Experimental indications for a critical relaxation time in spin-glasses

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Long-time (5–10^4 sec) superconducting-quantum-interference-device magnetometry measurements on a Au–8 at.% Fe spin-glass imply the existence of a well-defined characteristic time, beyond which no additional change of magnitude or position of the susceptibility cusp occurs. This characteristic time is 5×10^2 sec at an applied field of 0.1 mT (1 G) and estimated to be 10^5 sec at zero field. It is inferred that this characteristic time is coupled to a critical value of the cluster relaxation times where intercluster coupling becomes strong enough to initiate cluster recombinations.

Experimental observations on spin-glass materials are generally interpreted in terms of phenomenological models prescribing a wide spectral distribution of relaxation times. These observations range in time scale from magnetization measurements[1] to neutron scattering experiments.[2] Recently, within this phenomenological model, we[3] derived the complex susceptibility, $\chi=\chi'+i\chi''$, without the need of specifying any expression for the relaxation times. In particular, it was found that χ'' was proportional to the density of relaxation times around $1/\omega$, where ω is the applied angular frequency. χ'' was also found to be simply related to the frequency dependence of χ' through $\chi''=(\pi/2)\partial\chi'/\partial\ln\omega$. This relation was obeyed in the *amorphous* metallic spin-glass $(Fe_{0.06}Ni_{0.94})_{75}P_{16}B_6Al_3$ and later verified in *crystalline* Au(Fe) (Ref. 4) and other architypical spin-glasses,[5] experimental results which strongly support such a descriptive model. In view of the "unphysical" results generally obtained when the relaxation times are assumed to have the Arrhenius form, Tholence[6] analyzed ac susceptibility data of several spin-glass systems according to the Vogel-Fulcher law

$$1/\tau=f_0\exp[-E/k(T-T_0)]\ , \tag{1}$$

where $f_0=10^9$–10^{13} sec^{-1} is a characteristic frequency and E the height of the cluster energy barriers. T_0 is proposed by Shtrikman and Wohlfarth[7] to arise from intercluster coupling. The general idea in the present phenomenological model is that the relaxation-time spectrum exhibits a reasonably well-defined maximum value τ_{max}. As the temperature is lowered the spectrum continuously evolves towards longer relaxation times. A cusp occurs in χ' at the spin-glass freezing temperature (T_g), when τ_{max} equals the *time of observation* t ($1/\omega$ in ac susceptibility) due to "blocking" of clusters[1] with relaxation times larger than t. Thus Eq. (1) may approximately be rewritten

$$(\ln f_0+\ln t)^{-1}=(T_g-T_0)k/E\ . \tag{2}$$

ac susceptibility data analyzed according to Eq. (2) give physically acceptable values of E/k but the extrapolated value of T_0 is commonly found to be at a significantly lower temperature than the position of the cusp of the field-cooled dc curve.[4,6,8] This implies a breakdown of the Vogel-Fulcher law at long observation times. Such an anomalous behavior is indicated by the very-low-frequency measurements by Guyot *et al.*[9] on $PrP_{0.85}$ and the results of Ferré *et al.*[10] on $Eu_{0.4}Sr_{0.6}S$. Malozemoff and Imry[8] (later called MI) made long-time field-cooled measurements on Cu–4.6 at.% Mn and Al–37 at.% Gd but found no change of the position of the susceptibility cusp with cooling rates. Their results do not fit with an extrapolation of Tholence's earlier ac susceptibility data according to Eq. (2).

Here we report high-resolution long-time magnetization measurements on a Au–8 at.% Fe spin-glass. We investigate the *approach* of the susceptibility cusp towards equilibrium position at infinite time. We discuss the physical origin of the cusp of the field-cooled curve and indicate possible reasons for the conflicting results found from different time-scale observations.

The measurements were performed in a superconducting-quantum-interference-device (SQUID) magnetometer[11,12] on a rod-shaped sample (2.4 mm diam and 5 mm length). The initial measurements were made by cooling the sample in a static field (3 μT– 0.1 mT). Thereby, the sample temperature was decreased in steps of 50–100 mK and the time dependence of the magnetization at thermal equilibrium was recorded. For a 100-mK step decrease in temperature the sample attains the equilibrium temperature within 1 mK in an *overdamped* way in less than 20 sec. The long-time temperature stability is approximately 0.1 mK.[12] A detectable (>10 ppm at 0.1 mT for a time period of 20–10^3 sec) time dependence of the magnetization at thermal equilibrium occurred from about 0.5 K above T_g. The magnetization was then found to *increase* with time. Below T_g the magnetization *decreases* with time at

