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Improvements in Current-density of $YBa_2Cu_3O_{7-\delta}$ Films on Sapphire Buffered with Atomically flat CeO_2 Having High Density of Nanodots

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Experimental evidence of a cooperative self-assembled process was obtained where high temperature O_2 annealing (1000°C) induced a surface reconstruction in CeO_2 deposited on R-cut sapphire substrates. When the CeO_2 layer exceeded the critical thickness of 10 nm, an atomically flat CeO_2 surface with improved crystalline quality and a high density of nanodots was formed by the reconstruction. $YBa_2Cu_3O_{7-\delta}$ (YBCO) films grown on such CeO_2-buffered sapphire substrates had a high transition temperature ($T_{c\,\rho=0}$ > 90 K) and a high critical current density ($j_c > 3.0\times10^6$ A/cm^2 at 77.3 K and a zero applied magnetic field). A high density of nanodots are considered capable of creating extended defects, which result in strong pinning centers, in superconducting films. Magnetic field and angular dependence of j_c at various temperatures were investigated. YBCO film with the annealed CeO2 buffer layer showed a high j_c peak when the applied field was directed along the c-axis of YBCO. This j_c peak is considered to be caused by strongly c-axis correlated pinning sites, possibly due to the formation of nanodots on top of the CeO_2 wetting layer.
Key words: YBCO, CeO_2, Critical Current Density, Nanodots, C-axis Correlated Flux Pinning

1. INTRODUCTION

$YBa_2Cu_3O_{7-\delta}$ (YBCO) films deposited on sapphire (Al_2O_3) has generated much interest because of their reproducibly high film quality, high critical temperature (T_c) and high critical current density (j_c). Sapphire is a preferred substrate due to its small dielectric constant and low loss tangent. Moreover, good thermal conducting characteristics of Al_2O_3 substrates enable rapid thermal quenching, such as necessary for fast switching elements in fault current limiters [1]. However, buffer layers are necessary for deposition of YBCO on Al_2O_3 to provide good lattice matching and to prevent inter-diffusion between these two materials. CeO_2 has been found to meet these criteria [2,3]. For high jc YBCO films, in-plane orientation of the film [4] and strong pinning centers in the film are required. Therefore, a high-density CeO_2 buffer layer with a smooth surface and high lattice perfection and introduction of flux pinning related defects are the key factors for growth of high j_c YBCO films on such layers. Work has been done to improve the surface smoothness and the superconducting properties of YBCO films by using high temperature annealing (1000°C) of CeO_2-buffered sapphire substrates before deposition of the films [5,6]. The high temperature O_2 annealing has promoted a cooperative self-assembly process for CeO_2 buffer layers to form an atomically flat surface. The crystalline quality of CeO_2 buffer layers and the subsequent YBCO films was found to be significantly improved by the self-assembly process [6]. However, how this high temperature annealing affects the microstructure of CeO_2 buffer-layers and the YBCO films remains unclear, and thus, how j_c of the YBCO films are affected is also unclear.

In this study, we report the enhanced j_c of YBCO films on sapphire with the self-assembled CeO_2 buffer layer and nanodots. In particular, magnetic field and angular dependence of j_c of the YBCO films deposited on the as-grown and annealed CeO_2-buffered sapphire substrates were measured and compared. YBCO film with the annealed CeO_2 buffer layer showed a high j_c peak when the applied field was directed along the c-axis of YBCO.

2. EXPERIMENTAL DETAILS

CeO_2 thin films were deposited at various thickness on R-cut sapphire substrates by pulsed laser deposition (PLD) utilizing a KrF excimer laser source (248 nm, Lambda Physik COMPex 205) operated at 300 mJ. Depositions were made at a substrate temperature of 760-780°C in O_2 at 300 mTorr. The deposition rate was about 0.028 nm/pulse at a laser repetition rate of 1 Hz. The resulting as-grown CeO_2-buffered sapphire were further annealed *ex-situ* at 1000°C in an O_2 flow for 1 hour and then cooled to room temperature [6]. Then YBCO films were grown on these substrates by epitaxial deposition by PLD. The laser fluence of ~ 4 J/cm^2 (525

