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# Hybrid superconducting mesa-heterostructure with manganite-ruthenate interlayer

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**Abstract.** We present experimental data on Josephson effect in hybrid superconducting mesa-heterostructures (HSMH) with composite manganite-ruthenate interlayer. The HSMH base electrode consisted of the cuprate superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_7$  grown epitaxially on a NdGaO substrate using laser ablation. The interlayer was composed from in-situ deposited  $\text{SrRuO}_3$  (F1) and  $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$  (F2) thin films, each characterized by different directions of magnetization. The top electrode was Nb/Au thin film. A superconducting current was observed when the interlayer thickness was well above the correlation length, determined by the exchange field in F1 and F2. Obtained  $I_C(H)$  dependences and non-sinusoidal current-phase relation evaluated from microwave measurements are discussed in terms of generation of long-range spin triplet superconducting current component in heterostructures with interfaces of singlet superconductors and bilayer ferromagnetic materials with different spatial directions of magnetization.

## 1. Introduction

In superconducting heterostructures with a non-uniform magnetized ferromagnetic layer (F) between two singlet superconductors (S) a long-range triplet superconducting correlations may occur [1]. In SFS structures with uniform magnetization the spin projection on direction of magnetization is conserved and the both singlet and triplet superconducting correlations with zero spin projection appear [1, 2], penetrating into F with a characteristic length  $\zeta_F = (\hbar D / E_{ex})^{1/2}$  determined by the magnetic exchange energy  $E_{ex}$ , where in the dirty limit  $D = v_F l / 3$  is the diffusion coefficient,  $v_F$  is the Fermi velocity, and  $l$  is the mean free path. The long-range triplet superconducting correlations with nonzero ( $\pm 1$ ) spin projection are not suppressed by the exchange interaction and is determined by temperature  $T$ :  $\zeta_N = (\hbar D / k_B T)^{1/2}$  as for SNS junctions with normal metal (N) interlayer. If  $k_B T \ll E_{ex}$  then a long-range triplet superconducting correlations leads to existence of superconducting current in SFS structures at sufficiently large distances between superconductors [3-10]. Here we report on experimental studies of S/M/S<sub>d</sub> hybrid superconducting mesa-heterostructures (HSMH), where S is the Nb/Au bilayer, S<sub>d</sub> is the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) cuprate superconductor, and M is a composite magnetic oxide interlayer consisting of two thin F-films,  $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$  and  $\text{SrRuO}_3$  with noncollinear magnetization. SFS structures with spin triplet superconducting correlations were theoretically considered in [11-15].



## 2. Samples and measurements

Square shape HSMH with in-plane dimensions  $L = 10 - 50 \mu\text{m}$  were fabricated from  $S_d$  hybrid superconducting heterostructures deposited on (110)  $\text{NdGaO}_3$  substrates using photolithography, plasma chemical, and ion etching [16, 17]. The base electrode was  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , the interlayer M consisted of two epitaxially grown ferromagnetic films: F1 –  $\text{SrRuO}_3$  (SRO) and F2 –  $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$  (LSMO) with thicknesses from 5 to 30 nm (see inset in figure 1). The upper electrode was Nb/Au bilayer as the LSMO/YBCO interface exhibits insufficient transparency [16]. The magnetization vector of the LSMO is lying in the plane of the substrate [17], whereas the magnetization vector of SRO is directed at  $\sim 23^\circ$  from the normal to the substrate [18]. Ferromagnetic resonance in LSMO/SRO heterostructure was studied at 10 GHz and showed a uniaxial magnetic anisotropy that is characteristic of LSMO films deposited on (110)NGO substrate [17]. However, the ferromagnetism of the SRO film was not revealed from ferromagnetic resonance studies because of high, about 1 T, saturation field of SRO. Note, peaks in X-ray spectrum from YBCO, LSMO, SRO were observed indicating epitaxial growth of all oxide films and the absence of materials mixing at the interfaces.

