

Time Decay of the Remanent Magnetization in Spin-Glasses

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The time decay of the thermoremanent magnetization (σ_{TRM}) has been measured in 1.0% Cu:Mn and 2.6% Ag:Mn spin-glasses. It is shown that σ_{TRM} is neither an algebraic nor a logarithmic function of time, but it is found that σ_{TRM} can be characterized by a "stretched" exponential: $\sigma_{\text{TRM}} = \sigma_0 \exp[-C(\omega t)^{1-n}/(1-n)]$. Similar time dependences appear in the disorder-diffusion theory of Grassberger and Procaccia and the cooperative-relaxation theory of Ngai, but neither theory in its present form is directly applicable to spin-glasses.

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When measured in a small static field, the temperature dependence of the magnetization of a spin-glass changes abruptly at the glass temperature (T_g): Above T_g the magnetization obeys the Curie-Weiss law, attributable to weakly interacting paramagnetic spins, whereas below T_g the magnetization is nearly independent of temperature, indicative of the spin-glass state. The time dependence of the magnetization also changes dramatically in the vicinity of T_g : In the paramagnetic region the entire magnetization responds rapidly to a change in field, but in the spin-glass region some of the magnetization responds much more slowly.¹ One technique of investigating this viscous behavior is to apply a field (H) when the sample is in the paramagnetic region, field cool it through T_g , then remove H and measure the field-cooled remanence or "thermoremanent magnetization" (σ_{TRM}).²

The exact form of the time dependence of σ_{TRM} has not been previously established. Many investigators³ have reported a logarithmic decay: $\sigma_{\text{TRM}} = \sigma_0 [1 - (1-n)\log(t)]$, where σ_0 and n are constants. Such a decay is unbounded, however, and must be merely an approximation, valid over some finite interval of time. The Sherrington-Kirkpatrick mean-field model⁴ has been successful in describing many of the observed properties of spin-glasses. Calculations based on this model⁵ suggest that the magnetization should decay algebraically: $\sigma_{\text{TRM}} = \sigma_0/t^{1-n}$. This suggestion seems plausible since for $n < 1$ it approaches the correct equilibrium at long times (zero magnetization in zero field⁶), but no data have yet been published⁷ supporting an algebraic decay for σ_{TRM} .

We have made magnetization measurements in the interval from 0.2 to 1000 sec after removing H and conclude that σ_{TRM} has neither an algebraic nor a logarithmic time dependence. We do, however, find that the time dependence is accurately charac-

terized by a "stretched" exponential of the form

$$\sigma_{\text{TRM}} = \sigma_0 \exp[-C(\omega t)^{1-n}/(1-n)]. \quad (1)$$

Here the exponential factor (C) and relaxation frequency (ω) can be chosen to be independent of temperature throughout the spin-glass region, whereas the prefactor (σ_0) and time-stretch exponent (n) are temperature-dependent constants.

We have made time-decay measurements on three different samples: 1.0% Cu:Mn, 2.6% Ag:Mn, and 2.6% Ag:Mn + 0.46% Sb, all of which show qualitatively similar behavior. Here we will present only the 2.6% Ag:Mn + 0.46% Sb data. Measurements were made on a stack of thin ($\sim 25 \mu\text{m}$) foils with a total mass of 0.223 g. The glass temperature for this sample ($T_g = 9.30 \text{ K}$) was determined from the maximum in the magnetization in a static field of 3 Oe. We used a SQUID magnetometer to measure σ_{TRM} as follows: (1) a magnetic field ($H = 30 \text{ Oe}$ or $H = 15 \text{ Oe}$) was applied to the sample when in the paramagnetic region, (2) the sample was field cooled through T_g to a temperature in the spin-glass region, (3) H was removed and the time dependence of the magnetization was recorded by a computer-based data acquisition system; (4) after 1000 sec the remaining remanence was measured by warming the sample through T_g to establish the base line.

Figure 1(a) is a plot of σ_{TRM} vs $\log(t)$ at four temperatures within the spin-glass region. The fact that the slope changes with time shows that σ_{TRM} does not decay logarithmically. Figure 1(b) is a log-log plot of the same data, here demonstrating that σ_{TRM} does not decay algebraically. In Fig. 2 we show that σ_{TRM} decays by a stretched exponential function of time. The time-stretch exponent (n) is most easily determined by plotting $\log[-(d/dt)(\ln \sigma_{\text{TRM}})]$ as a function of $\log(t)$. This is done in Fig. 2(a) where a fit to the data after 5 sec yields:

