

TILTED BI-CRYSTAL SAPPHIRE SUBSTRATES IMPROVE PROPERTIES OF GRAIN BOUNDARY $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ JUNCTIONS AND EXTEND THEIR JOSEPHSON RESPONSE TO THZ FREQUENCIES

E. STEPANTSOV^{1,2,A)}, M. TARASOV^{1,3,A)}, A. KALABUKHOV^{1,4}, L. KUZMIN¹, AND
T. CLAESON¹

¹ *Microtechnology and Nanoscience, Chalmers Univ. of Technol., SE 41296 Gothenburg, Sweden*

² *Institute of Crystallography RAS, 117333, Moscow, Russia*

³ *Institute of Radio Engineering and Electronics RAS, 101999, Moscow, Russia*

⁴ *Department of Physics, Moscow State University, 119899, Russia*

We have studied the very high frequency response of high critical temperature Josephson junctions. High quality $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ epitaxial films were fabricated by pulsed laser deposition on tilted (vicinal) sapphire substrates with CeO_2 buffer layers. YBaCuO films have smaller tilt angles, from 1.0° up to 10.3° , compared to inclination angles of the substrates from 1.5° to 13.6° . X-ray diffraction shows only a single orientation of the films in the a - b plane, as well as an absence of a -axis particles and outgrowths. Critical temperatures as high as $T_c=88.5$ – 89.0 K and $\Delta T_c \leq 1.5$ K were obtained in all films. The meandering of the artificial grain boundary in a tilted bicrystal film is three times less than in an in-plane (un-tilted) bicrystal. Josephson junctions of widths from 1.5 to $6 \mu\text{m}$ were tested at temperatures from 0.26 K to 77 K. $I_c R_n$ products of up to 4.5 mV were observed at $T=4.2$ K. Shapiro steps were observed at voltages over 3 mV under 300 GHz irradiation. Josephson radiation was measured at frequencies up to 1.7 THz by a cryogenic bolometer. Suppressing the critical current with a magnetic field can separate Josephson radiation and thermal radiation. A parabolic dependence of the response on bias voltage for thermal radiation corresponds to an increase of junction temperature from 260 mK at zero bias to 3 K at 1 mV bias.

1. Introduction

Receivers for radioastronomy, remote atmosphere monitoring, biomedical research, or communications require new highly sensitive detectors and broadband tunable oscillators at terahertz frequency. The novel high critical temperature superconductors (HTS) potentially lift the upper frequency limit to over 10 THz from 1 THz set by the superconductor energy gap of conventional low critical temperature devices based on Nb and NbN. Although not suitable for large scale integration, the technology of bicrystal junctions is the most developed one of Josephson junctions, and it has best performance compared to other types.

Usually, a grain boundary Josephson junction (JJ) has been epitaxially grown on a bicrystal substrate with an in-plane misorientation angle. The use of tilted substrates can improve film and junction characteristics. Then the film growth may change from island or spiral growth to step flow growth, which leads to less surface roughness and an increase of current density at low tilt angles [1,2]. Transport properties become more anisotropic for tilt angles over 5 degrees [2,3]. Tilted grain boundary junctions can have much better performance margins than those with in-plane misorientation [4,5]. The

^{a)} Electronic mail: Mikhail.Tarasov@mc2.chalmers.se, Stepantsov@ns.crys.ras.ru

tilted film grain boundary is straight and follows the bicrystal substrate boundary. It shows a low degree of meandering as compared to that of a conventional in-plane grain boundary. The pioneer work by Divin *et al.* [5] demonstrated excellent dc characteristics for YBaCuO Josephson junctions on [100]-tilt SrTiO₃ bicrystals. An $I_c R_n$ product of up to 8 mV at 4.2 K shows a potential of this technology for Terahertz frequencies (I_c is the Josephson critical current and R_n the normal state resistance). NdGaO₃ tilted bicrystal junctions developed in [6] show $I_c R_n$ up to 1.8 mV, and clear detector response was observed up to 3.1 THz. To increase the frequency margin further, we decided to use sapphire tilted bicrystal substrates.

2. Film and junction fabrication and characterization

Pulsed laser deposition was used to grow epitaxial YBaCuO films on Al₂O₃ and Y-ZrO₂ monocrystal substrates. These were tilted from a standard orientation of (10 $\bar{1}2$) and (001) by 2°-14°. Symmetric bi-crystal substrates of sapphire that had the same tilt of monocrystal parts towards the boundary direction were used. Thicknesses of all films are 250 nm. The tilt of the c-axis was in the <110> direction for all films except in one, where it was in the <100> direction. CeO₂ was deposited at 770°C in an oxygen atmosphere at 0.3 mbar as a buffer layer prior to the YBaCuO film deposition. An YBaCuO film was deposited in the same vacuum cycle at 780°C and oxygen pressure 0.6 mbar. Afterwards, an in-situ Au film was deposited. The crystalline structure of the films was investigated by x-ray diffraction, see Fig. 1.

