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Memory effect in $Dy_{0.5}Sr_{0.5}MnO_3$ single crystals

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Abstract

We have performed a series of magnetic aging experiments on single crystals of $Dy_{0.5}Sr_{0.5}MnO_3$. The results demonstrate striking *memory* and *chaos-like* effects in this insulating half-doped perovskite manganite and suggest the existence of strong magnetic relaxation mechanisms of a clustered magnetic state. The spin-glass-like state established below a temperature $T_{sg} \approx 34$ K originates from quenched disorder arising due to the ionic-radii mismatch at the rare earth site. However, deviations from the typical behavior seen in canonical spin glass materials are observed which indicate that the glassy magnetic properties are due to cooperative and frustrated dynamics in a heterogeneous or clustered magnetic state. In particular, the microscopic spin flip time obtained from dynamical scaling near the spin glass freezing temperature is four orders of magnitude larger than microscopic times found in atomic spin glasses. The magnetic viscosity deduced from the time dependence of the zero-field-cooled magnetization exhibits a peak at a temperature $T < T_{sg}$ and displays a marked dependence on waiting time in zero field.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Many examples of striking relaxation dynamics and memory phenomena primarily found in canonical spin glasses are observed in phase-separated and/or A-site disordered rare earth manganites, e.g., $R_{1-x}A_xMnO_3$ where R is a rare earth, and A is an alkaline earth ion. Notable among the reports are those on rejuvenation and memory effects in resistivity and magnetization by Freitas *et al* [1] and Levy *et al* [2]. These time-dependent phenomena were attributed to the phase-separated state resulting from competing ferromagnetic (FM) double-exchange and antiferromagnetic (AFM) superexchange interactions in manganites. In the class of half-doped manganites, long-time logarithmic relaxation rates are reported [3] in $La_{0.5}Ca_{0.5}MnO_3$ and $Nd_{0.5}Sr_{0.5}MnO_3$ and ascribed to the presence of FM and AFM mixed interactions among the Mn ions. Non-equilibrium behavior in the specific heat of $La_{0.5}Ca_{0.5}MnO_3$ was also attributed to the dynamics of co-existing phases [4]. On the other hand, by means of a scaling analysis for magnetization data of $Eu_{0.5}Ba_{0.5}MnO_3$ and $(La_{0.25}Nd_{0.75})_{0.7}Ca_{0.3}MnO_3$ a divergence of the nonlinear susceptibility has been observed which demonstrates freezing corresponding to a real spin glass state [5, 6]. Phase separation (or the formation of polaronic states) can lead to the co-existence of regions with different electronic or magnetic properties on a nanometer scale [7]. The particular character of the glassy magnetic state may be caused by the mixed-crystal disorder effects leading to a nanometer-sized domain state of the different 'phases'. The corresponding magnetic states might behave differently

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than conventional spin glass systems with atomic spins and frustrated exchange. Here it is to be noted that the distinction between a 'homogeneous' spin glass state with atomic spins and a nanoscale phase-separated state is very subtle and difficult to establish. For comparison we should mention that various unconventional systems [8–10] also exhibit signatures similar to those of a spin glass state such as *aging*, *memory effect*, and the effect of cooling rate on aging.

Interestingly, it was recently demonstrated by means of a model of non- or weakly-interacting superparamagnetic particles that certain aging and memory effects can arise even in assemblies of independent magnetic entities [11]. Thus, careful experiments must be performed to differentiate the slow dynamics of a thermodynamic spin glass ground state from the activated dynamics over anisotropy-energy barriers of superparamagnets. Among the many protocols available, the most commonly employed ones are: field cooled (FC) [12, 13] and zero-field-cooled (ZFC) protocols [14], protocols with a heating step instead of a quench to test the validity of droplet [15, 16] versus hierarchical models of spin glasses [17], and waiting time dependence of the magnetization measured within the ZFC protocol [18, 19]. In this paper we focus on the latter, i.e., we describe the magnetic aging experiments performed on single crystals of $Dy_{0.5}Sr_{0.5}MnO_3$ and discuss the *spin-glass-like* state found in this system. In our recent investigation on $Dy_{0.5}Sr_{0.5}MnO_3$, we noticed a magnetic transition at low temperature from a paramagnetic to a spin-glass-like state that occurs at ∼34 K [20]. Electron paramagnetic resonance studies at low fields also display signatures of a spin-glass-like state in this system [21]. The system belongs to the class of half-doped manganites with a small R ion which is believed to cause a spin-glass-like state for random occupation of the A sites [22, 23]. However, by probing the magnetization dynamics through ac susceptibility measurements, we found an uncommonly large characteristic microscopic relaxation time τ_0 of 10^{-8} s [20]. Similarly high values for τ_0 have been obtained in a $La_{1-\delta}Mn_{0.7}Fe_{0.3}O_3$ cluster-glass compound [24]. This is several orders of magnitude larger than typical relaxation times of about 10^{-12} s found for spin glasses ruled by single-spin-flip dynamics. Also, in most half-doped manganites and Fe-doped cobaltates that exhibit spin glass behavior the microscopic time τ_0 yields values $\approx 10^{-13}$ s [5, 25]. Such small values are typically observed in canonical spin glasses. The large value of τ_0 found here indicates the presence of interacting clusters of spins in $Dy_{0.5}Sr_{0.5}MnO_3$. The power-law dynamical scaling is also well observed in the ac susceptibility data which rules out superparamagnetism of independent clusters. Here we conclusively characterize the glassy magnetic state in $Dy_{0.5}Sr_{0.5}MnO_3$ by carrying out *memory*, *aging*, and *waiting time* measurements which are described in the following sections. $Dy_{0.5}Sr_{0.5}MnO_3$ displays slow glassy dynamics and memory effects which can be explained only by cooperative spin-glass-like dynamics. Our experiments suggest that in this half-doped manganite a spin-glass-like or cluster-glass-like magnetic state exists that deviates from the properties of canonical spin glass systems.

