Strain effect on electronic transport and ferromagnetic transition temperature in $La_{0.9}Sr_{0.1}MnO_3$ thin films

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We report on a systematic study of strain effects on the transport properties and the ferromagnetic transition temperature T_c of high-quality $La_{0.9}Sr_{0.1}MnO_3$ thin films epitaxially grown on (100) $SrTiO_3$ substrates. Both the magnetization and the resistivity are critically dependent on the film thickness. T_c is enhanced with decreasing the film thickness due to the compressive stain produced by lattice mismatch. The resistivity above 165 K of the films with various thicknesses is consistent with small polaronic hopping conductivity. The polaronic formation energy E_P is reduced with the decrease of film thickness. We found that the strain dependence of T_c mainly results from the strain-induced electron-phonon coupling. The strain effect on E_P is in good agreement with the theoretical predictions.

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I. INTRODUCTION

The discovery of the colossal magnetoresistance effect in epitaxial manganite thin films has renewed interest in the doped manganite perovskite materials $L_{1-x}B_xMnO_3$ (L = trivalent rare-earth ions, B = divalent alkaline-earth ions) for potential sensor and magnetic recording applications as well as the need to understand the mechanisms underlying their behavior.[1–3] It has been found that properties such as ferromagnetic transition temperature T_c, resistivity ρ, and magnetoresistance are sensitive to the epitaxial strain due to lattice mismatch of the film with substrate.[4–10] When the film is grown on a substrate whose lattice parameter is smaller or larger than that of the bulk material, the epitaxial strain is expected to be compressive or tensile, respectively. Compressive strain usually reduces the resistivity and shifts T_c towards higher temperature. These effects have been confirmed in $La_{0.7}Ca_{0.3}MnO_3$ films[6] and $La_{0.7}Sr_{0.3}MnO_3$ films[7] grown on various substrates.

The observed strain effect is usually interpreted qualitatively within the double-exchange model,[11] since the hopping matrix element t could be altered by epitaxial strain through changing the Mn-O bond length d and the Mn-O-Mn bond angle θ. It has been also proposed that the Jahn-Teller electron-phonon coupling plays an important role in the strain effect on T_c.[12] However, recent detailed studies show that compressive strain does not always lead to enhancement of T_c,[9] while the cationic vacancies due to the oxygen annealing significantly enhance the T_c values much higher than any bulk values in the series compounds.[5,10] In most cases, tensile strain suppresses ferromagnetism and reduces T_c in manganite films. But some anomalous results have also been reported, showing T_c enhanced by tensile strain.[13–15] Most interestingly, there are reports of multiple phase segregation in fully strained epitaxial films.[16] The ferromagnetic coupling within the metallic regions accounts for the changes of T_c and conductivity. Thus, the strain effect in manganite films is far from being fully understood and challenging.

Lightly doped $La_{1-x}Sr_xMnO_3$ shows a great variety of intriguing phenomena originating from a pronounced interplay between spin, lattice, charge, and orbital degrees of freedom. As a result many phenomena like charge order,[17,18] orbital order,[19] and phase separation[20] have been recently observed in this regime of the phase diagram. $La_{0.9}Sr_{0.1}MnO_3$ is in the phase boundary of a spin-canted antiferromagnetic insulator and a ferromagnetic insulator.[21–24] This material has the lowest T_c among the series compounds,[22,25] which makes it possible to perform systematic investigations of the resistivity in the paramagnetic regime over a broad temperature range without using specialized equipment to extend the temperature range. Meanwhile, the pressure derivative of T_c, dT_c/dP, in this material is highest among the manganese perovskites.[25–27] It has been generally believed that pressure changes T_c and ρ in a similar manner as epitaxial strain. Thus, transport properties, transition temperatures, and phase transitions are expected to be significantly affected by epitaxial strain in $La_{0.9}Sr_{0.1}MnO_3$ films. Moreover, these investigations are most important for the understanding of fruitful phenomena and the use of these films as magnetic devices as well as air electrodes in high-temperature solid oxide fuel cells.[28,29]

In this work we investigate the transport properties by measuring the resistivity and magnetization of epitaxial $La_{0.9}Sr_{0.1}MnO_3$ films on $SrTiO_3$. The data clearly show that the high-temperature resistivity of the films can be well ascribed by a model for small-polaron hopping in the adiabatic limit. We experimentally find that the small-polaronic formation energy E_P decreases with the reduction of film thickness, which can account for the strain effect on T_c. We suggest that the electron-phonon coupling is responsible for the strain effect on the high-temperature electronic transport and the ferromagnetic transition temperature.

