Superconductor Ferromagnet bilayers; Experimental investigation of spin polarized transport across the interface

S.K. Wanchoo1, John J.2, V.C. Bagwe2, S. P. Pai2, A. M. Narsale3 and R. Pinto4

1School of Applied Physics & Mathematics, SMVD University, Katra, India-182 320
2Tata Institute of Fundamental Research, Mumbai-400 005
3WRIC, University of Mumbai, Mumbai – 400 098
4Department of Electrical Engineering, IIT Bombay, Mumbai - 400076.

E-mail: sunilkwanchoo@gmail.com

Abstract. Deposition of high temperature superconducting thin films by pulsed laser deposition (PLD) started a flurry of activities in the area of rare earth class of oxide manganites. Studies have been carried out by injection of the polarized spins from a ferromagnetic metal, into the superconductor via a thin insulating barrier. However because of the presence of an insulating layer possible role of joule heating in limiting the spin polarized transport across the interface cannot be ruled out. We would like to present some of the results of our work carried out on LaAlO3/La0.7Ca0.3MnO3/YBa2Cu3O7−δ (LAO/LCMO/YBCO) LCMO on top and LaAlO3/YBa2Cu3O7−δ/La0.7Ca0.3MnO3 (LAO/YBCO/LCMO) YBCO on top heterostructures deposited on <100> LAO substrates without any insulating layer separating the two. PLD technique was used to deposit a 500 micron wide line of LCMO followed by deposition of YBCO layer which was subsequently patterned into a 500 micron line directly on top of the LCMO line. Here we report the suppression in the critical current \( I_c \) of the superconductor due to pair-breaking phenomenon when polarized spins are injected into YBCO in case of LAO/LCMO/YBCO (LCMO on top) sample. Surface and interface characterization of these bilayers was performed by SQUID, AFM, SIMS and four probe techniques.

1. Introduction:

The coexistence of superconductivity and ferromagnetism is an interesting problem in physics which has attracted the attention of many workers. After the discovery of high temperature superconductors and their realization in thin film form, current interest is focused to look out for materials compatible with them. Deposition of high temperature superconducting thin films by pulsed laser ablation started a flurry of activities in the area of rare earth class of oxide manganites as the same are structurally compatible with the high temperature superconductors. These materials, due to their half-metallic nature, exhibit spin polarization close to unity and thus can act as reservoirs of polarized spins. These manganites poses a lot of promise for high temperature superconductor based devices involving spin dependent transport of the charge carriers. Devices patterned out of multilayered heterostructures provide considerable potential for applications. The results of the recent studies have put forward the idea that spin polarized electronic transport bringing about suppression of superconductivity[1]. These studies have been motivated by the idea of having three terminal superconducting devices[2] based on polarized quasiparticle injection. It is clear that a study of spin-polarized electron transport from ferromagnetic materials to superconductors is important for the understanding of spin dependent electronic properties and for the realization of possible spin-injection devices.

A number of reports[3,4] involving spin dependent transport in case of low temperature superconductors have been reported in literature. Similar studies in case of high temperature superconductors have also been reported[5,6]. These devices are expected to work at very high speeds. Various studies on suppression of the superconductivity[5-7] by the injection of polarized spins have been phenomenal.
Various devices have been proposed and the experimental data pertaining to these studies has been widely published[6-9] These devices make use of the electron’s spin apart from its charge. Almost all these studies are based on trilayer structures of the form, ferromagnet-insulator-superconductor (F-I-S). These studies have been carried out by injecting polarized spins from a ferromagnetic metal into the superconductor via a thin insulating barrier. However because of the presence of an insulating layer possible role of joule heating in limiting the critical currents cannot be ruled out. In this paper we report the synthesis of LAO/LCMO/YBCO (YBCO on top) & LAO/YBCO/LCMO (LCMO on top) heterostructures using a pulsed laser deposition system. We have directly deposited the two layers on each other to avoid any joule heating due to the intentionally deposited insulating thin film separating the two.

2 Experimental Details

The bulk targets of YBCO and LCMO used for the present work have been synthesized via the standard solid-state reaction route. Pulsed laser deposition system was used to grow the LAO/LCMO/YBCO (YBCO on top) & LAO/YBCO/LCMO (LCMO on top) heterostructure on LAO substrates. An excimer laser was used to grow these samples with a wavelength of 248 nm and a repletion rate of 10 Hz. Both the targets were mounted on the target holders which could be rotated using automated motor control. We have used almost identical deposition conditions (after optimizing the same) for the in situ growth of both the two oxide layers. These heterostructures were deposited at a substrate temperature of 800°C. Oxygen partial pressure of 400 mTorr for LCMO and 250 mTorr for YBCO were maintained during their growth respectively. The superconducting transition temperature was established by using a homemade ac susceptibility measurement setup. We have probed the surface morphology of as grown thin films using atomic force microscope. The phase pure nature of the films was established by powder
x-ray diffraction technique. To carry out the resistivity measurements using a homemade resistivity measurement setup, films were patterned using UV photolithography technique. Contact pads of gold were patterned using the lithography technique after depositing a gold film on the sample by PLD. Another sister sample, which was grown in the same run, was used to carry out the resistivity measurements on the bottom LCMO layer. The interface of the LAO/LCMO/YBCO (YBCO on top) & LAO/YBCO/LCMO (LCMO on top) heterostructures was characterized using secondary ion mass spectroscopy technique (SIMS).