2. Experimental details

The samples used in the present study were single crystals of $Dy_{0.5}Sr_{0.5}MnO_3$ grown by the optical floating-zone method [20]. The quality of the grown crystals was confirmed by Laue photography, and their chemical composition ascertained by inductively-coupled-plasma-atomic-emission spectroscopy using a Perkin Elmer Optima 2000 spectrometer. The composition thus determined was very close to perfect stoichiometry. A cuboid of dimension $2 \times 2 \times 2$ mm^3 extracted from the boule was used for the magnetic experiments. The ac susceptibility for this crystal has been measured in greater detail, compared to the experiments reported in [20], in the frequency range from 10 Hz to 10 kHz. A critical scaling analysis of these detailed data firmly corroborates the behavior and parameters already reported [20]. The magnetic aging and waiting time experiments were performed using a MPMS SQUID magnetometer whereas the ac susceptibility was measured in a commercial PPMS (both made by Quantum Design).

3. Results and discussion

3.1. Memory effect in cooling cycle

To understand whether the slow dynamics of $Dy_{0.5}Sr_{0.5}MnO_3$ is a result of a collective behavior, we performed FC magnetization experiments with stops, as suggested for weakly-interacting magnetic nanoparticle systems. Similar experiments [6] have been performed for the out-of-phase susceptibility of $(La_{0.25}Nd_{0.75})_{0.7}Ca_{0.3}MnO_3$. In our case, the sample was field cooled at 100 Oe from 300 to 5 K at a constant cooling rate of 1 K min^{-1}. In the cooling cycle, temporary stops were administered at 50, 30 and 20 K. During these stops, the field was switched off and a waiting time $t_w = 4$ h was engaged. Thus, during t_w the system aged. The stops at 30 and 20 K are discernible in figures 1(a) and (b) (in the latter, only the cooling curve is presented for clarity) as step-like features indicating the relaxation of the system during t_w. Once the cooling cycle was completed, the sample was heated back to room temperature at the same constant rate of 1 K min^{-1} while measuring the magnetization. Note that in the heating cycle the same field of 100 Oe was applied but no stops were administered. The changes in magnetization during the cooling cycle are retraced in the heating cycle and the curves essentially fall on top of each other. This retracing effect is clearly manifested by the peaks in the derivative dM/dT, which re-appear as smoothed features and with a delay in the heating cycle for $T < 40$ K, as presented in figure 1(c). This implies that the system recollects its previous thermal history during the heating cycle, which corresponds to a proper '*memory effect*'. In contrast, the stop administered at $T = 50$ K—which is well above the spin glass freezing temperature—did not cause any significant dip in $M(T)$ in either the cooling or the heating cycle. Notice that when cooling resumes after the temporary stops, the temperature dependence of the magnetization continues with the same slope dM/dT as before the stop. Also, the slopes of the magnetization measured with