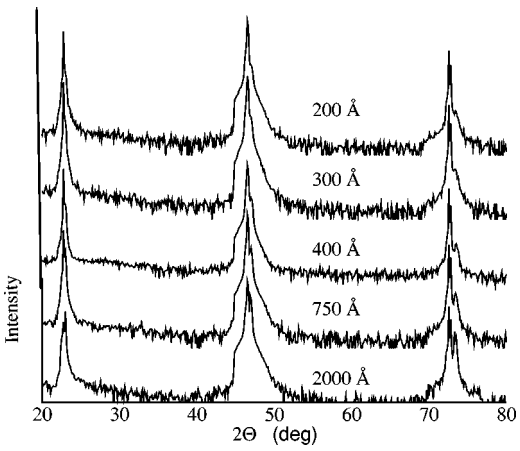

FIG. 1. Room-temperature XRD of $La_{0.9}Sr_{0.1}MnO_3$ films for various thicknesses.

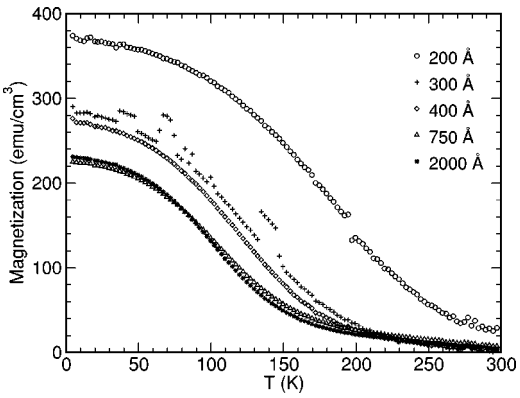

FIG. 2. Magnetization as a function of temperature measured in a field of 0.5 T of $La_{0.9}Sr_{0.1}MnO_3$ films with various thicknesses.

II. EXPERIMENTAL DETAILS

Thin films of $La_{0.9}Sr_{0.1}MnO_3$ were grown using the pulsed laser deposition technique. The target used had a nominal composition of $La_{0.9}Sr_{0.1}MnO_3$. The substrates were (100) single crystal of $SrTiO_3$. The laser energy density on the target was 2 J/cm^2 and the ablation rate was 5 Hz. The substrates were kept at a constant temperature of 850 °C during the deposition which was carried out at a pressure of 0.40 mbar of oxygen. The films were *in situ* annealed at 940 °C in oxygen at 1.0 bar for 30 min. This procedure always results in films of high crystalline quality and in very sharp film-substrate interfaces. The thickness of the films was varied from 200 to 2000 Å as measured by Dektak. The chemical composition of the films was determined by microprobe analysis, which showed a (La,Sr)/Mn ratio of 1:1 and a Sr content of $x=0.10\pm0.01$.

The structural study was carried out by x-ray diffraction (XRD) at room temperature by a Rigaku x-ray diffractometer with a rotating anode and Cu $K\alpha$ radiation, $\lambda=1.5406$ Å. The resistivity ρ was measured from unpatterned samples with sputtered chromium gold contacts using a standard four-probe technique. Magnetization M was recorded in a magnetic field parallel to the film plane using a Quantum Design MPMS superconducting quantum interference device (SQUID) magnetometer as a function of temperature.

III. RESULTS AND DISCUSSION

Figure 1 shows the evolution of the room temperature XRD data for $La_{0.9}Sr_{0.1}MnO_3$ thin films with thicknesses from 200 to 2000 Å. Each sample is single crystal and (l00) oriented without other impurity phases. Above $T=105$ K, $SrTiO_3$ has a perfect cubic perovskite structure with a lattice parameter $a=3.905$ Å. $La_{0.9}Sr_{0.1}MnO_3$ has a distorted perovskite structure due to the tilting of the MnO_6 octahedra and Jahn-Teller distortion, which results in a slightly orthorhombic structure. The bulk lattice parameters for this compound at room temperature are[30] $a=5.5469$ Å, $b=5.560\,33$ Å, and $c=7.7362$ Å. The in-plane lattice mismatch between the film and the substrate is given by $\epsilon=[d_{bulk}-d_{strained}]/d_{bulk}$, with d a lattice parameter. Epitaxially grown $La_{0.9}Sr_{0.1}MnO_3$ film on (100) $SrTiO_3$ substrates is under compressive strain since $d_{bulk}>d_{strained}$ with the bulk value $d_{bulk}=3.927$ Å. With decreasing the film thickness, the in-plane lattice parameter of the film decreases and the compressive strain is then enhanced.

