Epitaxial and highly electrical conductive La0.5Sr0.5TiO3 films grown by pulsed laser deposition in vacuum

Wenbin Wu, Fei Lu, K. H. Wong, Geoffrey Pang, C. L. Choy et al.

Citation: J. Appl. Phys. 88, 700 (2000); doi: 10.1063/1.373724
View online: http://dx.doi.org/10.1063/1.373724
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v88/i2
Published by the American Institute of Physics.

Related Articles
Surface rippling on bulk metallic glass under nanosecond pulse laser ablation

Negative ions: The overlooked species in thin film growth by pulsed laser deposition

High-frequency electromagnetic properties of epitaxial Bi2FeCrO6 thin films grown by pulsed laser deposition

Al and Fe co-doped transparent conducting ZnO thin film for mediator-less biosensing application
AIP Advances 1, 042112 (2011)

Effects of stress on the optical properties of epitaxial Nd-doped Sr0.5Ba0.5Nb2O6 films
AIP Advances 1, 032172 (2011)

Additional information on J. Appl. Phys.
Journal Homepage: http://jap.aip.org/
Journal Information: http://jap.aip.org/about/about_the_journal
Top downloads: http://jap.aip.org/features/most_downloaded
Information for Authors: http://jap.aip.org/authors

Advertisement

Explore AIP’s new open-access journal
- Article-level metrics now available
- Join the conversation! Rate & comment on articles

Submit Now
Epitaxial and highly electrical conductive La$_{0.5}$Sr$_{0.5}$TiO$_3$ films grown by pulsed laser deposition in vacuum

Wenbin Wu$^a$ and Fei Lu

Structure Research Laboratory, University of Science and Technology of China, Hefei 230026, China
and Department of Applied Physics, The Hong Kong Polytechnic University, Kowloon, Hong Kong, China

K. H. Wong, Geoffrey Pang, and C. L. Choy
Department of Applied Physics and Materials Research Center, The Hong Kong Polytechnic University, Kowloon, Hong Kong, China

Yuheng Zhang
Structure Research Laboratory, University of Science and Technology of China, Hefei 230026, China

(Received 4 January 2000; accepted for publication 17 April 2000)

The target material with nominal composition of La$_{0.5}$Sr$_{0.5}$TiO$_3$ sintered in air is an insulator and not a single-phase compound. By pulsed laser ablation in vacuum at the multiphase La–Sr–Ti–O target, however, highly electrical conductive and epitaxial La$_{0.5}$Sr$_{0.5}$TiO$_3$ films have been fabricated on LaAlO$_3$(001) substrates. Structural characterization using three-axis x-ray diffraction ($\theta$–2$\theta$ scan, $\omega$-scan rocking curve, and $\phi$ scan) reveals that the films have a pseudocubic structure and grow on the substrates with a parallel epitaxial relationship. Atomic force microscopy images show the films have quite smooth surface, for a film 200 nm thick, the roughness $R_a$ is about 0.31 nm over the 1 $\mu$m$\times$1 $\mu$m area. Resistivity versus temperature measurements indicate that the films are metallic at 2–300 K and have resistivity of 64 $\mu$Omega cm at 300 K, which is about one order lower than that of the single-phase La$_{0.5}$Sr$_{0.5}$TiO$_3$ bulk materials. After the same deposition procedure, epitaxial La$_{0.5}$Sr$_{0.5}$TiO$_3$ films have also been grown on TiN buffered (001) Si substrates.

