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Interfacial Properties of $YBa_2Cu_3O_{7-x}$ Thin Films on Al_2O_3 Substrates Prepared by Pulsed Laser Deposition

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The interfaces of $YBa_2Cu_3O_{7-x}$ (YBCO) superconducting thin films grown on $(1\bar{1}02)$ r-plane Al_2O_3 by pulsed laser deposition have been investigated by a transmission electron microscopy and an Auger electron spectroscopy depth profile. We used the $PrBa_2Cu_3O_{7-x}$ (PBCO) buffer layer to prevent the interdiffusion and compared the interfaces of YBCO/Al_2O_3 and YBCO/PBCO/Al_2O_3. The intermediate layer in the YBCO film deposited on bare sapphire is visible between the film and the substrate but no boundary layer in the film grown on PBCO buffered sapphire was observed directly by the cross-section image of TEM. The thickness of the intermediate layer in the film on bare sapphire is about 30 nm. This result of TEM observation is consistent with that of AES depth profile.

Key words: Buffer layer, interfacial properties, pulsed laser deposition, superconducting thin film

INTRODUCTION

Since the discovery of the cuprate superconductors, there has been notable progress in the ability to grow high quality superconducting thin films on various substrates. For the applications of the high-T_c superconducting thin films, especially microwave applications, such as filters, resonators, delay lines, and transmission lines, specific requirements on the substrate materials should be considered.[1,2] The requirements on the properties of substrate materials for high temperature superconducting microwave applications are mechanical strength, good lattice matching for the possibility of high quality film growth, and dielectric properties of the substrate.[3,4] Among different substrates, sapphire is a desirable dielectic substrate for the microwave application of high temperature superconducting thin films at high frequency due to its low dielectric constant, good mechanical strength, and low loss tangent.[5–7] However, a suitable buffer layer is required on sapphire substrates since sapphire reacts more easily with $YBa_2Cu_3O_{7-x}$(YBCO) thin film than any other substrates. For the formation and the microwave application of high quality superconducting thin film on sapphire, blocking interfacial reaction between the films and substrates should be considered because interfacial reaction may cause the degradation of YBCO superconducting thin film. Therefore, it is important to investigate accurately the interfacial properties of YBCO superconducting thin films on Al_2O_3 substrates with or without $PrBa_2Cu_3O_{7-x}$ (PBCO) buffer layer for high temperature superconducting microwave application.

In this paper, we present the results of the study of the interdiffusion between the YBCO thin films and Al_2O_3 substrates with or without PBCO buffer layer

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by observing the cross-section images of transmission electron microscopy (TEM) and Auger electron spectroscopy (AES) depth profiles. The results of AES observation were compared with those of the cross-section TEM. Resistivity measurements as a function of temperature were also performed to investigate the superconducting properties of YBCO thin films.

EXPERIMENT

Superconducting YBCO thin films were deposited on r-plane $(1\bar{1}02)$ Al_2O_3 substrates by a pulsed laser deposition method. A multi-target holder was installed to mount a YBCO target and a PBCO target simultaneously in the vacuum chamber as described before.[8] The sapphire substrates were cleaned in ultrasonic aceton, methanol, and deionized water before placing into the vacuum chamber. Prior to every film deposition, the target surface was slightly polished by scraping for the reproducibility of the laser plume and the film quality. In order to clean the YBCO target surface, 200 shots of focused laser beam were continuously fired on rotated YBCO target prior to film deposition. The PBCO buffer films of 20~500Å thickness were deposited at a substrate temperature of 750°C. Afterward, the YBCO thin film was deposited in the same condition of the PBCO deposition. A XeCl excimer laser with the wavelength of 308 nm was used to deposit both YBCO and PBCO layer. The excimer laser was fired at 5 Hz repetition rate for both YBCO and PBCO film deposition. The substrate temperature during deposition was measured using a thermocouple inserted into the substrate holder and a pyrometer focused on the substrate surface. These two different techniques gave consistent reading of the substrate temperature. Pulsed laser deposition was carried out in an oxygen ambient, which was controlled through a precision mass flow controller. An oxygen pressure of 200 mTorr was maintained during the deposition.

The films on sapphire have been investigated by conventional four-point probe method to measure the resistivity as a function of temperature. The cross-section TEM was carried out to observe directly the boundary layer between the film and the substrate. The interfacial properties were further examined by the AES which showed the interdiffusion between YBCO films and sapphire substrates with PBCO buffer layer and without PBCO buffer layer.

