Post-Deposition Cooling in Oxygen is Critical for $YBa_2Cu_3O_{7-d}$ Films Deposited by Eclipse Pulsed Laser Deposition Method

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$YBa_2Cu_3O_{7-d}$ thin films were deposited on MgO single crystals by means of an eclipse pulsed laser deposition method. Deposited films are cooled down *in situ* under an oxygen atmosphere at a given oxygen pressure. The relationship between critical temperature and oxygen deficiency was investigated by means of electrical resistance R(T) and x-ray diffraction measurements. Post-deposition cooling is critical and the high pressure of oxygen during cooling is favorable.

I Introduction

Much effort has been devoted in the research for the applications of $YBa_2Cu_3O_{7-d}$ (YBCO) thin films to superconducting devices such as Josephson junctions [1,2], flux flow transistors [3,4], and superconducting quantum interference devices [5]. It is surely a key issue to obtain YBCO thin films with a good quality. Deposition of YBCO thin films have been obtained for various methods such as thermal coevaporation [6], activated reactive evaporation [7], chemical vapor deposition [8], ion beam deposition [9], sputtering, and pulsed laser deposition (PLD). Among a variety of deposition methods, the last two methods, sputtering and PLD, are more often used nowadays.

The annoying problem of sputtering for the deposition of YBCO thin films is mainly resputtering; oxygen negative ions are accelerated and bombard the deposited film. The resputtering causes deterioration of the film morphology. On the other hand, PLD is basically free from resputtering because electric field does not exist in the deposition chamber. But PLD have a peculiar problem for i.e. droplets deteriorate the film morphology. A droplet is defined as a comparatively large cluster of atoms or ions. In order to avoid the problem, a new PLD method - eclipse PLD - has been recently used and reported [10].

In this article YBCO thin films were deposited on MgO substrates by eclipse PLD followed by cooling of the samples *in situ* in an oxygen atmosphere at a given pressure. The relationship between critical temperature (Tc) and the oxygen deficiency (d) was investigated. The effect of oxygen pressure during post-deposition cooling is discussed.

II Experimental details

The target used in the experiment was a commercially available stoichiometric single target, 25 mm in diameter with a thickness of 2 mm. The target was rotated at 100 r.p.m. in order to avoid the rapid deterioration that would be caused by always ablating the same place. The laser for the ablation was a KrF excimer laser, which radiates at 248 nm. The output pulse energy and repetition rate was 100 mJ and 8 Hz, respectively.

A (100) MgO single crystal was used as a substrate. The substrate was prepared in the form of a square of 100 mm^2 with a thickness of 2 mm. MgO crystals have been widely used as a substrate for YBCO thin films regardless of their large misfit of 7.7% with respect to the c lattice parameter of YBCO, because their dielectric properties are excellent: low dielectric constant and loss tangent.

The substrate was cleaned in acetone and in ethanol one after another repeatedly using an ultrasonic cleaner. A cleaned substrate was dried in air and put in the deposition chamber in such a way that the substrate was parallel to the target: on-axis configuration. The substrate was fixed onto a sample holder on which an SiC heater was mounted. The substrate was heated up to 740 °C during the deposition, which was monitored with a thermocouple attached to the sample holder.

Prior to deposition, the deposition chamber was evacuated to 3×10^{-6} Torr with a turbo pump. The deposition was performed under oxygen atmosphere for an hour. The thickness of the films was 90 nm. The samples were then cooled down in the chamber that was f1lled with oxygen gas after the deposition. A slow cooling rate was maintained in such a way that it took 30 minutes to cool from 740 down to 400 °C. During the cooling period, oxygen atoms were included in YBCO films. From 400 °C to room temperature, cooling was carried out without artificial control of the cooling rate. The deposition conditions render a post anneal in oxygen atmosphere unnecessary.

It is of interest how oxygen pressure during cooling affects the oxygen deficiency and Tc of the obtained YBCO f1lms. Samples A, B, and C were prepared at different oxygen pressures of 0.7, 0.8, and 9 Torr, respectively. The oxygen deficiency was estimated based on the known relationship between oxygen deficiency and c lattice constant [11], as described in the next section. The c lattice constant was determined by x-ray diffraction using a conventional $\theta - 2\theta$ Rigaku diffractometer with Cu kα radiation.

The resistance R(T) of the deposited film was obtained by an ordinary four-probe method. The temperature dependence of the resistance R(T) was obtained using a liquid helium cryostat and a computer-controlled heater attached to the sample holder. The critical temperature was defined as that where the gradient of the resistance R(T) with respect to temperature is maximum.

III Results and discussion

A typical x-ray diffraction pattern is shown in Fig. 1. Assignments of the observed structures are labeled in the figure; ten (001) YBCO peaks as well as a strong peak ($2\theta = 42.8$ degree) corresponding to an MgO substrate. The result shows that the YBCO thin film is preferably oriented so that c axis is normal to the surface. The c lattice parameters were determined to be 1.1738, 1.1713, and 1.1684 nm for the samples A, B, and C, respectively. The film thickness of each sample is larger than 100 nm, and therefore an influence of the strain at the interface is negligible [12].

