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Josephson Spectroscopy of Terahertz Losses in [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ Bicrystal Junctions

Y Divin$^1$, M Lyatti$^{1,2}$

$^1$Institute of Solid State Research, Research Centre Juelich, Juelich 52425, Germany
$^2$Institute of Radio Engineering and Electronics of RAS, Moscow 125009, Russia

E-mail: Y.Divin@fz-juelich.de

Abstract. Terahertz losses in the [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ grain-boundary junctions were studied using admittance Josephson spectroscopy. The $I$-$V$ curves of the [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ junctions, when annealed in atomic oxygen, were described by the resistively shunted junction model (RSJ) with an accuracy of better than 0.5% at the temperature range, where the characteristic voltage $I_cR_n < kT/2e$. The losses were shown to increase with the decrease of the oxygen content in the junction and corresponding decrease of junction conductance $R_n^{-1}$. At low temperatures, where $I_cR_n \gg kT/2e$, the absorption of Josephson radiation by optical phonon modes in YBa$_2$Cu$_3$O$_{7-x}$ was found to be reflected in the $I$-$V$ curve of the [100]-tilt junctions. The most prominent structure is situated at the voltages $V \approx 9.5$ mV, which gives the corresponding Josephson frequency of 4.6 THz in agreement with the frequency of the strongest IR active optical phonon mode in YBa$_2$Cu$_3$O$_{7-x}$. Assignment of additional lines in the derived losses is discussed according to available data on lattice dynamic calculations and experimental data for a dynamic conduction $\sigma_{1}(f)$ of YBa$_2$Cu$_3$O$_{7-x}$. Josephson spectroscopy might be useful for study of low-energy excitations in high-$T_c$ materials.

1. Introduction
Grain boundaries (GBs) in high-temperature superconductors are of great interest because they limit the critical currents even in the case of low misorientation angles and the large-angle GBs behave like Josephson junctions. The main experimental results are obtained for artificial high-$T_c$ GB Josephson junctions, which are fabricated by epitaxial growth of high-$T_c$ thin films on a bicrystal substrate. Conventional [001]-tilt high-$T_c$ GB junctions, due to an island growth of the c-axis high-$T_c$ films, demonstrate a faceted meandering of their GB around the substrate GB, inhomogeneous current transport, large parameter spread and low values of characteristic voltages $I_cR_n$[1].

Significant improvement was reached in [100]-tilt high-$T_c$ GB junctions with a tenfold decrease of the amplitude of GB meandering and a threefold increase of the $I_cR_n$-values [2]. The current transport is more uniform in these junctions, as follows from recent study of low-frequency noise [3]. The low-frequency fluctuations of normal-state resistance $\delta R_n(t)$ and critical current $\delta I_c(t)$ were found to be completely antiphase correlated in these junctions and their normalized spectral densities $S_{\delta R_n}(f)/R_n^2$ and $S_{\delta I_c}(f)/I_c^2$ were equal, i.e. both the quasiparticle and supercurrent tunnelling in the [100]-tilt junctions take place through the same parts of the barrier [3].

To whom any correspondence should be addressed.
However, improved quality and increased $I_c R_n$-values of the [100]-tilt junction did not result in a corresponding increase of the high-frequency limit of Josephson oscillations, compared with that of the [001]-tilt junctions. The highest frequency of 5.2 THz has been demonstrated for Josephson oscillations in [100]-tilt junctions [4], but, being a record value for Josephson frequencies, it is still too low from the expectations grounded on the gap frequencies. We can expect that some losses in the junction and its environment are responsible for weakening of the Josephson oscillations.

Here, we report on spectroscopic study of losses in the [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ grain-boundary junctions, which were modified by annealing in atomic oxygen.

2. Josephson admittance spectroscopy

An elegant way to the spectral dependencies of losses in the environment of the Josephson junction was suggested by A.F. Volkov [5]. An autonomous Josephson junction, described by a resistively-shunted-junction (RSJ) model [6], demonstrates a nonlinear static $I$-$V$ curve:

$$I(V) = \left( I_c^2 + V^2/R_n^2 \right)^{1/2},$$

where $I_c$ is the critical current and $R_n$ is the normal-state resistance of the junction. The difference between this $I$-$V$ curve and the $I$-$V$ curve of the junction in a normal state $I_n(V) = V/R_n$ is due to the time averaging of Josephson oscillations $V(t)$.

