The efficient production of MOCVD-derived double-sided 2-in YBCO thin films

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Abstract

A rotating double-sided deposition device was designed and applied in our home-made MOCVD system to efficiently prepare double-sided YBCO thin films. With it, at most 12 pieces of double-sided 2-in YBCO thin films could be prepared simultaneously. In this paper, 500 nm thick YBCO thin films were simultaneously deposited on both sides of 2-in $LaAlO_3$ single crystalline substrates at an average preparation rate of 23 min per piece. The YBCO thin films made in this way maintained good consistency both in-plane and double-sided. Meanwhile, the critical density has reached 2.4 MA • cm^{-2} (77K,0T), and the microwave surface resistance is 0.323 mΩ (77K,10GHz) that has meet the commercial demand of microwave devices.

Keywords: MOCVD YBCO double-sided efficient

1.Introduction

$YBa_2Cu_3O_{7\text{-delta}}$ (YBCO) thin films on single crystal ($LaAlO_3$, MgO, sapphire) has been widely used in the field of microwave devices, because of its excellent property of high current capacity and low microwave losses[1-10]. The microwave filter on double-sided high temperature superconductor (HTSC) YBCO thin films has much lower insertion loss and stronger anti-interference capability compared to traditional filters[11-14]. At first, many groups prepared double-sided YBCO thin films by depositing the second side after the first side is deposited[15-17], which has negative impacts on the double-sided consistency. Gradually, more and more researchers have been adopting simultaneous deposition route to prepare double-sided YBCO thin films and has made progress[18-20].

Although, double-sided YBCO thin films with good performance had been prepared successfully all over the world as early as more than ten years ago[18-22], the price is still expensive due not only to the high cost of preparation, but also to the low preparation efficiency. Nowadays, the main preparation methods of YBCO thin films on single crystal include evaporation[16, 21], sputtering[18, 19], pulsed laser deposition (PLD)[9, 20] and metal organic deposition (MOD)[22-24]. Evaporation method can be used to deposit single-sided YBCO thin films rapidly, but it cannot be used to simultaneously deposit double-sided thin films due to its dependence on gravity. The traditional sputtering method, of which the depositing rate is only about several nm min^{-1}, usually takes 13 hours to prepare a piece of 500 nm thick double-sided YBCO thin films and the PLD method takes 5 hours to make it. As for MOD method, just the decomposition process takes approximately 10 hours.

In recent years, metal organic chemical vapor deposition (MOCVD) method performs well in the preparation of YBCO high temperature superconducting (HTS) tapes[25-28], so we transplanted it into the preparation of YBCO thin films on single crystal. As is reported, the deposition rate of YBCO thin films on tapes has reached up to 1 μm·min^{-1}[25]. However, during the actual preparation process, the pre-preparation, such as heating and preparation of the precursor solution, and post-annealing costs lots of time that cannot be ignored. The efficiency of the preparation of YBCO thin films would greatly enhanced if multiply pieces of YBCO thin films rather than only one were deposited at one time.

In the paper, a rotating double-sided deposition device, which contained a planetary turntable for substrates and a shower having two opposite symmetrical narrow slits, was designed to prepare multiply pieces of double-sided YBCO thin films simultaneously and efficiently. The planetary turntable having 12 circumferentially shaped openings, in which substrate wafers were maintained, was applied to deposit YBCO thin films on multiply pieces of LaAlO3 substrate wafers. With it, at most 12 pieces of YBCO thin films could be deposited at one time, significantly shortening the preparation progress of each YBCO thin films. And the shower having two opposite symmetrical narrow slits was used to deposit YBCO thin films on both sides of a LAO wafer simultaneously. In this way, the efficient preparation of MOCVD-derived double-sided YBCO thin films was realized.

