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Current-Phase Relation of Superconductor-Ferromagnet-Superconductor Junctions with a Composite Interlayer

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Abstract. We report on microwave measurements of hybrid superconductor-ferromagnet-superconductor (SFF'S') junctions with a composite oxide ferromagnetic bilayer with non-collinear magnetizations. Current-phase relation (CPR) of the junction superconducting current has been evaluated as a function of magnetic field. A significant part of the 2^{nd} harmonic in CPR, about 50%, was measured.

Introduction

Over the past decade triplet superconducting correlations in superconductor-ferromagnet-superconductor (SFS) junctions became a topic of active research. The spin-triplet superconducting correlations have to be generated in case of spatial inhomogeneity in magnetic interlayer [1, 2]. A progress in experimental and theoretical studies has been summarized in a review [3]. However, an important question about the current-phase relation still remains unanswered, with the exception of a recent work [4], in which CPR of the junction was studied via measurements of the magnetic field dependencies of critical current at dc $I_C(H)$. Theoretically, in both clean [5] and diffusive [6] limits the dominant spin-triplet 2^{nd} harmonic in CPR of SFF'S' junctions was predicted. Moreover, author of [5] claims that 2^{nd} harmonic could exceed the 1^{st} one up to 10^3 times. Authors of [6] predict the 2^{nd} harmonic to be long-ranged, while the 1^{st} one is also triplet, but corresponds to short-ranged superconducting correlations. These theoretical predictions require experimental verification. We present experimental results of determination of CPR of hybrid SFF'S' superconducting junctions with non-collinear magnetizations in a composite interlayer via microwave measurements of Shapiro steps, varying also the external magnetic field.

Samples and experimental methods

Hybrid superconducting heterostructures with a composite magnetic interlayer have been fabricated utilizing epitaxial growth of oxide $YBa_2Cu_3O_{7-x}$ (YBCO) superconducting film on (110) $NdGaO_3$ or (001) $LaAlO_3$ substrates with *in-situ* laser ablation of ferromagnetic interlayer consisting of $SrRuO_3$ (SRO) and $La_{0.7}Sr_{0.3}MnO_3$ (LSMO). The top of the multilayer surface was *in-situ* covered by 20 nm Au film, and then Nb upper electrode was deposited *ex-situ* by RF magnetron sputtering after pre- sputtering of Au film and additional Au deposition again with 20 nm thickness. Samples were patterned using photolithography, ion-beam etching and lift-off to obtain mesa-heterostructures with in-plane sizes from 10 to 50 μm

In order to reveal the 2nd harmonic in CPR, measurements of Shapiro steps or selective detector response functions were used as described in [7]. In high-frequency limit deviations from Bessel functions in oscillating dependences of Shapiro steps vs. microwave power allowed us to estimate the fraction of 2nd harmonic. Another method for CPR measurement is based on examining the $I_C(H)$ dependencies (see, e.g. [4]), but it tells too little about high-frequency dynamics of Josephson junction.

Results and discussion

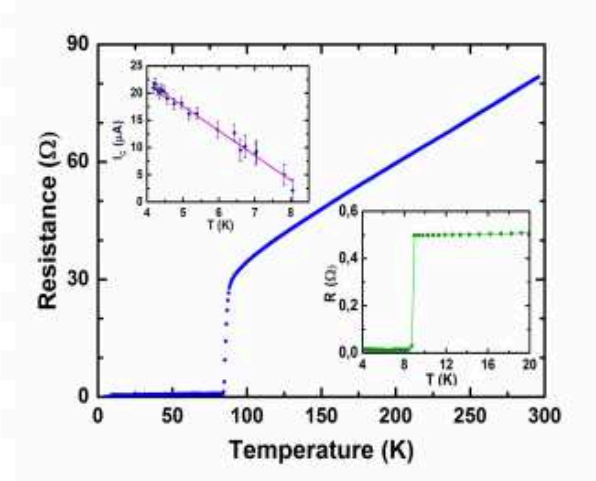

Fig. 1. (colour online) Temperature dependence of resistance $R(T)$ of mesa-heterostructure with in-plane size L=30 μm, d_{LSMO} =9 nm, d_{SRO} =10 nm. Lower inset: $R(T)$ in the vicinity of superconducting transition of Nb-Au electrode. Upper inset: temperature dependence of critical current.

