Direct Dynamical Evidence for the Spin Glass Lower Critical Dimension $2 < d_\ell < 3$

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A dynamical method is introduced to study the effect of dimensionality on phase transitions. Direct experimental measurements for the lower critical dimension for spin glasses is provided as an example. The method makes use of the spin glass correlation length $\xi(t,T)$. Once nucleated, it can become comparable to sample dimensions in convenient time and temperature ranges. Thin films of amorphous Ge:Mn alloys were prepared with thickness $\mathcal{L} \approx 15.5$ nm. Conventional behavior is observed as long as $\xi(t,T) < \mathcal{L}$. At the measurement time t_{co}, when $\xi(t_{co},T) \approx \mathcal{L}$, the time dependence is observed to cross over to exponential. These results are interpreted using spin glass dynamics, and are consistent with a lower critical dimension for spin glasses, d_ℓ, between $2 < d_\ell < 3$.

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The relation between the spin glass phase transition and dimensionality has previously been addressed through static experiments. For example, the spin glass transition temperature T_g has been measured for thin film materials in order to change the spatial dimension from $d = 3$ to $d = 2$ to probe the spin glass lower critical dimension, d_ℓ. However, extrapolations had to be introduced in order to demonstrate that $T_g \to 0$ as one approaches $d = 2$. For the first time, experiments presented in this Letter make use of dynamical measurements to examine the effect of dimensionality on the spin glass transition directly through the known properties of the spin glass correlation length.

The spin glass correlation length has already been shown to be a useful dynamical variable to probe length scale dependent quantities [1]. Recent experiments by Sahoo *et al.* [2,3] investigated aging and memory effects in a super-spin-glass, a discontinuous metal-insulator multilayer (DMIM), through temperature cycling magnetic relaxation measurements. They were able to extract the spin glass correlation length, "which seems to imply a crossover from three- to two-dimensional growth of the correlation length." In particular, they suggest experiments that could lead to the influence of dimensionality.

We make use of the growth of the spin glass correlation length $\xi(t,T)$ with time t at temperature T. In addition, the exponential increase of occupied states in the ultrametric theory for spin glass dynamics [4] weighs most heavily those states at the highest occupied barriers $\Delta(t,T)$, with the barrier heights exhibiting a direct relationship to $\xi(t,T)$ [5]. This Letter uses these relationships to probe the dynamical behavior of a spin glass from dimensions $d = 3$ to $d = 2$ using zero field cooled (ZFC) magnetization measurements on amorphous Ge:Mn thin films. This range of spatial dimensions spans the theoretical estimates of the lower critical dimension d_ℓ for spin glasses, both for Ising spins for which $d_\ell = 2.5$ [6], and for Heisenberg spins for which Lee and Young [7] state that d_ℓ "is close to, and possibly equal to, 3."

As noted above, previous (static) studies of the effect of dimensionality on spin glasses have used thin films of different thicknesses to probe the decrease of the spin glass transition temperature T_g as a function of film thickness. Grandberg *et al.* [8] studied variable thickness films of Cu:Mn (13.5 at.% Mn) from 10^4 to 20 Å. Their Fig. 3 displays a reduction in the *freezing* temperature from roughly 60 to 25 K over their thickness range. They extracted a crossover from typical behavior for bulk spin glasses "…to a slowing down for the very thin films that obeys a generalized Arrhenius law with a zero-temperature critical point," leading to "…a crossover from three- to two-dimensional spin glass dynamics when one spatial dimension is gradually diminished to a finite size." While these studies are suggestive, they depend upon extrapolations for their conclusions.

