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Spin glasses: model systems for non-equilibrium dynamics

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Abstract
Spin glasses are frustrated magnetic systems due to a random distribution of ferro- and antiferromagnetic interactions. An experimental three dimensional (3d) spin glass exhibits a second order phase transition to a low temperature spin glass phase regardless of the spin dimensionality. In addition, the low temperature phase of Ising and Heisenberg spin glasses exhibits similar non-equilibrium dynamics and an infinitely slow approach towards a thermodynamic equilibrium state. There are, however, significant differences in the detailed character of the dynamics as to memory and rejuvenation phenomena, and the influence of critical dynamics on the behaviour. In this paper some aspects of the non-equilibrium dynamics of an Ising and a Heisenberg spin glass are briefly reviewed and some comparisons are made to other glassy systems that exhibit magnetic non-equilibrium dynamics.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Dilute magnetic alloys exhibit a second order phase transition at concentrations of magnetic constituents well below the percolation limit. This transition—the spin glass transition—has, since its discovery [1], caused a lot of research activity and provided us with new magnetic phenomena. The conspicuous sharp cusp and frequency dependence of the ac-susceptibility is exemplified in figure 1 by measurements on the 3d Ising system $Fe_{0.5}Mn_{0.5}TiO_3$. In low field dc magnetization experiments, the spin glass transition is revealed by a maximum in the zero field cooled (ZFC) magnetization, an irreversibility between the ZFC and the field cooled (FC) magnetization and a continuous decay of the thermo remanent (TRM) magnetization to zero at the temperature where the irreversibility between the ZFC and FC magnetization appears (see figure 2). Static and dynamic scaling analyses of the critical behaviour support the occurrence of a second order phase transition for both 3d Ising and Heisenberg spin glasses. As to the dynamics, it is of certain interest to note that, in contrast to ordinary ferro- or antiferromagnetic

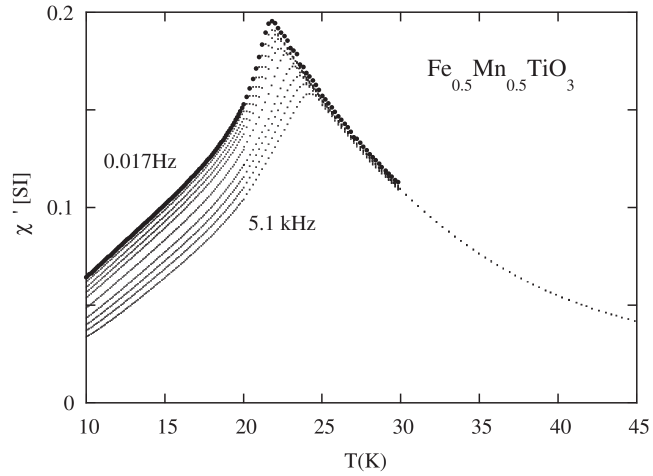

Figure 1. The ac-susceptibility of $Fe_{0.5}Mn_{0.5}TiO_3$ at logarithmically evenly spaced frequencies from 0.017 to 1.7 kHz (top to bottom).

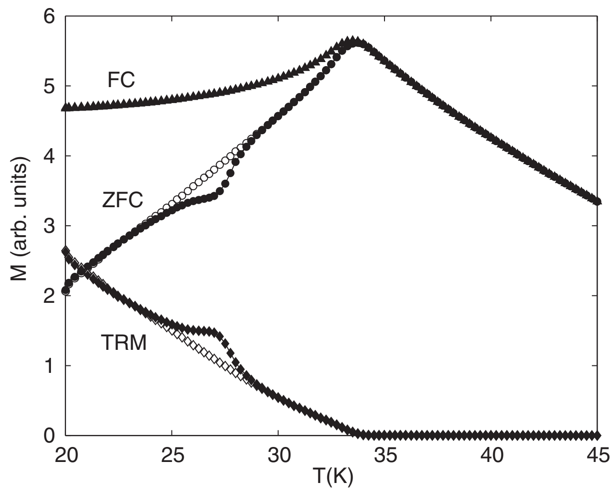

Figure 2. The ZFC, FC and TRM magnetization of Ag(11 at.% Mn) measured on continuous heating at the same rate after having cooled the system continuously (open circles) and after cooling it with stops for some hours at 27 K (filled circles). Reproduced from [2].

phase transitions, the character of the spin glass transition allows experimental determination of the critical slowing down exponent, $z\nu$, on the timescales of low frequency ac-susceptibility experiments. In addition, the low temperature spin glass phase owns intriguing non-equilibrium dynamics, which is the main subject of the current paper.

