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Large electric-field effects on the resistance of $La_{0.67}Ca_{0.33}MnO_3$ microstructures

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We investigate electric-field effects in thin film microbridges of $La_{0.67}Ca_{0.33}MnO_3$ with the focus on the regime of metal-insulator transition. A mechanically milled $SrTiO_3$ substrate is used as a backgate dielectric. Inside the metal-insulator transition we find a strong unipolar field-induced reduction in resistance, as well as a suppression of the nonlinear features in the *I-V* curves we observed earlier. We associate the observed effects with a phase separated state in which metallic regions coexist with short range correlated polaron regions. When the glassy polaron phase has fully developed and closes off the microbridge, the field effects disappear leaving the strongly nonlinear behavior of the transport current unaltered.

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I. INTRODUCTION

Doped manganites such as $La_{0.7}Ca_{0.3}MnO_3$ (LCMO), obtained by hole doping the antiferromagnetic insulating parent compound $LaMnO_3$, show a large variety in physical properties.[1,2] The Ca-doping introduces mixed Mn valence, with both Mn^{3+} present, which is Jahn-Teller (JT) active, and Mn^{4+}, which is not. This leads to competing interactions, trapping of electrons in JT distortions (polarons), and itinerancy of the electrons in the double exchange (DE) mechanism[3] when spins become polarized. Depending on the doping, this results in a metal-to-insulator transition at a characteristic temperature T_{MI}, with the formation of a small *d* band at low temperatures and a polaron liquid at high temperatures.

In view of the way the physical properties of these materials depend on doping, it is not surprising that the effects of applying electric fields have received a lot of attention. There is a promise that such systems, based on Mott insulators,[4,5] may be effective building blocks for field effect devices, such as transistors, because of the abundant amount of potential carriers (*d* electrons) and the ability to control the band gap. Numerous experiments on electric-field effects in various perovskite heterostructures have therefore been reported. For example a 3 nm $La_{0.8}Sr_{0.2}MnO_3$ film (screening length less than 1 nm[6]) combined with a ferroelectric material ($PbZr_{0.2}Ti_{0.8}O_3$, PZT) as the gate dielectric, shows *E*-field induced modulation of T_{MI} and the magnetoresistance. These effects[6,7] are *bipolar* (i.e., the sign of the resistance change is opposite for opposite signs of the applied gate voltage) and attributed to a straightforward modulation of the charge carrier density in the manganite. In a different investigation, large in-plane resistance variations (∼76%, bipolar) were observed in 50 nm thick LCMO films[8] with the PZT-gate geometry, and smaller effects for devices deposited on a $SrTiO_3$ (STO) film as the backgate dielectric. Since the thickness of the film is much larger than the *E*-field screening length, the effects were attributed to the presence of a phase separated state, the more so since there was clear asymmetry for the two signs of the gate voltage. For phase separation, fully *unipolar* field effects were also observed, for example. in $La_{0.8}Ca_{0.2}MnO_3$ thin films on mechanically thinned STO substrates.[9]

What has as yet received little attention is the detailed response of the electron system to an applied *E*-field when going through the metal-insulator transition, even though the physics there is of particular interest. Neutron scattering experiments performed on LCMO single crystals[10] show that a static polaron glass state consisting of nanometer-sized pockets of correlated polarons forms just above T_{MI}. In macroscopic transport measurements on thin films no special behavior is seen, but we demonstrated before that in microbridges structured in ultrathin and fully strained LCMO thin films, strongly nonlinear current *I* - voltage *V* characteristics could be found in the MI transition.[11] We attributed these nonlinearities to the formation of correlated polaron regions, aggravated by the strain, and now large enough to encompass the entire width of the micron-sized structure.

Here we investigate the effects of an electric field on the electrical conductance in the MI transition and on the nonlinear *I-V* characteristics in particular, of similar strained LCMO microbridges using the STO substrate as a backgate dielectric. We find strong *unipolar* field effects in the onset of the transition, which we associate with the occurrence of a phase separated state. In this state, metallic regions coexist with short-range correlated polaron regions.[10] As the system is warmed through the MI transition, the field effects disappear when the more or less homogeneous correlated polaron (glass) phase is fully developed.

