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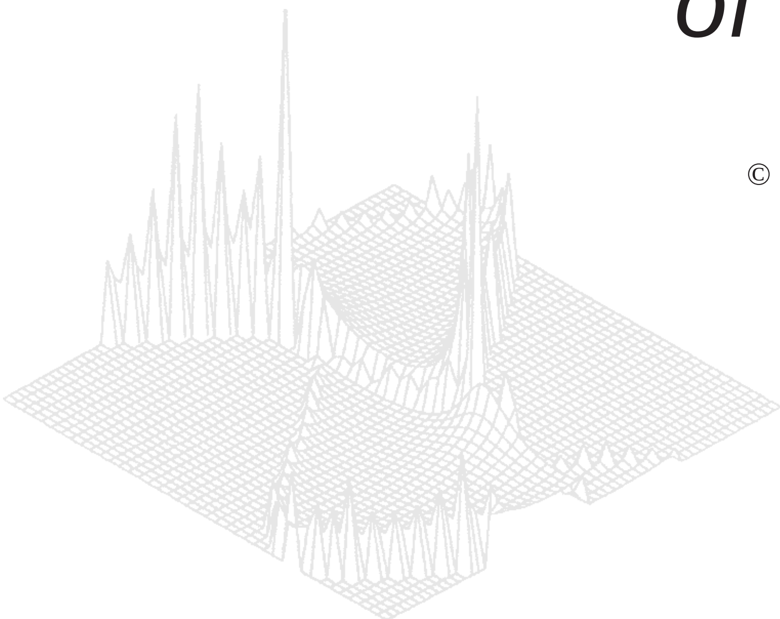

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Fabrication of $YBa_2Cu_3O_y$ Thin Films on Textured Buffer Layers grown by Plasma Beam Assisted Deposition*

M. Fukutomi, S. Kumagai[A] *and H. Maeda*

National Research Institute for Metals, Tsukuba Laboratories,
1–2–1 Sengen, Tsukuba-shi, Ibaraki 305, Japan.
[A] On leave from Mitsuba Electric Mfg Co. Ltd,
Kiryu-shi, Gumma 376, Japan.

Abstract

A new technique named plasma beam assisted deposition (PBAD) is proposed to grow in-plane textured yttria-stabilised zirconia (YSZ) thin films on polycrystalline metallic substrates as a buffer layer for deposition of $YBa_2Cu_3O_y$ (YBCO) films. The in-plane texturing of the YBCO films obtained is decisively governed by that of the YSZ buffer layer on which the YBCO grows. Because of a reduction of the weak links at high-angle grain boundaries, a marked increase in the critical current density J_c is observed with improved texturing of the YBCO films. So far, it has been demonstrated that YBCO films with J_c above $10^5\ \mathrm{A\,cm^{-2}}$ (77 K, 0 T) can be successfully deposited by a laser ablation technique. The PBAD process proposed here is found to be valuable technologically because it offers a very convenient method to grow textured films on long tape or large area substrates. An attempt was also made to grow textured films simultaneously on one side or both sides of various pieces of tape substrates. The results indicate that PBAD is one potential technique for future large scale application of YBCO films.

1. Introduction

The deposition of high T_c superconducting films on metallic substrates is technically important from the point of view of their future large scale applications, including conductors for high-field magnets, electromagnetic shields and devices like accelerator cavities or antennas. These applications, however, rely on the availability of high quality films on metallic substrates with high transport critical currents at 77 K. Recently, it has been shown that by depositing a YSZ buffer layer using an obliquely impinging ion-beam assisted deposition (IBAD) process, high J_c YBCO thin films can be grown on polycrystalline metallic substrates (Iijima *et al.* 1992; Reade *et al.* 1992; Sonnenberg *et al.* 1993; Wu *et al.* 1994). The J_c increase for these films is due to a biaxially aligned YSZ buffer layer produced by the IBAD process, resulting in a reduction of the weak links at high-angle grain boundaries in the subsequently grown YBCO films. This IBAD work indicating the occurrence of grain alignment in the films led us to develop a new plasma beam assisted deposition (PBAD) process, which enables us to deposit biaxially aligned YSZ thin films as a buffer layer on a metallic substrate for deposition of YBCO thin films (Fukutomi *et al.* 1994*a*).

