Observation of a time-dependent spatial correlation length in a metallic spin glass

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The aging process in a Cu (10 at.% Mn) spin glass has been investigated by means of zero-field-cooled magnetization measurements. A small persistent temperature shift immediately prior to the field application is found to have a very specific influence on the measured relaxation of the magnetization. It is inferred that the aging process may be interpreted in terms of a spatial correlation length that increases with the age of the system. The correlation length also governs the time scales where a crossover from equilibrium dynamics to nonequilibrium dynamics occurs.

A profound signature of spin glasses is aging,[1] which implies nonequilibrium dynamics. When a spin glass is cooled in zero field to a temperature below the spin-glass freezing temperature the spin configuration initially attains an energetically unfavorable state; a state with random spin configuration. The initial state changes towards an energetically most favorable spin configuration.[2] A new theory treating the nonequilibrium dynamics of spin glasses has recently been proposed by Koper and Hilhorst (KH).[3] KH use some ideas that also appear in a paper by Bray and Moore[4] where a description based on the droplet model[5] is presented. In the approach by KH a spin-glass system possesses domains within which the spins are correlated. At a constant temperature T the domains grow without limit at a rate determined by T. It turns out that the response function is a functional of the characteristic domain size leading to a functional form of the magnetization that is dependent on the age of the spin-glass system.

Experimentally the aging process can be studied by zero-field-cooled (ZFC) magnetization measurements. In such measurements the sample is cooled in zero field to a temperature T_m. After a wait time t_w at T_m a magnetic field is applied and the relaxation of the magnetization versus the logarithm of time is recorded. A general feature of the relaxation curve is the existence of an inflection point at an observation time of the order of the wait time. In the regime of linear response the application of a magnetic field can be considered as a weak perturbation on the system. Thus, the aging process proceeds unaffected in spite of the applied magnetic field.

In this Brief Report we report measurements where the sample is cooled according to the ZFC procedure and in addition is subjected to a *small temperature shift* immediately prior to the field application. It is found that the temperature shift has a very specific influence on the subsequent relaxation. Our experimental results indicate that the aging process in spin glasses is associated with a growing spatial correlation length ξ which determines a crossover length scale where the spin-glass dynamics change from equilibrium dynamics to nonequilibrium dynamics. For sufficiently small temperature shifts (positive or negative) the correlation length is unaffected, but a dramatic change of the growth rate with temperature is observed. Furthermore, it is found that a relaxation curve, measured at a specific temperature, is only dependent on the achieved correlation length.

The experiments were performed in a SQUID magnetometer on a Cu (10 at.% Mn) metallic spin glass. Initially, conventional ZFC magnetization measurements were performed. The sample is cooled in zero field from a

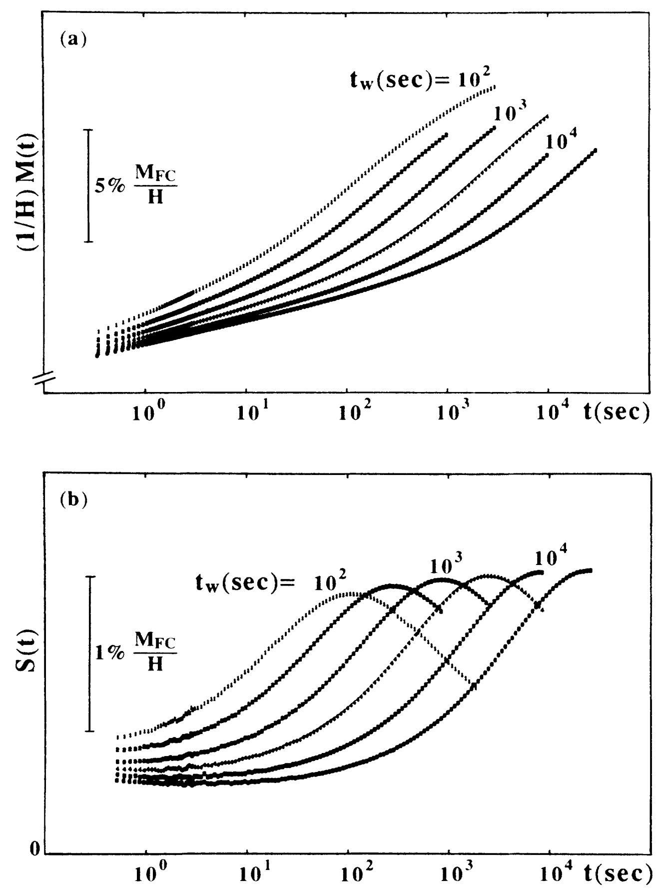

FIG. 1. Zero-field-cooled susceptibility $(1/H)M(t)$ and the corresponding relaxation rate $S(t)=(1/H)\partial M/\partial \ln(t)$ at different wait times $t_w=10^2$, 3×10^2, 10^3, 3×10^3, 10^4, and 3×10^4 sec, plotted vs $\log_{10}(t)$. $T_m=41.2$ K, $T_g=45.3$ K ($T_m/T_g=0.91$), $H=0.8$ G. (a) $(1/H)M(t)$, 5% of $(1/H)M_{FC}$ indicated. (b) $S(t)$, 1% of $(1/H)M_{FC}$ indicated.

