Temperature chaos in a Ge:Mn thin-film spin glass

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Temperature changes in thin-film Ge:Mn spin-glass dynamics are presented that exhibit temperature chaos (TC) when the spin-glass correlation length $\xi(t,T)$ grows to its thickness $\mathcal{L}$. For small $\mathcal{L} \approx 15.5$ nm, the transition to chaos takes place over a temperature range ΔT sufficiently large to exhibit both reversible and chaotic behavior. The value of ΔT can be related to the critical exponent for TC, ζ. Experimentally, ζ is found to be $\approx$1.06, in the range of recent simulations. The presence of a specific length scale $\mathcal{L}$ allows the transition to chaos to be examined over measurable laboratory temperature changes. The transition is found to be abrupt. Bulk materials, with a distribution of crystallite sizes, will smear out the transition, resulting in a very slow crossover. The abruptness of the transition and its nature are compared with recent theoretical calculations.

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I. INTRODUCTION

Temperature chaos (TC) in spin glasses is associated with the memory effect and is typical of polymers, random magnets, interacting nanomagnetic systems, and glasses in general. In this paper, we explore experimentally the length-scale dependence of TC, first introduced by Bray and Moore [1], then examined theoretically by a number of other authors [2–8] and applied to (qualitatively) explain experimental results [9–14]. It has been (theoretically) shown that the spin-glass ground state is unstable on length scales larger than $L^* \approx (J/\Delta T)^{1/\zeta}$, where J is a measure of bond energy between spins, ΔT is the temperature change, and ζ is the critical exponent for TC [1]. Spin configurations for changes in temperature ΔT are similar for length scales $<L^*$ and different for length scales $>L^*$. In our experiments, ΔT can be adjusted so that L^* can be greater than the spin-glass thin-film thickness $\mathcal{L}$, with no TC (equivalently, reversible behavior), or less than $\mathcal{L}$, resulting in TC. This enables us to estimate $\zeta \approx 1.06$, which agrees very well with numerical estimations [6–8]. We shall also demonstrate that the onset of TC is abrupt, in contrast to previous [15] assertions but in accord with recent [8] theoretical predictions.

TC in spin glasses has been reported for a number of years. Measurements of *rejuvenation* have exhibited TC in spin glasses, and more recent experiments have (qualitatively) examined memory effects [10] and interference effects [14] in bulk spin-glass samples, with a distribution of crystallite sizes. Overall, the picture that has emerged is that of a gradual phenomenon. The purpose of this paper is to examine TC in a sample where the length scale is fixed, namely, in a thin film of thickness $\mathcal{L}$. By working with a sufficiently thin sample, the transition to chaos can be studied over a temperature range ΔT sufficiently large that the nature of the transition itself can be studied. In particular, a fixed length scale $\mathcal{L}$ enables the study of the width of the reversible response to ΔT and the nature of the actual transition to chaos. Experimentally, it is found that the transition to chaos is abrupt, enabling evaluation of the temperature width for reversible behavior. This width can be used to evaluate the TC critical exponent ζ. The value extracted ($\zeta \approx 1.06$) is in much closer agreement with theory [6–8] than previous values from experiment [15].

A previous attempt [15] to estimate ζ arrived at $1/\zeta = 2.6 \pm 0.5$ through a scaling analysis (however, see Ref. [16] for a dissenting view of their analysis). Their experiments were conducted on bulk materials, presumably polycrystalline in nature. The distribution of crystallite sizes, and hence length scales [17], smoothed out their observation of the onset of chaos, causing them to assert that the transition to chaos was a "very slow crossover." In contrast, this paper uses a thin film of fixed thickness, providing a specific length scale for the onset of TC. As noted above, TC is related to a changing length scale for temperature changes ΔT. Having only a single characteristic length (the film thickness) allowed the observation of an abrupt onset of TC, in contrast to previous work.

