Features and Properties of $YBa_2Cu_3O_{7-x}$ Films Grown on $SrTiO_3$ by High Frequency PLD

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Abstract

YBCO films were deposited with various thicknesses from 100nm to 1.6μm on single crystal $SrTiO_3$ substrates by pulsed laser deposition (PLD). The effects of different deposition conditions, especially different deposition rates by means of changing the pulsed laser frequency up to 200Hz, on the J_c value were studied. For YBCO film with the thickness of 200nm, the J_c value of 2.1MA/cm^2 has been achieved under the high deposition rate of 3.2nm/s (190nm/min). The J_c can be maintained greater than 1M/cm^2 with the thickness less than 1μm. The X-ray analysis was used to examine the texture, crystallization and surface quality. The SEM was employed to analyze the surface of YBCO, and it was shown the surface of YBCO film became rougher with increasing the thickness. There were many large singular outgrowths and networks of outgrowths on the surface of YBCO films which lowered the density of thick YBCO film. The outgrowth network was probably the a-axis YBCO corresponding to XRD θ-2θ scan and χ-scan which were used to characterize a-axis orientation of YBCO film. The reason for J_c declining with increasing the thickness was studied and discussed.

Keywords : pulsed laser deposition, YBCO film, critical current density, texture

I. Introduction

Over the last several years significant efforts have been made to prepare long length YBCO coated conductors which is also known as the second generation high temperature superconducting (HTS) tape. The fast deposition of YBCO films greater than 1 μm in thickness for obtaining high critical current (I_c) through maintaining high critical current density (J_c) has been a critical issue in producing high quality coated conductor for practical application. However, fast growth rates can lead to the crystalline disorder, poor crystallinity and possible formation of metastable phases during deposition, with reduction in J_c [1]. Meanwhile, there are many difficulties in growing high quality YBCO film whose superconducting properties do not degrade with increasing film thickness. Typical problems in producing high quality thick YBCO films can include the development of misoriented YBCO grains or porosity, formation of non-superconducting phases, oxygen deficiencies, and changes in stoichiometry [2-4].

High quality YBCO films (with excellent values of T_c, J_c, surface resistance) have been routinely obtained using the PLD technique. Many studies have been carried out to optimize the deposition parameters of the PLD process for YBCO films, including the substrate temperature, the deposition rate, the laser energy, and the deposition pressure [5, 6].

Many studies on the relationship between the

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thickness/deposition rate and the properties of YBCO have been reported [1,7]. In this work, we studied the effects of the deposition rate and the thickness on the critical current density of YBCO films. The features and the properties of thick YBCO films were studied by XRD and SEM.

II. Experiment

A stoichiometric YBCO target of 2 inch diameter was ablated by an exicmer KrF pulsed laser (LPX220i with wavelength 248nm from Lambda Physik). The 3 × 10 and 3 × 20 mm^2 single crystal $SrTiO_3$ (STO) substrates were attached with a silver paste on the target holder (also the heater) which was directly facing the target.

The particulars of the deposition system were: fixed laser beam at an angle of 60° to the normal of target; target-substrate distance 65mm; target rotation 25rpm; background pressure 1×10^{-6} Torr. The deposition conditions were: laser repetition rate 40~200Hz; the size of the laser spot on target ~ 5 × 1 mm^2; and the pulsed laser energy density on the target ~ 2 J/cm^2. During deposition the heater temperature was 830~850°C, and the oxygen pressure in the chamber was 300mTorr. Following deposition, the YBCO film was quickly cooled to 550°C under deposition pressure, and then kept for 20 min under oxygen pressure of 500 Torr. All thickness values of the films used in this paper were measured using a stylus profilometer. All the values of J_c reported in this study were calculated from the four-point probe measurements of I_c conducted at 77K in self-field without patterning.

The X-ray diffraction system of D8 DISCOVER with GADDS (general area detector diffraction solution) from Bruker was used to analyze the orientation of films. XRD θ-2θ scan, ω-scan, ϕ-scan and χ-scan have been done with sample oscillation using a 1/4-circle Eulerian cradle xyz stage. SEM was used to investigate the surface morphology, microstructure, and defects in the YBCO films.

