Properties of SFS heterostructures prepared by a focused-ion-beam technique

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Abstract. We present a study of superconductor-ferromagnet (SF) bilayers and superconductor-ferromagnet-superconductor (SFS) heterostructures of nanometer dimensions prepared by a gallium focused-ion-beam (FIB) technique. The SFS heterostructures were implemented on the basis of high-\(T_c\) superconducting YBa\(_2\)Cu\(_3\)O\(_x\) (YBCO) and ferromagnetic La\(_{0.67}\)Sr\(_{0.33}\)MnO\(_3\) (LSMO) thin films deposited by magnetron sputtering on SrTiO\(_3\) (100) single crystal substrates. SFS weak link junctions require weak link dimensions in the range of nanometer size realizable by FIB patterning. On the other hand, the focused gallium ion beam might cause unacceptable degradation of the superconducting and ferromagnetic thin film properties. Our results confirm such influence of the FIB technology. However, protection of the structures by a gold thin film may effectively solve this problem, as is presented in the paper.

1. Introduction
Superconductor-ferromagnet (SF) bilayer and SFS heterostructures, in a form of weak links or Josephson junctions, are very attractive objects for the study of the interplay between superconductivity and ferromagnetism [1]. In addition, their potential applications are very promising in cryoelectronic or superconducting spintronic circuits [2] (e.g. qubits, 0-, pi-junctions, spin valves, etc.). However, utilization of high-\(T_c\) superconductors (HTS) has until now been only partially successful due to the very small coherence length of HTS. In the case of SFS junctions with high-\(T_c\) cuprate superconductor YBa\(_2\)Cu\(_3\)O\(_x\) (YBCO), manganites appear as convenient ferromagnetic materials (e.g. La\(_{0.67}\)Sr\(_{0.33}\)MnO\(_3\) (LSMO) - ferromagnetic perovskite half metal, which can be totally spin polarized) enabling one to create high-quality thin films and SF interfaces necessary for the study of their physical properties. Nevertheless, there are two essential differences in the transport properties of SF and SN (N-normal metal) heterostructures in close proximity. In SN structures, the penetration depth of Cooper pairs (CP) into the N metal is determined by the N metal coherence length \(\xi_N = (\hbar D_N/2\pi k_B T)^{1/2}\), where \(D_N = v_F l/3\) is the diffusion coefficient of N, \(v_F\) is the Fermi velocity of carriers, \(l\) the mean free length, \(T\) is the temperature, and \(\hbar\) and \(k_B\) are the Planck and Boltzmann constants. In case of SF bilayers, the...
penetration depth of CP into a ferromagnet is much shorter \( \xi_F \approx (\hbar D_F/2\pi E_{ex})^{1/2} \) since the magnetic exchange energy \( E_{ex} \gg k_B T \) in a ferromagnet. Fortunately, in addition to the short coherence length \( \xi_F \approx (1-2 \text{ nm}) \ll \xi_N \), it was found [3] that in case of presence of magnetic inhomogeneity \( \mathcal{F} \), there may exist a triplet component (with spins of CP oriented in the same direction) with a penetration depth close to \( \xi_N \) (the so-called long-range proximity effect (LRPE) in FS structures).

Nevertheless, the realization of high-quality high-\( T_c \) SF structures or SFS Josephson weak links 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. It was found recently [4] that, in comparison with the serial geometry, the amplitude of the triplet current component may be considerably enhanced in the so-called lateral geometry (figure 1), whereby the magnetic inhomogeneity may not be necessarily localized near the SF interface. In this paper, we report on the first attempt to prepare and study the properties of YBCO/LSMO/YBCO nanometer heterostructures using gallium focused ion beam (FIB) patterning in view of establishing whether this technology is a suitable tool for implementing such superconductor weak link structures.

2. Experimental

RF and DC magnetron sputtering were used for growing the ferromagnetic \( \text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \) (LSMO) and superconducting \( \text{YBa}_2\text{Cu}_3\text{O}_x \) (YBCO) bilayer heterostructures [5]. The polycrystalline thin films were directly deposited by RF off-axis single magnetron sputtering on \( \text{SrTiO}_3 \) (STO) single-crystal substrates. During the sputtering in \( \text{Ar}:\text{O}_2 \) (1:1) atmosphere with total pressure of 5.3 Pa, magnetron RF power of 30 W and substrate temperature of \( 780^\circ \text{C} \) were used. The LSMO layer thickness was about 30 nm. DC off-axis double magnetron sputtering was used for deposition of YBCO films on top of the LSMO layer. The film deposition was carried out at substrate temperature \( T_{\text{dep}} = 780^\circ \text{C} \) followed by annealing at temperature \( T_A = 530^\circ \text{C} \). The YBCO layer thickness was 70 nm.

The patterning of the basic structure was carried out by photolithography and argon ion beam etching with cooling of the substrate. The cross section of the lateral geometry is shown in figure 1. Figure 2 is a SEM image of the realized YBCO/LSMO/YBCO nanostructure with YBCO laterally removed within a length \( L < 100 \text{ nm} \) [6]. Before patterning, the FS bilayer heterostructure was investigated by the dc four-point method, as well as by a contactless inductive method. After patterning, only the dc method was used. X-ray diffraction analysis (XRD) was used to characterize the growth of both films.

