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TITLE INFLUENCE OF ION BEAM MIXING ON THE GROWTH OF HIGH
TEMPERATURE OXIDE SUPERCONDUCTING THIN FILM

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INFLUENCE OF ION BEAM MIXING ON THE GROWTH OF HIGH TEMPERATURE OXIDE SUPERCONDUCTING THIN FILM

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ABSTRACT

The superconducting properties of high temperature superconductor thin films are dependent on the quality of the substrate used to grow these films. In order to maximize the lattice matching between the superconducting film and the substrate, we have used a $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin film deposited on $\langle 100 \rangle \text{SrTiO}_3$ as a template. The first film was prepared by co-evaporation of Y, BaF_2 and Cu on $\langle 100 \rangle \text{SrTiO}_3$, followed by an anneal in "wet" oxygen at 850°C . This film showed a sharp transition at about 90 K. A thicker layer of about 5000 Å was then deposited on top of this first 2000 Å film, using the same procedure. After the post anneal at 850°C , the transition took place at 80 K and no epitaxy of the second film was observed. Ion beam mixing at 400°C , using 400 keV O ions was done at the interface of the two films (the second one being not annealed). After the post anneal, the film displayed an improved T_c at 90K. Moreover, epitaxy was shown to take place from the interface SrTiO_3 -123 film towards the surface and was dependent of the dose. These results will be discussed from the data obtained from Rutherford backscattering spectroscopy (RBS) combined with channeling experiments, x-ray diffraction (XRD) and scanning electron microscopy (SEM) observations.

INTRODUCTION

The quality of thin film high temperature superconductor (HTS) is dependent on the crystallographic orientation and the quality of the template¹. Because of the difference in lattice parameters between the HTS and the template, one cannot expect a perfect epitaxy of the film. A possible solution to the lattice mismatch problem is to use a high quality HTS film as a template for the growth of a second thicker film. In the present work, we compare the quality and the properties of two 7000 Å thick HTS films grown in two different ways: one by deposition of one thick HTS film and the other by deposition of two HTS layers, the first film being a template for the second one.

EXPERIMENTAL DETAILS

In the single step process, Y, BaF_2 and Cu were co

-evaporated up to a thickness of 7000 Å on single crystals of <100> SrTiO₃. The as-deposited stoichiometry and the thickness were determined by RBS measurements using 6 MeV alpha particles to be Y₁Ba_{1.98}Cu_{2.92} and 7000 Å respectively. The film was annealed at 850 °C. In the two step process, a 2000 Å film with a composition Y₁Ba_{1.77}Cu_{2.96} was first deposited on SrTiO₃ in a similar way. This film was then annealed at 850 °C in order to produce a highly oriented crystalline superconducting film. A second film was then evaporated on top of the first producing a total thickness of 7000 Å. The stoichiometry of this second film was determined to be Y₁Ba_{1.79}Cu_{2.96}.

Prior to oxygen annealing the 2-layered structure, half of the samples were ion irradiated in an attempt to ion mix the interface between the HTS film and the deposited Y-BaF - Cu film. Interface irradiation was carried out using 400 keV O ions with the sample held at 400 °C in a vacuum of 5x10⁻⁷ torr. Irradiation doses of 5x10¹⁴ and 1x10¹⁵ ions/cm² were employed.

The 2-layered samples were then annealed in oxygen at either 850 or 900 °C. The 850 °C anneal consisted of leaving the film at 750 °C for 1 hour in dry O₂, followed by 850 °C for 1 hour in flowing O₂ which had been bubbled through de-ionized water ("wet" oxygen anneal^{4,5}) and cooling slowly down to room temperature in dry O₂. In the 900 °C anneal, the film was left for 45 minutes at 900 °C in "wet" O₂; the sample was cooled down to room temperature in "wet" O₂ until 750 °C and then, in dry O₂. The annealing notations used in the rest of the paper will be T₁/T₂: T₁ being the anneal temperature of the 2000 Å thick film and T₂ the anneal temperature of the 5000 Å thick film.

Characterization was carried out using RBS, high energy backscattering^{2,3} and ion channeling techniques, resistivity versus temperature measurements and SEM observations.

RESULTS AND DISCUSSION

The four-point probe results are summarized in Table I

	T _{anneal}	T _c	c:a ratio
2000 Å	850 °C	89	34:1
2000 Å / 7000 Å	850 °C / 850 °C	76	17:1
	850 °C / 900 °C	82	30:1
7000 Å	850 °C	86	3:1

Table I. Critical temperature and c:a ratio for the non irradiated films.

