ISSN 1674-1056
http://cpb.iphy.ac.cn
http://iopscience.iop.org/cpb
中国物理 B
Chinese Physics
中国物理 Chinese Physics B

Enhancing superconductivity of ultrathin $YBa_2Cu_3O_{7-\delta}$ films by capping non-superconducting oxides

Hai Bo(薄海), Tianshuang Ren(任天爽), Zheng Chen(陈峥), Meng Zhang(张蒙), Yanwu Xie(谢燕武)

Citation: Chin. Phys. B . 2019, 28(6): 067402. **doi:** 10.1088/1674-1056/28/6/067402
Journal homepage: http://cpb.iphy.ac.cn; http://iopscience.iop.org/cpb

What follows is a list of articles you may be interested in

Particle-hole fluctuations and possible superconductivity in doped α-$RuCl_3$

Bin-Bin Wang(王斌斌), Wei Wang(王巍), Shun-Li Yu(于顺利), Jian-Xin Li(李建新)

Chin. Phys. B . 2019, 28(5): 057402. **doi:** 10.1088/1674-1056/28/5/057402

Tunable superconductivity in parent cuprate $Pr_2CuO_{4\pm\delta}$ thin films

Xinjian Wei(魏鑫健), Ge He(何格), Wei Hu(胡卫), Xu Zhang(张旭), Mingyang Qin(秦明阳), Jie Yuan(袁洁), Beiyi Zhu(朱北沂), Yuan Lin(林媛), Kui Jin(金魁)

Chin. Phys. B . 2019, 28(5): 057401: **doi:** 10.1088/1674-1056/28/5/057401

High-throughput research on superconductivity

Mingyang Qin(秦明阳), Zefeng Lin(林泽丰), Zhongxu Wei(魏忠旭), Beiyi Zhu(朱北沂), Jie Yuan(袁洁), Ichiro Takeuchi, Kui Jin(金魁)

Chin. Phys. B . 2018, 27(12): 127402. **doi:** 10.1088/1674-1056/27/12/127402

A single-crystal neutron diffraction study on magnetic structure of $CsCo_2Se_2$

Juanjuan Liu(刘娟娟), Jieming Sheng(盛洁明), Wei Luo(罗伟), Jinchen Wang(汪晋辰), Wei Bao(鲍威), Jinhu Yang(杨金虎), Minghu Fang(方明虎), S A Danilkin

Chin. Phys. B . 2018, 27(11): 117401. **doi:** 10.1088/1674-1056/27/11/117401

Superconductivity with peculiar upper critical fields in quasi-one-dimensional Cr-based pnictides

Guang-Han Cao(曹光旱), Zeng-Wei Zhu(朱增伟)

Chin. Phys. B . 2018, 27(10): 107401. **doi:** 10.1088/1674-1056/27/10/107401

Enhancing superconductivity of ultrathin $YBa_2Cu_3O_{7-\delta}$ films by capping non-superconducting oxides*

Hai Bo(薄海)[1], Tianshuang Ren(任天爽)[1], Zheng Chen(陈峥)[1], Meng Zhang(张蒙)[1], and Yanwu Xie(谢燕武)[1,2,†]

[1] *Interdisciplinary Center of Quantum Information, Zhejiang Province Key Laboratory of Quantum Technology and Device, and Department of Physics, Zhejiang University, Hangzhou* 310027, *China*

[2] *Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing* 210093, *China*

(Received 21 February 2019; revised manuscript received 5 April 2019; published online 8 May 2019)

In this study, we have explored the ways to fabricate and optimize high-quality ultrathin $YBa_2Cu_3O_{7-\delta}$ (YBCO) films grown on single-crystal (001) $SrTiO_3$ substrates. Nearly atomic-flat YBCO films are obtained by pulsed laser deposition. Our result shows that the termination of $SrTiO_3$ has only a negligible effect on the properties of YBCO. In contrast, we found that capping a non-superconducting oxide layer can generally enhance the superconductivity of YBCO. $PrBa_2Cu_3O_7$, La_2CuO_4, $LaMnO_3$, $SrTiO_3$, and $LaAlO_3$ have been examined as capping layers, and the minimum thickness of superconducting YBCO with capping is ~ 2 unit cells–3 unit cells. This result might be useful in constructing good-performance YBCO-based field effect devices.

