Relaxation in spin glasses at weak magnetic fields

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The low-field time-dependent magnetic susceptibility of the amorphous metallic spin glass $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ has been studied in a superconducting-quantum-interference-device magnetometer. Both ac susceptibility and zero-field-cooled susceptibility measurements have been utilized to cover more than ten decades in time, 3×10^{-6} - 10^{5} sec. At short observation times the equilibrium relaxation is observed, while at long observation times the influence of the aging process dominates the relaxation. The aging behavior is shown to persist through the spin-glass freezing temperature and it remains to have a vital influence on the relaxation until the spin system has been allowed to age for times comparable to the longest relaxation time of the system. The functional form of the time-dependent susceptibility is also illuminated.

I. INTRODUCTION

Spin-glass dynamics have lately received immense interest among both experimentalists and theoreticians. From experiments¹ it has been established that the characteristic slow nonexponential relaxation of spin glasses starts to develop at temperatures well above the spin-glass freezing temperature T_g . The relaxation occurs continuously from atomic time scales to a temperature and magnetic-field-dependent characteristic time, au_{max} , which signifies the end of the dynamic process. The dependence of $\tau_{\rm max}$ on temperature and magnetic field has recently been studied in dynamic scaling analyses.^{2–5} These studies have involved experiments on short-range^{2,3} (insulating) and long-range⁴ (metallic) spin glasses as well as short-range Ising spin glass models,⁵ investigated by Monte Carlo simulations. In the zero-field limit the results of these analyses seem to favor a divergence of $\tau_{\rm max}$ at T_g . Below T_g , τ_{max} is inaccessible to laboratory time scales. Furthermore, in the low-field regime the spinglass relaxation is highly influenced by an aging phenomenon.⁶ This implies that the spin-glass phase is a thermodynamic nonequilibrium state and that the response to a small field step is dependent on the wait time, t_w , at constant temperature prior to the field change. The relaxation only resembles spin-glass dynamics at thermodynamic equilibrium at observation times much smaller than t_w .⁶ The influence of the aging process on the relaxation is the main cause of the present confusion as to the functional form of the spin-glass relaxation. At the same time as the aging phenomenon was discovered and found to be of importance for the relaxation in spin glasses, other experimental results were reported implying that a stretched exponential functional form accurately described the spin-glass dynamics below T_g .⁷ Inspired by this work, theoreticians managed to construct spin-glass models with a stretched exponential functional form for the relaxation at long times.^{8–10} However, recent experimental results^{11–13} at temperatures below T_g show that a stretched exponential form may only be used to characterize the influence of the aging process. Aging is a thermodynamic nonequilibrium phenomenon not included in spin-glass theories of today. Hence, to make a correct comparison between experiments and theoretical models feasible it is necessary to properly account for the influence of the aging process on the total relaxation. This can only be accomplished using experimental data in as wide a time perspective as possible and combining results from different measuring probes covering different time intervals.

In this paper we report measurements of the low-field time-dependent magnetic susceptibility of an amorphous metallic spin glass. Both ac susceptibility and zero-fieldcooled susceptibility measurements have been utilized to obtain an experimental time window covering more than ten decades, 3×10^{-6} -10⁵ sec. At short observation times the equilibrium relaxation is observed while at long observation times there is a strong influence of the aging process on the relaxation. A special study in the immediate vicinity of the spin-glass freezing temperature shows that the aging process persists until the spin system has been allowed to age for times comparable to the longest relaxation time of the spin system. Furthermore, it is shown that the final approach of the relaxation towards equilibrium is extremely sensitive to the magnitude of the applied magnetic field.

The paper is organized as follows. In Sec. II the experimental procedure is described. Section III is devoted to the coupling between results from ac and dc susceptibility measurements and explains the conception timedependent susceptibility. In Sec. IV the results are presented and Sec. V contains a discussion on the results.

