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Scanning Tunneling Microscopy Study on $YBa_2Cu_3O_{7-x}$ Films with Growth Stop by observing Reflection High–Energy Electron Diffraction Oscillations

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Abstract

The growth of $YBa_2Cu_3O_{7-x}$ thin films was stopped on a maximum or a minimum of an intensity oscillation in reflection high- energy electron diffraction(RHEED). The resulting film surfaces were observed by scanning tunneling microscopy. Films stopped on a maximum have smoother surfaces, closer to the completion of a monolayer of unit cells, than those on a minimum. The width of monolayer terraces was almost the same as that of the substrates determined from the misorientation. On the terraces, islands of one unit cell height were observed, so that the diffusion length of $YBa_2Cu_3O_{7-x}$ must be shorter than the terrace size.

1. Introduction

The growth of high Tc superconducting thin films has been studied intensively to obtain smooth surfaces and excellent superconducting properties for applications. Recently, intensity oscillations of the specularly reflected beam in reflection high- energy electron diffraction (RHEED) have been found during the growth of $YBa_2Cu_3O_{7-x}$ by Terashima et al.[1]. They found that the period of the oscillations corresponds to the growth of one unit cell in the c direction. On the other hand, the surface morphology of $YBa_2Cu_3O_{7-x}$ has been studied on an atomic scale by scanning tunneling microscopy(STM) [2,3]. Screw dislocations were observed on most of the films, irrespective of the deposition methods (co- evaporation[4], sputtering [5] and laser ablation[5- 7]). As far as we know, there is no report to correlate the RHEED intensity to the surface morphology of $YBa_2Cu_3O_{7-x}$ films. It is very important to study the early stages of the film surface, which may provide informations for the formation of screw dislocations.

2. Film Preparation and STM Measurements

Films were deposited by reactive thermal co-evaporation. The oxygen pressure near the substrate was 10^{-4} mbar[4]. The deposition rates were 0.077- 0.108 nm/s. $SrTiO_3$(001) was used as a substrate, which had a misorientation of 0.67 - 1.85 degrees obtained by X- ray diffraction measurements, as shown in Table I. The substrate temperature was 650 °C. The electron beam for the in- situ RHEED observation had 18.8 kV and was incident along the [100] direction of the $SrTiO_3$. The angle of the incident beam is given in Table I.

Table I. Deposition conditions of the films and the results of STM measurements.

Sample No.	RHEED Oscill.	Incident Beam Angle [deg.]	Deposition Rate [nm/s]	Misorientation of the Substrate [deg.]	Calcu.Average Terrace Width [nm]	Meas. Average Terrace Width [nm]
A	max. 10	1.49	0.083	0.92	24	50
B	min. 6	1.25	0.108	1.85	12	30
C	max. 11	1.18	0.084	0.28	80	80
D	max. 9	0.59	0.077	0.98	23	40
E	min. 8	2.24	0.078	0.67	34	70

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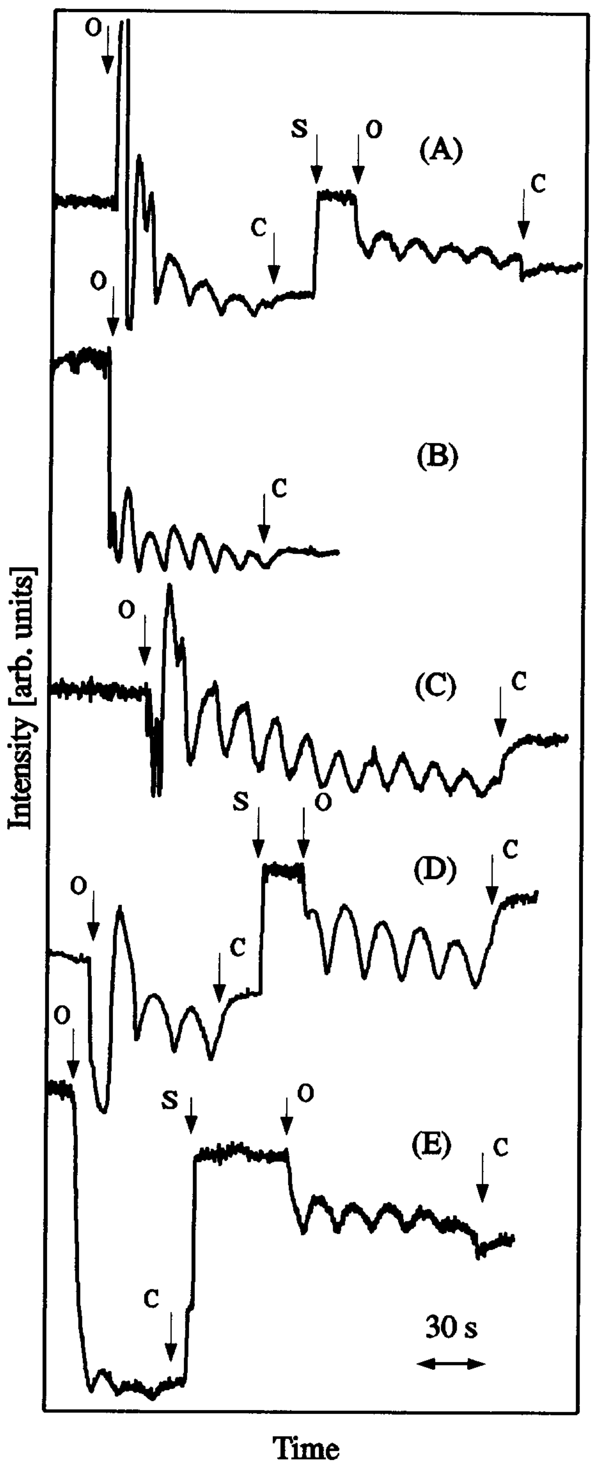

