Electric field control of the small-polaron hopping conduction in spatial confined $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$/PMN-PT heterostructure

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Electric field control of the small-polaron hopping conduction in spatial confined $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$/PMN-PT heterostructure

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The electric field dependent high-temperature small-polaron hopping conduction was investigated in patterned $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ strips. The small-polaronic activation energy E_A and the carrier localization were found to decrease with the reduction of the strip size. Meanwhile, a similar dependence on the strip size was also obtained for the calculated small-polaron coupling constants, which could be related to the strain relaxation in strips. These results indicate that the spatial confinement prefers to delocalize the carrier and reduce the electron-phonon interaction. Furthermore, opposite variation trends of E_A under negative and positive electric field were found in the strips with small size, which could be attributed to the enhancement of polarization effect induced by the reduction of strip size. [http://dx.doi.org/10.1063/1.4974887]

Recently, manganites have been intensively studied due to their rich phenomena, such as colossal magnetoresistance (CMR), metal-insulator transition (MIT), antiferromagnetic-ferromagnetic (AFM–FM) phase transition, charge ordering (CO), and phase separation (PS), etc.[1–3] It is generally agreed that the strong coupling among charge, orbital, spin and lattice degrees of freedom is the key factor for a comprehensive understanding of these novel phenomena. Especially, the strong electron-phonon coupling, i.e. Jahn-Teller distortion, plays a key role in driving the extraordinary phenomena like charge orbital ordering and phase separation via splitting the occupied e_g level in Mn^{3+} and affecting the orbital occupation.[4,5] Meanwhile, studies have shown that the lattice distortion in manganites can trap e_g electrons on Mn site and lead to the formation of small polarons.[6,7] In manganite thin films, strong polaronic effects are expected at temperature above MIT, $T>T_{MI}$. In fact, at high temperature the magnetic correlation can be neglected. The thermal activated motion of such polarons dominates the electric transport, so the small polaron hopping plays a key role in the conduction mechanism at high-temperature region.[8] Hence, by studying the characteristic of the resistivity in paramagnetic insulating phase, we can get easy access to the information about e_g electron localization and electron-phonon coupling strength that directly connects with the process of polaron activation.[9]

On the other hand, phase separation, which is referred as the coexistence and competition of the energy degenerated ferromagnetic metallic phase and charge orbital ordering insulating phase, has been recognized as a common phenomenon and a key factor to understand various novel physical properties in manganites.[10–13] Recently, a series of unique phenomena were observed in spatial confined phase-separated manganite films.[14,15] Several experiments showed that the spatial confinement

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breaks the percolation channel in manganite films and introduces a single charge/orbital ordering insulating domain in the sample, which provides a convenient and good platform to investigate the physical properties of the charge ordering insulating phase and related phenomena. However, most of the works on spatial confined manganite focus on the phase-separated region[14–17] and little is known at the high-temperature region in the paramagnetic insulating phase where the polaron hopping dominates the conduction behavior and contains the information of electron-phonon coupling. Since the lattice distortion and thus electron-phonon coupling is important to the charge orbital ordering phase and the phase coexistence and competition, the investigation of the evolution of small-polaron hopping conduction in the spatial confined film will help to reveal the role of electron-phonon coupling in the modulation of phase separation by spatial confinement.

In this paper, we study the influence of spatial confinement on the high-temperature conduction properties and small polaron hopping in micro-striped $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ (PCSMO) film by measuring the high temperature conduction properties of the stripe-patterned film. During these measurements, various electric bias were applied on PMN-PT substrate to introduce extra lattice strain in the film so that we can clarify the influence of spatial confinement on lattice distortion. Recent studies have shown that lattice strain can effectively modulate the lattice distortion and the electron-phonon coupling.[9,18] The thin film of PCSMO was used owing to its well-known micrometer-scale phase separation and moderate competition of two separated phases due to the substitution of Sr^{2+} for Ca^{2+}.[4] The single crystal PMN-PT was used as a substrate due to its excellent converse piezoelectric effect[13,19] so that the polaron hopping and thus the transport properties in PCSMO film can be easily manipulated by tuning the lattice strain (or electric field applied on the substrate).

