Aust. J. Phys., 1983, **36**, 519–36

Spin Glasses in Transition?*

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Abstract

Spin glasses have variously been described as apparently frozen but very slowly relaxing spin systems, and also as magnetic phases below a second order transition. The structural and dynamic properties of metallic spin glasses are surveyed with emphasis on neutron scattering and low field magnetic measurements. Spin glass regions in phases with magnetic long range order are included, and an attempt is made to assess spin glass theories with respect to the various transitions.

1. Introduction

Since the observation of a cusp in the low field a.c. susceptibility of metallic alloys containing dilute magnetic impurities (Canella and Mydosh 1972) (Fig. 1), there has been a suggestion that below a critical temperature there is a spin glass phase in which the impurity spins are aligned in fixed but random directions. This view was bolstered by the mean field theory of Edwards and Anderson (1975) which describes such a spin glass phase with a 'local order parameter' q, and which predicts a second order phase transition (see Section 5).

This paper will review low field magnetic and neutron scattering measurements, on mainly metallic spin glasses, which elucidate some of the dynamic and structural properties of these materials. In fact much of the data presented will be for *Cu*Mn alloys in order to restrict the wealth of phenomena and expose some principles. In this spirit an attempt will also be made to assess the current status of the Edwards–Anderson 'order parameter' in the theoretical modelling of spin glasses. The survey of spin glass phenomena will also include the more recently discovered spin glass phases in magnetically long range ordered alloys close to their critical concentration, and in which the 'glassiness' is thought to be in the transverse spin components.

Therefore, even in this restricted survey, there is still a richness of phenomena associated with the structure, dynamics and transitions of spin glasses.

2. Spatial Correlations in Spin Glasses

In a pure magnetic system the approach to the critical point is characterized by the divergence of the correlation length. This can be illustrated quite simply by

* Paper presented at the Seventh AIP Solid State Physics Meeting, Wagga Wagga, N.S.W., 9–11 February 1983.

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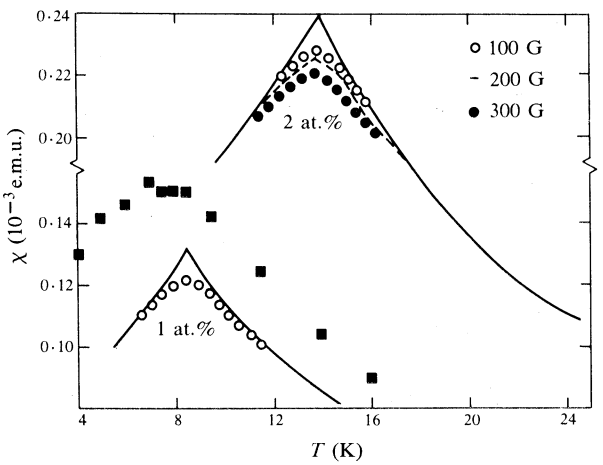


Fig. 1. Low field a.c. susceptibility of *Au*Fe spin glasses against temperature showing the curves for zero (solid curves) and other small fields (Cannella and Mydosh 1972). Squares refer to work by Lutes and Schmit (1962) for 1 at.% samples.

considering the wavelength dependent susceptibility of a localized moment ferromagnet above its critical temperature in the mean field approximation. The susceptibility is

$$\chi(\boldsymbol{\kappa}) = M(\boldsymbol{\kappa})/H(\boldsymbol{\kappa}) = \chi_0/\{1-\lambda(\boldsymbol{\kappa})\chi_0\},$$

with $M(\boldsymbol{\kappa})$ the wavevector dependent magnetization induced by a spatially varying field $H(\boldsymbol{\kappa})$; the susceptibility in the absence of any interactions is

$$\chi_0 = \{g^2\mu_B^2 S(S+1)\}/3k_B T,$$

and $\lambda(\boldsymbol{\kappa})$ is the mean field parameter, which has a $\boldsymbol{\kappa}$ dependence because the exchange coupling has a finite range. The critical temperature is when $\chi(0)$ diverges,

$$T_c = \{g^2\mu_B^2 S(S+1)\lambda(0)\}/3k_B,$$

where k_B is Boltzmann's constant.

The behaviour of $\chi(\boldsymbol{\kappa})$ for small $\boldsymbol{\kappa}$ can be obtained by considering that $\lambda(\boldsymbol{\kappa})$ must be an even function of $\boldsymbol{\kappa}$ with a maximum at $\kappa = 0$. So we have

$$\lambda(\boldsymbol{\kappa}) \approx \lambda(0) - \lambda_1 \kappa^2, \qquad \chi(\boldsymbol{\kappa}) \approx \chi_0/\{1-\lambda(0)\chi_0 + \chi_0\lambda_1\kappa^2\}.$$

The function $\chi(\boldsymbol{\kappa})$ is a Lorentzian whose half-width

$$\kappa_0 = [\{1-\lambda(0)\chi_0\}/\lambda_1\chi_0]^{\frac{1}{2}}$$

goes to zero at the critical temperature. The real space correlation length is proportional to the reciprocal of κ_0, and thus diverges at the critical temperature. This behaviour is well documented in ferromagnets and antiferromagnets (see e.g. Spooner and Averbach 1966), but in antiferromagnets it is the correlation length of the staggered susceptibility which diverges.

