

Temperature dependence of the response time of dilute metallic spin glasses

R. Hoogerbeets, Wei-Li Luo, and R. Orbach

Department of Physics, University of California, Los Angeles, California 90024

(Received 21 January 1986)

The temperature dependence of the time decay of the thermoremanent magnetization has been measured for four dilute metallic spin glasses: Ag:Mn_{2.6} at. %, Ag:Mn_{4.1} at. %, Ag:Mn_{2.6} at. % + Sb_{0.46} at. %, and Cu:Mn_{4.0} at. %. After cooling in a constant applied field, the field was cut to zero and the time decay of the thermoremanent magnetization was observed over a period of 0.2–500 sec. The time dependence of the thermoremanent magnetization can be described well by a stretched exponential: $\sigma_{\text{TRM}}(t) = \sigma_0 \exp[-(t/\tau_p)^{1-n}]$, with an apparent rate, $1/\tau_p$, which varies exponentially with the inverse reduced temperature over nearly the entire temperature range of measurement below T_g , the glass temperature. Moreover, the temperature dependence of $1/\tau_p$ is given by a universal function, $1/\tau_p = A \exp(-2.5T_g/T)$, with $A = 10^{-3} \text{ sec}^{-1}$. Very near to T_g , the apparent rate $1/\tau_p$ decreases much more rapidly as T diminishes, and the scaling with T_g breaks down. All of these results can be mapped onto a recent calculation of the dynamics of the infinite-range Ising spin-glass model by De Dominicis *et al.* We are able to fit the observed temperature dependence of $1/\tau_p$ over the full temperature range ($T \leq T_g$) using either the experimentally determined values for the quantities in the theory, or those values extracted from the Sherrington-Kirkpatrick model.

I. INTRODUCTION

The time decay of the thermoremanent magnetization of spin glasses has been studied experimentally in great detail in recent years.^{1–13} The time decay of the field-cooled thermoremanent magnetization (TRM) of both dilute metallic^{1–8} and insulating^{9–12} spin glasses has been found to be accurately characterized by the stretched exponential form: $\sigma_{\text{TRM}} = \sigma_0 \exp[-(t/\tau_p)^{1-n}]$, at least for relatively small applied fields and times in the ranges 5–10³ sec. Moreover, careful measurements of Chamberlin³ showed that the field-cooled spin-glass state is not an equilibrium state. Chamberlin found that the apparent response time τ_p increased exponentially with the “waiting time,” i.e., the time after the temperature falls below the glass transition temperature until the external field is cut to zero. The magnetization is found in our experiments to be nearly constant, however, during this same time period. The waiting-time dependence of τ_p clearly indicates that the system is relaxing between states of nearly the same magnetization, possessing ever increasing values of τ_p . This feature had been observed for the zero-field-cooled spin-glass state for metallic spin glasses by Lundgren *et al.*,^{13,14} and has been observed more recently in measurements on insulating spin glasses.^{8,11,12}

A fully satisfactory explanation for the stretched exponential form of the time decay of the TRM, and the waiting-time dependence of the characteristic response rate, when available, will certainly illuminate the true character of the spin-glass state. Recent theoretical developments appear to be making substantial progress in this regard. Parisi showed in a seminal paper¹⁵ that the order-parameter function, $q(x)$, associated with the replicate symmetry-broken solution of the infinite-range Ising spin glass model (Sherrington-Kirkpatrick model), is

directly connected to the overlap of any two pure states of the spin-glass state. Then, using the Parisi ansatz,¹⁶ Mézard *et al.*¹⁷ showed that the order-parameter function, interpreted as a probability law, is not a self-averaging quantity. Instead, it depends on the specific distribution of the coupling constants (“the realization”) of the specific sample, even in the thermodynamic limit.

Furthermore, Mézard *et al.* found that the space of pure equilibrium states has ultrametric topology, and thus a hierarchical structure. They showed¹⁷ that the fluctuations of the free energies associated with the pure states are independent random variables with an exponential distribution. Independently, and at the same time, Derrida and Toulouse¹⁸ derived an identical exponential free-energy distribution for the pure states without the replica trick, but using the random energy model introduced by Derrida.¹⁹ Finally, De Dominicis *et al.*²⁰ used a simple model for relaxation to show that $x(q)$, the inverse of the Parisi order-parameter function $q(x)$, relaxes in time according to a stretched exponential form. These new theoretical developments have considerably enlarged our understanding of the infinite-range Ising model, and appear to have significant experimental relevance. Our purpose in this paper is to show how our recent time-dependent measurements of a number of metallic spin glasses can be understood on the basis of the model of De Dominicis *et al.*²⁰ Though its applicability is by no means obvious, its successful description of measured spin-glass dynamics^{4,5} can hardly be ignored.

In a recent Letter⁴ we reported the results of our measurements of the temperature dependence of the apparent response time τ_p for two dilute metallic spin glasses, Ag:Mn_{2.6} at. % and Ag:Mn_{4.1} at. %. Our measurements showed that the apparent response rate, $1/\tau_p$, of these two spin glasses is approximately exponential in the inverse re-

duced temperature, T_r^{-1} ($T_r^{-1} = T_g/T$, where T_g is the glass transition temperature), over nearly the entire temperature range of measurement (viz., $T_r^{-1} \geq 1.1$). It increases much more rapidly than exponentially as the glass transition temperature is approached from below for $1.0 \leq T_r^{-1} \leq 1.1$. It was also found that the characteristic response rate for both alloys scaled with T_r only over the former temperature range $1.1 \leq T_r^{-1} \leq 5.65$.

We report the results of an extended study of the temperature dependence of the apparent response rate for both regimes. We have measured the temperature dependence of $1/\tau_p$ of two additional dilute metallic spin glasses, namely Ag:Mn_{2.6 at. %} + Sb_{0.46 at. %} and Cu:Mn_{4.0 at. %}. The former is known to have a larger magnetic anisotropy than Ag:Mn_{2.6 at. %} because of the antimony doping,²¹⁻²⁴ while the latter has a higher glass transition temperature ($T_g = 22.9$ K) and a smaller magnetic anisotropy than Ag:Mn_{2.6 at. %}. Our measurements on these two additional samples also exhibit an apparent response rate which scales with the glass temperature, and varies approximately exponentially with the inverse reduced temperature over a large temperature range ($1.5 \lesssim T_r^{-1} \lesssim 5.35$). However, as before, $1/\tau_p$ increases much more rapidly than exponentially in T_r^{-1} as the glass transition temperature is approached from below.

We shall show that the temperature dependence of $1/\tau_p$ for all four spin glasses over the entire temperature range of measurement is in excellent agreement with the temperature dependence of the quantity $x(t)$ derived recently by De Dominicis *et al.*²⁰ This will be true using experimental values for the quantities which appear in the theory, or using the values for these quantities obtained for the Sherrington-Kirkpatrick infinite-range spin glass model.

The experimental methods are outlined in Sec. II of this paper. We present our new experimental results in Sec. III, including some of our previous measurements (including the recently reported measurements⁵ of the temperature and field dependence of the apparent response rate of Ag:Mn_{2.6 at. %} + Sb_{0.46 at. %}) for completeness. An extended analysis of our results in terms of the Sherrington-Kirkpatrick infinite-range Ising model is presented in Sec. IV, while we summarize our results in Sec. V.