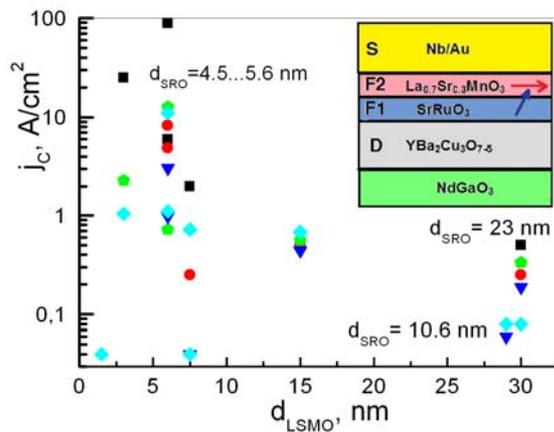
## 3. Results and discussion

The superconducting current was observed in all HSMH with a total thickness  $d$  of the composite M-interlayer up to 53 nm, which is much larger than the coherence lengths in F1 and F2, determined by the exchange field. We estimated the coherence length  $\xi_{\text{LSMO}} = 7 \text{ nm}$  in the LSMO film using the exchange energy  $E_{\text{ex}} = 2.3 \text{ meV}$  taken from electron specific heat [19], and  $v_F = 2 \cdot 10^7 \text{ cm/s}$  in dirty limit for the mean free path  $l_{\text{LSMO}} \approx 10 \text{ nm}$ . For SRO the  $E_{\text{ex}} = 13 \text{ meV}$  was taken from the data on proximity effect at SRO/YBCO interface [20], and in the dirty limit  $v_F = 10^7 \text{ cm/s}$  taking mean free path  $l_{\text{SRO}} \approx 10 \text{ nm}$  we estimate  $\xi_{\text{SRO}} = 4 \text{ nm}$ , close to the value obtained in [21].

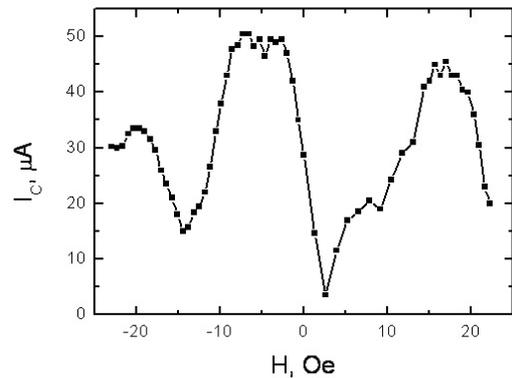
Control measurements of the HSMH with only the LSMO [16] or the SRO interlayer, showed [22] that the critical current is absent if the SRO and LSMO films are thicker than  $d_1 = 14 \text{ nm}$  and  $d_2 = 2 \text{ nm}$ , respectively. The critical current density  $j_C$  of the HSMH decreases by an order of magnitude, when increasing the total thickness of  $d = d_1 + d_2$  from 8.5 to 53 nm. The value  $j_C = 9 \text{ A/cm}^2$  was observed for a sample with  $d = 11.5 \text{ nm}$ ,  $L = 10 \mu\text{m}$ . Figure 1 shows that critical current density  $j_C$  depends non-monotonically on the thickness  $d_2$  of the LSMO. A relatively low  $j_C$  at small  $d_2$  (1.5 and 3 nm) could be explained by existence of a “dead” nonmagnetic layer at the interface. Note, a non-monotonic thickness dependence of the critical current was predicted [14, 15] for structures with long-range triplet superconducting correlations with a maximum of  $j_C(d)$  at  $d \approx \xi_F$  [14]. In our HSMH further significant increase in  $d$  resulted in a decrease of  $j_C$  as expected from theoretical calculations [11–14].

The measurements of the critical current  $I_C$  as a function of magnetic field  $H$  (see figure 2) show that maxima of  $I_C$  are observed in the range 5 – 15 Oe. Note the HSMH and the  $H$ -field coil were screened by the cooled  $\mu$ -metal shield. Similar  $I_C(H)$  dependences were observed for SFS junctions where long-range triplet superconducting component was induced [5, 10]. In a wider variation of the external magnetic field, a hysteretic behaviour was observed, indicating the existence of ferromagnetism in the M-interlayer [23]. The  $I_C(H)$  dependence shown in figure 2 does not exhibit hysteresis because the total range of  $H$  variation is much lower than the saturation magnetic field of the ferromagnetic layer [23].