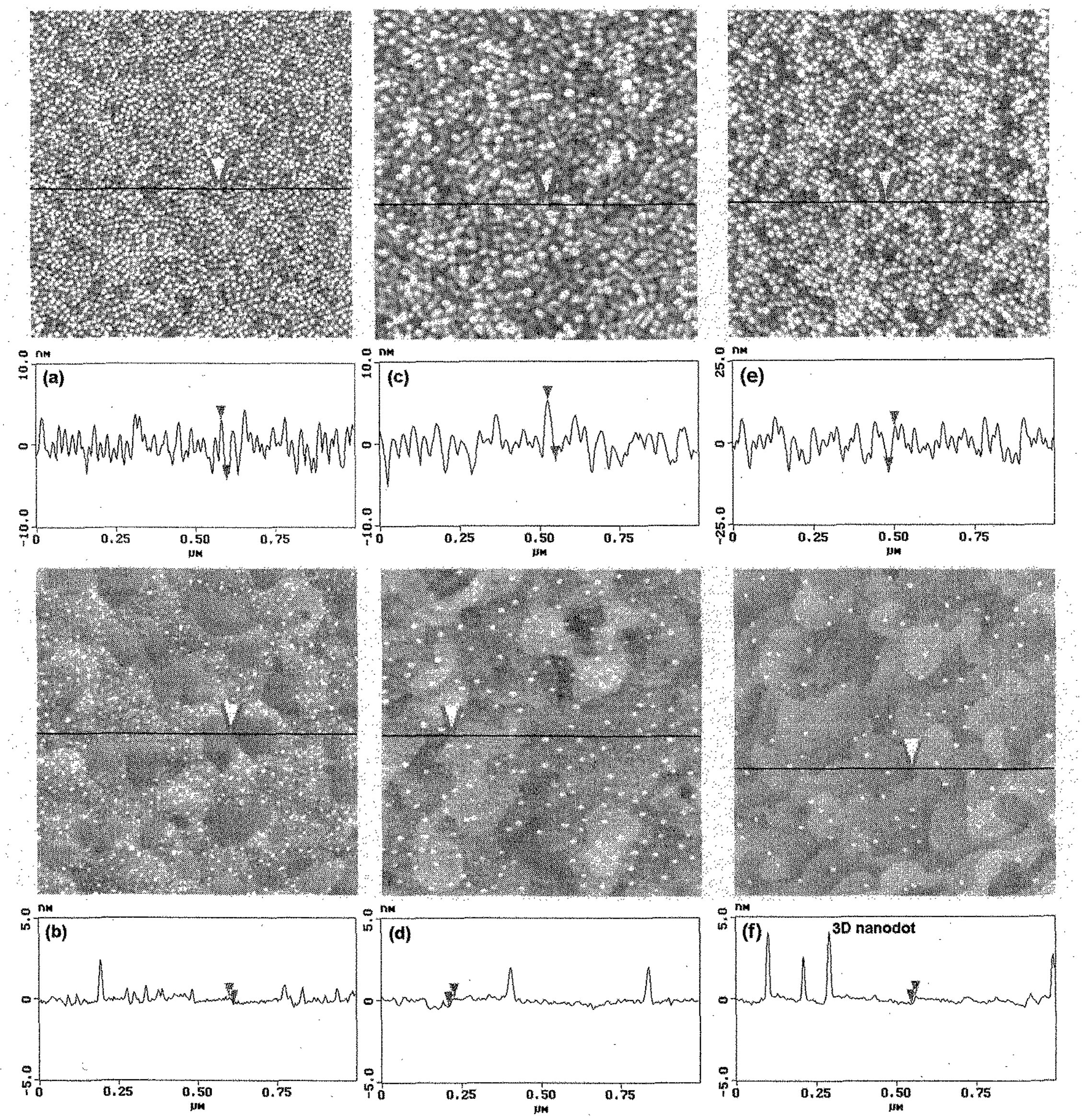

Figure 1. AFM images of three CeO_2 films with thicknesses of (a) (b) 10.0 nm, (c) (d) 23.6 nm and (e) (f) 36.6 nm, respectively, on R-cut sapphire substrates before and after the high temperature O_2 annealing.

mJ) on a YBCO target at 5 Hz laser repetition rate was used for the YBCO deposition, with the O_2 pressure at 300 mTorr and substrate temperature at 730°C. After YBCO deposition, the samples were annealed at 430°C in O_2 at 300 Torr for 1 hour and then cooled to room temperature.