II. EXPERIMENTAL METHODS

The measurements of the time decay of the thermoremanent magnetization (TRM) were performed on four dilute metallic spin glasses: Ag:Mn_{2.6 at. %} ($T_g = 10.18$ K), Ag:Mn_{4.1 at. %} ($T_g = 14.40$ K), Ag:Mn_{2.6 at. %} + Sb_{0.46 at. %} ($T_g = 9.30$ K), and Cu:Mn_{4.0 at. %} ($T_g = 22.90$ K). The samples were made by arc-melting 99.99%-pure Ag or Cu with 99.9999%-pure Mn in an argon atmosphere. The alloys were next annealed (again in an argon atmosphere) at about 900°C for more than eight hours, and subsequently quenched to room temperature in a time shorter than a minute. The silver manganese alloys were rolled out into foils (about 150- μ m thick) and again annealed and quenched. Finally, samples were made out of about four equally sized foils ($15 \times 5 \times 0.15$ mm³). A powder sample, with an average

grain size of about 1.5 mm, was also obtained by filing a lump of the Ag:Mn_{2.6 at. %} alloy. The Cu:Mn_{4.0 at. %} sample which was used in our experiments has a spherical shape with an approximate diameter of 3 mm.

The measurements of the time decay of the thermoremanent magnetization were performed as before.^{1,3,4,9} The magnetization was measured with a Superconducting Helium Electronics (SHE) multifunction probe and SHE 330 superconducting quantum interference device electronics. The experimental procedure was as follows. The sample was placed in one of the coils of a first-derivative pick-up-coil system. It was field cooled ($H \cong 6$ Oe) from a temperature above T_g to the measurement temperature T below T_g . After a certain waiting time, t_w , the external magnetic field was cut to zero, and the time decay of the TRM was measured over a period of no more than 500 sec. The waiting time is defined as the time difference between the instant that, while cooling, T_g is reached, and the instant that the applied field is cut to zero. After the decay of the TRM is measured, the sample is warmed up (in zero field) to a temperature above T_g and the baseline is established. Next, the procedure is repeated with the sample placed in the other coil of the pick-up-coil system. Finally, subtraction of the upper- and lower-coil magnetization signals yields the time decay of the TRM, free from any systematic background signal.

The values of σ_0 , the initial value of the TRM, n (the stretched exponential exponent), and t_p (the apparent response time) were obtained from fits of the stretched exponential form, $\sigma_{\text{TRM}}(t) = \sigma_0 \exp[-(t/\tau_p)^{1-n}]$, to the experimental data.

III. EXPERIMENTAL RESULTS

The waiting-time dependence of the apparent response rate $1/\tau_p$ for both the powder and foil sample of Ag:Mn_{2.6 at. %} at $T = 4.35$ K, is plotted in Fig. 1 as $\log_{10}(1/\tau_p)$ versus t_w . The data points in Fig. 1 are the average values of many measurements for fixed t_w ; the error bars were estimated from the scatter in the experimental results. Both the exponent, n , and the initial TRM, σ_0 , were found to be independent of waiting time, to within experimental accuracy ($n \cong 0.66$). The waiting time dependence of $1/\tau_p$ for $t_w \geq 10$ min can clearly be characterized by the exponential relation: $1/\tau_p = \omega_0 \exp(-t_w/t_0)$. The exponential waiting-time dependence of the apparent response rate has previously been reported for Ag:Mn_{2.6 at. %} + Sb_{0.46 at. %} (Ref. 3) and Eu_{0.54}Sr_{0.46}S.⁹ Inspection of Fig. 1 shows that, for $t_w = 5$ min, the value of $1/\tau_p$ for both samples deviates considerably from the exponential form. This is likely to be caused by a temperature instability, because it takes our system several minutes to come into thermal equilibrium. Moreover, the shorter the waiting time, the more important is the aging process during the observation time.^{7,8,11,12} The best fits of the exponential form to the experimental data for $10 \text{ min} \leq t_w \leq 40 \text{ min}$ yield: $\omega_0 = 5.6 \times 10^{-6} \text{ sec}^{-1}$ for the sample consisting of foils, and $\omega_0 = 1.2 \times 10^{-6} \text{ sec}^{-1}$ for the powder sample. For both samples, $t_0 = 860$ sec. Careful measurements at low reduced temperatures (down to $T_r = 0.2$) show that n does

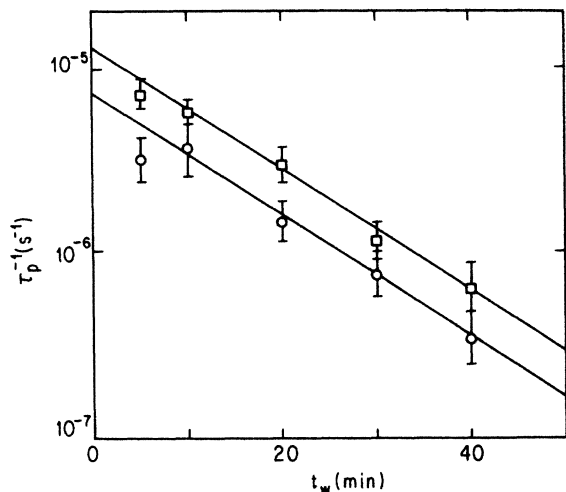


FIG. 1. The waiting-time dependence of the apparent relaxation rate of Ag:Mn_{2.6} at. % at $T=4.35$ K ($T_r=0.43$). The squares represent the data points for the powder sample, the circles represent the data for the sample consisting of foils. The solid lines are the best fits of the exponential form, $1/\tau_p = \omega_0 \exp(-t_w/t_0)$, to the experimental results. For the powder sample $\omega_0 = 1.2 \times 10^{-6}$ sec⁻¹, while the fit for the foil sample yields $\omega_0 = 5.2 \times 10^{-6}$ sec⁻¹. For both samples $t_0 = 860$ sec.

not depend upon the waiting time to within experimental accuracy over the entire temperature range of measurement.

The dependence of the exponent, n , on the reduced temperature $T_r = T/T_g$ for the four spin glasses is exhibited in Fig. 2 for $t_w = 10$ min. Again, the error bars in Fig. 2 were constructed from the scatter in the experimental results at a fixed temperature. Figure 2 shows that the value of n for the four alloys is, to within experimental accuracy, constant ($n \cong 0.65$) for $0.5 \leq T_r \leq 0.8$. Above a certain temperature, T_r^* , which depends on the specific spin glass, n was observed to increase as T_g was approached from below. For Ag:Mn_{2.6} at. % + Sb_{0.46} at. %, T_r^* is about 0.85, for Cu:Mn_{4.0} at. % $T_r^* = 0.91$, while for both

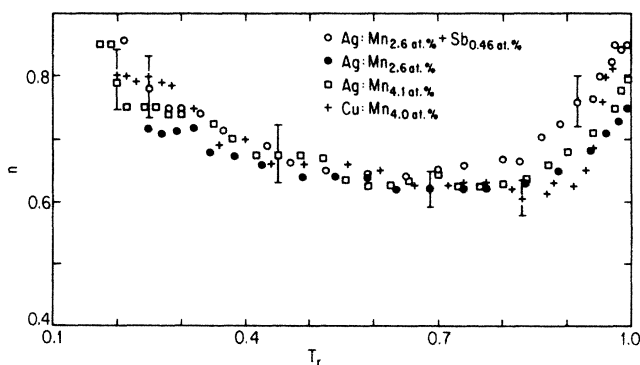


FIG. 2. Temperature dependence of the stretched exponential exponent, n , of the four spin glasses, plotted as a function of the reduced temperature for $t_w = 10$ min.