When the junction is additionally shunted by external admittance $Y_e(f)$, Josephson oscillations are modified and modified static $I$-$V$ curve is situated below that of autonomous junction. In the case of small external admittances $Y_e(2eI_cR_n/h) << R_n^{-1}$, this effect is described by Volkov’s result of perturbation analysis:

$$\Delta I(V) = \frac{V}{I_c^2 + V^2} V \Re Y_e(2eV/h),$$

where $\Delta I(V)$ is a difference between the $I$-$V$ curve, perturbed by the external admittance $Y_e(f)$, and the RSJ-like autonomous $I$-$V$ curve, $Y_e(0) = 0$, $V_c = I_c R_n$ [5]. In this approach, the deviations of the $I$-$V$ curve are determined by a real part of the external admittance $\Re Y_e(2eV/h)$. So, one can derive a spectral distribution of losses in the external admittance from the modification of the experimental $I$-$V$ curve, using (2). Equation (2) is a basic equation for an admittance Josephson spectroscopy, which can be applied to any modification of junction environment. An example of modification of the $I$-$V$ curve by a hypothetical external admittance $Y_e(f)$ is shown in figure 1.

![Figure 1](image)
Figure 2. Schematic presentation of the valley-type [100]-tilt high-$T_c$ junction (above) and a corresponding TEM cross-section of one of the fabricated [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB junctions (below).

Figure 3. Microphotograph of the 1µm-wide YBa$_2$Cu$_3$O$_{7-x}$ GB junction (centre), connected with YBa$_2$Cu$_3$O$_{7-x}$ contact lines for four point electrical measurements and sinuous Pt antenna for coupling with external radiation.

3. Experimental results

3.1. Junction fabrication

In this study we used the [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB junctions, which were deposited on bicrystal substrates by dc sputtering from a YBa$_2$Cu$_3$O$_{7-x}$ target, patterned by UV lithography and Br/ethanol etching [7], [8]. A schematic presentation of the valley-type [100]-tilt high-$T_c$ junction and a corresponding TEM cross-section of one of the fabricated [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB junctions are shown in figure 2. The GB, separating two epitaxial YBa$_2$Cu$_3$O$_{7-x}$ thin films with mutually tilted c-axis’s, is clearly seen as a vertical line in the TEM image in figure 2.

A micrograph of one of the fabricated junctions in transmitted light is shown in figure 3. The deposited films were patterned in the form of a bridge, crossing the bicrystal boundary of the substrate, situated vertically in figure 3, and contact lines for four-probe electrical measurements. To minimize possible electrical contributions of the parts of the tilted films in the bridge region, the voltage contact YBa$_2$Cu$_3$O$_{7-x}$ lines (vertically situated in figure 3) were patterned at a distance of ±1.5 µm from the GB. The current YBa$_2$Cu$_3$O$_{7-x}$ lines are horizontally situated and not visible because they are covered by an antenna. The broadband sinuous antenna is fabricated by lift-off technique from a Pt thin film, sputtered on the patterned YBa$_2$Cu$_3$O$_{7-x}$ layer. In this work we used platinum for the antenna and electrical contacts because this metal coating, in a contrast to silver coating, does not oxidise even in atomic oxygen.

As-fabricated junctions were subjected to annealing in an ozone-oxygen mixture at a temperature of 140°C in the presence of ultraviolet radiation. An oxidation for half an hour was sufficient for around two-fold increase the normal-state resistance of the junctions. After following annealings for 1 and 2 hours the junction resistance decreased only by few percents. The structural barrier and oxygen in the Cu-O chain in the YBa$_2$Cu$_3$O$_{7-x}$ junction electrodes are might be considered as the most probable sites of oxygen enhancement at this low annealing temperature [9]. The $I_cR_n$-product of the junctions was not reduced even after seven subsequent annealings with total time of 13.5 hours. The junctions were kept in pure oxygen at room temperature between the measurements.

