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Observation of strong reduction of $1/f$ noise in $Pr_{0.67}Sr_{0.33}MnO_3$ films

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Abstract

The resistance and $1/f$ noise in two representative $Pr_{0.67}Sr_{0.33}MnO_3$ films were recorded continuously from liquid nitrogen temperature to room temperature. Film A has a sharp resistance peak near 160 K and a strong reduction of $1/f$ noise was observed at slightly below the peak temperature. Film B has a moderate resistance peak near 233 K and instead of noise reduction a noise peak was observed at a higher temperature. Bipolaron theory is used to discuss the order of the phase transition. Differences in the order of phase transition, a broadened transition width and crossing of energy levels are discussed as possible explanations for observed noise reduction and the difference between two samples.

1. Introduction

In recent years, colossal magnetoresistance (CMR) has been one of the most intriguing topics in condensed matter physics. The CMR manganites are characterized by a peak in their temperature dependence of resistivity. The peak temperature T_p is close to the Curie temperature T_C [1], which is the transition point from a paramagnetic semiconductor at high temperatures to a ferromagnetic metal at low temperatures. Near T_p a huge negative magnetoresistance is observed. Much attention has been paid to the nature of the transition and the process occurring near the resistance peak, because the fundamental physics of the transitions remains unclear [2]. From an experimental point of view, it is important to find out whether this transition is homogeneous or inhomogeneous, and whether it is first-order or second-order.

$1/f$ noise behaviour is important for understanding the microscopic mechanism of low-frequency dynamic processes in a material, and it is also an important tool for studying the nature of the transition. Experiments have shown that CMR materials have large $1/f$ noise [3–6]. For the high-temperature paramagnetic state $1/f$ noise is usually thought to come from the localization of charge. Recently, it has been suggested that $1/f$ noise may result from the hopping of carriers between different energy levels [7]. For the low-temperature ferromagnetic

phase, Raquet *et al* [8] and Merithew *et al* [9] recently reported an observation of giant random telegraph noise in $La_{2/3}Ca_{1/3}MnO_3$, indicating a dynamic phase separation in that system. Podzorov *et al* [10] observed sharp increases in noise power near peak temperature in some high-resistivity low-T_p bulk manganites. Their data strongly suggest a percolation-type transition.

$1/f$ noise is generally described by Hooge's empirical relation and the derived Hooge parameter is used to compare the noise levels in different materials. Hooge's equation is:

$$S_V/V_{dc}^2 = \gamma/n_c\Omega f^\alpha \tag{1}$$

where S_V is the excess noise spectral density, V_{dc} the applied dc voltage across the sample, γ the Hooge parameter, n_c the density of carriers and Ω the sample volume.

A spectrum analyser is usually used to measure noise power spectral density. For $1/f$ noise the lower-frequency part is more pronounced than the background and is preferred for measurements. To acquire the low-frequency points sufficient time is required to accumulate enough data for the analysis and average processes. When there is a fast change of the noise amplitude, this method may only give an averaged result. Similar in principle, a lock-in amplifier can also be used to measure noise at a particular frequency. The spectral noise density is recorded continuously for noise in the pass band (equivalent noise band width, ENBW) by using the mean average deviation mode. This is described as [11]:

$$S_V(t|\bar{f}, \Delta f) = \langle[\delta V(t|\bar{f}, \Delta f)]^2\rangle, \tag{2}$$

where $\bar{f}$ is the centre frequency, Δf is the bandwidth of the lock-in filter, t is time and $\langle\rangle$ means the average over an ensemble of identical noise sources. With the ergodic assumption, the average can be done over some time interval selected by the moving average algorithm of the lock-in amplifier. The voltage noise measured by the mean average deviation mode can be described as:

$$V_n(t|\bar{f}, \Delta f) = (\overline{[\delta V(t|\bar{f}, \Delta f)]^2})^{1/2}, \tag{3}$$

where the bar above means the average over the time interval ranging from 10 to 80 time constants. For simplicity the symbols S_V, δV, V_n will be used in the following text. While the lock-in amplifier is doing the dynamic calculation, the averaged voltage ($\bar{V}$) and the deviation ($\delta V = V - \bar{V}$) are calculated from data accumulated in the averaging time. When there is a fast change of the noise amplitude V, $\bar{V}$ will not change too much, hence the deviation (δV) will show the change instantly but the variance (V_n) will not. So δV measurement may provide a clearer view for some fast processes.

