Magnetic relaxation phenomena and cluster glass properties of La$_{0.7-x}$Y$_x$Ca$_{0.3}$MnO$_3$ manganites

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The dynamic magnetic properties of the distorted perovskite system La$_{0.7-x}$Y$_x$Ca$_{0.3}$MnO$_3$ ($0 \leq x \leq 0.15$) have been investigated by ac-susceptibility and dc magnetization measurements, including relaxation and aging studies. All investigated samples display a metal-insulator transition. As yttrium is added in the compounds the overall results show evidence for the gradual appearance of a cluster glass behavior. For the $x = 0.15$ sample, magnetization measurements as a function of time at various temperatures show that the magnetic relaxation is maximum at a given temperature, well below the ferromagnetic transition. This maximum coincides in temperature with a frequency dependent feature in the imaginary part of the ac susceptibility, associated with a freezing process. This is interpreted as due to ferromagnetic clusters, which grow with decreasing temperature down to a temperature at which they freeze due to severe intercluster frustration.

PACS numbers: 75.50.Lk, 75.30.Vn, 72.80.Ga

I. INTRODUCTION

Extensive study of the doped perovskite manganites La$_{1-y}$Ca$_y$MnO$_3$ revealed a very rich and complex phase diagram, which includes colossal magnetoresistance (CMR) behavior near the transition temperature of the metallic ferromagnetic compositions, $0.15 < y < 0.50$. The ferromagnetic (FM) interaction appears as a consequence of the manganese mixed valence state. Substitution of La$^{3+}$ by Ca$^{2+}$ in antiferromagnetic LaMnO$_3$ changes the Mn$^{3+}$/Mn$^{4+}$ ratio and produces holes in the e$_g$ orbitals. The simultaneous para-ferromagnetic and metal-insulator transitions are basically understood within the framework of the double-exchange theory, which considers the transfer of one e$_g$ electron between neighboring Mn$^{3+}$ and Mn$^{4+}$ ions through the path Mn-O-Mn.

For a given e$_g$ concentration the tolerance factor of the perovskite structure, proportional to the average ionic size of the A site, was shown to have a drastic influence on the physical properties of manganites. Compounds of the type (La$_{0.7-x}$R$_x$)Ca$_{0.3}$MnO$_3$ (R = Pr, Y, Dy, or Tb), where the Ca concentration is close to an optimum value in relation to the FM interaction, are particularly suited to investigate this effect: replacing La$^{3+}$ with smaller rare-earth ions decreases the Mn-O-Mn bond angle, and thus affects the hopping of the e$_g$ electrons and weakens the double-exchange interaction, while the antiferromagnetic superexchange term is only marginally affected and therefore increase in importance. Consequently, both double exchange and superexchange interactions will compete more strongly depending on the structural distortion, giving rise to a magnetically disordered state. A magnetic and electronic phase diagram of several manganite systems plotted against the tolerance factor has been published on several investigations.

Lowering the value of the average ionic size of the A site initially reduces the transition temperature $T_C$, and below a critical value the low temperature phase changes to a spin-glass insulating state. Preliminary studies have additionally shown that the FM metallic phase of La$_{0.7-x}$Y$_x$Ca$_{0.3}$MnO$_3$ ($x = 0.15$) display signatures of a magnetic cluster glass state.

