Giant in-plane anisotropy in manganite thin films driven by strain-engineered double exchange interaction and electronic phase separation

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We investigate epitaxial Pr0.65(Ca0.7Sr0.3)0.35MnO3 thin film grown on orthorhombic (110) NdGaO3 substrate which breaks the lattice symmetry and affects the phase separated ground state. As a result of the anisotropic substrate strain, giant in-plane magnetic and magnetotransport anisotropy are observed, which is related to the anisotropic coupling and competition between the double-exchange interaction and the Jahn-Teller distortion. Furthermore, the in-plane anisotropy shows a distinct enhancement near the metal-insulator transition, implying a significant contribution from the phase separation to the anisotropic transport behaviors.

Clearly, the complex physics involving anisotropy in manganate thin films warrants further in-depth investigations.

In this letter, we address this issue by studying in details the structure, magnetism, and magnetotransport behaviors of Pr0.65(Ca0.7Sr0.3)0.35MnO3 (PCSMO) thin film epitaxially grown on (110) NdGaO3 (NGO) substrate. PCSMO is a typical PS manganite with coexisting and competing charge-ordered (CO) and FM phases. Our results reveal giant IP magnetic and transport anisotropy in the film, which can be associated with the anisotropic coupling and competition between the DE interaction and the JT distortion. Importantly, the observation of strain effects which are amplified by the electronic phase separation puts constraints on the potential theoretical models that can be used to elucidate the physics of competing energetics in complex oxides.

PCSMO thin films with a thickness of 80 nm were epitaxially deposited on (110) NGO substrates using pulsed laser deposition (PLD) at 750 °C in 150 mTorr oxygen. A KrF excimer laser (λ = 248 nm) was used with an energy density of ~1.3 J/cm² and a repetition rate of 3 Hz. The topographic imaging measurement was carried out on an atomic force microscope (AFM) (Asylum Research, MFP-3D). The crystalline structure of the film was examined using a high-resolution x-ray diffraction (HRXRD) system. Magnetic properties as a function of temperature (T) and magnetic field (H) were measured using a Quantum design MPMS. The magnetotransport behaviors were measured using a standard four-probe method in a quantum design physical property measurement system (PPMS).

Fig. 1(a) presents the HRXRD pattern of PCSMO/NGO thin film. The pure c-axis orientation indicates the good crystallinity and epitaxy of the film. The pseudocubic PCSMO has lattice parameters a = 3.851 Å, b = 3.840 Å, and c = 3.848 Å, while NGO substrate has an orthorhombic crystalline structure with lattice parameters a = 5.43 Å.
FC curve shows a sudden increase at 100 Oe. As shown in Fig. 2(b), along the [100] direction, the field-cooled (FC) conditions with an applied field of which were measured under both the zero-field-cooled (ZFC) and the [100] direction, which can be taken as the easy axis. This clear IP uniaxial magnetic anisotropy can be observed with physical properties of the PCSMO thin film, detailed magnetic anisotropy, i.e., $T_{\text{MI}}$ is higher and resistivity is smaller in the [100] direction compared with those in the [010] direction, indicating that the hopping of the $e_g$ electrons is much easier along [100]. The difference of $T_{\text{MI}}$ under zero magnetic field between the two IP directions is about 3 K.

In order to understand the role of PS in the transport anisotropy in strained manganite films, we used magnetic fields to tune PS. As shown in Fig. 3(a), the zero field $\rho(T)$ curve exhibits a distinct hysteresis, which is often identified in manganites with PS. The thermal hysteresis in the $\rho(T)$ curves is reduced as the applied field increases, and the values of $T_{\text{MI}}$ during cooling and warming are very close to each other as $H > 1.6$ T, indicating the weakening of the quenched-disorder-induced macroscopic PS. The $H$-dependence of $T_{\text{MI}}$, for both the cooling and warming processes, is plotted in Fig. 3(b). With increasing $H$, $T_{\text{MI}}$ monotonously shifts toward higher $T$, while the difference between the cooling and warming data starts to shrink as $H > 0.5$ T and eventually overlap with each other at $H > 1.6$ T, which suggests a strong suppression of the PS state at high magnetic fields. However, the gap between the values of $T_{\text{MI}}$ measured along the two IP directions does not shrink at all up to $H = 2.5$ T (Fig. 3(c)), which implies that the transport anisotropy is retained even when PS is substantially suppressed. Therefore, other factors besides PS contribute to the observed giant anisotropy in strained manganite films.

The schematic drawing in Fig. 4 illustrates the emergence of uniaxially elongated FM-metallic (FMM) clusters randomly distributed in the CO insulating (COI) matrix (left part of the figure), where the anisotropic strain promotes the growth of FMM phase along the [100] direction. Thus, in the PCSMO film with PS, an anisotropic percolation at the MIT as well as the consequent response to external fields would be expected. On the other hand, to understand the physics underlying these observed anisotropic behaviors, we have to consider the strain mediated orbital properties which have been shown to play important roles in strained manganite films. For the unstrained pseudocubic case with a high crystalline symmetry, equal weights of different orbitals (such as $|3y^2 - r^2>$ and $|3x^2 - r^2>$) and uniform overlaps between electronic clouds in different directions are...
expected, which lead to the isotropic DE interaction and JT distortions. Thus, identical transport and magnetic properties in different crystalline directions are expected. However, in the current case of PCSMO films grown on (110) NGO substrates, the IP anisotropic tensile strain causes different bending conditions of Mn-O-Mn bonds along two IP directions, i.e., compared to the [010] direction, the larger tensile strain along [010] gives rise to a stronger overlap of the electronic clouds (Fig. 4 (right)). Thus, the reduced bending of Mn-O-Mn bond in the [010] direction will promote the Mn-O-Mn bonds along the [100] direction compared to the [010] direction, which gives rise to anisotropic double-exchange and Jahn-Teller distortions.

In the theoretical study by Dong et al., the anisotropic double-exchange and John-Teller distortions were found to be responsible for the anisotropic transport in strained manganites. Our results on PCSMO films on (110) NGO substrates indeed reveal giant in-plane anisotropic magnetism and resistivity, giving support to the theory. Furthermore, it is clear that PS significantly contributes to the strain-induced anisotropy, and the anisotropy of physical properties is “amplified” at the phase boundary. Thus, the long range strain field is effective to not only influence the competition between separated phases, but also couple to the spin and orbital degrees of freedoms in manganite thin films.

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FIG. 3. (Color online) (a) $T$-dependence of resistivity measured along [010] direction under different magnetic field ($H = 0, 0.6, 1.0, 1.6, 5.0, 9.0$ T). The cooling processes are indicated by arrows except for cases of $H = 5.0$ and $9.0$ T, in which $\rho(T)$ curves measured during cooling and warming overlap with each other. (b) $H$-dependence of the metal-insulator transition temperature $T_{MI}$ in the PCSMO film measured along the [010] direction during both cooling and warming. $T_{MI}$ measured in a wider range of $H$ is shown in the inset. (c) $H$-dependence of $T_{MI}$ measured in both in-plane directions during warming.

FIG. 4. (Color online) Schematic illustrating the effect of anisotropic substrate strain on the manganite films with phase separation. Left: the uniaxially elongated FMM phase within the COI matrix contributes to the observed anisotropic transport. Right: the anisotropic strain causes a stronger overlap of the Mn orbitals along the [010] direction compared to the [010] direction, which gives rise to anisotropic double-exchange and Jahn-Teller distortions.