The PCSMO film was deposited on (001) oriented PMN-PT substrate by pulsed laser deposition. The deposition was performed with a substrate temperature of 700 °C and an oxygen pressure of 100Pa. After deposition the sample was cooled down to room temperature in 1500Pa pure oxygen. The thickness of the film was set to 100nm by controlling the growth time. The crystalline structure was determined by high-resolution Bruker D8 Discover X-ray diffraction (XRD) using Cu-Kα radiation. The film was patterned into narrow strips with the width varying from 50μm down to 3μm. Au layers were deposited on the surface of the strips and bottom of the substrate to serve as electrodes. The transport properties were measured by the superconducting quantum interference device (SQUID)–VSM), equipped with a home-made electric-measurement module. A kethley 6517 electrometer was used to apply the gate electric field on the substrate of PMN-PT.

The X-ray diffraction pattern of the film is shown in Fig. 1(a), which demonstrates that the film is highly oriented along the out-of-plane direction and exhibits no impure phases. It is determined from the diffraction pattern that the as-prepared film undergo out-of-plane compress strain of −0.62%. The temperature dependent resistance for the as-prepared film exhibits a typical phase separation characteristic with a thermal hysteresis about 7K due to the percolation transport behavior, as shown in Fig. 1(b).

The temperature dependent resistivity for all strips were measured during cooling process over a temperature range from 20K to 300K. During measurements, various electric fields were applied

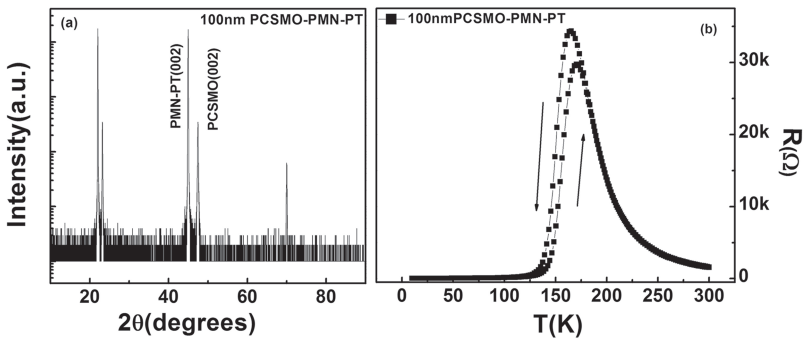

FIG. 1. (a) X-ray diffraction pattern of $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ film on PMN-PT substrate. (b) Temperature dependent resistance of $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ film.

on the PMN-PT substrate to change the lattice strain and thus the lattice distortion in the patterned strips. Firstly, due to the strong electron–phonon (e–ph) coupling, the polaron hopping behavior is expected above T_{MI} in hole-doped manganite. For our samples, we do find a similar thermally activated form in the high-temperature conduction region of strips with different widths, which can be well fitted by the adiabatic small polaron hopping (SPH) model. According to the SPH model the resistivity can be expressed in the following equation: $\rho(T) = AT\exp(E_A/k_BT)$ with k_B the Boltzmann constant, and $A = 2k_B/(3ne^2a^2\nu_{ph})$ a constant that depends on the polaronic concentration n, the hopping distance a, and the frequency of the longitudinal optical phonon ν_{ph}.[20,21] E_A is the activation energy for the polaron hopping, and approximately half of the polaron binding energy in the adiabatic limit. The inverse temperature dependence of $\ln(\rho/T)$ for all strips (with width of 3, 5, 10, 50μm) under zero electric field is presented in Fig. 2. A clear linear behavior is seen in the high temperature range above $\theta_D/2$ with θ_D being the Debye temperature. The solid lines in the plot are linear fits to the experimental data. The activation energy E_A of each strip under zero electric field is calculated from the fitting parameter, which is lower than that of the as-prepared film (see the inset of Fig. 2). This result implies that the spatial confinement has a significant effect on the e_g electron localization and thus the polaron hopping in PCSMO film. The reduction in the width of the strip would break the crystalline symmetry of the film and leads to the strain relaxation in the strip, especially at the edge of the strip.[22] As the lattice strain could change the bending condition of the Mn-O-Mn bond and induce compression or elongation of MnO_6 octahedra, the relaxation of lattice strain due to spatial confinement will weaken the Jahn-Teller distortion and thus release the trapped electrons, resulting in the reduction of the polaronic hopping potential barrier and, hence, the decrease in the activation energy. Moreover, one could find that the activation energy decreases monotonously with the width of strip (the inset of Fig. 2). In fact, with the reduction of the strip width, the edge proportion in the surface of the strip increases, leading to a continuous decrease in activation energy.

