THIN FILMS OF YBaCuO PREPARED BY MULTILAYER EVAPORATION PROCESS

X.K. Wang, D.X. Li, J.Q. Zheng, J.B. Ketterson, and R.P.H. Chang

Materials Research Center and Science & Technology Center for
Superconductivity, Northwestern University, Evanston, IL 60208

ABSTRACT

Thin films of YBaCuO were prepared as a superlattice of three constituents from three electron guns using a computer-controlled evaporator. After annealing, the multilayer films are converted to the homogeneous superconducting phase. Highly epitaxial thin films with: (1) the a-axis perpendicular to (100) $SrTiO_3$; (2) the c-axis perpendicular to (100) $SrTiO_3$; and (3) the [110] axis perpendicular to (110) $SrTiO_3$ were confirmed by x-ray diffraction as well as scanning electron microscopy and high resolution electron microscopy. Both the a-axis oriented and the c-axis oriented films exhibit zero resistance at 91K. The [110] oriented film shows the sharpest transition with a transition width of 1K and zero resistance at 85K. The zero field critical current density, J_c, determined magnetically, is in excess of $10^7 A/cm^2$ at 4.4K and $1.04 \times 10^6 A/cm^2$ at 77K for the c-axis oriented film; for the a-axis oriented film we obtained $6.7 \times 10^6 A/cm^2$ at 4.4K and $1.2 \times 10^5 A/cm^2$ at 77K. The orientation dependence of the critical current density in the basal plane of the a-axis oriented film was studied. The largest J_c's occur along the in-plane <100> axes of the substrate.

INTRODUCTION

The preparation of the high quality superconducting YBaCuO thin films with high T_c and J_c is of great importance not only for technological applications but also for fundamental studies. A considerable activity has been focused on this area.[1-5] A primary goal of most high T_c thin film efforts is to increase the critical current.[6-9] It is known that critical current density is affected by stoichiometry, the atomic arrangement at grain boundaries, intergrain coupling, alignment of the crystal grains, and the critical current anisotropy.[10] Accordingly, it is expected that samples consisting of grains with the desired stoichiometry, with good intergrain contact and a high degree alignment will show high value of J_c. Therefore, the preparation and characterization of essentially epitaxial thin films with well defined but different orientations should be of considerable interest.

We have prepared epitaxial, oriented thin films of YBaCuO with: (1) the a-axis perpendicular to (100)$SrTiO_3$ (a-axis oriented); (2) the c-axis perpendicular to (100)$SrTiO_3$ (c-axis oriented); and (3) the [110] axis perpendicular to (110)$SrTiO_3$ ([110] axis oriented). A computer-controlled, three e-gun, multisubstrate evaporator was employed. Our films were characterized by x-ray diffraction (XRD), scanning electron microscopy (SEM), high resolution electron microscopy (HREM), conventional four probe resistivity, and magnetization measurements.

PREPARATION OF THE THIN FILMS

The most important requirement for high quality thin films is to achieve strict stoichiometry in order to avoid second phase formation and to minimize interdiffusion between the substrate and the film. The composition of a multilayer film can be controlled simply by adjusting the thickness of each sublayer. With our system, thin films with an artificial-superlattice structure were deposited from three e-guns containing Y, BaF_2, and Cu in an atmosphere of 5×10^{-5} Torr of O_2. The substrates were mounted on a substrate wheel and maintained at 450°C. The wheel was driven by a computer-controlled stepping motor. Any of the (up to 20) substrates could be positioned over any of the three e-guns. A second computer-controlled stepping motor drove a shutter wheel which allowed the flux from any of the three e-guns to reach the substrate directly above it. The flux from each of the three e-guns was monitored by individual quartz crystal sensors which controlled their respective fluxes via feedback to the e-gun power supply. In addition, when the accumulated thickness of the sensor associated with the e-gun depositing a given layer of the multilayer structure reached a preset thickness, the computer was activated to advance the substrate to the next e-gun (and the monitor was rest to zero thickness). A complete superlattice was deposited on a given substrate before commencing deposition on the next substrate.

