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Time decay of the remanent magnetization in a Cu*Mn* spin glass

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The functional form of the relaxation of the thermoremanent magnetization of a Cu*Mn* spin glass has been investigated. A remarkable influence of the aging process of the spin-glass state, which can give an apparent stretched exponential form of the total relaxation, is demonstrated. The spin-glass relaxation can be described by a nearly logarithmic function superimposed by an additional relaxation, due to the aging process. The superimposed relaxation can be described by a stretched exponential function: $M_a = M_{a0}(T,t_w)\exp[-(t/t_w)^{1-n}]$, where t_w is the wait time at constant temperature in the field-cooled state.

It has been alleged by Chamberlin, Mozurkewich, and Orbach[1] (henceforth referred to as CMO), that the time dependence of the thermoremanent magnetization (M_R) of spin glasses is accurately characterized by a stretched exponential of the form

$$M_R = M_0\exp[-(t/t_p)^{1-n}] \quad . \tag{1}$$

A similar time dependence for $M_R(t)$ in an increased temperature range has recently been reported.[2] This functional form has been theoretically justified in a cooperative-relaxation model for the time decay in dielectrics.[3] This concept has also been used, to simulate the ac magnetic susceptibility, $\chi = \chi' - i\chi''$, of some insulating spin glasses.[4] Palmer, Stein, Abrahams, and Andersson[5] have exploited an idea based on hierarchically constrained dynamics for the relaxation process in complex, strongly interacting materials. Also, this approach can give a stretched exponential form for the relaxation function. Murani[6] has recently analyzed neutron diffraction and ac-susceptibility data on a Cu*Mn* spin glass. He finds a power-law decay at short times plus a logarithmic time dependence at long times, at some temperatures below T_g. In this Rapid Communication we report on measurements of the relaxation of the thermoremanent magnetization on a Cu*Mn* spin glass clearly showing that the stretched exponential time dependence for the total remanent magnetization obtained by CMO is artificially caused by the influence of an aging process of the spin-glass state on the relaxation of the magnetization. A stretched exponential only describes the relaxation in a *specific* time interval and does not accurately describe the total relaxation of the remanent magnetization.

Experimental results on the relaxation of spin glasses ranging from the shortest experimental time scales available 10^{-12} sec (neutron spin echo), to the longest 10^5 sec [zero-field-cooled (ZFC) and thermoremanent magnetization (TRM) measurements] show that below T_g the relaxation process occurs *at least* in the entire time window experimentally available. The time interval in which relaxation occurs starts to broaden at temperatures way above T_g, reaches the time scales of ac susceptibility (10^{-6}–10^0 sec) at temperatures just above T_g, and continuously evolves towards longer times on lowering the temperature.[7] It is therefore of paramount importance to use data in as large a time interval as possible when trying to find the true functional form for the total relaxation of the magnetization in spin glasses. The formulas relating the frequency and time domain susceptibilities are[8]

$$(1/H)M(t) \approx \chi'(\omega), \quad t = 1/\omega \ , \tag{2}$$

$$(1/H)\partial M/\partial \ln t \approx \partial\chi'/\partial\ln\omega \approx 2/\pi\chi''(\omega), \quad t = 1/\omega \ . \tag{3}$$

These relations provide the tools to combine relaxation data from ZFC and TRM experiments with ac-susceptibility data.

In addition to the very broad time interval for the relaxation of the magnetization, an aging process of the spin-glass state occurs, at temperatures below T_g.[9] This aging is revealed by a pronounced wait-time (t_w) dependence of the relaxation of the magnetization.[10] The $M(t)$ vs $\log_{10}t$ curves exhibits an inflection point at $t \approx t_w$, showing a similar behavior both in ZFC and TRM measurements.[11]

The time decay of the remanent magnetization of a Cu–5-at.%-Mn spin glass was investigated in a superconducting quantum interference device (SQUID) magnetometer[12] as a function of temperature and wait time at constant temperature prior to the field change. The measurements were performed by cooling the sample in an applied magnetic field ($H = 2$ G) from a reference temperature ($T_{ref} = 28.5$ K) above the spin-glass temperature ($T_g = 28$ K) to the temperature for the relaxation measurement, T_m. At T_m the external field is switched off after a wait time at constant temperature, and the relaxation of the magnetization is measured in the time interval 1–10^4 sec. The sample is then heated to T_{ref}, where the zero level of the remanence is established. The external field is produced by a small superconducting magnet (wound of pure Nb wire) operating in the persistant mode. The time-dependent background signal arising from a field change was measured at T_{ref}, where the intrinsic spin-glass relaxation is negligible. This background effect is found to be very small in comparison with the spin-glass relaxation below T_g.