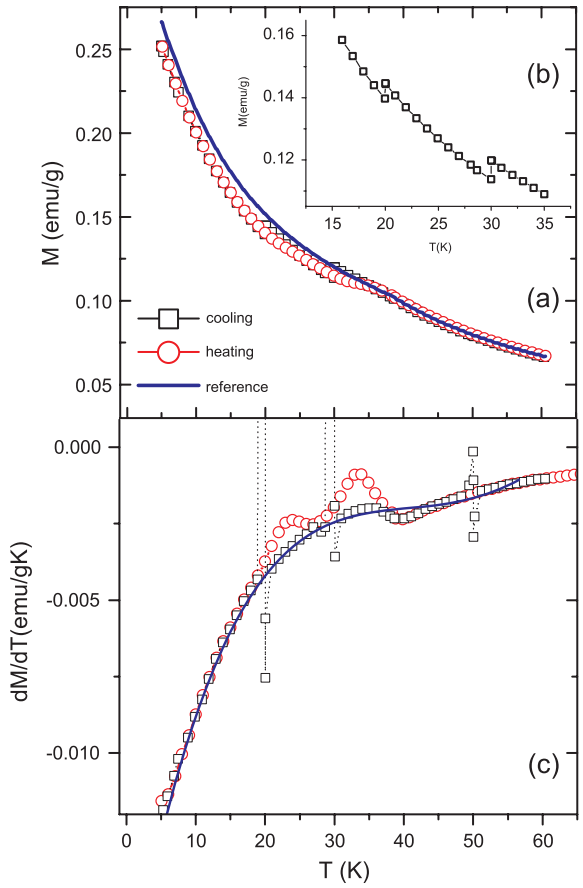


Figure 1. Memory effect in $Dy_{0.5}Sr_{0.5}MnO_3$ observed in the temperature dependence of the magnetization. (a) Squares represent the magnetization measured while cooling under FC conditions (100 Oe) at a rate of 1 K min^{-1}. Drops in M during the stops at 30 and 20 K are obvious and magnified for clarity in (b); the stop at 50 K cannot be recognized. Circles are data subsequently measured after the FC cycle during uniform heating without stops. The solid line is a reference cooling at 100 Oe with no stops during the measurement. (c) Derivatives dM/dT for the same cycles as in (a). The stops during cooling result in dM/dT data partially off scale. The memory effect gives rise to the broadened peaks on heating.

and without stops are the same with the exception of those temperatures close to the stops. Hence, the system behaves as if aging at higher temperatures (here at 20 and 30 K) which has had no effect on the low temperature behavior of the system except for the slightly reduced value of M. This is suggestive of the so-called *chaos effect* as described by Jonason *et al* [26]. However, the merging of the magnetization curves for the different cooling and the heating cycles is much slower than that observed in canonical spin glasses like Cu:Mn, or in a spin-glass-like system with a dense magnetic sublattice such as chromium thiospinels where memory effects were studied in detail [12]. Further, the comparison to the ac susceptibility measurements of Jonason *et al* [26] is not straightforward. In their ac susceptibility studies, the measurements were carried out in an ac-field of 1 Oe. Hence, the zero-field non-equilibrium state is only weakly perturbed during the measurements. In our dc magnetization measurements, however, a field of 100 Oe is applied during cooling. In the temporary stops when the field is switched off, the system relaxes back to the ground state. However, when the field is applied and cooling is reassumed, the system does not attain immediately the original FC magnetization as one would expect in the chaotic behavior of pure spin glasses. Instead, a certain amount of relaxation is observed implied by the lower value of M. A possible explanation could be that the internal field acting on the spin glass phase does not remain constant during cooling protocol due to a progressive increase of polarization of the paramagnetic Dy-spins. In fact, the presence of strongly interacting magnetic sublattices has been observed in $Dy_{1-x}Sr_xMnO_3$ ($0 \leqslant x \leqslant 0.4$) [27]. Such an interaction between Dy and Mn spins cannot be ruled out in $Dy_{0.5}Sr_{0.5}MnO_3$ as well. Also, manganites are known for intrinsic phase separation [7], which could yield intrinsically inhomogeneous magnetic states with different relaxation behavior. Further, even though the samples have been characterized by x-ray diffraction, small secondary phases might be present, since the method can hardly reveal clusters on a nanometer scale. Sophisticated structural characterization techniques such as high resolution cross section electron microscopy [28] or advanced synchrotron spectroscopic methods [29] might be needed to reveal these subtle effects. If such phase separation occurs in the present system, they may lead to spurious relaxations. Thus, the observed effects in the present case are likely more complicated than those in the canonical spin glasses.