A typical film was deposited under the following conditions: the deposition rates were 1.1 Å/sec., 1.3 Å/sec., and 2.9 Å/sec. for Cu, Y, and BaF_2 respectively. The sublayer deposition time was 40 seconds. The number of cycles in a complete deposition was 60. The run-to-run reproducibility of the composition was within the resolution of the energy dispersive x-ray spectroscopy (EDAX) which was of the order of two percent.

As-deposited films, with thickness approximately of 1 μm, were smooth, insulating and disordered. They were annealed in flowing O_2 saturated with H_2O at 860 °C to 900 °C for 1/2 hour and then maintained at this temperature for 1/2 hour in dry flowing oxygen; this was followed by a slow cool down at 2 °C/min.

CHARACTERIZATION OF THE FILMS

Figure 1 shows scanning electron micrographs of the microstructures of some highly oriented films. Fig. 1(a) shows that the a-axis oriented film (confirmed by XRD[6]) consists of an array of orthogonal, interconnecting, rectangular grains with mean in-plane dimensions of 0.3 μm by 3.0 μm. This aspect ratio of ~ 10 is consistent with that found for single crystals of YBaCuO.[11] The SAED pattern and the HREM image of this a-axis oriented film confirm that each grain is a single crystal of the 123 phase. The b-axis and c-axis are aligned with the long and short dimensions of the rectangular grains, respectively, and are also aligned with the two in-plane a-axes of the substrate; i.e., they are epitaxial. Furthermore, the 90° junctions between the grains are free of any second phase formation or other type of decoration. A HREM image of a typical a-axis oriented film is shown in Fig. 2. Cross-section results confirm that the rectangular grains, shown in Fig. 1(a), nucleate directly on the surface of the (100)$SrTiO_3$ substrate and grow through to the film surface. Further details regarding the microstructure studies are described in Ref. [12]. Fig. 1(b) shows a micrograph of a c-axis oriented film consisting of columns having their c-axis perpendicular to the substrate (again confirmed by x-ray data). Although it displays very well oriented c-axis crystallites, the x-ray diffraction pattern and a HREM micrograph show that small amounts of other phases are present in this c-axis oriented film.

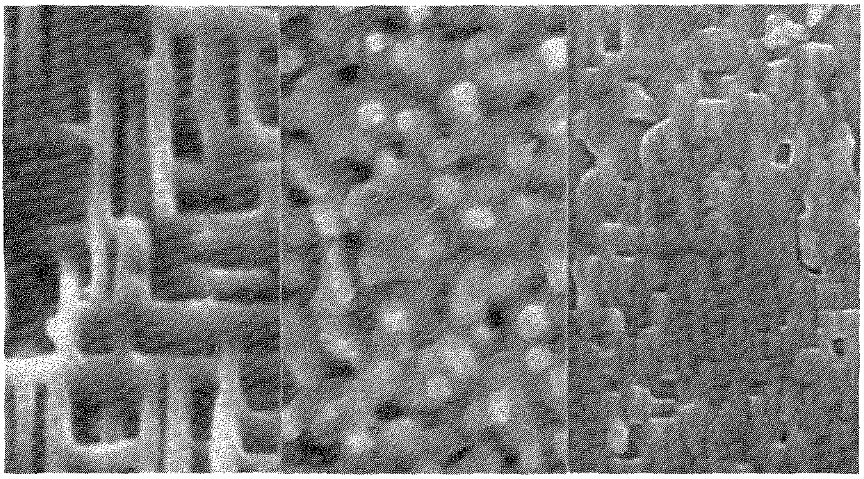

Figure 1.(a) The SEM micrograph of the a-axis oriented film showing a morphology consisting of an array of orthogonal, interconnecting, rectangular single crystal grains with well developed junctions, (b) The SEM micrograph of the c-axis oriented film, (c) The SEM micrograph of the [110] oriented film.