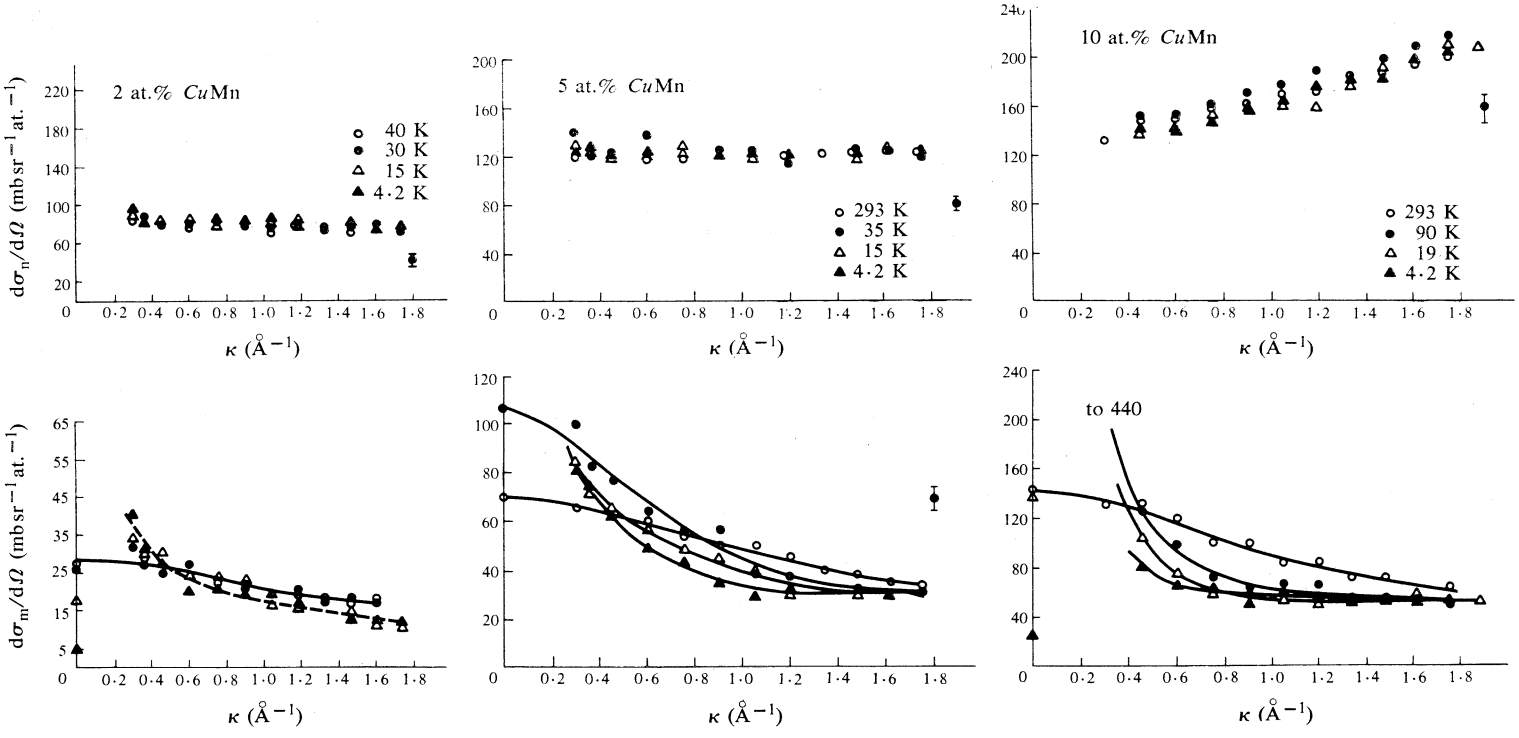


Fig. 2. Neutron scattering cross sections for three *Cu*Mn spin glasses for $\kappa = (4\pi \sin\theta)/\lambda$ Å^{-1}. The upper graphs show the nuclear scattering which indicates that Mn is randomly distributed for the 2 and 5 at.% alloys. The lower graphs show the wavevector and temperature dependence of the magnetic scattering. Also included at $\kappa = 0$ are points corresponding to the bulk susceptibility at the various temperatures measured on the same samples. The glass temperatures for the 2, 5 and 10 at.% alloys are approximately 14, 29 and 43 K respectively. [Ahmed and Hicks (1974).]

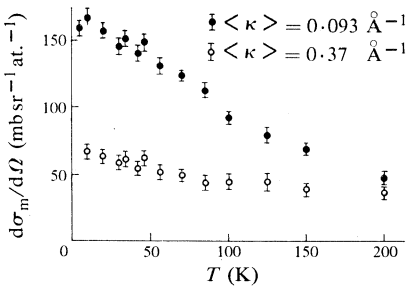
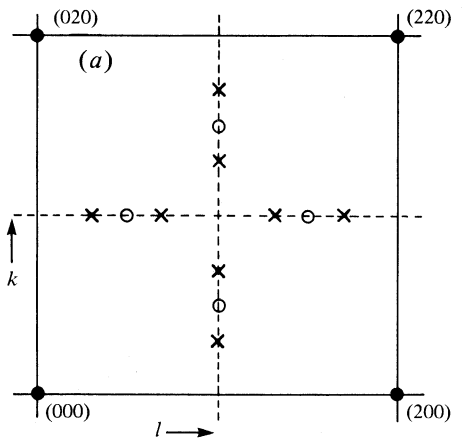


Fig. 3. Variation of the magnetic neutron scattering cross section with temperature for 5 at.% *Cu*Mn at two scattering vectors. [Mezei and Murani (1979).]



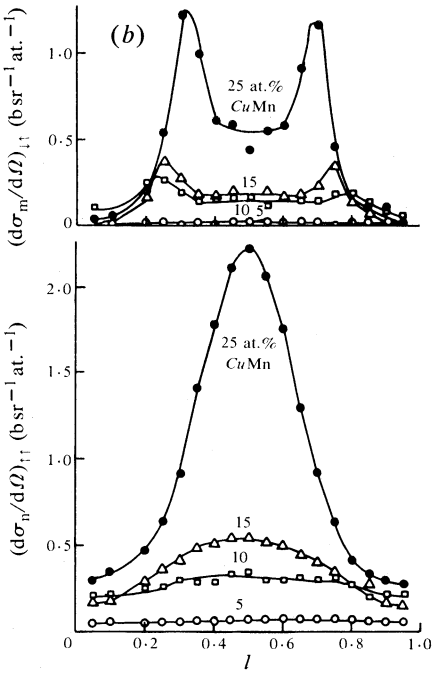


Fig. 4. (*a*) Positions in reciprocal space of features observed in (*b*) for *Cu*Mn alloys at 4·2 K. (*b*) Magnetic (upper) and nuclear (lower) neutron cross sections for *Cu*Mn alloys along the line $[l, 1, 0]$ in (*a*). [Werner and Cable (1981).]

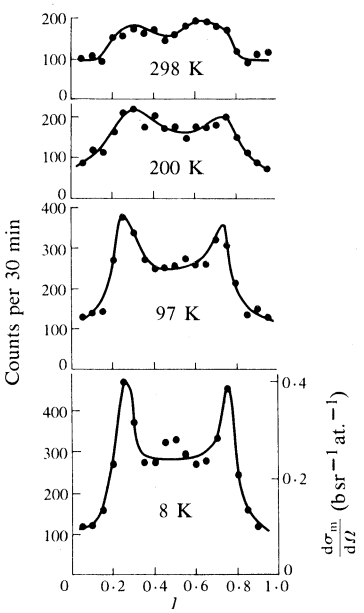


Fig. 5. Temperature variation of the features in the magnetic neutron scattering cross section for 15 at.% *Cu*Mn. [Werner and Cable (1981).]