Ag:Mn_{2.6} at. % and Ag:Mn_{4.1} at. % T_r^* is approximately 0.85. As the temperature is lowered below $T_r = 0.5$ the value of n exhibits a distinct increase for all four spin glasses.

The temperature dependence of the apparent response rate for the four spin glasses, measured at $t_w = 10$ min, is plotted for $\log_{10}(1/\tau_p)$ versus the inverse reduced temperature, $T_r^{-1} = T_g/T$, in Fig. 3. The error bars in Fig. 3, estimated from the scatter in the value of $1/\tau_p$ at a fixed temperature, become larger as the temperature decreases because the relaxation rate decreases while the observation time (500 sec) was kept constant. The drawn line in Fig. 3 represents the best fit to the exponential form: $1/\tau_p = A \exp(-\alpha T_r^{-1})$, with $A = 10^{-3}$ sec⁻¹ and $\alpha = 2.5$. Figure 3 shows that, as the glass transition temperature is approached from below, $1/\tau_p$ begins to deviate from the exponential form at a value of T_r^{**} which depends on the specific spin glass. Inspection of Fig. 3 shows that the

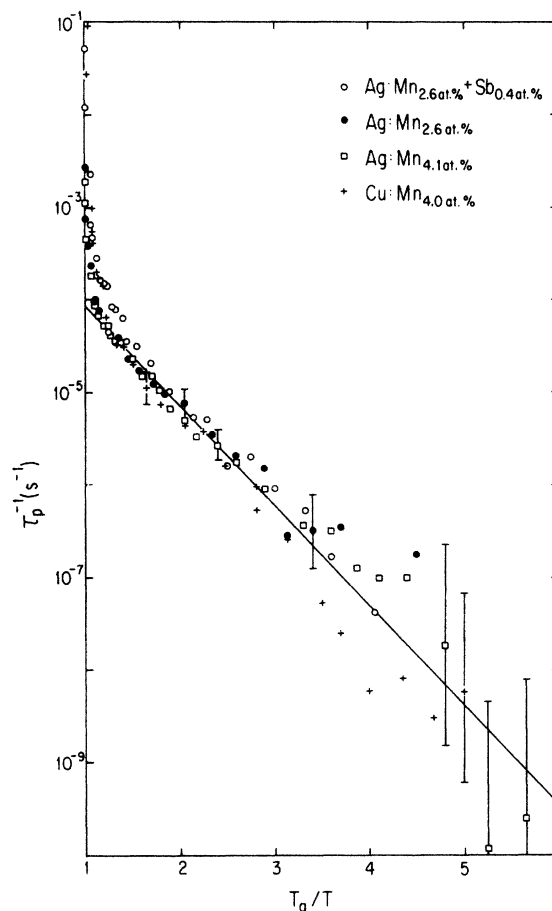


FIG. 3. Temperature dependence of the characteristic relaxation rate, $1/\tau_p$, plotted as $\log_{10}(1/\tau_p)$ vs T_r^{-1} for $t_w = 10$ min. For $T_r^{-1} \geq 1.5$, the values of the apparent relaxation rates of the four spin glasses scale to within experimental accuracy and vary exponentially with the inverse reduced temperature. The solid line represents the fit of the exponential form, $1/\tau_p = A \exp(-\alpha T_r^{-1})$, to the experimental data for $T_r^{-1} \geq 1.5$. The fit yields $A = 10^{-3}$ sec⁻¹ and $\alpha = 2.5$. As T_g is approached from below, the apparent relaxation rate of all four spin glasses increases much more rapidly than exponentially and the scaling with T_r^{-1} breaks down.

temperature at which $1/\tau_p$ starts to increase much more rapidly than exponentially is approximately $T_r^{**} = 0.69$ for the antimony-doped $\text{Ag:Mn}_{2.6 \text{ at. \%}} + \text{Sb}_{0.46 \text{ at. \%}}$ sample, about 0.80 for the copper manganese spin glass, and about 0.87 for both the $\text{Ag:Mn}_{2.6 \text{ at. \%}}$ and the $\text{Ag:Mn}_{4.1 \text{ at. \%}}$ spin glass.

Figure 4 shows the field dependence of the stretched exponential exponent, n , of $\text{Ag:Mn}_{2.6 \text{ at. \%}} + \text{Sb}_{0.46 \text{ at. \%}}$ at four temperatures just below T_g ($T_r \approx 0.91, 0.95, 0.97$, and 0.98). From Fig. 4 it is clear that n increases with increasing field (remember that by field, we mean the magnetic field in which the sample is cooled initially; the final field value is zero in all of the data reported in Fig. 4), the increase being larger the closer T is to T_g from below. At temperatures below about $0.9T_g$, n is independent of H ($H = 4.5\text{--}30$ Oe) to within experimental error, down to reduced temperatures of the order of $0.2T_g$. For yet lower temperatures, our preliminary results exhibit a small decrease of n with increasing field. The entire question of the field dependence of n is complex, and will be treated in a separate paper.²⁵ Finally, Fig. 5 exhibits the field dependence of the apparent response rate, $1/\tau_p$, of $\text{Ag:Mn}_{2.6 \text{ at. \%}} + \text{Sb}_{0.46 \text{ at. \%}}$ at four temperatures (again at $T_r \approx 0.91, 0.95, 0.97$, and 0.98). Figure 5 shows that, just below T_g , the apparent response rate depends strongly on the cooling field, while the field dependence of $1/\tau_p$ gradually decreases as the temperature is lowered.

IV. ANALYSIS

The exponential dependence of the apparent relaxation rate on the inverse reduced temperature appears consistent with thermally activated intervalley transitions between spin-glass energy levels with relaxation times given by an Arrhenius law. In such a model, the specific shape of the time decay of the TRM will depend on the distribution of relaxation times, i.e., on the specific distribution of energy levels and potential barriers separating them. In fact, already in 1980, Préjean and Souletie⁶ showed that the time and temperature dependence of the saturated TRM (STRM) of $\text{Cu:Mn}_{1.2,8 \text{ at. \%}}$ can be characterized by a single unique function of $(T/T_0)\ln(t/\theta_0)$. They found that the time decay of the STRM of the Cu:Mn spin glasses

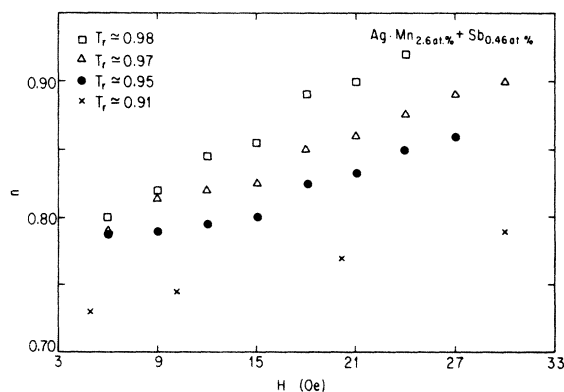


FIG. 4. The field dependence of the stretched exponential exponent, n , of $\text{Ag:Mn}_{2.6 \text{ at. \%}} + \text{Sb}_{0.46 \text{ at. \%}}$ at four temperatures just below T_g .

