

Effects of ferroelectric-poling-induced strain on the quantum correction to low-temperature resistivity of manganite thin films

R. R. Jia,¹ J. C. Zhang,^{1,*} R. K. Zheng,² D. M. Deng,¹ H.-U. Habermeier,³ H. L. W. Chan,² H. S. Luo,⁴ and S. X. Cao¹

¹Department of Physics, Shanghai University, Shanghai 200444, China

²Department of Applied Physics and Materials Research Center, The Hong Kong Polytechnic University, Hong Kong, China

³Max Planck Institute for Solid State Research, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

⁴State Key Laboratory of High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 201800, China

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Based on the complexity and difficult understanding on low-temperature resistivity minimum in manganites, the effect of ferroelectric-poling-induced strain on Kondo-type transport behavior was systemically investigated as a function of magnetic field for $\text{La}_{0.7}\text{Ca}_{0.15}\text{Sr}_{0.15}\text{MnO}_3$ manganite thin films grown on ferroelectric $0.67\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - 0.33PbTiO_3 (PMN-PT) single-crystal substrates. The results show that the low-temperature resistivity upturn is mainly caused from quantum correction effects driven by electron-electron interaction and inelastic scattering. Whether the PMN-PT substrate is in unpoled or poled state, the temperature where the resistivity shows an upturn near 15 K shifts to a higher temperature under magnetic field. The ferroelectric poling induces a reduction in the in-plane tensile strain and thus the lattice distortion of the film, which suppresses the resistivity upturn. These prove that the local lattice distortion relevant to the strain of the film is one of the main disorders that influence the resistivity upturn. The present results will be meaning to understand the physical mechanism of Kondo-type behavior at low temperature in colossal magnetoresistance manganites.

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I. INTRODUCTION

In perovskite manganites, the low-temperature resistivity minimum with a Kondo-type behavior has been widely studied both experimentally and theoretically. As it is well known, this Kondo-type behavior at low temperature is very complex and has not far been understood. It is said that the research about the low-temperature resistivity minimum is still under debate due to the complex of manganites system. So far, in order to explain these interesting and abnormal phenomena, various models have been proposed in terms of several different mechanism, such as spin-polarized tunneling through grain boundaries, Kondo-type effect due to spin disorder, and quantum corrections effect including e-e interaction enhanced by disorder and weak localization effect. Whereas from experiments, most obtained results include various factors above and strongly depend on the experimental sample's form. Thus, it is very important to investigate intrinsically the different and single factor which is related to the low-temperature resistivity minimum. In this process, it will be important to find the experimental sample with a single factor induced to low-temperature resistivity minimum and it was found that the magnetic fields have different effects on the low-temperature resistivity upturn for polycrystalline samples and thin films, and was attributed to different electronic conduction mechanisms in polycrystalline samples and thin films. The low-temperature resistivity minimum and upturn in polycrystalline samples was found to be suppressed by magnetic field and was interpreted in terms of the intergrain spin-polarized tunneling through grain boundaries.¹⁻³ The intergrain tunneling model was experimentally confirmed by Rozenberga and co-workers^{1,2} and Xu *et al.*³ The low-temperature resistivity upturn in intrinsically

disordered (e.g., magnetic/charge phase separation) samples⁴⁻⁶ was attributed to the Kondo-type effect which was previously observed in dilute magnetic alloys.⁷ It is noteworthy that Lee *et al.*⁸ theoretically pointed out the possible presence of quantum corrections to conductivity (QCC) in manganite single crystals and thin films.⁹⁻¹² Maritato *et al.*¹³ studied the effects of thickness (t) on the low-temperature (<60 K) transport properties of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ ($3.5 \leq t \leq 40$ nm) thin films, and found that the resistivity upturn appears with decreasing film thickness and interpreted this phenomena as due to the QCC effect which corrects the resistivity from two sources: (1) electron-electron (e-e) interactions enhanced by disorder and (2) weak localization effect due to the finite dimensions of the systems. Mukhopadhyay *et al.*¹² studied the coexist of quantum corrections and intergranular transport on $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ and $\text{Nd}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ thin films and ascribed it to e-e interaction in two dimensions. For manganite thin films, it is possible to study the QCC effect by varying different parameters (e.g., magnetic field and strain) which play important roles in determining the low-temperature electronic conduction mechanisms.

