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

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(May 3, 2018)

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 step-like changes of the resistance with temperature, observed for single crystals, suggest that the size of the ferromagnetic domains depends on the size of crystallites.

PACS numbers: 75.30.Vn, 73.50.Td, 64.60.Ak, 71.30.+h

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 \cite{1}. The CMR was attributed to the magnetic-field-induced shift of this transition \cite{2}. The nature of the transition changes drastically when the transition temperature $T_c$ is varied with the chemical pressure \cite{3}. While the transition in the high-$T_c$ materials bears a resemblance with the second-order transition \cite{4}, the low-$T_c$ manganites demonstrate many features which are intrinsic to the first-order transitions, including a strong thermal hysteresis of the resistivity $\rho$ 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 band-like carriers become important below $T_c$ \textsuperscript{5, 6}. Recently, it was suggested that these two types of carriers coexist near $T_c$ \textsuperscript{7}. 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 \textsuperscript{10}.

In this Letter, 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 \textsuperscript{11}.

We have measured the 1/f noise in the poly- and single crystal bulk samples of $La_{0.5}Sr_{0.5}Ca_{0.3}MnO_3$ with $x = 0.35$ \textsuperscript{12}. The sample preparation is described elsewhere \cite{4}. Typically, the polycrystalline samples were $4 \times 1 \times 1 \ mm^3$, single crystals - $3 \times 1 \times 0.5 \ mm^3$. The spectral density of the 1/f noise, $S_V$, and $\rho$ have been measured in the four-point configuration at the temperature $T = 4.2 - 300K$ 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 $1Hz$. 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 $f = 1 - 10^3Hz$ with $\gamma$ 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 accord with prior publications \cite{11, 12}, $\rho$ increases with cooling below the CO transition ($T_{CO} \sim 210K$ 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 $35K$ for $x = 0.4$ to $75K$ for $x = 0.35$. At lower temperatures, $\rho$ 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 $\rho$, 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 $\rho$ 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 $\rho$. Indeed, in the linear regime, $S_V$ is proportional to $V^2$, or, for a fixed dc current $I$, to $\rho^2$ \textsuperscript{13}:

$$ S_V \propto \frac{\alpha}{f \cdot n \cdot v_s}, \quad \alpha = \frac{\rho}{f \cdot n \cdot v_s}, $$

where $\alpha$ 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 \((S_V/V^2) \cdot f \cdot v_s = \alpha/n\). Figure 2 shows the temperature dependences of \(\rho\) and \(S_V \cdot f \cdot v_s/V^2\) 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 \(\rho\) 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 \(\alpha/n\) are \(10^{-21} - 10^{-25} \text{ cm}^3\) for disordered metals and \(10^{-18} - 10^{-21} \text{ cm}^3\) 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 \(\alpha/n\). There is a correlation between the magnitude of the noise peak and the ratio \(\rho(T_c)/\rho(300K)\). For example, for the polycrystalline sample (Fig. 2), \(\rho\) increases by 4 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 higher value of \(\rho(T_c)\), e.g. with 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 \(La_{2/3}Y_{x}Ca_{1/3}MnO_3\) [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 [14,15] attributed the increase of the noise below \(T_c\) to the magnetic domains formation. In principle, an abrupt increase of the normal-

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

FIG. 2. The temperature dependence of the resistivity and the normalized spectral density of the 1/f noise, \(\alpha/n\), for the polycrystalline sample \(La_{5/8-x}Pr_xCa_{3/8}MnO_3\) (\(x = 0.35\)) for cooling. The noise was measured at \(f = 10Hz\); 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 \(\rho\) in the interval \(T = 61K - 73K\) (below \(T_c = 73K\)). The solid line corresponds to the power law fit \(\alpha/n \propto \rho^{2.9}\).
The normalized magnitude of the 1/f noise by $1 \div 2$ orders of magnitude at magnetic transitions has been observed for many macroscopically homogeneous magnetic materials \[^{[10][15]}\]. 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 $T >> T_c$, 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 \[^{[11][15]}\]. At $T >> T_c$, 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 $T_c < T << T_{CO}$. However, the magnitude of the 1/f noise diverges with approaching the percolation threshold. The formation of the infinite percolation FM cluster at $T_c$ is also consistent with the observation of the maximum of $dp/dT$ exactly at the same temperature. Notice that $T_c$ is lower than the temperature of the maximum of $\rho$: this is expected for a percolating mixture of two phases where the “insulating” phase has a finite $\rho$ 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 $\rho$ on the "metallic" side of the CO-FM transition. In the vicinity of a percolation metal-insulator transition, the scaling behavior of $\rho$ and $S_V/V^2$ is expected \[^{[11][14][15]}\]:

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

$$\rho \propto (p - p_c)^{-t}.$$  \hspace{1cm} (2)

Here $p$ is the concentration of the metallic phase, $p_c$ is the critical concentration, $k$ 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 $\rho$ (in this case, no assumption on the value of $p_c$ is necessary):

$$S_V/V^2 \propto \rho^{-k/t}.$$  \hspace{1cm} (3)

The normalized magnitude of the 1/f noise versus $\rho$ 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) \[^{[11]}\].

Previous, 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 ($t = 1.9 \pm 0.03$).

![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 = 0.35).](image)

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 $t = 2 \pm 0.3$ on the metallic side of the transition. Previously, similar values $t = 2.3 \pm 0.4$ and $k = 5 \pm 1$ has been measured for the conducting particles in an insulating matrix \[^{[23][24]}\]. The experimental values
of the critical exponents $t = 2 \pm 0.3$ and $k = 5.9 \pm 1.5$ for our samples are consistent with the predictions of the inverted random-void model of the continuum percolation.

For high-quality single crystals of $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ ($x \sim 0.35$) [2], we have also observed a dramatic increase of the 1/f noise at the transition: $\alpha/n$ reaches $10^{-7}$ cm$^3$ for cooling and $10^{-10}$ cm$^3$ for warming (Fig. 3). An evidence of 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 $\rho$ and $S_V/V^2$ for poly- and single crystals. Although the magnitude of $\rho$ is similar for both types of samples (see Fig. 1), $\rho$ 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 $\rho$ by more than an order of magnitude, observed for this single crystal at $T \sim 72K$, can be interpreted as formation of a chain of a few connected FM domains between the voltage leads. The step-like behavior of $\rho$ 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 $\rho$ 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 $\alpha/n = S_V \cdot f \cdot v_s/V^2 \sim 10^{-10} \div 10^{-7} cm^3$ observed in the low-$T_c$ manganites at the transition, is the largest normalized magnitude of the 1/f noise for the condensate matter systems. A well-pronounced step-like 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.

We thank Sh. Kogan for helpful discussions. This work was supported in part by the NSF grant No. DMR-9802513.

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