2.Experiment

In our experiment, 2-in LaAlO3 (LAO) single crystal wafers were used as substrates for YBCO thin films. Before depositing thin films, the deposition chamber was heated to 845 ℃, while the evaporation chamber and transport pipeline were heated to 300 ℃. Meanwhile, $Y(tmhd)_3$, $Ba(thmd)_2$·(1,10-heptanedionate), $Cu(thmd)_2$ were dissolved into tetrahydrofuran solution in a proper proportion, where 'thmd' is the abbreviation of 2,2,6,6, -tetramethy-3,5-heptanedioline. The pre-preparation for the deposition of YBCO thin films took about 140 minutes. Figure 1 shows the schematic diagram of the rotating double-sided deposition device for the efficient preparation of double-sided YBCO thin films. As is shown in figure 1(a), the planetary turntable has 12 circumferentially shaped openings which are used to place the LAO wafers. The diameter of the openings was a little bigger than 2 inches (the diameter of LAO substrates). Upon rotation of the substrate turntable, the substrates self-rotated within each opening due to the force of friction between the substrates and the openings. This will significantly reduce the variation of thickness in the plane. In figure 1(b), a shower having two opposite symmetrical narrow slits was designed to deposit double-sided YBCO thin films simultaneously. The position relationship of the planetary turntable and the shower is shown in figure 1(c). During depositing, the precursor solution was evaporated to from vapor in the evaporator. Then the vapor was mixed up with the nitrous oxide, oxygen, argon in a proper gas flow ratio, and was transferred into the depositing area from the two narrow slits. Along with the rotation of the planetary turntable, YBCO thin films was deposited on both side of every LAO wafer uniformly. In order to obtain 500 nm thick YBCO double-sided thin films, the deposition was set for 75 minutes. Next, the temperature of the deposition chamber was reduced to 500 ℃ by adjusting heating current manually. After annealing for 20 minutes in pure oxygen, the YBCO thin films were cooled to a temperature below 100℃ slowly. On average, the post processing took about 60 minutes.

The thickness of YBCO thin films was measured by a step profiler (Veeco Dektak 150). The critical current density (Jc) was measured through the Leizig Jc-scan system and the microware surface resistant was measured through a sapphire resonance[29]. The texture was characterized by an x-ray diffraction system (XRD, DanDong DX-2700) with θ-2θ scan for crystal phase and orientation and ω-scan for out-of-plane orientation The surface morphology was characterized by a scanning electronic (SEM, JEOL7500F). The composition was characterized by an energy dispersive spectrometer (EDS, Oxford INCA).

3.Results and discussion

3.1 The average preparation rate of double-sided YBCO thin films on LAO substrates

Due to the planetary turntable having 12 openings for substrates and the shower having two opposite symmetrical narrow slits, up to 12 pieces of 2-in double-sided YBCO thin films can be prepared in a single experiment. Thus, the average preparation time of pre piece of double-sided YBCO thin films ($\overline{R}_{pre}$) can be calculated by the formula:

$$\overline{R}_{pre} = \frac{T}{12} , \tag{1}$$

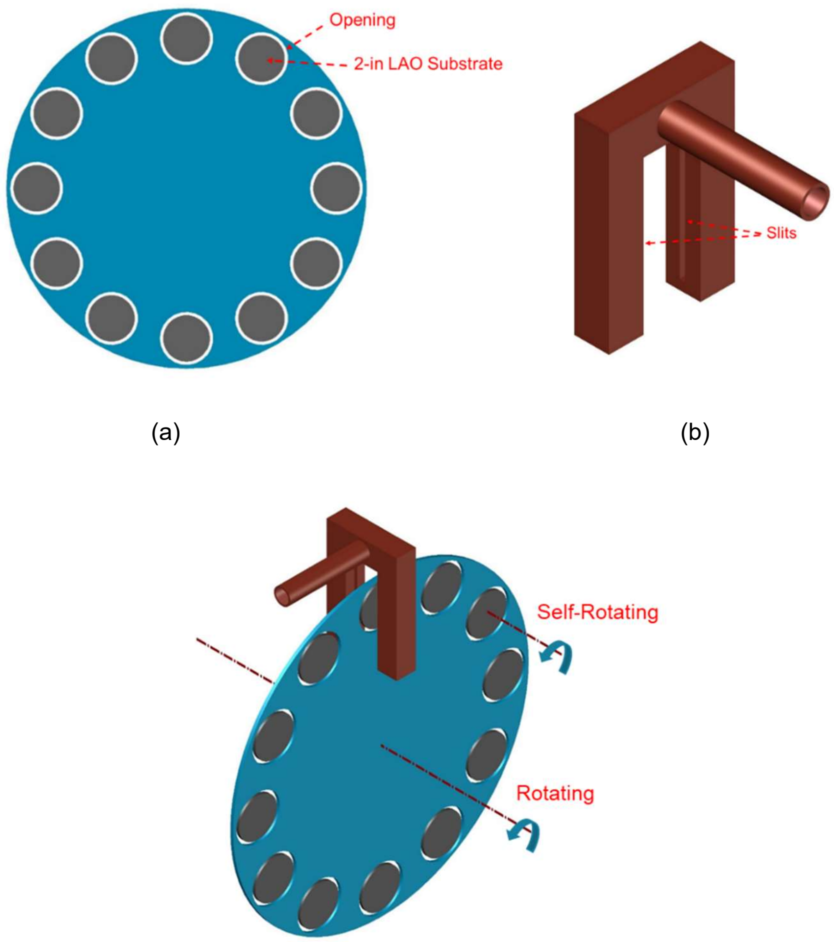
where T is the total time of a single experiment. It could be divided into the pre-preparation time (T_{pre}), depositing time(T_{dep}) and post processing time (T_{pos}) as is described in chapter 2. The T_{pre} which includes the heating of reaction chamber, evaporation chamber and pipes as well as preparation of precursor solution is about 140 minutes. The T_{dep} depends on the thickness of the required thin films, such as 75 min for 500 nm thick thin films. And the T_{pos} is usually 60min.Then, the T can be obtained by the calculation of the formula:

$$T = T_{pre} + T_{dep} + T_{pos} , \tag{2}$$

The calculated results are shown in table 1. Thus, the average preparation rate ($\overline{R}_{pre}$) of pre piece of double-sided 500nm thick YBCO thin films, which is calculated according to the measured results of experiments, is as fast as around 23 min per piece.

Table 1 Time consumption and the average preparation time of pre piece of double-sided 500nm thick YBCO thin films in a single experiment

Pre-preparation time (min)	Deposition time for 500 nm thick thin films (min)	Post preparation time (min)	Total time in a single experiment (min)	Average preparation rate (min pre piece)
140	75	60	275	23



(c)

Figure 1. The rotating double-sided deposition device: (a) the planetary turntable; (b) the shower having two opposite symmetrical narrow slits; (c) the position relationship of the planetary and the shower

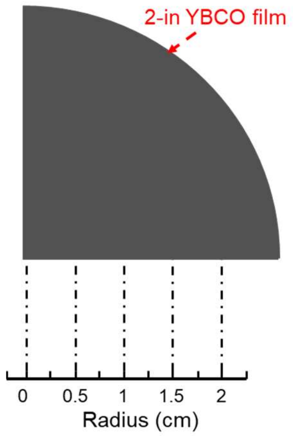


Figure 2. The testing radii of 2-in YBCO thin films

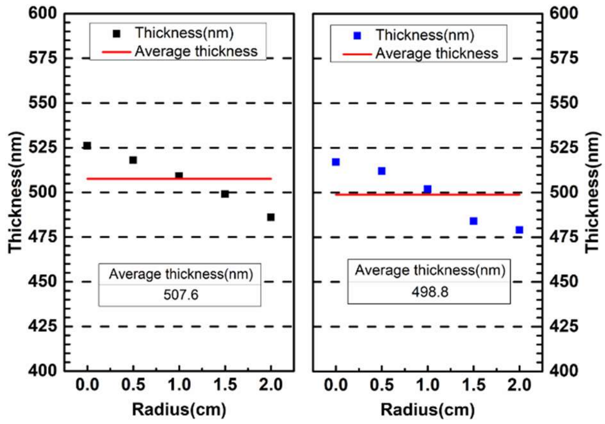


Figure 3. The variation of thickness of double-sided YBCO thin films at radii of 0, 0.5, 1, 1,5 ,2 cm

