Spin-Glass Response Time in Ag:Mn: Exponential Temperature Dependence

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The time dependence of the magnetization of the dilute spin-glass Ag:Mn (2.6 and 4.1 at.%) prepared in the field-cooled state has been measured after the field has been cut off at a constant waiting time. The time dependence is found to be of the stretched exponential form, with a rate constant which varies exponentially with inverse temperature. This result appears consistent with an exponential distribution of independent, random free-energy levels in the spin-glass phase.

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The field-cooled state of the dilute metallic spinglass Ag:Mn is known¹ not to be the equilibrium state. "Waiting-time" experiments of Chamberlin¹ have shown that the system is relaxing to states with everincreasing time constants as one "waits" during the time that the material has been cooled below the glass temperature T_g while holding the field constant. Similar effects have been observed by Lundgren *et al.*² in the zero-field-cooled state. Chamberlin, Mozurkewich, and Orbach³ found that when the field was cut off the magnetization recovered according to a stretched exponential

$$\sigma_{\text{TRM}}(t) = \sigma_0 \exp[-(t/\tau_n)^{1-n}],$$

where TRM stands for thermoremanent magnetization, and we have used Ngai's form for the time dependence of the magnetization recovery.⁴ This form agrees with an independent and remarkable recent calculation of De Dominicis, Orland, and Lainée,⁵ who calculate the time evolution of the squares of the spin-glass-state occupancies within the mean-field approximation (see below).

We have extended Chamberlin's experiments to considerably lower reduced temperatures. Our experiments exhibit the property that the characteristic rate $1/\tau_p$ varies exponentially with inverse temperature: $1/\tau_p = A \exp[-\alpha T_g/T]$, with $A \simeq 10^{-3} \sec^{-1}$ and $\alpha \simeq 2.6$. Remarkably, this same temperature dependence is contained in the work of De Dominicis, Orland, and Lainée.⁵ Their calculation is based on the recent work of Mézard *et al.*,⁶ Derrida and Toulouse,⁷ Mézard, Parisi, and Virasoro,⁸ and Sourlas.⁹ We shall offer an interpretation which claims that $1/\tau_p$ mirrors the calculated exponentially increasing "sparsity" of thermally accessible independent random free-energy levels as the temperature is lowered.

The measurements of the time decay of the TRM of Ag:Mn_{2.6 at.%} ($T_g = 10.18$ K) and Ag:Mn_{4.1 at.%} ($T_g = 14.40$ K) were performed on (annealed) samples consisting of about four equally sized foils with approximate dimensions of $15 \times 5 \times 0.15$ mm³. The magnetization was measured with an S.H.E. multifunction

probe and the S.H.E. 330 SQUID electronics. The SQUID magnetometer, cryogenics, and sample preparation have been described extensively elsewhere.^{3,10} The measurements were made in the temperature range 2.25-14.30 K. The experimental procedure has been described previously,¹¹ but will be described briefly here for convenience. The sample, positioned in one of the coils of the (first derivative) pickup coil system, is warmed up to a temperature above T_g and subsequently field cooled in a field of 6 Oe to a temperature below T_g . After a certain waiting time, t_w , the external magnetic field is switched off and the time decay of the TRM is observed over a time period of 500 sec. The waiting time, t_w , is defined as the time lag between the instant that, while cooling, T_g is reached, and the instant that the field is turned off. Next, the sample is warmed up to a temperature above T_{e} in order to establish the base line, and the entire procedure is repeated with the sample placed in the lower coil of the two-coil system. Subtraction of the upper- and lower-coil magnetization signals finally yields the time decay of the TRM, free from systematic background drift.

In Fig. 1, the time decay of the TRM (in arbitrary units) is exhibited for the Ag:Mn_{2.6 at.%} sample for $t_w = 10$ min and at various temperatures. Excellent fits, as represented by the solid lines in Fig. 1, of the stretched exponential,

$$\sigma_{\text{TRM}}(t) = \sigma_0 \exp[-(t/\tau_p)^{1-n}] \tag{1}$$

to the experimental data could be made over almost the entire observing period (0.2-500 sec), i.e., over three decades in time. From these fits, the value of the initial TRM, σ_0 , the characteristic time constant, τ_p , and the stretched exponential exponent, *n*, were obtained. The values of σ_0 , τ_p , and *n* for the Ag:Mn_{4.1 at.%} sample were also obtained from fits by Eq. (1). The dependence of *n* on the reduced temperature, $T_r = T/T_g$, for $t_w = 10$ min is exhibited in Fig. 2 for both alloys. The error bars in Fig. 2 were determined from the scatter in the experimental results for *n* (about 10% of *n*) at fixed temperature.



