Giant Discrete Steps in Metal-Insulator Transition in Perovskite Manganite Wires

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Optical lithography is used to fabricate LPCMO wires starting from a single $(La_{5/8-0.3}Pr_{0.3})Ca_{3/8}MnO_3$ (LPCMO) film epitaxially grown on a $LaAlO_3(100)$ substrate. As the width of the wires is decreased, the resistivity of the LPCMO wires exhibits giant and ultrasharp steps upon varying temperature and magnetic field in the vicinity of the metal-insulator transition. The origin of the ultrasharp transitions is attributed to the effect of spatial confinement on the percolative transport in manganites.

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The strong spin-charge-lattice interaction in transition metal oxides (TMOs) often leads to a striking phenomenon called electronic phase separation (PS), which is identified with the coexistence of a range of exotic electronic and magnetic phases despite the single crystalline structure [1]. The role of the PS in the related physical properties such as high-T_c superconducting and colossal magnetoresistance (CMR) is a hotly debated issue in the field of condensed matter physics. Spatial confinement is a very useful route to gain deeper insight into the nature of PS. In particular, when the spatial dimension of the TMOs is reduced to the characteristic PS length scale, one would expect that changes in the transport properties of the TMOs could be quite dramatic.

A model system for this study is $La_{5/8-x}Pr_xCa_{3/8}MnO_3$ (LPCMO), a perovskite manganite that is famously known for its large-scale PS. Transmission electron microscopy has revealed submicrometer-scaled ferromagnetic (FM) and charge ordered (CO) domains [2]. The coexistence of FM/CO phases in LPCMO reflects the competition between the intrinsic properties of its two starting materials, i.e., FM $La_{5/8}Ca_{3/8}MnO_3$ ($T_C \sim 275$ K) and CO antiferromagnetic (AFM) $Pr_{5/8}Ca_{3/8}MnO_3$ ($T_{CO} \sim 220$ K). Considering the fact that micrometer-scale charge separation would cost too much Coulomb energy, the driving mechanism causing such large-scale PS has attracted great attention. There have been several proposals for the origin of this exotic behavior, related to quenched disorder [3], intragranular strain [4,5], and long-range elastic strain [6]. In any event, it is generally agreed that the metal-insulator transition (MIT) in the LPCMO system occurs via percolation of FM phases, which gives rise to the colossal magnetoresistance.

In this Letter, we report spectacular changes of resistivity featuring giant and ultrasharp steps upon changing temperature or magnetic field in micron-scaled wires of LPCMO ($x = 0.3$) manganites. The steplike features have not been reported in either bulk [7] or thin films [8] of LPCMO in which the percolative MIT smears out the discontinuity of the resistivity change that is generally associated with the first-order phase transition. Remarkably, these steplike changes already show up in LPCMO wires that are 1.6 μm wide, indicating that these features are unlikely to be associated with any effect of quantum confinement. Based on transport measurements, a simple model is proposed, suggesting that the 1.6 μm LPCMO wires have reached the size limit where only a single conducting channel can be established in the percolative network. Consequently, any local event of FM percolation will lead to a sharp change of the global resistivity of the wires.

For consistency, a series of LPCMO wires with different width were fabricated from a single epitaxially grown LPCMO film using optical lithography. The parent LPCMO thin film (thickness ~70 nm) was grown on a (100)-oriented $LaAlO_3$ (LAO) substrate using laser (248 nm, 1 J/cm^2 fluence) molecular beam epitaxy in an ultrahigh vacuum chamber (base pressure $<1 \times 10^{-10}$ Torr). The detailed growth procedure has been described elsewhere [9]. The quality of the epitaxy was examined by atomic force microscope (AFM), x-ray diffraction, and by Rutherford Backscattering (RBS) or channeling experiment in a 2×1.7 MV Tandem accelerator using 2.1 MeV alpha particles. Figure 1(a) shows the (RBS) or channeling results for both the LPCMO film and the LAO substrate. The ratio between the RBS signal collected from a well-aligned incident direction [001] and from a random direction (5° away from that axis), χ_{min}, is determined to be 2.78% for the LPCMO film. Considering even the best Si single crystal has a χ_{min} value of about 2%, the ultralow χ_{min} value for the LPCMO films indicates an excellent epitaxy and high crystalline quality. AFM images (not shown here) also confirm that the surfaces of the LPCMO films are atomically flat with large terraces (~50 nm).