In order to establish the interface quality a depth profiling was carried out using a Cameca 34f SIMS machine. In this measurement $O_2^+$ was used for ionization at a current of 50nA and at an effective incidence angle of $42^\circ$. Finally an analysis zone of 33 micron diameter was used in the center of the erosion zone of 150 micron x 150 micron to carry out the depth profiling.

Further, in order to investigate the effect of the spin polarized current on the critical current of a superconductor another experiment was performed. This device was synthesized by first depositing a LCMO layer (1000 Å thick) in the form of a thin line (500 micron wide) on LAO substrate. This thin line was deposited on a single crystalline (001) oriented LAO substrate. The optimized deposition conditions were used for the growth of the LCMO line. The line was deposited using two other LAO substrates to cover the substrate as a substrate-mask. The substrate-mask was then dropped in situ by a mechanical arrangement (In situ substrate mask drop technique) as shown in figure 1. After dropping the substrate-mask, a YBCO layer (2000 Å thick) was deposited in situ on the LAO substrate. Then a 500 micron wide line of YBCO was patterned out of the YBCO layer deposited in situ using the PLD technique. The YBCO layer was patterned using the UV photolithography technique. In this case independent currents, perpendicular to each other, were passed through the LCMO and YBCO layers respectively. Figure 2 shows the connection scheme used for the measurements. $S_1$ and $S_2$ denote the two current sources used to send current through the LCMO and YBCO layers respectively. $V$ denotes the voltmeter used to determine the $I_c$ of the YBCO layer. $S_1$ was connected between 5 and 6 and $S_2$ was connected between 1 and 4 respectively. The voltmeter $V$ was connected between 2 and 3.

3 Results and Discussion

XRD results of the bilayer grown by pulsed laser deposition technique showed that most of the reflections are along the (001) direction suggesting the epitaxial c axis oriented growth of the both the layers. Figure 3 shows the AFM images of the constituent layers of the LAO/LCMO/YBCO (YBCO on top) sample. The AFM micrograph reveals the granular and morphologic nature of the growth. Figure 4 shows the temperature dependence of resistance for LAO/LCMO/YBCO (YBCO on top) sample. It can be seen from the graph that the metal – insulator transition temperature ($T_p$) of the LCMO layer is 250K.

Figure 3. Atomic force microscope micrographs LAO/LCMO/YBCO (YBCO on top) (a) a bilayer and (b) single LCMO layer.
At 250K LCMO layer shows a clear MI transition while as at 85 K since the resistance of the YBCO microbridge drops to zero and the same conclusively indicates the presence of superconductivity in the microbridge. Hence the four-probe measurements clearly indicate that the LAO/LCMO/YBCO (YBCO on top) sample essentially displays both the properties in a single sample.

The four-probe data though does not indicate any change in $T_p$ of LCMO as the same is found to be equal to that of a single layer of LCMO, which was grown for comparison. However the superconducting transition temperature $T_c$ of YBCO does show a decrease from standard 90K to 85 K. In case of LAO/YBCO/LCMO (LCMO on top) sample we do not see any superconducting transition in the fourprobe measurements carried out on the sample. However the top LCMO layer does show a clear metal insulator transition at 250K. The most obvious reason for this seems to be the oxygen deficiency in the bottom YBCO layer. The YBCO layer is not able to get properly oxygenated during the venting of the deposition chamber after the growth of the two layers in situ as it is fully covered by the top LCMO layer. In order to probe this top LCMO layer was chemically etched. The sample was then subjected to 3 hrs annealing at 650°C with oxygen flowing in the furnace. Resistivity measurements were then performed on the bottom YBCO layer. The YBCO layer showed a $T_c$ of 88 K which clearly indicates that the film was oxygen deficient.

