

Towards high-temperature quasi-two-dimensional superconductivity

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(Received 21 February 2018; revised manuscript received 7 August 2018; published 27 September 2018)

The demonstration of a quasi-two-dimensional electron gas (2DEG) and superconducting properties in LaAlO₃/SrTiO₃ heterostructures has stimulated intense research activity in the last ten years. The 2DEG has unique properties that are promising for applications in all-oxide electronic devices. The superconductivity in such heterostructures has been observed below 300 mK. For superconductivity applications it is desirable to have more wide temperature of the existence range and the ability to control superconducting properties by external stimulus. Based on first-principles calculations and theoretical consideration we show that all-oxide heterostructures incorporating a ferroelectric constituent, such as BaTiO₃/La₂CuO₄, allow creating 2DEG. We predict a possibility of a high-temperature quasi-two-dimensional superconducting state. This state could be switchable between superconducting and conducting states by ferroelectric polarization reversal. We also discuss that such structures must be more simple for preparation. The proposed concept of a ferroelectrically controlled interface superconductivity offers the possibility to design novel electronic devices.

DOI: [10.1103/PhysRevB.98.094522](https://doi.org/10.1103/PhysRevB.98.094522)

I. INTRODUCTION

The creation of quasi-two-dimensional superconducting phases at the interface and the ability to control such states by magnetic and electric fields is impossible without the use of new materials and development of new interfaces. Unique properties of functional materials may be achieved due to effects associated with complex composition of the interface structure. Such new materials include oxide heterointerfaces between two nonconducting oxides in which, owing to strong electronic correlations, unique transport properties are observed. A quasi-two-dimensional electron gas (2DEG) has been discovered at the interface between two oxide insulators LaAlO₃ and SrTiO₃ by Ohtomo and Hwang [1] and it has attracted significant attention [1–11] due to a wide range of other physical phenomena observed in this system. Demonstration of a high carrier mobility and high electron density of the 2DEG [1,3,7–9] makes it promising for applications in all-oxide field-effect devices. Subsequently, the coexistence of a two-dimensional superconductivity and ferromagnetism was discovered in this system [3,4]. It was found [3] that the system passes into the superconducting state below 300 mK. The density of the charge carriers in such a heterostructure reaches the value of $3 \times 10^{13} \text{ cm}^{-2}$.

The most common mechanism for describing the 2DEG formation phenomena is the polarization catastrophe model [1,10]. The polar discontinuity at the interface leads to the divergence of the electrostatic potential. Along the [001] direction, LAO slab can be considered as an alternation of the differently charged layers of (LaO)⁺¹ and (AlO₂)⁻¹. As it was shown experimentally, in the heterostructure with the TiO₂ interface layer the electric potential along the [001] direction appears due to the polarity disruption at the interface. Thus, the atomically flat quality of the interface between two

components is utterly necessary since the effect is related to the strictly defined sequence of layers inside each slab. However, a transition to the superconducting state is observed at very low temperatures. 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 suggest to use the parent compound of a high-temperature superconductor (PCHTSC) as a substrate and ferroelectric oxide in the heterostructure as an overlayer. Such a heterostructure might provide an opportunity to the electron system formation in the substrate layers close to the interface as in the LaAlO₃/SrTiO₃ 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 polarization. Moreover, using the PCHTSC is promising for the superconducting state formation. In order to test this idea, we present *ab initio* calculations of the structural and electronic properties of BaTiO₃/La₂CuO₄ (BTO/LCO) heterostructure consisting of ferroelectric material BTO and LCO as a PCHTSC. We consider the possibility of a high-temperature quasi-two-dimensional superconducting (HT2DSC) state appearance in that heterostructure. We discuss the Kosterlitz-Thouless critical temperature T_{KT} for the transition to the superconducting state due to preformed Cooper pairs. It appears that maximal value of T_{KT} is higher than the mean field critical temperature of the superconducting transition. Therefore, we conclude that the transition of the interface layer to the superconducting state will be governed by the mean field T_c . We also suggest that such structures must be more simple for experimental realization. Moreover, using a ferroelectric material as an upper slab of the heterostructure might allow us to change the interface properties by polarization switching in the ferroelectric slab. Therefore, this type of heterostructure

will provide a unique object for solving fundamental problems in the physics of high-temperature superconductors. Indeed, switching of the direction of polarization of the ferroelectric film in the heterostructure allows us to change the type of carriers in HT2DSC from a hole doped to an electron doped. This opens the opportunity to investigate electron and hole superconductivity under completely identical conditions, with the same lattice, phonon, and defect properties.

