End of Aging in a Complex System

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Aging phenomena in complex systems have been used as an important tool to investigate the physics of complexity. In particular, aging effects in spin glasses, measured using the thermoremanent magnetization decays, have been instrumental as a probe of complex equilibrium and nonequilibrium dynamics. In this Letter, we show that aging found in spin glass materials has a finite lifetime. After the aging part of the decay has ended, we find a post-aging decay which is apparently logarithmic in nature. This decay is independent of the waiting time and part of the same mechanism that produces aging.

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Physical aging effects are a very interesting and intrinsic property of many types of complex materials [1], including polymers, colloidal gels, molecular glasses, spin glasses, and glass systems. An important and open question in the study of aging dynamics concerns the temporal extent of aging dynamics. To date, only speculation on this issue exists, and descriptions include aging dynamics extending to geological time scales [2] or infinite times [3]. By probing the very long time epoch of spin glass thermoremanent magnetization (TRM) decays, we experimentally address these questions. The answers have broad consequences for the limiting structure of the phase space as well as limitations on the configuration space.

The classic measurement of aging effects in spin glasses is the thermoremnant or complementary zero field cooled magnetization decay measurements. In the TRM experiments, the sample is cooled, in a small constant magnetic field, through its transition temperature, to a measuring temperature $T_m < T_g$. After waiting a time t_w, in the magnetic field, the field is rapidly removed, and the consequent magnetization decay of the sample measured. The decay is long lived and strongly dependent on the waiting time. The dependence of the magnetization decays on t_w is called aging. Recently, the authors have observed [4] that, for a spin glass cooled sufficiently rapidly to a particular measuring temperature, the magnetization decays scale as a function of t/t_w. The short effective waiting times (≈ 19 s) achieved opened the possibility of making measurements in time regimes much greater than the waiting time, $t \gg t_w$.

While aging has been the primary focus of TRM measurements, it is not the only contribution to the TRM decay. Current belief [5–7] is that the full magnetization decay can be described by a power law plus an aging term. In this study, we show that there exists another part of the decay which is independent of the waiting time and occurs in the time regime $t \gg t_w$. This is a new region of the decay and is not related to previous discussions of $t \gg t_w$ [8,9]. Analysis of this long time regime suggests that it is independent of the waiting time and is not a continuation of the initial stationary (power law) term.

All measurements in this work were performed on a $Cu_{0.94}Mn_{0.06}$ alloy that has a transition temperature $T_g = 31.5$ K. The sample has been the focus of many previous studies and as such has been well-characterized [10].

We report very long time TRM decay measurements (10^5 s) for very short effective waiting times (7 s $< t_w^{\text{eff}} <$ 110 s) at a temperature of $0.83T_g$. The decays were obtained using identical fast cooling protocols with a small additional waiting time added before the magnetic field is shut off. It is this small additional waiting time that changes the age of the curves in Fig. 1(a). A slightly different protocol was employed to obtain the $t_w^{\text{eff}} = 7$ s decay. The sample was cooled at a faster rate from a slightly higher temperature (than the previously described measurements), and the temperature bottoms out closer to the measuring temperature. The ages (effective waiting time) of the individual curves were determined from the peak in the relaxation curve $[S(t) = -\frac{1}{H} dM/d\ln(t)]$ plot shown in the inset in Fig. 1(a). It can be observed that, as the effective waiting time increases, portions of the curves separate. We plot the difference between the 7 s waiting time curve and the longer waiting time curves in Fig. 1(b). When the decay curves overlap, the difference in Fig. 1(b) goes to zero. We observe for these short effective waiting times that the decays overlap each other at very long measuring times. It should be pointed out that the most accurate data point, in terms of an absolute value, is the data point at 100 000 s. It is with respect to this point that we define our baseline. Therefore, the very long time portions of the decays are the region in which we have the most confidence. The data at shorter times are more subject to sources of error including "wobbles" over long time scales (possibly due to small temperature fluctuations <5 mK).

