Time-dependent ac susceptibility in spin glasses

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The zero-field ac susceptibility $\chi(\omega)=\chi'(\omega)+i\chi''(\omega)$ of the short-range Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$ has been investigated in the vicinity of the spin-glass temperature T_g in the frequency interval 5×10^{-3}–5×10^{1} Hz. Because of the influence of aging, $\chi(\omega)$ is time dependent. After a temperature quench to a temperature $T\leq T_g$, $\chi''(\omega)$ is found to decrease with time. The decay towards equilibrium is logarithmic in time and the corrections to the equilibrium values of $\chi''(\omega)$ decrease slowly with increasing frequency. At temperatures $T>T_g$ in the vicinity of the freezing temperature of $\chi(\omega)$, a weak increase of $\chi''(\omega)$ with time is found. By use of linear response relations, the observed time dependence of $\chi''(\omega)$ is shown to be consistent with results from time-dependent magnetization measurements. Furthermore, the existence of an overlap length scale in spin glasses has been verified by $\chi''(\omega)$ and time-dependent magnetization experiments.

INTRODUCTION

A spin glass will at experimental time scales be in a thermodynamic nonequilibrium state close to and below the spin-glass temperature T_g. The nonequilibrium character of the spin-glass material is particularly clear when at constant temperature, after a temperature quench from a temperature $T>T_g$, a small magnetic field is applied; the response to the field depends on the waiting time at constant temperature before applying the field.[1,2] During the waiting time, there are no detectable changes in the magnetization. Nevertheless, the response of the spin system to the applied field becomes slower and slower with increasing waiting time. This implies that the spin glass is in a nonequilibrium state during the waiting time and only slowly evolves towards equilibrium; the spin system is said to undergo *aging*. Despite this nonequilibrium character of a spin-glass state, experimental studies have firmly established that linear response relations hold as long as the magnetic field changes are sufficiently small.[3] The magnetic response function will, however, depend on the *age* of the spin system which, in turn, implies that the spin-glass response is described by a nonequilibrium response function. Such a response function has implications for the ac susceptibility. The ac susceptibility will also depend on the age of the spin system and will therefore be time dependent. A time dependence has indeed been observed in experiments on metallic[4] as well as on insulating[5,6] spin-glass compounds. A typical experimental observation is that both components of the ac susceptibility decrease in magnitude with the time spent below the spin-glass temperature. Furthermore, the time-dependent effects are stronger for lower frequencies. It should be noted that a time dependence of the ac susceptibility has also been found in experiments on a two-dimensional spin glass,[7] showing that these effects are detectable even when there is no finite-temperature spin-glass transition.

In this paper we present an extended study of the time-dependent ac susceptibility on the short-range Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$. The objectives of this study were to (i) get a better understanding of the time-dependent ac susceptibility in spin glasses and (ii) to investigate the influence of a time-dependent ac susceptibility on dynamic scaling analyses.[8]

EXPERIMENT

The measurements were performed on a single crystal of $Fe_{0.5}Mn_{0.5}TiO_3$ (Ref. 9) using a SQUID magnetometer. For the ac susceptibility measurements, $\chi(\omega)=\chi'(\omega)+i\chi''(\omega)$, a magnetizing coil was wound directly onto the sample, generating a small oscillating magnetic field $h=h_0\sin(\omega t)$ ($h_0\sim0.1$G) along the crystallographic **c** axis. $\chi(\omega)$ was measured in the temperature range $0.9<T/T_g<1.1$ ($T_g=20.9$ K) and in the frequency interval 5×10^{-3}–5×10^{1} Hz (5×10^{-3} Hz, 1.7×10^{-2} Hz, 5×10^{-2} Hz, 5 Hz, 17 Hz, and 55 Hz). Typically, a phase stability of $\leq0.01°$ could be maintained for 10^4 s in the ac susceptibility measurements.

To compensate for the direct contribution from the ac field to the measured signal, a second and similar coil was wound onto the sapphire sample holder. The distance between the magnetizing and compensating coils was adjusted to match the distance between two sections of the pickup coil (a third-order gradiometer is used as a pickup coil). In the beginning of the ac susceptibility experiments, the sample holder was inserted into the pickup coil in such a way that the sample was centered in one of the pickup coil sections. This procedure minimizes the influence of displacements of the sample, caused by thermal expansion of various parts in the experimental setup, on the measured signal. The distance between the magnetizing and compensating coils was, however, not exactly the same as the distance between two sections of the pickup coil and the compensating coil was therefore less well centered. This implies that the measured signal is more sensitive to displacements of the compensating coil. Such displacements will only influence the in-phase component of the measured ac susceptibility and the long-time stability of $\chi'(\omega)$ was therefore not as good as for $\chi''(\omega)$.

