Properties of $YBa_2Cu_3O_7$ Thin Films Deposited on Substrates and Bicrystals with Vicinal Offcut and Realization of High I_cR_n Junctions

U. Poppe, Y.Y. Divin, M. I. Faley, J. S. Wu, C. L. Jia, P. Shadrin, and K. Urban

***Abstract*—The surface morphology, microstructure and transport properties of epitaxial $YBa_2Cu_3O_7$ and $PrBa_2Cu_3O_7$ thin films and heterostructures deposited on slightly vicinal substrates of $SrTiO_3$ by high pressure oxygen sputtering were studied. The vicinal angles of the substrates and bicrystals were less then 13°. Depending on the tilt angle of the substrate a transition from spiral or island to step-flow growth leading to an improvement of the surface roughness was observed. Atomic force and transmission electron microscopy were used for these investigations. Furthermore, electrical and structural properties of $YBa_2Cu_3O_7$ thin films on vicinal offcut $SrTiO_3$ bicrystals with different grain boundary types were studied. This included junctions with a 2x 12° tilt or twist of the $YBa_2Cu_3O_7$ c-axis across the grain boundary. In comparison to conventional [001]-tilt grain boundaries bicrystal Josephson junctions [100]-tilt grain boundaries showed high I_c R_n –products of up to 1.2 meV at 77 K and up to 8 meV at 4.2 K. IV-curve instabilities, probably of magnetic origin due to flux flow in the electrodes, often could be observed for junctions biased with high current densities.**

***Index Terms*—High-temperature superconductors, Josephson junctions, bicrystals, grain boundaries, vicinal substrates.**

I. INTRODUCTION

THE use of tilted substrates for the growth of high-T_c thin films can lead to improved properties for different applications. The c-axis of the film usually follows the c-axis orientation of the substrate for vicinal substrates with small tilt angles. It has been shown by several groups [1], [2] that the growth mode changed from island or spiral growth to step flow growth already at tilt angles below 5° leading to less surface roughness and improved transport properties for $YBa_2Cu_3O_7$ films on perovskite substrates like $SrTiO_3$. It is very important for device application, especially for planar type multilayer tunnel junction devices, to prepare high-T_c thin films with flat surfaces and interfaces.

For larger tilt angles, due to the layered structure of high-T_c materials, the transport properties [3], [4], [5] become more anisotropic and the surface roughness again increases due to step bunching and the appearance of grains elongated along the ab-planes of $YBa_2Cu_3O_7$. While [001]-tilt grain boundary junctions with c-axis oriented $YBa_2Cu_3O_7$ thin films are frequently used for fundamental and applied research (e. g. [6], [7], [8], [9]), -apart from step edge junctions- only a few reports on Josephson junctions with different types of grain boundaries have been investigated. Beside early work of Dimos et al [6], who studied the properties of very differently oriented bicrystal junctions, Tafuri et al [10] reported on biepitaxial tilt and twist boundaries with a 45° c-axis tilt of $YBa_2Cu_3O_7$.

We report on properties of epitaxial $YBa_2Cu_3O_7$ and $PrBa_2Cu_3O_7$ thin films and heterostructures deposited on 1.7° vicinal substrates of (100) $SrTiO_3$ by high pressure oxygen sputtering, as well as the growth of 12° c-axis tilted $YBa_2Cu_3O_7$ films on bicrystal substrates and the properties of corresponding Josephson junctions. Furthermore a comparison of [100]-tilt boundary junctions with conventional [001]-tilt grain boundary junctions is presented.

II. EXPERIMENTAL

The $YBa_2Cu_3O_7$ thin films were deposited by dc sputtering from a stoichiometric target at high oxygen pressure [11]. The sputtering parameters were optimized to obtain high-quality c-axis $YBa_2Cu_3O_7$ thin films with a transition temperature T_c above 90 K. The oxygen pressure was about 3.2 mbar, and the heater temperatures were held at about 920°C during the sputtering procedure. $YBa_2Cu_3O_7$ films with tilted c-axis were grown on 1.7° vicinal cut $SrTiO_3$ or $NdGaO_3$ substrates as well as 2x 12° tilt or twist type $SrTiO_3$ bicrystals [12]. Films were patterned for the IV-measurements of the bicrystal Josephson junctions by deep UV photolithography and nonaqueous etching with a 0.1% bromine solution in ethanol [13]. Several bridges with a width between 1 and 16 μm and a length of 6 or 10 μm were patterned across the bicrystal grain boundary for the junction measurements. A schematic diagram showing the crystallographic relations of the bicrystal junctions is given in Fig. 1. High resolution electron microscopy (HREM) studies of the films were carried out on a JEOL 4000 EX microscope. The surface morphology of the film and the grain boundary were investigated with a Molecular Imaging Picoscan atomic force microscope (AFM) in the acoustic mode.