Since all of the decay curves either follow or approach this long time decay, we conclude that it is independent of

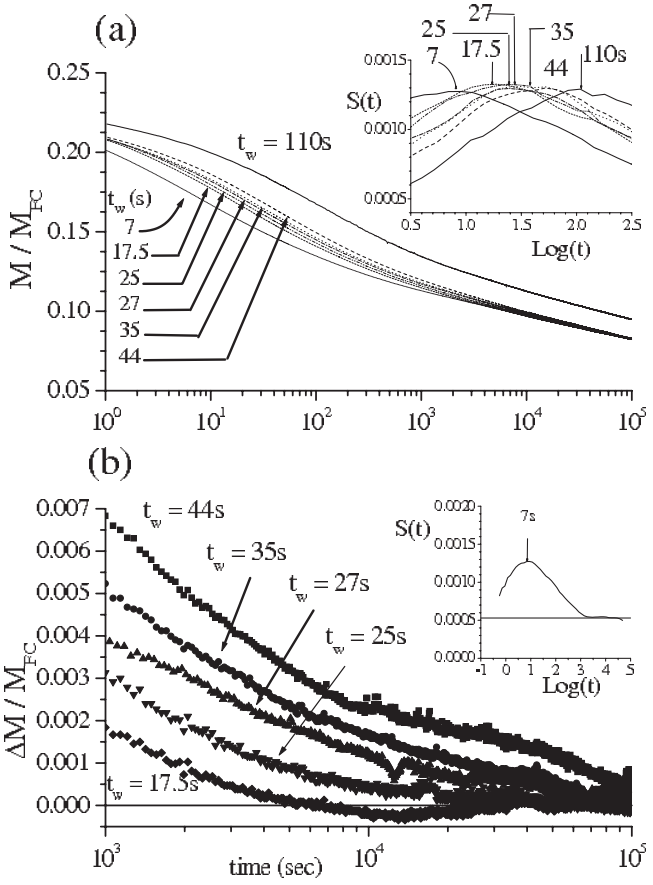

FIG. 1. (a) Decay curves for effective waiting times of 7,17.5, 25, 27, 35, and 44 seconds. In the inset, the $S(t)$'s for each curve used to determine the effective waiting time. (b) The subtraction of the $t_w^{\text{eff}} = 7$ s from the other curves, $\Delta M = M(t, t_w) - M(t, t_w^{\text{eff}} = 7\text{ s})$, for $t_w = 17.5$ (◆), 25 (▼), 27 (▲), 35 (•), and 44 s (■). It can be seen that the $M(t_w = 17\text{ s})$ curve begins to overlap $M(t_w = 7\text{ s})$ at approximately 6000 s. The inset shows the $S(t)$ function of the 7 s curve. The curve becomes horizontal at $t \sim 10^3$ s, indicating a crossover to logarithmic behavior.

the waiting time and, hence, distinct from the aging regime. We do not expect the curves to scale as $(t/t_w)^\mu$ for two reasons. (1) The initial state distribution in these short ''waiting time'' measurements are dominated by the cooling process [a very different mechanism than the isothermal evolution of states envisioned during the waiting time for which $(t/t_w)^\mu$ is generally applied]. In a recent variation on the real space droplet model, Jonsson and Takayama [11] proposed a two-step aging process where short-range-ordered clusters are thermally blocked and their size depends on the cooling rate. When the cooling is halted at T below T_c, the clusters, now under the influence of the stiffness energy, grow and reach a particular size in waiting time t_w. When the field is shut off, there exist two typical length scales and, hence, a two-step aging process. (2) At long times, the overlap of curves of different waiting times is contraindicative to scaling.

The $t_w = 44$ and 110 s curves do not overlap the shorter waiting time curves in the 10^5 s of measuring time, although the 44 s curve approaches the overlap at just over 10^5 s. Extrapolating the $t_w = 110$ s curve, we find that it approaches the post-aging decay at approximately 10^9 s.

It therefore appears that the aging effect is finite in time extent, and the time at which it ends is dependent on the waiting time. At long enough time scales, the aging component of the decay gives way to a long time post-aging magnetization decay that is independent of the waiting time. To determine the form of the post-aging decay, we can examine the overlap region (6000–100 000 s) between the 7 and 17.5 s decay curves. This region includes approximately 25 000 measured points. We find that we can fit this region to a power law, a stretched exponential, and a logarithmic function. The fitting functions $[F(t)]$ are evaluated by minimizing the reduced chi-square value, $\chi_N^2 = (M(t) - F(t))^2/N$ (N is the number of points) within an ORIGIN™ plotting program. The stretched exponential function with optimized exponent produces the worst fit (of the three fitting functions) with $\chi^2 = 5.8 \times 10^{-7}$. The power law, with an optimized exponent, produces a fitting with $\chi^2 = 7.0 \times 10^{-8}$, while the logarithm produces the best fit with $\chi^2 = 1.2 \times 10^{-8}$. The $S(t)$ function for the 7 s curve is plotted in the inset in Fig. 1(b). A horizontal line on this plot corresponds to a logarithmic decay.