Wires of different sizes have been fabricated from a single LPCMO film using optical lithography within an area of about 3 mm $\times$ 3 mm, as shown by scanning electron microscope (SEM) images in Fig. 1(b). In every unit (~0.5 mm $\times$ 2.5 mm), four gold pads (gold thickness is 200 nm) have been coated onto the LPCMO film with two

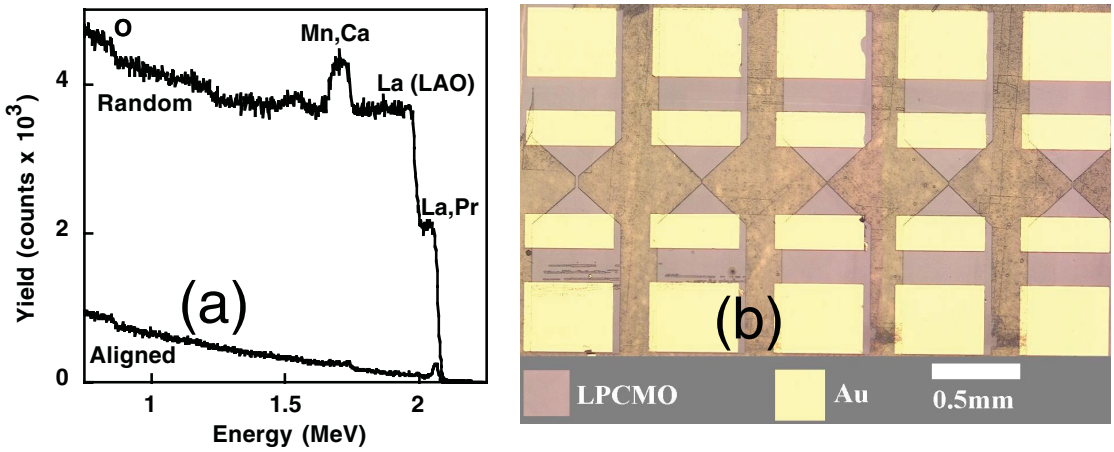

FIG. 1 (color). (a) Rutherford Backscattering or channeling spectra show excellent crystallinity of an epitaxillay grown LPCMO film on a LAO(100) substrate. The minimum yield $\chi_{\min}$ for the LPCMO film is determined to be 2.78%. 2.1 MeV α particles were used as probe; the subatomic (0.1 Å) displacement or vibrations can be detected along the aligned (001) direction, (b) SEM images of LPCMO wires fabricated from a single LPCMO/LAO(100) film with different sizes. The gold contacts are in 200 μm to 500 μm scale, which is considerably larger than the 20 μm diameter of the wire bonding thread. Current flows from top to bottom during the transport measurements.

500 μm $\times$ 400 μm pads serving as current contacts and two 500 μm $\times$ 200 μm pads as voltage contacts. The electrical connections between the Au pads and the puck of a physical property measurement system were made by Kulicke and Soffa wire bonders (model 4123 for Al and 4124 for Au). Twenty microns diameter Au or Al wires were directly engaged without using any paint and solder, which removes all possible contact problems for the transport measurements. Magnetic field was applied along the easy magnetization axis, i.e., the direction perpendicular to the substrate surface during the transport measurements.

