Dynamical breakdown of the Ising spin-glass order under a magnetic field

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The dynamical magnetic properties of an Ising spin glass Fe$_{0.55}$Mn$_{0.45}$TiO$_3$ are studied under various magnetic fields. Having determined the temperature and static field dependent relaxation time $\tau(T; H)$ from ac magnetization measurements under a dc bias field by a general method, we first demonstrate that these data provide evidence for a spin-glass (SG) phase transition only in zero field. We next argue that the data $\tau(T; H)$ of finite $H$ can be well interpreted by the droplet theory which predicts the absence of a SG phase transition in finite fields.

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One of the recent interests in the spin-glass (SG) study is the nature of Heisenberg spin glasses under a magnetic field [1, 2]. There exists, however, a still unsettled problem even on the conceptually much simpler Ising spin glasses. The mean-field theory predicts that the SG phase remains up to the de Almeida-Thouless (AT) line in the $(H, T)$ plane [3]. The droplet theory, based on the short-range Edwards-Anderson (EA) model, instead predicts that any applied magnetic field will break up the equilibrium SG long-range order [4, 5]. Experimental investigations on Ising spin glasses give evidence both for the existence of an AT-line [3] and against critical dynamics under a field [6, 7]. Some recent simulation and experimental results indicate that the SG phase does not exist under a magnetic field [8, 9, 10].

In the present paper, we address the problem through ac susceptibility measurements on the Ising spin glass Fe$_{0.55}$Mn$_{0.45}$TiO$_3$ under a dc bias field $H$ at temperature $T$. We first specify, in a manner explained in detail below, sets of $(T, H)$, at which the characteristic relaxation time of the system, $\tau(T; H)$, coincides with the inverse of the ac field frequency $\omega$, i.e., $1/\omega = \tau(T; H)$. We then examine whether or not these $\tau(T; H)$ obey a dynamical critical behavior represented by

$$\tau(T; H) \sim t_0(\xi_c/L_0)^z \sim t_0/T_c/(H_0 - 1)^{-2\nu},$$

(1)

at $T > T_c(H)$. Here we suppose that the SG replica symmetry breaking (RSB) phase predicted to appear below the AT line, $T_c(H)$, by the mean-field theory is accompanied with a high-temperature disordered phase which exhibits critical divergences of the correlation length $\xi_c(T; H) \sim L_0(T/T_c(H) - 1)^{-\nu}$ and of the correlation time given by the above equation, where $t_0$ and $L_0$ are respectively microscopic units of time and length, and $z$ and $\nu$ are critical exponents. It turns out that the data $\tau(T; H)$ are not compatible with the expected dynamical critical behavior except for the case with $H = 0$.

In the droplet theory, on the other hand, the so-called magnetic correlation length, $\xi_H$, is introduced. It specifies the behavior of droplet excitations with a linear size $L$ in a SG state under a field $H$ [4]. For droplets with $L < \xi_H$ their behavior is governed by the SG stiffness free energy $\mathcal{Y}(T)(L/L_0)\eta$, while for those with $L > \xi_H$ their behavior is dominated by the Zeeman energy $\sqrt{\eta_{EA}H}(L/L_0)^{d/2}$, where $\mathcal{Y}(T)$ is the SG stiffness modulus, $\theta$ the stiffness exponent, $\eta_{EA}(T)$ the EA order parameter, and $d$ the spatial dimension. Explicitly, at $T$ less than $T_c(\eta = T_0(0))$ which is a unique critical point of the system, $\xi_H$ is written as

$$\xi_H \sim L_0 \left(\frac{\mathcal{Y}(T)}{\sqrt{\eta_{EA}(T)}}\right)^{\delta} \sim L_0 \left(\frac{(1 - T/T_c)^{\alpha_{EA}}}{H}\right)^{\delta},$$

(2)

where $\delta = 2/(d - 2\theta)$. In the last expression the temperature dependence of $\mathcal{Y}/\sqrt{\eta_{EA}}$ is represented by $(1 - T/T_c)^{\alpha_{EA}}$. At $T < T_c$, $\alpha_{EA} = \nu - \beta/2$ is expected, where $\beta$ is the critical exponent of $\eta_{EA}$. In the present analysis on the ac susceptibility measurements of frequency $\omega$ under $H$, we identify $\xi_H$ with $L_T(t = 1/\omega)$, which is the mean size of droplets that can respond to the ac field of frequency $\omega$ at temperature $T$, i.e.,

$$\xi_H \approx L_T(1/\omega).$$

(3)

