Transport anisotropy in biaxially strained La$_{2/3}$Ca$_{1/3}$MnO$_3$ thin films

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Due to the complex interplay of magnetic, structural, electronic, and orbital degrees of freedom, biaxial strain is known to play an essential role in the doped manganites. For coherently strained La$_{2/3}$Ca$_{1/3}$MnO$_3$ thin films grown on SrTiO$_3$ substrates, we measured the magnetotransport properties both parallel and perpendicular to the substrate and found an anomaly of the electrical transport properties. Whereas metallic behavior is found within the plane of biaxial strain, for transport perpendicular to this plane an insulating behavior and non-linear current-voltage characteristics (IVCs) are observed. The most natural explanation of this anisotropy is a strain induced transition from an orbitally disordered ferromagnetic state to an orbitally ordered state associated with antiferromagnetic stacking of ferromagnetic manganese oxide planes.

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It is well known that the physics of the doped perovskite manganites is determined by a complex interplay of structural, magnetic, electronic, and orbital degrees of freedom. While the classical double exchange model can qualitatively explain the transition from a paramagnetic insulating to a ferromagnetic metallic state [1], for a more complete understanding of the physics of the manganites electron-lattice coupling has to be included [2]. Recently, Millis et al. have pointed out that uniform compression, as realized by hydrostatic pressure, increases the electron hopping amplitude favoring a ferromagnetic metallic state [3]. In contrast, biaxial strain, as realized in epitaxial thin films grown on substrates with significant lattice mismatch, enhances the Jahn-Teller distortions favoring an insulating state due to the tendency of the electrons to become localized [3]. Fang et al. have calculated the phase diagram of the almost tetragonal doped manganite La$_{1-x}$Sr$_x$MnO$_3$ as a function of biaxial strain by studying the instabilities of the ferromagnetic state [3]. Their results are in agreement with experiments on biaxially strained La$_{1-x}$Sr$_x$MnO$_3$ thin films [3]. In their work, it has been shown that the orbitally disordered ferromagnetic state (F) is unstable against orbital ordered states with layer (A) and chain (C) type antiferromagnetism. In turn, it is expected that these different magnetic states are associated with different magnetotransport behavior via the double exchange mechanism. The further investigation of the validity of the strain phase diagram, the corresponding magnetotransport properties, and the extendability to doped manganites with strong tilt of MnO$_6$ octahedra [3] and phenomena as charge ordering as for example La$_{1-x}$Ca$_x$MnO$_3$ is of great interest to gain more insight into the physics of these materials and its dependence on lattice distortions. For the purpose of this study, it is important to verify the coherency of the strained state of the doped manganite, in order to be able to determine properly the intrinsic properties within the biaxial strain phase diagram, and to exclude effects of nonuniform strain distribution or relaxation effects [3-8].

In this Letter we present a careful study of the structural, electronic, and magnetic properties of coherently strained La$_{2/3}$Ca$_{1/3}$MnO$_3$ (LCMO) thin films and LCMO-La$_{2/3}$Ba$_{1/3}$MnO$_3$ (LBMO) heterostructures on SrTiO$_3$ substrates. In addition to previous studies, transport properties have been measured both parallel and perpendicular to the plane of biaxial strain. The key result of our study is that biaxial strain results in highly anisotropic transport properties: Whereas insulating behavior and non-linear IVCs are observed perpendicular to the biaxially strained plane (parallel to the $c$ axis), the in-plane transport is metallic below the Curie temperature $T_C$. The saturation magnetization of biaxially strained LCMO is strongly reduced compared to the bulk material or the less strained LBMO films. It is shown that this behavior is not due to interface effects between different layers [3], but is an intrinsic property of the biaxially strained LCMO arising most likely from a strain induced orbital ordering. Another important result is that in strained LCMO a low-resistance state can be induced by applying either a magnetic field or a high current density. A similar behavior is for example observed in Pr$_{0.7}$Ca$_{0.3}$MnO$_3$ and Nd$_{0.5}$Sr$_{0.5}$MnO$_3$ and has been attributed to a magnetic field or current induced (local) melting of a charge resp. orbitally ordered ground state [3-6].