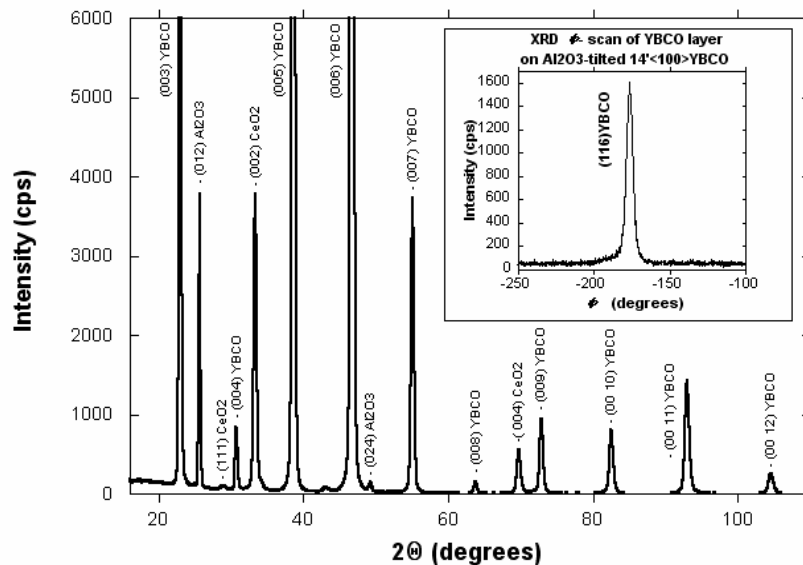


Figure 1. XRD $\theta/2\theta$ -scan of an YBaCuO thin 250 nm film grown on a CeO₂ buffered sapphire substrate with a 14° tilt of the normal to a (10 $\bar{1}2$) plane relatively the normal to the surface in direction [2 $\bar{2}01$]. This direction corresponds to a <100> YBaCuO. The inset shows a ϕ -scan around the (116) YBaCuO peak.

The exact offset of θ was determined by the x-ray measurement for the YBaCuO film, the CeO₂ buffer layer and the substrate, corresponding to the tilt angles of their crystalline lattices to the surface of the film. This offset is different for the buffer and the YBaCuO films. The main curve in Fig. 1 corresponds to the offset YBaCuO film. There are no additional peaks that can be due to CuO and α -particles. A ϕ -scan is presented in the inset. It corresponds to the (116) peak and covers 150 degrees. It shows no peaks due to other orientations of YBaCuO in the a-b plane. It means that the film is single-domain. In the main curve there are practically no substrate peaks that are determined by non-parallel orientation with deflection up to 3.5° between crystal lattices of film and substrate. Tilt angles of film and buffer layers are always less than the substrate angle. We observed a similar difference for substrates with lower angles.

Film surfaces were examined by a scanning probe microscope (SPM). Subgrains form at the surface and they are elongated in the a-b plane in a direction perpendicular to the tilt direction. The surface profile in the direction of a tilt of 11° on a sapphire substrate tilted by 14° shows a valley depth about 11–14 nm and an average roughness R_a not exceeding 1.4 nm. These values correspond to a film thickness of 250 nm. The SPM profile in the vicinity of the bicrystal boundary (Fig. 2) shows a V-shaped surface suppression of about 40 nm depth. This boundary is more straight than one for a conventional non-tilted bicrystal film. The average sub-grain length along the boundary is 3-5 times longer than its width contrary to conventional untilted bicrystal films, in which they are equal. Our SPM data show that the roughness of a 250 nm thick YBaCuO tilted film on sapphire is 1.5 times less than the values for 100 nm thick films on tilted SrTiO₃ and NdGaO₃ substrates [4,5]. As the roughness increases with increasing thick-

