# Copper Oxide Superconducting/Antiferromagnetic Interface

Yulii Kislinskii<sup>1,4,a\*</sup>, Karen Constantinian<sup>1,b</sup>, Gennady Ovsyannikov<sup>1,2,c</sup>,

Anton Shadrin<sup>1,e</sup>, Igor Borisenko<sup>1,d</sup>, Yuri Khaydukov<sup>3,f</sup>,

Alexander Sheyerman<sup>1,g</sup>, Aleksandr Vasiliev<sup>5,h</sup>

<sup>1</sup>Kotel'nikov IRE RAS, 125009 Moscow, Russia

<sup>2</sup>Chalmers University of Technology, SE-41 296 Gothenburg, Sweden

<sup>3</sup>Max-Plank Institute for Solid State Research, 70 569 Stuttgart, Germany

<sup>4</sup>Shubnikov Institute of Crystallography, 119333 Moscow, Russia

<sup>5</sup>Kurchatov National Research Center, Moscow, Russia

<sup>a\*</sup>yulii@hitech.cplire.ru, <sup>b</sup>karen@hitech.cplire.ru, <sup>c</sup>gena@hitech.cplire.ru, <sup>e</sup>anton\_sh@hitech.cplire.ru, <sup>d</sup>iboris@hitech.cplire.ru, <sup>f</sup>yury.khaydukov@frm2.tum.de, <sup>g</sup>sasha@hitech.cplire.ru, <sup>h</sup>a.vasiliev56@gmail.com

**Keywords:** accumulation and depletion of holes, band diagram, capacitance, high  $T_c$  Josephson junction, mesa-heterostructures, proximity effect, superconducting/antiferromagnetic interfaces.

**Abstract.** Superconducting Nb/Au/Ca<sub>1-x</sub>Sr<sub>x</sub>CuO<sub>2</sub>/YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> mesa-heterostructures were investigated. Dependencies of electrical parameters versus inverse capacitance were measured. A band diagram which takes into account an accumulation of holes in Ca<sub>1-x</sub>Sr<sub>x</sub>CuO<sub>2</sub> interlayer and band bending due to difference of work functions was proposed. The dependencies of electrical parameters were analyzed by examining the quasipartical and superconducting currents.

# Introduction.

Processes in interfaces are important for carrier transport in high critical temperature ( $T_C$ ) Josephson junctions. In YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>/PrBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>/YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO/PBCO/YBCO) junctions an accumulation of mobile holes into the p-type PBCO takes place, which results in conversion of hopping conductor PBCO into a metal up to 50 nm in depth [1]. In YBCO/La<sub>0.7</sub>Sr<sub>0.3</sub>MnO<sub>3</sub>/YBCO junctions the manganite barrier provides depletion of the holes in p-type YBCO electrode and dielectric layer was formed [2]. Procedure to calculate hopping conductivity parameters from electrophysical properties of junctions was developed [3, 4]. Such high –  $T_C$  junctions usually exhibit I-V curves SNS – type [1, 5] and products of normal resistance  $R_N$  by critical current  $I_C$  are not very high. But up to now models of carrier transport and band diagrams for high –  $T_C$  junctions are qualitative [1,2] and require in-depth investigation.