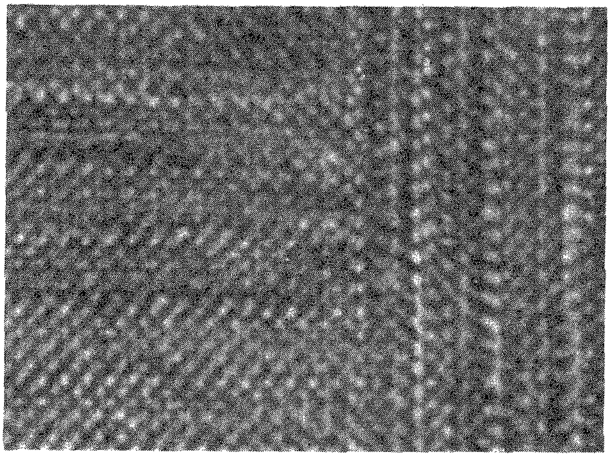

Figure 2. HREM image along [100] obtained from two interconnecting grains in the a-axis oriented film.

Fig. 1(c) shows a SEM micrograph, corresponding to a [110] oriented film (confirmed by XRD) in which only (hh0) reflections are observable. From the XRD data and SEM, as well as HREM micrographs, we conclude that the a-axis oriented film shows the best crystallinity with a high degree of grain alignment, both normal to and within the plane of the film. The crystallinity of the [110] oriented film is more perfect than that of the c-axis oriented film. We note in passing that films prepared in our lab, which were deposited on substrates such as MgO, ZrO_2, and Al_2O_3, where a considerable lattice constant mismatch exists, were never highly oriented. It is clear that lattice constant matching greatly affects the crystallographic orientations of the films.

Fig. 3 shows the resistive transition curves for the three oriented films. Transport measurements, performed by the conventional four probe technique, show that both the a-axis oriented and the c-axis oriented films exhibit zero resistance at 91K. The [110] oriented film shows the sharpest transition with a transition width of 1K, but achieves zero resistance only at 85K.

In addition to the characterizations mentioned above, we examined the films by performing extensive magnetic measurements using an SHE magnetometer equipped with a 50KG superconducting solenoid. The c-axis oriented film was measured with the field applied perpendicular to the plane of the film. The a-axis oriented film was measured with the magnetic field both perpendicular to and parallel to the plane of the film. In order to study the orientation dependence of the in-plane critical current, the film was positioned so that the magnetic field was at angles of 0°, 30°, and 45° to a substrate <100> axis, as shown in Fig. 4. The procedure described by Chaudari et al.,[13] was used to determine J_c. On removing the applied field, a magnetic moment is observed that is associated with trapped flux and circulating currents in the film. The critical current density, J_c, can be deduced using Bean's or Kim's model.[14-16] The critical current density $J^{c}_{c\perp}$ of the c-axis oriented film is in excess of 10^7A/cm^2 at 4.4K and 1.04 x 10^6A/cm^2 at 77K. The critical current density $J^{a}_{c\perp}$ of the a-axis oriented film is 6.7 x 10^6A/cm^2 at 4.4K and 1.2 x 10^5A/cm^2 at 77K (here the symbol $\perp$ indicates that the magnetic field perpendicular to the plane of the film). The critical current density $J^{a}_{c\theta=0^\circ}$ ($H\|$bars or equivalently the in-plane <100> substrate axis) is 1.6 time larger than $J^{a}_{c\theta=45^\circ}$. We could not distinguish a difference between $J^{a}_{c\theta=45^\circ}$ and $J^{a}_{c\theta=30^\circ}$.

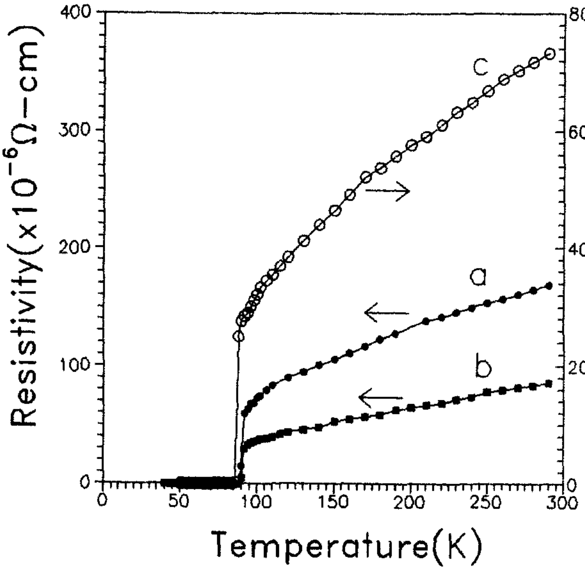

Figure 3. The resistive transition curves for: a) the a-axis, b) the c-axis, and c) the [110] oriented films.

