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Low-frequency noise in $La_{0.7}Sr_{0.3}Mn_{1-x}Fe_xO_3$ thin films

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Abstract

The low-frequency noise in epitaxial $La_{0.7}Sr_{0.3}Mn_{1-x}Fe_xO_3$ thin films with $x = 0$, 0.08 and 0.12 has been studied as a function of magnetic field (up to 1500 G). At zero external magnetic field ($B = 0$ G), there is no appreciable peak in the normalized noise power spectral density (PSD) for $x = 0$ thin films; however, a peak does appear for $x = 0.08$ and 0.12 thin films near the metal–insulator transition temperature (T_p), which is below the Curie temperature. Unlike the resistivity, the noise PSD does not change with the doping level (x) at $B = 0$ G; however, at $B = 1500$ G the noise PSD decreases with x. At a low external magnetic field of $B < 1500$ G, there is no or a small thermal hysteresis in the noise PSD for all three differently doped samples; however, at $B = 1500$ G there appears a large thermal hysteresis for all samples. The contribution of magnetic domain fluctuations is a possible origin for the noise PSD.

1. Introduction

Colossal magnetoresistance (CMR)—that is, an unusually large change of resistivity upon application of a magnetic field—has recently been extensively investigated in manganese oxides (manganites) and other transition metal oxides. This enthusiasm is prompted not only by the scientific interest of many anomalous features revealed in CMR oxides, but also by the potential for application of these materials in electrically readable magnetic field sensors, which are in great industrial demand [1–5]. In applications of manganite CMR materials, the intrinsic low-frequency noise of the sensors could limit the sensitivity of the devices in the detection of resistance changes induced by a magnetic signal. Thus, measurements of the $1/f$ noise may suggest applications of CMR materials and provide insight into their electronic transport properties [6–13].

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Among the studies of low-frequency noise in CMR materials, that by Podzorov *et al* [6] discovered an unprecedented magnitude of the $1/f$ noise near the Curie temperature (T_C) in low-T_C manganites: polycrystalline and single-crystal bulk samples of $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ with $x = 0.35$. Podzorov *et al* suggested that the so-called Curie temperature in the low-T_C materials is a percolation transition temperature rather than the long-range ferromagnetic phase transition temperature. The scaling analysis of the $1/f$ noise is consistent with the percolation model of conducting domains randomly distributed in an insulating matrix. Recently, giant random telegraph noise (RTN) was observed in $La_{2/3}Ca_{1/3}MnO_3$ (LCMO) thin films [9, 10] and bulk single-crystal samples [10]. It is suggested that the giant RTN is related to the fluctuation of material between more and less conductive local states. Reutler *et al* [13] measured the noise in LCMO thin films on different substrates and found that different amounts of lattice mismatch can significantly influence the noise properties.

Studies of Fe-doped manganites focusing on the $La_{1-y}Ca_yMnO_3$ and $La_{1-y}Sr_yMnO_3$ systems are scarce. Moreover, as far as we know, all research on Fe-doped manganites has been on polycrystalline samples. In such samples, grain boundaries (GB) not only contribute to the resistivity but also play an essential role in modulating the magnetoresistance (MR) by adding an extrinsic term to the MR, in addition to the intrinsic one arising from the grains [14]. However, high-quality epitaxial thin-film samples allow us to minimize the effects of GB; thus we can establish the effects of Fe doping with high reliability. In particular, it is very interesting to study the MR properties and phase transitions for typical inhomogeneous manganite systems. It has been suggested that the Fe-doped manganites should be good candidate systems for consideration. Because it is known that most of the Fe ions are nearly antiferromagnetically coupled with the Mn ions, act as trapping centres and block the conduction path of e_g electrons. Moreover, as far as we are aware there have been no noise measurements made on B-site-doped samples of manganite CMR materials and Fe doping is a typical B-site doping process. In order to study in detail both the low-frequency noise mechanism and the effect of disorder in CMR materials, we prepared high-quality epitaxial thin films of $La_{0.7}Sr_{0.3}Mn_{1-x}Fe_xO_3$ (LSMFO) with $x = 0$, 0.08 and 0.12 on $SrTiO_3$ substrates by pulsed laser ablation and measured the low-frequency noise as a function of magnetic field.