The resistivity versus temperature (R-T) curves of the LPCMO wires begin to exhibit sharp steplike features as the width is decreased. Figure 2(a) shows R-T curves for wires of 20 μm, 5 μm, and 1.6 μm measured under a magnetic field of 3.75 Tesla. All R-T curves show hysteresis behavior under cooling-warming cycles, a characteristic feature of the coexistence of FM and CO phases. The R-T curve of the 20 μm LPCMO wire indicated a smooth MIT (blue). For the 5 μm wire (green), the R-T curve starts to show small kinks, although the overall metal-insulator transition remains rather smooth. The kinklike features are dramatically enhanced in the case of 1.6 μm (red curve) LPCMO wire. In particular, after reaching the maximum at about 105 K, the resistivity drops to a very low value in two sharp steps in the cooling curve. The sharpness of each step is beyond the measuring step, i.e., 200 mK. In the warming curve, the resistivity again shows a steplike increase, reaching the maximum in one giant step.

The steplike features have also been observed in the 1.6 μm wire when the resistivity is measured against magnetic field at fixed temperatures in the vicinity of the MIT. Figure 2(b) shows resistivity versus magnetic field (R-H) curves for the 20 μm, 5 μm, and 1.6 μm wires measured at 110 K with magnetic field ramping-up and ramping-down. While the 20 μm and 5 μm wires exhibit rather smooth R-H curves, steplike jumps with sharpness beyond that of the measurement step size (250 Oe) are evident in the 1.6 μm wire [10]. Similar field-induced resistivity jumps were observed in a range of temperatures around and below MIT. At temperatures above T_c ($\sim$150 K) the R-H curves become smooth and no field-induced sharp steps can be observed in any of the LPCMO wires (not shown here).

What is the mechanism(s) for the observed ultrasharp steps in the MIT? First it is important to point out that sudden jumps of magnetization or resistivity have been reported in several other TMO systems including polycrystalline manganites [11,12], bulk single crystal $Sr_4Ru_3O_{10}$ [13], and $Nd_{0.5}Sr_{0.5}MnO_3$ [14]. The origins, depending upon the system, include Barkhausen effect [12], metamagnetic transition via electronic phase separation process with magnetic domain formation [13], and field-driven first-order phase transition at very low temperatures [14]. A sharp drop of resistivity has also been predicted by theoretical work based on the formation of bound polaron pairs and the collapse of charge carrier density at the critical temperature [15].

The ultrasharp steps in the MIT in the present study differ distinctly from the aforementioned cases in that the steps clearly originated from spatial confinement effect on transport properties. By fabricating all wires from a single LPCMO thin film, the wire width becomes the only parameter that can be responsible for the emerging steps. The fact that sharp steps start to appear in the 1.6 μm wire suggests that the stepped transitions are likely the consequences of the reduction of conducting channels down to a single pathway, i.e., spatial confinement. A simple model is schematically shown in Fig. 3. In a two-dimensional (thin