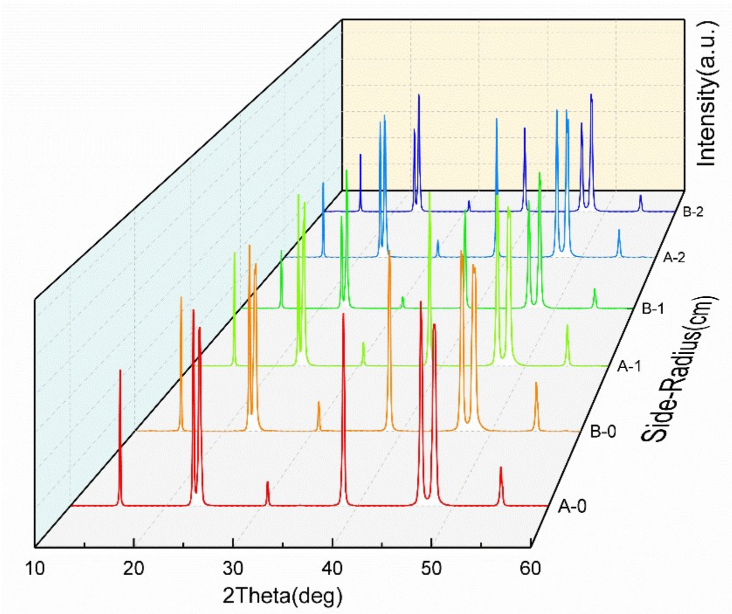


Figure 4. The XRD θ-2θ scanning patterns of double-sided YBCO thin films at radii of 0, 1, 2 cm of side A and B

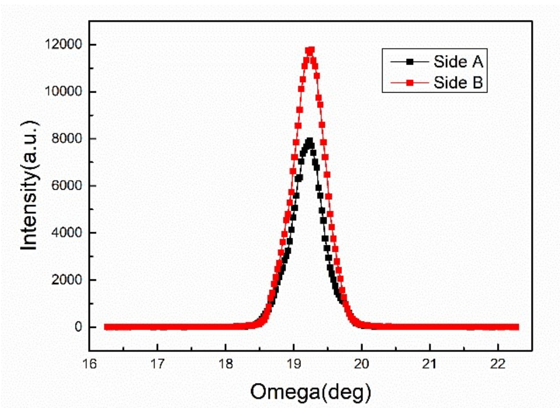


Figure 5. The XRD ω scanning patterns of double-side YBCO thin films

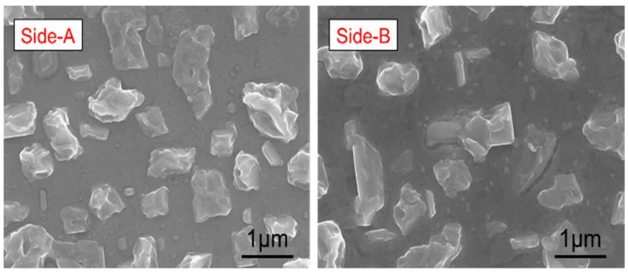
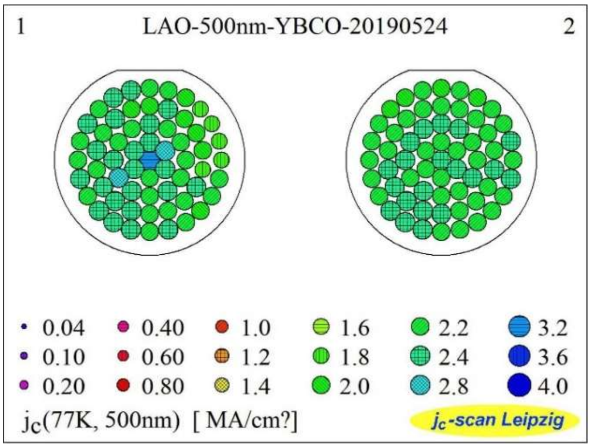