It has been shown that aging in spin glasses can end on laboratory time scales for length scales at the mesoscale [18]. In thin films of thickness $\mathcal{L}$, a quasiequilibrium state can be created at that length scale. One can then explore dynamical phenomena that are length-scale dependent, for example, the spin-glass dynamical susceptibility [17].

The temperature of the spin glass is quenched rapidly from above the freezing temperature T_g to the measurement temperature (also known as *quench* temperature) $T_q < T_g$. If it is cooled in the presence of a magnetic field H, the magnetization is referred to as the field-cooled (FC) magnetization $M_{\rm FC}(T_q)$. If after a waiting time t_w the magnetic field is cut to zero, there is an instantaneous drop in the magnetization, with a slow remnant that eventually decays to zero. The latter is referred to as the thermoremanent magnetization (TRM) $M_{\rm TRM}(t,T_q)$, where the time t begins at the time the magnetic field H is cut to zero. Its decay is dependent upon the waiting time, the rate being slower the longer t_w is. This is termed the memory effect, for the spin glass *remembers* how long it was kept at H before H was cut to zero.

Concomitantly, if the spin-glass temperature quench to T_q occurs in zero magnetic field and a field H is applied after a time t_w, there is a sudden increase in magnetic moment and an additional slow increase in magnetization, the two combined being the zero-field-cooled (ZFC) magnetization

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$M_{\rm ZFC}(t,T_q)$. The increase of the slow component is dependent upon the waiting time, the rate being slower the longer t_w is. The combination of the two magnetizations $M_{\rm TRM}(t,T_q) + M_{\rm ZFC}(t,T_q) = M_{\rm FC}(T_q)$.

The dependence of the rate of change of $M_{\rm TRM}(t,T_q)$ and $M_{\rm ZFC}(t,T_q)$ upon t_w has been interpreted as arising from successively increasing energy barrier heights associated with the diffusion from the initial state prepared at time $t = 0$, and temperature $T = T_q$ (*after* the waiting time t_w) to spin-glass states with ever-decreasing overlap with the initial state. Reference [17] contains a detailed discussion of spin-glass dynamics.

The experiments were performed on thin amorphous Ge:Mn (11 at. % Mn) films of thickness 155 Å [18,19]. Previous experiments have shown this insulating system to exhibit spin-glass properties [19,20], not unlike $Eu_xSr_{1-x}S$ [21], an insulating canonical spin-glass system. Further, the behavior of the field-cooled magnetization is very similar to that found for the thinnest Cu:Mn films by Kenning *et al.* [22]. All the dynamical measurements on these films [18] are consistent with the usual spin-glass systems, establishing confidence in the generality of effects seen in Ge:Mn films.

II. ANALYSIS

The enabling concept behind the experiments is the growth of the spin-glass correlation length $\xi(t,T)$ with time t after the spin glass has been rapidly quenched from a temperature above the spin-glass transition temperature T_g to a temperature $T < T_g$ [23,24]:

$$\xi(t,T) = c_1 a_0 \left(\frac{t}{\tau_0}\right)^{(T/T_g)c_2}, \tag{1}$$

where $c_1 \simeq 0.375$ and $c_2 \simeq 0.125$ are fitted numerical coefficients, $a_0 \simeq 5.3$ Å is the average interatomic spacing of the magnetic species, and τ_0 is an exchange time of the order of $\hbar/(k_B T_g)$, with T_g being the spin-glass transition temperature $\approx$24 K. Experiments [18] showed that after a time $t = t_{\rm co}$, defined by the time it takes for the correlation length $\xi(t,T)$ to grow to the sample thickness $\mathcal{L}$ so that $\xi(t_{\rm co},T) = \mathcal{L}$, the spin glass transitioned to a two-dimensional state. Because the lower critical dimension $d_\ell \approx 2.5$ [25–27], $T_g = 0$ and spin-glass ordering for length scales greater than $\mathcal{L}$ vanished. However, there remain spin-glass correlations for length scales less than $\mathcal{L}$.

