Vol. 92 (1997) *ACTA PHYSICA POLONICA A* No. 1

Proceedings of the ICSC–F'96 Jaszowiec '96

GROWTH SPIRALS APPEARED IN *c*-AXIS ORIENTED YBCO FILMS GROWN ON MgO SUBSTRATE BY SPUTTERING

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The early stages of growth and the evolution of surface microstructure of epitaxial *c*-axis $YBa_2Cu_3O_{7-x}$ thin films were studied as a function of film thickness, growth temperature, and growth rate. The films were grown *in situ* on as-polished or annealed MgO(100) substrates by off-axis magnetron sputtering. Atomic force microscopy was used to observe the surface microstructure. From the results at early stages, we proposed a growth model of spirals for the films grown on as-polished substrate, i.e. spirals are formed around the surface roughness of the substrate. Growth temperature and growth rate dependence on the density of spirals support this model. That is, the density of spirals does not change according to the change of growth temperature (600–740°C) or growth rate (7–80 nm/h).

PACS numbers: 74.76.Bz, 74.72.Bk, 68.55.-a

1. Introduction

It is very important to know the growth mechanism to obtain high-quality films with atomic surface flatness. The surface morphology of $YBa_2Cu_3O_{7-x}$ (YBCO) films depends on the deposition method, the substrate, as well as the preparation conditions, for example substrate temperature, deposition rate, film thickness, etc. [1–4]. Lattice mismatch to substrate and supersaturation during film growth are the essential parameters for thin film growth modes [3, 5]. MgO(100) single crystal is one of several commonly used substrates, but it has a large lattice misfit, –9%, with YBCO. In the case of MgO as substrate, the screw dislocations appear immediately upon the nucleation of islands and spiral growth mode becomes dominant [6, 7]. But, so far, two key questions: (1) When is the spiral growth initiated?, and (2) What is the origin of the spirals? have not been solved.

We observed growth spirals by scanning tunneling microscopy (STM) which appeared in *c*-axis oriented YBCO thin films grown by off-axis magnetron sputtering on as-polished and annealed MgO(100) substrates [8]. The density of spirals did not change by the growth temperature, but it was strongly influenced by the annealing of the substrates. We assumed that screw dislocations are originated at the nucleation centers on substrate surface and emanate to growth spirals when YBCO films are grown on as-polished, rough surface.

In this study, we observed the atomic force microscopy (AFM) images of substrate and YBCO thin films grown by off-axis sputtering on as-polished MgO(100) substrate and investigated the early stages of growth and the evolution of surface microstructure as a function of film thickness, growth temperature and growth rate. From the results, we proposed a growth model of spirals for the films grown on as-polished substrate, i.e. spirals are formed around the surface roughness of the substrate.

2. Experimental

YBCO films were deposited *in situ* by off-axis magnetron sputtering from a stoichiometric target on commercially available, as-polished MgO(100) single crystal substrate. Ratio of O_2/Ar and total pressure of sputtering gas were 50/50 and 200 mTr respectively. The nominal ultrathin film thickness was calibrated from the deposition rate determined for thicker films using Dektak3030 profilometer.

The surface morphology of the substrate and the as-grown YBCO films were investigated by atomic force microscopy in air. The scan size of all images was 1 μm×1 μm. Density of roughness of the substrate and that of spirals of YBCO films were determined by counting all islands which were recognized as them on the images.

3. Results and discussion

In this study we focused our interest on the solution of the question: whether there is a roughness of the substrate in the center of the spirals or not. The surface of as-polished substrate used in this study was very rough and no terrace or step structure was observed [8]. The variance of the surface (Ra) of the substrate was 4.13 nm and the density of roughness was 3.4×10^{10} cm^{-2}. AFM images in Fig. 1 show the early stage of the growth of YBCO on as-polished MgO substrate when nominal thickness of YBCO films was less than 3 nm. When thickness was 1 nm (Fig. 1a), existence of YBCO was not confirmed. This image is similar to that of as-polished MgO substrate. When 2 nm (Fig. 1b), it was clearly observed that YBCO was growing just around the roughness of the substrate in the lower right corner of the image. When 3 nm (Fig. 1c), the coverage of YBCO was nearly 1, but cone-shaped peaks of substrate were also observed. It seems that there are peaks of roughness of the substrate in the center of islands.

