ELSEVIER

Physica C 224 (1994) 179–184

PHYSICA C

Electric-field effect devices made of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{SrTiO}_3$ epitaxial multilayers

K. Joosse *, Yu.M. Boguslavskij, G.J. Gerritsma, H. Rogalla

University of Twente, Department of Applied Physics, PO Box 217, 7500 AE Enschede, The Netherlands

J.G. Wen

National Center for HREM, Laboratory of Materials Science, Delft University of Technology, Rotterdamseweg 137, 2628 AL Delft, The Netherlands

A.G. Sivakov

B. Verkin Institute for Low Temperature Physics and Engineering, Academy of Sciences of Ukraine, 47 Lenin ave., Kharkov 310164, Ukraine

Received 6 July 1993; revised manuscript received 26 February 1994

Abstract

Three terminal superconducting electric-field effect devices, consisting of a bufferlayer of $\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$, an ultrathin layer of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and a SrTiO_3 gate isolation layer were fabricated and successfully operated. Transmission electron microscopy and laser scanning microscopy showed that all layers are highly epitaxial and uniform over the device area. This is essentially important in the analysis of the mechanism of the electric field effect and for the reproducible fabrication of devices. With a 5 nm thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ layer and an applied electric field of 0.85 MV/cm the critical current was decreased by 5% at low temperatures and up to 36% close to T_c . Also enhancement was obtained.

1. Introduction

Superconducting three-terminal devices can be realized by changing the carrier density of the surface layer of a superconducting thin film by applying a strong electric field perpendicular to the surface of the layer [1]. Although several groups already reported on electric-field effects in thin layers of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO), not much consensus has been reached on the physical mechanisms causing the observed effects. Mannhart et al. argue that the su-

percurrent in their samples is limited by depinning, and conclude that the modulation of the critical current is caused by a change in the vortex pinning potential [2]. Walkenhorst et al. indicate that thermal fluctuations in ultrathin superconducting films give rise to a Kosterlitz–Thouless transition, which can be tuned by the electric field [3]. This mechanism has also been worked out theoretically [4]. Matsui et al. interpret their data by assuming that the effective thickness of the superconducting channel is determined by the electric field, which influences the critical current and the flux-flow resistance [5]. Finally, we proposed a combined effect on both the pinning

* Corresponding author.

potential and the Kosterlitz–Thouless vortex–anti-vortex interaction, where the former mechanism is mainly important at low temperatures and the latter dominates around T_c [6]. All these conclusions are mainly drawn from an analysis of the electric-field dependent current–voltage (I – V) characteristics of the YBCO thin film. However, the electric-field effect is much more pronounced in YBCO films which contain many weak links [7]. Consequently, the presence of grain boundaries in the sample due to island-like growth, screw dislocations, stress effects etc., can easily mask the electric-field effect on the single-crystalline YBCO itself. Also thickness variations of the YBCO layer will prevent a correct interpretation if not taken into account. For a correct interpretation of electric-field effect experiments it is important to have a “clean” system, consisting of perfectly epitaxial multilayers. From a practical point of view this will enable one to achieve a more reproducible fabrication of electric-field effect devices.

In this paper we analyze the epitaxy and uniformity of the subsequent layers of the device by transmission electron microscopy and laser scanning microscopy. Secondly we will show some electric-field effect measurements on these devices.

2. Microstructure of the devices

The fabrication of our PBCO/YBCO/SrTiO₃/Au multilayers and device structures has been described previously [8]. Briefly, the YBCO films, in-situ covered by SrTiO₃, were prepared by off-axis magnetron sputtering. Electrical contacts to the ultrathin YBCO film were made via a ramp-type connection to a thick YBCO layer on which the gold contacts were deposited. Polymethylmethacrylate (PMMA) was used to isolate the electrical leads to the gate electrode from the sides of the YBCO layer. These methods enabled the reproducible fabrication of electric-field effect devices on a small scale. The area of the superconducting bridge is typically $20\text{--}200 \times 50 \mu\text{m}^2$, and no complications are expected in further scaling down the dimensions. It was shown that the interdiffusion between the layers is negligible and that a PBCO (PrBa₂Cu₃O_{7-x}) buffer layer considerably improved the properties of the YBCO ultrathin films. In ref. [8] also the electrical properties of the differ-

ent layers were discussed. Here we focus on the uniformity and epitaxial growth of the layers, as investigated with transmission electron microscopy (TEM) and laser scanning microscopy.