The influence of the spatial confinement on the electric field control of the high-temperature conduction were also studied. A series of electric field was applied and maintained for measurements of transport properties for each strip. The $\ln(\rho/T)$ vs 1/T curves of each strip with different electric field shows that the high-temperature resistance of all the strips can be well described by the adiabatic SPH model. The electric field dependence of the activation energy for each strip is obtained by parameter fitting and is displayed in Fig. 3(a). It can be seen that in the 50μm strip, E_A decreases with increase of the electric field of both polarity. The magnitude of modulation by the electric field is further calculated and plotted in Fig. 3(b). It is suggested that the magnitude of modulation under the positive bias is quite similar for all the strips, indicating that the physical origin of the modulation by positive field is identical for the strips with different width. It's known that applying an electric field on the PMN-PT substrate reduces the lattice parameter of the substrate. The compressive in-plane strain-field can be transferred to the film through the interface between PCSMO film and substrate.

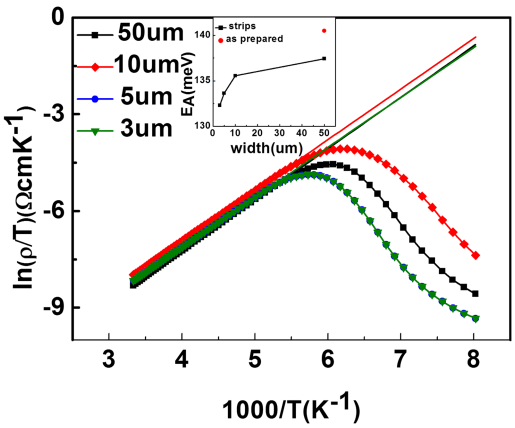

FIG. 2. Variation of $\ln(\rho/T)$ of strips with different width as a function of inverse temperature. Solid lines are the best fit to the SPH model, inset shows the activation energy of the strips and the film from the parameter fitting.

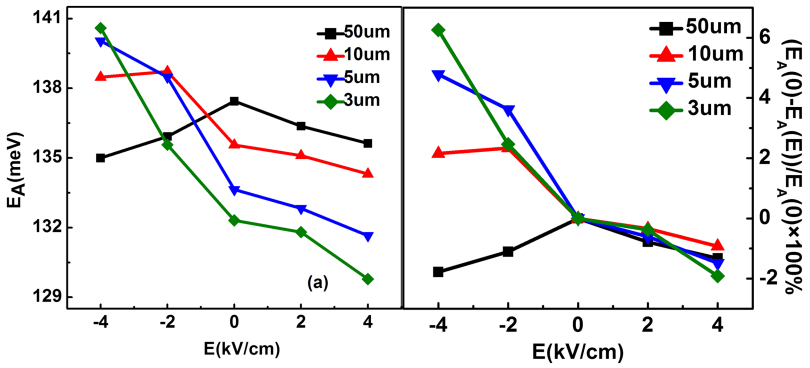

FIG. 3. (a) dependence of the activation energy on the electric field for strips with various width.(b) the electric field modulation of the activation energy.