2. Experiments

The LSMFO thin films with typical thickness of 2500 Å were deposited on (001)-oriented $SrTiO_3$ substrates using a 248 nm KrF excimer laser (Lambda Physik). The as-deposited films were post-annealed at 680 °C for 30 min under O_2 pressure of 400 mbar prior to a cooling down to room temperature at a rate of 15 °C min^{-1}. The crystal structures of these thin films were checked by x-ray diffraction and strong (00l) reflections were seen. We found that the structure does not change with the Fe doping and the lattice mismatch between the LSMFO thin film and $SrTiO_3$ substrate is small (<1%). The other detailed results on the microstructure can be found in [14].

The magnetotransport properties were measured using the standard four-probe technique. Electrical contacts were attached on gold pads by indium soldering. The contact resistance was much less than 1 Ω and the contact noise was shown to be negligible in this experiment. In the noise measurement, we used the battery as a current source and the biasing current was 0.1 mA (the corresponding current density was 25 A cm^{-2}). The spectra of the $1/f$ noise in the voltage fluctuation were measured between 1 and 100 Hz. The ac signal (because the dc signal was filtered by a large capacitor) between the voltage probes was fed through an EG&G 1900 low-noise transformer into an EG&G 5113 preamplifier and the output was measured by a HP 35670A spectrum analyser. The direction of the applied magnetic field was

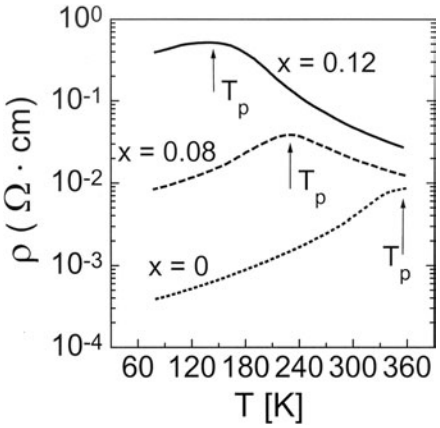

Figure 1. The temperature dependence of the resistivity, ρ, for three different samples, with $x = 0$ (dotted curve), 0.08 (dashed curve) and 0.12 (solid curve). The maximum values of the resistivities, which reveal T_p, are obtained at 360, 238 and 140 K for $x = 0$, 0.08 and 0.12, respectively, and marked by arrows.

perpendicular to the sample surface. The background noise (thermal noise and preamplifier noise) was measured when the sample current was zero and all noise signals shown in this paper were background-subtracted ones. The temperature stability during each measurement was better than 0.01 K and, in order to remove thermal fluctuations and drifts, all noise data were measured when the system was in thermal equilibrium.

3. Results and discussion

3.1. Noise power spectral density at $B = 0$ G

Figure 1 shows the temperature dependence of the resistivity for three differently doped samples ($x = 0$, 0.08 and 0.12). The maximum values of the resistivities, which reveal the metal–insulator (MI) transition temperatures T_p, are obtained at 360, 238 and 140 K for $x = 0$, 0.08 and 0.12, respectively. As x increases from 0 to 0.12, a rapid increase in ρ and a clear discrepancy between T_p and T_C are observed; i.e. ΔT $(=T_C - T_p) = 0$, 20 and 64 K for $x = 0$, 0.08 and 0.12, respectively [14]. It is suggested that the disorder introduced by the Fe doping combined with the replacement of some of the Mn–O–Mn bonds by Mn–O–Fe bonds causes an increase of resistivity. Unlike in the double-exchange (DE) scheme, these properties can be related to the disorders caused by Fe doping, which will lead to a fluctuation of the on-site energy experienced by the carriers. The on-site energy fluctuation plays a key role in determining the electronic state of the manganites for the disorder model [15–17].

Figure 2 shows the generic temperature dependence of the normalized noise PSD, $S_n(f)$ $(=S_v(f)/V^2)$, at $f = 9$ Hz and $B = 0$ G, for three different samples ($x = 0$ (■), $x = 0.08$ (□), $x = 0.12$ (●)). The temperature dependences of the normalized noise PSD are similar for all three differently doped samples except the noise peaks of the $x = 0.08$ and 0.12 samples near T_p. At $x = 0$, we could not find any peak in the noise PSD up to $T = 380$ K. We suggest that the temperature dependence of the noise PSD in these samples contains two terms, one due to the phase transition near T_p and the other a temperature-dependent term. The shape of the temperature-dependent one increases smoothly as temperature increases and this is also seen in other perovskite oxides [8–13]. We call this contribution $S_1(T)$. On top of