2. Critical dynamics

Experimentally it has been amply demonstrated that both Ising and Heisenberg systems exhibit dynamic critical behaviour on approaching the spin glass temperature T_g. Analyses of data from low frequency ac-susceptibility experiments indicate a divergence of the relaxation time as the glass temperature is approached, with a critical exponent, $z\nu$, that is of order 8–10. However, when comparing results from different experimental systems, it is yet not possible to distinguish significantly different values of the exponents on systems of different spin dimensionality. To illustrate the behaviour we quote analyses of the slowing down of the dynamics close to T_g of the model Ising system $Fe_{0.5}Mn_{0.5}TiO_3$ [3, 4]. The result of these studies is that the dynamic exponent $z\nu$ should be about 10 for the 3d Ising system. Quite similar values are also obtained from analyses of measurements on dilute magnetic alloys (Heisenberg systems) as well as on strongly interacting magnetic nanoparticle systems [5] (super spin glasses [6]). It is striking to learn from these experiments that the disordered and frustrated spin glass allows studies of critical behaviour at timescales and reduced temperatures that, for a pure ferro- or antiferromagnetic system, would reach far from the critical region. One could then expect that the critical behaviour of spin glasses should yield very reliable and unambiguous results as to the values of the critical exponents and possible differences between systems of different universality class. This is however not the case, due to strong limitations in how close to the transition it is possible to perform meaningful experiments; the slowing down of the dynamics and entrance into non-ergodic regions limit the studies to only orders of 0.01 in reduced temperature. The conclusion from dynamic scaling analyses is that there is a spin glass transition in a zero applied magnetic field, but is this also true in an applied magnetic field?

2.1. In-field criticality?

To understand spin glasses one needs useful and predictive modelling of the systems. There are two main approaches to describe spin glasses—starting from phase space (mean field models) and real space (droplet scaling models). The mean field model predicts a finite spin glass temperature and a persistence of the phase also in applied magnetic fields, the spin glass and the paramagnetic phase being separated by the AT-line [7]. The droplet scaling theory of 3d Ising spin glasses on the other hand predicts that, in the thermodynamic limit, any finite magnetic field destroys the spin glass phase, i.e. the in-field thermodynamic equilibrium phase is paramagnetic. Within this picture, this does not imply that critical dynamics cannot be seen in weak enough magnetic fields. A crucial point in this model is the correspondence between time and length scales, i.e. an experimental probe that measures at a certain frequency or timescale also probes the system on a length scale set by the observation time (and the temperature). A finite field sets an upper limit to the correlation length in the spin glass: on shorter length scales the system appears unaffected by the field and on larger length scales the system is at equilibrium (paramagnetic). The ac-susceptibility measured at low fields is dynamically limited by the frequency, ω, of the varying ac field and the susceptibility (at observation time, $t = 1/\omega$) increases with decreasing frequency. At temperatures above the freezing temperature (cusp in the real part of the susceptibility), the susceptibility becomes frequency independent on lowering the frequency. When a dc field is superposed on the system, the ac-susceptibility is unaffected if the field is weak enough. However after a certain field magnitude, the susceptibility cusp becomes suppressed and the freezing temperature is pushed to lower temperatures. However, at lower temperatures, the ac-susceptibility remains unaffected, i.e. one cannot distinguish the in-field from the zero field data (see figure 3). An analysis of the in-field slowing down of the dynamics of the 3d Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$ suggests that critical slowing down does not describe the behaviour, and that the spin glass transition is destroyed by any magnetic field [4]. It should, in this context, be mentioned that a careful study using torque magnetometry of the spin glass transition on systems with varying degrees of anisotropy is supportive of the existence of an in-field phase transition

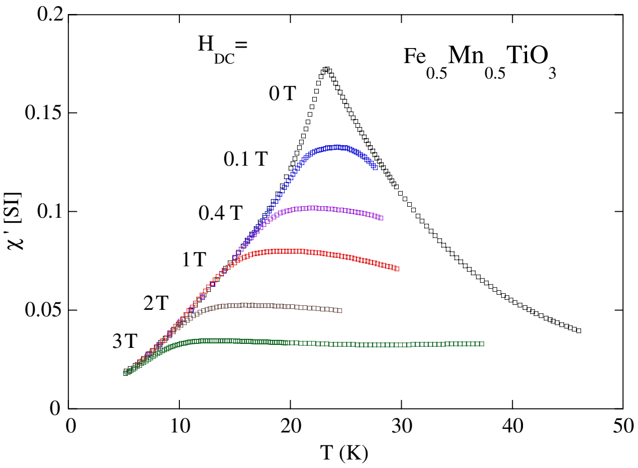

Figure 3. The ac-susceptibility of $Fe_{0.5}Mn_{0.5}TiO_3$ measured in a weak 125 Hz ac-field in superposed dc magnetic fields as indicated in the figure.

for Heisenberg spin glasses [8]. Such a difference between the in-field behaviour of Ising and Heisenberg systems is of course remarkable, and is not expected within a droplet scaling picture of the spin glass.