Figure 1 shows the time decay of the remanent magnetization (M_R) for different wait times at three temperatures below T_g. The curves are drawn relative to the zero level of the remanence at T_{ref}. As is clearly seen from Fig. 1 there is a pronounced t_w dependence of the relaxation. The most striking feature is the existence of an inflection point at $t \approx t_w$ for all wait times, at all temperatures. The existence of an inflection point for the $t_w = 0$ curves in the measured time interval, at 21 and 25 K, is due to the finite cooling rate (1 K/min) resulting in an effective wait time of about 200 and 50 sec, respectively. An increased cooling rate pushes the inflection points towards shorter times. The in-

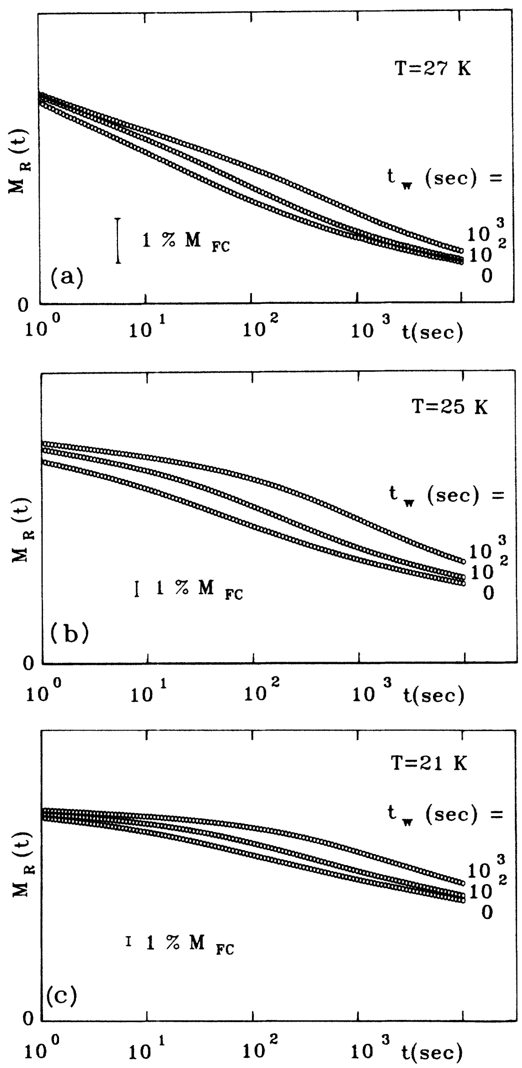

FIG. 1. Remanent magnetization curves at different wait times (t_w) plotted vs $\log_{10}(t)$. 1% of the field-cooled magnetization value is indicated. Cu-5 at.% Mn. (a) $T_m = 27$ K ($T_m/T_g = 0.96$), (b) $T_m = 25$ K ($T_m/T_g = 0.89$), (c) $T_m = 21$ K ($T_m/T_g = 0.75$).

