Biaxial strain and orbital order in $La_{2/3}Ca_{1/3}MnO_3$ thin films

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Abstract

Biaxially strain is an important parameter in the complex phase diagram of the doped manganites that can lead to a different groundstate compared to bulk single crystals at the same doping level. For coherently strained $La_{2/3}Ca_{1/3}MnO_3$ thin films on $SrTiO_3$ substrates, we report an unusual direction dependence of the electrical transport properties. For transport perpendicular to the substrate induced strained plane, an insulating behavior associated with non-linear current-voltage characteristics is observed. We propose an A-type antiferromagnetic groundstate for the biaxially strained films as is consistent with the observed strongly reduced saturation magnetization.

Key words: doped manganites; biaxial strain; orbital order; $La_{2/3}Ca_{1/3}MnO_3$

It is well known that the physics of the hole 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 state 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 on single crystals, increases the electron hopping amplitude leading to a more cubic metallic state [3]. In contrast, biaxial strain, as realized in lattice mismatched epitaxial thin films, tends to enhance the Jahn-Teller distortion resulting in more insulating behavior [3]. Fang *et al.* [4] have shown by density functional calculation that for the system $La_{1-x}Sr_xMnO_3$ the groundstate magnetism changes as a function of biaxial strain c/a. The groundstate magnetism in turn is associated with different orbitally (dis)ordered states.

Here, a 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 is presented. The details of the thin film fabrication and analysis are published elsewhere [5,6]. The most important result of our study is that biaxial strain can induce an *uniaxial* metal-insulator-transition: Perpendicular to the biaxially strained plane (parallel to the c axis), insulating behavior associated with non-linear electrical transport is observed, while the in-plane transport remains metallic below the Curie temperature T_C. This is not an interface or surface effect between different layers, but is an *intrinsic* property of the strained LCMO thin films [6]. Furthermore, the magnetization of the LCMO thin films is strongly reduced compared to the bulk material or the less strained LBMO thin films.

In Fig. 1 the main result of our study is summarized. We show resistance vs. temperature curves for

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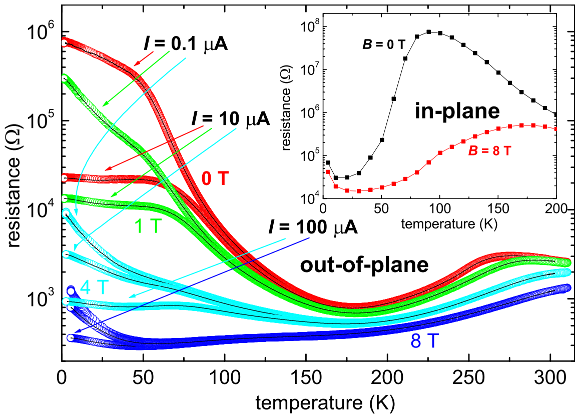

Fig. 1. Resistance vs temperature of LCMO thin films measured out-of-plane and in-plane (inset) for different magnetic fields and currents. The symbols (full lines) represent data measured during cooling (heating).

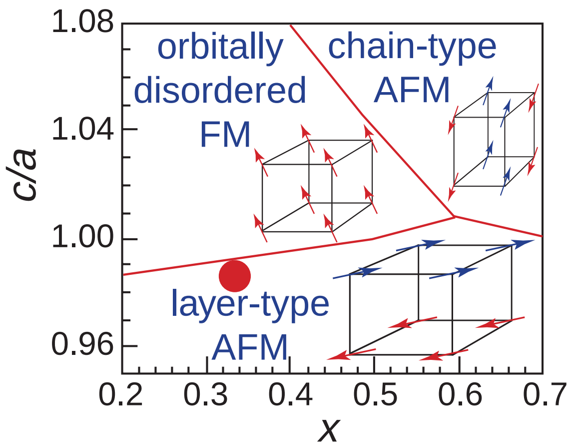

Fig. 2. Proposed phase diagram for $La_{1-x}Ca_xMnO_3$ as a function of the lattice strain c/a following references [4,9].

different directions of the electrical current: (i) in-plane of the thin film, and (ii) out-of-plane within a mesa-type structure [5]. The in-plane resistance for a thin LCMO film is shown in the inset of Fig. 1. Clearly, the transition from the paramagnetic-insulating state to the ferromagnetic-metallic state is observed at around 100 K. It is well known that the reduced transition temperature compared to the bulk material is due to the biaxial strain [7].

The out-of-plane resistance for a trilayer junction with a thickness of the middle LCMO layer of 7.2 nm is shown in the main panel of Fig. 1. The base electrode is formed by a 27 nm thick LBMO thin film, the top part of the mesa structure consists of 12 nm LBMO. The large difference in resistivity between LCMO and LBMO ensures a homogeneous current feed into the mesa structure. In the whole temperature range, no transition to a metallic like behavior of the LCMO layer is observed. Below 100 K the $R(T)$-curves become current dependent i.e. non-linear current-voltage characteristics appear. An external magnetic field reduces the resistivity strongly within the whole temperature range. In summary, a clear *transport anisotropy* is observed at a doping level ($x = 1/3$) corresponding to an isotropic metallic state in bulk material at low temperature.

We believe that the phenomen of phase separation which plays an important role in the doped manganites cannot account for the observed anisotropy [8]. From phase separation in metallic and insulating patches one would expect *isotropic* behavior of the transport properties.

As explanation of the transport anisotropy we propose a phase diagram as shown in Fig. 2 following the diagram of Fang et al. [4,9] for $La_{1-x}Sr_xMnO_3$. Due to the lattice strain in $La_{1-x}Ca_xMnO_3$ the orbital occupation is changed favoring the occupation of in-plane e_g $d_{x^2-y^2}$-orbitals at the Mn sites reducing strongly the overlap in z-direction. While in-plane double-exchange mediated ferromagnetic ordering persists, the individual layers couple antiferromagnetically in z-direction. This groundstate can account for the out-of-plane insulating behavior and the strongly reduced saturation magnetization to about half the value of the unstrained film [6]. We conclude that for $x \approx 1/3$ and $c/a \approx 0.985$ $La_{1-x}Ca_xMnO_3$ is slightly below the border between the ferromagnetically orbitally disordered groundstate and the A-type antiferromagnetic orbitally ordered groundstate.

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