Current-induced conductance switching in epitaxial

$[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]$ multilayers

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Abstract

We report on the non-linear in-plane electrical transport in coherently grown $[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]$ multilayers with ultrathin ($< 3$ nm) single layers. Current-induced switching of the conductance, with low conductance at larger currents, is demonstrated. The conductance switching is modified under a magnetic field, resulting in an extremely large magnetoresistance of negative, or in a special case even positive, sign. Our results suggest a percolative nature of transport where a large local current density gives rise to a spin-polarizing and a thermal effect of current.

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The narrow conduction band of metallic manganites $R_{1-x}A_xMnO_3$ ($R = $ La or rare earth metal, $A =$ di- or monovalent metal, $x \sim 0.3$) shows a very high spin polarization at the Fermi energy. Therefore, pronounced phenomena of spin-polarized transport can be observed, including a very large tunneling magnetoresistance in ferromagnet-insulator-ferromagnet junctions. The double exchange (DE) model successfully explains the associated appearance of metallicity and ferromagnetism in this class of materials although it is not sufficient to elucidate the complexity of phases because any coupling of charge or spin to the lattice (e.g. the strong Jahn-Teller effect) is neglected. Low doping $x$ or lattice distortions lead to competition of DE mediated by itinerant electrons with antiferromagnetic superexchange interactions of localized Mn 3d states. These compounds tend to be antiferromagnetic or spin-canted insulators, but some of them have been proven to show phase separation into metallic ferromagnetic and insulating clusters. Additional to charge localization, some insulating manganites show the characteristic feature of charge ordering, in particular for a commensurate fraction of charge carriers (e.g., $x = 0.5$).

Another scenario of competing double exchange and superexchange is found at surfaces of ferromagnetic manganites. In the first two or three layers of perovskite cells (of $\sim 3.9$ Å height), reduced lattice symmetry at the surface suppresses the DE. This probably causes the semiconducting nature of ultrathin ($\leq 2$ nm) manganite films, even if epitaxial strain is very low (0.1%). Thin films can be positioned in an epitaxial multilayer, where their lattice structure and magnetization can be evaluated more precisely.

Recent work on charge-ordered $Pr_{1-x}Ca_xMnO_3$ ($x = 0.3$ to 0.5) crystals and charge-ordered thin films of several $x = 0.5$ compounds has demonstrated the induction of metallic conductivity by application of an electric field or by creation of carriers via irradiation with light or x-rays. A metallic path develops through the charge-ordered background, in some cases showing switching to high conductance at a threshold voltage. On the other hand, a spin current might well have an effect on the magnetic order of localized Mn spins (for theoretical description of this interaction see e.g. Ref.15). Indeed, there are experiments which have been interpreted in this way. They are characterized by a current...
of high density between ferromagnetic regions of manganite (grains, or particles in a tunnel junction).

In this work we report on the non-linear in-plane electrical transport of coherently grown \([\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]\) multilayers with ultrathin manganite layers. The results indicate a phase separation into metallic and insulating clusters and percolative transport in the ultrathin \(\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3\) films. Near the critical thickness of the manganite layers below which metallic conduction is lost, a current-induced switching of the conductance is observed. Additional application of a magnetic field leads to an extremely large, discontinuous, negative or even positive magnetoresistance in a certain range of the parameters temperature and current. Systematic dependence of the transport behavior on the thickness of manganite layers has been found. The observations are discussed within the framework of a spin-polarizing and a thermal effect of the current.

The \([\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]_{20}\) multilayers used in the experiments have been grown by off-axis pulsed laser deposition as described previously\(^\text{18}\), using a substrate temperature of 825 °C and a deposition rate of 1 Å s\(^{-1}\). The lattice mismatch of the insulating \(\text{SrTiO}_3\) is +0.9 %. From the numerous satellite peaks in standard x-ray diffractograms the superlattice period of the samples of \(4.0 \text{ nm} \leq \Lambda \leq 6.35 \text{ nm}\) has been determined. Thickness of manganite layers has been varied between 1.6 nm (about 4 perovskite cells) and 2.9 nm (about 7 cells). Images of the sample cross sections obtained from high resolution transmission electron microscopy (Fig. 1) prove a coherent film growth. Some local thickness variations by one unit cell are present, as well as a certain curvature of the layers, that increases from substrate to surface of the multilayers. Resistance and magnetoresistance have been measured in current-in-plane (CIP) geometry, using four in-line contacts made of silver paint, with the magnetic field oriented along the current direction. Current-voltage (I-V) curves were registered at several temperatures and magnetic fields using a current source. Magnetization has been determined in a SQUID magnetometer, including its dependence on a dc current flowing between the two sample ends.