In one type of experiment the sample was stepwise cooled from a temperature above T_g and at each temperature the time dependence of the ac susceptibility was registered for $3\times10^3-10^4$. For a temperature decrease of 0.2–1 K, temperature stability was obtained within ~ 10 s. This experimental procedure, which mimics the procedure used to collect data for dynamic scaling analyses, was performed using cooling rates in the interval $2.5\times10^{-5}-5\times10^{-4}$ K/s.

In another type of ac susceptibility experiment the sample was quenched to a temperature T_m below T_g and a time decay of $\chi''(\omega)$ was registered for 3×10^3s. Then the sample was subjected to a temperature cycling ΔT (the duration of the temperature cycling $\sim$10s), and when T_m was recovered $\chi''(\omega)$ was registered for another 10^4s.

In addition to the ac susceptibility measurements, zero-field-cooled (ZFC) magnetization experiments have been performed. The sample was quenched in zero field to $T_m < T_g$ and it kept a constant temperature for a waiting time t_w. It was then subjected to a temperature cycling ΔT, and when T_m was recovered a small magnetic field was applied and the relaxation $M(t)$ was recorded. These measurements were carefully checked to ensure that the fields used were well within the linear response regime. All results presented are given in units of the field-cooled susceptibility, $\chi_{\rm eq}=M_{\rm FC}/H$, where $M_{\rm FC}$ is the measured field-cooled magnetization.

LINEAR RESPONSE

As mentioned above, even if the spin glass is in a nonequilibrium state, results from time-dependent magnetization measurements clearly show that linear response relations hold provided that the field changes are sufficiently small. This implies that the magnetization $M(t)$ responds linearly to a magnetic field $H(t)$ applied after a waiting time t_w,

$$M(t)=\int_0^{t-t_w} dt'\chi(t,t-t')H(t-t') \ . \tag{1}$$

The response function $\chi(t,t-t')$ gives the magnetic response at time t to a unit magnetic field impulse at time $t-t'$. The time $t=0$ is generally set equal to the instant of time when the spin glass is quenched to a temperature below the spin-glass temperature. However, a spin glass will be out of equilibrium on experimental time scales already at temperatures above the spin-glass temperature.[10] Therefore, $t=0$ should be set equal to the instant of time when the spin glass is quenched to any temperature where it is in a nonequilibrium state, irrespective of if it is below or above the spin-glass temperature. The ac susceptibility $\chi(\omega;t)$ is measured by applying a small oscillating magnetic field $H(t)\sim e^{i\omega t}$. $\chi(\omega;t)$ can then be found by a simple Fourier transform of the magnetization over a time segment $t_m(\sim 2\pi/\omega)$ centered around t,

$$\chi(\omega;t)=\frac{1}{t_m}\int_{t-(1/2)t_m}^{t+(1/2)t_m} dt''e^{-i\omega t''} \times\int_0^{t''-t_w} dt'\chi(t'',t''-t') \times e^{i\omega(t''-t')} \ . \tag{2}$$

Assuming that the magnetic response function varies only little over the time segment t_m, which for spin glasses is valid as long as $t_m \ll t_w$, this equation can be simplified to

$$\chi(\omega;t)=\int_0^{t-t_w} dt'\chi(t,t-t')e^{-i\omega t'} \ . \tag{3}$$

Most of the results presented in this paper are concerned with the time dependence of the out-of-phase component of the ac susceptibility $\chi''(\omega;t)$, which can be written

$$\chi''(\omega;t)\sim -\int_{\tau_0}^{t-t_w} d\ln(t')\left[\frac{\partial M}{\partial \ln(t')}\right]_{t_w}\sin(\omega t') \ , \tag{4}$$

where $t-t_w \ll t_w$ is assumed, which implies that the response function can be set equal to $(\partial M/\partial t')_{t_w}$, i.e., the time derivative of the magnetization for a spin glass of age t_w. Furthermore, the lower limit of the integration is set equal to a microscopic time τ_0. Assuming that the relaxation rate $[\partial M/\partial\ln(t')]_{t_w}$ is a slowly varying function of $\ln(t')$ close to $t'\approx 1/\omega$, one finally obtains the following approximate relation:

$$\chi''(\omega;t)\approx -C\left[\frac{\partial M_{t'\approx 1/\omega}}{\partial \ln(t')}\right]_{t_w} \ . \tag{5}$$

The proportionality constant C will be close to $\pi/2$. This equation, which is valid for $t-t_w \ll t_w$, tells us that $\chi''(\omega;t)$ will decrease with time if the relaxation rate $[\partial M/\partial\ln(t')]_{t_w}$ at constant observation time $t'\approx 1/\omega$ is a decreasing function of t_w. If $[\partial M/\partial\ln(t')]_{t_w}$ instead increases with t_w, $\chi''(\omega;t)$ is expected to increase with time.

