Properties of LSMO/YBCO cross-strip type junctions

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Abstract. The properties of a La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO)/YBa$_2$Cu$_3$O$_{7-δ}$ (YBCO) interface in thin film LSMO/YBCO cross-strip type junctions were investigated by means of electrical transport measurements. Resistance vs. temperature and current-voltage dependences, as well as conductance spectra, were used to characterize the electrical parameters of the interface. The results indicated a low resistance (below 10 $\Omega$), while the dielectric properties of the interface pointed to the presence of a 10-nm wide and 40-meV high dielectric potential barrier. The oxygen vacancies in both LSMO and YBCO films at the interface and the charge transfer through the interface were both considered to explain the insulating character of the LSMO/YBCO interface.

1. Introduction

Depending on their composition and structural properties, perovskite materials can behave as dielectrics, conventional metals, or high-temperature superconducting (HTS), colossal magnetoresistive (CMR), ferroelectric or multiferroic materials. Therefore, such materials, and especially the CMR/HTS interface, are attractive for studying the interplay between two fundamental condensed-matter phenomena, superconductivity (S) and ferromagnetism (F). Recent developments in fabricating atomically smooth (flat) CMR/HTS interfaces in superlattices [1, 2] provided an ideal system to investigate intriguing phenomena, such as a long-range proximity effect [3, 4], spin-polarized quasiparticle injection into the HTS layer within a spin-diffusion length $\xi_{FM}$ [5], giant modulation of the CMR-layer magnetization induced by superconductivity [6], or charge transfer from CMR into HTS [7,8,9]. A variety of effects and phenomena at the SF interface offer possibilities of using these materials in different microelectronic and spintronic applications [10].

In this work we studied a La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO) / YBa$_2$Cu$_3$O$_{7-δ}$ (YBCO) interface in the form of cross-strip type junction by means of electrical characterization, namely, the resistance vs. temperature ($R(T)$) dependences, and the current-voltage ($I(V)$) and differential conductance vs. voltage ($dI/dV(V)$) characteristics. The results obtained indicate an insulating (dielectric) character of the interface in spite

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of the metallic/superconducting (YBCO) and ferromagnetic half-metallic (LSMO) materials involved. We estimated the width of the barrier at about 10 nm and its height at about 40 meV. We also discuss a possible explanation of the appearance of an insulating barrier in the LSMO/YBCO interface.

2. Experimental
The LSMO and YBCO layers were prepared by pulsed laser deposition (PLD) under the same conditions. A KrF excimer laser was used oscillating at 248 nm, with a pulse width of 20 ns, repetition rate of 10 Hz and energy density of 6 J/cm² (spot size on the target ~2 mm²). During the deposition, the substrate holder temperature was kept at 850 °C and the oxygen pressure, at 53 Pa. After deposition, the films were cooled down at a rate of 20 °C/min in O₃ (4×10⁴ Pa). The growth rates of the LSMO and the YBCO were about 6.5 nm/min. First, a 25-40-nm-thick LSMO film was deposited on a MgO single-crystal substrate. The as-prepared ferromagnetic LSMO thin film was patterned as a base electrode of a junction using standard photolithography processes and etched by a solution of potassium iodide, hydrochloric and ascorbic acid [11]. Subsequently, a superconducting YBCO thin film with a thickness of about 50-70 nm was prepared on the MgO substrate with patterned LSMO. Then, standard photolithography processes and wet etching (0.1 % solution of nitric acid) were used to pattern the YBCO as a counter electrode [11]. A schematic view of the sample with three junctions of different areas on one chip is shown in the inset of figure 1. The area of the junctions varied from 1600 μm² to 15000 μm², corresponding to junction dimensions from 40×40 μm² to 120×120 μm².

The R(T) dependences and I(V) characteristics were measured by the standard 4-probe method, while the conductance spectra (i.e. dI/dV(V) curves) were numerically calculated from the I(V) data.

3. Results and discussions
The R(T) dependences of the LSMO junctions exhibited a “negative” resistance mainly for temperatures above T_C0 (the zero-resistance critical temperature of YBCO) (figure 1). Such a property can arise in junctions when the resistance of the barrier (in our case, the interface resistance R_IF) is lower than the resistances of the adjacent YBCO and LSMO layers (R_S and R_F, respectively). A negative value of the junction resistance R_J measured by the 4-probe method was first reported by Giaever [12]; we recently analyzed one in more detail for a LSMO/YBCO interface [13]. Our analyses showed that in the case of superconducting YBCO (R_S = 0), the junction resistance measured should always be positive. When the resistance of the interface R_IF is small (R_IF ≤ 1 Ω), then the junction resistance measured is positive, but R_J assumes very low values ~ 10⁶ R_IF; when R_IF ≥ 10 Ω, R_J increases and shifts to positive values, closer to R_IF (figure 1).