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reference temperature $T_{ref}=46.1$ K, above the spin-glass temperature $T_g=45.2$ K ($T_{ref}/T_g=1.02$) to the measurement temperature $T_m=41.2$ K ($T_m/T_g=0.91$). At T_m the sample is allowed to age for t_w seconds. The magnetic field $H=0.8$ G is applied and the magnetization as a function of time $M(t)$ is recorded in the time interval 0.3–3×10^4 sec. The sample is then heated to T_{ref} and a reference value of the magnetization is measured.

Figure 1 shows $M(t)$ and the corresponding relaxation rate $S(t)=(1/H)\,\partial M/\partial \ln t$, measured at T_m for different t_w. The $M(t)$ curves are drawn relative to the magnetization value at T_{ref}. A significant feature is the inflection point in M vs $\log_{10}(t)$ and a corresponding maximum in S vs $\log_{10}(t)$ at an observation time around t_w. The wavelike shape of $S(t)$, which with increasing wait time shifts towards longer observation times, reflects the influence of the aging process on the relaxation in the spin-glass system. A similar behavior is found in semiconducting[6] as well as insulating[7] spin-glass systems and is a general feature of spin glasses.

To further investigate the nature of the aging process, the system is subjected to a *small positive temperature shift* immediately prior to the field application. The sample has thus been aged at a temperature slightly lower than T_m. At the same wait time $t_w=10^4$ sec, five measurements were performed for different temperature shifts $\Delta T=0$, 0.15, 0.30, 0.45, 0.60 K. All curves are measured at one and the same temperature $T_m=0.91T_g$. Figure 2 shows M vs $\log_{10}(t)$ and the corresponding relaxation rate S vs $\log_{10}(t)$. The salient feature is that the inflection point in $M(t)$ and the corresponding maximum in $S(t)$ occur at times *shorter* than t_w. With successively larger ΔT the location of the inflection point gradually shifts towards shorter times. The location of the inflection point in $M(t)$ [or equivalently the maximum in $S(t)$] is denoted the apparent wait time $t_{w_{app}}(\Delta T)$. The curves in Fig. 2 show similar features as the curves in Fig. 1. It is of particular interest to note that two different curves with inflexion points occurring around the same observation time are virtually identical. This is illustrated in Fig. 3 where a conventional ZFC curve with $t_w=3\times10^3$ sec is plotted together with a curve with $\Delta T=0.15$ K and