Keywords: $YBa_2Cu_3O_{7-\delta}$ (YBCO), superconductivity, oxides, film

PACS: 74.72.–h, 74.78.–w **DOI:** 10.1088/1674-1056/28/6/067402

1. Introduction

The high-temperature superconductor $YBa_2Cu_3O_{7-\delta}$ (YBCO) is still attracting intense attention.[1,2] Particularly, ultrathin films of YBCO have attracted significant research interest since the discovery of YBCO.[3–7] In the early days, a fundamental question as to what is the minimum thickness needed for the occurrence of superconductivity was raised because the layered structures of YBCO suggest a high degree of anisotropy.[3] Interestingly, it has been demonstrated that while single YBCO thin films lose superconductivity quickly when their thickness decreases to a few unit cells (uc), superconductivity has been achieved in as thin as 1-uc-thick YBCO layer embedded in superlattices that comprise YBCO and nonsuperconducting $PrBa_2Cu_3O_7$ (PBCO)[5–7] or sandwiches of PBCO/YBCO/PBCO.[3,8,9] In addition to the above fundamental question, the major interest of ultrathin YBCO films lies in its promising application of electric-field-effect devices such as three-terminal high-temperature superconductor transistors.[10] Compared with conventional Bardeen–Cooper–Schrieffer (BCS) superconductors, YBCO has a much smaller carrier density, $3 \times 10^{21}/\text{cm}^3$–$5 \times 10^{21}/\text{cm}^3$, which makes it intriguing to control superconductivity by electric field.[11] For this purpose, ultrathin YBCO films are needed because the tuning ability of the field effect is generally small. In addition, ultrathin YBCO films can also be used to construct an ultra-sensitive YBCO-based single-photon detector.[12]

As far as field effect application is concerned, unfortunately, superlattices[5–7] and sandwiches[3,8,9] are not very useful because an electric field can hardly penetrate these structures. For the optimization and future applications of YBCO-based field-effect devices, a high-quality ultrathin YBCO film that is directly accessible to gating materials is crucial.[10] Over the past two decades, electric-field effects on YBCO have been realized by either back gating through $SrTiO_3$[13–15] or top gating through $SrTiO_3$,[16,17] ferroelectrics,[18–20] or ionic liquids.[21–23] The ultrathin YBCO films used in these studies are either single[13,14,23] or buffered with PBCO.[17–20,22] Compared with the single YBCO, the PBCO-buffered YBCO has much better superconducting properties, and thus has been widely used in top-gating devices.[17–20,22] However, this strategy cannot be applied to the $SrTiO_3$-based back gating devices because PBCO, rather than YBCO, is directly contacting with $SrTiO_3$, the gating material. Instead, one would expect that a high-quality ultrathin YBCO film that is made directly on $SrTiO_3$ and covered with a suitable capping layer should work well for the back gating. Despite the intense previous studies of YBCO,[24] little attention has been paid to optimizing ultrathin YBCO films in this way.

In this work, we explore the possibility of optimizing ultrathin YBCO films directly grown on $SrTiO_3$ by controlling the termination of $SrTiO_3$,[25] and by capping nonsuperconducting oxides whose structures are compatible with YBCO. Our result shows that the superconductivity of YBCO is insen-

*Project supported by the National Key Research and Development Program of the Ministry of Science and Technology of China (Grants Nos. 2017YFA0303002 and 2016YFA0300204) and the Fundamental Research Funds for the Central Universities, China.

†Corresponding author. E-mail: ywxie@zju.edu.cn

© 2019 Chinese Physical Society and IOP Publishing Ltd

sitive to the terminations of $SrTiO_3$, but can be generally enhanced by capping different oxides, including dielectrics such as $SrTiO_3$ and $LaAlO_3$.

2. Experiment

All the films were grown on TiO_2-terminated (001)-oriented $SrTiO_3$ substrates by pulsed laser deposition using sintered polycrystalline targets. A KrF excimer laser (248 nm) was used with a laser fluence of 1.0 J/cm^2 and a repetition rate of 1 Hz. SrO-terminated $SrTiO_3$ was obtained by inserting 1-uc SrO on the TiO_2-terminated $SrTiO_3$ before growing YBCO (as shown in Fig. 1). The growth of YBCO was performed at 750 °C, under a mixed gas atmosphere of 0.03-mbar N_2O and 0.10-mbar O_2 (1 bar = 10^5 Pa). To avoid disturbing the properties of YBCO, all the capped oxides were deposited using the same growth conditions as YBCO. After growth, the samples were postannealed *in situ* at 500 °C, under 500-mbar O_2, for 30 minutes or 60 minutes, and then cooled to room temperature under the same atmosphere. The transport properties were measured using a standard four-probe method with evaporated silver electrodes.

