values were required in different field ranges. In the light of equation (4) this is to be expected.

We have shown that an empirical analytic expression provides a very good fit to experimental spin-glass magnetic isotherms. We have found that in three cases there is

no evidence for a divergence in the non-linear susceptibility and that the critical isotherm can be described on exactly the same footing as data taken at higher temperatures. This result suggests that in the terms of current theoretical models there is no static phase transition in real spin glasses since the predictions of these models are not borne out by experiment. Furthermore we have demonstrated that results that have until now been taken as strong evidence for a static phase transition at T_g are also consistent with an alternative hypothesis in which χ_{nl} varies approximately as T^a at temperatures below about (1.5–2) T_g.

Although this work, if confirmed, may be a step forward, it does not really enhance our understanding of spin glasses since our expression is entirely empirical with no obvious physical basis for the parameter K. If it is to have a physical reality, then K must be related to $k_b T/\mu$; possibly it provides a measure of cluster size. Perhaps some clues are contained in its temperature dependence shown in figure 6. All the curves appear to follow an approximate power law in T at low temperatures but curve over towards the results for the Brillouin function at high temperatures. This is in qualitative accord with the data on inverse susceptibility reported by Morgownik and Mydosh (1981). They showed for a series of Cu–Mn alloys of different compositions that there were large ferromagnetic deviations from Curie–Weiss behaviour existing up to at least $2T_g$. Our analysis in terms of equation (4) and hence $K(T)$ provides a compact description of the way in which spin-glass isotherms evolve from the high-temperature paramagnetic state. It is possible that our empirical expression could provide guidance in the construction of a microscopic theory of interacting and evolving clusters.

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