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Effects of low-temperature ozone annealing on current-voltage and low-frequency noise characteristics of [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ grain-boundary Josephson junctions.

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Abstract. The dc and noise characteristics of YBa$_2$Cu$_3$O$_{7-x}$ [100]-tilt bicrystal Josephson junctions have been studied before and after annealing in ozone. Junctions with a misorientation angle 2x14° have been characterized in the temperature range from 7 K to 75 K, and their $I_R^2$-product values were equal up to 5.4 mV at 7 K. Ozone annealing of the junctions at 140°C results in a two-fold decrease of the normal-state resistances $R_n$. The values of the $I_R^2$-product have been increased not more than 7% after annealing. The spectral densities $S_V(f)$ of the voltage fluctuations measured at various currents were of $1/f^\alpha$ -type with $\alpha = 0.8$-1.0. The normalized spectral densities $S(f)=S_V(f)/R_n^2$ and $S(f)=S_Ic(f)/I_c^2$ of normal-state resistance fluctuations $\delta R_n$ and critical current fluctuations $\delta I_c$, respectively, plus a coefficient $k$ of the correlation between $\delta R_n$ and $\delta I_c$ fluctuations have been derived from the measured spectra $S(f)$ before and after annealing. The modification of transport and noise characteristics were consistent with a junction model of the [100]-tilt junction, where the supercurrent and quasiparticle current flow through the same junction area, the annealing in ozone results in a decrease of the barrier thickness and the barrier is free from localized states. We suggest that the low-frequency noise arises at the interface between superconductor and the barrier.

1. Introduction.

At present time, due to a simple fabrication and good repeatability of the parameters, high-temperature grain-boundary Josephson junctions (GB JJ) are widely used both for fundamental and applied purposes. But, despite numerous studies, the GB JJ barrier origin is still unclear. There are various models of current transport in high-$T_c$ GB JJ [1,2,3,4], but the current transport study alone cannot distinguish between these models. The low-frequency noise supposed to be originated from fluctuations of the barrier thickness, which cause critical current fluctuations $\delta I_c$ and normal-state resistance fluctuations $\delta R_n$ [5]. Hence, the low-frequency noise properties can be used as a valuable tool in clarifying the validity of different GB JJ barrier models. Most experimental results have been obtained for [001]-tilt high-$T_c$ bicrystal GB JJ. Due to the island growth of the c-axis high-$T_c$ thin films, a real grain boundary in a [001]-tilt bicrystal junction demonstrates a complicated microstructure with a meandering around the substrate bicrystal boundary and nanofacets with various local misorientations. This complex spatially inhomogeneous microstructure of the [001]-tilt GB JJ complicates the interpretation of the low-frequency noise data. Recently, [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB JJ have been fabricated with an order of magnitude less meandering, better current homogeneity and a
three-fold increase of the $I_R$-product values, compared with the conventional [001]-tilt GB JJ [6]. In this paper we report on our study of current transport and low-frequency noise characteristics of [100]-tilt GB JJ before and after modification of oxygen concentration in the grain boundary region.

2. Experimental results and discussion.

Four [100]-tilt YBa$_2$Cu$_3$O$_{7-x}$ GB JJ on two NdGaO$_3$ bicrystal substrates with a misorientation angle of 2x14° have been characterized before and after annealing in an ozone-oxygen (O$_3$-O$_2$) atmosphere mixture. Each junction has been successively annealed for 1/2 hour, 1 hour and 2 hours in the ozone-oxygen atmosphere at a temperature of 140 °C. In a separate study, an annealing at this temperature was found to result in a modification of the grain-boundary electrical properties, but not the oxygen content inside the grains. The junctions were heated in a glass chamber at an ozone-oxygen pressure slightly above 1 bar in the presence of UV radiation. The ozone-oxygen mixture has been produced by a commercial ozone generator with an output ozone concentration of 90 g/m$^3$.

