Superconductor Science and Technology

Batch production of large-area double-sided $YBa_2Cu_3O_{7-\delta}$ thin films by DC magnetron sputtering

IOP PUBLISHING SUPERCONDUCTOR SCIENCE AND TECHNOLOGY

Supercond. Sci. Technol. **22** (2009) 055019 (6pp) doi:10.1088/0953-2048/22/5/055019

Batch production of large-area double-sided $YBa_2Cu_3O_{7-\delta}$ thin films by DC magnetron sputtering

Fazhu Ding[1,3], Hongwei Gu[1,3], Tao Li[1], Jiangli Cao[2], Xudong Lv[1] and Yuegang Lei[1]

[1] Superconductor Research Center, General Research Institute for Non-ferrous Metals, Beijing 100088, People's Republic of China

[2] Advanced Materials and Technology Institute, University of Science and Technology Beijing, Beijing 100083, People's Republic of China

E-mail: dingfz2008@163.com and guhw@grinm.com

Received 12 January 2009, in final form 18 February 2009
Published 7 April 2009
Online at stacks.iop.org/SUST/22/055019

Abstract

Using a novel DC magnetron sputtering system, high quality $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films were deposited on both sides of twelve pieces of 3-inch $LaAlO_3(00l)$ single crystal substrates in one batch. This sputtering system has a rotatable disc sample holder with the $LaAlO_3$ substrates being positioned in the middle of upper and lower YBCO targets. These two sputter guns are arranged to simultaneously deposit YBCO thin films on both sides of the substrate. The rotatable sample holder is an important approach to large scale deposition of high quality YBCO thin films. In this manner, the sputtered species can diffuse well around the substrates during one rotation cycle. Furthermore, the anode is installed in the inner of rotatable disc sample holder to avoid negative ion bombardment. Thus, a good uniformity over the whole area for all the films has been realized. The full-width at half-maximum (FWHM) values of the (005) rocking curves typically range from 0.49° to 0.56°. The superconducting critical temperatures fall in the range of 89–90 K and the critical current densities are between 3.75 and 4.0 MA cm^{-2} at 77 K. The microwave surface resistance, R_S (77 K, 10 GHz), of the YBCO thin films range typically from 0.56 to 0.71 mΩ, which make these films suitable for microwave applications.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Epitaxial $YBa_2Cu_3O_{7-\delta}$ (YBCO) high temperature superconducting (HTS) thin films are considered to be prospective materials for practical applications due to their high critical current density and low microwave losses even at the liquid nitrogen temperatures. Microwave applications generally require large-area double-sided YBCO thin films with low microwave surface resistance. $LaAlO_3$ (LAO) exhibits a low loss tangent and a dielectric constant value which makes it a suitable substrate for development of microwave devices. Currently, there are a few successful methods of preparing large-area double-sided YBCO thin films on LAO substrates such as sputtering, pulsed laser deposition (PLD) [1], thermal co-evaporation (TCE) [2, 3], metalorganic chemical vapor deposition (MOCVD) [4] and metalorganic deposition (MOD) [5–7]. Among these processes, magnetron sputtering is a promising technique for YBCO thin film deposition because of its ease of process control and the repeatability of the resulting sample characteristics [8]. Many research groups have prepared double-sided YBCO coatings by sputtering. Müller *et al* [9] prepared double-sided YBCO coatings by breaking the vacuum after one side is deposited, before starting the deposition of the other side. In order to prepare double-sided YBCO thin films more efficiently, Liu *et al* [10] developed a new system in which a single inverted cylindrical sputter gun was arranged to simultaneously deposit YBCO thin films on both sides of the substrate. The substrate was rotated around the rod of the substrate holder.

[3] Authors to whom any correspondence should be addressed.