The relationship between $\xi(t,T)$ and the largest barrier height $\Delta_{\rm max}$ encountered when $\xi(t,T)$ approaches $\mathcal{L}$ is given by [28]

$$\frac{\Delta_{\rm max}}{k_B T_g} = \frac{1}{c_2}\left[\ln\left(\frac{\mathcal{L}}{a_0}\right) - \ln c_1\right], \tag{2}$$

where c_1 and c_2 are the constants in Eq. (1). The important aspect of Eq. (2) is that $\Delta_{\rm max}$ is *independent* of the temperature and only a function of the film thickness $\mathcal{L}$. Further, $\Delta_{\rm max}$ is the largest barrier encountered for *all* times $t > t_{\rm co}$. This means that there is no further aging of the correlated spin-glass state. The occupancy of all the states between barriers is given by the Boltzmann distribution. Hence, it is appropriate to regard this initial state as a quasiequilibrium state in which TC can be measured.

The experiments of Ref. [18] showed that the conventional ($d = 3$) time-dependent increase of the zero-field-cooled magnetization $M_{\rm ZFC}(t,T)$ crossed over to activated behavior at $t = t_{\rm co}$, with activation energy given by Eq. (2). There was an end to aging, as $\xi(t,T)$ could not continue to grow. It was cutoff at the value $\xi(t_{\rm co},T) = \mathcal{L}$. This means that the spin-glass-correlated states with magnetization $M_{\rm ZFC}(t,T)$ were in equilibrium among themselves with an absence of aging and an activated magnetization increase to the field-cooled magnetization $M_{\rm FC}(T)$. These dynamics will be central to our observation of TC.

Having prepared the initial state with a length scale $\mathcal{L}$, one can now explore experimentally the length-scale dependence of TC. The temperature range for which $L^* > \mathcal{L}$, and therefore for reversible dynamics, can be estimated from Bray and Moore [1]. They showed that the spin glass ground state was unstable on length scales larger than $L^* \approx (J/\Delta T)^{1/\zeta}$, where J is a measure of bond energy between spins [1]. Setting $J \approx k_B T_g$, with a_0 being the average spacing between magnetic atoms, dimensional analysis would suggest

$$L^* = a_0 \left|\frac{T_g}{T - T_q}\right|^{1/\zeta}. \tag{3}$$

Thus, in the experiments presented in this paper, $|\Delta T = T - T_q|$ can be adjusted so that L^* can be greater or less than the thin-film thickness $\mathcal{L}$. If $L^* > \mathcal{L}$, then the spin-glass dynamics should be reversible, with no TC. Conversely, for $L^* < \mathcal{L}$ temperature chaos should appear. Notice that there are no adjustable parameters. The fixed length scale $\mathcal{L}$ specifies a specific temperature difference $|T - T_q|$ according to Eq. (3), the only variable being a possible multiplicative factor in front of a_0.

The experimental procedure is as follows. The temperature is rapidly reduced from well above T_g to a quench temperature $T_q < T_g$ in the absence of a magnetic field. A magnetic field is applied when the temperature is stabilized at T_q (usually in a few minutes), and the system is *aged* for times longer than $t_{\rm co}$. During this aging period, the magnetization increases towards $M_{\rm FC}(T_q)$, but the increase is so slow that the magnetization after the end of aging still is well short of $M_{\rm FC}(T_q)$. At this point in time, the temperature is reduced to $T = T_q - \Delta T$, where ΔT is positive. If ΔT is sufficiently small, so that $L^* > \mathcal{L}$, the dynamics of the system should remain reversible in the sense that they will depend upon the initial preparation of the system before the change in temperature. Further, aging is over. Thus, the dynamics will only exhibit activated behavior associated with the value of $\Delta_{\rm max}$ at $T_q - \Delta T$.