The film thickness of GB JJ was 0.07 μm and the junction widths were in the range of 1-3 μm. Details of fabrication have been reported elsewhere [7]. The junctions were current biased. To provide noise measurements a low-noise preamplifier with a liquid-nitrogen-cooled first stage with input voltage noise of 230 pV/Hz$^{1/2}$ and small signal bandwidth of 150 kHz has been used. The low-frequency spectral analysis was performed by a SR760 spectrum analyzer in the frequency range of 32 Hz-25.6 kHz. More details of the experimental setup have been reported earlier [8].

Following the ideas suggested in paper [3], the low-frequency noise parameters of the GB JJ have been calculated using a revised equation for measured voltage noise spectral density $S_v$:

$$S_v(f) = (V-R_dI)^2 S_r(f) + V^2 S_i(f) + 2k(f)(V-R_dI)V[S_r(f)S_i(f)]^{1/2},$$

where $R_d = dV/dI$ is the differential resistance, $S_r(f) = S_r(f)/R_n^2$ and $S_i(f) = S_i(f)/I_c^2$ are the normalized spectral densities of the fluctuations $\delta R_n$ and $\delta I_c$, correspondingly. $k(f) = |\gamma_{ir}(f)| \cos \theta_{ir}(f)$ is a correlation coefficient between the fluctuations $\delta R_n$ and $\delta I_c$. $\theta_{ir}$ is a phase angle of the cross-spectral density $S_{ir}$, $\gamma_{ir}^2(f) = |S_{ir}(f)|^2/S_r(f)S_i(f)$.

The as-fabricated junctions had the values of the $I_R$-product up to 5.4 mV at a temperature of 7.5 K and a normal-state resistance in the range of 4.7-9.6 Ω. As the noise analysis is based on the RSJ model, the current transport and low-frequency noise characteristic were studied in the interval of 55-75K, because at lower temperatures, the $I$-$V$ curves showed deviations from this model [7].

The typical effect of ozone-UV annealing on the $I$-$V$ curve of [100]-tilt GB JJ is shown in figure 1a. The critical current of the junction at a temperature of 55K increased from 0.5 mA to 1.1 mA after ozone-UV annealing, while the normal-state resistance decreased from 4.7 to 2.3 for a net

![Figure 1](image-url)
increase in the $I_R$-product value from 2.35 mV to 2.5 mV. The first ozone-UV annealing for a half an hour resulted in 2.0±0.2 times decrease in normal state resistance of the junctions independently on their widths. The $I_R$-product value of the junctions was increased for 4±7 % after the first annealing. The following annealings for one hour and two hours resulted in an only insignificant decrease of the junction resistances compared with the first modification. The $I_R$-product value of the junctions remained the same as after the first annealing. The modification of the current-voltage characteristics was likely due to the decrease of the junction barrier thickness, because the decrease of the junction resistance was independent of junction width. The current-voltage characteristics were in good agreement with RSJ model including thermal noise rounding without any excess current both for as-fabricated and for ozone-UV annealed junctions in the temperature range of 55-75K.

The voltage noise spectra have been measured at various current biases $I_b$ in the temperature range of 55-75K. The voltage noise spectra of as-fabricated junctions showed only small deviations from $1/f^\alpha$ dependence with $\alpha = 0.8-1.0$. As it was reported, these deviations are believed to be associated with lorentzian-like contribution of individual fluctuators [2]. The ozone-UV annealing resulted in the decrease of these deviations and the voltage noise spectra of ozone-annealed junctions were close to the featureless spectra of $1/f^\alpha$ type as it shown in figure 1b.

To calculate the values of normalized spectral densities of the fluctuations $\delta R$, and $\delta I$, and the correlation coefficient $k$, the experimental data were fitted by equation (1). Both as-deposited and ozone-annealed junction demonstrated the good agreement with equation (1). The dependencies of voltage noise spectral density at a frequency of 3.2 kHz on the square of voltage bias for as-fabricated and ozone-UV annealed junctions are shown in figure 2a.

