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Title: Temperature-Dependent Growth of $LaAlO_3$ Films on $YBa_2Cu_3O_7$ C-Axis Films for Multilayer Structures

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TEMPERATURE DEPENDENT GROWTH OF LaAlO3 FILMS ON $YBa_2Cu_3O_7$ C-AXIS FILMS FOR MULTILAYER STRUCTURES

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ABSTRACT

Fabrication of ultra smooth films, free of micro-shorts, is essential to the development of High Temperature Superconducting (HTS) thin film devices. One such example is a SNS junction consisting of two HTS layers separated by a uniformly smooth continuous barrier material. Other schemes under consideration require multilayer structures of up to 5 - 7 epitaxially grown layers of complex oxide material. Successful fabrication of such devices necessitates understanding the epitaxial growth of polycrystalline oxide films on polycrystalline film templates. Toward this end we have developed a set of deposition parameters that produce high quality epitaxial insulating layers suitable for HTS device applications. All films in this study were grown by off-axis RF magnetron sputter deposition. $LaAlO_3$ films were deposited over MgO grown $YBa_2Cu_3O_7$ (YBCO) c-axis thin films at temperatures ranging from 200 to 700C and on virgin substrates at 600C.. Atomic Force Microscopy, eddy current measurements, and x-ray diffraction techniques were used to monitor the effect of growth conditions on the resulting film crystallinity, nanostructure, and electrical properties.

Ex-situ interrupted growth characterization of these materials has yielded new insight into the processes that control the growth mechanism and resulting microstructure. All films were polycrystalline. Below 600C, $LaAlO_3$ films were not epitaxial while films grown at 650C showed some <200> orientation. The shape of the underlying YBCO film is most clearly evident for the film grown at 400C. Surface roughness depended on the appearance of crystals on the film surface. The superconducting properties of the underlying YBCO film required O_2 annealing prior to deposition of the $LaAlO_3$ layer.

INTRODUCTION

Fabrication of ultra smooth films, free of micro-shorts, is essential to the development of high temperature superconductor (HTS) based technology. These criteria necessitate understanding and controlling the growth processes that affect structural properties such as crystalline quality, stoiciometry, hetero-epitaxy, substrate-film interfacial structures, inter and intra grain or island interfaces, and defect structures. All of these features impact directly on the resulting device transport properties. For the synthesis of multilayer polygranular complex oxide based devices, a major stumbling block to reproducibly making high quality structures is understanding the role of each parameter in a multicomponent interdependent parameter space. Each succeeding incorporated layer needs to be grown on a granular template without degradation of preceding layers. Equally important is the effect of lattice mismatch between each component materials.

A number of potential device designs, e.g. SNS (ramp type)[1,2,3] and SIS[4] schemes, require a uniformly smooth barrier layer with the desirable dielectric properties. Typical candidate barrier materials,e. g. $LaAlO_3$ (LAO)[2,5,6], graded Pr doped $Y_{1-x}Pr_xBa_2Cu_3O_7$[3], Y stabilized ZrO_2 (YSZ)[4], $SrTiO_3$ (STO)[7], and MgO[8,9], are not surprisingly the same ones that have already been proven to be good substrates for the growth of high quality $YBa_2Cu_3O_7$ (YBCO) c-axis films[10,11]. One such multilayer SNS ramp junction, under development at our laboratory and discussed elsewhere[1,2], is shown in figure 1. This device relies on four features: 1) a low temperature (LT) YBCO seed layer to minimize 45° rotated grains in the bottom YBCO electrode 2) a smooth

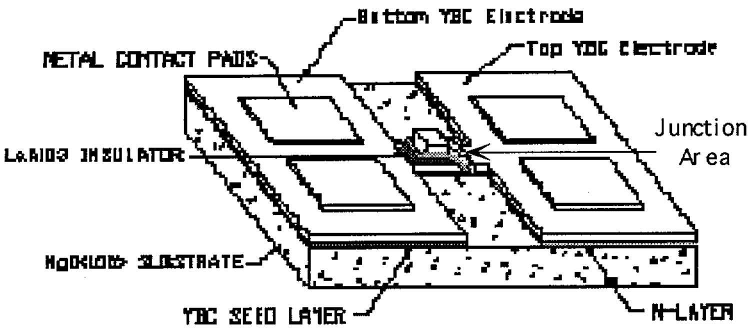

Figure 1. Diagram of SNS junction device which consists of 2 YBCO layer separated by a low temperature deposited YBCO layer. The $LaAlO_3$ layer functions as a barrier between the YBCO electrodes except within the immediate YBCO-nYBCO-YBCO junction region.

hole-free LAO insulating but not necessarily epitaxial layer to isolate the superconducting electrodes from each other, 3) a variable thickness LT-YBCO (670C) normal layer to fine tune junction properties, and 4) well developed sputter deposition and dry milling techniques to ensure a smooth ramp edge on which to form the actual YBCO-nYBCO-YBCO SNS junction. Because the actual junction does not involve the LAO layer, the epitaxial requirement for this layer is lifted. The critical considerations for the LAO layer are that it be smooth, hole-free, and ion mill to form a smooth ramp base area for the growth of the normal and final YBCO layers.