RESULTS AND DISCUSSIONS

Figure 1 shows the time dependence of $\chi''(\omega)$ at different temperatures in an experiment where the temperature was stepwise decreased. For each temperature the time dependence of $\chi''(\omega)$, registered for 10^4s, is visualized on a logarithmic time scale. The time $t=0$ has in these measurements been set equal to the instant of time when temperature stability was achieved. *Close to and below* T_g [at temperatures below the peak of $\chi''(\omega)$ vs T], there is a decrease with time of $\chi''(\omega)$. The decay towards equilibrium is very slow, close to logarithmic in time. At temperatures $T>T_g$, close to the inflection point temperature of $\chi''(\omega)$, there is a tendency for $\chi''(\omega)$ to increase with time. However, the size of this time-

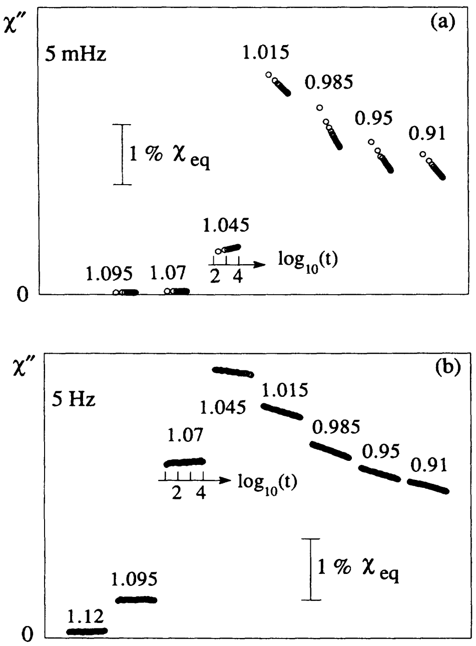

FIG. 1. $\chi''(\omega)$ vs $\log_{10}(t)$ at different temperatures. The temperature was stepwise decreased and at each temperature a recording of the time decay was performed for 10^4 s. The numbers correspond to the temperatures (T/T_g) where the decay was measured. The effective cooling rate was 5×10^{-5} K/s. $T_g=20.9$ K. (a) $\omega/2\pi=0.005$ Hz and (b) $\omega/2\pi=5$ Hz.

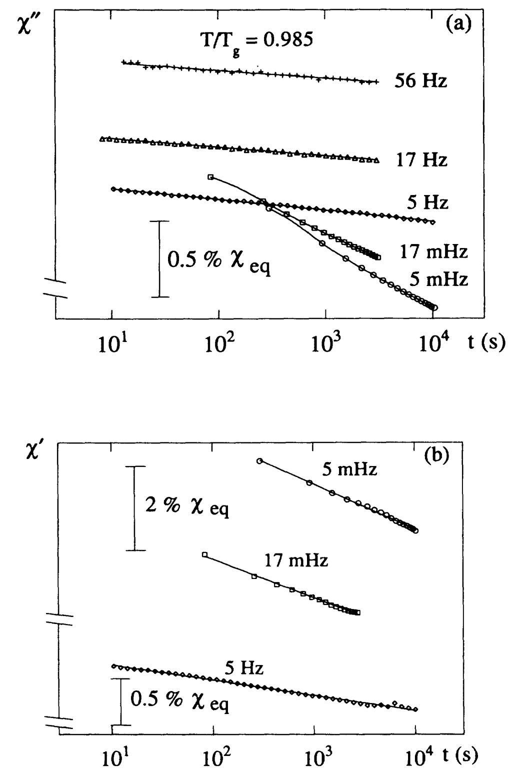

FIG. 2. Decay of $\chi''(\omega)$ and $\chi'(\omega)$ at $T=0.985T_g$ for different frequencies. The effective cooling rate was 5×10^{-5} K/s. $T_g=20.9$ K. (a) $\chi''(\omega)$ and (b) $\chi'(\omega)$.

dependent effect is almost an order of magnitude smaller as compared to the decay found at the lower temperatures. For the lowest frequencies investigated ($\omega/2\pi\leq0.05$ Hz) this time dependence is close to logarithmic while for higher frequencies our results indicate that the equilibrium value of $\chi''(\omega)$ is obtained while monitoring its time dependence. A similar time dependence, as shown in Fig. 1, was observed using different cooling rates in the interval 2.5×10^{-5}–5×10^{-4} K/s (the cooling rate in Fig. 1 was $\sim5\times10^{-5}$ K/s).

