Spin-triplet Superconducting Current in Metal-oxide Heterostructures with Composite Ferromagnetic Interlayer

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Abstract— Superconducting heterostructures fabricated from oxide superconductor YBa$_2$Cu$_3$O$_{7-\delta}$ and a composite ferromagnet La$_{0.7}$Sr$_{0.3}$MnO$_3$/SrRuO$_3$ interlayer and Au/Nb counter electrode were studied experimentally. Superconducting current was observed at magnetic field $H$ raised up to 2000 Oe which is greater than saturation magnetic field of manganite La$_{0.7}$Sr$_{0.3}$MnO$_3$ (of order 100 Oe) and greater by a few orders than the value of magnetic field corresponding to penetration of one magnetic flux quantum. Microwave measurements of integer and half-integer Shapiro steps in conditions when relatively low external magnetic field $H=30$ Oe was applied showed that the second harmonic in the current-phase relation of superconducting current becomes as big as the first harmonic. Fourier analysis of $I_s(H)$ dependencies allows extracting the components of fractional periods in $I_s(H)$ function that also confirms a deviation from the sinusoidal current-phase relation. The obtained experimental data are explained by theoretical models which predict a huge enhancement of the second harmonic of the spin-triplet component in the superconducting current. The current-phase relation could be controlled by an external magnetic field, changing the directions of magnetization in the composite bilayer ferromagnet, inserted between two spin-singlet superconductors.

Index Terms— superconducting heterostructure, composite ferromagnet, long-range proximity effect, spin-triplet pairing, current-phase relations.

I. INTRODUCTION

O VER last decade studies of controlled manipulation of spin-triplet superconducting currents attract growing interest for their possible applications in spintronics and quantum computing [1]–[3]. However, an important question about the superconducting current-phase relation (CPR) in experimental Josephson junctions with spin-triplet pairing still remains unanswered. A distinctive feature of spin-triplet superconducting correlations is their insensitivity to the magnetic exchange field $E_{EX}$ in superconducting proximity effect junctions [4]–[6]. Theory [7], [8] showed that the second harmonic of spin-triplet superconducting current dominates in $S/F_1/F_2/S$ Josephson junctions, where $S$ is a superconductor with spin-singlet pairing, and $F_1/F_2$ is a ferromagnetic bilayer with non-collinear magnetizations. Here we present experimental results on studies of spin-triplet superconducting current in $S/F_1/F_2/S$ Josephson junctions in order to examine the CPR using measurements at microwave frequencies. In order to insure high enough transparency of $S/F_1$ and $F_1/F_2$ interfaces we used metal-oxide superconductor for $S_1$ and oxide ferromagnetic materials for ferromagnetic $F_1/F_2$ bilayer which well match each other by their crystal structure and could be fabricated using epitaxial thin film growth.

II. EXPERIMENTAL SAMPLES AND MEASUREMENTS

Hybrid superconducting heterostructures with a composite magnetic interlayer have been fabricated utilizing epitaxial growth of oxide YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) superconducting films on (110) NdGaO$_3$ or (001) LaAlO$_3$ substrates and in-situ laser ablation of ferromagnetic interlayers consisting of SrRuO$_3$ (SRO) and La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) thin epitaxial films. The thicknesses of oxide films, particularly of ferromagnetic films $d_{SRO}$ and $d_{LSMO}$, were controlled by the number of pulses of an excimer Kr-laser with 248 nm wavelength. The top of the multilayer surface was covered by a 20 nm in-situ Au film. An ex-situ pre-sputtering of additional gold preceded the RF magnetron sputtering of the top Nb electrode followed by another 20 nm thick contacting gold layer.
The magnetization vector of the LSMO film lies in the plane of the substrate, whereas the magnetization vector of the SRO film was directed at an angle of about 23° from the normal to the plane of the substrate. Note, in order to turn the vector of the SRO magnetization and make it collinear to the direction of LSMO magnetization, one need to apply an in-plane magnetic field of order 1 T.

Samples were patterned using photolithography, ion-beam etching and lift-off which allows to obtain 5 heterostructures on chip with in-plane sizes varied from $L=10\ \mu m$ to $50\ \mu m$. Normal state resistance was $R_N=8\ \Omega - 300\ \Omega$.

We observed a superconducting critical current $I_C$ for most of investigated heterostructures with a total thickness $d_{LSMO}=15\ \text{nm}$, $d_{SRO}=5.6\ \text{nm}$, $L=50\ \mu m$, $I_{C_R}=0.22\ \mu V$.

In order to reveal the second harmonic weight in CPR, measurements of Shapiro steps were performed at negative voltage biasing. Sample parameters: $d_{SRO}=5.6\ \text{nm}$, $d_{LSMO}=15\ \text{nm}$, $L=50\ \mu m$, $I_{C_R}=0.22\ \mu V$.