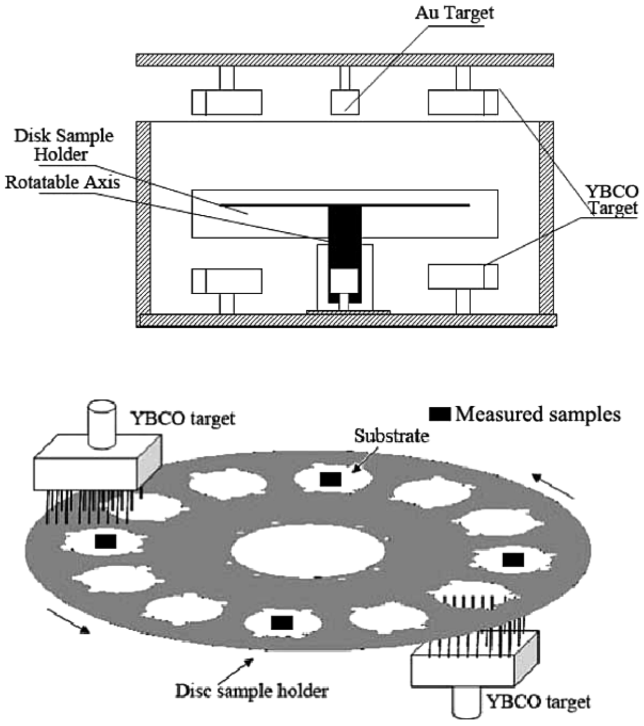

Figure 1. Schematic illustration of the apparatus used in this work.

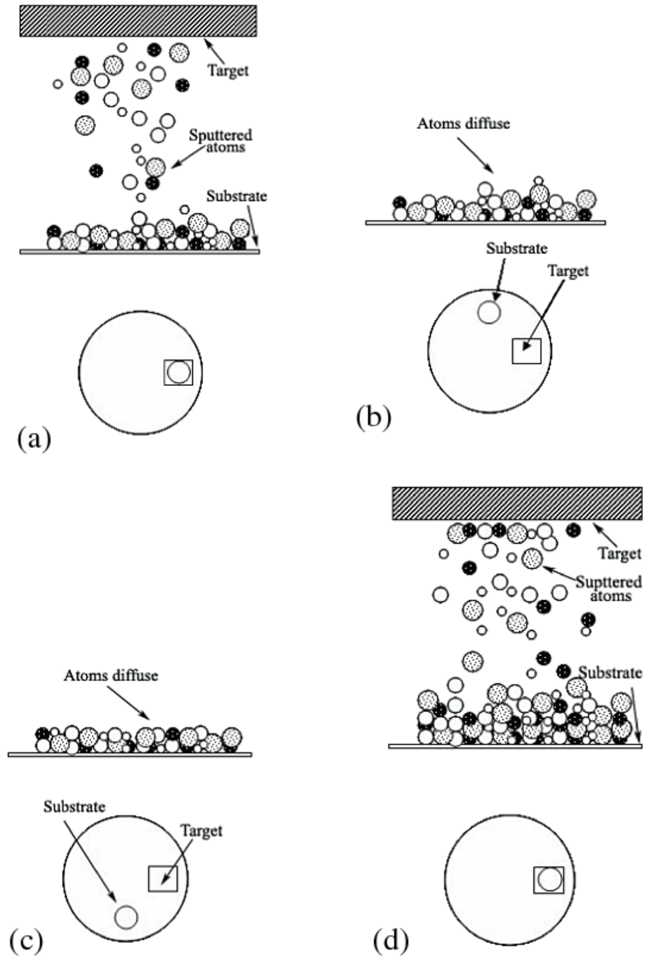

Figure 2. The whole deposition process of YBCO films in one cycle.

2-inch double-sided YBCO thin films were deposited by simultaneous sputtering from a single target. Roeck *et al* [11] also fabricated high quality large-area YBCO films on single crystal MgO by rotatable magnetron sputtering. They believe that the rotatable target is an important approach to the large scale fabrication of YBCO coated conductors. Though these research groups have prepared high quality large-area YBCO films, they can fabricate only one piece of YBCO film in one batch. However, in order to lower manufacturing costs, HTS microwave devices require high productivity, reproducibility and uniformity. Hence, it is necessary to develop a low-cost and easily scalable method, which holds out the promise for the production of large-area double-sided YBCO films by sputtering.