In contrast to the well-known metallic behavior of thicker epitaxial \(\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3\) films,
the macroscopically observed in-plane resistivity \( \rho_{\text{eff}} \) of the present multilayers shows a complicated dependence on the measuring current \( I \) (examples shown in Fig. 2). With decreasing thickness \( d \) of the manganite layers, the overall character of \( \rho_{\text{eff}}(T) \) changes, as expected, from metallic (Fig. 2a) to semiconducting (Fig. 2b). The non-linear behavior can be characterized as follows: For low currents, the resistance decreases with increasing current. Thus, the enhancement of the conductance is a consequence of current flow. However, for currents above a threshold value, the resistance increases. This is also obvious in the I-V characteristics (example in Fig. 3), where a hysteretic switching towards higher voltage is seen at a switching current \( I_0 \), after a gradual increase of the differential conductance \( dI / dV \) below \( I_0 \). Striking jumps have also been observed in \( \rho_{\text{eff}}(T) \) curves (Fig. 2). The jump temperature, \( T_0 \), always shifts with increasing current in a way that the high-resistance part of the \( \rho_{\text{eff}}(T) \) curve expands. (For instance, in Fig. 2a, \( T_0 \) moves down with increased current, while in Fig. 2b it shifts upwards.)

The jumps are well reproducible if the sample is kept well below the ferromagnetic ordering temperature, \( T_C \). As an example, Fig. 2b shows the same \( \rho_{\text{eff}}(T) \) jump measured for the two directions of current, with +I during cooling and -I during warming. After heating above \( T_C (< 300 \text{ K}) \) or change of contact positions at room temperature, slight shifts of \( I_0 \) (in the range of 20 \%) and \( T_0 \) (by few K) have been registered.

The Curie temperature of the samples reduces with decreasing \( d \), from \( T_C = 260 \text{ K} \) for \( d = 2.9 \text{ nm} \) to 165 K for 1.9 nm, and the saturation magnetization (44 to 38 emu g\(^{-1}\)) is much lower than expected for the Mn spins being collinear (91 emu g\(^{-1}\)). In order to check the magnetic response to current, the magnetization \( M \) has been measured under applied current. Within an accuracy of better than 0.2 \%, there was no systematic change of the magnetization in dependence on the applied current (not presented in a figure). In particular, no anomalies of \( M \) are associated with the conductance jumps. This result rules out a magnetic coupling phenomenon of adjacent manganite layers, as well as a phase transition, since both would be reflected in a change of magnetization. Thus, the striking variations of conductance do not significantly affect the magnetic order of the sample volume.
Instead, assuming a magnetic phase separation for the ultrathin manganite layers, as it is also suggested for the interface region by recent tunneling experiments\(^3\), the electrical transport seems to be dominated by metallic paths within the manganite layers. These paths will be interrupted by nm-size high-resistance “barriers” if the thickness of the manganite layers is slightly below the value where percolation appears. For the current-induced increase of conductance, an aligning of Mn spins within the barriers, mediated by a spin-polarized current of high density seems likely. Also, magnetic coupling of ferromagnetic clusters via the spin current\(^{15,20}\) might be involved. Current-dependent switching of a manganite cluster surrounded by ferromagnetic manganite layers has been described in that way recently\(^4\).

The microscopic origin of the observed polarizing effect of current needs further clarification.

In the following we suggest a mechanism explaining the effect of larger currents to reduce the conductance. Assuming a dominating role of small barriers (with reduced magnetic order resulting in increased resistivity) within the conduction path, the thermal energy produced by the current at these barriers might be substantial. The temperature profile within the manganite layers depends on the distribution of both, the dissipated energy density and the thermal conductivity; furthermore, it also depends on the substrate temperature. When either heat dissipation becomes large enough or thermal conductivity becomes low enough that the local temperature approaches the (local) \(T_C\), the resistivity of the considered barrier strongly increases. As an example, for a local resistivity of 10 \(\Omega\text{cm}\) within a barrier of \((2.5 \text{ nm})^3\) in extension and a thermal conductivity of 1 \(\text{W K}^{-1} \text{m}^{-1}\) for the manganite\(^{21}\), a current of 500 nA would produce a temperature difference of \(\sim\)40 K to the environment. Therefore, the thermal effect is unlikely in single films: The thermal resistance from the barrier to the sample surface (typically being at He atmosphere during measurement) would be much smaller. Furthermore, the thermal effect of current shows an inherent tendency to produce switching of the conductance, being well-known as “thermal switching”\(^{22}\). The strong rise of the resistance of metallic manganites when \(T_C\) is approached from below\(^1\) might well result in a process where a cluster switches from the low-resistance metallic state to the high-resistance semiconducting state due to Joule microheating. The existence of
threshold values for the current or the sample temperature can be expected. The magnitude of the resulting resistance jump will depend on how much the concerned switching barrier contributes to the resistance. The mechanism of thermal switching provides a consistent explanation of the discontinuous changes of conductance in our data. In particular, the $\rho_{eff}$ jump observed at 55 K in Fig. 2b can be ascribed to the reduced thermal conductivity at low temperatures. It is important to note that, in contrast to the current-induced formation of a metallic path, the thermal effect is always characterized by $d\rho_{eff}/dI > 0$.

