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Is $LaAlO_3$ a viable substrate for the deposition of high quality thin films of $YBa_2Cu_3O_{7-\delta}$?

Gad Koren and Emil Polturak

Physics Department, Technion—Israel Institute of Technology Haifa, 32000, Israel

E-mail: gkoren@physics.technion.ac.il

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Abstract
A systematic study of the surface morphology of epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ on (100) $LaAlO_3$ wafers is reported. The films were prepared by high pressure dc sputtering or laser ablation deposition, on wafers of 0.5–2.8 mm thickness and 2 or 3 inch diameter. Optical and atomic force microscopy (AFM) were used to characterize the surfaces, while transport was used to verify the high quality of the films. For films prepared under the same conditions, we found a systematic increase in size and number of extended defects in the films with wafer thickness. In some cases, a clear correlation was observed between the defect structure and the twin boundaries of the $LaAlO_3$ substrate. We specify the conditions for minimizing these defects.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Crystalline $LaAlO_3$ (LAO) with its low dielectric constant and loss tangent at microwave frequencies had been used extensively as an epitaxial substrate for the deposition of high temperature superconducting thin films [1–3]. Most of the work was done on epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) on (100) LAO for passive microwave applications, and in the present study we shall deal only with this system. Crystalline LAO has a good lattice match with YBCO to within ~1%, and can be grown up to reasonably large sizes. YBCO films on LAO have a transition temperature T_c of about 90 K and critical current densities at 77 K in the range of 1–4 × 10^6 A cm^{-2}. These numbers are similar to those obtained for the commonly used substrates $SrTiO_3$ and MgO, but unlike these substrates, LAO is generally heavily twinned due to its many structural phase transitions [1]. While heating the LAO wafers to the deposition temperature of YBCO, which is typically 800 °C, one can easily observe the motion, formation and annhilation of these twins with a long distance microscope. These phenomena can introduce strain and defects in the overlying YBCO films, and this is the subject of the present study.

2. Experimental details

The YBCO films in this study were prepared by either high pressure dc sputtering or laser ablation deposition. A schematic drawing of the dc sputtering system is given in the inset to figure 1. This system is similar to that described previously [4], but has a much larger size. It contains a water-cooled YBCO target of 5 inch diameter and a heater wound on a ceramic structure for wafers of 2 or 3 inch diameter. The wafers are held at the bottom of the heater by gravity while facing the target and plasma from below. Thus gravity forces most of the particulates or debris to fall on the target and leave the coated side of the wafer free from these objects. For two-side deposition, two LAO wafers were mounted one on top of the other at the bottom of the heater in order to keep the top side of the bottom wafer clean for a second deposition step on its backside. The thickness of the top (cap) wafer when used, was always 0.5 mm. Our standard deposition conditions are 1.5 Torr of oxygen pressure, 1.2 A current and a heater temperature of 980 °C in the oxygen gas at its centre. The actual wafer temperature is about 780 °C when the second cap wafer is used. Both wafers are heated by infrared radiation from the hot wires of the heater, while the cap wafer is also heated

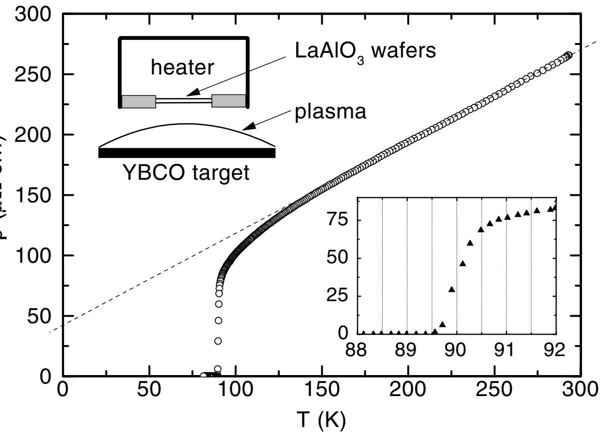

Figure 1. Resistivity versus temperature of a typical YBCO film on 0.5 mm thick and 2 inch diameter LAO wafer.

by convection of the hot gas inside the heater. The thermal contact between the bottom and cap wafers is poor due to a separation of a few μm between them. Thus the former is also heated by convection but has a strong convective cooling to the nearby sputtering target which is held at 50 °C. As a result, the top (cap) wafer is hotter than the bottom wafer, and since most of the infrared absorption occurs in the first 1 mm thickness of both wafers, the temperature of the bottom wafer is almost the same for all wafers used in the present study which are of 0.5, 1, 1.4 and 2.8 mm thicknesses. Typically, the deposition rate was 50 nm h^{-1}, and films of about 200 nm thickness were used in the present surface morphology investigation. The 'as-deposited' YBCO films had a transition temperature T_c of 86–87 K, and only after post-annealing in 0.3 atm of oxygen at 400 °C for 2 h did their T_c increase to 89–90 K, as seen in figure 1.