II. EXPERIMENTAL

The LCMO films were grown by dc sputtering in an oxygen atmosphere of 3 mbar and at a growth temperature of 840 °C. The films are patterned into microbridges[12] using electron-beam lithography and Ar etching [see Fig. 1(a)]. The STO substrate was subsequently mechanically milled down to 100 μm and used as a gate dielectric. Measurements before and after the milling show that the bridges remain undamaged during this process. The geometry of the measured devices is shown in Fig. 1(b). The gate voltage V_g is applied between the back of the STO substrate (through a silver paint contact) and one of the voltage contacts of the microbridge. We measured *I-V* curves as function of temperature and in high magnetic fields using a Physical Properties Measurement System (Quantum Design) for temperature and magnetic field control (T = 20–300 K; H_a = 0–9 T). We found the leak currents through the gate to be negligible compared to the currents used for the *I-V* measurements.

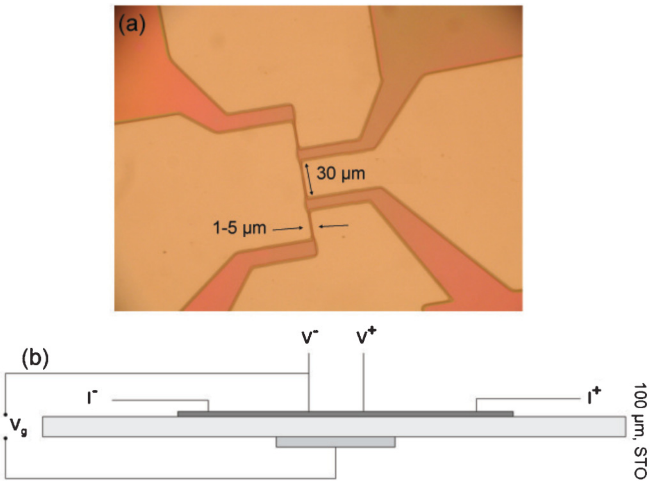

FIG. 1. (Color online) (a) The microbridge patterned into the LCMO thin film. Dimensions: width is 5 μm and the distance between the voltage contacts is 30 μm. (b) FET device geometry.

III. RESULTS

Typical resistance (R) vs temperature (T) behavior for microbridges patterned in 10 nm thick LCMO films grown on STO substrates is shown in Fig. 2(a), with $T_{\rm MI}$ = 120 K and typical magnetoresistance behavior at the transition upon application of a high magnetic field.

In the paramagnetic state $R(T)$ is expected to be activated. Indeed, Fig. 2(b) shows the high temperature resistance behavior in a $\log_{10}$ R vs T^{-1} plot. A linear fit to the data provides an activation energy E_A = 112 meV for the small polaron hopping process, which is the accepted value in this temperature regime.[13] The resistance values in Figs. 2(a) and 2(b) are determined form the I-V curves measured at every temperature. Two of those curves taken at T = 200 K (open triangles) and at T = 110 K (closed squares) are presented in Fig. 2(c). In all our microbridges we observe strongly nonlinear behavior in the steep part of the transition[11] and linear curves at low and high temperatures. We previously associated this behavior with the formation of an intervening glassy polaron phase as the microbridge is warmed through the transition.[10,11]

In the following we present the effect of an applied electric field on the $R(T)$ behavior and on the nonlinearities (i.e., formation of the glassy phase). The resistance behavior of the same 5 μm bridge as in Fig. 2, now on the mechanically milled STO substrate, in zero field and upon application of ±75 V (i.e., 7.5 × 10^5 V/m) is shown in Fig. 3. In zero field we find the same R vs T behavior as before the milling, with the same $T_{\rm MI}$ = 120 K. The value of R at low T (30 kΩ) is a bit higher than before [10 kΩ, see Fig. 2(a)]. In zero field we find typical R vs T behavior for strained LCMO films.[15] Furthermore, we observe a strong E-field effect quite sharply peaked around T = 90 K ($T_{\rm MI}$ = 120 K). The resistance is reduced by a factor of 5 upon application of V_g = 75 V, but quite asymmetric, with only a small effect for gate voltages of the opposite sign. Still, the effect is *unipolar*, the resistance decreases irrespective of the sign of the gate voltage. It is important to note that the position of $T_{\rm MI}$ remains unchanged when a gate voltage is applied and that the main effect is sharply peaking in the regime of the MI transition and not beyond (i.e., quite different from the observations by Wu *et al.*[8]). The width of the E-field effect is about 10 K. Moreover, we also observe a reduction in resistance at low temperatures with both signs of the gate voltage resulting in similar resistance changes. Although the change is smaller than in the transition, it is still of the order of 50%, indicating the presence of inhomogeneities even at low temperatures, and resulting in increased metallicity upon application of an E-field.