Now we turn to the results of transport in an external magnetic field $H$. In the whole temperature range, from 5 K to 250 K, a large magnetoresistance (MR) in a field of 5 T is observed, as can be expected from the reduced magnetic order in the manganite layers. Concerning the field dependence of the transition temperatures, an interesting observation is made: While the metal-insulator transition (if any) shows the usual shift towards higher temperatures in a magnetic field, the temperature $T_0$ of the conductance jump decreases. Consequently, a large positive MR is found in a range below $T_0$ for metallic-like samples (Fig. 2a). The MR for this peculiar case will be described later. In the high-resistance state, the observed negative MR shows a hysteretic switching (Fig. 3a) in a certain range of current. Most measured resistance loops $R(H)$ are reproducible for repeated field cycling, e.g. the loop in Fig. 3a, where the resistance reproducibly switches by a factor of 8 in a field of 20 mT. Moreover, switching fields $H_S$ increase with current ($dH_S / dI > 0$, compare also I-V curves under magnetic field, Fig. 3). This agrees with the above discussed general rule that larger currents stabilize the high-resistance state. The latter property is also found for the striking positive MR obtained just below $T_0$ for metallic-like samples (Fig.4b). The hysteretic switching towards large resistance in high magnetic field has been observed for several samples at appropriate current. As expected, it is characterized by $dH_S / dI < 0$, in contrast to the switching with negative MR. The strong sensitivity to current is worth noticing: $H_S$ varies by more than 1 T at a 10 % change of I. The function of $H_S(I)$ is roughly linear for both types of switching, with positive or negative MR.

The discussion of the striking field effect on the non-linear transport remains hypothetical...
at present. It is known that the magnetic field enhances the magnetic order of the Mn core spins everywhere throughout the sample volume. Thereby, an insulator-to-metal transition can be induced, without any additional effect of current. (In this case, transition fields should be insensitive to current.) Also for the non-linear transport, the changes of size and distribution of the metallic phase brought about by the field will be essential. Probably, the local current density $j$ at the conductance path will be reduced due to the increased magnetic order. Thereby, a thermal effect being proportional to $j^2$ would be more suppressed than a polarizing one. This might contribute to the stabilization of the low-resistance state in a magnetic field (case of Fig.4a). However, the origin of the positive MR and the downshift of $T_0$ in the field must be different. It can be speculated that the electrical transport in a multilayer in a large magnetic field resembles that of another multilayer with somewhat thicker manganite layers in zero field, since the effect of the external field adds to the ferromagnetic double exchange. This very simple idea is supported by experimental results: Both, increase of $d$ and application of a magnetic field typically lead to a drop of $T_0$ and to larger values of $I_0$. However, the complex interplay of current and field effects on the conduction process certainly needs further investigation.

In conclusion, we have observed non-linear in-plane electrical transport in coherently grown $[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]$ multilayers with ultrathin single layers. Current-induced switching of the conductance, with low conductance at larger currents, has been demonstrated. Furthermore, large discontinuous changes of the conductance also appear in dependence on temperature and magnetic field. Our results indicate a magnetic phase separation within the ultrathin manganite layers and a percolative nature of transport. Non-linear transport is related to a large local current density. The latter is suggested to give rise to a spin-polarizing and a thermal effect, which favor a metallic and a high-resistance state, respectively.

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REFERENCES

1. J. M. D. Coey, M. Viret, and S. von Molnár, Adv. in Phys. 48, 167 (1999).
2. J.-H. Park, E. Vescovo, H.-J. Kim, C. Kwon, R. Ramesh, and T. Venkatesan, Nature 392, 794 (1998).
3. Moon-Ho Jo, N. D. Mathur, N. K. Todd, and M. G. Blamire, Phys. Rev. B 61, R14905 (2000).
4. M. Uehara, S. Mori, C. H. Chen, and S.-W. Cheong, Nature 399, 560 (1999).
5. M. Fäth, S. Freisem, A. A. Menovsky, Y. Tomioka, J. Aarts, and J. A. Mydosh, Science 285, 1540 (1999).
6. B. Raquet, A. Anane, S. Wirth, P. Xiong, and S. von Molnár, Phys. Rev. Lett. 84, 4485 (2000).
7. K. Dörr, J. M. de Teresa, K.-H. Müller, D. Eckert, T. Walter, E. Vlakhov, K. Nenkov, and L. Schultz, J. Phys. Cond. Mat. 12, 7099 (2000).
8. M. J. Calderón, L. Brey, and F. Guinea, Phys. Rev. B 60, 6698 (1999).
9. J. Z. Sun, D. W. Abraham, R. A. Rao, and C. B. Eom, Appl. Phys. Lett. 74, 3017 (1999).
10. T. Walter, K. Dörr, K.-H. Müller, D. Eckert, K. Nenkov, M. Hecker, M. Lehmann, and L. Schultz, J. Magn. Magn. Mater. (to be published)
11. V. Kiryukhin, D. Casa, J. P. Hill, B. Keimer, A. Vigliante, Y. Tomioka, and Y. Tokura, Nature 386, 813 (1997).
12. M. Fiebig, K. Miyano, T. Satoh, Y. Tomioka, and Y. Tokura, Phys. Rev. B 60, 7944 (1999).
13. H. Oshima, K. Miyano, Y. Konishi, M. Kawasaki, and Y. Tokura, Appl. Phys. Lett. 75, 1473 (1999)
14 S. Srivastava, N. K. Pandey, P. Padhan, and R. C. Budhani, Phys. Rev. B 62, 13 868 (2000).