3. Results and discussion

First, we examine the effect of $SrTiO_3$ termination on the properties of ultrathin YBCO films. Previous studies showed that the termination of $SrTiO_3$ has a dramatic influence on the transport properties of $LaAlO_3/SrTiO_3$ heterostructures[26] and manganite films,[27] because it changes the sequence of atomic layers in $LaAlO_3$ or manganite, which in turn changes charge transfer within the heterostructures or films. By analogy, in YBCO, charge transfer between different atomic layers plays a key role in superconductivity. Therefore, changing the termination of $SrTiO_3$ may also have an important influence on the properties of YBCO.[28]

Figure 1 shows the typical growth processes and surface topology of 5-uc YBCO films grown on TiO_2- and SrO-terminated $SrTiO_3$ substrates, respectively. The thickness of each layer is monitored by reflection high energy electron diffraction (RHEED) (Figs. 1(a) and 1(c)). Both types of surface are very smooth (Figs. 1(b) and 1(d)). The root mean square roughness of the TiO_2- and SrO-terminated samples are 0.41 nm and 0.52 nm, respectively. The cross-line profiles show that the roughness mainly comes from the height difference around 1 uc or 0.5 uc of YBCO. These characterizations confirm that our ultrathin YBCO films are of good quality.

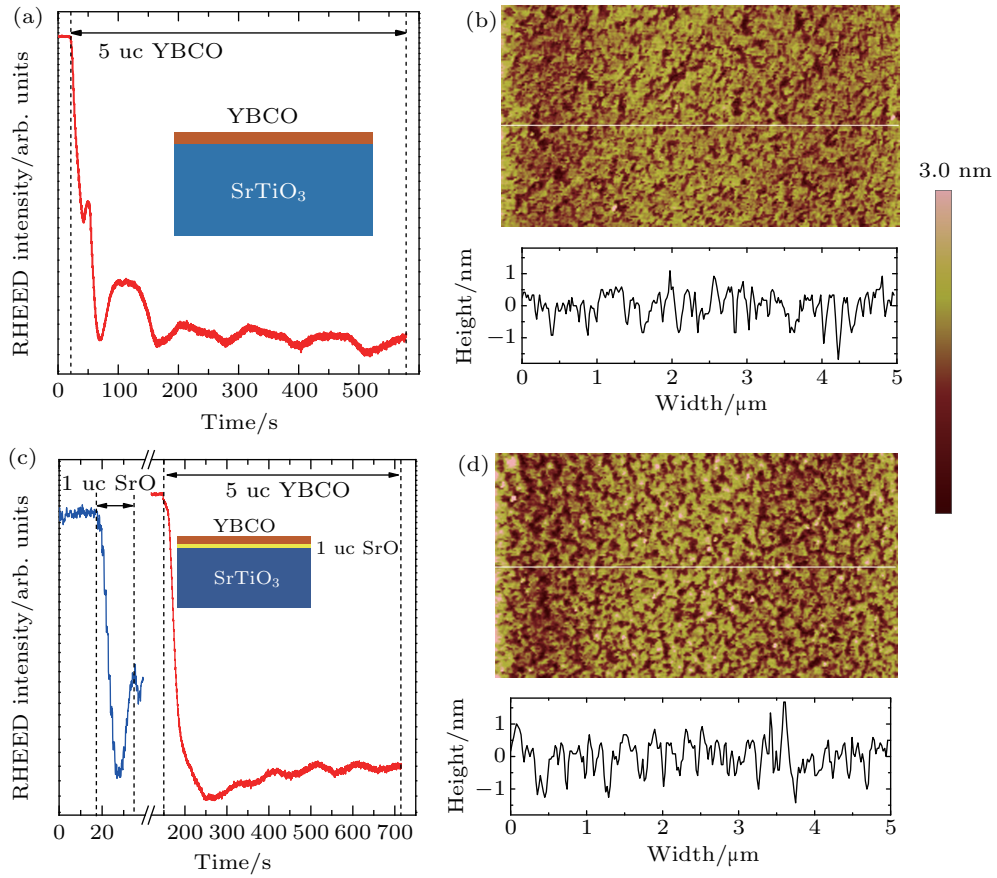


Fig. 1. (a) RHEED intensity oscillations and (b) atomic force microscope (AFM) image of a 5-uc YBCO film grown on TiO_2-terminated $SrTiO_3$ substrate. (c) RHEED intensity oscillations and (d) AFM images of a 5-uc YBCO film grown on SrO-terminated $SrTiO_3$ substrate that is made by inserting 1-uc SrO layer on the TiO_2-terminated $SrTiO_3$. Insets in panels (a) and (c) show the sample configuration. The root mean square roughness of panels (b) and (d) is 0.41 nm and 0.52 nm, respectively. Lower panels in panels (b) and (d) show the typical height profiles along the white cross lines.

In Fig. 2, we present the dependence of resistance on temperature for YBCO films of different thicknesses. As expected, as the YBCO thickness decreases, the critical temperature T_c decreases and the transition region broadens. This kind of thickness-dependent behavior can be attributed to several factors such as Kosterlitz–Thouless (KT) transition,[3] strain,[29] or defects. The superconductivity is fully suppressed when YBCO is thinner than 4 uc. One important observation is that the YBCO films grown on SrO-terminated $SrTiO_3$ (dotted lines) actually have quite similar transport properties as those grown on TiO_2-terminated $SrTiO_3$ (solid lines). Therefore, we conclude that simply changing the termination of $SrTiO_3$ cannot enhance superconductivity of the ultrathin YBCO films. In the following surface-capping studies, we will only focus on the YBCO films grown directly on the TiO_2-terminated $SrTiO_3$ substrates.

