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PREPARATION OF a-AXIS ORIENTED $YBa_2Cu_3O_x$ THIN FILMS*

NIE JIA-CAI(聂家财), CHEN LIE(陈 烈), YANG TAO(杨 涛), GUO LI-PING(郭丽萍), WU PEI-JUN(吴培钧), LIU GUI-RONG(刘贵荣), LI LIN(李 林), and ZHAO ZHONG-XIAN(赵忠贤)
National Laboratory for Superconductivity, Institute of Physics, Academia Sinica, Beijing 100080, China

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The a-axis oriented $YBa_2Cu_3O_x$ (YBCO) thin films could be grown on (100) $SrTiO_3$ (STO) substrates with STO buffer layers by dc and rf magnetron sputtering either by lowering the deposition temperature, or by using a self-template technique. For the latter, the resistivity of the thin film at 290 K along the substrate [001] direction is about four times larger than that in the [010] direction. The zero resistance temperatures T_{c0} are 89 K in both directions. So high quality a-axis oriented YBCO thin films can be prepared by the self-template technique. Also the T_{c0} increase monotonously with the reduction of the thickness of the YBCO seed layer.

PACC: 7475; 6150J; 6116; 6110

I. INTRODUCTION

The superconducting properties of high-T_c superconductors (HTSCs) are highly anisotropic. In particular, the coherence lengths in ab planes are much larger than the coherence lengths normal to the ab planes.[1] It is well known that high quality epitaxial $YBa_2Cu_3O_x$ (YBCO) thin films with the c-axis perpendicular to the substrate have in- plane electrical properties comparable to the best single crystals.[2] However, the c-axis oriented films are not suitable for making sandwich tunnel junctions. One of the reasons is that the coherence lengths in the directions of the a- and b-axes are several times larger than that of the c-axis. In recent years, preparation and application of a-axis oriented YBCO thin films have been the focus of many researches.[3-13]

In this paper, we describe two ways to prepare a-axis oriented YBCO thin films: (1) by lowering the deposition temperature approximately 100°C lower than that for making c-axis oriented YBCO thin films, and (2) by using a "self-template" method. These films are then characterized by X-ray diffraction and transmission electron microscopy.

II. EXPERIMENTAL

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The YBCO thin films were grown on (100) $SrTiO_3$ (abbreviated as STO) substrates by the magnetron sputtering method. A Leybold produced model Z550 magnetron sputtering apparatus was used, which was equipped with both dc and rf power sources. Sintered YBCO and STO pallets of 70 min in diameter were used as the sputtering targets. The distance between the substrate and the target was in the range of 20 to 30 mm. The substrates were adhered to a heater with silver paste and a thermocouple was used to monitor the heater's temperature. In order to improve the substrate's roughness a thin layer of STO with a thickness of 30 nm was deposited first on the STO (100) substrate using the rf magnetron sputtering technique. On doing this, the heater's temperature was held at 690°C, and the sputtering atmosphere consisting of 20 Pa of Ar and 5 Pa of O_2 was used with the rf power being 100 W. The deposition rate was 3.0 nm/min. After deposition, in situ heat treatment was carried out in 1 atm. of O_2 at 400–500°C for 10–30 min, and subsequently the heater together with the substrate was rotated to the position facing the YBCO target to deposit the YBCO thin film.

When using the first method, the heater's temperature was lowered to 590°C. A dc power source was used to sputter the YBCO target, with the voltage and current varying in the range of 100–130 V and 440–460 mA; the deposition rate was held at 3.3 nm/min. The second method, which is usually referred to as the self-template technique, was carried out by a two-step deposition process: first, a t_1-thick YBCO seed layer was sputtered at 590°C on STO (100) substrate with the STO buffer layer. After deposition, the thin film was annealed in 1 atm. of O_2 at 400–500°C for 10–30 min. Then the heater's temperature was gradually increased to 690°C, and further deposition of YBCO with a thickness of t_2 was carried out at this temperature. Finally a YBCO films was obtained with a thickness of $t(= t_1 + t_2)$ by the self-template technique.

The superconducting transition temperature T_c and transport critical current density j_c were measured using the standard four-probe technique. The strip lines of 50 μm in width and 500 μm in length were patterned by means of photolithography. A criterion of 1 μV was used for j_c measurement. The thickness of the thin films and the area of the bridges were measured with the DAKTAK IIA profilemeter. In order to investigate the thin film structure, X-ray diffraction (XRD) and transmission electron microscopy (TEM) were used.

