Comment on "Temperature dependence of the response time of dilute metallic spin glasses"

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In a recent article Hoogerbeets, Wei-Li Luo, and Orbach (HWO) advocate that the time decay of the thermoremanent magnetization below the spin-glass freezing temperature T_g can accurately be described by a stretched-exponential function form: $M_{\mathrm{TRM}}=M_0\exp(-(t/\tau_p)^{1-n})$. In this Comment we show that a pure stretched-exponential form is *never* sufficient to describe the relaxation of the magnetization in spin glasses below T_g. *Inter alia* we demonstrate that the exponential temperature and wait-time dependence of the "apparent response time" τ_p obtained by HWO from limited experimental observations, yields large deviations from the real behavior of the relaxation at long observation times.

In a recent article Hoogerbeets, Wei-Li Luo, and Orbach[1] (henceforth referred to as HWO) report extensive experimental studies of the time decay of the thermoremanent magnetization (TRM) below the spin-glass freezing temperature T_g of several metallic spin glasses. The bulk of the measurements by HWO have been performed in the experimental time window 0.2 to 500 sec and at a specific wait time of 10 min before the removal of the external field $H=6$ Oe. From these measurements and earlier investigations by the same group[2-6] it is stated that the time-dependent response of the field-cooled magnetization after the field cutoff is of the stretched-exponential form:

$$M_{\mathrm{TRM}}=M_0\exp(-(t/\tau_p)^{1-n}) \ . \qquad (1)$$

The general form of a stretched-exponential function is shown in Fig. 1 together with its relaxation rate $S=-dM_{\mathrm{TRM}}/d\ln t$. The relaxation rate exhibits a maximum at $t=\tau_p$ with a maximum value of $M_0(1-n)e^{-1}$.

In this Comment we demonstrate that a pure stretched-exponential form is *never* sufficient to describe the time decay of the TRM at temperatures below T_g. The resemblance to a stretched-exponential form is only due to the influence of the aging process on the relaxation which imposes a maximum to the relaxation rate at observation times of the order of the age of the studied spin-glass state.[7-10] The stretched-exponential functional form is by nature alien to the spin-glass dynamics at thermodynamic equilibrium. We show that the exponential wait time and temperature dependences of the parameter τ_p as obtained by HWO from a fit of experimental data to Eq. (1) is of little physical relevance.

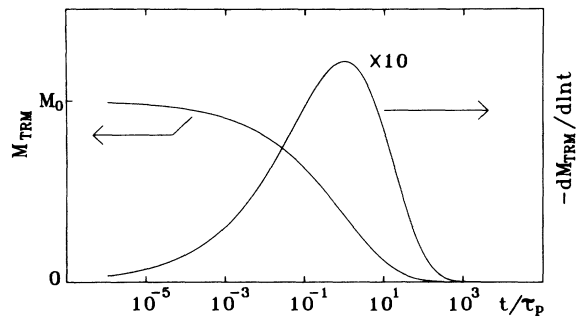

FIG. 1. Functional form of a stretched exponential [Eq. (1)] and its relaxation rate $-dM_{\mathrm{TRM}}/d\ln t$. The $\log_{10}(t)$ axis is drawn in units of t/τ_p. $n=0.66$.

The relaxation of the magnetization in spin glasses occurs *continuously* in an exceedingly large time window ranging from atomic time scales (10^{-13} sec) to a strongly temperature- and field-dependent maximum relaxation time, which at temperatures close to T_g goes beyond experimental time scales. The functional form of the relaxation is governed by the *temperature* and the *age* of the studied spin-glass state and the *magnitude of the applied field.*[11] An experimental search for the true low-field functional form of the relaxation in spin glasses using dc magnetization measurements thus has to meet the following essential requirements.

(i) A large time interval should be covered and results from experiments at short observation times (neutron scattering,[12] muon spin resonance,[13] ac susceptibility,[14] and Monte Carlo simulations of spin-glass models[15,16]) must be incorporated in the interpretation of the results.