As can be seen in Fig. 1, the decay is larger for $\omega/2\pi=5\times10^{-3}$ Hz [Fig. 1(a)] than what it is for $\omega/2\pi=5$ Hz [Fig. 1(b)]. Figure 2(a) shows the time dependence of $\chi''(\omega)$ at $T/T_g=0.985$ for different frequencies. In the investigated frequency range, the frequency dependence of the decay rate $\partial[\chi''(\omega)]/\partial\ln(t)$ can be fitted to a power law $\sim\omega^{-\alpha}$ with $\alpha\approx0.25$. A similar frequency dependence of $\partial[\chi''(\omega)]/\partial\ln(t)$ was found for all temperatures $T<T_g$. Figure 2(b) shows the time dependence of $\chi'(\omega)$ for different frequencies. The magnitude of the decay of $\chi'(\omega)$, expressed in units of χ_{eq}, is ~3 times as large as for $\chi''(\omega)$. We will in the following limit ourselves to discussing the $\chi''(\omega)$ results, partly because of a better long-time stability for this component of the ac susceptibility. It should be noted, however, that most of the things said in the following about the time dependence of $\chi''(\omega)$ also apply to the in-phase component of the ac susceptibility.

Recently, an alternative approach to dynamic scaling was proposed by Geschwind, Huse, and Devlin (GHD),[8] where $[\chi''(\omega)T]\omega^{-\beta/z\nu}$ is scaled vs $(T-T_g)/\omega^{1/z\nu}$. Using this approach it is possible to extend the scaling plot to temperatures below T_g. However, as already noticed by GHD, deviations from scaling are found at temperatures near and below T_g. The departures from scaling for $\omega/2\pi=1$ Hz at low temperatures were found to be $\sim15\%$. The time dependence of $\chi''(\omega)$ discussed above clearly shows why the scaling performance will be worse in this temperature regime. The decay of $\chi''(\omega)$ at low temperatures is $\sim25\%$ for $\omega/2\pi=5$ mHz (measured over 1.5 decades of time) and $\sim8\%$ for $\omega/2\pi=5$ Hz (measured over 3 decades of time). Furthermore, since the decay is close to logarithmic in the experimental time window, there is no obvious way to extrapolate the time dependence to longer times to determine the equilibrium values of $\chi''(\omega)$. While, in principle, it should be possible to scale the ac susceptibility at higher temperatures [temperatures higher than the peak in $\chi''(\omega)$ vs T], one has to be careful (especially at low frequencies) to equilibrate the spin glass for a sufficiently long time in order to measure equilibrium values of $\chi''(\omega)$.

The spin-glass transition is frequently analyzed in terms of critical slowing down. In this kind of analysis, the critical correlation time τ is related to the critical properties of the correlation length $\xi\sim|T-T_g|^{-\nu}$ via $\tau\sim\xi^z\sim|T-T_g|^{-z\nu}$. Several criteria have been used to deduce the $\tau(T)$ dependence from ac susceptibility measurements. One often used method is to choose the inflection point temperature of the $\chi''(\omega)$ curves and set τ equal to $1/\omega$. In view of the observed time dependence of $\chi''(\omega)$ at temperatures close to the inflection point temperature (which will be difficult to avoid for very low frequencies), a better choice will be to use the criterion $\lim_{\omega\to 0}\chi''(\omega)/\chi'(\omega)=\omega\tau_{av}$, where τ_{av} corresponds to the average relaxation time.[11,12] If it is difficult to establish the zero-frequency limit in the ac susceptibility measurements, it might even be advisable to use a criterion $\phi=\chi''(\omega)/\chi'(\omega)=\omega\tau$ with ϕ small, typically $\phi\leq 0.01$.[13]

Several models have been proposed to describe the nonequilibrium character of the spin-glass phase.[14–16] The models can be divided into (i) hierarchical phase-space models where the dynamics below the transition temperature is coarse grained into a Markov process on a tree structure[16] and (ii) real-space "droplet" models where the fundamental nonequilibrium process is the growth of spin-glass-ordered domains.[14,15] The hierarchical phase-space picture has been able to reproduce some of the experimental findings, e.g., a maximum in the relaxation rate at an observation time $\sim t_w$ [cf. Figs. 3 and 5(a)], but give no predictions as to what to expect in time-dependent ac susceptibility measurements. The models of Fisher and Huse[14] (FH) and Koper and Hilhorst[15] (KH) have much in common, even though some differences exist. FH assume thermally activated dynamics and a logarithmic growth law of the domains while KH make the assumption that the domains grow with time according to a power law. Furthermore, to describe the long-time behavior of the magnetic relaxation, FH find a power-law relationship between the magnetization and the domain size while KH have an exponential relationship. A power-law relationship is more consistent with results from time-dependent magnetization measurements[17] and we will therefore, in the following, focus on the droplet model by Fisher and Huse.[14] In this model, at fixed temperature below the spin-glass transition temperature, only two pure states exist related by a global spin reversal. The dominant excitations are large-scale coherent spin-glass fluctuations, droplets, which occur on a length scale L. The dynamics of these droplets is governed by thermal activation over barriers of characteristic magnitude $\Delta(T)L^{\Psi}$, where $\Delta(T)$ sets the free-energy scale of the barriers and Ψ is the barrier exponent. After a temperature quench to a temperature below T_g, the spin system will be out of equilibrium and the spin configuration will be divided into spin-glass domains. The spin system will lower its energy by the growth of larger and larger domains, a process which is logarithmically slow in time, $R(t)\sim[T\ln(t/\tau_0)/\Delta(T)]^{1/\Psi}$ (at $T\ll T_g$, τ_0 is a microscopic spin flip time). If the system is equilibrated for a waiting time t_w and then probed in a ZFC magnetization measurement, the response to the field application can be divided into two different time regimes. At observation times $t_{obs}\ll t_w$ ($t_{obs}=t-t_w$), the magnetization grows through the polarization of active droplets of size

