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Growth and characterization of superconductor-ferromagnet thin film heterostructure $La_{1.85}Sr_{0.15}CuO_4/SrRuO_3$

Youngdo Kim[a,b], Byungmin Sohn[a,b], and Changyoung Kim[a,b,*]

[a] *Center for Correlated Electron Systems, Institute for Basic Science, Seoul 08826, Korea*
[b] *Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea*

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Abstract

Superconductor-ferromagnet thin film heterostructure is an ideal system for studying the interplay between superconductivity and ferromagnetism. These two antagonistic properties combined in thin film heterostructure create interesting proximity effects such as spin-triplet superconductivity. Thin film heterostructure of optimally doped $La_{2-x}Sr_xCuO_4$(LSCO) cuprate superconductor and $SrRuO_3$(SRO) ruthenate ferromagnet has been grown by pulsed laser deposition. Its temperature-dependent resistivity and Hall effect measurements show that our LSCO/SRO heterostructure has both superconductivity and ferromagnetism. In the Hall effect measurement results, we find additional hump-like structures appear in the anomalous Hall effect signal in the vicinity of superconducting transition. We conclude that giant magnetoresistance of the LSCO layer distorts the AHE signal, which results in a hump-like structure.

Keywords: superconductor-ferromagnet heterostructure, $La_{2-x}Sr_xCuO_4$ (LSCO), $SrRuO_3$(SRO), pulsed laser deposition(PLD)

1. INTRODUCTION

The interplay between superconductivity (SC) and ferromagnetism (FM) can induce novel phenomena such as spin-triplet SC[1–3] or Majorana bound states[4]. These exotic properties are usually studied and observed in artificially constructed superconductor-ferromagnet (S-F) thin film heterostructures. Furthermore, SC-FM interaction in S-F heterostructures is the key to dissipationless spintronics devices[5].

Generally, SC and FM have antagonistic relation because most of the known superconductors have spin-singlet Cooper pairing, consisting of two electrons with opposite spins - singlet Cooper pair cannot survive in the high exchange field of ferromagnet layer. However, since F.S. Bergeret *et al.*[1] proposed the possibility of the formation of triplet Cooper pairs in the S-F interface, numerous works have been performed to observe triplet Cooper pairs in S-F heterostructures.

S-F-S ferromagnetic Josephson junction is the most used system in the search for triplet SC. Triplet supercurrent through long-range ferromagnetic barrier between two S layers, which is an evidence for triplet SC, has been reported in several experiments[6–11]. Multilayers of high-T_c cuprates and ferromagnetic oxide perovskites are also drawing attention for the study of novel SC-FM interaction phenomena in S-F heterostructure systems. Suppression of both SC and FM[12], giant magnetoresistance effect[13], and antiferromagnetic long-range coupling[14] have been reported in various oxide S-F systems.

In order to expand the boundary of the material choices for novel S-F heterostructure studies, we have grown oxide S-F heterostructures of $La_{2-x}Sr_xCuO_4$ (LSCO) high-T_c cuprate for S layer and $SrRuO_3$ (SRO) ruthenate for F layer. LSCO has a simple K_2NiF_4 crystal structure which makes it suitable for building heterostructures with other oxide perovskites. In addition, hole doping of LSCO is easily controlled via tuning of Sr concentration *x*. SRO is an itinerant ferromagnet with a cubic oxide perovskite structure. SRO remains ferromagnetic in the ultrathin regime, down to the thickness of 3 unit-cell (UC)[15].

In this paper, we demonstrate the growth of the LSCO/SRO thin film heterostructure system with pulsed laser deposition (PLD) technique. We show via electrical transport measurements that our LSCO/SRO thin film heterostructure has both FM and SC.

2. METHODS

LSCO/SRO thin films were deposited on (001)-oriented $SrTiO_3$ (STO) substrates in a PLD system (Pascal Co., Ltd.). STO substrates were treated with deionized water and annealed at a temperature of 1070 ℃ before the growth. Stoichiometric SRO and $La_{1.85}Sr_{0.15}CuO_4$ polycrystalline targets were used for deposition, ablated with Coherent KrF excimer laser (λ=248 nm). Laser intensity was 2.0 J/cm^2 for SRO and 0.5 J/cm^2 for LSCO, both with a repetition rate of 2 Hz. During the deposition, the substrate temperature was kept at 670 ℃ for SRO and 600 ℃ for LSCO. All deposition processes were performed in an oxygen partial pressure of PO_2 = 100 mTorr.

The thin film growth was monitored with an in-situ reflection high energy electron diffraction (RHEED) system. Both growth mode and growth rate were determined by monitoring Bragg peak intensity oscillation.