3. Results

Figure 1 presents the resistivity versus temperature results measured on a bridge forming a part of a microwave filter (a pole line) after post-annealing. This figure shows that the film is still underdoped, its absolute resistivity at 100 K is less than 100 $\mu\Omega$ cm, and its critical current density at 77 K is 2×10^6 A cm^{-2}. This demonstrates that the quality of our films is satisfactory, and in the following we focus on the detrimental defect structures that occasionally developed. We also worked with laser ablated films on 3 inch LAO wafers [5]. In laser ablated films however, the grains are much smaller than those in the dc sputtered films, and therefore the strain release process is much easier. As a result, these films are less likely to develop extended defects, as is also obvious from their capability of transmitting high microwave power of 1 kW at 0.5 GHz [5].

Figure 2 shows AFM images of four as-deposited films prepared by dc sputtering under the same conditions on (100) LAO wafers of 2 inch diameter but of a different wafer thickness. In all deposition runs pertaining to figure 2, a second LAO cap wafer of 0.5 mm thickness was mounted on top of the coated wafer. In figure 2(*a*) one can see large and small outgrowths or particulates of 500 nm and 100 nm height, respectively, spread over the film area. These are minority *a-axis* grains in the film which grow faster than the majority *c-axis* grains as was observed in many TEM studies of YBCO films. In figure 2(*b*) many of the small particulates are gone and small holes are visible in the places of these particulates which did not stick to the substrate surface and consequently fell off. This effect is even more pronounced in figure 2(*c*) where the substrate is thicker. In figure 2(*d*), a lesser number of bigger particulates is left, and a disordered quadro pattern of needle-like outgrowths is developed.

To better understand this behaviour versus the wafer thickness, we used optical microscopy with backside illumination. This allowed us to follow the defects which penetrate through the whole thickness of the films, or at least where the films are thinner and more transparent. Thus, in figure 3 the holes are the bright spots. Figure 3(*a*) shows the microhole pattern of a typically good film where the hole density is about 1 per 8000 μm^2. Figure 3(*b*) shows a film grown without the cap wafer and thus at a higher deposition temperature of about 830 °C. One can easily see that in addition to the much denser microholes (1 per 60 μm^2), transparent needle-like structures of 20–50 μm length appear. This type of defect is also observed in figures 3(*c* and *d*) in a film grown on a 2.8 mm thick wafer under our standard conditions (with the cap wafer). Although only small regions of a few mm^2 near the edges of the wafer showed this dense pattern of needle-like defects, one can already see the same kind of defects in figure 2(*d*) but with a much smaller density.

We now focus on the origin of the needle-like defects shown in more detail in figure 4. First we would like to distinguish them from the elongated microcracks seen in figure 4(*a*). In this image, we see a film that was grown in two separate deposition runs on the same side of the wafer. Exposure to a thermal shock of the first coating by a sudden removal of the shutter at the deposition temperature led to the observed microcracks. We shall not discuss this effect further in the present study. Figure 4(*b*) shows that the grains in our dc sputtered film are very big (2–4 μm) and have the typical screw dislocation growth pattern. This is very different from what is observed in laser deposited films where the grain size is an order of magnitude smaller. Figure 4(*b*) also shows that needle-like particulates grow inside the holes in the films, possibly on twin boundaries of the LAO wafer and is due to the higher temperature at the holes. Figure 4(*c*) shows elongated holes due to fallen needle particulates, while figure 4(*d*) shows some more needle particulates and also a faint zigzag structure of needles. Since the zigzag structure of these outgrowth needles apparently follows the zigzag structure of the underlying twins in the LAO wafer, we conclude that the twins of the LAO induce the growth of the needle-like defects in the YBCO films. This effect is obviously more pronounced when the wafer is thicker, since the strain in the film is then larger. The fact that we did not find the needle-like defects in laser deposited films is probably due to their much smaller grains which allow better strain relief. In contrast to small localized defects such as the microholes in figure 3(*a*), the extended size defects such as the 'needles' or the 'zigzag' are obviously detrimental to the current carrying capability of the films, because they allow for easy motion of the flux lines [6].