As a next step, we studied the ZFC magnetic relaxation and the effect of a temperature quench on relaxation. For this purpose, the sample was first cooled in zero field to $T_1 = 30$ K. A magnetic field of 100 Oe was applied at this temperature, and the magnetization was recorded as a function of time for a period $t_1 = 42$ min. Hereafter, the sample was field-quenched to a lower temperature $T_2 = 15$ K, and the sample was held for a time $t_2 = 42$ min at this lower temperature while recording the magnetization. Finally, the temperature was restored to T_1, and magnetization was again recorded for a time $t_3 = 42$ min. The magnetization $M(t, T)$ obtained from such a relaxation experiment is presented in figure 2(a). We observe that the magnetization returns to nearly the same value after the temporary halt at the lower temperature $T_2 = 15$ K. This is a clear confirmation of the memory effect observed in the previous experiment. Figure 2(b) shows the result of the same measurement repeated at 500 Oe. To further illustrate this phenomenon the magnetization obtained during the aging periods t_1 and t_3 is plotted sequentially, in figure 3(a) (for 100 Oe) and (b) (for 500 Oe), leaving out the intermittent time evolution $M(t_2, T_2)$. The plot displays *perfect memory*, i.e., the magnetization within the time period t_3 appears as a perfect continuation to the curve during t_1 for both measurements taken at 100 and 500 Oe. Thus, figure 3 is a clear indication that the system recollects its initial configuration when it returns to the initial temperature after a temperature quench. In the insets of figure 3, the same data are presented on a logarithmic scale (the logarithmic time dependence of M will be discussed below).

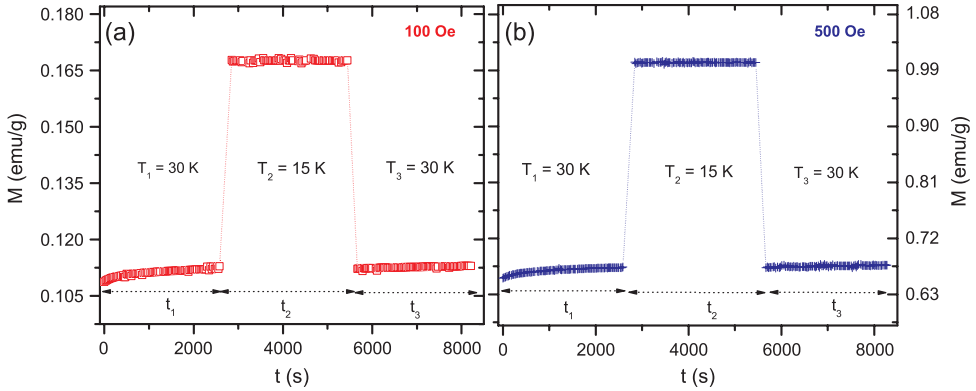


Figure 2. Response of the magnetization to a relaxation experiment employing the ZFC protocol during which a temporary quench to 15 K is applied under 100 Oe (a) and 500 Oe (b).

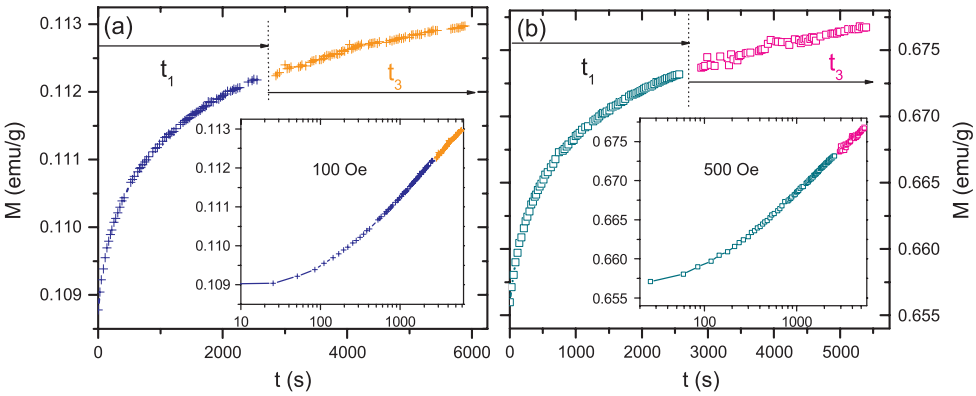


Figure 3. Magnetic relaxation as a function of time during period t_1 and t_3 under 100 Oe (a) and 500 Oe (b). Both data sets are obtained at $T = 30$ K, cf figure 2. Insets show the same data on a logarithmic timescale. An excellent merging of the magnetization curves $M(t_1)$ and $M(t_3)$ is observed which indicates perfect memory.