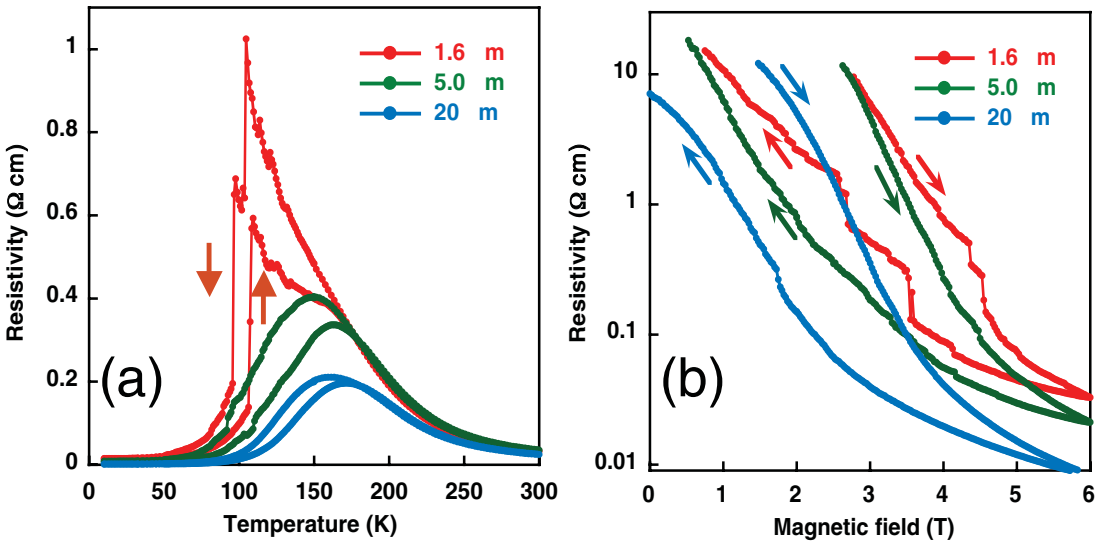

FIG. 2 (color). (a) Resistivity vs temperature (R-T) curves for the LPCMO wires under a 3.75 T magnetic field. Arrows indicate the direction of the temperature ramp. The R-T curves all exhibit hysteresis behavior in cooling-warming cycles, which is consistent with the coexistence of FM and CO domains in the LPCMO system. The MIT is rather smooth for both the 20 μm and the 5 μm wires. Ultrasharp and giant steps are clearly visible for the 1.6 μm wire; (b) resistivity vs magnetic field curves for the LPCMO wires measured at 110 K. Sudden steplike jumps are again visible in the 1.6 μm wire. Arrows indicate the sweeping directions of the magnetic field for each curve.

film) system below T_c, the electrical transport from α to β can be easily established through multiple channels within the percolative network of the FM phase (dark regions). The MIT, although still with first-order nature, is globally smooth due to the multiple conducting channels. However, if the system is cut into a wire geometry (black rectangular box) with a width that is comparable to the length scale of the FM and CO domains, the electrical transport from A to B is left with a single pathway that is interrupted by CO domains (indicated by the arrows). Both temperature and magnetic field can drive the CO to FM transition resulting in the completion of the conducting pathway. With such a single transport pathway, each local CO to FM transition will lead to a sudden change of resistivity, as observed in our experiments. In the LPCMO system, due to the large-scale PS, the single transport pathway is reached already in the 1.6 μm wire. In other TMO systems with smaller PS, one should still observe these sudden jumps of resistivity, albeit in wires with correspondingly smaller size.

Because of the randomness of the formation process of FM and CO domains, the conducting pathway should be different whenever the system goes through a new temperature cycle. This implies that it is unlikely that the resistivity jumps will occur at the same critical temperature or field when repeating the measurements. Indeed, Fig. 4(a) shows the R-T curves of the 1.6 μm wire measured repeatedly in three successive temperature cycles under a 3.75 T field. Although sudden jumps can be observed in all three runs, their positions and magnitude are rather irregular, confirming the random formation process of FM and CO domains upon cooling. This result also rules out the possibility that the jumps are associated with any large-scale structural defects in the LPCMO wire, which should produce reproducible jumps for each temperature cycle.

Finally, it is observed that the sharp jumps of resistivity in the R-T curves of the 1.6 pun wire disappear at higher magnetic fields, as shown in Fig. 4(b). With increasing fields, the MIT shifts to higher temperature, and no jumps can be observed at 6 T and above. A likely cause is that the high magnetic field prevents the formation of large-scale FM/CO domains in the LPCMO wire, resulting in the

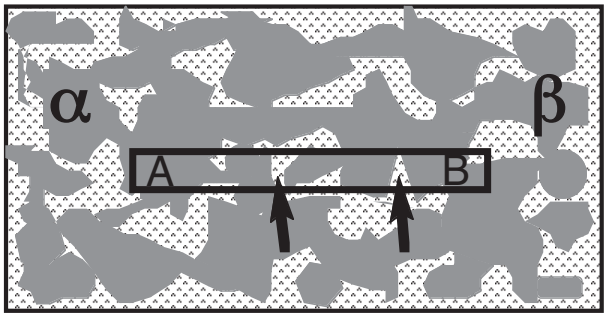

FIG. 3. Schematic picture showing the mechanism for the observed sudden jumps in the R-T and R-H curves of the 1.6 μm LPCMO wire. The dark and dotted regions represent FM and CO domains, respectively. When the width of the LPCMO wire (rectangular box) is comparable to the FM and CO domain size, the electrical transport within the wire may be conducted in a single channel that is interrupted by the CO barriers (indicated by arrows). Both temperature and magnetic field can drive a local CO to FM transition, which causes a sudden change of the resistivity.

