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## ADVERTISEMENT



# High quality $\text{YBa}_2\text{Cu}_3\text{O}_x$ films prepared in air using pulsed laser deposition

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High quality  $\text{YBa}_2\text{Cu}_3\text{O}_x$  superconducting films have been deposited by laser deposition on yttrium stabilized cubic zirconia (YSZ) substrates using a partial pressure of air instead of oxygen. The  $T_c$  of these films was over  $2^\circ$  higher than films made in the same deposition system using oxygen. X-ray diffraction indicated that the films were oriented with the  $c$  axis normal to the substrate surface. The critical current densities of the films were on the order of  $10^6$  A/cm<sup>2</sup> at 77 K and self-magnetic field, and the room-temperature resistivities were about  $245 \mu\Omega$  cm. A unique aspect of these films was that the normal state resistivities showed nonlinear behavior with respect to temperatures. No secondary phase was detected by x-ray diffraction and SQUID magnetometry. © 1994 American Institute of Physics.

Since the discovery of high-temperature superconducting materials,<sup>1</sup> the preparation of epitaxial high-temperature superconducting films on single crystal substrates has been established by a variety of different deposition techniques.<sup>2-5</sup> The *in situ* pulsed laser deposition (PLD) technique is one of the simplest and most versatile methods. The relatively low number of control parameters, as well as the stoichiometric removal of constituent species from the target during deposition, makes the PLD technique particularly attractive for growth of complex multicomponent systems. High quality  $\text{YBa}_2\text{Cu}_3\text{O}_x$  films<sup>6-8</sup> (in terms of  $T_c$ ,  $J_c$ , surface resistance, and room temperature resistivity) have been routinely obtained by many groups using the *in situ* PLD technique. Many studies have been devoted to the optimization of the PLD process, particularly for  $\text{YBa}_2\text{Cu}_3\text{O}_x$  films.<sup>9-11</sup> The important deposition parameters include the substrate temperature  $T_s$ , the rate of deposition, the target to substrate distance  $D_{t-s}$ , as well as the oxygen pressure  $P_{\text{O}_2}$ .

Otis *et al.*<sup>11</sup> used  $\text{NO}_2$  and  $\text{N}_2\text{O}$  to make YBCO in their laser deposition system and found that these gases increase the formation of CuO in the plume. This indicates that the presence of nitrogen is not detrimental to the formation of  $\text{YBa}_2\text{Cu}_3\text{O}_x$ . Phillips *et al.*<sup>12</sup> have sputter deposited  $\text{Y}_2\text{O}_3$ , CuO, and  $\text{BaF}_2$ , then annealed the film in wet oxygen and obtained good results. This indicates that the presence of water vapor (at high temperatures) also is not detrimental. Furthermore, bulk YBCO can be made by solid state reaction in air. These results motivated us to investigate the effects of using a partial pressure of air (composed of  $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ , etc.) instead of oxygen during deposition. We report here our results and present what we found to be the optimal deposition parameters for reproducibly obtaining high quality  $\text{YBa}_2\text{Cu}_3\text{O}_x$  films.

An excimer KrF laser (with wavelength 248 nm) was focused onto a stoichiometric  $\text{YBa}_2\text{Cu}_3\text{O}_x$  target to produce a fluence of about  $1.5 \text{ J/cm}^2$ . The size of the laser spot was about 1 mm. The laser pulse frequency was set at 7 Hz.

(100)-oriented yttrium stabilized cubic zirconia (YSZ) substrates were used in this study. Substrates were attached with silver paint to the center of a 2 in. diam heater which was placed directly opposite to the target. The deposition temperature  $T_s$  was monitored using a thermocouple embedded in a hole in the heater block. A calibration measurement was taken to determine the substrate temperature, and it was found that at  $700^\circ\text{C}$  the actual substrate temperature was about  $35^\circ\text{C}$  lower than  $T_s$ . The chamber pressure was controlled by a throttle valve.

In the first set of experiments, the deposition temperature,  $T_s$  (measured at the heater block) was held at  $740^\circ\text{C}$ , and the chamber air pressure  $P_{\text{air}}$  was varied from 150 to 800 mTorr. Since the size of the laser plume was related to the deposition pressure (at the higher pressure, the plume size is smaller), we varied the distance between the substrate and target  $D_{t-s}$ , so that the plume envelope was on the substrate. In the second set of experiments we held the chamber air pressure at 590 mTorr, and varied the deposition temperatures in the range of  $600\text{--}760^\circ\text{C}$ . After each deposition, the chamber was filled with air to 1 atm and held for 8 min before the heater was turned off and the film was cooled.

The transition temperatures were measured using a four point probe. Critical current densities were measured using microbridges with width about  $15 \mu\text{m}$  and length between two voltage probes of about 3.5 mm made by using wet etching. We lowered the photoresist baking temperature from  $100^\circ\text{C}$ , as recommended for the photolithography, to  $60^\circ\text{C}$  to prevent possible degrading of superconductivity. The criterion we used to determine the critical current density was  $3 \mu\text{V/cm}$ . The film thickness was typically 300–600 nm.