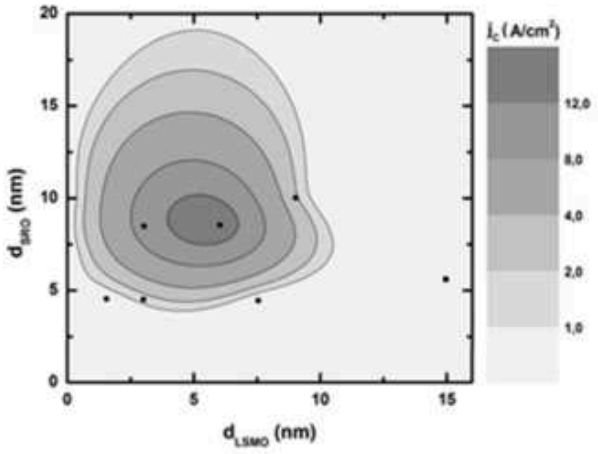

Fig. 2. Distribution of critical current density j_C values on the $d_{SRO} - d_{LSMO}$ plane, T = 4.2 K, H=0. Black points show j_C averaged over data for several junctions on the same wafer.

In heterostructures with only one ferromagnetic interlayer (either SRO or LSMO) superconducting current was not observed even though the thickness of interlayer was reduced to the coherence length in the ferromagnet. But when we used a composite ferromagnetic interlayer containing both SRO and LSMO (the SRO **M**-vector has an out-of-plane component, and LSMO has **M** lying in-plane), the superconducting current was registered even in case of $d_{LSMO} >> \xi^{LSMO}$. Particularly,sample with d_{SRO} = 5.6 nm, d_{LSMO} = *15* nm and L = 50 μm had critical current I_C = 17 μA. Fig. 1 shows temperature dependence of resistance $R(T)$ of SFF'S' mesa-structure and the temperature dependence of critical current. This observation could be explained by spin-triplet superconducting correlations in SFF'S mesa-heterostructures. Critical current dependence on manganite thickness d_{LSMO} shows maximum at $d_{LSMO} \sim \xi^{LSMO}$ for the main part of experimental samples. Analyzing data obtained for a few tens values of critical current density $j_C = I_C/L^2$ of mesa-heterostructers with different d_{SRO} and d_{LSMO} thicknesses a distribution of j_C on $d_{SRO} - d_{LSMO}$ plane was obtained, shown in Fig.2. It is seen that as d_{SRO} and d_{LSMO} thicknesses approach the coherence lengths of ferromagnets, the j_C data demonstrate a maximum. Note, theory [6] also predicts non-monotonic dependence with a maximum for spin-triplet superconducting current. Reduced critical current densities j_C were observed at lower thicknesses of LSMO and could be explained by presence of a so-called "dead" nonmagnetic layer of manganite.

We have recently reported on half-integer Shapiro steps [8] and the CPR with sufficient 2nd harmonic component. Here we study Shapiro steps under the influence of external magnetic field and observed them for a few different samples. Half-integer Shapiro steps were registered for some of the samples when they were not washed up by background noise of experimental setup. This

clearly points at the nonsinusoidal CPR as the experimental conditions well correspond to the high-frequency limit [7] when influence of capacitance is negligible. Critical current and Shapiro steps amplitudes were measured at either f_e = 3 GHz using a coaxial cable system, or at 40 GHz using 8-mm rectangular waveguide (see Fig. 3). Data present critical current and Shapiro steps vs. normalized RF current $x = I_{RF}/\omega I_C$, where $\omega = hf_e/2eI_CR_N$ is normalized frequency (h is a Planck's constant, e – electron charge, R_N – normal resistance. Microwave measurements also confirm absence of pinholes in experimental samples because the presence of pinholes would result in a significant reduction of Shapiro steps heights from the ones expected in resistively shunted junction model. To determine the 2nd harmonic fraction q, we used approach described in [7]. For simplicity we took $I_C \approx I_{C1}$ assuming small values of q<1. This may have resulted in somewhat overestimated I_{C1} amplitudes and reduced q values.