Surface profiles of each sample are shown in Fig. 1d–f by solid lines. Of course, z(height)-direction is enhanced more than ten times as x(distance)-direction, cone shape is the majority in Fig. 1d. We thought that these cone shapes were the surface of the substrate, and boldly drew the interface between substrate and growing film as shown by broken lines in the figure. Hereafter in this article, cone shaped surface roughness of substrate is called a "cone". Note that these broken lines are still matter of speculation. The height of cone varied from a few nm to several tens nm. Here shadowed areas are the growing YBCO. At a glance of Fig. 1d–f, it is vividly seen that YBCO grows making spiral structures around the cones of the substrate. Namely, when nominal film thickness was 1 nm, i.e. about 1 unit cell of YBCO (Fig. 1d), YBCO seemed to be deposited only at the bottom

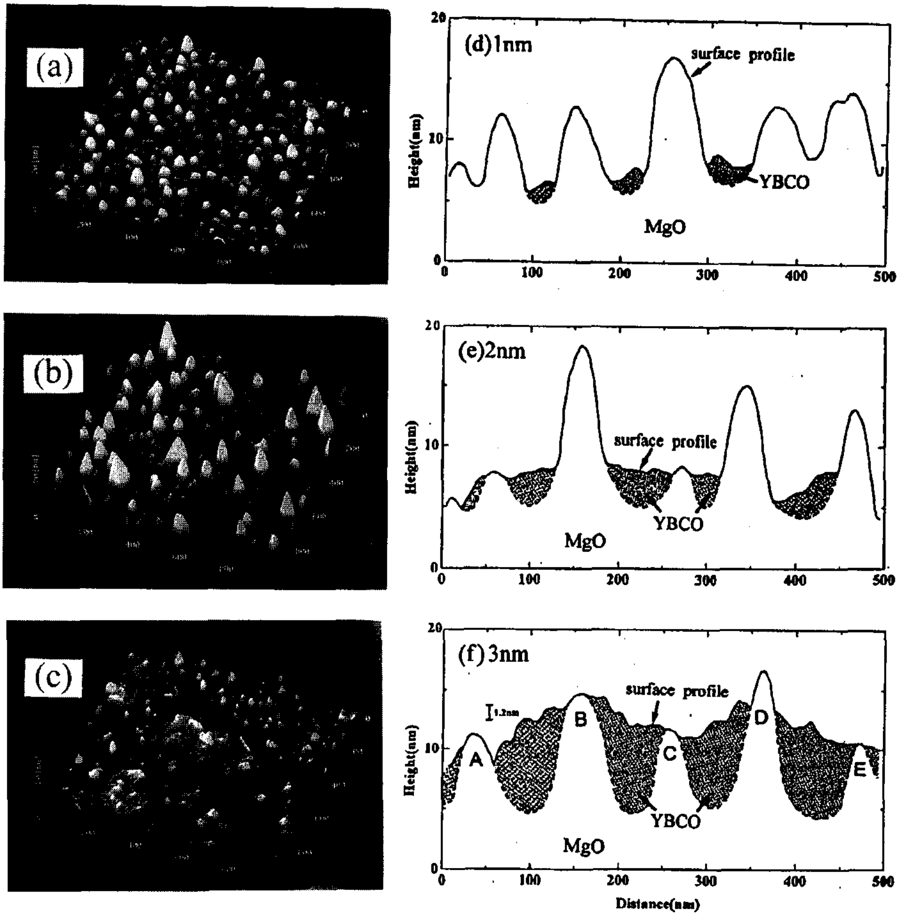

Fig. 1. AFM images and cross-sectional structures speculated from line scans of ultra-thin YBCO films deposited on as-polished MgO substrate. The nominal film thickness are 1 nm ((a) and (d)), 2 nm ((b) and (e)) and 3 nm ((c) and (f)).

of the roughness of the substrate. When 2 nm (Fig. 1e), it seems that YBCO film grows burying the low cones of the substrate. Figure 1f shows us clearly that YBCO grows around tall roughness of the substrate making spiral structures when nominal film thickness is 3 nm. Around cone B and D, 1.2 nm high steps which correspond to one unit cell of YBCO were observed. The height from the substrate of them between right side and left side of each cone is different about half step (0.6 nm). This means that spiral structures are growing around cones B and D.