3.2. I-V curves at $2eI_cR_n/kT \leq 1$

The I-V curves of fabricated junctions were studied at the temperatures $T$ from the critical temperature $T_c \approx 90$ K to 4.2 K. At low temperatures, where $2eI_cR_n/kT >> 1$, some localized peculiarities were
observed at the I-V curves and a first derivative dI/dV vs. V. At the temperature range from the critical temperature \( T_c \approx 90 \text{ K} \) up to \( T \approx 55 \text{ K} \), where the ratio \( 2eI_cR_n/kT \leq 1 \) for our junctions, the I-V curves are smooth and even first derivative of the I-V curve does not contain sharp peculiarities.

The I-V curves of a typical [100]-tilted grain-boundary junction at the temperature of 55 K are shown in figure 4. The I-V curves of as-fabricated junctions (dashed line) had visible deviations, up to 10\%, from the I-V curves predicted by the RSJ-model with similar values of the critical current \( I_c \) and the normal-state resistance \( R_n \) (dotted line). The same junction, modified by annealing in the ozone-oxygen mixture for half an hour and measured immediately after annealing, demonstrated more than a twofold decrease in the resistance \( R_n \) from 4.75 Ohm to 2.25 Ohm, while the \( I_cR_n \)-value increased less than 1\% (solid line). The I-V curve of the oxygen-annealed junction with an accuracy of a 0.4\% was close to that of the RSJ-model. The I-V curves of the junctions were even more close to those of the RSJ model, when the junction temperatures were increased from 55 K to the critical temperature \( T_c \) of 90 K.

The observed proximity of the I-V curves of oxygen-annealed junctions to those of the RSJ model might be considered as an indication that an “ideal” Josephson junction has the RSJ-like I-V curve and observed deviations from ideality is due to shunting of Josephson oscillations by some admittance, modified by oxygen treatment of the junction. With this approach in mind, we derived two spectral dependences of \( \text{Re}Y_e \) applying equation (2) to the difference between the experimental I-V curves (figure 4a) and corresponding RSJ-like ones. The results are presented in figure 4b for an as-fabricated junction (dashed line) and oxygen-annealed junction (solid line). The \( \text{Re}Y_e(f) \)-dependence for as-fabricated junction is more broadband and has the values more than one order higher than those of oxygen-anealed junction. The maximum values of \( \text{Re}Y_e \), decreased from 7.6\( \times \)10\(^{-2} \) \( \Omega^{-1} \) for as-fabricated junction to 7.7\( \times \)10\(^{-3} \) \( \Omega^{-1} \) for oxygen-annealed junction.

Modification of low-frequency noise in the same junction before and after oxygen-annealing is shown in figure 5, where experimental spectral densities \( S \) vs. \( V \) together with nonlinear fit data [3] for as-fabricated (circles and dashed line) are 3-5 times larger than corresponding data for oxygen-annealed (squares and solid line) junction.

Modification of losses, junction resistance and 1/f voltage noise with low-temperature annealing in atomic oxygen might be understood in a model where the junction barrier is formed by a band bending [10]. Actually, the junction barrier is considered to consist of three layers: the structurally distorted interface in the middle and two adjacent charge-depleted layers of undistorted material [11]. Oxygen annealing results in a reduced amount of vacancies in the Cu-O chains, a decreased mobility of oxygen, an increased doping and a corresponding decrease of the length of depletion layers.

\[ S_f(\text{Hz}) \]

\[ S_f(\text{Ohm}^{-1}) \]

\[ S_f(\text{V}^{-2}) \]

Figure 4. The experimental I-V curves (a) of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) GB junction before (dashed) and after (solid) low-temperature annealing in oxygen and corresponding \( \text{Re}Y_e(f) \) dependencies (b), derived from data in figure 4a by using equation (2). The RSJ-like I-V curves in figure 4a are shown as dotted lines. \( T = 55 \text{ K} \).

Figure 5. Spectral noise densities \( S \), at 3 kHz vs. junction voltage for as-fabricated (circles) and oxygen-annealed (squares) junction.
3.3. I-V curves at $2eI_cR_n/kT \gg 1$

The I-V curves of fabricated junctions at low temperatures, where $2eI_cR_n/kT \gg 1$, demonstrated enhanced deviations from that of RSJ model with some localized peculiarities. Reproducible fine structures are observed on the differential conductance $dI/dV$ vs. voltage $V$ for the [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB junctions at the voltages $V \approx 7.3$ mV, 9.5 mV and 11.8 mV (figure 6, below). The visibility of these structures increases for low-resistance junctions. The most prominent feature is situated around voltages $V \approx 9.5$ mV. We might suspect that this feature is related to an interaction of tunneling carriers with some strong mode in the barrier or YBa$_2$Cu$_3$O$_{7-x}$ itself.

