Giant 1/f noise in perovskite manganites: Evidence of the percolation threshold

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We discovered an unprecedented magnitude of the $1/f$ noise near the Curie temperature (T_c) in low-T_c manganites. The scaling behavior of the $1/f$ noise and resistance provides strong evidence of the percolation nature of the ferromagnetic transition in the polycrystalline samples. The steplike changes of the resistance with temperature, observed for single crystals, suggest that the size of the ferromagnetic domains depends on the size of crystallites.

In the beginning of the colossal magnetoresistance (CMR) research on manganites, it was realized that the ferromagnetic (FM) transition occurs simultaneously with the metal-insulator transition.[1–3] The CMR was attributed to the magnetic-field-induced shift of this transition.[4] The nature of the transition changes drastically when the transition temperature T_c is varied with the chemical pressure.[4] While the transition in the high-T_c materials bears a resemblance with the second-order transition,[5] the low-T_c manganites demonstrate many features which are intrinsic to the first-order transitions, including a strong thermal hysteresis of the resistivity ρ and magnetization M. There is a growing theoretical and experimental evidence that the transport properties of the insulating state above T_c are dominated by small polarons or magnetic polarons, and that the bandlike carriers become important below T_c.[6–8] Recently, it was suggested that these two types of carriers coexist near T_c.[9] In addition, the static coexistence of the metallic FM phase and the insulating charged-ordered (CO) phase was found to play an important role in the CMR effects, especially in the low-T_c materials.[10]

In this paper, we report on measurements of the temperature dependence of the $1/f$ noise in polycrystalline and single crystal samples of low-T_c manganites. Our data strongly indicate that the so-called Curie temperature in the low-T_c materials is, in fact, a percolation transition temperature rather than the temperature of the long-range ferromagnetic phase transition. The scaling analysis of the $1/f$ noise is consistent with the percolation model of conducting domains randomly distributed in an insulating matrix.[11]

We have measured the $1/f$ noise in the poly- and single-crystal bulk samples of $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ with $x=0.35$.[12] The sample preparation is described elsewhere.[4] Typically, the polycrystalline samples were $4\times1\times1$ mm^3, single crystals—$3\times1\times0.5$ mm^3. The spectral density of the $1/f$ noise, S_V, and ρ have been measured in the four-point configuration at the temperature $T=4.2-300$ K for both cooling and warming. For the S_V measurements, we used the Stanford Research 830 lock-in amplifier in the mean average deviation mode with an equivalent noise bandwidth 1 Hz. The dc voltage applied to the sample, V, and S_V were recorded simultaneously as a function of T for the samples biased with a fixed dc current ($10^{-5}-10^{-3}$ A). It has been verified that the spectrum of the noise has a power-law form $1/f^\gamma$ in the frequency range $f=1-10^3$ Hz with γ close to unity for all temperatures. All the data discussed below were obtained in the linear regime, where the rms noise was linear in current.

The upper panel of Fig. 1 shows the dependences $\rho(T)$ for the poly- and single-crystal samples. In accordance with prior publications,[4,10] ρ increases with cooling below the CO transition ($T_{CO}\sim210$ K for $x>0.3$), reaches the maximum and decreases rapidly when the system undergoes the transition into the FM state. The FM transition temperature is strongly x dependent: T_c increases from 35 K for $x=0.4$ to 75 K for $x=0.35$. At lower temperatures, ρ is almost T independent, its value is anomalously large even for the single-crystal samples. The transition into the FM phase is accompanied by the increase of the magnetization, which saturates at $T<T_c$ (see the lower panel of Fig. 1). In contrast to ρ, the magnetization changes gradually at T_c for both poly- and single-crystal samples. This smooth dependence $M(T)$, which is observed even in low magnetic fields, is unusual for the FM transition in a homogeneous system. A strong temperature hysteresis of ρ and M was observed for all the samples discussed in this paper.

