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Effect of thermal annealing on the properties of the YBCO films grown by DC magnetron sputtering♣

M. BESHKOVA*, B. BLAGOEV, D. KOVACHEVA[a], G. MLADENOV, T. NURGALIEV
Institute of Electronics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee Blvd., 1784 Sofia, Bulgaria.
[a] *Institute of General and Inorganic Chemistry, Bulgarian Academy of Sciences, Acad. G. Bonchev str., bl.11, 1113 Sofia, Bulgaria.*

The effect of thermal annealing on the electrical, structural and morphological properties of YBCO ($YBa_2Cu_3O_{7-\delta}$) superconducting films grown on $LaAlO_3$ (100) substrates by DC off axis magnetron sputtering were investigated. The deposited samples were thermal annealed at 530 ºC for 40 min under oxygen pressures of 200 and 500 Torr. The critical temperature (T_c) and current density (J_c) enhance and the transition width of temperature ΔT narrows with increasing oxygen pressure. A higher T_c 89.2 K and J_c 1.38 MA/cm^2 is exhibited for an annealing oxygen pressure of 500 Torr and c-axis length of 11.70 Å which corresponds to an oxygen content of 6.84. The effect of the property variations of YBCO films was established with changes in the annealing oxygen pressure.

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1. Introduction

In spite of the rather advanced status of high-T_c superconducting thin film technology, intensive research still continues to grow better quality films for use in future technological applications [1].

To produce superconducting YBCO films, two stages are needed. The first one is YBCO (123) growing on a suitable substrate. This stage forms a stoichiometric tetragonal $YBa_2Cu_3O_{7-\delta}$ phase with poor oxygen content ($7-\delta < 6.5$) [2]. Tetragonal YBCO is an antiferromagnetic insulating material, and is not superconducting [3]. In the second stage, the YBCO (123) tetragonal phase is converted to a YBCO (123) orthorhombic superconducting phase by an oxidizing procedure (thermal annealing in oxygen ambient) [4-6]. During oxygen annealing, important parameters are the oxygen pressure, the annealing temperature of the substrate and the duration of annealing. The oxidizing procedure increases the oxygen content and it stands at over 6.5 in the (123) orthorhombic phase.

In this work, we investigated the influence of various annealing oxygen pressures on the critical parameters, structure and morphology of submicron YBCO films (~ 100 nm), magnetron sputtered onto $LaAlO_3$ (100) substrates.

2. Experimental

The films (100 nm thickness) were deposited by DC off axis magnetron sputtering on 5 mm × 10 mm × 0.5 mm $LaAlO_3$ (100) using two (situated face to face) stoichiometric YBCO targets. The films were grown at a substrate temperature of 780 °C and an oxygen/argon (1:3) ambient of 0.4 mbar (0.3 Torr). A sketch of the experimental configuration of growth assembly is shown in Fig. 1. The sputtering conditions were described in detail in our previous work [7].

After the deposition was finished, pure oxygen was vented into the growth chamber, in the range 200 or 500 Torr, the samples were cooled to 530 °C at a rate of about ~ 5 deg/sec, maintained at this temperature for 40 min and cooled to room temperature at a rate of 0.38 deg/sec.

AC contactless inductive methods were used to measure the critical temperature – T_c and the critical current density – J_c of the films [8]. According to these methods, information about T_c and J_c is obtained from the magnetic response of the superconducting film to an AC magnetic field. The sample is situated between two flat

♣ Paper presented at the International School on Condensed Matter Physics, Varna, Bulgaria, September 2008 coils. To one of them is applied a sinusoidal magnetic field. The AC magnetic field of the currents induced in the YBCO layer is registered by the other coil. To measure T_c, the basic harmonic is used, and for J_c, the third harmonic. Measurements were made without a DC magnetic field ($B = 0$).

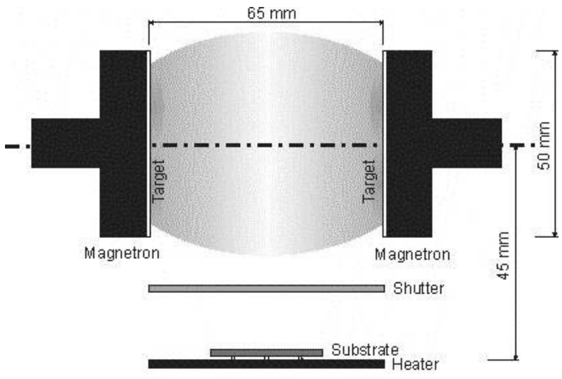

Fig. 1. Magnetron sputtering system

The crystalline structure of the YBCO films were analysed using a Bruker D8 Advance Diffractometer (XRD) with Cu-Kα radiation and a SolX detector.

The surface morphology of the films was studied by Scanning Electron microscopy (SEM), using a JEOL Superprobe 733. The images were recorded at an accelerating voltage of 25 kV.

3. Results and discussion

Fig.2 shows XRD θ-2θ measurements for YBCO films annealed at different oxygen pressures.