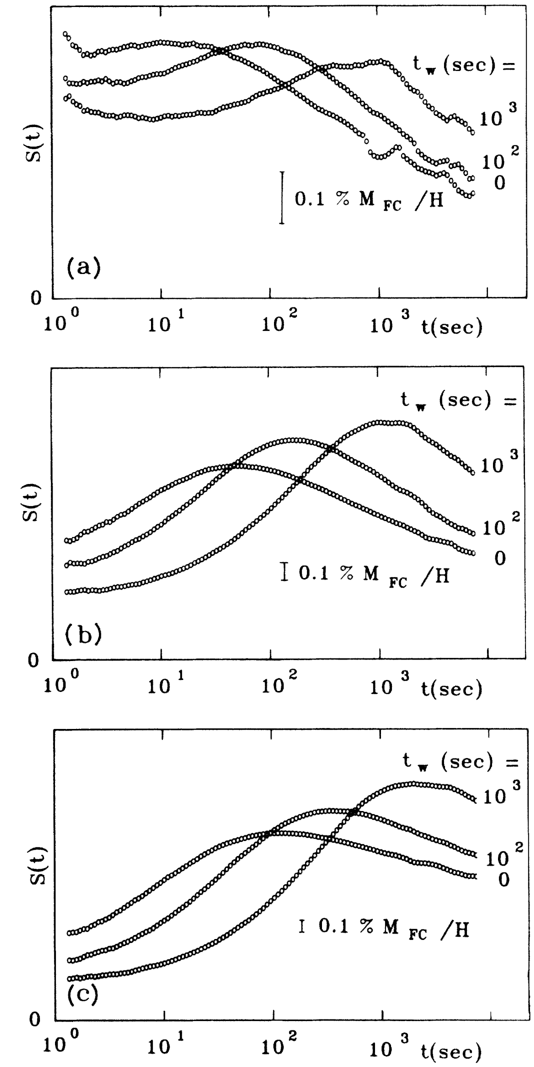

FIG. 2. Relaxation rates $[S(t)]$ of the remanent magnetization at different wait times plotted vs $\log_{10}(t)$. A relaxation rate of 0.1% of the field-cooled susceptibility value is indicated the figure. Cu-5 at.% Mn. (a) $T_m = 27$ K, (b) $T_m = 25$ K, (c) $T_m = 21$ K.

fluence of t_w is further emphasized by Fig. 2, where the relaxation rate, $S(t) = (1/H)(\partial M_R/\partial \ln t)$, is plotted for different wait times at the same temperatures as in Fig. 1. The time at which the maximum of the relaxation rate occurs is closely equal to t_w. The maximum is broadened by the effect of the finite cooling rate when t_w is smaller or comparable to the magnitude of the cooling time. At 27 K the influence of the aging process on the measured curves is rather small and is only revealed as a bump at $t \approx t_w$. At 25 and 21 K the influence of the aging process *dominates* the relaxation rate in the measured time interval. It should be emphasized that all these curves extrapolate[10] to a *finite* relaxation rate at $t << t_w$, at all temperatures. This finite value, which is several times smaller[10] than the experimentally observed value at $t \approx t_w$, is of the same magnitude as the corresponding values obtained at shorter times from ac-susceptibility measurements[13,14] through Eq. (3). After a presumed time t_{eq} (Ref. 9) the spin glass attains a dynamic equilibrium situation characterized by an almost time-independent relaxation rate in the time interval covered by ac- and dc-susceptibility measurements (this relaxation rate is experimentally observed at $t << t_w$). The time t_{eq} reaches astronomic time scales already at temperatures just below T_g. It is only in the dynamic equilibrium situation that one finds a unique relaxation function for the total relaxation of the magnetization. If this function yields a stretched exponential, the coefficient n must already at temperatures above T_g be close to 1, since the relaxation occurs in a very wide time interval and the relaxation is closely logarithmic in large time intervals.[6,7]

In conclusion, we analyze our data according to the procedure used by CMO to obtain the parameters characterizing a stretched exponential decay of the magnetization. In Fig. 3(a) we have plotted $-d/dt[\ln(M_R)]$ vs t in a log-log diagram for a wait time of 10^3 sec at 21 and 25 K (this wait time is of the same order of magnitude as the one used by CMO, Hoogerbeets, Luo, and Orbach,[2] and by Chamberlin[15] when analyzing the wait-time dependence of the relaxa-

