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## Laser deposition of biaxially textured yttria-stabilized zirconia buffer layers on polycrystalline metallic alloys for high critical current Y-Ba-Cu-O thin films

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Pulsed laser deposition of yttria-stabilized zirconia (YSZ) layers on polycrystalline metallic alloy substrates is used to produce an intermediate layer for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (YBCO) thin-film growth. The desired (001) YSZ texture is obtained at 1.0 mTorr oxygen pressure and 70 °C. Significant improvement in (001) texturing is demonstrated by using an ion beam to assist growth. Argon-ion-assisted growth produces layers with alignment of the in-plane crystal axes in addition to the (001)-normal texture. Highly *c*-axis-oriented biaxially aligned YBCO thin films can be deposited on these layers, with  $T_c(R=0)=92$  K and  $J_c$  (77 K, B=0 T)= $6 \times 10^5$ A/cm<sup>2</sup> and  $J_c$  (77 K, 0.4 T)= $8 \times 10^4$  A/cm<sup>2</sup>. With further improvement of the YSZ texture, the YBCO current-carrying capacity of films on polycrystalline metallic alloys may approach that of films on single-crystal substrates.

The growth of high quality thin films of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (YBCO) on polycrystalline metallic alloys, such as stainless steel or nickel based superalloys, is desirable for electric power and energy storage applications. It has been shown, however, that YBCO films deposited directly on these substrate materials exhibit poor superconducting properties.<sup>1,2</sup> It has been demonstrated that a yttriastabilized zirconia (YSZ) intermediate layer can yield YBCO films of higher quality, as demonstrated by improved *c*-axis alignment with higher critical temperatures  $(T_c)$  and currents  $(J_c)$ .<sup>3-5</sup> In order to further improve these films, we have focused on the improvement of the YSZ intermediate layer texture.

Several groups are investigating the use of YSZ intermediate layers for the deposition of YBCO thin films on silicon and gallium arsenide.<sup>6-8</sup> Critical currents  $(J_{r})$ higher than  $10^6$  A/cm<sup>2</sup> at 77 K have been reported for these films. Unfortunately, previously reported  $J_c$ 's for YBCO films deposited under similar conditions on polycrystalline metallic alloys with YSZ intermediate layers have been two to three orders of magnitude lower.<sup>3-5</sup> The reason for this discrepancy is that the YSZ intermediate layers grow highly oriented on single-crystal semiconductors due to lattice registry with these substrates, whereas this is not the case, and is in fact undesirable, for YSZ growth on the randomly oriented polycrystalline metallic alloys. If, however, deposition conditions can be found to grow the YSZ layers on these alloys with texture similar to that obtained on the single-crystal semiconductors, there is no reason that YBCO films of as high a quality should not be expected.

Recently, enhanced texturing of YSZ has been reported for layers grown by an ion-assisted sputtering technique.<sup>9</sup> Using these layers, the authors have reported an order of magnitude improvement in  $J_c$  for YBCO thin films deposited on the YSZ by pulsed laser deposition. Our pre-

vious studies included the investigation of ion assistance to the YBCO laser deposition process,<sup>10</sup> so we expanded our effort to include *in situ* ion-assisted pulsed laser deposition of YSZ layers.

The configuration of our laser deposition system has been described previously.<sup>1,2,4</sup> A KrF (248 nm) excimer laser is focused into the vacuum deposition chamber to an energy density of 2–3 J/cm<sup>2</sup> at the target. The substrate is attached to a resistively heated block opposite the target using a silver epoxy. Target-substrate spacings range from 4 cm for YSZ deposition to 5–6 cm for YBCO deposition. A 3-cm diam ion gun is positioned to provide an ion beam at 30°–60° incidence to the substrate for ion-assisted deposition tests. Argon and oxygen are available for the deposition chamber background atmosphere or as ion source gases.

Substrates of nickel-based superalloy (Haynes Alloy No. 230) are cut from 0.6 mm sheet and polished using aluminum oxide lapping powders to a final grit size of 0.05  $\mu$ m. The substrates are rinsed in deionized water and organic solvents, and then preheated to 500 °C in a vacuum of  $\leq 10^{-6}$  Torr. YSZ layers are deposited from a commercially available ceramic sputtering target (~10 at. % Y<sub>2</sub>O<sub>3</sub> content).

Our previous work showed that (001)-textured YSZ films result from depositions in oxygen pressures near 1.0 mTorr and temperatures near 70 °C, while (111)-texture results at higher pressures and temperatures.<sup>4</sup> An x-ray diffraction pattern for a (001) layer is shown in Fig. 1(a). YBCO thin films on the (001) YSZ layers exhibited better *c*-axis texture as well as higher  $T_c$  and  $J_c$  than those on the (111) YSZ layers or untextured layers.