The first measurement of $\chi(\kappa)$ for a spin glass was that of Ahmed and Hicks (1974) on *Cu*Mn alloys. Their results are shown in Fig. 2. The magnetic part of the neutron scattering cross section is directly proportional to $\chi(\kappa)T$ under the assumption that the scattering is quasi-elastic. For all three alloys examined there is a change in the κ dependence of $\chi(\kappa)$ with temperature, but in a gradual and continuous manner. In particular, there is no large difference in $\chi(\kappa)$ above and below the spin glass temperature defined by the cusp in the low field a.c. susceptibility.

Support for the above observation comes from the data of Mezei and Murani (1979) who monitored the temperature variation of $\chi(\kappa)T$ for two κ values (Fig. 3). The variation is continuous through the spin glass temperature. Both experiments can be criticized for being too coarse either in κ or in temperature, and indeed significant features in $\chi(\kappa)T$ were missed until the work of Werner and Cable (1981) was reported on *Cu*Mn single crystals. Fig. 4 shows the features observed by Werner and Cable and their positions in reciprocal space. It should be pointed out that these features are not Bragg-like and therefore only refer to correlations of the order 30–40 Å, even at a concentration of 25 at.% Mn. They also develop continuously as the temperature is decreased (Fig. 5). Recently, Harders and Wells (1983) have confirmed the existence of these features.

To date, at least in 5 at.% *Cu*Mn, most of the first Brillouin zone has been scanned for $\chi(\kappa)T$ (Davis and Hicks 1979; Werner and Cable 1981) and its temperature dependence for all κ appears to be continuous through the spin glass temperature.

3. Dynamics of Spin Glasses

If the spatial correlation of the spins in a spin glass shows no anomalous behaviour at the spin glass temperature why is there a cusp in the susceptibility? The beginning of a crude answer can be deduced from the experiment of Ahmed and Hicks (1974), who compared the extrapolation of $\chi(\kappa)$ ($\kappa \to 0$) obtained from the magnetic neutron scattering cross section with the bulk susceptibility measured on a magnetometer. At high temperatures the two susceptibilities agree very well, but below the spin glass temperature the bulk susceptibility is small compared with that extrapolated from the neutron scattering cross section. In Fig. 2, points for the bulk susceptibilities for the various temperatures are shown at $\kappa = 0$.

The discrepancy between the bulk and neutron susceptibilities can be reconciled if the relaxation time of the magnetism is much longer than the time of the bulk susceptibility measurement. This means that on the time scale of a bulk susceptibility measurement most of the moments in the material appear frozen. Of course we cannot tell whether they are completely frozen because the experiment contains no information on the time dependence of the susceptibility at times longer than that of the bulk susceptibility measurement. If the moments are *completely* frozen, however, they will scatter the neutrons *strictly* elastically. Only that fraction not completely frozen, and scattering quasi-elastically, can then be interpreted as contributing to $\chi(\kappa)$.

Murani and Tholence (1977) analysed the energy of neutrons scattered from an 8 at.% *Cu*Mn sample. In their analysis they only counted neutrons outside their energy resolution as contributing to $\chi(\kappa)$, and effectively treated the rest of the scattered intensity as *strictly* elastic. They compared $\chi(\kappa)$, obtained in this way, with the low field a.c. susceptibility measured on the same sample at 16 Hz, and Fig. 6

shows the results for several small values of κ. The main feature is the cusp in $\chi(\kappa)$ obtained at a temperature very much higher than for the a.c. susceptibility. The invariance of temperature of the cusp in $\chi(\kappa)$ with κ suggests that the unusually high temperature would probably be a property of $\chi(0)$ derived in the same way.

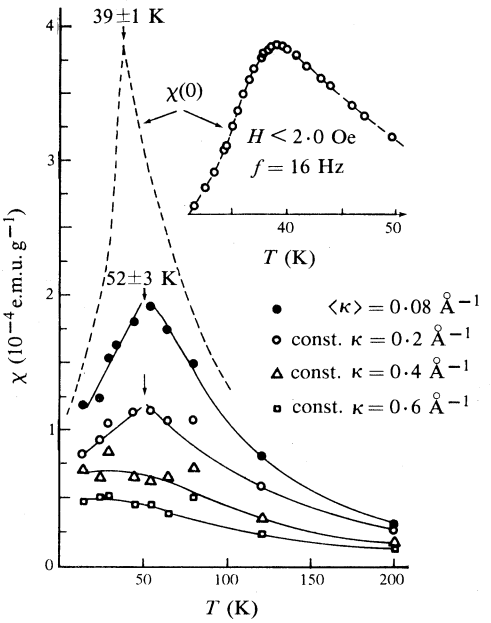


Fig. 6. Variation with temperature of the susceptibility calculated from that part of the quasi-elastic neutron scattering lying outside the energy resolution of the experiment (corresponding to a time constant of 10^{-11} s) for 8 at.% *Cu*Mn. The dashed curve and inset show for comparison the low frequency susceptibility measured on the same sample. [Murani and Tholence (1977).] (Note: 1 Oe $\equiv 10^3/4\pi$ A m^{-2}.)

The most probable explanation for the neutron derived susceptibilities having a cusp temperature of 52 K rather than 39 K is that the missing part of the susceptibility is included in that part of the scattering not resolvable from the strictly elastic scattering (or there may be no strictly elastic scattering). The energy resolution of this experiment is equivalent to being able to excite those parts of the susceptibility with relaxation times of less than 10^{-11} s. This would indicate that at 52 K, parts of the system acquire relaxation times of longer than 10^{-11} s and appear frozen on that time scale, whereas the same thing happens at 39 K on a time scale of seconds to minutes.

This experiment then raises the possibility that the glass temperature is not an equilibrium phase transition but a non-equilibrium artifact of the time scale of the experiment, and suggests that a profitable line of investigation might be to monitor the susceptibility cusp as a function of frequency. Unfortunately, the cusp temperature is widely reported to be independent of frequency (Dahlberg *et al.* 1979; Gray 1980) for metallic spin glasses. Tholence (1980), however, does report frequency dependent cusp temperatures for metallic spin glasses. His results are shown in Fig. 7

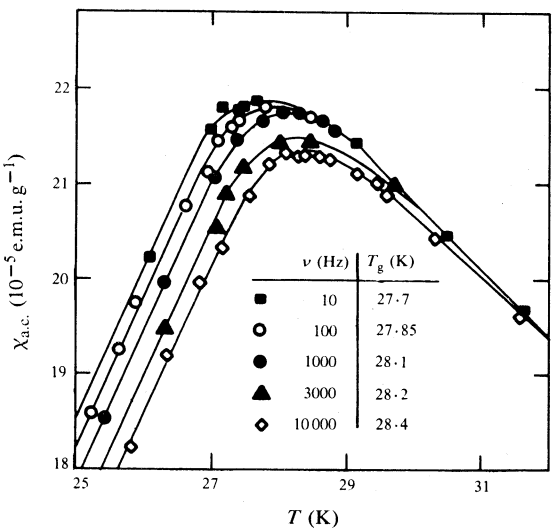


Fig. 7. Frequency dependence of the a.c. susceptibility of a 4·6 at.% *Cu*Mn spin glass near T_g. [Tholence (1980).]