The dynamics of the spin glass phase offers a much more direct approach for study of the effects of dimensionality. In particular, one can examine the effects on the ZFC magnetization of the growth of the spin glass correlation length $\xi(t,T)$, from nucleation to the thickness of a thin film, $\mathcal{L}$. Let t_{co} be the time for $\xi(t_{co},T) = \mathcal{L}$. For times $t < t_{co}$, the system behaves as though $d = 3$. When $t > t_{co}$, the spin glass dynamics are those of $d = 2$, and presumably should vanish for $\xi(t,T) \geq \mathcal{L}$ if $d_\ell > 2$. However, in analogy with percolation theory [9], regions for $\xi(t,T) < \mathcal{L}$ do not *know* of the limiting dimension $\mathcal{L}$, and, hence, remain in the $d = 3$ spin glass state. There should be a largest barrier associated with $\xi(t_{co},T) = \mathcal{L}$, $\Delta_{max}(t_{co},T)$, through the relationship established in [5],

$$\frac{\Delta(t,T)}{k_B T_g} = \frac{1}{c_2}\left\{\ln\left[\frac{\xi(t,T)}{a_0}\right] - \ln c_1\right\}, \quad (1)$$

where the constants c_1, c_2 are defined by the growth expression for the spin glass correlation length $\xi(t,T)$,

$$\frac{\xi(t,T)}{a_0} = c_1 \left(\frac{t}{\tau_0}\right)^{(T/T_g)c_2}. \quad (2)$$

This form for $\xi(t,T)$ follows from theoretical expressions for the spin-glass correlation length: $\xi(t,T) \sim t^{\alpha(T)}$ [10–12]. Kisker *et al.* [11] found $\alpha(T=0.2)=0.026$ and $\alpha(T=0.7)=0.081$, with their $T_g = 0.9$, leading to $0.12 < c_2 < 0.13$ for $0.22 \le T/T_g \le 0.78$. The coefficient c_1 is of order unity [10–12], a_0 is the average spacing between spins, and the characteristic exchange time, $\tau_0 \approx \hbar/(k_B T_g)$. For comparison with spin-glass experiments on other materials, Joh *et al.* [5] found $c_1 \approx 0.53$ and $c_2 \approx 0.132$ for the thiospinel $CdCr_{1.7}In_{0.3}S_4$.

As interpreted experimentally [13], after rapid cooling in a ZFC experiment (see below for the experimental protocol) to the measurement temperature $T < T_g$, diffusion from the initial state is expressed as taking place along an ultrametric tree [4,13], with ever higher free energy barriers corresponding to increasing Hamming distances surmounted as the measurement time increases. When the magnetic field H is applied, the free energy barrier heights are reduced by the change in Zeeman energy E_z (expressed as a reduction in trap depth in Ref. [14]). For the states with free energy barrier heights $\Delta(t,T) < E_z$, an instantaneous transition to the (now) lowest energy manifold with magnetization $M_{\rm FC}$ takes place, $M_{\rm FC}$ denoting the field-cooled (FC) magnetization appropriate to the applied field H. This instantaneous increase is interpreted as the reversible magnetization, $M_{\rm rev}(T)$.

As time continues to progress, there is diffusion in the $M=0$ ZFC manifold to states with ever higher barriers $\Delta(t,T)$. There is also diffusion in the reverse direction to the *hole* in the lowest barrier energy states (smallest Hamming distances) created by the change in Zeeman energy E_z being larger than the respective free energy barrier heights. This results in the gradual buildup of states in the $M_{\rm FC}$ manifold, causing the magnetization to increase towards equilibrium as $M(t,T) \to M_{\rm FC}$. As a consequence of the bidirectional diffusion in the ZFC ($M=0$) manifold, the buildup of the magnetization $M(t,T)$ is a complex function of time, described by scaling similar to the strain creep function for polymers [15–20]. If $d_\ell > 2$, then this picture should vanish at $t_{\rm co}$ when $\xi(t_{\rm co},T) \gtrsim \mathcal{L}$.