Although the substrate-induced lattice strain effect in manganite thin films has been studied extensively, a quantitative understanding of substrate-induced lattice strain effect is still needed from physical understanding viewpoints. As is known, the oxygen content, defects, and structural disorder strongly influence the properties of manganite thin films,¹⁴⁻¹⁶ and it is quite difficult to prepare thin films with the same oxygen content, defects, and microstructure, irrespective of their thicknesses. In order to obtain the intrinsic effects of the substrate-induced strain on the resistivity upturn at low temperatures, the extrinsic effects induced by the above-mentioned variables (e.g., oxygen nonstoichiometry) have to

be excluded. Recently, Thiele *et al.*¹⁷ and Zheng *et al.*¹⁸ reported that the strain state of manganite thin films epitaxially grown on ferroelectric $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ single-crystal substrates can be *in situ* dynamically modulated via ferroelectric poling or the converse piezoelectric effect.^{17,18} In this work, we obtained an ideal sample with low-temperature resistivity minimum resulted from single factor, i.e., quantum correction effect. Here, the poling intrinsically modifies the strain state of the $\text{La}_{0.7}\text{Ca}_{0.15}\text{Sr}_{0.15}\text{MnO}_3$ (LCSMO) and thereby alters e-e interaction enhanced by disorder underlying the resistivity upturn. This is an interesting and direct method to examine such quantum correction effect. The effects of the strain induced by the ferroelectric poling on the resistivity upturn give a direct evidence of the quantum correction effect at low temperature for manganite thin films. The effects of magnetic field and ferroelectric-poling-induced strain on the low-temperature resistivity upturn were systemically studied for epitaxial LCSMO thin films grown on the ferroelectric $0.67\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.33\text{PbTiO}_3$ (PMN-PT) single-crystal substrates. The ferroelectric poling *in situ* induces an in-plane compressive strain in the PMN-PT substrate, which modifies the strain state and hence the resistivity upturn of the LCSMO film. The correlation of the ferroelectric poling strain with low-temperature resistivity minimum was also given. This result will be meaning to understand the physical mechanism of low-temperature resistivity minimum.

II. EXPERIMENTAL DETAILS

LCSMO films were deposited on the (001)-oriented and polished PMN-PT substrates using dc magnetron sputtering.¹⁹ The deposition was carried out in an argon-oxygen flow with 60% Ar and 40% O_2 at a pressure of 5 Pa. The substrate temperature was kept at 700 °C during deposition. After deposition, the films were cooled to room temperature and postannealed in 1 atm of flowing O_2 at 700 °C for 30 min using a rapid thermal processor furnace. The thickness of the films is estimated to be ~ 100 nm. X-ray diffraction (XRD) patterns of the PMN-PT substrate and LCSMO film were recorded using a four-circle Bruker D8 Discover x-ray diffractometer equipped with $\text{Cu K}\alpha 1$ radiation. The results indicate that the films are *c*-axis preferentially oriented and have no secondary phases. The inset (a) of Fig. 1 is a schematic diagram of the LCSMO/PMN-PT structure and the electrical measurement circuit. A physical property measurement system (PPMS, Quantum Design) was used to measure the resistivity of the film between the two top-top gold electrode in the temperature range 2–35 K and in a magnetic field (up to 8 T) applied parallel to the film plane. The poling of the PMN-PT substrate was achieved by applying a dc poling field E to the LCSMO/PMN-PT structure through the LCSMO film (top electrode) and the bottom gold electrode.

III. RESULTS AND DISCUSSION

Figure 1 displays the temperature dependence of the resistivity for the LCSMO film when the PMN-PT substrate is