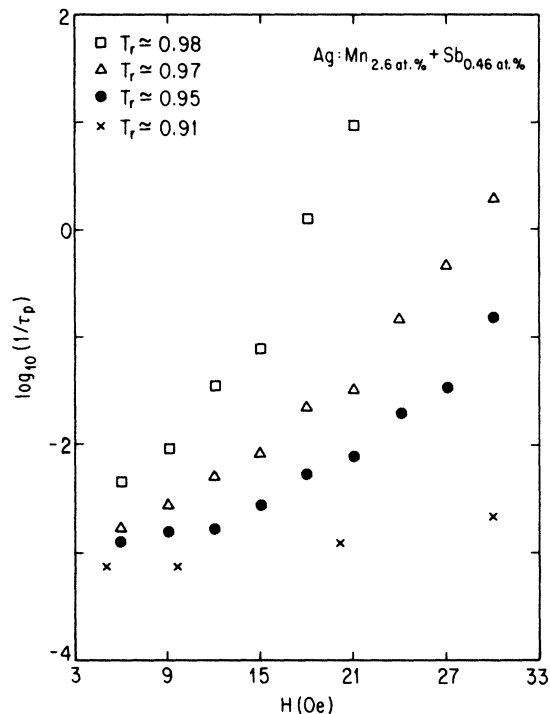


FIG. 5. The field dependence of the apparent relaxation rate, $1/\tau_p$, of $\text{Ag:Mn}_{2.6 \text{ at. \%}} + \text{Sb}_{0.46 \text{ at. \%}}$ at four temperatures just below T_g .

followed a power law: $\sigma_{\text{STRM}}(t) = \sigma_0(t/\theta_0)^{-T/T_0}$, where $\theta_0 \approx 10^{-11}$ sec, and $T_0 \approx 5T_g$, independent of the manganese concentration. They explained this scaling behavior of σ_{STRM} in terms of a distribution of relaxation times arising from a distribution of asymmetrical double-well potentials between which the system can make thermally activated transitions. Our experimental results confirm the scaling of the effective relaxation rate for $T_r^{-1} \geq 1.5$.

However, it should be noted that our experiments are performed in very small magnetic fields, as compared to Ref. 6. In particular, we have found that the prefactor σ_0 in the stretched exponential expression for the time decay of σ_{TRM} increases roughly linearly with increasing magnetic field at temperatures below $T/T_g \sim 0.9$. In this temperature regime, the different magnetic fields we employ cause very little change in the apparent response rate, $1/\tau_p$. At temperatures closer to T_g , the variation of σ_0 with magnetic field is steeper, approaching a quadratic form very near to T_g ($T/T_g \sim 0.97$). Thus, the remanent magnetization is not saturated in our experiments anywhere in the H - T plane of measurement. It is possible, therefore, that our experiments measure different quantities than are reported in Ref. 6, in so far as they report the time dependence of the saturated remanent magnetization (SRM). For example, as the glass transition temperature is approached from below, our results show that the scaling property of the apparent relaxation rate only holds for the $\text{Ag:Mn}_{2.6 \text{ at. \%}}$ and $\text{Ag:Mn}_{4.1 \text{ at. \%}}$ spin-glass samples. Scaling is not exhibited for the Cu:Mn and Sb-doped

Ag:Mn samples. The amount of magnetic anisotropy is known to depend strongly on the type of nonmagnetic host lattice (e.g., Ag,Cu,Au), and on doping with nonmagnetic impurities with a relatively large spin-orbit scattering cross section (e.g., Sb). This suggests that the spin-glass dynamics and the scaling property just below T_g may be closely connected with the amount of magnetic anisotropy. In any case, the lack of saturation of σ_0 in our measurements makes a direct comparison with Ref. 6 somewhat problematic. If we do adopt their model, both the exponential temperature dependence and the scaling of the apparent relaxation rate can be explained over part of our measurement range ($T_r^{-1} \geq 1.5$). It is not clear to us, however, how such a model could account for the nonexponential temperature dependence, and the rapid increase of $1/\tau_p$, just below T_g .

Recently, De Dominicis *et al.*^{20,26} reported the first results of their calculations of the *dynamics* of the infinite-range Ising spin-glass model using a "simple model." Their results appear to be consistent with our experimental results over the full temperature range. De Dominicis *et al.* calculated the approach to equilibrium of the inverse of the Parisi order-parameter function, $x(q)$, after a change in temperature or a change in applied magnetic field. Using the results of Mézard *et al.*¹⁷ and of Derrida *et al.*¹⁸ (the nonextensive fluctuations of the free-energy valley minima of the spin-glass state are independent random variables with an exponential distribution), they found that the sum of the perturbed spin-glass occupancies, related to the length of the right plateau of the $q(x)$ plot,^{15,17,18,20} relaxes towards equilibrium according to a stretched exponential form. They found for the long-time relaxation of $x(t)$ after a change δh of the applied field:²⁷

$$- [x(t)]_{\text{pure}} + [x(t)]_{\delta h} = (1-x') \exp[-v_0 \Gamma(1-x)(2t/\tau_0)^x], \quad (1)$$

where $[x(t)]_{\text{pure}}$ is the value of $x(t)$ of the system prepared in a pure thermodynamic state. In Eq. (1), $x = 1 - y(q_{EA})$, i.e., the value of x for the Edwards-Anderson order-parameter value (x' is the field-shifted value), $v_0 = M \exp[-f_c x/kT]$, where f_c is an upper cut-off of the free-energy fluctuations (M , the number of states, and f_c are sent to infinity such that v_0 remains finite), and $\Gamma(1-x)$ is the factorial function. Although Eq. (1) was derived within the framework of the infinite-range Ising model, and the relation between $x(t)$ and σ_{TRM} of a real spin glass is not obvious, it will be shown below that our experimental results for the time dependence of σ_{TRM} over the entire temperature range of measurement are well described by the right-hand side of Eq. (1). Assuming that $\sigma_{TRM}(t) \propto -[x(t)]_{\text{pure}} + [x(t)]_{\delta h}$, comparison of the stretched exponential form of σ_{TRM} with Eq. (1) yields for the apparent relaxation rate, $1/\tau_p$, of the TRM:

$$1/\tau_p = 2M^{1/(1-\bar{y})} \Gamma(\bar{y})^{1/(1-\bar{y})} \exp(-f_c/kT)/\tau_0, \quad (2)$$

with the identification $\bar{y} = 1 - x = n$ in the simplest model.²⁰ In order to test the validity of the conjecture that $1/\tau_p$ is given by Eq. (2), (weighted) multiple linear regression fits of the theoretical expression for $\log_{10}(1/\tau_p)$ were made to our experimental data. The fitting param-

eters were M , τ_0 , and f_c . As the experimental results for Ag:Mn_{2.6} at. % and Ag:Mn_{4.1} at. % scale over the entire temperature range of measurement, the theoretical expression for $\log_{10}(1/\tau_p)$ was fitted to the total of our experimental results for these two spin glasses. The fits to the data for the antimony doped and the copper spin glasses were made separately. For each set of data two fits were made: one using the experimental values of n , as shown in Fig. 2, and one using the theoretical values of \bar{y} . In the latter case the temperature dependence of \bar{y} was calculated in two steps. First, the temperature dependence of the Edwards-Anderson order parameter, q_{EA} , was calculated from the self-consistency equation for q_{EA} derived by Sherrington and Kirkpatrick:^{28,29}

$$q_{EA}(T) = 2 \int_0^\infty dz (2\pi)^{-1/2} \exp(-\frac{1}{2}z^2) \times \tanh^2[q_{EA}^{1/2}(T)zT_g/T]. \quad (3)$$