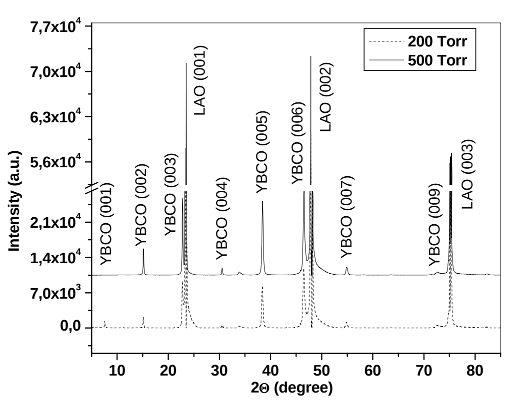

Fig. 2. X-ray diffraction pattern for YBCO films annealed at 200 and 500 Torr oxygen pressures.

The spectrum exhibits only (00*l*) peaks, indicating that the films were grown epitaxially with the c-axis perpendicular to the substrate surface.

With increasing oxygen annealing pressure, a shift of the (005) peak positions is observed, indicating c-axis shortening from 11.72 to 11.70 Å which corresponds to oxygen contents of 6.67 and 6.84 [9, 10].

The full widths at half maximum (FWHM) of the 005 ω-curves also decrease from 94 to 81 arcsec with increasing annealing oxygen pressure, suggesting a reduction in the density of misaligned grains.

The relative intensities of the diffraction peaks increase with increasing annealing oxygen pressure, which clearly reveals an improvement of the crystalline quality.

The results of measurements of the magnetic response of the samples, which allow one to evaluate their J_c and T_c, are shown in Fig. 3 and Fig. 4 respectively.

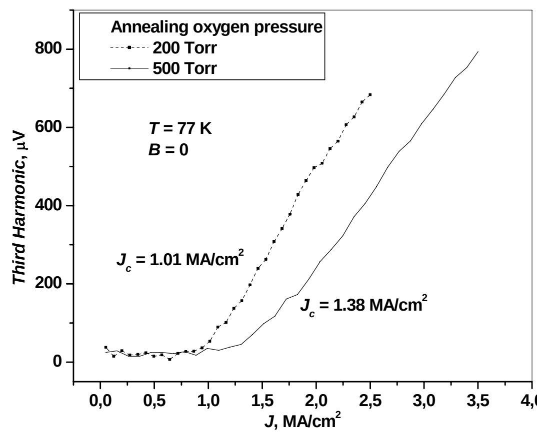

Fig. 3. Dependence of the third harmonic response signal amplitude on the screening current density J induced in YBCO films at 77 K by an AC magnetic field (<5 mT). The third harmonics of the response signal arise at J = 1 MA/cm² and J = 1.4 MA/cm², which correspond to the critical current densities of the samples annealed at 200 and 500 Torr respectively.

It is seen that J_c increases when the films are annealed at a higher oxygen pressure ~ 500 Torr, which can be explained by an increase in the superconducting carrier concentration with improved crystalline structure. In this case, the superconductive (SC) temperature transition is quite abrupt (the transition width of the temperature ΔT, i.e. the width of temperature interval where a transition between superconducting and normal states occurs, is narrower) (Fig.4). This implies a better spatial homogeneity of the sample annealed at 500 Torr. The critical temperature T_c is not affected strongly by the annealing oxygen pressure at p ~ 200 and 500 Torr. With increasing annealing oxygen pressure, the *Tc* value increases, which is attributed to removing the oxygen deficiency.

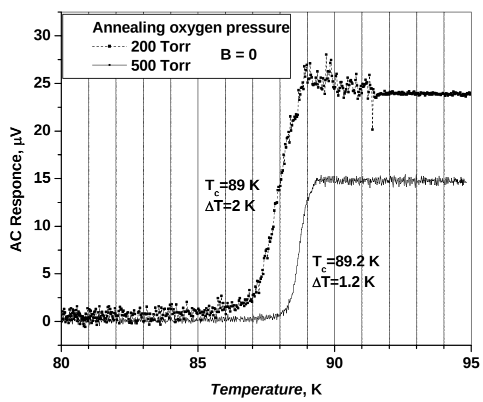

Fig. 4. Temperature dependence of the amplitude of the basic harmonic of the response signal of YBCO films exposed to the action of an AC magnetic field (<0.01 mT). A decrease in the response signal amplitude starts at T = T_c = 89 and 89.2 K, for the samples annealed at 200 and 500 Torr oxygen pressures respectively.

It is known that even epitaxial YBCO films consist of "islands" (i.e. grains) separated by weak links (the separation is between 0.01 and 1 μm) [11] and in any event, currents have to pass through these weak links in a measurement process. For this reason, the critical temperature and current density, measured in our experiments, reflect the T_c and J_c not only of the grains, but the parameters of the weak links as well. It can be noted that in epitaxial YBCO films, the T_c and J_c of such weak links may be great enough [11] and may approach the critical parameters of the grains. The effect of weak links can be seen as steps in the experimental dependence for both samples, as illustrated in Fig. 4.

On the one hand, the enhanced annealing pressure can accelerate the oxidizing process in YBCO materials with large specific surfaces (like thin films) [12], thus yielding films with less oxygen deficiency. On the other hand, in single monocrystals and thick YBCO films at very high values of the oxygen annealing pressure (10 bar for monocrystals), the critical parameters worsen [12, 13].