Figure 6 (a) shows the $M$ Vs $H$ hysteresis loop for LAO/LCMO/YBCO sample was carried out at 40K (well below the $T_c$ of YBCO). The plot clearly demonstrates the excellent display of superconducting ordering in these bilayers. Figure 6 (b) shows the temperature dependence of magnetic moment for the said sample under the FC and ZFC conditions. Once again the plot shows perfect diamagnetic behaviour below 85K (same $T_c$ as was obtained in from the fourprobe measurements). This clearly indicates that the said sample shows excellent diamagnetic behaviour. In order to probe the decrease in $T_c$ we have carried out secondary ion mass spectroscopy (SIMS) studies on the two types of samples to characterize the sharpness of the interface between LCMO and YBCO. Figure shows the SIMS profiles for
LAO/LCMO/YBCO (YBCO on top) & LAO/YBCO/LCMO (LCMO on top) heterostructures. The interface quality depends primarily on the surface roughness of the bottom layer and on the possible inter-diffusion of cations at the interface during the growth of top layer. Atomic force microscopy showed the LCMO roughness (determined partly due to the twinned LAO substrate) to be ~200 Å. We evaluated the cationic inter-diffusion using SIMS. Shown in figure 7 are the SIMS profiles of (a) LAO/YBCO/LCMO (LCMO on top) & (b) LAO/LCMO/YBCO (YBCO on top) heterostructures. If we account ~200 Å due to surface roughness in the ~400 Å inter-diffused layer (SIMS), we see only a nominal inter-diffusion of ~100 Å on each side of the junction, except for diffusion of Cu into the LCMO layer in case of LAO/LCMO/YBCO (YBCO

Figure 6. (a) M-H hysteresis loop recorded for LCMO/YBCO heterostructure at 40 K. (b) M vs T curve recorded for LCMO/YBCO heterostructure at a magnetic field of 500 Oe.

Figure 7. (a) SIMS profiles of a typical LAO/YBCO/LCMO (LCMO on top) sample. The full scale of the x-axis corresponds to 2757 Å. (1820 s in the time axis correspond to an etch crater depth of 2537 Å). (b) LAO/LCMO/YBCO (YBCO on top) sample. The full scale of the x-axis corresponds to 3954 Å. (2150 s in the time axis correspond to an etch crater depth of 3416 Å)
on top) sample. Longer diffusion of Cu into the LCMO layer does not appear to affect the spin carrier density as the $T_p$ was found to be 250 K. This clearly indicates that the minimal diffusion of Cu in case of LAO/LCMO/YBCO (YBCO on top) sample does not affect the properties of LCMO. We believe that the inter-diffused Cu may be sitting at the grain boundaries and does not have any impact on the LCMO properties.

When we probed the LAO/YBCO/LCMO (LCMO on top) sample we could not find any Cu diffusion into the LCMO layer. The interface is sharp and clean in this case however the problem of deoxygenating of the bottom YBCO layer remains a major concern. Therefore it is clear that due to the loss of Cu and also due to a 100 Å inter-diffused layer, the $T_c$ of YBCO shows a decrease of 5 K (in case of LAO/LCMO/YBCO (YBCO on top) sample). When the LCMO is deposited on top we do not see any superconducting transition in the bilayer.

In case of the device as shown in figure 2 (LAO/LCMO/YBCO (YBCO on top)) initially a sensing current of 10 µA just enough for the four-probe measurements was passed through the YBCO line and the LCMO current was progressively increased. For a particular value of LCMO current ($I_m$), the YBCO line became normal (lost superconductivity) even when only 10 µA current was flowing through the YBCO line. To investigate this further, the YBCO current ($I_L$) was also increased in small steps. It was observed that the YBCO line lost superconductivity at progressively lesser LCMO currents. The observed phenomenon was independent of the direction of the injection current ($I_m$) which is clearly visible in figure 8. Figure shows the plot of $I_m$ vs $I_L$ at 77K and 85K respectively. This result leads to an interesting device application as a “superconducting switch”. For relatively large current (less than $I_L$) flowing through the superconductor (YBCO), even a small current through LCMO ($I_m$) is enough to cause a voltage drop across the superconductor and kill the superconductivity.

4 Conclusion

In conclusion, we have fabricated LAO/LCMO/YBCO (YBCO on top) & LAO/YBCO/LCMO (LCMO on top) heterostructures in situ by PLD on LAO substrates. Measurements showed that $T_c$ of YBCO microbridge on top of LCMO layer is equal to 85 K. $T_p$ of the bottom LCMO layer was found to be 250 K. Contribution of ~100 Å due to surface roughness (as seen by AFM) and ~100 Å due to inter-diffusion (as verified by SIMS) does not seem to affect the material properties of LCMO. The LCMO-YBCO heterostructure displays both superconductivity and ferromagnetism simultaneously. On probing the YBCO-LCMO heterostructures we observed that the bilayer shows excellent ferromagnetic properties with a $T_p$ of 250K however there is now superconducting transition which could be observed. On analyzing the heterostructure with SIMS no cationic interdiffusion could be observed. In case of LAO/YBCO/LCMO (LCMO on top) after etching the top LCMO layer and annealing the bottom YBCO layer in flowing oxygen a $T_c$ of 88K could be
observed indicating that the YBCO layer was oxygen deficient. An interesting switching experiment was performed and it was observed that the YBCO line lost superconductivity at progressively lesser LCMO currents. In conclusion suppression of superconductivity due to the injection of polarized spins from LCMO into YBCO is successfully demonstrated.

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