In the present work, a novel sputtering system for producing large-area double-sided YBCO thin films has been developed. Twelve pieces of 3-inch diameter double-sided YBCO thin films can be prepared by this method in one batch. The crystallization, uniformity and superconducting properties of these films are reported.

2. Experimental details

Figure 1 shows the schematic of the set-up used for deposition of double-sided YBCO thin films. The disc sample holder with 12 pieces of ϕ3-inch $LaAlO_3$ (LAO) (00*l*) substrate is positioned in the middle of two YBCO targets. The disc sample holder rotates around the rod of the substrate holder. Two sputter guns are arranged to simultaneously deposit YBCO thin films on both sides of the substrates. When the disc sample holder rotates, all the substrates can be deposited with YBCO films. The whole deposition process during one cycle is illustrated in the figure 2. For each cycle, the process can be divided into two steps according to the relative position between the targets and the samples. The sputtered species are first deposited on the substrate when the substrate is right under the YBCO target, as shown in figure 2(a). These sputtered materials then diffuse around on the substrate surface at high temperature in an oxidation atmosphere to form the Y-123 phase, as shown in figures 2(b) and (c). When the substrate returns to the sputtering window, the sputtered materials from the target are deposited again, as shown in figure 2(d).

Another problem encountered in YBCO thin film growth by sputtering is the negative ion bombardment effect. The negative ions bombard the growing films. It causes strong deviation of the thin film composition from stoichiometry as well as poor superconductivity and degradation of the crystal quality. To avoid resputtering, the anode was installed in the inner of the rotatable sample holder in our system so that the negative ions bombard the disc sample holder rather than the growing films.

The ceramic $Y_1Ba_2Cu_3O_{7-\delta}$ targets of 75 mm $\times$ 150 mm were prepared by solid-state reaction from mixed powders of Y_2O_3, $BaCO_3$, and CuO with an initial cation ratio of Y:Ba:Cu = 1:2:3. The LAO substrates were