This follows from the relationship between $\Delta_{\rm max}$ and the Hamming distance $D(T)$ associated with the ultrametric geometry of the overlap q of the spin-glass states [29–31]. The Hamming distance for our situation, $\xi(t_{\rm co},T_q) = \mathcal{L}$, is defined for Ising spin glasses by the relationship

$$2D(T) = q_{\rm EA}(T) - q_{\rm min}, \tag{4}$$

where $q_{\rm EA}(T)$ is the Edwards-Anderson self-overlap [32] of the spin-glass states at temperature T and $q_{\rm min}$ is the minimum overlap of the spin-glass states set by the condition that $\xi(t_{\rm co},T_q)$ is cut off at the length scale $\mathcal{L}$.

If the temperature is *now* lowered from T_q to $T_q - \Delta T$, $q_{\min}$ does not change, but $q_{\rm EA}(T)$ increases from $q_{\rm EA}(T_q)$ to $q_{\rm EA}(T_q - \Delta T)$, thereby increasing the Hamming distance $D(T)$ from Eq. (4). The value of Δ is a monotonic increasing function of the Hamming distance [33], so that $\Delta_{\max}$ will increase. Previous measurements [29–31] exhibited this behavior, with an extrapolation of $\Delta_{\max}$ growing to a large value for decreasing temperature. This behavior has also been observed to be reversible, with reversion to the value associated with the initial state created at T_q when the system is warmed back to T_q. In TC terms, the system *remembers* the initial state when it is cooled by ΔT.

However, if ΔT is sufficiently large, so that $L^* < \mathcal{L}$, the system at $T_q - \Delta T$ is chaotic. It bears no relationship to the system prepared after aging at T_q. In this case, the dynamics should start over, and $M_{\rm ZFC}(t, T_q - \Delta T)$ should increase towards $M_{\rm FC}(T_q - \Delta T)$ as though it were initially prepared at $T = T_q - \Delta T$. Said another way, it is *rejuvenated* [10,34–36], and its activated dynamics start over. From Eq. (2), this means that $\Delta_{\max}$ will revert to that value *independent of temperature* that it had at the initial quench temperature T_q.

This analysis displays the fundamental difference between reversible and chaotic behavior in the experimental results. Again, in the former, there should only be $d = 2$ activated dynamics, with a temperature-dependent $\Delta_{\max}$. In the latter, there should initially be dynamics associated with $d = 3$, then a crossover to $d = 2$ dynamics at times $t > t_{\rm co}$. In terms of observation in the activated region of dynamics, $\Delta_{\max}(T_q - \Delta T)$ vs $T_q - \Delta T$ should initially rise in the reversible region as ΔT increases and then drop back to the value of Eq. (2) for ΔT sufficiently large that TC has taken place.

The situation for a positive change in temperature is quite different. When the temperature is raised, $T > T_q$, not only is $D(T)$, and hence $\Delta_{\max}(T)$, reduced, but also the dynamics are faster because the temperature is higher. The combination of the two effectively re-initializes the distribution and hence is indistinguishable from TC. This reinitialization was exhibited in the experiments of Refregier *et al.* [29] and Lederman *et al.* [30]. As a consequence, the experiments described below are only performed at $T < T_q$, with one exception for T slightly larger than T_q, the results of which will be presented here.

III. RESULTS AND DISCUSSION

The 155 Å amorphous Ge:Mn thin-film sample was subjected to the TC protocol. Because of the small exponent in Eq. (1) ($c_2 \simeq 0.125$) and the relatively thick sample ($\mathcal{L} =$ 155 Å), the accessible temperature range over which measurements could be made was rather narrow. For temperatures below about 20 K, the growth of $\xi(t,T)$ to $\mathcal{L}$ is much too slow for laboratory time scales, and for temperatures above about 22 K, the response time is too fast for meaningful measurements. As a compromise, the quench temperature was set at $T_q = 21.5$ K.

The sample was cooled from ~55 K to $T_q = 21.5$ K, and a 50 G in-plane magnetic field was applied immediately after the temperature stabilized at T_q. Figure 1 plots conventional ZFC magnetization increase and the corresponding TRM decay curves at 21.5 K. From Fig. 1, the estimated crossover time $t_{\rm co}$ is $\sim 6.8 \times 10^4$ s, or about 19 hours at 21.5 K. So after a wait time of 20 hours, the sample is in the quasiequilibrium state, with any chaos induced by the change in magnetic field no longer relevant.