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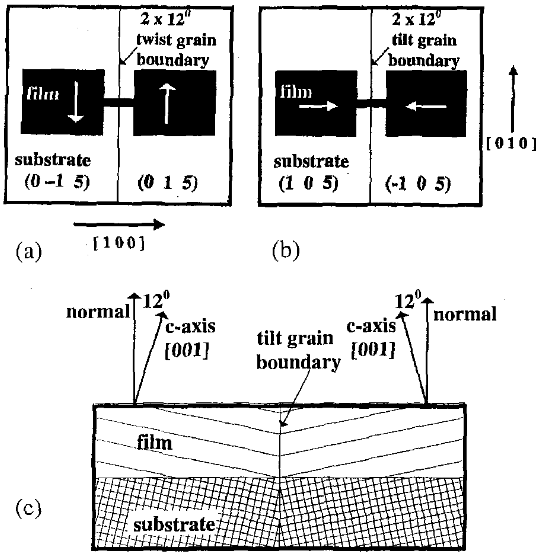

Fig. 1. Schematic diagram showing the crystallographic relations of the bicrystal junction. The white arrows in (a) and (b) show the projection of the film c-axis onto the substrate for the twist and tilt boundary.

III. RESULTS

A. *Growth and microstructure of films on vicinal substrates*

In agreement with studies by e. g. Vassilojannis [1] or Mechin et al [2] the epitaxial $YBa_2Cu_3O_7$ and $PrBa_2Cu_3O_7$ thin films and heterostructures deposited on 1.7° vicinal substrates of (100) oriented $SrTiO_3$ or $NdGaO_3$ by high pressure oxygen sputtering show a significantly lower surface roughness compared to c-axis oriented films. This effect is more pronounced for $PrBa_2Cu_3O_7$ films which have a better microstructure with less stacking faults. Fig. 2 shows an example of an AFM image for a 200 nm thick $PrBa_2Cu_3O_7$ film on a 1.7° vicinal substrate with unit cell steps.

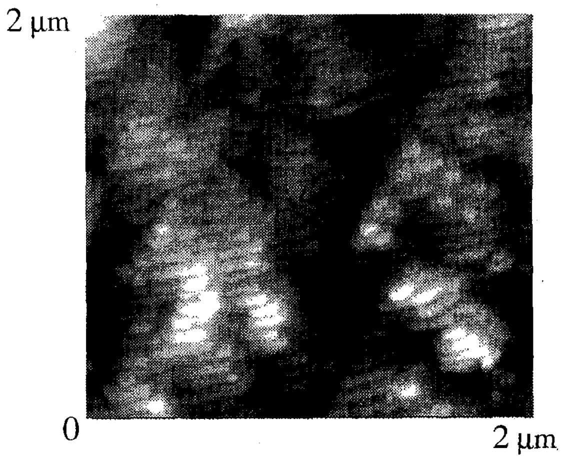

Fig. 2. AFM image of the surface of a 200 nm thick $PrBa_2Cu_3O_7$ film on a 1.7° vicinal $SrTiO_3$ substrate demonstrating a low surface roughness of about 3nm over an area of 2 μm x2 μm due to step-flow growth. Surface steps of one unit cell height (c~ 1.2 nm) can be recognized.

A cross-sectional HRTEM image of an epitaxial $PrBa_2Cu_3O_7$ /$YBa_2Cu_3O_7$ heterostructure film on a 1.7° vicinal $SrTiO_3$ substrate is shown in Fig. 3. One can see antiphase boundaries in the $PrBa_2Cu_3O_7$ film emerging from substrate steps (white arrows). They stop at the $YBa_2Cu_3O_7$ interface where stacking faults are introduced.