$$L(t_{obs})\sim[T\ln(t_{obs}/\tau_0)/\Delta(T)]^{1/\Psi}\ .$$

Since the droplet size in this case is much smaller than the characteristic domain size $R(t_w)$, the droplet excitations will, apart from a small fraction close to domain walls, be those of the pure states. In this time regime, quasiequilibrium dynamics will be probed. For observation times $t_{obs}\gg t_w$, the magnetization cannot grow through the polarization of droplets, since the droplets are limited in size by the size of the domains. Instead, the magnetization will increase with the growth of domains and the dynamics can be described as nonequilibrium dynamics. At observation times $t_{obs}\sim t_w$, $L(t_{obs})\sim R(t_w)$, there is a crossover from quasiequilibrium to nonequilibrium dynamics. In experiments, this crossover is visible as a maximum in $\partial M/\partial\ln(t_{obs})$ [cf. Figs. 3 and 5(a)].

Results from temperature cycling experiments can, in the droplet model, be explained using the overlap correlation function (defined as the overlap between the equilibrium states at two temperatures T and $T+\Delta$)

$$\Xi(i,j,T,T+\Delta T)=\overline{\langle S_iS_j\rangle_T\langle S_iS_j\rangle_{T+\Delta T}}\ ,$$

which will decay with increasing distance $r_{ij}=|\mathbf{r}_i-\mathbf{r}_j|$ with the characteristic length scale $l_{\Delta T}(T)$. This correlation function tells us that the positions of large-scale active droplets will differ from one temperature to another and thus that the equilibrium states at temperatures T and $T+\Delta T$ will differ at length scales larger than $l_{\Delta T}(T)$. $l_{\Delta T}(T)$ will, of course, decrease with increasing ΔT. Figure 3 shows results from ZFC relaxation experiments, $\partial M/\partial\ln(t_{obs})$ vs $\ln(t_{obs})$, where the sample has been aged 3×10^3 s and immediately prior to the field application subjected to a temperature cycling ΔT. On the one hand, for ΔT sufficiently small, $l_{\Delta T}>R(t_w=3\times 10^3$ s), the spin-glass domains continue to grow as if no temperature cycling has been performed and a maximum in the relaxation rate is expected at $t_{obs}\sim t_w$. On the other hand, if

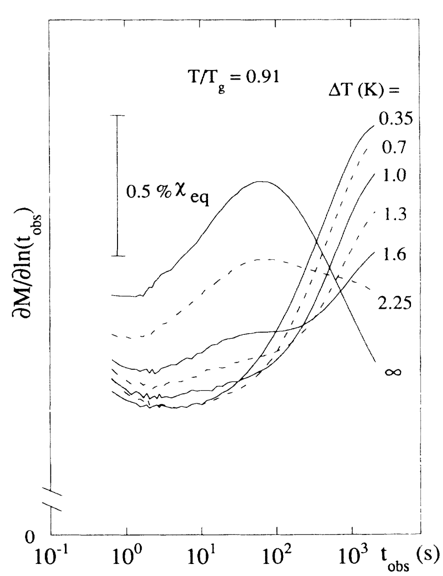

FIG. 3. $\partial M/\partial\ln(t_{obs})$ vs (t_{obs}) at $T/T_g=0.91$. The sample has been aged 3×10^3 s at T and afterwards subjected to a temperature cycling ΔT immediately prior to the field application. $T_g=20.9$ K.

