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Scanning tunneling microscopy of laser-deposited YBCO thin films

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High-quality YBCO thin films deposited on yttria-stabilized zirconia substrates by laser ablation have been studied by scanning tunneling microscopy in air. The films have been grown at different substrate temperatures, and the measured critical current density (J_c) and surface resistance (R_s) have been shown to vary with deposition temperature; where the J_c decreased with deposition temperature, the R_s increased. These variations were related to a change in the film surface morphology, showing screw dislocations for the films with lower deposition temperature, while at higher temperatures these dislocations were absent.

1. Introduction

Since the discovery of the high-temperature superconductors (HTS) in 1986 [1] there has been an extensive effort to refine the materials, where special emphasis has been put on the thin films. However, a number of important questions related to the microstructure of the films remains unresolved. One is the unknown nature of the defects responsible for flux pinning. Another question, that needs to be further addressed, concerns the growth and nucleation mechanisms.

Scanning tunneling microscopy (STM) [2] has proved to be an important method in characterizing these materials. Early STM studies have been concentrated on single crystals [3]. More recently, sputtered [4] and laser-deposited [5] YBCO thin films have been studied. The sputtered films contained a high density of screw dislocations, and it was proposed that some of the flux pinning was due to these dislocations. The laser-deposited films exhibited a similar surface morphology.

In this contribution, we present an STM study of YBCO thin films on yttria-stabilized zirconia substrates. The films have been grown by laser-depositing at different temperatures.

2. Experiment

2.1. STM

STM imaging was carried out on as-received films. The STM used here is described in greater detail elsewhere [6]. The microscope was operating in air and at room temperature. A standard piezo tube scanner assured a maximum scan length of 2.8 μm. For the coarse motion a commercial inch-worm motor was used (Burleigh Instruments, Inc.). To reduce the mechanical and acoustical noise the tunneling unit was hanging in thin rubber bands inside a closed box. The box was placed on a platform, which was suspended from the ceiling with rubber bands. A Macintosh workstation was used to control the measurements and for the data presentation. All images were measured in the constant-current mode, with an analog feedback system. The tips were commercially available etched Pt/Ir tips (Longreach Scientific Resources). A low-bias voltage was used (50 mV, tip negative), because at higher-bias voltages (700 mV) surface modifications occurred. The current set point was between 0.05 and 0.1 nA. The scanning speed was usually 2 μm/s. No

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filtering was employed in producing the final images.

2.2. Sample preparation

The films were deposited in a conventional laser-deposition system, consisting of a KrF excimer laser and a vacuum chamber (for more details see ref. [7]). The energy density at the target was 1.5 J/cm^2, the substrate-to-target distance 5 cm, the O_2 pressure 0.2 mbar and the laser pulse rate was 10 Hz. The deposition was 0.04 nm/pulse giving a deposition rate of 0.4 nm/s. After deposition, the O_2 pressure was increased to 1 atm and the temperature was ramped down to 100°C at 20°C/min. To avoid a possible target degradation the target was rotated during deposition and polished between each run. The substrates were YSZ (12% Y_2O_3) with (001) orientation. Five films were grown at substrate temperatures (T_{sub}) ranging from 710 to 765°C. The film thickness was 300 nm except for the film deposited at 740°C which had a thickness of 240 nm. All films were investigated by X-ray diffractometry and their critical temperature and microwave surface resistance (R_s) were measured. Three of these films were studied by STM. The temperature dependence of the resistance was measured by a four-point AC method, and the T_c ($R = 0$) was determined. The R_s was measured at 4.2 K by a parallel plate resonator method [8] at a frequency of 21.5 GHz. To measure the critical current density (J_c), the films were patterned using photolithography and ion-beam milling [9], whereby 6 μm wide bridges were defined. The J_c values of the bridges were determined using a 1 μV criterion. An evaporated Ag layer was used for contacts.

3. Results

All films had a T_c between 89.5 and 92 K (the highest was obtained for $T_{sub} = 740$°C). The X-ray diffractometry showed that all films were highly c-axis oriented. The two films made at the lowest temperatures, however, showed some traces of a-axis oriented grains. Fig. 1 shows R_s and J_c versus T_{sub}, and the data indicate that higher substrate temperature gives lower J_c and higher R_s values. The measurements are consistent in the sense that a higher J_c corresponds to a lower R_s.