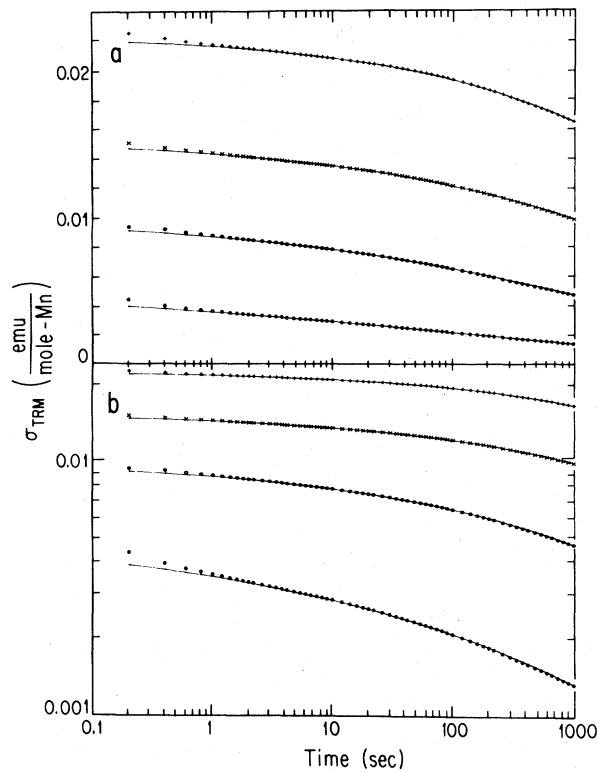


FIG. 1. (a) Semilog and (b) log-log plot of the thermoremanent magnetization (σ_{TRM}) of 2.6% Ag:Mn + 0.46% Sb as a function of time at $T/T_g = 0.771, 0.856, 0.897,$ and 0.966 , from top to bottom, respectively. The solid curves are the best stretched-exponential fits to the data.

$n = 0.694, 0.740, 0.766,$ and 0.831 for $T/T_g = 0.771, 0.856, 0.897,$ and 0.966 , respectively. The stretched exponential nature of σ_{TRM} is verified in Fig. 2(b) by the linear dependence of $\log(\sigma_{\text{TRM}})$ on t^{1-n} . The quality of agreement is also shown in Fig. 1, where we plot the best stretched exponential fit to each set of data. The deviation before 5 sec can be attributed to the decay of induced eddy currents in the metallic samples as is evidenced by a measurable magnetic absorption,⁸ even in the paramagnetic region, at frequencies above 0.2 Hz. After 5 sec, σ_{TRM} is accurately characterized with the four parameters $\sigma_0, C, \omega,$ and n . Note that no adjustment is allowed in the base line of the data since it is unambiguously established in step 4 of our procedure.

The temperature dependence of the prefactor and the time-stretch exponent is shown in Fig. 3. At low temperatures ($T < 0.75T_g$) σ_0 decreases linearly with increasing temperature whereas $n \approx \frac{2}{3}$ independent of temperature. For $T > 0.75T_g, n$ in-

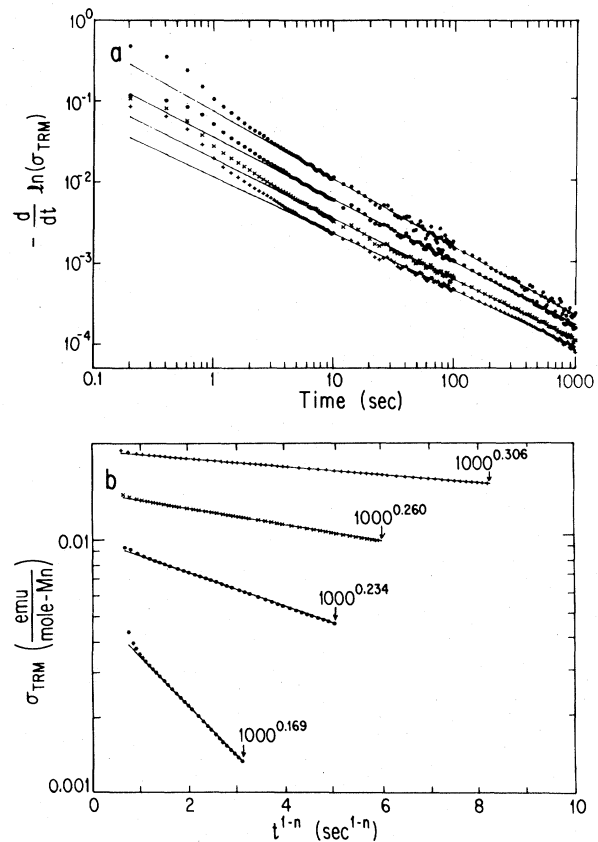


FIG. 2. Determination of the time-stretch exponent, n . (a) Log-log plot of $-d \ln(\sigma_{\text{TRM}})/dt$ as a function of time. The slope gives $-n$ and the $t=1$ -sec intercept gives $C\omega^{1-n}$. (b) Semilog plot of σ_{TRM} as a function of t^{1-n} . The solid lines are the best fits to the data.