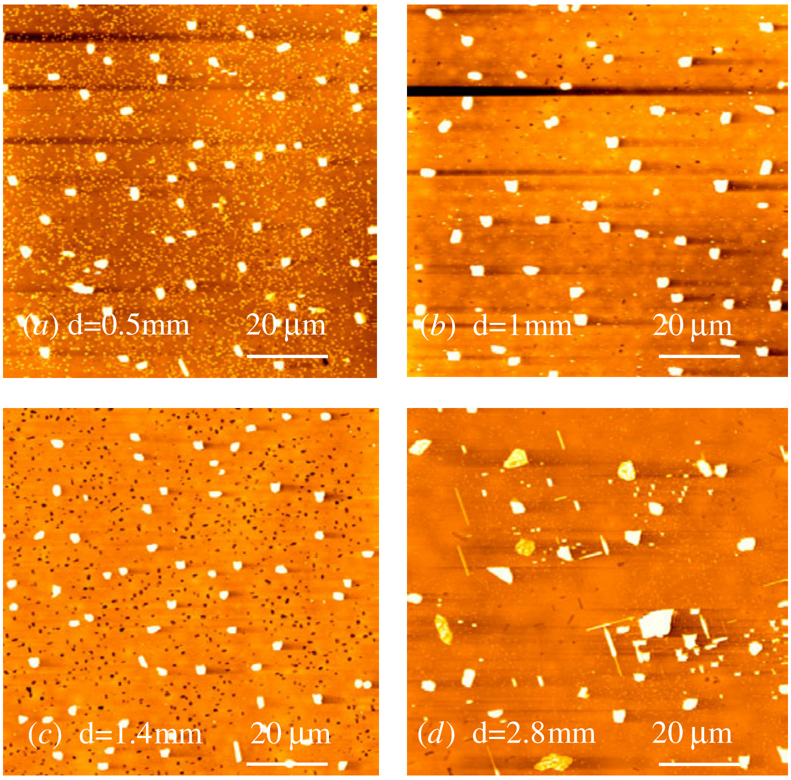

Figure 2. Typical AFM images of the surface morphology of four YBCO films prepared under the same conditions as in the film of figure 1, for four different LAO wafer thicknesses. The horizontal darker bands in (*a*) and (*b*) are artefacts caused by big particulates in the flattening process of the pictures.

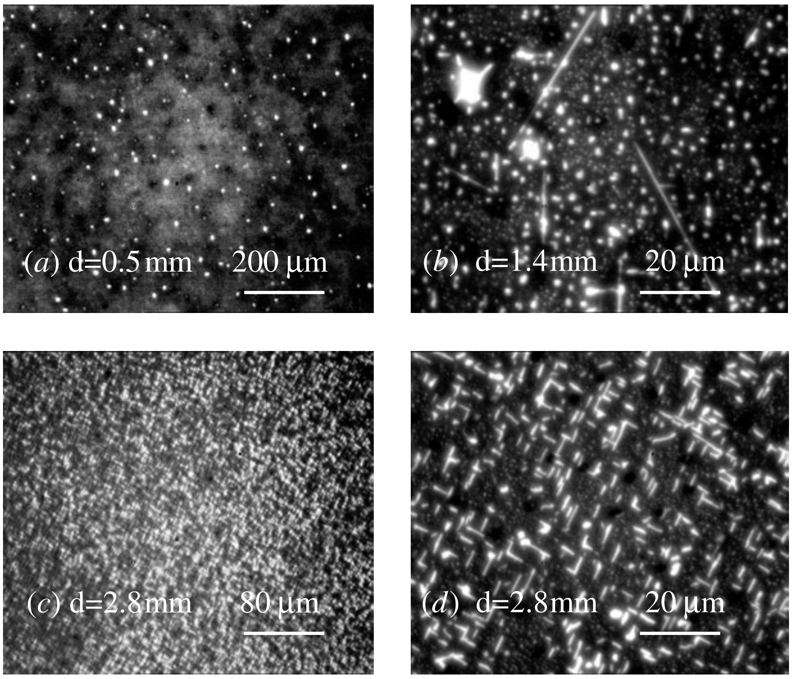

Figure 3. Optical microscope images with backside illumination of four films: (*a*) of the film in figure 2(*a*) (a standard good film), (*b*) of a film grown at a higher substrate temperature, (*c*) and (*d*) of the film in figure 2(*d*) but near the edges of the wafer.