III. Results and discussion

A. Relationship between critical current density and deposition rate for the thin YBCO films

In PLD process, two concepts of the deposition rates need to be considered, i.e. instantaneous and average deposition rates. Since the PLD occurs in a pulsed mode, the pulse time (~20ns) compared with interval time (~10 – 100 ms) is very small. Thereby with each pulse there is a very high instantaneous deposition rate (~10^4Å/s) [4]. If the laser energy density is kept constant, the instantaneous deposition rate is not changed. But by varying the repetition rate, the average deposition rate can be controlled. In this paper, deposition rate was varied by adjusting laser pulse frequency and was determined by dividing the total film thickness by deposition time. This gave an average rate for film deposition.

Two sets of YBCO films have been deposited under similar conditions except the frequency of pulsed laser and thickness. One group was YBCO films with 120 nm thickness, and the other was those with 240 nm thickness. Fig. 1 shows the J_c values of these samples. For both group samples, the J_cs of YBCO films decreased as the repetition rate was increased from 40Hz to 200Hz, meanwhile the deposition rate increased from 0.56nm/s (33nm/min) to 2.87nm/s (170nm/min). The J_cs of YBCO films with 120nm thickness were greater than those

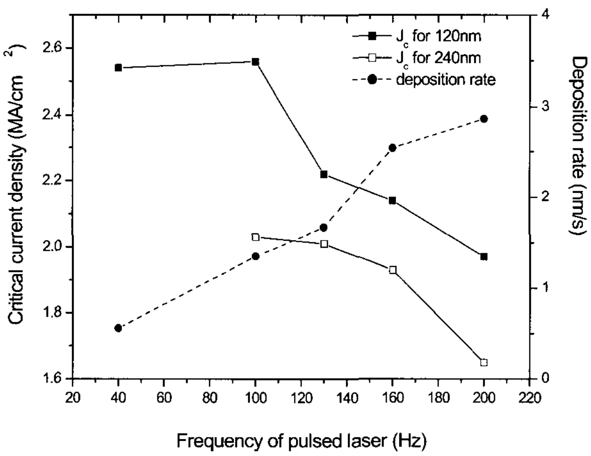

Fig. 1. The relationship between the critical current density of YBCO films, the pulsed laser frequency, and the deposition rate of PLD.

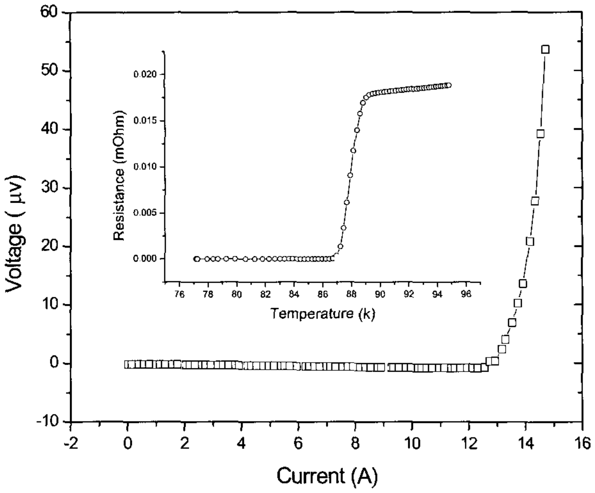

Fig. 2. I_c and T_c measurements for a 200nm YBCO film deposited at the rate of 3.2nm/s.