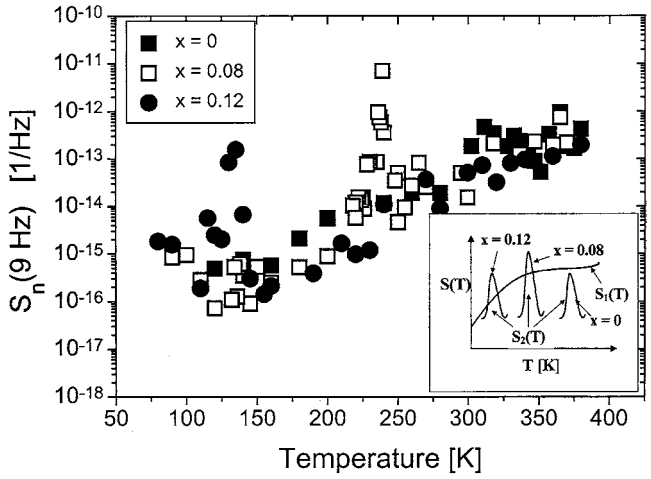

Figure 2. The temperature dependence of the normalized noise PSD, $S_n(f = 9\ \text{Hz})$, for three different samples, with $x = 0$ (■), $x = 0.08$ (□), $x = 0.12$ (●). Here all data were obtained while reducing the temperature and at $B = 0$ G. The inset shows sketches of the noise terms $S_1(T)$ and $S_2(T)$. Here $S_1(T)$ is the temperature-dependent one and $S_2(T)$ is the contribution arising from phase separation.

$S_1(T)$ there is a contribution connected to the transition at T_p, which is the contribution arising from the electronic phase separation and similar causes. We call this $S_2(T)$. Sketched graphs of $S_1(T)$ and $S_2(T)$ are shown in the inset of figure 2.

For $x = 0$, T_p is high and is located in a region where $S_1(T)$ is rather high and for reasons mentioned below $S_2(T)$ is rather low and may not even exist. So in this case we have $S_2 \ll S_1$ and the observed noise is essentially $S_1(T)$, i.e. one cannot observe a noise peak at T_p. For $x = 0.08$ and 0.12, T_P is low and in this temperature region $S_1(T)$ is also low and for reasons mentioned below $S_2(T)$ is high. So for these samples $S_2 > S_1$ and one will observe a noise peak at T_p.

The noise peak near T_P is due to coexisting phases, which have different average conductivities, and the region fluctuates between the two limits. The observed magnitude of the noise will thus depend critically on the ratio of the conductivities of the two coexisting phases. For $x = 0$, DE in Mn^{4+}–O–Mn^{3+} is dominant and the metallic conduction is achieved through the onset of a ferromagnetic ordering. Thus the $x = 0$ film is a bad metal, the transition is not at all sharp and the noise component S_2 is low and may not even exist. It is also submerged within S_1 as explained in the inset of figure 2. For $x = 0.08$ and 0.12, however, we found a clear discrepancy between T_C and T_p and the noise peak found near T_p for $x = 0.08$ and 0.12 is highly correlated with the MI phase transition [14]. Moreover, for $x \neq 0$, Fe^{3+} acts as an e_g-trapped Mn^{3+} and produces insulating Fe–O–Mn sites besides the Mn^{3+}–O–Mn^{4+} conducting channel. According to Ahn *et al* [18], the top of the Fe e_g band is almost located at the bottom of the Mn e_g band, with an overlap of <3%. Intrinsically, Fe^{3+} cannot act as the DE partner with Mn^{4+}. Fe doping depopulates the hopping electrons and weakens the DE interaction between Mn^{4+} and Mn^{3+} ions. Furthermore, the randomness of the Fe substitution will result in various near-neighbour configurations of Mn and Fe ions; i.e., as x increases the degree of disorder also increases and thus the conductivity changes are very prominent near T_p; therefore there is a large S_2 for $x \neq 0$.