Fig. 1 shows a low-magnification TEM image of 3–5 unit cells PBCO, 2–3 unit cells YBCO and 150 nm SrTiO₃, grown on a SrTiO₃ substrate. This picture covers a length of ~ 250 nm, and similar images were made at different places. This shows that all layers are excellently uniform in thickness and structure. Laser scanning microscopy (LSM) [9] confirms this for larger length scales. With this method, the structure is locally heated by a focussed laser beam, while the voltage is measured over the current-biased sample. The sample is below the transition temperature, and biased slightly above the critical current. This technique has proven to be a suitable method to visualize inhomogeneities of the transition temperature or the critical current density due to stress, impurities, grain boundaries etc. The resolution of this method is close to $1 \mu\text{m}$ at this moment. Fig. 2 shows a LSM image of our sample, consisting of 10 nm PBCO, 10 nm YBCO and 50 nm SrTiO₃ and structured into a $100 \times 50 \mu\text{m}^2$ bridge. The response indicates that the current density varies over the sample width. This distribution is perfectly uniform over the sample length.

We conclude that the layers grow layer-by-layer, since the coalescence of growth islands, and also screw dislocations, would certainly give rise to grain boundaries and thickness variations that would be visible on the length scales measured by low-resolution TEM or LSM.

We also performed high-resolution TEM to analyze the epitaxial growth and the presence of grain boundaries at a small scale. However, during the preparation of the TEM samples by conventional ion-milling, the ultrathin YBCO layers sandwiched between the SrTiO₃ substrate and the SrTiO₃ toplayer are found to very easily become amorphous. Only in the case of ultrathin YBCO films grown on YSZ this could be avoided, using special techniques, like gold-coating the cross sections after grinding, to provide a better heat removal.

Fig. 3 shows a HRTEM image of a sample prepared in this way. It is a 8–9 nm YBCO film covered with SrTiO₃, on a YSZ substrate. A BaZrO₃ reaction layer is seen between the substrate and the YBCO film

Fig. 1. Low-magnification TEM image of 3–5 unit cells PBCO, 2–3 unit cells YBCO and 150 nm SrTiO_3 on a SrTiO_3 substrate. The YBCO layer has become amorphous during the sample preparation.

Fig. 2. Laser scanning microscopy showing the voltage response over a current-biased YBCO bridge after local laser beam heating.

[10]. Partly because of this reaction layer, YSZ is not a suitable substrate for the preparation of ultrathin YBCO layers. Still this picture can be used to analyze the interface between YBCO and the sputtered SrTiO_3 layer. The growth is epitaxial with a sharp boundary between the layers, although the interface between the two sputtered layers seems to be more defect-rich than the rest of the layers. The Cu–O planes appear straight and continuous in this image, with no grain boundaries present, but we should keep in mind that this is a very local picture, and that the growth of YBCO depends strongly on the substrate.

Unfortunately, no good ion-milled sample could be obtained with a SrTiO_3 substrate. After cleaving a

grinded sample into a wedge-like shape, TEM can be performed at the edge of the sample. Fig. 4 shows a sample prepared in this way, consisting of 3–5 unit cells PBCO, 2–3 unit cells YBCO and 150 nm SrTiO_3 on a SrTiO_3 substrate, and taken in the [100] direction. Only the region very close to the edge of the sample, roughly indicated by the arrows, should be considered. Outside this area the wedge is too thick to draw conclusions from this TEM image. Some imperfection may be seen at the interface with the substrate, which stresses the advantage of using a buffer layer.

