“Glassy Dynamics” in Ising Spin Glasses — Experiment and Simulation

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The field-cooled magnetization (FCM) processes of Ising spin glasses under relatively small fields are investigated by experiment on Fe0.55Mn0.45TiO3 and by numerical simulation on the three-dimensional Edwards-Anderson model. Both results are explained in a unified manner by means of the droplet picture. In particular, the cusp-like behavior of the FCM is interpreted as evidence, not for an equilibrium phase transition under a finite magnetic field, but for a dynamical (‘blocking’) transition frequently observed in glassy systems.

KEYWORDS: Ising spin glass, field-cooled magnetization, droplet picture, glassy dynamics

Slow relaxational dynamics of nonexponential type has been observed in various disordered systems, and is now called ‘glassy dynamics’.1 The most interesting slow dynamics is the one observed in systems, such as structural glasses and spin glasses, whose relaxation processes are determined cooperatively through interactions between constituent elements of the system. In this case, the distribution of relaxation times exhibits a peculiar temperature dependence besides the one attributed to the ther- modynamic origin which, at the same time, involves thermal ‘blocking’ at low temperatures. We argue in the present letter that the field-cooled-magnetization (FCM) process in Ising spin glasses is one typical example of such ‘glassy dynamics’.

In spin-glass (SG) studies, one of most notable problems yet unsettled concerns the stability of the equilibrium SG phase under a finite magnetic field h.2 This is the case even for Ising spin glasses which are the conceptually most simple SG systems. The mean-field theory predicts the stability of the SG phase up to a certain critical magnitude of h specified by the de Almaida-Thouless (AT) line,3 while according to the droplet theory,4–6 the SG phase is unstable even in an infinitesimally small h. Recently, Takayama and Hukushima (TH)7 have carried out numerical simulations of the field-shift aging protocol on the three-dimensional (3D) Ising Edwards-Anderson (EA) model, the results of which strongly support the droplet picture. They have claimed that the AT-transition-like behavior observed experimentally8 is well interpreted as a dynamical crossover from the SG behavior to the paramagnetic one upon application of a field h.9 Further numerical works in favor of the droplet picture have been reported more recently.10,11

Since early SG studies,12 it has been observed that the zero-FCM (ZFCM) deviates from the FCM at a certain temperature, denoted here as \( T_{\text{irr}} \), and that the FCM exhibits a cusp-like behavior at a certain temperature, denoted here as \( T^* \), below which the FCM becomes nearly independent of \( T \). Here, the ZFCM (and FHM used below) is the magnetization measured in a reheating process after a ZFC (FC) process. Without consideration of cooling/heating rate effects, these phenomena have often been considered to be evidence for the SG phase transition at \( T_{\text{irr}} \approx T^* \) under a finite field. The main purpose of the present study is to address this problem by performing detailed experiments on the Ising spin glass Fe0.55Mn0.45TiO3 with \( T_c \approx 22.3 \text{ K} \),13 by extending the Monte Carlo simulation on the same Gaussian Ising EA model as in the study of TH with \( T_c \approx 0.95 \).14,15 (in units of the variance of interactions), and by comparing the results of the two analyses. The consequence is that the above-mentioned FCM behavior, a most fundamental phenomenon in spin glasses, can in fact be interpreted as the ‘glassy dynamics’ in the sense described above.

Let us begin with two sets of FCM, FHM and ZFCM curves, measured with two different cooling(heating) rates, and shown in Fig. 1(a). It is clear that \( T_{\text{irr}} \), the temperature at which the irreversibility between the FCM and ZFCM occurs, does depend on the cooling rate. Furthermore, we can see that \( T_{\text{irr}} \) is significantly higher than \( T^* \), the cusp-like temperature of the FCM.16 The corresponding FCM and ZFCM obtained in our simulation are presented in Fig. 1(b). The rate-\( n \) in the figure means the cooling process with \( n \) MC steps (mcs) at each \( T \), which is changed in steps of \( \Delta T = 0.01 \). When the rates are measured in units of \( T_c/t_0 \), with \( t_0 \) being the microscopic spin flip time (which is 1 mcs for simulation and 10−12 s for experiment), the rate of the process with rate-100 in the simulation is \( 1 \cdot 10^{-4} \), which has to be compared, for example, with \( 2 \cdot 10^{-17} \) of that in the experimental process denoted as 1.7 mK/s. The results in Fig. 1 imply that the occurrence of the irreversibility between the FCM and ZFCM corresponds by no means to a certain equilibrium phase transition.

