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Optimization of electrical and structural parameters of $YBa_2Cu_3O_{7-x}$ thin-film bicrystal Josephson junctions with chemical and thermal treatments of substrates

I Gundareva[1,2] and Y Divin[1,2]
[1]Peter Grünberg Institute, Forschungszentrum Jülich, Jülich, Germany
[2]Kotel'nikov Institute of Radio Engineering and Electronics, Russian Academy of Sciences, Moscow, Russian Federation

E-mail: i.gundareva@fz-juelich.de

Abstract. The [100]-tilt high-T_c Josephson junctions are characterized by high characteristic voltages I_cR_n but their current range is limited by low critical currents I_{cf} of the thin-film electrodes with tilted *c*-axes. The impact of chemical and thermal treatments of $NdGaO_3$ substrates on morphology and *IV* curves of the $YBa_2Cu_3O_{7-x}$ thin films and the bicrystal thin-film junctions have been studied. It was found that *IV* curves of the films above a critical current I_{cf} are described by a power-law dependence $V = V_0(I/I_{cf} - 1)^n$, where I_{cf}, V_0 and n are dependent on the *c*-axis tilt of the film and treatment of the substrate. Critical current densities j_c(78K) up to $1.4 \cdot 10^7$ A/cm^2 have been reached in 1.7°-tilt $YBa_2Cu_3O_{7-x}$ films perpendicular to the tilt. Bicrystal junctions have been fabricated with optimized $YBa_2Cu_3O_{7-x}$ thin films, and RSJ-like *IV*-curves in the current ranges from I_c up to 5 I_c with I_cR_n (78K)-values of 1 mV have been achieved.

1. Introduction

High-T_c Josephson junctions are often fabricated by thin-film deposition on bicrystal substrates [1]. When compared with the *c*-axis $YBa_2Cu_3O_{7-x}$ thin-film bicrystal junctions, the bicrystal junctions made from the $YBa_2Cu_3O_{7-x}$ films with mutually tilted *c*-axes have demonstrated higher structural homogeneity, higher critical current density j_{cj}, and higher characteristic voltage I_cR_n up to 10 mV [2]. Such junctions can be used in THz detection and spectral analysis [3, 4] even at the voltages V above I_cR_n and corresponding Josephson frequencies exceeding 5 THz. However, due to the tilt of the *c*-axes the critical current densities j_{cf} of the tilted thin films along the tilt [5] are lower than that of the *c*-axis films. The motion of magnetic vortices in thin-film electrodes at $j > j_{cf}$ might contribute to the total voltage and this phenomenon limits the maximum current I where only the junction contributes to the measured voltage to the values below I_{cf}. Thus, optimization of substrate surface treatment, a crucial step for fabrication of high-quality epitaxial thin films, should be carried out.

Previous studies [6] on chemical and thermal treatments of substrates and epitaxial $YBa_2Cu_3O_{7-x}$ films did not report on electrical characterisation. Here, we present electrical characteristics of $YBa_2Cu_3O_{7-x}$ thin films, both *c*-axis and with tilted *c*-axes, fabricated on $NdGaO_3$ substrates after such treatments. The electrical characteristics of bicrystal junctions fabricated with optimized thin films are also presented.

2. Experimental details

The $NdGaO_3$ substrates, with the orientations of the main planes of (110), (110) with 1.7°-tilt around [001], (320) and (230), were used in fabrication of $YBa_2Cu_3O_{7-x}$ thin films. Bicrystal substrates made from (320) parts were used for junction fabrication. After chemical cleaning in acetone and ethanol, the substrates were rinsed with deionized water in an ultrasonic bath for 10 min, etched for 0.5 - 8 min in a buffered hydrofluoric (BHF) solution prepared with 10 ml commercial BHF (12.5 vol% HF + 87.5 vol% NH_4F) + 4 ml NH_4OH (37 vol%) + 90 ml H_20, after ref. [6]. The substrates were subsequently annealed in oxygen [6], [7], but the temperature of the heater was varied from 800°C to 1000°C. The annealing was made under constant oxygen flow during 20- 240 min and at pressure of 1 bar.

