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$YBa_2Cu_3O_{7-\delta}$ *a*-axis films and planar junctions

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***Abstract*—Among the concepts for Josephson junctions with artificial barriers, planar junctions are most favorable, because they can be deposited *in situ*.**

In order to prepare planar junctions, multi-layers were sputtered, using $YBa_2Cu_3O_{7-\delta}$ as superconductor and $PrBa_2Cu_3O_{7-\delta}$ as barrier material. The sandwiches are *a*-axis oriented. Out of these, single junctions were etched, planarized with CeO_2. Finally gold contacts were sputtered. The junctions are squares of size 20×20 to $100 \times 100\,\mu m^2$.

A prerequisite for a successful preparation of junctions out of *a*-axis oriented multi-layers is a study of such films. We deposited the films with RF off-axis sputtering. The films were characterized electrically. Their morphology was investigated by XRD, AFM and TEM. The films are very smooth and have a grain size of below 100×100 nm^2. To improve T_c and crystal quality, template layers were used. The T_c of a single film is about 62 K, using a template layer up to 78 K were reached. For *a*-axis oriented growth not only a reduction of the deposition temperature is important, but also the growth rate must be high enough. Best results were obtained at rates higher than 150 nm/h.

The first planar junctions show a supercurrent, but otherwise rounded *I*-*V*-curves.

I. INTRODUCTION

Both, the structural and the superconducting properties of high-T_c superconductors are anisotropic. The superconducting coherence length, for instance, is much shorter in the *c*-axis direction than in the *a*-axis direction. Therefore, in most high-T_c Josephson devices the supercurrent is coupled along the CuO-planes, *i. e.* the *ab*-planes.

On the other hand, only one junction configuration comprising artificial barriers offers the possibility of depositing the superconducting electrodes and the barrier *in situ*: the planar junction. Firstly, a multi-layer sandwich consisting of superconductor, barrier and superconductor is deposited. Secondly, Josephson junctions and the electrical contacts are formed without damaging the device. This has been demonstrated by different groups [1]–[3].

Only with *a*-axis oriented sandwiches can the advantages of *in situ* growth and coupling in *ab*-direction be combined in one device [4].

In order to achieve reproducible junction characteristics, all layers must be smooth (to avoid pinholes) and be of high crystal quality. The anisotropy of the growth rate of the cuprates gives the possibility of growing films with different orientations, with the *c*-axis perpendicular to the surface (*c*-axis oriented films) or with the *a*-axis perpendicular to it (*a*-axis oriented films). *a*-Axis oriented $YBa_2Cu_3O_{7-\delta}$ thin films can be obtained by lowering the substrate temperature during deposition by about ~ 100 °C compared to the optimum temperature for *c*-axis films [5]. The use of a template layer can improve the crystal quality and the superconducting properties. $YBa_2Cu_3O_{7-\delta}$ (YBCO), as a self-template, and $PrBa_2Cu_3O_{7-\delta}$ (PBCO) are mainly used for this purpose [6], [7]. After deposition of a template layer the superconducting film can be deposited at the optimum temperature for *c*-axis growth. Nevertheless, it will continue growing in the *a*-axis orientation. As we will show, however, not only the deposition temperature, but also the deposition rate, is an important parameter in this context. If the rate is too low, a fairly large amount of *c*-axis material will still be present in the film.

In this paper we present our preliminary results on planar junctions. These are made of *a*-axis oriented $YBa_2Cu_3O_{7-\delta}$ films, which are very smooth and show high crystal quality.

II. *a*-AXIS FILMS

A. Preparation and results

1) The sputter process: The films are deposited with RF magnetron sputtering in an off-axis configuration on $SrTiO_3$ (100) substrates. The total pressure of the argon-oxygen gas-mixture is typically about 6 Pa and the volume ratio of argon to oxygen 1:2. Some water vapor (0.1 Pa) was added to the sputter gas in order to improve the crystal quality. Films were prepared, both with and without a template layer. Single YBCO layers and template layers were deposited at a temperature of about 640 °C. The template materials are YBCO and mostly

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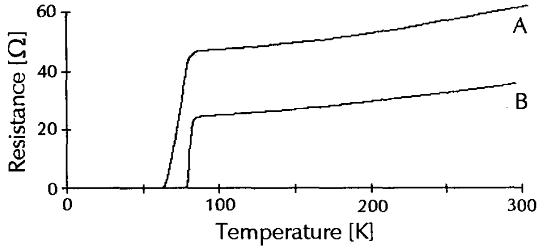


Fig. 1. Resistance vs. temperature curves of optimized *a*-axis oriented YBCO films. Sample A is *without* template layer and sample B *with* YBCO self-template. No difference between YBCO and PBCO template layers has been seen.