In the following discussion, we will show that (1) the post-aging decay is a new, previously undetected magnetization decay; (2) the post-aging decay is intrinsically related to the same mechanisms that give the aging effects in this system; and (3) the decay is related to the initial state set up by the cooling procedure.

(1) The logarithmic decay observed in Fig. 1 is the first observation of the post-aging decay. However, there are at least two previously discovered magnetization decay components which, at first glance, may be responsible for this long term decay. These include the short time stationary decay which has a power law form [5] and the long time logarithmic decay of the field cooled magnetization [12]. Both of these decay mechanisms appear to be independent of the aging effects. The power law decay is much too small at the long times under consideration to account for the large long time decay. Our measurements of the logarithmic decay of the field cooled magnetization in the $Cu_{0.94}Mn_{0.06}$ sample give a coefficient 30 times less than the post-aging coefficient. These significant differences alone suggest that these decays are not directly responsible for the post-aging logarithmic decay.

(2) We now address the question as to whether the long time, post-aging decay is an independent contribution to the decay or an intrinsic part of the same mechanism that produced the aging. In Fig. 1, it can be observed that the onset of the post-aging decay is strongly dependent on the waiting time, varying from approximately 6000 to 100 000 s for waiting times ranging from 19 to 44 s. The initial observed value of the magnetization at the onset of the post-aging decay also decreases.

If the post-aging decay were independent of the aging decay, then we would expect the post-aging decay to be an additive term, or possibly multiplicative, to the aging decay. In this case, we would expect that subtraction or division of the post-aging decay from the aging term would improve scaling. Subtraction or division of any of the fitting functions from a complete set of the aging decays ($t_w = 50$–$10\,000$ s) produces sets of curves that no longer scale as t/t_w. In fact, scaling utterly fails, for the subtracted or divided data, using any of the standard scaling techniques [13]. Unless we are to abandon the concept that the aging decays scale as t/t_w, or that the decays scale at all, we are led to the conclusion that this long time decay is not simply an additive or multiplicative term like the short time stationary decay. This scaling analysis implies that this long time post-aging decay is intrinsically related to the same mechanism responsible for the aging process and is the decay due to this mechanism, after the aging component ends.

(3) Isothermal remanent magnetization (IRM) experiments on the $Cu_{0.94}Mn_{0.06}$ sample show no evidence of a post-aging decay. Using the principle of superposition, it has been shown [14] that the IRM decay for a pulse time t_w is essentially the difference between the TRM with waiting time t_w and the magnetization decay set up as a result of the cooling process. Since the IRM has no long time post-aging decay, we can conclude that the post-aging decay comes from the cooling process.

In a previous Letter [4], we have shown that the effects of even short cooling times can produce state occupations of large barrier states corresponding to long time dynamics. In this Letter, we have shown that the post-aging logarithmic decay is formed out of the cooling process. Using a variety of initial state distributions, we have probed the spin glass barrier model (SGBM) [15] and find a very interesting result in the case of a uniform initial distribution. A post-aging logarithmic decay is observed which satisfies many of the physical properties we have observed in the experimental data. All simulations were performed on a 600 barrier system with a branching ratio $r = 1.03$ and minimum barrier $\Delta_0 = 0.1$. Figure 2 shows the TRM decays for a set of four different initial distributions within the SGBM. The initial distribution was modeled as a weighted point at the origin (delta function) and a uniform distribution over all barriers. The total area under the distribution was normalized to one. To understand the logarithmic decay, observed in the barrier model, we have performed further analysis on the decay dynamics from individual wells located between two arbitrary, but successive, barriers. We find that decay from an individual well (1) begins at a time approximately equal to the time required for Boltzmann hopping over the smaller of the two barriers which bracket the well and (2) is power law in nature. The logarithmic nature is a direct consequence of the fact that successive wells are separated from each other by linearly growing barriers. The onset of decays from successive wells are therefore separated by a constant amount on a log time scale. This produces a cascading effect on a course grain scale (small number of total barriers) evolving into a smooth logarithmic decay as the time distance between barriers approaches the continuum limit (large number of total barriers). In the barrier model, the time at which the log term goes to zero (i.e., the maximum aging time) determines the maximum barrier height within the system.