(ii) The temperature should be accurately measured and controlled.

(iii) The influence of the age of the system must be measured (for some orders of magnitude in wait time) and properly accounted for.

(iv) The measurements should be performed in a field regime where there is a linear response of the relaxation.[11]

The macroscopic observables that directly reflect the time variation of the spin-spin time correlation function $q(t)$ are the frequency dependence of the real part of the ac susceptibility and the time dependence of the zero-field-cooled (ZFC) magnetization. The time decay of the thermoremanent magnetization is closely equal to the time dependence of the ZFC magnetization but has an additional component in the relaxation from the time decay of the field cooled (FC) magnetization that to some extent affects the results (see Refs. 9 and 17). Figure 2 shows the time decay of the FC magnetization for a Cu (10 at.% Mn) spin glass in an applied field of 3 G at $T/T_g=0.88$.

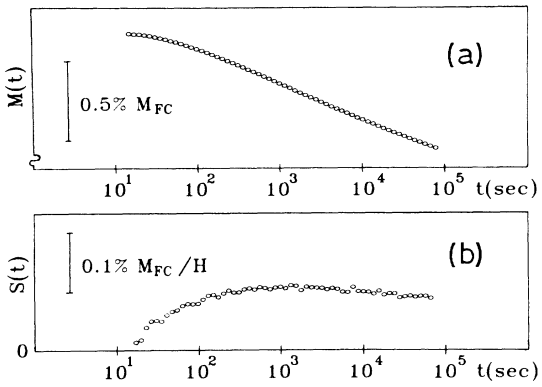

FIG. 2. Time decay of the field-cooled magnetization (M_{FC}) at constant temperature ($T/T_g=0.88$, $T_g=45.3$ K) of a Cu(10 at.% Mn) spin glass at $H=3$ G. (a) $M_{FC}(t)$. 0.5% M_{FC} indicated. (b) $S(t)=(1/H)\partial M_{FC}/\partial \ln t$. 0.1% M_{FC}/H indicated. The apparent small value of S at short times is due to the finite cooling rate (1 K/min).

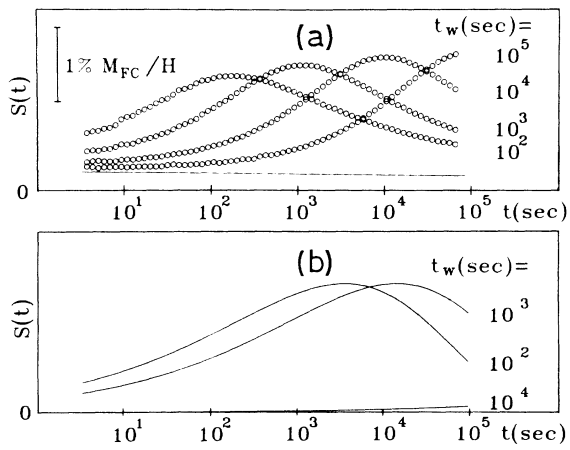

FIG. 3. (a) Relaxation rate $S=1/H\partial M_{ZFC}/\partial \ln t$ of a Cu(10 at.% Mn) spin glass at different wait times t_w. The solid line is a weak power-law decay representative of the equilibrium relaxation ($t_w=\infty$). $T/T_g=0.88$. $H=3$ G. 1% M_{FC}/H indicated. (b) Wait-time dependence of the relaxation rate of spin glasses according to HWO. The curves are drawn from a direct differentation of Eq. (1) using the wait-time dependence of τ_p given in Eq. (2) with parameters from Ref. 3 for a Ag(2.6 at.% Mn+0.45 at.% Sb) spin glass at $T/T_g=0.81$. The amplitude of S is scaled to be of comparable size to the curve at $t_w=10^3$ sec in (a).