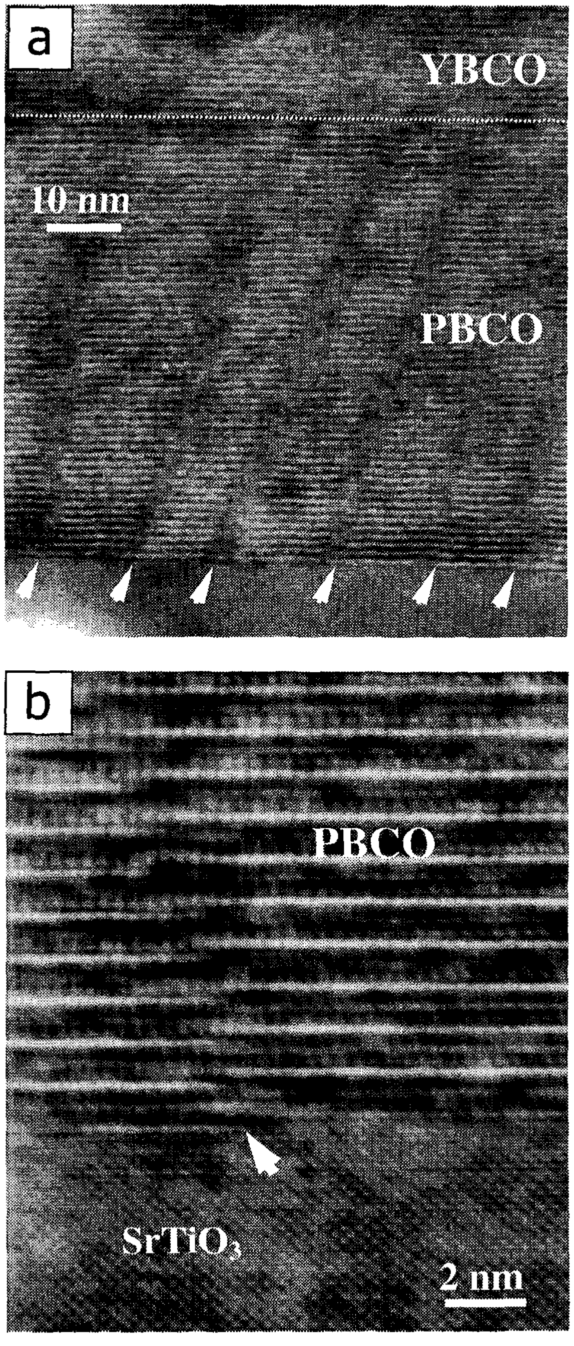

Fig. 3. Cross-sectional HRTEM image of an epitaxial $PrBa_2Cu_3O_7$ /$YBa_2Cu_3O_7$ heterostructure film on a 1.7° vicinal $SrTiO_3$ substrate. In picture (a) one can see equidistant antiphase boundaries in the $PrBa_2Cu_3O_7$ (PBCO). They emerge from substrate steps (white arrows)and stop at the $YBa_2Cu_3O_7$ (YBCO) interface. (b) shows a detail near the substrate step. The viewing direction is along [010].

For tilted c-axis $YBa_2Cu_3O_7$ films with higher tilt angles the surface roughness increases and elongated grains along the ab-planes of $YBa_2Cu_3O_7$ appear. An AFM image (2 µm x2 µm) of such a (1 0 15) oriented film on a (1 0 5) oriented $SrTiO_3$ substrate (see also Fig. 1) is shown in Fig. 4a. In Fig. 4b (4 µm x4 µm) one can recognize a [100]-tilt grain boundary (arrows) of the "valley" type, where the c-axes of both crystal parts were 12° tilted towards the grain boundary (see also Fig. 1c). The meandering width was of the order of the grain width which was about 100 nm. Such bicrystals were used for the junction measurements presented in the next section. [100]-tilt grain boundary of the "roof" type where the c-axes of both crystal parts were 12° tilted away from the grain boundary showed a much wider and deeper surface suppression at the boundary and were not electrically characterized.