Concerning our experimental results, we suppose that the sample annealed at p = 200 Torr is not saturated with enough oxygen, and it is characterized by greater spatial inhomogeneity and includes nanograins with lower critical parameters (where the SC carrier concentration is lower due to the oxygen deficiency) together with nanograins with optimal ones. This demonstrates a greater ΔT_c and a lower J_c, regardless of its relatively high critical temperature. A high value of T_c may be due to the current paths consisting of the "best" grains and the "best" contacts between them. At a higher annealing pressure (~500 Torr), the oxidization conditions are preferable and the samples demonstrate better critical parameters – high values of T_c and J_c and a narrow SC transition in the temperature scale.

The morphology of the YBCO films is not seen to depend essentially on the annealing pressure in this interval of pressures (200 – 500 Torr). Fig. 5 shows the surface morphology of YBCO films annealed at a 500 Torr oxygen pressure.

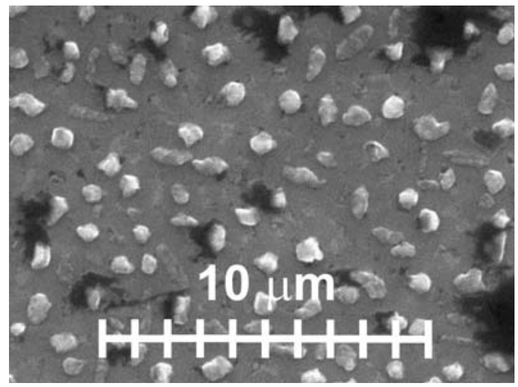

Fig. 5. Surface morphology of a YBCO film annealed at 500 mbar oxygen pressure.

Scanning electron microscope (SEM) revealed a set of outgrowths distributed almost uniformly over the surface area for both YBCO films. We suppose that these outgrowths could be CuO and Y_2O_3 particles [14]. Their lateral dimensions are 0.5-1 μm. According to [14], these outgrowths do not prevent the production of YBCO layers with good electrical properties.

4. Conclusions

Thermal annealing was performed on superconducting YBCO films at 530 ºC for 40 min, at oxygen pressures of 200 and 500 Torr.

The increase of J_c from 1.01 to 1.38 MA/cm^2 at 77 K and T_c from 89 to 89.2 K for the annealed YBCO at 500 Torr oxygen pressure can be attributed to an increase in the superconducting carrier concentration in the grains and an improvement in the weak link qualities due to the reduction of oxygen deficiency in the sample. The more abrupt SC temperature transition at the higher oxygen annealing pressure indicates a better spatial homogeneity of the sample. The XRD spectra show a reduction of the c-axis parameter from 11.72 to 11.70 Å, which corresponds to an increased oxygen content from 6.67 to 6.84.

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References

[1] L. Rostila, L. Söderlund, R.Mikkonen, J. Lehtonen, Physica C **467**, 91 (2007).
[2] J. D. Jorgensen, B. W. Veal, W. K. Kwok, G. W. Crabtree, A. Umezawa, L. J. Nowicki and A. P. Paulikas, Phys. Rev. B **36**, 5731 (1987).
[3] J. L. Tallon, C. Bernhard and C. Niedermager, Supercond. Sci. Technol. **10**, A38 (1997).
[4] H. Huhtinen, E. Lähderanta, P. Paturi, Journal of Low Temperature Physics **117,** 795 (1999).
[5] C. Cancellieri, A. Augieri, V. Boffa, G. Celentano, L. Ciontea, F. Fabbri, V. Galluzzi, U. Gambardella, G. Grassano, T. Petrisor, R. Tebano, IEEE Transactions on Applied Superconductivity **15**, 3038 (2005).
[6] X. G. Qiu, H. Koinuma, M. Iwasaki, M. Kawasaki, Y. Segawa, Phys. Rev. B **60**, 707 (1999).
[7] M. Beshkova, B. Blagoev, D. Kovacheva, G. Mladenov and T. Nurgaliev, Journal of Physics, Conference series **113**, 012021 (2008).
[8] T. Nurgaliev, Proc. European Conf. Appl. Supercond., The Netherlands, Inst. Phys. Conf. Series IOP Publishing Ltd **158**, 801 (1997).
[9] J. Grybos, D. Hohlwein, F. Kubanek, Physica C, **244**, 349 (1995).
[10] A. Sequeira, H. Rajagopal, J. V. Yakhmi, Solid State Commun. **65,** 991 (1988).
[11] J. Halbritter, Journal of superconductivity, **8**, 691 (1995).
[12] High temperature superconductivity, Fundamental and applied studies 1, A. A. Kiseleva, Leningrad (1990).
[13] Z. Aslanoglu, Y. Akin, M. El-Kawni, W. Sigmund, Y. Hascicek, Physica C **384,** 501 (2003).
[14] R. Krupke, Z. Barkay, G. Deutscher, Physica C **315**, 99 (1999).

*Corresponding author: mbeshkova@yahoo.com