The paper is organized as follows: in Sec. II we present *ab initio* results, in particular, we briefly introduce the computational method that we employ; we present the results on the electronic structure of bulk components, as well as for the LCO slab. In the last part of Sec. II we show the electronic properties of the BTO/LCO heterostructure. In Sec. III we present some estimations and discuss the possibility of a high-temperature quasi-two-dimensional superconducting state formation in the considered heterointerface. In Sec. IV we discuss our DFT results together with theoretical estimations and make some conclusions. Section V summarizes our results.

II. DFT RESULTS

For density of states calculations and structural optimization we use density functional theory (DFT) [12]. Exchange and correlation effects were accounted for by generalized gradient approximation (GGA) [13]. Kohn-Sham equations were solved using the plane-wave basis set (PAW) [14], realized within the VASP code [15], which is a part of the MedeA[®] software of Materials Design [14]. The cut-off energy was chosen to be 400 eV. The force tolerance was 0.05 eV/Å 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 is a strong correlation between *d* and *f* electrons in our system, the GGA+*U* correction were included in our computational scheme [16]. The *U* parameter was added to La 4*f*, Ti 3*d*, and Cu 4*d* orbitals (*U* = 8, 2, and 10 eV, respectively). The choice of *U* for Ti and La values was based on our previous research [17]. The choice of *U* for Cu was based on comparison with LSDA+*U* calculations and experimental data for band gap and Cu local magnetic moment (Table 2 from Refs. [18,19]). In the case of LCO/BTO, heterointerface components represent two insulating oxides with different structure and cell parameters of the elementary cell. Due to a high mismatch between BTO and LCO ($\approx 26\%$) the BTO unit cell was rotated by 45° before merging with LCO substrate, since the $a_{\text{BTO}} \times \sqrt{2} = 5.657$ Å (what results in $\approx 5\%$ mismatch with LCO substrate). This means that the substrate is compressive with respect to the film. As a result, the polarization axis of the ferroelectric BTO will be directed perpendicular to the interface. For modeling the heterostructure the LCO central slab was enlarged by a factor of 1.5 and bounded by a varying number of BaTiO₃ layers with interface BaO or TiO₂ layers on both sides. Such a unit cell guarantees the absence of artificial dipoles and additional polarity which might arise due to nonsymmetric structure. In order to avoid interaction of the surfaces and slabs with their periodic images, a 20 Å vacuum region was added. In planes *a* and *b* cell parameters were fixed to lattice parameters of the LCO substrate, whereas atom positions were allowed to relax during the optimization procedure.

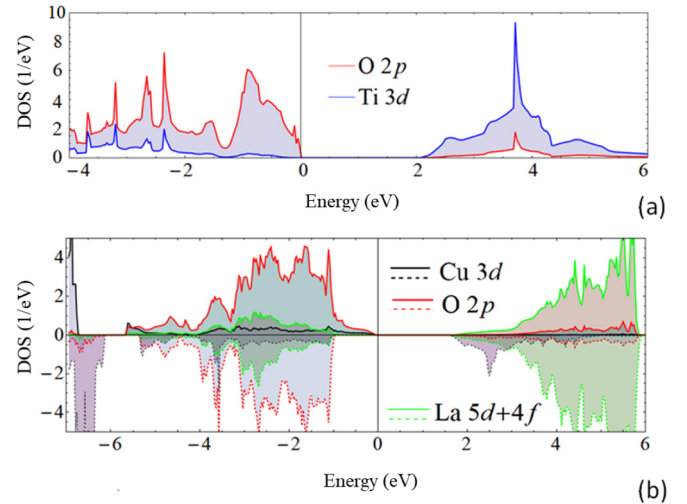


FIG. 1. (a) The atom-resolved electronic density of states of the bulk BaTiO₃ in the tetragonal phase. (b) Spin- and atom-projected density of states of the bulk La₂CuO₄ in the orthorhombic phase. Solid and dashed curves correspond to spin up and spin down contributions, respectively.