Following the first ozone-UV annealing for a half an hour, the amplitude of low-frequency noise was reduced as one can see in figure 2a. Both the $S_v$ and $S_r$ values decreased after the first annealing. But, the $S_v/S_r$ ratio remained approximately the same. The following two ozone-UV annealings for one hour and two hours did not result in the further decrease of $S_v$ and $S_r$ values. The transport and low-frequency noise characteristics of the junctions were measured in two different ways. The characteristics of two junctions were studied immediately after annealing. The other two junctions were placed in the pure (5N) oxygen atmosphere at room temperature for 12 hours after annealing before their characteristics were studied. For the twojunctions studied immediately after ozone-UV annealing the decrease of $S_v$ value was 2.8 and 3.2 times correspondingly. For the two junctions studied after 12 hours after ozone-UV annealing the decrease of $S_v$ value was only 1.4 times for both.

As the resistance reduction was the same for all junctions, the difference in $S_v$ modification for

Figure 2. The spectral density of the voltage noise of [100]-tilt junction at $f = 3.2$ kHz as a function of square of voltage bias at the temperature of 55K before (square) and after (circle) ozone-UV annealing (a). The spectral density of the voltage noise of ozone-UV annealed [100]-tilt junction at $f = 3.2$ kHz as a function of square of voltage at the temperature range of 55-75K (b). The solid lines represent a least-square fit based on equation (1).
Junctions, studied immediately after annealing, and for junctions, studied 12 hours after annealing, can be explained by an increase of the fluctuator concentration [9] in the junctions, studied 12 hours after annealing, caused by oxygen concentration relaxation in the junction region. Thus, the fluctuators can be probably associated with oxygen vacancies. The concentration of the fluctuators, responsible for the low-frequency noise, was reduced only after the first annealing, while the following annealing did not result in further decrease of the $S_I$ value. Such dependence of fluctuator concentration on annealing duration can be explained, if we suppose that these fluctuators are located at insulator-superconductor (I-S) interface.

For the ozone-UV annealed junction with the highest $I_R\cdot R$-product value of 2.5 mV at $T= 55$ K the $(S/S_I)^{1/2}$ ratio was found to be equal to 1.0±0.1 and the correlation coefficient $k$ to be equal to -1.1±0.1. Thus, the direct tunneling mechanism dominates both the quasiparticle current transport and the supercurrent transport in this junction. Complete anti-phase correlation of the critical-current and normal-resistance fluctuations corresponds to the uniform distribution of the current across the junction. Due to the high $I_R$ value, the influence of correlation term $2k(V-I_R)\sqrt{S(f)S(f)}^{1/2}$ on the magnitude of voltage noise spectral density is clearly seen in figure 2b. One can see, that the spectral density curves at different temperatures do not fall into single line at high current biases, but go parallel to each other. The reason is an anti-phase correlation between fluctuations of $\delta I_c$ and $\delta R_n$. As values of $S(f)$ and $S(f)$ were shown to be independent of temperature and voltage across the junction [2], the correlation term in equation (1), according to the RSJ model, is independent on voltage bias and proportional to the square of $I_R$-value: $2k(V-I_R)\sqrt{S(f)S(f)}^{1/2}=-2k(V-I_R)\sqrt{S(f)S(f)}^{1/2}$.

The $(S/S_I)^{1/2}$ ratio value was found in the range of 1.0-1.5 for other studied junctions. Thus, the transport and low-frequency noise characteristics of the studied [100]-tilt junctions are consistent with the S-I-S Josephson junction model with an insulating barrier without any localized states. The transport and noise characteristics of studied [100]-tilt junctions are in contradiction with the predictions of the ISJ model [2] and the filament model [3]. The assumption, that oxygen vacancies, located at I-S interface, are responsible for low-frequency noise, is in agreement with the JJ model with a barrier composed by a double Schottky barrier [1]. In the frame of this model the low-frequency noise can be caused by fluctuations of the build-in charge at the I-S interface.

3. Conclusions.
The current transport and low-frequency noise of [100]-tilt GB JJ with high $I_R\cdot R$-values have been studied before and after ozone-UV annealing. The junction resistances were decreased by two times after annealing, while the $I_R\cdot R$-values have been enhanced only by 4-7%. The ozone-UV annealing resulted in a decrease of the fluctuator concentration by 2.8-3.2 times. The results of successive ozone-UV annealings demonstrated that these fluctuators are likely associated with the oxygen vacancies at the superconductor-isolator interface. The studied current transport and noise characteristics are consistent with a double Schottky barrier junction model.

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