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The present paper reports this PBAD process which makes possible fabrication of textured buffer layers without a separate ion-beam gun. The growth quality of YBCO thin films laser deposited on PBAD-YSZ buffer layers is then discussed. Furthermore, using PBAD, attempts to deposit textured films on both sides of the substrate and on a large area substrate are also described.

2. Film Preparation on Tape Substrates by PBAD and Pulsed Laser Deposition

The principle of developing an in-plane texturing by PBAD is basically the same as that of IBAD; an in-plane texturing occurs by off-normal ion-beam bombardment, probably because of the higher sputtering yields of all orientations other than the channeling direction. In a PBAD process, a flux of energetic particles impinging on the growing film is generated using a bias sputtering technique instead of a separate ion source in IBAD. Fig. 1 shows a pair of specially-devised electrodes (a substrate holder electrode and an auxiliary electrode) installed in a conventional magnetron sputtering system. The auxiliary electrode consists of two plates, each being $2{\times}10\ \mathrm{cm}^2$, facing each other with a separation of 2 cm. This was vertically installed below the substrate holder electrode. The distance d between the two electrodes was 5–10 mm. Here the bias voltage applied to the substrate holder and auxiliary electrode are referred to as V_{s} and V_{a} respectively. Substrates of Hastelloy tape (Hastelloy C-276), with dimensions $3{\times}100{\times}0{\cdot}3\ \mathrm{mm}^3$, were mechanically clamped on a tiltable specimen holder which was incorporated in the substrate holder electrode at an off-centred position, as shown in Fig. 1*b*. The specimen tilt angle θ measured from a horizontal level was fixed at the desired angle between $-50°$ and $90°$; the positive direction of θ was taken as counterclockwise. The distance between the specimen holder and target was about 7 cm. The target composition was an 8 mol% Y_2O_3–ZrO_2 sintered disk. Films were deposited at ambient temperature. The substrate temperature, however, rose to about 250°C during deposition because of energetic particle bombardment. Fig. 1*b* shows a characteristic parabola-shaped plasma also, which was generated in the space surrounded by the electrodes. This plasma was found to play an important role in achieving in-plane texturing of the films; the argon ion flux extracted from this plasma appeared to impinge on the growing film at a channeling direction of the YSZ crystal structure, resulting in the occurrence of in-plane texturing. As reported in a previous paper (Fukutomi *et al.* 1994*b*), our YSZ thin films had the strongest (200) preferred orientation when films were grown at an rf power of 200 W and an Ar–2%O_2 pressure of $0{\cdot}13$ Pa, while V_{s} and V_{a} were fixed at -200 V. Accordingly, all experiments in the present work were carried out under these deposition conditions while θ was varied over a wide range. It should be noted here that the degree of in-plane texturing depends strongly on the specimen tilt angle θ. This means that the YSZ buffer layer with a different degree of in-plane texturing can be grown under control on Hastelloy substrates.

The Hastelloy tapes thus pre-coated with the YSZ buffer layer were then used for deposition of YBCO films using a laser ablation technique. A KrF excimer laser ($\lambda = 248$ nm) with $\sim$350 mJ per pulse operated at $\sim$10 Hz was used. The laser beam was focused onto a 30 mm diameter rotating $YBa_2Cu_3O_y$ target at an energy density of about $4\ \mathrm{J\,cm^{-2}}$. The heated substrates were placed 3 cm from the target. After deposition, oxygen was introduced into the chamber to bring the chamber pressure close to 1 atmosphere. The films were then cooled to room temperature at a rate of 20°C per minute. The best films were grown at a substrate temperature of 700°C in an oxygen pressure of 20 Pa.