The primary emphasis of this paper will be on the temperature and substrate dependent structure and roughness of these LAO barrier layers grown over YBCO c-axis material but a brief description of the results of LAO buffer layer growth on single crystal STO, $NdGaO_3$ (NGO), and CeO_2 buffered R-plane sapphire substrates as characterized by atomic force microscopy (AFM) and x-ray diffraction analysis will also be presented. Although LAO growth on YBCO, perovskite substrates[5,6] and on Si[12,13,14] has been presented previously, the SNS junction structure presented here, in which a smooth 400C LAO layer is incorporated, represent the first device fabrication scheme that has reproducibly yielded high quality junction characteristics [1].

EXPERIMENTAL

Deposition parameters have been established that produce epitaxial LAO films on STO(100) and NGO(110) substrates as well as on top of epitaxial c-axis oriented YBCO thin films. Additionally, deposition parameters were established that produced exceptionally smooth non-epitaxial LAO films on top of YBCO films. In this study LAO films were grown in a 90° off-axis geometry by RF magnetron sputter deposition (Figure 2). Two inch planar magnetron guns with stoichiometric targets were used to deposit all films. A more detailed description of the experimental apparatus and the YBCO deposition parameters were reported earlier[2]. LAO films were grown over a bottom YBCO electrode on MgO (100) at deposition temperatures ranging from 200 to 700C or directly onto STO(100), NGO(110), and CeO_2 buffered R-plane sapphire surfaces at 600C. In all cases virgin substrates were employed. A P_{total} sputter gas pressure of 40mtorr (30mtorr Ar to 10mtorr O_2) was used with the RF power supply maintained at 100 watts. Substrates were positioned ~ $2^1/_2$ inches vertically and $1^1/_2$ inches horizontally, center to center, from the target. This combination of deposition parameters yielded a LAO growth rate ≈13.3Å/min. 600Å films were grown on the bare substrates while ~2400Å films were used to overcoated the YBCO films (~2600Å thick). Both the YBCO seed and normal layers (125Å) were grown at 670C as described elsewhere[2].Typically, the YBCO films were annealed for 30 min. at 450C in 250 torr oxygen prior to the LAO deposition. The LAO anneal was identical.

LAO and YBCO films were characterized by x-ray θ-2θ scans and AFM microstructural characterization. The YBCO film growth and structures have been studied previously[15,16]. The HTS superconducting properties were monitored by eddy current measurements.

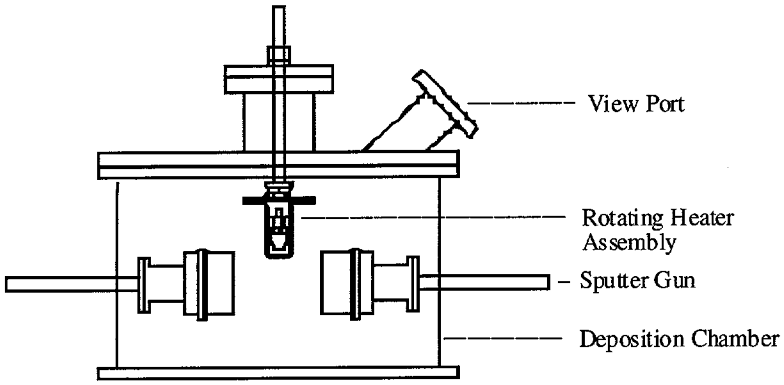

Figure 2. 90° off-axis RF magnetron sputter deposition system used to grow YBCO and LAO films. The substrate is mounted with Ag paint at the bottom of the heater assembly which is equipped with buried thermocouple for monitoring the substrate temperature.

