

High quality $\text{YBa}_2\text{Cu}_3\text{O}_x$ ultra-thin films and Y/Pr/Y multilayers made by a modified RF-magnetron sputtering technique

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High quality \hat{c} -axis oriented thin and ultra-thin films have been grown in situ on (100) surfaces of ZrO_2 , SrTiO_3 and MgO . Sharp transitions were observed with $T_{c,zero}$ of 87–91 K for films thicker than 70 Å. On atomically polished MgO substrates films as thin as 15 Å revealed a full transition to superconductivity above 45.5 K. The critical current density at 77 K was found to be strongly dependent on film thickness. A maximum value was found for a 100 Å film with $8 \times 10^6 \text{ A/cm}^2$ at 77 K. Also, YBCO/PBCO/YBCO multilayer thin films have been fabricated in situ by the same technique. The epitaxy is maintained throughout the whole multilayer system. The superconducting properties of $\text{YBa}_2\text{Cu}_3\text{O}_x$ layers do not change compared to single layers. Interdiffusion and possible chemical reaction close to the interfaces can be neglected.

1. Introduction

Due to its importance for both basic research as well as applications much effort has been spent on the preparation and study of high T_c superconducting thin films in the last few years. Much attention has been spent to obtain thin films with high critical current density at the boiling temperature of liquid nitrogen and offering a smooth film surface. Potential applications of such thin films in superconducting electronics require high quality multilayers of superconducting and non-superconducting materials. For this, the epitaxial growth should be maintained throughout the multilayer system without detectable interdiffusion. In addition, the properties of the superconducting layers should not be reduced too much. Recently some groups reported possible heterostructures on the basis of isomorphic $\text{ReBa}_2\text{Cu}_3\text{O}_x$ (Re: rare earth element) [1,2]. In this paper we present results on the in situ preparation of epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_x$ ultra-thin and $\text{YBa}_2\text{Cu}_3\text{O}_x/\text{PrBa}_2\text{Cu}_3\text{O}_x/\text{YBa}_2\text{Cu}_3\text{O}_x$ (Y/P/Y) multilayer films by a modified off-axis RF magnetron sputtering technique.

2. Experimental

The modified off-axis sputter configuration is sketched in fig. 1. A ground plate was mounted, facing the target at a 45° angle. This geometry makes the direct substrate bombardment improbable. Similar to ion beam sputtering, the sputter parameters can be controlled independently. Stoichiometric sintered $\text{YBa}_2\text{Cu}_3\text{O}_x$ (YBCO) and $\text{PrBa}_2\text{Cu}_3\text{O}_x$ (PBCO) targets with a diameter of 50

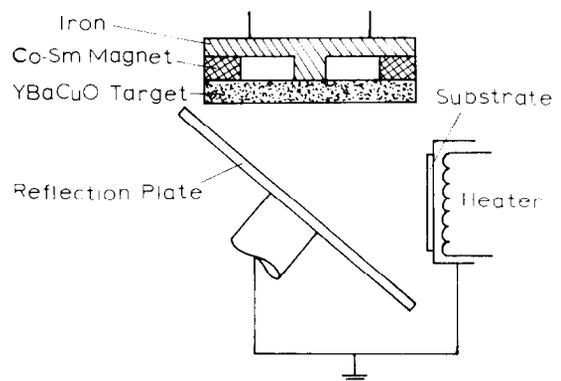


Fig. 1. Schematic diagram of the modified off-axis RF-magnetron sputtering system.

mm were used. For the substrates, (100) surfaces of ZrO_2 , SrTiO_3 and MgO have been used. The sputtering gas was a mixture of argon and oxygen which consisted, in most cases, of 50% oxygen and 50% argon. Typical sputter pressures are between 5×10^{-2} and 3×10^{-1} mbar. During deposition the substrates were heated to temperatures between typically 640 and 700 °C. The incident RF sputter power was between 50 and 100 W.

To determine resistivities and critical currents the films have been structured into small bridges of 100 μm length and 10 μm width. Both, argon ion beam sputtering and wet chemical etching in diluted phosphoric acid with photoresist masks as well as single-shot laser ablation using suitable metal masks have been used, all techniques causing less than 1 K reduction of T_c .

3. Results and discussions

By means of the modified off-axis sputter process, reproducible superconducting films with a $T_{c,\text{zero}}$ of about 90 K have been obtained on ZrO_2 , SrTiO_3 and MgO (100) oriented single crystalline substrates. Usually the width of the resistivity transition ΔT (10%–90%) is less than 3 K. Above the superconducting transition temperature the films showed linear metallic behaviour with resistance ratios $R(300\text{ K})/R(100\text{ K})$ (RRR) between 2.7 and 3.2. Normal state resistivities at 100 K (film thickness > 100 Å) are typically between 35 and 100 $\mu\Omega\text{ cm}$, which is comparable to that of the single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_x$ [3].