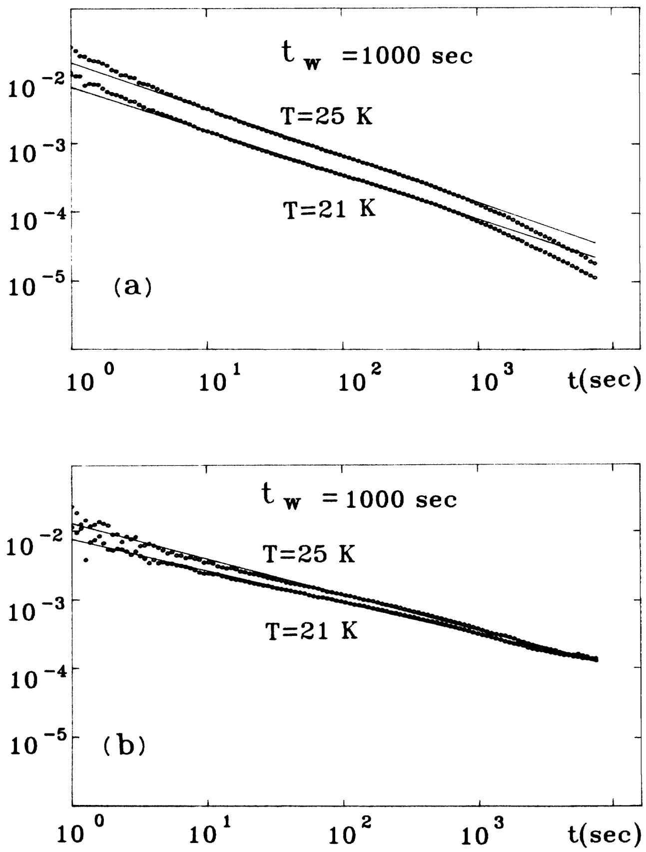

FIG. 3. (a) $\log_{10}\{-d/dt\ln[M_R(t)]\}$ plotted vs $\log_{10}t$. $t_w=10^3$ sec. Temperatures 21 K (bottom curve) and 25 K (top curve). Fitted straight lines in the time interval $5<t<10^3$ sec are indicated. $n=0.68$ (25 K) and $n=0.64$ (21 K). (b) $\log_{10}\{-d/dt[\ln(M_R-SH\ln t-M_1)]\}$ vs $\log_{10}t$. $t_w=10^3$ sec. Temperatures 21 K (bottom) and 25 K (top). Fitted straight lines in the time interval $1<t<10^4$ sec are indicated. $n=0.51$ (25 K) and $n=0.46$ (21 K). Parameters: $S=0.24\%$ M_{FC}/H, $M_1=19\%$ M_{FC} (21 K); $S=0.29\%$ M_{FC}/H, $M_1=9.4\%$ M_{FC} (25 K).

tion). If the data fall on a straight line when plotted as in Fig. 3 the relaxation follows a stretched exponential. The slope of the line gives the parameter n and the intercept at $\log_{10}t=0$ gives $(1-n)(1/t_p)^{1-n}$. As is seen from the figure deviations from a straight line occur both at *short* and at *long* times. It is only possible to achieve a reasonable fit to a straight line in the limited time interval $5<t<10^3$ sec. These fitted lines are indicated in Fig. 3(a). The values of the parameter n obtained from these fittings are in agreement with those obtained by CMO in the same time interval at corresponding temperatures. The deviations at short times, which are attributed to eddy current effects by CMO, are due to true relaxation of the spin-glass magnetization (a frequency dependence of the ac susceptibility of the same magnitude as this relaxation rate [Eq. (13)] is measured below T_g,[13,14] whereas a frequency-independent ac susceptibility is observed well above T_g). The deviations at long times are due to the inflection point in the relaxation at $t\approx t_w$, which occurs at too high a level of the remanence to possibly be described by only a stretched exponential decay. For the shorter wait times it is not possible to achieve a reasonable fit to a stretched exponential, in any major part of the measured time interval. At 27 K the relaxation is essentially different from only a stretched exponential decay.

We find that the total relaxation of the magnetization may accurately be described by empirically assuming a pure logarithmic decay superimposed by a stretched exponential form that accounts for the influence of the aging process