In this paper, we report measurements of the temperature dependence of both V_n and δV in two representative $Pr_{0.67}Sr_{0.33}MnO_3$ films.

2. Experimental details

Films of $Pr_{0.67}Sr_{0.33}MnO_3$ 200 nm thick were grown by pulsed laser deposition at 700 °C using a stoichiometric target on rectangular single-crystal $LaAlO_3(100)$ substrates of size 0.25×1 cm^2. All data discussed henceforth will be for two samples prepared under different deposition conditions. The first one, sample A, was kept at the same temperature for half an hour after deposition at a pressure of 2×10^{-5} Torr and was then cooled down, which resulted in a lower peak temperature with rapid resistance drop because of oxygen deficiency and the extra annealing. The second one, sample B, was cooled down to room temperature without any further treatment and had a broad resistance peak at higher temperature. Similar behaviour has been found for other samples in the respective batches. X-ray diffraction patterns indicate

that the films show epitaxial features and that the c-axis is perpendicular to the film surface. The details of sample preparation are described elsewhere [12].

The resistance and noise measurements were performed with a standard four-probe dc configuration. Four stripes of silver parallel to the shorter side of the rectangular substrate were made by evaporation and patterned by photolithography. Electrical contacts were made with indium solder. Batteries and ballast resistances were connected to make a near constant current source. The measuring current is about 10–100 μA for all the measurements with different set-ups. The voltage fluctuations were dc-filtered by a large capacitance, amplified by a PAR113 preamplifier, and then received by an HP35665A spectrum analyser as well as a Stanford Research 850 lock-in amplifier. The spectrum analyser generated S_V in the frequency range 1–100 Hz. Calibration by measuring the excess noise of a several hundred ohm carbon resistance was usually conducted before measuring the samples.

The lock-in amplifier recorded V_n and δV at the central frequency $\bar{f} = 10$ Hz with bandwidth $\Delta f = 250$ mHz. The time constant τ is set to 1 s and the recording rate is about 0.67 s^{-1}. These parameters were chosen bearing in mind the magnitude of the noise signals and the required speed of measurements. In principle, the lower the centre frequency, the higher the magnitude of the noise signal and the easier the measurement, but data average time is also longer and this affects the speed of measurement. Since the sample resistance changes drastically near T_p, the temperature sweeping rate was chosen to be 2 K min^{-1} by adjusting the thermal exchange between the sample and the liquid nitrogen bath. Use of the electronic temperature controller was avoided to minimize noise interference. Although the dc voltage drop across the sample $V_{dc}(t)$ changes a little in the averaging time when the temperature is sweeping, the ac component $\bar{V}_{ac}(t, 10\text{ Hz}, 250\text{ mHz})$ will most probably still be the same and close to 0. The magnitude of δV at 10 Hz will change proportional to $V_{dc}(t)$ as described in Hooge's equation. Within the sampling time the change of $V_{dc}(t)$ and hence the change of δV at 10 Hz is about one thousandth and is negligible compared with fluctuations of δV itself. So as stated in the introduction δV shows real-time fluctuations and V_n shows an averaged result.

3. Results

The temperature dependence of the resistance of samples A and B is shown in figure 2. The sharp resistance peak of sample A occurs at $T_p \sim 160$ K. At higher temperature the resistance decreases exponentially with increasing temperature. The part from 195 to 270 K was fitted by using equation $R(T) \propto T \exp(-E_\rho/kT)$, according to small polaron theory [13, 14]. The calculated activation energy E_ρ is 543 K. Below T_p the resistance decreases dramatically with decreasing temperature and then varies slowly in the metallic regime. A noticeable thermal hysteresis of resistance was observed from 140 to 190 K.

For sample B, a moderate resistance peak occurs at $T_p \sim 233$ K. Compared with sample A, the resistance of sample B is lower and the peak is broad and indistinct. No thermal hysteresis of resistance was observed. Below T_p the resistance decreases more slowly with decreasing temperature. Above T_p, the activation energy is calculated to be 480 K by the same fit procedure used for sample A.