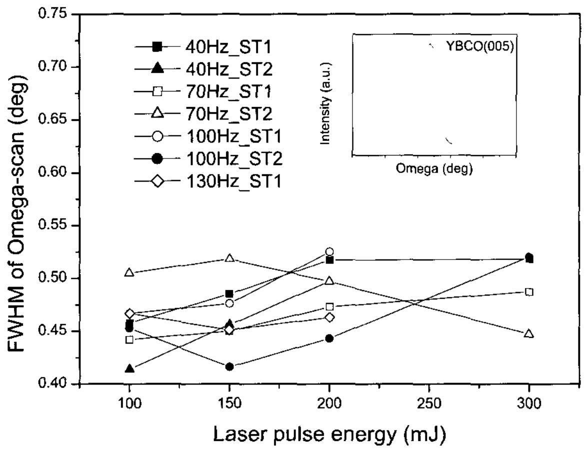

Fig. 3. FWHM values of XRD ω-scans of YBCO films with 120nm thickness deposited at different repetition rates and different laser energies (ST1: $3\times10mm^2$ substrate; ST2: $3\times20mm^2$ substrate)

deposited at the same frequency with 240nm thickness, and all of the J_cs of these two group of samples were greater than $1.5MA/cm^2$. The maximum value of J_c of YBCO film with 200nm thickness we have obtained was $2.1MA/cm^2$ under the high deposition rate of 3.2nm/s (190nm/min) (this value was not included in Fig. 1). Fig. 2. is the measurement plots of I_c and T_c of this sample at 77K in self field. The values of the full width at half maximum (FWHM) of XRD ω-scan for (005) peak of 120nm YBCO films deposited at different repetition rates and different laser energies are in the range of 0.41°-0.53° (shown in Fig. 3) and ϕ-scan for (103) plane were about 1.50°. The amount of c-axis component of YBCO was about 100% characterized by X-ray χ-scan. All of these mean there is no significant difference in the crystallinity of these thin films. However, the growth process can be influenced directly by the deposition rate. At low deposition rate which has a long interval time, adatoms are allowed sufficient time to migrate to some of the local free energy minimum, thus, perfect c-axis oriented orthorhombic YBCO domains with long-range ordering of Y and Ba atoms will be formed. However, at high deposition rate which has very short interval time, the encounter probability between migrating adatoms on substrate surface is very high. Therefore, not sufficiently c-axis oriented orthorhombic YBCO domains with short-range ordering of Y and Ba atoms will be formed, which may lead to the declination of Jc.

B. Properties of YBCO films with different thicknesses

A set of YBCO films, which were deposited at 200Hz, 850°C, and 300mTorr oxygen, with thickness range of 100 nm~1.6 μm were studied.

The relationship between J_c and thickness for these YBCO films is shown in Fig. 4. The deposition rate of these films was about 2.9 nm/s (170 nm/min). The J_c was decreased from 3.0 to 0.69 MA/cm^2 while the thickness was increased from 0.1 μm to 1.6 μm.

XRD ω-scan and ϕ-scan were used to examine the orientation of YBCO films. The FWHM value of ω-scan for (005) peak was increased from 0.45° at the thickness of 0.22 μm to 0.63° at the thickness of 1.6 μm, which suggests an increased dispersion of c-axis mosaic of YBCO grains, possibly increasing gradually from the bottom to the top of the YBCO film. Furthermore, the FWHM value of XRD ϕ-scan for (103) plane was about 1.5° for the thickness less than 0.4 μm, and was about 1.9° for those thicker than 0.6 μm (shown in Fig. 5), indicating that there is some deterioration in the in-plane texture of YBCO films as film thickness increases from 0.6 μm to 1.6 μm. These deteriorations of crystallinity are one of the reasons for the degradation of superconductivity

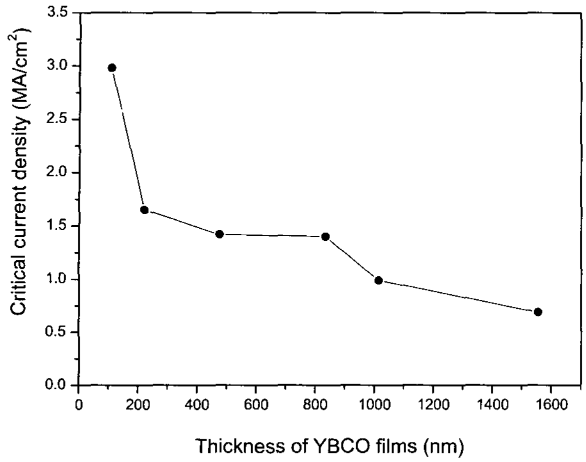

Fig. 4. The relationship between critical current density and thickness for YBCO films.