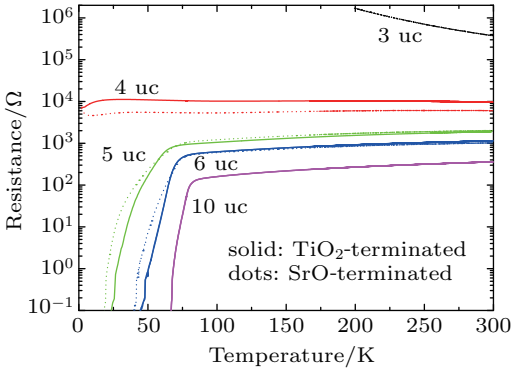


Fig. 2. The dependence of resistance on temperature for uncapped YBCO thin films of different thicknesses. The solid and dotted lines represent samples grown on TiO_2- and SrO-terminated $SrTiO_3$ substrates, respectively. The postannealing time for all these samples is 60 minutes.

Next, we examine the effect of surface capping on the properties of ultrathin YBCO films. Unlike the buffer layer, the choice of the capping layer is almost unlimited because it is on the top and thus does not obviously influence the crystalline structure of the underneath YBCO. In the present study, in order to obtain a high-quality capping layer, we limit our choice to a few oxides whose crystalline structures are compatible with YBCO. In addition, we have reduced the postannealing time from 60 minutes to 30 minutes to optimize the surface quality of the capped samples. Most of these capped samples still have a quite flat surface with a root mean square roughness below 1 nm (not shown). Figure 3 shows the typical transport results of capping 5-uc YBCO with $\sim$ 7-nm different oxides. The most important observation is that almost all of these capped oxides enhance the superconductivity of the YBCO films, more or less.

The dashed line in Fig. 3(a) represents the $R(T)$ curve of the uncapped 5-uc YBCO film. Note that it has a lower T_c and a broader transition region than the 5-uc YBCO shown in Fig. 2. This difference is due to the different postannealing time in oxygen because YBCO is very sensitive to oxygen content. As stated above, all the capped samples shown in this study are postannealed for 30 minutes. Therefore, we compare them with an uncapped sample that has been postannealed in the same condition. We emphasize that this uncertainty in the uncapped 5-uc YBCO film does not affect our main conclusion. To give a more quantitative comparison, we define T_c as the transition onset that is determined as the position of the maximum of the derivative curve, $dR(T)/dT$.[30] The result is summarized in Fig. 3(b). As shown in Figs. 3(a) and 3(b), after capping PBCO or La_2CuO_4, the superconducting properties become much better as indicated by a much higher T_c and a narrower transition region. Similar but smaller enhancements are observed in samples capped with $LaMnO_3$ and $La_{0.67}Ca_{0.33}MnO_3$. The transition regions of the samples capped with $La_{1.56}Sr_{0.44}CuO_4$, and $SrTiO_3$ are relatively broad, but their T_c values are clearly enhanced. An enhancement of T_c is also observed for the $LaAlO_3$-capped sample, although the transition is very broad. This point can be better appreciated if the data are plotted in the linear coordinates (inset of Fig. 3(a)).