The surface morphology of the films was measured by using atomic force microscopy (AFM, Nanoscope III) and transmission electron microscopy (TEM, Model JEM-2000EX) operated at 200 kV. Specimens for plan-view and cross-sectional TEM were prepared by standard techniques including Ar ion milling. The microscopic composition of YBCO film was analyzed by energy dispersive X-ray spectroscopy (EDS, Oxford Instruments, ISIS).

To measure the j_c of the deposited YBCO films, 2 mm-long and 20 µm-wide bridges were etched in YBCO films by pulsed laser etching. The current-voltage curves of these bridges were measured by a four-probe transport method using pulse currents. A voltage criterion of 1 µV/mm was used to define j_c.

3. RESULTS AND DISCUSSION

It has been demonstrated that the high temperature (1000°C) O_2 annealing could induce a surface reconstruction in CeO_2 deposited on R-cut sapphire substrates [6]. When the CeO_2 film exceeded a critical thickness (~ 10 nm), an atomically flat surface was formed by the reconstruction. Fig. 1 shows AFM images of CeO_2 films with various thicknesses before and after the high temperature O_2 annealing. The as-grown CeO_2 films show small, round-shaped islands, typical of a three-dimensional growth mode (Figs. 1a, 1c, and 1e). For the annealed samples, atomically flat surfaces are clearly evident in Figs. 1b, 1d, and 1f as atomically flat terraces and atomic steps. AFM also revealed irregularly distributed nanodots, 10-20 nm in diameter and up to 10 nm in height. Both their dimensions and surface mean density depend strongly on the nominal thickness of the CeO_2 film, as shown in Fig. 1. On the other hand, both the surface mean density and the dimensions of nanodots depend also strongly on the misorientations (by miscut) of the underlying sapphire plane around $(1\underline{1}02)$. If the vicinal angle is over 1°, nanodots will not form, frequently observed features, instead, are trains of parallel steps of atomic height aligning along one direction (data not shown). Similar to the sputtered nanodots [7,8], imperfectly aligned CeO_2 nanodots is considered capable of introducing extended defects, which result in strong pinning centers, in superconducting films and should therefore increase j_c of the film. Also, the surface distribution of the nanodots is irregular, implying an enhancement of j_c in a wide range of applied magnetic field, not only up to a matching field.

YBCO films grown on the annealed CeO_2 buffer layers reveal [6] only one epitxial orientation with YBCO(001)‖CeO_2(001)‖Al_2O_3($1\underline{1}02$) & YBCO [110]‖CeO_2[100]‖Al_2O_3[$11\underline{2}0$]. The 200-nm-thick YBCO film on the annealed CeO_2 buffer layer showed a superior j_c compared with that for the film on the as-grown CeO_2 buffer layer. Figure 2 shows the field dependence of j_c of these two typical YBCO films at various temperatures (20, 60 and 77.3 K) in applied magnetic field up to 8 T. The applied field was aligned in the configuration of H//c-axis of YBCO. For the film on the annealed CeO_2 buffer layer, j_c of 3.05×10^6 A/cm^2 in zero magnetic field and at 77.3 K was obtained. At 40 K, j_c reaches 2.24×10^7 A/cm^2 and is reduced monotonically to 3.36×10^6 A/cm^2 under the magnetic field of 8 T. For the film on the as-grown CeO_2 buffer layer, j_c is about 7.34×10^5 A/cm^2 in zero magnetic field and at 77.3 K and increased to 9.28×10^6 A/cm^2 at 40 K. At 77.3 K, a j_c value of ~ 10^3 A/cm^2 was maintained even at 8 T for the film on the annealed CeO_2 buffer layer, which is superior to that for the film on the as-grown layer, j_c vanished at 4 T.