thermal equilibrium. These time effects are shown in Fig. 1 at some temperatures around T_g. Below T_g the time dependence reaches a broad maximum roughly 1 K below T_g. Here a step decrease in temperature of 0.1 K gives a decay in magnetization of 0.4% (at 0.1 mT) in the time interval $20-5\times10^3$ sec with a logarithmic time dependence in the interval $10^3-5\times10^3$ sec. However, in view of the small-time dependence in the vicinity of T_g we believe that equilibrium ($t=\infty$) values can be determined, thus implying an accurate localization of the cusp. Two equilibrium curves at 0.03 and 0.1 mT (marked DC) are shown in Fig. 2. Since the magnitude of the susceptibility cusp is linearly suppressed with field (3 μT–0.1 mT) we find the cusp of the zero-field curve to have a 0.4% higher value than the 0.03-mT curve in Fig. 2 with the cusp at $T_g(\mathrm{DC})=28.13$ K.

Since we have observed virtually identical time effects in the field-cooled curve of Cu–4 at.% Mn, we believe that the characteristic features described above are quite typical for many spin-glass systems. Clearly, experimental field-cooled curves cannot be characterized as "equilibrium" or "reversible" curves. The undetected time effects of the field-cooled curves in the experiments by MI[8] are likely due to the following crucial points: (i) In their SQUID magnetometer an intended step decrease in temperature of 0.2–0.4 K starts out with an undershoot of 1.5 K in temperature followed by an overdamped approach from below towards the equilibrium temperature, which is reached after 6 min. According to our experiments, temperature oscillations around the equilibrium temperature reduces the time effects, at least below T_g. (ii) In contrast to the belief by MI, external fields smear out the time effects. (iii) Since the time effects are small at times above 6 min, the shortest observation time by MI, the effects are not easily detected. MI compared the position of the cusp of the field-cooled curves at various cooling rates to the frequency dependence of the cusp in ac susceptibility by Tholence. We find such a comparison quite ambiguous. A more adequate comparison is obtained when the sample is cooled in zero field, the external field then applied, and the change of magnetization with time observed (at constant temperature). Hence, in the phenomenological model prescribing a wide spectrum of relaxation times, we assign to each relaxation time τ_i a magnetic moment, which at constant applied field attains the value $m_0(\tau_i)$ at $t=\infty$. The approach to equilibrium is

$$m=m_0(\tau_i)[1-\exp(-t/\tau_i)]\ . \tag{3}$$

Assuming that the density of relaxation times $g(\tau)$ varies slowly in $\ln\tau$ the total moment is, with $g(\tau)$ normalized,

$$m=\int_{\tau_{\min}}^{\tau_{\max}} m_0(\tau)(1-e^{-t/\tau})g(\tau)\,d\ln\tau\ . \tag{4}$$

Taking the partial derivative of m with respect to $\ln t$ we obtain

$$\frac{\partial m}{\partial \ln t}=\int_{\tau_{\min}}^{\tau_{\max}} m_0(\tau)g(\tau)\frac{t}{\tau}e^{-t/\tau}d\ln\tau\ . \tag{5}$$

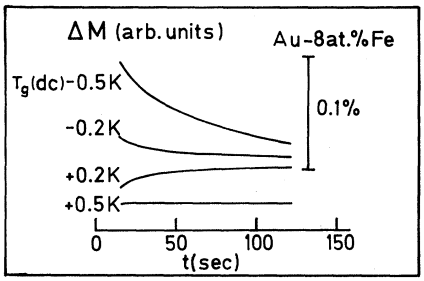

FIG. 1. Relative change of magnetization with time at thermal equilibrium after a step decrease in temperature of 80 mK. Constant applied field equal to 0.1 mT. $T_g(\mathrm{DC})=28.38$ K.

