Very Long Time Magnetization Decays in Spin Glasses

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It is currently believed that the decay of the thermoremanent magnetization in spin glasses is composed of two terms; The "stationary" term which does not depend on the sample history and dominates the short time decay (< 1s) and a long time aging term which depends on the samples history. The sample history includes both the thermal history and the time spent in a magnetic field (waiting time) before that field is removed. We report finding a third component of the decay at times much larger than the waiting time. This decay is independent of the waiting time, but part of the same mechanism that produces aging. We explain this decay in terms of the initial state distribution that is developed during the cooling process. This explanation is tested by performing Iso-thermoremanent magnetization decay measurements. Follow up calculations using the Spin Glass phase space barrier model suggest that the long time decay is logarithmic and that the maximum aging time is approximately three thousand years, in this sample, at a measuring temperature of .83 $T_g$.

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Understanding the effects of disorder on states of matter has been the subject of intense study for almost 100 years. In the early 1970s the realization that highly disordered magnetic materials with random interactions undergo an apparent phase transition, sparked considerable interest and excitement in the condensed matter community. This discovery of the so-called spin glass phase was marked by a large experimental effort by many researchers in an attempt to try to understand the physical parameters of the underlying phase space. One very interesting property of the spin glass phase was the observation that there exists a large distribution of relaxation times extending from atomic fluctuation time scales through what are believed to be geological time scales.

In 1983 it was shown that materials in the spin glass phase have memory effects on time scales ranging from milliseconds, to seconds, to days and beyond. To date much information has been obtained about the structure of the spin glass phase space from the study of aging effects. The classic measurement of these memory effects is the Thermoremanent (TRM) or complementary Zero Field Cooled (ZFC) 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 $T_m$. 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 strongly dependent on the waiting time ($t_w$). This is called aging. It has recently been observed that the magnetization decays scale with $t_w$ for a spin glass cooled, sufficiently rapidly, to a particular measuring temperature.

While aging has been the primary focus of TRM measurements, it is not the only contribution to the decay. Upon cooling the spin glass sample from a high temperature, in a magnetic field, the magnetization generally follows a Curie or Curie Weiss like behavior. Cooling the spin glass further, through its transition temperature, the magnetization appears to remain approximately constant, at a value $M_{fc}$. This is an indication that the spins freeze in approximately random directions with a net bias due to the field. Upon removal of the field there is a rapid decrease in the sample magnetization, from a value of $M_{fc}$, before entering the aging regime. Various measurements including AC susceptibility, muon decay, neutron spin echo, and NMR each provide some evidence that this rapid decay is power law in nature. The power law term is independent of the waiting time and has been called the stationary term (i.e. stationary with respect to the waiting time).

Current belief is that the full magnetization decay can be described by a power law plus an aging term

$$M_{TRM} = A t^{-\alpha} + M(t/t_w)$$

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 $t >> t_w$. This is a new region of the decay and is not related to previous discussions of $t >> t_w$. Analysis of this long time regime suggests that it is independent of the waiting time and is
FIG. 1: A(a) Decay curves for effective waiting times of: 7, 17.5, 25, 27, 35, 44 seconds. In the inset, the S(t)'s for each curve used to determine the effective waiting time. (b) the subtraction of the fastest decay from the other curves, \( \Delta M = M(t_w = 7s) - M(t_w), \) for \( t_w = 17.5(\bullet), 25(\triangledown), 27(\Delta), 35(\bullet), 44(\blacksquare) \). It can be seen that the \( M(t_w = 17) \) curve begins to overlap \( M(t_w = 7) \) at approximately 5000s.

We report very long time TRM decay measurements (10^5 seconds) for very short effective waiting times (7s < \( t_w < 110s \)). The data is plotted in Figure 1. 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 Figure 1a. The ages of the individual curves were determined from the peak in the relaxation curve \( S(t) = -\frac{dM}{d\ln(t)} \) plot shown in the inset of Figure 1a. It can be observed that as the effective waiting time increases the aging portion of the curves separate. This is a well-known effect of aging \[17]\. We observe for the short waiting times curves that the decays overlap each other at very long times. Since all of the decay curves either follow or approach this long time decay, we conclude that it is independent of the waiting time and hence distinct from the aging regime. We plot the difference between the 7s waiting time curve and the longer waiting time curves in Figure 1b. The range over which this difference goes to zero determines the onset of the third component of the decay for each of the waiting times. It can be observed that, within error, the decay of the 17s waiting time decay begins overlapping the 7s waiting time decay at approximately 5,000 seconds. It can also be observed that the 25s waiting time decay overlaps, within error, at approximately 35,000 seconds while the 27s waiting time decay overlaps at approximately 75,000 seconds. The 35s and 45s waiting time decays systematically approach the 7s waiting time decay, but the magnetization difference does not go to zero within a measuring time of 100,000 seconds. This analysis is subject to the errors inherent within the measuring system. Sources of error include a high frequency point-to-point fluctuation and low-frequency "wobbles" (possibly due to small temperature fluctuations < 10mK). The signal to noise ratio, at 100,000 seconds, is approximately 100 to 1.