3.2. Asymmetry in temperature cycle

The droplet model of spin glasses [15, 16] predicts a symmetric memory effect in response to heating and cooling during magnetic relaxation, whereas the hierarchical model favors an asymmetric response [17]. Experimentally, effects of temperature changes on the magnetic aging have been studied in thiospinels by Lefloch *et al* [30]. In order to gain deeper insight into the magnetic relaxation mechanism in $Dy_{0.5}Sr_{0.5}MnO_3$ we also performed relaxation measurements with intermittent heating cycles. The results are presented in figures 4(a)–(d). Here, the sample was initially zero-field-cooled and subsequently a field of 500 Oe was applied while measuring $M(t)$. Comparing the heating ($T_1 \rightarrow T_2$; 10–25 K in figure 4(a) and 15–30 K in figure 4(b)) and cooling steps ($T_2 \rightarrow T_3$; 25–10 K in figure 4(a) and 30–15 K in figure 4(b)), the response of the system is clearly asymmetric, and there is no memory effect. As can be seen in figures 4(c) and (d), asymmetry is also observed when the temperature difference ΔT is as small as 5 K. A similar experiment in the intermittent heating cycle, but with an applied field of 100 Oe, was also performed (not shown) wherein, again, no memory effect was observed. The different behavior under an intermittent temperature change during the relaxation experiments is in agreement with the hierarchical model. This model explains memory effects in terms of multi-valley structures in the free-energy surface of the system at a given temperature. Upon quenching the system from temperature T to $T - \Delta T$, the free-energy valleys split into new sub-valleys. If the temperature quench ΔT is large, then the barriers separating the originally existing valleys become too high. Hence, the magnetic system can only access states within the newly formed sub-valley structure but cannot reach any other of the originally existing valleys during finite waiting times. This confinement of the magnetic system to the set of new sub-valleys formed at $T - \Delta T$ out of *one* original valley at T is believed to be at the heart of the memory effect. When the temperature T is restored, the sub-valleys merge back to the original free-energy valley, and the relaxation at T resumes without being influenced by the excursion to the lower temperature $T - \Delta T$. Thus, the hierarchical model predicts a memory effect for temperature quenches but certainly not for increasing temperature. This asymmetry is clearly observed in our system: the memory effect is found after intermittent cooling (cf subsection 3.1), but not for intermittent heating (figure 4). This type of memory effect shows that the magnetic system in $Dy_{0.5}Sr_{0.5}MnO_3$ owns complexity which must arise as a consequence of frustrated couplings and cooperative dynamical processes that create a complex free-energy landscape. The effect of positive and

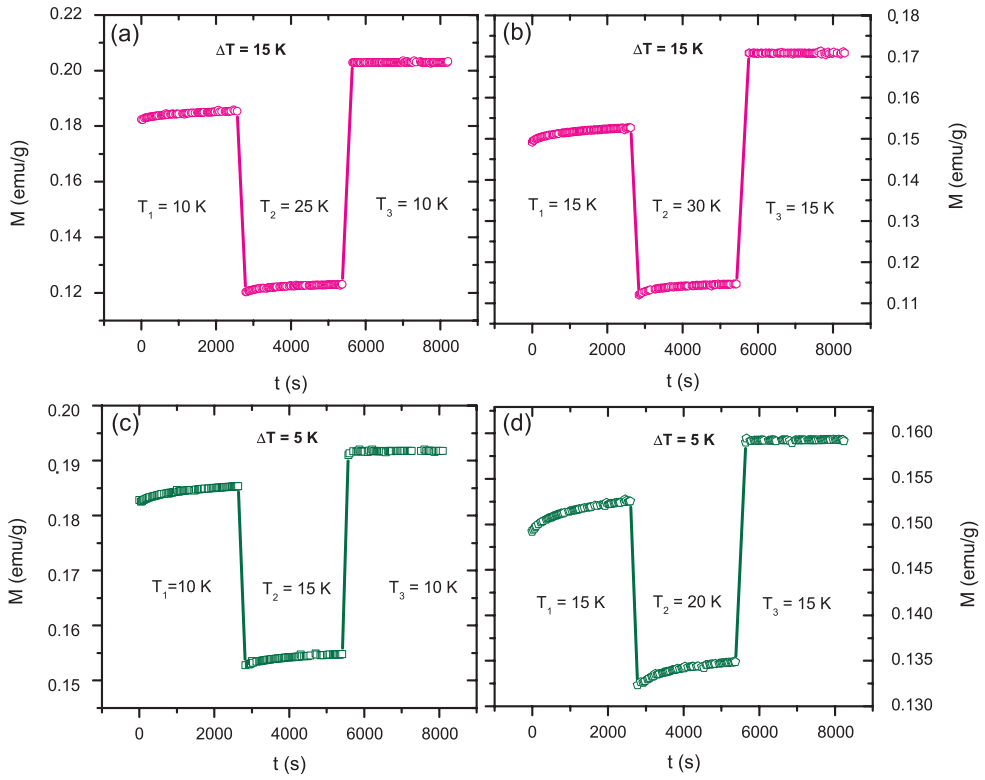


Figure 4. Asymmetry in the magnetic response to an intermittent heating cycle. After zero-field cooling, a field of 500 Oe was applied for $M(t)$ measurements. The magnetic relaxation is asymmetric after the second temperature change back to (a) $T_3 = T_1 = 10$ K, (b) 15 K, (c) 10 K, and (d) 15 K.

negative temperature cycling in the glassy state has been investigated in many systems including spin glasses [30], relaxor ferroelectrics [31], and orientational glasses [32].