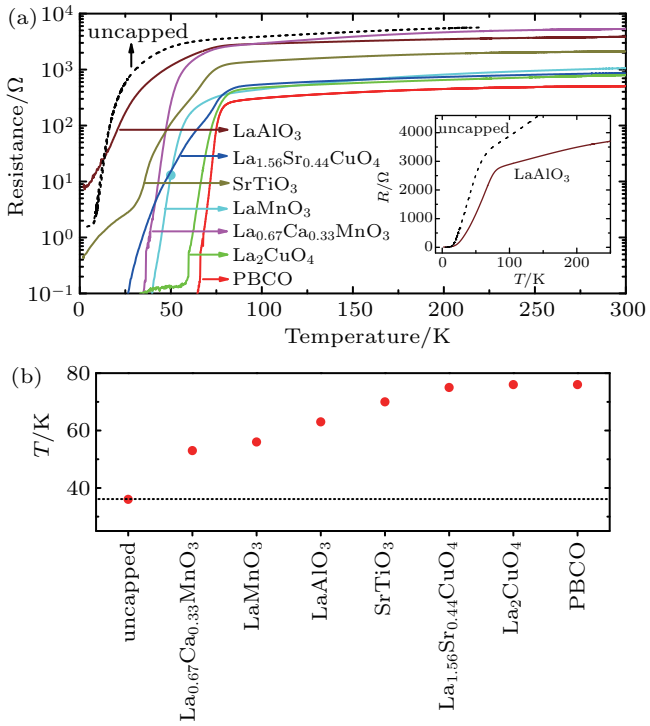


Fig. 3. (a) The dependence of resistance on temperature for 5-uc YBCO films capped with different oxides. The thickness of all the capping layers is around 7 nm. The result of the uncapped YBCO is also shown for comparison (the dashed line). The inset replots the result of the uncapped and the $LaAlO_3$-capped samples in the linear coordinates. (b) The critical temperature, T_c, generated from the R–T curves shown in panel (a). Here, T_c is defined as the transition onset that is determined as the position of the maximum of the derivative curve, $dR(T)/dT$. The postannealing time for all these samples is 30 minutes.

Why can the superconducting properties of ultrathin YBCO films be enhanced by these capped oxides? One general possibility is that the capped oxides can work as a protection layer. It is well known that the surface of YBCO is

chemically instable when it is exposed to air, mainly due to the reaction with water humidity and carbon dioxide. These deteriorations can be avoided by capping a protection layer. However, if this mechanism dominates, one would expect that all the capped oxides will enhance the superconductivity to a similar extent, which is clearly inconsistent with the result shown in Fig. 3. Capping-induced strain or optimization of the surface structure of YBCO may be excluded since it has been shown that the effect of capping is similar for crystalline or amorphous PBCO.[8] Therefore, the most likely scenario is charge transfer. That is, surface capping increases the hole concentration of YBCO. This scenario is supported by the observation that even the normal-state resistance of the capped samples drops a lot (Fig. 3), which cannot be simply attributed to the parallel conduction from the capped oxides.

We consider two charge-transfer processes. For the single YBCO film, the defects on its surface can easily react with water humidity and carbon dioxide in air, which can generate high-energy-level electrons that will be transferred into the YBCO.[31] This process will reduce the hole concentration of YBCO and thus deteriorate the superconductivity. This process can be largely suppressed by surface capping. This is the reason why almost all the capped oxides can enhance superconductivity. The second charge-transfer process takes place between the capped layer and the YBCO. This process determines the apparent difference of capping different oxides. Hole injection by capping PBCO has been discussed in previous studies.[3] Besides PBCO, La_2CuO_4, $La_{1.56}Sr_{0.44}CuO_4$, $LaMnO_3$, and $La_{0.67}Ca_{0.33}MnO_3$ are either a parent insulator of hole-type conductor or a hole-type conductor. They can provide holes to, or at least will not annihilate too much holes in, the YBCO. Therefore, the net effect of capping these materials is a significant enhancement of superconductivity in both T_c and transition region. The slightly lower T_c of $LaMnO_3$- and $La_{0.67}Ca_{0.33}MnO_3$-capped samples may be partially attributed to the interaction between superconductivity and magnetism.[32] $SrTiO_3$ and $LaAlO_3$ have different physical properties from others and cannot supply holes to YBCO. Particularly, the surface of $LaAlO_3$ tends to react with water humidity. The high-energy-level electrons generated at this surface can still transfer across $LaAlO_3$ into YBCO.[31] Therefore, it is reasonable that the net enhancement of superconductivity by capping $SrTiO_3$ and $LaAlO_3$ is relatively weak. The broad transition region observed in $La_{1.56}Sr_{0.44}CuO_4$-, $SrTiO_3$-, and $LaAlO_3$-capped YBCO samples is a signature of inhomogeneity of superconducting region. It might be related to the capping-induced local inhomogeneity in structure, charge transfer, and oxidization.