$l_{\Delta T} < R(t_w = 3\times10^3$ s), some domains of size $R(t_w = 3\times10^3$ s) will break up and continue to grow from the size $l_{\Delta T}$. The ZFC relaxation rate is thus expected to show two maxima, one corresponding to old domains of size $R(t_w = 3\times10^3$s) and another corresponding to young domains. For ΔT sufficiently large, the spin glass will be completely reborn in the sense that the spin-glass correlations, established during the waiting time t_w at T, will be fully destroyed. From Fig. 3 it is seen that $l_{\Delta T} \sim R(t_w = 3\times10^3$ s) for a temperature cycling $\Delta T = 1$ K. Comparing the relaxation rate curve for $\Delta T = 0.7$ K with the corresponding curve for $\Delta T = 0.35$ K, there is a small shift of the maximum in the relaxation rate curve for $\Delta T = 0.7$ K to longer-time scales. This effect is not related to $l_{\Delta T}$, instead it is an effect of the higher growth rate for domains at $T+\Delta T$.

The consequences of an overlap length scale should also be seen in the time decay of $\chi''(\omega)$ [and $\chi'(\omega)$]. In temperature cycling experiments one would expect an increased magnitude of $\chi''(\omega)$ if $l_{\Delta T} < R(t_w)$. Figure 4 shows results from measurements of the time dependence of $\chi''(\omega)$ where the sample has been quenched to $T/T_g = 0.91$ and the decay of $\chi''(\omega)$ was registered for 3×10^3 s. The sample was then subjected to a temperature cycling and the decay registered for another 10^4 s.

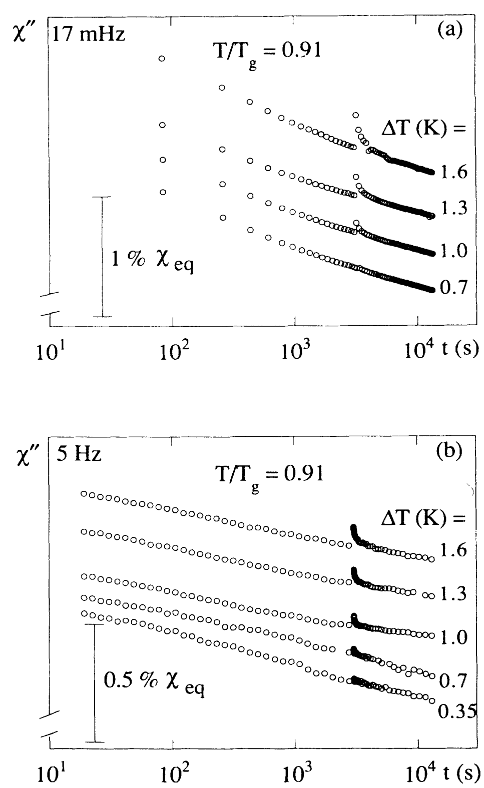

FIG. 4. $\chi''(\omega)$ vs t at $T/T_g = 0.91$. The time dependence was registered for 3×10^3s after which the sample was subjected to a temperature cycling ΔT and the time decay was registered for another 10^4s. The different curves have been displaced along the y axis. $T_g = 20.9$ K. (a) $\omega/2\pi = 0.017$ Hz and (b) $\omega/2\pi = 5$ Hz.

As expected, the decay of $\chi''(\omega)$ remains unaffected for $\Delta T < 1$ K, while the effect of a finite $l_{\Delta T}$ is clearly seen for larger ΔT. For the higher frequency [$\omega/2\pi = 5$ Hz, Fig. 4(b)] there is an indication that the effect of a finite overlap length is seen already at $\Delta T = 0.7$ K. We have also performed temperature cycling experiments at the temperature $T/T_g = 0.985$ where $T+\Delta T$ (for $\Delta T > 0.3$K) will correspond to a temperature above T_g. The naive guess in this case would be that the domains grown at $T/T_g = 0.985$ will be fully destroyed when increasing the temperature above T_g. Our results indicate otherwise, however. The spin system is not completely reborn until $(T+\Delta T)/T_g \geq 1.05$.

All of our temperature cycling experiments have been performed in the vicinity of the spin-glass temperature, with $T+\Delta T \to T_g$ or even exceeding the spin-glass temperature. According to the droplet theory, as $T+\Delta T \to T_g$ the overlap length becomes of order the critical correlation length ξ, but never less. However, recent theoretical progress[18] has revealed a new critical exponent ζ_c which appears in the scaling laws for $l_{\Delta T}$ close to T_g. The implication of this exponent is that $l_{\Delta T}$ becomes smaller than ξ in the critical region and that quasi-droplets of size smaller than ξ will exist close to T_g. Our temperature cycling experiments seem to support this view since for the data shown in Fig. 3 there is a gradual change of the relaxation rate curves with increasing ΔT (towards the curve corresponding to a completely reborn spin-glass state). More extensive temperature cycling experiments are, however, needed in order to gain a better understanding of the behavior of $l_{\Delta T}$ in the critical region.

