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

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***Abstract*— Superconducting heterostructures fabricated from oxide superconductor $YBa_2Cu_2O_{7-\delta}$ 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 $I_C(H)$ dependencies allows extracting the components of fractional periods in $I_C(H)$ 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

OVER 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}>> k_BT (*T* is ambient temperature, k_B is the Boltzmann constant) allowing penetration into a ferromagnet over much longer distances than the characteristic length $\xi_F \sim (1/E_{EX})^{1/2}$. Instead, the characteristic length $\xi_N \sim (1/k_BT)^{1/2}$ becomes applicable for spin-triplet superconducting correlations as in the case of superconducting proximity effect in S/N junctions, where N is a normal nonmagnetic metal.

Experimentally spin-triplet superconducting current was detected in several types of weak linked superconductors coupled by a ferromagnet characterized by spatially non-uniform magnetization [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 $S_1/F_1/F_2/S_2$ Josephson junctions in order to examine the CPR using measurements at microwave frequencies. In order to insure high enough transparency of S_1/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 $YBa_2Cu_3O_{7-x}$ (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.

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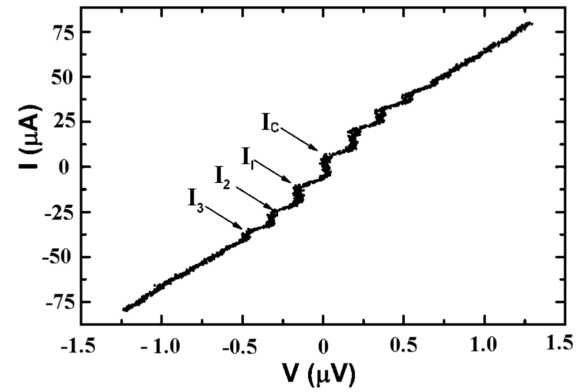

Fig. 1. *I-V* curve under external RF signal at frequency f_e=80 MHz Arrows point on positions of critical current I_C and Shapiro steps I_n for n= 1, 2, 3 at negative voltage biasing. Sample parameters: d_{SRO}=5.6 nm, d_{LSMO}=15 nm, L=50 µm, I_CR_N=0.22 µV.

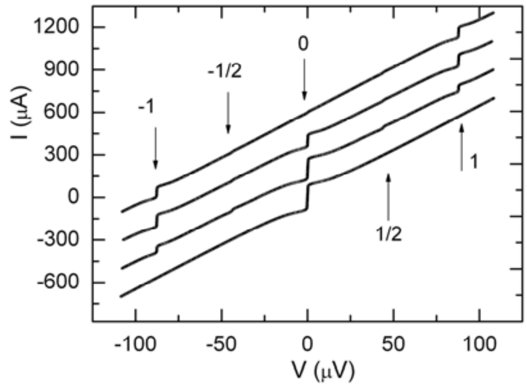

Fig. 2. *I-V* characteristics under external microwave signal at f_e=40 GHz, increasing microwave powers upwards. Arrows point on positions of critical current I_C (n=0) and Shapiro steps I_n with integer n= $\pm$1, and half-integer n=$\pm$1/2. T=4.2 K, H=0.

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 µm to 50 µm. Normal state resistance was R_N =8 mΩ – 300 mΩ. We observed a superconducting critical current I_C for most of investigated heterostructures with a total thickness d_{SRO}+d_{LSMO} of the composite interlayer up to 50 nm. For comparison, we prepared structures with only one LSMO or SRO ferromagnetic interlayer. These structures had no superconducting current if the thickness of the ferromagnetic film exceeds 5 nm. For smaller thicknesses of ferromagnetic film some of samples featured characteristics which could be attributed to pinholes.

In order to reveal the second harmonic weight in CPR, measurements of Shapiro steps were used as described in [9]. Recently a deviation of the CPR from sinusoidal, obtained by measurements of critical current dependencies from magnetic field, was reported in [10]. Measurements discussed in this paper were performed at T=4.2 K using both microwave signals and application an external magnetic field. Spin-triplet nature of superconducting correlations of our heterostructures was recently proved by studies [9], [11]. For measurements in the mm wave frequency band, f_e = 37 GHz – 78 GHz, we used frequency-tunable backward wave oscillators with rectangular waveguide output. At lower frequencies, for f_e <3 GHz, the probe signal was applied via coaxial cable. Magnetic field dependences in relatively weak fields, H<100 Oe, were measured using sample screening by a multi-turn µ-metal foil. At stronger fields, magnetic shield was removed, that resulted in some rise of external noise.

