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COMMUNICATIONS

EFFECT OF GALLIUM FOCUSED ION BEAM IRRADIATION ON PROPERTIES OF $YBa_2Cu_3O_x/La_{0.67}Sr_{0.33}MnO_3$ HETEROSTRUCTURES

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We present initial investigation of the superconductor-ferromagnet-superconductor (SFS) heterostructures of nanometer dimensions prepared by the gallium focused ion beam (FIB) technology. The SFS heterostructures were realized on the basis of high-Tc superconducting $YBa_2Cu_3O_x$ and ferromagnetic $La_{0.67}Sr_{0.33}MnO_3$ thin films. SFS weak link junctions require dimensions of the weak link connection in the range of nanometer size realizable by FIB patterning. On the other side the gallium focused ion beam might bring about unacceptable degradation of the superconducting as well as ferromagnetic thin film properties. The presented results show that FIB offers a suitable procedure for realization of nanometer size devices but some degradation of the ferromagnetic and superconducting properties was observed. Solution of this problem will be achieved in the next stage of our investigations.

Keywords: superconductor-manganite junctions, proximity effect, magnetic domain wall

1 INTRODUCTION

Superconductor-ferromagnet (SF) bilayer and SFS trilayer heterostructures, in the form of weak links or Josephson junctions, are very attractive objects for the study of mutual interplay between superconductivity and ferromagnetism [1]. In addition, their potential applications are very promising in cryoelectronic or cryospintronic circuits [2] (*eg*, qubits, 0-, pi-junctions, spin valves, *etc*). However, utilization of high-Tc superconductors in this area was not successful until now. In the case of the high-Tc cuprate superconductor $YBa_2Cu_3O_x$ (YBCO), as convenient ferromagnetic (F) materials are manganites, *eg* $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) — ferromagnetic perovskite half metal, which may be totally spin polarized in one spin direction. YBCO/LSMO heterostructures, prepared on single crystal MgO or $SrTiO_3$ substrate, are able to create high quality thin films and SF interfaces necessary for the study of the physical properties. In addition, present advanced technologies allow realization of structures of nanometer dimensions which open new possibilities for preparation of high-Tc superconducting weak links with properties similar to low-Tc Josephson junctions, however operating at much higher temperatures. In this paper we describe the first stage of preparation and the properties of YBCO/LSMO/YBCO nanometer heterostructures, using the technology of gallium focused ion beam (FIB) patterning, to find out whether this technology is a suitable tool for realization of superconductor weak link structures.

Generally, electron transport through the interface between the superconductor (S) and normal (N) or ferromagnetic (F) metal in close proximity with S, is mediated by Andreev reflection [3], the process in which an electron with energy smaller than the energy gap of S is reflected as a hole preserving in N (F) phase coherence of Cooper pairs (CP) over some distance from the interface. There are essential differences between the proximity effect in SN and SF structures. In SN connection the penetration depth of singlet spin ($\uparrow\downarrow$) CP into the N metal is determined by the coherence length $\xi_N = (\hbar D_N/2\pi k_B T)^{1/2}$, where D_N is the diffusion coefficient in N, T is temperature, and $\hbar$ and k_B are the Planck and Boltzmann constants. At very low temperatures ξ_N can reach several micrometers. In the case of a SF bilayer the penetration depth of singlet spin CP into the ferromagnet is much shorter $\xi_F = (\hbar D_F/2\pi E_{ex})^{1/2}$ provided that the magnetic exchange energy $E_{ex} \gg k_B T$ is rather large, which is fulfilled in the case of perovskite (half metal) materials. Fortunately, in addition to the short coherence length ξ_F ($\sim$1–2 nm) there was discovered a long range proximity effect (LRPE) in FS structures, in case inhomogeneous magnetization in the vicinity of SF interface is present [4]. It was also shown [5] that such inhomogeneity (*eg*, domain wall, spin active interface) generates a triplet spin CP, with amplitude comparable to the singlet one, containing phase correlations between electrons with the same spin projections ($\uparrow\uparrow$), on the coherence length ξ_{FL}. The penetration depth in the case of LRPE should be of the order ξ_N as in the case of singlet spin CP into a normal metal. Large attention is therefore dedicated to the investigation of Josephson effects in cuprate SFS struc-

