INFLUENCE OF DEPOSITION PARAMETERS ON PROPERTIES OF LASER ABLATED $YBa_2Cu_3O_{7-\delta}$ FILMS

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Abstract - $YBa_2Cu_3O_{7-\delta}$ films have been laser ablated under a variety of conditions, onto four different substrate materials. Using scanning tunneling microscopy it was observed that the films grow by an island nucleation and growth mechanism. The properties of the films were studied as functions of the deposition conditions. Films on $LaAlO_3$ had the best and most reproducible properties. The superconducting transition temperatures of films deposited on $LaAlO_3$ proved to be fairly insensitive to the substrate temperature during deposition; $T_c > 90$ K were obtained for films deposited over a temperature range from 760° to 850° C. The oxygen partial pressure during the deposition had a large effect on the transition temperature; the highest T_cs were obtained for films deposited in 26.7 Pa (200 mTorr) oxygen. Measurements of transport critical current indicate that films deposited at lower temperatures are less sensitive to magnetic fields, suggesting that they may contain more defects which act as flux pinning sites.

I. INTRODUCTION

The excellent properties of laser ablated $YBa_2Cu_3O_{7-\delta}$ films have resulted in many possible device applications. For most of these applications, film morphology is almost as important as electrical properties. Since the film morphology is determined by the deposition conditions, it is useful to examine the influence which various deposition parameters have on the properties of the films. In this study the influence of substrate material, substrate temperature and oxygen pressure during deposition, on the properties of $YBa_2Cu_3O_{7-\delta}$ thin films are examined.

II. EXPERIMENTAL

A frequency tripled Nd-YAG laser (355 nm) was used to ablate YBCO films onto four different substrate materials: single crystal (001) oriented $LaAlO_3$, $SrTiO_3$, MgO and Y_2O_3 stabilized ZrO_2 (hereafter referred to as LAO, STO, MgO and YSZ). Substrate preparation and standard deposition procedures are described elsewhere.[1] A single YBCO target (>90% dense) was used for all depositions. The substrate heater block temperature during deposition was varied from 730 to 875°C and the oxygen pressure from 6.7 to 40 Pa (50 to 300 mTorr). The deposition rate was 60 nm/min and the films were grown to thicknesses of ~200 nm. Throughout the study, depositions were periodically made under a standard set of conditions (substrate temperature 790°C, oxygen pressure 26.7 Pa (200 mTorr)) to ascertain that uncontrolled changes in the system had not altered the films.

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Manuscript received August 24, 1992.

The as-deposited films were examined by scanning electron microscopy, SEM, and scanning tunneling microscopy, STM. The STM images were taken in air with Pt/Ir tips. The resistive transition of each film was measured using a four probe ac technique with an applied current of 50 μA; the transition temperature, T_c, was taken to be the temperature at which the resistance was less than 1 mΩ. Transport critical current density, J_c, measurements were made in liquid nitrogen, on films patterned into 100 μm strips. Magnetic field was applied parallel to the c axis and perpendicular to the current flow. An electric field criterion of 1 μV/cm was used.

III. RESULTS AND DISCUSSION

A scanning electron micrograph of a 200 nm thick film deposited on LAO at 790°C, under 26.7 Pa (200 mTorr) oxygen is shown in Fig. 1. The image is typical of the films

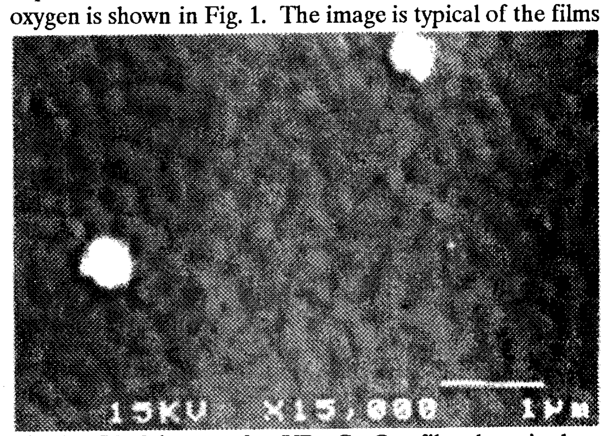

Fig. 1 SEM image of a $YBa_2Cu_3O_{7-\delta}$ film deposited on LAO at 790°C in 26.7 Pa (200 mTorr) oxygen for 3 min.

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studied. There are several large features, commonly referred to as boulders, which were seen in most of the films. The size and density of the boulders varied from film to film and frequently within a single film. At a much lower contrast level a granular structure is evident. It was necessary to increase the contrast of the SEM almost to its maximum to image these features.