Figure 6. The SEM images of double sides of the YBCO thin films



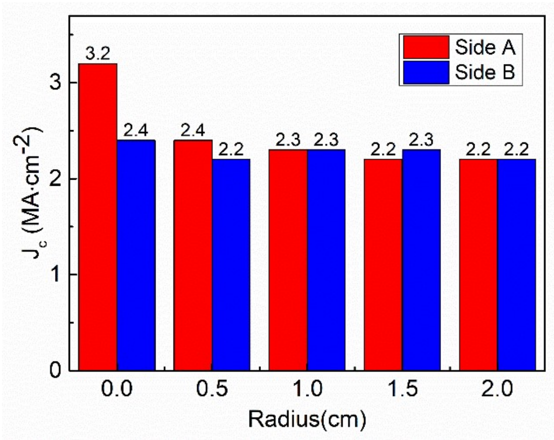


Figure 7. Homogeneity of the critical current density (77K,0T) of a piece of 2-in double-sided YBCO thin films on LAO substrate

Figure 8. Transformation of the critical current density (77K, 0T) at radii of 0, 0.5, 1, 1.5 ,2 cm on both sides of a 2-in YBCO thin films on LAO substrate

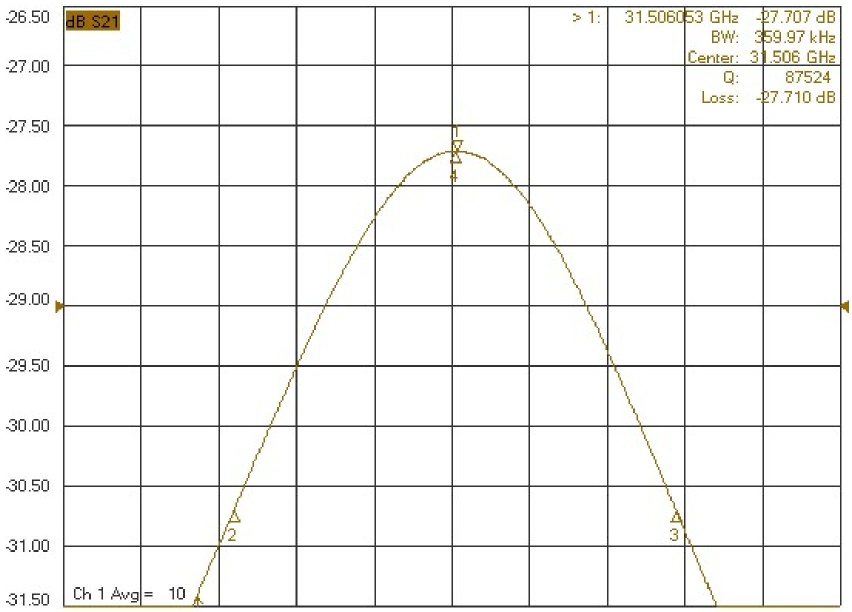


Figure 9. The microwave surface resistance at the center of the 2-in 500nm thick YBCO films

Table 2. The calculation results of the microwave surface resistance of the 500nm thick YBCO films at 77K

Q	*A*	*B*(mΩ $^{-1}$)	f_0(GHz)	*f* (GHz)	R_{s0}(mΩ)
87524			10	31.056	0.323

3.2 The measurement of the double-sided YBCO thin films

The thickness of both sides of the 2-in YBCO thin films was measured by a step profiler. As is shown in figure 3, on each side, radii of 0, 0.5, 1, 1.5, 2 cm were selected for thickness measurement in order to test the consistency of double-sided YBCO thin films. According to the results in figure 4, the average thickness of the two sides are approximately 507.6 nm and 498.8 nm, respectively. Meanwhile, the deviation of thickness of each side is lower than 10%, which indicates that the double-sided YBCO thin films is of good consistency.