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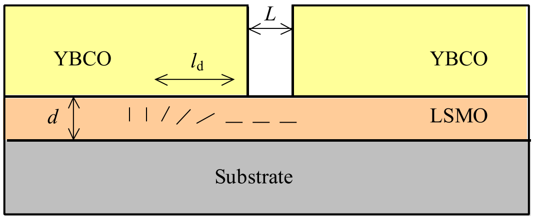

Fig. 1. Lateral geometry of the SFS microstrip structure. Magnetic inhomogeneity in LSMO of thickness d is intended to be realized by domain wall of width l_d. The two YBCO electrodes are separated in length L as shown

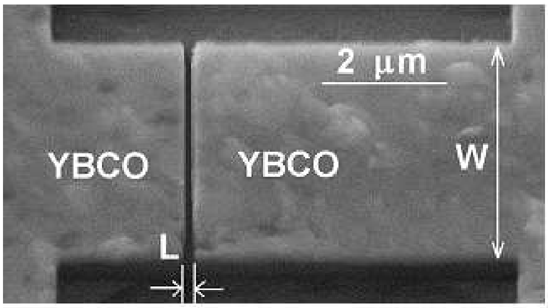

Fig. 2. The YBCO/LSMO microstrip ($7 \times 4\,\mu\mathrm{m}^2$) with a gap in YBCO thin film (length $L \approx$ 70–80 nm) created by FIB etching. This SFS structure represents YBCO/LSMO/YBCO junction of *lateral geometry*

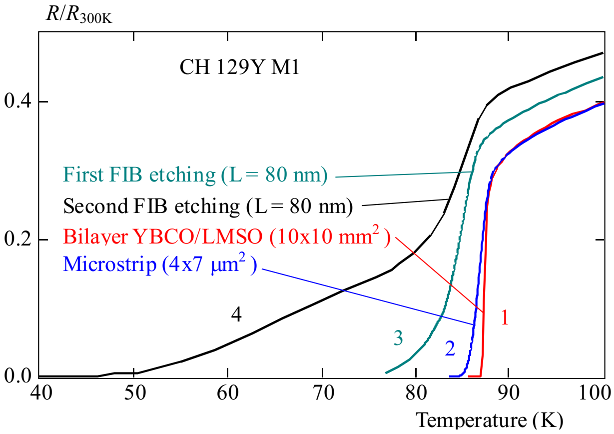

Fig. 3. The normalized R–T dependences of four sequences of SFS structure preparation: 1– as prepared bilayer film, 2– bilayer microstrip ($7 \times 4\,\mu\mathrm{m}^2$), 3– after the first FIB etching of the gap in YBCO, and 4– after the second FIB etching of the gap

tures due to possible larger variety of Josephson junction properties realized on these materials combination [6].

Realization of high quality high-Tc SF or SFS structures manifesting LRPE is a complicated task. The magnetic inhomogeneity, in the so-called *series geometry*, must be localized immediately at the SF interface, which is experimentally extremely difficult, otherwise the triplet current amplitude is negligibly small. Recently, it was analysed [7] that, in comparison with the *serial geometry*, the amplitude of the triplet current component may be in the so- called *lateral geometry* (Fig. 1) enhanced by factor l_d/d, where l_d is the domain wall width and d is the thickness of F thin film. The domain wall width [8] $l_d = \pi(A/K)^{1/2}$, where A is the exchange energy acting to keep spins parallel and K is the energy of magnetization anisotropy. Competition of these energies results in a finite width of the domain wall. Recently, the values of A and K were extracted for LSMO from LSMO/SrRuO$_3$ bilayers and superlattices [9] as $A = 5 \times 10^{-12}\,\mathrm{J/m}$ and $K = -5 \times 10^3\,\mathrm{J/m}^3$, thus the domain wall width may receive values in the range of 100 nm. In reality the values of A and K depend on composition, temperature, stress in the F film, consequently, for $l_d/d \gg 1$ the high quality and small thickness (10-20 nm) of the ferromagnetic film is required to bring LRPE coherence length ξ_{FL} closer to ξ_N.