The curve is recorded by starting the clock the moment the measurement temperature (T) is reached. This decay is by no means insignificant when viewed in a large logarithmic time perspective that is indispensable for accurately picturing the spin-glass relaxation. Additionally, the absolute value of TRM exhibits a nonlinear field dependence.[11] The best way to display data of the relaxation of the magnetization is through a plot of the relaxation rate $(1/H)\partial M/\partial \ln t$ in TRM and ZFC[18] measurements or $\partial \chi'/\partial \ln \omega$ in ac susceptibility measurements.[19] In such plots spurious effects of ill-defined equilibrium levels are avoided and the influence of the age of the system on the functional form is stressed.

In light of our own investigations we compare the results obtained by HWO to the experimental requirements stated above and critically examine their summarizing conclusions regarding: (i) the feasibility of a pure stretched-exponential functional form for the relaxation, (ii) the influence of wait time on the relaxation, (iii) the temperature dependence of the relaxation, (iv) the relaxation close to T_g, and (v) the agreement to theoretical predictions.

Dating from the initial discovery[7,20] of aging phenomena in spin glasses, it was realized that the aging process has a profound influence on the low-field experimental ZFC and TRM curves. The aging process is revealed as a wait-time (t_w) dependence of the experimental curves. In Fig. 3(a) we show measurements of the relaxation rate of the ZFC magnetization at various t_w on a Cu(10 at.% Mn) spin-glass sample. The measurements are performed at $T/T_g=0.88$ ($T_g=45.3$ K) and in an applied field of 3 G. From measurements on CuMn samples, with Mn concentrations ranging from 1 to 10 at.%, it is found that the relaxation-rate curves look very similar at the same reduced temperature. The relaxation-rate curves of Fig. 3(a) exhibit a wavelike character with the crest of the wave progressively moving toward longer times with increasing wait time. The most significant feature of the curves is the existence of a maximum at an observation time closely coinciding with the wait time. Judging from the evolution of the relaxation rate curves with wait time, it is not hard to imagine that the curves at equilibrium (i.e., $t_w=\infty$) level down on the solid line in Fig. 3(a) which represents a weak power-law decay. A power-law decay of the spin-glass relaxation at thermal equilibrium is also suggested from recent computer simulations of $q(t)$ by Ogielski.[16] A comparison between the relaxation rate curves in Fig. 1 and Fig. 3(a) clearly show that an *apparent* stretched-exponential relaxation is imposed by the influence of the aging process and is only observed when the exprimental observation time is of the order of the wait time. The relaxation-rate curves may crudely be described by a stretched-exponential relaxation with $\tau_p \sim t_w$ superimposed on a weak power-law relaxation.[10] The curves in Fig. 3(a) cannot accurately be described by a pure stretched-exponential form.

HWO report that the influence of the age of the system is described by only a wait-time dependence of the "apparent response time" τ_p according to a formula suggested by Chamberlin:[3,4]

$$\tau_p = B\exp(t_w/t_0) \ . \tag{2}$$

Figure 3(b) shows the wait-time dependence of the relaxation rate obtained by differentiation of Eq. (1) and using the wait-time dependence of τ_p as given by Eq. (2). The parameters are chosen according to the behavior at $T/T_g=0.81$ reported by Chamberlin in Ref. 3, i.e., $B=3\times10^3$ sec, $t_0=650$ sec, and with constant values of M_0 and n. For instance, Eq. (2) implies that already at the moderate wait time $t_w=10^5$ sec, the maximum relaxa-

tion rate should be obtained at 10^{70} sec, and thus no relaxation should be observed at experimental time scales. As is evident from Fig. 3(a) this exponential increase of τ_p with t_w gives an unphysical description of the influence of aging on the spin-glass relaxation. The maximum relaxation rate is experimentally always found at an observation time closely equal to the wait time. Measurements by Alba, Ocio, and Hammann[21] on a Ag(Mn) spin glass show results very similar to ours on Cu(Mn) spin glasses. In the time interval 5–500 sec their data are in agreement with the data of HWO at a similar wait time. Alba *et al.*, analyze their results adopting a formalism originally used to describe aging in polymers.[22] This formalism accurately describes their data in a large time interval ($1-10^5$ sec) and a wide range of wait times (10^3-10^5 sec). In addition, it predicts a reasonable behavior at short observation times (a weak power-law decay).