Figure 2 shows the temperature dependence of the magnetization measured in 0.5 T of films with various thicknesses after correction for the magnetization of the substrate. The curves have been measured by warming up in a magnetic field after zero-field cooling. The features of the M-T curves are ferromagnetic with $M\sim230$–360 emu/cm^3 at 10 K. The magnetization was taken at 0.5 T to avoid the variation due to magnetic domain rotation at lower fields. Both T_c and M increase with decreasing film thickness. The value of T_c for 200 Å thin film is 50 K higher than the bulk value.[26] We had not observed a magnetization jump occurring at a characteristic temperature T_{CA} as appeared in the $La_{0.9}Sr_{0.1}MnO_3$ single crystals,[26,31] which indicates a canted antiferromagnetic state as confirmed by neutron scattering experiments.[17] This is not surprising since the strained films usually show properties much different from the bulk compounds in manganites.[5]

Although the reduction of film thickness should enhance T_c under compressive strain as we observed in Fig. 2, there are few measurements in other manganites films to support this phenomenon. The experiments on $La_{0.8}Ca_{0.2}MnO_3$ films grown on $LaAlO_3$ do not always show a correlation between the compressive strain and T_c.[9] Interestingly, anomalously high T_c and metal-insulator transition temperature T_{MI} (100 K higher than bulk values) have been observed in this strained film with 1000 Å thickness after annealing under oxygen.[10] For this $La_{0.8}Ca_{0.2}MnO_3$ film, T_{MI} is 30 K higher than the highest $T_{MI}=260$ K found for $x=0.33$ bulk compound. Thus, the large enhancement of T_c and T_{MI} in this film should be dominated by compressive strain. The lack of this enhancement observed previously in $La_{0.8}Ca_{0.2}MnO_3$ thin film may be due to oxygen deficiency.

The results of the temperature dependence of the resistivity are shown in Fig. 3. The resistivity of our films displays semiconducting behavior at high temperatures and metallic behavior for $T_{CA}\leqslant T\leqslant T_{MI}$. It has an upturn at T_{CA} and then

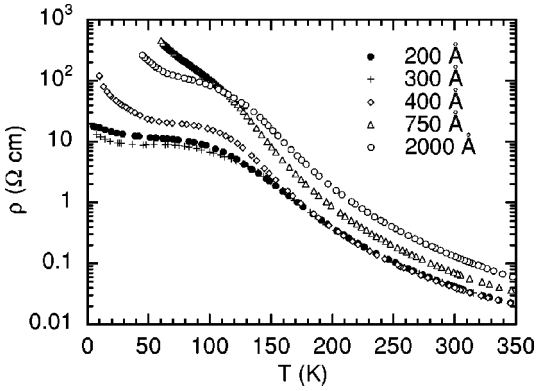

FIG. 3. Temperature dependence of resistivity of $La_{0.9}Sr_{0.1}MnO_3$ films with various thicknesses.

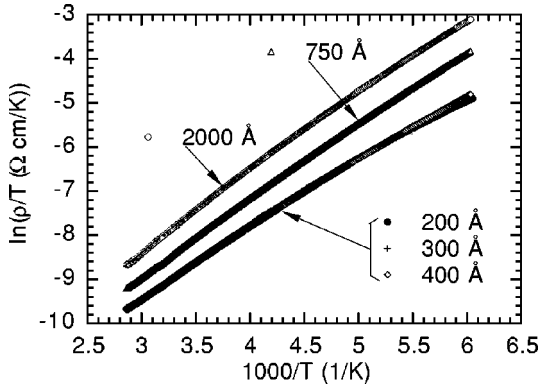

FIG. 4. Plot of $\ln(\rho/T)$ vs $1000/T$ of $La_{0.9}Sr_{0.1}MnO_3$ films with various thicknesses.