We have grown LCMO films and LBMO-LCMO-LBMO heterostructures on SrTiO$_3$ ($a \approx 3.905$ Å) substrates using pulsed laser deposition [3-4]. The lattice mismatch between the SrTiO$_3$ substrate and LCMO ($a_{\text{bulk}} \approx 3.864$ Å in pseudocubic notation) is -1.2% resulting in in-plane tensile strain, whereas the mismatch...
between SrTiO$_3$ and LBMO ($a_{\text{bulk}} \approx 3.910$ Å) is only about 0.13% resulting in very small compressive strain. Note that in the LBMO-LCMO-LBMO heterostructures the LBMO layers provide low resistance contacts to the (ultra)thin LCMO films allowing for a homogeneous current feed to the transport perpendicular to the film. Furthermore, effects of a possible surface “dead layer” [13] are avoided in the multilayer structure. Layer-by-layer growth of the films was confirmed by a high pressure reflection high energy electron diffraction (RHEED) system showing clear growth oscillations [14, 15].

As has already been stressed it is important to prove the coherency of the strained state. The coherent film thickness was determined from Laue oscillations in $\theta - 2\theta$ x-ray scans. It was found that both LCMO and LBMO films grow coherently strained on SrTiO$_3$ substrates up to a thickness of at least 60 nm, consistent with literature [14, 18]. The out-of-plane ($c$ axis) and in-plane ($a$ axis) lattice parameters of the LCMO thin films grown on SrTiO$_3$ have been determined from the (002) and (103) reflections. The in-plane lattice parameters coincide throughout the whole layer with the substrate lattice parameters. That is, the in-plane lattice parameter of LCMO is enlarged, while the out-of-plane lattice constant is reduced, leading to a tetragonal lattice distortion with $c/a \approx 0.985$. The tetragonal distortion can be viewed as a Jahn-Teller like distortion resulting in an increased Jahn-Teller splitting of the Mn $e_g$ levels and, in turn, in a tendency of the electrons to become localized.

In order to measure the electrical transport properties perpendicular to the film plane, mesa structures (see inset of Fig. 2) were patterned using optical lithography and Ar ion beam etching. The mesas have typical area of several $\mu$m$^2$. Fig. 1 shows the resistance vs temperature, $R(T)$, curves for LBMO-LCMO-LBMO mesa structures with different thickness $d_{\text{LCMO}}$ of the LCMO layer. For comparison, a sample without LCMO layer ($d_{\text{LCMO}} = 0$) is shown. It is evident that the resistance increases with increasing $d_{\text{LCMO}}$. We estimate the involved resistivities $\rho_c$ to about several $\Omega$m at 10 K. Note that due to the non-linear IVCs, it is difficult to obtain a meaningful $\rho_c$ or thickness dependence $\rho_c(d)$. In the inset we show the sheet resistance as a function of $d_{\text{LCMO}}$. The strong increase of resistance with increasing layer thickness can partially be due to tunneling through the barrier. The important point is that the resistivity does not show any tendency to saturation meaning the resistance is intrinsic to the barrier and not to the interface. Furthermore, the metallic $R(T)$ behavior below $T_C$ that is observed for the sample with $d_{\text{LCMO}} = 0$ turns into a semiconducting or insulating $R(T)$ behavior with increasing $d_{\text{LCMO}}$. The low-temperature plateau (below 60 K) of the resistivity corresponds to a plateau in the temperature dependence of the magnetization (see upper panel of Fig. 3) as expected for double exchange materials. This thickness dependence clearly shows that the insulating $R(T)$ behavior is not due to the patterning process, due to a contact resistance between the gold contact layer and the manganite film, nor due to interface effects. One can conclude that the insulating behavior for transport along the $c$ axis observed at low temperatures is an intrinsic property of the coherently strained LCMO thin films. We note that in the work of Bibes et al. interfaces between magnetic LCMO and non-magnetic SrTiO$_3$ are shown to favor phase segregation [8]. Inhomogeneous transport properties were found in La$_{0.7}$Ca$_{0.3}$MnO$_3$/SrTiO$_3$ heterostructures and interpreted as arising from magnetic interface disorder [19]. In our case, the interfaces are between two differently doped manganites. While it remains to be investigated whether interdiffusion is present at such interfaces, the effects observed here are related to the magnetic order of the whole thin film layer.

In Fig. 2 we show the IVCs of several mesa structures with different values of $d_{\text{LCMO}}$ measured in $c$ axis direction, i.e. with current perpendicular to the biaxially strained plane. While for low enough current densities $V \propto I$, all mesas show highly non-linear IVCs for higher current densities following a $V \propto I^n$ dependence with $n \approx 0.2 - 0.3$. In contrast, the IVCs measured with the current in-plane are ohmic. For comparison, mesa structures have been patterned into single LBMO films ($d_{\text{LCMO}} = 0$) that are well lattice matched to the SrTiO$_3$ substrate. For these samples, ohmic IVCs have been obtained both for the current applied in- and out-of-plane. From this and the fact that the measured voltage clearly increases with increasing $d_{\text{LCMO}}$ (see discussion before), preparation or interface effects can be excluded as the origin of the nonlinear IVCs. Hence, we can conclude that for current perpendicular to the plane of biaxial strain...
the insulating behavior of the coherently strained LCMO films is associated with non-linear current transport. We note that the non-linearity becomes smaller with increasing $T$ and vanishes at the $T_C$ of the LCMO thin films ranging between 100 and 150 K. $T_C$ was determined from magnetization measurements of the bi/trilayer films before mesa patterning. This strongly suggests that the electronic anisotropy is coupled to the magnetic properties of the LCMO films.