The R(T) dependences of the adjacent ferromagnetic and superconducting films are presented in figures 2 a and 2 b. In figure 2 a, the R(T) dependences of the as-prepared LSMO film with a patterned LSMO/insulator transition (T_MI = 360 K), which corresponds to the Curie temperature of a paramagnetic-ferromagnetic transition [14,15]. The YBCO strip exhibited the rather low T_C0 value of 84 K (figure 2 b). To investigate the LSMO/YBCO interface, we measured the current-voltage characteristics at temperatures below T_C0, namely at 83 K, 60 K, 40 K, 21 K, 11 K and 4.2 K (figure 3, inset).
The detailed plot of the $I(V)$ characteristic taken at 60 K (figure 3, dotted line) is compared with the tunnel current through a dielectric barrier at low voltage (figure 3, solid line), as derived by Simmons [16] in the form $I = \alpha(V + \gamma V^2)$, where $\alpha$ and $\gamma$ are constants characteristic for the given junction. The estimation of these constants is simple using the differential conductance dependence on the voltage $dI/dV(V)$ (figure 4) [17]. The differential conductance $dI/dV = G(V) = \alpha(1+3\gamma V^2)$ is a parabolic function of the voltage (figure 4). Then, $\alpha$ is the differential conductance at $V = 0$, and $\gamma$ can be estimated from the voltage corresponding to conductance $G(V) = 2\alpha$, according to the relation $V_{2\alpha} = (1/3\gamma)^{1/2}$. Knowing the constants $\alpha$ and $\gamma$, we can solve the next two relations [16, 17]

$$
\gamma \approx \frac{0.0115}{\varphi} t^2 ,
$$

$$
3.16 \times 10^{10} (\varphi^{1/2}/t) \exp(-1.025 \varphi^{1/2}) = \alpha
$$

and estimate important parameters of the barrier, such as the barrier thickness $t$ (in angstroms) and the potential barrier height $\varphi$ (in eV). The same procedure of estimating $t$ and $\varphi$ was applied for all $I(V)$

Figure 2 a. Normalized temperature dependences of the resistance of an as-prepared LSMO film (solid line) and an LSMO strip (dotted line).

Figure 2 b. Temperature dependence of the YBCO strip resistance. The zero-resistance critical temperature $T_{C0}$ is 84 K.

Figure 3. Current-voltage characteristic of the LSMO/YBCO junction taken at 60 K (dots) approximated by the tunnel current according to Simmons [16] (solid line). Set of $I(V)$ curves at various temperatures (inset).

Figure 4. Parabolic differential conductance dependence on the voltage. The $dI/dV(V)$ curve is numerically calculated from the $I(V)$ data in figure 3. The estimates of the junction constants $\alpha$ and $\gamma$ are indicated.
The insulating character of the interface can be explained by at least two effects. First, it is known that the properties of both YBa$_2$Cu$_3$O$_{7-\delta}$ and La$_{1.8}$Sr$_0.2$MnO$_3$ are very sensitive to the oxygen content. For example, if $\delta \gtrsim 0.7$, YBCO exhibits insulating properties [18]; also, if $y \gtrsim 0.02$, La$_{0.85}$Sr$_{0.15}$MnO$_3$ behaves as an insulator [19]. Therefore, some oxygen deficiency at the LSMO/YBCO interface can create an insulating layer acting as a dielectric barrier. Sawa et al. [20] investigated a similar structure of YBCO/LSMO cross-strip type junctions (the opposite placing of YBCO and LSMO layers in a junction); they also found insulating, moreover, ferromagnetic character of the YBCO/LSMO interface. The existence of a ferromagnetic insulator layer was ascribed to oxygen vacancies in the LSMO film at the interface, while the presence of some peculiarities (a zero-bias conductance peak (ZBCP) and a gap-like structure) in the conductivity spectra was accepted as indicating small (or no) degradation of the YBCO layer at the interface. In our case, the LSMO seems to be optimally oxygenated ($T_{MI} = 360$ K), but the YBCO, with $T_{CO}$ of 84 K, indicates a lower degree of oxygenation. Therefore, we assume that the insulating layer at the interface is due to oxygen vacancies created more on the YBCO side, since, besides the suppressed superconductivity in this region, we did not observe any peculiarities, such as a ZBCP or an YBCO gap-structure in the conductivity spectra.

The second effect that should be taken into account is the charge transfer through the CMR/HTS interface ranging in distance from a few tenths of nm to several nm [7, 8, 9]. Gray et al. [7] found a loss of 0.67 electrons per CMR monolayer in their CMR/HTS superlattices, which leads to conversion of the CMR layer to a paramagnetic or antiferromagnetic insulator. On the other hand, the electrons transferred through the CMR/HTS interface dope the Cu and Ba atoms in the YBCO and transform the superconducting YBCO to a Mott insulator. Holden et al. [9] also registered dramatic metallicity suppression at a distance of 10-20 nm from the interface due to a massive charge transfer between the HTS and CMR layers in the CMR/HTS superlattice. In both cases, the charge transfer creates an insulating interface of a thickness close to the one estimated in this paper.