Finally, let us determine what is the thinnest YBCO film that is directly grown on $SrTiO_3$ and still shows superconductivity. Here, PBCO is used as the capping material because it enhances the superconductivity most effectively (Fig. 3). Figure 4 shows the $R(T)$ curves of PBCO-capped YBCO thin films of different thicknesses. As expected, the superconducting properties in all these films are significantly enhanced. The capped 3-uc YBCO still shows a complete superconducting transition. But the capped 2-uc YBCO only shows an onset of superconductivity transition. Therefore, a YBCO film of at least 2 uc–3 uc is necessary for back gating. Although it is still quite challenging to gate this YBCO with the $SrTiO_3$ substrate, Mannhart *et al.*[13,14] have demonstrated a revised back-gating structure in which much larger tuning range can be obtained through the pre-deposited $SrTiO_3$ film. In addition, our present study suggests that depositing gating materials on the top of YBCO will enhance, rather than reduce, its superconducting properties. Therefore, dual gating, from both back and top, could be a useful strategy to control ultrathin YBCO by an electrical field.

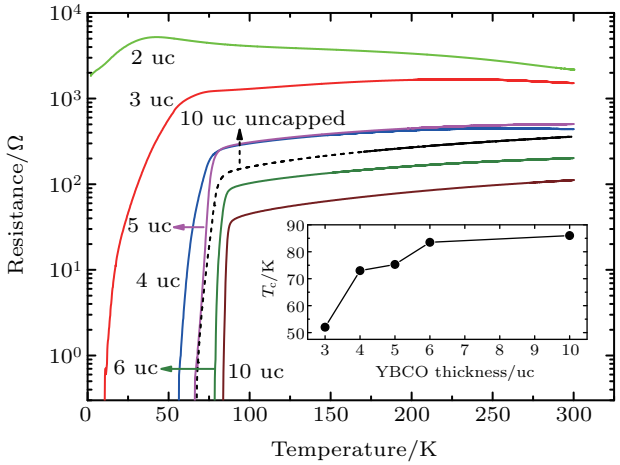


Fig. 4. The dependence of resistance on temperature of PBCO (∼ 7 nm)-capped YBCO thin films of different thicknesses. The inset shows the onset of T_c, determined as the position of the maximum of the derivative curve, $dR(T)/dT$, for the PBCO-capped YBCO thin films. The postannealing time for all samples is 30 minutes. The "10-uc uncapped" sample (dashed line) is regenerated from Fig. 2 for comparison.

4. Summary

In summary, we have studied the effect of both the $SrTiO_3$ termination and the nonsuperconducting capping layer on the properties of ultrathin YBCO films grown directly on $SrTiO_3$. Our result demonstrates that the superconducting properties of the ultrathin YBCO films are insensitive to the termination of $SrTiO_3$, but can be significantly enhanced by surface capping. This enhancement effect is very general and even applicable to dielectric materials. The minimum thickness of the superconducting YBCO ultrathin film obtained by capping is ∼ 2 uc–3 uc. This result may be of interest for designing more effective YBCO-based field effect devices.