The typical frequency dependence of power spectral density S_V and the dependence of S_V on V^2 for sample A are shown in figure 1. It has been verified that both samples produce clean $f^{-\alpha}$ spectra in the frequency range 1–100 Hz with the exponent α ranging from 0.8 to 1.2 for nearly all the temperature range measured. The noise power spectral density $S_V(f)$ always scales with V^2, indicating that the measured noise power spectra result solely from fluctuation in the conductance of the samples, and self-heating due to the current flow is negligible. The contact resistance of voltage contacts is usually less than 1 Ω. Compared

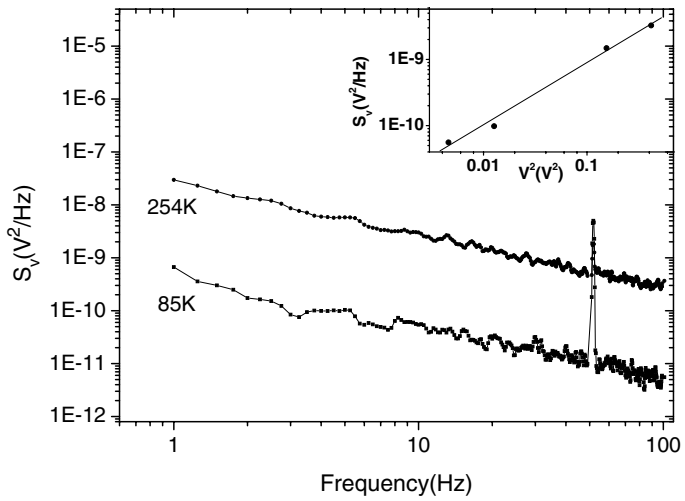

Figure 1. Frequency dependence of power spectral density S_V for sample A at 254 and 85 K, both showing $1/f$ dependence. Inset: linear V^2 dependence of S_V at room temperature and 10 Hz.

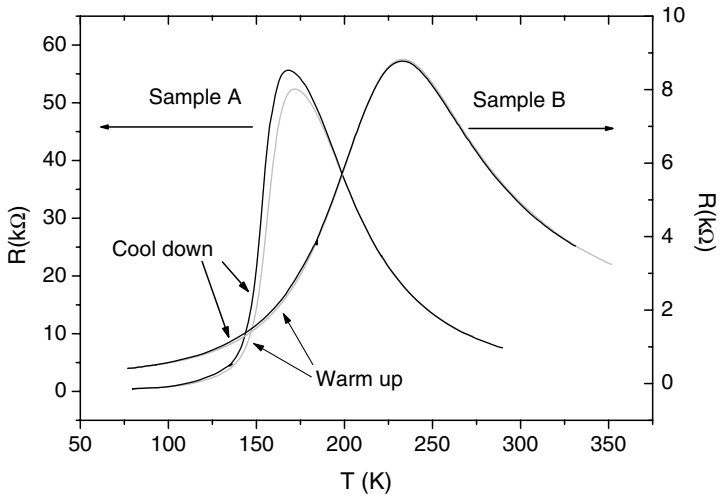

Figure 2. Temperature dependence of the resistance of samples A and B. For sample A a sharp resistance peak presents near 160 K and clear hysteresis can be found in the peak regime from 140 to 190 K. For sample B a moderate peak rises near 233 K and almost no hysteresis can be found.

with the huge sample resistance and huge γ of CMR samples its contribution to the noise is also negligible. The fluctuation of thermal noise measured by the lock-in amplifier is $\sqrt{4k_BTR\Delta f} \approx 0.13\sqrt{R}\sqrt{T/300}\sqrt{ENBW}nV$ and is about 0.1 μV for the peak resistance and 1 Hz ENBW. This is also negligible for most cases except in the regime where excess noise is strongly reduced.

The Hooge parameter γ can be estimated from the room-temperature power spectral density of sample A,. The density of carriers is $n_c = 5 \times 10^{21}$ cm^{-3} when we assume $1/3$ hole per formula unit. We obtain $\gamma \sim 10^9$, which is three orders higher than that of previous measurements [4].