The XRD θ-2θ scan maps of double-sided YBCO thin films, which were measured at radii of 0, 1, 2 cm are shown in figure 5. Except the peaks of LAO at 23.4° and 47.9°, there are only YBCO *(00l)* peaks other than *(h00)* peaks in the θ-2θ curve, which indicates that almost all YBCO grains are c-axis-oriented as we expected.

The XRD ω-scan of the double-sided YBCO thin films, which were used to characterize the out-of-plane texture, were performed at the equivalent position of each side. As is shown in figure 5, the full width of half maximum (FWHM) of each side is 0.498° and 0.507° respectively that shows little difference between the two sides.

The SEM was used to characterize the surface morphologies of the equivalent position of each side, as is shown in figure 6. Scarcely any a-axis-oriented YBCO grains can be observed among the SEM images of both sides, which is consistent with the measured results of XRD. There are some outgrowths on each side, which were identified to be CuO and Cu-Y-O phases by the EDS. Thus, the morphologies of the two sides of the double-sided YBCO thin films are very similar.

Figure 7 shows the measured results of the critical current density (Jc) at 77K and 0 T of a piece of 2-in double-sided YBCO thin films on LAO substrate. The Jc (77K, 0T) at every point measured is between 2.2 MA cm^{-2} and 2.4 MA cm^{-2}. As is shown in figure 8, the variation of Jc along the radius of each side of a 2-in double-sided YBCO thin films is small and the distinction between the two sides is not obvious.

In order to obtain the microwave surface resistant (R_s), a sapphire resonance was used to measure the quality factor(Q) of the YBCO films firstly [29]. As shown in figure 9, the quality factor of 500nm YBCO films prepared is 87524 at the frequency of 31.506 GHz. The relationship between Q and Rs is as follow:

$$\mathrm{Q} = \frac{1}{A + B*R_s} , \tag{1}$$

where A, B are only related to the electromagnetic fields distribution and can be determined by calibration. Therefore, the microwave surface resistant can be calculated by the formula:

$$R_s = \frac{\frac{1}{Q} - A}{B}, \tag{4}$$

Meanwhile, the relationship of the microwave surface resistant of YBCO films and the operating frequency (f) is as follow:

$$R_s = k * f^2, \tag{5}$$

where k is a constant. Thus, the microwave surface resistant (R_{s0}) at the frequency of 10GHz (f_0) can be calculated by the formula:

$$R_{s0} = \frac{\left(\frac{1}{Q} - A\right)*f_0^2}{B*f^2}, \tag{6}$$

and the calculation results are shown in table 2. The microwave surface resistant at the frequency of 10GHz is as low as 0.323mΩ(77K,10GHz) which indicates that the YBCO thin films prepared has met the commercial demand of surface resistant of microwave devices.

4.Conclusion

In this paper, we presented a rotating double-sided deposition device in our home-made MOCVD system. Results indicated that the average preparation rate of pre piece of 2-in double-sided 500 nm thick YBCO thin films was as fast as 23 minutes. Meanwhile, the double-sided YBCO thin films maintained good consistency both in-plane and double-sided. And the critical current density of 500nm thick YBCO thin films with good biaxial texture reached 2.4 MA cm^{-2}(77K, 0T). Moreover, the microwave surface resistant was as low as 0.323mΩ(77K,10GHz) which met the commercial demand of microwave devices. Thus, it illuminated that our design to prepare double-sided YBCO thin films efficiently was feasible.

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