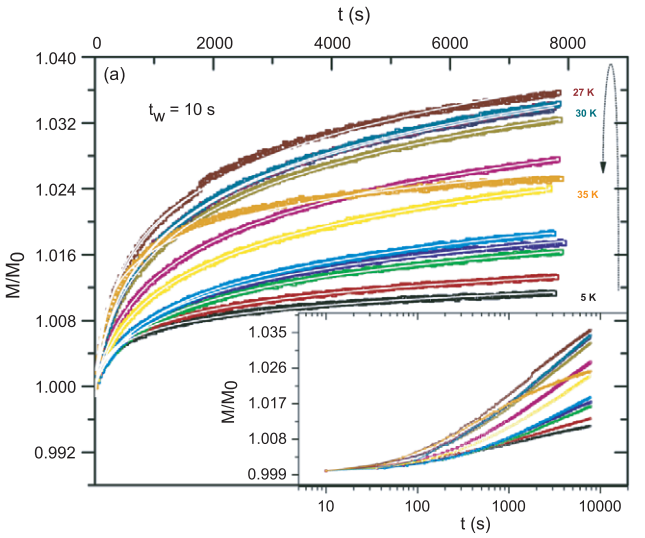
3.3. Waiting time dependence of $M(t)$

The effect of waiting time on the slow dynamics was studied through ZFC measurements. To this end, the sample was heated to a temperature $T = 300$ K well above the spin glass freezing temperature, and zero-field-cooled to a temperature below T_{sg}. This was followed up by waiting times of $t_w = 10$ or 3600 s, in zero field. Subsequently, a field of 100 Oe was applied, and the magnetization was recorded as a function of time.

Time dependences of the magnetization $M(t)$ are commonly described by the well-known expression for magnetic viscosity

$$M(t, T) = M(0, T) + S \ln(1 + t/t_0), \qquad (1)$$

where $M(0)$ is the magnetization at $t = 0$ and S is the magnetic viscosity. The reference time t_0 is typically orders of magnitude larger than the observed microscopic spin flip times τ_0. Experimental data $M(t)$, normalized to the first data point $M(t = 0)$, for $t_w = 10$ and 3600 s at various temperatures below the spin glass freezing are presented in figures 5(a) and (b). We find that the magnitude of $M(t)$ below the freezing temperature strongly depends on the waiting time t_w spent at that temperature before switching on the field. This effect is a consequence of non-equilibrium dynamics. The applicability of equation (1) to these data is evidenced by the inset of figure 5 in which the same data are plotted on a logarithmic timescale. Note that—according to equation (1)—good linearity of $M(t)/M(0)$ in such a representation can only be expected for $t \gg t_0$. Consequently, we attempted to fit our $M(t)$ data to equation (1). The resulting fit curves are included in the main panel of figure 5 as solid white lines. The values of t_0 obtained from the fittings at different temperatures and for the two waiting times are presented in figure 6. Figure 7 correspondingly displays $S(T, t_w)$ as a function of temperature for two waiting times. The logarithmically slow relaxation for the experiments with both short and long waiting time can be described by equation (1), but with different viscosity coefficients S. Clearly, the different $M(t)$ dependences reflect the fact that waiting time in zero field strongly affects the relaxation behavior and hence the magnetic viscosity. This behavior might indicate that there is a wider distribution of collectively relaxing magnetic entities than in a canonical homogeneous spin glass. Also, in canonical spin glasses a cross-over in the relaxation behavior for times $t \simeq t_w$ is usually found [12]. That is, $S(t) = \mathrm{d}M/\mathrm{d}\ln(t)$ displays a maximum at $t \simeq t_w$. In the present case, however, we see either a saturation of $S(t)$ for later times in the case of the data with $t_w = 10$ s or a broad maximum in the case of the data with $t_w = 3600$ s. Thus, we do not see a clear cross-over effect in our measurements. This is probably due to a broad distribution of relaxation times caused by inhomogeneous distribution of magnetization in the sample.