Equation (3) was solved numerically for several reduced temperatures in the range $0 \leq T_r \leq 1$, and a sixth-order polynomial was fitted to the calculated values of $q_{EA}(T_r)$. In fitting the polynomial, the limiting behavior of $q_{EA}(T_r)$, as predicted by the Sherrington-Kirkpatrick model,^{28,29} was used:

$$q_{EQ}(T_r) = 1 - (2/\pi)^{-1/2} T_r \text{ for } T_r \rightarrow 0, \quad (4)$$

$$q_{EA}(T_r) = 1 - T_r \text{ for } T_r \rightarrow 1. \quad (5)$$

The calculated temperature dependence of q_{EA} is shown in Fig. 6. Secondly, the polynomial expression for the temperature dependence of q_{EA} was inserted into the differential equation which relates q , \bar{y} , and T_r , derived by Vannimenus *et al.*³⁰ on the basis of a scaling assumption for $q(x, T)$:

$$\frac{1-q}{T_r} + \bar{y} \frac{dq}{dT_r} = 0, \quad (6)$$

(though they find $\bar{y} = \frac{1}{2}$ at $T=0$, not $\bar{y}=1$ as we find). The theoretical temperature dependence of \bar{y} on T_r is exhibited in Fig. 7. Comparison of Figs. 2 and 7 shows indeed that the stretched exponential exponent, n , and the theoretically calculated values of \bar{y} have the same qualitative temperature dependence.

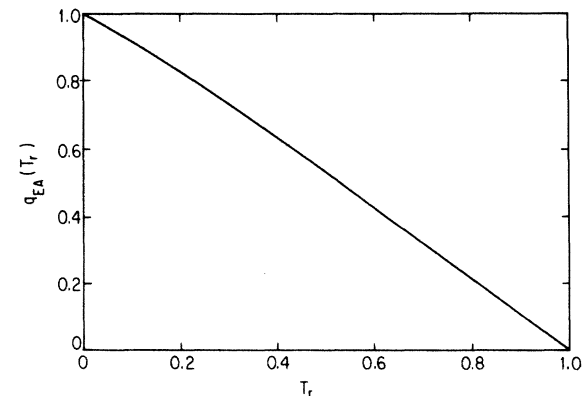


FIG. 6. The temperature dependence of the Edwards-Anderson order parameter, q_{EA} , calculated for the Sherrington-Kirkpatrick model.

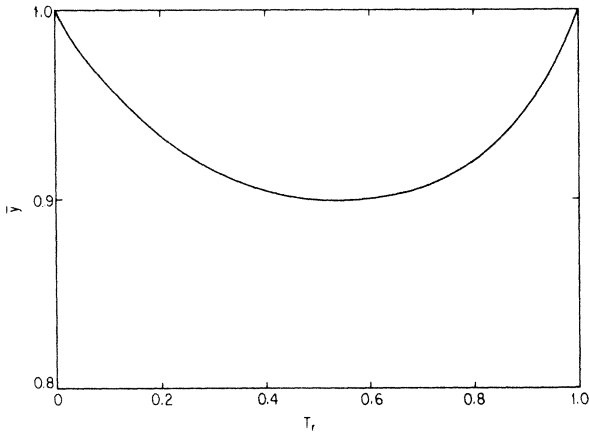


FIG. 7. The temperature variation of the stretched exponential exponent, \bar{y} , with the reduced temperature, calculated for the Sherrington-Kirkpatrick model (see text). The overall variation of \bar{y} is qualitatively the same as the variation of the experimental values of n , plotted in Fig. 2.

Because the error in the experimental data increases as the temperature decreases, the experimental data in the temperature region where $1/\tau_p$ varies exponentially with T_r^{-1} were weighted in all fits with the reduced temperature. The data in the nonexponential region, just below T_g , were all given a weight of 0.5 in order to reduce the effect of an error in the reduced temperature.

In Fig. 8, the fit of the theoretical expression for $\log_{10}(1/\tau_p)$, using the experimental values of n , for Ag:Mn_{2.6} at. % and Ag:Mn_{4.1} at. % is shown. The quality of the fit is reasonably good over the whole temperature range, and yields $\tau_0 = 10^5$ sec, $M = 5.5$, and $f_c = 3.2k_B T_g$. Clearly, the small deviation from the exponential temperature dependence at low temperatures ($T_r^{-1} > 4$) is caused by the variation of n with T_r . The fit for the calculated values of \bar{y} for the same two spin glasses is shown in Fig. 9, and gives $\tau_0 = 3.8 \times 10^3$ sec, $M = 1.02$, and $f_c = 2.5k_B T_g$. Again, the quality of the fit is quite good over the whole temperature range and, moreover, the value of f_c is equal to the value of α , which was obtained from a fit of the exponential expression for $1/\tau_p$.

Figure 10 shows the fit to the experimental data for Ag:Mn_{2.6} at. % + Sb_{0.46} at. %, for the experimental values of n . The fit yields $\tau_0 = 2.2 \times 10^3$ sec, $M = 3.9$, and $f_c = 3.3k_B T_g$. The fit for the theoretical values of \bar{y} is plotted in Fig. 11; $\tau_0 = 4.6 \times 10^2$ sec, $M = 1.01$, and $f_c = 3.3k_B T_g$.

Finally, Figs. 12 and 13 exhibit the fits of the theoretical expression for $\log_{10}(1/\tau_p)$ to the experimental results for Cu:Mn_{4.0} at. %, for the experimental and the calculated values of n and \bar{y} , respectively. The fit for the experimental values of n (Fig. 10) yields $\tau_0 = 4.2 \times 10^4$ sec, $M = 10$, and $f_c = 4.7k_B T_g$, while the fit for the calculated \bar{y} values yields: $\tau_0 = 4.5 \times 10^3$ sec, $M = 1.16$, and $f_c = 3.0k_B T_g$. The strong curvature of the fit shown in Fig. 12 is due to the rather strong variation of the stretched exponential exponent with the reduced temperature (Fig. 2). However, the fit for the calculated values of \bar{y} is surprisingly good

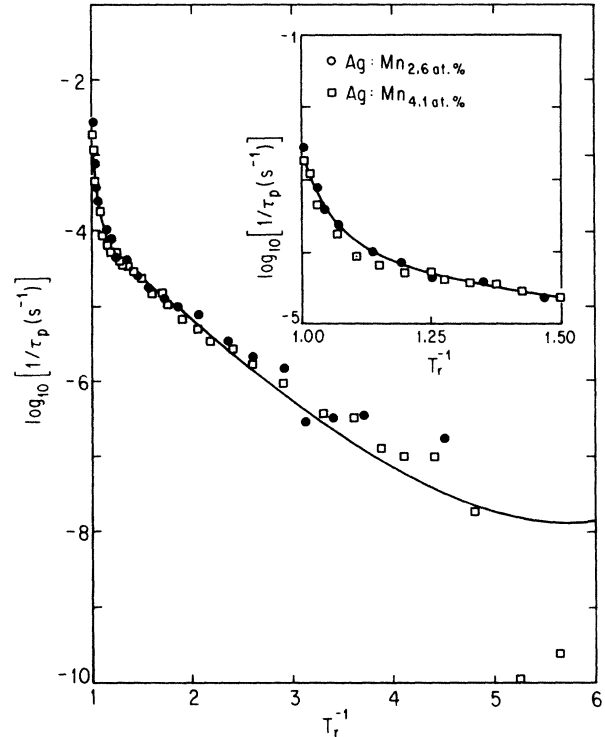


FIG. 8. Fit of the theoretical expression for the apparent relaxation rate to the experimental data for Ag:Mn_{2.6} at. % (solid circles) and Ag:Mn_{4.1} at. % (squares). The weighted multiple linear regression fit was done using the experimental values of n and yields: $\tau_0 = 10^5$ sec, $M = 5.5$, and $f_c = 3.2k_B T_g$. The insert shows the fit just below T_g in detail.