Furthermore it is considered that $L_T(t)$ has the same functional form as that of the growth law $R_T(t)$ of the SG correlation length which grows with time $t$ after the system is kept at a constant temperature $T$ under $H = 0$. Explicitly, we adopt here an algebraic growth law

$$L_T(t) \sim R_T(t) \sim L_0(t/t_0)^{\theta T/T_c},$$

(4)

which is commonly observed in numerical simulations [13, 14] as well as in experiments [15, 16]. Interestingly, the present data $\tau(T; H)$ for relatively large $H$ and so relatively low $T$ turn out to be consistent with the scenario represented by Eqs. (3) and (4). We interpret this result as evidence for the presence of the magnetic correlation length $\xi_H$ predicted by the droplet theory.
We measure the ac susceptibility, $\chi(\omega, T, H) = \chi'(\omega, T, H) + i\chi''(\omega, T, H)$, of the Ising spin glass Fe$_{0.55}$Mn$_{0.45}$TiO$_3$ in the frequency range of $0.001 \, \text{s} \lesssim 1/\omega \lesssim 1 \, \text{s}$. All measurements are performed on a MPMS-XL squid magnetometer equipped with the low-field option. The susceptibilities $\chi'$ and $\chi''$ measured under different bias fields are shown in Fig. 1 while shown in Fig. 2 are the dc and ac susceptibilities of different frequencies in zero bias field and in a bias field of $H = 1000$ Oe. The equilibrium susceptibility $\chi_{eq}(T, H)$ can be determined in the temperature range within which the dc field-cooled (FC) and zero-field-cooled (ZFC) magnetization $M$ (measured with a slow cooling rate) coincide with each other, as $\chi_{eq}(T, H) = \frac{dM_{eq}(T, H)}{dT} \approx \frac{\Delta M_{eq}(T, H)}{\Delta T}$. The field $h$ should be small in order to probe the linear response.

$$\chi_{eq}(T, H) = 0.98 \chi_{eq}(T, H).$$

To explain our idea behind this condition we present in Fig. 2 $\chi_{eq} - \chi'(\omega)$, which is proportional to the dynamic spin correlation function $q(t)$ with $t = 1/\omega$, for different bias fields at $T = 20$ K. It has been found to follow the empirical formula $q(t) = Ct^{\alpha t}((T)\alpha)$ at $T$ above $T_c$ and a pure power law below $T_c$ for $H = 0$ both in numerical simulations on the EA Ising model[15] and in experiments on Fe$_{0.55}$Mn$_{0.45}$TiO$_3$[16]. The results are interpreted to indicate that the distribution of relaxation times is bounded from above by a certain value around $\tau^{*}(T)$ at $T > T_c$, while $\tau^{*}(T)$ is infinite at $T \leq T_c$. Although our experimental timewindow for a fixed set of $(T, H)$ is rather limited, Fig. 3 indicates that it is possible to bring the Ising SG to equilibrium by applying a magnetic field. In particular, for the set of $(T, H)$ for which Eq. 5 is found to be satisfied, the corresponding $q(t)$ exhibits a stretched-exponential form and so the corresponding $\tau^{*}(T; H)$ is definitely finite. We have therefore simply introduced the condition of Eq. 5 to specify the upper bound of relaxation times without fitting our $q(t)$ to a stretched-exponential form explicitly. We consider that the present method for specifying $\tau(T; H)$ is systematic and preferable to other methods used previously[20]. It is also noted that the condition is satisfied in the temperature range where we can determine $\chi_{eq}$ as seen in
that if both critical slowing down.

FIG. 4: (Color online) Test for critical dynamics assuming that the spin glass transition temperature \(T_g(H) = T(\tau; H)/[1 + (\tau/\theta)^{-1/z\nu}]\) [Eq. (3)] changes with \(\tau\) for each \(H\) as explained in the text. The inset shows \(\tau\) vs \(T(\tau; H = 0)/T_c - 1\) with \(T_c = T_g(H = 0) = 22.3\) K on a log-log scale.