The purpose of the present work is to investigate the dynamic properties of this disordered phase by means of magnetic relaxation measurements. One characteristic feature of glassy systems is that the magnetization of a sample cooled in zero field to a certain temperature depends on how long it is held in zero field before the magnetic field is applied. This is the so called aging effect. Furthermore, if $M(t)$ is differentiated with respect to $\ln t$, the derivative $S$
= \partial M/\partial(\ln t) \) displays a maximum which shifts to higher \( t \) for longer values of the wait time, \( t_w \). These effects have been observed on a well characterized cluster glass compound, the cobaltite \( \text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3 \), where glassy behavior and intracluster ferromagnetism were shown to coexist. More recently, on \( \text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \), it was shown that disorder and frustration may occur in the ferromagnetic phase. Within this context, this paper presents low temperature magnetic relaxation measurements on \( \text{La}_{0.7-x}\text{Y}_x\text{Ca}_{0.3}\text{MnO}_3 \), with \( 0 \leq x \leq 0.15 \). As yttrium is added in the compound, the results evidence for the first time the gradual appearance of glassy signatures within the ferromagnetic metallic phase, prior to the existence of the insulating spin glass phase which occurs with higher rare-earth doping at the La site. Relaxation measurements probe the out-of-equilibrium state of the system, where the clear observation of a peak in the magnetic relaxation as a function of temperature and the observation of aging effects further characterize the cluster glass properties of this system.

II. EXPERIMENTAL

The investigated compounds are polycrystalline samples of \( \text{La}_{0.7-x}\text{Y}_x\text{Ca}_{0.3}\text{MnO}_3 \), with \( x = 0, 0.07, 0.10 \) and 0.15, prepared by standard solid state reaction. X-ray analysis confirmed a single phase orthorhombic perovskite structure. Magnetic measurements were made with an extraction magnetometer (Quantum Design PPMS) when the applied field was greater then 5 mT. Low field data, \( H = 0.3 \) mT, was taken with a SQUID magnetometer (Quantum Design MPMS). Zero field cooled relaxation measurements were performed on all samples at different temperatures between 10K and 250 K, and for applied fields varying from 5mT to 0.5T. The samples were cooled from a reference temperature in the paramagnetic state with a constant cooling rate, and kept at a target temperature for a certain wait time \( t_w \). Thereafter, a dc field was applied and the magnetization was recorded vs. the elapsed time, \( t \). The samples were additionally characterized by transport measurements, made by a conventional ac four-probe method. The results are shown on Fig. 1. All samples display a metal-insulator transition, below which a metallic-like electrical conductivity is observed. A similar behavior was previously reported for these compounds.

III. RESULTS AND DISCUSSION

To recall the characteristic magnetic behavior of \( \text{La}_{0.7-x}\text{Y}_x\text{Ca}_{0.3}\text{MnO}_3 \) compounds, Fig. 2 displays the zero field cooled (ZFC) and field cooled (FC) temperature dependence of the dc magnetization of the samples \( x = 0, 0.07, 0.10 \) and 0.15, measured at \( H = 5 \) mT. The small hump visible at low temperatures in most of the FC data is due to a problem in the experimental apparatus. The results of Fig. 2 clearly show a standard FM transition for the \( x = 0 \) sample at \( T_C = 252 \) K, as observed on numerous investigations. For the doped compounds \( (x > 0) \) the ZFC curves evolve continuously, with higher yttrium concentration, to a cusp-like anomaly at a temperature \( T_A \) just below the Curie-like temperature \( T_C \), defined as the maximum inflection of the FC data. The \( T_C \) values obtained are 143, 114, and 89 K, for \( x = 0.07, 0.10, \) and 0.15 respectively. An important point to be noted here is that the FC magnetization continues to increase strongly below the irreversibility temperature, \( T_r \), at which ZFC and FC curves merge, a typical feature of various cluster glass systems. This effect is more pronounced on the \( x = 0.15 \) sample. On the other hand, in canonical spin glass systems the FC magnetization shows a nearly constant value below \( T_r \). It worth noting that due to the fact that cluster glasses exhibit finite range ferromagnetic ordering below \( T_C \), this system may show some features similar to those found in reentrant spin glass systems (RSG), which undergo a PM-FM transition at \( T_C \) and have a lower freezing temperature. Nevertheless, it may be noted that in several RSG systems, such as \( \text{NiMn} \), \( \text{FeMn} \), \( \text{AuFe} \), and \( \text{FeZr} \), the irreversibility temperature \( T_r \) occurs at temperatures well below \( T_C \) (when the measuring field is higher then the coercive field), whereas in a cluster glass the irreversibility arises just below \( T_C \), as observed in Fig. 2 for the yttrium doped samples.