$YBa_2Cu_3O_{7-x}$ thin films were deposited on $NdGaO_3$ substrates at a heater temperature of 950°C by dc sputtering in pure oxygen at pressure of 3.4 mbar. These sputtering parameters were optimized to decrease *a*-axis inclusions in the predominantly *c*-axis thin films. The thickness t of the films was in the range of 60-200 nm. The films and junctions were patterned in the form of bridges by UV lithography with wet etching in 0.1% bromine-ethanol solution. The widths w of the bridges were measured by AFM in the range from 2 to 18 μm with a width error of ±0.5μm, which was due to inhomogeneous etching of the film edges.

3. Results and discussion

The effect of substrate treatment on the morphology of the *c*-axis thin films was studied by varying etching time in the BHF solution from 0.5 to 8 min. The smoother film surface was obtained with an etching time of 2 min (Figure 1a). The films sputtered on the substrates without chemical treatment as well as on the substrates subjected to longer BHF treatment (more than 2 min) had up to 5 times more *a*-axis grains. So, short-time chemical etching of $NdGaO_3$ substrates in the BHF solution improves the quality of the substrates as in [6]. The thermal treatment used in [6] did not result in improvement of the morphology of the films. After substrate annealing at temperatures T_a of 950-1000°C and at T_a = 910°C the concentrations of the *a*-axis grains in the *c*-axis thin films were 8 and 3 times larger, correspondingly, than those in films without thermal treatment of the substrates.

The $YBa_2Cu_3O_{7-x}$ thin films grown on (110) $NdGaO_3$ substrates with 1.7°-tilt demonstrated a transition from island growth to step-flow growth (Figure 1b). The morphology of an $YBa_2Cu_3O_{7-x}$ thin film deposited on (320) $NdGaO_3$ substrate is shown in Figure 1c. The (320) $NdGaO_3$ substrate was etched in BHF and, additionally, annealed in oxygen atmosphere. A set of terraces and steps between them was observed on the surface of the $YBa_2Cu_3O_{7-x}$ film (Figure 1c), with the step heights much larger than the $YBa_2Cu_3O_{7-x}$ cell size. This step-bunching on the surface of the $YBa_2Cu_3O_{7-x}$ films with tilted *c*-axes was found to depend on the duration and temperature of substrate annealing and could be varied from 10 nm for low-temperature (T = 800°C) or short-term (about 20 min) high-temperature (T = 950°C) annealing to 20-30 nm for high-temperature annealing (T = 950°C) for more

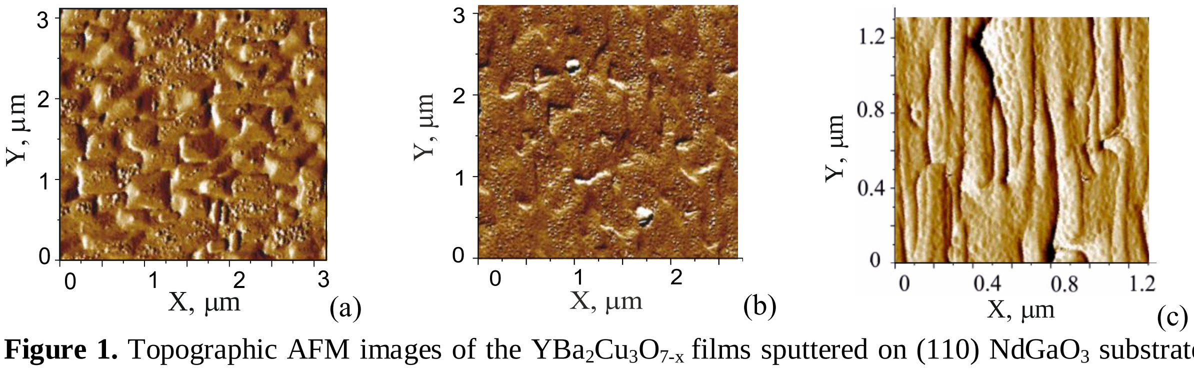

Figure 1. Topographic AFM images of the $YBa_2Cu_3O_{7-x}$ films sputtered on (110) $NdGaO_3$ substrate after 2′-BHF treatment (a); (110) $NdGaO_3$ with 1.7°-tilt after 5′-BHF treatment (b); (320) $NdGaO_3$, after 2′-BHF treatment and 20′-annealing at T=950°C (c). The tilt direction is to the left in (b) and (c).

than 0.5 hour, and this step-bunching is a consequence of the step-bunching on the surface of the substrate due to high-temperature annealing in oxygen atmosphere [7].