At about 500 times higher frequency, we measured a set of $I-V$ characteristics registered at $f_e=80\ \text{MHz}$ for a heterostructure with parameters $I_C=27.5\ \mu A$, $R_N=8\ \text{mO}$. The external RF signal was weakly coupled through air to the junction. In spite of large impedance mismatch, occasional high-Q resonant coupling leads to better transmission from the open-ended coaxial output to the wiring leads. When using resonant coupling, additional print-on filters at desired frequency bands will reduce the impact of external noise. However, although no filters were used the Shapiro steps are seen very well in Fig.1.

At about 500 times higher frequency, we measured a set of $I-V$ characteristics registered at $f_e=40\ \text{GHz}$, increasing microwave powers upwards. Arrows point on positions of critical current $I_C$ and Shapiro steps $I_n$ with integer $n=\pm 1$, and half-integer $n=\pm 1/2$. $T=4.2\ \text{K}, H=0$.

### III. RESULTS AND DISCUSSION

Obtained heterostructures had a relatively low Josephson critical frequency $f_C=(2e/h)I_C R_N$ and low normal state resistance $R_N$. Note, it is a common disadvantage of Josephson junctions with a magnetic barrier [12]. At the same time as other types of Josephson junctions these heterostructures should be easily manipulatated by a weak external microwave signal. In order to answer the question how our heterostructures behave at high frequencies $f_e \gg f_C$ we performed measurements of $I-V$ characteristics in a wide range of applied microwave signals.

Fig. 1 shows Shapiro steps in the $I-V$ characteristics registered at $f_e=80\ \text{MHz}$ for a heterostructure with parameters $I_C=27.5\ \mu A$, $R_N=8\ \text{mO}$, $T=4.2\ \text{K}$. The external RF signal was weakly coupled through air to the junction. In spite of large impedance mismatch, occasional high-Q resonant coupling leads to better transmission from the open-ended coaxial output to the wiring leads. When using resonant coupling, additional print-on filters at desired frequency bands will reduce the impact of external noise. However, although no filters were used the Shapiro steps are seen very well in Fig.1.

At about 500 times higher frequency, we measured a set of $I-V$ characteristics under external microwave signal at $f_e=40\ \text{GHz}$, Fig. 2 shows $n=\pm 1$ Shapiro steps, $I_n$, and half-integer steps $n=\pm 1/2$ as well. Parameters of the heterostructure were $L=10\ \mu m$, $I_C=88\ \mu A$, and $R_N=0.16\ \Omega$. The maximum of the first Shapiro step was $I_1=94\ \mu A$ and, correspondingly, the ratio $I_1/I_C=1.1$ is in well agreement with the resistively shunted junction (RSJ) model.

In order to evaluate the ratio $q=I_{C_2}/I_{C_1}$ of second harmonic amplitude of critical current $I_{C_2}$ to the amplitude of the first harmonic $I_{C_1}$ we used numerical approximation of the dependences of the experimental critical current amplitudes $I_{C_1}$ and Shapiro steps, utilizing the approach in Ref. [13] which takes into account impact of both the junction capacitance $C$ and $q$. At high frequency limit $\omega= f_e/ f_C \gg 1$ and applied microwave currents $I_{IMW} > I_C$ the half-integer Shapiro steps

![Fig. 1: I-V curve under external RF signal at frequency $f_e=80\ \text{MHz}$ Arrows point on positions of critical current $I_C$ and Shapiro steps $I_n$ for $n=1, 2, 3$ at negative voltage biasing. Sample parameters: $d_{SRO}=5.6\ \text{nm}$, $d_{LSMO}=15\ \text{nm}$, $L=50\ \mu m$, $I_{C_R}=0.22\ \mu V$.](image1)

![Fig. 2: I-V characteristics under external microwave signal at $f_e=40\ \text{GHz}$, increasing microwave powers upwards. Arrows point on positions of critical current $I_C$ and Shapiro steps $I_1$ with integer $n=\pm 1$, and half-integer $n=\pm 1/2$. $T=4.2\ \text{K}, H=0$.](image2)
may appear [14] if McCumber parameter $\beta_c = 2 \pi \kappa R_0 C \sim 1$.

For the heterostructure shown in Fig. 2 parameters were $\omega = 5.9$, and $\beta_c < 0.02$ estimated taking the barrier capacitance $C < 1 \text{pF}$, clearly points on negligible influence of capacitance $C$ on CPR. Note, because of moderate barrier transparencies of our heterostructures, especially between the LSMO and Au/Nb, we rule out mechanism of appearance of the second harmonic, predicted for point-like contacts due to multiple Andreev reflection process [15].