II. EXPERIMENTAL PROCEDURE

Ribbons of the amorphous metallic system $(Fe_x Ni_{1-x})_{75}B_{16}P_6Al_3$ have been prepared by the centrifugal spin quenching technique.¹⁴ The ribbons have a typical cross section of $0.02 \times 1 \text{ mm}^2$. The magnetic phase diagram and general magnetic properties of this system have been extensively investigated previously.¹⁵ Specifi-

cally it has been shown that spin-glass properties are found for $x \leq 0.17$. In this study ribbons with a nominal concentration of x = 0.15 cut in pieces, 5 mm of length, were used. The sample consisted of 20 pieces packed together. The measurements were performed in a superconducting quantum-interference device (SQUID) magnetometer¹⁶ utilizing the SHE Corp. (San Diego) model 30 electronic system. The ac susceptibility measurements were performed in the frequency range 0.51 Hz- 51 kHz and in the temperature range $0.9 < T/T_g < 1.16$, where $T_g = 22.6$ K as determined from static scaling analyses on the same spin-glass system.¹⁷ The amplitude of the oscil-lating field was ~50 mG. The zero-field-cooled (ZFC) susceptibility measurements were performed in the time interval $3-10^5$ sec and in the temperature range $0.9 < T/T_g < 1.01$ using a small probing magnetic field, H = 0.1 G. A magnetizing coil consisting of a 0.025mm-diam copper wire was wound directly onto the sample. A similar but antiparallel coil was placed nearby the sample for compensating purposes. The coils were attached to a sapphire rod, constituting part of the sample holder, and centered in one of the pickup coils of the flux transformer of the SQUID magnetometer. The SQUID electronic unit was connected to a lock in amplifier where the real and imaginary parts of the ac susceptibility were simultaneously detected. By using both the normal output jack, 0.51 Hz-2 kHz, and the high-frequency output jack, 2-51 kHz, of the SQUID electronic unit it is feasible to make measurements in the above given frequency range. The temperature was measured with a copper thermometer¹⁸ consisting of a 0.04-mm-diam copper wire, 1 m in length, bifilarly wound onto the sample holder. The ac susceptibility measurements were performed by changing the temperature in steps of 0.2 K, and at constant temperature the real and the imaginary parts were recorded. The extracted data are averages from heating and cooling cycles. In the ZFC measurements the sample was cooled in zero applied field from the reference temperature, $T_{ref} = 24.1$ K ($T_{ref}/T_g = 1.07$). At a constant temperature T, and after a wait time t_w , a small probing field was applied and the relaxation of the magnetization was detected. t_w is defined as the time interval between the moment T is reached and the time the probing field is applied. Afterwards the sample was heated to T_{ref} where the reference level of the ZFC susceptibility was recorded.

III. THE TIME-DEPENDENT SUSCEPTIBILITY

The time-dependent susceptibility offers a unique possibility to cover an unusually wide time window of the spin-glass dynamics. The susceptibility at a given observation time can be measured by both ZFC and ac susceptibility experiments. The ZFC susceptibility at the observation time t equals $\chi_{ZFC}(t) = (1/H)M(t)$. In ac susceptibility measurements, where $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$ is recorded, the observation time equals $1/\omega$, ω representing the angular frequency of the external oscillating field. In the low-field regime it has previously been shown that the ZFC and ac susceptibilities are related through,¹⁹

$$(1/H)M(t) \simeq \chi'(\omega), \quad t = 1/\omega$$
 (1)

Similarly, it is possible to relate the relaxation rate, $S(t) = (1/H)\partial M/\partial \ln t$, of the ZFC susceptibility to the imaginary part of the ac susceptibility by,¹⁹

$$(1/H)\partial M/\partial \ln t \simeq -(2/\pi)\chi''(\omega), \quad t = 1/\omega$$
 (2)

Thus, combining our results of the ac susceptibility $(3 \times 10^{-6} < 1/\omega < 3 \times 10^{-1} \text{ sec})$ with the time dependence of the ZFC susceptibility $(3 < t < 10^5 \text{ sec})$ we cover some 10 decades in time of the spin-glass dynamics.

