High critical current densities in $YBa_2Cu_3O_{7-x}$ films on polycrystalline zirconia

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We report the growth on polycrystalline yttria-stabilized zirconia substrates of $YBa_2Cu_3O_{7-x}$ films with J_c (77 K) = 11 000 A/cm^2 and J_c (4.2 K) = 122 000 A/cm^2 by pulsed laser ablation. These J_c values are among the highest reported for $YBa_2Cu_3O_{7-x}$ on any polycrystalline substrate and approach the intrinsic upper limit for films with large-angle grain boundaries, as indicated by recent bicrystal experiments. We find that the substrate temperature during film growth is most important in obtaining high J_c polycrystalline films. Although the magnetic field dependence of J_c indicates the presence of weak links, the behavior of J_c (4.2 K, H) suggests that a percolative path consisting of low-angle grain boundaries exists in the films, resulting in J_c (4.2 K, 60 kOe) = 4100 A/cm^2.

Since the discovery of high-temperature superconductors, efforts have been made to obtain practical conductors with high critical current densities. These efforts include Ag-sheathed $YBa_2Cu_3O_{7-x}$ (Y-123), Tl-Ba-Ca-Cu-O and Bi-Sr-Ca-Cu-O superconducting wires and tapes,[1-3] magnetically and melt-textured bulk materials,[4,5] as well as Y-123 on polycrystalline metallic substrates.[6] For these systems, some degree of crystalline orientation results in improved current carrying capabilities. We recently reported the growth by pulsed laser ablation of c-axis oriented Y-123 thin films on randomly oriented polycrystalline yttria-stabilized zirconia (YSZ) with $T_c \sim 89$ K.[7] Their c-axis perpendicular orientation resulted in J_c higher than for randomly oriented bulk material; however, J_c (77 K) was only ~ 1400 A/cm^2.

In this letter we report the growth of Y-123 thin films with relatively high J_c on randomly oriented polycrystalline yttria-stabilized zirconia (YSZ) substrates. These films are c-axis oriented with $85\text{ K} < T_{c0} < 89$ K, J_c (77 K, $H=0$) = 11 kA/cm^2, and J_c (4.2 K, $H=0$) = 122 kA/cm^2. We find that substrate temperature is very important in obtaining high J_c Y-123 films, possibly because of grain boundary diffusion and substrate/film interactions. These values for J_c are among the highest reported for Y-123 films on polycrystalline substrates and approach the intrinsic upper limit for Y-123 films with high-angle grain boundaries, as determined by recent bicrystal experiments.[8] In addition, the magnetic field dependence of J_c (4.2 K) suggests that a percolative path of low-angle grain boundaries exists in the films, resulting in J_c (4.2 K, 60 kOe) = 4.1 kA/cm^2.

The Y-123 thin films were grown *in situ* by pulsed laser ablation as has been described elsewhere.[7,9] A KrF excimer laser beam [$\sim$350 mJ, 38 ns full width half maximum (FWHM) pulse duration] was focused to a horizontal line on a $\sim$25-mm-diam rotating Y-123 target. The focused energy density was 2.5–3.0 J/cm^2. The heated substrates were placed 6.5 cm from the Y-123 pellet. The best films were grown at a substrate temperature of 680 °C (calibrated by infrared thermometry) in an oxygen pressure of 200 mTorr. After deposition, the films were cooled in 600 Torr of oxygen at a rate of 10 °C/min in order to convert the as-deposited tetragonal films into fully superconducting orthorhombic Y-123 films. The films were patterned for critical current density measurements using standard photolithographic techniques with a bridge dimension of 100 μm by 3 mm.

