Deposition and characterization of high temperature superconducting YBa$_2$Cu$_3$O$_{7-\delta}$ films obtained by DC magnetron sputtering and thermal annealing modification

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Abstract. C-axis oriented 100-nm thick YBCO films were deposited on LaAlO$_3$ (100) substrates at substrate temperature of 780 °C in a mixed oxygen/argon atmosphere (1:3) of 0.3 Torr by DC off-axis magnetron sputtering. The samples deposited were thermally annealed in oxygen ambient of 600 Torr at 530 °C for 40 min. Superconductivity with zero resistance 89.1K was observed for the YBCO films after annealing. These results show that thermal annealing is an important technique for improving the parameters of thin superconducting films. A correlation between the YBCO layers properties before and after annealing was established.

1. Introduction
Thin superconducting YBCO films have been grown by various techniques including electron beam deposition [1], magnetron sputtering [2,3], laser ablation [4,5].

Post-deposition annealing is often required to overcome the oxygen deficiency in the as-deposited films [6-8].

We describe here the formation of YBCO layers by DC off-axis magnetron deposition and the thermal annealing effect on the transition temperature $T_c$, the critical current density $J_c$, and the films structure and morphology.

2. Experimental details
High-temperature superconducting YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) thin films (~100 nm) were grown on 5 mm $\times$ 10 mm $\times$ 0.5 mm LaAlO$_3$(100) (LAO) substrates using 50 mm diameter stoichiometric YBa$_2$Cu$_3$O$_{7-\delta}$ targets. The deposition was carried out at a total oxygen/argon (1:3) gas pressure of 0.3 Torr and substrate temperature of 780 °C. The typical magnetron current and voltage were 200 mA and 170 V, respectively, which corresponds to a deposition rate of 20 nm/h. Figure 1 shows schematically the magnetron sputtering configuration.

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Following deposition, the films were cooled in oxygen ambient of 600 Torr and held at 530°C for annealing times of 0 and 40 min. The samples were cooled down to room temperature at a rate of 0.38 deg/sec.

The electrical characteristics (the critical temperature, $T_c$, and the critical current density, $J_c$) of the YBCO films were investigated using AC contact-less methods [9], whereby the information about $T_c$ and $J_c$ is obtained from the magnetic response of the film to an applied AC field.

![Figure 1. Magnetron sputtering configuration.](image)

The structure of the films was investigated by using a Bruker D8 Advance Diffractometer with Cu-Kα radiation and SolX detector within the 2θ range 5 - 80 deg and a constant step 2θ of 0.05 deg. The surface morphology of the films was examined using a scanning electron microscope (SEM) Philips 525M. The images were recorded at accelerating voltage of 8 kV.

3. Results and discussion

Figure 2 shows $J_c$ values before and after annealing.

![Figure 2. Critical current density for YBCO films before and after annealing at 530°C.](image)

![Figure 3. Transition temperature for YBCO films before and after annealing at 530°C.](image)
The improvement in the $J_c$ after thermal annealing can be attributed to the better oxygen distribution in the films, leading to an increase of the superconducting carrier concentration. Figure 3 shows $T_c$ values before and after annealing. The increase of $T_c$ after thermal annealing can also be attributed to the better oxygen distribution in the films.

Only the $(00l)$ reflections were observed for all samples in a symmetric 0-20 scan indicating that the films were grown epitaxially with the $c$-axis perpendicular to the substrate surface (figure 4).

The relative intensities of the YBCO $(00l)$ peaks increase with the annealing time, which suggests improvement of the crystallinity.

The $c$ constant was calculated using d-spacing of the $(005)$ peak. The accuracy is estimated as 0.01Å. The $c$ constant for both YBCO films is 11.67Å, which is responsible for oxygen content 6.97 [10]. The crystallites size (mean coherent domain size) calculated from XRD using the Scherrer formula [13] was 35 nm and 50 nm for YBCO film before and after 40 min annealing. The larger crystallites size also favors improvement of electrical properties.

Scanning-electron-microscope (SEM) photographs revealed some “stones” for all samples, which are normal for YBCO films. The origin of these stones is the formation of Y- and Cu- oxides that precipitate on the layer surface during epitaxy. These precipitates are non-conductive and their sizes define the quality of the superconducting layer [11, 12]. After annealing, the size of these stones decreases, which favors the electrical properties of YBCO films (figure 5).
4. Conclusions

Thermal annealing was performed on superconducting YBCO films at 530°C in oxygen ambient of 600 Torr for time periods of 0 and 40 min.

The increase of $J_c$ from 0.633 to 1.446 MA/cm$^2$ and the $T_c$ from 87.5 to 89.1 K for the annealed YBCO film can be attributed to better oxygen distribution. The XRD spectra confirm the better monocrystalline structure for the thermally annealed sample. The SEM images reveal a smaller size of the “stones” in the annealed sample, probably due to partial dissolution in the crystal structure of the film that suggests a superconducting properties improvement.

The results obtained show that thermal annealing in oxygen ambient is an important process in improving the electrical and structural properties of YBCO films.

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