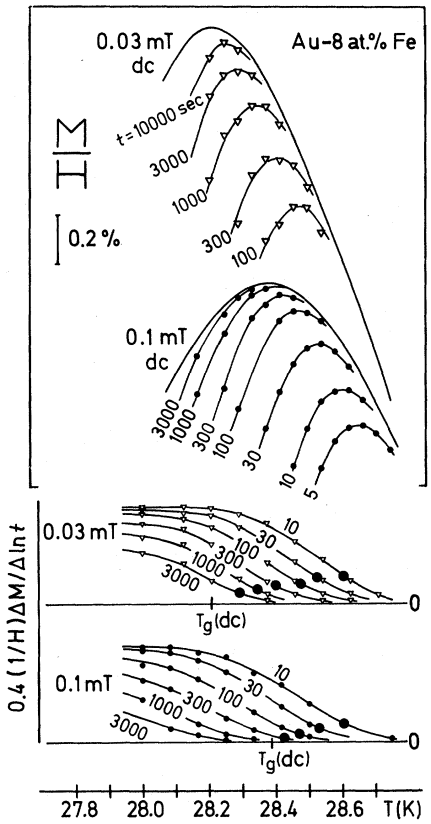

FIG. 2. M/H and $(1/H)\Delta M/\Delta\ln t$ at observation times $t=5-10^4$ sec at 0.03 and 0.1 mT. The large filled circles at the $\Delta M/\Delta\ln t$ curves indicate the location of the cusp of respective M/H curve. The curves marked DC are the field-cooled curves. All M/H curves have a common zero level.

Since the integrand is peaked around $t=\tau$ we bring out $g(\tau)$ from the integral attaining the value $g(\tau_m)$, where $\tau_m \approx t$. After integration and assuming $t >> \tau_{\min}$ we get

$$\frac{\partial m}{\partial \ln t} = m_0(\tau_m) g(\tau_m) e^{-t/\tau_{\max}};\quad \tau_m < \tau_{\max} \ . \qquad (6)$$

Thus, by observing the change of magnetization with time at constant temperature, the relaxation-time spectrum may, in principle, be obtained. However, by plotting the experimental data of M/H and $(1/H) \times \Delta M/\Delta \ln t$ at constant t a close equivalence to $\chi'(\omega)$ and $\chi''(\omega)$ in ac susceptibility measurements[3] at observation time $1/\omega$ is found. These plots are presented in Fig. 2 at observation times $5-10^4$ sec and at 0.03- and 0.1-mT fields. As can be seen in the figure the $\Delta M/\Delta \ln t$ curves evolve continuously to lower temperatures with increasing t values. At low-t values the cusp of the M/H curves coincides in temperature with the onset of a finite $\Delta M/\Delta \ln t$ at respective time, i.e., when $\tau_{\max} \approx t$. This is analogous to ac susceptibility results of χ' and χ''.[3-5,9] However, as is particularly notifiable on the 0.1-mT curves, there is a marked cutoff of the change of magnitude as well as of location of the susceptibility cusp at long observation times in spite of the continuous evolution of respective $\Delta M/\Delta \ln t$ curves towards lower temperature. This readily implies a reduction of the total magnetization before $\tau_{\max} \approx t$ and the cluster blocking occurs. A similar behavior is quite likely found in the zero-field curve, though at considerably longer times. In Fig. 3 we have plotted the variation of T_g vs observation time t according to Eq. (2). We have also included $T_g(\mathrm{DC})$, assumed to be the position of the susceptibility cusp at infinite time. As is clearly indicated in Fig. 3 there is a virtual collapse of the Vogel-Fulcher law (or *any* other law of similar form) within an extremely narrow temperature interval above $T_g(\mathrm{DC})$. This sudden breakdown strongly implies a *collective transition phenomenon,* signified by a characteristic time. This characteristic time is 5×10^2 sec at the 0.1-mT field and extrapolated from Fig. 3 to be 10^5 sec at zero field. We propose that this characteristic time is coupled to a critical value of the relaxation times (τ_{crit}), where intercluster coupling becomes strong enough to initiate cluster recombinations. Possibly (but not ultimately) these larger clusters attain infinite relaxation time in a way of local ordering. Due to the collective nature of the transition these ordered regions suggestively percolate the sample like infinite chains.[13] A cusp of the susceptibility curve at observation time $t > \tau_{\mathrm{crit}}$ occurs if these cluster recombinations give a significant net reduction of the total magnetic moment of the sample. As is apparent from the finite $\Delta M/\Delta \ln t$ at $t > \tau_{\mathrm{crit}}$ below $T_g(\mathrm{DC})$ (see Fig. 2, 0.1-mT curves) finite relaxation times larger than τ_{crit} exist below $T_g(\mathrm{DC})$. As the temperature is decreased a sharp drop of the susceptibility occurs when $\tau_{\max} \approx t$ due to the onset of cluster blocking. Time effects simultaneously appear. The relaxation time spectrum at short times ($<\tau_{\mathrm{crit}}$) is unaffected by the ordering at $T_g(\mathrm{DC})$ and no anomaly at this temperature is observed at observation times $t < \tau_{\mathrm{crit}}$. Susceptibility curves compatible with the above description are shown in Fig. 4.