As we have reported elsewhere, the large anisotropy in growth kinetics for Y-123 can result in c-axis perpendicular oriented thin films with an x-ray diffraction rocking curve width of only 1.0° on randomly oriented polycrystalline substrates.[8] However, because there is no in-plane epitaxial alignment, high-angle grain boundaries are present. From the work reported by Dimos *et al.* on the effect of grain boundary angle on J_c there appears to be an intrinsic limit J_c (77 K) $\sim$40–80 kA/cm^2 for films with high-angle grain boundaries.[7] In this study, we have grown polycrystalline Y-123 thin films in which J_c approaches this intrinsic limit.

Figure 1 shows the critical current density as a function of temperature for Y-123 thin films grown on randomly oriented polycrystalline as well as single-crystal (100) YSZ substrates. The film thicknesses are approximately 500 nm. The intrinsic upper limit for polycrystalline films dominated by large-angle grain boundaries can be approximated by reducing J_c for epitaxial Y-123 films by a factor of 1/50. This yields J_c values that are close to the highest observed by Dimos *et al.* for large-angle grain boundaries.[8] As seen in the figure, the film grown on polycrystalline YSZ at 680 °C is superior to the film grown at

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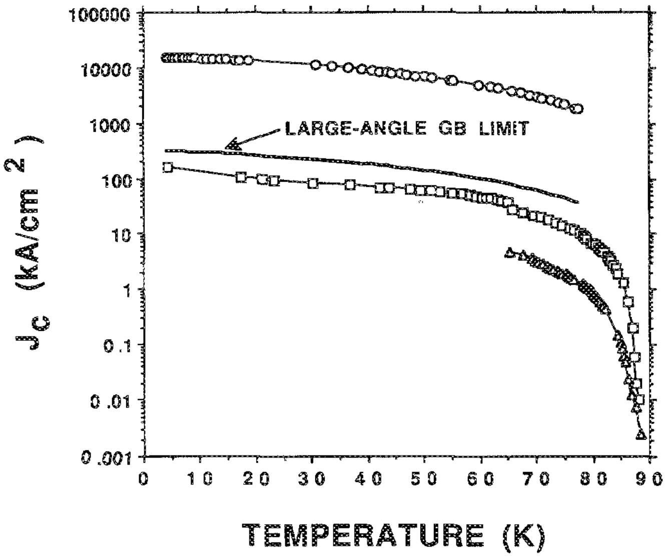

FIG. 1. Critical current density vs temperature for Y-123 thin films grown on (100) YSZ (○), as well as on polycrystalline YSZ at 680 °C (□), and 730 °C (△). Also shown is the intrinsic upper limit for $J_c(T)$, based on transport measurements across large-angle grain boundaries.

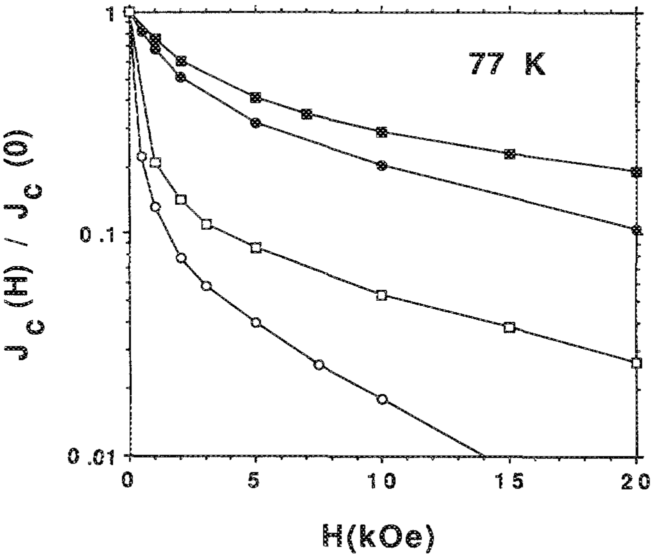

FIG. 2. Reduced critical current density vs magnetic field at 77 K for Y-123 grown on (100) YSZ (▨,●), and on polycrystalline YSZ (□,○). $J_c(H)$ is shown for the magnetic field applied parallel to (▨,□) and perpendicular to (●,○) the Y-123 *a-b* planes. The current is perpendicular to H for all cases.