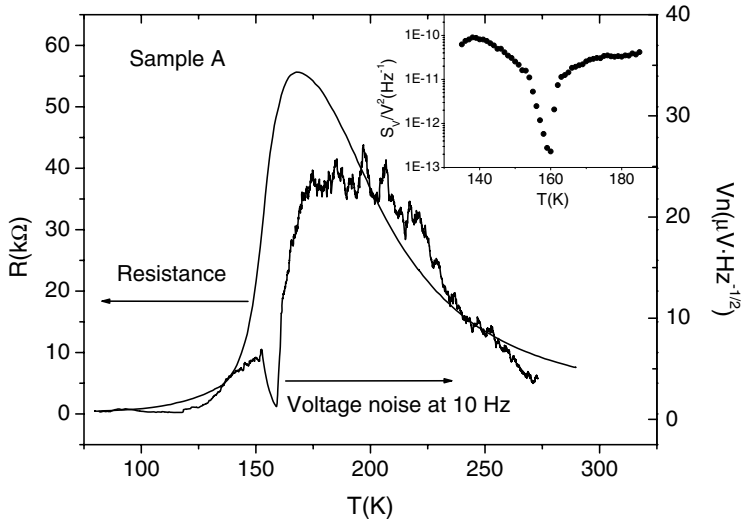

Figure 3. Temperature dependence of the resistance and $1/f$ noise of sample A. The latter was measured by a lock-in amplifier with slowly increasing temperature at 10 Hz. Inset: normalized power spectral density near the strong reduction regime. The noise reduction is clear near the resistance peak temperature in both figures.

Figure 3 shows the temperature dependence of V_n of sample A measured by the lock-in amplifier as well as the temperature dependence of the resistance. In the metallic regime the voltage noise is small. To perform continuous measurement the input range of SR850 is set to be large enough to tolerate the huge noise near the resistance peak. But with a large input range the background noise of SR850 increases and the small noise can be blurred by the background. Here we focus on the relatively high resistance part starting from 130 K. V_n begins to drop significantly at temperatures about 5 K below T_p. The drop is much more prominent in normalized power spectra, as shown in the inset of figure 3. At temperatures very close to T_p, V_n starts to increase dramatically. At higher temperatures it decreases slowly as resistance decreases with increasing temperature. The reduction of V_n is repeatable when cooling the sample through T_p as long as the temperature sweeping rate is slow. And when measured by the spectrum analyser, a clear strong reduction of S_V was also observed in the same regime with a smaller exponent α. This deviation may be explained by the average of smaller $1/f$ noise and flat noise background (i.e. thermal noise).

The reduction of V_n is clearer when the real-time voltage deviation δV is recorded. Figure 4 shows that δV drops instantly and is very small in the 5 K temperature range below T_p. To show the magnitude clearly, the absolute value of fluctuations is presented. In the strong reduction regime the magnitude of δV is about two orders smaller than it is outside this regime. As shown in the inset of figure 4, the tiny δV is less than 0.5 μV and is comparable to thermal noise. This provides evidence why in this regime the noise power spectrum deviates from $1/f$ type.

Measurements were made with lower gain by the PAR113 preamplifier to make sure there is no overloading of instruments by a large noise signal, which might block the measuring system and result in artificial observations; the reduction of noise is found to be the same. This strong noise reduction is also found in measurements of other samples from the same batch as sample A.

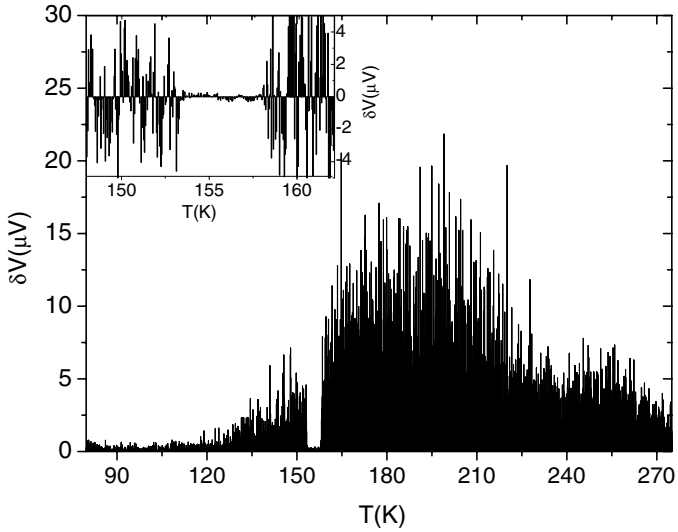

Figure 4. The absolute value of the real-time voltage deviation δ_V with slowly increasing temperature of sample A. A strong reduction of δ_V in the temperature range about 5 K below T_p is clearly displayed. Inset: the real-time δ_V from 145 to 165 K. The magnitude inside the noise reduction regime is less than 0.5 μV and close to the magnitude of thermal fluctuation.