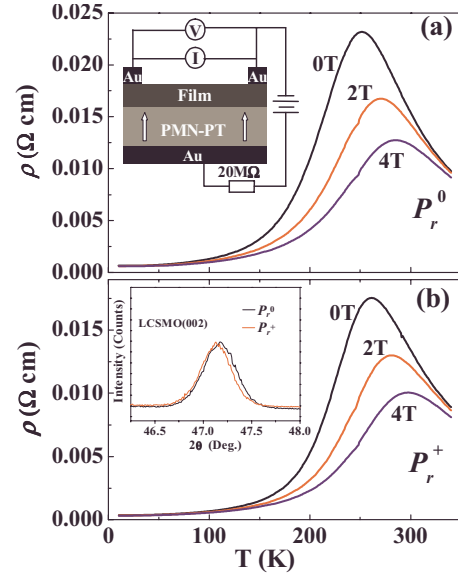


FIG. 1. (Color online) Temperature dependence of the resistivity for the LCSMO film when the PMN-PT substrate in (a) P_r^0 and (b) P_r^+ states at $H=0, 2,$ and 4 T. Inset in (a) shows a schematic of the LCSMO/PMN-PT structure and the electrical measurement circuit. Inset in (b) shows the XRD patterns of the LCSMO(002) reflection when the PMN-PT substrate is in P_r^0 and P_r^+ states, respectively.

in unpoled state (referred to as P_r^0) and positively poled state (referred to as P_r^+), respectively. It can be seen that the ferroelectric poling causes the insulator-to-metal transition temperature T_p shifts from 252 to 262 K and reduces the resistivity in the whole temperature range. Whether the PMN-PT substrate is in P_r^0 or P_r^+ state, when a magnetic field is applied, T_p increases and resistivity decreases, exhibiting the typical electrical behaviors of CMR materials. The inset (b) of Fig. 1 shows the XRD patterns of the LCSMO(002) reflection when the PMN-PT substrate was in P_r^0 or P_r^+ state, respectively. The results indicate that, associated with the ferroelectric poling, the LCSMO(002) reflection shifts from 47.18° to 47.13° , corresponding to an increase in the lattice constant c from 3.8497 to 3.8535 Å, with the latter value still smaller than that ($c \sim 3.8878$ Å) of the LCSMO ceramic target. With respect to the strain state of the LCSMO film when the PMN-PT substrate is in P_r^0 state, the in-plane tensile strain of the LCSMO film is reduced due to the ferroelectric poling. Such a change in the strain state of the film would reduce the tetragonal distortion of the MnO_6 octahedra, and thus reduces the electron-lattice coupling strength, thereby favoring the active hopping of electrons.^{19–21}

Figure 2 shows the resistivity (ρ) of the LCSMO film as a function of temperature (T) at various magnetic fields in the temperature range from 2 to 35 K when the PMN-PT substrate is in P_r^0 or P_r^+ state, respectively. All ρ - T curves show resistivity minimum at T_{\min} , which can be more clearly seen from Fig. 3 where the resistivity was normalized to that at 35 K. It is worth noting that, the resistivity upturn is strongly enhanced by the applied magnetic field and T_{\min} shifts to a higher temperature for both P_r^+ and P_r^0 states. As is known, for an intrinsically disordered system, the low-temperature resistivity is significantly affected by QCC, resulting from

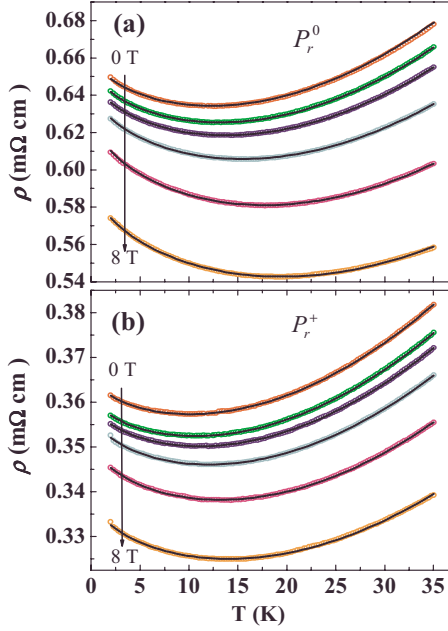


FIG. 2. (Color online) Resistivity as a function of temperature under magnetic fields for the LCSMO film when the PMN-PT substrate in the (a) P_r^0 and (b) P_r^+ states. The solid lines are fitted results using Eq. (6) as the $p=2$.

the weak localization effect and the e-e interaction.¹⁰ The total resistivity of the system in the first-order correction is given by

$$\rho(T, H) = \rho_0 + \rho_m(T, H) - \rho_0^2 [\sigma_{ee}(T, H) + \sigma_{wl}(T, H)] \quad (1)$$

with ρ_0 ($\rho_0 = 1/\sigma_0$) is the residual resistivity, σ_0 is the residual conductivity, $\rho_m(T, H)$ represents the magnetic resis-