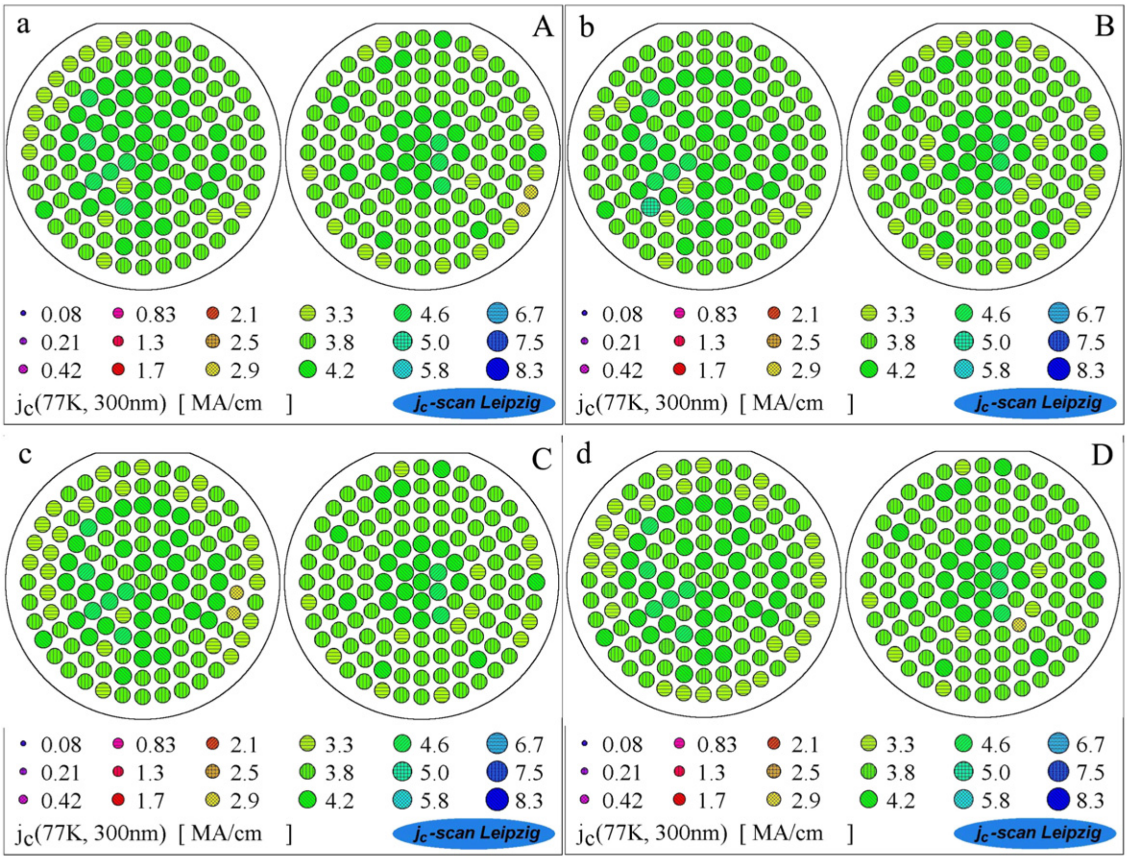

Figure 3. The J_C mappings of the 3-inch YBCO thin films as measured by J_C-scan measurement system (Leipzig system).

3 inches in diameter, 0.5 mm in thickness and double-side polished (commercial products from Hefei Kejing Materials Technology Co. Ltd). The target–substrate distance was optimized at 40–70 mm. In this study, the substrates were first cleaned in an ultrasonic cleaner with acetone for 30 min and subsequently with alcohol for 30 min. After cleaning, the substrate was fixed to the sample holder. Then oxygen gas and argon gas with a ratio of 1.5:1 were introduced into the chamber. The total pressure inside the chamber was 20 Pa. The target was pre-sputtered for 15 min, during which the temperature of the sample holder was increased up to the deposition temperature of 750 °C. This pre-sputtering is required to eliminate contamination on the target surface. At the same time, the stainless steel baffle was inserted between the target and the substrate to avoid substrate pollution. The substrates were heated in tube-like heaters. The deposition rate was about 15 nm min^{-1}. After the deposition, the substrate temperature was maintained at 600 °C for 30 min with an oxygen pressure of 8.0×10^4 Pa. Finally, YBCO thin films with a thickness of about 300 nm were obtained.

The surface morphology of the YBCO film was observed by atomic force microscopy (AFM). The phase purity and texture of the as-grown film was characterized by x-ray diffraction (XRD) using Cu Kα radiation and pole figure analysis (Philips X' Pert MRD). An x-ray ω-scan was carried out to evaluate the out-of-plane texture. The in-plane orientation of the film was investigated by means of φ-scans. The resistive T_C value was measured by a standard four-probe method and J_C was measured by a J_C-scan Leipzig system [12]. The microwave surface resistance R_S of the films was measured by a dielectric resonator method operated in the TE_{011} mode at 12.06 GHz.

3. Results and discussion

Four pieces of double-sided YBCO films within one batch were chosen for evaluation of the sample uniformity, as shown in figure 1. In the following descriptions, the film facing up during the heat treatment process is named the 'upside film', which is labeled in lower case. The film facing down is named the 'downside film', which is labeled in capitals. Table 1 gives the measured parameters of four pieces of double-sided YBCO films within one batch.

The J_C mappings at 77 K for the double-sided films (300 nm in thickness) on the four samples are presented in figure 3. It can be seen that the spatial distributions of J_C in the upside films are consistent with those in the downside films. The majority of J_C values in all the samples are in the range 3.75–4.0 MA cm^{-2} and the standard deviation of J_C is about 0.30. This indicates a good J_C uniformity over the whole area of the YBCO films. These results are quite impressive when compared with those previously reported [13–15]. The superconducting transition temperatures of the films were measured by the standard four-probe method. The T_C values of both films are generally in the range 89–90 K.