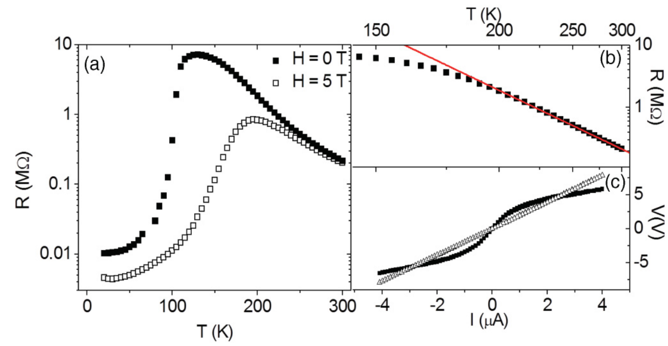

FIG. 2. (Color online) (a) $R(T)$ behavior of a 5 μm × 10 nm LCMO bridge determined from I-V curves at an applied current of I = 0.1 μA at H = 0 T (closed symbols) and H = 5 T (open symbols) for the film on the unmilled STO substrate. (b) $\text{Log}_{10} R$ vs T plot in the temperature range 150–300 K for the same sample. Note: the scale on the T axis (top) is reciprocal in order to show the $1/T$ behavior. The solid (red) line indicates the fit to extract the activation energy (E_A = 112 meV) of the small polaron hopping process. (c) I-V characteristics at T = 110 K (closed squares) and at 200 K (open triangles).

We check the reproducibility of the effect by repeating the same experiment but with application of 100 V. In Fig. 4 we compare the field-induced resistance changes for measurements at V_g = 75 V [Fig. 4(a)] and at V_g = 100 V [Fig. 4(b)]. The 100 V measurement shows very similar behavior around the transition albeit with slightly reduced magnitude of the field-induced resistance drop. A point to

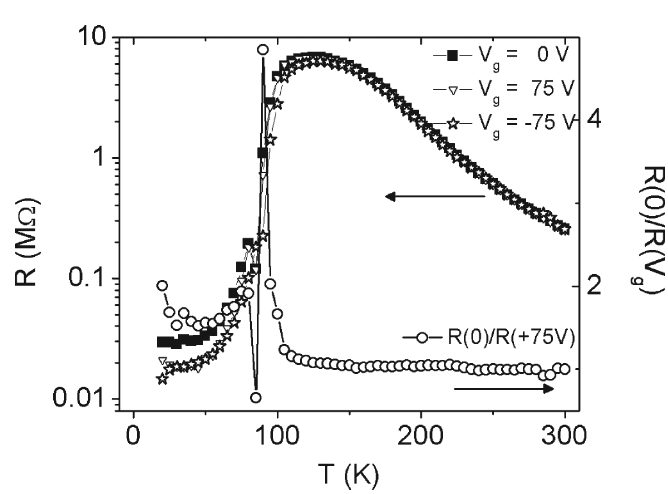

FIG. 3. $R(T)$ behavior (left axis) of a 5 μm × 10 nm LCMO bridge (same sample as Fig. 2) on the mechanically milled STO, measured at I = 0.1 μA for three applied gate voltages: V_g = 0, ±75 V. On the right axis the ratio between $R(0)$ and $R(+75\text{V})$ is shown.