* Corresponding author: changyoung@snu.ac.kr

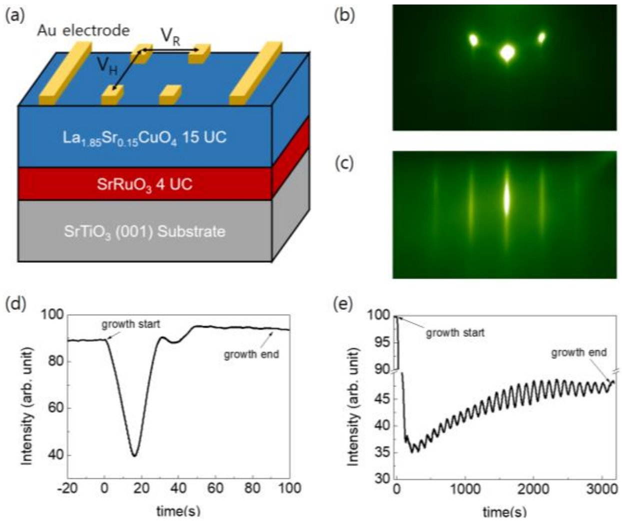

Fig. 1. (a) Schematic illustration of 15 UC LSCO/4 UC SRO heterostructure (b)(c) RHEED image after growth of SRO layer and LSCO layer (d)(e) RHEED intensity oscillation of SRO and LSCO layer growth..

After the growth, Au electrodes with Hall bar geometry were ex-situ deposited on top of LSCO/SRO with an electron beam evaporator system for electrical transport measurements. Temperature-dependent resistivity and Hall effect were measured by using Quantum Design Physical Property Measurement System.

3. RESULTS

15 UC LSCO/4 UC SRO thin film heterostructure was grown on STO substrates. For each growth, the film surface was monitored by RHEED and the thickness was determined based on the RHEED intensity oscillation. RHEED images after growth of SRO and LSCO layers are shown in Fig. 1(b)(c). Both RHEED patterns of SRO and LSCO layers indicate flat surfaces, especially so for SRO with single crystalline surface considering the spot pattern. RHEED intensity is measured at the 00 Bragg peak, and is also plotted in Fig. 1(d)(e). RHEED oscillation pattern shows that the growth of the SRO layer is in step-flow mode while that of the LSCO layer is in a layer-by-layer mode.

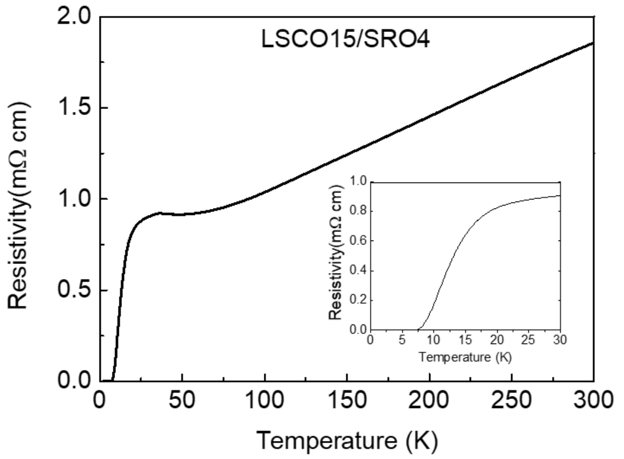

Fig. 3. Temperature-dependent resistivity of 15 UC LSCO/4 UC SRO.

Temperature-dependent resistivity and Hall effect measurements were performed by depositing Au electrodes on the LSCO/SRO heterostructure. Fig. 2 shows the result of temperature-dependent resistivity measurements. The T_c onset is about 20 K, and resistivity becomes zero at around 8 K (see the inset). The reduction of T_c compared to the optimally doped LSCO single crystal from 36 K to 8 K may be due to the strain effect from the STO substrate[16, 17] and also to existence of FM in a close proximity. Furthermore, the broad superconducting transition may be attributed to the effect of the