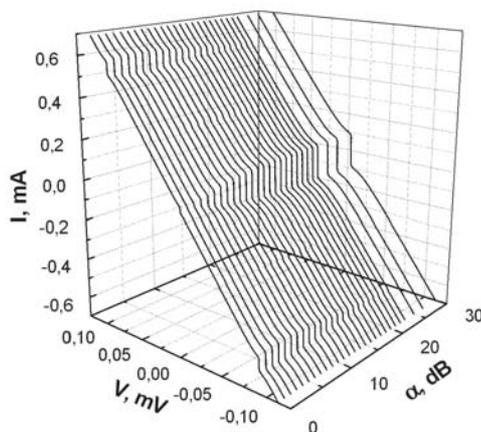
The theoretical calculations reported in [12, 13, 15] predict a strong increase of the second harmonic in the current-phase relation of the superconducting current  $I_S(\varphi) = I_{C1}\sin(\varphi) + I_{C2}\sin(2\varphi)$  for the asymmetric M-interlayer ( $d_1 \neq d_2$ ) under the variation of the angle between the directions of the magnetizations of the F films, giving  $I_{C2} \gg I_{C1}$ . The measurements of Shapiro steps of HSMH demonstrate deviation from the sinusoidal current-phase relation. The  $I$ - $V$  characteristics of the HSMH with  $L = 10 \mu\text{m}$ ,  $I_C = 88 \mu\text{A}$ , and  $R_N = 0.16 \Omega$  under microwave radiation at  $f_e = 41 \text{ GHz}$  exhibit both integer and fractional Shapiro steps (see figures 3 and 4). The maximum of the first Shapiro step was  $I_1 = 94 \mu\text{A}$  and, correspondingly, the ratio  $I_1/I_C = 1.1$  is in well agreement with the resistively shunted junction (RSJ) model, as the critical frequency  $f_C = (2e/h)I_C R_N = 6.8 \text{ GHz}$ , which



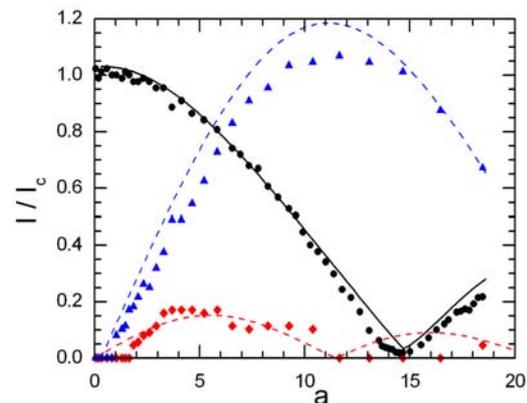
**Figure 1.** Superconducting current density in HSMH vs. thickness of the LSMO film at  $T = 4.2$  K and  $H=0$ . The squares, circles, pentagons, triangles, and diamonds mark  $L$  equal to 10, 20, 30, 40, and 50  $\mu\text{m}$ , respectively. The inset shows the cross section of the HSMH with different directions of the magnetization vectors.



**Figure 2.**  $H$ -field dependence of the critical current for HSMH with  $L = 50 \mu\text{m}$ ,  $d_1 = 4.5 \text{ nm}$ , and  $d_2 = 3 \text{ nm}$ ,  $T = 4.2 \text{ K}$ . The range of  $H$ -field is much below the level of saturation field of the ferromagnetic interlayer.



**Figure 3.** Family of the current–voltage characteristics of the HSMH under microwave irradiation at 41 GHz, where  $\alpha$  is the inserted damping by external polarizing attenuator.



**Figure 4.** Shapiro steps for a HSMH with  $d_1 = 6 \text{ nm}$ ,  $d_2 = 5.5 \text{ nm}$ , and  $L = 10 \mu\text{m}$ ,  $T = 4.2 \text{ K}$ .  $I_c$  (circles),  $I_1$  (triangles), and  $I_{1/2}$  (diamonds), are plotted vs.  $a = I_{RF}/I_c$

gives the ratio  $f_e/f_c = 6$ , very well corresponds to the high frequency limit of the RSJ model. The maximum of the half-integer Shapiro step was  $I_{1/2} = 15 \mu\text{A}$  and within the modified RSJ model [24], which takes into account a non-sinusoidal current–phase relation, the fraction of the second harmonic  $I_{c2}/I_{c1}$  is about 13%. It is worth noting that the direct comparison of experimental results with theory [11–15] is complicated because of the presence of a barrier between the manganite and superconducting electrodes. The authors of paper [14] stated that a long-range triplet superconducting

correlations hardly may exist in a structure with only two ferromagnetic layers and the “third component” is required. In our case, likely, one of the S/M barriers is magnetically active and serves as the “third component.” Also, it is not excluded that an antiferromagnetic barrier may appear at the SRO/LSMO interface [25] providing the function of the “third component” of a spin active interface.

### 3. Conclusions

We have experimentally observed the superconducting current in hybrid mesa-heterostructures with a composite oxide ferromagnetic bilayer with non-collinear directions of the 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 structures is explained by the penetration of the long-range triplet component of the superconducting order parameter into the magnetic interlayer. The deviation of the current–phase relation from a sinusoidal dependence has been measured having 13% fraction of the second harmonic that can also be explained by the triplet component of the superconducting order parameter.

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