RESULTS AND DISCUSSION

Epitaxial (100) oriented $LaAlO^3$ films were grown on STO(100) and NGO(110) substrates at a deposition temperature of 600C as indicated by x-ray θ–2θ scans, Figure 3a andb. However, the LAO film grown on CeO_2 buffered R-plane sapphire was nonepitaxial, Figure 3c. This is probably due to the lack of strong epitaxy of the CeO_2 buffer layer. All three films were extremely smooth as seen in Figure 4, with AFM root -mean-square (RMS) surface roughness measurement data ≈ 11Å, 15Å and 28Å, respectively. In addition to the slightly higher roughness of the CeO_2 grown film, the grain sizes revealed in the AFM images in Figure 3 are also slightly larger in this film. The 600C growth temperature for these epitaxial LAO films grown on single crystal substrates is in general about 100 degrees lower than previously reported for LAO films grown on Si[12,13,14], which could be attributed to the presence of an amorphous SiO_2 surface coating. No growth conditions were reported in reference [6] for LAO growth on STO or LAO for comparison.

At deposition temperatures at or above 600C, epitaxial <100> oriented LAO films were grown on YBCO film on MgO(100) single crystal substrates. Substrate temperatures below 600C resulted in polycrystalline or amorphous films. X-ray θ–2θ scans indicated that there was no epitaxially oriented LAO for films deposited at either 200C or 400C. However, in the case of the 400C film, x-ray data taken after deposition of the second YBCO film at 695C, showed the appearance (100) oriented LAO peaks diffraction data (see Figure 6). This was not true for the 200C film.

The maximum RMS surface roughness we observed was around 390Å for epitaxial films grown at 700C. In general, the nonepitaxial films deposited at temperatures below 600C, Figure 5a and b, were smoother than the epitaxial ones due to the appearance of LAO crystallites on the surface, Figure 5c and d. The AFM roughness data is summarized in Table I.

Table I. Surface roughness of $LaAlO_3$ films as a function of T

Temperature (C)	200C	400C	650C	700C
RMS	41Å	46Å	108Å	387Å

As can be seen in Table I, the RMS surface roughness was ≈ 40Å for LAO films deposited at 400C. This value is not unlike that expected for relatively smooth c-axis YBCO films alone. The RMS data for the 200C film is a bit deceptive since the roughness value of 41Å calculated over a $4\mu^2$ area does not tell anything about the local detail that contributes to the RMS value. In the latter case, the AFM $10\mu^2$ images revealed both surface particulates and possible voids.

All LAO films consisted of nano-sized grains scaling in size with temperature, increasing from 20nm at 400C film to ~40nm at 600C. Although the grains in the 200C film, Figure 5a, seem to have some orientation, this is probably due an AFM tip artifact. This films also differs slightly

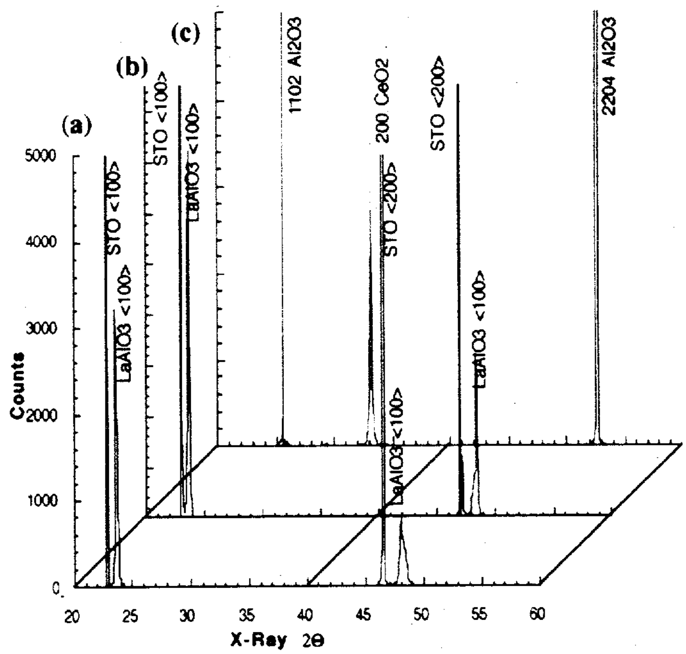

Figure 3. X-ray data for $LaAlO_3$ films grown at 600C on (a) $SrTiO_3$, (b) $NdGaO_3$, and (c) CeO_2 on R-plane sapphire.