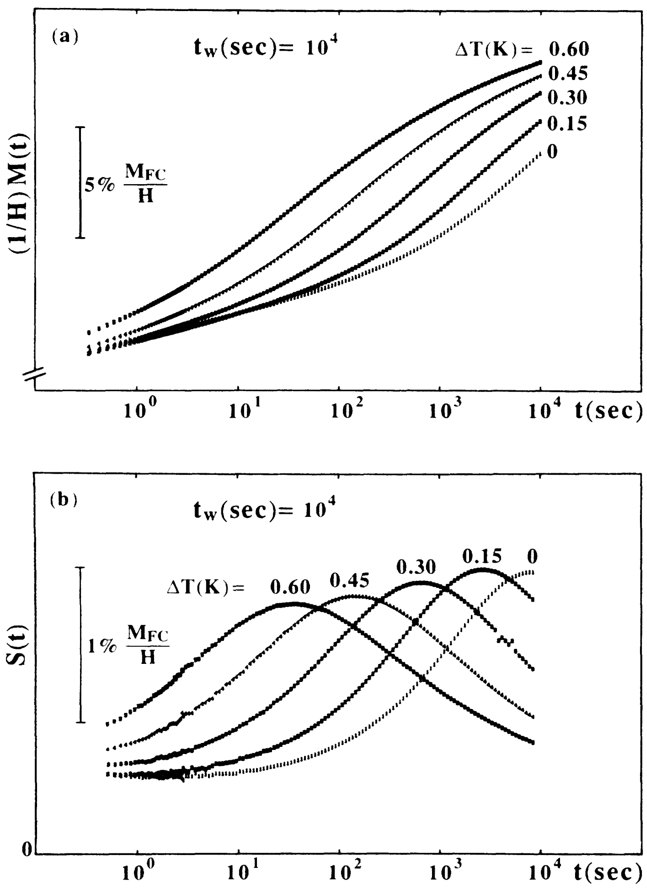

FIG. 2. Zero-field-cooled susceptibility $(1/H)M(t)$ and the corresponding relaxation rate $S(t)=(1/H)\partial M/\partial \ln(t)$ plotted vs $\log_{10}(t)$. The sample has been aged for $t_w=10^4$ sec at the temperature $T_m-\Delta T$. The different curves refer to different temperature shifts ΔT. Immediately prior to the field application the temperature is raised to T_m. $T_m/T_g=0.91$, $H=0.8$ G. (a) $(1/H)M(t)$, 5% of $(1/H)M_{FC}$ indicated. (b) $S(t)$, 1% of $(1/H)M_{FC}$ indicated.

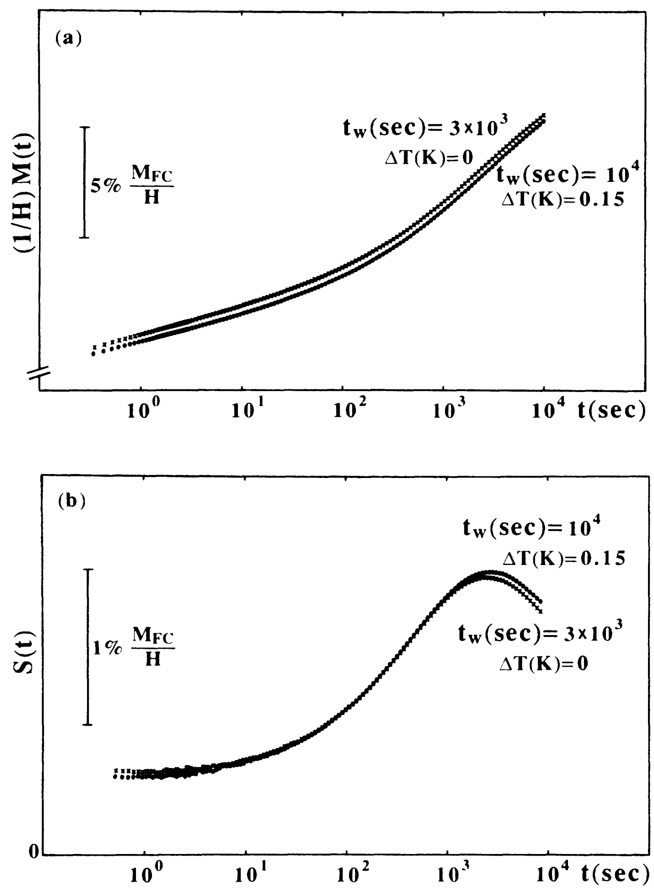

FIG. 3. Zero-field-cooled susceptibility $(1/H)M(t)$ and the corresponding relaxation rate $S(t)=(1/H)\partial M/\partial \ln(t)$ plotted vs $\log_{10}(t)$. The curves refer to $t_w=3\times10^3$ sec, $\Delta T=0$ K and $t_w=10^4$ sec, $\Delta T=0.15$ K, respectively. $T_m/T_g=0.91$, $H=0.8$ G. (a) $(1/H)M(t)$, 5% of $(1/H)M_{FC}$ indicated. (b) $S(t)$, 1% of $(1/H)M_{FC}$ indicated.

$t_w = 10^4$ sec. Both experimental curves yield an apparent wait time $t_{w_{app}} = 3\times10^3$ sec.

The apparent wait time $t_{w_{app}}(\Delta T)$, extracted from the maximum of $S(t)$, characterizes an apparent age of the spin-glass system. To investigate any wait-time dependence on $t_{w_{app}}/t_w$, measurements were carried out for different t_w up to 3×10^4 sec. In Fig. 4 the logarithm of the reduced apparent wait time $\log_{10}(t_{w_{app}}/t_w)$ versus temperature shift ΔT is plotted. In this semilogarithmic plot there is a linear dependence of the reduced apparent wait time on ΔT with an increasing slope with t_w. At ΔT larger than 0.8 K the effect saturates. This is due to the finite heating rate to T_m which results in a smallest attainable apparent wait time in the experiments.