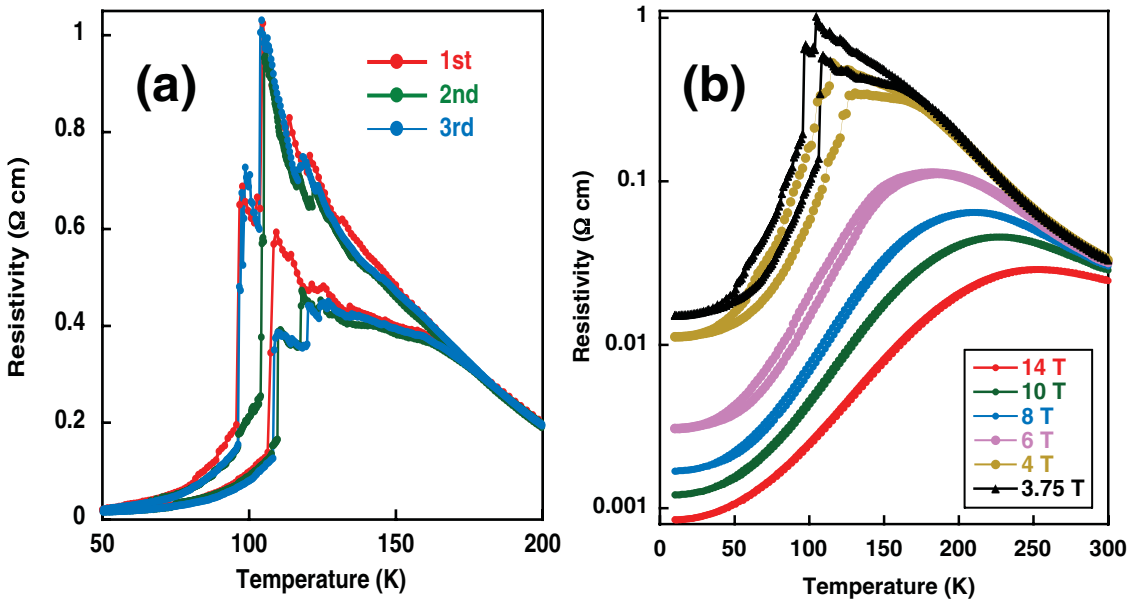

FIG. 4 (color). (a) *R-T* curves of the 1.6 μm wire measured repeatedly in three temperature cycles under the same magnetic field (3.75 T). While sharp jumps appear in all three cases, their location and magnitude are clearly random. (b) *R-T* curves of the 1.6 μm wire measured at different magnetic fields. The sudden jumps disappear at 6 T and higher fields. Note the resistivity in (a) and (b) is shown in linear and log scale, respectively.

disappearance of the sharp jumps. This is supported by the fact that the thermal hysteresis of the *R-T* curves disappear at higher fields. This speculation needs to be verified by charge and/or magnetic imaging under higher magnetic fields.

In summary, we have observed ultrasharp jumps of resistivity upon changing temperature or magnetic field in the vicinity of the MIT in a LPCMO wire. The ultrasharp jumps are clearly associated with the spatial confinement, which is comparable to the length scale of the PS in the system. The ultrasharp jumps imply that a small change of temperature or magnetic field can cause a large change of resistivity, which may have significant impact on the potential applications of manganite materials. With properly selected magnetic field bias or temperature, the favorite CMR materials may see practical applications for the first time with the sampling magnetic field variation capability in the same scale of the current information recording devices.

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