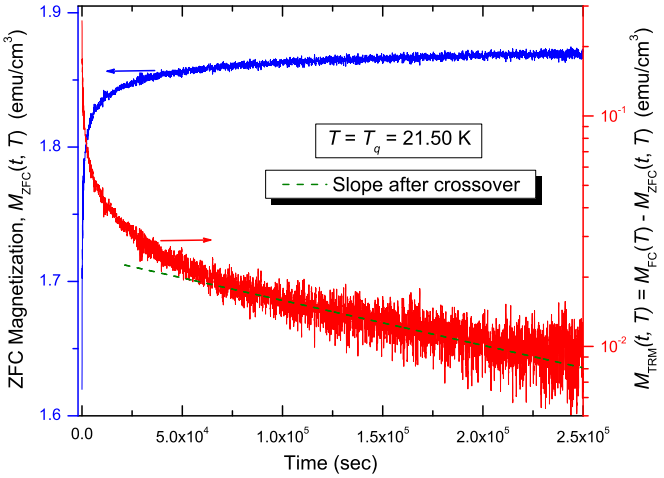

FIG. 1. (Color online) The left axis plots the time dependence of the ZFC magnetization of the Ge:Mn thin-film sample, quenched to a temperature $T_q = 21.5$ K, with an in-plane 50 G magnetic field applied. The right axis plots the time dependence of the difference between the FC and the ZFC magnetizations, displaying explicitly the activated nature of the dynamics [18].

The temperature was *then* changed to $T = T_q - \Delta T$, and the increase of the magnetization $M_{\rm ZFC}(t,T)$ was recorded for a range of temperatures from 21.60 to 20.00 K. Examples from the raw data are shown in Figs. 2(a)–2(c) for three different temperature ranges: $|T - T_q| \geqslant 0.6$ K, $|T - T_q| \leqslant 0.6$ K, and $T > T_q$. The difference, $M_{\rm FC}(T) - M_{\rm ZFC}(t,T)$, is included in Figs. 2(d)–2(f), which display the different activation energies for the three temperature regimes. The behaviors are quite different. For $|T - T_q| \geqslant 0.6$ K, the slopes are steeper than for $|T - T_q| \leqslant 0.6$ K, indicating that $\Delta_{\max}$ is smaller in the former temperature range than in the latter. Likewise, for $T > T_q$, the slope is similarly steeper than for $|T - T_q| \leqslant 0.6$ K.

There is another feature in Fig. 2 that is worth noting. In the chaotic region, the memory of the initial state ($T = T_q$) is erased, and rejuvenation takes place [10,34–36]. The correlation length $\xi(t,T)$ grows from nucleation, resulting in a rapid initial increase in $M_{\rm ZFC}(t,T)$ at short times before $\xi(t,T)$ has reached $\mathcal{L}$. At that point and after, the increase of $M_{\rm ZFC}(t,T)$ follows an activated behavior [Fig. 2(d)]. However, in the reversible regime, the memory of the initial state ($T = T_q$) remains. There is no further growth of $\xi(t,T)$ because it has already reached $\mathcal{L}$. Hence, at $t = 0$ in Fig. 2(e), the increase of $M_{\rm ZFC}(t,T)$ *begins* with activated dynamics. It is striking to compare the raw data in the two regimes and see the difference between chaotic and reversible behaviors.