Here we propose the growth model of YBCO film on rough MgO substrate as follows; the spirals which are made around tall roughness of the substrate such as cones B and D will remain as spirals to the last, but those made around low cones such as A, C or E may be coalesced increasing the film thickness and according to the growth conditions. In other words, there is a cone of substrate in the center of a spiral. In order to make sure the adequacy of this assumption, we observed the dependence of the surface morphology on growth conditions such as film thickness, growth temperature, and growth rate.

At first we discussed the influence of film thickness on the surface morphology. When YBCO films were grown at 740°C and thickness was more than 5 nm, clear spiral structures were observed in all films. Figure 2 shows the thickness dependence of the density of spirals. The density of spirals decreased with increasing the film thickness. And it became constant (3.5×10^9 cm^{-2}) when film was thicker than 40 nm. This value was almost the same with the density of roughness whose height was more than 6 nm. This result strongly suggests that the formation of spirals closely correlated to the roughness of the substrate. That is, in case of rough surface, YBCO is initially deposited around the cones of the substrate and grows making spiral structures. And with increasing the film thickness, the weak spirals which are initiated at low cones will be coalesced by strong ones initiated at tall cones. This means that there is a cone of substrate in the center of a spiral. If this assumption is correct, the surface morphology of YBCO films grown on such rough substrate will not be affected by the growth conditions such as growth temperature or growth rate.

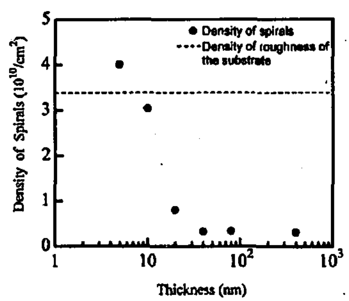

Fig. 2. Thickness dependence of the density of spirals.

Then we observed the temperature dependence of the surface structures between 600°C and 800°C for 80 nm thick YBCO films. By varying the growth temperature it is possible to change the diffusion length and, in effect, to alter the amount of supersaturation of constituent atoms under which film growth occurs [6]. When growth temperature was between 600°C and 740°C, the size and the density of spirals did not change significantly. On the other hand, when growth temperature was 800°C, the size of spirals became fairly large and the density of them decreased to 1×10^9 cm^{-2}. The growth steps became quite broad, but the shape was still rhombic. Screw dislocation is reported to decrease with increasing growth temperature and misorientation of the substrate [9]. But the fact that the density of spirals was not changed when growth temperature was varied from 600°C to 740°C means that the mechanism of growing spiral structures of our case is different from the usual case emanating from the screw dislocations on smooth surface. This supports our assumption that spirals are formed around the roughness of the substrate when deposited on as-polished MgO substrate.

Fig. 3. AFM image of 80 nm thick YBCO film grown at a growth rate of 7 nm/h on as-polished MgO substrate. The growth temperature is 740°C. The scan size is 1 μm×1 μm.

Finally, we observed the effect of growth rate on the surface morphology of YBCO films. Growth rate controls the supersaturation on the substrate, which is an essential parameter dominating thin film growth modes [5]. All films shown above were grown at 100 W rf power and growth rate as 80 nm/h. With decreasing rf power, growth rate decreased linearly, and when rf power was 20 W it was reduced to 7 nm/h. Figure 3 shows the AFM image of YBCO film grown at a growth rate of 7 nm/h for 10 hours. Growth temperature was 740°C. Shape of islands became unclear with decreasing growth rate from 80 nm/h to 7 nm. But the size and the density of island did not change significantly when growth rate was varied from 80 nm/h to 7 nm/h. We think that this result also supports our model.

4. Conclusion

We grew *c*-axis oriented epitaxial YBCO thin films on as-polished MgO(100) substrates by off-axis magnetron sputtering and observed the surface morphology by AFM. From the results of the observation of early stage of the growth and the film thickness dependence of the density of spirals we proposed a growth model of YBCO film on rough MgO substrate as follows; initially spirals are formed around the substrate roughness, and those which are formed around tall roughness of the substrate will remain as spirals to the last, but those made around low roughness will be coalesced with increasing the film thickness and according to the growth conditions. We thought that there is a roughness of substrate in the center of a spiral. The facts that the density of spirals does not change according to the change of growth temperature from 600°C to 740°C or to the change of growth rate from 7 nm/h to 80 nm/h support our model.

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