Spin-glass systems are, within laboratory time scales, captured in thermodynamic nonequilibrium states below T_g . They are thus subjected to aging phenomena which can be revealed in both ac (Ref. 20) and ZFC (Ref. 6) susceptibility measurements. In ac susceptibility measurements, at constant temperature and constant frequency, aging is detected as a time dependence of $\chi'(\omega)$ and $\chi''(\omega)$. In ZFC experiments aging is revealed by a strong t_w dependence of the time-dependent ZFC susceptibility curves. Only at observation times much smaller than t_w the experimental data are representative of spin-glass relaxation at thermodynamic equilibrium. This implies, however, that most ac susceptibility data can be considered as being representative for spin-glass relaxation at thermodynamic equilibrium.

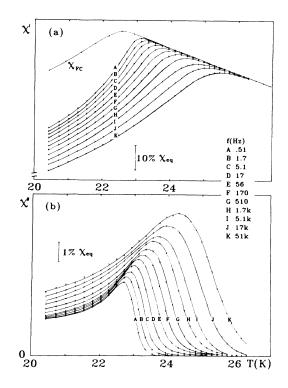


FIG. 1. Complex ac susceptibility $[\chi(\omega) = \chi'(\omega) + i\chi''(\omega)]$ versus temperature at different frequencies of the applied oscillating field. (a) $\chi'(\omega)$. The field-cooled susceptibility (χ_{FC}) at an applied field of H = 50 mG is also plotted. Ten percent of the equilibrium susceptibility $(\simeq\chi_{FC})$ is indicated. (b) $\chi''(\omega)$. One percent of the equilibrium susceptibility $(\simeq\chi_{FC})$ is indicated.

IV. RESULTS

In Figs. 1(a) and 1(b) we have visualized the temperature dependence of the equilibrium real $[\chi''(\omega)]$ and imaginary $[\chi''(\omega)]$ components of the ac susceptibility for different frequencies of the applied oscillating field. For comparison the field-cooled (FC) susceptibility curve has been included. The FC curve was obtained by cooling the sample in a constant applied magnetic field of 50 mG. The general features of the zero-field ac susceptibility²¹ in spin glasses can from Fig. 1 be stated as follows. The cusp in χ' shifts towards higher temperatures with increasing frequency. χ'' exhibits a sharp anomaly with an inflection point at a temperature closely coinciding with the cusp temperature of the χ' curve. The magnitude of χ'' increases continuously with increasing frequency.

In Figs. 2(a) and 2(b) the time dependence of the ZFC susceptibility (1/H)M(t) and the corresponding relaxation rate [S(t)] at 0.1 G are visualized for different wait times at $T = 0.9T_g$. S(t) has been obtained from a sliding fivepoint differentiation of the raw data. The ZFC curves in Fig. 2(a) are drawn relative to the susceptibility value at T_{ref} . As is clearly seen from Fig. 2, there is a pronounced t_w dependence of the curves. The most striking feature is an inflection point in (1/H)M(t) and a corresponding

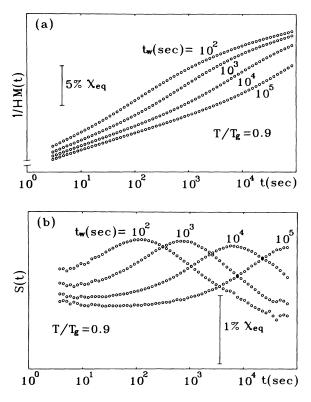


FIG. 2. Zero-field-cooled susceptibility [(1/H)M(t)] and corresponding relaxation rate $[S(t)=(1/H)\partial M/\partial \ln t]$ at different wait times $(t_w=10^2, 10^3, 10^4, \text{ and } 10^5 \text{ sec})$ plotted versus $\log(t)$. T=20.3 K, $T_g=22.6$ K, $T/T_g=0.9$, H=0.1 G. (a) (1/H)M(t). Five percent of the equilibrium susceptibility $(\simeq \chi_{FC})$ is indicated. (b) S(t). One percent of the equilibrium susceptibility ($\simeq \chi_{FC}$) is indicated.