Figure 3 plots the activation energies $\Delta_{\max}$ extracted from plots similar to those of Figs. 2(d)–2(f) for all the temperatures $T = T_q - \Delta T$ probed in these experiments. Remarkably, $\Delta_{\max}$ is found to lie at the value extracted in Fig. 1 at the quench temperature $T_q = 21.5$ K for both $|T - T_q| > 0.6$ K and $T > T_q$. That is, as noted above, $\Delta_{\max}$ reverts back to its value for the range of temperatures consistent with available laboratory time scales, $20.00 \leqslant T \leqslant 20.88$ K, indicating that

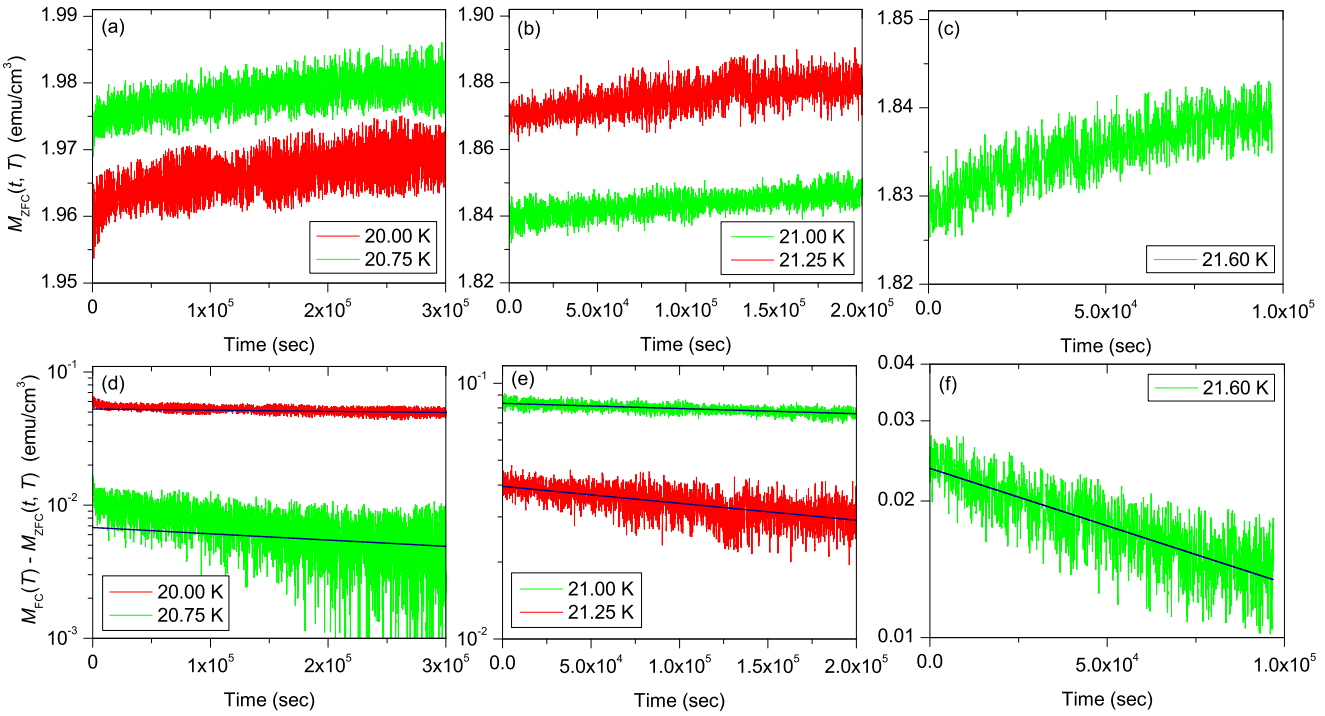

FIG. 2. (Color online) Representative plots of (a)–(c) the time dependence of the zero-field magnetization $M_{\rm ZFC}(t,T)$ and (d)–(f) the differences $M_{\rm FC}(T) - M_{\rm ZFC}(t,T)$, plotted on semilog scales. The three temperature regimes of interest are contained in $|T - T_q| \geqslant 0.6$ K (chaotic regime) in (a) and (d), $|T - T_q| \leqslant 0.6$ K (reversible regime) in (b) and (e), and $T > T_q$ (chaotic regime) in (c) and (f). Note that for the chaotic regimes [(a), (c), (d) and (f)], rejuvenation causes an initial decay towards the activated regime, associated with the growth of $\xi(t,T)$ towards $\mathcal{L}$, followed by an activated time dependence when $\xi(t,T)$ has reached $\mathcal{L}$. For the reversible regime [(b) and (e)], the time dependence is activated from $t = 0$.

rejuvenation has taken place within this temperature range. Under these conditions, $\Delta_{\rm max}(T)$ is set by the sample thickness according to Eq. (2) and must equal $\Delta_{\rm max}(T_q)$.