FH also discuss the approach to equilibrium of $\chi''(\omega;t_w)$ in the limit $L_\omega \ll R(t_w)$, where $L_\omega \sim [T|\ln(\omega\tau_0)|/\Delta(T)]^{1/\Psi}$ is the probing length scale in an ac susceptibility measurement. Probing length scale means that the ac susceptibility will be determined by the dynamics of excitations (droplets) of the length scale L_ω. The approach to equilibrium will be concerned with the density of frozen-in domain walls since a region of size L_ω passing through a domain wall will have a reduced excitation free energy. FH predict that $\chi''(\omega;t_w)$ will approach equilibrium as $|\ln(\omega\tau_0)|/\ln(t_w/\tau_0) \to 0$. Even though our measurements might not fulfill the condition $L_\omega \ll R(t_w)$,[19] we would like to emphasize the results of FH; (i) the decay towards equilibrium is logarithmically slow in time and (ii) the corrections to the equilibrium values of $\chi''(\omega)$ decay slowly with increasing frequency.

It is interesting to investigate whether the observed waiting time dependence of $\chi''(\omega)$ is consistent with results from time-dependent magnetization measurements. In time-dependent ZFC magnetization measurements, the sample is quenched from an equilibrium state at high temperature to a nonequilibrium state at low temperature. The sample is then equilibrated for a waiting time t_w, after which the magnetization is probed by applying a small magnetic field. As discussed above, $\chi''(\omega)$ relates to the relaxation rate of the ZFC experiment through $\chi''(\omega;t_w) \propto [\partial M/\partial \ln(t_{obs})]_{t_w}$ with $t_{obs} \approx 1/\omega$. In Fig. 5(a) the ZFC relaxation rate is shown at $T/T_g = 0.95$ for

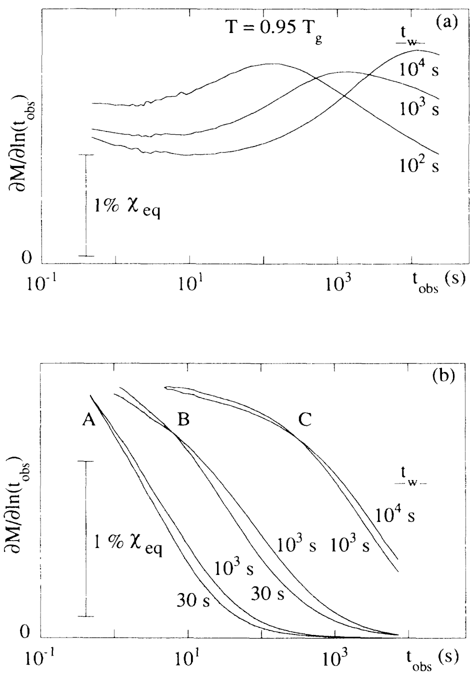

FIG. 5. $\partial M/\partial \ln(t_{obs})$ vs (t_{obs}) for different waiting times t_w. $T_g=20.9$ K. (a) $T/T_g=0.95$ and (b) A, B, and C correspond to $T/T_g=1.045$, 1.035, and 1.025, respectively.

different waiting times. Since in ac susceptibility measurements $1/\omega < t_w$, this figure can be used to discuss the expected time dependence of $\chi''(\omega;t_w)$ for observation times less than 10^2s ($\omega/2\pi > 1.7$ mHz). At a constant observation time t_{obs}, the ZFC relaxation rate decreases with increasing t_w, in accordance with the observed waiting time dependence of $\chi''(\omega)$. Not only qualitatively but also quantitatively, the agreement is good. When increasing t_w from 10^2 to 10^4s, the change of $[\partial M/\partial \ln(t_{obs})]_{t_w}$ is $\sim 0.6\%$ χ_{eq} for $t_{obs}=10$s ($\omega/2\pi=17$ mHz). This can be compared to the decay of $\chi''(\omega)$ shown in Fig. 4(a) (using the $\Delta T=0.7$ K curve since the decay in this case is unaffected by the temperature cycling). The magnitude of this decay is $\sim 0.8\%$ χ_{eq}, which is close to what can be calculated using Eq. (5). Figure 5(b) shows the relaxation rate at different temperatures above T_g. At each temperature, the ZFC relaxation has been measured for two different waiting times. The characteristics of the relaxation rate curves can be described as follows: At short observation times, the relaxation rate curve corresponding to a longer waiting time has a lower relaxation rate. The two curves cross at an observation time t^* close to the "knee" in the $\partial M/\partial \ln(t_{obs})$ vs $\log_{10}(t_{obs})$ curve and for observation times $t_{obs} > t^*$, the curve corresponding to the longer waiting time has a higher relaxation rate; $[\partial M/\partial \ln(t_{obs})]_{t_w}$ increases with increasing t_w. When increasing t_w from 30 to 10^3s, the change of $[\partial M/\partial \ln(t_{obs})]_{t_w}$ at $T/T_g=1.045$ is $\sim 0.1\%$ χ_{eq} for $t_{obs}=30$s ($\omega/2\pi=5$ mHz). To make a quantitative comparison with the observed increase of $\chi''(\omega)$ with time, the data shown in Fig. 1(a) will be used. This comparison is not strictly correct since the results displayed in this figure were obtained in a measurement where the sample temperature was stepwise decreased. This implies that the observed decay of $\chi''(\omega)$ at low temperatures will depend on the equilibration of the sample performed at higher temperatures as well as on the overlap length $l_{\Delta T}(T)$. However, at $T/T_g=1.045$ the thermal history of the sample is less important since the sample was in an equilibrium state at the higher temperature. The magnitude of the increase of $\chi''(\omega)$ with time (measured over 1.5 decade of time) is $\sim 0.1\%\chi_{eq}$, in reasonable agreement with the prediction of Eq. (5).