The treatment of the substrate surfaces had a large effect on the electrical characteristics of the $YBa_2Cu_3O_{7-x}$ thin films. The critical current densities j vs. voltages V for $YBa_2Cu_3O_{7-x}$ thin-film bridges deposited on $NdGaO_3$ substrates of various orientations and treatments are presented in Figure 2. Current densities $j = I/wt$ have been calculated for all fabricated bridges from the experimental current I values. The resulting $j(V)$-curves of fabricated $YBa_2Cu_3O_{7-x}$ thin films were fitted to the following expression:

$$V = V_0 \,\mathrm{sign} j\, (|j|/j_{cf} - 1)^n, \quad \text{if } |j| > j_{cf};$$
$$V = 0, \quad \text{if } |j| \le j_{cf}. \qquad (1)$$

Using (1) and a nonlinear-fit program [8], an accuracy of a few percent have been reached for j_{cf} and n. The c-axis $YBa_2Cu_3O_{7-x}$ films sputtered on the untreated (110) NGO substrates show j_{cf} values around $4{\cdot}10^6$ A/cм^2 (Figure 2, curve 1), and these values were increased up to $5{\cdot}10^6$ A/cм^2 for the c-axis $YBa_2Cu_3O_{7-x}$ films sputtered on the substrates, which were shortly (2 min) etched in BHF (Figure 2, curve 2). The index n also increased from 3.2 to 4.1. The $YBa_2Cu_3O_{7-x}$ films with 1.7°-tilt of the c-axes showed a considerable increase of the j_{cf} up to $1.4{\cdot}10^7$ A/cm^2 (Figure 2, curve 3) in the direction perpendicular to the tilt, with n = 1.4. The j_{cf} -values for $YBa_2Cu_3O_{7-x}$ films sputtered on unannealed (320) $NdGaO_3$ substrates were around $4.5{\cdot}10^5$ A/cm^2 along the tilt (curve 4), for the films on shortly annealed substrates these values dropped to $2{\cdot}10^5$ (curve 5) and the j_c-values increased to $4{\cdot}10^5$ A/cm^2, when (320) substrates were annealed in oxygen for 4 hours (curve 6). The n-indices were equal to 2.5; 1.9 and 2.9 for curves 4, 5 and 6, correspondingly.

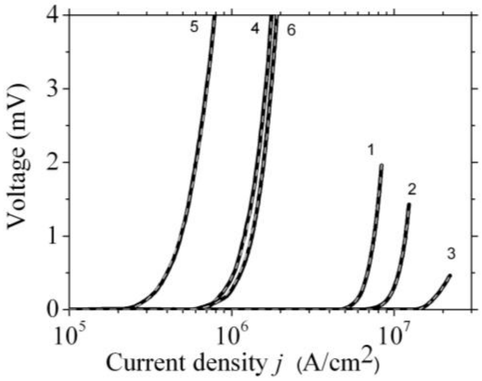

Figure 2. Experimental (solid) and fitted (dash) dependences of voltage vs. current density for $YBa_2Cu_3O_{7-x}$ thin films on the substrates: 1 - (110) NGO untreated, 2 - (110) NGO etched in BHF, 3 - 1.7°tilted (110) NGO etched in BHF, 4 - (320) NGO untreated, 5 -(320) NGO annealed 20 min., 6 - (320) NGO annealed 4 hours. T= 77.8 K