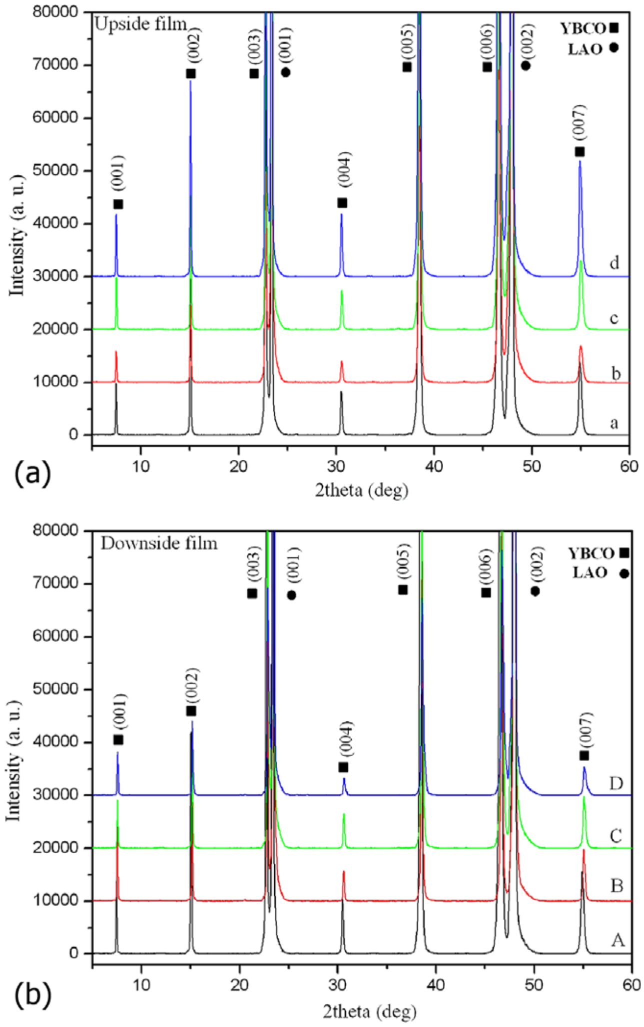

Figure 4. XRD patterns of the YBCO films deposited on LAO substrates: (a) upside films and (b) downside films.

