High $J_c$ YBCO thin films and multilayers grown by chemical solution deposition

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Abstract. Chemical solution deposition (CSD) has emerged as a very competitive technique to obtain superconducting films of high quality. However, there is still few knowledge about how underlying microstructure can affect the performances of the YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) films. Here we will summarize our recent progress in this subject. We have grown YBCO thin films by the so called trifluoroacetate route on top of buffer layers also grown by CSD. By modifying the growth conditions of SrTiO$_3$ and BaZrO$_3$ buffer layers we have investigated the influence of surface roughness and grain size. The role of lattice parameter mismatch has been studied by producing multibuffered architectures as SrTiO$_3$/BaZrO$_3$/LAO. It has been proved that YBCO thin films with critical currents in excess of 1MA/cm$^2$ at 77K in self field can be achieved, thus demonstrating the abilities of the CSD technique

INTRODUCTION

Coated conductors have emerged as potential conductors for high-temperature high-field applications [1] although their spread use is linked to developing competitive low-cost deposition methods. One of the most promising architectures consists of YBa$_2$Cu$_3$O$_7$ (YBCO) films prepared by the trifluoroacetates (TFA) route [2,3] onto buffer layers grown by chemical solution deposition (CSD). However, due to their particular ex-situ growth mechanisms, CSD layers usually exhibits higher roughness and porosity than vacuum-processed layers and the feasibility of such a CSD multilayered epitaxial structure needs to be demonstrated.

In this work we have studied the growth of YBCO thin films by the so-called trifluoroacetates route onto CSD oxide layers. We have concentrated on the relationship between the buffer oxide microstructure and the final superconducting properties of the YBCO film. By growing different heterostructures we have investigated the influence of parameters such as porosity, roughness and lattice mismatch. Our results show that growth of epitaxial multilayers is also feasible by CSD leading to high critical current superconducting thin films.

EXPERIMENTAL DETAILS

The samples studied in this work are epitaxial c-axis oriented YBCO thin films grown by chemical solution deposition onto SrTiO$_3$-buffered LaAlO$_3$ and SrTiO$_3$
single crystalline substrates and onto double-buffered (SrTiO$_3$/BaZrO$_3$) LaAlO$_3$ single crystalline substrates. The buffer layers were prepared by spin-coating a methanol-based precursor solution on the substrate and then heating at 700-900°C for 4h in a 95% Ar/5% H$_2$ atmosphere. Typical thicknesses were of 30-40nm for SrTiO$_3$ and 25-30 nm for BaZrO$_3$. The YBCO films were prepared by the TFA-MOD route from methanol-based trifluoroacetate precursor solutions. The coated wafers were calcined in three steps following a heating treatment similar to the one reported in Ref 4 where more details can be found. In the present work the crystallization conditions were chosen to be T=795°C and P(H$_2$O)=0.6% and P(O$_2$)=200ppm. Typical YBCO film thicknesses were 300 nm.

Microstructure of the YBCO films and buffer layers were systematically studied by x-ray diffraction, scanning electron microscopy (SEM), transmission electron microscopy (TEM) and atomic force microscopy (AFM). The temperature dependence of the dc-resistivity of the YBCO films was measured by a usual four-point technique. Critical currents were obtained from inductive measurements (SQUID magnetometer and ac susceptometer).

![Graph](image)

**FIGURE 1.** $\phi$-scans of the three layers grown by CSD in a YBCO/STO/BZO/LAO multilayer. It corresponds to the (202) reflection in the case of the oxide buffer layers and to the (102) reflection in the case of YBCO.

**RESULTS AND DISCUSSION**

X-ray diffraction patterns showed that YBCO layers grow epitaxially on top of the buffered single crystals exhibiting excellent out-of-plane and in-plane texture with FWHM of the $\phi$-scan for the (102) reflection of less than 1° in the case of YBCO/STO$^{\text{CSD}}$/STO. For the double buffer structure YBCO/STO$^{\text{CSD}}$/BZO$^{\text{CSD}}$/LAO it
is interesting to notice that the final in-plane texture of the YBCO films improves that of the underlying buffer layers, $\Delta \phi_{YBCO}^{(102)} = 1.4^\circ$ whereas $\Delta \phi_{STO}^{(202)} = 1.6^\circ$ and $\Delta \phi_{BZO}^{(202)} = 2.1^\circ$ as it is showed in the scans of Figure 1. Another observed result it is that the lattice mismatch among the different layers is preserved and hence the interface stress is fully relaxed at these thicknesses, probably through the creation of misfit dislocations [5]. These results confirmed the high potentiality of CSD to grow epitaxial multilayers.

![Graph](image)

**FIGURE 2.** Temperature dependence of the critical current density (a) and the electrical resistivity (b) for the three architectures here studied.

The slight increase in the YBCO texture spread for the case of the double buffered structure is also reflected in the superconducting properties. Figure 2 shows the temperature dependence of the critical current (a) and of the resistivity (b) for YBCO films grown in the three architectures here analyzed. From the results of this figure several comments are in order. First, chemical solution deposition can lead to epitaxial thin films with high critical currents. The obtained $J_c$ value in the case of YBCO/STO$^{\text{CSD}}$/STO is $J_c(77K) \sim 1 \times 10^6$ A/cm$^2$ which is comparable to that achieved onto single crystalline substrates. Second, there is a slight decrease of $J_c$ and an increase of normal state resistivity in the case of the heteroepitaxial sample and of the double-buffered sample. The enhancement of the resistivity can be attributed to an increase of the sample porosity which reduces the effective cross section of the sample and increases the effective percolation length through the sample [6]. Recently we have shown that this porosity also controls $J_c$ and, in fact, there is a close link between
the critical current density and the normal state resistivity [6]. In the present case, however, besides this increase of porosity, we can also observe that there is some residual resistivity as indicated by the linear extrapolations in Figure 2(b) for the heteroepitaxial and double-buffer samples in agreement with the observed texture values. This could suggest that a second mechanism for the reduction of $J_c$ is present, as for example, a slight loss of grain boundary quality. We should note that preliminary TEM results and SEM observations confirm the presence of some a/b-axis grains and a somewhat deteriorated interface in the double buffered architecture. AFM measurements also show that surface roughness of buffer layer is higher in this case. An improving of the buffer layers quality should lead then to an improvement of superconducting properties.

CONCLUSIONS

We have analyzed the feasibility of YBa$_2$Cu$_3$O$_{7-\delta}$ superconducting multilayers grown by chemical solution deposition (CSD). The studied models are suitable architectures for coated conductors and they consist in YBCO thin films grown by the trifluoroacetates route onto CSD homoepitaxial and heteroepitaxial SrTiO$_3$ buffer layers and onto double buffered, SrTiO$_3$/BaZrO$_3$ buffer layers. We have conclusively shown that heterostructures can be indeed prepared by CSD with very high critical currents.

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