One of the possible scenarios for the noise peak at T_p for $x = 0.08$ and 0.12 is a percolative nature of the charge transport in the ferromagnet [6, 9, 19, 20]. In percolation

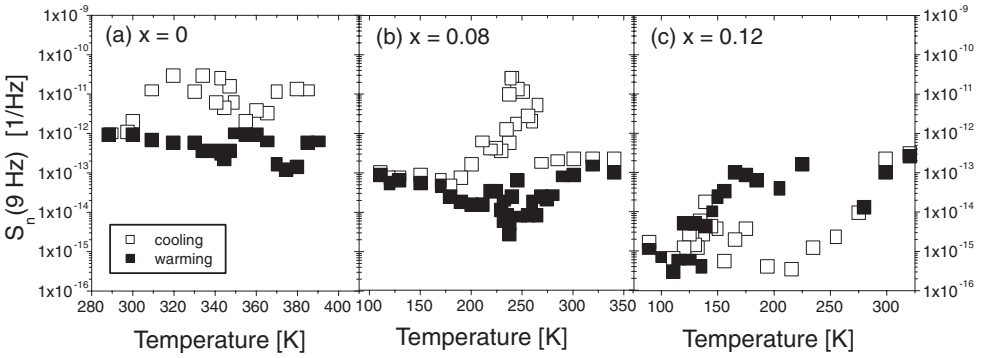

Figure 3. The temperature dependence of the normalized noise PSD, $S_n(f = 9\ \text{Hz})$ for three different samples, with $x = 0$ (a), $x = 0.08$ (b), $x = 0.12$ (c) at $B = 1500$ G. Here, open symbols indicate cooling and closed symbols indicate warming.

theory, the normalized noise PSD and resistivity increase as the fraction (p) of metallic particles approaches the percolation threshold (p_C) [21, 22]:

$$S_n \propto (p - p_c)^{-\kappa} \tag{1}$$

$$\rho \propto (p - p_c)^{-t}. \tag{2}$$

Here, κ and t are the critical exponents of the noise and the resistivity, respectively. However, from the fitting of our experimental results for S_n and ρ, we found that even though the critical exponent of S_n is in good agreement with the 3D random-void percolation model [21, 22], where $\kappa = 1.56 \pm 0.1$ for 3D, the exponent of ρ does not agree with the percolation model [23].

Another possible origin of the noise peak is related to the charge hopping between domains in inhomogeneous, disordered magnetic materials [4]. Moreover, it is known that the low-frequency fluctuations of domains give rise to an anomalous high level of low-frequency noise PSD. In our data, the abrupt increase of the noise PSD at T_p for $x = 0.08$ and 0.12 is by 2–3 orders of magnitude. It is known [13] that the lattice mismatch between the film and substrate causes local structural disorder in the doped manganites. Thus the strain in our film [14], even though it is small (<1%), is responsible for defect fluctuations which couple to the local magnetization and thus cause resistance fluctuations [12].

3.2. Noise PSD at $B \neq 0$ G

In a low magnetic field of $B < 1500$ G, there is no thermal hysteresis and the generic shape of the noise PSD is similar to that for $B = 0$ G as shown in figure 2. At $B = 1500$ G, however, a large thermal hysteresis appears in the noise PSD for all three samples, as shown in figure 3. For $x = 0$ and 0.08, the thermal hysteresis appears in the temperature region of T_p, but for $x = 0.12$, the thermal hysteresis appears only in the temperature region of $T > T_p$. Moreover, at $x = 0$ and 0.08 for $B = 1500$ G, the magnitude of $S_n(f)$ when cooling is one or two orders of magnitude larger than that when warming; however, at $x = 0.12$, the magnitude of the $S_n(f)$ when cooling is one or two orders of magnitude smaller than that when warming.

Raquet *et al* [8] suggested that the magnetic spin fluctuation is the main origin of the noise in the ferromagnetic phase of $La_{0.7}Sr_{0.3}MnO_3$ and this is also consistent with the drastic initial decrease of the electrical noise level when a magnetic field is applied. However, in our data, the noise PSD increases with applied magnetic field. This reveals that the magnetic spin fluctuation is not the origin of the noise in LSMFO thin films; the origin of noise may be related to the magnetic domain fluctuations considered below.

At $B = 1500$ G, the large thermal hysteresis, in all three differently doped samples, can be explained by magnetic domain fluctuations (thermal fluctuations of magnetic domains) [10]. Thermal hysteresis suggests that the noise originates from magnetic fluctuations, which could be magnetic domain fluctuations and fluctuations of the magnetic clusters. Moreover, in our data, the noise PSD depends both on the magnitude and on the history of the magnetic field, i.e., field cooling or zero-field cooling [23]. The magnetic domain structure has a magnetic field hysteresis and the large noise PSD arises from thermal fluctuations of magnetic domains [4, 24]. All of these things suggest that magnetic domain fluctuations may be the main noise source for our LSMFO thin films.