A positive temperature shift yields a relaxation curve characterized by an apparent wait time $t_{w_{app}} < t_w$. To extend the measurements we have explored the response after a negative ΔT. Hence, the sample is aged a wait time t_w at a temperature above the measurement temperature and is then cooled to T_m where the magnetic field is applied. In Fig. 5 we have plotted the relaxation of the magnetization and the corresponding relaxation rate for $\Delta T = -0.15$ K and $t_w = 1000$ sec at $T_m = 0.91T_g$. Also drawn in this figure are the corresponding curves at $T_m = 0.91T_g$ and $t_w = 1000$ sec for $\Delta T = 0.15$ K and $\Delta T = 0$. As seen in Fig. 5 the curve obtained after a negative temperature shift yields an apparent wait time which is *longer* than t_w.

In conclusion we promote an interpretation of the spin-glass dynamics in terms of a time-dependent spatial correlation length ξ for the spin system. Figure 6 shows a tentative plot of ξ vs time (time=age of the system) in a semilogarithmic diagram at three temperatures $T_m+\Delta T$, T_m, and $T_m-\Delta T$. We also assume that a measured relaxation curve probes the dynamics of the spin system on a length scale $l(t)$. $l(t)$ increases with observation time in a similar manner as ξ evolves with the increasing age of the system at the same temperature. Using Fig. 6 this interpretation yields the following results and conclusions.

(i) A relaxation measurement on a system that has been aged a wait time t_{w2} at the temperature T_m, probes at observation times $t \ll t_{w2}$ a system with a correlation length

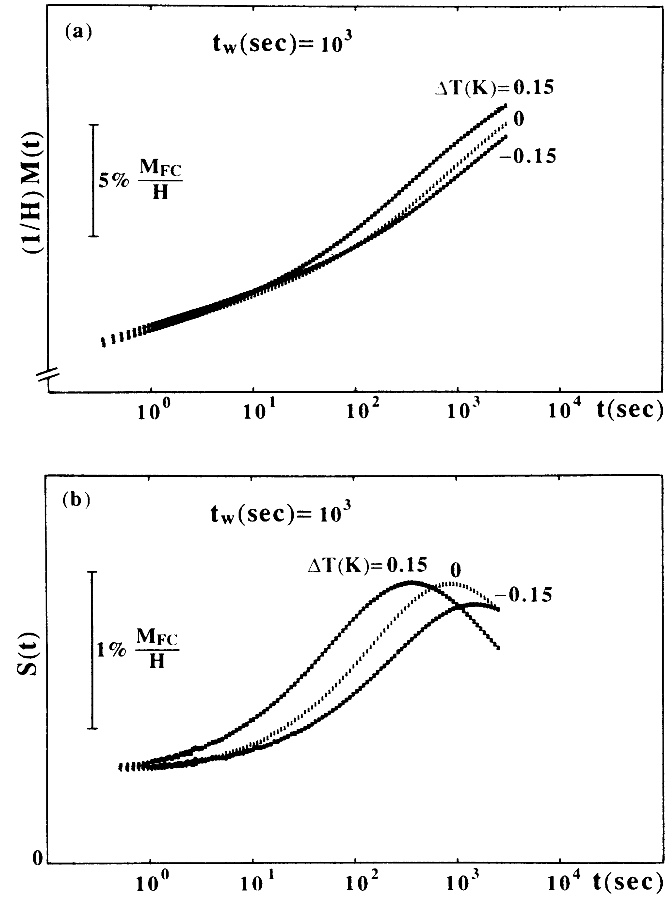

FIG. 5. Zero-field-cooled susceptibility $(1/H)M(t)$ and the corresponding relaxation rate $S(t)=(1/H)\partial M/\partial \ln(t)$ at $t_w = 10^3$ sec plotted vs $\log_{10}(t)$. The sample has been aged below ($\Delta T = 0.15$ K) and above ($\Delta T = -0.15$ K) the measurement temperature. Also plotted is a conventional ($\Delta T = 0$ K) ZFC curve. $T_m/T_g = 0.91$, $H = 0.8$ G. (a) $(1/H)M(t)$, 5% of $(1/H)M_{FC}$ indicated. (b) $S(t)$, 1% of $(1/H)M_{FC}$ indicated.