In the inset of Fig. 3 we show the \(H - T\) relations which yield common values of \(\tau(\tau; H)\). They are roughly consistent with the AT-lines, i.e., of the form \(H \propto [1 - T(\tau; H)/T(\tau; 0)]^{1/\nu}\). However, such apparent AT-lines dependent on measuring time scales \(\tau\) are by no means a proof of an equilibrium SG phase under a field \(H\). We require the dynamical critical slowing down represented by Eq. (4) to hold at \(T > T_g(H)\) where \(T_g(H)\) is an assumed critical temperature under a field \(H\). As shown in the inset of Fig. 4 \(\tau(T; 0)\) in our timewindow are in fact fitted to this expression with \(T_c = T_g(0) \approx 22.3\) K, \(\theta_0 \approx 3 \cdot 10^{-13}\) s, and \(z\nu \approx 11\). The value of \(z\nu\) is in accordance with the previous values obtained for the Ising spin glass Fe\(_{50}\)Mn\(_{50}\)TiO\(_3\) [8]. This result gives evidence for an equilibrium SG phase transition in zero field. The main frame of Fig. 4 is, on the other hand, the result of an attempt to see if the system also exhibits critical slowing down in small dc fields assuming that \(t_0\) remains the same as in zero field. The best fit to Eq. (4) under this constraint is found with \(z\nu \approx 22\) for all \(H > 0\). To demonstrate the quality of the fit we show a \(H - T\) diagram whose \(T\)-axis is \(T_g(H)\) calculated for each \(\tau\) using the obtained value of \(z\nu\). A unique \(T_g(H)\) is found only for \(H = 100\) Oe (and \(H = 0\)) and the data in \(H > 100\) Oe exhibit systematic dispersion, giving evidence against critical dynamics for \(H > 100\) Oe. Even for \(H = 100\) Oe, however, due to the large value of \(z\nu \approx 22\) we consider that the relatively good fit to Eq. (4) has no physical meaning. We also note that if both \(\theta_0\) and \(z\nu\) are adjusted in the fitting, the resultant \(t_0\) takes unphysically large values. We therefore conclude that, under a magnetic field, the system does not exhibit a phase transition which is accompanied with critical slowing down.

Next, let us examine the data \(\tau(T; H)\) for relatively large \(H\) based on the droplet picture, namely, by regarding \(\tau(T; H)\) as a relaxation time of the largest SG clusters of a mean size \(\xi_H(T; H)\), which is determined by Eq. (4) combined with Eqs. (2) and (4). For this purpose we plot \(\tau(T_c; H) [\ln(\tau/\theta_0)\) for \(H \geq 5000\) Oe as a function of \(a_{\text{eff}} \ln(1 - T/T_c) - \ln H\), thereby adjusting \(a_{\text{eff}}\) but keeping \(\theta_0\) fixed to the value obtained above. As shown in Fig. 5 the best collapse of the data at \(T/T_c < 0.7\) is obtained with \(a_{\text{eff}} \approx 0.25\). The slope of the fit, which is equal to \(b/\delta\), gives \(b \approx 0.11\), where \(\delta = 0.77\) are used. Here we examine \(\tau(t; H)\) in a rather narrow temperature range (the lower bound of our observation of \(\tau\) is 0.55\(T_c\)), since the approximation \(\Upsilon/\sqrt{q_{EA}} \sim (1 - T/T_c)^{a_{\text{eff}}}\) used to derive the last expression of Eq. (4) with a constant \(a_{\text{eff}}\) is not expected to work in a wider temperature range. In fact when we analyze the \(\tau(T; H)\) data obtained for Fe\(_{50}\)Mn\(_{50}\)TiO\(_3\) by Mattsson et al. [7] by the present method, we obtain \(a_{\text{eff}} \approx 0\) and \(b \approx 0.09\) for their data at \(T \lesssim 0.5T_c\), and \(a_{\text{eff}} \approx 0.2\) and \(b \approx 0.09\) for their data at \(0.5T_c \lesssim T \lesssim 0.7T_c\). The circumstances become more subtle at \(T\) closer to \(T_c\), since the critical behavior of \(\Upsilon(T)\) and \(q_{EA}(T)\) has not been well established yet. We can only mention that the same analysis on our data \(\tau(T; H)\) at \(H \lesssim 0.95T_c\) and for \(H \lesssim 5000\) Oe yields \(a_{\text{eff}} \approx 0.45\) and \(b \approx 0.13\). This strongly implies that the critical exponent, \(a_{\text{eff}}\) at \(T = T_c\), is positive. In spite of such subtlety concerning with the temperature dependences of \(\Upsilon(T)\) and \(q_{EA}(T)\), the results shown in Fig. 5 are sufficient for us to conclude that the system is in the paramagnetic phase, with \(\xi_H\) and \(\tau(T; H)\) being the upper bounds for the SG correlation length and relaxation time, respectively.