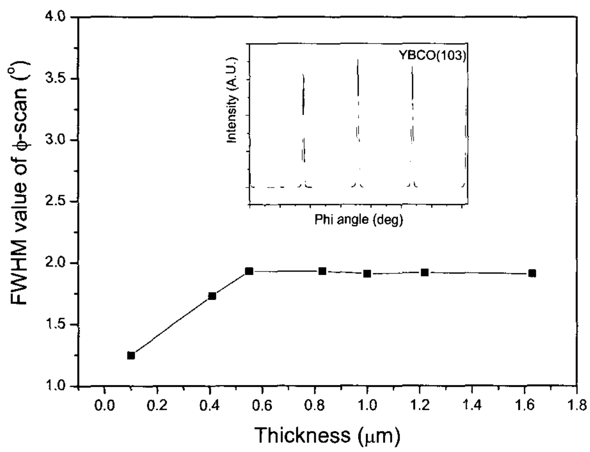

Fig. 5. FWHM values of XRD ϕ-scans of YBCO films with different thicknesses from 0.1μm to 1.6μm.

of YBCO films.

Fig. 6 (A) and (B) show the SEM images of YBCO films with the thickness of 1.6 μm. In this case, the surface YBCO film has large singular and rectangular outgrowths perpendicular to each other to form an outgrowth network. From Fig. 6(A), it can be seen the outgrowth networks are overlapped with each other, and some cavities are left between the layers, which surely lower the density of YBCO films. XRD χ-scan was used to examine the a-axis part of YBCO films. Fig. 7 shows the χ-scans for (102) plane of 1.6μm YBCO film. There is plenty of a-axis YBCO corresponding to the outgrowth

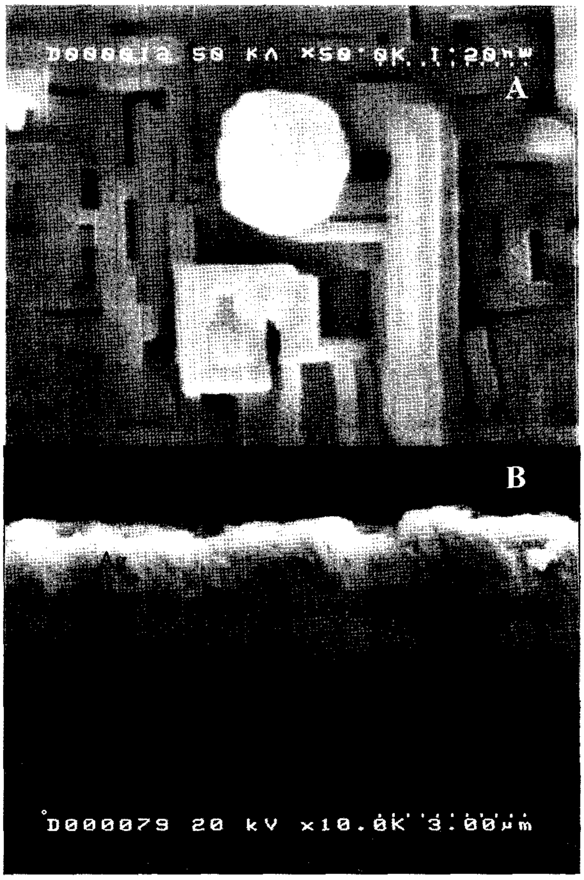

Fig. 6. SEM images of YBCO film with 1.6μm thickness; (A) surface image, (B) cross-section image.