3. Non-equilibrium dynamics

At temperatures below the spin glass temperature, the zero field spin glass never reaches equilibrium; the equilibration time is infinite. Experiments on this phase are thus looking at a non-equilibrium system and the results are age dependent. The non-equilibrium character can be experimentally observed from an age dependence of the magnetic response [9]. In certain temperature ranges the ageing imposes a magnetic response of instructive and almost simplistic character. There is an inflection point in a plot of the magnetization (M) relaxation versus log t at an observation time equal to the wait time, t_w, before the application (or removal) of a weak magnetic field, and the relaxation rate ($S(t) = (1/H)\,\mathrm{d}M(t)/\mathrm{d}\log t$) exhibits a maximum at the same observation time. This behaviour reveals much of the physics behind the ageing phenomenon in spin glasses and together with results from experiments according to somewhat altered thermal protocols it has been possible to relate effective ageing times to the sizes of equilibrium spin glass domains. This length-timescale (droplet model) approach to the spin glass problem will be adopted below to interpret the non-equilibrium dynamics as measured in low frequency ac-susceptibility and dc-magnetic relaxation experiments.

The timescale for spin reversal of an atomic spin is about 10^{-13} in spin glasses. On approaching the glass temperature, correlation between spins occurs on longer and longer length scales and the response time of the system diverges at T_g. At lower temperatures the response time again decreases according to critical slowing down, however, the random spin structure that is obtained after a quench is not compatible with the competing interaction pattern, and there is a slow restructuring of the system on larger and larger length scales. In terms of the droplet scaling model this is governed by thermal activation over an energy barrier that increases with decreasing temperature and the growth occurs logarithmically in time [10]. However, the equilibrium configuration at one temperature is different from the equilibrium structure at any other temperature (temperature chaos).

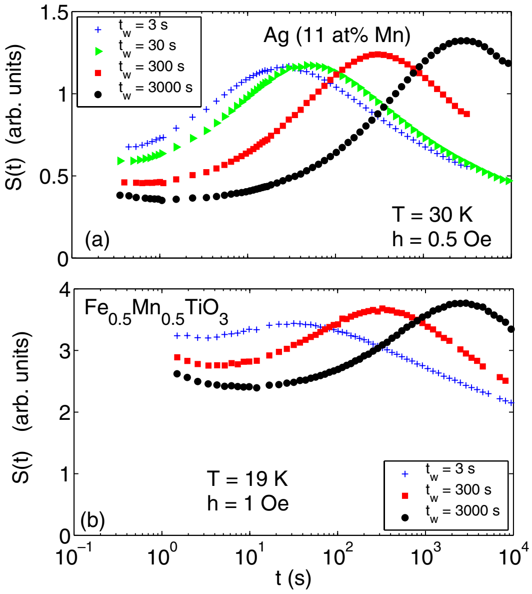

Figure 4. Relaxation rates for the ZFC relaxation of Ag(11% Mn) and $Fe_{0.5}Mn_{0.5}TiO_3$ after different wait times at the measurement temperature. The experiments are in both cases performed at $T/T_g = 0.9$ [11].

3.1. Isothermal ageing

Quenching the system from a temperature above T_g to a measurement temperature may seem to be a simple procedure, but experimentally, quenching in a magnetometer implies cooling rates of only some kelvin per minute. This is of course a slow cooling compared to a spin flip time of only 10^{-13} s. We are, however, fortunate in the sense that experiments in a magnetometer system are also confined to long timescales compared to the spin flip time. Figure 4 shows results from an isothermal ageing experiment on an Ising and a Heisenberg system; plotted in the figure is the magnetization relaxation and the relaxation rate versus time after the field application in a semi-logarithmic diagram. Both systems show a maximum relaxation rate at a time of the order of the wait time before applying the magnetic field. A striking difference between the behaviour of the two systems is that the ageing has a much larger influence on the relaxation of the Heisenberg system than of the Ising system. In the measured time window, the relaxation rate rises a factor of about five above an estimated equilibrium relaxation rate in the Ag(Mn) spin glass, but only a factor of less than two for the Ising system. Additionally, for both systems it does not matter at what temperature we choose to make our measurement; an ageing behaviour is always observed. In fact, provided the temperature step is large enough, one can make two sequential stops at a higher and a lower temperature, and the observed ageing at the lower temperature is the same as if the system was immediately cooled from above T_g without the stop. The ageing is non-accumulative in both systems, however a larger separation between the two temperatures is required for the Ising than the Heisenberg system.