# **Experimental.**

We have fabricated Nb/Au/Ca<sub>1-x</sub>Sr<sub>x</sub>CuO<sub>2</sub>/YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> mesa-structures. Most of the data in this paper concern mesa-heterostructures with an interlayer made from G-type antiferromagnetic Ca<sub>0.5</sub>Sr<sub>0.5</sub>CuO<sub>2</sub> (AMS). Ca<sub>0.5</sub>Sr<sub>0.5</sub>CuO<sub>2</sub> (CSCO) could be treated as hopping conductor with reistivity at 300 K  $\rho(300)\sim10 - 100 \text{ m}\Omega \cdot \text{cm}$ , which is more than for PBCO  $\rho(300)\sim1 \text{ m}\Omega \cdot \text{cm}$  [1]. The YBCO films of 100 nm thick were deposited on NdGaO<sub>3</sub> substrate by laser ablation. Without vacuum breaking CSCO layer was deposited at the same run. An epitaxial growth of YBCO and CSCO films was provided. A thin protection layer of 15 nm Au was deposited *in-situ*. Top layers of 200 nm Nb and Au contacts were made in other chamber. A square junctions with areas *A* from 10x10 to 50x50  $\mu$ m<sup>2</sup> and with CSCO interlayer with thickness d<sub>M</sub>=5 – 80 nm were made [6].

#### Model and uniformity of mesa-heterostructures.

Superconducting current was observed for AMS with CSCO thickness  $d_M=12 - 50$  nm with critical current density  $J_C=I_C/A$  of 2 - 600 A/cm<sup>2</sup>. Magnetic field dependencies  $I_C(H)$  were studied. Fig. 1 demonstrates dependence of  $I_C$  vs. magnetic field at 4.2 K for AMS with relatively thin  $d_M=20$  nm, A=10x10 µm<sup>2</sup>. The up triangles are positive current biased critical currents and down triangles – negative ones. Singularities on  $I_C(H)$  are caused by antiferromagnetic (AF) interlayer and are less pronounced than in [7] due to large  $J_C=350$  A/cm<sup>2</sup>. The main  $I_C$  oscillation period is of  $B_0\approx45$  µT. It has double width at H=0 that points on uniformity of critical current distribution at least for AMS with  $d_M\geq20$  nm. Field period  $B_0$  estimated by differences between  $I_C(H)$  minima in Fig. 1 gives an effective area for magnetic field penetration of 5 µm<sup>2</sup>. The period was of 10 times smaller than the  $B_0$  periods for mesa-structures without CSCO interlayer (MS). Small  $B_0$  of AMS compared to  $B_0$  of MS were observed earlier for other CSCO thicknesses [7]. This was explained by giant magnetic oscillations of critical currents of S-AF-S junctions [8].

A band diagram of the AMS is presented in Fig. 2. A YBCO electrode has high work function  $\Phi_{YBCO} 5 - 6 \text{ eV} [2]$ . The work functions of metals are low  $\Phi_{Au}=4.3 \text{ eV}$ ,  $\Phi_{Nb}=4 \text{ eV}$ . We suppose that electron affinity of CSCO  $\chi_{CSCO}$  is between:  $\Phi_{YBCO} > \chi_{CSCO} > \Phi_{Nb}$ . So, free holes from YBCO accumulate in p-type CSCO [1]. It is shown by up band bending in Fig. 2. By reference [2] if condition  $\chi_{CSCO} > \Phi_{Nb}$  is fulfilled a depletion of holes appears in CSCO at Au interface, band bending is down. Total bending in AMS is equal to difference in work functions  $\Phi_{YBCO}-\Phi_{Nb}=1-2 \text{ eV}$ .





Fig. 1.  $I_C$  versus magnetic field dependence at 4.2 K for AMS with  $d_M=20$  nm, A=10x10  $\mu$ m<sup>2</sup>. Up triangles are positive biased critical currents and down triangles – negative ones.

Fig. 2. Band diagram of AMS with CSCO barrier layer. Valence band  $E_V$  in YBCO and CSCO is shown as the bold line. Fermi level (dash line) is a constant in whole structure. Hopping sub-band of CSCO with an average barrier height  $E_0$  is thin dash lines. Difference in work functions  $\Phi$  is shown.