It therefore appears that the aging effect is finite in time extent and at long enough time scale gives way to a long time component of the magnetization decay that is independent of the waiting time. Unfortunately the region of the overlap (5,000-100,000s) between the 7s and 17s waiting time decays gives only one and a half decades of measurement time with which to fit a function, precluding an absolute determination of the form of the fitting function. We find that we can fit this range to a power law, a stretched exponential and a logarithmic function(Figure 2). Values of the fitting coefficients and exponents are given in the figure caption. We take a data point every 3 seconds for a total of approximately 20,000 measurements in the overlap range. The large number of measured points and small signal to noise ratio give a very small value for the \( \chi^2 \) of the fitting functions. The stretched exponential is the worst fit with \( \chi^2 = 5.8 \times 10^{-7} \), while the logarithm gives the best fit with \( \chi^2 = 1.2 \times 10^{-8} \).

It is possible that Eq[14] is the correct description of the decay and that we are observing at long times, the short time stationary term after the aging decay has ended. To test this we have determined the upper limit of the power law exponent for the sample, measured at 83 \( T_g \), using a TRM magnetic field of 20G and a waiting time of 10,000 seconds (data is displayed in Figure 3). The short time decay in the TRM should be dominated by the power law decay of the short time stationary term, Eq[14]. Fitting over the first 3 seconds of this TRM decay we have determined that the upper limit for the exponent \( \alpha \) is 0.01. This

All measurements in this work were performed on an alloy \( Cu_{0.94}Mn_{0.06} \) that has a transition temperature \( T_g = 31.5K \). The sample has been the focus of many previous studies and as such has been well-characterized[16]. Measurements were performed on a home built DC SQUID magnetometer that allowed for good control of the rapid temperature quench and subsequent waiting time. All measurements in this study were performed at \( T = 26K \) which corresponds to reduced temperature of \( 0.83T_g \).

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compares with an exponent of the power law $\alpha = 0.072$ found for the very long time decay. This value is also much larger than the value reported from ac susceptibility experiments\cite{17} for the short times stationary term. This significant difference suggests that the short time and long time decays are of different origin.

FIG. 2: Three fits to TRM($t_w = 7s$), fitted to $t > 5000s$. The three fitting functions are: power law, stretched exponential and logarithmic. For power law ($M = M_0 t^{-\alpha}$), $M_0 = 0.1881$ and $\alpha = -0.072$. For stretched exponential ($M = M_0 \exp[t^{-\alpha}/\tau_0]$, $M_0 = 1.48, \tau_0 = 10^{-13}$ and $\alpha = -0.02559$. For logarithmic fit ($M = M_0 + \log(t^\alpha)$), $M_0 = 0.156$ and $\alpha = -0.0148$.

We now address the question as to whether this long time decay is an additive term. Subtraction of any of the fitting functions from the general decay curves produce sets of curves that no longer scale as $t/t_w$. In fact, we are not able to scale the subtracted data using any of the standard scaling techniques\cite{20}. Unless we are to abandon the concept that the aging decays 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 term like the short time stationary decay. This analysis implies that this long time stationary decay is intrinsically related to the aging process and does not occur until the effects of aging are effectively over. An understanding of both, the mechanisms responsible for, and form of, this long-term decay can be found through analysis of the barrier model.

The Barrier Model of the spin glass phase space was developed to explain aging in spin glasses. Inspired by the Parisi\cite{18} solution of the first principles Sherrington Kirkpatrick\cite{19} model of spin glasses and grounded in experimental analysis, the barrier model implies that spin glass phase space can be described by diffusion of the system within a phase space, defined by a set of energy barriers which grow linearly as a function of Hamming distance\cite{21}. Generally a minimum barrier $\Delta_0$ is defined and the height of barrier $N$ is equal to the product of $N$ and $\Delta_0$. The model simplifies the hierarchically ordered tree structure of the Parisi solution by collapsing local branches of the tree(states) into a single state with the appropriate degeneracy multiplication factor(this corresponds to the product of the barrier number $N$ and the branching ratio $r$). Diffusion within the Barrier Model is essentially a trade-off between the geometrically growing number of states as a function of increasing barriers and an exponentially decreasing probability for Boltzman hopping over the increasing size of the barriers. This model has proven to be very successful in quantitatively modelling aging decays, including temperature cycling.