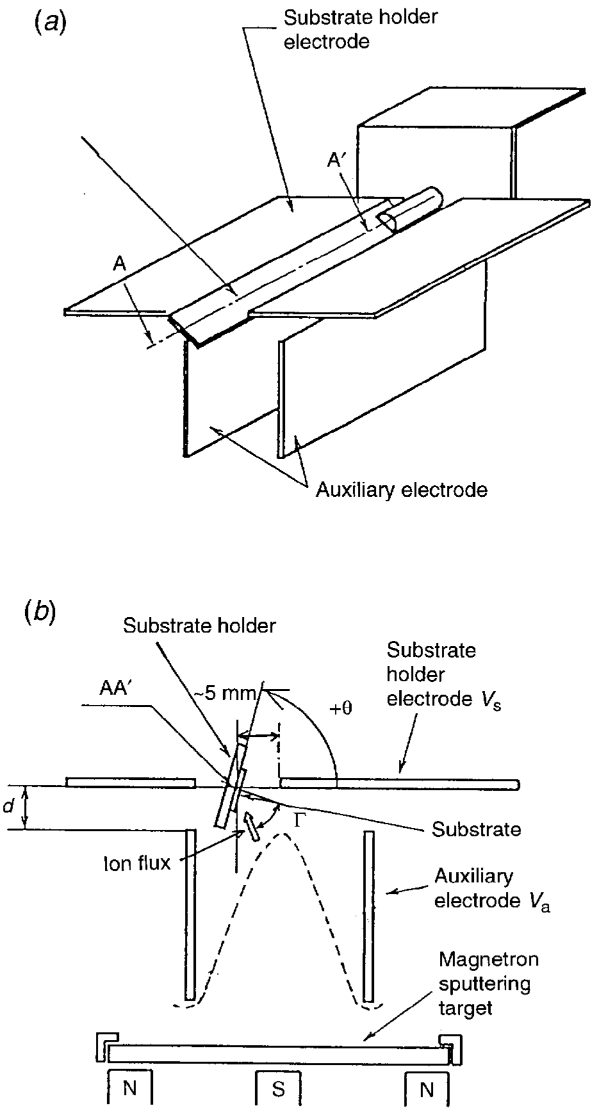

Fig. 1. (*a*) Configuration of electrodes installed in a sputtering system. The substrate holder is rotatable around a horizontal axis AA′ lying in the holder. (*b*) Cross-sectional view showing the off-centred mounting of the specimen.

(2a) Characterisation of In-plane Texturing of YSZ and YBCO Thin Films

X-ray pole figure measurements were conducted to characterise the in-plane texturing of the films. Fig. 2*a* gives a (111) ϕ-scan of the cubic YSZ films obtained at a specimen tilt angle θ of 75°. The result indicates an apparent in-plane grain orientation in the YSZ film. On the other hand, Fig. 2*b* gives the (103) ϕ-scan of the YBCO film laser-deposited on the textured YSZ buffer layer shown in Fig. 2*a*. A high degree of in-plane texturing in our YBCO films is evident, with strong fourfold symmetry of the (103) peak. A set of results on the ϕ-scan measurements for YSZ and YBCO films given in Fig. 2 indicates that YBCO [100] axes in our films were aligned to the YSZ [110] axes at the interface of the films.

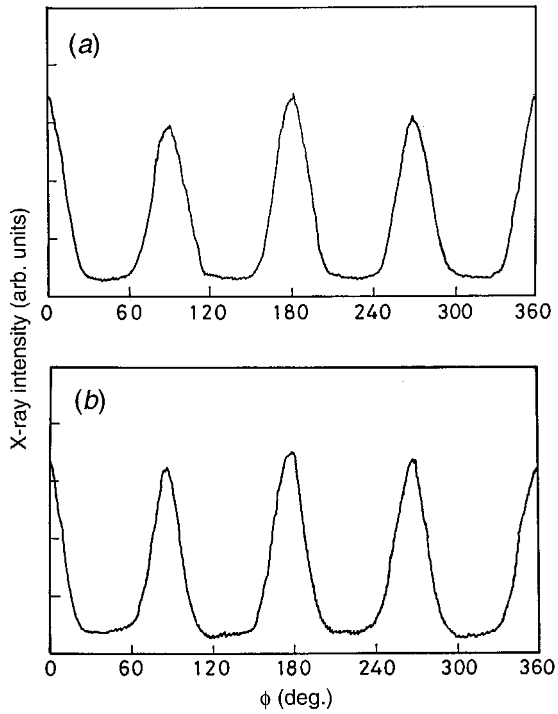

Fig. 2. (*a*) X-ray (111) ϕ-scan of a PBAD deposited YSZ film. (*b*) A (103) ϕ-scan of YBCO grown on the YSZ buffer layer shown in (*a*).