becomes of semiconducting character. A neutron scattering study demonstrates that the point of resistivity upturn is consistent with the onset temperature of the polaron order.[17] The magnitude of resistivity of our films is smaller than those of single crystals.[22,24,32] For example, the resistivity of the 2000 Å film at $T=100$ K is 83.7 Ω cm. Note that the compressive strain decreases the resistivity in our thin films. This behavior is typical for manganites films under compressive strain.[6,7] The observed T_{MI} (defined as the temperature where $d\rho/dT$ changes sign) of ~100–150 K in our films are comparable to those of $La_{0.9}Sr_{0.1}MnO_3$ single crystals.[22,24,32] For films with thicknesses $d=750$ and 2000 Å, T_{MI} almost coincides with T_c. However, T_{MI} is significantly smaller than T_c for ultrathin films. The scenario to correlate with this observation could be the existence of microscopic phase segregation due to the formation of small ferromagnetic clusters, which are large enough to give a magnetic contribution in ultrathin films but not to allow metallic conductivity appearing in zones of ferromagnetic insulating behavior. The smaller T_{MI} value compared to T_c has reported previously in $La_{0.67}Sr_{0.33}MnO_3$ thin films.[33] Recent nuclear magnetic resonance measurements in $La_{2/3}Ca_{1/3}MnO_3$ films on $SrTiO_3$ give strong evidence in favor of the existence of microscopic phase separation.[16]

An additional increase of resistivity on cooling can be seen at low temperatures proceeded by a minimum at T_{CA}. The structural data of single crystals show that a phase transformation from a pseudocubic O''-type to an orthorhombic O'-type structure occurs near T_{CA}.[23,34] The low-temperature phase is known to be a spin-canted antiferromagnetic phase for $0\leqslant x\leqslant 0.1$,[21] which results from competing antiferromagnetic superexchange interactions between half-filled t_{2g} orbitals along the *c*-axis Mn-O-Mn bonds and ferromagnetic double-exchange interaction via e_g conduction electrons. With the reduction of the film thickness, T_{CA} shifts towards low temperatures and ρ decreases in the insulating low-temperature phase. It has been reported that T_{CA} increases and ρ decreases under pressure in $La_{0.9}Sr_{0.1}MnO_3$ single crystals.[26] Although ρ behaves in a similar manner under compressive strain and external pressure, the observed variation of T_{CA} is in sharp contrast with the pressure measurements. It has been established[20] that pressure influences T_{CA} in the same way as an increase in x with a maximum within the range $0.12<x<0.15$ for slightly doped $La_{1-x}Sr_xMnO_3$. At low pressures, the thermoelectric power through T_{CA} is sensitive to the charge carrier density. It is indicated that pressure induces the change of carrier concentration, which should account for the dependence of T_{CA} on pressure. The growth conditions such as film deposition and oxygen annealing are the same for all films studied here. The carrier concentration in these films should not be different. Therefore, the dependence of T_{CA} on strain is possibly different from the pressure effect on T_{CA}.

An interesting feature is the absence of the jump in resistivity in films near $T\sim 330$ K. Structural analyses of $La_{0.9}Sr_{0.1}MnO_3$ crystals[24,30,34] reveal that the system undergoes another structural transition around the characteristic temperature $T_s=330$ K from an orthorhombic *O* phase having a dynamic Jahn-Teller distortion to a orthorhombic O' phase at lower temperatures where Jahn-Teller distortion becomes static and cooperative. The jump in resistivity at T_s in single crystals has been reported by Urushibara *et al.*[22] The absence of jump indicates that the compressive strain in films either suppresses the structural phase transition or shifts T_s towards higher temperatures above 350 K. There is competition between the charge mobility and the structural phase transition in the slightly doped $La_{1-x}Sr_xMnO_3$.[18] The change tendency of T_s and T_{CA} is usually different under pressure or magnetic field.[18,20] In our films, T_{CA} decreases with decreasing film thickness due to the compressive strain. Thus, the increase of T_s is possible under compressive strain.

The preconditions for polaron formation—namely, large electron-lattice coupling and low electronic hopping rates—appear to be satisfied for manganites.[35] In Fig. 4 we have represented $\ln(\rho/T)$ versus inverse temperature. A linear behavior is obtained between 165 and 350 K. This is strong support for the mechanism of adiabatic small-polaron conduction. The resistivity as a result of the hopping of adiabatic small polarons is, within Emin-Holstein theory,[36] given by

$$\rho = AT\exp\left(\frac{E_A}{k_BT}\right). \tag{1}$$

Here the prefactor *A* depends on the polaronic concentration, the hopping distance, and the frequency of the longitudinal optical phonon. The activation energy E_A has the form[37] $E_A=E_P/2+\epsilon_0-J$, where ϵ_0 is the energy required to generate intrinsic carriers and *J* is the transfer integral.