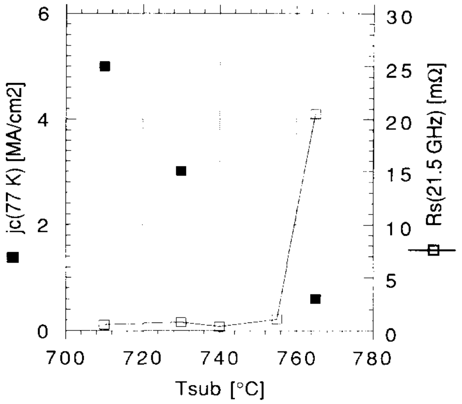

Fig. 1. Critical current density (J_c) at 77 K and surface resistance (R_s) at 4.2 K and 21.5 GHz, versus substrate holder temperature during deposition. J_c decreases and R_s increases with increasing deposition temperature.

Fig. 2. STM image of a film deposited at 730°C, showing the island growth with unit cell steps. Two screw dislocations (arrowed) are also visible. (Image size: 2 by 2 μm.)

Fig. 2 shows a film that was deposited at 730°C. The dominating growth process is island growth with unit cell steps. Two screw dislocations are also clearly visible, but the density is not high. The film with a deposition temperature of 740°C, had a similar growth but no screw dislocations were found. Fig. 3 shows a smaller area, where atomically flat terraces are separated by unit cell steps. At an even higher temperature (755°C), the surface obtained a smoother appearance (fig. 4).

4. Discussion

It is evident that the change in J_c and R_s are due to the change in substrate temperature (fig. 1). This change is expected to be related to a change in the surface morphology. The present STM images also show a related change; at lower temperature, the films contain screw dislocations (fig. 2), while at higher temperature the surface gradually becomes smoother and the screw dislocations disappear (figs. 3 and 4). These results support the proposal [4] that screw dislocations partially contribute to the pinning force.

Fig. 3. STM image of a film deposited at 740°C, with a higher magnification (image size: 200 nm by 200 nm). Atomically flat terraces are separated by unit cell steps.

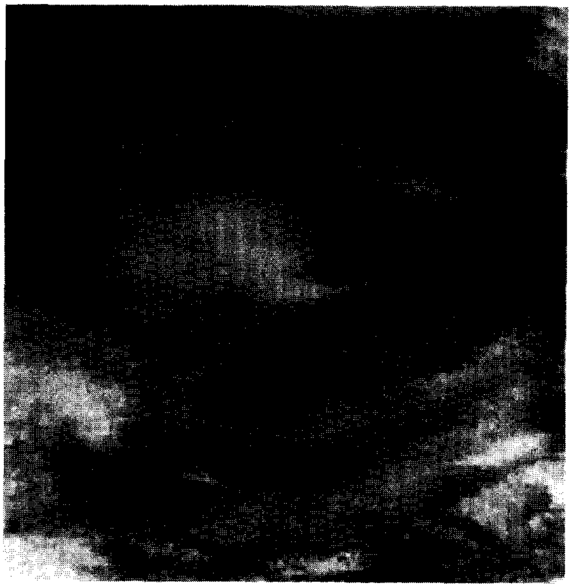

Fig. 4. STM image of a film deposited at 755°C, showing a smoother surface as compared to the films deposited at lower temperature (image size: 340 nm by 340 nm).

A recent transmission electron microscopic (TEM) study [10] on the present films, showed that a change in the film morphology occurred at a temperature of about 750°C. Above 750°C the films consisted of grains connected by low-angle grain boundaries, where the grain size was about 200 nm. At lower temperatures the films were single crystals. It is hard to see this transition to a polycrystalline film in the STM image in fig. 4, but when comparing figs. 2 and 3 with fig. 4 there is a significant change. The most striking appearance is the less good resolution, reflecting the finding that it was much harder to get stable tunneling conditions with this sample. One explanation for this phenomenon can be the less good coupling between the grains in the polycrystalline films.

The importance of the substrate misorientation for the film morphology has been shown recently [11]; the screw dislocation density decreased with increasing misorientation. In our case we did not study this behavior but we know that the misorientation angle in our samples was less than 1°, so it should not invalidate our present results.

5. Conclusions

We have studied high-quality laser-deposited YBCO thin films on YSZ substrates by scanning tunneling microscopy in air. The present STM results show that the substrate temperature during deposition plays an important role in determining surface morphologies and transport properties, e.g. J_c and R_s.

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