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Off-axis unbalanced magnetron sputtering of $YBa_2Cu_3O_7$ thin films

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Abstract

An unbalanced magnet assembly is used in DC off-axis sputtering to grow $YBa_2Cu_3O_7$ superconducting thin films. The assembly with a stronger central magnet than the annular magnet directs plasma to bombard the off-axis substrate, resulting in deteriorated thin films. In contrast, the assembly with a weaker central magnet directs plasma away from the off-axis substrate and results in a film with a higher transition temperature and a smoother surface. Furthermore, the latter type of unbalanced DC magnetron sputtering gives a higher deposition rate and enables the grown film to be uniform in composition on static substrates over a range of 8 cm in length. These results reveal the feasibility of using the unbalanced magnetron for thin film growth.

Keywords: Unbalanced magnetron; YBCO; Thin films; Sputtering

1. Introduction

Various thin film deposition techniques have been used to prepare high quality $YBa_2Cu_3O_7$ (YBCO) films grown from a stoichiometric target. In particular, sputtering serves as a highly effective technique for depositing YBCO films. Using off-axis sputtering or high pressure sputtering can eliminate the resputtering effect on as-deposited films. Additionally, Savvides and Katsaros successfully grew YBCO films by using an unbalanced magnet assembly on an on-axis sputtering system [1,2]. Using that approach, the magnetic field directs the plasma away from the substrate and avoids resputtering effects commonly observed in on-axis sputtering. The magnets used in their experimental setup were a stronger central magnet and a relatively weaker annular magnet around it. If the central magnet is weaker than the annular magnet, the plasma is directed to the substrate [3]. In addition to this modified on-axis sputtering, off-axis sputtering seems to be a more popular choice for growing YBCO films with large areas [4]. Rao et al. used off-axis sputtering with rotational substrates to prepare YBCO thin films up to 5 in in diameter with uniform characteristics [5]. We consider here how the unbalanced magnetron be used in an off-axis geometrical configuration. In this study, we explore the effects of various magnet assemblies with respect to the deposited YBCO thin films' morphology and composition. The unbalanced magnet assembly with a weaker central magnet allows YBCO films to be grown with better superconductive properties and surface morphologies. In addition, films with a uniform composition distributed over 8 cm on a stationary substrate can be grown at 100 mTorr from a 2 in target.

2. Experimental

Fig. 1 depicts the experimental apparatus with the normal off-axis geometrical configuration. The sputter gun was located perpendicular to the substrate holder. YBCO thin films were grown at an argon to oxygen flow rate ratio of 4/1 and a substrate temperature of 700°C with various total chamber pressures. The sputter current was maintained at 150 mA and the cathode voltage was about 120–140 V. The growth rate for balanced magnetron sputtering was 0.93 nm min^{-1} at 100 mTorr and decreased to 0.31 nm min^{-1} at 300 mTorr. The grown films' thickness ranged from 160 nm to 200 nm. As shown in Fig. 1, three types of magnet assemblies were used inside the gun to change the magnetic field distribution, which affects the plasma distribution. In a balanced magnetron, all the field lines originating from the central magnet finish on the annular magnet. In one extreme (type I), two central magnets are set and thus some field lines do not pass into the annular magnet. In these circumstances, the plasma is directed off-axis (side-directed). In the other extreme (type II), a weaker central magnetic field is used so that some field lines from the annular magnet do not pass into the central

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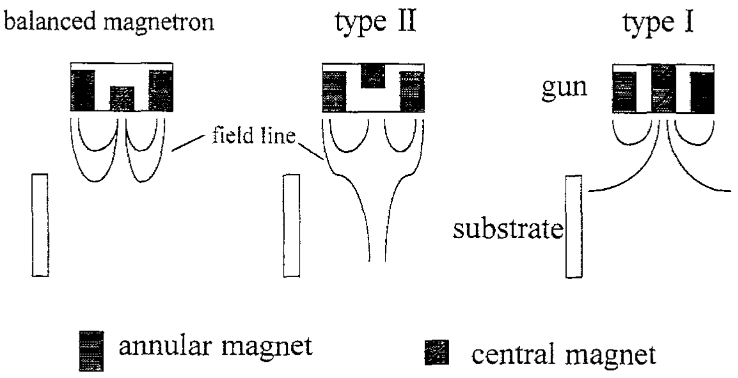

Fig. 1. Schematic diagram of the balanced, type I and type II magnetrons.

magnet. Such a configuration renders the plasma to be directed on-axis as shown in Fig. 1.