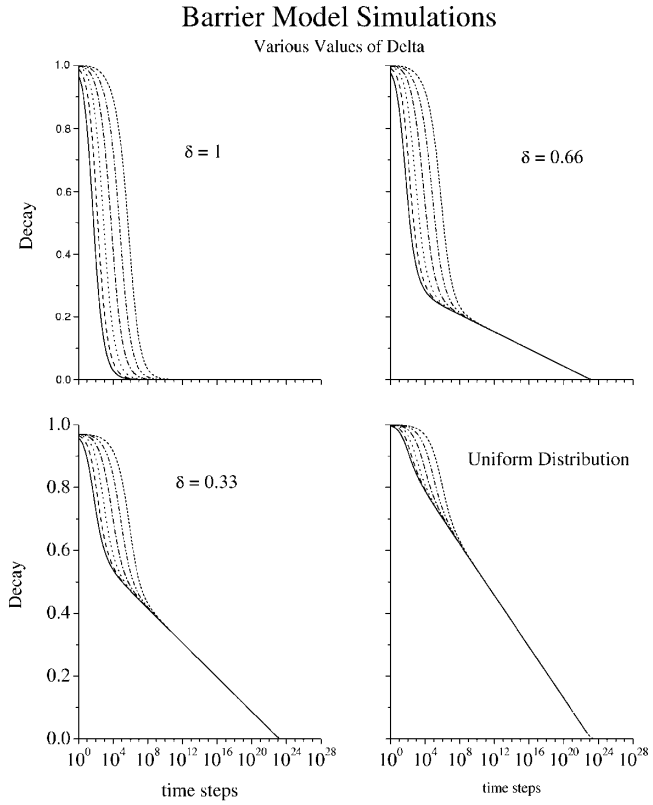

FIG. 2. Simulations with the barrier model for various initial conditions. The total initial distribution is normalized to 1. δ states begin evolution from the origin, while $1 - \delta$ states are initially distributed uniformly over all other barriers.

An initial uniform distribution within the SGBM arises quite naturally within the multivalleyed free energy landscape of the hierarchical picture. As a spin glass sample is cooled from the paramagnetic state to the transition temperature, barriers begin to grow. Lowering the temperature below the transition temperature, a system with many energy minima is formed, and, as a particular temperature (for example, the measuring temperature) is approached, there is no *a priori* reason for the system to choose any particular minimum. This leads to a random occupation probability for occupation of any particular minimum. Our sample is approximately 1–2 mm^3 in size. There is strong evidence from multilayer film experiments [16] and also from the nonlinear susceptibility [17] that in bulk samples of this material there are correlated regions within the samples on the order of 100–500 nm. This implies (assuming nonoverlapping correlated regions and reasonable space filling) that within our sample we have between

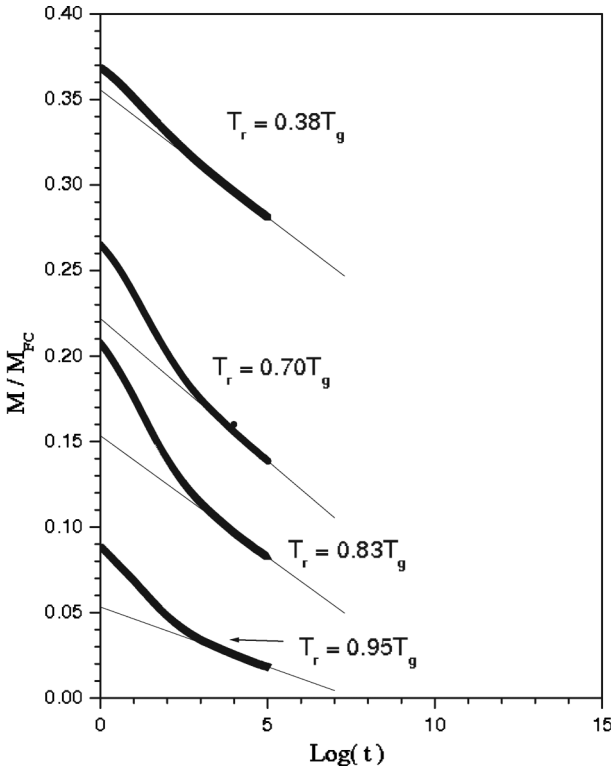

FIG. 3. Logarithmic fitting to the long time post-aging decay for various temperatures. Comparable ($t_w^{\text{eff}} \sim 20$ s) decay curves are used for fitting a log decay.