Figure 3 shows the field-angle dependence of j_c at 60 K for the films deposited on R-cut sapphires with the as-grown and the annealed CeO_2 buffer layers. Extremely high j_c values are observed when the applied magnetic field is aligned parallel to the CuO planes. j_c peaks can be seen at 90° and 270° and are partly asymmetric. Slightly higher j_c values are obtained if the Lorentz force $\boldsymbol{F} = \boldsymbol{j_c} \times \boldsymbol{B}$ is pointing to the film substrate interface, which is consistent with the previous report [9]. The intrinsic pinning mechanism [10] is mainly responsible for the high and sharp j_c peaks in the H//a-b plane case. The CuO plane distance is comparable to the coherence length along the c-axis. The maximal pinning is given if the flux line is aligned exactly

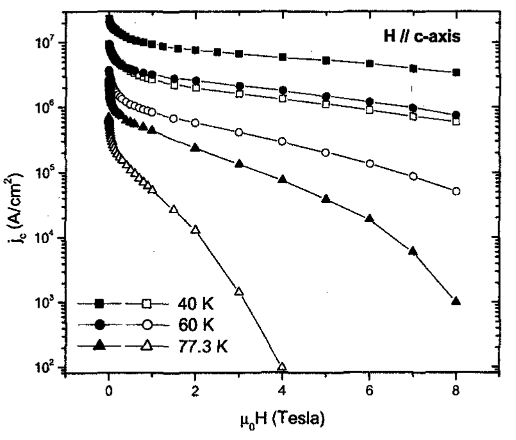

Figure 2. Critical current density j_c as a function of applied magnetic field (H) at various temperatures for two typical 200-nm-thick YBCO films on R-cut sapphire with a 36-nm-thck annealed (solid) and as-grown (open) CeO_2 buffer layer, respectively.

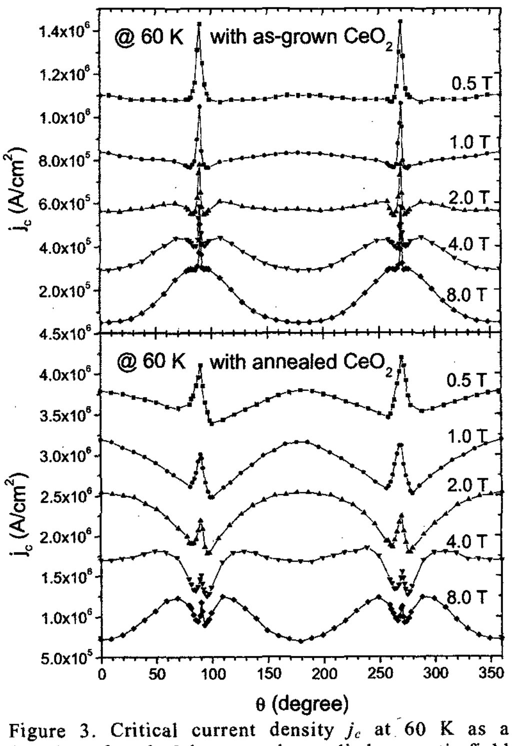

Figure 3. Critical current density j_c at 60 K as a function of angle θ between the applied magnetic field (H) and the film normal. Two typical films deposited on R-cut sapphires with the as-grown and the annealed CeO_2 buffer layers were measured and compared.

parallel to the a-b plane of YBCO, thus interacting with the weakly superconducting layers between the CuO planes along the whole flux-line length. When the flux-line direction deviates by only a few degrees, they intersect these planes only at discrete points.

