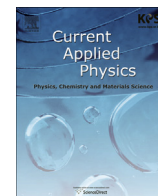




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## Structure and functionalities of manganite/cuprate thin film

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### ABSTRACT

Bilayer thin film samples consisting of colossal magnetoresistance manganite  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  (LCMO) and superconducting cuprate  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) were stabilized by means of pulsed laser deposition techniques on single crystal  $\text{SrTiO}_3$  (001) substrate. The X-ray diffraction measurements confirm the epitaxial relationship of grown bilayer samples. The functional properties of the LCMO/YBCO bilayer were explored through magnetic and electrical transport measurements. The magnetization curve of LCMO/YBCO bilayer sample retains the characteristic ferromagnetic-paramagnetic transition of LCMO and superconducting transition of YBCO. The electrical resistivity was found to show different trend around the superconducting transition depending upon the four probe electrical contacts configuration on bilayer samples and the obtained results lead to direct visualization of proximity effect. A change in sign of magnetoresistance near the superconducting and metal to insulator transition temperature is observed which is attributed to the intrinsic property of YBCO and LCMO layers.

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### 1. Introduction

In the current realm of technology, the aspiration to design the smart devices has steered the demand of functional materials. Manganites and cuprates are two renowned class of functional materials owing to their fascinating functional properties such as colossal magnetoresistance (CMR), multiferrocity and high temperature superconductivity [1–8]. Manganites are manganese based compounds, having the general formula  $\text{RMnO}_3$  (R is rare earth ion) with perovskite crystal structure while the cuprate superconductors generally have quasi-two-dimensional layered crystal structure comprising  $\text{CuO}_2$  planes separated by charge reservoir layers.  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  (LCMO) is a ferromagnetic (FM) CMR material showing the highest magnetoresistance among manganites family while  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) cuprate is known to be the first superconductor with superconducting transition temperature well above liquid nitrogen boiling point [2,7]. Co-occurrence of singlet ferromagnetism and superconductivity in bulk materials is highly improbable owing to their incompatible nature but this can be realized in bilayer thin film structure. Apart from retaining their individual functional properties, integrated thin film structure may show additional functional properties as

well [9–12]. The integration of FM manganite LCMO with superconducting cuprate YBCO in thin film form is further acknowledged for another interesting property known as proximity effect [13,14]. In proximity effect, the FM of LCMO and the superconductivity in YBCO are affected upon their integration in bilayer or superlattice structure. Spin polarized quasiparticle injection from CMR layer to YBCO superconductor has shaped a lot of attention too for spin-injection device technology [15]. For device realization, all the above mentioned mesmerizing functional properties call for additional research work on integrated structure of LCMO and YBCO. In this work, we have stabilized the epitaxial LCMO/YBCO bilayer thin film samples and studied their functional properties. The electrical transport properties of LCMO/YBCO bilayer samples were studied in detail to figure out the intrinsic effect of constituent layers as well as the effect of external parameters like temperature and magnetic field.

### 2. Experiment

Two bilayer thin film samples were prepared using pulsed laser ablation in a multi-target deposition chamber using dense pellets of LCMO and YBCO targets. For ablation, we have used KrF excimer laser ( $\lambda = 248$  nm, pulse-width  $\approx 20$  ns). During the sample preparation, laser pulse rate was kept at 4 Hz with energy density  $2 \text{ J/cm}^2$  of laser beam at the target surface. Chemically cleaned  $\text{SrTiO}_3$  (001) single crystal substrates were mounted in the

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deposition chamber and a base pressure of  $10^{-6}$  Torr was achieved prior to deposition. At first, YBCO layer (~50 nm) was grown at a substrate temperature of 820 °C. The oxygen partial pressure was kept at 350mTorr during the deposition. After the deposition, the deposition chamber was filled with ambient oxygen pressure and then the films were slowly cooled down to room temperature. Then half part of one of the grown YBCO layer sample was masked prior to grow LCMO layer. The chamber was again evacuated to achieve the base pressure and the LCMO layer (~50 nm) was grown on top of YBCO layer at 750 °C substrate temperature by keeping all other deposition parameters same. Stylus profilometer was used to measure the thickness of the grown films. The film orientation and phase purity of heterostructure was checked by X-ray diffraction (XRD) technique. Out of plane XRD measurements were performed at 5A beamline ( $\lambda = 1.0716$  Å) of Pohang Accelerator Laboratory synchrotron source (PLS-II) in South Korea. In plane XRD measurements were carried out using Bruker D8 Discover diffractometer (Cu  $K_{\alpha}$  radiation,  $\lambda = 1.5414$  Å). The magnetization nature of the grown sample was examined using SQUID-vibrating sample

magnetometer (SVSM; Quantum Design Inc., USA). Four probe resistivity technique was employed to study the electrical and magneto-transport properties at magnetic field (H) up to 8 T (H || to the films plane) and temperatures down to 5 K.

### 3. Results and discussion

Fig. 1(a) shows the normal  $\theta$ - $2\theta$  out of plane XRD pattern of LCMO/YBCO bilayer grown on STO (001) substrate. Apart from substrate peaks, the observed XRD peaks match to the [001] planes for both YBCO and LCMO orthorhombic structures. The peak around  $16^{\circ}$  refers to the sum of overlapping YBCO, LCMO, and STO signals and one can easily notice the increasing separation among these for higher order planes near  $49^{\circ}$ . Further, the observation of identical XRD reflections for the top LCMO layer and the STO substrate indicates the epitaxial nature of the grown layers. To explore such epitaxial stabilization of grown thin film structure, we have performed in-plane azimuthal angle measurements. Fig. 1(b) shows  $\Phi$ -scans (azimuthal angle scans) from the LCMO (161), YBCO (103) and STO (012) planes. The four symmetry peaks, anticipated for the cubic structure appear in the  $\Phi$ -scan of grown bilayer thin film. The observed four-fold in-plane symmetry confirms the epitaxial growth of LCMO and YBCO layers.