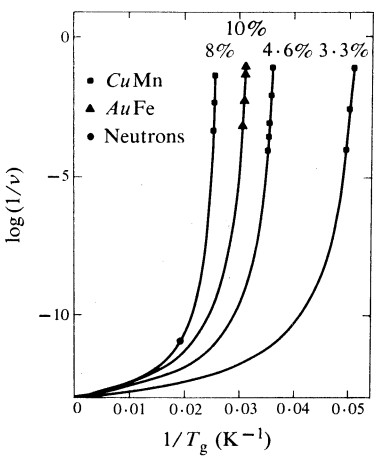


Fig. 8. Variation of T_g with frequency for some metallic spin glasses plotted as a Fulcher law. [Tholence (1980).]

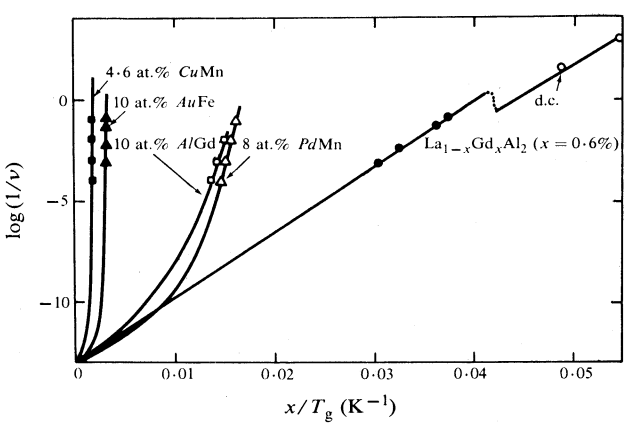


Fig. 9. Fulcher law plots of the variation of T_g with frequency for a range of metallic spin glasses. The plots shown range from extreme Fulcher law (*Cu*Mn, *Au*Fe) to Arrhenius behaviour ($La_{1-x}Gd_xAl_2$). For the latter $T_0 \approx 0$. [Tholence (1980).]

for 4·6 at.% *Cu*Mn, but he makes the point that the relation between the glass temperature T_g and the measuring time τ is not given by the Arrhenius law

$$\tau = \tau_0 \exp(E_a/k_B T_g),$$

with E_a the activation energy. The change of T_g with τ is so small that the characteristic time τ_0 would have to be an unphysical $\sim 10^{-37}$ s. Tholence rather fits his results to a Fulcher law $\tau = \tau_0 \exp\{E_a/k_B(T_g - T_0)\}$ with some success (Fig. 8), even including the result from neutron scattering. His survey of other spin glass systems (Fig. 9) shows that there is a complete spectrum from extreme Fulcher to simple Arrhenius behaviour.

The Fulcher law suggests that at long measurement times there is a temperature T_0 below which the glass temperature does not fall. Such measurement times and temperature precision are very hard to achieve and no data exist which would test the Fulcher law predictions at long times.

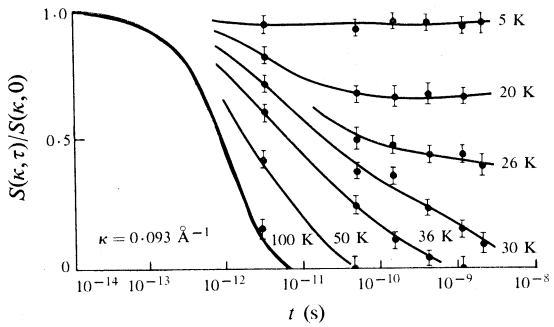


Fig. 10. Variation of the spin correlation function with temperature and time as measured by the neutron spin echo technique for 5 at.% *Cu*Mn. The curve to the left corresponds to a simple exponential decay $\exp(-\gamma t)$ ($h\gamma = 0{\cdot}5$ meV), and for different decay rates would simply move along the time axis with unaltered shape. The other curves merely connect points taken at the same temperature. [Mezei and Murani (1979).]

Data on relaxation times, however, do exist for short times from Mezei and Murani (1979) using the neutron spin echo technique. This technique directly measures the spin correlation function

$$1 - \pi \int_0^{\tau} \mathrm{d}t\, \chi(\mathbf{\kappa}, t)/\chi'(\mathbf{\kappa}, \omega = 0),$$

where χ' is the real part of the frequency dependent susceptibility. The results for 5 at.% *Cu*Mn are shown in Fig. 10. Over most of the temperature and time range the relaxation is nowhere near exponential, and is better described as being proportional to $\ln\tau$. If such a dependence continues to long times it is understandable that there is such a small change in the susceptibility over many frequency decades.

4. The 'd.c.' Susceptibility

With the advent of squid magnetometers and sensitive vibrating sample magnetometers, it is now possible to measure the 'd.c.' susceptibility of spin glasses in low

magnetic fields. The 'd.c.' susceptibility is written within quotation marks to emphasize that there is always a finite measuring time.

An example of 'd.c.' measurements is that of Nagata *et al.* (1979) taken with a squid magnetometer (Fig. 11). Two branches of the susceptibility can be seen for each sample below the glass temperature. The higher branches (A and C) are taken with the field always applied and cooling down through T_g. The lower branches (B and D) are taken by first cooling below T_g and then applying the field. The a.c. susceptibility more closely corresponds to the lower branches. In addition to the two susceptibilities, there is a remanent magnetization very closely equal to the difference between the upper and lower branches of the susceptibility at all temperatures; this will be discussed below.

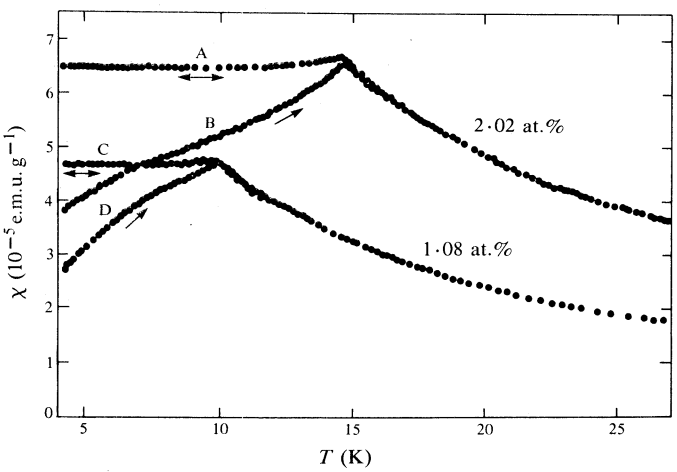


Fig. 11. The 'd.c.' susceptibility of two *Cu*Mn samples taken in a field of 5·9 Oe. The two branches below T_g are explained in the text. [Nagata *et al.* (1979).]