(2b) Variation of Critical Current Density with Degree of In-plane Texturing of YBCO Thin Films

As stated earlier, we found that by varying the specimen tilt angle θ indicated in Fig. 1*b*, we could control the degree of in-plane texturing of YSZ buffer layers. For example, a clear in-plane texturing was observed for the films grown at around $\theta = 80°$ and $-30°$, while the film deposited between 20° and 40° exhibited only a weak in-plane alignment. One possible explanation was offered in a separate paper (Aoki *et al.* 1994) for the occurrence of a marked in-plane texturing at these specific angles. In short, when the specimen was tilted by $\theta = 80°$ or $-30°$, a flux of energetic Ar ions appeared to impinge on the film surface with an average incident angle Γ of 55° measured from the specimen normal, as indicated in Fig. 1*b*. This angle of 55° corresponds to the [111] channeling direction from [001] axes in cubic YSZ films with the (200) plane parallel to the specimen surface. As a result, this 'channeling effect' of the impinging ions was considered to cause the in-plane grain alignment of the YSZ films.

The in-plane texturing was measured for YBCO films laser-deposited on YSZ buffer layers with a different degree of in-plane texturing. The results show that the in-plane texture of YBCO films is decisively governed by that of the YSZ buffer layer on which YBCO grows.

Next the correlation between the critical current densities and degree of texture of the YBCO films was examined. Transition temperature and critical current densities were resistively measured by a standard four-probe method. Using a steel scriber, a constricted region about 0·5 mm wide was made on the film with about 2 mm between the voltage taps. The criterion for J_c in the present study was 1 μV mm^{-1}. In Fig. 3 the transport J_c of the films is plotted against the degree of in-plane texture of YBCO. Here the degree of texturing was evaluated from the reciprocal of the full width at half maximum (FWHM) of the peak, $\Delta\phi$, in the ϕ-scan of the YBCO films. An increase in J_c with an improved texturing is due to a reduction of weak links at high-angle grain boundaries of YBCO films.

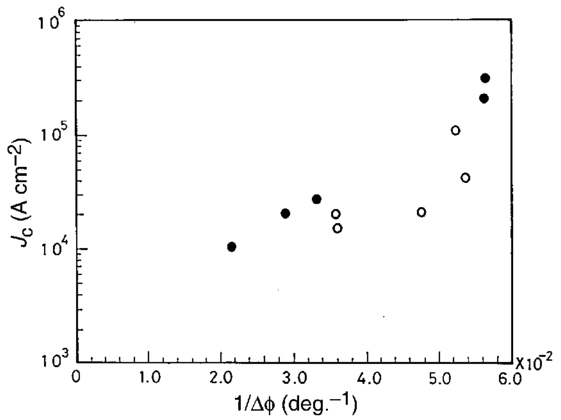

Fig. 3. Transport J_c at 77 K as a function of in-plane texturing of YBCO thin films. The solid circles are for films where a Pt underlayer was inserted at the boundary between YSZ and Hastelloy (see Fukutomi *et al.* 1994*a*).

The best film thus prepared to date gives a T_c of 90 K and a J_c of 3×10^5 A cm^{-2} (77 K, 0 T). This J_c value is one order higher than that reported in our earlier paper (Saitoh *et al.* 1991), where YBCO was grown on Hastelloy with an untextured YSZ buffer layer.

3. Various Modifications of the PBAD Process

(3a) Film Fabrication on Both Sides of Tape Substrate

An attempt was made to grow textured films on one side or both sides of a plurality of tape substrates. Fig. 4 shows a typical configuration of electrodes used for this purpose. Technological fundamentals of this modified PBAD were basically the same as those already described. We can see the characteristic parabolic plasma edge in each discharge space formed by a pair of adjacent auxiliary electrodes and a substrate electrode. In Fig. 4*a*, the substrates of

Hastelloy C tape, about 10 cm in length, were attached to substrate holders in such a way that the longer tape direction becomes parallel to the auxiliary electrode. The tape is fixed normal to the substrate electrode, leaving both surfaces open for simultaneous deposition. Fig. 4*b* shows a substrate electrode having a plurality of substrate holders capable of causing a substrate to tilt by a desired angle relative to the horizontal plane, and a plurality of auxiliary electrodes arranged vertically below the substrate electrode. This new electrode configuration enabled us to fabricate textured thin films on plural pieces of tape substrates simultaneously.