All the films made in this way have mirror-like surfaces. No structure could be seen as investigated with a scanning electron microscope (SEM). The cross section, analysed by transmission electron microscopy (TEM), clearly presents a well defined interface between film and substrate. No signs of a pronounced diffusion could be observed.

The main advantage of this deposition technique is that it is especially suitable to make high quality ultra-thin films. As shown in fig. 2, an ultra-thin film with a thickness of only 15 Å deposited on an atomically polished (100) surface of MgO showed rather good superconducting

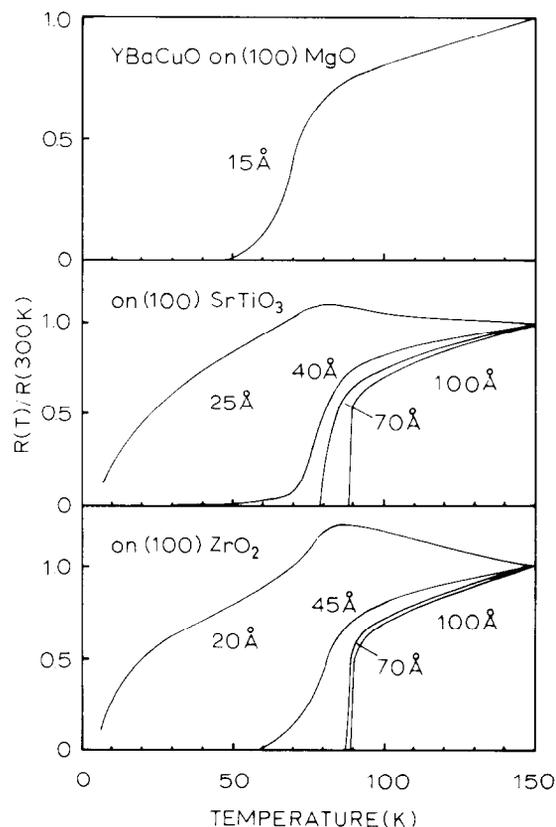


Fig. 2. Normalized transition curves of $\text{YBa}_2\text{Cu}_3\text{O}_x$ films of various thicknesses on different substrates.

properties with $T_{c,\text{onset}}$ and $T_{c,\text{zero}}$ of 84 and 45.5 K, respectively. For films on ZrO_2 , with thicknesses down to 70 Å, no variation in the superconducting transition temperature has been found and 87–91 K transitions have been achieved. On a film with a thickness of 45 Å the $T_{c,\text{zero}}$ drastically decreased to 58 K. A film of 20 Å thickness showed a semiconducting behaviour and no reasonable transition could be observed above 4.2 K. For SrTiO_3 substrate, the results are similar to that of ZrO_2 .

The film thickness dependence of the critical current density at 77 K of films with $T_{c,\text{zero}}$ between 87 and 90 K on ZrO_2 substrates is shown in fig. 3. A value of $J_c(77\text{ K}) = 8 \times 10^6\text{ A/cm}^2$ was achieved on a film with a thickness of 100 Å. In the thickness range of 1000–100 Å it was found that the critical current density at 77 K increases with decreasing thickness, being almost inversely proportional to the film thickness. This suggest

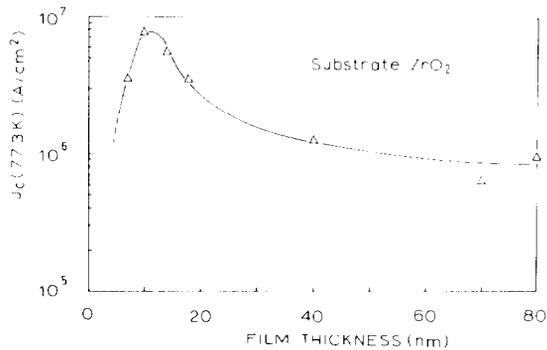


Fig. 3. Critical current density at 77 K of films on ZrO_2 as a function of film thickness. A flux flow resistance criterion of 1×10^{-6} of the normal state resistance was used.

that the high critical current density is caused by lattice defects at the interface between substrate and film. As shown in fig. 3, when the film thickness is over 400 Å, $J_c(77\text{ K})$ decreases fast. In this case the bulk pinning is believed to play a dominant role.

The influence of the sputter parameters have been investigated carefully. Changing the oxygen partial pressure has almost no effect on T_c , as long as the partial pressure of 10 Pa is exceeded. Varying the sputter power density from 8 to 1 W/cm^2 has very little influence on T_c . Also the substrate temperature was changed over a wide range (from 600 to 740 °C). It turned out that the transition temperature was independent of the substrate temperature if the latter was above 650 °C. An upper bound could not be found due to the limited heater temperature. Details of this study are published elsewhere [4].