A hopping conductor has conductivity dependency  $G(T)=G_0exp[-(T_0/T)^{1/4}]$  that exponentially decreases with lowering the temperature. Experimental constant T<sub>0</sub> depends on carrier localization radius *a* and density of states at Fermi level *g* as  $T_0=24/(\pi kga^3)$ , k is Boltzmann constant. Details of the calculations for junctions with hopping conductor were described in [3]. By localization radius CSCO *a*=5±2 nm and by T<sub>0</sub>~10<sup>6</sup> K we have calculated g=(0.2 – 5)·10<sup>18</sup> (eV)<sup>-1</sup>cm<sup>-3</sup> in [6].

Conductivity by resonant tunneling with average barrier height  $E_0$  is  $G_{res} = (\pi e^2/\hbar)E_0 \cdot ga \cdot exp(-d/a)$ . For dielectric thickness  $d \rightarrow 0$  it is  $G_{res} = (\pi e^2/\hbar)E_0 \cdot ga$ . By measured G/A versus d dependency and known values g and a barrier height in YBCO/PBCO/Au junctions was calculated  $E_0=51 \text{ meV}$  [3]. We calculate the average height as  $E_0 = (h/2\pi^2 e^2)/[ga \cdot R_N A(d \rightarrow 0)]$ . The  $R_N A(d \rightarrow 0) \approx 0.18 \mu \Omega \text{ cm}^2$  was extrapolated from experimental dependency of resistance on area products versus CSCO thickness:  $R_N A(d_M)$  [6]. The calculated value for CSCO is  $E_0=10 - 20 \text{ meV}$ . The band diagram shows that there are metallic - type layer with thickness  $d_N$  and dielectric-type layer with thickness  $d_0$ .

#### Dependencies of electrical parameters and interface capacitance.

A dielectric thickness  $d_0$  may be less than interlayer thickness  $d_M$  if the accumulation takes place [1]. Dependence  $d_0$  versus  $d_M$  was calculated from C - capacitance of AMS [6]. By hysteresis of voltage-current curves McCumber parameters were calculated by Zappe formula [9]:  $\beta_C = [2 - (\pi - 2)\alpha] \alpha^{-2}$ ,  $\alpha = I_{RETURN}/I_C$ . By McCumber definition  $\beta_C = 4\pi e I_C R_N^2 C/h$  and for  $C = \varepsilon_0 A/d_0$ :  $\frac{d_0}{\varepsilon} = \varepsilon_0 \frac{A}{C} = \frac{4\pi\varepsilon_0}{h} \cdot \frac{A I_C R_N^2}{\beta_C}$ . (1)

We have calculated dielectric thickness  $d_0$  from experimental A/C values. Calculation by (1) shows increase of  $d_0/\varepsilon$  from  $d_0/\varepsilon=0.24\pm0.08$  at  $d_M=12$  nm to  $d_0/\varepsilon=11.7\pm2.9$  nm at  $d_M=50$  nm. Layer which does not contribute to capacitance could be extracted by the  $d_0/\varepsilon$  dependency vs.  $d_M$  [6]. This layer with a thickness of  $d_N\approx20$  nm has metallic type behavior. For samples with  $d_M>20$  nm there is a wide dielectric layer  $d_0\approx d_M$ - $d_N$ . Dependence of  $R_NA$  versus  $d_0/\varepsilon$  ratios is shown in Fig. 3.



Fig. 3. Dependency of  $R_NA$  of AMS versus  $d_0/\varepsilon$ . Open symbols are data for AMS with  $d_M \le 20$  nm, exponential approximation for the data is dotted line. Closed symbols show data for samples with  $d_M \ge 20$  nm, exponential approximation – solid line, it's standard deviation – dashed lines.



Fig. 4. Dependency of current densities versus  $d_0/\varepsilon$ . Open symbols are data for AMS with  $d_M \le 20$  mn, exponential fit is dotted line. Closed symbols – data for  $d_M \ge 20$  nm, it's exponential approximation – solid line, standard deviation shown as dashed lines.