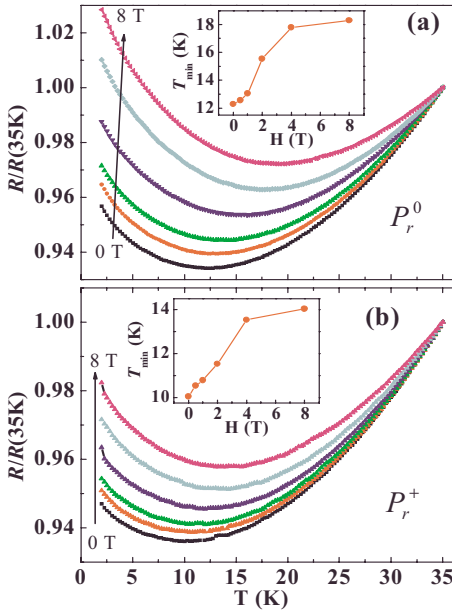


FIG. 3. (Color online) Normalized resistivity as a function of temperature under magnetic fields for the LCSMO film when the PMN-PT substrate in the (a) P_r^0 and (b) P_r^+ states. The insets show T_{\min} as a function of H .

tivity contributed from the anisotropic magnetoresistance and magnon scattering, σ_{wl} and σ_{ee} are the conductivity due to the weak localization effect, and e-e interaction enhanced by disorder,¹⁰ respectively. Generally, weak localization is expected to be negligible for two reasons. First, the weak localization results from the interference of complementary electron waves. However, the phase coherence of electron waves has already been destroyed by the strong spontaneous ferromagnetic field in these compounds.^{22,23} Second, Maritato *et al.*¹³ observed that the weak localization effect plays an increasingly important role in influencing the low-temperature resistivity upturn only for very thin films ($t < 20$ nm). For our sample, the film thickness is about 100 nm, which is relatively thick. According to Refs. 10 and 24, the weak localization effect can be easily suppressed by the application of a magnetic field. It is said that the dependence of low temperature resistivity on applied field is in accordance with previous studying in which the electron-electron interaction enhanced by disorder is dominated. So the weak localization effect can be deviated with it whereas the electron-electron interaction enhanced by disorder plays a key role.

Next, let us analyze the e-e correction model at low temperatures. In general, the resistivity at low temperatures can be described by

$$\rho(T, H) = \rho_{el}(T, H) + \rho_{in}(T, H), \quad (2)$$

where ρ_{el} and ρ_{in} are the resistivity contributed from elastic (electron-impurity and Coulomb interaction) and inelastic (e.g., electron-phonon interaction, electron-magnon interaction) scattering,¹ respectively. According to the theory of e-e correction to the residual conductivity in three-dimensional systems developed by Altshuler *et al.*,²⁵ the conductivity due to elastic scattering can be expressed as

$$\sigma_{el}(T, H) = \sigma_0 + \delta\sigma_{ee}(T, H), \quad (3)$$

where $\delta\sigma_{ee}(T, 0) = 0.0309 \frac{e^2}{\hbar L_T}$ and $L_T^{-1} = \sqrt{k_B T / \hbar D} = A\sqrt{T}$, where D is the carrier diffusion constant. So, Eq. (3) can be expressed as^{1,2}

$$\rho_{el}(T, H) = \rho_0 - a\sqrt{T}. \quad (4)$$

The resistivity due to quantum correction, resulting from the combined effect of e-e interaction and disorder, gives $T^{1/2}$ term and $a = 0.0309 A \rho_0^2 e^2 / \hbar$. On the other hand, the resistivity due to the inelastic scattering can be expressed as

$$\rho_{in}(T, H) = bT^p. \quad (5)$$

Substituting Eqs. (4) and (5) into Eq. (2), the total resistivity $\rho(T, H)$ can be obtained as

$$\rho(T, H) = \rho_0 - a\sqrt{T} + bT^p. \quad (6)$$

Using Eq. (6) and taking $p=2$,^{1,22} we fitted the temperature dependence of the resistivity under different magnetic fields shown in Fig. 2. The values of the fitting parameters obtained are shown in Table I. The solid lines in Figs. 2(a) and 2(b) are the fitted results, which agree well with the experimental data.