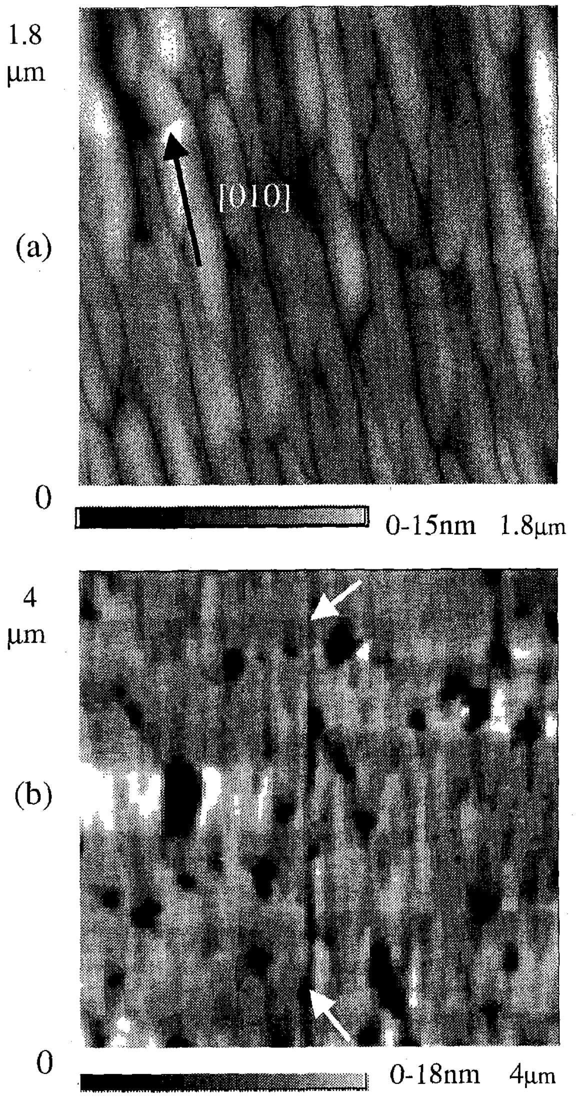

Fig. 4. AFM image of the surface of a $YBa_2Cu_3O_7$ film on a 2x12° bicrystal substrate. (a): film, (b): electrodes with [100]-tilt grain boundary (arrows).

B. Properties of grain boundary Josephson junctions on tilted bicrystals

In order to prevent cracking we sputtered $YBa_2Cu_3O_7$ films which only were about 100nm thick for the grain boundary Josephson junctions on tilted bicrystals. Different types of boundaries namely [100]-tilt grain boundaries and [100]-twist grain boundaries were investigated for Josephson junctions of different width. In table I some junction properties for several junctions on several chips (A, B, C, D, E) –which were produced in three different sputtering systems- are listed.

TABLE I

BICRYSTAL JUNCTION PROPERTIES

Sample Number	Ic Rn [mV]		Rn [Ohm]	Junction width [µm]	Type 2x12° [100]-
	4.2 K	77 K			
A1	6.75	0.46	1.3	5	tilt
A2	7	0.63	1.8	3	tilt
A3	7	0.3	1.3	5	tilt
B1	8.3	1.1	1.8	2.5	tilt
B2	7	1.0	1.5	2.5	tilt
B3	8	1.2	1.9	2.5	tilt
B4	7.5	1.1	1.5	2.5	tilt
C1		0.3	1.0	8.5	tilt
C2		0.23	1.5	6	tilt
C3		0.6	2.7	3.5	tilt
C4		0.72	0.6	16	tilt
E1	3	0.2	12	4	twist
E2	3.4		26	4	twist

All films were approximately 100 nm thick. As the patterning of the film was performed by chemical etching with resulting sloping edges the given junction width is an approximate value only. The junction resistance R_n did not significantly change with temperature. Three bicrystals (samples A, B, C) containing 3 or 4 junctions of the [100]-tilt type and one bicrystal chip (sample E) with 2 twist junctions were investigated.

Some of the junctions showed RSJ-like IV-characteristics (samples B1-B4, samples A1-A3 at 4.2K, samples E1and E2) while others exhibited excess current contributions (samples A1-A3 at 77K, samples C1-C4 at 77K).

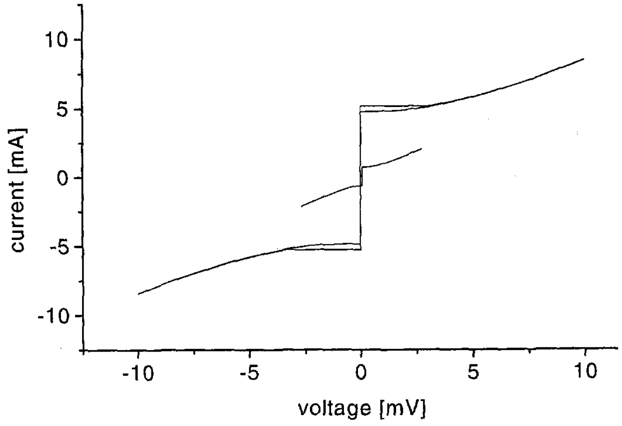

Fig. 5. IV-characteristics of the 2.5 µm wide junction B2 at 4.2K with a slightly hysteretic curve in the range up to 10 mV and at 77K in the range up to 2.5 mV (small curve in the center).