creases while σ_0 decreases more rapidly than at lower temperatures. The temperature dependence of n allows us to extract the relative values of ω and C by plotting $\log(C\omega^{1-n})$, obtained from Fig. 2(a), as a function of $1-n$. This is done in Fig. 4 where linear regression to the data yields $C = 0.59 \pm 0.05$ and $\omega = (3 \pm 1) \times 10^{-6} \text{ sec}^{-1}$. The only difference we measure between cooling the sample in $H = 15$ Oe and in $H = 30$ Oe is a linear dependence σ_0 on H . $n, C,$ and ω do not depend on the cooling field for these relatively small values of H ; thus, the changes in n and σ_0 for $T > 0.75T_g$ cannot be attributed to the saturation of σ_{TRM} in H .⁹

Two separate theories of relaxation, which have recently appeared in the literature, give a stretched exponential time behavior similar to that of Eq. (1). Neither theory has yet been applied to spin-glasses, so we can only point out the possibility of a connection. The first theory, due to Grassberger and Pro-

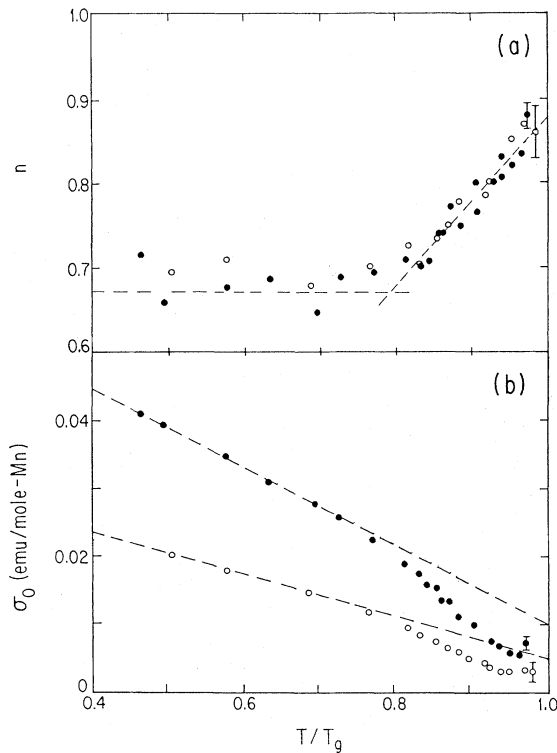


FIG. 3. (a) The temperature dependence of n . (b) The temperature dependence of the prefactor (σ_0). The solid circles are for a 30-Oe cooling field and the open circles are for $H=15$ Oe. The dashed lines are guides for the eye.

caccia,¹⁰ considers the diffusion of particles through a d -dimensional space interspersed with randomly distributed traps. A connection to spin-glasses can be made by using the model of Bantilan and Palmer¹¹ in which the energy of a spin-glass is pictured as a labyrinthine function in spin-configuration space containing many maxima and local minima, and several quasidegenerate ground-state minima. The configuration of a group of spins travels randomly through configuration space until it is trapped into one of the ground-state minima. Grassberger and Procaccia find that the number of untrapped "configurations" (N_c) is given by

$$N_c \approx f(t) \exp[-g(n)(\omega t)^{1-n}/(1-n)],$$

where the time-stretch exponent is related to the dimension of the diffusive space by $n = 1 - d/(d + 2)$. Although this interpretation is interesting, three experimental facts emphasize the need for further development before this model may apply to spin-glasses. First, σ_{TRM} obeys a simple stretched exponential of time; we do not measure any time dependence to the prefactor [$f(t)$].