FIG. 3: TRM experiment interpreted using a schematic of the Barrier model. During the cooling process, there is a "seeding" of the phase space. An initial distribution is set up. During the waiting time, states begin to diffuse into the phase space. The barrier associated with waiting time is $\Delta_{tw}$. The initial state occupation is formed upon cooling the spin glass through its transition temperature to some measuring temperature, in a small magnetic field (Fig-
Magnetic field cycling during the waiting time has previously been employed to probe the redistribution of states in going from the field cooled manifold to the zero field manifold and back. The initial state occupation was determined to be a delta function located at zero energy barrier with a possible background contribution from states close to the delta function. In reality, this study probed the initial distribution within the zero field manifold (after the field was cut) and this distribution was also taken to be the initial distribution in the field cooled manifold after initial cooling. In the present study we find that it is not the case that the initial distribution in the field cooled manifold is a delta function at the origin. Instead, we are only able to fit the long-time decay data by assuming that the states formed during the cooling process are distributed uniformly over all possible states (i.e. over all barriers). This has several significant implications. First, it has a self similar background which is necessary for understanding the temperature dependence of the TRM. Secondly, it suggests that correlations of all possible strengths are formed during the cooling process. Finally, the uniform background produces a logarithmic decay, implying a limiting barrier height and hence a limit on aging.

In Figure 2 we plot a fit of the whole 7s waiting time TRM decay using the results of the barrier model (\(\Delta_0 = 1.2\), \(r = 1.048\) and \(N_{\max} = 800\) barriers), evolved from a constant background initial state, coupled with a power law term to model the “short time” stationary part of the decay (Eq. 1). Values for the power law were those determined from the 10,000 second waiting time data, previously mentioned. Extrapolating (straight line logarithmic dependence of the barrier model) to zero magnetization, we find a limiting age (corresponding to a maximum barrier) of \(t_{\max} \approx 1.1 \times 10^{11}\) seconds. This corresponds to approximately 3000 years. This remarkable result implies that the phase space, at least in this sample, is limited in the time domain.

We can test the assumption of an initial uniform state distribution over all barriers by performing an Isothermal Remnant Magnetization (IRM) measurement. In this experiment the sample was cooled in zero magnetic field, producing an initial distribution of states within the zero field (zero magnetization) manifold. The sample is kept in zero field for a time \(t_{w1} = 10,000\) seconds. A small magnetic field (20G) is then applied for a time \(t_{w2}\). After \(t_{w2}\) the magnetic field is removed and the decay observed. The data is plotted for several different values of \(t_{w2}\) in Figure 3.

We start our analysis of the data with the assumption that the field cool (FC) and zero field (ZF) manifolds are equivalent. This assumption has strong experimental backing. The cooling procedure in the IRM should therefore produce an equivalent distribution of states in ZF manifold as the TRM produces in the FC manifold. Waiting for a long time \(t_{w1}\) allows enough time for states, which contribute to aging, to diffuse over significantly large barriers in the ZF manifold. When the magnetic field is applied a small number of these aging states diffuse back to the zero barrier and jump to the FC manifold. These states then begin to age, starting from the zero energy barrier, in the FC manifold for a time \(t_{w2}\). Removal of the magnetic field then produces a measurable decay of the states back from the FC manifold to the ZF manifold. If our assumption that the background is due to the cooling effect, then the uniform background should be located deep within the ZF manifold and should not be observed in the IRM experiment. This is exactly what is observed. We do find an aging decay followed by a very small power law decay with an exponent compatible with that observed in the short times stationary measurements.

![FIG. 4: IRM decays. We use \(t_{w1} = 10000s\) and vary \(t_{w2} : 10(\square), 25(\bigcirc), 100(\bigtriangleup), 150(\bigtriangledown), 300(\bigtriangledown), 1000(\bigtriangleleft)\) and \(3000(\blacktriangleright)\) seconds. The fitted curve is a power law with the power, \(\alpha = -01\).](image)

In conclusion, we have measured very long time TRM measurements with rapid cooling protocols and small waiting times. We find that that the aging time is finite in extent and that there is a longtime decay which is independent of the waiting time. This decay is different from the short times stationary term and related to the mechanisms responsible for producing aging in spin glasses.
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