Figure 5 shows the temperature dependence of V_n for sample B as well as the temperature dependence of the resistance. No strong noise reduction was observed near T_p. The fluctuation of V_n itself in the metallic regime is bigger than for sample A. Several spikes are clearly seen in the broadened resistance peak regime. At higher temperature, a sharp noise peak is observed. Although the noise peak can be found in repeated measurements, its position and magnitude are not very repeatable. At room temperature, from S_V measured by the spectrum analyser, the Hooge parameter γ is found to be 10^7, which is two orders smaller than that of sample A but close to previous measurements [4].

4. Discussion

$1/f$ noise of a $Pr_{0.67}Sr_{0.33}MnO_3$ film with a similar T_p ($\approx$160 K) to our sample A has been measured before by Rajeswari *et al* [4]. In their case, a shallow dip in power spectral density near T_p was observed and the room-temperature Hooge parameter is $\gamma \sim 10^6$. Compared with their sample, the slope of $\rho(T)$ near T_p on the side of the metallic regime is much steeper for our sample A. If we defined the transition width as the minimum temperature difference within which 80% of the total change of resistance occurs from the metallic regime to the peak, the transition width is about 20 K in our sample A and 62 K in their case. The broad transition there might be related to their lower deposition temperature (625 °C) and no further annealing after deposition. Therefore, a possible explanation for the different noise behaviours is that our sample A has a smaller transition width and as a result the noise dip cannot be smeared.

As for the origin of the strong noise reduction we do not have a clear picture at present. Raquet *et al* [7] proposed that the activation energy for fluctuations may be associated with electronic excitations in the band structure when applying the Dutta–Horn model to explain the temperature dependence of $1/f$ noise in $La_{2/3}Sr_{1/3}MnO_3$. Especially prevalent are the excitations from the Fermi level to the bottom of the spin-down t_{2g} band and from the top of the filled 2p band to the Fermi level.

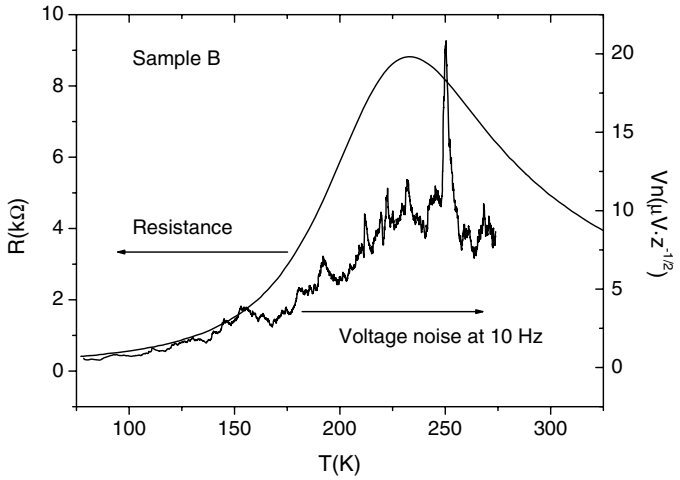

Figure 5. Temperature dependence of the resistance and $1/f$ noise of sample B. The latter was measured by a lock-in amplifier with slowly increasing temperature at 10 Hz. No clear noise reduction was observed near the resistance peak at 233 K. Spikes and one noise peak can be seen in the broad peak regime.

For $Pr_{0.7}Sr_{0.3}MnO_3$, Toulemonde *et al* [15] found that upon cooling below the metal–insulator transition temperature the dynamic Jahn–Teller effect reduced and then the splitting of the energy levels of the manganese ion became smaller. This made the energy of the $t_{2g}^{\downarrow}$ orbital change from higher than $e_g^{\uparrow 1}$ to lower than $e_g^{\uparrow 1}$ as shown in figure 8 of [15]; electrons will also change orbital to occupy the lower energy orbital. If we assume that $1/f$ noise does originate from the electronic excitations in the band structure, then when the two energy levels cross, the activation energy distribution of $1/f$ noise might be reduced and the $1/f$ noise will be correspondingly strongly reduced.

The noise sources can also change when temperature changes. For $La_{0.67}Ca_{0.33}MnO_3$ at temperatures below T_P Raquet *et al* [8] and Merithew *et al* [9] observed giant random telegraph noise originating from a few switching clusters located in critical bonds of the narrowest current paths. They mentioned there is no evidence for the random telegraph noise [8] above 180 K while the Curie temperature T_C is ≈210 K. So it is possible when the temperature is close to the peak temperature ($T \lesssim 210$ K) that certain noise sources disappear above 180 K. In our case, we can then interpret the noise reduction in our sample A as a delay caused by a switch between two regimes with different noise sources.