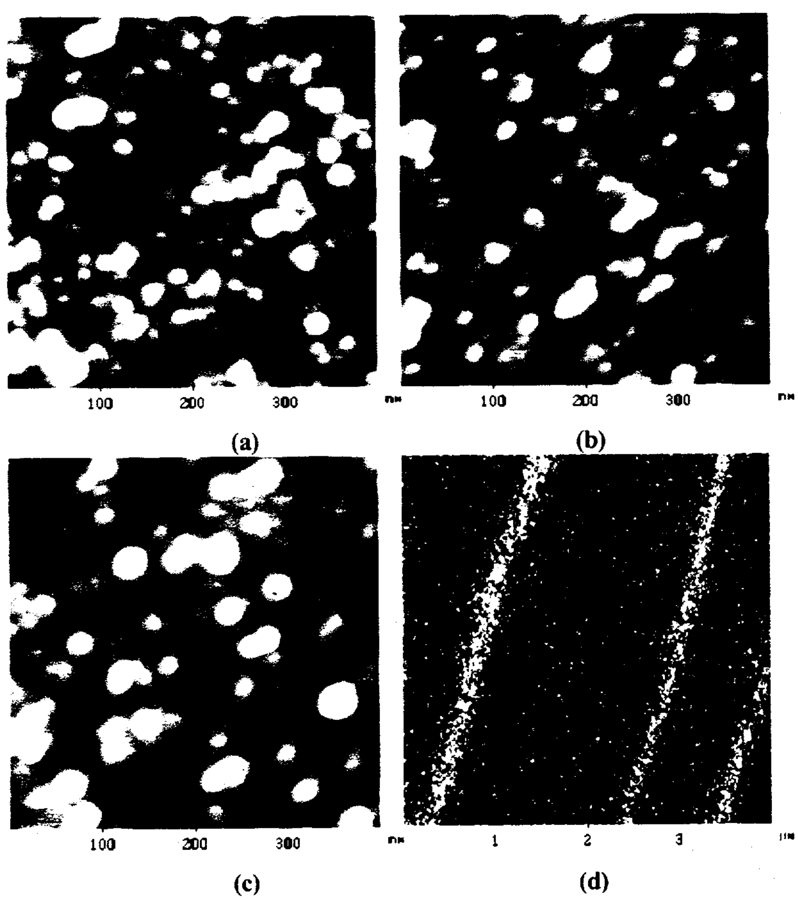

Figure 4. Tapping mode AFM images of $LaAlO_3$ films deposited on (a) $SrTiO_3$, (b) and (d) $NdGaO_3$, and (c) CeO_2 buffered R-plane sapphire. Note the parallel cracks on film (d). The scan area in images (a), (b), and (c) is $400nm^2$. Although the scan area in (d) is only a $4\mu^2$ sampling of the film surface, it is representative of the entire film.