3. Results and discussion

All the thin films grown at pressures above 100 mTorr exhibit *c*-axis orientations. However, *c*-axis crystalline films can also be obtained at pressure of 65 mTorr for the type I magnetron and the balanced magnetron but they cannot be grown for the type II magnetron at 65 mTorr. The substrate temperature is sufficiently high and the oxygen pressure at 65 mTorr is suitable to grow *c*-axis YBCO films. However, the available oxygen, which consists of injected molecular oxygen and sputtered atomic oxygen, may change near the substrate owing to the varied plasma guided by magnet assemblies. For type II magnetron sputtering, this available oxygen near the substrate could be decreased because the plasma is directed away. Thus, the inability to grow *c*-axis films at 65 mTorr for type II magnetron is possibly ascribed to insufficient oxygen pressure over the substrate. On the contrary, the appearance of *c*-axis film grown by type I magnetron at 65 mTorr implies that the side-directed plasma may yield a higher oxygen pressure near the substrate. However, the side-directed plasma may also introduce possible bombardments. Scanning electron microscopy (SEM) images of the films in Fig. 2(a) confirm this bombardment. As the figure reveals, the surface is distributed with bombarded concavities. A high chamber pressure can reduce the bombardment effect. Figs. 2(b) and 3(b) show that the as-

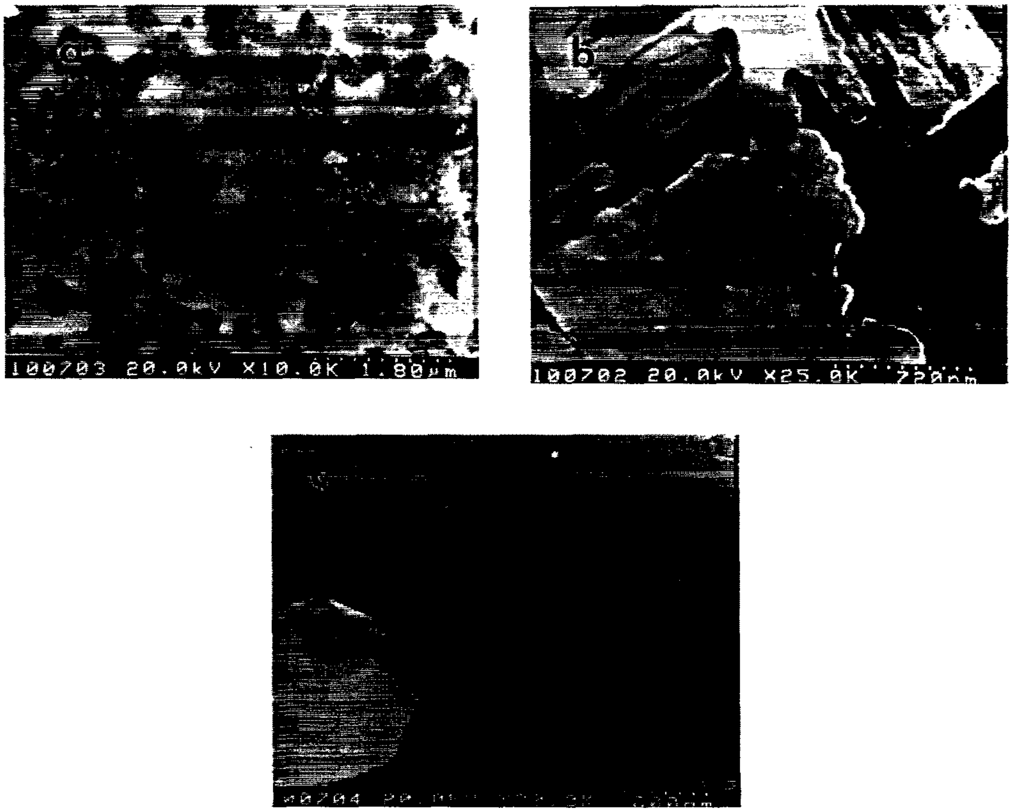

Fig. 2. SEM images of the films grown with type I magnetron at (a) 30 mTorr, (b) 100 mTorr and (c) 300 mTorr.