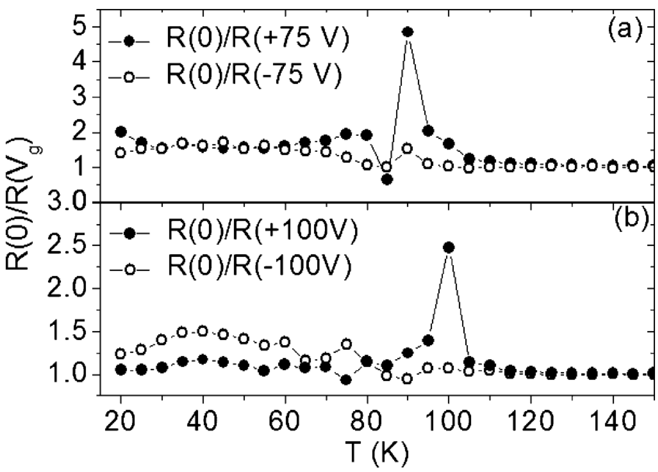

FIG. 4. (a) Ratio between $R(0)$ and $R(V_g)$ (closed circles: $V_g = +75$ V; open circles: $V_g = -75$ V) determined at $I = 0.1$ μA. (b) Ratio between $R(0)$ and $R(V_g)$ (closed circles: $V_g = +100$ V; open circles: $V_g = -100$ V) determined at zero bias.

note is that the maximum in the E-field effect shifts to slightly higher temperature going from 75 to 100 V.

Next we turn to the I-V curves. In accordance with previous findings[11] they are linear for most temperatures but show strong nonlinear behavior in the steep part of the transition. Here we investigate the influence of the applied electric field on these nonlinearities. The left panels of Fig. 5 show the I-V curves of the 5 μm bridge for temperatures $T = 85$, 95, 100, and 110 K for $V_g = 0$ and $V_g = +100$ V; the panels on the right show the corresponding (numerical) derivatives. Around 95 K the nonlinearities which we associated with the formation of a homogeneous glassy polaron phase start to appear, with a full width of the peak in dV/dI of 0.3 μA. The full width increases with temperature to 1 μA at 100 K. From the data it becomes clear that at these temperatures the nonlinearity is suppressed upon application of an E-field. At $T = 110$ K, where the nonlinearity has developed strongly (peak width: 4 μA), the strength and shape of the peak remains fully unaltered when an E-field is applied.

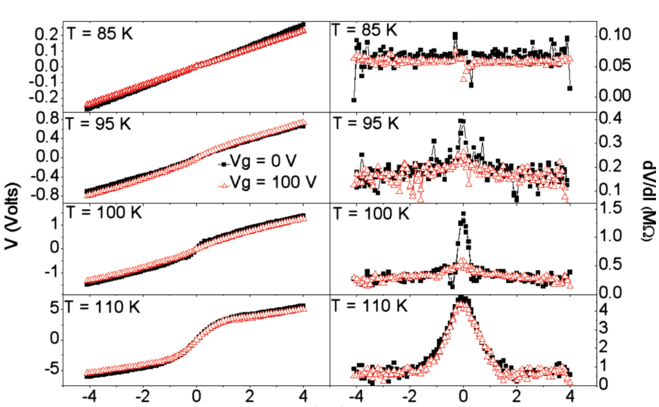

FIG. 5. (Color online) (Left) I-V curves for 5 μm bridge at $T = 85$, 95, 100 and 110 K. (Right) corresponding numerical derivatives. $V_g = 0$ V (closed black squares) and $V_g = +100$ V (open red triangles).

IV. DISCUSSION

Similar to our earlier report,[11] in these microbridges we find strongly nonlinear behavior in the steep part of the MI transition. Here we show that for a small temperature range these nonlinearities can be suppressed by an applied E field. However, one concern with respect to the current observations might be that they are connected to the cubic-to-tetragonal phase transition in STO around 105 K. A small effect was actually reported to exist in thin films of LCMO on STO, not in the resistivity, but in the temperature coefficient (TC) $1/\rho(d\rho/dT)$ (with ρ the specific resistance), which showed a variation of 0.5% in a 9 nm film.[14] In that film T_{MI} was at 160 K, and the TC variation was observed in the flat metallic part of the resistance. Our thin films have lower T_{MI}, which we have argued is the effect of homogeneous strain,[15] and a 1% variation is not visible in the strong decrease in ρ below the MI transition. We could detect a variation in the TC in the microbridge of a 20 nm thick film where $T_p \approx 160$ K. As shown in Fig. 6 this film even showed a variation in $R(T)$, which is probably due to the larger film homogeneity in the small structure. The resistance peak in this case is exactly at the phase transition temperature of 105 K, and the variation in $R(T)$ has disappeared around 90 K, where we find the large E-field induced effects discussed above. Moreover, there is hardly any change in the dielectric constant of STO at the phase transition.[16] It is also important to note that the position of the transition temperature remains unchanged as the field is applied indicating that strain relaxation in the film due to electrostriction in the substrate is not an issue in our microbridges. Hence, our E-field effects appear to be intrinsic features of the fully strained LCMO microstructures.