It was found that the superconducting properties of the films made in air were very sensitive to the deposition pressure while less sensitive to the substrate temperature. Figure 1 shows the transition temperature  $T_c$  and the normal resistance ratio  $R_{300}/R_{100}$  versus the deposition pressure of air for films made at  $740^\circ\text{C}$ . Both curves peak around 580–590 mTorr; which equivalent to oxygen partial pressure of about 120 mTorr. This equivalent partial pressure of oxygen was lower than what we found to be optimal to deposit

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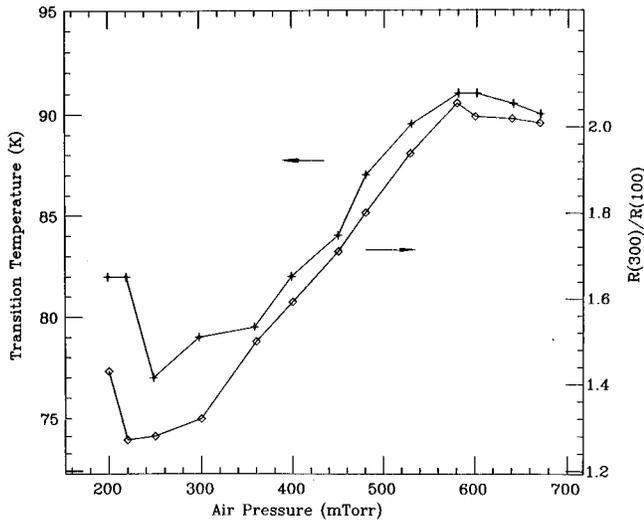


FIG. 1. The transition temperatures  $T_c$  and the normal resistance ratios  $R_{300}/R_{100}$  of a film deposited at  $740^\circ\text{C}$ , as a function of the deposition air pressure  $P_{\text{air}}$ .

$\text{YBa}_2\text{Cu}_3\text{O}_x$  films when pure oxygen was used (200–300 mTorr), but it was still within the Faupel–Hehenkamp thermodynamic phase stability line,<sup>13</sup> if we take the substrate temperature correction into account. X-ray diffraction indicated that films deposited in the above conditions were  $c$ -axis oriented, no  $a$ -axis orientation or secondary phase was detected, see Fig. 2. The films showed semiconductor behavior, however, when air pressure was higher than 800 mTorr, while had lower and broad transition temperatures when air pressure was lower than 450 mTorr.

Films deposited in the temperature range of  $730$ – $760^\circ\text{C}$  and at air pressure of 590 mTorr were highly  $c$ -axis oriented and no significant differences in transition temperatures were noticed. Films grown at temperatures lower than  $650^\circ$  generally showed  $a$ -axis orientation feature. Figure 3 shows two films deposited at optimal substrate temperature ( $730^\circ\text{C}$ ) but at low air pressure (300 mTorr), and at optimal air pressure (590 mTorr) but at low substrate temperature ( $600^\circ\text{C}$ ). Both

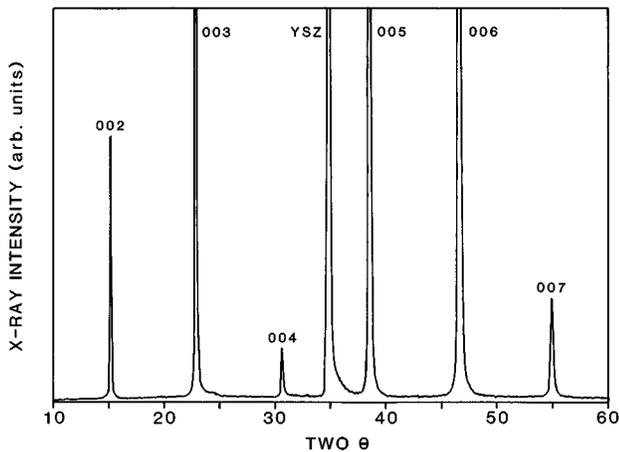


FIG. 2. X-ray diffraction pattern from a film made in partial pressure of air.

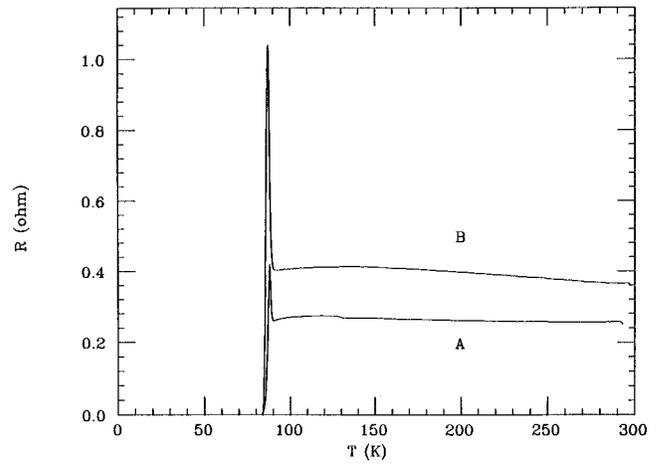


FIG. 3. Resistance–temperature curves of  $a$ -axis oriented films. The deposition parameters were  $T_s=730^\circ\text{C}$ ,  $P_{\text{air}}=300$  mTorr (curve A); and  $T_s=600^\circ\text{C}$ ,  $P_{\text{air}}=590$  mTorr, (curve B), respectively.

of them showed a large resistance peak before the transition which is generally seen for  $a$ -axis oriented films.