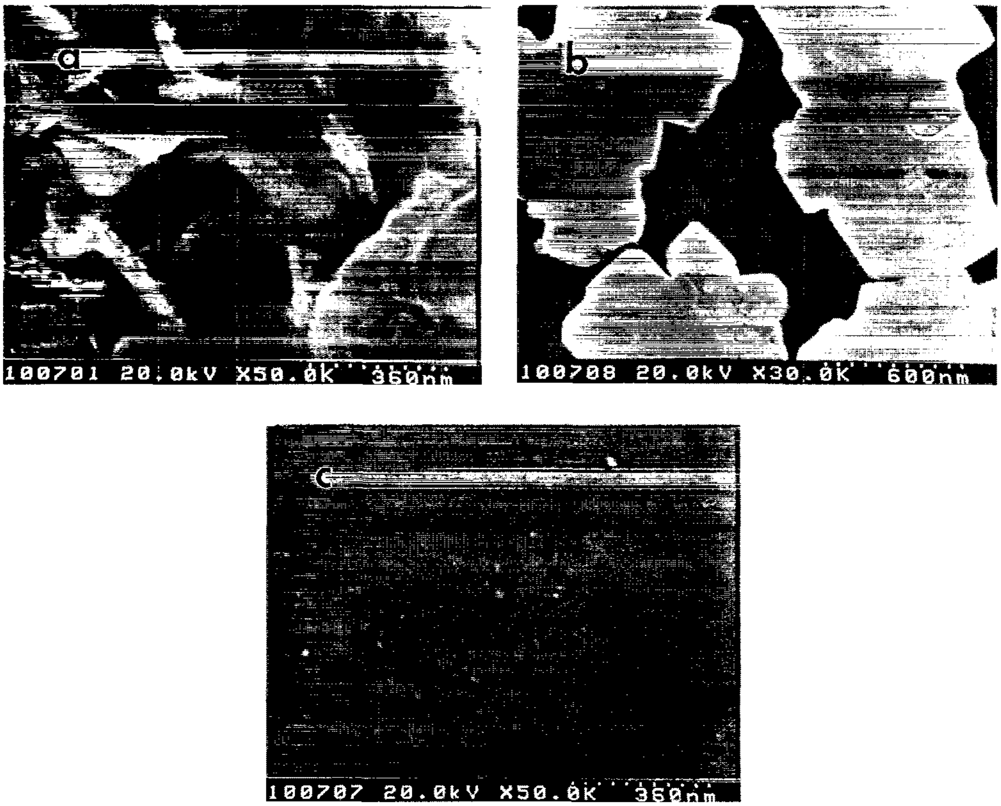

Fig. 3. SEM images of the films grown with type II magnetron at (a) 30 mTorr, (b) 100 mTorr and (c) 300 mTorr.

grown films at 100 mTorr have the typical YBCO rectangular mosaic morphologies (underlayer). Nevertheless, YBCO grains in Fig. 2(b) grown at 100 mTorr with type I magnetron do not have as precise rectangular YBCO outlines as the grains grown with type II sputtering as illustrated in Fig. 3(b). Probably, minor plasma bombardment continues during the film's growth with type I magnetrons at 100 mTorr. The more irregular grain outline in Fig. 2(b) than that in Fig. 3(b) suggests that the film grown at 100 mTorr with a type II magnetron should have better crystallinity than that grown at 100 mTorr with a type I magnetron. Figs. 2(c) and 3(c) depict the surfaces of the films grown at 300 mTorr with type I and type II magnetrons. The surfaces are smooth except for the large particle in Fig. 2(c). The film's surface becomes smoother as the total pressure increases for all the magnetrons. Next, the rocking curve of the film's X-ray diffraction was examined to identify the film's crystallinity. Table 1 lists the full width at half-maximum (FWHM) values of (005)YBCO diffraction of the films. As this table reveals, the crystallinity of the film grown with a type II magnetron is better than that of the film grown with a type I magnetron. This finding corresponds to the observations from SEM images.

