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Magnetic aging above the freezing temperature in $La_{0.82}Sr_{0.18}CoO_3$

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Abstract
We have performed a detailed study of magnetic relaxation in $La_{0.82}Sr_{0.18}CoO_3$ which shows spin-glass-like behavior at low temperature. Contrary to the case in a classical spin-glass where the collective relaxation behavior is observed only below the freezing temperature T_f, the magnetic relaxation in $La_{0.82}Sr_{0.18}CoO_3$ is found to be of the stretched exponential form not only below but also above T_f. Moreover, the aging effect is also observed above T_f. These results are interpreted in terms of the interactions between ferromagnetic clusters.

1. Introduction

Many glassy systems exhibit the feature of relaxation dynamics, i.e. so-called 'ergodicity breaking'. When the sample is cooled, it begins to fall out of thermodynamic equilibrium close to the freezing temperature, T_f, and be left in a state corresponding to a higher temperature. Through monitoring the time-dependent variation of physical quantities after quenching the sample below T_f, a nonstationary behavior described as 'physical aging' takes place. The physical quantities vary gradually with time when the sample is maintained in some fixed external conditions for a waiting time t_w, and they will relax towards its new equilibrium value in a way that reflects the aging or waiting time t_w when the external conditions are changed [1]. Generally, the aging phenomenon is observed below the freezing temperature T_f. In contrast, above T_f no aging can be observed in many glassy systems. That is, the waiting time dependence of relaxation vanishes in the experimental waiting time and observation time windows [2, 3].

The perovskite cobaltites $La_{1-x}Sr_xCoO_3$ have recently attracted considerable attention due to their intriguing magnetic behaviors, such as the existence of spin state transitions, and the unusual magnetic ground state and spin dynamics [4–9]. Substitution of La^{3+} by Sr^{2+} induces Sr-rich clusters and an La-rich matrix. Caciuffo *et al* [10] have observed Sr-rich clusters with sizes ranging from 8 to 40 nm. The Sr-rich clusters represent ferromagnetic metal due to double exchange between Co^{3+} and Co^{4+}, while the La-rich matrix is non-magnetic. Itoh *et al* [5] suggested that there exist spin-glass ($0 < x \lesssim 0.18$) and cluster-glass ($0.18 \lesssim x \leqslant 0.5$) regions for cobaltites. Furthermore, the spin relaxation behavior has been studied by some groups [5, 11–13]. In previous studies by Itoh *et al*, the long-time relaxation was observed not only in spin-glass regions but also in cluster-glass regions. However, the aging effect was observed only in spin-glass regions but not in cluster-glass regions [5]. In $La_{0.85}Sr_{0.15}CoO_3$, magnetic aging is found to exist in the susceptibility both above and below the frequency-dependent maximum temperature similarly to the re-entrant spin-glass phase, but the slowing down of the dynamics does not support the existence of a re-entrant spin-glass phase [11]. Recently, long-time relaxation and the aging phenomenon were reported in $La_{0.5}Sr_{0.5}CoO_3$. In particular, the aging effect is more pronounced at high temperature [12].

These results indicate that there exists an unusual temperature dependence of relaxation in $La_{1-x}Sr_xCoO_3$. However, the details of the temperature dependence of magnetic relaxation and the origin of aging at high temperature are still unclear. In this work, we carried out a detailed study on the relaxation behavior of $La_{0.82}Sr_{0.18}CoO_3$ at various temperatures. It was found that the magnetization as a function of time can be described well by a stretched exponential form both above and below T_f. Based on this study, we suggest that the interactions between ferromagnetic (FM) clusters are responsible for the aging effect above T_f in $La_{0.82}Sr_{0.18}CoO_3$.

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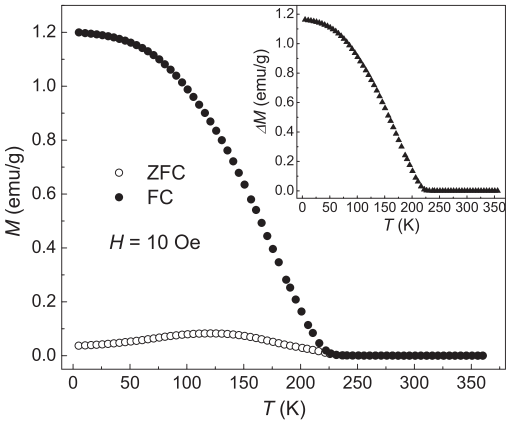

Figure 1. Field-cooled and zero-field-cooled dc magnetizations, measured with 10 Oe. The inset shows $\Delta M = M_{FC} - M_{ZFC}$.