First, we study the heterostructure components separately. The structure optimization has been performed for the bulk BTO in the quasicubic tetragonal phase and for the bulk LCO with orthorhombic structure. Cell parameters obtained after optimization were $a = b = 4.00$ Å, $c = 4.02$ Å for BTO and $a = 5.42$ Å, $b = 5.42$ Å, $c = 13.23$ Å for LCO (experimental values are $a = b = 3.999$ Å, $c = 4.022$ Å and $a = 5.331$ Å, $b = 5.339$ Å, $c = 13.150$ Å, respectively [20,21]). In Fig. 1(a) the density of states spectrum (DOS) of the bulk BTO is presented. The obtained band gap of 2.1 eV is somewhat lower than the experimental value of 3.2 eV [22]. This is a well known fact [23] that the local and semilocal exchange-correlation functionals as provided by the local density approximation (LDA) and the generalized gradient approximation (GGA) underestimate the band gap of semiconductors. In order to analyze the electronic properties of the studied bulk LCO structure, DOS spectrum has been calculated taking into account magnetic properties of LCO and presented in Fig. 1(b). It is clearly seen that LCO is a magnetic semiconductor with a band gap of bulk LCO approximately 1.55 eV. The biggest contribution to magnetism is from Cu atoms in agreement with Ref. [18].

In Fig. 2(a) half of the unit cell of the studied system BTO/LCO with a BaO interface layer is presented, whereas the second part is a mirror copy with respect to the central LaO layer. After the optimization of both interface types (first corresponds to BaO interface layer, second to the TiO₂ layer) it was found that in the first case the total energy of the system is lower, meaning that the structure is more stable. That is why all further reasoning will be presented for the most stable configuration. It is seen from Fig. 2(a) that in the near-surface TiO₂ layer the Ti atoms move out of the oxygen planes by $\delta z \approx 0.15$ Å. That leads to a dipole moment induction towards the interface. Calculations involving a higher number of the BTO layers are required to get a full picture of structural distortions, which will be done in our future

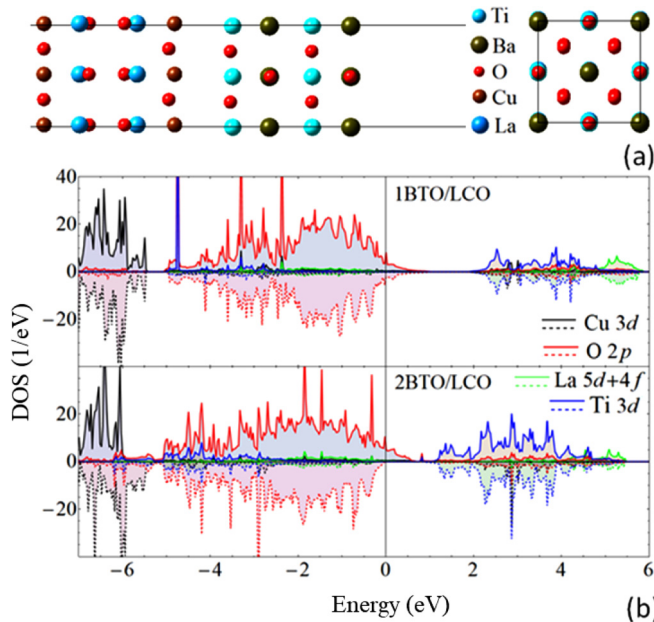


FIG. 2. (a) The unit cell of the heterostructure BTO/LCO (a) (only the right part of the slab is displayed). (b) Partial electronic density of states for the BTO/LCO heterostructure with one and two BaTiO₃ overlayers, respectively. Solid and dashed curves correspond to spin up and spin down contributions, respectively.

publications. In order to determine the electronic properties of the studied structure, the density of states (DOS) spectrum has been calculated taking into account magnetic features of LCO. Figure 2(b) show the atom-resolved DOS for the BTO/LCO heterostructure with one and two BTO overlayers. It is seen that already with one BTO layer the system becomes conducting. Cu atomic levels cross the Fermi level. Besides, the magnetic moment induction takes place which mainly corresponds to Cu atoms.