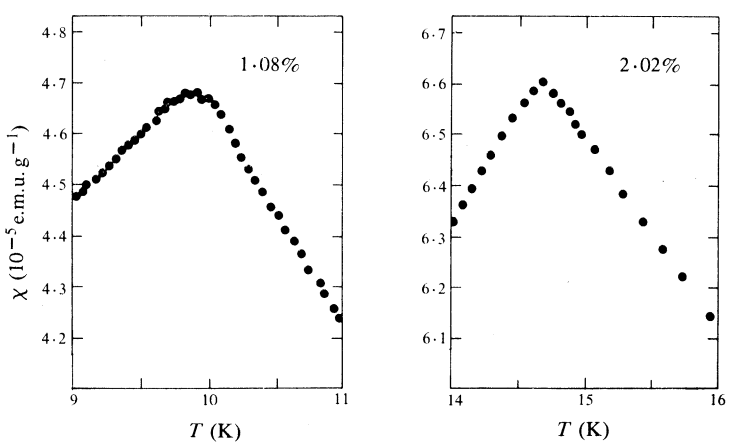


Fig. 12. Detailed view near T_g for the initial 'd.c.' susceptibility of *Cu*Mn samples. [Nagata *et al.* (1979).]

Fig. 12 shows an expanded view of the glass temperature region of Fig. 11 and the cusp is sharp enough to be able to identify T_g to much less than 0·1 K. At this level it is difficult to say whether any broadness observed in the cusp is due to slight inhomogeneities in the alloy or whether it is inherent. Nagata *et al.* (1979) also showed that the susceptibility above T_g is a very good Curie–Weiss law, even close to the glass temperature. This is surprising in view of the well-known deficiencies of the mean field theory.

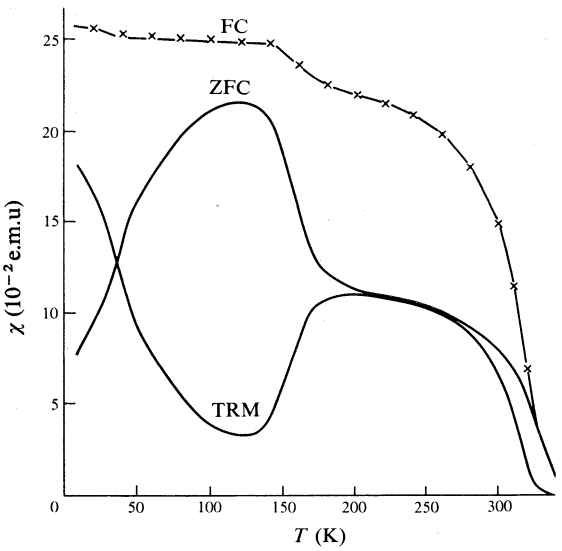


Fig. 13. The d.c. susceptibilities of 20 at.% *Au*Mn measured in a field of 3 Oe. The FC (field cooling) branch is measured while cooling in the field. The TRM (thermo-remanent magnetization) branch is measured in zero field while warming after the sample has been cooled in the field. The ZFC (zero field cooling) branch is measured while warming in the field after the sample has been cooled in zero field. [Gray (1983).]

Gray (1983) has recently measured the 'd.c.' susceptibilities of a more complicated spin glass, 20 at.% *Au*Mn, which is a good illustration of the sum rule between the remanent magnetization and the magnetizations induced from the two susceptibilities (see Fig. 13). The susceptibility is large enough so that demagnetizing fields have to be taken into account, especially as the applied field was only 3 Oe. The strange dip in the remanent magnetization (TRM) at ~120 K is explained as follows. The back field from the remanent magnetization acts on the reversible part of the susceptibility, so that the measured total magnetization is less than the remanent magnetization. When the reversible susceptibility has a peak, then the apparent remanent magnetization has a minimum, whilst the variation of the actual remanent magnetization is monotonic with temperature. Analysed in this way (Gray 1983), the remanent magnetization (TRM) and induced reversible magnetization (ZFC) do indeed sum to the magnetization (FC) observed in the upper branch. Further, the volume fraction of irreversible material is a monotonically decreasing function of temperature, to the accuracy of the demagnetization corrections.

The above analysis encourages one to think in terms of volumes of reversible and irreversible material. However, the diffraction evidence, at least for simple dilute

spin glasses (Ahmed and Hicks 1975), is that they are microscopically homogeneous and, apart from the complication of gross demagnetizing effects, show the same reversible and irreversible effects. If the time dependence of the relaxation, which is observed for short times by the neutron spin echo technique, extends to laboratory times, it is quite possible to observe this as reversible and irreversible parts. In Fig. 14 the function $y \propto \ln \tau$ is plotted linearly with a constant of proportionality which would keep the relaxation going over many decades of τ. As can be seen the curve can be easily decomposed into a fast relaxing part and a slow relaxing part.

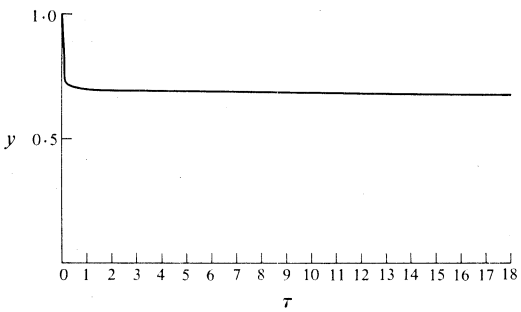


Fig. 14. Linear plot of $y = 0{\cdot}01 \ln \tau + 0{\cdot}7$. For this function $y = 1$ when $\tau \approx 10^{-13}$, $y = 0$ when $\tau \approx 10^{30}$.