In the reversible range, $|T - T_q| < 0.6$ K, Fig. 3 exhibits a $\Delta_{\rm max}$ that increases sharply with decreasing temperature. This is consistent with previous experiments [29–31]. However, the increase is cut off sharply as one enters the TC range for $|T - T_q|$. The abruptness of the transition from reversible to chaotic regimes is reminiscent of the nature of the TC state as described in Ref. [8] but opposite to previous conjectures [15]. Chaos is the result of rare, but large, excursions and is not describable from an equilibrium approach. Hence, the transition to temperature chaos is not smooth but abrupt as $|T - T_q|$ increases, as demonstrated rather clearly in Fig. 3.

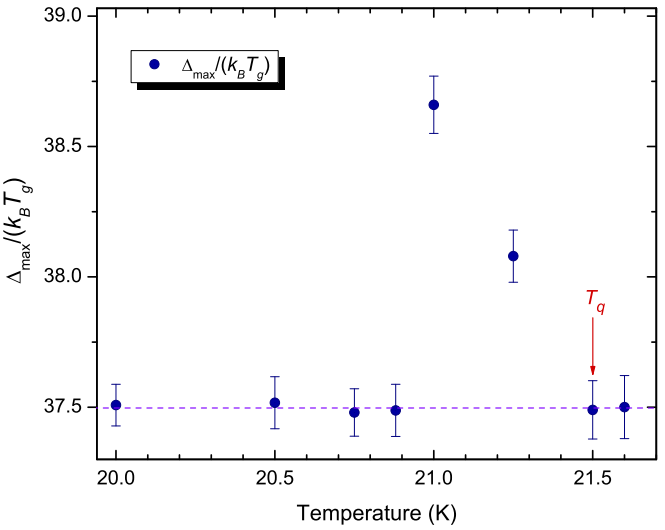

FIG. 3. (Color online) Plot of $\Delta_{\rm max}/(k_B T_g)$ versus T extracted from data represented by the difference $M_{\rm FC}(T) - M_{\rm ZFC}(t,T)$, as plotted in Figs. 2(d)–2(f).

In order to convince readers that the activation fit to the long time difference of $M_{\rm FC}(T) - M_{\rm ZFC}(t,T)$ in Fig. 2 is reasonable and that our interpretation is correct, we have plotted in Fig. 4 the value of $\Delta_{\rm max}/(k_B T_g)$ for a series of waiting times t_q at T_q, from 10 to 40 hours, before we lowered the temperature from $T_q = 21.5$ K to $T = T_q - \Delta T = 21.0$ K. For shorter waiting times, such that $t_q < t_{\rm co}$ at T_q [equivalently, $\xi(t,T_q) < \mathcal{L}$], $\xi(t,T_q - \Delta T)$ grows to $\mathcal{L}$ as the time progresses. The values of $\Delta_{\rm max}/(k_B T_g)$ observed at long times were the same as if the system had been quenched initially to $T_q - \Delta T = 21.0$ K, namely, *independent* of the quench temperature according to Eq. (2). However, when the waiting time $t_q > t_{\rm co}$ at T_q, the spin-glass correlation length had reached $\mathcal{L}$, the growth was over, and the subsequent reduction in temperature by $\Delta T = 0.5$ K resulted in the enhanced value for $\Delta_{\rm max}/(k_B T_g)$ of $\sim$38.7, in accordance with the argument following Eq. (4).