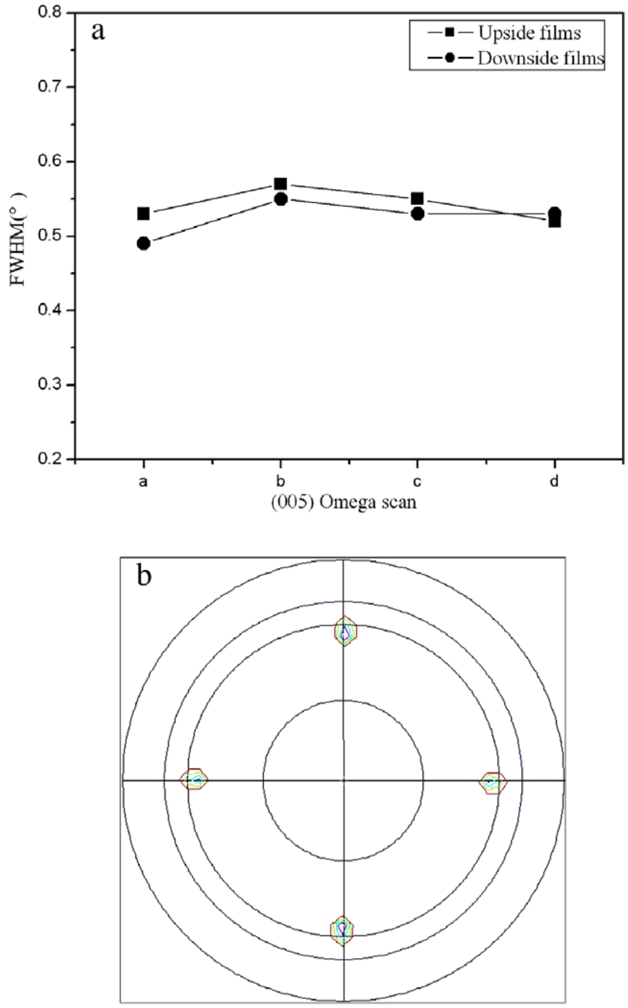

Figure 5. XRD patterns for the YBCO films deposited on LAO substrate (a) FWHM of x-ray(005) ω-scan and (b) x-ray (102) φ-scans.

Table 1. The measured parameters of YBCO films at different positions in the same batch.

Sample	J_{CM}^{a} (77 K, 0 T) (MA cm^{-2})	J_{CS}^{b}	T_C (K)	FWHM (ω-scan) (deg)	RMS (nm)	R_S (77 K, 10 GHz) (mΩ)
a	3.97	0.31	89.2	0.49	6.19	0.59
A	3.78	0.27	89.5	0.52	7.02	0.68
b	3.89	0.28	89.8	0.53	6.23	0.62
B	3.78	0.31	90.0	0.56	7.15	0.71
c	3.83	0.30	90.0	0.51	6.16	0.56
C	3.81	0.25	89.9	0.53	7.08	0.68
d	3.80	0.32	90.0	0.54	6.32	0.65
D	3.83	0.25	89.7	0.53	7.07	0.70

[a] J_{CM} is the mean value of the critical current density.
[b] J_{CS} is the standard deviation of the critical current density.

Figure 4 shows the XRD θ–2θ diffraction patterns for the same samples as in figure 3. It can clearly be seen that all the peaks can be indexed as (00*l*) reflections of YBCO phase and LAO substrate for all the samples, as labeled. It suggests that all the samples completely crystallize with a pure (00*l*) phase. No impurity phases are observable. To characterize the divergence of out-of-plane orientation of LAO (00*l*) and YBCO (00*l*) planes, an x-ray ω-scan was performed for the YBCO (005) reflections. The full-width at half-maximum (FWHM) of the (005) peaks for the upside and downside films of the samples range typically from 0.49° to 0.60°, as shown in figure 5(a). These results indicate that the YBCO films have a good out-of-plane texture. The in-plane orientations of the YBCO films were investigated by means of φ-scans. All the samples have similar results, as shown in figure 5(b). A fourfold symmetry can be clearly seen, which confirms the presence of a primarily single in-plane orientation. Both patterns show that the YBCO films are highly *c*-axis-oriented, with no secondary phases or off-orientations.

An AFM study was used to examine the surface morphology of the same samples as in figures 4 and 5. The scanning area of the AFM images was 20 μm × 20 μm. The root-mean-square (RMS) surface roughness was measured, as shown in figure 6. The RMS for the upside films of the samples are 6.19 nm, 6.23 nm, 6.16 nm, and 6.32 nm, respectively, while the RMS data for the downside films of the samples are 7.02 nm, 7.15 nm, 7.08 nm, and 7.07 nm, respectively. It is evident that the YBCO films are smooth and uniform.