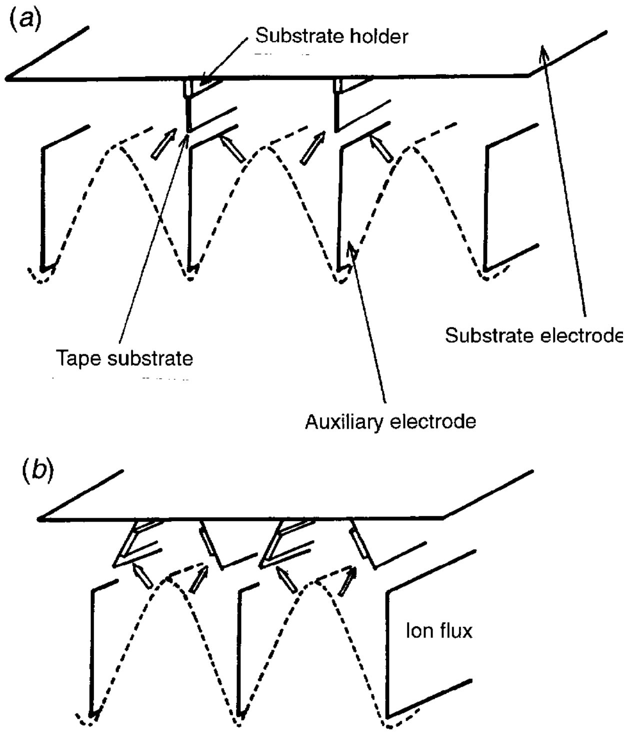

Fig. 4. Various modifications of the PBAD process: (*a*) electrode configuration for double-sided deposition and (*b*) simultaneous deposition on plural pieces of tape substrates.

Fig. 5 is a typical pole figure for the (111) reflection plane of the YSZ film grown using the electrode configuration shown in Fig. 4*a*. The existence of an in-plane texture of YSZ is apparent from this figure. It should be noted here that identical pole figure results were obtained for films on both sides of the tape substrate. The off-axis laser deposition geometry was used for the in-situ double-sided preparation of YBCO films. The films obtained on both sides of the tape were found to have quite similar superconducting properties, showing zero resistance at around 88 K. Transport J_c values for the films were, however, somewhat scattered between 5×10^4 and $1\cdot5\times10^5$ A cm^{-2} at zero field and 77 K (Fukutomi *et al.* 1994*c*). For further improvement in J_c, the growth quality of double-sided YSZ buffer layers, particularly with respect to the degree of in-plane texturing, must be improved. At the present time, the lowest value for $\Delta\phi$ in our double-sided YSZ films is about 35°, which is somewhat larger than the film grown on one side of the tape substrate. Various modifications of the electrode configuration are currently being examined for further improvement in texturing of YSZ buffer layers.

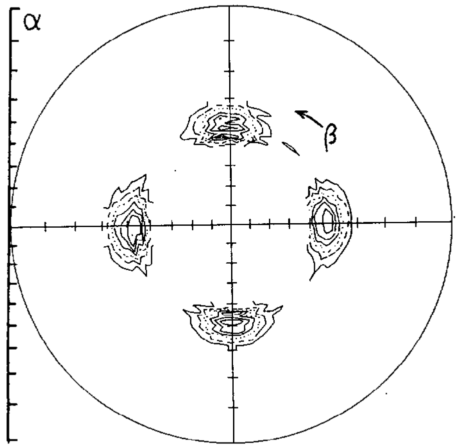

Fig. 5. The (111) pole figure for a YSZ film grown on both sides of the substrate.