730 °C, with J_c approaching the intrinsic limit for large-angle grain boundaries. We postulate that the dependence of critical current density on film-growth temperature is related to the interaction between Y-123 and YSZ. This results in the formation of $BaZrO_3$ which diffuses up through the grain boundaries and degrades intergranular conduction.[7,10] At reduced growth temperatures, the degree of substrate/film interaction is reduced, along with the rate of diffusion along the Y-123 grain boundaries, resulting in improved coupling of the supercurrent between individual grains. This leads to higher critical current densities, despite the fact that this reduction in film-growth temperature probably results in lower intragranular critical current densities, since our highest J_c Y-123 films on single-crystal substrates are obtained at 730 °C.

Figure 2 shows the anisotropic magnetic field dependence of J_c at 77 K for a high J_c Y-123 thin film grown on polycrystalline YSZ. A rapid decrease in J_c with magnetic field is observed, indicative of weak-link behavior. In addition, J_c decreases more rapidly when H is perpendicular to the Y-123 *a-b* planes. Two mechanisms contribute to this anisotropy. As is shown in the figure, a similar anisotropy exists for epitaxial Y-123 thin films on (100) YSZ. For epitaxial Y-123 films, it has been suggested that this anisotropy results from intrinsic flux pinning due to the layered structure.[11,12] The fact that the polycrystalline films also contain weak links (large-angle grain boundaries) introduces additional anisotropy, since weak link decoupling is more effective with the magnetic field direction parallel to the weak-link boundary. For vertical grain boundaries, H will be parallel to the entire grain boundary when it is perpendicular to the *a-b* planes. For the most favorable orientation, magnetic field parallel to the *a-b* planes, J_c (77 K) drops to 1800 A/cm^2 at 1 kOe and to 460 A/cm^2 at 10 kOe. Some hysteresis of J_c was observed, indicating flux trapping within the grains.

Figure 3 shows J_c (H) at 4.2 K for Y-123 grown on single-crystal (100) YSZ as well as polycrystalline YSZ. As was the case at 77 K, J_c initially decreases rapidly with magnetic field, as is expected for a film with weak links. However, for $H >$ 10 kOe, J_c is much less dependent on H, with J_c (4.2 K, $H = 60$ kOe) = 4.1 kA/cm^2. In fact, the dependence of J_c on H for $H >$ 10 kOe is similar to that found for epitaxial Y-123 films, as shown in the figure. This insensitivity of J_c to H at high fields suggests that a percolative path, consisting of low-angle grain boundaries, exists in the film. At high magnetic fields, the weak links in the

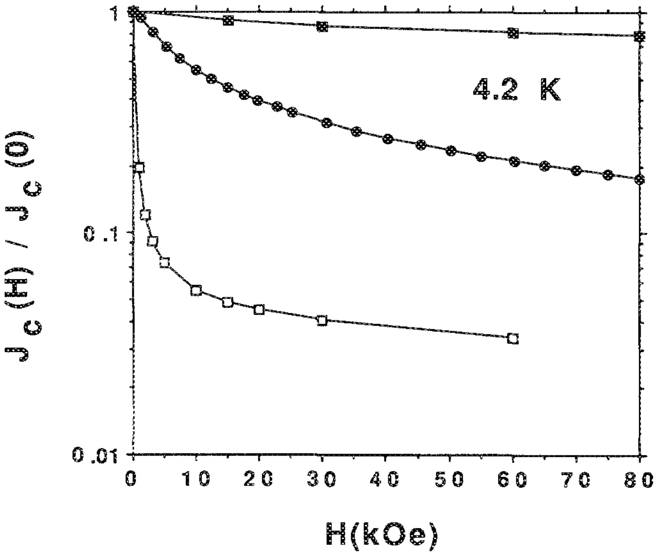

FIG. 3. Reduced critical current density vs magnetic field at 4.2 K for Y-123 grown on (100) YSZ (▨,●) and on polycrystalline YSZ (□). $J_c(H)$ is shown for the magnetic field applied parallel to (▨,□) and perpendicular to (●) the Y-123 *a-b* planes. The current is perpendicular to H for all cases.

film are effectively decoupled, and the super current flows only through the low-angle grain boundaries that do not behave as weak links.

