High temperature superconductivity at the interface 
Ba$_{0.8}$Sr$_{0.2}$TiO$_3$/La$_2$CuO$_4$

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Abstract. We present the results of the investigations of high conducting area and superconductivity at the interfaces between ferroelectric oxide and insulating oxide in heterostructures, isostructural to BaTiO$_3$/La$_2$CuO$_4$. The performed numerical simulations of BaTiO$_3$/La$_2$CuO$_4$ heterostructure show the possibility of high conductivity state at the interface. The temperature dependence of the measured electrical resistance of the Ba$_{0.8}$Sr$_{0.2}$TiO$_3$/La$_2$CuO$_4$ heterostructure interface have been studied and the superconducting behavior with transition temperature $T_c$ about 30K has been found. Therefore, the transition to the state with 2DEG at the interface is demonstrated. The results offer the possibility to design novel electronic devices.

1. Introduction

The creation of high-mobility quasi-two-dimensional electron gas (q2DEG) at the interface and the ability to control such states by magnetic and electric fields is impossible without the use of new materials and without the development of new design of interfaces. Unique properties of functional materials are achieved due to the effects associated with the complex composition of the interface structure. Such new materials include oxide heterointerfaces between two nonconducting oxides, in which unique transport properties are observed due to strong electronic correlations [1-11]. A high-mobility electron gas has been discovered at the interface between two oxide insulators LaAlO$_3$ (LAO) and SrTiO$_3$ (STO) by Ohtomo and Hwang [1]. In particular, it was found that the metallic phase (quasi-two-dimensional electron gas, 2DEG) is formed in the STO layers at the LAO/STO interface, when the number of LAO layers is larger than three [2]. The density of the charge carriers in such heterostructure reaches the value of $3 \times 10^{13}$ cm$^{-2}$. Subsequently, the coexistence of a two-dimensional superconductivity and ferromagnetism was discovered in this system [3,9] and in other systems with the atomically perfect interfaces [12,13]. The LAO/STO system undergoes a transition to a superconducting state at temperatures below 300 mK [3,9]. That is why it is essential to develop technical approaches to create quasi-two-dimensional superconductivity at higher temperatures and to study the processes of...
superconductivity switching. In our work, we used the parent compound of a high-temperature superconductor (PCHTSC) La$_2$CuO$_4$ (LCO) as a substrate and ferroelectric oxide Ba$_{0.8}$Sr$_{0.2}$TiO$_3$ (BSTO) as an overlayer in the heterostructure. We chose the BSTO/LCO heterostructure, because we have a great experience in working with BSTO films [20,21]. Epitaxial films of ferroelectric BSTO were deposited by reactive sputtering of stoichiometric targets by RF plasma (RF-sputtering) [20,21] at 650°C on antiferromagnetic LCO single crystals. The polarization of the ferroelectric film was perpendicular to the surface of interface. Such heterostructure might provide an opportunity to the electron system formation in the substrate layers close to the interface as in the LaAlO$_3$/SrTiO$_3$ case. The difference is that the polar discontinuity at the interface is created not by the charge sequence in the upper slab, but by the bulk polarization. Therefore, we expect that quasi-two-dimensional superconductivity could be observed at much higher temperatures in the BSTO/LCO heterostructure. The temperature dependences of the electrical resistance and magnetic properties were studied. The numerical simulations were performed for the BaTiO$_3$/La$_2$CuO$_4$ (BTO/LCO) heterostructure.

2. Modelling and calculations
The BaTiO$_3$ (BTO) compound is close in its physical properties to ferroelectric oxide BSTO. Thus, the numerical simulations [14-20] of the structural and electronic characteristics have been performed for the BaTiO$_3$/La$_2$CuO$_4$ heterostructure, because the calculations become much simpler for this case. For density of states calculations and structural optimization, we use density functional theory (DFT) [14]. Correlation effects were accounted by generalized gradient approximation (GGA) [15]. Kohn-Sham equations were solved using the plane-wave basis set (PAW) [16] realized within the VASP code [17], which is a part of the MedeA® software of Materials Design [16]. The cut-off energy was chosen to be 400 eV. The force tolerance was 0.5 eV/nm and the energy tolerance for the self-consistency loop was $10^{-5}$ eV. The Brillouin zones were sampled including $5 \times 5 \times 1$ k-points. Since there are strong correlations between $d$ and $f$-electrons in our system, the GGA+U correction were included to our computational scheme [18]. The $U$ parameter was added to La 4$f$ and Ti 3$d$ orbitals ($U = 8$ eV and 2 eV, respectively). The choice of $U$ for Ti and La atoms was based on our previous results [19].