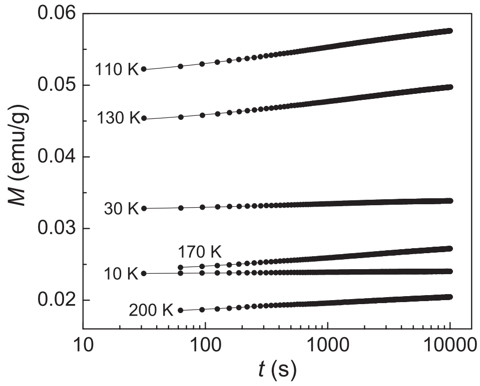

Figure 2. M versus t at different temperature. The solid lines are the best fits to equation (1).

2. Experiment

Experiments were performed on a ceramic sample of $La_{0.82}Sr_{0.18}CoO_3$ which was prepared by solid-state reaction method. A stoichiometric mixture of $SrCO_3$, Co_3O_4, and La_2O_3 powders was well ground and calcined twice at 800 and 950 °C for 24 h. Then, the resulting powder was pressed into pellets and sintered at 1100 and 1150 °C for 24 h, respectively. X-ray diffraction (XRD) shows that, at least within the sensitivity of XRD, the sample is single phase with rhombohedral structure. Also, the $M(T)$ behaviors shown below are similar to the results of other groups [5, 8], which further evidences that the sample is single phase. Using a superconducting quantum ineterference device (SQUID) magnetometer, dc magnetization measurements were performed. In order to obtain a low field, the superconducting magnets of the apparatus were demagnetized before measurements.

3. Results and discussion

Figure 1 shows the temperature dependence of magnetization measured on heating in a low magnetic field ($H = 10$ Oe) with the zero-field-cooled (ZFC) and field-cooled (FC) processes for $La_{0.82}Sr_{0.18}CoO_3$. Both the rate of cooling and the rate of heating are 5 K min^{-1} during measurement. The sample displays a bifurcation of the ZFC and FC magnetization at an irreversibility temperature T_{irr} of 230 K. Both the ZFC magnetization and FC magnetization rise sharply on lowering the temperature below 240 K, which is the Curie temperature T_C of the FM clusters. Also, the $M(T)$ behaviors obey the Curie–Weiss law above 240 K. The ZFC magnetization curve exhibits a maximum at ≈120 K, which is usually regarded as the freezing temperature T_f. The ZFC magnetization below T_{irr} corresponds to nonequilibrium. However, the effect of magnetic relaxation could not influence the measured $M(T)$ dependences and could not mask the freezing temperature due to the small measuring time, which is only 1 min for every data point. Two other important points to be noted here are that the FC magnetization is much larger than the ZFC magnetization below T_{irr} and that the FC magnetization continues to increase strongly below T_f rather than showing a plateau. These characteristics can be expressed explicitly by ΔM, which is defined as $\Delta M = M_{FC} - M_{ZFC}$, where M_{FC} and M_{ZFC} are the FC and ZFC magnetizations respectively, as shown in the inset of figure 1. All these results indicate that there exists a collective freezing process of the moments of FM clusters. Also, the freezing process begins even above the freezing temperature T_f, as suggested by the difference between M_{FC} and M_{ZFC} in the temperature range $T_f \leqslant T \leqslant T_C$.