TABLE I. Thickness dependence of the activation energy E_A, the resistivity coefficient A, and the ferromagnetic transition temperature T_c in $La_{0.9}Sr_{0.1}MnO_3$ films.

Thickness (Å)	E_A (meV)	A (10^{-6} Ω cm/K)	T_c (K)
200	119.1	1.86	194.9
300	124.8	1.28	150.0
400	126.8	1.19	116.9
750	139.6	1.28	100.0
2000	141.3	2.39	105.6

From the fit to Eq. (1), the values of A and E_A are obtained. These data are summarized in Table I. The fitting for ρ is valid for temperatures larger than half the Debye temperature Θ_D. This is fulfilled for the present films since specific heat measurements show Θ_D in the 255–360 K range.[38,39] We noted that the fitting adiabatic prefactor A is in the range from 1.19×10^{-6} to 2.39×10^{-6} Ω cm/K, which is typical for small polaronic conduction as observed in $La_{0.67}Ca_{0.33}MnO_3$ films[40] as well as $(La_{1-x}Gd_x)_{0.67}Ca_{0.33}MnO_3$ films.[41]

There have been studies of the high-temperature resistive behavior in $La_{0.9}Sr_{0.1}MnO_3$ bulk materials.[42–44] The reported conduction mechanisms are controversial. Early measurements on the ceramic $La_{0.9}Sr_{0.1}MnO_3$ show that the high-temperature resistivity obeys the small-polaron transport behavior in the nonadiabatic limit with an activation energy $E_A=0.2$ eV.[42] In single crystals, some groups found that their data can be well fitted by a variable-range-hopping model $\rho=\rho_0(T/T_0)^{1/2}\exp[(T_0/T)^{1/4}]$ with $T_0=1.72\times10^8$ K in the paramagnetic regime,[43] while others[44] reported the resistivity follows a small-polaron model in the adiabatic limit above T_{MI} with activation energy $E_P=0.3$ eV. The high-temperature resistivity of our films with various thicknesses is consistent with adiabatic small-polaron hopping conductivity. It has been generally accepted that the conductivity can be well ascribed by adiabatic small-polaron model in $La_{1-x}Ca_xMnO_3$ films.[37,40,41,45–47] Our present data provide clear support for the existence of this conductivity mechanism in $La_{1-x}Sr_xMnO_3$ films.

At high temperatures and in the adiabatic limit where the contribution from ϵ_0 and J may be neglected, the variation of E_P is approximately affected by the change of E_A. Taking $E_P=2E_A$, we have plotted the thickness dependence of E_P in Fig. 5. The thickness dependence of T_c is also plotted for comparison. It is interesting to notice that the variation of T_c with thickness can be well reflected by the thickness dependence of E_P. For the thick films, the strain is relaxed. Both T_c and E_P scarcely change with the thickness. Below 750 Å, with a reduction of the film thickness, E_P decreases, whereas T_c increases. It is therefore indicated that the electron-phonon coupling possibly dominates the strain effect on T_c.

The polaronic formation energy E_P is usually related to the effective bandwidth W_{eff} in polaronic models. Zhao *et al.*[48] proposed an effective bandwidth of the form $W_{eff}=W\exp(-\gamma E_P/\hbar\nu)$, where W is the electronic "bare" bandwidth, ν is the characteristic vibration frequency of the optical phonon mode, and γ depends on the ratio E_P/W. According to the model proposed by Varma,[49] T_c can be written as

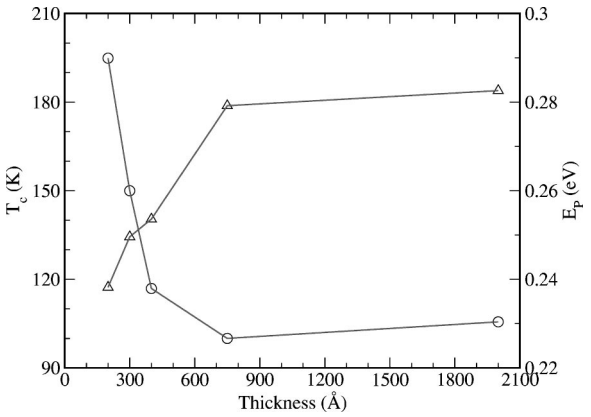

FIG. 5. Thickness dependence of the ferromagnetic transition temperature T_c (circles) and the polaronic formation energy E_P (triangles) in $La_{0.9}Sr_{0.1}MnO_3$ films.

