Electron Microscopy and Microanalysis Study of La$_{1.89}$Ce$_{0.11}$CuO$_4$/Ba$_{0.5}$Sr$_{0.5}$TiO$_3$/La$_{0.67}$Sr$_{0.33}$MnO$_3$ Heterostructure

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Currently, all-perovskite oxide thin film heterostructures have attracted more and more attentions due to their potential applications in new kinds of devices. Their performances depend on interfacial structures of the heterostructures, and the interfacial structures have a strong correlation with thin film growth processes, especially for multilayer structures. In this study, we investigate the interfaces of an all-perovskite oxide heterostructure to reveal the correlation between interfacial structures and the growth processes. The p-i-n heterostructure is composed of manganite La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO), ferroelectric Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ (BST) and electron-doped high-$T_c$ superconductor La$_{1.89}$Ce$_{0.11}$CuO$_4$ (LCCO), which function as p-type layer, barrier layer (i), and n-type layer, respectively.

The epitaxial LCCO/BST/LSMO p-i-n heterostructure was deposited on SrTiO$_3$ (001) (STO) substrates by using pulsed laser deposition (PLD) method with a XeCl excimer laser of 308 nm [1]. The LSMO, BST and LCCO have pseudo-cubic, cubic and tetragonal $T'$-phase structure, respectively. The lattice parameters of LSMO, BST and LCCO are 0.3873 nm, 0.3950 nm and 0.4007 nm, respectively. The STO substrate has a cubic cell with $a = 0.3905$ nm. Small lattice mismatches between the substrate and the three rare materials ensure crystal growth of tri-layers. The surface of each individual layer and interfaces of tri-layer samples are characterized by scanning electron microscopy (SEM) and transmission electron microscopy (TEM), respectively.

As demonstrated in Figure 1a-b, grains with uniform size can be found over the surface of LSMO and BST thin films, indicating that the two layers have smooth surfaces for the growth of LCCO layer. However, large particles distribute on the LCCO film surface (Figure 1c), which can be attributed to the precipitation of Cu$_x$O ($x = 1$ or 2) during the growth process [1-2]. Figure 1d shows TEM dark-field cross-sectional image of the LCCO/BST/LSMO p-i-n heterostructure. Both LSMO and LCCO layers contain some columnar structures. The inset of Figure 1d is a [0 1 0] selected area electron diffraction (SAED) pattern taken from an area covering the STO substrate and the three thin films, which indicates crystalline structure of the three thin films. Detail information on interface structures are investigated by high-resolution TEM (HRTEM) and Fourier-filtered images. As given in Figure 2, yellow dashed lines reveal sharp LCCO/BST and BST/LSMO interfaces. It can be seen that the growth of three thin films is along [001]. On the left, the amplified HRTEM images of the LCCO/BST and BST/LSMO interfaces demonstrate epitaxial growth of the films on the STO substrate. Therefore, electron microscopy characterizations above clearly show that all-perovskite oxide LCCO/BST/LSMO p-i-n heterostructure can be epitaxially formed on the SrTiO$_3$ (001) substrate by using pulsed laser deposition method.

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Figure 1. (a-c) SEM images of the surface of LSMO, BST, and LCCO thin films; (d) TEM dark-field cross-sectional image of LCCO/BST/LSMO on STO (0 0 1) substrate (Inset: [0 1 0] SAED pattern).

Figure 2. Cross sectional HRTEM images of the LCCO/BST and BST/LSMO interfaces as well as their corresponding Fourier-filtered images (Insets).