On the other hand, the epitaxial all-perovskite heterostructures usually need to be grown on silicon substrates to realize their applications in microelectronics. For the integration, buffer layer(s) will be inserted between the Si and the multilayered oxide system due to structural, thermal, and chemical mismatches between them.$^{10,15–18}$ Until now, there are few reports on epitaxial growth of perovskite-type oxide films directly onto the Si wafer. In general, a prerequisite of growing an epitaxial film on Si is that the film could be deposited in a reduced or inert-gas atmosphere to avoid oxidizing the surface of the Si wafer at the deposition temperature.$^{16–18}$ As has been reported by several groups, single-phase La$_{1-x}$Sr$_x$TiO$_3$ compounds could be synthesized only in reduced atmosphere,$^{12,14}$ indicating the possibility to grow La$_{1-x}$Sr$_x$TiO$_3$ directly onto Si substrates. In this article, we try to grow La$_{0.5}$Sr$_{0.5}$TiO$_3$ (LSTO) on the lattice-matched single crystal LaAlO$_3$ (LAO) substrates first. Our results will demonstrate that by pulsed laser deposition (PLD) in the vacuum atmosphere, epitaxial and highly electrical conductive LSTO films of perovskite-like structure could be grown on the LAO and TiN buffered Si substrates.

I. INTRODUCTION

The family of perovskite oxides displays a broad range of technologically important phenomena, including superconductivity, magnetism, and ferroelectricity. Due to the underlying structural and chemical similarities of these materials, and because of recent advances in their vapor deposition in thin film form, it is now possible to take advantage of such diverse behavior in epitaxial heterostructures. Epitaxial and highly electrical conductive perovskite-type oxide films have been found useful for electrodes and junctions in such heterostructures, and because of recent advances in their vapor deposition technologies, this opens the possibility to fabricate epitaxial La$_{0.5}$Sr$_{0.5}$TiO$_3$ compounds.

II. EXPERIMENT

The target used for PLD of the LSTO films was made by conventional solid state reactions in air with starting materials La$_2$O$_3$ (99.9% purity), SrCO$_3$ (99.5% purity), and TiO$_2$ (99.99% purity). The nominal composition was La$_{0.5}$Sr$_{0.5}$TiO$_3$. The sintering temperature was 1200–1300$^\circ$C, and during the target preparation, grinding, press,
and sintering were repeated three times. The LSTO films were fabricated on LAO(001) (perovskite subcell indices) or the TiN buffered Si (001) substrates by PLD for 15 min, using KrF excimer laser (λ = 248 nm, Lambda Physik, Complex 205) of 10 Hz in repetition frequency. The laser energy density irradiated on the rotating target was 4 J/cm². The target-substrate distance was 45 mm. Before the deposition the chamber was evacuated by a cryopump to a base pressure of 6 Torr, the growth temperature was 550–700 °C and during deposition the chamber pressure was 2 × 10⁻⁶ Torr. After deposition the films were cooled also at the base pressure. The thickness of the LSTO films is about 200 nm.

X-ray diffraction (XRD) patterns were obtained with Cu Kα radiation (Ni filter) by a four-circle diffractometer. Scanning electron microscopy and atomic force microscopy (AFM) operated in air at room temperature were employed to characterize the surface morphology of the films. A standard four-probe method was used to measure the resistivity of the films between 2 and 300 K. X-ray photoemission spectra (XPS) were taken by using a monochromatized x-ray source of Mg Kα radiation (hν = 1253.6 eV) and the energy resolution was about 0.8 eV.

III. RESULTS AND DISCUSSION

Figure 1 shows the XRD pattern of the air-sintered La–Sr–Ti–O target. It is seen that the target is not a single-phase compound. Apart from reflections that could be indexed according to a pseudocubic perovskite phase, some reflections from other unknown phase(s) appeared. Because in air atmosphere the most stable oxide of the titanium is TiO₂ with Ti⁴⁺, when replacing the Sr²⁺ by the La³⁺, a reduced ambient is usually indispensable to decrease the Ti valence state from +4.0.12–14 The resistivity of the target material at room temperature is larger then 10⁸ Ω cm.

To create a reduced atmosphere for the growth of stoichiometric La₃/₅Sr₃/₅TiO₃ (LSTO) compound, we made use of the multiphase target and the vacuum atmosphere (2 × 10⁻⁶ Torr) to deposit LSTO films by the PLD method. Figure 2 shows XRD patterns of a typical LSTO film deposited at 650 °C. Although the target is not of single phase, no impurity phases were recorded for the as-deposited film. In Fig. 2(a), strong reflections from LSTO(001) diffraction planes appeared and were indexed according to a pseudocubic structure. Some weak peaks due to the Cu Kβ radiation were also indexed. The inset shows the XRD rocking curve on the LSTO(002) reflection. In (b) and (c), XRD φ scans on the LSTO(220) and LAO(220) reflections are shown, respectively.