On the other hand, magnetization curves measured as a function of field, \( M(H) \), do show signatures of a RSG behavior. The low field portion of a typical hysteresis loop measured after ZFC condition at 2.0 K, a temperature well below \( T_r \), is shown in the inset of Fig. 3. The curve displays an \( S \) shape in the virgin branch, with a positive curvature at low fields, a typical characteristic of both canonical SG and RSG systems. In the same way, the variation of the coercive field \( H_C \) with temperature, plotted in Fig. 3, is quite similar to those usually observed in RSG systems, where \( H_C \) increases rapidly as the temperature is lowered below \( T_C \). Such conflicting results evidence a more complex glassy behavior in these compounds.

Susceptibility measurements are a very efficient way to evidence glassy behavior. The temperature dependence of the ac susceptibility of \( \text{La}_{0.55}\text{Y}_{0.15}\text{Ca}_{0.3}\text{MnO}_3 \) is presented on Fig. 4. The variation with temperature of the in-phase susceptibility \( \chi' \), shown on the inset of the figure, is comparable to that of the dc \( M_{ZFC} \), with an onset around
120K related to the paramagnetic to ferromagnetic transition, followed by a maximum at T = 79 K, close to the MZF C cusp temperature Tc = 72 K. This maximum is not frequency dependent, which suggests that the FM state originates from intracluster ferromagnetism. Moreover, if the χ′ maximum and Tc are related, the cusp on the MZF C curve is the dc signature of these FM interactions, rather than a consequence of cluster freezing. The out-of-phase susceptibility χ′′ also shows a maximum at T = 87 K, which is frequency independent. It has been suggested [8] that the temperature at which this maximum occurs is related to the irreversibility temperature, Tr = 88 K, obtained on the dc magnetization curves. Therefore the maximum in χ′′ at higher temperatures is also related to the intracluster FM interactions. However, an additional feature clearly visible in χ′′ is the presence of a hump, at Tf ∼ 40K. This broad peak is frequency dependent and shifts towards higher temperatures with increasing frequency, a characteristic feature of the dynamics of spin glass systems. A similar behavior has previously been reported in other manganite samples [20]. This double peak structure in χ′′ is progressively less visible for lower Y doping, and it is not present in the Y=0 sample.

In order to gain further information on the underlying nature of this cluster glass system, we have measured the long time relaxation of the magnetization, with a time scale greater than 10^4 s. Figure 5a displays the normalized M(t)/M(0) data for all samples measured at a reduced temperature T/Tc = 0.2, with H = 5 mT (this probing field is within the linear response regime). As Y is added in the compounds the data show a gradual increase of M(t)/M(0), at any given time. Quantitatively, after 10^4 seconds, the fractional change of the magnetization is 0.18, 0.93, 4.5, and 12% in the samples with x = 0, 0.07, 0.10, and 0.15 respectively. Relaxation data at different temperatures for La0.55Y0.15Ca0.3MnO3 are shown in Fig 5b. The slope of M(t)/M(0) increases below Tc, reaches a maximum at T/Tc ≃ 0.4, and decreases for lower temperatures. This evolution of the magnetization relaxation behavior with temperature is indicative of the development of the magnetic clusters as temperature is lowered. It is noteworthy that relaxation is observed not only below the freezing temperature Tc, but also up to the FM transition temperature Tc, which indicates that the system is not in a true ferromagnetic state. This is reinforced with measurements of M(H) above and below Tc (not shown), where Arrot plots M² vs. H/M do not yield straight lines. Previous observations in RSG system [21] also show that relaxation is present above the freezing temperature of the spin glass state.