Properties of the films grown with a balanced magnetron are exactly characterized intermediately between the results of type I and those of type II magnetron. The surface morphologies observed from Figs. 2(b) and 3(b) denote a smooth overlayer and an oriented mosaic spread of YBCO underlayer, respectively. The distinct difference in surface morphology between the underlayer and the overlayer excludes the possibility of layer-by-layer growth. The overlayer in Fig. 2(b) shows clusters of YBCO islands having dimensions of 150 nm × 150 nm. Such clustering is a typical characteristic of island growth [6]. The overlayer exhibits grain coalescence characteristics. The films in Figs. 2(c) and 3(c) are smooth and do not exhibit mosaic morphology. Fig. 4 shows atomic force microscopy (AFM) images of the films grown at 300 mTorr with a type II magnetron. The film grown to a thickness of 400 Å reveals preferably aligned grains with respect to the substrate. The surface roughness is about 35 Å. Section analysis of the surface shows a step height of 30–42 Å. As Fig. 4(b) indicates, similar surface morphology is observed for the film grown to a thickness of 2000 Å. This observation indicates that the growth of YBCO is not two-dimensional and tends to be island growth in this thickness range.

As indicated in Table 1, the resistance–temperature measurements (*R*–*T*) of the films reveal that the films grown with a type II magnetron exhibit the highest transition temperature (T_{co}). The T_{co} superiority of the films grown with a type II magnetron is apparent at low pressures; however, it becomes unclear for those grown at high pressures. Further annealing of those films grown with a type I magnetron at 1 atm oxygen and 450°C does not improve the *R*–*T* curves. Moreover, the oxygen pressure near the substrate is higher for films grown with a type I magnetron as suggested above. Thus the inferior *R*–*T* property of type I magnetron grown films is not due to the possible relative oxygen deficiency of the films. The infe-

Table 1
Properties of the films grown with type I, type II and balanced DC magnetrons

Magnetron type of the samples	Growth pressure (mTorr)	FWHM	Electrical property T_{c0} (K)
type I	30	[a]	–
type I	65	0.63	–
type I	100	0.8	76
type I	200	0.6	85
type I	300	0.22	86
type II	30	[a]	–
type II	65	[a]	–
type II	100	0.52	82
type II	200	0.44	87
type II	300	0.20	86
balanced	50	0.68	70
balanced	100	0.69	81
balanced	200	0.55	86
balanced	300	0.22	84

[a] The film does not exhibit a reliable c-axis oriented X-ray spectrum.

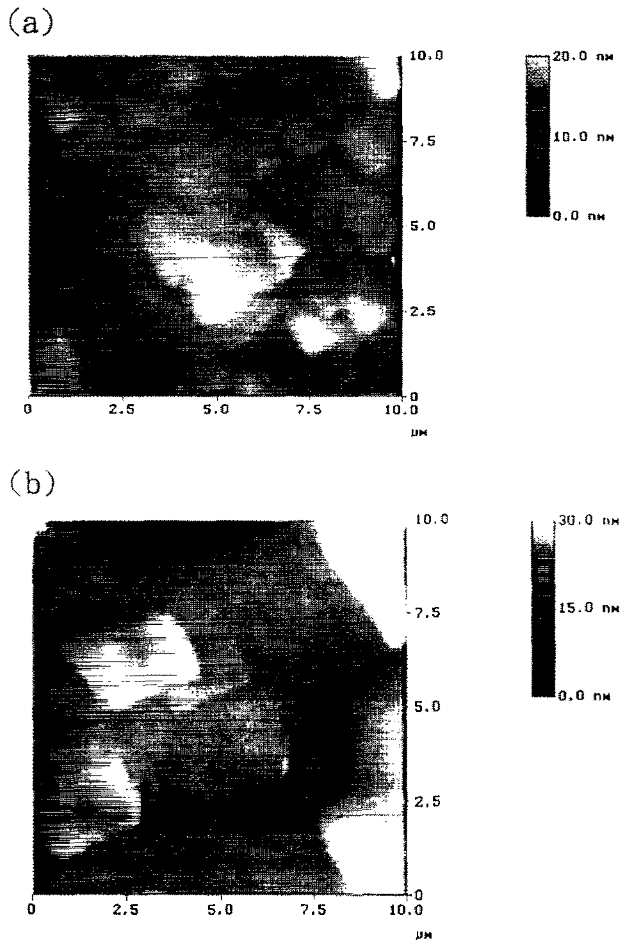

Fig. 4. Atomic force microscopy images of the (a) 400 Å and (b) 2000 Å thick films grown at 300 mTorr with type II magnetron.

rior T_{c0} property of the films grown with a type I magnetron compared with those for the other magnetrons seems to be related to the poorer crystallinity of the films caused by plasma bombardment.