In the case of *lateral geometry* the LRPE should be independent of the domain wall localization [7], in the immediate vicinity of SF interface, what enables considerable simplification of technological conditions. In the ongoing experimental investigation we will henceforth verify these analyzes.

2 EXPERIMENTAL

The dc magnetron sputtering was used for in situ growing the bilayer heterostructure YBa$_2$Cu$_3$O$_x$ (YBCO) and La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO) on single crystal MgO (100) substrate. The single LSMO thin films showed transition to the metallic ferromagnetic state at about 200 K and their resistivity at liquid nitrogen temperature was $\rho \approx 10^{-3}\,\Omega\mathrm{cm}$, [10]. The thickness of LSMO layers was in the range 20–50 nm where the LSMO thin film should be ferromagnetic [11]. The LSMO crystallizes as pseudocubic perovskite, it has a fully spin-polarized conduction band and bulk material exhibits ferromagnetic transition around room temperature. The YBCO films were deposited applying high pressure on-axis dc magnetron sputtering carried out at oxygen pressure 300 Pa, substrate temperature $T_s = 810\,°\mathrm{C}$, and dc power 200 W, with a deposition rate of 1 nm/min [12], [13]. The thickness of the YBCO superconducting films was about 150 nm.

Patterning of the basic bilayer structures for four point measurement of the transport properties was carried out by optical photolithography and wet (1 % H$_3$PO$_4$) or Ar ion beam etching (300 eV, $20\,\mathrm{mA/cm}^2$), with substrate cooled to about minus 20 °C. Subsequently Ga^{+3} focused ion beam patterning was applied (Quanta 3D 200i) to receive the convenient *lateral geometry* (Fig. 1).

3 RESULTS AND DISCUSSION

Zero resistance critical temperature T_{C0} of single c-axis oriented YBCO films, deposited directly on single crystal MgO substrate, was typically somewhat below 90 K, the critical current density at 77 K $j_C(77\,\mathrm{K}) \approx$

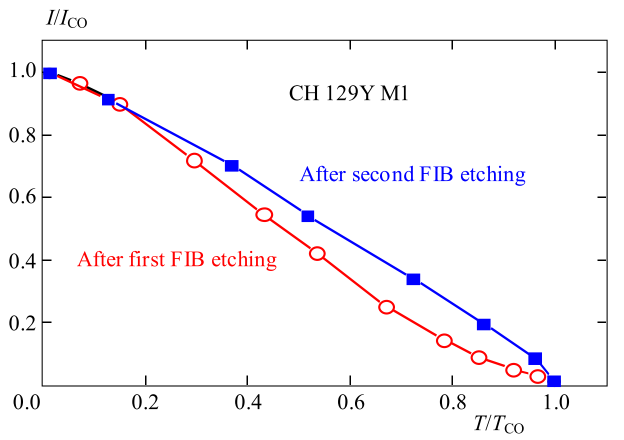

Fig. 4. Normalized dependences of the critical current *vs* temperature for YBCO/LSMO/YBCO structure after the first (open circles) and the second (filled squares) FIB etching of the gap in YBCO

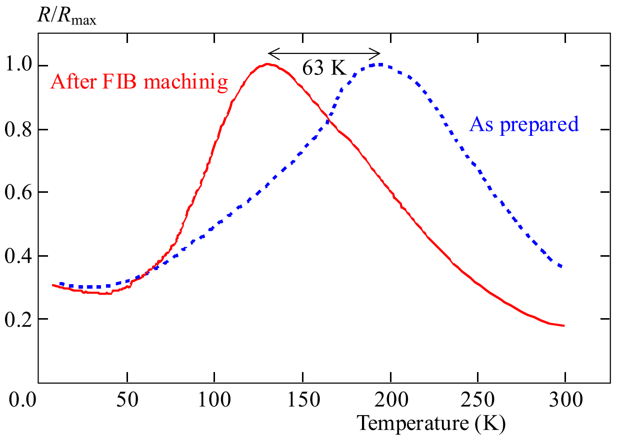

Fig. 5. R–T dependences of SFS structure after removing the YBCO film in the gap (full line) and LSMO film not influenced by FIB irradiation (dashed line)