FIG. 1. The time decay of the thermoremanent magnetization (TRM) for Ag:Mn_{2.6 at. %} at a constant 10-min waiting time and for various temperatures. The magnetization (in arbitrary units) is plotted vs log t. The lines represent the fits of the data by the stretched exponential form $\sigma_{\text{TRM}}(t) = \sigma_0 \exp[-(t/\tau_p)^{1-n}]$.

Inspection of Fig. 2 shows that, for both alloys, *n* is to within experimental error constant $(n \sim 0.63)$ for $0.5 \leq T_r \leq 0.8$ while *n* increases as T_r increases from 0.8 or as T_r decreases below about 0.5. Measurements at different waiting times (5 min $\leq t_w \leq 60$ min) show that, to within experimental accuracy, *n* (and σ_0) is independent of t_w , in agreement with previous observations.^{1,3,10} Finally, we exhibit in Fig. 3 the temperature dependence (at fixed $t_w = 10$ min) of the logarithm of the characteristic response rate, τ_p^{-1} , plotted versus the inverse reduced temeprature $T_r^{-1} = T_g/T.^{12}$

The most remarkable aspect of our results can be seen in Fig. 3. The characteristic rate, $1/\tau_p$, scales with inverse reduced temperature over the entire temperature range of measurement, and varies itself over 7 orders of magnitude for a temperature change of less than an order of magnitude. The variation for $T_g/T > 1.1$ accurately follows the form

$$1/\tau_p = A \exp[-\alpha (T_g/T)], \qquad (2)$$

with $A \cong 10^{-3} \sec^{-1}$ and $\alpha \cong 2.5$.

Completely independent of our experimental findings, De Dominicis, Orland, and Lainée⁵ have obtained a time dependence for the sum of the squares of the spin-glass-state occupancies which is identical in form to the stretched-exponential dependence exhibited in Eq. (1). They made explicit use of the Mézard, Parisi, and Virasoro⁸ study of the free energies of the pure states in the spin-glass phase for the Sherrington-Kirkpatrick model.¹³ Letting the free energy per spin be denoted by f_{α}/N , Mézard, Parisi, and Virasoro found the f_{α} to be independent random vari-



FIG. 2. A plot of the dependence of the stretchedexponential exponent, *n*, against the reduced temperature $T_r = T/T_g$. The solid circles are the results for the Ag:Mn_{2.6 at.%} sample, and the squares are the results for the Ag:Mn_{4.1 at.%} sample.



FIG. 3. A plot of the characteristic rate $1/\tau_p$ vs inverse reduced temperature T_g/T , on a semilogarithmic plot, for the Ag:Mn_{2.6 at.%} (solid circles) and the Ag:Mn_{4.1 at.%} (squares) dilute alloys. Note that the results for the two concentrations lie on top of one another for all temperatures, indicating a scaling behavior according to the relationship $1/\tau_p = A \exp[-\alpha T_g/T]$, with $A \simeq 10^{-3} \sec^{-1}$ and $\alpha \simeq 2.5$. The solid line is the best fit with this form.

ables with an exponential distribution

$$\mathscr{P}_{\rho}(f_{\alpha}) = \rho \{ \exp[\rho(f_{\alpha} - f_{c})] \} \theta(f_{c} - f_{\alpha})$$

where ρ is a function of the temperature and the external field *H* and equals $\beta(1-y)$, with $\beta = 1/k_B T$, and *y* the width of the right plateau in the q(x) plot¹⁴, and f_c is a cutoff energy. Conversely, starting from the random (free) energy model of Derrida,¹⁵ Derrida and Toulouse⁷ have independently obtained the same distribution. It has also been derived previously in Ref. 6 by use of the Parisi¹⁶ mean-field solution.