However, as noted above, there will exist states in the ZFC ($M=0$) manifold with $\xi(t,T) \lesssim \mathcal{L}$ associated with the occupied states lying between free energy barriers $E_z \le \Delta \le \Delta_{\rm max}(t_{\rm co},T)$. These states will continue to diffuse to the *hole* created by E_z, leading to a continued increase in magnetization with time. However, because of the exponential increase of the number of states in an ultrametric geometry with increasing Hamming distance, the diffusion in the other direction will be dominated by the highest barrier $\Delta_{\rm max}(t_{\rm co},T)$. Thus, the increase of the magnetization for $t \ge t_{\rm co}$ will be *exponential* in time, with an activation energy given by $\Delta_{\rm max}(t_{\rm co},T)$. This argument obeys a crucial test. Namely, Eq. (1) relates $\Delta_{\rm max}(t_{\rm co},T)$ to $\xi(t_{\rm co},T)$, or more succinctly, to Eq. (1) with $\xi(t_{\rm co},T)$ replaced by $\mathcal{L}$. We shall see that the experiments reported here obey this condition.

The Ge:Mn thin films were prepared by 20 keV energy Mn^+ ions implanted into a 3-inch diameter, 0.5 mm thick Ge(100) wafer with a dose of $1.5 \times 10^{16}/\,{\rm cm}^2$ at a 7° tilt angle and constant temperature of 300 °C. Cross-sectional high-resolution tunneling electron microscope images show formation of a ~15.5 nm thick, uniform, amorphous Ge:Mn layer. Grazing-incidence synchrotron x-ray diffraction studies did not detect any significant presence of binary phases of Ge and Mn. Moreover, Mn $2p$ synchrotron x-ray absorption spectra shows that there is no long-range crystallographic order and localized Mn moments are located in a spherically symmetric environment [21]. The average Mn concentration in the amorphous region is $\sim 6.71 \times 10^{21}/{\rm cm}^3 \approx 11$ at.%, as determined from secondary ion mass spectrometry data (not shown), equivalent to an average Mn-Mn spatial separation of $a_0 \approx 5.3$ Å. Temperature dependent magnetization experiments exhibit a spin glass phase, with $T_g \approx 24$ K at 50 G field (Fig. 1, inset).

This material has a phase diagram [21,22], not unlike that of $Eu_xSr_{1-x}S$ for $x > 0.5$ [23]. For amorphous Ge:Mn, at the concentration of our sample, ≈11% Mn, a transition from paramagnetism to ferromagnetism takes place as the temperature is lowered, followed by a transition to the spin glass state at 24 K (for 50 G field). The ferromagnetic state is characterized by an S-shaped hysteresis with a very small coercive field (≲2 G). The inset in Fig. 1 displays a FC magnetization that rises rather rapidly with decreasing

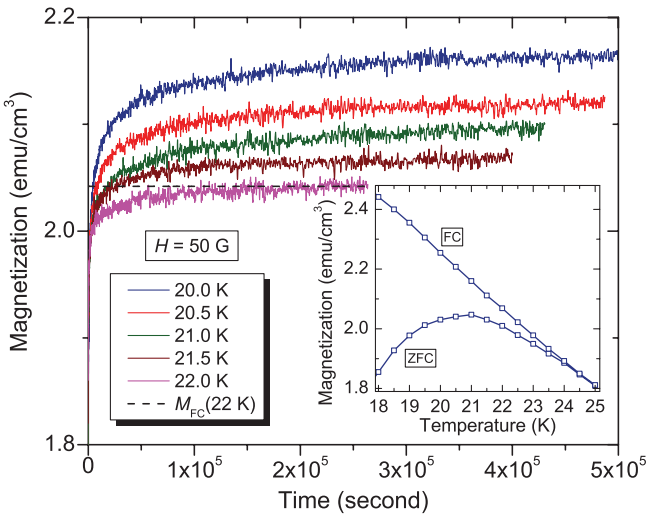

FIG. 1 (color). The ZFC magnetization $M(t,T)$ of the Ge:Mn amorphous thin film plotted against time, with the zero of time taken at the time the temperature is stabilized. The inset displays the ZFC and FC magnetizations as a function of T.

temperature below T_g, most probably associated with the remnants of the ferromagnetic network destroyed by the random field generated by the spin glass structure [24–27]. We presume that the response time of this contribution is rapid on the time scale of our measurements, so that they would not show up in the difference between the ZFC and FC measurements. The vanishing of this difference defines the glass temperature T_g, and all of our experiments and analysis depend solely on that difference.