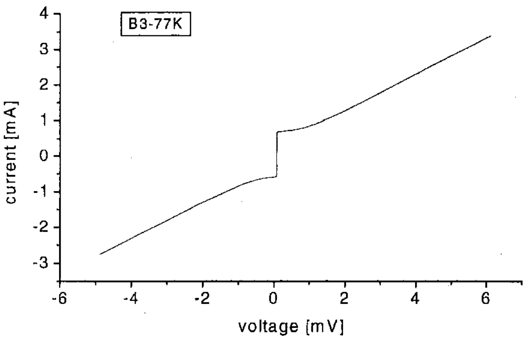

Fig. 6. IV-characteristic of junction B3 at 77K.

All junctions were slightly hysteretic at a temperature of 4.2K and non-hysteretic at 77K (Fig. 5,6). The critical current densities of the tilt junctions (estimated from table) varied between 1-2 x 10^6 A/cm^2 at 4.2K and between 0.3-2 x 10^5 A/cm^2 at 77K. Only at higher temperatures (77K) and at voltages above about 7 mV the differential resistance started to increase, indicating flux flow behavior of the c-axis tilted film or a heating effect on the junction. IV-curve instabilities with hysteretic behavior -probably of magnetic origin due to motion of vortices in the electrodes- could often be observed for junctions biased with high current densities.

IV. DISCUSSION

The scattering of the values for I_c and R_n is not too large for junctions of comparable width on one chip as can be seen for the junctions B1-B4 in table I. This might be explained by the fact that the [100]-tilt grain boundary is quite straight showing not much faceting in comparison to conventional [001]-tilt boundaries as can be seen from Fig. 4b.

If one compares the I_c R_n values at 77 K from table I with the data of bicrystal junctions with [001]-tilt boundaries [7], [8] one finds similar results but for junctions at much lower temperatures. While the critical current densities j_c of about 1-2x10^6 A/cm^2 are higher than usual and only comparable with the results for Ca-doped $YBa_2Cu_3O_7$ films [14], the junction resistance R_n is higher.

The mentioned instabilities of the IV-curve at high current densities is presumably due to the motion of vortices in the electrodes. It was actually observed [15] that c-axis tilted films show more easy flux motion in comparison with c-axis oriented ones. The fact that after several IV-curve sweeps or by limiting the bias voltage to a few millivolts these instabilities often disappeared, also indicates the hopping of vortices. While the vortex motion might be beneficial for some devices like flux flow transistors it is usually important for practical applications of junctions to enhance the flux pinning in tilted films. This might be established by surface, edge or substrate induced pinning. Actually Czerwinka et al [4] found high values of critical currents with nearly no anisotropy for 10° tilted films. Also by using layouts for the patterning of the junction electrodes which avoid high transport current densities in directions which are along the tilt might help to avoid this problem in the future.

As discussed by Mannhart et al [16] or Tafuri et al [10] due to symmetry reasons the order parameter at the grain boundary should be not so much suppressed in the case of [100]-tilt or [100]-twist boundaries in comparison to conventional [001]-tilt boundaries. Furthermore the local strain of the grain boundary should be lower from crystallographic reasons for the first kind of junctions. The same holds for the grain boundary faceting, as growth islands are usually faceted along the [100] and [010]-direction of the film one of them being parallel to the grain boundary. HRTEM studies of the structural details of the [100]-tilt boundary might give more insight to this open question.

V. CONCLUSIONS

It has been shown that use of slightly tilted substrates for the growth of high-T_c films leads to an improved surface roughness. Josephson junctions produced on vicinal bicrystals of the 2x12° [100]-tilt type show very high I_c R_n – products of up to 1.2 meV at 77 K and up to 8 meV at 4.2 K. The reasons might be attributed to the grain boundary microstructure and the different symmetry relations of the order parameter across the boundary in comparison with conventional [001]-tilt boundaries.

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