TABLE I. Summary of the fitted values of the parameters for the LCSMO film at $H=0, 0.5, 1, 2, 4,$ and 8 T when the PMN-PT substrate is in P_r^0 and P_r^+ state.

H (T)	P_r^0					P_r^+				
	ρ_0 (10^{-4} Ω cm)	a (10^{-5} Ω cm $K^{-1/2}$)	b (10^{-8} Ω cm K^{-2})	D (10^{-2} cm^2/s)	$N(E_F)$ (10^{-49} $\text{J}^{-2} \text{m}^{-3}$)	ρ_0 (10^{-4} Ω cm)	a (10^{-5} Ω cm $K^{-1/2}$)	b (10^{-8} Ω cm K^{-2})	D (10^{-2} cm^2/s)	$N(E_F)$ (10^{-49} $\text{J}^{-2} \text{m}^{-3}$)
0	6.70	1.00	6.78	1.49	0.39	3.70	40.80	3.17	0.83	1.26
0.5	6.60	1.00	6.50	1.41	0.42	3.60	43.39	3.10	0.67	1.61
1	6.50	1.00	6.22	1.32	0.45	3.60	44.84	3.04	0.62	1.75
2	6.50	1.00	5.83	1.32	0.45	3.60	49.46	2.96	0.51	2.13
4	6.30	2.00	5.54	0.29	2.12	3.50	54.06	2.82	0.38	2.93
8	6.00	2.00	4.84	0.24	2.71	3.40	53.32	2.53	0.35	3.29

The e-e interaction is only weakly dependent on magnetic field since the resistivity minimum still remains under a high magnetic field of 8 T. As is known, the conductivity due to e-e interaction under magnetic field can be expressed as

$$\sigma_{ee}(T, H) - \sigma_{ee}(T, 0) = -[e^2/(4\pi^2\hbar)]\sqrt{(k_B T)/(2\hbar D)}g_3(h), \quad (7)$$

where $g_3(h)$ is a function of the argument $h = g\mu_B\mu_0 H/(k_B T)$ and can be computed numerically and is a positive constant.^{8,10} So the e-e interaction would result in a positive magnetoresistivity. This could be understood in terms of magnetic-field-induced suppression of inelastic scattering whereas the elastic scattering is hardly affected by the magnetic field. This is consistent with the theoretical predictions by Rana *et al.*¹¹ who showed that T_{\min} increases with increasing magnetic field.

From the above discussion, the e-e interaction enhanced by disorder is possibly the main cause of the resistivity upturn at low temperatures. One of the disorders could be the random potential fluctuations due to different charge and sizes of the La^{3+} , Ca^{2+} , and Sr^{2+} ions.²⁶ The other one could be the strain-induced lattice distortion. In the presence of orbital degree of freedom, there are two degenerate orbital, i.e., $3d_{x^2-y^2}$ and $3d_{3z^2-r^2}$, of the e_g electron on each Mn site if the JT distortion is absent. Thus the orbital disorder leads to an additional off-diagonal disorder in the effective hopping integrals.²⁷

We now focus on the influence of ferroelectric-poling-induced strain on the QCC at low temperatures for the LCSMO film. Figure 4 shows the normalized resistivity $\rho(T)/\rho(35 \text{ K})$ under $H=0$ and 8 T when the PMN-PT substrate is in P_r^0 and P_r^+ state, respectively. It can be seen that the resistivity upturn is suppressed by the ferroelectric poling. For both states, we found similar variation in T_{\min} with magnetic field H , which is particularly prominent at high magnetic field. According to Ref. 28, the ferroelectric poling reduces the in-plane tensile strain and thus the lattice distortion of MnO_6 octahedra of the LCSMO film. This would reduce the orbital disorder, thereby suppressing T_{\min} and resistivity upturn.