The density of states of BTO/LCO heterostructure for the case when 2 layers of BTO were deposited on top of LCO substrate is shown in Figure 1. Due to a high mismatch between BTO and LCO ($\approx30\%$) the BTO unit cell was rotated by a 45° before merging with LCO substrate. Because the substrate is compressive with respect to the film, the polarization axis of the ferroelectric BTO will be directed perpendicular to the interface. The density of states of BTO/LCO heterostructure at the Fermi-level becomes also sufficiently large, approximately 5 eV$^{-1}$. The band gap vanishes, and we expect the system to be a good metal at the interface. Thus, the possibility of q2DEG state in this heterostructure has been shown. BTO/LCO heterostructure can be superconductive at low temperatures.

![Figure 1](image_url)

**Figure 1.** Density of states of BaTiO$_3$/La$_2$CuO$_4$ heterostructure for the case of 2 BaTiO$_3$ layers on the top of La$_2$CuO$_4$. 
3. Experimental results and discussion

The electrical resistance of the BSTO/LCO heterostructure at the low temperatures has been measured. The results are shown in Figure 2. The temperature dependence of the resistance of the BSTO/LCO heterostructure above 50 K shows typical semiconducting behaviour. At temperatures above 50 K the main current flows through the substrate. And only a small part of the current is localized in the interface area. This cannot affect the current in the large volume of the substrate at high temperatures. At low temperatures, the resistance rapidly decreases and superconducting behaviour is observed. Most of the current flows in the interface region, because at these temperatures the resistivity of the substrate increases, but the interface is in the superconducting state. Therefore, the interface between the ferroelectric and insulating oxides demonstrates the superconducting behaviour with the critical temperature about 30 K (Fig. 2). The resistivity starts to drop around 40 K demonstrating the superconducting transition, similar to what is observed in La_{2−x}Sr_xCuO_4 (LSCO) single crystals at optimal doping [12].

In order to demonstrate the superconducting state in the heterostructure, we have measured the zero field cooled and field cooled magnetic susceptibility in different magnetic fields (Fig. 3). Observation of the diamagnetic susceptibility (Fig. 3) confirms the existence of superconducting phase at the interface. A very rough estimate of the superconducting layer thickness from diamagnetic susceptibility measurements provides value of 4-100 nm.

Figure 2. Temperature dependences of the resistance for Ba_{0.8}Sr_{0.2}TiO_3/La_2CuO_4 heterostructure for film polarization directed perpendicular to ferroelectric film (red squares). Space distribution of surface charge density $\sigma_s$ in the vicinity of the interphase is shown in the insert.

Figure 3. Temperature dependence of the magnetic susceptibility of Ba_{0.8}Sr_{0.2}TiO_3/La_2CuO_4 heterostructure.

Figure 4. Magnetic field dependence of the resistance of the Ba_{0.8}Sr_{0.2}TiO_3/La_2CuO_4 heterostructure.
The magnetic field dependence of the resistance of the Ba$_{0.8}$Sr$_{0.2}$TiO$_3$/La$_2$CuO$_4$ heterostructure is shown in Figure 4. When a weak magnetic field is applied to the heterostructure in the direction perpendicular to the surface of the interface, the finite resistance of the interface appears and it increases with increasing the field (Fig. 4) as it was predicted [22]. The magnetic field was applied perpendicular to the surface and parallel to the c axis of the LCO substrate at T = 22.3 K. The magnetic field dependence of the heterostructure resistance shows that nonzero resistance appears at a very low field. The $H_{c1}$ for a thin layer of superconductor is very small and the magnetic field penetrates in the superconducting layer. In that case, the system demonstrates flux-flow resistance.

