

Spin-Glasses

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For about ten years now there has been a wealth of activity in an area of magnetism denoted by the catch-word "spin-glass". This popularity stems from the current interest in randomness and random materials and from the possibility of a novel phase transition which would provide a means to test and extend the theories of critical phenomena. Moreover, with the many unusual experimental effects observed in spin-glasses, a number of practical applications are becoming conceivable. Here we introduce the main concepts of a spin-glass and illustrate their general and fundamental significance.

Consider a collection of point magnetic moments or localized spins randomly distributed in a dilute magnetic alloy: *CuMn* or *AuFe*. In such systems, the magnetic exchange interaction $J(r)$ between pairs of moments is a damped, but oscillatory function of the distance r , i.e. ferromagnetic ($\uparrow\uparrow$) for a certain separation and antiferromagnetic ($\uparrow\downarrow$) as the distance increases. This "competition" among the interactions operating at various separations will cause a random alignment to result with respect to the assemblage of spin orientations. There is no preferred direction and no long range spatial order. Such a situation is illustrated in Fig. 1 for a two dimensional lattice with competing ferro/antiferro interactions. At high temperatures, $k_B T \gg J(r)$, thermal disorder causes spins to fluctuate locally resulting in a paramagnetic state, just as in any other magnet.

Now let us slowly cool our system down to low temperatures such that $k_B T < J(r)$. What will happen to the spin orientations as $T \rightarrow 0$ K? At first sight, our intuition tells us that as the temperature is reduced the spin system should

gradually freeze out, and finally at $T = 0$ K we shall have a static collection of randomly oriented spins. Here there should be no translational invariance so that the net magnetization is zero. However, the key part to the above question is how the system enters the frozen state and sometimes our intuition is only partially correct and we must turn to experiment to observe the situation more clearly.

Susceptibility

A good way to probe the freezing process is to measure the response function. For magnetic systems such as spin-glasses, we fortunately have the magnetization M and its conjugate magnetic field H . Thus the ratio $\chi \equiv M/H$ for $H \rightarrow 0$ defines the response or susceptibility of the system. To obtain maximal experimental sensitivity we usually measure the AC susceptibility in a small oscillating field $h \approx 0.1$ Oe at some audio frequency. In Fig. 2 we plot this generic AC susceptibility as a function of the temperature which spans a rather large range. Note the sharp cusp in $\chi(T)$ at a well-defined temperature, now called



Fig. 1 — A two-dimensional spin-glass analogue. The dashed lines indicate the local clustering.

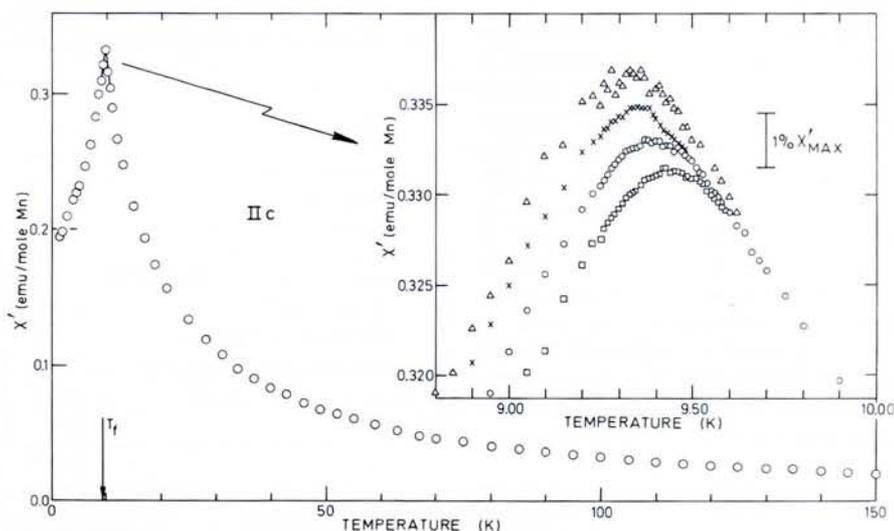
the freezing temperature T_f . This characteristic temperature increases with increasing concentration of magnetic atoms and the cusp becomes greatly rounded when even a weak external DC field is applied. Certainly the behaviour illustrated in Fig. 2 is typical of a phase transition and early theoretical models have treated the spin-glass freezing as a critical phenomenon leading to a new magnetic phase. So it would seem that in contrast to our original intuition we have here a sort of order out of randomness.