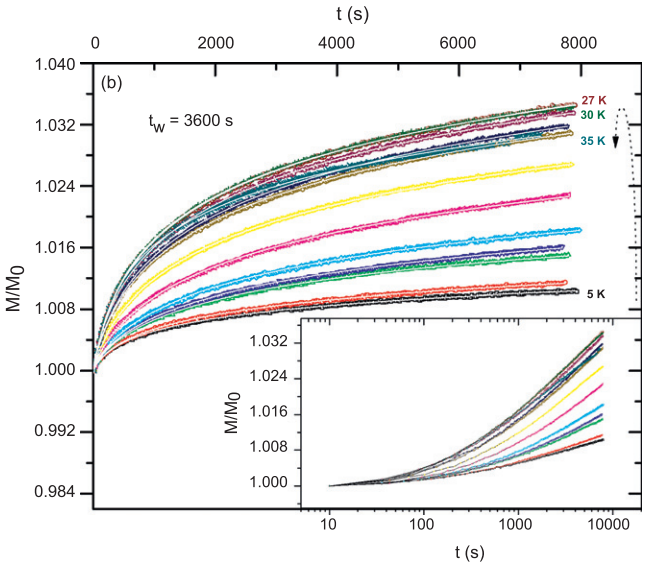


Figure 5. Normalized magnetization $M(t)/M(0)$ as a function of time for $5 \leqslant T \leqslant 35$ K. The measurements were conducted in an applied field of 100 Oe, subsequent to a waiting time (a) $t_w = 10$ s and (b) $t_w = 3600$ s in zero field. Solid white lines are the results of fits to equation (1) as described in the text. Insets present corresponding data on a logarithmic timescale (lower axis).

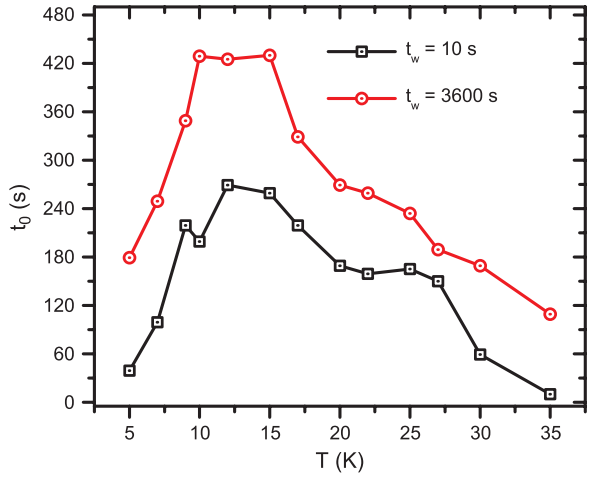


Figure 6. The temperature dependence of t_0 for two different waiting times obtained from a fit according to equation (1) to the $M(t)$ data.

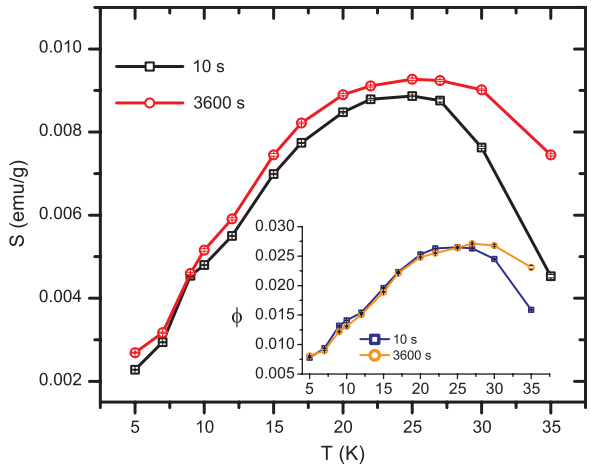


Figure 7. Magnetic viscosity S obtained from a fit according to equation (1) to the $M(t)$ data. Inset shows the parameter ϕ obtained from fitting $M(t)$ to equation (2). Both S and ϕ exhibit a peak at $T < T_{sg}$.

Alternatively, we note that an expression for a logarithmically slow relaxation of power-law distributed entities,

$$M(t) = M(0)[1 + \ln(1 + t/\hat{t}_0)]^{\phi}, \qquad (2)$$

also yields a satisfactory fit to the magnetization data, $M(t)$. However, these fits gave slightly poorer χ^2 (variance of residuals) when compared to those of equation (1). Equation (2) is inspired by similar expressions derived within the framework of the droplet-scaling theory [16]. Nevertheless, we here use this expression only to emphasize that the long-time $M(t)$ relaxation data are open to interpretation within the crude framework of thermally activated dynamics, described by a single viscosity coefficient S, or that some more complex distributions of barriers and magnetic entities may underlie the experimental observations. The inset of figure 7 depicts the dependence of ϕ on temperature obtained from fits to equation (2). Obviously, both parameters S and ϕ exhibit a similar dependence on temperature and, in particular, peak at $T \approx 25$ K $< T_{sg}$. A maximum in $S(T)$ below the spin glass freezing temperature T_{sg} is, however, a characteristic feature not only of cooperative dynamics of magnetic clusters, as earlier found in a dilute frozen ferrofluid with dipolar couplings [10], but also in systems of non-interacting magnetic nanoparticles [33]. However, glassy magnetic systems with cooperative dynamics show marked differences in $M(t)$ for different t_w, as is indeed observed in our experiments. Notably, the magnitude of the time-dependent magnetization response $M(t)$ below T_{sg} depends crucially on the waiting time. This can clearly be seen from figure 8, where $M(t)$ is plotted for temperatures below as well as above the spin glass transition temperature. In the spin glass state $T < T_{sg}$, the magnetization depends crucially on the time t_w period