The temperature range for which $L^* > \mathcal{L}$, and therefore for reversible dynamics, can be estimated from Eq. (3). A value for the critical exponent, $\zeta = 1$, was found by Kondor and Végso [5] for dimensions $d \geqslant 8$. More recently, Sasaki *et al.* [6], Katzgraber and Krzakala [7], and Fernandez *et al.* [8] have found in $d = 3$ that $\zeta = 1.04$ and 1.07(5), respectively, close

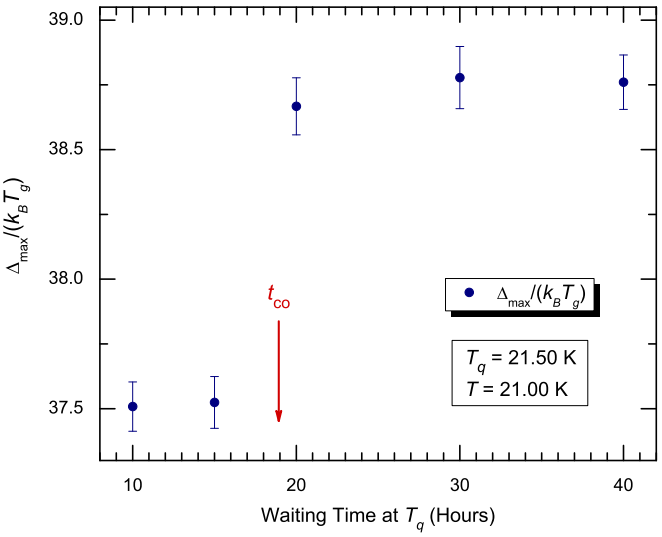

FIG. 4. (Color online) Plot of $\Delta_{max}/(k_B T_g)$ as a function of waiting times before the temperature is changed from $T_q = 21.5$ K to $T = 21.0$ K.

to the $d \geqslant 8$ "mean-field" value of unity reported by Kondor and Végso [5]. Taking $\zeta = 1.06$, the midpoint of the estimates [6–8], setting $L^* = \mathcal{L}$, and using the parameters appropriate to the Ge:Mn sample, one can evaluate the reversible range for $|T - T_q|$ from Eq. (3). We find $|T - T_q|_{\text{theo}} = 0.67$ K. This value is of the order of that estimated from Fig. 3, $|T - T_q|_{\text{expt}} \approx 0.6$ K.

That these estimates agree is a consequence of $\zeta \approx 1.06$. Previous estimates [15] arrived at $1/\zeta = 2.6 \pm 0.5$ through a scaling analysis (however, see [16] for a dissenting view). Such a value would destroy the agreement between theory and experiment for our results. We believe, therefore, that our experiments have established the value for ζ in the range of theoretical estimates for temperature chaos in spin glasses.

IV. SUMMARY

In summary, Figs. 2 and 3 are direct observations of temperature chaos in Ge:Mn spin glass. The thin-film thickness sets a length scale $\mathcal{L}$ in relation to the chaos length L^*. When the temperature change is small, so that $L^* > \mathcal{L}$, the system shows reversible behavior. When, however, the temperature change is sufficiently large, so that $L^* \approx \mathcal{L}$, an abrupt transition to chaotic behavior is exhibited and maintained for $L^* < \mathcal{L}$ for larger temperature changes. These properties are exhibited for the first time explicitly in Figs. 2 and 3. The abruptness of the transition to chaotic behavior, as opposed to a smooth equilibrium transition, is in agreement with the recent work of Fernandez *et al.* [8].

Further, following the expression presented by Bray and Moore [1] and using the theoretical estimate for $\zeta = 1.06$ from Refs. [7,8], we estimate $|T - T_q| \approx 0.67$ K for the crossover from reversible to chaotic behavior, very close to our experimental value of $|T - T_q| \sim 0.6$ K, as deduced from Fig. 3. The agreement between theory and experiment establishes the value of $\zeta \approx 1.06$, as opposed to a previous [15] estimate of $1/\zeta \approx 2.6 \pm 0.5$.

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