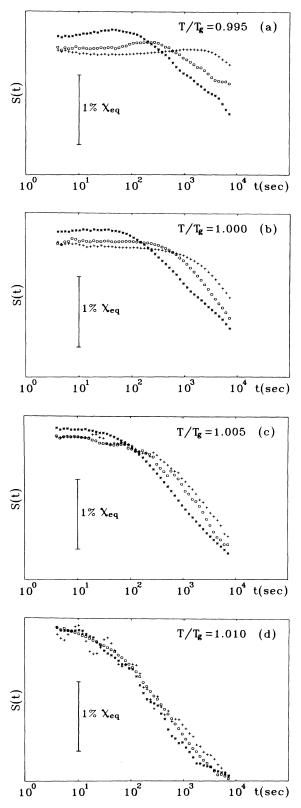


FIG. 3. Relaxation rate $[S(t)=(1/H)\partial M/\partial \ln t]$ vs $\log(t)$. The various curves refer to different wait times, $t_w = 10^2(*)$, $10^3(\odot)$, and $10^4(+)$ sec. One percent of the equilibrium susceptibility ($\simeq \chi_{FC}$) is indicated. (a) $T/T_g = 0.995$, (b) $T/T_g = 1.000$, (c) $T/T_g = 1.005$, (d) $T/T_g = 1.010$.

maximum in S(t) at $t \simeq t_w$ for all wait times. These attributes are entirely due to the aging process which, as is particularly evident from the relaxation rate curves, has a dominating influence on the spin-glass relaxation in this time window. It is only the S(t) curve corresponding to the largest wait time ($t_w = 10^5$ sec) that at short observation times levels down close to the equilibrium relaxation rate which can be estimated from the imaginary component of the ac susceptibility (see below). The appearance of the ZFC susceptibility shown in Fig. 2 is universal for spin glasses at temperatures below T_g and has been experimentally observed in other metallic, ^{11,12} insulating¹³ and semiconducting²² spin-glass systems. To establish whether the aging process is a unique property of the spin-glass state below T_g or if it exists at all temperatures where the relaxation takes place in a broad time interval, we have made an experimental survey of the t_w dependence of the ZFC susceptibility in the immediate vicinity of T_g . Figure 3 shows the relaxation rate versus logarithm of time at four temperatures around T_g . At each temperature the relaxation rate has been measured at three different wait times, $t_w = 10^2$, 10^3 , and 10^4 sec. This sequence of figures clearly shows that the aging process persists through T_g and that it disappears when the total ag-ing time is of the order of the largest relaxation time

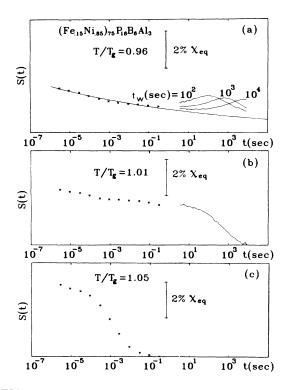


FIG. 4. Relaxation rate versus $\log(t)$ obtained from ac susceptibility $[S(t) = -(2/\pi)\chi''(\omega), 3 \times 10^{-6} < 1/\omega < 3 \times 10^{-1} \text{ sec}]$ and zero-field-cooled susceptibility $[S(t) = 1/H)\partial M/\partial \ln t, 3 < t < 10^4 \text{ sec}]$ measurements. Two percent of the equilibrium susceptibility $(\simeq\chi_{FC})$ is indicated. (a) $T = 0.96T_g$. The various ZFC curves refer to different wait times, $t_w = 10^2$, 10^3 , and 10^4 sec. The solid line represents the best power-law fit $(S \propto t^{-0.05})$ to the ac susceptibility data. (b) $T = 1.01T_g$. (c) $T = 1.05T_g$.