In Fig 3 data for AMS with small CSCO thicknesses  $d_M=12$ , 20 nm and for thicker CSCO with  $d_M=28$ , 40, 50 nm were fitted separately by steep and smooth sloping exponents. The fits are:

$$R_N A = k_{RS} \exp(\frac{d_0/\varepsilon}{a_{RS}}) \qquad d_M \le 20 \text{ nm}, \qquad R_N A = k_{RG} \exp(\frac{d_0/\varepsilon}{a_{RG}}) \ d_M \ge 20 \text{ nm}.$$
(2)

For steep exponent by a least squares fit method we obtain coefficients  $k_{RS}=0.5 \ \mu\Omega \text{cm}^2$ ,  $a_{RS}=0.6 \text{ nm}$  that is shown by dotted line in Fig 3. For AMS with  $d_M>20 \text{ nm}$  the exponential increase was more smooth:  $k_{RG}=9 \ \mu\Omega \text{cm}^2$ ,  $a_{RG}=6.6 \text{ nm}$  that is solid line. An error of calculation of the coefficient gives interval 5< $a_{RG}$ <9 nm that is shown as dashed lines.

Dependency of critical current density  $J_C$  versus ratio of  $d_0/\epsilon$  is shown in Fig 4. The fits are:

$$J_C = k_{JS} \exp\left(-\frac{d_0/\varepsilon}{a_{JS}}\right) \qquad d_M \le 20 \text{ nm}, \qquad J_C = k_{JG} \exp\left(-\frac{d_0/\varepsilon}{a_{JG}}\right) \qquad d_M > 20 \text{ nm}.$$
(3)

A steep exponential decrease with coefficients  $k_{JS}=380 \text{ A/cm}^2$ ,  $a_{JS}=0.5 \text{ nm}$  was obtained by the least squares fit. Dependence for AMS with  $d_M>20 \text{ nm}$  gives the parameters:  $k_{JG}=12.5 \text{ A/cm}^2$ ,  $a_{JG}=8.0 \text{ nm}$ . It is shown as solid line in Fig. 4. The error interval  $5 < a_{JG} < 14 \text{ nm}$  is shown as dashed lines.

From Fig. 3 and Fig. 4 we conclude that transport mechanism in AMS changes during increase of thickness approximately at  $d_M=20$  nm. In case of  $d_M\leq 20$  nm the steep slopes in  $R_NA(d_0/\varepsilon)$  and  $J_C(d_0/\varepsilon)$  dependencies may be explained by direct tunneling through thin barrier at CSCO/Au interface (Fig. 2). The barrier thickness may be estimated by  $\varepsilon \sim 2$  and  $d_0/\varepsilon = 0.24 - 1$  nm (Fig. 3) that gives  $d_0\approx 0.5 - 2$  nm. The rest of CSCO layer is metallic – type due to the accumulation. The height of a rectangular barrier may be estimated as:  $E_b\approx h^2/(8\pi em_e a_D^2)$  with  $m_e=9\cdot 10^{-31}$  kg. Characteristic length  $a_D$  may be obtained by dependencies for direct tunneling:  $J_C\sim exp(-2d/a_D)$ ,  $R_NA\sim exp(2d/a_D)$ . Compared the dependencies with formula (2) and (3) one obtains  $a_D\approx 2\varepsilon \cdot a_{RS}$ ,  $a_D\approx 2\varepsilon \cdot a_{JS}$  that gives  $a_D\sim 4a_{JS}=2$  nm and  $E_b\sim 10$  meV. The coefficients are the same  $a_{RS}\approx a_{JS}$ , thus  $I_CR_N(d_0)\approx$ const.