III. RESULTS AND DISCUSSION

An *a*-axis oriented YBCO thin film could be obtained by sputtering at 590°C. The typical XRD pattern of the sample YS013 ($t = t_2 = 300$ nm) is shown in Fig. 1(a). Four peaks can be detected around 2θ of 23° and 47°, two of which are the (100) and (200) peaks from the STO substrate, and the other two correspond to the (100) and (200) reflections of YBCO.

For comparison, Fig. 1(b) shows the XRD pattern of the sample YST01 prepared by the self-template technique, with $t_1 = 20\,\text{nm}$ and $t_2 = 280\,\text{nm}$. Except the four peaks shown in the pattern, no other peaks from YST01 are detectable even if the background was magnified by a factor of 50, while the peaks in the background from YS013 are noticeable (Fig. 1(a)). A full width at half maximum (FWHM) of 0.10° was obtained for the (200) peak of YST01, while a FWHM of 0.25° was obtained for the (200) peak of YS013.

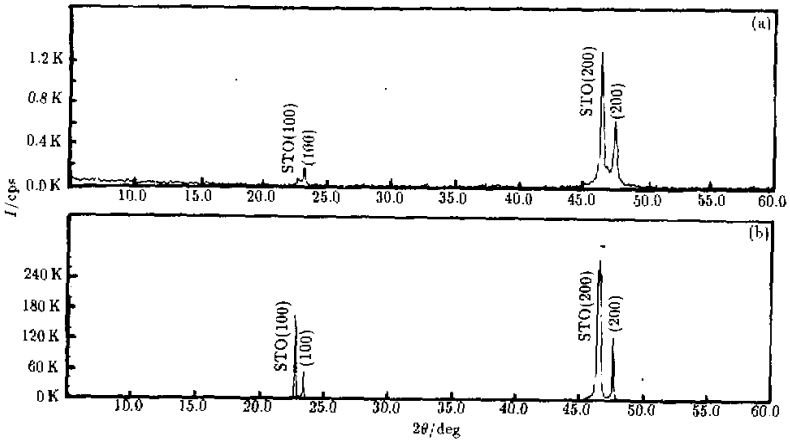


Fig. 1. X-ray diffraction patterns of the a-axis oriented films deposited on STO (100) substrates (a) with a STO buffer layer at a substrate temperature of 590°C; or (b) using the self-template technique.

Figure 2 shows the dependence of resistance upon temperature for the directions perpendicular and parallel to the direction of [001] of the STO substrate for the sample YS013. The inset is a schematic of the arrangement for the electrical measurement on the sample YS013. The dependence of resistance upon temperature in both directions is similar, however, above T_c (onset) the dependence of resistance upon temperature shows a semiconducting behavior. Figure 3 shows the resistivity vs temperature curves in the directions of [001] and [010] of the STO substrate for the sample YST01. The inset is a schematic of the arrangement for the electrical measurement on the sample YST01. The values of zero resistance temperature T_{c0} are 89 K in both directions, the dependence of resistivity upon temperature in both directions shows a metallic behavior and the resistivity in the direction of [001] at 290 K is 2.49 mΩ·cm, which is about four times larger than that in [010] direction (590 μΩ·cm). This anisotropy of resistivity was seldom reported for the a-axis oriented films with twin boundaries.

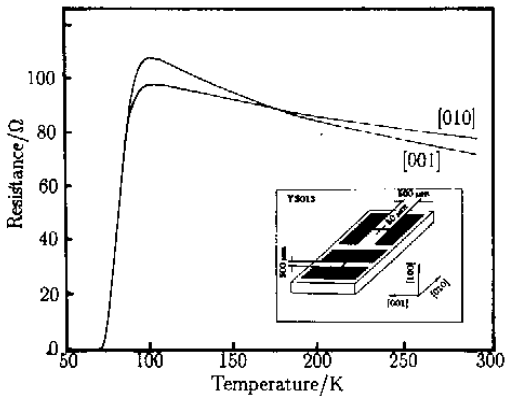


Fig. 2. Dependence of resistance upon temperature in the substrate [001] and [010] directions for an *a*-axis oriented YBCO film grown on a STO (100) substrate at a deposition temperature of 590°C.