III. Results and Discussion

Obtained heterostructures had a relatively low Josephson critical frequency f_C=(2e/h)I_CR_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 manipulated by a weak external microwave signal. In order to answer the question how our heterostructures behave at high frequencies f_e >> 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 MHz for a heterostructure with parameters I_C=27.5 µA, R_N=8 mΩ at T=4.2 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 GHz. Fig. 2 shows n=±1 Shapiro steps, I_1, and half-integer steps n=±1/2 as well. Parameters of the heterostructure were L= 10 µm, I_C = 88 µA, and R_N = 0.16 Ω. The maximum of the first Shapiro step was I_1 = 94 µA 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_{C2}/I_{C1} of second harmonic amplitude of critical current I_{C2} to the amplitude of the first harmonic I_{C1} we used numerical approximation of the dependences of the experimental critical current amplitudes I_C 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 ω= f_e/f_C >1 and applied microwave currents I_{MW} > I_C the half-integer Shapiro steps

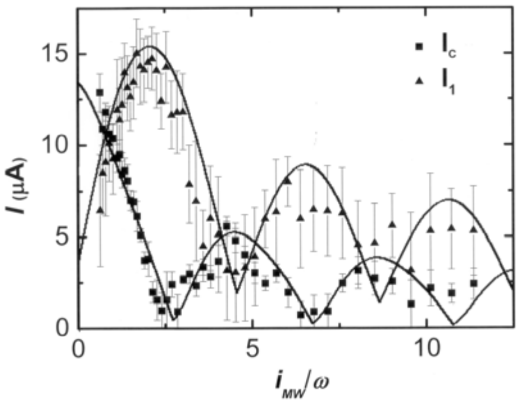

Fig.3. Amplitudes of critical current I_C and first Shapiro step I_1 of the vs. normalized microwave current at f_e=3 GHz, T = 4.2 K and magnetic field H=133 Oe. Sample parameters: d_{SRO}=5.6 nm, d_{LSMO}=15 nm and L=50 μm, λ_J=220 μm. Solid lines show extended RSJ digital simulation for q=0.2.

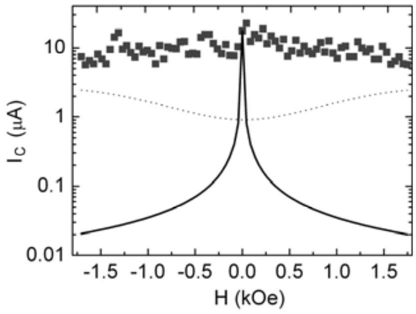

Fig. 4. Magnetic field dependence of critical current for a heterostructure with d_{SRO}= 8.5 nm, d_{LSMO}=3 nm, L=10 μm, λ_J=51 μm, measured in wide range of span. Dashed line shows noise floor. Solid line shows expected decay of maxima of I_C amplitudes oscillating by Fraunhofer dependence.

may appear [14] if McCumber parameter $\beta_C = 2\pi f_C R_N C \sim 1$. For the heterostructure shown in Fig.2 parameters were ω=5.9, and β_C<0.02 estimated taking the barrier capacitance C< 1 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_e = 3 GHz. Fig. 3. shows dependencies of I_C and I_1 vs. normalized microwave current i_{MW}/ω, where $i_{MW}=I_{MW}/I_C$. As we mentioned earlier at this level of magnetic field μ-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 Oe, which is much larger than $I_C(H)$ oscillation period ΔH=1 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 of n=1 Shapiro step, which became very close to the theoretical maximum I_1/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 kOe. The data in Fig.4 have been obtained changing magnetic field by the steps expanding over a few ΔH periods of $I_C(H)$ oscillation. $I_C(H)$ dependence was different from Fraunhofer pattern, and oscillates with a period ΔH=6 Oe. Note, the width L=10 μm is smaller than λ_J=51 μm, the Josephson penetration depth of magnetic field. An other heterostructure with parameters d_{SRO}= 8.5 nm, d_{LSMO}=6 nm, and L=20 μm, had Ic = 16.5 μA at H = –1.3 kOe which is 94% of $Ic(H{=}0)$ and 70% of the maximal peak at H = –6.5 Oe.