Because  $a_{RG} \approx a_{JG}$  one obtains  $V_C(d_0) \approx \text{const}$  for  $d_M > 20$  nm also. Large values of  $a_{RG} \sim 10a_{RS}$  and  $a_{JG} \sim 10a_{JS}$  lead to a barrier height  $E_b \sim a_D^{-2} \approx 0.1$  meV small compared to kT $\approx 0.36$  meV at 4 K. So direct tunneling can not explain the smooth exponential slopes. Other explanation may be proximity effect which originates from resonant tunneling through pair states in CSCO barrier [10]. In case of narrow widths of energy states  $\Gamma = E_0 exp(-d_0/a) < kT_C$  the pair breaking occurs. In opposite limit  $\Gamma > kT_C$  if Cooper pairs tunnel via resonant levels [10], it also yields  $V_C(d_0) = \text{const.}$ 

Note, at localization radius  $a\approx 1$  nm no supercurrent was observed through PBCO interlayer 7.5 nm in thickness [3], but for larger  $a\approx 3$  nm and PBCO thickness 20 nm a Josephson current was reported in [4]. In our AMS with superconducting/antiferromagnetic interfaces large radii a in Ca<sub>0.5</sub>Sr<sub>0.5</sub>CuO<sub>2</sub> antiferromagnetic interlayer support long proximity effect and result in experimentally observed decrease of quasipartical current with  $d_M$ , keeping  $V_C$  very slightly dependent from CSCO thickness.

# Acknowledgment

This work was supported partially by the RAS, RFBR projects 14-07-00258, 14-07-93105, Scientific School grant NSH-4871.2014.2. P.V. Komissinki is grateful for fruitful discussions.

# References

[1] M.I. Faley, U. Poppe, C.L. Jia, K. Urban, Order and interface effects in  $YBa_2Cu_3O_7$ - $PrBa_2Cu_3O_7$ - $YBa_2Cu_3O_7$ -Josephson junctions, IEEE Trans. on Appl. Super. 7 (1997) 2514 – 2517.

[2] M. Van Zalk, A. Brinkman, J. Aarts, H. Hilgenkamp, Interface resistance of  $YBa_2Cu_3O_7/La_{0.7}Sr_{0.3}MnO_3$  ramp-type contacts, Physical Rev. B 82 (2010) 134513-1 – 134513-9.

[3] J. Yoshida, T. Nagano, Tunneling and hopping conduction via localized states in thin  $PrBa_2Cu_3O_7$  barriers, Physical Rev. B 55 (1997) 11860 – 11871.

[4]J. Yoshida, T. Nagano, T. Hashimoto, Current transport amd electronic states in a, b- axisoriented  $YBa_2Cu_3O_7/PrBa_2Cu_3O_7/YBa_2Cu_3O_7$  sandwich type junctions, Physical Rev. B 53 (1996) 8623 - 8631.

[5] S. Charpentier, G. Roberge, S. Godin-Proulx, P. Fournier, Proximity effect in electron-doped cuprate Josephson junctions, Applied Phys. Lett. 99 (2011) 032511-1 – 032511-3.

[6] K.Y. Constantinian, Yu.V. Kislinskii, G.A. Ovsyannikov, A.V. Shadrin, A.E. Sheyerman, A.L. Vasiliev, M.Yu. Presnyakov, P.V. Komissinskiy, Interfaces in superconducting hybrid heterostructures with an Antiferromagnetic Interlayer, Physics of the Solid State. 55, (2013) 461 – 465.

[7] Yu.V. Kislinskii, K.Y. Konstantinian, G.A. Ovsyannikov, P.V. Komissinskiy, I.V. Borisenko, A.V. Shadrin, Magnetically dependent superconducting transport in oxide heterostructures with an antiferromagnetic layer, JETP 106 (2008) 800 – 805.

[8] L.P. Gorkov, V.Z. Kresin, Mixed-valence manganites: fundamental and main properties, Physics reports 400 (2004) 149 – 208.

[9] H.H. Zappe, Minimum current and related topics in Josephson tunnel junction devices, Journal of Appl. Phys. 44 (1973) 1371 – 1377.

[10] I. A. Devyatov, M.Yu. Kupriyanov, Resonant tunneling and long-range proximity effect, JETP Letters, 59 (1994) 200 – 205.