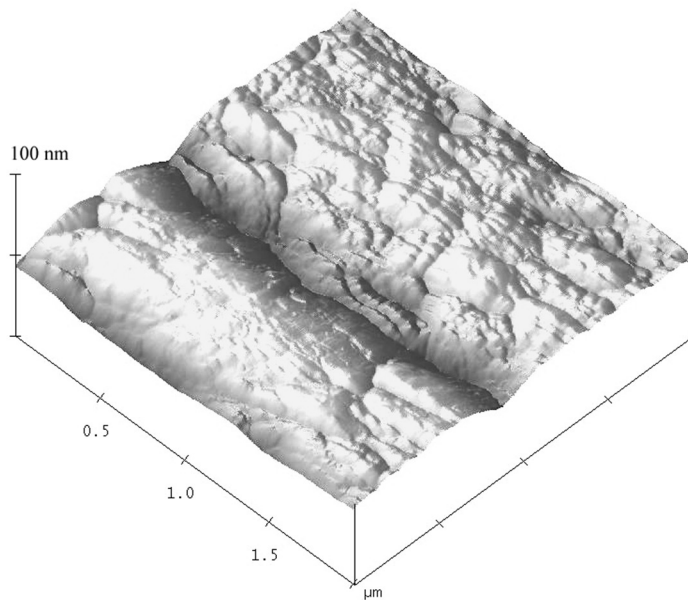


Figure 2. Three-dimensional SPM picture of an YBaCuO thin film grown on a symmetric sapphire bi-crystal substrate with a 14°-tilt of the normals to (1012) planes of both single crystalline parts in directions [2201] to the grain boundary relatively the normal to the surface.

ness, the difference in roughness should be even higher in favor of our films on sapphire, if the films were of equal thickness. Hence, a low roughness adds to the advantages of sapphire tilted substrates.

3. Josephson effects

“Valley”-type tilt grain boundary junctions, of widths from 1.5 to 6 μm , had critical current densities of about 1500 kA/cm^2 at 4.2 K and 20-40 kA/cm^2 at 77 K. $I_c R_n$ -products of up to 4.5 mV were observed at 4.2 K and up to 150 μV at 77 K.

A Josephson oscillator chip consists of five junctions integrated with planar antennas. The log-periodic antenna is designed for frequencies 100-1000 GHz. Radiation at 300 GHz gave up to 5 Shapiro steps in IV curves at $T=4.2$ K. Critical current and Shapiro steps oscillate with applied microwave power.

Josephson radiation was measured in two ways. A cryogenic bolometer [7] was used as a detector in both cases. In the first experiment it was clamped to the substrate with the Josephson junction and cooled to 260 mK. The critical current of the bicrystal junction varied from 20 to 2 μA at different values of magnetic field. The detected response (Fig. 3) consists of two components. The first one is independent of magnetic field and corresponds to infrared radiation from an overheated Josephson junction. The second component is largest for the highest critical current and can be suppressed by a magnetic field. By suppressing the critical current of a Josephson junction one can not only decrease the oscillation output power, but also reduce the frequency range of Josephson oscillations. When the critical current is suppressed below 2 μA , the response voltage is clearly proportional to the square of the JJ bias current. This means that we can separate Josephson radiation at frequencies below 1 THz and thermal radiation of overheated junction for bias voltages over 1 mV.

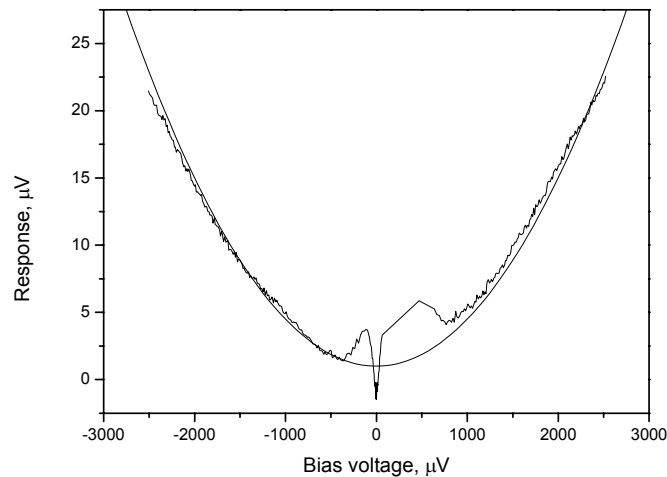


Figure 3. Detected response of the bolometer as a function of voltage in a Josephson oscillator clamped to its back side, and a fitting parabola (smooth line). The initial temperature is 260 mK.

Overheating in a Josephson junction is described by a model given in Ref. [8] for a variable thickness microbridge. It results in an equivalent electron temperature of about 3 K at 1 mV bias. One should take into account that this power is radiated and then received. It means that Planck's radiation law should be applied. For our experiment we can take the bandwidth of the double dipole antenna $\Delta f=0.3f$. For a design frequency 300 GHz and a bias range of 5 mV, the radiated power can be approximated as

$$P_{rad} = \frac{0.6}{4\pi^2} \cdot \frac{e^2 V^2}{h} \quad (1)$$

This approximation is presented in Fig. 3 as a fitting parabola.