However, nature is not so simple, and more subtle effects can be discerned from a closer examination of the susceptibility cusp. As shown in the insert of Fig. 2, when the frequency ω is varied, there is a small, but significant, shift of the susceptibility curves with decreasing frequency, and T_f is a function of frequency, albeit a weak one with $\Delta T_f/T_f \approx 5 \times 10^{-3}$ per decade of frequency. This gives us a clue that a more complex, dynamic behaviour is occurring and the simple phase transition models using mean field approximations are not completely correct. Yet there is still this unique and unexpected cooperativeness in the freezing process exemplified by the cusp.

Specific Heat

In order to delve more deeply into the freezing processes we can measure another important thermodynamic quantity, namely the specific heat. Fig. 3 exhibits a set of magnetic (the other contributions have been subtracted) specific heat curves as a function of temperature for different applied DC fields. Here one should note, in contradistinction to $\chi(T)$, the smooth variation of $C_m(T)$, even in zero field, through the spin-glass freezing temperature. There is no clear criterion to determine T_f from specific

Fig. 2 — Zero-field AC susceptibility for *CuMn* (1 at. %). Insert: enlargement of the cusp region. Measuring frequencies: \square , 1.33 kHz; \circ , 234 Hz; \times 10.4 Hz; and \triangle , 2.6 Hz. After Mulder et al., Phys. Rev. **B23** (1981) 1384.



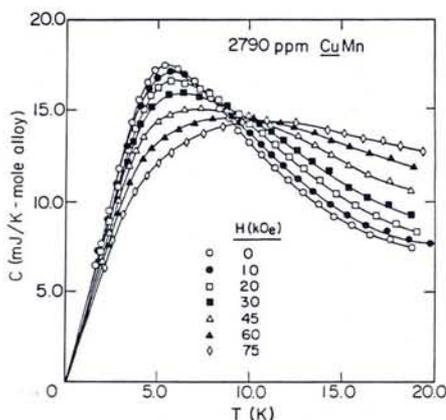


Fig. 3 — Temperature and field dependences of the magnetic specific heat for CuMn (0.3 at. %). Note that $T_f = 3.89$ K. After Brodale et al., *J. Magn. Magn. Mater.* **31-34** (1983) 1331.

heat measurements alone. Two other salient features are evident in Fig. 3. First, $C_m(T)$ is simply proportional to T at very low temperature. This linear behaviour, signalling a special type of low- T excitations (tunnelling) between configurations with almost coincident energies is a general characteristic of amorphous, glassy and random materials. Second there is the obvious high temperature tail which becomes even more pronounced as the DC field is increased.

This effect intimates that short-range correlations are beginning to occur far above (5 to 10 times) T_f and that the resulting "clustering" (see Fig. 1) is an important precursor to the freezing at T_f . So solely from the specific heat measurements, there are no indications of critical phenomena, just a lot of short-range order corresponding to a slow removal of the magnetic entropy. Now we need to construct a theory which incorporates the sharpness and dynamics of the susceptibility along with the gradualness and special excitations of the specific heat.

Other Systems — Other Experiments

Before moving on to sketching some of the basic theoretical approaches, we should emphasize the generality of the spin-glass phenomenon. The concept of a cooperative freezing creating a special ground state is applicable not only for magnetic behaviour where a few hundred different real systems (both metallic and insulating) have been claimed to be spin-glasses, but also for numerous non-magnetic analogues. For the former, the spin-glass state is now thought to be the third fundamental type of magnetic ordering after ferromagnetism and antiferromagnetism. For the latter, we can mention such diverse spin-glass analogues as dilute ferroelectrics, structural or orientational as in molecular

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crystals or ortho-para hydrogen, polar, and even at this time real window glasses. Furthermore, many of the ideas necessary to describe spin-glasses, e.g. a multiply degenerate ground state with very large numbers of degrees of freedom and various hierarchical levels of broken symmetry, are finding their way into biophysics and computer design.