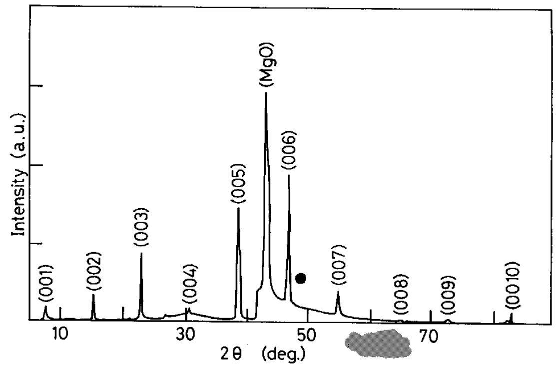

Figura 1. A typical x-ray diffraction pattern of an YBCO film on MgO. The c axis of YBCO is oriented normal to the surface.

The relationship between the c lattice constant (c) and the oxygen deficiency (d) is drawn in Fig. 2 on the basis of JCPDS data. The figure shows that there is a linear relationship between them. With the help of the relation, the oxygen deficiencies of samples A, B, and C were determined to be 0.44, 0.30, and 0.14, respectively.

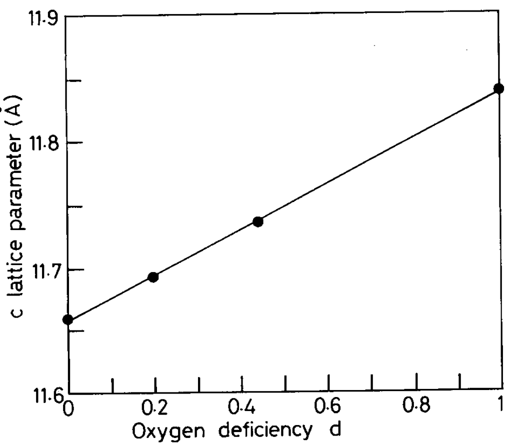

Figura 2. A relationship between c lattice parameter and oxygen deficiency on the basis of JCPDS card data (closed circles). A linear relation of d=5.556c-64.78 exists between them.

The temperature dependence of the resistance R(T) is shown in Fig. 3, 4, and 5 for the sample A, B, and C, respectively. A deviation from a straight line above 200K in Fig. 4 is due to trivial experimental errors. The superconducting transition occurs at the highest temperatures when the oxygen deficiency is the smallest. The relationship between Tc and the oxygen deficiency is shown in Fig. 6, together with the data reported by W. Prusseit *et al.* [13]. They prepared YBCO films by thermal co-evaporation. Our obtained result is roughly similar to that reported by Prusseit *et al.* Based on the result, the cooling in oxygen atmosphere is an effective method to suppress oxygen deficiencies in case of an eclipse PLD.

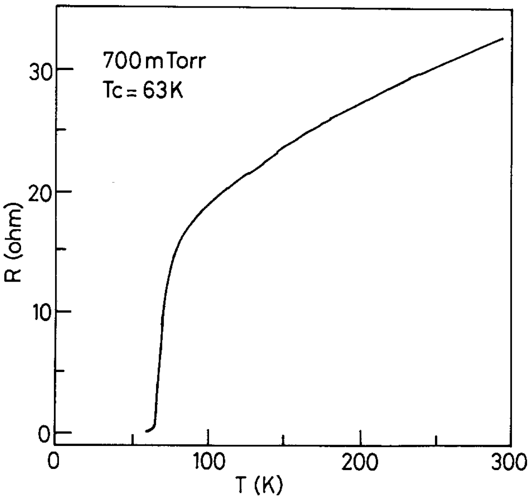

Figura 3. Temperature dependence of the resistance (sample A). The oxygen pressure during the cooling stage was 0.7 Torr. The critical temperature is 63 K.

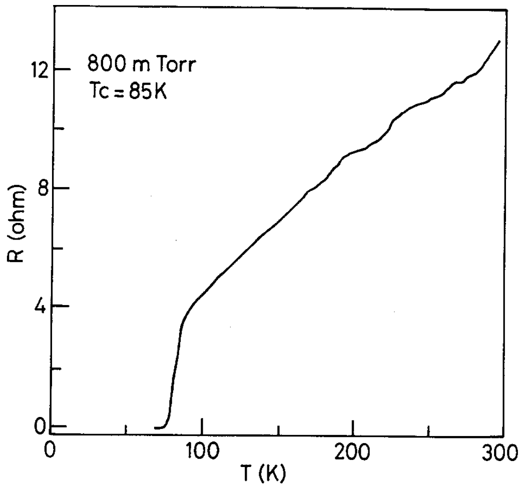

Figura 4. Temperature dependence of the resistance (sample B). The oxygen pressure during the cooling stage was 0.8 Torr. The critical temperature is 85 K.