Amongst the various functional forms that have been proposed to describe magnetization as a function of time, one of the most popular is a stretched exponential of the form:

\[ M(t) = M_0 - M_r \exp \left( -\left( \frac{t}{\tau_r} \right)^{1-n} \right) \]  

(1)

where M₀ relates to an intrinsic FM component, and Mᵣ to a glassy component mainly contributing to the relaxation effects observed. The time constant τᵣ and the parameter n are related to the relaxation rate of the spin-glass like phase. The values of n are scattered between 0.48 and 0.6, in agreement with previous results [20]. The variations of M₀ and Mᵣ with reduced temperature are shown on Fig. 6 for the samples with x = 0.10 and 0.15. As expected, M₀ and Mᵣ depend strongly upon temperature. The dependence of M₀ with T/Tc evidence the FM transition at T/Tc ≃ 1. As observed on the temperature dependence of MZF C, the FM component first reaches a maximum at T/Tc ≃ 0.8 that corresponds to Tc (MZF C cusp temperature) and decreases further with decreasing the temperature. The FM component M₀ is larger for x = 0.10 compared to x = 0.15, in agreement with its stronger FM behavior. The variation of Mᵣ with T/Tc shows that for x = 0.15 the relaxing component first increases from T/Tc ≃ 1 to T/Tc ≃ 0.45, as a consequence either of an increase of the clusters number or of the growth of the clusters size, and decreases for T/Tc <0.45. The resulting maximum in Mᵣ coincides with the temperature of the frequency dependent lower maximum in the out-of-phase susceptibility, which supports the idea that it is related to the beginning of cluster freezing. A similar behavior is observed for x = 0.10, but at a lower temperature, with the maximum in Mᵣ occurring around T/Tc ≃ 0.20. The relaxing component Mᵣ in x = 0.10 sample is also much lower than in x = 0.15, due to a less pronounced glassy behavior. The relaxation time τᵣ is extremely sensitive to any noise in the data. However, our fitting (not shown) indicate that τᵣ increases with decreasing temperature for both x = 0.15 and x = 0.10, suggesting a regular stiffening of the spin relaxation, which is compatible with an increase of the clusters size and enhanced intercluster frustration.

To further investigate the dynamic magnetic behavior of these metallic glass compounds the wait time tᵣ dependence of the long time relaxed magnetization, or aging effect, was studied. The results were obtained with a small dc field, H = 0.3 mT. Additional measurements (not shown) as a function of the applied field revealed that the magnitude of aging effect is reduced when the field increased, and totally disappears at H = 10 mT. Magnetization vs. time was measured with wait times tᵣ = 100, 1000, and 10000 s, before application of the magnetic field. Results for the x = 0.10 sample, measured at T/Tc = 0.2, are plotted in Fig. 7a. This temperature was chosen because it corresponds to the maximum in the relaxing component Mᵣ. It is clear from the figure that the measured magnetization strongly
depends on \( t_w \), which confirms the existence of aging processes. Macroscopically, aging means that the system becomes “stiffer” for larger wait time, i.e., the measured magnetization is lower for higher \( t_w \), as observed in the data. The corresponding time dependent relaxation rate, \( S(t) = \partial M/\partial (\ln t) \), is plotted in Fig. 7b. The \( S(t) \) curves were obtained by taking the derivative of a polynomial fit of the magnetization data. It is readily observed that \( S(t) \) reaches a maximum, corresponding to an inflection point on the \( M(t)/M(0) \) curves, that shifts to longer observation times for longer values of \( t_w \). These results are similar to those reported for various glassy systems. \([3],[12],[22],[23]\] It may be pointed out that the maximum in \( S(t) \) is not centered on \( t_w \) as expected. However, it is believed that this is related to the additional time required for stabilizing the temperature and the magnetic field applied.