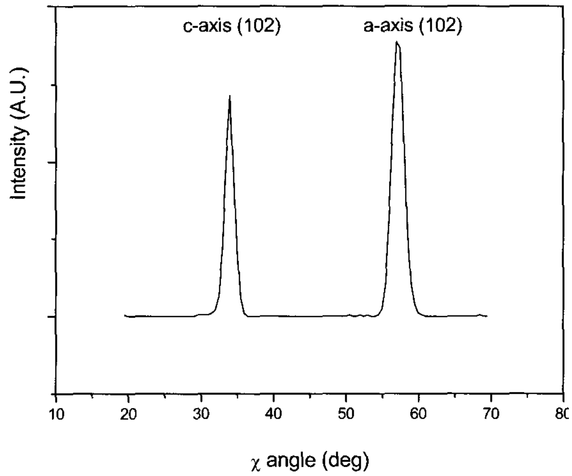

Fig. 7. XRD χ-scan for (102) plane of YBCO film with1.6 μm thickness.

networks on the YBCO surface. In the SEM cross-section image (Fig.6(B)), it can be seen the surface of YBCO film is very rough. Sum up the above analysis, The deterioration of crystallinity, the lower density of YBCO film and a-axis YBCO part caused the degradation of J_c value of the YBCO film with the thickness over 1μm.

IV. Conclusion

The YBCO films deposited by PLD at different deposition rate and with different thickness were investigated by XRD and SEM techniques in this work. The J_c of YBCO film was decreased as the deposition rate was increased. For YBCO film with the thickness of 200nm, the J_c value of 2.1MA/cm^2 has been achieved under the high deposition rate of 3.2nm/s (190nm/min). The J_c can be maintained greater than 1MA/cm^2 with the thickness less than 1μm. The reason for J_c degradation with increasing the thickness was studied and discussed. Through the SEM analyses for the surfaces of YBCO films, it was found that the surface of YBCO film became rougher and void with increasing the thickness. The outgrowth network on the surface of 1.6μm YBCO film was probably a-axis YBCO which corresponds to the XRD χ-scan which was used to determine the amount of the a-axis oriented YBCO component of thick YBCO film. The increased a-axis YBCO component, the lowered density and the deterioration of crystallinity of thick YBCO film most probably caused the degradation of the J_c with increasing the thickness.

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References

[1] A. Berenov, A. Purnell, A. Zhukov, N. Malde, Y. Bugoslavsky, L. F. Cohen, J. L. MacManus-Driscoll, S. J. Foltyn and P. Dowden, "Microstructural characterization of high J_c, YBCO thick films grown at very high rates and high temperatures by PLD", Physica C 372-376, 683-686 (2002).

[2] J. H. Park and S.Y. Lee, "Orientation transition in a thick laser deposited $YBa_2Cu_3O_{7-x}$ film observed by glancing angle X-ray diffraction", Physica C 314, 112-116 (1999).

[3] K. D. Develos, H. Yamasaki, A, sawa, Y. Nakagawa, S. Oshima and M. Mukaida, "Microstructure of $YBa_2Cu_3O_{7-y}$ films on CeO_2-buffered Al_2O_3", Physica C 357, 1353-1357 (2001).

[4] S. R. Foltyn, E. J. Peterson, J. Y. Coulter, P. N. Arendt, Q. X. Jia, P. C. Dowden, M. P. Maley, X. D. Wu and D. E. Peterson, "Influence of deposition rate on the properties of thick $YBa_2Cu_3O_{7-\delta}$ films", J. Mater. Res. 12, 2941-2946 (1997).

[5] M. Lorenz, H. Hochmuth, D. Natusch, H. Borner, G. Lippold, K. Kreher and W. Schmitz, "Large-area double-side pulsed laser deposition of $YBa_2Cu_3O_{7-x}$ thin films on 3-in. sapphire wafers", Appl. Phys. Lett. 68, 3332-3334 (1996).

[6] Douglas B. Chrisey and Arun Inam, "Pulsed Laser Deposition of High T c SuperconductingThin Films for Electronic Device Applications", MRSBULLETIN 17, 37-43 (1992).

[7] Mikhail Strikovski and John H. Miller, Jr., "Pulsed laser deposition of oxides: Why the optimum rate is about 1 Å per pulse", Appl. Phys. Lett. 73, 1733-1735 (1998).