Figure 1. Intensity oscillations of the specularly reflected beam during growth of $YBa_2Cu_3O_{7-x}$. Arrows O and C indicate that the shutter was opened or closed, and S marks an increase of the sensitivity.

The whole RHEED pattern was taken by a charge- coupled device camera and stored with a video recorder. Observing the oscillations at the position of the specularly reflected beam, the growth of the films was stopped on the maximum or the minimum intensity by closing the shutter. On these films, STM measurements were made in a constant current mode with a current of 50- 100 pA and a bias voltage of 1.0- 2.0 V.

3. Results and Discussions

Figure 1 shows a series of intensity oscillations of the specularly reflected beam as a function of time during the growth of $YBa_2Cu_3O_{7-x}$ on $SrTiO_3(001)$ substrates. After the shutter is opened, strong damping and recovery is observed at the initial deposition. This behavior depends strongly on the angle of the incident beam, because the formation of crystalline Cu metal and Cu_2O islands causes some extra spots in the RHEED pattern, which influence the intensity oscillations. Details will be published elsewhere[8]. After these irregular oscillations, periodic oscillations are observed. Whenever the deposition is stopped by closing the shutter, the intensity recovers. This suggests that the migration of adatoms to surface steps increases completion. When the shutter is opened again, the regular oscillations continue. The fact that these oscillations are regular implies stable deposition rates. The intensity decays. This feature is due to the increasing roughness as will be discussed below.

The growth was eventually stopped as indicated in fig.1. Films (A), (C) and (D) were stopped on a maximum, and (B) and (E) on a minimum. STM images of the resulting surfaces are shown in fig.2. On the terraces of the surfaces stopped on the maximum, there are only a few small islands formed. On the other hand, many islands of various sizes formed on the terraces stopped on the minimum. These islands have a height of one unit cell of $YBa_2Cu_3O_{7-x}$. This means that the surface of the films stopped on the maximum is closer to monolayer completion and smoother. The existence of a few islands on the terraces may indicate that we have not hit the point of a maximum smoothness exactly. We may get smoother films by an appropriate timing for the shutter.

We compared the average width of almost complete terraces on the films with that on the substrates. The terrace width of the substrates is calculated from the angle of the misorientation,