For La\(_{0.55}\)Y\(_{0.15}\)Ca\(_{0.3}\)MnO\(_3\), \( S(t) \) has been calculated at various temperatures, and the results are shown in Fig. 8. Aging effects are observed at \( T/T_C = 0.2, 0.4 \) and \( 0.6 \). The wait time dependence of the peak in \( S(t) \) clearly increases with temperature. Aging effects are more pronounced at higher temperatures, i.e., when the FM state is stronger, as reported previously for La\(_{0.5}\)Sr\(_{0.5}\)CoO\(_3\) \([8]\) or for the two-dimensional ferromagnet Rb\(_2\)Cu\(_{0.89}\)Co\(_{0.11}\)F\(_4\). \([24]\) This may be explained by a more steady cluster growth in the FM phase. Larger clusters give rise to a slower response due to larger free-energy barriers and, correspondingly, a larger number of spins to be simultaneously flipped; the smaller the cluster, the more rapidly it will relax. At \( T/T_C = 0.2 \) on the contrary, the relaxation is slow (Fig. 6b) and the aging effect small, because the regular cluster growth with decreasing temperature lead eventually to a blocking of the clusters at \( T_f \) and below. It may also be noted in the data that the maximum in \( S(t) \) occurs at higher observation times for lower temperatures. This effect is mostly experimental in origin at low \( t_w \) (10\(^2\) seconds), because \( t_w \) is of the same order of the time it takes to reach the measuring temperature, which effectively increases the wait time at low temperatures. However, at high \( t_w \) (10\(^4\) seconds), the wait time is much higher than the time for temperature stabilization, and the shift in the maximum of \( S(t) \) with temperature may be associated with the slower response time of the clusters at low temperatures. It is worth mentioning that the measuring field of 0.3 mT may not be in the linear regime, which could affect these results. However, larger fields, outside the linear regime, mostly change the intensity but not the position of the maximum in \( S(t) \).

**IV. CONCLUSIONS**

The peak observed in the temperature dependence of the relaxing component of the magnetization, \( M_r \), or equivalently a maximum in the slope of \( M(t)/M(0) \) at a given temperature (Fig. 5b), combined with the aging effects observed for La\(_{0.55}\)Y\(_{0.15}\)Ca\(_{0.3}\)MnO\(_3\), and, to a lesser extend for La\(_{0.6}\)Y\(_{0.10}\)Ca\(_{0.3}\)MnO\(_3\), evidence the importance of magnetic frustration in these samples and establish the existence of cluster glass properties over a broad temperature range. The enhanced lattice distortion resulting from the presence of cationic size mismatch on the perovskite A-site weakens the Mn\(^{3+}\)-O-Mn\(^{4+}\) FM double-exchange interactions and favors frustration between the ferromagnetic and antiferromagnetic superexchange interactions. This competition is sufficient to suppress long range FM order, giving rise to the appearance of spatially confined ferromagnetic clusters, which in turn are responsible for the observed glassy behavior of the system due to frustrated interaction amongst their magnetic moments. It is important to stress out that the observed cluster glass behavior occurs within the metallic phase, as opposed to the standard spin glass phase of the manganites, which occurs in the insulating phase. From the behavior of \( M_r \), it is inferred that clusters start to form just below the FM transition \( T_C \). As the temperature is lowered, the size and/or number of these magnetic clusters increase, leading to a maximum in the relaxing component of the magnetization. The slower (weaker) relaxation at lower temperatures can be attributed to the freezing of the clusters, similar to the blocking of the magnetic moments in a superparamagnet, due to an enhanced intercluster frustration resulting from oversized domains.

The overall relaxation results presented here demonstrate the existence of a magnetic glassy behavior within the metallic phase, which plays an important role in the physics of these compounds. An important issue which remains to be verified is whether the observed cluster glass properties in La\(_{0.7-x}\)Y\(_{x}\)Ca\(_{0.3}\)MnO\(_3\) develop uniformly distributed due to disorder or enhanced canting within the FM phase, or possibly due to phase separated spin glass regions in a FM background. It is also worth noting that \( M \) vs. \( H \) measurements are similar to that obtained in RSG systems, indicating the existence of more complex frustration effects and disorder in this system. Within the phase segregation scenario, it is possible that the FM clusters and the FM background give rise to different glassy behavior.