$$T_c=\frac{0.1}{2}W\exp\left(-\frac{\gamma E_P}{\hbar\nu}\right)n(1-n), \tag{2}$$

where n denotes the carrier concentration. Considering that ν is related to the isotope mass M through $\nu\propto M^{-1/2}$, the oxygen isotope exponent α ($\equiv -d\ln T_c/d\ln M$) is then given by $\alpha=0.5\gamma E_P/\hbar\nu$. The strain coefficient of T_c, $d\ln T_c/d\epsilon$, is readily obtained from Eq. (2):

$$\frac{d\ln T_c}{d\epsilon}=\frac{d\ln W}{d\epsilon}-2\frac{d\alpha}{d\epsilon}. \tag{3}$$

For $La_{0.9}Sr_{0.1}MnO_3$, the pressure coefficient of T_c has been found by Senis *et al.*[26] to be $d\ln T_c/dP=0.16$ GPa^{-1}. Using the lattice compressibility $\kappa_a=2.32\times10^{-3}$ GPa^{-1},[50] we obtain $d\ln T_c/d\epsilon=69$. The electronic bandwidth W of the manganites can be estimated by the average Mn-O bond distance d and the Mn-O-Mn angle θ by using the relation:[51] $W\propto\cos\phi/d^{3.5}$, where $\phi=(\pi-\langle\theta\rangle)/2$. The pressure dependence of $\cos\phi$ has been determined by neutron diffraction measurements[50] to be $(\cos\phi)^{-1}d\cos\phi/dP=2.1\times10^{-4}$ GPa^{-1}. Taking the value of κ_a as the bond compressibility κ_d, the calculated $d\ln W/d\epsilon$ is 3.6. Thus, $d\alpha/d\epsilon=-32.7$ is obtained from Eq. (3). In $La_{0.9}Sr_{0.1}MnO_3$, the oxygen isotope exponent $\alpha=0.2$ reported previously by Zhao *et al.*[48] Based on the above-determined parameters, one estimated the pressure derivate of α, $d\alpha/dP=-0.076$ GPa^{-1}. This value is very close to the reported value of -0.05 GPa^{-1} in $La_{0.65}Ca_{0.35}MnO_3$.[52]

According to the expression for α, $d\alpha/d\epsilon$ is then expressed as

$$\frac{d\alpha}{d\epsilon}=\alpha\left(\frac{d\ln E_P}{d\epsilon}-\frac{d\ln\nu}{d\epsilon}\right). \tag{4}$$

The Raman spectra of $La_{0.9}Sr_{0.1}MnO_3$ have been collected previously by Podobedov *et al.*[53] The sharp peaks at the top

of the wide band are located at 243, 493, and 609 cm^{-1}. The high-frequency B_{1g} mode at 609 cm^{-1} is suggested as a stretching Mn-O vibration. Recent high-pressure studies[54] show that this stretching mode is the most sensitive to pressure with an initial pressure coefficient $d\ln\nu/dP = 0.01\ GPa^{-1}$. Thus $d\ln\nu/d\epsilon = 4.4$. Equation (4) gives $d\ln E_P/d\epsilon = -159$. It follows that E_P decreases with increasing compressive strain. This is in good agreement with our experimental fitting parameters as shown in Fig. 5. Combining Eqs. (3) and (4), we can conclude that the strain dependence of T_c mainly results from the strain dependence of the polaronic formation energy though there are also contributions from the electronic bandwidth W and the characteristic phonon frequency ν.

IV. CONCLUSIONS

We have epitaxially grown $La_{0.9}Sr_{0.1}MnO_3$ thin films on $SrTiO_3$ substrates. The high-temperature resistivity of the films with various thicknesses obeys the small-polaron hopping conductivity in the adiabatic limit. We experimentally found that the small-polaronic formation energy E_P decreases with the reduction of the film thickness, which mainly accounts for the the strain effect on T_c. By theoretical analysis, we found that the contribution from the electronic bandwidth is much smaller than that from the electron-phonon interaction. We therefore concluded that electron-phonon coupling is responsible for the strain effect on the high-temperature electronic transport and the ferromagnetic transition temperature in our films.

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