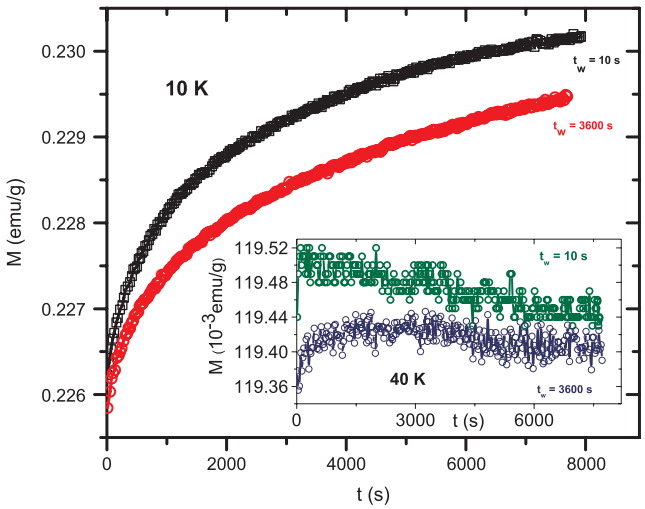


Figure 8. Magnetization $M(t)$ versus time measured at 10 K and for different waiting times $t_w = 10$ and 3600 s. Inset presents the data for similar ZFC waiting time experiments at 40 K, a temperature which is above the spin glass freezing temperature.

spent at ZFC condition before applying a magnetic field and commencing measurement. In contrast, at $T > T_{sg}$ there are only tiny differences (note the scale) which likely are due to artifacts of the measurement. We emphasize that this method of characterizing the spin glass state is highly reliable. It clearly demonstrates that the time evolution of the magnetization at temperatures below T_{sg} takes place through slow collective spin dynamics.

4. Conclusions

$Dy_{0.5}Sr_{0.5}MnO_3$ exhibits many characteristics of a real spin glass system like critical slowing down, memory and aging effects. However, closer inspection of these dynamic properties reveals distinct deviations from the properties seen in canonical spin glasses or other spin-glass-like systems with atomic spins and frustrated couplings. In particular, the microscopic spin flip time τ_0 in $Dy_{0.5}Sr_{0.5}MnO_3$ obtained from dynamic scaling is orders of magnitude larger than the usually reported values both for manganite spin glasses or for other canonical spin glass systems [20]. If we interpret this characteristic 'microscopic' timescale of about 10^{-8} s as an attempt frequency to flip a magnetic entity in the Mn–O sublattice, then it becomes clear that the dynamics in $Dy_{0.5}Sr_{0.5}MnO_3$ are not ruled by individual flips of atomic spins. Rather, relatively large, strongly-coupled clusters seem to undergo magnetic flips. We must emphasize that the observed magnetic phenomena derived from the Mn sublattice, are largely operating in a background which is a paramagnetic Dy lattice. In addition, the memory effects in this material appear to be much slower than similar relaxation effects in canonical spin glasses. This is evident, e.g., by the absence of any clear cross-over of the relaxation rate for the waiting time experiments.

Dynamical scaling and various memory and aging effects observed in the relaxation of the magnetization confirm the collective dynamics. Further features of the magnetic relaxation dynamics, such as the incomplete memory after large temperature quenches and the behavior of magnetic viscosity in remagnetization experiments after zero-field cooling also indicate a magnetically-clustered state. The dependence of $M(t)$ on the waiting time after zero-field cooling rules out the presence of an assembly of superparamagnets which may otherwise explain the observed memory and aging effects. Our results suggest that the half-doped manganite $Dy_{0.5}Sr_{0.5}MnO_3$ with small rare earth and, therefore, strong lattice distortions undergoes freezing into a magnetically inhomogeneous clustered state. It remains an open issue to what extent structural inhomogeneities, induced by the large size mismatch of the Dy and Sr ions on the A-site and electronic inhomogeneities in the sense of a nanometer-scale phase separation conspire in this material to induce such a magnetic state.

Acknowledgments

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