In order to improve the c-axis YBCO film texture, further improvement of the (001) texture of the YSZ was needed. As can be seen in Fig. 1(a), the (002) x-ray diffraction peak demonstrating (001) texture is broad and shallow, likely indicating incomplete (001)-texturing and small grain size. Annealing treatments resulted in only

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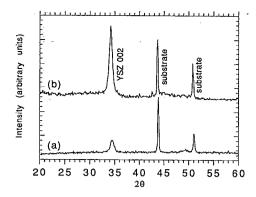


FIG. 1.  $2\theta$  x-ray diffraction patterns for YSZ layers deposited on Haynes Alloy No. 230 (a) without and (b) with ion-beam assistance.

slight improvements in the (001) YSZ texturing. A 700 °C anneal of a (001) layer for 30 min in oxygen resulted in a (002) x-ray peak intensity improvement of only 10%–20%. Longer anneals did not result in further improvement and higher temperature anneals destroyed the (001) texture.

Ion-assisted depositions were carried out to investigate the possibility of ion-enhanced texturing. Oxygen or argon gas was supplied to the ion gun at a rate of 10 sccm. The chamber background pressure was held at 1.0 mTorr, as in previous (001) YSZ depositions. For depositions using argon as the ion-beam gas, a total pressure of 1.0 mTorr was achieved by first flowing argon through the gun and adjusting the pumping rate to give a pressure of 0.5 mTorr and then admitting oxygen through a separate valve to 1.0 mTorr. Ion-beam voltages were varied from 50 to 200 V and currents from 5 to 60 mA. The gun was placed 5–8 cm from the substrate. All other deposition parameters remained the same, though the substrate temperature typically increased from 70 to 130 °C during a 15 min deposition due to ion-beam heating.

A x-ray diffraction pattern from a film grown with oxygen-ion-assistance is shown in Fig. 1(b). Considering that the films have similar thicknesses, strongly enhanced (001) texturing of the YSZ is apparent with no evidence of any unwanted (111) or (011) texturing in this film. The selective enhancement of the growth of (001) grains may occur because an energetic beam striking the growing film at an angle corresponding to the angle of a channeling direction for a particular crystal orientation can enhance the growth in this orientations.<sup>11</sup> The [111] channeling direction in cubic YSZ is 54.7° from the [001] direction, so ions directed near this angle to the substrate normal during layer growth may enhance (001) texture.

Further characterization of films grown with the oxygen ion-beam showed that although the desired (001)normal texture was achieved, there was no evidence for alignment of the in-plane crystal axes. Therefore, subsequently deposited YBCO thin films did not exhibit in-plane grain alignment, allowing the existence of a large percentage of high-angle boundaries between the grains. Highangle grain boundaries in YBCO lead to weak-link behav-

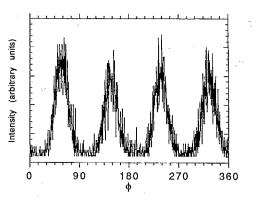


FIG. 2. (111)  $\phi$ -scan for a YSZ layer grown with Ar<sup>+</sup>-beam assistance.

ior, limiting critical current densities.<sup>12-14</sup> Thus, it was not surprising that  $J_c$  (77 K) for a YBCO layer of this type was only 10<sup>4</sup> A/cm<sup>2</sup>, far lower than for films on biaxially aligned YSZ layers on silicon.

In-plane texturing occurred when argon was substituted for oxygen as the ion-beam gas. Unlike oxygen, the argon ion is nonreactive and is also a different size and mass. Thus, the use of argon ions was expected to change the energetics of the ion-assisted growth. The use of argon affected the in-plane YSZ texturing without detriment to the other layer properties; the  $2\theta$  x-ray diffraction pattern for the argon-ion-assisted layer is nearly identical to that for the oxygen-ion-assisted layer shown in Fig. 1(b). Inplane texturing is demonstrated by the (111)  $\phi$ -scan of Fig. 2, for a film grown using a 175 V, 10 mA ion beam. Clear fourfold symmetry proves the alignment of the in-plane axes. Also, we note that a peak of (111) intensity occurs at the  $\phi$  angle corresponding to the incoming ion direction. This in-plane texturing is due to the use of the argon-ion beam, as no in-plane texture was evident for layers grown without ion assistance or with oxygen-ion assistance.