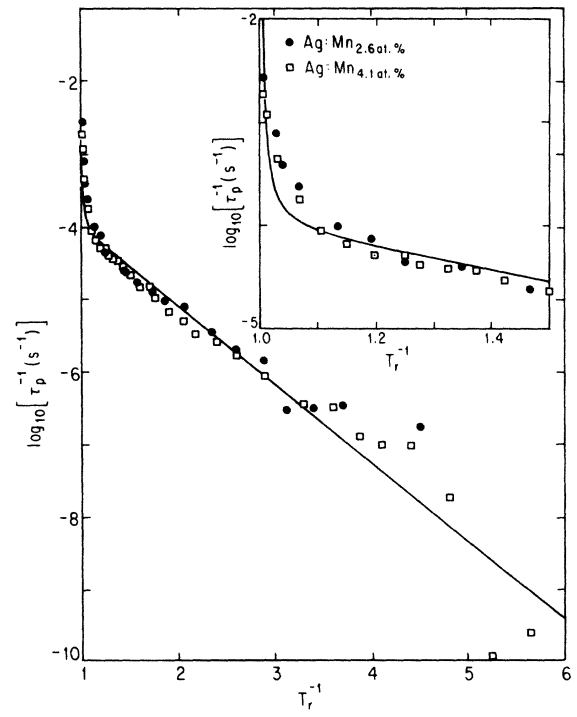


FIG. 9. The weighted multiple linear regression fit of Eq. (2), using the calculated temperature dependence of \bar{y} (Fig. 5), to the experimental for Ag:Mn_{2.6} at. % (solid circles) and Ag:Mn_{4.1} at. % (squares). The fit yields: $\tau_0 = 3.8 \times 10^3$ sec, $M = 1.02$, and $f_c = 2.5k_B T_g$. The insert shows the fit just below T_g in detail.

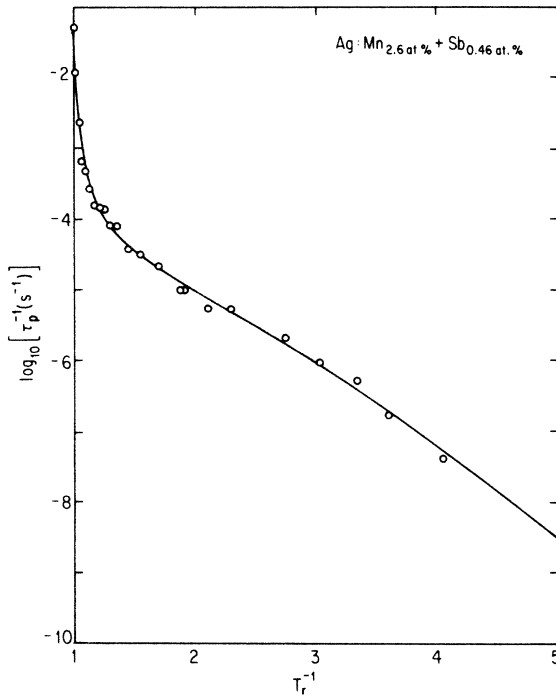


FIG. 10. The temperature dependence of $1/\tau_p$ of Ag:Mn_{2.6} at. % + Sb_{0.46} at. %. The solid line represents the fit of Eq. (2) for the experimental values of n , to the experimental data and corresponds to $\tau_0 = 2.2 \times 10^3$ sec, $M = 3.9$, and $f_c = 3.3k_B T_g$.

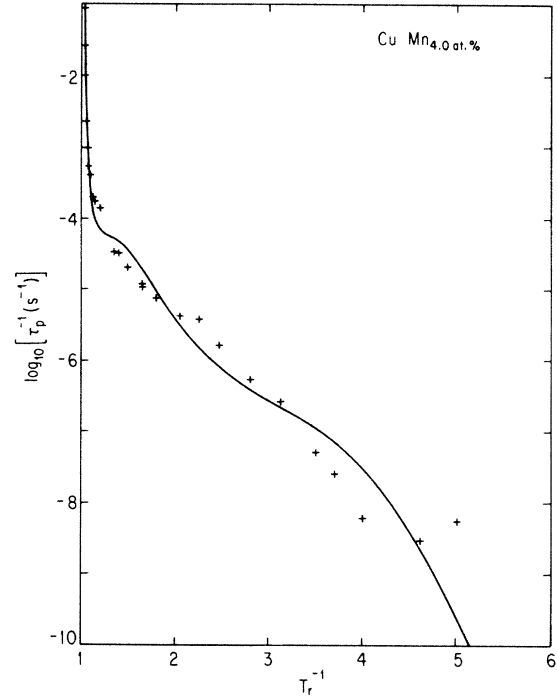


FIG. 12. Fit of the theoretical expression for $1/\tau_p$ [Eq. (2)] to the experimental data for Cu:Mn_{4.0} at. %. The fit was done for the experimental values of n , and gives $\tau_0 = 4.2 \times 10^4$ sec, $M = 10$, and $f_c = 4.7k_B T_g$.

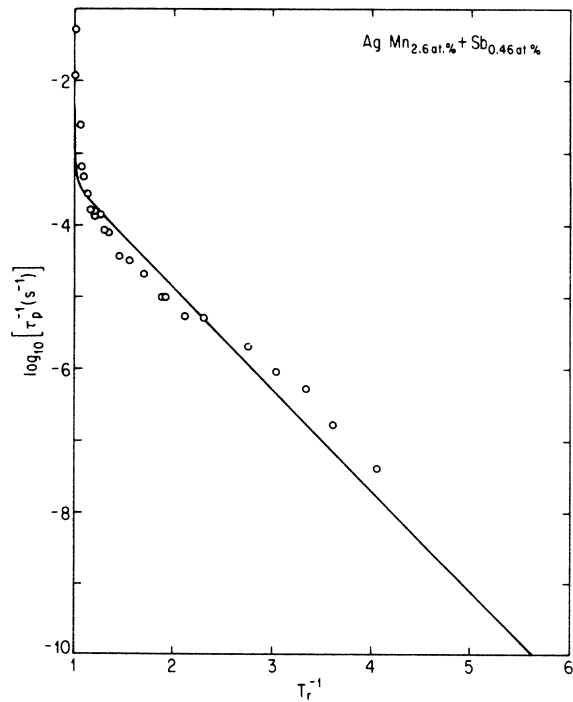


FIG. 11. The fit of Eq. (2) to the experimental data for Ag:Mn_{2.6} at. % + Sb_{0.46} at. % for the theoretical values of \bar{y} . The best fit gives $\tau_0 = 4.6 \times 10^2$ sec, $M = 1.01$, and $f_c = 3.3k_B T_g$.