Another feature of our measurements is the use of the rise in the ZFC magnetization as opposed to the decay of the thermoremanent magnetization (TRM). We are interested in finding when the conventional ($d = 3$) rise of the ZFC magnetization ends [presumably when $\xi(t_{co}, T) = \mathcal{L}$ for $d_\ell > 2$]. Stability of the superconducting quantum interference device (SQUID) and other backgrounds make absolute measurements difficult. It proves more accurate to relate the measured rise of the ZFC magnetization to the FC magnetization measured at the same temperature. Subtraction of the asymptote of the exponential rise of the ZFC magnetization from the *difference* between the measured time dependence of the ZFC magnetization and the measured FC magnetization would give (and does) a zero magnetization reading when the crossover from $d = 3$ to $d = 2$ takes place for conventional spin glass dynamics, allowing us to extract the crossover time t_{co} from our measurements.

The experiments were performed using a Quantum Design dc-SQUID magnetometer following the no-overshoot (NOS) cooling protocol of Rodriguez *et al.* [20]. This SQUID has a sensitivity of $\sim 10^{-7}$ emu and a temperature stability of ± 1 mK. A paramagnetic Sm_2CuO_4 sample was used to measure the superconducting magnet's offset magnetic field ($\sim$1.40 G), with that field offset defined as $H = 0$.

The sample temperature was quenched rapidly from well above T_g, to the measurement temperature T, without ever exceeding T throughout the cooling protocol, in accordance with the NOS protocol of [20]. This ensures that the effective waiting time, t_w^{eff}, is a minimum, and much less than the measurement times in our experiments. Immediately after the sample temperature was stabilized at T, an in-plane magnetic field $H = 50$ G is applied and the time-dependent magnetization measured. At zero waiting time, we estimate $t_w^{\text{eff}} \sim 56$ s, arising primarily because of the time for the superconducting magnet to reach its final field value.

Because of the small exponent c_2 in Eq. (1), the accessible temperature range over which measurements could be made was very narrow. For temperatures below about 20 K ($T/T_g < 0.83$), the response times were much too long to probe on ordinary laboratory time scales, while for temperatures above about 22 K ($T/T_g > 0.92$), the response time was much too short for meaningful measurements, in addition to a small magnetization difference leading to large experimental error. Figure 1 exhibits the growth of the ZFC magnetization as a function of time for temperatures 20 K $\leq T \leq$ 22 K.

If the arguments surrounding $\xi(t, T) \approx \mathcal{L}$ are correct, the long time extrapolation of the rise of the ZFC magnetization at a given temperature should be exponential in time. It is more convenient and accurate to plot the difference $\log_{10}[M_{FC}(T) - M(t, T)]$ vs t, as asymptotically $M(t, T) = M_{ZFC}(t, T) + M_{rev}(T) \to M_{FC}(T)$. The results are exhibited in Fig. 2, where we use the measured values for $M_{FC}(T)$.

Using an Arrhenius law, we express $\tau(T)$, the time constant of spin glass dynamics for $t > t_{co}$, as $\tau(T) = \tau_0 \exp[\Delta_{max}/(k_B T)]$. We extract values of $1/\tau(T)$ for the different measurement temperatures from the slope of the data in Fig. 2. The values of $\log_{10}[\tau(T)]$ vs T^{-1} are plotted in Fig. 3, and are linear as expected. The values of $\Delta_{max}(t_{co}, T)/(k_B T_g)$ extracted from $\tau(T)$ are also plotted in Fig. 3 and are *independent of* T. Importantly, they fall within the range reported for other spin glasses in the experimentally accessible time window [13]. The very large error bars at $T = 22$ and 21.5 K in Figs. 2 and 3 are partly an artifact of the logarithmic scale, and intrinsic because of equilibrium magnetic fluctuations as observed by Reim *et al.* [28] for an insulating spin glass, and Svedlindh *et al.* [29] for a metallic spin glass.