Spin-glasses have also proved themselves to be a testing ground for many new experimental techniques. We refer to the important experimental investigations of spin-glasses using SQUIDS, EXAFS, muon spin relaxation, perturbed angular correlation and neutron spin echo. For certain spin-glasses, measurements have been performed on time scales from 10^{-12} to 10^5 s in fields of a few μ T to 40 T and at temperatures down to 0.1 mK. A great abundance of experimental data exists and many experimentalists are forced to interpret their results phenomenologically owing

to the complexity of the behaviour observed and the lack of a global theory.

Models and Theories

An ingenious way of modelling the real but complex situation illustrated in Fig. 1 is the Edwards-Anderson model. Here the random site occupancy of spins and their competing interactions are replaced by a regular spin lattice, i.e. every site has a spin. The spins S_i interact with each other through exchange couplings J with a Gaussian distribution $P(J)$, i.e. random bonds. For a Gaussian centred about zero there is an equal probability for ferromagnetic (+) as for anti-ferromagnetic (-) couplings.

A popular method for calculating the thermodynamic properties of this model is the Sherrington-Kirkpatrick approach which involves a mean field-like approximation where every spin interacts with every other spin, regardless of the distance, and the so-called replica trick

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where the logarithm of the partition function Z is calculated by constructing n -replicas of the system and using the identity

$$\ln Z = \lim_{n \rightarrow 0} (n-1)/n.$$

The results show a phase transition to occur with the appearance at the critical temperature of a spin-glass order parameter $q(T) \equiv \langle \langle S_i >_{\tau}^2 \rangle \rangle$, where first a thermal (T) average is performed, then squared, and configurationally averaged over the $P(J)$. Furthermore, the theory gives clear indications of this phase transition in both the susceptibility and the specific heat, the latter not being in agreement with experiment. Moreover, there was the non-physical result of a negative entropy arising at $T=0$. Since 1975 when these first theories appeared, many hundreds of theoretical papers have followed employing the above model and studying the properties of the mean field solution.

A few years ago, after enormous theoretical effort, the consensus was reached that the replica symmetry trick was invalid below a transition line relating H and T , and a scheme to break the replica symmetry was required. Recently a number of very clever schemes have been proposed and a new spin-glass order parameter function $q(x)$ has emerged in which the variable is rather difficult to interpret physically. In one such interpretation (Parisi), $q(x)$ is related to the probability distribution of the overlap of the magnetization in different states of the system. In another (Sompolinsky), a dynamical meaning is given to $q(x)$. Here $q(x) = \langle \langle S_i(0)S_i(t) \rangle \rangle_{\tau} >_{\tau}$ measures the amount of spin correlation which has not decayed after time t . And so today the controversy continues and no prevailing view has yet come forth.

On the other hand, the spin-glasses can be readily simulated via the Monte Carlo method. All that is needed is a very large or a special purpose (now being developed at Bell Labs.) computer. The results of many computer simulations over many years using a variety of starting models, even one mimicking the real

situation of Fig. 1, suggest no phase transition. Certainly there are large amounts of short range order and strong dynamical fluctuations, but no unambiguous critical phenomenon or order parameter has been obtained. Rather do these studies highlight the need to consider the dynamical properties of spin-glasses in order to arrive at a proper description. Most experimentalists are using a phenomenological "distribution of relaxation times" model along the above lines to describe their various measurements.