3.2. Memory

The non-accumulative nature of the ageing and a preserved memory of the cooling process is best illustrated by a zero field cooled magnetization versus temperature experiment under two different cooling protocols. The behaviour has already been introduced in figure 2, where the low field ZFC, FC and TRM magnetization of the Ag(Mn) sample was plotted versus temperature. The protocol including a stop at a certain temperature shows in the ZFC case a rather narrow dip that has a logarithmically increasing depth with increasing stop time. A corresponding, but rather more shallow and broader dip is observed in experiments on the Ising sample. A memory of the spin structure attained at the stop temperature is imprinted and preserved during the succeeding cooling–heating process. The understanding of the non-equilibrium dynamics may be further elucidated by low frequency ac-susceptibility using the same thermal protocol as in the dc-magnetization studies described here. Using such studies, the temperature memory behaviour [12] was first explicitly reported by Jonason *et al* [13] in the measurement of the out of phase component of the ac-susceptibility versus temperature on a $CdCr_{1.7}In_{0.3}S_4$ spin glass.

Using the functional form of the decay of the out-of-phase component of ac-susceptibility under isothermal ageing at different temperatures and frequencies, a quantitative measure of the increasing spin glass domain size in a Ag(Mn) spin glass in terms of the droplet scaling model has recently been derived by Jönsson *et al* [14]. One ingredient of the description of the ageing behaviour that these and other experimental results imply is the existence of an overlap on short length scales between a spin glass domain structure developed at one temperature, and the domain structure at a different temperature $T_m + \Delta T$. The concept accounting for this is the overlap length [15], the validation and influence of which can be further studied by experiments after applying perturbations (temperature steps or cycles) to the system after an ageing period.

3.3. The overlap length and ghost domains

There is quite extensive literature on the influence of perturbations (temperature, field or bond) on spin glass dynamics both from the point of view of experiment and simulation. One recent theoretical development of the droplet model introducing the ghost domain concept [16] provides a visual illustration of how an original domain structure is recovered after a perturbation that intuitively should destroy any memory of the preceding ageing process. The procedure and process are illustrated in figure 5 for bond perturbations on a 2d Mattis model. In terms of a temperature cycling experiment, the important features are: the system is first aged at a constant temperature and spin glass domains of 'up' and 'down' sign grow. After a wait time at this temperature, the temperature of the system is shifted and the system is allowed to evolve under these new conditions. At this temperature, domains start to grow, starting from the overlap length and having a completely different structure than that of the original domains on larger length scales. The temperature is then shifted back to the original one. However, at this temperature, a (ghost) pattern of the original domain structure remains and the perturbation is seen as patches of the wrong domain kind within the original domains. Keeping the system at this temperature, the patches are gradually washed away and simultaneously the original domains continue to grow. The experiments that reflect and promote that this kind of internal processes occur are magnetic relaxation and relaxation of the ac-susceptibility recorded after the perturbation protocols [11].

4. Conclusions

The non-equilibrium dynamics of spin glasses is generic to both Ising and Heisenberg systems, but with some significant differences in the detailed behaviour. In a recent article by Bert *et al* [17] a systematic study of the effects of perturbations on the ageing behaviour of spin glasses

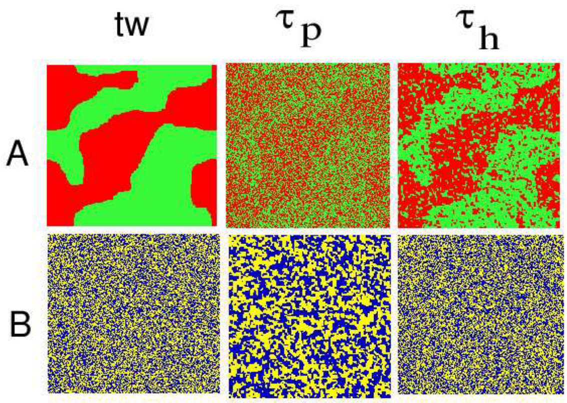

Figure 5. The evolution of domains in a 2d Mattis model [16] during a bond perturbation experiment. The system is allowed to evolve under two different interaction patterns having completely different ground states (A and B). The spin structure of the system is projected onto the ground states A and B at three different stages of a perturbation simulation. Left figures: after the first wait time under interaction pattern A; grown plus and minus domains are seen in the projection on A, whereas the projection on B yields a random pattern. Middle figures: after the perturbation under conditions B, a domain pattern has evolved in the projection onto B, and in the projection onto A one can still resolve the sign and size of the original domains (ghosts), but in a lot of random noise. Right figures: after evolving under the interaction pattern A again, the noise in the projection onto A is largely washed away and the original domain structure recovered. Reproduced from [11].

with different anisotropy ranging from purely Ising to closely isotropic systems was reported. Also other magnetic systems show spin glass like non-equilibrium dynamics; characteristic of Ising systems is the critical dynamics [5] and memory behaviour [18] of a near monodispersed interacting nanoparticle system, whereas certain features of the ageing and memory behaviour of a high temperature superconductor showing the paramagnetic Meissner effect [19] are more Heisenberg like.

Acknowledgments

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