The "most remarkable"[5] result obtained by HWO is an exponential temperature dependence of $1/\tau_p$:

$$1/\tau_p = A\exp[-\alpha(T_g/T)] \ , \tag{3}$$

with $A = 10^{-3}$ sec^{-1} and $\alpha = 2.5$.

The measurements by HWO refer to a wait time of 10 min and the experimental data are fitted to a stretched exponential in the time interval 5–500 sec. Figure 4(a) shows the temperature dependence of the relaxation-rate curves for a Cu(10 at.% Mn) spin glass at $t_w = 10^3$ sec in the time interval $3-3\times10^4$ sec. As seen in Fig. 4(a) at short observation times the relaxation rate monotonically decreases with decreasing temperature. A maximum is always (at least down to $T/T_g = 0.1$) occurring at $t \sim t_w$. The magnitude of the maximum value grows to a maximum height at $T/T_g \sim 0.8$, followed by a monotonic decrease with decreasing temperature. Figure 4(b) shows the corresponding behavior of the relaxation-rate curves using the parameters obtained by HWO for metallic spin glasses from a fit to Eq. (1). The temperature dependence of M_0 is taken from Fig. 1 in Ref. 5 [data from an Ag(2.6 at.% Mn) spin glass]. The curves in Figs. 4(a) and 4(b) are scaled at $T/T_g \sim 0.78$ in the time interval 5–500 sec. A comparison between the curves in Figs. 4(a) and 4(b) strikingly illustrates the limitations of fitting limited experimental data to only a stretched-exponential functional form. The temperature dependence of τ_p as given by Eq. (3) implies that the maximum relaxation rate at $T/T_g = 0.1$ occurs at $\tau_p = 10^{14}$ sec and with a very large amplitude, while the real experimental observation is a weak, shallow maximum at an experimental observation time closely equal to the wait time (10^3 sec), i.e., a deviation of more than ten orders of magnitude between the experimentally observed "apparent response rate" and the value indicated by HWO. The fundamental observation that a maximum in the relaxation rate is always observed at an observation time closely equal to the wait time immediately rules out the physical relevance of both Eqs. (2) and (3).

At temperatures close to T_g the functional form of the time decay of TRM does not even resemble a pure stretched-exponential form [see, e.g., Figs. 1(a) and 2(a) in Ref. 10]. The field and temperature dependence of the parameters n and τ_p deduced at these temperatures by HWO are thus of less physical significance than those obtained at $T/T_g < 0.95$.

We illustrate in Fig. 5 a plausible form of the relaxation rate of a typical spin glass in a major observation time interval ranging from the experimental short time limit 10^{-12} sec (neutron diffraction) to the current experimental long time limit of some 10^5 sec, which can be reached in dc magnetization measurements. The curve (thick solid line in Fig. 5) is drawn compatible to data at $T/T_g = 0.55$, $t_w = 10^3$ sec, and in the region for a linear response of the spin-glass relaxation. As shown in Fig. 5 the relaxation rate exhibits a weak power-law decay at