At \( T > T_{\text{irr}} \), the FCM, which coincides with the
ZFCM, is the magnetization \( M \) in equilibrium under \( h > 0 \). The small deviation of this equilibrium \( M/h \) from the Curie law may be attributed to the nonlinear field effect in the simulation with \( h = 0.1 \) (see Fig. 4(b) below), and to a small nonvanishing Curie-Weiss constant for the experiment with \( h = 5 \) Oe. Here, we note that \( h = 0.1 \) in the simulation corresponds to the order of 1 T in the experiment on \( \text{Fe}_{0.55}\text{Mn}_{0.45}\text{TiO}_3 \). In this temperature range, the SG short-range order, represented by the SG coherence length \( \xi(T) \), and the corresponding correlation time, \( \tau(T) \), increase, and at \( T \approx T_{\text{irr}} \) the latter reaches the time scale proportional to the inverse of the cooling rate. Actually, if this proportionality constant is set to unity, and \( T_{\text{irr}} \) is estimated using the peak temperature of the ZFCM, our simulation results can be fitted to an expression of the critical slowing down

\[
\tau(T) \approx a_0 [(T - T_0(h))/T_0(h)]^{-z\nu},
\]

with \( a_0 \approx 30, z\nu = 11.0 \) and \( T_0(h = 0.1) \approx 0.81 \), where \( T_0(h) \) is a supposed transition temperature under a finite \( h \). Here, we don’t claim that \( T_0(h) \) is an equilibrium transition temperature. Instead, we want to emphasize that the SG short-range order starts to increase at temperatures as high as \( 2T_0 \), with the value of \( z\nu \) comparable to that obtained for the equilibrium transition under \( h = 0.13.17 \) We therefore consider that the SG correlation of the order of \( \xi^* \sim \tau^{1/z}(T_{\text{irr}}) \) has appeared at \( T \approx T_{\text{irr}} \) on average, and that locally correlated regions of relatively large size start to be thermally blocked around \( T_{\text{irr}} \). We call them spin clusters.

At \( T < T_{\text{irr}} \), not only the ZFC state but also the FC state is out of equilibrium. This is clearly seen in FCM processes, where the cooling is stopped at a certain temperature, denoted as \( T_{\text{stop}} \). As shown in Fig. 2, the FCM increases during a stop made in the temperature range \( T_{\text{irr}} > T_{\text{stop}} > T_c \) (\( T_{\text{stop}} = 22.8 \) K in the experiment and \( T_{\text{stop}} = 1.2 \) K in the simulation). Because of a fairly large dynamical exponent \( z \) in eq. (1), the size of the SG short-range order is not expected to appreciably increase further during the stop within this temperature range. Instead, spin clusters thermally blocked at around \( T_{\text{irr}} \) tend to equilibrate, or further polarize, yielding the FCM increase observed both in the experimental and simulation results. In an FC process without such a stop, spin clusters of a smaller size than the already blocked spin clusters are blocked, and at around \( T^* \), these blocked clusters are considered to come into contact with each other.