In order to compare the behavior of the first heterostructure (see Fig. 1) at higher frequencies we measured it at $f_r = 3 \text{GHz}$. Fig. 3 shows dependencies of $I_C$ and $I_1$ vs. normalized microwave current $i_{\text{MW}}/\omega$, where $i_{\text{MW}}=i_{\text{MW}}/I_c$. As we mentioned earlier at this level of magnetic field $\mu$-metal shield was removed and measurements were done in less precise conditions, shown by error bars in Fig. 3. These measurements were performed under applied magnetic field $H=133 \text{Oe}$, which is much larger than $I_C(H)$ oscillation period $\Delta H = 1 \text{Oe}$, resulting just in 2.1 times reduction of critical current $I_c$. Note, in a usual Josephson junction, characterized by Fraunhofer $I_C(H)$ dependence, the number of penetrating flux quanta increase with $H$ resulting in a decrease of the $I_c$ peaks proportional to $1/H$. Another feature caused by influence of magnetic field was the enhancement of maximal amplitude $n=1$ Shapiro step, which became very close to the theoretical maximum $I_{C1}/I_c = 1.16$, predicted by RSJ-model. Taking into account results of theory [8] the both, very weak reduction of $I_C$ at large levels of magnetic field, which influenced the magnetization of LSMO, and the enhancement of Shapiro step amplitudes $I_1$ could be explained by contribution of higher harmonics of CPR.

Fig. 4 shows that critical current was observed at $H$-fields, up to $2 \text{KOE}$. The data in Fig. 4 have been obtained changing magnetic field by the steps expanding over a few $\Delta H$ periods of $I_C(H)$ oscillation. $I_C(H)$ dependence was different from Fraunhofer pattern, and oscillates with a period $\Delta H=6 \text{Oe}$. Note, the width $L=10 \mu\text{m}$ is smaller than $\lambda_r=51 \mu\text{m}$, the Josephson penetration depth of magnetic field. An other heterostructure with parameters $d_{\text{SRO}}=8.5 \text{nm}$, $d_{\text{LSMO}}=6 \text{nm}$, $L=20 \mu\text{m}$, and $H=6.5 \text{Oe}$.
heterostructures #1, #2, #3, correspondingly. Fig 5 shows decrease of $\Delta H_{HF_Y}$ with 1/L. Heterostructures #1 and #3 show linear decrease, while $\Delta H_{HF_Y}$ for #2 are shifted toward larger widths. Deviation from sinusoidal CPR could be also observed in Josephson junction networks [17]. Taking into account almost the same $R, L^2$ products (13 $\mu$m)$^2$, 0.11, and 0.13) for #1, #2, and #3, correspondingly, this explanation hardly could be applied. However, taking into account differences of experimental $\Delta L_0(H)$ from regular Fraunhofer diffraction pattern we omit making conclusions about the impact 3rd and 4th harmonics on the CPR of our S1/F1/F2/S2 heterostructures.

It should be noted that occurrence of the second harmonic in the CPR in a Josephson junction made from -wave component of the superconducting order parameter. Our direction of YBCO, the critical current is determined by the exchange field. The Josephson effect length of superconducting correlations in ferromagnetic layers, magnetizations in the layers. It has been shown that the total superconducting current in hybrid heterostructures with a composite oxide heterostructures with noncollinear magnetization, "Evidence for spin-triplet superconducting correlations in oxide Heterostructures with a Composite Ferromagnetic Interlayer", JETP Letters, vol. 97, pp. 145-148, 2013.


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IV. CONCLUSION

We have experimentally observed the superconducting current in hybrid heterostructures with a composite oxide ferromagnetic bilayer with non-collinear directions of magnetizations in the layers. It has been shown that the total thickness of the magnetic interlayer is much larger than the length of superconducting correlations in ferromagnetic layers, determined by the exchange field. The Josephson effect observed in these heterostructures is explained by penetration of the long-range triplet component of the superconducting order parameter into the magnetic interlayer. The magnetic field dependence of the current-phase relation also could be explained by the spin-triplet component of the superconducting current. At relatively low magnetic fields, $H=30$ Oe, the second harmonic component was estimated to be larger than 50% of the first one. Experimental results are based on measurements of Shapiro step amplitudes at microwave frequencies, as well on Fourier analysis of magnetic field dependencies of critical current amplitudes.

Fig. 5. Periods $\Delta H_{HF_Y}$ of the Fourier components of $L_0$($H$) dependencies vs. parameter $1/L$ for 3 heterostructures on a chip ($d_{SRO}$ = 8.5 nm, $d_{LSMO}$ = 6 nm) with widths $L = 10 \mu$m, 20 $\mu$m, 40 $\mu$m, having $\lambda_p$ = 120 $\mu$m, 110 $\mu$m, and 190 $\mu$m, correspondingly. Harmonic numbers point on linear $\Delta H_{HF_Y}(1/L)$ functions.

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