10^9–10^{12} correlated regions. Since bulk measurements are an average over all correlated regions, the random single particle distribution leads to a uniform distribution over the averaged free energy landscape. Using the argument above in the barrier model, as it exists, a uniform distribution occurs in the case of a branching ratio $r = 1$. Recent simulations and calculations [18] have shown that, in the limit of large numbers of barriers, the barrier model branching ratio approaches a value of one. Following the results obtained from the barrier model, we have fit logarithmic best fit functions to the post-aging decay for several reduced temperatures in Fig. 3. While extrapolations of the log fit are clearly highly speculative, we include it, as it is suggested by the barrier model and may provide an indication of the ultimate time scales in the problem. The extrapolated logarithmic post-aging decays go to zero on finite time scales ranging from approximately a year at $0.95T_g$, the highest reduced temperature measured, to many times the age of the Universe for the lowest reduced temperature measured $0.38T_g$. Of course, it is also possible that the post-aging term is only an intermediate mechanism and that other decay mechanisms are possible at much longer times. We, however, have no evidence for other very long time mechanisms.

While this simple picture may only suggest a direction for understanding "post-aging," it is nonetheless clear that the post-aging regime is an important and large component of the thermoremanent magnetization decay in spin glasses. As such, a first principles understanding of the effect is clearly desirable.

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[1] For examples, please see *Slow Relaxations and Nonequilibrium Dynamics in Condensed Matter*, edited by J.-L. Barrat, J. Dalibard, M. Feigelman, and J. Kurchan (Springer, Berlin, 2003), and references therein.
[2] D. Herisson and M. Ocio, Phys. Rev. Lett. **88**, 257202 (2002).
[3] T. Jonsson, K. Jonason, P. Jonsson, and P. Nordblad, Phys. Rev. B **59**, 8770 (1999).
[4] G. F. Rodriguez, G. G. Kenning, and R. Orbach, Phys. Rev. Lett. **91**, 037203 (2003).
[5] E. Vincent, J. Hammann, M. Ocio, J. P. Bouchaud and L. F. Cugliandolo, cond-mat/9607224.
[6] L. F. Cugliandolo and J. Kurchan, Phys. Rev. Lett. **71**, 173 (1993); Philos. Mag. B **71**, 501 (1995).
[7] J. P. Bouchaud and D. S. Dean, J. Phys. I (France) **5**, 265 (1995); J. P. Bouchaud, L. F. Cugliandolo, J. Kurchan, and M. Mezard, *Spin Glasses and Random Fields*, edited by A. P. Young (World Scientific, Singapore, 1998).
[8] M. Alba, M. Ocio, and J. Hammann, Europhys. Lett. **2**, 45 (1986).
[9] Heiko Rieger, J. Phys. A **26**, L615 (1993).
[10] G. G. Kenning, D. Chu, R. Orbach, Phys. Rev. Lett. **66**, 2923 (1991).
[11] P. E. Jonsson and H. Takayama, J. Phys. Soc. Jpn. **74**, 1131 (2005).
[12] L. Lundgren, P. Nordblad, and L. Sandlund, Europhys. Lett. **10**, 529 (1986).
[13] See, for example, Refs. [1,5] and references contained therein for a description of scaling techniques.
[14] V. S. Zotev, G. F. Rodriguez, G. G. Kenning, R. Orbach, E. Vincent, and J. Hammann, Phys. Rev. B **67**, 184422 (2003).
[15] Y. G. Joh, R. Orbach, J. Hammann, Phys. Rev. Lett. **77**, 4648 (1996).
[16] G. G. Kenning, J. M. Slaughter, J. A. Cowen, Phys. Rev. Lett. **59**, 2596 (1987); G. G. Kenning, J. Bass, W. P. Pratt, Jr., D. Leslie-Pelecky, L. Hoines, W. Leach, M. Wilson, R. Stubi, and J. A. Cowen, Phys. Rev. B **42**, 2393 (1990).
[17] Laurent P. Levy and Andrew T. Ogielski, Phys. Rev. Lett. **57**, 3288 (1986).
[18] J. Bowen and G. G. Kenning (unpublished).