The effects of ion beam current and voltage on the YSZ in-plane texture were investigated. Changing the current was found to have little effect on the in-plane texture and high currents (>50 mA) reduced growth rates significantly, presumably by sputtering the growing layer. Conversely, variation of the beam voltage was determined to have a major impact on the in-plane texture. For beam voltages lower than about 50 V,  $\phi$ -scans showed little or no evidence of in-plane texture. Higher beam voltages increased texture, up to the limit imposed by our present configuration and deposition parameters (~200 V).

A 0.4  $\mu$ m thick YBCO thin film was deposited on the ion-assisted (001) YSZ layer shown in Fig. 2. The resulting film is highly *c*-axis oriented, as shown by  $2\theta$  x-ray diffraction in Fig. 3(a). The YBCO peak intensities for this film are an order of magnitude greater than for films on YSZ layers without in-plane texture, indicating an improved degree of *c*-axis alignment. A (005) rocking curve measurement showed a  $\Delta \omega \sim 1.3^{\circ}$  full width half maximum, only slightly higher than the best reported for YBCO thin films on epitaxial YSZ on single-crystal silicon (0.6°-1.0°).<sup>6</sup> A YBCO (103)  $\phi$ -scan for this film is shown

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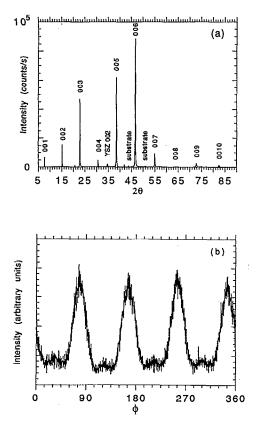


FIG. 3. X-ray diffraction data for a YBCO thin film deposited on a biaxially aligned YSZ layer: (a)  $2\theta$  pattern and (b) (103)  $\phi$ -scan.

in Fig. 3(b). A high degree of in-plane texturing is evident, with strong fourfold symmetry of the (103) peak and a low background level.

A 50  $\mu$ m wide bridge approximately 1 mm in length was patterned using the excimer laser. Resistance and critical current measurements were made with a standard four-point probe technique. Contacts were made with silver paint. The normalized resistance versus temperature data appear in the inset to Fig. 4.  $T_c(R=0)$  is approximately 92 K. dc critical current densities (1  $\mu$ V/mm criterion) as a function of temperature are shown in Fig. 4. The  $J_c$  at 77 K without field is  $6 \times 10^5$  A/cm<sup>2</sup>, while with field parallel, perpendicular, and at 45°, the  $J_c$ 's fall to 8

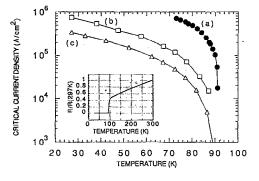


FIG. 4. YBCO critical current densities (a) without magnetic field, and with 0.4 T field (b) parallel to the film and (c)  $45^{\circ}$  to the film normal, the field direction for which  $J_c$  is lowest. Inset shows normalized resistance vs temperature.

 $\times 10^4$ ,  $5 \times 10^4$ , and  $3 \times 10^4$  A/cm<sup>2</sup>, respectively. This unusual angular dependence of  $J_c$  (lowest with field at 45° rather than perpendicular) can be attributed to increased flux pinning for fields perpendicular to the film, compared with epitaxial films.<sup>15</sup> These values are higher than any previously reported for films on polycrystalline substrates and are of levels suitable for applications to high-current tapes for current leads, solenoids, etc. Past films without in-plane texture had far lower  $J_c$ 's, presumably due to weak link behavior at high-angle grain boundaries. The  $J_c$ 's in the present film are closer to those demonstrated for films on single crystals, suggesting a sharp reduction in weak links at high-angle boundaries.

In summary, we have successfully demonstrated *in situ* ion-assisted laser deposition of biaxially textured YSZ buffer layers suitable for YBCO thin-film growth on polycrystalline metallic alloys. In-plane texturing of subsequently laser-deposited YBCO films was shown to give high critical current densities  $[J_c (77 \text{ K}, 0 \text{ T}) \sim 6 \times 10^5 \text{ A/cm}^2]$ . With further improvement of the YSZ texture, the YBCO current-carrying capacity of films on polycrystalline metallic alloys may approach that of films on single crystals. Also, we have deposited biaxially textured YSZ layers on fused silica and anticipate that these layers may grow on other substrates of technical interest, such as other amorphous oxides, other randomly oriented polycrystals, or single crystals on which epitaxy is difficult.

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