In order to obtain further information on the spin dynamics of this sample, we have carefully measured the long-time relaxation of the magnetization. The sample was cooled from 360 K to a measurement temperature in zero field. Then a small field ($H = 5$ Oe) was applied and the magnetization M was recorded versus time t. For clarity, only some representative results are plotted in figure 2. It is noteworthy that the glassy-relaxation behavior is present not only below the freezing temperature T_f, but also above T_f up to the Curie temperature T_C of the FM clusters. This behavior was also previously observed in some manganites and cobaltites [11, 12, 14]. Among the various functional forms proposed to describe the relaxation behavior in spin-glass, one of the most popular relations is the stretched exponential function

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

where M_0 relates to an intrinsic FM component. The glassy component M_r and the time constant τ_r depend on the temperature and waiting time t_w. l is only a function of temperature. We found that all of the magnetic relaxation curves $M(t)$ can be described perfectly by the stretched exponential function. The solid lines in figure 2 are the best fits with equation (1). The fitting parameters are shown in figure 3. As expected, M_0, M_r and τ_r depend strongly on temperature. All of them first increase until reaching a maximum and then decrease further with decreasing temperature. The resulting maxima in M_0 and M_r coincide with the freezing temperature T_f. Because the measured value of M_0 is decided by the measuring condition, such as the measuring process, applied field and temperature, it decreases with a decrease in

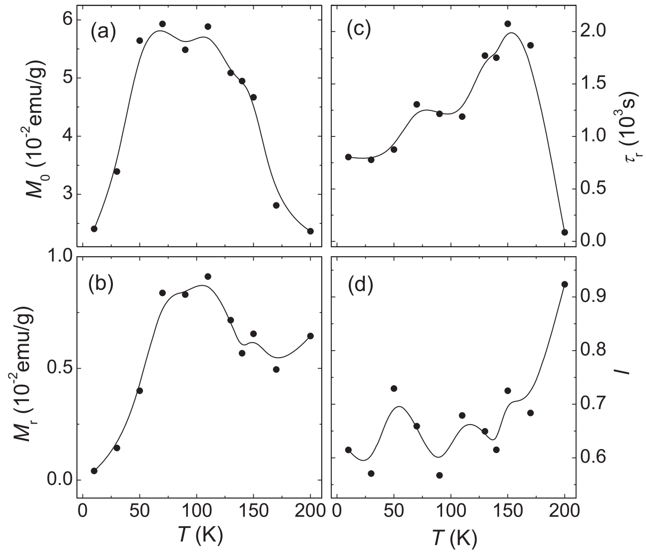

Figure 3. The fitting parameters M_0, M_r, τ_r and l as a function of temperature.

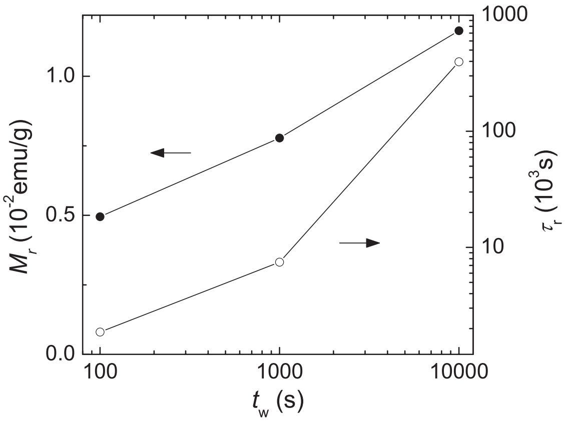

Figure 4. The fitting parameters M_r, and τ_r as a function of the waiting time t_w.

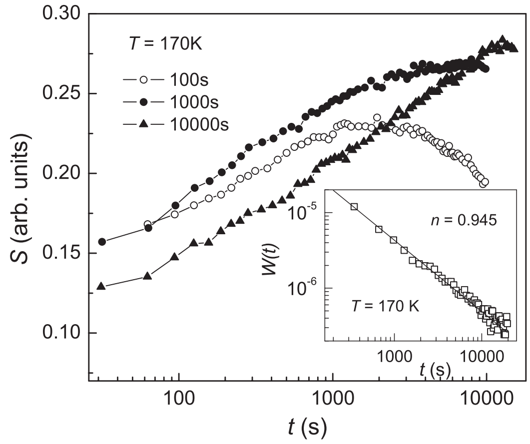

Figure 5. S versus log t at 170 K for different waiting times. The inset shows W versus t at 170 K.

temperature below 100 K, although M_0 relates to the intrinsic FM component. We also noticed that M_r at high temperature is not too small and the maximum in τ_r is at about 150 K, which is higher than T_f. The large value of M_r at high temperature further supports the idea that the spin-freezing process takes place even above T_f. The large value of τ_r above T_f indicates that there still exists a strong intercluster interaction above T_f. For l, the values fluctuate around 0.65, and a rapid increase is observed at 200 K, in agreement with previous observations [14, 15].