It was necessary to place the Josephson oscillator separately from the bolometer to avoid overheating in measurements with high critical current junctions, $I_c > 0.5$ mA. This was arranged in a quasioptical configuration where the substrate with the Josephson junction was attached to a sapphire hyper-hemisphere lens at a 1.8 K stage and the bolometer with its Si lens was placed at the 260 mK stage in the cryostat with lenses facing each other. The results presented in Fig. 4 show signal maxima at voltages 0.8 mV, 1.9 mV, 2.8 mV, and 3.5 mV that correspond to the spectral dependencies of the antennas and the quasioptical beamguide. The signal maximum at 3.5 mV corresponds to frequency 1.7 THz. The maxima can be suppressed by a magnetic field, which proves that the received signal has its origin in Josephson radiation.

4. Conclusion

The tilt angle of a buffer or an YBCO film is different from the tilt angle of the crystal lattice of a tilted sapphire substrate. The difference in lattice tilt angles relative to the surface can be explained by the thermodynamic tendency to minimize the surface energy of the film by tilting it towards the surface of a low-index plane. The high value of the $I_c R_n$ -product, and its small spread, for a symmetric bicrystal junction, can be explained by a more regular boundary in a [100]-tilt grain boundary compared to a conventional [001]-

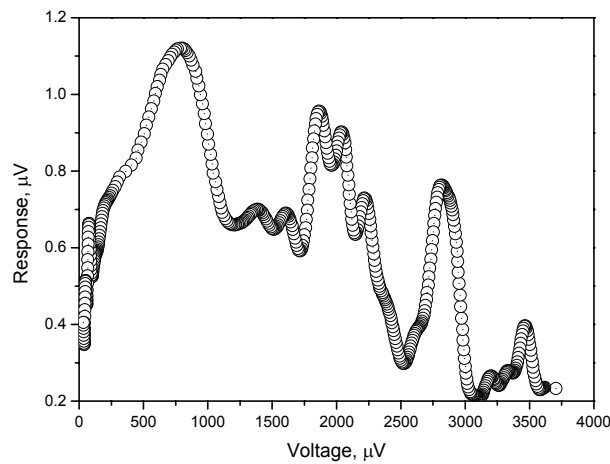


Figure 4. The bolometer response versus bias voltage of the Josephson oscillator (bi-crystal YBaCuO on $2 \times 14^\circ$ tilted sapphire). $T=1.8$ K for the oscillator, 0.28 K for the bolometer.

tilt grain boundary, at which a significant faceting exists. Direct heating of HTS Josephson junctions at bias voltages over 1 mV can be an important limiting factor for applications at terahertz frequency. The bias voltage is about 10 mV for a 5 THz signal and overheating can go up to 30 K. The actual critical current for such a bias voltage will be the same as the one measured at 30 K. This is much less than at 4 K, the $I_c R_n$ product will be reduced proportionally, and, as a result, the high-frequency response will be suppressed. To reduce such overheating, a heat sink should be used, e.g., a substrate with a high thermal conductivity or a thick-film, normal metal quasiparticle trap (gold in our case) deposited on top of YBaCuO and used as a planar antenna.

Acknowledgement

This work was supported in part by the Swedish Research Council (VR), Royal Academy of Sciences (KVA), Foundation of Strategic Research (SSF), STINT, the Swedish Institute, and by EU INTAS-01-686. TC thanks NTT-BRL for hospitality.

References

1. L. Mechin, P. Berghus, and J. Evetts, *Physica C* **302**, 102 (1998).
2. P. Czerwinka, R. Campion, K. Horbelt *et al.*, *Physica C* **324**, 96 (1999).
3. Y. Divin, U. Poppe, J. Seo, B. Kabius, K. Urban, *Physica C* **235-240**, 657 (1994).
4. U. Poppe, Y. Divin, M. Faley *et al.*, *IEEE Trans. Appl. Supercond.* **11**, 3768 (2001).
5. Y. Divin, U. Poppe, C. Jia, P. Shadrin, K. Urban, *Physica C* **372-376**, 115 (2002).
6. Y. Divin, O. Volkov, M. Liatti, and V. Gubankov, *IEEE Trans. Appl. Supercond.* **13**, 676 (2003)
7. M. Tarasov, M. Fominsky, A. Kalabukhov, and L. Kuzmin, *JETP Lett.* **76**, 507 (2002).
8. M. Tinkham, M. Octavio, and W. J. Skocpol, *J. Appl. Phys.* **48**, 1311 (1977).