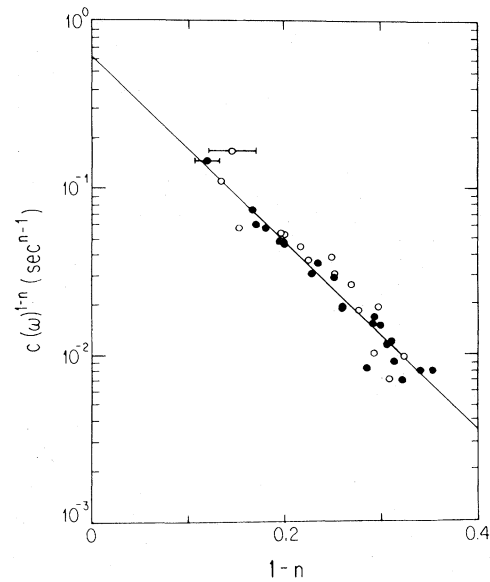


FIG. 4. Semilog plot of $C\omega^{1-n}$ as a function of $1-n$ for $H=30$ Oe (solid circles) and $H=15$ Oe (open circles). The solid line is the best fit to the data. The slope of this line gives $\omega = (3 \pm 1) \times 10^{-6} \text{ sec}^{-1}$ and the intercept gives $C = 0.59 \pm 0.05$.

Second, we can choose ω so that the exponential factor [$g(n)$] is independent of n . And third, we find $n \geq \frac{2}{3}$, which implies a nonphysical ($d \leq 1$) configuration space.

The second theory giving a stretched exponential time decay is due to Ngai.¹² The Ngai theory treats the cooperative relaxation of a primary system of dipoles perturbatively coupled (coupling constant V) to a secondary continuum of low-energy excitations whose density of available levels is linear in energy: $N(E) = \alpha E$. In the Ngai model the measured susceptibility of a sample is due entirely to the particular microstate of the dipolar system, but the rate at which the dipoles approach equilibrium is influenced by the continuum. If the coupling could be ignored, the dipoles would make transitions at some time-dependent rate, $1/T_0$. But if this coupling is not negligible, the dipole transitions will excite the continuum, which in turn will influence the transition rate. Ngai gives the susceptibility as a function of time [$\phi(t)$] from which the magnetic response to the removal of H at $t=0$ is easily found:

$$M(t) = H \int_0^t \psi(t') dt' = H \psi_0 \exp\left[-\left(\frac{e^{-\gamma}}{T_0 E_c^n}\right) \frac{t^{1-n}}{1-n}\right], \quad (2)$$

where E_c is a cutoff energy for the low-energy excitations and the time-stretch exponent is given by $n = \alpha V^2$. If E_c and $1/T_0$ are set equal to ω , then Eq. (2) has the same form as Eq. (1). The Ngai theory gives a specific value for the exponential factor in terms of Euler's constant: $C = e^{-\gamma} = 0.5615\dots$ which, to within experimental accuracy, is the value we measure. The Ngai model empirically characterizes the observed time dependence of σ_{TRM} , but a significant inconsistency still exists: Eq. (2) is an approximation supposedly valid only at long times ($E_c t \gg 1$), which for $E_c = \omega \sim 10^{-5} \text{ sec}^{-1}$ is never achieved in our measurements. We cannot yet explain why Eq. (2) seems to be valid throughout the time regime of our measurements. Furthermore, the source of the low-energy excitations, why $1/T_0$ should equal E_c , and the temperature dependences of σ_0 and n are not yet understood.

In conclusion, we have shown that the decay of σ_{TRM} in spin-glasses is neither a logarithmic nor an algebraic function of time. It is, however, accurately characterized at all temperatures within the spin-glass region by a stretched exponential [Eq. (1)] with four adjustable parameters. The prefactor (σ_0) and time-stretch exponent (n) are temperature dependent, whereas the exponential factor (C) and relaxation rate (ω) can be chosen to be independent of temperature throughout the spin-glass region. In addition, we find σ_0 to depend linearly on the cooling field, whereas n , C , and ω are independent of H for $H \leq 30 \text{ Oe}$. We point out possible connections to two recent theories of relaxation, but emphasize that further development is necessary before either theory may be applied to the decay of σ_{TRM} in spin-glasses.

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⁷J. Ferré, J. Rajchenbach, and H. Maletta, *J. Appl. Phys.* **52**, 1697 (1981), find that σ_{TRM} in $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ is better characterized by an algebraic decay than by a logarithmic decay, but they conclude that both forms are still only approximations.

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⁹The data and analysis of H. Bouchiat and P. Monod, *J. Magn. Magn. Mater.* **30**, 175 (1982), give $H_{\text{TRM}} = (2400 \text{ Oe}) \exp(-2.1T/T_g)$ for 2.8% Ag:Mn, where H_{TRM} is a rough measure of the field above which σ_{TRM} becomes nonlinear. Thus for $T = 0.75T_g$, $H_{\text{TRM}} \sim 500 \text{ Oe}$, which supports our evidence that the change in n and σ_0 for $T > 0.75T_g$ is not due to the saturation of σ_{TRM} .

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