The values of the exponents extracted above from \(\tau(T; H)\) at 0.55\(T_c\) \(\leq T \leq 0.7T_c\) can be compared with those obtained in the simulation on the Ising EA model in the corresponding temperature range; \(b \approx 0.16\) and \(a_{\text{eff}} \approx 0.14\), where the latter value is extracted from \(\xi_H\) which is obtained and denoted as \(l_T H^{-b}\) in [5]. These
figures are in turn compatible with the ZFC magnetization of Fe_{0.5}Mn_{0.5}TiO_{3} observed in heating processes with intermittent stops \[13\] as well as with the crossover scenario \[8\] for the AT-like transition observed also in Fe_{0.5}Mn_{0.5}TiO_{3} \[8\]. Here we emphasize that the values of $b$ extracted from the simulation and the experiments, over more than 10 decades difference in time scales in units of $t_0$, agree with each other reasonably well.

One more comment is in order on the growth law of the SG correlation at a constant $T$. In the original droplet theory \[1\], instead of Eq. \[1\], a logarithmic form of

$$L_T(t) \sim L_0 \left[ \frac{T \ln(t/t_0)}{\Delta} \right]^{1/\psi}, \quad (6)$$

has been proposed, where $\Delta$ is the characteristic energy scale of the free-energy barrier. For Fe_{0.5}Mn_{0.5}TiO_{3} Eq. \[6\] has been applied to its various phenomena with the resultant exponent $\psi$ ranging from 0.03 to 1.9. \[7\], \[10\], \[13\], \[14\]. When Eq. \[4\] is replaced by Eq. \[6\] with a $T$-independent $\Delta$ in the present analysis, we obtain $\psi \approx 0.6$ and the same $a_{eq}$ as obtained above from the $\tau(T; H)$ data at $0.55T_c \lesssim T \lesssim 0.7T_c$. We consider, however, that the power-law growth is superior to the logarithmic growth in the sense that the values of $b$ extracted from various phenomena are less dispersive than those of $\psi$. \[22\]. The power-law growth implies that the free-energy barrier of droplets excitations of a size $L$ is proportional to $\ln L$ which becomes smaller than $L^\theta$, the free-energy gap of the corresponding droplet excitations, for a sufficiently large $L$, say $L^*$. Therefore, our conclusion that the power-law is a more plausible description of various SG glassy dynamics mentioned above implies that the SG short-range of less than $L^*$ is involved in such slow-dynamical phenomena observed even by laboratory experiments. In particular, $\chi_{eq}$ and $\chi'(\omega)$ analyzed in the present work involve length scales $\xi(T)$ and $L_T(1/\omega)$ or $\xi_H$ which are much shorter than $L^*$ so that they can be regarded as SG equilibrium properties (see \[11\] for non-equilibrium phenomena associated with similar length scales much less than $L^*$). We believe that, in the field of glassy dynamics, proper understanding of the length and time scales of phenomena we observe is very important, though it is often not an easy task.

To summarize, we have found experimental evidence against the existence of an equilibrium phase transition in Ising spin glasses under a bias magnetic field, i.e. evidence against the AT phase transition. From the present analysis we learn that one has also to be careful to draw conclusions about the equilibrium phase diagram of Heisenberg spin glass under a field; the occurrence of the irreversibility alone may not be evidence for the presence of an equilibrium phase transition.

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