Determining the order of the phase transition of CMR is also an intriguing problem. The low-T_p manganites demonstrate many features that are intrinsic to first-order transitions [16]. Although the phase transition might be broadened by a distribution in T_C, the ferromagnetic transition is frequently considered to be second order. According to a recent theory of Alexandrov and Bratkovsky [17, 18], there is a criterion for determining the order of the transition. For second-order transitions, the dimensionless binding energy $\delta \equiv \Delta / J_{pd} S$ is less than a critical value δ_c, which depends on the doping concentration only. Here Δ is the polaron binding energy and $J_{pd}S$ the strength of an exchange interaction of polaronic carriers with localized spins. For first-order transitions, $\delta > \delta_c$, the behaviour in conduction and magnetization will be discontinuous. The critical value δ_c is decided by equation (9) in [17]:

$$n_c^{1/2} \ln \frac{2(x - n_c)}{n_c^2} = 2^{3/2}\delta, \tag{4}$$

where n_c is the polaron density at T_C, and $t_c = 2k_B T_C / J_{pd} S$ is the dimensionless temperature at T_C. In zero field $t_c = (n_c/2)^{1/2}$.

If we know $J_{pd}S$ and Δ we can get δ and compare it with $\delta_c \sim 0.42$ for $x = 0.33$. However we do not know the exact value of $J_{pd}S$, although a possible range of 1000–3000 K for similar CMR samples can be found in [14, 19]. Another way to apply this equation is by assuming it is solvable and letting $J_{pd}S$ be the unknown, then with the values of $T_C(T_p)$ and Δ we can solve the equation and compare the solution with possible values of $J_{pd}S$ and see whether it is reasonable.

The activation energy E_ρ from the resistance fit process consists of two parts. One part is the bipolaron binding energy and corresponds to the carrier density. The other part is related to carrier mobility and is derived from small polaron hopping [14]. The upper limit for Δ is $2 \times E_\rho$ when the energy of formation of small polarons is negligible. When bipolaron theory is applicable, most of the carriers (polarons) are formed even in the relatively lower-temperature part corresponding to the metallic state. In the higher-temperature part the change of mobility is very small compared with the change of carrier density. So the upper limit is close to the real case. The lower limit is arbitrarily set as E_ρ. For sample A with $T_p = 160$ K and $E_\rho = 543$ K, we got $J_{pd}S = 7481$ K for the upper limit ($\Delta = 1086$ K) and $J_{pd}S = 1399$ K for the lower limit ($\Delta = 543$ K). For the upper limit $J_{pd}S$ is much higher than a reasonable value. In this case a first-order transition for our sample A may be justified. Although this is a rough estimation, the clear thermal hysteresis and the sharp change of the resistance in the phase transition area shown in figure 2 is consistent with it.

For sample B, with $T_p = 233$ K and $E_\rho = 480$ K, we got $J_{pd}S = 2897$ K for the upper limit and $J_{pd}S = 1107$ K for the lower limit. Both are in the range of reasonable values. So the transition is more likely a second-order one according to the theory. As shown in figure 2, no clear thermal hysteresis of resistance was observed and the resistance decreases slowly with decreasing temperature from T_p. This behaviour resembles the $\rho(T)$ of a second-order transition with $\delta < \delta_c$ as shown in figure 1 of [17]. Also the noise spikes and peak near T_p may suggest a second-order phase transition. A brief explanation is as follows. Since the correlation length appears to be infinite for a second-order phase transition in the phase transition area, the typical energy associated with correlation length is small. If this energy distribution covers the low-energy window corresponding to the activation energy of $1/f$ noise, there may be large $1/f$ fluctuations near the transition area.

5. Conclusion

The temperature dependences of resistance and $1/f$ noise of two representative $Pr_{0.67}Sr_{0.33}MnO_3$ films were measured from liquid nitrogen temperature to room temperature. For the sample with a sharp resistance peak at $T_p \approx 160$ K, by recording the real-time voltage noise δV a strong reduction of noise was clearly observed in the metal–semiconductor transition regime. For the sample with a moderate resistance peak such noise reduction is absent, and some spikes and a peak are observed. Possible explanations are discussed. The bipolaron model for CMR is applied to fit the temperature dependences of resistance and to determine the order of phase transition. Basic agreement with theoretical predictions is found. Further work is under way.

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