The size of the granular features observed in the SEM, ~0.2 μm, corresponds to the size of the growth features imaged by STM. A scanning tunneling micrograph of the same film imaged in Fig. 1 is shown in Fig. 2. With the enhanced vertical resolution of the STM image, it is evident that the granular structure of Fig. 1 is the result of island nucleation and growth features which have not completely coalesced. This island type of growth, which has been reported previously for laser ablated $YBa_2Cu_3O_{7-\delta}$ films,[2,3] was observed in all of the films examined in this study. No screw dislocation growth features were observed. The diameter of the islands was found to vary with the substrate material and the deposition temperature, lower

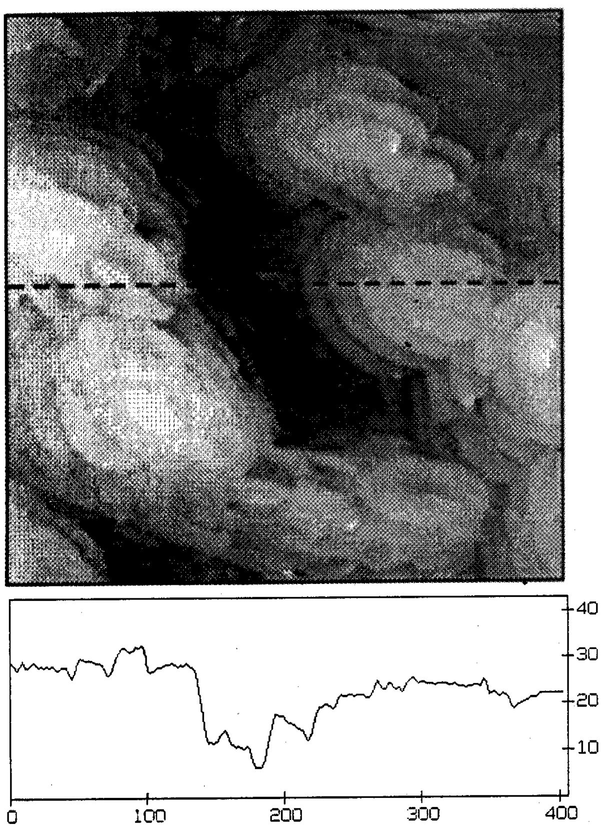

Fig. 2 Scanning tunneling micrograph of a 400 nm x 400 nm area in the same film imaged in Fig. 1. The graph below the image is a surface profile taken along the dashed line; the scales are in nm.

substrate temperatures during deposition resulted in films with smaller islands. A detailed report of the morphology of these films is given elsewhere.[1]

In all of the films deposited on LAO and STO no second phases or misoriented grains were observed by SEM or STM. However, *a*-axis grains were found in films deposited on MgO and YSZ at extremely high and low temperatures. In general the morphology of most of the films examined indicates that the films are of high quality, as is reflected by the electrical properties of the films.

The superconducting transition temperatures of the films were frequently as high as 92 K. The influence of the substrate temperature during deposition, on the T_cs of films grown on LAO, is shown in Fig. 3. All of the films represented here were deposited under 26.7 Pa (200 mTorr) oxygen. Films deposited at temperatures above 760°C appear to be fairly insensitive to the substrate temperature. Over a range of deposition temperatures of 100°C (760° to 875°C) the films had T_cs above 91 K. It was not possible to deposit films at temperatures above 875°C due to heater limitations. Films deposited at temperatures below 760°C had T_cs supressed by 4 or 5 degrees.

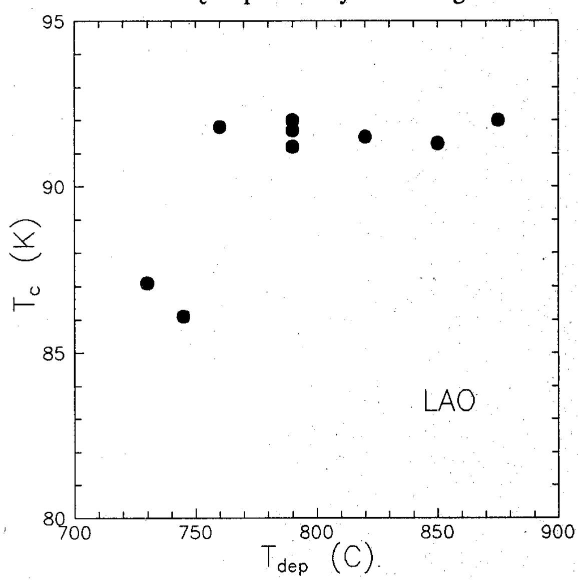

Fig. 3 Resistive transition temperatures of films grown on LAO substrates under 26.7 Pa (200 mTorr) oxygen as a function of the substrate temperature during deposition.