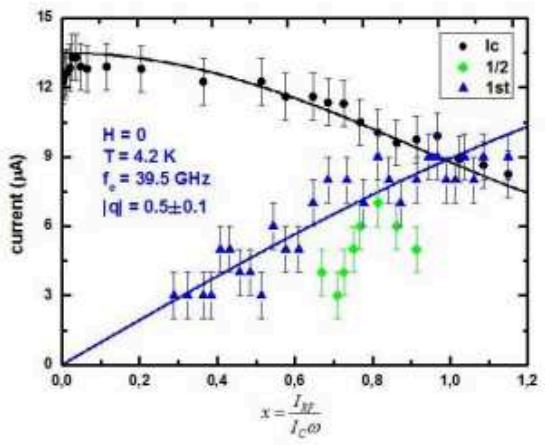

Fig. 3(a). (colour online) Amplitudes of critical current, first and half-integer ½ Shapiro steps of the sample with d_{SRO} = 8.5nm, d_{LSMO} = 6nm and L = 20 μm under microwave irradiation of 39.5 GHz at T = 4.2 K. Solid lines show dependencies $I_C(x)$ and $I_1(x)$ calculated within the modified resistively shunted junction model taking into account nonsinusoidal CPR.

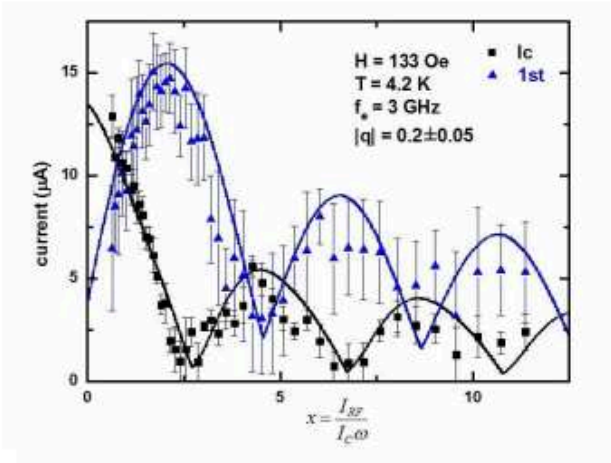

Fig. 3(b). (colour online) Amplitudes of critical current and first Shapiro step of the sample with d_{SRO} = 5.6nm, d_{LSMO} = 15nm and L = 50 μm dependencies on normalized RF current of 3 GHz at T = 4.2 K. Solid lines show calculated $I_C(x)$ and $I_1(x)$ dependencies.

The fraction of second harmonic q was smaller when we applied magnetic field H. For the sample with d_{SRO} = 10 nm, d_{LSMO} = 9 nm and L = 30 μm we obtained q = 0.25±0.05 for H = 0 and q= 0.15±0.05 for H = 50 Oe. Fig. 3a demonstrates the case of registered well pronounced half-integer Shapiro steps and q = 0.5±0.1. Measurements of $I_C(H)$ functions showed oscillating behaviour with very high unexpected maxima under relatively strong magnetic fields. We suppose that obtained $I_C(H)$ dependencies are the product of conventional Fraunhofer pattern influenced by the long-range spin-triplet proximity effect: a significant critical current was observed at in-plane directed H up to 2.5 kOe in zero-field cooled mode. Our measurements were also performed in field cooled mode started from temperature of 160 K, but restricted by H level when we used superconducting coil.

Additionally, $I_C(H)$ had hysteretic behaviour due to magnetization switching of LSMO layer which has a coercive field of about 100 Oe. Change in amplitudes of critical current and deviation of CPR from sinusoidal type are caused by the change of angle between magnetizations in ferromagnets forced by external magnetic field. Note, H field rotates only the LSMO **M**-vector, while the SRO magnetization stays in the same direction since its coercive field is about 1 T.

Theories [5, 6] predict dominant 2nd harmonic in CPR for the certain thicknesses of FF' interlayer and angles between magnetizations. We applied magnetic field which may change direction of magnetization in weaker ferromagnet in FF' bilayer. We did not apply too strong magnetic fields to try making the angle between magnetization of π/2, around which according to [6] the 2nd harmonic of long-range triplet superconducting component should reach its maximum.

Conclusion

We have experimentally observed the superconducting current in hybrid mesa-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 mesa-heterostructures is explained by penetration of the long-range triplet component of the superconducting order parameter into magnetic interlayer. The deviation of the current-phase relation from a sinusoidal dependence has been measured having at least 50% fraction of the second harmonic that can also be explained by the triplet component of the superconducting order parameter.

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