It is very instructive to study the arising conducting state by means of the layer-resolved densities of states. The corresponding results for the BTO/LCO heterostructure with two BTO overlayers are shown in Fig. 3. Here we clearly observe that the first and second BTO layers are transparent, having ≈ 2.2 and ≈ 1.1 eV band gaps, respectively. The maximum of DOS at the Fermi level corresponds to the next interfacial CuO₂ layer (DOS ≈ 5 eV⁻¹). The next two LaO layers have a negligibly small contribution to the Fermi level. The next CuO₂ layer has even smaller DOS. In other words, the main part of charge carriers is localized near the interfacial CuO₂ layer, towards the center of the LCO slab intensity sharply decreases after the interface, then subsides slower, and does not reach zero due to relatively thick substrate slab used in our simulations.

III. THEORETICAL ESTIMATIONS

Let us analyze the results and perform some estimates of the parameters of the arising state. We could estimate the width of the area with metallic conductivity as 0.7–3 nm. Then for a two-dimensional density n_s we got the value $n_s \approx 10^{14} - 4 \times 10^{14}$ cm⁻². Thus we expect that the system will

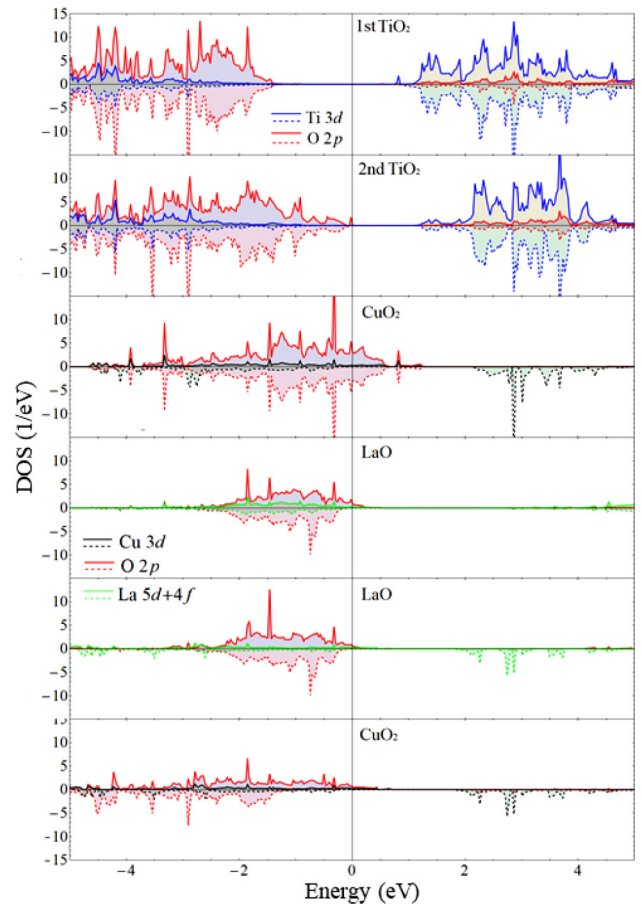


FIG. 3. Layer-resolved DOS for the 2BTO/LCO heterostructure. TiO₂ layers correspond to the BTO slab, whereas CuO₂ and LaO to the LCO substrate (the last CuO₂ layer is a central one).

become superconducting. Taking into account that the thickness of the conducting layer is small the superconducting properties may be governed by the Kosterlitz-Thouless transition. Let us estimate the maximal Kosterlitz-Thouless transition temperature of the interface layer assuming that superfluid density is equal to the total density in the interface layer:

$$T_{KT} = \frac{A \hbar^2 n_s}{4k_B m^*}, \quad (1)$$

where m^* is the effective mass of the current carriers, A is a coefficient ($A \approx 0.9$ for the two-dimensional case) [24]. Taking into account that 2D density is $n_s = 10^{14}$ cm⁻² and assuming that $m^* = 3m_e$ [25], where m_e is the free electron mass. We obtain $T_{KT} \approx 70$ K. It appears that this temperature is higher than the mean field critical temperature of the bulk LCO with optimal doping ($T_c \approx 40$ K). Since the thickness of the conducting layer at the interface is larger than the c -axis coherence length of optimally doped LCO superconductor, the critical temperature of the interface will be determined by the mean field critical temperature of LCO with optimal doping.

