Nucleation and growth of $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ barrier layers on ramps in $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$ studied by atomic force microscopy

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We studied the microstructure of Ar ion-beam etched ramps in epitaxial $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films by atomic force microscopy. Generally, ramps were well aligned ($\phi \approx 0^\circ$) with one of the crystal axes of the (001) SrTiO_3 substrate. In those cases we observed a surface reconstruction into a regular pattern of very long (up to $0.5 \mu\text{m}$), about 20 nm wide facets, parallel to the ramp edge. For high misorientation angles ($\phi > 0^\circ$), the reconstruction appeared much more complex and less regular. Furthermore, we investigated the nucleation and growth of very thin barrier layers of $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ on well aligned ramps. We found that nucleation takes place in the shallow trenches in the ramp, where facets meet. When more material is deposited, islands coalesce to form closed domains parallel to the trenches. This causes a thickness modulation close to 100% for barrier layer thickness up to 6 nm. The conclusions drawn from this research allow for an important improvement of the technology for the fabrication of ramp-type junctions. © 1996 American Institute of Physics. [S0003-6951(96)03409-1]

Ramp-type high- T_c superconducting (HTS) Josephson junctions, rather than other junction types, allow for adjustment of the junction performance by changing the barrier thickness or the barrier material itself. We found for the $I_c R_n$ product, which is an important figure of merit for Josephson junctions, a systematically higher value for lower $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (PBCO) barrier thickness.¹ However, when we decreased the barrier thickness to below 10 nm, the junction properties tended to be rather irreproducible and the current-voltage characteristics showed large excess currents. Probably, for low barrier thickness, the influence of pinholes becomes dominant.

Ramp-type junction structures are prepared by heteroepitaxy of a barrier layer and a top electrode on ramped edges created in a $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (DBCO) base electrode. A well-known source of trouble for the high T_c materials is their extremely short coherence length ξ . Obviously, details on the scale of unit cells of the ramp surface morphology, as well as the growth mode of the barrier material on the ramp will severely influence the transport properties of these junctions. Barrier layers with a thickness of only a small number of unit cells have to be used to obtain significant critical dc Josephson current. As a reference, we found in an earlier study a decay length of the supercurrent in the order of only 2 nm for PBCO barriers.¹

Therefore more detailed insight is needed in the nucleation and growth of PBCO on ramps, to estimate at which nominal thickness complete coverage is obtained. If growth proceeds layer-by-layer, the lower limit is obviously the PBCO unit cell. If the barrier material first tends to form islands, thin barriers will be inhomogeneous in thickness, enhancing the probability of pinholes. The ultimate height resolution of atomic force microscopy (AFM), well within the unit-cell dimensions, provides a unique analytical tool to

study this growth and is useful in optimizing the fabrication process of ramp-type junctions.

We deposited 100 nm thick epitaxial DBCO layers onto $5 \times 10 \text{ mm}^2$ SrTiO_3 substrates using off-axis rf-magnetron sputtering from a single stoichiometric target.^{1,2} The films were deposited at conditions where we reproducibly obtain optimal T_c and J_c values. The texture was checked using x-ray diffraction (XRD) measurements; rocking curve analysis of the (005) DBCO reflection typically revealed a full width at half-maximum (FWHM) of below 0.4° . The film surfaces were grossly free of boulders and outgrowths, and on lateral dimensions of microns, peak-to-peak height differences were not larger than $\sim 3 \text{ nm}$.

Before ramp structuring, Shipley S-1813 photoresist was spun for 30 s at 4000 rpm, and baked during 10 min on a hot plate at 90°C . The photoresist stencil then had a thickness of about $1.2 \mu\text{m}$. Special care was taken to align the mask with misorientation angle $\phi \approx 0^\circ$ with respect to one of the crystal axes of the SrTiO_3 substrate. The etching was done at a background pressure of $6 \times 10^{-2} \text{ Pa}$ with a Kaufmann Ar ion gun ($V = 500 \text{ V}$, $I = 10 \text{ mA}$, 3 cm in diameter) in a pulsed mode (8 s on, 10 s off) to prevent overheating of the sample. The plane of incidence was always perpendicular to the edge of the photoresist and the angle of incidence $\psi = 45^\circ$ with the surface was kept constant for each sample, yielding an etch rate of 30 nm/min for DBCO and 60 nm/min for photoresist. As a result of the difference in etch rates, the ramp angle α will be smaller than ψ : $\psi = 45^\circ$ will give $\alpha \approx 16^\circ$ and $\psi = 30^\circ$: $\alpha \approx 8^\circ$. The photoresist was removed by ultrasonic stirring in acetone and rinsing in 2-propanol. The bare ramps studied in this work got no further treatment. The time between sample preparation and AFM observation was typically about 1 h. However, even after a few days storage, no changes in surface morphology were visible.

For the epitaxial PBCO layers, the time between ramp structuring and bringing it into vacuum again, was minimized to a few minutes. Directly after photoresist stripping

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FIG. 1. (a) Sketch of the geometry during etching. (b) and (c): AFM images of a ramp after ion milling. (c) shows a zoomed part of the ramp.

the sample was brought into the Kaufmann etching chamber for cleaning the ramp surface from contamination during the *ex situ* process. The cleaning procedure we used was performed at identical conditions as used for ramp structuring and consisted of four pulses (8 s on, 10 s off) at 500 V/10 mA and two pulses at 200 V/7 mA. Without breaking the vacuum the sample was transported into the sputter deposition chamber. The chamber was filled with the usual sputter atmosphere (20 Pa, Ar:O₂ = 1:1, 0.1% H₂O gas), the temperature was raised to 790 °C and kept at this value during 1 h before sputter deposition was started. The value for the barrier thickness d used, is the nominal thickness in the direction perpendicular to the substrate surface as inferred from the calibrated growth rate (90 ± 10 nm/hr). We prepared samples with $d=2, 4, 6$, and 10 nm, each on a separate substrate. After deposition the chamber was filled with oxygen to 200 mbar and the sample was cooled down to room temperature within 1 h.