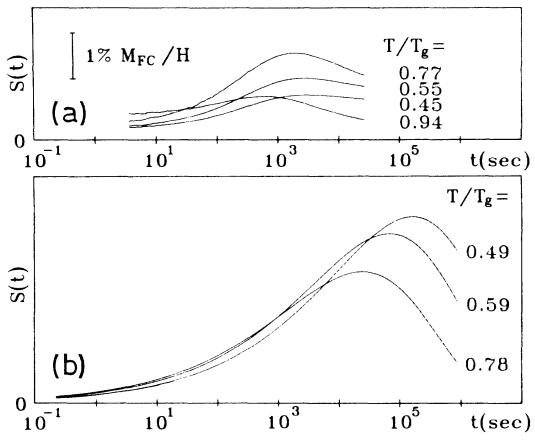

FIG. 4. (a) Temperature dependence of the ZFC relaxation rate S of Cu(10 at.% Mn) at $T/T_g = 0.94$, 0.77, 0.55, and 0.45. $T_g = 45.3$ K. $t_w = 10^3$ sec. $H = 3$ G. 1% M_{FC}/H indicated. (b) Temperature dependence of the relaxation rate S_{TRM} according to HWO. The curves are drawn from a direct differentation of Eq. (1) using the temperature dependence of τ_p obtained from Eq. (3). The amplitude of S is scaled at $T/T_g = 0.78$ to the corresponding amplitude in (a) at $T/T_g = 0.77$ in the time interval 5–500 sec. $n = 0.66$. The temperature dependence of M_0 [Eq. (1)] is taken from Fig. 1 in Ref. 5.

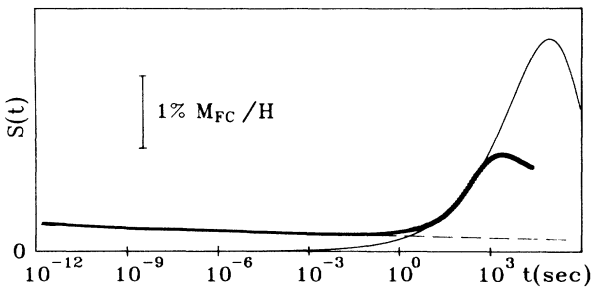

FIG. 5. The form of the relaxation rate S is the experimentally achievable time window 10^{-12}-10^5 sec. The thick solid line is a weak power law which is continued with the measured $S(t)$ of a Cu(10 at.% Mn) spin glass at $T/T_g = 0.55$ and $t_w = 10^3$ sec [as in Fig. 4(a)]. The dashed line represents a continuation of the weak power law to long times. The thin solid line corresponds to the relaxation rate of a pure stretched-exponential form with parameters compatible to a spin-glass relaxation according to HWO, at $t_w = 10^3$ sec and $T/T_g = 0.55$.

short observation times followed by a large wave at observation times of the order of the wait time. This wave, which is a nonequilibrium phenomenon, progressively moves along the dashed line with increasing wait time. The dashed line is the continuation of the power-law decay, which represents the behavior of the relaxation at equilibrium ($t_w = \infty$). For comparison we have also plotted the relaxation-rate curve according to HWO (thin solid line in Fig. 5) using their parameters at $T/T_g = 0.55$ and scaling the amplitudes in the observation time interval 5–500 sec, where their data agree with our results. This figure strikingly illustrates the necessity to use a wide time perspective to appreciate the real functional form of the relaxation in spin glasses.

A major part of the article by HWO is devoted to a comparison of their results and the dynamics of an infinite-range Ising spin-glass model reported by De Dominicis, Orland, and Lainée.[23] The latter authors obtain a time dependence for recovery of the perturbed spin-glass-state occupancies described by a power law times a stretched-exponential form. In this functional form the stretched-exponential part characterizes the final approach toward equilibrium at long times. In measurements on spin glasses at low fields and at temperatures below T_g, not even a precursor of an equilibrium approach is ever detectable at experimental time scales. The apparent stretched-exponential form is a nonequilibrium effect caused by the influence of aging on the spin-glass relaxation. The relevance of the results by HWO as to the credibility of this theory is thus not clear.

The experimental data obtained by HWO are unusually well defined compared to many other published results on the time decay of the thermoremanent magnetization of spin glasses. However, their statement that the relaxation in spin glasses is of a pure stretched-exponential form is misleading. Earlier works by the same group have been cited by many theoreticians[23,24] and by experimentalists[25] in the field of glassy dynamics, which has a tenuous relation to the actual experimental observations by HWO.

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