In summary, time-dependent ac susceptibility measurements show that the spin glass is in a nonequilibrium state at temperatures close to and below the spin-glass temperature. The corrections to the equilibrium values of $\chi''(\omega)$ [and $\chi'(\omega)$] decay very slowly with time as well as with frequency, which has implications for dynamic scaling analyses. Our results clearly show why deviations from scaling are expected in this temperature range. The existence of an overlap length scale has also been verified through time-dependent ac susceptibility measurements. Using linear response relations, the observed time dependence of $\chi''(\omega)$ is shown to be consistent with results from time-dependent magnetization measurements.

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[1]M. Alba, M. Ocio, and J. Hammann, Europhys. Lett. **2**, 45 (1986).

[2]P. Svedlindh, P. Granberg, P. Nordblad, L. Lundgren, and H. S. Chen, Phys. Rev. B **35**, 268 (1987).

[3]L. Lundgren, P. Nordblad, and L. Sandlund, Europhys. Lett. **1**, 529 (1986).

[4]L. Lundgren, P. Svedlindh, and O. Beckman, J. Magn. Magn. Mater. **31-34**, 1349 (1983).

[5]Ph. Refregier, E. Vincent, J. Hammann, and M. Ocio, J. Phys. (Paris) **48**, 1533 (1987).

[6]Ph. Refregier, M. Ocio, J. Hammann, and E. Vincent, J. Appl. Phys. **63**, 4343 (1988).

[7]A. G. Schins, A. F. M. Arts, H. W. de Wijn, L. Leylekian, E. Vincent, C. Paulsen, and J. Hammann, J. Appl. Phys. **69**, 5237 (1991).

[8]S. Geschwind, D. A. Huse, and G. E. Devlin, Phys. Rev. B **41**, 4854 (1990).

[9]A. Ito, H. Aruga, E. Torikai, M. Kikuchi, Y. Syono, and H. Takei, Phys. Rev. Lett. **57**, 483 (1986).

[10]P. Svedlindh, K. Gunnarsson, P. Nordblad, L. Lundgren, A. Ito, and H. Aruga, J. Magn. Magn. Mater. **71**, 22 (1987).

[11]A. T. Ogielski, Phys. Rev. B **32**, 7384 (1985).

[12]K. Gunnarsson, P. Svedlindh, P. Nordblad, L. Lundgren, H. Aruga, and A. Ito, Phys. Rev. Lett. **61**, 754 (1988).

[13]D. Bertrand, J. P. Redoulès, J. Ferré, J. Pommier, and J. Souletie, Europhys. Lett. **5**, 271 (1988).

[14]D. S. Fisher and D. A. Huse, Phys. Rev. B **38**, 373 (1988); **38**, 386 (1988).
[15]G. J. M. Koper and H. J. Hilhorst, J. Phys. (Paris) **49**, 429 (1988).
[16]P. Sibani and K. H. Hoffmann, Phys. Rev. Lett. **63**, 2853 (1989); K. H. Hoffmann and P. Sibani, Z. Phys. B **80**, (1990) 429.
[17]P. Nordblad, P. Granberg, P. Svedlindh, L. Sandlund, and L. Lundgren, Nucl. Phys. B (Proc. Suppl.) **5A**, 86 (1988).
[18]M. Nifle and H. J. Hilhorst, Phys. Rev. Lett. **68**, 2992 (1992).
[19]For temperatures not too close to the spin-glass transition temperature τ_0 is a microscopic time which implies that the condition $L_\omega \ll R(t_w)$ will be difficult to attain in ac susceptibility measuremets. For temperatures $T \rightarrow T_g^-$, FH suggest that the critical correlation time should be used in place of τ_0 in the expressions for L_ω and $R(t_w)$ which implies that $L_\omega \ll R(t_w)$ will be easier to attain experimentally.