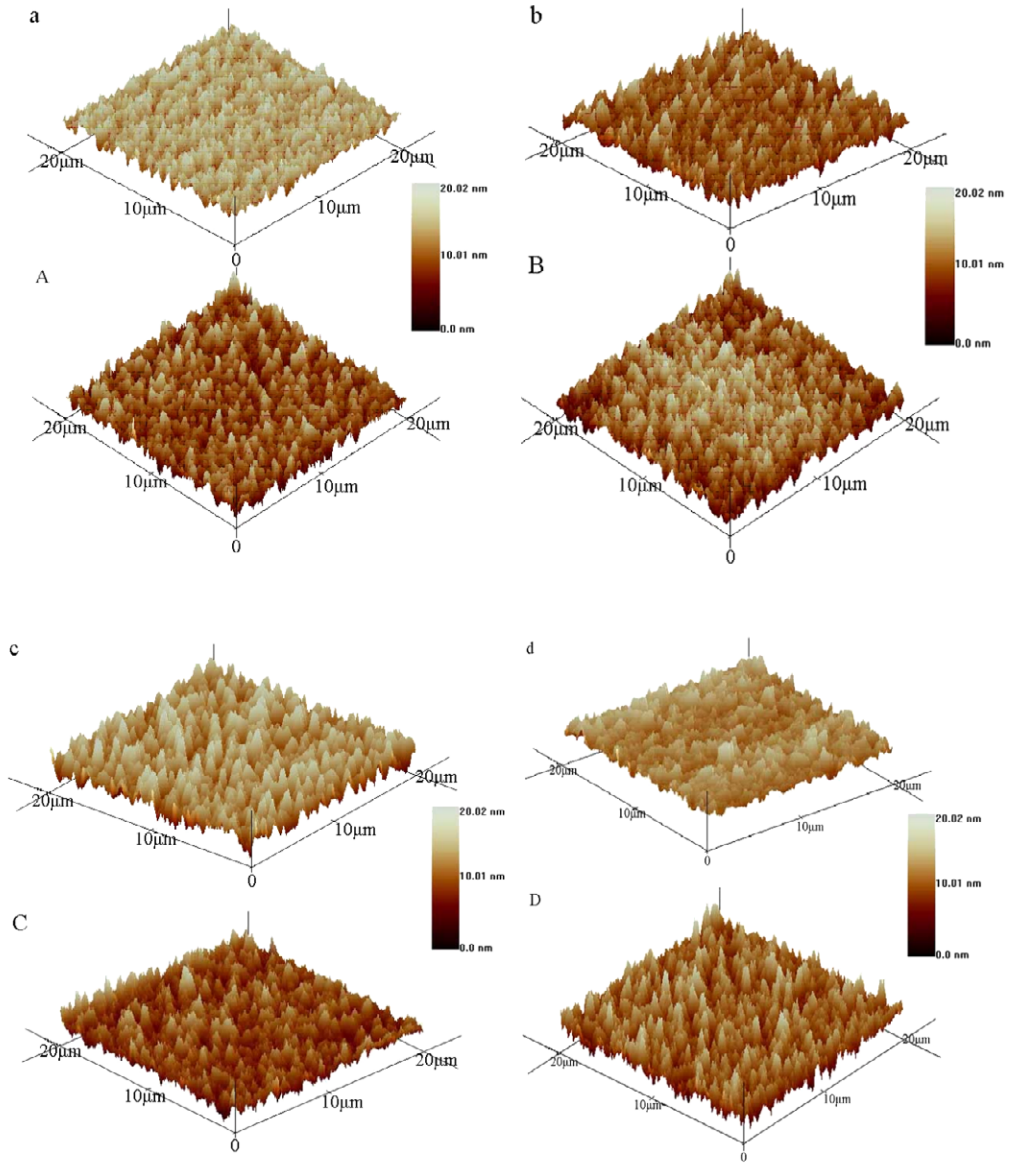

Figure 6. AFM micrographs of the upside and downside films of the same samples as in figure 2.

The microwave surface resistance R_S of the films was measured by a dielectric resonator method. The frequency response of a dielectric resonator with a short end of the YBCO film is shown in figure 7. It operates in the TE_{011} mode with a resonance frequency $f_0 = 12.06$ GHz. The microwave surface resistance at 10 GHz in low-power fields is extrapolated from the experimental data by assuming $R_S \propto f^2$. The R_S (77 K, 10 GHz) values of YBCO thin films on both sides of the samples range typically from 0.56 to 0.71 mΩ. This is homogeneous and the microwave surface resistance is low enough to meet the requirements for microwave applications well.