The surface of the LSTO films grown at the vacuum atmosphere is very smooth. Scanning electron microscopy images show no features of the film surface even with a magnification of 10⁵ (not shown). Figure 3(a) shows a typical AFM image taken for the same LSTO film shown in Fig. 2. The roughness R₉ over the 1 μm × 1 μm surface is only 0.31 nm. Some of this roughness is attributable to the pres-
ence of a few voids with about 200 nm in diameter and 1.8 nm in depth, as reflected by the line scan across the dark area in the image. From the line scan analysis of the image the area is 0.5 mm × 0.5 mm shown in Fig. 3(a), steps with height of about just one lattice unit length (4.03 Å) were also recorded. At present, the low roughness observed for the epitaxial LSTO films is not clearly understood. In general, it is believed that at reduced oxygen pressures laser–solid interactions produce species with energies large enough to damage the solid surfaces upon which they impinge.19 For a material system that could be grown in vacuum ambient such as the LSTO, the case may be quite different.

Figure 4 shows the normalized Ti2p and O1s (the inset) spectra from the epitaxial LSTO film (solid line) and the target material (dashed line). Compared with the target material, the absorbate (O atom) intensity in the O1s spectrum from the film is significantly reduced, as denoted by the arrow. For the Ti2p spectrum, apart from the peak at 458.85 eV, which is usually attributed to the atoms with Ti4+, a new component at a lower binding energy of 456.6 eV appeared. These spectra clearly reflect that the valence-state and chemical environment of the Ti atom in the films and the target material are different. The composition of the epitaxial LSTO films was also checked by the XPS measurements and the ratio of La:Sr:Ti was 0.44:0.56:1.02, near the nominal ratio of the target.

Figure 5 shows the temperature dependence of resistivity measured for the epitaxial LSTO films grown at 650 °C. A metallic transport behavior was observed over the whole temperature range of 20–300 K. The resistivity at room temperature was about 64 μΩ cm, which is about one order lower than that of the single-phase La1−xSr0.5TiO3 (x = 0.5) bulk materials.12 Tokura et al. found that for bulk LSTO, the temperature dependence of resistivity could fit well by the $\rho = \rho_0 + AT^2$ relation and they described the transport behavior by the Fermi liquid model.12 For the epitaxial LSTO film, the resistivity-temperature profile was found fit well by $\rho = \rho_0 + AT^{2.5}$ at higher temperatures, as shown in the inset. At temperatures lower than 150 K, however, the relation does not.
Figure 7 shows the XRD linear scan of the LSTO/TiN/Si heterostructure. The inset shows a ϕ scan on the LSTO(220) reflection of the same heterostructure.

[100]TiN(001)/[100]Si(001). The growth of LSTO films directly onto the Si(001) substrates is currently underway.

IV. CONCLUSIONS

In summary, epitaxial and highly electrical conductive LSTO films have been fabricated on LAO(001) substrates by PLD at vacuum of $2 \times 10^{-6}$ Torr. Three-axis XRD and AFM studies indicate that the as-grown LSTO films have excellent structural and surface properties. The epitaxial LSTO films show a metallic transport behavior at 2–300 K and a low resistivity of 64 $\mu \Omega \cdot \text{cm}$ at room temperature. These films could be employed for both basic scientific research and the fabrication of all-perovskite heterostructures for actual applications. The PLD procedure and the target preparation process used in this work may also be applicable to deposit thin films of other oxide materials that need to be synthesized in reduced atmosphere, such as the La–Sr–V–O system.

ACKNOWLEDGMENTS

The work described in this article was supported by the Chinese Natural Science Foundation and a grant from the Research Grants Council of the Hong Kong Special Administrative Region (Project No. PolyU5160/98P).

11. J. Z. Sun, W. J. Gallagher, P. R. Duncombe, L. Krusin-Elbaum,