 (τ_{max}) of the spin system which is defined as the observation time where the relaxation rate drops to zero (cf. Fig. 3d, where $\tau_{\text{max}} \simeq 10^3$ sec). However, on increasing the temperature further above T_g the influence of the aging process is never detectable since τ_{max} becomes much smaller than the time it takes to achieve thermal equilibrium in an experiment.

In Fig. 4 we have visualized the relaxation rate [S(t)]versus the logarithm of time for three temperatures around T_g . The data points in the observation time range $3 \times 10^{-6} - 3 \times 10^{-1}$ sec have been obtained from the ac susceptibility [Eq. (2)] while the solid lines in the observa-tion time range $3-10^4$ sec refer to the relaxation rate obtained from the ZFC susceptibility. The different ZFC curves at $T = 0.96T_g$ have been recorded at different wait times, $t_w = 10^2$, 10^3 , and 10^4 sec. They nicely illustrate the effect of aging in a large time perspective. The best fit of the relaxation rate to a power-law behavior using only the ac susceptibility data, $S(t) \propto t^{-0.05}$, is shown by a solid line in Fig. 4(a) and reflects the equilibrium relaxation rate to which the curves for different wait times will level down in the long time limit $(t_w \rightarrow \infty)$. The choice of a power-law fit is justified below. At $T = 1.01T_g$ it is possible to out wait the effect of aging [cf. Fig. 3(d)] and the measured ZFC curve in Fig. 4(b) reflects the equilibrium relaxation. On further increase of the temperature, the maximum relaxation time of the spin system drastically moves towards shorter times. This is illustrated at $T = 1.05 T_g$ [Fig. 4(c)] where $\tau_{\rm max}$ has moved roughly five decades in time in comparison with $\tau_{\rm max}$ at $T = 1.01 T_g$.

V. DISCUSSION

In low magnetic fields a linear response of the spinglass relaxation is found.^{23,24} Experiments of the ac and ZFC susceptibilities, in this field regime, mirror the time variation of the zero-field spin-spin time correlation function q(t). The time dependence of the q parameter can also be studied in Monte Carlo (MC) simulations of spinglass models. Thus a check of the validity of these models is feasible by a direct comparison with experiments on real spin glasses. This requires, however, that the experiments are well defined as regards the influence of the aging process and the magnitude of the external magnetic field.

In Fig. 4 we have chosen to present the time dependence of the relaxation rate instead of the time-dependent susceptibility. One advantage of displaying the relaxation rate instead of the susceptibility is that the effect of aging becomes more pronounced in such a plot. The general features of the equilibrium spin-glass relaxation and the additional effect of the aging process can from Fig. 4 be summarized as follows. At temperatures above T_g , the equilibrium relaxation rate slowly decreases with increasing observation time in a logarithmic time perspective. In a narrow time segment, centered around the observation time that equals τ_{max} , the relaxation rate drops to zero. τ_{max} drastically increases when approaching T_g and possibly in the zero-field limit diverges at this temperature.²⁻⁴ The final approach towards equilibrium is strongly dependent on the applied field. In Fig. 5 we have plotted the re-

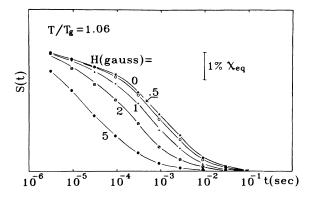


FIG. 5. Relaxation rate $[S(t) = -(2/\pi)\chi''(\omega)]$ vs $\log(t)$ at different dc magnetic fields (*H*). One percent of the equilibrium susceptibility $(\simeq\chi_{FC})$ is indicated. $T/T_g = 1.06$.