The crucial test is to see whether Eq. (1) is satisfied. To best access the value of t_{co}, we subtract the asymptotic exponential fit of $M(t, T)$ in Fig. 2, $M_{asym}(T)$, from $M_{FC}(T) - M(t, T)$. The results are plotted in Fig. 4 for each of the measured temperatures. The value of t_{co}, extracted from when the curves first reach zero, are also plotted as the filled squares in the inset. Separately, the log of the time t_{FC}, defined by $M(t_{FC}, T) = M_{ZFC}(t_{FC}, T) + M_{rev}(T) = M_{FC}(T)$ from Fig. 1, is plotted as the filled

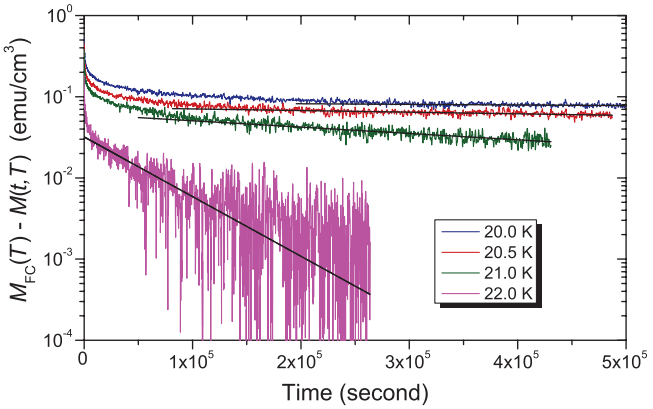

FIG. 2 (color). The difference $\log_{10}[M_{FC}(T) - M(t, T)]$ plotted against t for temperatures within the measurement window. The plot for $T = 21.5$ K is omitted because the large fluctuations at $T = 21.5$ and $T = 22.0$ K would overlap one another. The reason for the much larger noise at $T = 22.0$ K as compared to $T = 21.0$ K is partly an artifact of a logarithmic scale, and intrinsic because of equilibrium magnetic fluctuations [28,29].

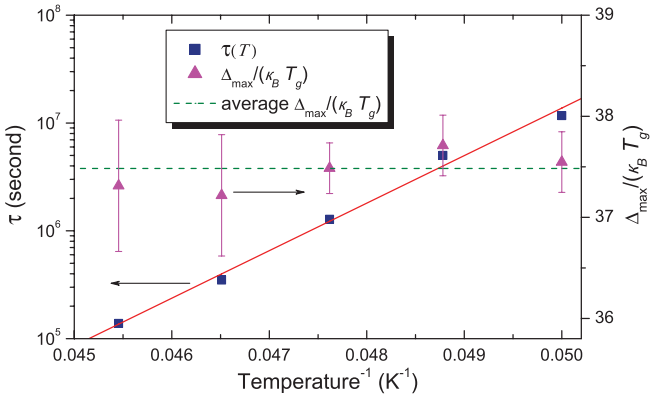

FIG. 3 (color). The logarithm of $\tau(T)$ and the associated maximum barrier height $\Delta_{\max}(t_{\rm co}, T)$, plotted vs T^{-1}.

circles in the inset. The time difference between the two plots is the time for the remaining ZFC states with length scales less than $\mathcal{L}$ to completely empty to the states with magnetization $M_{\rm FC}$.

A quantitative fit to Eq. (1) with $\xi(t_{\rm co}, T) = \mathcal{L}$ and $\Delta_{\max}(t_{\rm co}, T)$ from Fig. 3 depends on the constants c_1 and c_2. Because their values are not known, we plot consistency with Eq. (1) of $\xi(t_{\rm co}, T) \approx \mathcal{L}$ in the inset in Fig. 4. We have chosen the range for c_2 to be that found by Kisker *et al.* [11], resulting in c_1 lying roughly between 0.3 and 0.45. These values are not far off from those found for the thiospinel $CdCr_{1.7}In_{0.3}S_4$ [5], as previously noted.