We now discuss the fitting parameters shown in Table I. One can find that ρ_0 and b is decreased by the applied mag-

netic field while a is increased. The parameter a which represents the e-e interaction enhanced by disorder for P_r^0 state is larger than that for P_r^+ states. b for P_r^0 state is slightly higher than that for P_r^+ state. This indicates that the lattice distortion is one of the disorders that influence the low temperature upturn significantly. Using the Einstein formula $\sigma_0 = e^2 N(E_F) D$,⁸ the values of electron diffusion constant D and density of state E_F are calculated and also shown in Table I. As can be seen in Fig. 2, the theoretical fits are in quantitative agreement with the experimental data. The values of D for the LCSMO film is close to those of $\text{La}_{0.7}\text{A}_{0.3}\text{MnO}_3$ (A: Ca, Sr, and Ba) manganites²² but is much smaller than those of normal metals [e.g., Cu ($D_{\text{Cu}} \sim 2.2 \times 10^2 \text{ cm}^2/\text{s}$)], indicating that the charge carriers of the LCSMO films have a lower diffusivity than those of metals. It is known that strong e-e interaction can result in a change in the density of state near

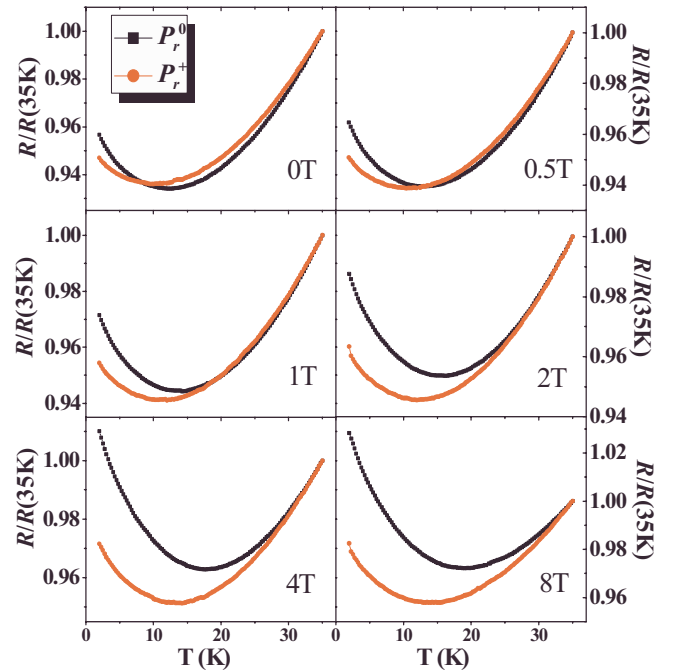


FIG. 4. (Color online) Normalized resistivity $\rho(T)/\rho(35 \text{ K})$ as a function of temperature for the LCSMO film when the PMN-PT substrate is P_r^0 and P_r^+ states, respectively.

the Fermi energy, which has been suggested for the possible origin of the low-temperature resistivity upturn. In our samples, $N(E_F)$ for P_r^+ state is larger than that for P_r^0 state. The scattering amplitude interferes coherently, leading to an increased Coulomb coupling in the density of state at the Fermi energy. Our results demonstrate that the e-e interaction enhanced by disorder may play an important role in determining the resistivity upturn at low temperatures. The effects of the strain induced by the ferroelectric poling on the resistivity upturn give a direct evidence of the QCC effect at low temperatures for manganite thin films.

IV. SUMMARY

In order to understand the complex behavior of Kondo-type resistivity minimum at low temperature, the effects of the ferroelectric-poling-induced strain and magnetic fields were systemically studied for the low-temperature resistivity upturn of $\text{La}_{0.7}\text{Ca}_{0.15}\text{Sr}_{0.15}\text{MnO}_3$ thin films grown on ferroelectric PMN-PT single-crystal substrates. For P_r^0 and P_r^+ states, T_{\min} shifts to a higher temperature under magnetic field. Whether the film is under a low or high magnetic field, the ferroelectric poling suppresses the low-temperature resistivity upturn of the film, which is interpreted as due to strain-induced reduction in the local structural disorder. We explain the low-temperature resistivity upturn in terms of QCC effect originating from inelastic scattering and electron-electron interaction enhanced by disorder which is closely related to the local lattice distortion relevant to the strain induced by ferroelectric poling. This is an interesting and direct method to examine such quantum correction effect. The effects of the strain induced by the ferroelectric poling on the resistivity upturn give a direct evidence of the QCC effect at low temperatures for manganite thin films.

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*Corresponding author; jczhang@staff.shu.edu.cn

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