Fabrication of hybrid thin film structures from HTS and CMR materials

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Abstract. We present the preparation of bilayers from high-temperature superconductors (HTS) and half-metallic ferromagnetic (FM) manganite with a colossal magnetoresistance (CMR). We used YBa2Cu3O7-x (YBCO) and Tl2Ba2CaCu2O8-y (TBCCO) thin films as a HTS material and La0.67Sr0.33MnO3 (LSMO) film as a CMR material. In the case of YBCO/LSMO, we prepared FM/HTS heterostructure for studying the spin-polarized current injection effect on the electrical properties of the YBCO strip in dc or low-frequency regimes and on the microwave characteristics of the strip. For the first time, we report the preparation of a TBCCO/LSMO bilayer. In some applications, the TBCCO offers better parameters (higher working temperature, lower surface resistance, lower 1/f noise) than YBCO.

1. Introduction
Superconducting spintronic devices use the spin-dependent properties of superconductors in superconductor (SC)/ferromagnet (FM) heterostructures. The SC/FM heterostructures are of considerable practical importance. Layered heterostructures could be used for tunable magnetic microwave devices, for dynamic modification of SC critical parameters and for spin-polarized injection [1-3]. The perovskite high Tc superconductors (HTS) and the doped rare-earth manganese perovskite oxides L_{1-x}A_xMnO_3 (L = La, Pr…, A = Ca, Sr, Pb…) are promising candidates for preparing such heterostructures. These two perovskites are also interesting because of their ability to form high-quality epitaxial multilayers with each other and they can grow on conventional monocrystalline substrates, such as SrTiO_3 (STO), LaAlO_3 (LAO), La_{0.26}Sr_{0.74}Al_{0.61}Ta_{0.37}O_3 (LSAT), MgO and sapphire [4-7].

In this work, we describe the preparation of two types of HTS/FM bilayers, namely YBa2Cu3O7-x (YBCO)/La_{0.67}Sr_{0.33}MnO3 (LSMO) and Tl2Ba2CaCu2O8-y (TBCCO)/LSMO. A standard photolithography process, wet etching and lift-off techniques were used to prepare the YBCO/LSMO heterostructures. The basic electrical characterization of the layers and structures are presented. The

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motivation to use TBCCO layer is a higher critical temperature \( T_c \) (a higher operating temperature), a lower surface resistance \( R_s \) at microwave frequencies and, at a given temperature, lower \( 1/f \) noise caused by flux motion compared to the YBCO film [8].

2. Experimental

The YBCO and LSMO layers were prepared by laser pulsed deposition (PLD) by the same procedure. A KrF\(^+\) excimer laser operating at 248 nm with a pulse width of 20 ns, repetition rate of 10 Hz and energy density of 6 J/cm\(^2\) (spot size on the target \( \approx 2 \) mm\(^2\)) was used to grow the YBCO (LSMO) films. A temperature of the substrate holder of 850 °C and an oxygen pressure of 53 Pa were set during the deposition. After the deposition, the films were cooled down at a rate of 20 °C/min in \( \text{O}_2 \) (4x10\(^4\) Pa). The thickness of each LSMO and YBCO film on the LAO or MgO substrates was 50 nm. The growth rate of the prepared by laser ablation YBCO and LSMO was about 6.5 nm/min.

The LSMO films were deposited also using an on-axis dc commercial magnetron sputtering system (Lesker, TORUS 2C) from a stoichiometric ceramic target onto one-side polished MgO (0 0 1). The deposition was performed in an Ar (80%) + \( \text{O}_2 \) (20%) atmosphere at a total pressure of 30 Pa. During the deposition, the substrate was heated to 800 °C. The thickness and the growth rate of the LSMO films were about 500 nm and 8 nm/min, respectively. In order to increase the oxygen content, some LSMO films were subsequently annealed \textit{in-situ} in \( \text{O}_2 \) (10\(^4\) Pa) at 800 °C for an hour. After annealing, the temperature was lowered down to room temperature at a rate of 4 K/min [9].