4. Conclusions
We investigated the electrical properties of the interface between high $T_C$ superconductor YBCO and ferromagnetic manganite LSMO thin films in the form of an LSMO/YBCO cross-strip junction. The junctions were prepared on single crystal MgO substrates from LSMO and YBCO thin films deposited ex-situ by PLD. The films were patterned using optical photolithography and wet etching. The junction area ranged from 1600 to 15000 $\mu$m$^2$. The $R(I)$, $I(V)$ and $dI/dV(I)$ dependences of the strips and junctions were studied to characterize the LSMO/YBCO interface. While the $R(I)$ dependences of the individual strips creating a junction exhibited a standard behavior typical for the LSMO and YBCO materials, the junction resistance exhibited ‘negative’ values of the resistance for temperatures above the critical superconducting YBCO temperature. Such values are possible if the junction resistance is smaller than the resistances of the adjacent LSMO and YBCO films. Despite the low junction resistance, both the $I(V)$ dependence and the conductance spectra of the junctions pointed to their insulating character; the interface behaved as a 10-nm wide and 40-meV high potential barrier. The insulating nature of the interface can be explained by oxygen vacancies in the YBCO and LSMO materials at the interface due to an ex-situ process and/or by a charge transfer through the interface creating an insulating layer of paramagnetic LSMO and a Mott insulator on the YBCO side.

Acknowledgement
This work was supported by the Slovak Research and Development Agency under contracts Nos APVV-14-0613 and APVV-16-0315. We acknowledge the grant agency VEGA for the financial support to project No 2/0120/14. This publication is part of the results of the Joint Research Project “Perovskite heterostructures of nanometric thickness for sensors and spintronics” between the Institute of Electronics BAS, Bulgaria and the Institute of Electrical Engineering SAS, Slovakia.
References

[1] Varela M, Lupini R A, Pennycook S J, Sefrioui Z and Santamaria J 2003 Solid State Electr. 47 2245
[2] Biškup N, Das S, Gonzales-Calbet J M, Bernhard C and Varela M 2015 Phys. Rev. B 91 205132
[3] Peña V, Sefriouri Z, Arias D, León C, Santamaria J, Varela M, Pennycook S J and Martinez J L 2004 Phys. Rev. B 69 224502
[4] Štrbík V, Beňačka Š, Gazi Š, Španková M, Šmatko V, Knoška J, Gál N, Chromik Š, Sojková M and Pisarčík M 2017 Appl. Surf. Sci. 395 42
[5] Soltan S, Albrecht J and Habermeier H –U 2004 Phys. Rev. B 70 144517
[6] Hoppler J, Stahn J, Niedermayer C, Malik V K, Bouyanrif H, Drew A J, Rössle M, Buzdin A, Cristiani G, Habermayer H –U, Keimer B and Bernhard C 2009 Nature Mater. 8 315
[7] Gray B A, Middey S, Conti G, Gray A X, Kuo C T, Kaiser A M, Ueda S, Kobayashi K, Meyers D, Kareev M, Tung I C, Liu J, Fadley C S, Chakhalian J and Freeland J W 2016 Sci. report 6 33184
[8] Chien T Y, Kourkouts L F, Chakhalian J, Gray B, Kareev M, Guisinger N P, Muller D A and Freeland J W 2013 Nature Commun. 4 2336
[9] Holden T, Habermeier H U, Cristiani G, Golnik A, Boris A, Pimenov A, Humlíček J, Lebedev O I, Van Tendeloon G, Keimer B and Bernhard C 2004 Phys. Rev. B 69 064505
[10] Linder J and Robinson J W A 2015 Nature Phys. 11 307
[11] Sojková M, Štrbík V, Nurgaliev T, Chromik Š, Dobročka E, Španková M, Blagoev B and Gál N 2016 J. Phys.: Conf. Series 700 012022
[12] Giaever I 1969 Tunneling Phenomena in Solids ed E Burstein and S Lundqvist (Plenum Press New York) chapter 3 pp. 19-30
[13] Sojková M, Nurgaliev T, Štrbík V, Chromik Š, Blagoev B and Španková M 2017 Acta Physica Polonica B 48 842
[14] Španková M, Štrbík V, Dobročka E, Chromik Š, Sojková M, Zhen M and Li J 2016 Vacuum 126 24
[15] Španková M, Rosová A, Dobročka E, Chromik Š, Vávra I, Štrbík V, Machajdík D, Kobzev A and Sojková M 2015 Thin Solid Films 583 19
[16] Simmons J G 1963 Appl. Phys. 34 238
[17] Rowell J M 1969 Tunneling Phenomena in Solids ed E Burstein and S Lundqvist (Plenum Press New York) chapter 27 pp. 385-404
[18] Semba K and Matsuda A 2001 Phys. Rev. Lett. 86 496S
[19] De Léon-Guevara A M, Berthet P, Berthon J, Millot F, Revecolevschi A, Anane A, Dupas C, Le Dang K, Renard J P and Veillet P 1997 Phys. Rev. B 56 6031
[20] Sawa A, Kashiwaya S, Obara H, Yamashiki H, Koyanagi M, Yoshida N and Tanaka Y 2000 Physica C 339 287