The field cooled state of canonical spin-glass revisited

Sudip Pal, Kranti Kumar, A. Banerjee, S. B. Roy
UGC DAE Consortium for Scientific Research
Indore-700001, India

A. K. Nigam
Tata Institute of Fundamental Research
Mumbai-400005, India

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Canonical spin-glass (SG) is an enigmatic system in condensed matter physics. In spite of the intense activities of last five decades several questions regarding the nature of the SG phase transition and the SG ground state are yet to be resolved completely. In this backdrop we have revisited the field cooled state of canonical spin-glass. We have experimentally studied magnetic response in two canonical spin-glass systems AuMn(1.8%) and AgMn(1.1%), both in the field cooled (FC) as well as zero field cooled (ZFC) state. We show that the well known magnetic memory effect, which clearly established earlier the metastable nature of the ZFC state in SG, is also present in the FC state. The results of our experimental study indicate that the FC state also is a non-equilibrium state, and hence the energy landscape involved is a non-trivial one. This in turn seriously questions the picture of spin-glass transformation as a second order thermodynamic phase transition.

Spin-glass (SG) transition in magnetic alloy systems is a very interesting phenomenon in condensed matter physics and the concept has found applications in various other areas like colloids, granular media, structural glass, neural networks etc. Over the years there have been two distinct approaches to understand this SG transition in canonical spin-glass systems. First, the Parsi’s solution of infinite range Sherrington-Kirkpatrick (SK) model that predicts a thermodynamic second order phase transition from high temperature paramagnetic state to low temperature spin-glass state. Within this model cooling the system in presence of a finite field from paramagnetic state would bring the spin-glass to the “infinite time equilibrium state”. The other model, known as a short range droplet model, predicts a SG transition only at zero magnetic field (H) and absence of any phase transition for non-zero H in any finite dimension. Numerical simulations have found a SG phase transition in presence of external (random) field only above an upper critical dimension of nearly six; the debate, however, is yet to be resolved completely.

There exist some experimental reports, which showed that the field cooled (FC) magnetization being sensitive to the cooling rate and the time decay of thermoremanent magnetization depended on the waiting time before the cooling field was removed. This is in contradiction to the picture of a thermodynamic spin-glass transition to an equilibrium state. However, the FC state of spin-glass has not been subject of intense investigations as much as the zero field cooled (ZFC) state so far, presumably due to the popularity and the implicit acceptance of thermodynamic phase transition picture. In this light it is necessary to revisit the FC state of the canonical spin-glasses and subject it to fresh experimental scrutiny. Over the period of time memory effects observed in various glassy systems have emerged as effective experimental technique to characterize the energy landscape of such systems.

A system in equilibrium state does not show memory effect, whereas a system with rugged energy landscape can store the memory of a previous state passed while cooling, and it affects the physical properties of the system during the subsequent re-heating. The ZFC state of a canonical spin glass shows memory effect in dc magnetization measurement, thus clearly highlighting the metastable nature of this state. In the present work we have investigated the memory effect in two well-known canonical spin-glass systems AuMn(1.8%) and AgMn(1.1%) through dc magnetization measurements. In particular, we have studied the nature of the FC state of these canonical SG systems through the observation of memory effects in low field dc magnetization. We find that along with the ZFC state, the FC state also shows clear signature of memory effect below the spin-glass freezing temperature. This highlights the non-equilibrium nature of the FC state in canonical spin-glasses AuMn(1.8%) and AgMn(1.1%).

AuMn(1.8%) and AgMn(1.1%) samples have been prepared by induction melting in Argon atmosphere. Further details of the sample preparation and characterization have been reported earlier.

Magnetization measurements have been carried out in 7 Tesla SQUID magnetometer (M/S Quantum design, USA) using Reciprocating Sample Option (RSO) transport with scan length of 4 cm both in the ZFC and FC mode. In the ZFC mode the sample is cooled to the lowest measured temperature (here 2 K) before the applied magnetic field (H) is switched on, and the measurement is made while warming up the sample. In the FC mode the sample is cooled to the lowest measured temperature in the presence of applied H and the measurement is then made while warming up the sample. We have performed our measurements on both the samples and obtained qualitatively similar results. Therefore, for the sake of conciseness we will present data mainly for AuMn(1.8%) system.