The observed FCM behavior at temperatures comparable to and lower than \( T^* \) can be interpreted in terms of the droplet picture,\(^4\)-\(^6\) where the field crossover (corre-
equilibrium SG phase below $T_\mathrm{c}$ by their size $L$: it is dominated by the Zeeman energy ($\sim \sqrt{q_{\mathrm{EA}}h^2 L^2/2}$) for $L > L_h$ and by the SG stiffness energy ($\sim TL^\theta$) for $L < L_h$. Here, $q_{\mathrm{EA}}$ is the SG order parameter, $d$ the spatial dimension, $Y(T)$ the stiffness constant of SG domain walls, and $\theta$ the gap exponent. Explicitly, $L_h$ is written as

$$L_h \sim \frac{(T_c - T_c)}{h^2 q_{\mathrm{EA}}^{\delta}} \left[\frac{(T_c - T)/T_c}{\text{cm}/h}\right]^{\delta},$$  \hspace{1cm} (2)

where $\delta = (d/2 - \theta)^{-1}$ and the exponent $\delta_{\text{eff}} (>$ 0)\textsuperscript{7,13} is given by the temperature dependence of $T$ and $q_{\mathrm{EA}}$.

We now apply this droplet picture to spin clusters grown in the nonequilibrium FC process. At temperatures higher than $T^*$ ($\approx T_c$), spin clusters are thermally blocked independently of each other, or in other words, the SG stiffness energy introduced above is not effective at all. Thus, the configurations of the spin clusters are overbalanced to the Zeeman energy. At and below $T^*$, the SG stiffness energy becomes effective and competes with the overbalanced Zeeman energy. We then naturally expect reorientation of spins, which results in the growth of new spin clusters with SG short-range order for a given set of $(T, h)$ within old spin clusters.\textsuperscript{18} We denote the mean size of the new spin clusters simply as $\xi$. The consequences of this restoration of the SG stiffness energy, or the growth of $\xi$, are not only the FCM cusp-like behavior at $T^*$ but also a decrease in FCM by further cooling the system below $T^*$. For the same reason, an \textit{initial decrease} in FCM is expected during a stop of the FC process at $T_{\text{stp}}$ below $T^*$. All of these expectations were in fact observed in the experiment as can be seen in Figs. 1(a) and 2(a). In the simulation, the initial decrease in FCM at the stop at $T_{\text{stp}} = 0.8$ ($< T_c$) is clearly seen in Fig. 2(b), and the decrease in FCM below $T^*$ during cooling can be recognized in our slowest-cooling process with rate-10000 shown in the inset of Fig. 4(b) below. This FCM decrease below $T^*$ is contrary to the FCM increase commonly observed in noninteracting magnetic nanoparticle systems.\textsuperscript{19}

During a stop at a temperature below $T^*$, the SG short-range order of size $\xi$ continues to increase and so the FCM decreases until $\xi$ becomes comparable with $L_h$ given by Eq. (2). Then, $\xi$ no longer increases since the system is now in the paramagnetic state. The magnetization makes a turn and increases (due to the polarization of spin clusters) up to the equilibrium paramagnetic value. This is the dynamical crossover scenario proposed by TH. However, for $h$ of a few Oe, whose corresponding Zeeman energy in Fe$_{0.55}$Mn$_{0.45}$TiO$_3$ is about $10^{-4} \times T_c$, $L_h$ is in general much longer than $\xi^*$, and it will take an astronomical amount of time for $\xi$ to reach $L_h$. An exception is expected at $T_{\text{stp}}$ close to $T_c$, at which $L_h$ can be small (Eq. (2)) and the growth rate of $\xi$ is the fastest. Actually, we experimentally observed the expected FCM upturn behavior at such a stop as shown in Fig. 3. Similar behavior has been observed for Fe$_{0.5}$Mn$_{0.5}$TiO$_3$,\textsuperscript{20} while it is vaguely seen in the simulation with rate-100 at $T_{\text{stp}} = 1.05$ (see inset of Fig. 2(b)).

Another finding observed in both Fig. 1(a) and Fig. 4(b) is that the FHM is smaller than the FCM. This can be simply regarded as the cumulative memory effect of the SG short-range order.\textsuperscript{21} The SG order represented by $\xi$ increases at $T$ below $T^*$ and so the depolarization increases but $\xi$ is still much shorter than $L_h$ when the system is heated back to temperatures near $T^*$. Also one can see that the FCM at lowest temperatures is smaller, the slower the cooling rate. In the simulation, this is observed in the cooling processes slower than rate-333, as indicated in the inset of Fig. 1(b). In the experimental results shown in Fig. 1(a), crossing of the two FCM curves is observed. A possible interpretation for this is that, for the slower cooling process, $\xi^*$ is longer and the corresponding depolarization effect due to the SG stiffness
energy below $T^*$ is larger.