Distribution of Relaxation Times

Let us now consider this "experimentalist's" model as a first approximation to the freezing process. From the measured time correlations, e.g. the frequency dependences of the susceptibility shown in Fig. 2, we can define a relaxation time τ for a given spin. Since, except at the highest temperatures, the spins are randomly and non-uniformly interacting, a better approach necessitated by experiment is to determine a distribution of relaxation times $P(\tau)$ which evolves with temperature $P(\tau, T)$. In Fig. 4 we schematically sketch the qualitative character of such a $P(\tau, T)$ for a typical spin-glass. The true paramagnetic (non-interacting spins) regime ($T > 10 T_f$) is distinguished by a nearly unique relaxation time $\tau \approx 10^{-12}$ s. As T is lowered, local correlations begin to form between near neighbouring spins, and this biases the distribution function towards longer times. Then as $T \rightarrow T_f^+$ there is a sudden shift of $P(\tau)$ to much longer times due to longer-range, cooperative interactions. Finally, at T_f a very long-time tail appears. Here it should be mentioned that the local correlations usually become fixed, i.e. do not grow any further around T_f , and the dominant effects are mainly caused by the time dependences or relaxations of well-defined clusters. The extension of $P(\tau)$ to include static correlations ($\tau \rightarrow \infty$) signals the onset of the frozen spin-glass state.

The actual dependence of the shift

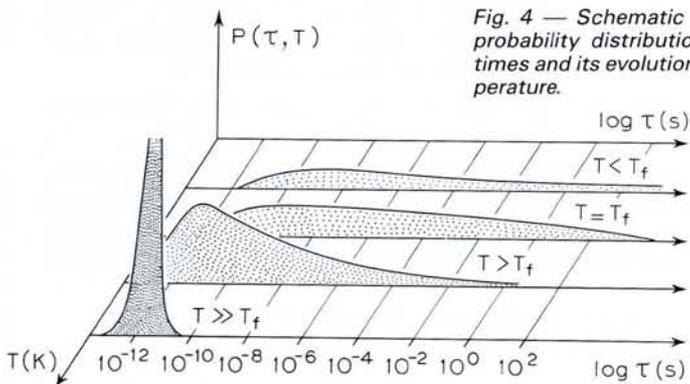


Fig. 4 — Schematic representation of the probability distribution for spin relaxation times and its evolution as a function of temperature.

and distortion of $P(\tau)$ on T is a function of the longer-range exchange couplings between the clusters and their anisotropy. For the metallic spin-glasses (CuMn , AuFe etc.) these are quite strong and we have a strong coupling limit where a sudden shift in $P(\tau)$ occurs in a small range of temperature near T_f . For a random superparamagnet with no interactions between the clusters, $P(\tau)$ is governed by the Arrhenius law:

$$\tau = \tau_0 \exp(E/k_B T)$$

and the distribution of the activation energy E , so a more gradual, completely "thermally activated" freezing occurs.

The frequency dependence of the AC susceptibility illustrated in Fig. 2 allows us to calculate $P(\tau, T)$ over the accessible frequency ($\omega \propto 1/\tau$) region. Conversely, the specific heat behaviour as shown in Fig. 3 reflects only the local spatial correlations, and in particular traces their growth as T_f is approached from very high temperature.

Conclusion

Based upon the preceding ideas, we can define a spin-glass as a random, mixed interacting magnetic system characterized by a sharp freezing behaviour at a well-defined temperature, below which a unique magnetic state exists without the usual types of long range order. This magnetic state has a variety of intriguing properties: irreversibilities, remanences, metastabilities, time dependences, and it can even coexist with the superconducting state. Unfortunately, a full theoretical description of the freezing process and this unique ground state is not presently available. However, with the intense interest in amorphous and random materials and the vast concentration of endeavour currently under way, we can certainly look forward to a more complete explanation of the spin-glass phenomenon. Additionally, ample possibilities exist for the practical applications of spin-glasses, not only in computer switching, memories and layouts, but also in the general modelling of random behaviour in, for example, biological systems and cosmology.

A final word has to do with the comparison of spin-glasses to real glasses. No doubt many properties are similar between these two different classes of material. So perhaps the choice of "glass" for the spin systems was not all that bad. Nevertheless, since much is also lacking in the theory of real glasses, it would be very advantageous to attempt a sort of theoretical symbiosis. The goal would be to describe as much random phenomena as possible through a unified approach.