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Effects of lattice modulation on magnetic properties of epitaxial ferromagnetic/paramagnetic manganite superlattices

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Abstract

Strained artificial superlattices of $(La_{0.7}Ba_{0.3})MnO_3/LaNiO_3$ (LBMO/LNO) were formed on SrTiO₃ (001) substrates by radio frequency magnetron sputtering. The result of (00*L*) crystal truncation rod intensity profiles reveals an enhancement of tetragonality of LBMO lattice in the designed superlattices due to the misfit strain at the hetero-interface. Absorption experiments give the evidence of the discrepancy among the in-plane nearest-neighbor Mn–O bond length of the LBMO/LNO superlattices with different modulation lengths; the oxidation state of Mn atoms for LBMO sublayers in a series of strained LBMO/LNO superlattices is independent of the thickness of LNO spacer. The strain in the superlattice structure contributes significantly to the suppression of Curie temperature of the LBMO/LNO superlattices.

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1. Introduction

Doped pervoskite manganites $R_{1-x}D_xMnO_3$, in which some of the trivalent rare-earth ions R^{3+} are replaced with divalent alkaline-earth metal ions D^{2+} , have been investigated extensively in recent years, as they exhibit interesting metal-insulator transitions or orbital ordering. Their complex crystal structures and the strong sensitivity of their physical properties to subtle structural changes make the preparation of both bulk and thin films difficult [1]. Epitaxial thin films and superlattice structures provide many opportunities to study and to manipulate the characteristics of doped manganites [2,3]. Additionally, a thin film structure is required by envisioned device applications. However, under strain caused by lattice mismatch, lattice distortions are well known to be caused by substrate imperfections and the thickness of the film can

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strongly affect the properties of manganite thin films [4]. Strained colossal magnetoresistance (CMR) thin films typically exhibit characteristics that differ greatly from those of bulk compounds. In most cases, tensile strain suppresses ferromagnetism, which fact is normally understood by considering the strain-induced distortion of MnO₆ octahedra [4,5]. However, some anomalous results have also been reported, showing that tensile strain promotes ferromagnetism [6,7]. Furthermore, CMR thin films exhibit some novel characteristics [8], suggesting that the strain effect can be exploited to tailor or optimize the magnetotransport properties of CMR thin films, although the strain effect is not yet thoroughly understood. A few studies of heterostructures and superlattices that incorporate CMR manganites and epitaxially compatible oxides have been conducted [9,10]. All of these investigations have suggested the occurrence of an anomalous strain effect, which may be responsible for some novel and interesting characteristics of (La_{0.7}Ba_{0.3})MnO₃ (LBMO) thin films. A detailed study of the strained LBMO thin film may promote under-

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standing of the strain effect. This work reports systematically studies the magnetic properties of the variety of strained LBMO thin films using an LBMO/LaNiO₃ (LNO) asymmetric superlattice structure in which LNO was incorporated for its satisfactory crystallographic compatibility with the heteroepitaxial growth of a perovskite layer [11]; a smaller lattice than that of LBMO, which introduces a lattice mismatch strain at the hetero-interface, serves as a stressor for the manganite LBMO sublayer. This work establishes the importance of the correlation between structural information and the magnetic properties based on X-ray scattering, absorption and magnetic measurements.

2. Experiments

The designed superlattices with $(LBMO_m/LNO_n)_{12}$ in which m = 60 Å and n = 15, 30, 45 Å were adopted. Deposition was performed using a radio frequency (rf) magnetron sputtering dual-gun system. During deposition, the substrate temperature was kept at 550 °C and the gas pressure of deposition was fixed at 15 mTorr with an Ar/O₂ ratio 3:1.

The superlattice structure was characterized on measuring the (00L) and (0K0) crystal truncation rod (CTR) intensities with a synchrotron X-ray source. The detailed experimental setup has been described elsewhere [11]. Manganese K-edge X-ray absorption spectra were recorded at the wiggler beamline Taiwan. A Si (111) double-crystal monochromator with a 1.0 mm entrance slit was used for energy scanning. The energy resolution, $\Delta E/E$, was about 1.9×10^{-4} . Measurements were performed at room temperature in fluorescence mode and the sample was positioned at 90° to the incident X-ray beam.

3. Results and discussion

The epitaxy between the LBMO and LNO sublayers in the superlattice is demonstrated by the in-plane orientation with respect to the major axes $\{102\}$ of the SrTiO₃ substrate. The azimuthal (102) diffraction patterns of the $(LBMO_{60}/LNO_{30})_{12}$ superlattice at the vicinity of main peak are shown in Fig. 1. No other peaks are observed in the intervals between the four peaks, indicating the well epitaxial nature between the LBNO and LNO sublayers grown on a SrTiO₃ substrate.

Fig. 2(a) presents (00L) CTR intensity profiles of superlattices, with LNO spacers that range from 15 to 45 Å thick. For simplicity, values of *H*, *K* and *L* given in this paper are expressed in reciprocal lattice units (r. l. u.) referred to the STO lattice parameter (3.905 Å near 295 K). In the (002) CTR spectra, a well-defined (002) zerothorder peak with well-resolved satellites on both sides reveals the high crystalline quality of the prepared superlattices. In the LBMO/LNO superlattice, the lattice mismatch is estimated to be ~0.85% (bulk lattice parameters: $a_{LBMO} = 3.92$ Å and $a_{LNO} = 3.887$ Å). In this



Fig. 1. Phi scans of the (102) zeroth-order peak of the (LBMO $_{60}$ /LNO $_{30}$)₁₂ superlattice.



Fig. 2. (a) X-ray (00L) CTR intensity profiles of LBMO/LNO superlattices with various modulation lengths and that of the single LBMO epilayer. The arrow represents the position of the zeroth-order peak of superlattices and the (002) Bragg reflection of the single LBMO epilayer. (b) The dependence of mean lattice parameter *c* of superlattices on LNO spacer thickness.

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