The reduction in T_c for lower deposition temperatures, was even more pronounced for films on other substrates. The T_cs for films deposited under 26.7 Pa (200 mTorr) oxygen, at three different temperatures on each of the four substrate materials studied are shown in Fig. 4. For each of the substrates a film with a T_c of 90 K or higher was obtained by depositing at 790°C. However, unlike the films grown on LAO, there was a large degree of variability between the T_cs for different films deposited on STO, MgO and YSZ at this temperature. The variability being largest for films on YSZ and smallest for those on STO. Depositing at 730°C reduced the T_cs of films on all substrates, while depositing at 875°C produced films with high transition temperatures on all substrates except MgO.

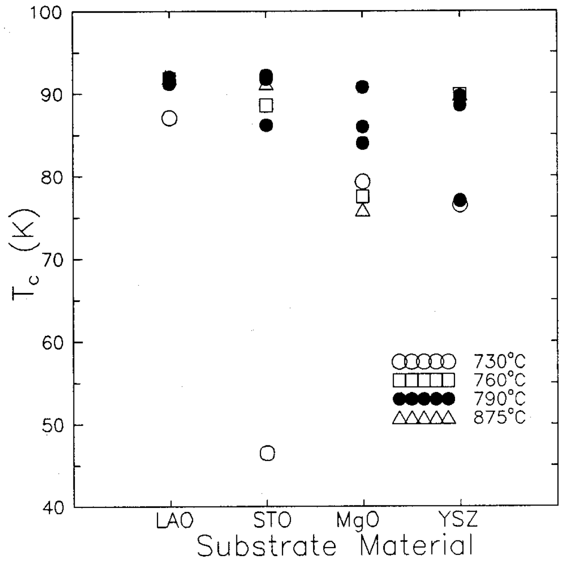

Fig. 4 T_cs of films grown under 26.7 Pa (200 mTorr) oxygen at three different temperatures on LAO, STO, MgO, and YSZ substrates.

It is possible that the variability in the T_cs of films deposited at 790°C on STO, MgO and YSZ, actually result from small changes in the substrate temperature. As shown in Fig. 3, the transition temperatures of films on LAO are quite insensitive to the deposition temperature above 760°C. It is possible that films on the other substrates are more sensitive to temperature. Consequently, relatively small changes in deposition temperature would result in larger variations in the T_cs. This effect may be magnified by differences in the emissivities of the substrate materials, which would cause further differences in the surface temperatures of the substrates. Likewise, differences in the substrates, such as defect content, polish angle, or cleaning, may also be responsible for the variations in the films on substrates other than LAO.

The oxygen pressure during the deposition was also found to have a substantial effect on the transition temperatures of films on the four substrate materials. As shown in Fig. 5, a pressure of 26.7 Pa (200 mtorr) produced films with the highest T_cs. All of the films represented here were grown at a substrate temperature of 790°C. For films grown on all substrates except YSZ, both higher and lower oxygen pressures resulted in lower transition temperatures. Films grown on LAO showed the greatest reproducibility, and the least reduction in T_c with changes in P_{O2}. All films deposited on LAO in 26.7 Pa (200 mTorr) oxygen had transitions above 90 K. Films on both STO and MgO showed more variability in the transition temperatures of films deposited at this pressure, and also larger drops in the transition temperature with changes in P_{O2}. Films on YSZ behaved similarly to those on STO and MgO, except that the transition temperature of a film deposited at 40 Pa (300 mtorr) had a T_c as high as any film deposited on YSZ. For all substrate materials, using only 6.7 Pa (50 mtorr) oxygen during the deposition produced films with greatly reduced transition temperatures.