The 
\[ \text{Ti}_2\text{Ba}_2\text{CaCu}_2\text{O}_{8+y} \]
thin films were prepared by a two-step process. In the first step, 300-nm thick precursor films with a nominal composition of \( \text{Ba}:\text{Ca}:\text{Cu} = 2:2:3 \) were deposited by RF magnetron sputtering from a single target of the same stoichiometry. The films were sputtered at a total pressure of 10 Pa in an Ar atmosphere at room temperature with a deposition rate of 0.3 nm/s corresponding to an RF power of about 100 W. The thallination of the precursor films (the second step) was at annealing temperature of 850 – 860 °C for 45 minutes in oxygen flow atmosphere [7].

The crystallographic structure and the growth orientation of the obtained films were examined by X-ray diffraction (XRD) using a BRUKER AXS D8 DISCOVER diffractometer with rotating Cu anode (CuK\( _\alpha \)). Scanning electron microscopy (SEM) was performed using FEI Inspect F50. The basic electrical characterization of the bilayers and structures were performed by the dc 4-point and a low-frequency inductive (contactless) methods.

The sketch of the HTS/FM heterostructure for studying the spin-polarized current injection effect from FM into HTS is shown in figure 1. The parallel FM strips are connected by a sinusoidal (meander) strip of HTS. The as-prepared ferromagnetic LSMO thin films were patterned using standard photolithography processes and etched by a solution of potassium iodide, hydrochloric and ascorbic acid [7].

Subsequently, a superconducting YBCO thin film was deposited on top of the patterned LSMO using PLD. Then the standard photolithography processes and wet etching (0.1 % solution of nitric acid) were used to pattern the YBCO meander. Several bilayer samples consisting of a HTS YBCO strip and FM LSMO electrodes were prepared to study the spin-polarized current injection effect (from the FM LSMO) on the electrical and microwave properties of the YBCO strip. The strip was prepared in the form of a meander to increase its geometrical length and to decrease its resonance frequency in measurements in the regime of microstrip resonator. In this configuration, the sample can be used to study the influence of the current injection effect on the microwave surface impedance in the YBCO structure. The sample was designed to be also suitable for investigation of the effect in low frequency or dc...
regimes. In this case, injection of the spin-oriented current from the LSMO into the YBCO could affect the critical temperature of the YBCO strip and the critical current density. The normal resistance of the YBCO strip in vicinity of $T_C$ (where a pseudogap exists) could be expected to be somewhat affected by the current injection process as well.

3. Results and discussions

The high-quality LSMO thin films of thickness about 50 nm were deposited on LAO or MgO single crystal substrates by PLD. The crystal structure of the LSMO films grown on the MgO (0 0 1) substrate was investigated by X-ray diffraction techniques $\theta$-2$\theta$ scans, $\omega$-scans and $\varphi$-scans. The $\theta$-2$\theta$ scan of the LSMO films indicated a preferred (0 0 l) oriented film growth. The in-plane growth properties were followed using $\varphi$-scans of the LSMO (4 0 2) and MgO (4 0 2) planes. We observed four sharp maxima indicating the four-fold symmetry of the LSMO lattice. Our XRD analyses confirmed that the LSMO grown on the MgO substrate fulfills the epitaxial relation LSMO (0 0 1)$_c//MgO$ (0 0 l)$_c//MgO$ (0 0 l)$_c//MgO$ [1 0 0] [10,11].

The resistivity vs. temperature ($\rho$–$T$) dependencies of the LSMO film and the LSMO strip are shown in figure 2. The typical feature of the dependence is a broad resistivity peak with a resistivity maximum at a temperature $T_{MI}$, which corresponds to the metal-insulator transition temperature and is above 400 K. This value is about 40 K higher than the bulk value of the LSMO target ($T_{MIbulk}$ = 374 K) [12]. The resistivity of the film is in this case a little higher ($\rho_{max} \approx 25 \text{ m}\Omega \text{ cm}$) than previously presented [11], but it remains the same after the wet etching as well (figure 2).

The LSMO prepared films exhibited also a high $T_{Curie}$ (Curie temperature) above 400 K [11]; at room temperature about 76 % and at 77 K (expected working temperature of HTS/FM heterostructures) even 97-98 % spin polarization was reached [11,12]. So, the LSMO films are suitable for spin-injection into the HTS film in the HTS/FM heterostructures.