Qualitatively, the temperature dependence of the spectral density of the $1/f$ noise is as follows: S_V increases upon cooling below T_{CO}, reaches the maximum, and decreases steeply on the metallic side of the CO-FM transition. In the CO phase, far from the CO-FM transition, the increase of S_V with decreasing T is due mostly to the growth of ρ. Indeed, in the linear regime, S_V is proportional to V^2, or, for a fixed dc current I, to ρ^2:[13]
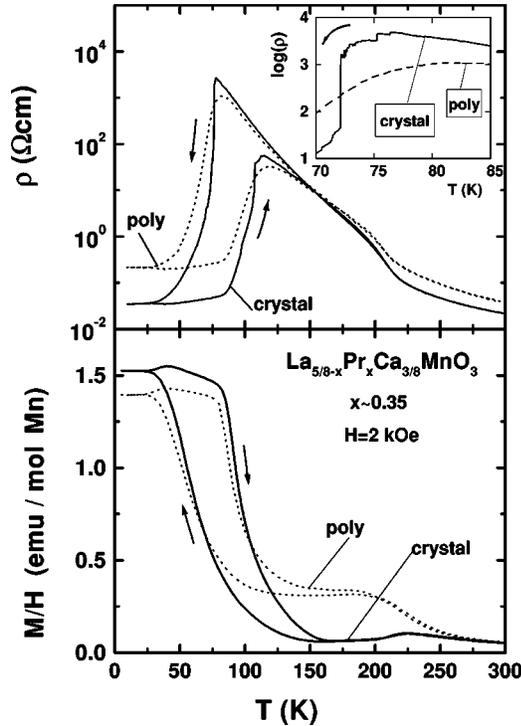
$$\frac{S_V}{V^2} = \frac{\alpha}{fnv_s},\qquad(1)$$

FIG. 1. The upper panel: the temperature dependences of the resistivity for $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ ($x=0.35$) (dashed line—the polycrystalline sample, solid line—the single crystal). The inset is the blowup of the $\rho(T)$ dependence for cooling near T_c. The lower panel: the temperature dependences of the magnetic susceptibility for the same samples (dashed line—polycrystalline, solid line—single crystal) measured at $H=2$ kOe. The arrows indicate the directions of the temperature change.
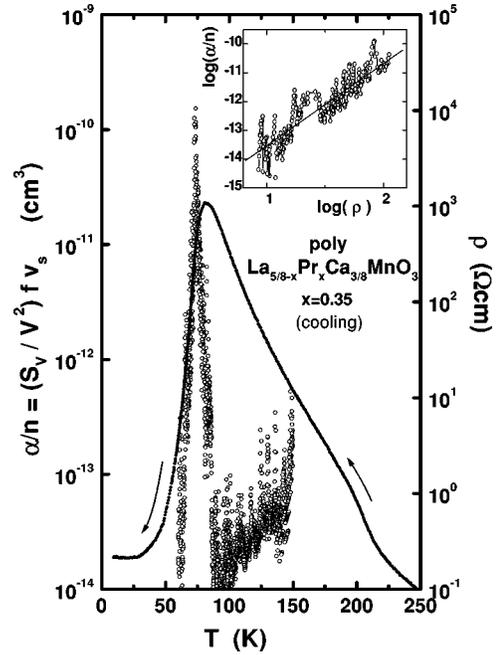

FIG. 2. The temperature dependence of the resistivity and the normalized spectral density of the $1/f$ noise, α/n, for the polycrystalline sample $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ ($x=0.35$) for cooling. The noise was measured at $f=10$ Hz; the background noise signal with no current through the sample has been subtracted. The inset shows the scaling dependence of the normalized magnitude of the $1/f$ noise versus ρ in the interval $T=61$ K–73 K (below $T_c=73$ K). The solid line corresponds to the power-law fit $\alpha/n \propto \rho^{2.9}$.

where α is the Hooge's parameter, f is the frequency at which the noise is measured, n is the concentration of the charge carriers or ''fluctuators,'' and v_s is the volume of the sample. Below we present the $1/f$ noise data in the normalized form $(S_V/V^2)fv_s=\alpha/n$. Figure 2 shows the temperature dependences of ρ and $S_V fv_s/V^2$ for the polycrystalline sample. The normalized magnitude of the $1/f$ noise is weakly T dependent in the CO phase: it varies by a factor of 2–3 over the range $T=90-150$ K, though ρ changes by 2–3 orders of magnitude over the same interval. Interestingly, the magnitude of the $1/f$ noise is anomalously large even far from the transition. (For comparison, the typical values of α/n are $10^{-21}-10^{-25}$ cm^3 for disordered metals and $10^{-18}-10^{-21}$ cm^3 for semiconductors.[13])

The magnitude of the $1/f$ noise increases dramatically in the vicinity of the CO-FM transition. The sharp peak of the $1/f$ noise enables to determine the transition temperature with a high accuracy; below we identify T_c with the temperature of the maximum of α/n. There is a correlation between the magnitude of the noise peak and the ratio $\rho(T_c)/\rho(300$ K$)$. For example, for the polycrystalline sample (Fig. 2), ρ increases by four orders of magnitude with cooling from room temperature down to 80 K; the normalized noise magnitude at the transition also increases by a factor of $\sim 10^4$. Since the high-temperature portion of the $\rho(T)$ dependences is approximately the same for all compounds with $x>0.35$,[10] the noise peak is more pronounced for materials with a higher value of $\rho(T_c)$, e.g., with a lower T_c.