$$M_R=SH\ln(t)+M_1+M_{a0}(T,t_w)\exp[-(t/t_p)^{1-n}]\ , \qquad (4)$$

where S is the relaxation rate at dynamic equilibrium,[16] M_1 the intercept at $\log_{10}(t)=0$ for the logarithmic decay, and M_{a0} the magnitude of the superimposed stretched exponential. In Fig. 3(b), we have plotted $-d/dt[\ln(M_R-SH\ln(t)-M_1)]$ vs t in a log-log diagram for $T_m=21$ and 25 K. The data fall on straight lines in the whole time interval measured. The values of the parameters are $n\approx0.5$ and $t_p\approx1500$ sec. Since the inflection point of the relaxation curves occurs at $t\approx t_w$ it is tempting to identify t_p with t_w and to take into account the aging by a stretched exponential, where $n=0.5$, $t_p=t_w$, and M_{a0} is a temperature and (weakly) wait-time-dependent parameter, superimposed on a logarithmic decay. Hence the time decay is closely logarithmic at $t \ll t_w$ as well as at $t \gg t_w$, and the aging process *superimposes* a stretched exponential term on the relaxation at $t\approx t_w$. To illustrate this we have in Fig. 4(a) ($t_w=10^3$ sec) and in Fig. 4(b) ($t_w=10^2$ sec) plotted the relaxation in a larger time interval, $10^{-4}<t<10^8$ sec. The

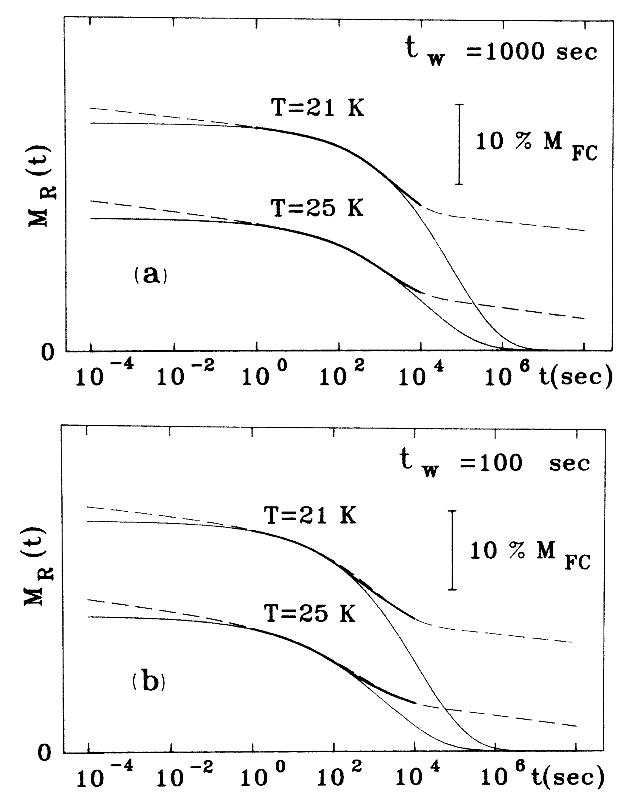

FIG. 4. Relaxation of the remanent magnetization (M_R) in the time interval $10^{-4}<t<10^8$. Thick full lines are experimental data. Dashed lines are the relaxation according to Eq. (4). Thin solid lines are the relaxation according to Eq. (1). 10% M_{FC} is indicated. (a) $t_w=10^3$ sec, temperatures 21 K (top) and 25 K (bottom). (b) $t_w=10^2$ sec, temperatures 21 K (top) and 25 K (bottom).

thick solid lines illustrate the experimental data, the thin solid lines the relaxation according to CMO with only a stretched exponential decay (the fitting for $t_w = 10^2$ sec is done in the time interval $1 < t < 10^2$ sec), and the dashed lines illustrate the relaxation according to Eq. (4). The relaxation of the magnetization given by Eq. (4) is at $t < 1$ sec in accordance with ac-susceptibility data on other Cu*Mn* spin glasses.[6,13,14] On the basis of the present results a continuity of the closely logarithmic decay at times beyond 10^4 sec is the most plausible extrapolation. Figure 4 illustrates the very peculiar behavior of the relaxation of the magnetization of spin glasses exhibiting an inflection point at $t \approx t_w$ in a logarithmic time perspective. It clearly shows that a pure stretched exponential decay is only illusively obtained in *limited* time intervals, *specific* for all wait times.

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[16]ac susceptibility (Ref. 6) and ZFC-magnetization measurements (Ref. 10) on Cu*Mn* spin glasses indicate that the equilibrium relaxation rate (S) weakly decreases with $\ln(t)$. Thus a logarithmic relaxation function gives a good approximation over a wide time interval with a constant S, although the limit $t \rightarrow \infty$ is not properly taken into account.