However, there are other strange results for the $x = 0.12$ sample for $B = 1500$ G, e.g.: (1) the magnitude of $S_n(f)$ for cooling is larger than that for warming for $x = 0$ and 0.08 (figures 3(a) and (b)), but at $x = 0.12$ the magnitude of $S_n(f)$ for warming is larger than that for cooling; and (2) at $x = 0$ and 0.08, the thermal hysteresis starts above T_p and ends far below T_p ($T_p - T \leqslant 60$ K), but at $x = 0.12$ the thermal hysteresis appears $T > T_p$ only; these cannot be explained by the thermal fluctuation of magnetic domains alone. Moreover, it is known that there are no domains above T_C; however, for $x = 0.12$ there appears large thermal hysteresis even above T_C (=204 K) as shown in figure 3(c). Thus, we must consider the structural disorder or oxygen deficiency with Fe doping in these $x = 0.12$ thin films. Recently we found a non-collinear magnetic structure (spin canting) for the highly doped LSMFO thin films with $x \geqslant 0.12$ [14]. When the doping concentration (x) is high, a given Fe ion will have both Fe and Mn ions as its nearest neighbours. Due to the competition between Fe–Fe, Fe–Mn and Mn–Mn magnetic interactions, the spins of Fe and Mn ions will deviate to misaligned directions and the result will be a canted structure. Thus the abnormal thermal hysteresis at $x = 0.12$ may be related to the spin canting.

3.3. Hooge constant, γ

In order to compare the magnitude of the $1/f$ noise PSD in CMR materials among different material systems, it is practical to determine the Hooge parameter [25], γ (a dimensionless quantity): $\gamma = fn\Omega\frac{S_V(f)}{V^2}$, where V is the dc voltage across the sample, n the charge-carrier density and Ω the sample volume. Typically γ is between 10^{-5} and 10^{-1} for metals and about 10^{-3} for semiconductors. In the calculation of $\gamma(T)$, we took the charge-carrier density, n, as 0.5×10^{22} cm^{-3} for $La_{2/3}Sr_{1/3}MnO_3$ thin film [8]. At room temperature, the γ-value for $x = 0$ thin film is 2.8×10^4, which is significantly larger than that for conventional metals [25–27]. For $x = 0.08$ and 0.12 thin films, we do not know the exact value of n; however, even though it decreases by two or three orders, the γ-value is still larger than that for conventional metals. Moreover, it is comparable to the γ-values for metallic multilayers exhibiting giant magnetoresistance (GMR) [28] and this also reveals that the enhanced noise in these CMR thin films is associated with the magnetic domain fluctuations. Recently, Reutler *et al* [13] found a very low value of γ for high-quality thin films with very little strain and disorder and a very high value of γ for strained thin films. They suggested [13] that the enhanced noise in CMR thin films is associated with the lattice mismatch between the thin film and substrate and this reveals that the intrinsic $1/f$ noise level in CMR materials may not be high.

4. Summary

The electronic and magnetic transport properties of high-quality epitaxial LSMFO thin films with $x = 0$, 0.08 and 0.12 are studied by measuring the resistivity and low-frequency noise as a function of low magnetic fields of up to 1500 G. In resistivity measurements, the temperature

of the maximum resistivity reveals the MI transition temperature (T_p), which is below the Curie temperature. At zero external magnetic field ($B = 0$ G), there is no noise peak near T_p for $x = 0$ thin films; however, near T_p, there is a noise peak for Fe doping ($x = 0.08$ and 0.12). The on-site energy fluctuation, which is related to the disorder caused by Fe doping, might be the origin of the noise peak for $x = 0.08$ and 0.12. The change in the noise PSD with doping rate is within one order of magnitude; however, the change in resistivity is by more than one order of magnitude. This reveals that the Fe doping does not contribute much to the dynamic fluctuations of charge carriers and only affects the static increase of the resistivity.

The large value of the Hooge parameter for our thin films is related to the strain, which arises from lattice mismatch between film and substrate. At $B = 1500$ G, there is a large thermal hysteresis in the noise PSD for all three samples and the contribution of magnetic fluctuations may be the origin of the thermal hysteresis in the noise PSD for $x < 0.12$. However, for $x = 0.12$ thin films, we must consider the structural disorder or oxygen deficiency with Fe doping.

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