A-site ordering and stripe phases in manganite films


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Abstract

Insulating and metallic stripes above and below the Curie temperature, $T_C$, respectively, were observed by a high-resolution scanning tunneling microscopy (STM) and/or spectroscopy (STS) in A-site ordered and macroscopically strain free epitaxial La$_{0.75}$Ca$_{0.25}$MnO$_3$ film grown on MgO substrate. The “insulating” stripes were found to be incommensurable to the lattice and aligned along (1 1 0) direction. Metallic stripes were commensurable with periodicity $2a_p\sim 0.8$ nm and aligned parallel to the crystallographic $a/b$-axis. Formation of these stripes involves competing charge, orbital, and lattice orders and is an outcome of an overlapping of electron wave functions mediated by the local lattice-strain distribution, existed even in A-site ordered film due to the difference in cation radii of La and Ca.

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In colossal magnetoresistant (CMR) [1] manganites, of the form La$_{1-x}$Ca$_x$MnO$_3$ (0.2 $\leq x \leq$ 0.33), the localizing effect of the charge due to the Coulomb repulsion and the Jahn–Teller (JT) coupling at the Mn-site [2] results in correlated polarons. Accommodation of the JT distortion through a strain relaxation process in the presence [3] or in the absence [4] of a disorder at the A-site influences the transport properties such as metal–insulator transition (MIT) and CMR to a large extent. Consequently, the electronic phase separation and the percolative mechanism of the CMR and MIT [5] are also associated with the structural distortions. In the absence of substrate-induced strain within the La$_{0.75}$Ca$_{0.25}$MnO$_3$ (LCMO) film grown on MgO(1 0 0) we have shown recently La/Ca cation-ordered rhombohedral superstructure that suppresses the electronic inhomogeneity down to 1 nm scale [6]. In this paper, we present low-dimensional stripe phases, similar to the charge–orbital density wave, due to structural modulation coming from uncompensated lattice-strain.

Epitaxial film with a La and Ca ordering was grown by metal-organic aerosol deposition technique [6]. Scanning tunneling microscopy (STM) and spectroscopy (STS) were performed at a base pressure of $\sim (1–5) \times 10^{-10}$ Torr. Mechanically cut Pt–Ir tips were used to get high-resolution images in the constant current mode ($I=0.1–0.3$ nA) with a tip bias ranging from 0.35 to 0.7 V [7]. At room and low temperatures, high-resolution images, within an area of 50 $\times$ 50 Å$^2$, were obtained on flat terraces of $\sim$500–2000 Å length-scales.

High-resolution STM at 61.7 K is visualized in Fig. 1a. One can clearly see the stripe-like features with the orientation perpendicular to one of crystallographic axis with a predominant peak-to-peak modulations of 6.5–8.5 Å in the ferromagnetic metallic state ($T_C\approx T_{MI}=273$ K). The room temperature STM images shown in Fig. 1b also demonstrate stripe-like features, oriented diagonal to the crystallographic axes, with a predominant periodicity of $\sim$5.8 Å. These stripes reveal two different quasi-periodic corrugations: a large (black arrows in Fig. 1b) corrugation with a width (spread) of 6 Å, quite in contrast to the smaller corrugation (orange arrows in Fig. 1b), which is 4 Å wide.

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Criteria for obtaining the atomic resolution by tunneling experiments in manganites were recently proposed based on the reduced screening of the charge due to a defect-induced confinement—a trapped polaron [8]. Polarization of the charge in manganites is shown to be induced by the strain field that determines the nature of overlapping electron wave functions, and in turn the direction of the charge–orbital stripe [9]. This can be rationalized by the change in the value of the term $C_{11} - C_{12} - C_{44}$ (where $C_{ij}$ is the elastic modulii) from less than zero, favoring diagonal charge–orbital modulation, to greater than zero, which favors bond-type charge–orbital modulations. Clearly, the shear ($C_{44}$) and uniaxial ($C_{11}$) deformations contribute to the stripe formation. The presence of such a strain in the LCMO film can be associated with the atomic displacements caused by the ordering of the La–Ca cations. Checkered-board arrangement of uniaxially dilated (La) and compressed (Ca) unit-cells can be deciphered from the cross-sectional high-resolution transmission electron microscopy images (HRTEM) [6]. The unique ordering of uniaxially strained ($e_1$) unit-cells generates shear ($e_2$) and ‘shuffle’ ($s_x$ and $s_y$) deformations shown in Fig. 2 in accordance with Ref. [4]. Although the strain is compensated macroscopically resulting in a rhombohedral structure, it should be uncompensated around defects, and dominate the charge and the orbital modulations. The nanometer length-scale of these stripe features, therefore, is a natural outcome of a microscopically uncompensated strain.

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References