Consequently, the lattice distortion will be reduced and thus the trapped e_g electron on Mn sites will be delocalized. As a result, E_A decreases as the electric field is applied and increases. Especially, for the 50μm strip, the modulation of E_A by the electric field shows similar decrease trend for both electric bias polarity, indicating that the electric field induced strain effect dominates the evolution of E_A at strip with large size. Moreover, the similar decrease trend and magnitude of E_A by increasing positive bias on strips with small size indicate that the electric field induced strain-effect is still predominant under the positive bias for those strips with small size.

Interestingly, as the width of the strip reduces, the variation of E_A with negative electric field gradually changes from decrease to increase while the varying trend of E_A under positive electric field keeps nearly unchanged (see Fig. 3(a)). Such a gradual change suggests that another mechanism gradually overwhelm the electric field induced strain effect under the negative electric field as the width of the strip reduces. Aside from the strain-induced effect, the polarization effect is another factor that could cause the modulation of the transport properties of the manganite films. As the electric field is applied to the device, electrons or holes will be injected in the film, resulting in a modulation in the carrier density. Meanwhile, the proportion of Mn^{3+} and Mn^{4+} is altered as the electrons or holes are injected in the film. For a negative bias field, such polarization effect will lead to an increase in the proportion of Mn^{3+}. Considering the Mn^{3+} Jahn-Teller active, more Mn^{3+} would enhance the lattice distortion and thus the electron-phonon coupling,[23,24] resulting in the electrons to be more localized and the increase of the activation energy. In the present case, the reduction of the strip width would enhance the polarization effect, leading to the increase of E_A with electric field for the negative bias polarity.

To further investigate the origin of the electric field modulation of the activation of small polarons in narrow strips, we calculate the value of small polaron coupling constant γ_p, which serves as an evaluation of the e-ph interaction strength. The values of γ_p are calculated from the relation $\gamma_p = 2E_H/h\omega_0$.[25,26] For a rough estimation, we use $E_H \approx E_A$ in the adiabatic limit and the calculated values are shown in Fig. 4. It can be seen that γ_p for all samples vary between 7.2 and 8.7, suggesting very strong e-ph interaction in the strips. The electric field dependence of γ_p is similar to that of E_A: decreases as the width of the strip reduces, which suggests that the electron-phonon coupling is weakened as the width of the strip reduces. These results demonstrate that the spatial confinement prefers to weaken the electron-phonon coupling, which could be ascribed to the strain relaxation in the spatial confined sample.

In summary, the electric field dependent high-temperature conduction of patterned $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films is illustrated by considering the adiabatic small polaron hopping conduction above MIT temperature. The small-polaronic activation energy, E_A, obtained from the fitting of temperature dependent resistivity in the high-temperature region, is found to decrease with the reduction of patterned-strip width. Meanwhile, large e-ph coupling constants (γ_p) is further calculated using SPH model for strips with various widths, demonstrating a similar size dependence. The strain relaxation in the strip is used to explain such behavior. Furthermore, an opposite variation trend of E_A under negative and positive electric field is found in the strips with small size, different from

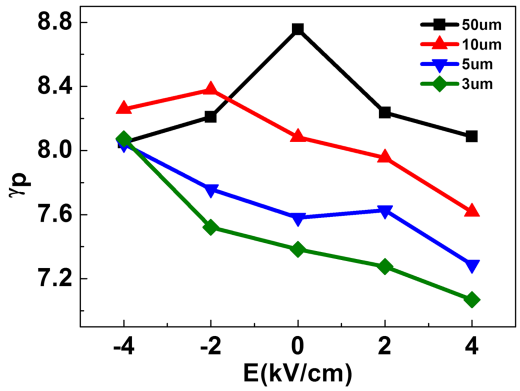

FIG. 4. dependence of the electron-phonon coupling constant γ_p on the electric field for strips with different width.

the behavior for large size. Such change in the evolution of E_A with electric field is attributed to the enhancement of polarization effect with strip width reduction. These results suggest that the spatial confinement prefers to reduce the electron-phonon coupling and the strain effect on the e-ph coupling in the phase separated PCSMO film.

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