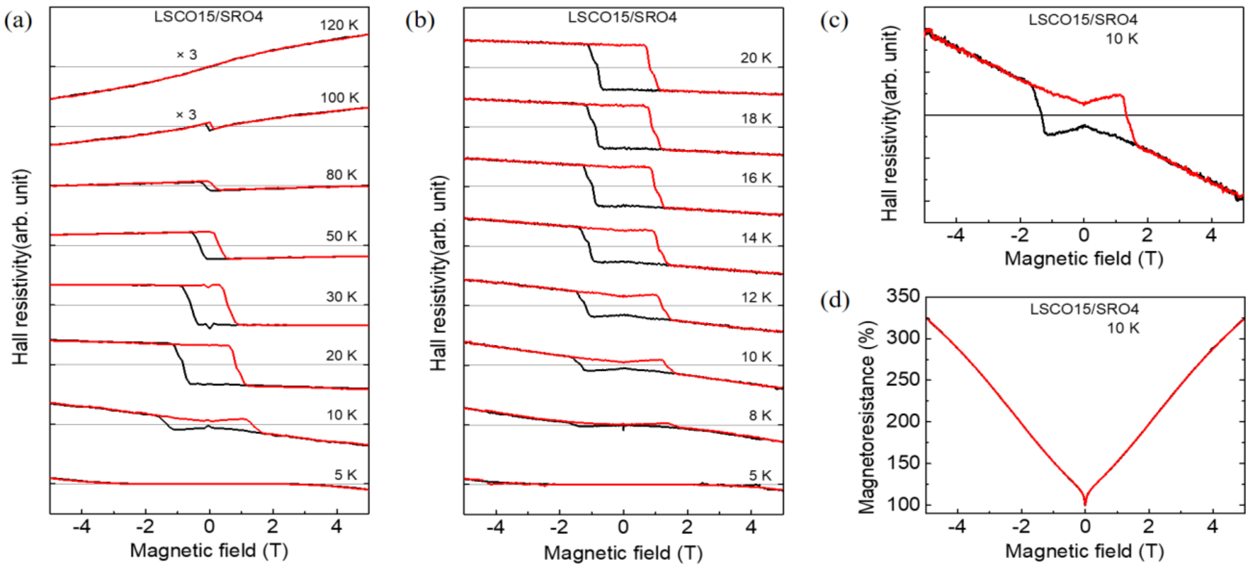

Fig. 2. Red: up-sweep, Black: down-sweep (a) Temperature-dependent Hall resistivity (5 K – 120 K). (b) Temperature-dependent Hall resistivity in the vicinity of superconducting transition (5 K – 20 K). (c) Hall resistivity at 10 K. (d) Magnetoresistance at 10 K, normalized to the value at 0 T.

inhomogeneity of the film. Large in-plane lattice mismatch and different unit-cell height between SRO/STO and LSCO cause disorders which create low-T_c domain in the film.

In order to investigate FM properties, we performed temperature-dependent Hall measurements and the results are plotted in Fig. 3(a) and 3(b). The field was applied in the out-of-plane direction which is the easy axis of the SRO film. Anomalous Hall effect (AHE) which comes from the momentum space Berry curvature has been used to study the magnetic state of ferromagnetic materials, including ferromagnetic SRO thin films[15, 18]. Existence of AHE can be interpreted as a sign of FM.

As the temperature increases, the AHE vanishes at a temperature between 100 to 120 K. Conversely, as the temperature decreases, the AHE signal and coercive field increase until the temperature reaches 20 K. Below the superconducting onset temperature of 20 K, the SC of the LSCO layer starts to affect the AHE. We assumed our LSCO/SRO heterostructure as a system with LSCO and SRO layers connected in parallel. In such a parallel-connected system, the current flowing through each layer is proportional to the reciprocal of the resistance of that layer. As the LSCO layer enters the superconducting phase, the current dominantly flows through LSCO and that through the SRO layer diminishes. Consequently, the magnitude of the AHE which requires current through SRO is also reduced. AHE disappears completely in zero resistivity regime below 5 K. The SC of the LSCO appears to only affect the current through the SRO, not the SRO's FM itself. Although the magnitude of AHE signal decreases, the coercive field of the SRO keeps increasing when the system enters the superconducting regime.

In the vicinity of superconducting transition, hump-like structures arise in the Hall effect signal. As shown in Fig. 3(c), the hump begins with changing field direction and continues to rise until the field reaches the coercive field. We find that these humps are caused by giant magnetoresistance (MR) of the LSCO layer resulting from the suppression of the SC by the magnetic field. MR of LSCO/SRO at 10 K is about 325% at 5 T magnetic field, as shown in Fig. 3(d). As the field increases, the resistivity of the LSCO layer increases rapidly while that of the SRO layer hardly changes. Therefore, more current flows to the SRO layer and, as a result, the AHE signal increases. So the AHE signal becomes minimum at zero magnetic field and maximum at the coercive field. This effect distorts the ordinary AHE signal to a ribbon-shape hysteresis curve, which looks like a hump structure.

We have investigated the transport properties of F-S heterostructure, especially near the superconducting transition. However, many of the transport signals vanish in the full superconducting regime as the resistivity vanishes while full SC is critical to investigate SC-FM proximity effects. Therefore, we plan to use angle-resolved photoemission spectroscopy in future studies to probe the electronic structure of the S-F heterostructure.

4. CONCLUSION

In conclusion, an S-F heterostructure with 15 UC of LSCO and 4 UC of SRO was grown by PLD. In-situ RHEED patterns show that the growth process is epitaxial and that the film has a flat surface. Both SC and FM exist in the heterostructure. Superconducting onset temperature was about 20 K and the critical temperature with zero resistivity was found to be 8 K. Anomalous Hall effect signal was visible below 100 K. Between the superconducting onset temperature of 20 K and the zero-resistivity temperature of 8 K, hump-like structures arise in the AHE signal. We conclude this is an effect of giant magnetoresistance in the LSCO layer.

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