PBCO. Additional layers were sputtered at 750 °C. These temperatures are measured in the heater block just below the substrate.

2) The results: Initially we tried to deposit *a*-axis oriented films *without* a template layer by using exactly the same deposition conditions as for *c*-axis films, except for a lower substrate temperature. The largest amount of *a*-axis material was obtained at a temperature about 110 °C below the optimal temperature for *c*-axis growth, but these films still contained *c*-axis oriented material.

For YBCO and PBCO we estimated the relative volumes of the different crystal orientations using X-ray diffraction (XRD). Only the *a*- and *c*-axis orientation were observed.

They were distinguished in a ψ-ϕ-scan. (102)-reflections occur at ψ-angles of about ~ 33° for *a*-axis growth and ~ 57° for *c*-axis growth (for both cuprates). The ϕ-scans of *a*-axis oriented YBCO films on $SrTiO_3$ (100) exhibit four-fold symmetry as a consequence of twinning. The relative amount of *c*-axis material is obtained by dividing the intensity of the *c*-axis signal by the sum of the *c*- and *a*-axis signals. Before this calculation the background is subtracted.

In these first experiments the deposition rate was not higher than 30 nm/h (= 0.08 Å/sec) and the amount of *c*-axis oriented material did not fall below 40 %. It turned out, however, that not only the sputter temperature, but also the sputter rate is a critical parameter in the fabrication of *a*-axis films. Because *a*- and *c*-axis phases grow in different modes, the films were very rough, as was observed by scanning electron Microscopy (SEM). The electrical properties of the films were dominated by the large amounts of *c*-axis material.

By changing the sputter gun we were able to increase the rate to more than 150 nm/h (= 0.4 Å/sec). This drastically changed the film growth.

$YBa_2Cu_3O_{7-\delta}$ films grown *without* a template layer exhibit a maximum T_c of 62 K (fig. 1); the transition onset lies at 86 K. The resistance decreases with decreasing temperature, before a very broad transition to the superconducting state occurs. This non-ideal behavior can attributed to the imperfections of the crystal structure.

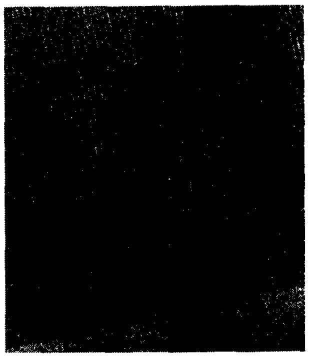
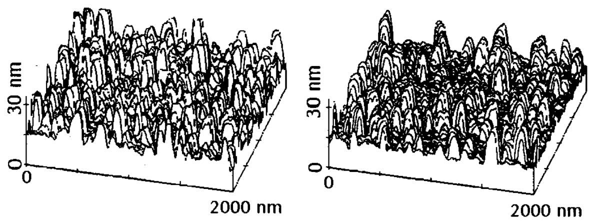


Fig. 2. TEM picture of an *a*-axis film with YBCO self-template. *c*-Axis growth is found at the interface, which can be assigned to stress relaxation.

To improve the crystal quality and the superconducting properties the use of a template layer is advantageous. Mostly used are YBCO (self-template) and PBCO, because the latter tends more to *a*-axis growth [8]. The template layer must not be too thin in order to give a good stress relaxation, but a too thick one can lead to roughening. We chose a thickness of 25 nm; a thicker template does not lead to an improvement of crystal quality and T_c.