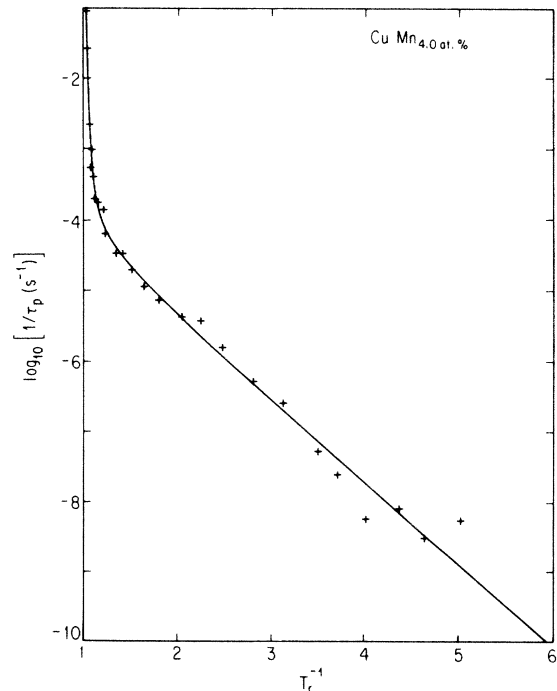


FIG. 13. Fit of Eq. (2) to the experimental data for Cu:Mn_{4.0} at. % using the theoretical temperature dependence of \bar{y} . The fit yields: $\tau_0 = 4.5 \times 10^3$ sec, $M = 1.16$, and $f_c = 3.0k_B T_g$.

over the whole temperature range.

It is clear that the theoretical expression for the effective relaxation rate of $x(t)$, which has been derived recently by De Dominicis *et al.*,²⁰ is in excellent agreement with our experimental results. Both the fits for the experimental values of n and the calculated values of \bar{y} yield, for all three data sets, values for τ_0 and f_c close to the values of $2/A$ and α , which were obtained from the fit of the exponential expression, $1/\tau_p = A \exp(-\alpha T_r^{-1})$, exhibited in Fig. 3 [notice the factor of 2 in the exponent of Eq. (1)]. The exponential temperature of the theoretical relaxation rate in Eq. (1) is a direct consequence of the fact that the density of states, ν_0 , is constant for any finite free energy and varies exponentially with the inverse temperature.^{17,20} Our experimental results suggest, therefore, that the apparent relaxation rate of σ_{TRM} reflects the temperature dependence of the density of states in the spin-glass state.

Inspection of Eq. (2) shows that the increasing value of \bar{y} , as $T_r \rightarrow 1$, gives rise to the rapid, nonexponential increase of $1/\tau_p$ just below T_g . As $T_r \rightarrow 1$, the factor $M^{1/(1-\bar{y})}$ in the right-hand side of Eq. (2) diverges for $M > 1$. It is therefore not surprising that our fits, particularly those for the calculated \bar{y} values, yield values for M close to unity. The nonexponential temperature dependence of the apparent relaxation rate clearly originates in the strong temperature dependence of the length of the right plateau in the $q(x)$ plot just below T_g . This sug-

gests that the strong field dependence of the apparent response rate just below T_g (Fig. 5) originates in the field dependence of n (Fig. 4). We have therefore plotted in Fig. 14 $\log_{10}(1/\tau_p)$ versus $1/(1-n)$ for several temperatures ($T_r \approx 0.91, 0.95, 0.97$, and 0.98) and fields (4.5–30 Oe). Figure 14 shows that $\log_{10}(1/\tau_p)$ varies indeed linearly with $1/(1-n)$. Moreover, the experimental data for $T_r \approx 0.91, 0.95$, and 0.97 fall, to within experimental accuracy, on a single line. The drawn line is the best fit of the form, $\log_{10}(1/\tau_p) = -c + [1/(1-n)] \log_{10} M$, to the data for the lowest three reduced temperatures. The broken line represents the fit to the data for $T_r \approx 0.98$, with $c = -5.5$ and $M = 4.4$, for the other temperatures $c = -5.5$ and $M = 3.8$. The latter value is in excellent agreement with the value obtained from the fit of Eq. (2) to the experimental data for the temperature dependence of $1/\tau_p$ at fixed field (Fig. 10). The increment of the slope of the data with temperature suggests that M increases as T_g is approached from below. The field and temperature dependence of $1/\tau_p$ clearly argues for the validity of Eq. (2). However, the unphysically small value of M is troubling. It may be associated with the differences between the theoretical infinite-range model (equivalent to a fully concentrated sample) and the small finite concentration of the samples actually measured in our experiments. Apart from this numerical distinction, the good qualitative agreement of Eq. (2) with our experimental results raises the hope that the infinite-range Ising model can account for the dynamics of metallic spin glasses, and that recent^{26,31,32} and future theoretical developments will improve substantially our understanding of these systems.

V. SUMMARY AND CONCLUSIONS

Our main experimental and conclusions are the following.

(1) The time decay of the field-cooled thermoremanent magnetization of the spin glasses (Ag:Mn_{2.6} at. %, Ag:Mn_{4.1} at. %, Ag:Mn_{2.6} at. % + Sb_{0.46} at. %, and Cu:Mn_{4.0} at. %) is accurately characterized by the stretched exponential form in the time range of measurement, 5–500 sec.

(2) The apparent relaxation rate of the time decay of the thermoremanent magnetization of the four spin glasses depends approximately exponentially on the waiting time, for waiting times in the range 10–40 min.

(3) The apparent relaxation rate depends exponentially on the inverse reduced temperature and scales with the glass transition temperature for inverse reduced temperatures larger than about 1.5.

(4) As the glass transition temperature is approached from below, the apparent relaxation rate increases much more rapidly than exponentially with inverse reduced temperature, and the scaling with T_g breaks down. The temperature at which the scaling breaks down is related to the difference in magnetic anisotropy of the spin glasses.

(5) Recently derived expressions for the relaxation of the perturbed sum over the square of the spin-glass-state occupancies is found to map onto our measurements of the time dependence of $\sigma_{\text{TRM}}(t)$: the stretched exponential form for the recovery of the perturbed spin-glass-state oc-

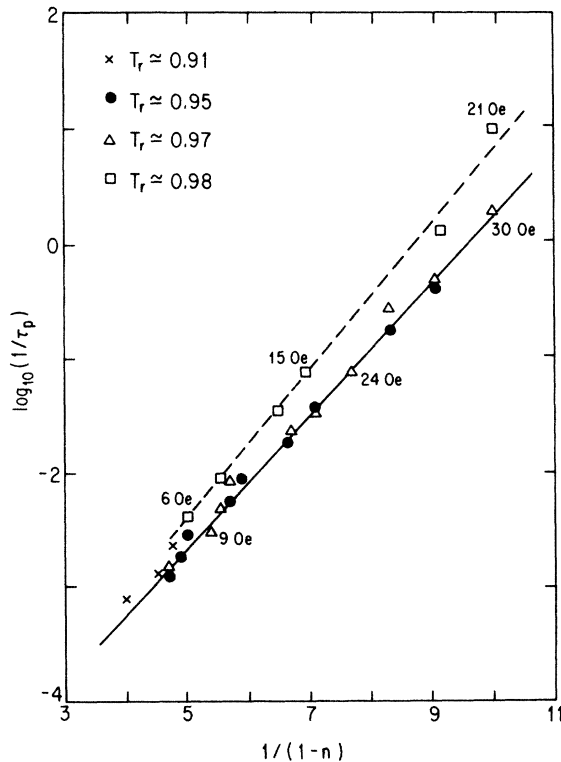


FIG. 14. The dependence of $\log(1/\tau_p)$ on $1/(1-n)$ at several reduced temperatures and magnetic fields (4.5–30 Oe). The solid line represents the best fit of the form $\log_{10}(1/\tau_p) = -c + [1/(1-n)] \log_{10} M$, to the experimental data for $T_r \approx 0.91, 0.95$, and 0.97 , with $c = -5.5$ and $M = 3.8$. The broken line represents the fit to the experimental data for $T_r \approx 0.98$, yielding $c = -5.6$ and $M = 4.4$.

cupancies is consistent with the stretched exponential time decay of the thermoremanent magnetization; the apparent relaxation rate occurring in the theoretical expression for the recovery of the spin-glass-state occupancies is in excellent agreement with our experiment value for the apparent relaxation rate.