Fig. 5 depicts the spatial composition variation of the films grown at 700°C with a type II magnetron at 100 mTorr and 200 mTorr. Composition for both the thin films grown by a type II magnetron is uniform over substrates distributed 8 cm in length along the holder. However, the films grown at 200 mTorr exhibit better metal stoichiometry than those grown at 100 mTorr. This stoichiometry partly explains why the films grown at 200 mTorr exhibit higher T_{c0} than those grown at 100 mTorr with type II magnetron. Furthermore, the films grown with an balanced magnetron and a type I magnetron at 100 mTorr or 200 mTorr do not have a similarly uniform composition. Minor plasma bombardment probably occurs for both magnetrons. For comparison, films are also grown by RF magnetron sputtering (balanced and type II). However, the compositions show a greater monotonic change with distance from the target for total pressures of 100 mTorr and 200 mTorr. This result is different from a previous result [4]. At lower pressures, the composition distribution generally changes slightly at distances far from the target and

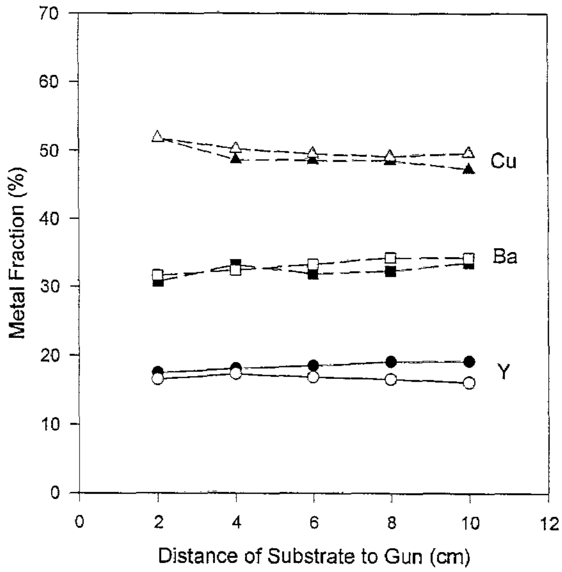

Fig. 5. Spatial variation of metal fraction of the films grown with type II DC magnetron at 100 mTorr (filled symbols) and 200 mTorr (open symbols).

changes to a greater extent near the target. This change may be due either to the mass effect of angular distribution of sputtered atoms or to the possible resputtering effects on the films near the target [7]. Thus, a substrate would not be preferred to locate near the target, although the deposition rate is high. Moreover, the composition change of films grown by type I and balanced magnetrons near the target is nonlinear with distance. Therefore, a rotational substrate would not be expected to transform the nonlinear composition variation with distance into a uniform distribution of the film's composition. Since a substrate holder was unavailable to support a uniform high temperature (at least 700°C for YBCO growth) over 5 cm in size, we grew YBCO thin films at 100 mTorr with type II DC sputtering for two experimental setups. The first setup placed the substrate holder close to the target. The other setup placed the substrate holder 2 in from the target. The overall electrical properties of the films for the two setups are uniform with $T_{co} = 82 \pm 2$ K. The electrical properties are believed to be improved with an increase in substrate temperature and oxygen pressure.

The weaker central magnet in type II magnetron sputtering drives the plasma concentration inward and thus the sputter etched track on the target is smaller. Therefore, sputtered area in the target is reduced, which might cause fewer sputtered atoms and a lower deposition rate. On the other hand, since the input power is maintained constant, the input power density is higher for the type II magnetron, which might increase the deposition rate. Experimental results show that the deposition rate for type II magnetron sputtering is around 0.37 nm min^{-1} at 300 mTorr, even slightly higher than those of the other magnetrons, in fact. Moreover, the smaller etched track on the target means that even a smaller target is usable for sputtering without decreasing the deposition rate. These results reveal the high feasibility of sputtering with type II magnetron.

4. Summary

In summary, the magnet assembly in the sputter gun plays an important role in the plasma distribution even for off-axis sputtering. Unbalanced magnetron sputtering with a stronger central magnet than the annular magnet directs plasma to the off-axis substrate and causes as-deposited YBCO films to be resputtered. In contrast, unbalanced magnetron sputtering with a weaker central magnet directs the plasma further away from the off-axis substrate, thereby producing films with a better T_{co} and uniformly distributed composition with distance. YBCO thin films with uniform compositions over 8 cm were obtained using the latter magnetron. This demonstrates the feasibility of using an unbalanced magnetron to grow large area YBCO thin films.

Acknowledgements

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