De Dominicis, Orland, and Lainée⁵ then introduced a simple choice for the relaxation rate which reduced the coupled equations for the time dependence of the occupation probabilities of the individual free-energy levels to an independent set of time dependences for each level but with a relaxation time which was random and depended on all available random free energies. Their final result for the time dependence of the sum of the squares of the perturbed spin-glass-state occupancies was of the stretched exponential form, with an exponent n [in the notation of our Eq. (1)] = y. They noted that $y \rightarrow 1$ as $T \rightarrow T_g$ from below for the Sherrington-Kirkpatrick model, in rough agreement with our results (see Fig. 2). The temperature dependence we have found for τ_p in Eq. (2) appears explicitly in their formulation.

A physical argument for our results, especially in Eq. (2), can be made as follows. It has been argued¹⁷ that changing the magnetic field after field cooling destroys the minimum in which the system "sits" (but remember: The system is still not in equilibrium because of the waiting-time effect). The system must then seek other minima to return towards equilibrium. However, the lower the temperature, the smaller the number of accessible states according to the form for $\mathcal{P}_{\rho}(f_{\alpha})$ given by Mézard, Parisi, and Virasoro.⁸ Hence, the characteristic response time would be expected to lengthen as the temperature is lowered. Why it should be nearly precisely proportional to the number density of available states is not clear, but our experiments seem to argue for such a proportionality.

In summary, our experiments exhibit properties which are not inconsistent with recent mean-field calculations for the Sherrington-Kirkpatrick model. The stretched exponential form for the recovery of the magnetization which we observe is identical in form to that derived in a recent calculation by De Dominicis, Orland, and Laniée⁵ for the time evolution of the squares of the spin-glass-state occupancies. The observed response rate, $1/\tau_p$, appears to mirror the calculated temperature-dependent distribution of the free-energy states of the spin-glass. An identical temperature dependence has been obtained explicitly by De Dominicis, Orland, and Lainée.⁵ They have not yet computed the time-dependent response of the magnetization under conditions analogous to our experimental conditions, but their results are highly suggestive that, for the first time, a microscopic theory can agree directly with experiment for a spin-glass.

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¹R. V. Chamberlin, Phys. Rev. B 30, 5393 (1984).

²L. Lundgren, P. Svedlindh, P. Norblad, and O. Beckman, Phys. Rev. Lett. **51**, 911 (1983); L. Lundgren, P. Svedlindh, and O. Beckman, Phys. Rev. B **26**, 3990 (1982).

³R. V. Chamberlin, George Mozurkewich, and R. Orbach, Phys. Rev. Lett. **52**, 867 (1984).

⁴K. L. Ngai, Comments Solid State Phys. **9**, 127 (1979), and **9**, 141 (1980); K. L. Ngai, A. K. Rajagopal, and C. Y. Huang, J. Appl. Phys. **55**, 1714 (1984).

 ${}^{5}C$. De Dominicis, H. Orland, and F. Lainée, to be published.

⁶M. Mézard, G. Parisi, N. Sourlas, G. Toulouse, and M. A. Virasoro, Phys. Rev. Lett. **52**, 1146 (1984), and J. Phys. (Paris) **45**, 843 (1984).

⁷B. Derrida and G. Toulouse, to be published.

⁸M. Mézard, G. Parisi, and M. A. Virasoro, to be published.

⁹N. Sourlas, private communication.

¹⁰R. V. Chamberlin, thesis, University of California at Los Angeles, 1984 (unpublished).

 11 R. V. Chamberlin, M. Hardiman, L. A. Turkevich, and R. Orbach, Phys. Rev. B **25**, 6720 (1982).

¹²The error bars in Fig. 3 were estimated from the scatter in the values of $1/\tau_p$ at fixed temperature. As the temperature is lowered, the error in $1/\tau_p$ increases, as indicated in Fig. 3, because of the relatively short observation time (500 sec).

 13 D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **32**, 1792 (1975).

¹⁴G. Parisi, Phys. Rev. Lett. **50**, 1946 (1983).

¹⁵B. Derrida, Phys. Rev. Lett. **45**, 79 (1980), and Phys. Rev. B **24**, 2613 (1981).

¹⁶G. Parisi, Phys. Rev. Lett. **43**, 1745 (1979), and J. Phys. A **13**, L-115, 1101, 1887 (1980).

¹⁷Gary G. Grest, C. M. Soukoulis, and K. Levin, J. Appl. Phys. 55, 1634 (1984).