Let us consider the effect of application of an external magnetic field to SC layers. First of all note that the effective penetration depth will be enhanced $\lambda_{\text{eff}} = \lambda^2/d$ (d is the effective thickness of the interface layer and λ is the bulk penetration depth). Therefore the lower critical field will be

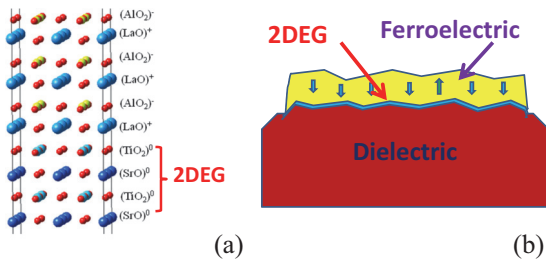


FIG. 4. An interface area of the LAO/STO heterostructure (a) and schematic illustration of the ferroelectric/dielectric with nonflat interface boundary (b).

strongly reduced. Depending on the intensity of the magnetic field H , it either penetrates or does not penetrate into the SC. The field does not penetrate into the SC at fields below H_{c1} . Above H_{c1} , the field begins to penetrate into the SC in the form of vortices, while the volume around the vortices remains in the SC state. Fields above H_{c2} completely penetrate the sample, the magnetic field in the sample becomes uniform, and the SC completely collapses. In the case of a thin SC film or a thin SC layer, there will be the feature, primarily associated with a demagnetizing factor. When the field is applied perpendicularly to the SC layer the field at the film boundary increases greatly by a factor of K ($K \approx 2\text{--}10 \times 10^6$ for a 1–5 nm thick film [26]) due to a large demagnetizing factor. As a result, when the field at the boundary $H_1 = K H$ exceeds H_{c1} , magnetic field vortices begin to penetrate the sample. Thus, the value of the external magnetic field at which the field begins to penetrate into SC is $H_{c1\text{eff}} = H_{c1}/K$, where H_{c1} is the value for the bulk sample. (In fact H_{c1} for films is smaller than that for bulk due to a transmission coefficient being bigger for films. But the coefficient K is so huge, that $H_{c1\text{eff}}$ is very small in any case.) Thus, the value of $H_{c1\text{eff}}$ for a thin SC layer turns out to be $H_{c1\text{eff}} \approx 0$ ($\ll 1$ Oe). The value of H_{c2} for a thin SC layer is the same as that for a bulk. The value of H_{c2} for a thin SC layer is still large (for example, $H_{c2} \approx 820$ kOe for $\text{La}_{0.85}\text{Sr}_{0.15}\text{CuO}_4$ [27]). Therefore, it is necessary to expect the appearance of flux flow resistivity in our heterostructure in any arbitrarily small fields, due to vortex motion.

It is supposed that the appearance of interface conductivity is related to the structural and, consequently, electronic reconstructions. The layers $(\text{BaO})^0$ and $(\text{TiO}_2)^0$ are “electrically neutral” in the simplest ionic limit, but there is a ferroelectric polarization due to the Ti^{+4} atoms displacements out of the octahedron center in the BTO. The direction of such a polarization can be switched by an external electric field. That is impossible to do in the case of LAO slab, because an external influence cannot change the sequence of $(\text{LaO})^{1+}$ and $(\text{AlO}_2)^{1-}$ layers. Moreover, it is very important that in this case there is no need to make a very high-quality interface boundary, because the polarization arises in the volume of the ferroelectric. This differs from the case of LAO where, for the appearance of polarization on the interface, it is necessary to obey strictly the sequence of $(\text{LaO})^{+1}$ and $(\text{AlO}_2)^{-1}$ layers [see Fig. 4(a)]. Besides, the BTO/LCO system attracts the interest because it contains an antiferromagnetic insulator La_2CuO_4 , which can be transferred to a conducting and superconducting state by increasing the concentration of the

free carriers [28,29]. That was realized by doping of the LCO by Sr or Ca [28,29]. It can be further expected that increasing the free charge carriers can lead to the 2D superconductivity in a system with 2DEG. Therefore, there is an opportunity to switch both conductivity and superconductivity by an electric field in the heterostructures similar to BTO/LCO.