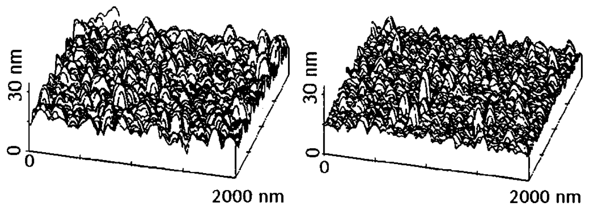
With the template layer technique T_c was raised to 78.4 K (fig. 1), although the onset temperature remained nearly constant (87 K). No variation between films with YBCO and PBCO template layers has been observed. The critical current density $j_\mathrm{c}(T)$ parallel to the substrate surface exceeded $6 \cdot 10^4$ A/cm^2 at a temperature of 10 K below T_c.

A transmission electron microscopy (TEM) picture was prepared (fig. 2) for such a sample. It shows nearly perfect *a*-axis growth. This is in agreement with the XRD results: the amount of *a*-axis material lies above 95%. At the surface of the $SrTiO_3$ (100) substrate a *c*-axis oriented layer is observed, which is between 30 and 70 Å thick. This layer is present over the whole cross-section. No direct *a*-axis growth on top of the substrate is found. A careful examination of the TEM picture reveals, that the thickness of the thin *c*-axis layer varies over the substrate. The starting layer is deposited at a temperature, where only the *a*-axis oriented phase can be formed, but the lattice mismatch induces strain and this may cause a cation rearrangement at growth temperatures of more than 700 °C. Other groups have also reported *c*-axis material at the substrate interface [9]–[11]; HAMET *et al.* also gave a possible mechanism for the cation rearrangement [6]. At the points of maximum strain thick *c*-axis layers are found. Where the lattice of substrate and film is better matched, fewer layers of *c*-axis material are formed. Because $SrTiO_3$ (100) offers a square unitcell, *a*-axis oriented YBCO grows twinned. This was also observed in the TEM pictures. The size of the *a*-axis grains is in



rate = 109 nm/h, Tc = 48.4 K
roughness = 4.7 nm (rms)

rate = 126 nm/h, Tc = 61.1 K
roughness = 4.1 nm (rms)



rate = 142 nm/h, Tc = 71.5 K
roughness = 2.8 nm (rms)

rate = 148 nm/h, Tc = 67.2 K
roughness = 2.0 nm (rms)

Fig. 3. AFM pictures for four films, that were sputtered in the same run, but with different deposition rates. The indicated numbers are the deposition rate, T_c and the roughness (rms).

agreement with the size estimated by AFM.

If the deposition conditions are off optimum, a mixture of *a*- and *c*-axis oriented material is deposited, as is found by XRD. However, it appears in those cases that the T_c is higher, the R(T)-curve is much steeper, and the ratio R(300 K)/R(100 K) lies closer to 3, than for films with small *c*-axis amount. Here the film characteristics are determined by the *c*-axis material, either in the underlying layer or in the grains. No conclusions concerning the crystal quality of the films may be taken from these film properties. This fact can explain some reported results on higher T_c for *a*-axis films.

3) Current status: Using the optimized process, we deposit films with a roughness of 1.6 nm (rms) at a total film thickness of 100 nm. When the deposition rate is reduced below 150 nm/h the properties of the films decrease drastically. Four samples were sputtered in the same run, but at different positions relative to the plasma, so the respective sputter rates vary strongly. For the samples with the lowest sputter rate T_c dropped and the roughness increased (fig. 3).

B. Discussion and interpretation

The orientation of a thin film is not only dependent on the substrate temperature, which determines the surface mobility, but also on the deposition rate, or the flux of material. When the rate is less than 150 nm/h at 640 °C not only *a*-axis, but also *c*-axis oriented grains are formed. Because of the different growth mechanisms such films will be much rougher, as discussed by PENNYCOOK *et al.* [11].

III. PLANAR JUNCTIONS

A. Preparation

We prepared planar junctions in the following way: directly on the substrate a template layer is deposited for proper growth of the first YBCO layer [7]. The whole sandwich of superconducting base electrode, barrier and superconducting top electrode is deposited *in situ*. The superconductor is YBCO and the barrier material PBCO. The typical thicknesses of the layers are 25 nm for the template, 150 – 300 nm for the base electrode, 10 – 20 nm for the barrier and 100 nm for the top electrode. The substrate temperatures were 640 °C for the template, and 750 °C for the other layers. Immediately after cooling down in 200 hPa oxygen, a covering layer of gold with a typical thickness of 60 nm is sputtered.