Magnetizations versus temperature (M-T) measurement were carried out in order to understand the magnetic behaviour of the grown LCMO/YBCO bilayer sample. Fig. 2 depicts the obtained M-T curve recorded at 200 Oe in the field cooled warming mode. One can clearly notice the appearance of magnetic transition at temperature around 250 K which belongs to the PM-FM transition of LCMO [16]. So the PM-FM transition of LCMO is retained in the LCMO/YBCO bilayer sample. Apart from this magnetic transition, another significant change in M-T curve is seen at lower temperature around 75 K. This magnetic transition at low temperature is possibly driven by the superconducting transition of underlying YBCO layer [17,18]. Due to FM LCMO layer induced proximity effect, the superconducting transition of YBCO may have shifted to lower temperature. To get more insight on this transition, we have also measured the magnetization in YBCO single layer (inset of Fig. 2) grown along bilayer. The observed magnetic transition around 80 K

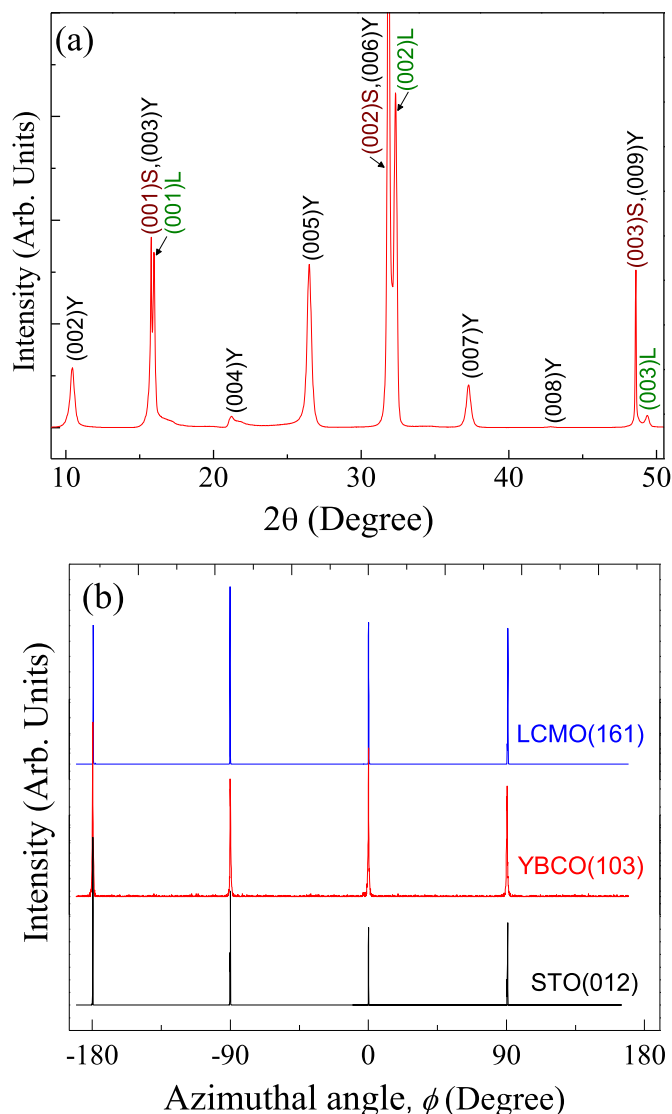


Fig. 1. (a) X-ray diffraction patterns of LCMO/YBCO film deposited on STO substrate. In this figure letters L, Y and S represent LCMO, YBCO and substrate respectively. (b) Asymmetric  $\phi$ -scan curves as a function of the azimuthal angle for substrate and deposited layers in bilayer sample.

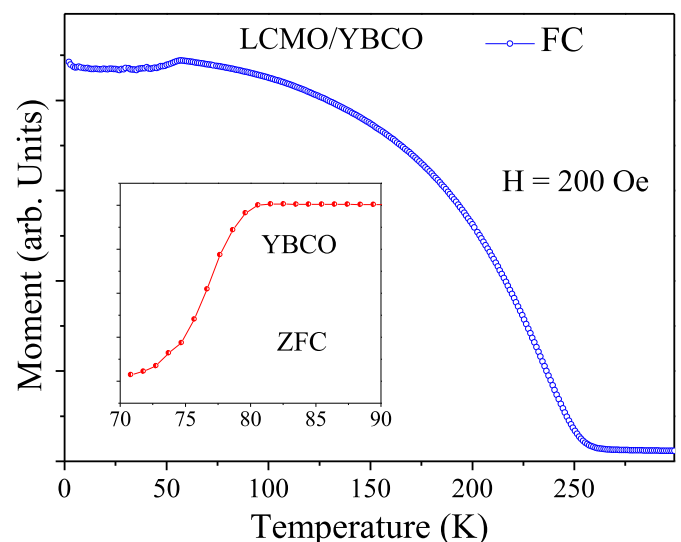


Fig. 2. Temperature dependent field cooled magnetization curves of LCMO/YBCO bilayer sample recorded during warming in 200 Oe external magnetic field applied || to the films plane. The inset shows the magnetization in single layer YBCO thin film sample.

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