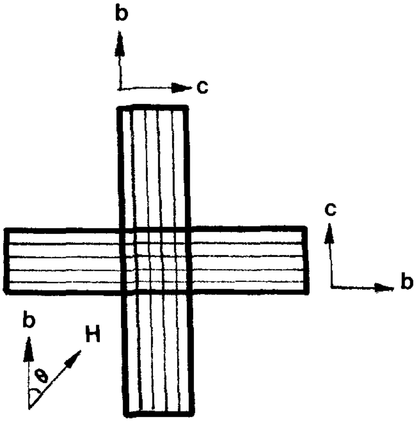

Figure 4. Schematic diagram showing the geometry of the crystal bars in the plane of the film and the reactive orientation of the magnetic field. Values of θ=0°, 30°, and 45° were studied.

Our results argue that the degree of alignment of crystal grains is crucial for achieving high critical current densities. Furthermore, our a-axis oriented film (with its high degree of alignment both normal to and within the plane of the film) has a rather high critical current density.

CONCLUSION

We have prepared highly oriented films of YBaCuO by multilayer deposition and characterized them. We have shown that the superconducting critical current densities can be rather high, not only in the c-axis oriented film, but also in the epitaxial a-axis oriented films, which have excellent intergrain contact.

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REFERENCES

1. B. Oh, M. Naito, S. Arnason, P. Rosenthal, R. Barton, M.R. Beasley, T.H. Geballe, R.H. Hammond, and A. Kapitulnik, Appl. Phys. Lett. 51, 852 (1987).

2. B.M. Clemens, C.W. Nieh, J.A. Kitt, W.L. Johnson, J.Y. Josefowicz, and A.T. Hunter, Appl. Phys. Lett. 53, 1871 (1988).

3. D.M. Hwang, T. Venkatesan, C.C. Chang, L. Nazar, X.D. Wu, A. Inam, and M.S. Hegde, Appl. Phys. Lett. 54, 1702 (1989).

4. C.H. Chen, J. Kwo, and M. Hong, Appl. Phys. Lett. 52, 841 (1988).

5. G. Linker, X.X. Xi, O. Meyer, Q. Li, and J. Greek, Solid State Communications 69, 249 (1989).

6. T.R. Dinger, T.K. Worthington, W.J. Gallagher, and R.L. Sandstron, Phys. Rev. Lett, 58 2687 (1987).

7. F.K. LeGoues, Philos, Mag. B57 167 (1988).

8. P. Chauhari, F.K. LeGoues, A. Segmüller, Science 238 324 (1987).

9. S. Nakahara, G.J. Fisanick, M.F. Yan, R.B. van Dover, T.Boone, and R. Moore, J. Crystal Growth 85 639 (1987).

10. K. Salama, V. Selvamanickam, L. Gao, and K. Sun, Appl. Phys. Lett. 54 5 (1989).

11. L.F. Scheneemeyer, J.V. Waszczak, T. Siegrist, R.B. van Dover, L.W. Rupp, B. Batlogg, R.J. Cava, and D.W. Murphy, Nature 328, 601 (1987).

12. D.X. Li, X.K. Wang, D.Q. Li, R.P.H. Chang, and J.B. Ketterson, J. Appl. Phys., **66**, 5505 (1989).

13. P. Chaudhari, R.H. Koch, R.B. Laibowitz, T.R. McGuire, and R.J. Gambino, Phys. Rev. Lett. **58** 2684 (1987).

14. C.P. Bean, Phys. Rev. Lett. 9 250 (1962).

15. Y.B. Kim, C.F. Hempstead, and A.R. Strand, Phys. Rev. Lett. **129** 528 (1963).

16. X.K. Wang, D.X. Li, S.N. Song, J.Q. Zheng, R.P.H. Chang, and J.B. Ketterson, MRS Fall Meeting Proceedings, Boston 1989.