It is worth mentioning that a much less pronounced increase of the $1/f$ noise at the FM transition has been reported for thin films of $La_{2/3-x}Y_xCa_{1/3}MnO_3$.[14,15] The temperature dependences of S_V/V^2 for the thin films differ qualitatively from the dependences we observe for bulk samples. The authors of Refs. 14 and 15 attributed the increase of the noise below T_c to the magnetic domains formation.

In principle, an abrupt increase of the normalized magnitude of the $1/f$ noise by $1-2$ orders of magnitude at magnetic transitions has been observed for many macroscopically homogeneous magnetic materials.[16–18] However, the T dependence and the magnitude of the $1/f$ noise in these materials differ significantly from our data for the CMR manganites.

Our $1/f$ noise measurements provide strong evidence of the percolation nature of the CO-FM transition in the polycrystalline bulk samples of the low-T_c manganites. These data indicate that the FM regions appear progressively with decreasing temperature in the CO phase, and the transition occurs when the concentration of the FM phase exceeds the percolation threshold. This is consistent with observation of the ferromagnetic regions in manganites at $T \gg T_c$, well beyond the conventional fluctuation regime (these regions were interpreted as magnetic polarons).[20] A diverging behavior of the $1/f$ noise is typical for the percolation metal-insulator transition.[11,19] At $T \gg T_c$, the transport properties of the sample are governed by a very large and, apparently, weakly fluctuating contribution of the CO phase. As a result, the $1/f$ noise is relatively low at $T_c < T \ll T_{CO}$. However, the magnitude of the $1/f$ noise diverges with approaching the perco-

lation threshold. The formation of the infinite percolation FM cluster at T_c is also consistent with the observation of the maximum of $d\rho/dT$ exactly at the same temperature. Notice that T_c is lower than the temperature of the maximum of ρ: this is expected for a percolating mixture of two phases where the ''insulating'' phase has a finite ρ that increases rapidly with cooling. Previously, spatially inhomogeneous FM state and percolation nature of the conductivity in thin films of manganites have been discussed in Ref. 21.

A clearly diverging behavior of the $1/f$ noise allows to determine T_c with a high accuracy and to perform the scaling analysis of S_V and ρ on the ''metallic'' side of the CO-FM transition. In the vicinity of a percolation metal-insulator transition, the scaling behavior of ρ and S_V/V^2 is expected:[11,13,19]

$$S_V/V^2 \propto (p-p_c)^{-k}, \qquad (2)$$

$$\rho \propto (p-p_c)^{-t}. \qquad (3)$$

Here p is the concentration of the metallic phase, p_c is the critical concentration, and k and t are the critical exponents of the noise and the resistivity. It is convenient to represent S_V/V^2 as a function of ρ (in this case, no assumption on the value of p_c is necessary):

$$S_V/V^2 \propto \rho^{k/t}. \qquad (4)$$

The normalized magnitude of the $1/f$ noise versus ρ for the polycrystalline sample is shown in the double-log scale in the inset of Fig. 2. Within the experimental accuracy, this dependence can be fitted by the power law (4) with $k/t = 2.9 \pm 0.5$. These values of k/t are close to the result $k/t = 2.4$ obtained theoretically for the continuum percolation model of conducting regions, randomly placed in an insulating matrix (the so-called inverted random-void model).[11] Previously, a similar value of $k/t=3$ has been observed experimentally for the mixed powders of conducting and insulating materials.[22] Notice that the ratio of critical exponents k/t in the continuum percolation exceeds significantly $k/t = 0.5-0.8$ for the discrete random models, though the values of the critical exponent t are almost the same for these models ($t=1.9\pm0.03$).