To clarify the mechanism of interaction, we performed photon-assisted tunneling experiments and illuminate the junction with 96GHz radiation at various levels of intensities. The modification of the conductance curves are shown in figure 7. The conductance curve at large attenuation (60 dB) of 94GHz source has a peak at the voltage of 9.6 mV. With an increased level of 96 GHz intensity, the peak at 9.6 mV goes to the background level (10 dB) and then increases again (6 dB). Several satellite peaks appear at the voltage differences of integral numbers of ±0.2 mV from the position of the main peak and also oscillate with increased power level. The difference in peak positions is corresponding to $hf/2e$ and this circumstance is a signature of photon-assisted Josephson tunneling.

Now we can compare our data $dI/dV$ vs. voltage $V$ with optical data for YBa$_2$Cu$_3$O$_{7-x}$, which are always the functions of frequency. In figure 6 (above) we presented the frequency dependence of phonon contribution to the dynamic conductance of $\sigma_{1c}(f)$ of YBa$_2$Cu$_3$O$_{7-x}$, which was calculated from infrared reflectance data of Genzel [12]. As one can see from a comparison of $\sigma_{1c}(f)$ and $dI/dV$ vs. $V$, the main peak at 152 cm$^{-1}$ corresponds to the pronounced odd-symmetric structure at $V = 9.5$ mV, and the lower peak at 191 cm$^{-1}$ corresponds to one of the features close to $V = 11.8$ mV. Possible assignments of ion displacements to the frequencies of phonon modes, according to [13], are presented as insets in upper part of figure 6.

![Figure 6](image1.png)

**Figure 6.** Differential conductance $dI/dV$ vs. voltage for 1µm- and 2µm-wide [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB junctions with the $I_cR_n$-products of 6.5 mV at 5 K (below) and phonon contributions to the dynamic conductance $\sigma_{1c}(f)$ of YBa$_2$Cu$_3$O$_{7-x}$. Insets show atomic displacements in YBa$_2$Cu$_3$O$_{7-x}$ for phonon modes at 152 cm$^{-1}$ (left) and 191 cm$^{-1}$ (right) [13].

![Figure 7](image2.png)

**Figure 7.** Effect of electromagnetic radiation with the frequency $f = 96$ GHz and various levels of power on the conductance $dI/dV$ vs. voltage $V$ near a peak at 9.6 mV. Satellite peaks appear at the voltage differences in integral numbers of $hf/2e = 0.2$ mV and oscillate with power level. Each curve is shifted on 0.2 Ohm$^{-1}$ down for attenuations 14, 10 and 6 dB.
Figure 8. The experimental $dI/dV$ vs. $V$ curves of 1µm-wide [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB junction with $I_c R_n = 6$ mV at $T = 5$ K before (grey) and after (black) low-temperature annealing in atomic oxygen.

Oxygen annealing of the [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ junctions results in an interesting modification of localized features on the differential conductance dependencies $dI/dV$ vs. $V$ at the voltages below 10 mV (figure 8). While amplitude of the main feature near $V \approx 9.5$ mV ($f \approx 153$ cm$^{-1}$) increases, the features at $V \approx 7.3$ mV ($f \approx 118$ cm$^{-1}$) and $V \approx 5.5$ mV ($f \approx 87$ cm$^{-1}$) becomes less visible after annealing. Low-temperature annealing in atomic oxygen, which we use, might increase only the oxygen concentration in CuO chains [9]. The low-frequency phonon modes, where these oxygen atoms (O(1)) are involved, are mainly in-plane IR modes according to lattice dynamic calculations [13]. Due to these circumstances, we can ascribe the features at $V = 5.5$ and 7.3 mV on the $dI/dV$ vs. $V$ to an interaction of Josephson oscillations with in-plane phonon modes.

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