In comparison, YBCO/Au/Nb structures without magnetic interlayer [13], or YBCO/CSCO/Au/Nb with the antiferromagnetic $Ca_{0.7}Sr_{0.3}CuO_2$ (CSCO) interlayer [16] demonstrated sharp decrease of critical current with increase of magnetic field, while in some of discussed $S_1/F_1/F_2/S_2$ heterostructures the critical current even increased at magnetic fields stronger than 1 kOe. Such unusual behavior of the critical current was also reported in references listed in [1], [10] for spin-triplet junctions with metallic ferromagnetic interlayer. We observed a significant critical current for in-plane oriented H-fields up to 2.5 kOe in zero-field cooled mode. Measurements in field cooled mode were started from temperature 160 K, which is higher than T_C of superconductors and lies in between of Curie temperature of SRO, 145 K, and 350 K for LSMO.

Theories [7], [8] predict a dominating second harmonic for the certain thicknesses of F_1 and F_2 films in composite interlayer and the angle between their magnetizations. In accordance to [8] the second harmonic increases when misorientation angle of magnetization in bilayer approaches π/2. According to measurements by SQUID magnetometer [11] an in-plane antiferromagnetic ordering of magnetizations takes place at the interface of LSMO and SRO layers. Consequently at H=0 the angle between magnetizations of the ferromagnetic layers at F_1/F_2 interface could be assumed near to π, and critical current is minimal in accordance with calculations in [8]. For the LSMO, which is characterized by uniaxial magnetic anisotropy, the angle between the direction of magnetization and the external magnetic field is determined by in-plane magnetic field for H>100 Oe, larger than saturation level of LSMO magnetization. Consequently, an increase of the second harmonic in the CPR could be observed even at relatively weak magnetic fields. For heterostructure with d_{SRO}= 8.5 nm, d_{LSMO}= 6 nm and L = 20 μm we estimated q=0.46 at H=0 when only intrinsic magnetism takes place. Harmonic components of CPR could be evaluated also from oscillatory dependences $I_C(H)$ [10]. We analyzed data recorded at $|H|$<30 Oe for three heterostructures #1, #2, #3 located on the same chip with L=10 μm, 20 μm, and 40 μm, and total F_1/F_2 thickness $d_{SRO}+d_{LSMO}$=14.5 nm. Fast Fourier transform (FFT) analysis give q values 0.71, 0.63, and 0.61 for

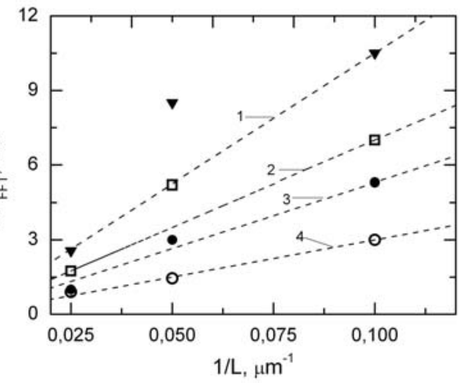

Fig. 5. Periods ΔH_{FFT} of the Fourier components of $I_C(H)$ dependences vs. parameter $1/L$ for 3 heterostructures on a chip (d_{SRO}= 8.5 nm, d_{LSMO}= 6 nm) with widths L = 10 µm, 20 µm, 40 µm, having λ_J= 120 µm, 110 µm, and 190 µm, correspondingly. Harmonic numbers point on linear $\Delta H_{FFT}(1/L)$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

heterostructures #1, #2, #3, correspondingly. Fig 5 shows decrease of ΔH_{FFT} with 1/L. Heterostructes #1 and #3 show linear decrease, while ΔH_{FFT} 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 $R_N L^2$ products (13 µΩ cm^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 $I_C(H)$ from regular Fraunhofer diffraction pattern we omit making conclusions about the impact 3rd and 4th harmonics on the CPR of our $S_1/F_1/F_2/S_2$ heterostructures.

It should be noted that occurrence of the second harmonic in the CPR in a Josephson junction made from *s*-wave and *d*-wave superconductors could be due to *d*-wave symmetry of the YBCO wave function in the *ab* plane. However, in the *c* direction of YBCO, the critical current is determined by the *s*-wave component of the superconducting order parameter. Our heterostructures were grown over *c*-oriented YBCO film and have small *s*-wave component of S_s electrode in experiment as manifested in small values of the critical frequency f_C as compared to the structures without magnetic interlayer [13].

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.

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