There is a rough justification for expecting a $\ln \tau$ law for relaxation, due originally to Street and Woolley (1949). If we take an initial density of metastable states $f_0(E)$, then after time t, the density of states will be

$$f_t(E) = f_0(E) \exp(-\lambda t), \qquad \text{with} \quad \lambda = c \exp(-E/k_B T).$$

As time proceeds the lower energy states will be activated first and the distribution will move to higher energies. Below a certain value of λt the metastable states will be intact. Above this value they will have been activated. We choose this value so that $\exp(-\lambda t)$ is small, $\lambda t = C$ say. Then this point is represented by

$$ct \exp(-E/k_B T) = C \qquad \text{or} \qquad \ln(ct) - E/k_B T = \ln C,$$

where $E = k_B T \ln(ct/C)$. If the distribution of metastable energies is broad then $f_0(E)$ is almost a constant and the number of states activated in a time between t_1 and t_2 is

$$\Delta N \propto \Delta E = k_B T \ln(t_2/t_1).$$

Thus, the logarithmic relaxation is characteristic of a broad distribution of activation energies.

Time dependent effects have been seen in *Au*Fe spin glasses by Tholence and Tournier (1974) and Guy (1978). Tholence and Salamon (1982) have recently followed the relaxation of the remanent magnetization in various fields and temperatures. They described the relaxation by a term $S(H, T) \ln t$ for 0·24 at.% *Cu*Mn. Decay of the remanent moment has been observed by Maletta and Felsch (1979) in $Eu_xSr_{1-x}S$ spin glasses, but these compounds are not metallic and all observations are at relatively high fields.

5. Theoretical Situation

Edwards and Anderson (EA) (1975) were the first to attempt to put the theory of spin glasses on a spin by spin statistical mechanics footing. Previous models (see e.g. Tholence and Tournier 1974) considered the behaviour of correlated regions of the alloy frozen in the local anisotropy field. However, while models of this variety explain many of the properties of spin glasses, it is difficult to delineate the regions in view of the observed lack of magnetic short range order in dilute metallic spin glasses (Ahmed and Hicks 1975). Such models are still useful in explaining time dependent effects especially, but hopefully these dependences will eventually be explained by the more microscopic treatments.

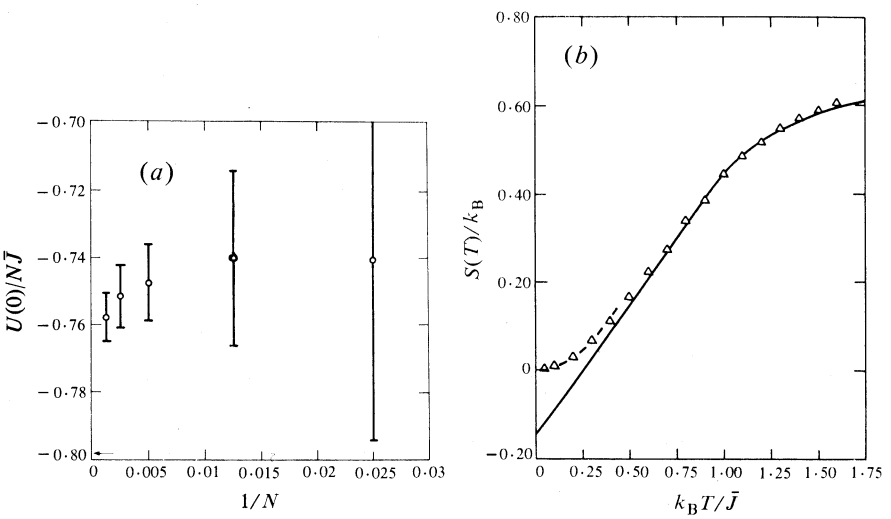


Fig. 15. (*a*) Ground state energies for Monte Carlo simulations of Ising spin glasses with zero average interaction. Here N is the number of spins, $\bar{J}$ is the average exchange energy and the arrow indicates the value predicted for an infinite number of spins from replica theory. (*b*) Entropy against temperature for a 500 spin Ising spin glass with zero average interaction. The solid curve is the replica theory prediction. [Kirkpatrick and Sherrington (1978).]

Edwards and Anderson derived a solution for a system of classical spins connected by first neighbour exchange interactions with a gaussian frequency distribution and a mean strength of zero. They found that at low temperatures each spin developed a correlation with itself in the thermodynamic limit and they defined an 'order parameter'

$$q = \lim_{t-t'\to\infty} \frac{1}{N}\sum_j \langle \boldsymbol{S}_j(t).\boldsymbol{S}_j(t')\rangle .$$

In the EA theory there is an ordering temperature (discontinuity in the specific heat/second order transition) at which there is a cusp in the susceptibility. There is no time dependence in the theory because only equilibrium properties can be calculated.

One of the objections to the EA theory is that it is a mean field theory which can predict transitions where there are none, e.g. a ferromagnetic alloy system below

its percolation concentration. Sherrington and Kirkpatrick (SK) (1975) hoped to remedy this by treating a situation where mean field theory should be exact, for example an Ising spin system with infinite range interactions distributed as in the EA theory. They also allowed the mean interaction to be nonzero. The solution obtained was similar to the EA theory but, as it was supposed to be exact, the occurrence of a negative entropy at low temperatures showed that something had gone wrong. Kirkpatrick and Sherrington (1978) also performed Monte Carlo tests on systems of up to 800 spins to compare with the SK theory. The Monte Carlo simulations produced values for the ground state internal energy and entropy significantly different from the theory (see Fig. 15). The entropy at 0 K is predicted by the simulation to be a satisfyingly physical zero. The tests could also simulate system dynamics, and in particular the time dependence of q, which was found to decay very slowly in the spin glass phase and not to have stabilized before effects due to the finite number spins became important. Binder (1977) also saw a slow decay of q in a similar Monte Carlo study of Ising spin glass systems.

The problem with the EA and SK solutions was pointed out by de Almeida and Thouless (1978). To calculate the free energy of the system a 'replica trick' was used in which the average of the logarithm of the partition function over bond configurations was

$$\langle \ln Z \rangle_c = \lim_{n \to 0} \frac{1}{n} (\langle Z^n \rangle_c - 1),$$

where Z^n is the product of partition functions of n identical replicas of the system and $q = \lim_{n \to 0} \langle q_j^{\alpha\beta} \rangle$ with $q_j^{\alpha\beta} = S_j^\alpha S_j^\beta$ ($\alpha \neq \beta$ are replica labels). The problem appears to be that for the method to be simply applied, all the $\langle q_j^{\alpha\beta} \rangle$ should be identical, but for those below the SK critical temperature this is not so, and the effective field at each spin is no longer gaussian.

The beginning of a solution to this 'replica symmetry breaking' has been found by Parisi (1980) who, by crudely approximating the variation in q, obtained values of the low temperature entropy and internal energy of the SK model predicted by Kirkpatrick and Sherrington's (1978) Monte Carlo simulation.