The structuring procedure consists of four steps, which are made using standard photolithography and argon ion-beam etching, with 500 V acceleration voltage. In the first step the junctions are formed: the etching is stopped in the base electrode, as is controlled by the etching time. Without removing the photoresist an isolating layer of 85 nm CeO_2 is deposited by pulsed laser deposition at room temperature. The lift-off of the photoresist in acetone clears the contact surfaces, especially the top electrodes of the junctions. Afterwards a second gold layer for electrical contacts is sputtered. The final etching stops in the top YBCO layer in order to separate the electrodes, for a proper four-point-measurement. The prepared junctions have sizes between $20 \times 20\ \mu m^2$ to $100 \times 100\ \mu m^2$.

CeO_2 is chosen as isolating material, because it can be deposited at room temperature and, even though it is amorphous, it isolates very well. Also the lift-off caused no problem. Earlier tests with sputtered SiO_2 degraded the underlying $YBa_2Cu_3O_{7-\delta}$ layer. A later annealing step was not possible in that case, because the Si would diffuse into the $YBa_2Cu_3O_{7-\delta}$ and degrade its properties. By contrast, CeO_2 does not attack leaves the superconducting properties. After degradation of the device due to thermal cycling the superconducting properties can be restored by annealing in 200 hPa oxygen at 500 °C.

B. Preliminary results

The 300 nm thick sandwich, consisting of template layer, superconductor, barrier and again superconductor also exhibits a roughness of only 2.3 nm (rms). From the AFM-scan the grain size was estimated to be smaller than $100 \times 100\ nm^2$ (fig. 4). This is smaller than in earlier samples. Those films were very rough and not usable for planar junctions. The etching of the junctions is critical and often degrades the superconducting properties. In fig. 5 we present the current-voltage-characteristic of a

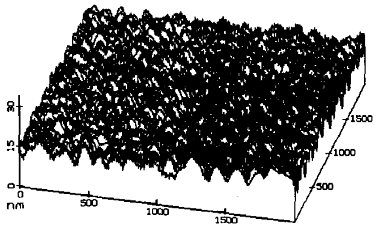


Fig. 4. AFM picture of a 300 nm thick multilayer of YBCO and PBCO for junction preparation, exhibiting a roughness of only 2.3 nm (rms).

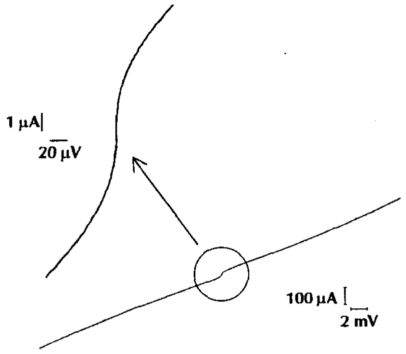


Fig. 5. I-V-curve of 50×50 μm^2 planar junction. The barrier thickness is 10 nm.

sample showing with a barrier thickness of 10 nm. This junction shows a critical current I_c of 3.9 μA and and a normalstate resistance R_N of 0.2 mV. With an area of 50×50 μm^2 a current density j_c of 0.16 A/cm^2 can be calculated. The magnetic field and microwave dependence has not been established yet.

In the multi-step process for the junction preparation the underlying YBCO layer and the junctions themselves are the most sensitive parts. A degradation due to the etching is observed in the R(T)-measurements. A further improvement of the fabrication process is necessary.

IV. SUMMARY

a-Axis oriented thin films and planar junctions have been prepared and characterized.

It was shown that by RF magnetron sputtering at high deposition rates, *i.e.* 150 nm/h or higher, high quality *a*-axis oriented YBCO layers or trilayers YBCO/PBCO/YBCO can be grown. In optimized *a*-axis oriented films no *c*-axis oriented material can be detected. If the flux of material lies below the above threshold, the resulting films contain a large amount of *c*-axis oriented material.

Carefully optimized films are very smooth, with a roughness of about 1.6 nm (rms) for 100 nm film thickness. For 300 nm thick YBCO/PBCO/YBCO multilayers for junction preparation the roughness increases only slightly to 2.3 nm.

The junctions exhibit small supercurrents, but otherwise rounded I-V-characteristics, due to degradation by the etching process.

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