Finally, we bring further evidence to our conclusion that extended defects in the YBCO films originate in twin boundaries in the underlying LAO wafers. Figure 5 shows an exceptionally long (and rare) zigzag kind of defect. Although not as clean as the previous needle-like defects, it seems to be composed of aggregates of many microholes surrounding

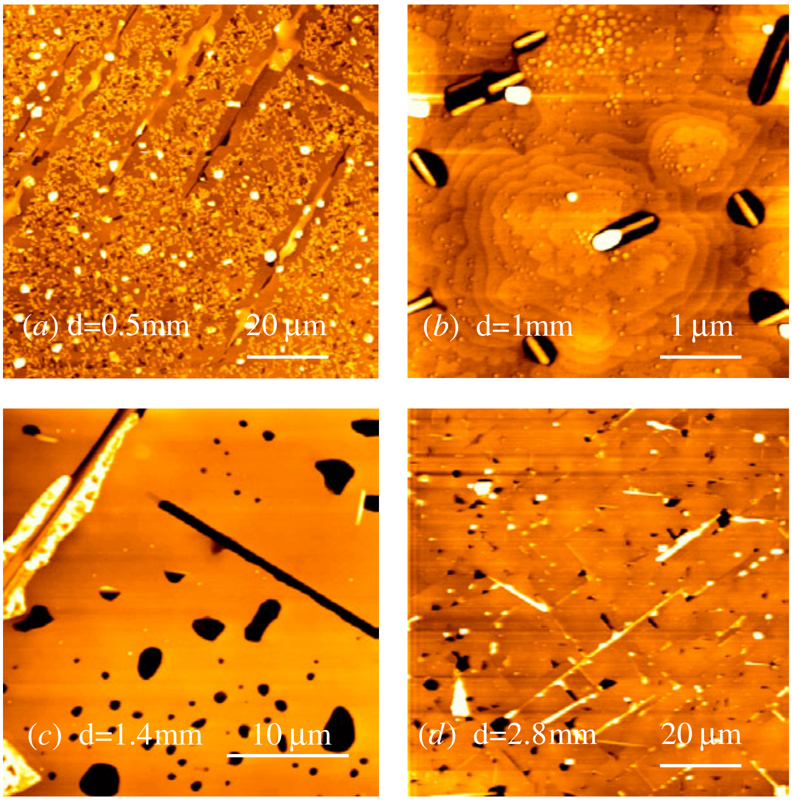

Figure 4. AFM images of (*a*) a film with microcracks due to a thermal shock, (*b*) of the film in figure 2(*b*) showing typical large grain size, screw dislocation growth and needles grown inside holes left behind by detached particulates, (*c*) of a film grown at a higher temperature showing holes due to fallen needles, and (*d*) of a film grown at a higher temperature showing intact needles and a zigzag of needles tracing some wafer's twins.

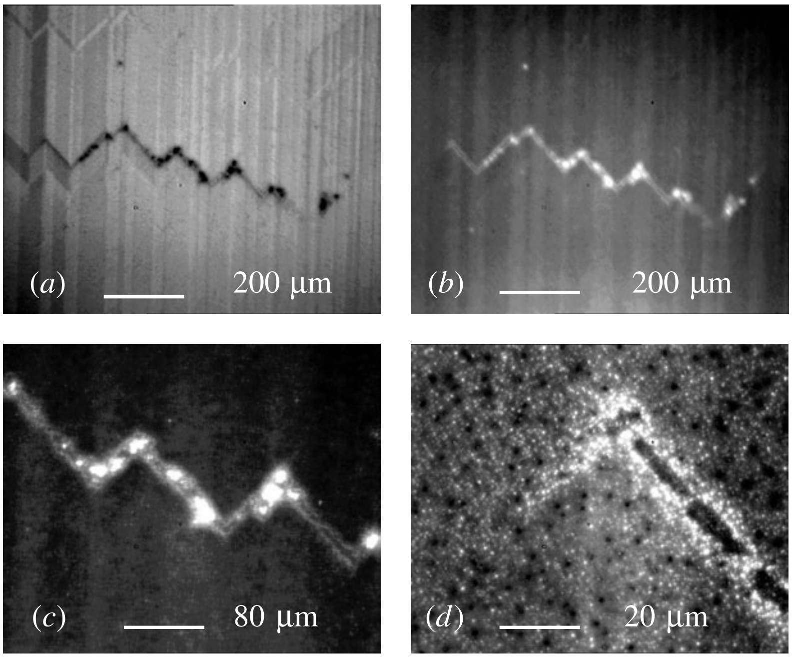

Figure 5. An example of a long zigzag defect in the film of figure 2(*c*) as seen in optical microscopy with frontside illumination (*a*), and backside illumination (*b*)-(*d*). This defect follows the underlying twin structure in the substrate.

elongated islands of YBCO film. The islands are the dark areas in figure 5(*d*). Clearly, such a zigzag defect in the YBCO film can only originate in the underlying twins of the substrate. Another zigzag structure *without* the defect in the YBCO film can be observed in the top part of figure 5(*a*).

4. Conclusions

In conclusion, LAO is a problematic substrate for the deposition of thin YBCO films. Especially when the films have large grains and a thick substrate is required, detrimental *extended* defects are developed. The use of thin LAO wafers of 0.5 mm thickness is advantageous, provided the deposition temperature is kept as low as possible.

Acknowledgments

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