Currently therefore it is not certain that even the highly simplified theoretical models undergo a phase transition, but the models are very useful in defining 'regions of behaviour'. For instance Gabay and Toulouse (1981) showed that for three-dimensional spins the transition from ferromagnetism to a spin glass phase is due to the freezing of the transverse components of the spins, although Cragg *et al.* (1982) identified this transition with problems of replica 'symmetry breaking'. Cragg and Sherrington (1982) also explored the spin glass phases for three-dimensional spins in the presence of uniaxial anisotropy and identified regions of transverse, longitudinal and mixed spin glass behaviour. It is true, however, that there is no spin glass phase exhibiting a nonzero EA order parameter which is free from the problems of replica 'symmetry breaking'.

6. Spin Glass and Long Range Order

Spin glass phases occur up to a composition corresponding to the establishment of long range magnetic order in many alloy systems. How the two phases join or merge is a very interesting problem.

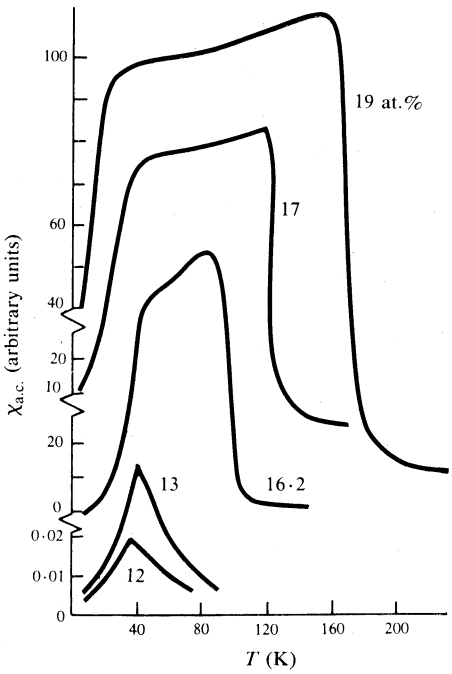


Fig. 16. Temperature variation of the a.c. susceptibility of *Au*Fe alloys with different iron content. [Coles *et al.* (1978).]

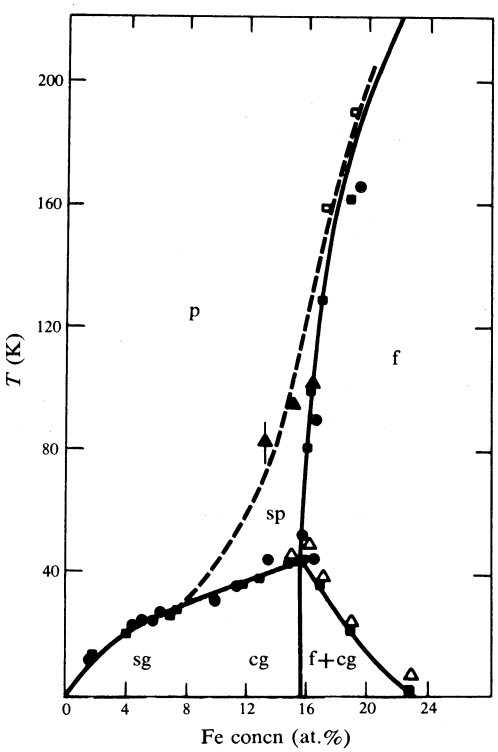


Fig. 17. Magnetic phase diagram for *Au*Fe alloys derived from susceptibility, neutron, specific heat and electron spin resonance measurements (f–ferromagnetic, p–paramagnetic, sp–superparamagnetic, sg–spin glass, cg–cluster glass). [Coles *et al.* (1978).]

The early solution of the SK model indeed predicted a transition from a spin glass phase to a ferromagnetic phase as the average interaction is increased from zero. It is interesting that a ferromagnetic to spin glass transition is expected as the temperature is *lowered* in a certain range of interaction. The de Almeida and Thouless (1978) correction to the SK solution shows that the ferromagnetic spin glass transition lies in the unstable region. Nevertheless, the question is raised whether one should expect the disappearance of spontaneous magnetization as the temperature is *lowered* in a composition range just inside the long range ordered phase.

The most extensively studied spin glass–ferromagnetic critical region is that of the *Au*Fe system (Coles *et al.* 1978). Fig. 16 shows low field a.c. susceptibility measurements spanning the critical concentration region. The changed character of the temperature dependence is clearly seen between 13 and 16·2 at.% *Au*Fe. The lower anomaly is continuous with T_g and the upper anomaly marks the Curie temperature. Fig. 17 shows the phase diagram assembled by Coles *et al.* (1978) from anomalies in various quantities detailed in the caption. It clearly shows a spin glass phase extending under the ferromagnetic phase.

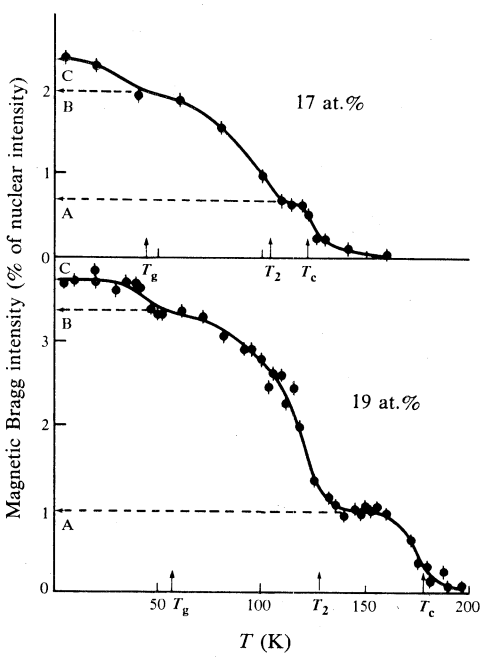


Fig. 18 Temperature variation of the (111) integrated magnetic Bragg intensity for two *Au*Fe alloys from neutron diffraction. [Murani (1980).]

The neutron experiment of Murani (1980) on 17 and 19 at.% *Au*Fe alloys identifies anomalies in the broad vicinity of the (111) Bragg peak with T_c and T_g. Fig. 18 shows the integrated intensity as a function of T. The high and low temperature anomalies are coincident with T_c and T_g. It is interesting to note that below T_g the integrated intensity still rises. This means that either Bragg peaks (reflecting the spontaneous ferromagnetic moment) increase at T_g, or other long range correlations (greater than $\sim$50 Å from the resolution used) grow. Murani also showed small angle neutron scattering results indicating that, as the temperature is lowered from 50 K, through T_g for the 17 at.% alloy, to 5 K, the correlations shorter than $\sim$400 Å grow in preference to those which are longer.