The relaxation times *decrease* with field, manifested by a shift of the $\Delta M/\Delta \ln t$ curves to lower temperatures (see Fig. 2). Thus "viscosity" effects at a *specific* observation time occur at lower temperature with increasing field. This has previously been observed in dc magnetization measurements ($t \sim 100$ sec) by Chamberlin *et al.*[14] A decrease of the relaxation times with field was already recognized by Néel,[15] but is not reflected in the change of the position of the susceptibility cusp as envisaged by Wohlfarth.[16] In fact, due to the reduction of τ_{crit} with field in Au(Fe) the cusp of the field-cooled curve occurs at an increasingly higher temperature. The observed *increase* of the density of relaxation

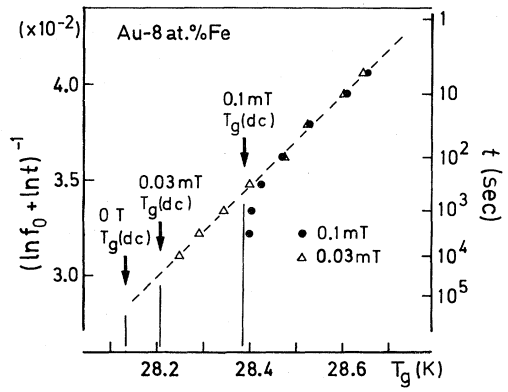

FIG. 3. Observation time t vs T_g according to the Vogel-Fulcher law [Eq. (2)] with $f_0 = 10^{10}$ sec^{-1}. The slope of the dotted line gives $E/k = 40$ K and $T_0 = 26.9$ K.

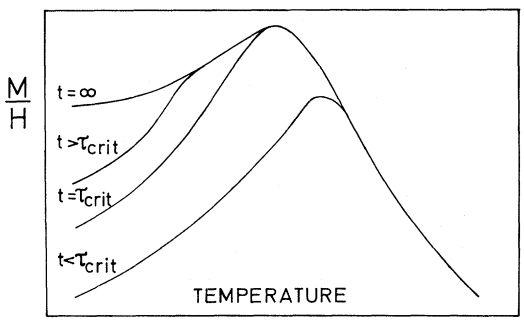

FIG. 4. Representative susceptibility (M/H) curves at various observation times with respect to the critical relaxation time (τ_{crit}). Note that τ_{crit} depends on the applied magnetic field (see text).

times at τ_{max} with field possibly contributes to the reduction of τ_{crit}.

The small increase of the magnetization with time above T_g(DC) of the field-cooled curve is in agreement with the general idea of a wide spectrum of relaxation times evolving towards longer times with decreasing temperature. Due to the suppression of the spectrum to shorter times with increasing field these time effects are gradually reduced. The decrease of the magnetization with time below T_g(DC) of the field-cooled curve is interpreted to originate from uncompensated moments at the surface of the ordered regions. As these ordered regions "melt" together frustration effects appear and rearrangements of the magnetic moments occur in order to lower the magnetostatic energy. External fields stabilize these metastable spin configurations but net moments will eventually be "trapped" creating an internal field. Hence, the field-cooled curve will at laboratory time scales exhibit a higher value than the hypothetical $t = \infty$ curve.

Conclusively, we propose a simplified picture, where the spin-glass state consists of two coexisting phases, one ordered phase and one phase characterized by a broad spectral distribution of relaxation times with a finite maximum value. The ordered phase appears at T_g(DC) due to a critical value of the relaxation times. At T_g(DC) the ordered phase occupies very small volume (like a spider's net) but will continuously grow in size with decreasing temperature. This growth occurs at the expense of clusters with the largest relaxation times, which will enter the ordered phase. The ordering probability depends not only on the value of the largest relaxation times but also on the concentration of large relaxation times within the sample. When measurements are made at observation times $t < \tau_{crit}$ no detectable anomaly at T_g(DC) will be found and the results may be interpreted with the *only* assumption of a broad spectrum of relaxation times. When measurements are made at observation times $t > \tau_{crit}$ the onset of ordering at T_g(DC) will be observed and time effects at a temperature below T_g(DC) will be found, i.e., when $\tau_{max} \approx t$.

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