The polarization discontinuity model is most often discussed [1,2,10,11] to describe the q2DEG formation phenomena in the LAO/STO heterostructure. The polar discontinuity at the interface leads to the divergence of the electrostatic potential. The polar discontinuity for LAO/STO case exists due to an alternation of the differently charged layers of (LaO)$_3^+$ and (AlO)$_2^-$. Therefore, the atomically flat quality of the interface between LAO and STO components is utterly necessary in this case. For the case of BSTO/LCO heterostructure, a volume ferroelectric polarization exists, and it is independent on the sequence of layers. As a result, it is possible to use more simple techniques in order to create BSTO/LCO heterostructure and to obtain quasi-two-dimensional superconductivity at much higher temperatures.

Let us discuss the physical processes at the interface. We take into account that a sharp discontinuity of the polarization $P$ at the interface leads to the formation of the surface charge. The surface charge density $\sigma_s = P_s$, where $P_s$ is the component of polarization perpendicular to the interface (see insert in Fig. 2). This surface charge should be screened. Therefore, the regions with the excess charge are formed at the opposite side of the interface. This charged region from La$_2$CuO$_4$ side may be considered as a surface doping. The characteristic surface carrier density $n_s$ in this region can be estimated as $n_s = \sigma_s / e = P_s / e$, where $e$ is the elementary charge. Assuming that the characteristic value of polarization in the bulk of ferroelectric is $P_s \approx 30$ $\mu$C/cm$^2$ we obtain $n_s \approx 1.87 \times 10^{14}$ cm$^{-2}$.

This charged region has a nonuniform charge distribution. The characteristic charge density decreases exponentially when we go away from the interface. The characteristic decaying length $d$ of the charge density is determined by the screening, and the screening length is defined as:

$$r_D = \sqrt{\frac{\varepsilon_0 E_F}{6\pi e^2 n_s}},$$

(1)

where $\varepsilon_0$ is the static dielectric constant, $E_F$ is the Fermi energy, and $n_s$ is the volume carrier density. Taking into account that $E_F$ is proportional to $n_s^{2/3}$ ($E_F = \hbar^2 (3\pi^2 n_s)^{2/3} / 2m$), the screening radius only weakly depends on carrier density $n_s$ ($r_D \sim n_s^{1/3}$). In order to estimate the characteristic decaying length of the charge density, we assume that $d = r_D$, and, therefore, $n_s = n_o / r_D = n_o / d$. Substituting this density in Eq. (1) and in the expression for $E_F$ we obtain equation for $d$. Solving this equation, we obtain the following expression for $d$:

$$d = \left( \frac{\hbar^2 \varepsilon_0 (9\pi)^{1/3}}{12e^2 mn_s^{1/3}} \right)^{3/5},$$

(2)

here $m$ is the effective mass of electron in parent compound, and $\hbar$ is the Planck constant. Taking into account that the effective mass is of the order of free electron mass and $\varepsilon_0 = 30$, we obtain $d \approx 4.5 \times 10^{-5}$ cm.

It should be noted that this length is slightly shorter than the distance between electrons in 2D plane $r = n_s^{-1/3} \approx 10^{-7}$ cm. Therefore, we believe that the characteristic thickness of the 2D electron gas would be more than 1-2 nm, which is slightly greater than the estimated value. The value of surface carrier density $n_s \approx 1.87 \times 10^{14}$ cm$^{-2}$ exceeds the value $n_o \approx 0.53 \times 10^{14}$ cm$^{-2}$, which corresponds to the carrier density of the CuO$_2$ layer in superconducting state for optimal doping ($x=0.15$) in La$_2$Sr$_x$CuO$_4$. Thus, superconducting state can be realized on the one of the CuO$_2$ layers in the interval of 1-2 nm in the vicinity of the interface.
4. Conclusion
In conclusion, the numerical simulations have shown the possibility of q2DEG state in the BaTiO$_3$/La$_2$CuO$_4$ heterostructure. We observed the transition to quasi-two-dimensional electron gas state with superconducting behaviour below 40 K in the similar Ba$_{0.4}$Sr$_{0.6}$TiO$_3$/La$_2$CuO$_4$ heterostructure. The polarization of the ferroelectric film was perpendicular to the interface surface. It was shown that the resistivity properties of the superconducting state change irreversibly, when a magnetic field was applied perpendicular to the surface of interface. The observation of diamagnetic susceptibility confirmed the formation of the superconducting state of BSTO/LCO heterostructure. Therefore, our results demonstrate that quasi-two-dimensional highly conductive and superconductive regimes can be observed at the ferroelectric/dielectric interfaces.

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