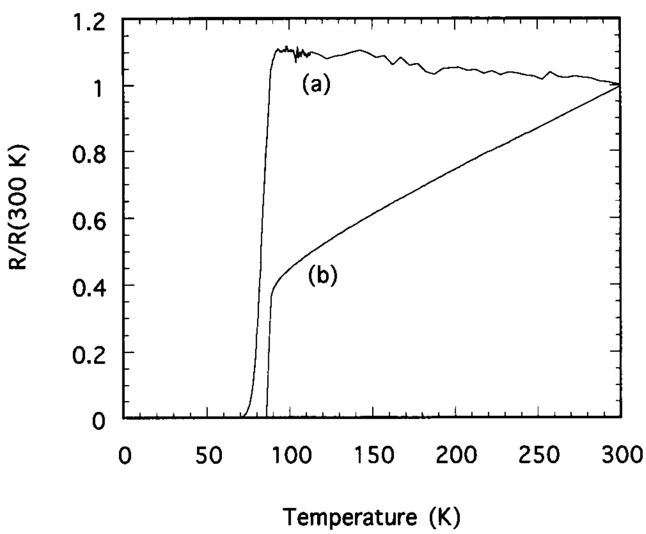
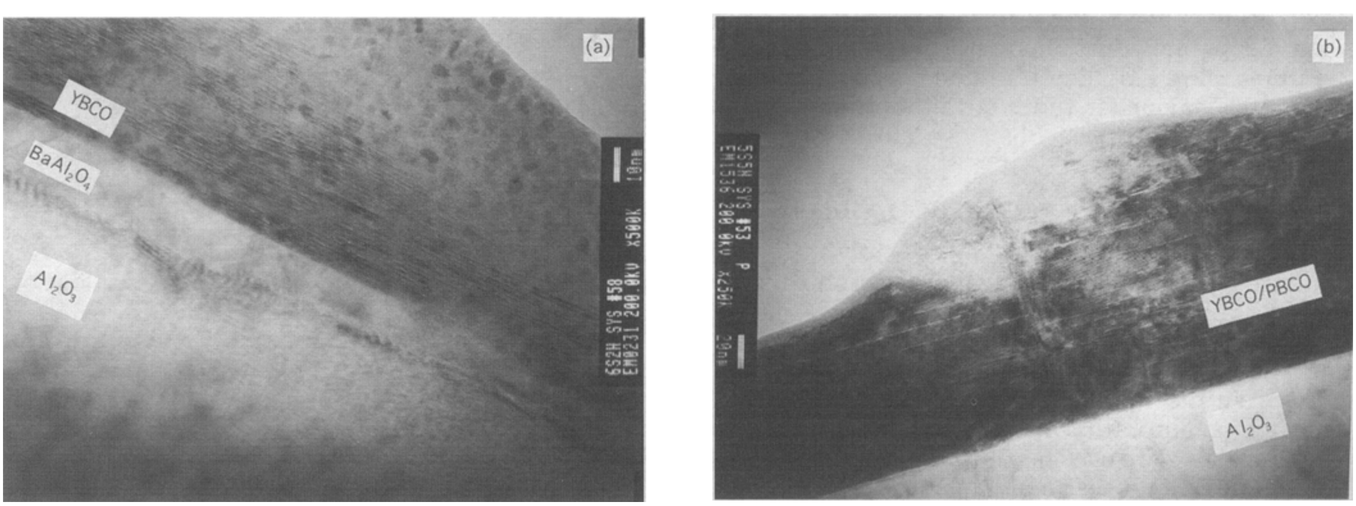


Fig. 1. Normalized resistivity vs temperature curves of (a) YBCO film on sapphire substrate, and (b) YBCO film on 200Å thick PBCO buffered sapphire.

RESULTS AND DISCUSSION

Resistance Measurements

The resistance curves as a function of temperature normalized to R(300K) were presented in Fig. 1. Figure 1a shows a typical four-probe resistance vs temperature (R-T) curve for a YBCO film deposited on bare sapphire. Most of the YBCO films on bare sapphire showed a T_c of about 70–75K. The R-T curve



a b

Fig. 2. Typical cross-section images of TEM of (a) $YBCO/Al_2O_3$ and (b) $YBCO/PBCO/Al_2O_3$. The interfacial boundary layer of about 300 nm thickness is observed clearly between YBCO thin film and sapphire substrate in (a). However, no diffusion layer was observed in (b).

showed a flat slope above the transition temperature and a long tail near T_c. However, the electrical properties of the YBCO film were greatly improved if a PBCO buffer layer was used. Figure 1b shows a typical R-T curve for a YBCO thin film on PBCO/Al_2O_3, where PBCO layer was deposited under identical conditions. The PBCO layer thickness is 200Å and the YBCO thickness is 2500Å. As can be seen from Fig. 1, it is obvious that the use of PBCO buffer has a very important influence on the YBCO film quality. Our best film on PBCO buffered Al_2O_3 showed a T_c of 87K with a metallic behavior.