![Figure 1. Cross section of the lateral geometry for long range proximity effect study. The domain wall width \( \ell_d \) and the YBCO separation \( L \), and LSMO layer of thickness \( d \) are shown.]

![Figure 2. SEM picture of a SFS junction consisting of a YBCO/LSMO nanostructure (5x0.3 \( \mu \text{m}^2 \)) with laterally removed YBCO within length \( L < 100 \text{ nm} \) (inset).]
3. Results and discussions

The $R$-$T$ measurement of the LSMO and YBCO single layers indicate very good properties of both thin films. The resistance of a 70-nm thick single YBCO film linearly decreases between 300-100 K with the ratio $R_{300K}/R_{100K} = 3$, and the resistance, extrapolated to temperature $T = 0$, was $R_{\text{ex}}(0) \approx 0$ as a rule. The zero resistance critical temperature $T_{\text{c0}}$ of the as-prepared single layer YBCO films reached 90 K. The LSMO single films exhibited a metal-to-insulator transition at temperature $T = 320$ K, indicating ferromagnetic properties at room temperature. The resistivity of the LSMO single layers at 77 K was $\rho_{\text{LSMO}} \approx 300 \mu\Omega$ cm, while the resistivity of the YBCO single layers just above the transition temperature was $\rho_{\text{YBCO}} \approx 40 \mu\Omega$ cm (figure 3).

The YBCO/LSMO bilayers, patterned by argon ion beam etching into 5-µm wide strips, had a zero resistance critical temperature of about 84.5 K; the width of the phase transition was $\Delta T_{\text{c}} < 1$ K. To realize nanostrips of length 5 µm and width below 0.5 µm (figure 2, without the cross line), we applied the FIB technique (using 30 keV focused Ga ions and current in the range of 10 pA). Strong deterioration of the YBCO film properties after FIB application was observed if the bilayer was not covered by a protecting film (figure 4). Covering the bilayer by a 50-nm thick gold layer was sufficient to suppress the impact of the focused Ga ion beam (figure 5).

![Figure 3. Normalized $R$-$T$ dependences of YBCO and LSMO single layers, both with good electrical parameters.](image)

![Figure 4. Normalized $R$-$T$ dependences of the YBCO/LSMO bilayer before and after etching by FIB when the microstrip was not protected by Au.](image)

To complete the SFS structure of lateral geometry (figure 1), cross-lines of length $L \leq 100$ nm were realized. The final aim of these attempts was to remove completely the YBCO film in the cross-line (figure 2, inset) to observe the LRPE in the SFS structure. This step is crucial as the local removal of YBCO is extremely difficult without removing the very thin LSMO film beneath the YBCO). This is the most critical step of the SFS sample preparation, because during the sample irradiation by 30 keV Ga$^3$ ions, the properties of superconductor and manganite films can be strongly affected. We applied this procedure on the 28 microstrip ($5 \times 5$ µm$^2$) motives on 1 cm$^2$ of STO substrate. After calibration of the FIB etching time (from the microstrip resistance measurement during the FIB application), it was possible to adjust the time for weak link preparation. Figure 6 shows the $R$-$T$ dependence of the SFS junction with critical temperature of the nanostrip $T_{\text{c0n}} > 76$ K, resistance of the weak link plateau $R \approx 4$ Ω and critical temperature of the weak link $T_{\text{cWL}} \approx 57$ K. In addition, the $R$-$T$ dependence of the structure above the microstrip critical temperature is similar to the LSMO $R$-$T$ dependence, in agreement with the result in [7]. Bearing in mind the estimated flux-flow type $I$-$V$ characteristics and the weak dependence of the junction critical current on the magnetic field, it is not clear whether the coupling of the YBCO through LSMO of length $L < 100$ nm is not the result of a residual YBCO film in the cross-line (the critical current density of the SFS structure is in the range of MA/cm$^2$) or indicates the presence of LRPE. Clarifying this will be the aim of following studies. The important
output of these preliminary results is that applying FIB to a chosen SFS motive did not affect the properties of the other 27 motives located on the STO substrate when the sample was covered by a 50 nm thick gold layer.

**Figure 5.** $R$-$T$ dependences of microstrip before and after FIB application when the microstrip was covered by a 50-nm thick Au film.

**Figure 6.** $R$-$T$ dependence of the SFS junction with resistance plateau ($R \approx 4 \ \Omega$) of the junction weak link part and $T_{cWL} \approx 57 \ \text{K}$.

**Conclusions**

The realization of high quality superconducting weak links or Josephson junctions based on high-$T_c$ superconductors is at present a very difficult technical task. Progress is expected using advanced microcircuit technologies (e.g. FIB) for preparation of nanometer-size SNS or SFS structures. The SFS heterostructures, in addition, offer the opportunity of new physical effects, LRPE, as well as new modes of operation in cryoelectronic or superconducting spintronic circuits. We presented preliminary results on the high-$T_c$ superconducting SFS structure in the so-called lateral geometry. We showed that FIB is a suitable procedure for implementing nanometer size devices, while the problems of films degradation due to FIB irradiation may be solved by protecting the structure by a gold layer with thickness about 50 nm.

**Acknowledgement**

This work was supported by the Slovak Grant Agency VEGA under projects No. 2/0144/10 and 2/0164/11 and by the European Social Fund under the project BG051PO001.3.3.04/54/2009.

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