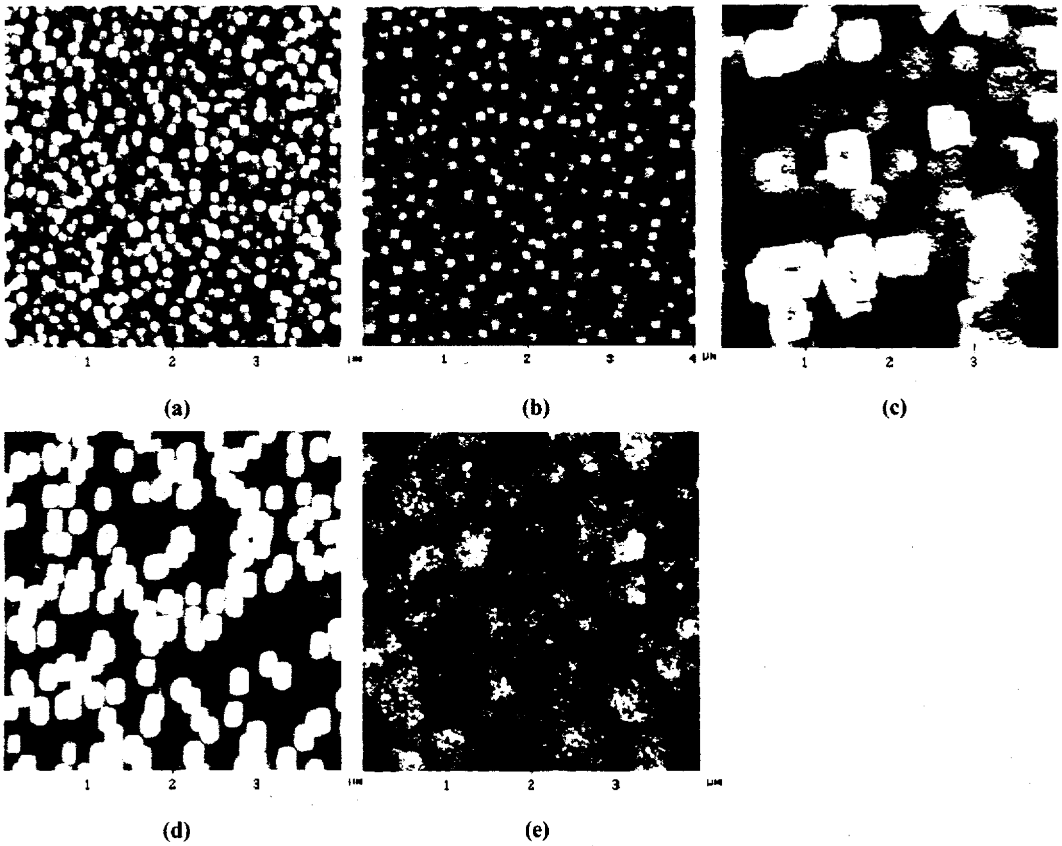

Figure 5. AFM 4 μ2 images of $LaAlO_3$ 45 minute depositions over YBCO films grown on MgO at different substrate temperatures (a) 200C, (b) 400C, (c) 650C, and (d) 700C. The last image (e) is a $LaAlO_3$ grown at 450C on a $GdBa_2Cu_3O_7$ sputter deposited on MgO at 40 degrees higher than the underlying YBCO film in image (b); all other conditions were the same.

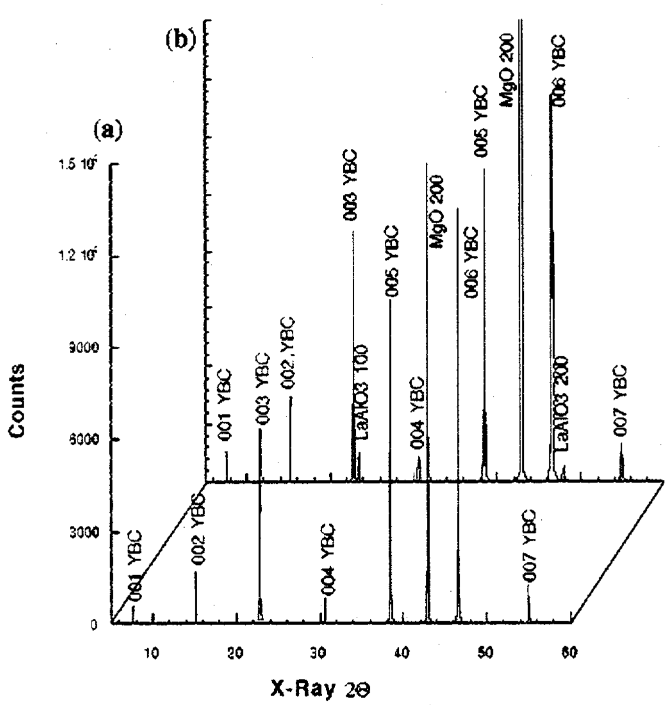

Figure 6. X-ray scan taken (a) after $LaAlO_3$ was sputtered over YBCO film and (b) following deposition of the second YBCO layer. The $LaAlO_3$ peaks appeared after the final layer was added to the multilayer structure.

from the rest of the films in this temperature series. The YBCO film was allowed to cool to room temperature before heating to 200C for deposition of the LAO insulating film. Outlines of the underlying YBCO islands are clearly visible in Figure 5b in spite of the 400C LAO coating. An AFM image of a 450C LAO film deposited over a 730C $GaBa_2Cu_3O_7$ (GBCO) film is included in Figure 5e for comparison. The RMS roughness and grain size of this LAO film are approximately the same as that of the 400C film grown on YBCO. The apparent larger size of the underlying HTS film is possibly due to its 40° high deposition temperature. A comparison of images 5b and 5c reveals a striking similarity between the surface nano-grains comprising the 400C film and the lower film surface of the 650C film. Similarly the surface crystals on the 650C film, which appear to possess a 4-fold symmetry, resemble the lower film surface of the 700C film. The progression in structure from one film to the next as the temperature is increased suggests the onset of a change in growth mode with temperature.

In conclusion, LAO films grown at 400 - 450C fulfilled the smoothness and hole-free criteria crucial to our SNS ramp junction devices. The deposition temperature is low enough to insure no degradation of the superconducting properties of the HTS material, although none was observed over the range of LAO depositions temperatures used in this study. The 450C deposition temperature also coincides with the normal YBCO annealing temperature. Annealing of the YBCO in O_2 prior to deposition of the LAO layer was essential for full oxygenation of the HTS material. Once deposited, the LAO served to protect the HTS material from O_2 loss. Smooth but not necessarily epitaxial LAO films were required to separate the HTS electrodes. Ion milling the 400C films yielded smooth ramp structures for the final normal and superconducting YBCO layers.

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