AuMn(1.8%) and AgMn(1.1%) undergo the spin-glass
FIG. 1. Magnetization versus temperature plots for AuMn(1.8%) and AgMn(1.1%) in applied magnetic fields $H = 200$ and $100$ Oe, respectively.

FIG. 2. Variation of irreversibility temperature $T_{ir}$ in $H$-T plane for AuMn(1.8%). Red line is the linear fit to the data.

FIG. 3. Zero field cooled (ZFC) magnetization memory effect in AuMn(1.8%) at $H_m = 200$ Oe. In one experiment the magnetization was measured under standard ZFC protocol (see text for details). In the other cooling was stopped and temperature kept constant at $T = 3$ and $5$ K for 3 hours while cooling in zero field.

or spin freezing transition around $T_f = 7$ and $5$ K respectively as evident from the cusp in the ZFC magnetization curve as shown in Fig. 1. The $T^*$s obtained in the present study agrees quite well with the earlier reported results. The ZFC and FC $M(T)$ curves bifurcate below an irreversibility temperature, $T_{ir}$ which lies slightly below $T_f$. As we increase the applied field (data not shown here), both $T_{ir}$ and $T_f$ gradually shift towards lower temperatures and the difference between FC and ZFC magnetization simultaneously reduces. The cusp at $T_f$ in the ZFC curve also broadens with applied magnetic field ($H$). In Fig. 2, we have shown the variation of $T_{ir}$ with respect to $H$ for the sample AuMn(1.8%). It approximately varies according to $T_{ir} \sim H^{2/3}$ showing the existence of de Almeida - Thouless (AT) line in the $H$-$T$ plane as predicted by the mean field theory of spin-glass.

In Fig. 3, we present the memory effect observed in AuMn(1.8%) following standard ZFC procedure. Here we have first cooled the sample in the absence of any magnetic field down to $2$ K and then measured the magnetization of the sample while heating in the presence of an applied $H$ of $200$ Oe. This provides the reference ZFC magnetization curve. The sample is then cooled again in zero magnetic field but with temporary halts at $T_m = 5$ K and $3$ K for 3 hours. The measurement of magnetization is then performed as before while heating. The magnetization shows distinct deep around each $T_m$ before merging with the reference ZFC curve at higher temperatures (see Fig.3). Thus it reflects the strong memory of cooling history and highlights the metastable nature of the ZFC state of the spin-glass. A model of complex hierarchical distribution of multiple potential minima in the free energy landscape has been widely used to explain these features. According to this model, potential minima are separated by finite energy barriers and get divided into more number of minima as temperature decreases. As the temperature decreases, barriers become more effective due to reduced thermal energy, and the heights of some of the barriers also increase. During the waiting time, the system gradually creeps through the lower energy states available nearby, which is known as “aging”. When cooling is resumed after aging, the system “rejuvenates” back to the earlier state and magnetization shows
a finite dip in the following reheating curve (see Fig. 3). Recently, it is further reported that, the application of a small magnetic field for finite time while zero field cooling, also results in memory effect. It indicates that a spin glass also remembers the evolution of its state due to external magnetic field. Here, we have used a different protocol to find the effect of field cycle on the ZFC memory, which is shown in Fig. 4. In this protocol, we have initially cooled the system in zero field down to 2 K, and applied the measuring field, \( H_m = 200 \text{ Oe} \) and then began recording the ZFC curve. At an intermediate temperature, \( T_m = 4 \text{ K} \) (< \( T_f = 7 \text{ K} \)), cooling was temporarily paused and the field was increased to 400 Oe for next \( t_w = 1.5 \text{ hour} \) and again reduced back to 200 Oe and warming was resumed. After the field cycle, magnetization starts from a higher value and merges back to the reference ZFC curve on further heating (see Fig. 4). During \( t_w \), magnetization increased with time, and this increase could be fitted with stretched exponential function (see the inset of Fig. 4, black curve). On the other hand, in another measurement where the field is reduced from \( H_m = 200 \text{ Oe} \) to zero at \( T_m \) instead of increasing it, magnetization starts form a lower value, however very small (as shown in the Fig. 4, red dotted circle,) and merges back with the reference curve. These measurements clearly show that a spin glass remembers the state that is perturbed by an external field. It is, however, interesting to note that both increase and decrease in field by same amount perturb the state by nearly same amount (evident from amount of relaxation, inset of Fig. 4), but increase in field causes a larger discontinuity.