In order to disentangle k and t, one has to measure either the concentration of the FM phase $p(T)$ as a function of T, or some quantity that is proportional to p, e.g., the magnetization. In the vicinity of the transition, M is approximately a linear function of (T_c-T) (Fig. 1). Hence, instead of $(p-p_c)$, we can use (T_c-T) as a variable in the scaling dependences (2) and (3). The dependence $\rho(T)$ was found to be close to the power law $\rho \propto (T_c-T)^{-t}$ with $t=2\pm0.3$ on the metallic side of the transition. Previously, similar values $t=2.3\pm0.4$ and $k=5\pm1$ have been measured for the conducting particles in an insulating matrix.[23,24] The experimental values of the critical exponents $t=2\pm0.3$ and $k=5.9 \pm1.5$ for our samples are consistent with the predictions of the inverted random-void model of the continuum percolation.

For high-quality single crystals of $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ ($x\sim0.35$),[12] we have also observed a dramatic increase of the $1/f$ noise at the transition: α/n reaches 10^{-7} cm^3 for cooling and 10^{-10} cm^3 for warming (Fig. 3). An evidence of
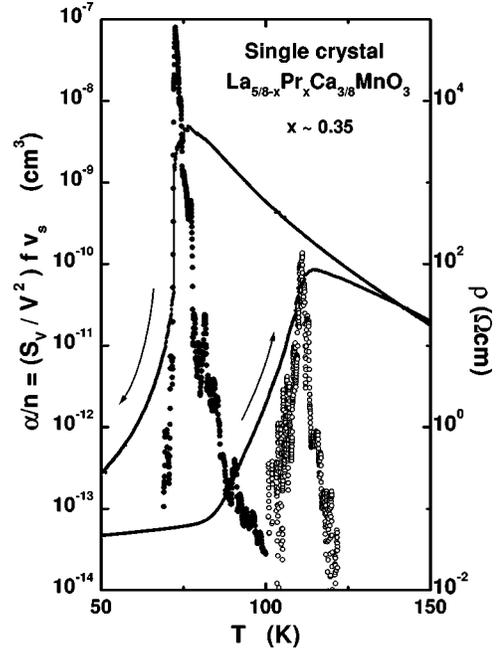

FIG. 3. The temperature dependence of the resistivity and the normalized spectral density of the $1/f$ noise for cooling (●) and warming (○) for the single crystal $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ ($x \sim 0.35$).

the phase inhomogeneity of the single crystal is provided by the smooth dependence of $M(T)$ (Fig. 1), which is similar to that for the polycrystalline samples. However, there are several important distinctions between the temperature dependences of ρ and S_V/V^2 for poly- and single crystals. Although the magnitude of ρ is similar for both types of samples (see Fig. 1), ρ for the single crystal exhibits reproducible steps as a function of T in the vicinity of T_c (see the inset of Fig. 1). The sharp drop of ρ by more than an order of magnitude, observed for this single crystal at $T\sim72$ K, can be interpreted as formation of a chain of a few connected FM domains between the voltage leads. The steplike behavior of ρ at the transition indicates that the size of the FM regions in the single crystal is significantly bigger than that in the polycrystalline samples. When the voltage leads become ''shortened'' by a chain of the metallic FM domains, an abrupt drop of the $1/f$ noise magnitude occurs. The percolation approach is not applicable in this case, since we probe the inhomogeneous system at the scale smaller than the percolation correlation length.

To summarize, we observed the dramatic peak of the $1/f$ noise in the CMR manganite $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ ($x =0.35$ and 0.375) at the transition between the charge-ordered and ferromagnetic states. The scaling analysis of the magnitude of the noise and the resistivity in the polycrystalline samples is consistent with the continuum percolation model of conducting FM domains randomly placed in the insulating CO matrix. Being combined with the data on the temperature dependence of ρ and M, these measurements provide strong evidence of the percolating nature of the CO-FM transition in the polycrystalline samples of the low-T_c manganites. To the best of our knowledge, the value of $\alpha/n=S_Vfv_s/V^2 \sim 10^{-10}-10^{-7}$ cm^3 observed in the low-T_c

manganites at the transition is the largest normalized magnitude of the 1/f noise for the condensed-matter systems. A well-pronounced steplike temperature dependence of the resistivity, observed for high-quality single crystals, suggests that the scale of the phase separation is much greater than that in the polycrystalline samples. This might indicate that the surface energy and/or the strain effects associated with the grain boundaries influence the size of the ferromagnetic domains in the low-T_c manganites.

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