In summation, dynamical measurements relying on the growth of the spin-glass correlation length $\xi(t, T)$ in a thin film have been used to probe the lower critical dimension for spin glasses, d_ℓ. For times sufficiently short, $\xi(t, T)$ is less than the film thickness $\mathcal{L}$, and conventional time dependences [16–19] for the growth of the magnetization were observed. When $\xi(t_{\rm co}, T) \approx \mathcal{L}$, the time dependence of the growth of the magnetization changes its character from conventional to exponential, with activation energy equal to the largest barrier $\Delta_{\max}(t_{\rm co}, T)$ surmounted during the growth of $\xi(t, T)$ to $\mathcal{L}$. This is dynamical evidence that a spin glass state with finite T_g does not exist at $d = 2$, and, hence, that $2 < d_\ell < 3$, consistent with the numerical findings of Franz *et al.* [6] for Ising spin glasses and Lee and Young [7] for Heisenberg spin glasses, and consistent with the extrapolation of the thin film spin glass experiments of Grandberg *et al.* [8].

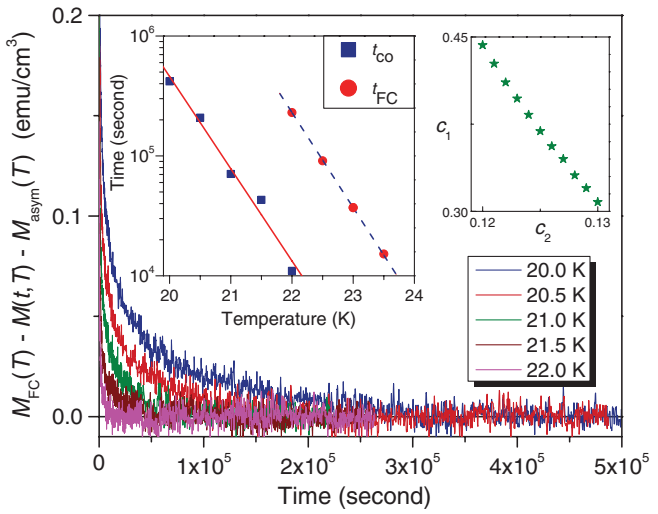

FIG. 4 (color). The "remainder" of the magnetization after subtraction of the exponential term, $M_{\rm FC}(T) - M(t, T) - M_{\rm asym}(t, T)$, plotted against time t. The logarithm of the cross over time to exponential rise, $t_{\rm co}$, extracted from the zero of the curves, is plotted vs T on the inset, as well as the time for the magnetization to reach $M_{\rm FC}$, $t_{\rm FC}$. The inset is a plot of the relationship of the constants c_1 and c_2 arising from the fit to Eq. (1) and (2).

In addition, the remaining rise in magnetization after $t_{\rm co}$ was accounted for quantitatively through transitions from the ZFC states occupied for $\xi(t, T) \leq \mathcal{L}$. The exponential increase in state degeneracy associated with ultrametric symmetry generates an exponential time dependence for transition of the ZFC states ($M = 0$) to the states in the $M_{\rm FC}$ manifold with an activation energy $\Delta_{\max}(t_{\rm co}, T)$. The critical test for this analysis, the relation between $\xi(t_{\rm co}, T) \approx \mathcal{L}$ and $\Delta_{\max}(t_{\rm co}, T)$ according to Eq. (1), is satisfied. Finally, explicit values for the two parameters present in the expression for $\xi(t, T)$ from Eq. (2), c_1 and c_2, were determined within the confines of the range calculated by Kisker *et al.* [11].

In summary, we have shown how one can use the growth of the correlation length with time to probe the lower critical dimension of a phase transition. Our direct determination of the length scale dependence of a phase transition is in contrast to static measurements that often depend upon extrapolations. Further, the ability to *tune* the dynamics to convenient laboratory temperature and time domains offers opportunities for probing the temporal and spatial nature of the transition. We have used a specific example of the verification of d_ℓ for spin glasses, but the method is a general one with, we believe, broad applications in other mesoscopic systems.

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