Tanneal	Dose	T _c	c:a ratio
850 °C / 900 °C	5x10 ¹⁴ O/cm ²	82	35:1
	1x10 ¹⁵ O/cm ²	88	

Table II. Critical temperature and c:a ratio for the irradiated films.

and II. They showed the 2000 Å film to be a good superconductor with a sharp transition (2 - 3 K) and a high critical temperature of about 90 K. The films made with the two-step process showed different T_c according to the ion beam and annealing procedures used. The T_c for irradiated films was 82 K and 88 K for low dose and high dose treatments respectively. No significant improvement in the onset of the transition was observed in the ion mixed films. The 7000 Å thick film produced in a single deposition step showed a transition at around 87 K.

Channeling techniques using 8.8 MeV alpha particles aligned with the <100> direction of the SrTiO₃, were used to examine the epitaxial quality of the films. The 2000 Å thick films showed reasonable alignment with the underlying substrate displaying a minimum yield (x_{min}) of 38 %. The thicker 7000 Å film grown in one single step, displayed a x_{min} of 70 %, suggesting that a better epitaxy is achieved with thinner films.

Channeling data obtained on the 850/900 films, both with and without ion mixing, are reported in Fig. 1. In the

Fig. 1. Ion channeling results in the non irradiated and

non-irradiated samples the channeling yield of Cu, Y, and Ba in the top layer is equal to the random yield while channeling is observed in SrTiO_3 . These data suggest non-alignment of the film with the substrate. Similar trends were observed for the other T_1/T_2 films. Figure 1 also displays the aligned spectra of the two irradiated films where significant changes in the Y and Ba peaks are apparent. Channeling in the top layer is visible with a maximum occurring at the interface between the two layers. Increased dechanneling occurs at shallower depths suggesting that some epitaxial growth has occurred in the top film which deteriorates near the surface. This effect is greater in the high dose sample.

The epitaxial quality of the HTS films was further examined using x-ray diffraction pole figure measurements carried out in a standard back-reflection goniometer. For each sample, the c-axis to a-axis orientation ratio was determined by scans over the maxima in the (102)/(012) pole figure. Table I and II list the c:a ratio measured for each film as well as the measured T_C . These data clearly show that there is a relationship between these two parameters. The 2000 Å film exhibited a high T_C and a high c:a ratio. However, the 7000 Å films deposited in a single step exhibited a poor c:a ratio and a reduced T_C .

An examination of the T_1/T_2 data shows that films which were annealed without ion mixing possess a varied quality (Table I) which improved with increasing processing temperature. A comparison of the 850/900 films shows greater c-axis orientation for ion mixed films relative to non-irradiated films with a c:a ratio of 35:1 versus 30:1 respectively. The c:a ratio measured for the ion mixed film corresponds to an average value of the ratio for the two doses.

The surface morphology of the HTS films has been examined using SEM and is reported on Figure 2. These data show evidence of a pebble structure on the 2000 Å thick film. The T_1/T_2 films display a needle structure which lacks the "basket-weave" pattern found in oriented films. The 850/900 film displays a more compacted structure with thicker needles while the 850/850 film has smaller needles. The 7000 Å film prepared in one step displays a morphology composed of needles oriented in a "basket-weave" texture and pebbles. The texture of the irradiated films is not different from the non irradiated 2-layer films. However, in the case of the high dose, some small areas of pebbles are present.

These results have shown that best quality films are obtained with thin films around 2000 Å and are consistent with other observations made on BaF_2 processed films^{4,6,7,8}. With increasing thickness, the amount of a-axis growth increases and the c:a ratio decreases. These experiments have shown that it is not possible to achieve good epitaxial layer by growing a second film on top of a high quality HTS film. Structures produced by this technique lack channeling and possess a degraded c:a ratio relative to films of a similar thickness grown in a single step. However, epitaxy can be improved by using ion irradiation to mix the interface prior to the final oxygen anneal. The c:a ratio is improved and the film starts channeling from the interface film/substrate towards the surface. Presently, experiments are being performed in order to study the dose and the temperature dependence on the

Fig. 2. SEM micrographs of the different HTS films.

CONCLUSION

Studies have been carried out on $Y_1Ba_2Cu_3O_7$ films evaporated in two steps on $SrTiO_3$. The data showed that it was not possible to grow a good film in this manner. Two films have been irradiated with 400 keV O ions while being heated at 400 °C. X-ray pole figure analysis, channeling experiments showed that ion beam mixing can improve the epitaxial quality of these films relative to the films that have not been irradiated.

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