15 C. Heide, R. J. Elliott, and N. S. Wingreen, Phys. Rev. B 59, 4287 (1999).

16 J. Z. Sun, J. Magn. Magn. Mater. 202, 157 (1999).

17 W. Westerburg, F. Martin, S. Friedrich, M. Maier, G. Jakob, J. Appl. Phys. 86, 2173 (1999).

18 T. Walter, K. Dörr, K.-H. Müller, B. Holzapfel, D. Eckert, M. Wolf, D. Schläfer, L. Schultz, and R. Grötzschel, Appl. Phys. Lett. 74, 2218 (1999).

19 Measurements repeated some months later showed enhanced conductance of the samples, probably due to a long-term diffusion process.

20 J. C. Slonczewski, Phys. Rev. B 39, 6995 (1989).

21 J. Hejtmánek, Z. Jirák, M. Marysenko, C. Martin, A. Maignan, M. Hervieu, and B. Raveau, Phys. Rev. B 60, 14057 (1999).

22 T. Burch, P. P. Craig, C. Hedrick, T. A. Kitchens, J. I. Budnick, J. A. Cannon, M. Lipsicas, and D. Mattis, Phys. Rev. Lett. 23, 1444 (1969).

23 J. M. De Teresa, M. R. Ibarra, C. Marquina, P. A. Algarabel, and S. Oseroff, Phys. Rev. B 54, R12689 (1996).
FIG. 1. HRTEM image of a section near the surface of a $[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3(M)/\text{SrTiO}_3(S)]_{20}$ multilayer with single layers of 2.9 nm / 3.2 nm thickness. The curvature of the layers decreases towards the bottom of the multilayer.

FIG. 2. Effective in-plane resistivity $\rho_{eff}$ vs temperature T of $[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]_{20}$ multilayers measured during warming at different dc currents I. Layer thicknesses in nm are (a) 2.3/2.7 and (b) 1.9/2.3. Lines represent straight connections of measuring points. $T_0$ is the (current-dependent) temperature of the conductance jump. At low currents, $\rho_{eff}$ shows time-dependent fluctuations. Inset of (b): Voltage response for the two opposite directions of I.

FIG. 3. In-plane current (I) - voltage (V) characteristics of a $[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3(1.6\text{ nm})/\text{SrTiO}_3(1.6\text{nm})]_{6}$ multilayer, measured at several longitudinal magnetic fields. Such characteristics are typical for samples and temperatures where negative magnetoresistance of switching type is found.

FIG. 4. In-plane resistance vs. magnetic field for two $[\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{SrTiO}_3]_{20}$ samples with layer thicknesses in nm of (a) 1.9/2.3 and (b) 2.9/3.2. After zero field cooling, the field sweep was $\mu_0H = 0 \to 5 \text{T (1.) } \to 0 \text{ (2.) } \to -5 \text{ T (3.) } \to 0 \text{ (4.)}$. (a) Negative magnetoresistance (MR) in the high-resistance state. (b) Positive MR observed at certain temperatures in the low-resistance state (see text), for different measuring currents. The 12 $\mu\text{A}$ curve has been shifted by +50 $\Omega$ for clarity.
Fig. 1
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Fig. 2a
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Fig. 2b
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Fig.3:  
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$d_{\text{LSMO}} = 1.9 \text{ nm}$

$I = 0.1 \mu\text{A}$

$T = 60 \text{ K}$
Fig. 4b
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\[ T = 5.5 \text{ K} \]

\[ 12 \, \mu\text{A} \]

\[ 11 \, \mu\text{A} \]

\[ 10 \, \mu\text{A} \]

\[ 1 \, \mu\text{A} \]

\[ R (\Omega) \]

\[ \mu_0 H (T) \]