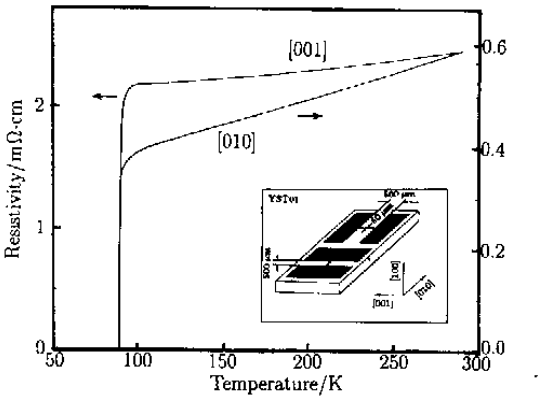
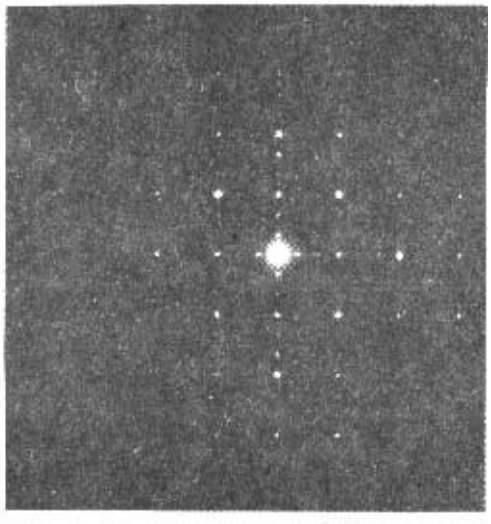


Fig. 3. Dependence of resistivity vs temperature in the substrate [001] and [010] directions for an *a*-axis oriented YBCO film grown on a STO (100) substrate by the self-template technique.

(a) TEM planar view of an *a*-axis oriented sample YS014 without a template layer, BPF and S boundaries being marked with corresponding arrows.



(b) The selected- area diffraction pattern obtained from the same planar region.

Fig.4. Transmission electron micrographes.

The microstructure of a YBCO film was characterized by TEM. The planar view TEM sample was prepared by grinding and ion milling on the substrate side only. A bright field image of an *a*-axis film (YS014, $t = t_2 \approx 300$ nm) prepared by simply lowering the deposition temperature is shown in Fig. 4(a), which clearly shows many strip-like structures, suggesting that the *ab* planes are perpendicular to the substrate surface and there exist many 90° [100] boundaries ([100] is the rotation axis which is a direction common to both grains). Two

types of boundaries could be observed: the symmetrical (S) and the basal-plane-faced (BPF) tilt boundaries, having the (103) (103) and the (100)(001) planes as their boundary planes respectively.[14] Some researchers have suggested that the S tilt boundaries may exhibit good transport properties because of an ideal one-to-one connection of CuO_2 planes across the boundary, whereas the BPF boundaries will be deleterious[15] due to the minimal connectivity of these planes.[16,17] This result is in agreement with our electrical measurement. A selected-area electron diffraction pattern obtained from the same planar region is shown in Fig. 4(b). Only diffracted spots instead of rings or arcs emerge, indicating that the sample is a single crystalline film.

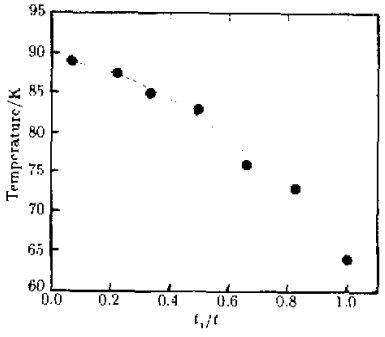


Fig. 5. The zero resistance temperature T_{c0} vs the thicknesses of the YBCO seed layer t_1 with identical total thickness of YBCO thin films $t \approx 300$ nm for the samples prepared with the self-template technique.

It was found that the thickness of the YBCO seed layer strongly affects the quality of the whole a-axis oriented YBCO thin film. Figure 5 shows the curve of T_{c0} vs the thickness of the YBCO seed layers t_1 with identical total thickness of YBCO thin films $t(\approx 300\,\mu m)$. The curve indicates that it is necessary to restrict the thickness of the YBCO seed layer, because the low deposition temperature of the seed layer will reduce the crystal quality of the films and subsequently T_{c0} will reduce monotonously with the increase of t_1.

IV. CONCLUSION

We have used two methods to prepare the a-axis oriented YBCO thin films. The first was simply reducing the deposition temperature about 100°C lower than that for preparing c-axis oriented films. These thin films were characterized by XRD and TEM. It was found that although the thin films are a-axis oriented, they are not very pure, and some defects exist in the thin films. The second was the self-template technique, by which we have obtained high quality a-axis oriented YBCO thin films. The electrical measurements showed that the quality of these a-axis oriented YBCO thin films are similar to that of c-axis oriented thin films. The resistivity along the [001] direction of the substrate at 290 K is about four times larger than that in the [010] direction. The zero resistance temperatures in both directions are 89 K. Thus, the a-axis oriented YBCO thin films prepared by the self-template method will be beneficial to both device preparation (such as biepitaxial grain boundary junctions and tunnel junctions) and basic research of the electrical anisotropy of YBCO.

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