The transition to a superconducting state of the deposited YBCO films exhibiting standard properties is shown in figure 3. The midpoint of the transition, measured by a contactless method, is the same for the film and the patterned YBCO.

![Figure 2](image.png)

**Figure 2.** $\rho$–$T$ dependencies of LSMO films on a MgO substrate before (full line) and after patterning (open squares).

![Figure 3](image.png)

**Figure 3.** Superconducting transitions of YBCO film (open circles) and patterned YBCO meander (full line) deposited on MgO with patterned LSMO.

![Figure 4](image.png)

**Figure 4.** Realized HTS/FM heterostructure.
The onset of this transition corresponds to the zero resistance critical temperature as measured by the 4-point dc method. The realized HTS/FM heterostructure for the spin injection is shown in figure 4. The preliminary results indicate that the resistance of the HTS/FM interface is low ($R < 10^{-4} \, \Omega$) despite the ex-situ process of YBCO deposition. Investigations of the spin-polarized current injection effect on the electrical and microwave characteristics of the YBCO strip are in progress.

Usually, LSMO and YBCO materials are used in HTS/FM heterostructures or, alternatively, a La$_{0.67}$Ca$_{0.33}$MnO$_3$ as FM material. Now, we present the application of a more exotic HTS material, namely, a Tl-based cuprate superconductor. The Tl-based HTS superconductors of several phases exhibit a higher $T_C$ (i.e., a higher operating temperature), a lower surface resistance $R_s$ at microwave frequencies and, at a given temperature, lower 1/f noise caused by flux motion compared to the YBCO film. The XRD analyses ($\theta$-2$\theta$ scan) revealed that the c-axis-oriented grown Tl-based superconductor was single-phase with peaks belonging to the Tl-2212 phase (Tl$_2$Ba$_2$Ca$_2$O$_{8-\delta}$-TBCCO) (figure 5). Besides the Tl-2212 phase, LSMO and MgO substrate peaks could be seen. We note that the LSMO in this case was prepared by dc magnetron sputtering [9], so that $T_{\text{MI}}$ of only about 300 K was achieved. The resistance vs temperature ($R$-$T$) dependence of the TBCCO/LSMO bilayer is shown in figure 6 (full line). This dependence is compared with the $R$-$T$ of the LSMO film (open circles) measured before the growth of the TBCCO layer in the two-step technological process. The resulting $R$-$T$ dependence can be interpreted in terms of the relation $R_C^{-1}(T) = R_{\text{HTS}}^{-1}(T) + R_{\text{FM}}^{-1}(T)$, where $R_{\text{HTS}}$ is the HTS resistance and $R_{\text{FM}}$, the resistance of the ferromagnetic films [13]. The zero resistance critical temperature of the TBCCO layer on LSMO reached about 95 K; therefore, improving the TBCCO layer is necessary.

**Figure 5.** $\theta$-2$\theta$ scan of a TBCCO/LSMO bilayer on a MgO substrate.

**Figure 6.** Dependences of a TBCCO/LSMO bilayer (full line) and a LSMO film before deposition of the TBCCO layer (open circles).

### 4. Conclusions

We prepared two types of bilayers from ferromagnetic manganite and high-temperature superconductors. In the first case, we present the bilayer YBCO/LSMO combination, where LSMO exhibits enhanced parameters – Curie temperature $T_{\text{Curie}}$ as well as temperature of metal-insulator transition $T_{\text{MI}}$, both above 400 K, and, thus, very high spin polarization (about 97-98 %) at 77 K. The wet-etching processes used (developed separately for each layer) allow one to pattern the LSMO and YBCO layers without degradation of their properties. We were able to fabricate YBCO/LSMO heterostructures to investigate the spin-polarized current injection effect on the electrical properties of the YBCO strip in the dc and low frequency regimes and on the microwave characteristics of the strip.
In the latter case, we prepared the bilayer with TBCCO/LSMO composition. We demonstrated the possibility to prepare also another HTS material with a complicated structure and technology of fabrication (the two-step process) on the LSMO film. The use of TBCCO films is convenient for HTS/FM bilayer applications where the TBCCO has better parameters than the YBCO.

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