AFM micrographs were recorded in “open air” conditions with a Nanoscope III in contact mode. Commercial Si₃N₄ tips mounted on a V-shape cantilever with a force constant of 0.06 N/m were used. All features presented here were observed with at least two different tips and checked at

different scan angles to rule out artifacts caused by the tip profile.

In Fig. 1(a) the geometry during the etching process is sketched. Figures 1(b) and 1(c) are AFM images of a well aligned ramp ($\phi=0^\circ$). While zooming in on a part of the ramp (c), the average plane was subtracted. As the ramp surface is somewhat curved, the difference in height and lateral scales then causes the concave appearance in Fig. 1(c). Clearly, the ramp surface consists of facets formed during the ion milling. For small misorientation angles $\phi \approx 0^\circ$, very long (several hundreds of nm, up to $\sim 0.5 \mu\text{m}$), about 20 nm wide facets are formed parallel to the ramp edge. For misaligned ramps ($\phi > 0^\circ$) the pattern of facets, this is constructing the macroscopic ramp surface, looks much less regular.

We checked the influence of the annealing effects during heating of the etched and cleaned ramp in sputter atmosphere. The general procedure was followed, except for the deposition of a PBCO layer. This 1 h anneal at 790 °C did not result in any visible change of the ramp surface morphology, indicating that there was no reconstruction of the facets at deposition conditions.

Figures 2(a) and 2(b) show AFM images after deposition of thin PBCO layers on the ramps. Nucleation takes place in the ~ 1 nm deep trenches in the surface, where the facets join. Nuclei become 3–4 nm thick, already for a nominal barrier thickness of $d=2$ nm (a). For a nominal thickness of 6 nm, a regular superstructure parallel to the trenches is visible (b), with a periodicity of about 60–80 nm and 8–10 nm thickness variation.

In Fig. 3, we superimposed the cross sections of the ramps covered with 6 and 10 nm PBCO, respectively, to that of the bare ramp. Vertically, the flat parts have been shifted

FIG. 2. Top: AFM images of a ramp covered with (a) 2 nm PBCO; (b) 6 nm PBCO. Bottom: corresponding line scans a-a' and b-b'.

FIG. 3. Cross sections of ramps covered with: (a) 6 nm PBCO; (b) 10 nm PBCO, compared to that of the bare ramp. The lines (on the ramped part: the average planes) are shifted vertically by their effective thickness to give an indication of the thickness and its variation across the ramp.

upward by 6 and 10 nm. Also the averaged local planes of the covered and bare ramps should be shifted in vertical direction by this amount. This then gives us the horizontal alignment. Because of the regular domain structure along the ramp [see Fig. 2(c)], here it is easily performed by determining the averaged planes from the cross sections of the ramped part. Figure 3(a) shows that the thickness modulation for $d=6$ nm should be close to 100%. This roughening is found to be less for 10 nm nominal thickness [Fig. 3(b)], indicating that there is secondary nucleation in the voids between the domains.

Epitaxial c -axis oriented DBCO films on SrTiO_3 substrates grow with the a (b) axis parallel to the $\text{SrTiO}_3(100)$ axis. During the ion-beam etching process a continuous surface reconstruction of the ramp results in the formation of facets. This explains why the ramp morphology, obtained by ion-beam etching, is strongly related to the orientation of the ramp edge with respect to the underlying crystal lattice of DBCO. As the macroscopic ramp surface appears to consist of preferred facets of the 123-crystal, a misorientation angle $\phi \neq 0^\circ$ results in a much more complicated pattern. Growth of 123-materials on such a surface will result in even more roughening, as a result of the anisotropy in growth speeds. This may explain the *saw-tooth-like* appearance we frequently observe by scanning electron microscopy on the ramps covered by a thick film in cases where $\phi \neq 0^\circ$.

In conclusion, the microstructure of Ar ion-beam etched ramps in epitaxial DBCO films was studied by AFM. For well aligned ramps ($\phi \approx 0^\circ$) we saw that the surface was reconstructed into a regular pattern of very long (up to $0.5 \mu\text{m}$), about 20 nm wide facets, parallel to the ramp edge. For misorientation angles ($\phi > 0^\circ$), the reconstruction looked much more complicated and less regular. This is due to the fact that facets are the preferred crystal lattice planes, and the macroscopic ramp surface is constructed out of these facets. We found that nucleation of material deposited on top of the ramp takes place in the shallow trenches, where facets meet. Islands coalesce to form a regular superstructure parallel to the trenches, with 100% thickness modulation up to $d=6$ nm. At $d=10$ nm it was found that the voids between the domains were filled up by secondary nucleation. For the use in ramp-type junctions, these results indicate that a minimum thickness of 8–10 nm should be applied for PBCO barriers to avoid pinholes through the barrier caused by intrinsic roughening of 123-epitaxial films.

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¹M. A. J. Verhoeven, A. A. Golubov, G. J. Gerritsma, and H. Rogalla, IEEE Trans. Appl. Supercond. **5**, 2095 (1995).

²J. Gao, W. A. M. Aarnink, G. J. Gerritsma, and H. Rogalla, Physica C **171**, 126 (1990).