In summary, we have shown that Y-123 superconducting thin films can be grown by pulsed laser ablation on randomly oriented polycrystalline YSZ substrates with J_c (77 K) = 11 000 A/cm^2 and J_c (4.2 K) = 122 000 A/cm^2. These values for J_c are among the highest reported for Y-123 films on polycrystalline substrates and approach the upper limit for J_c for Y-123 thin films containing large-angle grain boundaries, based upon recent bicrystal thin-film experiments.[8] For polycrystalline films, the possibility of grain boundary diffusion of film/substrate reaction products must be considered, and is consistent with the observed importance of reduced-temperature Y-123 film growth. Although we find that growth kinetics alone produce well-oriented c-axis perpendicular Y-123 films on polycrystalline substrates, these results imply that significant further improvements in J_c for c-axis perpendicular Y-123 polycrystalline thin films are likely to come about only through some degree of grain alignment in the a-b plane.

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[1] M. Okada, A. Okayama, T. Matsumoto, K. Aihara, S. Matsuda, K. Ozawa, Y. Morii, and S. Funahashi, Jpn. J. Appl. Phys. **27**, L1715 (1988).

[2] M. Mimura, H. Kumakura, K. Togano, and H. Maeda, Appl. Phys. Lett. **54**, 1582 (1989).

[3] M. Okada, R. Nishiwaki, T. Kamo, T. Matsumoto, K. Aihara, S. Matsuda, and M. Seido, Jpn. J. Appl. Phys. **27**, L2345 (1988).

[4] S. Jin, R. C. Sherwood, E. M. Gyorgy, T. H. Tiefil, R. B. van Dover, S. Nakahara, L. F. Schneemeyer, R. A. Fastnacht, and M. E. Davis, Appl. Phys. Lett. **54**, 584 (1989).

[5] J. Seuntjens, X. Cai, and D. C. Larbalestier, IEEE Trans. Mag. **25**, 2021 (1989).

[6] M. Ozaki, N. Harada, S. Akashita, and J. Chang, in *Science and Technology of Thin Film Superconductors*, edited by R. D. McConnell and S. A. Wolf (Plenum, New York, 1989), p. 363.

[7] D. P. Norton, D. H. Lowndes, J. W. McCamy, J. D. Budai, D. K. Christen, E. C. Jones, T. D. Ketcham, D. St. Julien, K. W. Lay, and J. E. Tkaczyk, J. Appl. Phys. **68**, 223 (1990).

[8] D. Dimos, P. Chaudhari, J. Mannhart, and F. K. LeGoves, Phys. Rev. Lett. **61**, 1653 (1988).

[9] D. H. Lowndes, D. P. Norton, J. W. McCamy, R. Feenstra, J. D. Budai, D. K. Christen, and D. B. Poker, Mater. Res. Soc. Symp. Proc. 169 (to be published).

[10] L. A. Tietz, C. B. Carter, D. K. Lathrop, S. E. Russek, R. A. Buhrman, and J. R. Michael, J. Mater Res. **4**, 1072 (1989).

[11] M. Tachiki and S. Takahashi, Physica C **162-164**, 241 (1989).

[12] D. K. Christen, C. E. Klabunde, R. Feenstra, D. H. Lowndes, D. Norton, H. R. Kerchner, J. R. Thompson, S. T. Sekula, J. Budai, L. A. Boatner, J. Narayan, and R. Singh, Mater Res. Soc. Symp. Proc. **169** (to be published).