In agreement with previous experiments we found a strain dependence of the saturation magnetization $M_S$ of the LCMO films. Zandbergen et al. reported $M_S \approx 2.5 \mu_B/{\text{Mn}}$ atom at 5 K for a 6 nm thick, coherently strained LCMO film on SrTiO$_3$. Consistently, we have measured a value of $M_S \approx 2.2 \mu_B/{\text{Mn}}$ atom at 5 K for a 57.5 nm thick LCMO film (see inset of Fig. 3). In contrast, almost strain free LBMO films on SrTiO$_3$ show the expected saturation magnetization of about $3.67 \mu_B/{\text{Mn}}$ atom. These findings are in agreement with the phase diagram predicted by Fang et al. where for tensile strain an instability to an A-type antiferromagnetic state is predicted. Another interesting observation is that the size of the hysteresis loop in $M(H)$ curves also depends on strain. While for almost strain free LBMO films on SrTiO$_3$ a coercive field of $\mu_0 H_c \approx 10$ mT is observed at 5 K, a much larger value of $\mu_0 H_c \approx 70$ mT is measured for the strained LCMO films.

Fig. 3 gives an overview on the field, current and temperature dependence of the resistance and magnetization in strained LCMO films by showing $R(T)$ curves recorded at different applied fields and currents. Applying high magnetic fields results in a strong suppression of the resistance at all $T$. Also, due to the non-linearity of the $IVCs$ the measured resistance is reduced when the applied current is increased below about 150 K. We note that for $T \lesssim 150$ K the resistance is dominated by the LCMO layer, only around 250 K it is dominated by the LBMO layer. Fig. 3 clearly shows that with increasing field, the non-linearity becomes weaker and also the onset temperature of the non-linear behavior is shifted from about 100 K at 0 T to 50 K at 8 T. For comparison, the inset of Fig. 3 shows the in-plane $\rho(T)$ curves of a LCMO film (4 T) measured for current applied in-plane. In the upper picture the magnetization $m$ is shown.

To further study the effect of the applied current, we have measured IVCs for successive current sweeps with increasing amplitude as shown in Fig. 4. After zero field cooling, the current is first increased to $10 \mu A$ (curve a) corresponding to a current density of about 100 A/cm$^2$.
On decreasing the current again (curve b), a lower voltage is measured at the same current values, i.e. the applied current of 10 μA has switched the sample to a state with lower resistance. Applying a high magnetic field (8 T) has the same effect as applying a high current (1 mA). After applying a field of 8 T, the measured IVC are stable and the resistance is independent on the applied current. Thus, both a high magnetic field and a high current density induce a state with reduced resistivity.

We now address the possibility of a phase separated state in fully strained LCMO. We note that in the case of inhomogeneously strained or relaxed films (e.g. due to island growth), it is plausible to assume a phase separated state with ferromagnetically and antiferromagnetically ordered clusters, as has been discussed recently for LCMO thin films grown on LaAlO$_3$. A similar phenomenon has been observed for bulk Pr$_{10.7}$Ca$_{0.3}$MnO$_3$ samples as well as for LCMO bulk and thin film samples. However, phase separation cannot explain the transport anisotropy present in our samples, but would be expected to lead to direction independent behavior. All our experimental observation can be naturally described by the assumption of strain induced orbital order as predicted by Fang et al.

Figure 4: IVCs of a LBMO-LCMO-LBMO mesa structure ($d_{LCMO} = 6.0$ nm) measured for current perpendicular to plane at 5 K and zero field. The different curves have been obtained in successive current sweeps with increasing amplitude. The mesa area was 9 μm$^2$. 

In summary, we have investigated coherently strained LCMO films. The biaxial strain was found to induce anisotropic transport properties at low temperatures with metallic and insulating behavior for current in- and out-of-plane. It has been shown that this behavior is not due to inhomogeneous interface effects or phase separation. We suggest strain induced orbital ordering as the origin of the observed behavior in agreement with theoretical predictions. We also have shown that by applying a high current density, a low-resistance state can be induced. This effect may be of interest with respect to magnetoelectronic devices.

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See references [1-12] for more details.
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