Interface Observed by the Cross-Section TEM

A cross-sectional TEM observation reveals the preferential c-axis growth of the YBCO film from the substrate if the PBCO buffer was used. The cross-section images of high temperature superconducting thin films are presented in Fig 2a and Fig. 2b. Figure 2a shows the cross-section image of TEM of YBCO thin film grown on bare sapphire. Interfacial layer is clearly visible between the substrate and the film in Fig. 2a. The thickness of the interfacial layer was directly measured to be about 30 nm from the cross-section image of TEM. This interfacial layer was produced due to substantial diffusion of Al into the film and formation of an uncontrolled intermediate layer of $BaAl_2O_4$.[9] Dovidenko et al. suggested that this layer impeding the epitaxy is partly responsible for the deterioration of crystalline quality of the YBCO thin film, which may lead to substantial degradation of YBCO properties.

In comparison with the YBCO film on bare sapphire, the film grown on PBCO buffered sapphire displayed no boundary layer at the interface and the interface between the YBCO film on PBCO buffer and the substrate is sharp. Since PBCO is structurally and chemically compatible with YBCO, hardly any interface between YBCO and PBCO could be observed. This result shows that PBCO buffer improves interfacial property of YBCO film, which enhances substantially the superconducting properties of the film on sapphire. Further interdiffusion analysis of the films using AES depth profile also showed very different behavior for the interface between YBCO and Al_2O_3 and the interface between YBCO and PBCO buffered Al_2O_3.

Interdiffusion Between the YBCO Film and the Sapphire Substrate

Figures 3 and 4 show the Auger depth profiles of the YBCO film on bare sapphire and the YBCO film on 200Å thick PBCO buffered sapphire, respectively. The AES was carried out by a Microlab 310D system of VG Scientific Ltd. The AES depth profile was taken using a filament LaB6 with a 10-keV electron beam. 5 keV Ar^+ ions were used for the ion etching. Accurate milling rates were estimated using a SiO_2 wafer which has known oxide thickness as a reference. Slow ion milling rate of 6.9 nm/min for the thin film was used for the precise measurements based on 6 nm/min milling rate for SiO_2. The depth was checked by measuring the film thickness directly from a cross-section image display in a TEM and a film thickness monitor of α-step profile. The Auger depth profiles of the YBCO films on sapphires are shown in Fig. 3 and Fig. 4. The YBCO film grown on bare sapphire is shown in Fig. 3 and the YBCO film on 200Å thick PBCO buffered sapphire is shown Fig. 4. In the AES crater edge profiles, the Y, Ba, Cu, Pr, and Al line profiles are observed. It should be noted that the Y:Ba:Cu:O ratios are constant in these films. For the YBCO film on the bare sapphire, the interface between film and substrate was not very sharp and Al, Ba, and Cu were observed within an interdiffusion range of 30 nm thickness as shown in Fig. 3. This result is consistent with the boundary layer thickness observation from the cross-section TEM. From the Auger spectrum of the YBCO/Al_2O_3, it reveals that the reaction between the thin film and the substrate is

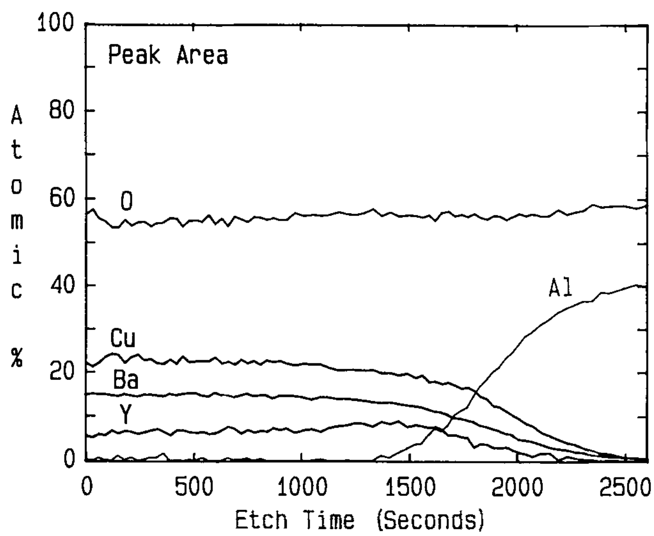


Fig. 3. Auger depth profile of YBCO/Al_2O_3. The long tail of aluminum into the YBCO film is clearly observed.