Finally we would like to put an emphasis on the use of a rotatable disc sample holder during the sputtering process, which, in our view, is very important for the batch production of high quality large-area double-sided films. On the one hand, the rotatable disc sample holder is a crucial step in the realization of a large scale deposition process. Due to the disc sample holder rotating at a fixed speed, all the samples can be deposited on YBCO films. On the other hand, it ensures the adatoms diffuse around the substrate surfaces very well in the deposition process. The YBCO thin film nucleation and growth process may occur as follows [16]. The sputtered atoms are deposited onto the substrate surface with a deposition flux. Once atoms adsorb onto the surface as adatoms, they can diffuse around on the substrate surface. These adatoms can meet other adatoms to form a dimer or to attach to existing islands. Once adatoms are attached to an island, they can diffuse along the island edge, enter into the kink or detach from the island edge. In our experiment, the deposition process can be divided into two steps according to the relative position between the target and the sample. In the second step, the adatoms have more time to diffuse to positions on the substrate surface with lower free energy. Therefore, high quality epitaxial YBCO thin films can be obtained.

Figure 7. The frequency response of the dielectric resonator with YBCO films.

4. Conclusions

In this study, a novel DC magnetron sputtering system for preparing large double-sided $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films has been developed. Twelve pieces of ϕ3-inch high quality double-sided YBCO thin films have been reproducibly fabricated by this method in one batch. The properties of the double-sided YBCO thin films are uniform and very similar on both sides of the wafer. The YBCO films exhibit a good *c*-axis texture and desirable surface morphology. The full-width at half-maximum (FWHM) values of the (005) rocking curves range typically from 0.49° to 0.56°. The superconducting properties T_C, J_C and R_S for typical films all fall into the range 89–90 K, 3.75–4.0 MA cm^{-2} (77 K) and 0.56–0.71 mΩ at 77 K and 10 GHz, respectively. The performances of our samples fulfil the requirements for microwave devices.

Acknowledgments

This work was supported by the National Nature Science Foundation of China under grant No 50672011 and the Hi-Tech Research and Development Program of China under grant No 2007AA03Z216.

References

[1] Schey B, Biegel W, Kuhn M, Klarmann R and Stritzker B 1998 *Appl. Surf. Sci.* **127–129** 540–3
[2] Kinder H 2001 Device for producing oxidic thin films *US Patent Specification* 6,294,025
[3] Jaekel C, Waschke C, Roskos H G, Kurz H, Prusseit W and Kinder H 1994 *Appl. Phys. Lett.* **64** 3326–8
[4] Selvamanickam V *et al* 2003 *IEEE Trans. Appl. Supercond.* **13** 2492–5
[5] Manabe T, Sohma M, Yamaguchi I, Tsukada K, Kondo W, Kamiya K, Tsuchiya T, Mizuta S and Kumagai T 2006 *Physica* C **445–448** 823–7
[6] Ding F, Gu H and Li T 2008 *Supercond. Sci. Technol.* **21** 095004
[7] Araki T, Hirabayashi I and Niwa T 2004 *Supercond. Sci. Technol.* **17** 135–9
[8] Liu X Z, Tao B W, Luo A, He S M and Li Y R 2001 *Thin Solid Films* **396** 225–8
[9] Zaitsev A G, Kutzner R, Wördenweber R, Kaiser T, Hein M A and Müller G 1998 *J. Supercond.* **11** 361–5
[10] Liu X Z, Tao B W, Deng X W, Zhang Y and Li Y R 2002 *Supercond. Sci. Technol.* **15** 1698–1700
[11] Roeck I D *et al* 2002 *Physica* C **372–376** 1067–70
[12] Hochmuth H and Lorenz M 1996 *Physica* C **265** 335–40
[13] Abbott F, Dégardin A F and Kreisler A J 2005 *IEEE Trans. Appl. Supercond.* **15** 2907–10
[14] Lemaitre Y, Marcilhac B, Mansart D, Siejka J and Mage J C 2002 *Physica* C **372–376** 667–70
[15] Avci I, Tepe M and Abukay D 2004 *Solid State Commun.* **130** 357–61
[16] Ratsch C and Venables J A 2003 *J. Vac. Sci. Technol.* **21** S96–S109