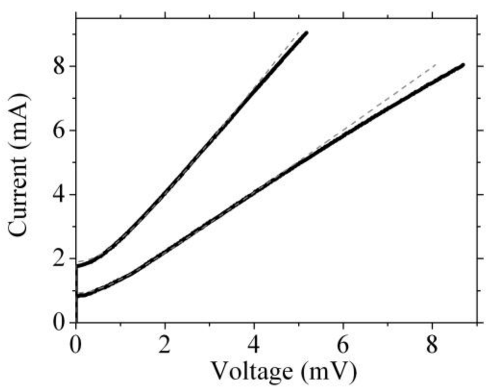

Figure 3. IV- curves of $YBa_2Cu_3O_{7-x}$ bicrystal junctions fabricated on (320) $NdGaO_3$ bicrystal substrates (solid lines) and corresponding IV-curves from the RSJ model (dash lines) with the following fitting parameters: 1 - I_cR_n = 1.08 mV, R_n = 0.56 Ω; 2 - I_cR_n = 0.93 mV, R_n = 1.0 Ω. T = 77.8 K

The resistive state of superconductors is explained by motion of the magnetic vortices. In the presence of static disorder in a superconductor, the IV-curve at $j \ge j_c$ has a non-ohmic form, which depends on the dimension of the superconducting material and on the topology of the induced flux motion [9]. The critical scaling applied to the IV-curves of the superconductors with pinning centers gives the power law $V \propto j^{\,n}$ [9], which is similar to the IV curves in Figure 2. The n-value of 3.2

obtained for our *c*-axis $YBa_2Cu_3O_{7-x}$ thin films is close to the corresponding value of n = (2.9±0.2) obtained earlier for the *c*-axis $YBa_2Cu_3O_{7-x}$ thin films on $SrTiO_3$ substrates [10].

The [100]-tilt $YBa_2Cu_3O_{7-x}$ bicrystal junctions consisting of optimized $YBa_2Cu_3O_{7-x}$ thin-film electrodes with 2x11.3° mutually tilted *c*-axes were fabricated on (320) bicrystal $NdGaO_3$ substrates. The measured *IV* curves of the junctions at T = 77.8 K are presented in Figure 3 by solid lines, and corresponding RSJ-like *IV* curves - by dash lines. The optimized bicrystal junctions are characterized by high I_cR_n -values of around 1 mV and follow the RSJ-like *IV* curves in the current range from I_c to $4I_c$ for the junction 1 and from I_c to $5I_c$ for the junction 2. The deviations of the experimental curves from the RSJ curves at the currents I, which are more than 4-5 times larger than the critical currents I_c of the bicrystal junctions, originate from the vortex motion in the junction thin-film electrodes with tilted *c*-axes at $I > I_{cf}$. These optimized junctions at the temperature of 78 K can be used as Josephson detectors for electromagnetic radiation up to the voltages of 4 mV and Josephson frequencies of 2 THz.

4. Conclusions

In this paper, electrical and structural characteristics of $YBa_2Cu_3O_{7-x}$ thin films fabricated on the $NdGaO_3$ substrates subjected to thermal and chemical treatments are presented. Optimization of the sputter deposition parameters and substrate treatment yielded critical current density j_c up to $1.4\cdot10^7$ A/cm^2 for $YBa_2Cu_3O_{7-x}$ films with 1.7°-tilt of the *c*-axis. Critical current densities for [100]-tilt $YBa_2Cu_3O_{7-x}$ films sputtered on (320) $NdGaO_3$ substrates strongly depended on the time and temperature of preliminary annealing of the substrates in oxygen and on the size of the bridges. The shape of the *IV*-curves of $YBa_2Cu_3O_{7-x}$ films was found to be described by a power law $V = V_0\ (I/I_c - 1)^n$, $I > I_c$, where the index n is in the range of $1.4 < n < 4$ and depends on the *c*-axis tilt and on the time and temperature of preliminary annealing of the substrate in oxygen atmosphere. Obtained dependencies and reproducibility of the electrical characteristics of $YBa_2Cu_3O_{7-x}$ thin films were used to fabricate high quality [100]-tilt $YBa_2Cu_3O_{7-x}$ bicrystal Josephson junctions with I_cR_n-values of 1 mV and RSJ-like *IV*-curves in the current range from I_c up to $5I_c$.

Acknowledgments

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