3×10^6 A/cm^2 and the FWHM for the rocking curves of the (005) YBCO peak of 0.2°. A small decrease of T_{C0} and $j_C(77\,\mathrm{K})$ is due to the lattice mismatch between YBCO and MgO. Zero resistance critical temperature of the YBCO/LSMO bilayers was above 80 K, in the presented sample $T_{C0} = 87.5$ K (Fig. 3, curve 1). In Fig. 2 we show SEM picture of YBCO/LSMO microstrip ($4 \times 7\,\mu\mathrm{m}^2$). The YBCO/LSMO bilayer was removed outside the microstrip by Ar ion beam. Subsequently the sample was transferred in Quanta 3D 200i and the gap of length L in the YBCO film was realized by FIB etching. In addition the FIB etching was used for smoothing the microstrip edges.

The corresponding R-T dependence of the YBCO/LSMO microstrip is schown in Fig. 3, curve 2, at which a small decrease of T_{C0} to 85 K was observed due to the gallium ion irradiation. In the next step it was crucial to remove YBCO (without removing the LSMO thin film) as a narrow lateral gap of length L, to realize *lateral geometry* of the SFS structure. This is the most critical step of sample preparation because during the sample irradiation adjusted on the 30 keV Ga^{+3} ions the properties of superconductor and manganite films can be influenced.

After the first run of FIB etching (current 10 pA and time duration of the procedure 7 min) we obtained SFS weak link with relatively strong coupling of the two parts of YBCO. Zero resistance critical temperature T_{C0} decreased to 77 K (Fig. 3, curve 3). In the next run of FIB etching (current 10 pA and time duration of the procedure 3 min) we further weakened the SFS weak link coupling of the two YBCO parts, T_{C0} decreased to 43 K, whereas the resistance of SFS structure increased by about 50 %.

The weakened coupling between both YBCO parts in *lateral geometry* can be documented by the temperature dependence of the critical current through the gap of length L. Extrapolated zero temperature value of I_{C0}, after the first FIB etching of the gap, was 3 mA and corresponding temperature dependence of the critical current is shown in Fig. 4 by the open circle curve. The extrapolated zero temperature value of I_{C0}, after the second FIB etching of the gap, reaches 0.3 mA and its temperature dependence is in Fig. 4 depicted by the curve with filled squares. The two $I_C - T$ dependences indicate a frequent character of SNS weak link junctions [14], [15] therefore we suppose so far that a very thin residual YBCO film remains in the gap of the SFS structure with the above mentioned critical currents.

After the next FIB etching (current 10 pA and duration 1.5 min) we did not observed superconducting properties through the gap, therefore we suppose that YBCO was completely removed but the LRPE was not realized. This confirms the R–T dependence of SFS structure corresponding to typical LSMO dependence (Fig. 5, full line). However, in comparison to as-prepared LSMO R–T dependence (Fig. 5, dashed line) one can see a large (63 K) shift of the resistance maximum to a lower temperature.

As it was mentioned above the cutting of YBCO film can be accompanied by degradation of the under-laying LSMO film. The decreased temperature of the resistance maximum (transition of LSMO to ferromagnetic state) can be explained by the influence of gallium FIB irradiation on the LSMO.

CONCLUSIONS

The realization of high quality superconducting weak links or Josephson junctions based on high-Tc superconductors is at present a very difficult task. Some achievements are expected using advanced microcircuit technologies (*eg*, FIB) for preparation SNS or SFS structures of nanometer dimensions. SFS heterostructures, in comparison with SNS weak links, offer an opportunity for new physical effects (LRPE) as well as new modes of operation in cryoelectronic and cryospintronic circuits (pi-junctions, spin valves). In the paper we present preliminary results on the high-Tc superconducting SFS structure in the so-called *lateral geometry*. In the YBCO/LSMO bilayer the top YBCO layer was disconnected by narrow ($L \approx 70-80$ nm) lateral gap in the YBCO microstrip using focused ion beam (FIB) patterning. Results show that FIB offers suitable procedure for realization of nanometer size devices but problems of films degradation by FIB radiation have to be solved. Another separate problem, generation of LRPE in SFS structure, is creation of local magnetic inhomogeneity in the half metal LSMO film. Solution of these problems is the aim of the next period of our investigations.

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