Apart from the intrinsic peaks, no second maximum has been observed for the film deposited on the as-grown CeO_2 buffer layer. For the film deposited on the annealed CeO_2 buffer layer, however, a high and relatively broad j_c maximum is found for $\theta = 0°$ and 180° when the applied field is parallel to the *c*-axis of YBCO. With increase of the applied magnetic field (> 1 T), the *c*-axis correlated peaks even exceed the intrinsic peaks and then were suppressed with further increase of the magnetic field (> 4 T). Two mechanisms might be responsible for the strong pinning in the film with annealed CeO_2 buffer layer in the *H//c*-axis case. One is the extended defects in line with the *c*-axis, such as dislocations enhanced by the CeO_2 nanodots. We do not ascribe the c-axis correlated j_c peaks to twins, because for both films the twin planes are visible as proved by plan-view transmission electron microscopy (data not shown). The second effect is the high density precipitates with sizes ranging from 20-50 nm, which are egg-shaped and elongated along c-axis of YBCO, most of them were found to be correlated to the presence of the CeO_2 nanodots at the CeO_2-YBCO interface. The broadening of the c-axis correlated j_c peaks is also consistent with the quasi-anisotropy or the misalignment of the precipitates, which are formed spontaneously during the growth of the film. On the other hand, the formation of misaligned YBCO could possibly be attributed to the high deposition rate (~ 0.18 nm s^{-1}) employed in the growth of YBCO films. Further details of microstructure of the YBCO films will be published elsewhere.

4. CONCLUSION

In conclusion, experimental evidence of an atomically flat CeO_2 surface with a high density of nanodots (formed by a cooperative self-assembly process) was obtained. The resultant YBCO films grown on such buffered sapphire substrates had a substantial enhancement in $j_c > 3.0\times10^6$ A/cm^2 (at 77.3 K and 0 T). Magnetic field and angular dependence of j_c at various temperatures were investigated. YBCO film with the annealed CeO_2 buffer layer showed a high j_c peaks when the magnetic field was aligned along the *c*-axis of YBCO. This j_c peak is considered to be caused by strongly *c*-axis correlated pinning sites, possibly due to the formation of CeO_2 nanodots on top the CeO_2 wetting layer. This simple method of high temperature annealing of the buffer layers to obtain YBCO films with high j_c is of interest for electronic applications.

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REFERENCES

[1] B. Gromoll, G. Ries, W. Schmidt, H. -P. Kraemer, B. Seebacher, B. Utz, R. Nies, H. –W. Neumeueller, E. Baltzer, S. Fischer, B. Heismann, *IEEE Trans. Appl. Supercond.* **9**, 656 (1999).
[2] X. Wu, R. C. Dye, R. E. Muenchausen, S. R. Foltyn, M. Maley, A. D. Rollett, A. R. Garcia, and N. S. Nogar, *Appl. Phys. Lett.* **58**, 65 (1991).
[3] M. Maul, B. Schulte, P. Häussler, G. Frank, T. Steinborn, H. Fuess, and H. Adrian, *J. Appl. Phys.* **74**, 2942 (1993).
[4] D Dimos, P. Chaudhari, J. Mannhart, and F. K. LeGroves, *Phys. Rev. Lett.* **61**, 1653 (1988).
[5] J. H. Lee, W. I. Yang, H. J. Kwon, V. A. Komashko, and S. Y. Lee, *Supercond. Sci. Technol.* **13**, 989 (2000).
[6] J. C. Nie, H. Yamasaki, H. Yamada, Y. Nakagawa, and K.Develos-Bagarinao,*Supercond. Sci. Technol.* **16**, 768 (2003).
[7] A. Crisan, P. Badica, S. Fijiwara, J. C. Nie, A. Sundaresan, Y. Tanaka and H. Ihara, *Appl. Phys. Lett.* **80**, 3566 (2002).
[8] A. Crisan, S. Fijiwara, J. C. Nie, A. Sundaresan, and H. Ihara, *Appl. Phys. Lett.* **79**, 4547 (2001).
[9] B. Roas, L. Schultz, and G. Saemann-Ischenko, *Phys. Rev. Lett.* **64**, 479 (1990).
[10] M. Tachiki, S. Takahashi, *Solid State Com.* **70**, 291 (1989).

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