IV. DISCUSSION

The idea to use ferroelectric oxide as an overlayer in the heterostructure of two insulating perovskid oxides was theoretically considered earlier [6,11]. We suggest to use ferroelectric oxide as an overlayer in the heterostructure when a substrate has the other type of elementary cell. In our case $a_{\text{BTO}} \times \sqrt{2} = 5.657$ Å and $a_{\text{LCO}} = 5.331$ Å. Because $a_{\text{LCO}}/a_{\text{BTO}} \times \sqrt{2} < 1$, the LCO substrate is compressive with respect to the BTO film. And, as a result, the polarization axis c of the ferroelectric film will be directed perpendicular to the interface. Recently, the occurrence of a quasi-two-dimensional state with a metallic character of the temperature behavior of the conductivity at the boundary of the ferroelectric $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ and the antiferromagnet LaMnO_3 [30] had been experimentally observed. A distinctive feature of these experiments was that the 2DEG state occurs on the boundary between different components with different types of elementary cells and in the heterostructure that was not an ideally flat interface. In contrast, LAO/STO 2DEG formation requires the atomically flat boundary and, in addition, a certain sequence of LaO and AlO_2 layers. Earlier it was predicted [6,31] that 2DEG can arise when a ferroelectric film is deposited at the insulator. In this paper we present an idea that two-dimensional superconductivity with relatively high critical temperatures can arise when a ferroelectric film is grown at the insulator substrate which could become a superconductor after doping.

Reference [30] was the first report on the low interface quality systems. Thus the proposed heterostructure seems to be exceptionally crucial. First, we investigate the emergence of a quasi-two-dimensional electron gas at a new type of $\text{BaTiO}_3/\text{La}_2\text{CuO}_4$ heterostructure interface in which there is neither LaAlO_3 nor SrTiO_3 . Second, PCHTSC can transform into a superconducting state with an increase in the number of carriers, so new physics and new possibilities appear here. And the most importantly, third, the use of a ferroelectric makes it possible to avoid the necessity of an extremely high quality of the interface, because the polarization in BaTiO_3 allows the polarization catastrophe to occur irrespective of the interface quality. We have demonstrated that increasing of the number of BTO overlayers [see Fig. 2(b)] increases the number of charge carriers since the polarization increases as well. Moreover, as it follows from Fig. 3 these carriers are localized near the interface. The possibility of a quasi-two-dimensional electron gas creation under less stringent conditions for the interface quality of the heterostructure and the realization of multifunctional conductivity switching regimes and magnetization can substantially increase the possibility of utilization of these systems in the mass production of technical devices based on such heterostructures.

Earlier, in the work of the Bozovic group, the giant proximity effect in cuprate superconductors had been observed [32].

Later, in Goldmann's work [33], the switching-off of the superconductivity when an electric field is applied to a sample with superconducting properties through an ionic conductor was considered. In our investigation, we propose the creation of a superconducting state due to the proximity effect with a ferroelectric, therefore turning it off and turning it on with electric or magnetic fields, by changing the direction of polarization in the ferroelectric film, or by changing the magnetic properties of the substrate, respectively. We also emphasize that when ferroelectric films are used in heterostructures, the less stringent requirements are imposed on the quality of the emerging interfaces. Therefore, the suggested interfaces with ferroelectric films are a completely new approach to the creation of 2DEG, which we have already tested [30]. And the idea of creating on the basis of such 2DEG states of two-dimensional superconductivity at the interface with LCO is an even more unique proposal, which has no analogs in the world.

V. CONCLUSION

In conclusion, in our paper we have presented the structural and electronic properties of the ferroelectric/PCHTSC

(BaTiO₃/La₂CuO₄) heterostructure obtained by *ab initio* calculations. We have shown that conducting phase arises in the case of slab geometry with surface conducting state, and in the case of BTO/LCO heterostructure with conducting state located mostly at the interfacial LCO layer. After the epitaxial ferroelectric BaTiO₃ film was deposited on the LCO sample using, for example, the magnetron sputtering technique, we can get HT2DSC in the interface. Note that HT2DSC have been observed in the Ba_{0.8}Sr_{0.2}TiO₃/La₂CuO₄ heterostructure with $T_c = 30$ K [34]. This T_c is 100 times larger than T_c in LaAlO₃/SrTiO₃ [3].

ACKNOWLEDGMENTS

R.F.M. acknowledges financial support from Slovenian Research Agency, Project BI-RU/16-18-021. V.V.K. acknowledges financial support from Slovenian Research Agency Program P1-0040. The authors from Kazan Federal University acknowledge partial support by the Russian Government Program of Competitive Growth of Kazan Federal University. This study was supported by the Supercomputing Center of Lomonosov Moscow State University.

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