The memory effect, we have followed two protocols. We have cooled the system from the paramagnetic state to \( T_m \), which is less than the respective \( T_{ir} \), the irreversibility temperature of the sample for the applied field \( H_m \). Then, in our first protocol, we have reduced the field to zero and after a waiting period of \( t_w = 1.5 \text{ hour} \), the field \( H_m \) is reapplied and cooling of the sample is resumed taking it down to 2 K and then subsequently reheated. In the second protocol, at \( T_m \) instead of reducing the field to zero, it is increased to 400 Oe and kept fixed for next 1.5 hour and then reduced back to \( H_m = 200 \text{ Oe} \). Then the cooling in presence of 200 Oe is resumed and subsequently re-heated. In both protocol we see a discontinuity in the cooling curve due to the field cycle. However, in the first protocol, magnetization after the field cycle (and wait for 1.5 hour) is smaller than the respective M value before performing the cycle. The subsequent warming curve also shows discontinuity above \( T_m \) and finally merges with the reference warming curve showing the memory effect. Whereas, in the second protocol, in

![FIG. 4. Zero filed cooled (ZFC) magnetization memory effect in AuMn(1.8%) at \( H_m = 200 \text{ Oe} \). While warming after zero field cooling, at \( T_m = 4 \text{ K} \), H is increased to 400 Oe (black filled circles) and reduced to zero (red circles) and wait for \( t_w = 1.5 \text{ hour} \). The field is then returned back to 200 Oe and heating is continued. Inset shows the aging of the spin glass during waiting time.](image1)

![FIG. 5. Field cooled (FC) magnetization memory effect in AuMn(1.8%) at \( H_m = 200 \text{ Oe} \). (a) at \( T_m = 4 \text{K} \), field is reduced from 200 to 0 (remnant) Oe (b) field is increased to 400 Oe for a time period of \( t_w = 1.5 \text{ hour} \) and returned back to \( H_m = 200 \text{ Oe} \). Then the system is subsequently cooled and re-heated.](image2)
which the field was increased from $H_m = 200$ to 400 Oe, M again shows a discontinuity but in this case, M after the field cycle is larger than the M before the cycle. Here also the consequent warming M-T cycle shows memory effect and merges with the reference M-T curve.

The above results clearly indicate that even in the FC state a spin-glass remembers the earlier history. According to the short range interaction model of SG, FC state is a paramagnetic state. Whereas, in the Mean field model, the field cooling brings the system to an equilibrium SG state. In the later case, the system is not supposed to have any memory effect, for example a long range ferromagnetic state reached from a high T paramagnetic state through a second order phase transition does not show any memory effect although it has paramagnetic state through a second order phase transition and droplet model, a cluster model of spin glass has also been considered for canonical spin glass systems.

In these systems, the memory effect arises from the wide distribution of relaxation time originating because of competing interaction between magnetic clusters as well as distribution in cluster size. It may be noted here that apart from the two main stream theoretical models namely SK model and droplet model, a cluster model of spin glass has also been considered for canonical spin glass systems. According to this model, the long range interactions between spins may encourage formation of the correlated spin clusters even above the spin freezing temperature. These clusters form at relatively higher temperature primarily due to concentration fluctuation of impurity spins and they grow in size with decreasing temperature. Below the characteristic temperature, $T_f$, an infinite cluster forms along with many other finite clusters with varying size and shape. Within one cluster each spin is connected with at least one of the other spins in the same cluster. This kind of system has broad distribution of relaxation spectrum and therefore can possibly be useful in explaining the observed memory effects.

In conclusion, we have studied memory effect in the ZFC and FC state of two canonical spin glass systems AuMn(1.8%) and AgMn(1.1%) through dc magnetization measurements. We find that ZFC state shows clear memory effect which is reminiscent of the rugged free energy landscape of spin glass. In addition, distinct memory effect is also observed in the FC state. Change in applied magnetic field perturb the FC state at a temperature $T_m$ below $T_{irr}$. In the subsequent re-heating cycle the system rejuvenates back to the earlier state above $T_m$ signifying that the FC state of a canonical spin glass also contains a rugged energy landscape, which is not consistent with the picture of a thermodynamic equilibrium state. This possibly indicates that the theoretical understanding of spin-glass phenomena is yet to be complete, and the cluster model for spin-glass may need some reconsideration for accounting such experimental observations of metastable behavior.

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