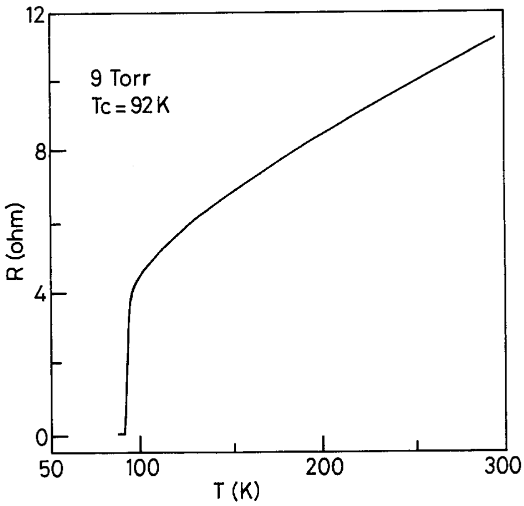

Figura 5. Temperature dependence of the resistance (sample C). The oxygen pressure during the cooling stage was 9 Torr. The critical temperature is 92 K.

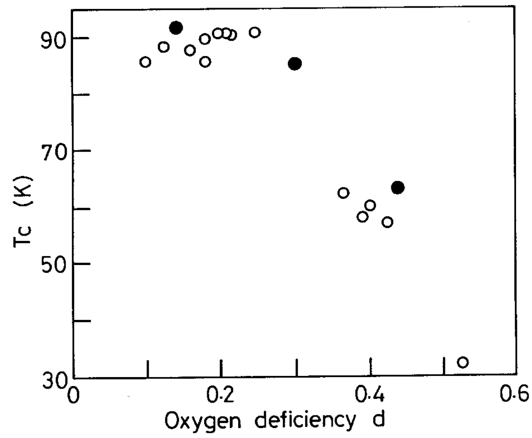

Figura 6. The relation between Tc and the oxygen deficiency. The closed circles denote our data, and open circles the data reported by W. Prusseit *et al.* [13].

Strictly speaking, our results show the higher transition temperature than their results. The fact implies that the transition temperature is not only determined by the oxygen deficiency but by other factors such as the structure of the film. We consider that disorder can not be excluded during cooling at low oxygen pressure otherwise a shadow mask in the chamber excludes droplet

Kinoshita *et al.* investigated the dependence of oxygen pressure during the deposition and obtained the largest Tc at 0.3 mTorr. But Tc becomes rather worse when the oxygen pressure is enharlced above 0.3 mTorr. On the other hand, Tc increases monotonically with increasing the oxygen pressure during cooling according to our results. The slow cooling in oxygen atmosphere is critical in order to obtain high Tc YBCO films for an eclipse PLD method.

The resistance just above Tc is 15 ohm in sample A though it is 4 ohm in samples B and C. Having a large resistance R(T), sample A can include more disorder. The close investigation of morphology or disorder of the films is the next interesting points.

IV Conclusion

YBCO thin films have been deposited on MgO substrates by means of an eclipse PLD method. The relationship between Tc and the oxygen deficiency is generally similar to that reported for the films deposited by thermal co-evaporation. The cooling in oxygen atmosphere followed by deposition is critical for an eclipse PLD method. The high oxygen pressure during cooling is favorable to high Tc. The oxygen pressure can be affected the film quality as well as the oxygen content in the film.

References

[1] J. E. Zimmerman, J. A. Beall, M. W. Cromar, and R. H. Ono, Appl. Phys. Lett. **51**, 617 (1987).

[2] M. S. Colclough, C. E. Gough, M. Keene, C. M. Muirhead, N. Thomas, J. S. Abell, and S. Sutton, Nature **328**, 47 (1987).

[3] K. Miyahara, S. Kubo, and M. Suzuki, Appl. Phys. Lett. **76**, 4772 (1994).

[4] K. Miyahara, S. Kubo, and M. Suzuki, J. Appl. Phys. **75**, 404 (1994).

[5] R. H. Koch, C. P. Umbach, G. J. Clark, P. Chaudhari, and R. B. Laibouritz, Appl. Phys. Lett. **51**, 200 (1987).

[6] P. Berberich, B. Utz, W. Prusseit, and H. Kinder, Physica C **219**, 497 (1994).

[7] T. Terashima, K. Iijima, K. Yamamoto, Y. Bando, and H. Masaki, Jpn. J. Appl. Phys. **27**, L91 (1988).

[8] K. Watanabe, H. Yamane, H. Kurosawa, T. Hirai, N. Kobayashi, H. Iwasaki, K. Noto, and Y. Muto, Appl. Phys, Lett. **54**, 575 (1989).

[9] J. Fujita, T. Yoshitake, A. Kamijo, T. Satoh, and H. Igarashi, J. Appl. Phys. **64**, 1292 (1988).

[10] K. Kinoshita, H. Ishibashi, and T. Kobayashi, Jpn. J. Appl. Phys. **33**, L417 (1994).

[11] JCPDS cards.

[12] M. Ohmukai, Appl. Surf. Sci. **152**, 49 (1999).

[13] W. Prusseit, A. Murk, K. Schlenga, G. Hechtfischer, and P. Müller, Proc. of 5th ISEC Nagoya, Japan, 11 (1995).