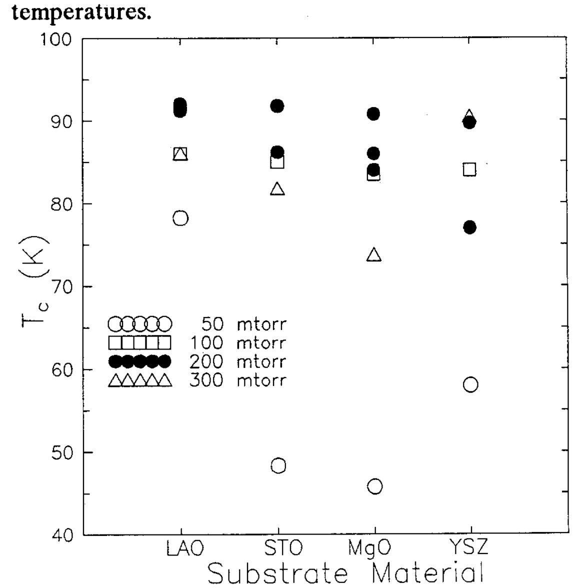

Fig. 5 T_cs of films grown on LAO, STO, MgO, and YSZ substrates at 790°C under different oxygen pressures.

It is anticipated that the $YBa_2Cu_3O_{7-\delta}$ films become fully oxygenated during the slow cool in oxygen after deposition. Therefore, the strong influence of the oxygen pressure during deposition is surprising. However, the fact that the film T_cs decrease as the oxygen pressure is increased as well as when it is decreased indicates that the change in pressure may be affecting something other than the oxygen content of the film. Attempts were made to improve the transition temperature of the material by depositing films under low pressure at high temperature, however, the change in the T_c was negligible.

The transport critical current densities of the films on LAO were measured in liquid nitrogen (76 K). The J_cs in an applied field of 0.5 T as a function of the film deposition temperature, are shown in Fig. 6. The three data points at 790°C are from the control samples deposited at various times during the period of the experiment; the scatter in these data represent the reproducibility of J_c. The film deposited at 760°C appears to have an anomalously low current density. This sample also had unusually large islands. Disregarding this data point, the general trend of the data indicates that as the deposition temperature of the film is decreased, the critical current density in field is increased.

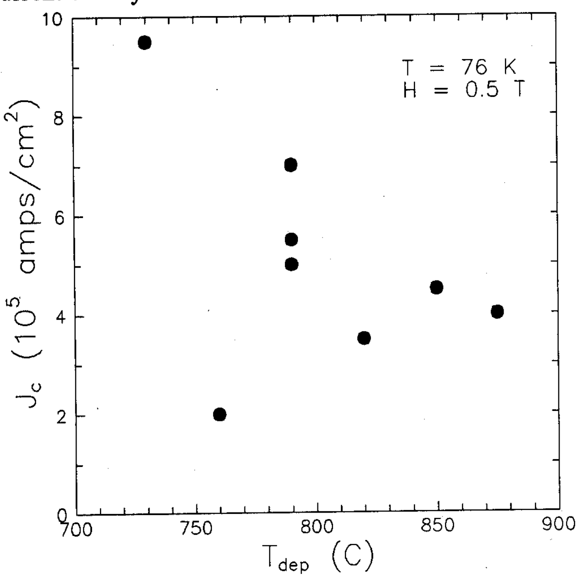

Fig. 6 Transport J_cs as a function of film deposition temperature. All the films were grown under 26.7 Pa (200 mTorr) oxygen on LAO substrates. The J_cs were measured at 76 K in 0.5 T applied field.

This is consistent with a study by Mannhart et al. of pinning in sputtered $YBa_2Cu_3O_{7-\delta}$ films on STO.[4] The films in their study exhibited a screw dislocation growth mechanism, and it was found that as the screw dislocation density increased, the transport J_c of the films, in 0.5 T applied field, also increased. A previous study had shown that the density of screw dislocations in these samples decreased with increased growth temperature.[5] Therefore, Mannhart et al. concluded that the pinning defects must be associated with the misoriented screw dislocation growth spirals. Our present results, on films which grow by an island nucleation and growth mechanism, indicate that the screw dislocations and defects associated with their growth spirals are not necessary for enhanced pinning. Instead it appears likely that the nonequilibrium nature of the film growth allows for incorporation of numerous defects, both line and point type, and that more defects are trapped in the films at lower deposition temperatures. The exact nature of the defects primarily responsible for the pinning are under study.

V. SUMMARY

The substrate material, substrate temperature and oxygen pressure used for deposition of $YBa_2Cu_3O_{7-\delta}$ thin films, all have a strong influence on the morphology and properties of the films produced. Films on LAO are the least sensitive to changes in deposition parameters. The substrate temperature can be varied over more than 100°C and still yield films with T_cs of 91 K. However, the transition temperature drops rapidly as the oxygen pressure is either increased or decreased. The highest transport critical current densities in field were obtained for films deposited at low temperatures.

ACKNOWLEDGMENTS
The authors would like to thank Ted Stauffer for help with transport critical current measurements and Kristie Kimminau for help with sample patterning.

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