Lastly, let us discuss the $h$-dependences of the FCM and ZFCM curves. For the experimental results shown in Fig. 4(a) one can notice the following characteristics. As $h$ increases from 5 Oe, which we have thus far investigated, $T_{\text{irr}}$ approaches $T^*$. This implies that a stronger field makes it easier for the ZFCM curve to merge with the FCM curve in the paramagnetic phase. Under a sufficiently large $h$, on the other hand, $T_{\text{irr}}$ becomes lower than $T^*$, whose associated cusp-like shape is much more rounded. The phenomena in this field range coincide with those observed for Fe$_{0.5}$Mn$_{0.5}$TiO$_3$ by Aruga Katori and Ito$^9$ and are interpreted as the dynamical crossover by TH; $\xi$ in the ZFCM process reaches a relatively shorter $L_h$ at $T \approx T_{\text{irr}}$, within the observation time at each temperature. In the processes under $h$ of a few hundred Oe, we see $T_{\text{irr}} \approx T^*$. A rounded cusp-like shape in the FCM curve in these processes can be regarded as a blocking phenomenon since a difference between the FCM curves with different cooling rates is still noticeable below $T^*$ though it decreases with increasing $h$. As shown in Fig. 4(b), $T_{\text{irr}}$ obtained by the simulation also decreases significantly as $h$ increases.

We have argued that the FCM behavior of Ising spin glasses observed in both the experiment and simulation can be interpreted in a unified way by a scenario based on the (extended) droplet picture, thereby we have emphasized the viewpoint of ‘blocking’ of the FCM, or ‘glassy dynamics’. The scenario proposed in the present work is rather intuitive and qualitative, but, at least at low temperatures under relatively large $h$, it is consistent with the results obtained in our related works$^{7,22}$ where we have argued that the experimental and simulation results, whose timescales in units of microscopic spin-flip time differ by more than ten orders of magnitude, can be understood in a unified way even semiquantitatively. Combined with our most recent work claiming the dynamical breakdown of the SG state under relatively small $h$, the present results strongly suggest that the slow dynamics in the FCM processes we have observed under moderate $h$ is ‘glassy dynamics’ far from equilibrium.

The viewpoint of ‘glassy dynamics’ is a natural consequence of the marginal stability of the SG phase predicted by both the droplet theory and the mean-field theory. It implies an infinitely wide distribution of relaxation times, at least under $h = 0$. In the 1980’s, the peculiar FCM phenomena discussed thus far had already been observed in various spin glasses and were reviewed by Lundgren et al.$^{26}$ They introduced a finite equilibration time, $t_{\text{eq}}$, at all temperatures; however, the above-mentioned marginal stability implies an infinite $t_{\text{eq}}$ at $T \leq T_c$ under $h = 0$. According to the droplet theory $t_{\text{eq}}$ is expected to become finite under $h > 0$ and can be estimated using eq.(2) combined with an appropriate growth law for the SG short-range order.$^{23-25}$ For a sufficiently small $h$, this $t_{\text{eq}}$ becomes of an astronomical order, and so the blocking of certain modes, spin clusters in the present argument, is inevitable in real FCM measurements. Practically, however, the FCM cusp is a good estimate of $T_c$, i.e., $T^* \approx T_c$ if $h$ is small and the cooling rate is sufficiently slow. A detailed scenario of ‘glassy dynamics’ for Heisenberg spin glasses is of importance, but it may become more complicated due to the presence of two types of degrees of freedom: continuous spin and (Ising-like) chirality.$^{27,28}$ It is also of interest to analyze the dynamic transition of structural glasses on the basis of the present scenario of ‘glassy dynamics’.

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