For the preparation of Y/Pr/Y multilayers, both YBCO and PBCO thin films have been deposited in situ. To avoid contamination of the interfaces of the sandwich structures, the substrates could be turned under different targets without breaking the vacuum. The deposition was followed by an anneal at 400 °C in 1 bar oxygen.

In fig. 4 the superconducting transitions of YBCO top and bottom layers are shown. For the top layer $T_{c,zero}$ was measured to be 91 K. Then the top and middle layers were sputtered away by an argon ion beam. The transition temperature of the bottom layer was found to be 90.1 K. Obviously the superconducting properties of YBCO

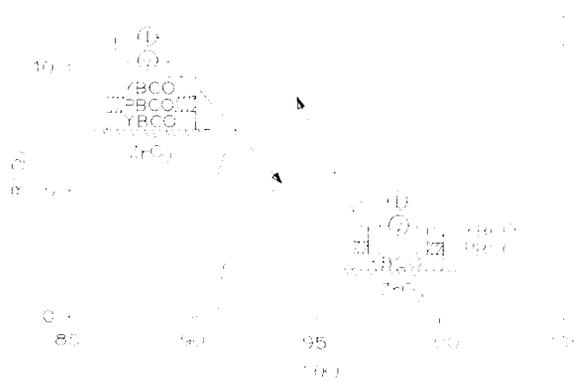


Fig. 4. Superconducting transitions of YBCO top and bottom layers of the same sample. For the latter measurement the second PBCO and the third YBCO layers were sputtered away by an Ar-ion beam.

layers are not affected by the absence of the PBCO middle layer.

X-ray diffraction was performed to determine the phases formed in the trilayer films. Besides the substrate's reflections, only (001) peaks are present. The full-width at half-maximum of the (005) peak was observed to be 0.35° . It is somewhat broader than that we found for single films (for YBCO thin films: $\Delta\theta_{50} = 0.28^\circ$; for PBCO thin films: $\Delta\theta_{50} = 0.30^\circ$), but still indicates that these multilayers are heteroepitaxially grown. In fig. 5

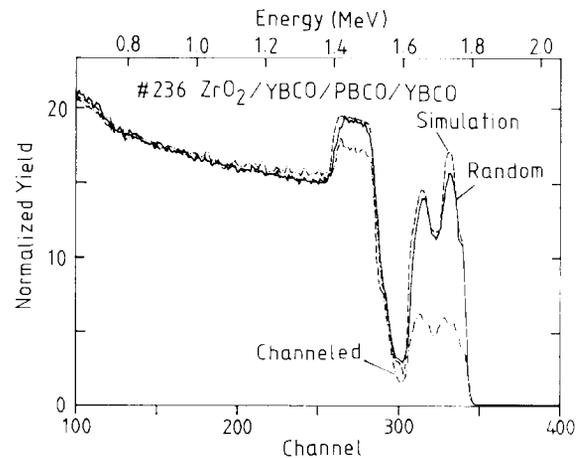


Fig. 5. RBS random and channeling spectra (2.0 MeV He ions) for a Y/P/Y trilayer sample with film thickness of 40, 53, and 107 nm, respectively, from substrate to top. The simulation curve fit to the random spectrum indicates good overall stoichiometry.

RBS and ion channeling measurements on a trilayer sample with 2 MeV He ions are presented. The broken line is a RBS computer simulation of a layer sequence of 40 nm YBCO, 53 nm PBCO and 107 nm YBCO. The simulation curve fits well to the experimental data. Within the depth resolution limits of RBS of about 5 nm, no interdiffusion between layers could be observed. A minimum yield of 37% throughout all three layers and substrate has been measured by the He ions channeling along the (001) direction. Also, similar results have been obtained by Auger sputter profiling of a trilayer with a 10 nm thick PBCO layer. Within the experimental resolution there is no interdiffusion visible between Y and Pr.

The surfaces of PBCO and YBCO top layer have been investigated by SEM on Y/P bilayer and Y/P/Y trilayer samples. Both showed very smooth surfaces. No pin-holes could be found. Thus these heterostructures provide a promising basis for superconducting electronic applications.

4. Conclusions

We have shown that the modified RF-magnetron sputtering process is a versatile and simple technique for the reliable production of high quality ultra-thin films of $\text{YBa}_2\text{Cu}_3\text{O}_x$. The dependence of J_c upon film thickness indicated that film surface or the interface with the substrate is important for flux-line pinning. High quality Y/P/Y multilayers have been made in situ by the same technique as well. It was found that the superconducting properties of YBCO layers do not change if a PBCO layer is present. No interdiffusion between Y and Pr could be observed by RBS and Auger depth profiling.

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