References

[1] Tao X and Jian X 2017 *Physics* **46** 514 (in Chinese)

[2] Ren M Q, Yan Y J, Zhang T and Feng D L 2016 *Chin. Phys. Lett.* **33** 127402

[3] Terashima T, Shimura K, Bando Y, Matsuda Y, Fujiyama A and Komiyama S 1991 *Phys. Rev. Lett.* **67** 1362

[4] Norton D P and Lowndes D H 1993 *Appl. Phys. Lett.* **63** 1432

[5] Triscone J M, Fischer, Brunner L, Antognazza L and Kent A D 1990 *Phys. Rev. Lett.* **64** 804

[6] Lowndes D H, Norton D P and Budai J D 1990 *Phys. Rev. Lett.* **65** 1160

[7] Li Q, Xi X X, Wu X D, Inam A, Vadlamannati S, McLean W L, Venkatesan T, Ramesh R, Hwang D M, Martinez J A and Nazar L 1990 *Phys. Rev. Lett.* **64** 3086

[8] Chan I N, Vier D C, Hasen J, Guimpel J, Schultz S and Schuller I K 1993 *Phys. Lett. A* **175** 241

[9] Kwon C, Li Q, Takeuchi I, Warburton P A, Doughty C, Mao S N, Xi X X and Venkatesan T 1996 *Physica C* **266** 75

[10] Mannhart J 1996 *Supercond. Sci. Technol.* **9** 49

[11] Ahn C H, Triscone J M and Mannhart J 2003 *Nature* **424** 1015

[12] Arpaia R, Golubev D, Baghdadi R, Ciancio R, DraŽić G, Orgiani P, Montemurro D, Bauch T and Lombardi F 2017 *Phys. Rev. B* **96** 064525

[13] Mannhart J, Bednorz J G, Müller K A and Schlom D G 1991 *Z. Phys. B: Condens. Matter* **83** 307

[14] Mannhart J, Schlom D G, Bednorz J G and Muller K A 1991 *Phys. Rev. Lett.* **67** 2099

[15] Matthey D, Reyren N, Triscone J M and Schneider T 2007 *Phys. Rev. Lett.* **98** 057002

[16] Xi X X, Li Q, Doughty C, Kwon C, Bhattacharya S, Findikoglu A T and Venkatesan T 1991 *Appl. Phys. Lett.* **59** 3470

[17] Kawahara T, Suzuki T, Komai E, Nakazawa K, Terashima T and Bando Y 1996 *Physica C: Supercond.* **266** 149

[18] Crassous A, Bernard R, Fusil S, Bouzehouane K, Briatico J, Bibes M, Barthelemy A and Villegas J E 2013 *J. Appl. Phys.* **113** 024910

[19] Ahn C H, Gariglio S, Paruch P, Tybell T, Antognazza L and Triscone J M 1999 *Science* **284** 1152

[20] Crassous A, Bernard R, Fusil S, Bouzehouane K, Le Bourdais D, Enouz-Vedrenne S, Briatico J, Bibes M, Barthélémy A and Villegas J E 2011 *Phys. Rev. Lett.* **107** 247002

[21] Horide T, Matsufuji T and Matsumoto K 2015 *IEEE Trans. Appl. Supercond.* **25** 1800204

[22] Fête A, Rossi L, Augieri A and Senatore C 2016 *Appl. Phys. Lett.* **109** 192601

[23] Leng X, Garcia-Barriocanal J, Bose S, Lee Y and Goldman A M 2011 *Phys. Rev. Lett.* **107** 027001

[24] Khanof S, Mannhart J and Boschker H 2018 *Appl. Phys. Lett.* **113** 022605

[25] Nakagawa N, Hwang H Y and Muller D A 2006 *Nat. Mater.* **5** 204

[26] Ohtomo A and Hwang H Y 2004 *Nature* **427** 423

[27] Wang X R, Li C J, Lü W M, Paudel T R, Leusink D P, Hoek M, Poccia N, Vailionis A, Venkatesan T, Coey J M D, Tsymbal E Y, Ariando and Hilgenkamp H 2015 *Science* **349** 716

[28] Huijben M, Koster G, Blank D H A and Rijnders G 2008 *Phase Transit.* **81** 703

[29] Zhai H Y and Chu W K 2000 *Appl. Phys. Lett.* **76** 3469

[30] Wu J, Pelleg O, Logvenov G, Bollinger A T, Sun Y J, Boebinger G S, Vanević M, Radović Z and Božović I 2013 *Nat. Mater.* **12** 877

[31] Zhang M, Chen Z, Mao B, Li Q, Bo H, Ren T, He P, Liu Z and Xie Y 2018 *Phys. Rev. Mater.* **2** 065002

[32] Sefrioui Z, Varela M, Peña V, Arias D, León C, Santamaría J, Villegas J E, Martínez J L, Saldarriaga W and Prieto P 2002 *Appl. Phys. Lett.* **81** 4568