We explain the observations in light of our previous report on the formation of a polaron glass phase in LCMO microbridges as it is warmed through the MI transition.[11] We argued before that the nonlinear I-V characteristics signal the formation of correlated polaron regions as were found to exist in single LCMO crystals.[10] Here we find that these (strong) nonlinearities are suppressed in a *unipolar* fashion

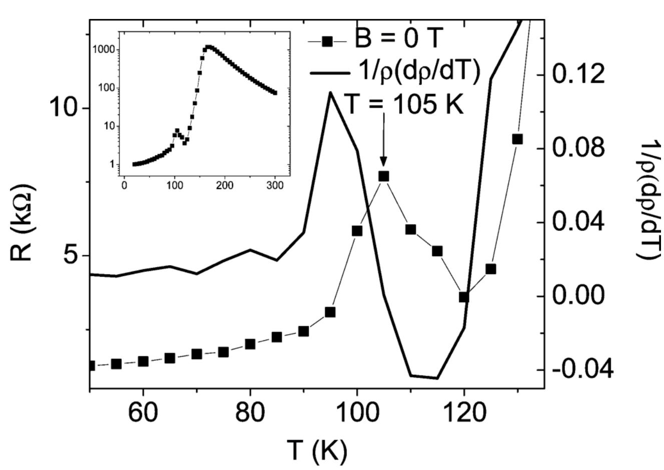

FIG. 6. $R(T)$ behavior of a 20 nm thick LCMO microbridge ($w = 5$ μm) in the range $T = 50$–135 K. Right axis: $1/\rho(d\rho/dT)$ (solid line). The arrow indicates the transition in the STO. Inset: full $R(T)$ curve.

through application of an E-field. The *unipolar* nature of the observed effects indicate that they are to be attributed to a state in which metallic and insulating regions coexist, as argued before by Eblen-Zayas *et al.*,[9] rather than due to a variation in carrier density. The insulating regions are formed by short range correlated polarons, as precursor to the polaronic state at high temperature. The applied field changes the relative volume fraction of the coexisting phases by accumulating charge at the interfaces between them, which can result in the dielectric breakdown of the inhomogeneities. Important to note is that the maximum in the effect occurs in the onset of the transition when the nonlinearities just start to appear in the I-V curves. When the nonlinear effect has fully developed, the electric-field effect disappears. Apparently, when the glassy polaron phase becomes homogeneous and closes off the bridge, the E-field becomes ineffective and cannot break it down anymore. We note that the effect in the transition is *unipolar* but asymmetric. It is possible that doping still plays a role and that asymmetry in hole and electron modulation of inhomogeneities in the microbridge leads to the observed asymmetric behavior.

We also observe E-field effects at low temperatures, indicating that this regime is also not homogeneous. Moreover, there is some hysteresis in the low T effects, which indicates and that the collapse and rebuilding of the insulating regions is not well controlled.[17] Still, the microbridge does relax back to its initial state since remeasuring the gate effect leads to similar if somewhat smaller effects in the transition. Furthermore, we observed a small increase in the temperature at which the effect peaks when gate voltage was increased. However, it is important to stress that even for the increased gate voltage the position of the MI transition remains unchanged. Therefore, we believe that the presence of the E-field alters the volume fractions of the metallic and glassy phases, which may hamper the formation of the more homogeneous polaron glass phase necessary to close the bridge for percolation. This in turn would explain the increased temperature at which the effect peaks. Hence, this temperature shift also strongly supports the conclusion that the observed E-field effects are not caused by field-induced changes of the substrate, since $T = 90$ K is well below the phase transition of STO.

V. CONCLUSIONS

In conclusion, going to micron-sized structures reveals a strong response of $La_{0.7}Ca_{0.3}MnO_3$ thin films to an applied electric field. The observed effects are clearly tied to the percolating behavior of the conductance in the bridge, which takes place on the scale of the width of the bridge.

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