However, the fits of the theoretical expression for the effective relaxation rate to our experimental data yield apparently unphysical values for the number of states that are available to the system. Nevertheless, the qualitative agreement of the theoretical results with our experimental data seem to indicate that the infinite-range Ising model

contains features which can explain the dynamics of metallic spin glasses.

ACKNOWLEDGMENTS

We have benefited considerably from many stimulating discussions with Dr. N. Bontemps, Dr. B. Derrida, Dr. C. De Dominicis, Dr. D. Fiorani, Dr. M. Mézard, Dr. C. Pappa, Dr. N. Surlas, and Dr. G. Toulouse. This work was supported by the National Science Foundation Grants Nos. DMR 81-21394 and No. INT 83-12985.

- ¹R. V. Chamberlin, G. Mozurkevich, and R. Orbach, *Phys. Rev. Lett.* **52**, 867 (1984).
- ²R. V. Chamberlin, M. Hardiman, L. A. Turkevich, and R. Orbach, *Phys. Rev. B* **25**, 6720 (1982).
- ³R. V. Chamberlin, *Phys. Rev. B* **30**, 5393 (1984).
- ⁴R. Hoogerbeets, Wei-Li Luo, and R. Orbach, *Phys. Rev. Lett.* **55**, 111 (1985).
- ⁵R. Hoogerbeets, Wei-Li Luo, R. Orbach, and D. Fiorani, *Phys. Rev. B* **33**, 6531 (1986).
- ⁶J. J. Préjean and J. Souletie, *J. Phys. (Paris)* **41**, 1335 (1980).
- ⁷P. Nordblad, P. Svedlindh, L. Lundgren, and L. Sandlund, *ICM 1984, J. Appl. Phys.* **57**, 3371 (1985).
- ⁸P. Nordblad, L. Lundgren, and L. Sandlund, *ICM 1985, J. Magn. Magn. Mater.* **54-57**, 185 (1986).
- ⁹R. Hoogerbeets, Wei-Li Luo, R. Orbach, and H. Maletta, *ICM 1985, J. Magn. Magn. Mater.* **54-57**, 177 (1986).
- ¹⁰J. Ferré, M. Ayadi, R. V. Chamberlin, R. Orbach, and N. Bontemps, *ICM 1985, J. Magn. Magn. Mater.* **54-57**, 211 (1986).
- ¹¹M. Ocio, M. Alba, and J. Hamman, *ICM 1985, J. Magn. Magn. Mater.* (to be published).
- ¹²M. Alba and J. Hamman, *ICM 1985, J. Magn. Magn. Mater.* (to be published).
- ¹³L. Lundgren, P. Svedlindh, P. Nordblad, and O. Beckman, *Phys. Rev. Lett.* **51**, 911 (1983).
- ¹⁴L. Lundgren, P. Svedlindh, and O. Beckman, *Phys. Rev. B* **26**, 3990 (1982).
- ¹⁵G. Parisi, *Phys. Rev. Lett.* **50**, 1946 (1983).
- ¹⁶G. Parisi, *Phys. Rev. Lett.* **43**, 1754 (1979); *J. Phys. A* **13**, L115 (1980); **13**, 1101 (1980); **13**, 1887 (1980).
- ¹⁷M. Mézard, G. Parisi, N. Surlas, G. Toulouse, and M. A. Virasoro, *Phys. Rev. Lett.* **52**, 1156 (1984); *J. Phys. (Paris)* **45**, 843 (1984); *J. Phys. (Paris) Lett.* **46**, 217 (1985).
- ¹⁸B. Derrida and G. Toulouse, *J. Phys. (Paris) Lett.* **46**, 223 (1985).
- ¹⁹B. Derrida, *Phys. Rev. Lett.* **45**, 79 (1980); *Phys. Rev. B* **24**, 2618 (1981).
- ²⁰C. De Dominicis, H. Orland, and F. Lainée, *J. Phys. (Paris) Lett.* **46**, L463 (1985).
- ²¹J. J. Préjean, M. Joliclerc, and P. Monod, *J. Phys. (Paris)* **41**, 427 (1980).
- ²²A. Fert and P. M. Levy, *Phys. Rev. Lett.* **44**, 1438 (1980).
- ²³P. M. Levy and A. Fert, *Phys. Rev. B* **23**, 4667 (1981).
- ²⁴M. Levy, C. Morgan-Pond, and A. Fert, *J. Appl. Phys.* **53**, 2168 (1982).
- ²⁵N. Bontemps, R. Hoogerbeets, Wei-Li Luo, R. Orbach, and C. Pappa (unpublished).
- ²⁶C. De Dominicis (unpublished); (private communication).
- ²⁷Equation (1) was derived by De Dominicis *et al.* (Ref. 20) for the difference of $x(t)$ for an Ising spin-glass system initially prepared in a single (pure) free energy state, and in a shifted magnetic field (hence, the subscript δh). Another expression, relevant to the difference between $[x(t)]_{\text{pure}}$ and x (the equilibrium value) in Ref. 20, contains a prefactor to the exponential, proportional to t^{-x} . We have attempted to fit our data using such a prefactor, but without success over nearly the full temperature range available to us. The effect of this prefactor can be ascertained by inspecting the time decay of the thermoremanent magnetization (TRM), as plotted against $\log_{10}(t)$ in Fig. 1 of Ref. 4. There, the change in slope is seen to be negative. The second derivative of the De Dominicis expression for $[x(t)]_{\text{pure}} - x$ with respect to $\log_{10}(t)$ is positive definite. The experimentally measured change in slope will be positive only when the measurement times exceed the apparent response time τ_p . But this occurs only in a very narrow temperature range near T_g , and corresponds to only the first two data points of Fig. 3, as an example. We therefore prefer a fit to Eq. (1), with no time prefactor. Of course, one must note that De Dominicis *et al.* (Ref. 20) did not calculate the time dependence of the TRM, but rather that of the Parisi parameter $x(t)$. One must wait for the former before a full comparison between theory and experiment is possible.
- ²⁸D. Sherrington and S. Kirkpatrick, *Phys. Rev. Lett.* **35**, 1792 (1975).
- ²⁹S. Kirkpatrick and D. Sherrington, *Phys. Rev. B* **17**, 4385 (1978).
- ³⁰J. Vannimenus, G. Toulouse, and G. Parisi, *J. Phys. (Paris)* **42**, 565 (1981).
- ³¹C. De Dominicis and H. J. Hilhorst (unpublished).
- ³²G. Paladin, M. Mézard, and C. De Dominicis (unpublished).