laxation rate at $T/T_g = 1.06$, obtained from ac susceptibility data [Eq. (2)] versus log(t) for different superposed dc magnetic fields. Largely, the only effect of the applied field is to suppress the edge of the relaxation rate towards shorter observation times. At low fields this suppression is proportional to H^2 , in accordance with results from dynamic scaling analyses on the same spin-glass system.⁴ When approaching T_g , the field dependence of the edge of the relaxation rate rapidly increases and its zero-field limit therefore becomes exceedingly difficult to achieve experimentally. Below T_g an end of the relaxation process is never observed in laboratory time scales. The equilibrium relaxation only displays a slow decrease with the logarithm of observation time. However, the effect of aging on the relaxation rate now becomes clearly detectable. It superimposes an extra part onto the equilibrium relaxation rate and the total relaxation rate exhibits a maximum at an observation time closely equal to the wait time (t_w) . At tempertures below T_g the general characteristics of the spin-glass relaxation are not compatible to the currently popular suggestion⁷ of a pure stretched exponential func-tional form $(M(t) \propto \exp[-(t/t_p)^{1-n}])$. A stretched exponential only enters the functional form of the spin-glass relaxation to characterize the aging phenomenon and must not be associated with the equilibrium relaxation. This has been amply demonstrated by Nordblad et al.¹¹ and Alba et al.¹²

Within a critical fractal cluster model⁹ the following expression for the equilibrium relaxation rate in the zero-field limit has been derived:²⁵

$$\partial M / \partial \ln t \propto (t/\tau_0)^{-\beta/z\nu} \exp[-(t/\tau_{\max})^{-\beta\delta/z\nu}]$$
 (3)

where β , δ , *z*, and ν are standard static and dynamic critical exponents. τ_{max} is expressed as

$$\tau_{\max} = \tau_0 (T/T_g - 1)^{-zv} , \qquad (4)$$

where τ_0 represents the minimum relaxation time of the spin system (τ_0 can be identified with the single spin-flip time, $\simeq 10^{-13}$ sec). Equation (3) yields a slow power-law decay [experiments and simulations give values of β/zv (Refs. 2-5) in the range 0.05-0.1] followed by a sudden disappearance of the relaxation rate at $t \sim \tau_{max}$ characterized by a stretched exponential form. Since τ_{\max} diverges at T_g [Eq. (4)], only a power law is found below T_g . Thus, the relaxation rate according to Eq. (3) possesses the same fundamental characteristics of the equilibrium relaxation rate as found experimentally in spin glasses. Adopting the values of the standard critical exponents and τ_0 as found from static¹⁷ and dynamic⁴ scaling analyses on the presently investigated spin-glass system ($\beta = 0.38 \ \delta = 10$, zv = 8.2, and $\tau_0 = 2 \times 10^{-13}$ sec) a qualitatively as well as quantitatively correct description of the time dependence of the experimental relaxation rate, valid through T_g , is found.25

The behavior of the equilibrium relaxation in real spinglass systems resembles the corresponding relaxation found in a computer simulation by Ogielski⁵ on a shortrange three-dimensional Ising spin-glass model. The time variation of the q parameter at thermodynamic equilibrium follows at temperatures below T_g a pure power law. This functional form and the value of the power-law exponent (-0.05 at $T/T_g = 0.96$) are in accordance with the results presented in this paper. In a logarithmic time perspective the computer simulations revealed at temperatures above T_g a slow decay of q at short times followed by a sudden drop to zero. This time dependence of q was by Ogielski described by a power law times a stretched exponential form where the latter part characterizes the sudden drop at the very end of the relaxation. A plot of the relaxation rate, $\partial q / \partial \ln t$, using parameters given from the computer simulations yield curves similar in form to those found in measurements of real spin-glass systems [cf. Fig. 4(b) and 4(c)]. Thus the relaxation in real spin glasses shows great similarity to that found by Ogielski. However, to make the comparison complete, simulations of the q parameter at nonequilibrium should be performed in order to incorporate the experimentally observed aging process.

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