Gabay and Toulouse (1981) claimed that Murani's results vindicate their phase diagram of the spin glass–ferromagnetic interface (Fig. 19). They solved the Heisenberg spin problem equivalent to the Ising system treated by Sherrington and Kirkpatrick (1975). The M_1 phase has a spontaneous magnetization and freezing of the transverse components of the Heisenberg spins; M_2 is in addition unstable in the manner of that reported by de Almeida and Thouless (1978). In Fig. 18, T_2 is thought to mark the transition from the ferromagnetic to M_1 phase and T_g marks the transition from the M_1 to M_2 phase. However, it is not clear how any order parameters can be defined for the M_2 region for the same reason that the status of q in the EA and SK models is unclear. In addition M_1 has also been shown to suffer from replica symmetry breaking instabilities (Cragg *et al.* 1982).

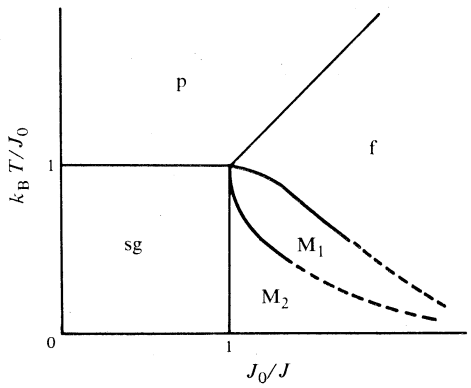


Fig. 19. Magnetic phase diagram of a spin glass system with three-dimensional spins as a function of average interaction J_0 normalized with respect to the standard deviation J. The phase M_1 is specified as having a nonzero EA order parameter in the transverse spin components, in the presence of a spontaneous moment. The phase M_2 has the replica 'symmetry breaking' instability (see Cragg *et al.* 1982) (p—paramagnetic, f—ferromagnetic, sg—spin glass). [Gabay and Toulouse (1981).]

Because Murani's results have three anomalous temperatures, Gabay and Toulouse identified these with the boundaries between f, M_1 and M_2. In addition Murani's results are taken as confirming the existence of a spontaneous moment below the anomaly at the lowest temperature. As we have seen Murani's results do not necessarily show this. In fact at low temperatures there appears to be a redistribution away from long correlation lengths.

A similar situation exists in the classic *Cu*Mn spin glass system. In this case the long range ordered phase is antiferromagnetic and does not occur until the manganese content is in excess of 70%. At this composition it is certain that the manganese moments are not localized. But again there is good evidence (Gibbs and Smith 1980) for the existence of a low temperature spin glass-like phase within what normally would have been regarded as the antiferromagnetic phase. Fig. 20 shows the susceptibility results for one of the alloys and Fig. 21 shows the phase diagram deduced. The reversible susceptibility has the same characteristics as for *Au*Fe alloys except that it shows a break at the Neel temperature T_N rather than a fall at T_c. The remanent magnetization and higher branch of the susceptibility are both again characteristic of a spin glass.

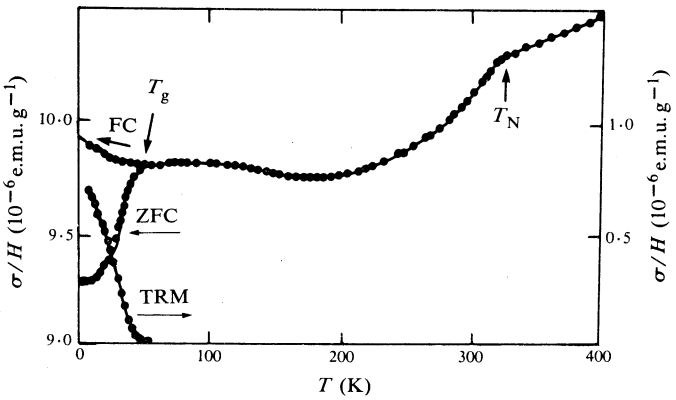


Fig. 20. The 'd.c.' susceptibility of an antiferromagnetic 83 at.% *Cu*Mn alloy showing the three branches typical of spin glass behaviour at low temperatures (see Fig. 13), where T_N is the Neel temperature. [Gibbs and Smith (1980).]

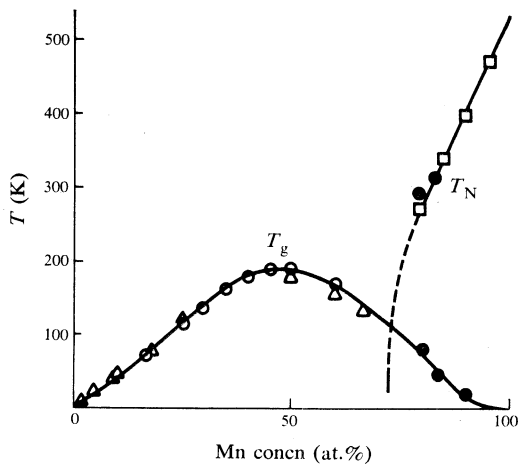


Fig. 21. Phase diagram for *Cu*Mn alloys. The spin glass behaviour spans the whole concentration range and long range antiferromagnetic order is established above ~70 at.% Mn. [Gibbs and Smith (1980).]

7. Conclusions

This paper has sought to bring together experimental and theoretical evidence for the nature of, and the nature of the transition to, the spin glass state. It has been restricted to looking at the low field susceptibility and magnetic neutron scattering cross section of metallic spin glasses.

The critical property used for the classification of phase transitions is, however, the specific heat, and the subject should not be left without reference to measurements of this property. No discontinuity has been detected in the specific heat or its temperature derivative, and typical of the results are those of Wenger and Keesom (1976) which show a very broad maximum above the glass temperature, and the very precise results reported by Fogle *et al.* (1981) on *Cu*Mn spin glasses. As must be clear from this paper it is extremely unlikely that we are seeing equilibrium properties even at laboratory times. It is therefore possible that if the experiments could be done slowly enough some discontinuity in the specific heat or its slope might be seen.

Whether or not the spin glass transition is a phase change therefore rests on very long time experiments which have very little chance of being definitive unless there is some clear method of extrapolation to infinite time. It all depends on whether some order parameter like the EA q parameter is nonzero in this limit.

Even if it were shown that a second order or higher transition is involved, the argument about whether it is a transition between two phases might not be over. As Pippard, writing in 'Elements of Classical Thermodynamics', puts it 'In a second order transition... the two phases (and at this stage we may doubt the wisdom of using this terminology to describe the two states on either side of the transition line) are identical in constitution, energy, entropy and volume'.

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Manuscript received 9 March, accepted 23 May 1983