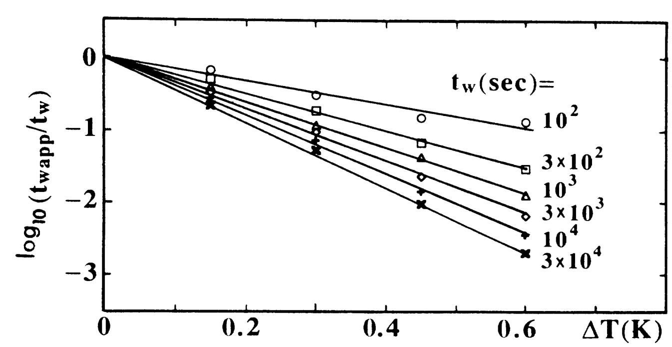

FIG. 4. Logarithm of the reduced apparent wait time $\log_{10}(t_{w_{app}}/t_w)$ plotted vs temperature shift ΔT. The data are extracted from the location of the maximum of $S(t)$ at the respective t_w and ΔT. All curves from where the data are extracted are measured at $T_m/T_g = 0.91$, $H = 0.8$ G.

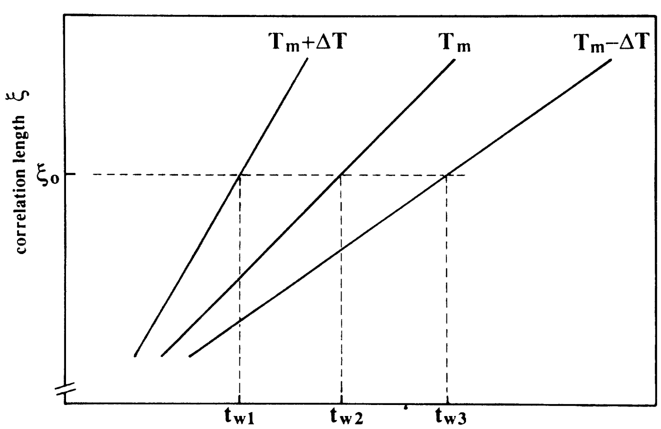

FIG. 6. Correlation length ξ vs age (logarithmic scale) at different temperatures $T_m-\Delta T$, T_m, and $T_m+\Delta T$. The different wait times t_{w1}, t_{w2}, and t_{w3} yield the same value ξ_0 of the correlation length.

of the order of ξ_0. Thus, at these short observation times the relaxation of the magnetiztion reflects the dynamics in a correlated spin system, or in other words, equilibrium dynamics are probed. When the observation time becomes of the order of t_{w2}, the length scale $l(t)$ and the correlation length have reached the same order of magnitude and thus a correlating spin system is probed; nonequilibrium dynamics are observed. The actual observation of the crossover from equilibrium dynamics to nonequilibrium dynamics is an inflection point in M vs $\log_{10}(t)$ and a corresponding maximum in S vs $\log_{10}(t)$ at an observation time $t \approx t_{w2}$.

(ii) A system that is subjected to a small positive temperature shift from $T_m - \Delta T$ to T_m at an age t_{w3} preserves the achieved correlation length ξ_0 and appears as a system of age t_{w2} at T_m. A relaxation curve measured immediately after the temperature shift thus yields an inflection point in M vs $\log_{10}(t)$ at an observation time $t \approx t_{w2}$.

(iii) A system that is subjected to a small negative temperature shift from $T_m + \Delta T$ to T_m at an age t_{w1} likewise preserves the achieved correlation length ξ_0 and thus appears as a system of age t_{w2} at T_m. A relaxation curve measured immediately after the temperature shift thus yields an inflection point in M vs $\log_{10}(t)$ at observation times $t \approx t_{w2}$.

(iv) For sufficiently small temperature shifts during the aging process, the measured relaxation curve at constant temperature is *only governed* by the magnitude of the spatial correlation length. A certain correlation length ξ_0 obtained at $T_m + \Delta T$, T_m, or $T_m - \Delta T$ cannot be distinguished from subsequent relaxation measurements at T_m. Such measurements reveal closely identical relaxation curves with inflection points in M vs $\log_{10}(t)$ at observation times $t \approx t_{w2}$. This is nicely illustrated in Fig. 3.

(v) The path towards an equilibrium spin configuration at a temperature T_m is closely similar to the path taken at a slightly different temperature. The main difference is a pronounced change with temperature of the growth rate of the spatial correlation length.

In recent papers[8] it has been shown that the aging process is insensitive to small temporal temperature variations around the measurement temperature. However, the experimental observations presented here show that a *persistent temperature shift* has a dramatic influence on the aging process.

Note added. After having submitted this manuscript we received a paper prior to publication by D. S. Fisher and D. A. Huse.[9] In this paper, within the droplet scaling theory, they ascribe the fundamental long-time nonequilibrium process in the spin-glass phase to the thermally activated growth of spin-glass-ordered domains. The physical interpretation of our paper for the observed nonequilibrium phenomena in the spin-glass phase can be readily incorporated in the more extensive model for the nonequilibrium behavior suggested by Fisher and Huse.

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