Since the spin-freezing process and magnetic relaxation take place even above the freezing temperature T_f, an aging effect is also expected. We studied the waiting time t_w dependence of the long-time-relaxed magnetization. The sample was cooled from 360 K to a measurement temperature of 170 K in zero field. The magnetization M versus time t was recorded with waiting times $t_w = 100$, 1000, and 10 000 s, before application of a small field ($H = 5$ Oe). All of the magnetic relaxation curves $M(t)$ (not shown) can also be described perfectly by the stretched exponential function. Figure 4 shows that both M_r and τ_r strongly depend on the waiting time t_w, in agreement with the behavior of a classical spin-glass below the freezing temperature. With an increase in the waiting time t_w, both M_r and τ_r increase rapidly, indicating a stiffening of the spin relaxation. To further illustrate a waiting time dependence of the magnetic relaxation, it is convenient to use the relaxation rate S, defined as $S = (1/H)(\partial M/\partial \ln t)$. S versus $\log(t)$ is plotted in figure 5 for different waiting times. It is clear that the magnetic relaxation depends on the waiting time. A maximum in $S(t)$ occurs at a time which is approximately equal to the waiting time t_w, implying an aging effect. It is noticed that the time of the maximum is larger than the waiting time t_w, especially for $t_w = 100$ and 1000 s. It is possible that this is related to the additional time required for stabilizing the temperature and the applied field when performing the relaxation measurements.

In our previous study, we demonstrated that both a spin-glass-like phase and the interactions between FM clusters contribute to the glassy magnetic behaviors at low temperature in $La_{1-x}Sr_xCoO_3$ [16]. It seems that the intercluster interactions begin to gradually appear below T_C and above T_f with the growth of FM clusters. Thus, these interactions induce spin freezing and magnetic relaxation. To prove these ideas further, we measured the thermo-remanent magnetization for our sample with the following procedure: cooling the sample in a 10 Oe field from 360 to 170 K and then switching off the field after waiting 10 000 s, and subsequently measuring $M(t)$. Recently, in studying interacting magnetic particles, Ulrich *et al* [17] proposed a model and demonstrated that for all particle densities the relaxation rate, $W(t) = -(\mathrm{d}/\mathrm{d}t) \ln M(t)$, decays by a power law, with a density-dependent exponent n:

$$W(t) = At^{-n}. \tag{2}$$

The theoretical prediction was recently corroborated by magnetic relaxation measurements in a superferromagnetic granular multilayer [18] and PS manganites [19]. It is found that our $M(t)$ data can also be described by the theory mentioned above. As shown in the inset of figure 5, these open squares present our experimental results, and the solid line is the best fit of equation (2). The fitting parameter n is 0.945, which is even larger than the value of n below the freezing temperature, reported in our other work [16]. Since n is a parameter reflecting the strength of interaction, the larger value of n indicates a much large strength of the intercluster interaction. This result is consistent with the result that a large τ_r value is obtained at 170 K. Furthermore, it confirms that the intercluster interactions are present above the freezing temperature T_f and induce spin freezing and magnetic relaxation.

4. Conclusion

The dynamics of the magnetic properties of $La_{0.82}Sr_{0.18}CoO_3$ have been studied by dc magnetization measurements, including ZFC and FC relaxation. The ZFC relaxation results can be described well by the stretched exponential relationship and the FC relaxation rate follows a power law, which describes the relaxation in single-domain ferromagnetic particles. It is important to note that these relaxation behaviors occur even above the freezing temperature, T_f, and a significant aging effect is also observed above T_f. The large values of the fitting parameters τ_r and n above T_f indicate strong intercluster interactions. It is suggested that the intercluster interactions are responsible for the magnetic relaxation and aging above the freezing temperature.

Acknowledgments

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