From Figs. 3 and 4 we can also conclude that the first 10–20 nm of the SrTiO_3 layer are practically defect-free. Then, there is a rather abrupt change towards a more defect-rich type of growth. This behavior was observed in all samples that were analyzed with TEM. From a practical point of view, the perfect growth of the SrTiO_3 layer directly from the interface is interesting. Electrical breakdown of the gate isolator is likely to start at imperfections, like grain boundaries or pinholes. The absence of such imperfections will probably enable one to use a very thin gate-isolator layer, which of course increases the voltage gain in these devices. By using energy dispersive

Fig. 3. HRTEM image of 8–9 nm YBCO on a YSZ substrate, taken in the YBCO [100] direction.

analysis of X-rays (EDX) the chemical composition of the layers could be locally determined. It was found that in the defect-free part of the SrTiO_3 layer Ti was partially substituted by Cu. It is, however, unclear what the effect of this doping is on the insulating properties of the gate-isolation layer.

3. Electric-field effect measurements

We used a standard four-point structure to measure the I – V characteristics of the YBCO bridges. The measurements were performed in an Oxford CF1200 continuous flow cryostat. The gate voltage was applied between the gate electrode and a sixth terminal with a low ohmic contact to the superconducting bridge. The leakage current through the gate-isolator was simultaneously measured and kept below 100 nA to avoid a large influence on the measurements. As the I – V characteristics of the SrTiO_3 layer are strongly asymmetric, this condition implies that the highest allowed gate voltages are +6 V and –2 V. The dielectric constant of the SrTiO_3 layer was determined from a capacitance measurement and was found to

be ~ 100 . Thus, the application of gate voltages of +6 V and –2 V corresponds to a charging of the YBCO surface of $-4.8 \times 10^{13} \text{ cm}^{-2}$ and $+1.6 \times 10^{13} \text{ cm}^{-2}$, respectively. The excess charge is accumulated within a thin layer characterized by the charge screening length of $4.5 \text{ \AA}$ from the YBCO/ SrTiO_3 interface [11]. Averaged over the total film thickness, the change in the carrier density is –2.1% and +0.7%, assuming $n = 4.5 \times 10^{21} \text{ cm}^{-3}$ in (bulk) YBCO. We observed electric-field effects in all samples in which the thickness of the YBCO film was 10 nm or less. A sample consisting of 10 nm PBCO, 5 nm YBCO (4–5 unit cells) and 70 nm SrTiO_3 had a T_c^{zero} of 40 K and J_c at 6 K was as high as 10^6 A/cm^2 , the latter being another indication for the absence of weak links. Fig. 5 shows a typical set of gate-voltage dependent I – V characteristics. By applying a bias voltage of +6 V and –2 V to the gate electrode, I_c was suppressed and enhanced by –5% and +1.8% at low temperatures, and –36% and +16% close to T_c , as shown in Fig. 6. Above T_c , the normal-state resistance increases with a positive gate voltage and decreases with a negative voltage. For the above mentioned gate voltages the change in the resistance was +50% and

Fig. 4. HRTEM image of 3–5 unit cells PBCO, 2–3 unit cells YBCO and 150 nm SrTiO_3 on a SrTiO_3 substrate, cleaved into a wedge-shaped TEM sample. Only the first few nm from the edge should be considered, as indicated by the arrows.

Fig. 5. I - V curves of a 5 nm YBCO film at zero applied electric field, ($\square$) at $V_G = +6$ V (Δ) and at $V_G = -2$ V ($\triangleright$), at $T = 20$ K.

Fig. 6. Relative change of the critical current I_c and the normal-state resistance R_n below and above T_c , respectively, upon applying gate voltages of +6 V and -2 V.