(3b) Fabrication of Textured Films on Large Area Substrates

Fig. 6 is a cross-sectional view of the electrodes used for obtaining in-plane textured YSZ films over the whole surface of the sample. As predicted from the results described in Section 2*a*, the degree of in-plane texturing varies depending upon the sample position; films grown on the part of the substrate located directly above the centre between a pair of adjacent auxiliary electrodes showed comparatively poor texture. This is because glancing-angle ion bombardment during deposition is one of the requirements for the achievement of in-plane texturing. At this area, however, Ar^+ ions impinge on a film not obliquely but at almost right angles. In order to avoid the growth of poorly-oriented films at this area, masks made of zirconium tape were placed at these positions, as illustrated in Fig. 6. In addition, we incorporated a movable substrate holder electrode which enabled us to slide the substrate horizontally during deposition. After a period of deposition, the substrate was moved horizontally so that a film with in-plane texturing could grow on the masked area. Consequently the whole area of the substrate was successfully covered with in-plane textured YSZ thin films. Using this electrode, we attempted to grow YBCO films on a copper disk substrate with a diameter of 36 mm and thickness 3 mm. Prior to YSZ deposition, a thin Cr underlayer was ion-plated on the Cu substrate to ensure good adhesion of YSZ films. The copper substrates thus precoated with the YSZ/Cr buffer layer were then used for deposition of YBCO films using the laser ablation technique. In order to obtain uniform large area YBCO films, the mirror was oscillated so that an excimer laser beam reflected from the mirror could be scanned on the rotating target surface. However, the film thickness was distributed on the entire substrate between 1·5 and 2 μm. Fig. 7*a* shows the pole figure for (103) peaks of the YBCO films deposited on the in-plane textured YSZ buffer layer, while that for the YBCO film grown on an untextured YSZ buffer layer was shown in Fig. 2*b* for comparison. The YBCO films obtained so far have T_c values higher than 86 K, regardless of their texturing. However, an apparent difference in transport J_c was observed between textured and untextured YBCO films. The film grown on an in-plane textured YSZ buffer layer gave a J_c of $1\cdot 0\times 10^5$ A cm^{-2}, whereas a comparable film on untextured YSZ buffer layers had values as low as 10^3–10^4 A cm^{-2} at zero field and 77 K. The YBCO films fabricated on copper substrate have a potential application to a particle accelerator cavity. Therefore, measurements of microwave surface resistance of the YBCO films obtained are now under way as a collaborative work with the National Laboratory for High Energy Physics (Liu *et al.* 1995).

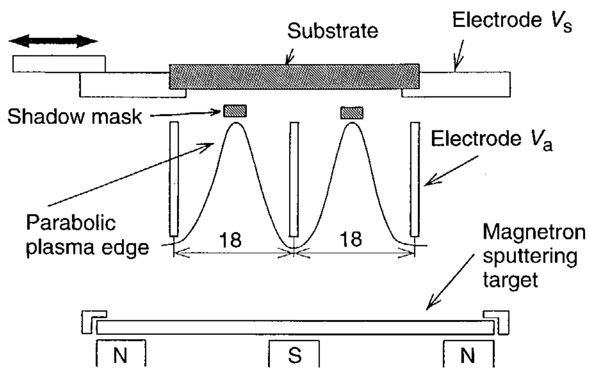

Fig. 6. Cross-sectional view of the configuration of electrodes.

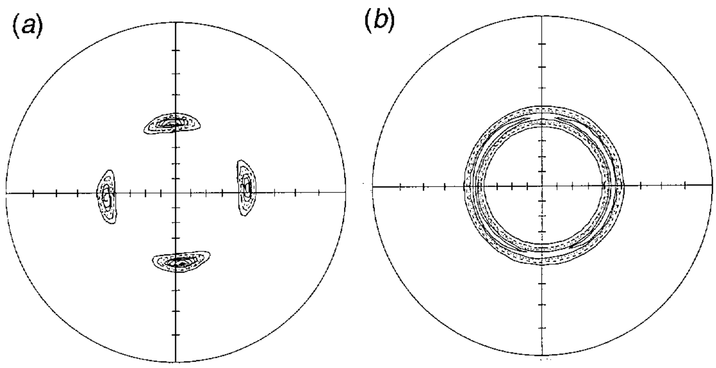

Fig. 7. The (103) pole figure for (*a*) in-plane textured YBCO and (*b*) untextured YBCO.

4. Conclusions

A new technique named plasma beam assisted deposition (PBAD) was proposed to grow biaxially textured YSZ thin films on polycrystalline metallic substrates as a buffer layer for deposition of YBCO films. The in-plane texturing of the YBCO films was decisively governed by that of the YSZ buffer layer on which the YBCO grows. Because of a reduction of the weak links at high angle grain boundaries, a marked increase in J_c is achieved with improved texturing of the YBCO films. The PBAD process proposed here is found to be valuable technologically because it offers a very convenient method to grow textured films on long tape or large area substrates. An attempt was also made to grow textured films simultaneously on one side or both sides of various pieces of tape substrates. Future work must concentrate on further improvements in the degree of texturing of buffer layers by optimising the deposition conditions.

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