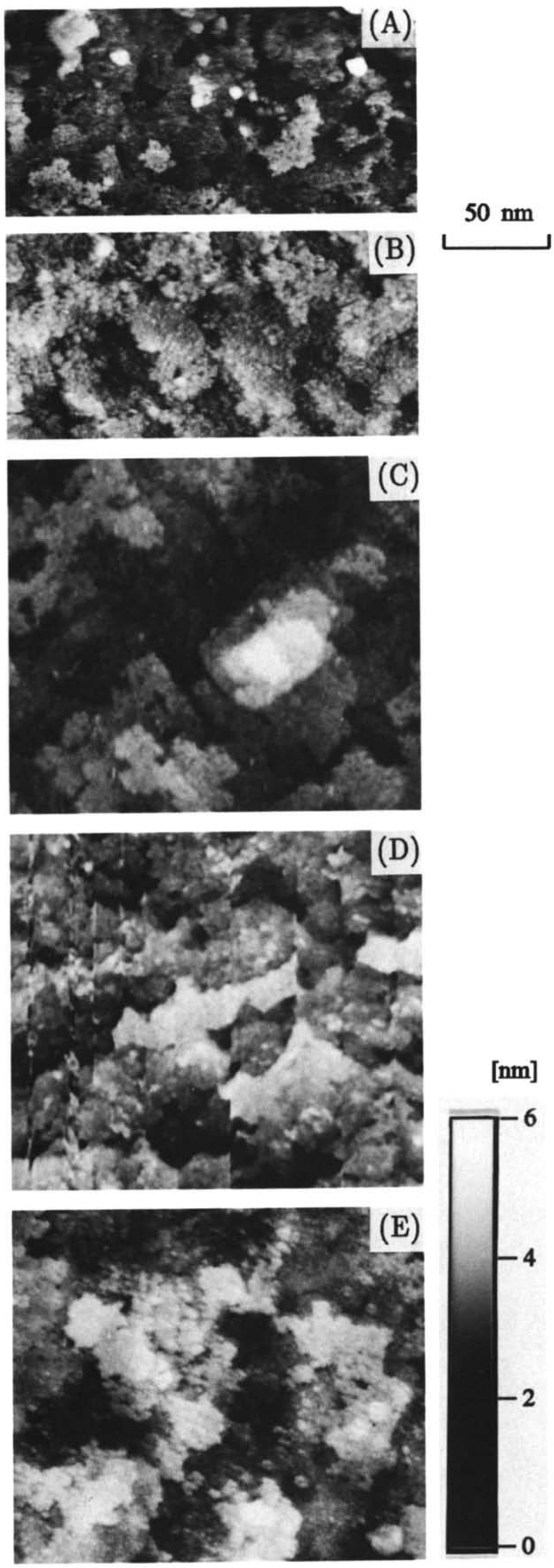

Figure 2. STM images of the $YBa_2Cu_3O_{7-x}$ films with growth stop as indicated in fig.1. The vertical scale on the upper- right side indicates the height variation.

assuming a constant slope. The calculated values are shown in Table I, together with the average terrace widths taken from fig.2. It is obvious that both quantities are closely correlated. This suggests that the terrace width of the film is almost determined by that of the substrate.

In general, the competition between the terrace width and the average diffusion length determines the formation of islands. Islands are nucleated when the terrace width is larger than the diffusion length. In this case, the succession of the island nucleation and layer completion leads to the RHEED oscillations as observed in our experiments. On the other hand, if the terrace width were shorter than the diffusion length, the film would grow by step flow without formation of islands and no RHEED oscillations would be seen. From the largest terraces without islands, we deduced a maximum diffusion length of about 15 nm. Therefore, the step flow can be expected only on misoriented surfaces of about 6 degrees. On our sample (B) with the largest misorientation of 1.85 degrees, we still observe the nucleations of islands. No attempt has been made to use larger misoriented substrates. Much disorder is expected to be induced in the $YBa_2Cu_3O_{7-x}$ films by steps.

We have not seen any screw dislocations on the samples discussed so far. Even on thicker films of 16 layers on $SrTiO_3$, we found only 0- 2 screw dislocations in an area of 10 x 10 μm^2. So, it is clear that screw dislocations are not initiated during the early epitaxial growth, but form later in the deposition process. This is in agreement with the observation of Zheng et al.[9] that screw dislocations in films made by laser ablation were still absent at 16 layers, but had appeared at 32 layers. However, they observed on their STM images that between 8 and 16 unit cells, the growth mode of $YBa_2Cu_3O_{7-x}$ changed from the layer by layer mode to the three- dimensional island formation (Stranski- Krastanov mode). Our results show that there is no such a transition in the RHEED observation and STM measurements between 6 and 16 unit cell layers. The height variation(peak to valley) in this thickness range is within three unit cells, which is much smaller than their results of 7- 10 nm in 16 unit cells thick film. This discrepancy may come from the difference in the deposition conditions between laser ablation and co- evaporation and hence the growth mechanisms of the films.

4. Summary

We have studied the surface of $YBa_2Cu_3O_{7-x}$ films by STM. The films were made with observing RHEED oscillations and stopping the growth on a maximum or a minimum. We found that the films stopped on the maximum were smoother and closer to monolayer completion of one unit cell than those on the minimum. The monolayer terrace width was almost determined by the average terrace width of the substrates. On the monolayer terraces, islands of one unit cell height form, which is considered to be due to the short diffusion length of $YBa_2Cu_3O_{7-x}$, compared with the average terrace width of the substrates. Screw dislocations are not present in the early growth of the film, but form rather later in the deposition process.

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