**V. ACKNOWLEDGMENTS**

We thank F. Wolff and P. Pureur for assistance with the SQUID measurements, and M.A. Gusmão for helpful discussions. This work was partially financed under the contract PRONEX/FINEP/CNPq no 41.96.0907.00. Additional
support was given by FUJB. The sample preparation was funded by the UK-EPSRC.

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Figure 1 - Temperature dependence of the resistivity of the studied compounds.

Figure 2 - Field-cooled (solid lines) and zero-field-cooled (dotted lines) dc magnetization of La$_{0.7-x}$Y$_x$Ca$_{0.3}$MnO$_3$, measured with 5 mT.

Figure 3 - Temperature dependence of the coercive field, $H_C$, obtained from M(H) measurements after the application of $H = 9$ T. The inset shows the low field portion of the M(H) measurements taken at 2.0 K. The lines are only guide to the eyes.

Figure 4 - Out-of-phase susceptibility ($\chi''$) of La$_{0.55}$Y$_{0.15}$Ca$_{0.3}$MnO$_3$, measured with an ac field $h_{ac} = 0.5$ mT and frequencies $f = 90$ (circles), 250 (squares), 700 (triangles), and 2000 Hz (diamonds). The position of a frequency dependent feature in the data is indicated by $T_f$. The inset shows the in-phase susceptibility ($\chi'$) of the same sample.

Figure 5 - (a) Normalized zero field cooled magnetization of La$_{0.7-x}$Y$_x$Ca$_{0.3}$MnO$_3$, measured as a function of time, with $H = 5$ mT at a reduced temperature $T/T_C = 0.2$; (b) The same measurement for La$_{0.55}$Y$_{0.15}$Ca$_{0.3}$MnO$_3$, at $T/T_C = 0.1, 0.4,$ and 0.8.

Figure 6 - Evolution of the fitting parameters $M_0$ and $M_r$ (see text for details) as a function of a reduced temperature $T/T_C$, for $x = 0.10$ and 0.15 samples.

Figure 7 - (a) Zero field cooled relaxation magnetization, and (b) the corresponding relaxation rate $S(t) = \partial M/\partial \ln t$, of La$_{0.60}$Y$_{0.10}$Ca$_{0.3}$MnO$_3$, measured at $T/T_C = 0.2$ with $H = 0.3$ mT, after different wait times $t_w = 10^2$, $10^3$, and $10^4$ s.

Figure 8 - Relaxation rate $S(t) = \partial M/\partial \ln t$ of La$_{0.55}$Y$_{0.15}$Ca$_{0.3}$MnO$_3$ measured after different wait times $t_w = 10^2$ (open circles) and $10^4$ (solid circles) at $T/T_C = 0.2, 0.4, 0.6$. The arrows indicate the peak position of each data curve.
La_{0.7-x}Y_{x}Ca_{0.3}MnO_{3}

\( x = 0.07 \)
\( x = 0.10 \)
\( x = 0.15 \)
$x = 0.15$

$\chi''(10^4 \text{ emu/g})$

$\chi'(10^2 \text{ emu/g})$

$T_f$

$T(K)$

$T(K)$
(a) $T/T_c = 0.2$

X = 0.15

(b) $T/T_c = 0.4$

X = 0.15

$T/T_c = 0.1$

$T/T_c = 0.8$
(a) $M (10^{-2} \text{ emu/g})$

- $x = 0.10$
- $t_w = 10^2 \text{ s}$
- $t_w = 10^3 \text{ s}$
- $t_w = 10^4 \text{ s}$

(b) $S (t)$

- $t_w = 10^2 \text{ s}$
- $t_w = 10^3 \text{ s}$
- $t_w = 10^4 \text{ s}$
$T/T_c = 0.6$

$t_w = 10^4 \text{s}$

$T/T_c = 0.4$

$t_w = 10^2 \text{s}$

$T/T_c = 0.2$