–20% in the vicinity of T_c , and decreased to saturation values of +0.8% and –0.3% above 80 K (see Fig. 6). Qualitatively, the temperature dependence of $\Delta I_c(V_G)$ and $\Delta R_N(V_G)$ agrees with measurements performed by others [12,13]; however, the magni-

tude of the electric-field effect is smaller. For example, we measured $\Delta I_c/I_c = -5\%$ for $\Delta N = -4.8 \times 10^{13} \text{ cm}^{-2}$ at $T = 6 \text{ K}$, compared to $\Delta I_c/I_c = -16\%$ for $\Delta N = -1.0 \times 10^{14} \text{ cm}^{-2}$ in a 5 nm thin film at $T = 5 \text{ K}$ in [12] and even $\Delta I_c/I_c = -50\%$ for $\Delta N = -4 \times 10^{13} \text{ cm}^{-2}$ in a 7 nm thin film at 4.1 K in ref. [13]. We assume that the thickness-homogeneity of our samples and the absence of weak links are the reason for the reduced electric-field effects compared to those reported by others.

4. Conclusions

Electric-field effect devices were made consisting of $\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}/\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{SrTiO}_3$ multilayers. Transmission electron microscopy and laser scanning microscopy showed that these layers were highly epitaxial and uniform over the device area. This means that the measurements on these samples may indeed be interpreted as electric-field effect measurements on epitaxial YBCO thin films, rather than on grain boundaries. Therefore these devices are expected to be suitable for the investigation of the mechanisms of the electric-field effect in high- T_c thin films and for a reproducible fabrication of high- T_c three-terminal devices. Electric-field effects were observed in all devices in which the thickness of the YBCO layer was 10 nm or less.

Acknowledgements

These investigations were performed in the program of the Foundation for Fundamental Research on Matter (FOM) and have been supported by the Netherlands Technology Foundation (STW).

References

- [1] R.E. Glover and D.M. Sherrill, *Phys. Rev. Lett.* 5 (1960) 248.
- [2] J. Mannhart, D.G. Schlom, J.G. Bednorz and K.A. Müller, *Phys. Rev. Lett.* 67 (1991) 2099.
- [3] A. Walkenhorst, C. Doughty, X.X. Xi, Q. Li, C.J. Lobb, S.N. Mao and T. Venkatesan, *Phys. Rev. Lett.* 69 (1992) 2709.
- [4] B.Ya. Shapiro, preprint.
- [5] K. Matsui, T. Awaji, T. Hirano, T. Fujii, K. Sakuta and T. Kobayashi, *Jpn. J. Appl. Phys.* 31 (1992) L1342.
- [6] K. Joosse, Yu.M. Boguslavskij, G.J. Gerritsma and H. Rogalla, *Appl. Supercond.* 2 (1993) 861 (proceedings EUCAS '93).
- [7] J. Mannhart, J. Ströbel, J.G. Bednorz and Ch. Gerber, *Appl. Phys. Lett.* 62 (1993) 630.
- [8] K. Joosse, Yu.M. Boguslavskij, G.J. Gerritsma and H. Rogalla, *J. Alloys and Comp.* 195 (1993) 723.
- [9] V.A. Konovodchenko, A.G. Sivakov, A.P. Zhuravel, V.G. Efremenko and B.B. Banduryan, *Cryogenics* 26 (1986) 531.
- [10] J.G. Wen, C. Træholt, H.W. Zandbergen, K. Joosse, E.M.C.M. Reuvekamp and H. Rogalla, *Physica C* 218 (1993) 29.
- [11] J. Mannhart, *Mod. Phys. Lett. B* 6 (1992) 555.
- [12] X.X. Xi, C. Doughty, A. Walkenhorst, S.N. Mao, Qi Li and T. Venkatesan, *Appl. Phys. Lett.* 61 (1992) 2353.
- [13] J. Mannhart, J.G. Bednorz, K.A. Müller, D.G. Schlom and J. Ströbel, *J. Alloys and Comp.* 195 (1993) 519.