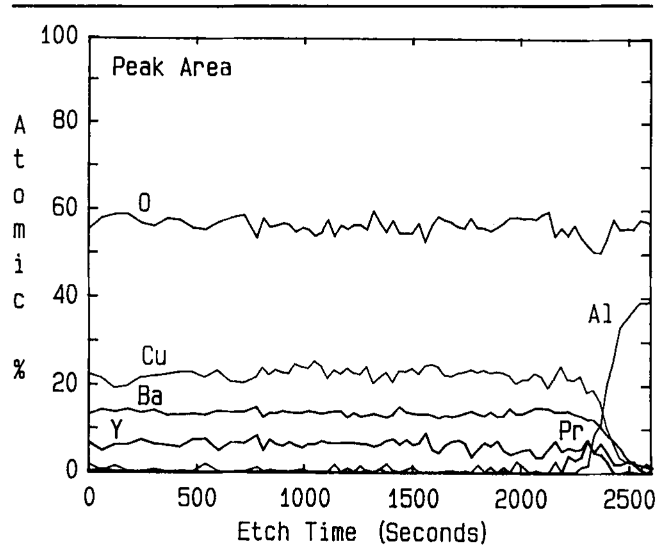


Fig. 4. Auger depth profile of YBCO/PBCO(200Å)/Al_2O_3. The interface between YBCO film and sapphire substrate becomes sharp by using a PBCO buffer.

observed by the diffusion of aluminum from the substrate into the film.

To prevent such a reaction, we used a PBCO intermediate layer as a diffusion barrier. Since PBCO has the same perovskite structure and similar lattice constant as YBCO, it enforces heteroepitaxial growth of the overlying YBCO layer without a significant structural disorder. By using such a buffer layer, the interaction between YBCO and sapphire can be effectively blocked. However, as we already mentioned, the dielectric properties of the buffer cannot be ignored for the microwave application. Therefore, we have systematically changed the PBCO buffer thickness to find a minimum thickness to preserve the YBCO superconducting properties.

In Fig. 4, Auger depth profile of YBCO superconducting thin film on sapphire with PBCO buffer layer is shown. The interfaces between YBCO/PBCO and PBCO/Al_2O_3 are sharp compared with the interface of YBCO/Al_2O_3 as shown in Fig. 3. In the YBCO superconducting thin film on 200Å thick PBCO buffered sapphire, aluminum peak-to-peak value abruptly decreases to the noise level in the PBCO buffer layer as shown in Fig. 4. No aluminum has diffused through the 200Å thick PBCO layer to the YBCO film and the Ba and Pr peak-to-peak values also decrease to the noise level in the sapphire substrate. This indicates that 200Å thick PBCO layer is sufficient to prevent the aluminum from diffusing into the YBCO layer.

CONCLUSIONS

We have compared the properties of the interface between YBCO thin film and sapphire substrate and the interface between YBCO film and PBCO buffered sapphire by an AES depth profile and a cross-section TEM. Superconducting YBCO thin films on $(1\bar{1}02)$ r-plane Al_2O_3 with or without PBCO buffer were grown by *in situ* laser deposition using a XeCl excimer laser. We used the PBCO buffer to prevent the interdiffusion and compared the interfaces of YBCO/Al_2O_3 and YBCO/PBCO/Al_2O_3. The intermediate layer in the YBCO film deposited on bare sapphire is clearly visible between the film and the substrate but no boundary layer in the film grown on PBCO buffered sapphire was observed from the cross-section images of TEM. The thickness of the intermediate layer in the film on bare sapphire is about 30 nm. This diffusion depth observed by TEM is well agreed with that observed by AES depth profile. Compared with the YBCO film directly grown on sapphire, the interfacial and superconducting properties of the films on sapphire with the PBCO buffer layer are greatly enhanced. From the AES depth profile, it has been observed that no diffusion occurred between the YBCO and sapphire substrate by the use of an intermediate buffer layer of PBCO. These improvements are attributed to the perovskite structure of the PBCO material same as that of the YBCO. It has been verified that the enhancement of the YBCO film property due to the use of PBCO buffer is caused by the clear interface between the film of YBCO/PBCO and the sapphire substrate with no boundary interdiffusion layer observed by TEM. High quality superconducting film with such a buffer layer on dielectric substrate can be useful for microwave device applications.

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