We also investigated different substrate to target distances  $D_{t-s}$ , for the deposition. All films deposited where the substrates were placed in the middle of the laser-induced plume showed no or poor superconductivity, while the ones deposited where the substrates were placed at the envelope of the laser-induced plume (typical  $D_{t-s}$  was about 4–5 cm at 580–590 mTorr) and optimal conditions had  $c$ -axis oriented structure and high  $T_c$  and  $J_c$ .

We have remeasured the films 7 months after the first measurements, no  $T_c$  and  $J_c$  drop were observed, however, the room temperature ( $300^\circ\text{C}$ ) resistances were found to be 5%–10% lower than the fresh films. All the films were stored in air without desiccator.

Films deposited at optimal conditions generally had room temperature resistances about  $245\ \mu\Omega\ \text{cm}$ . Transition temperatures  $T_c$  of these films were over  $2^\circ$  higher than films grown in the same deposition system using oxygen and optimal conditions (200–300 mTorr, 4 cm and  $740^\circ$ ). The films deposited in air under optimal conditions (580–590 mTorr, 4–5 cm and  $730$ – $760^\circ\text{C}$ ) had an on-set temperature higher than 93 K, and an off-set temperature of around 90 K with typical transition temperature width of less than  $1^\circ$ . Films made in oxygen had a typical on-set temperature around 90 K and an off-set temperature around 87 K. The critical current densities of the films made in air were larger than  $10^6\ \text{A}/\text{cm}^2$  at 77 K and self-magnetic field, which was the same as that of the films grown in oxygen. Figure 4 shows the temperature dependence of resistances and susceptibility for a  $\text{YBa}_2\text{Cu}_3\text{O}_x$  film made in air ambient. The following parameters were used to deposit this film:  $T_s$  was  $745^\circ\text{C}$ ,  $P_{\text{air}}$  was 590 mTorr, and  $D_{t-s}$  was 4 cm. As a comparison, the transition curve of a typical good  $\text{YBa}_2\text{Cu}_3\text{O}_x$  film made in pure oxygen in the same laser deposition system is shown with dashed line. The one grown in air has higher transition temperature (about  $3^\circ$ ). A bridge was made from this film with dimensions as  $3.5\ \text{mm} \times 15\ \mu\text{m} \times 310\ \text{nm}$ . The voltage–current characteristics of the bridge measured at 77 K and

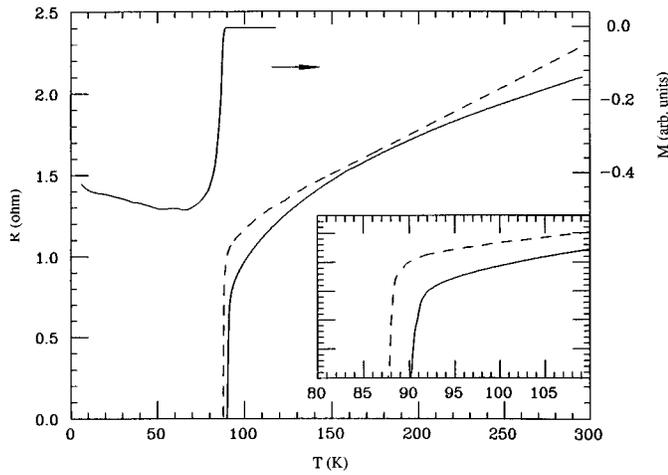


FIG. 4. The resistance and susceptibility of one  $c$ -axis oriented film (in Fig. 2) as a function of temperature. As a comparison, the  $R$ - $T$  curve of a good YBCO film deposited in oxygen is shown in dashed line. The inset figure gives a close comparison of the transition temperatures of the two films.

self-magnetic field are shown in Fig. 5. From this curve, the critical current density  $J_c$ , has been determined to be  $1.4 \times 10^6$  A/cm<sup>2</sup>. We have fitted the experimental data to the thermal activation process and quantum nucleation mechanism of vortex rings,<sup>14</sup> both models are equally good for fitting our results at 77 K. Quantum nucleation of vortex rings only takes place at low temperature ( $\ll T_c$ ), unless the film is of high quality with high critical current density.<sup>14</sup> The solid line in Fig. 5 is a fitting curve with the quantum nucleation mechanism of vortex rings.

The high  $T_c$  superconductors have a unique normal state linear temperature dependent of the in-plane resistivity in a broad range from well beyond room temperature down to few degrees before transition temperature. This inplane resistivity can be described as

$$\rho_{ab} = \alpha T + \rho_0, \quad (1)$$

where  $\alpha = d\rho_{ab}/dT$  is the slope of  $\rho_{ab}$ - $T$  curve. Typical  $\alpha$

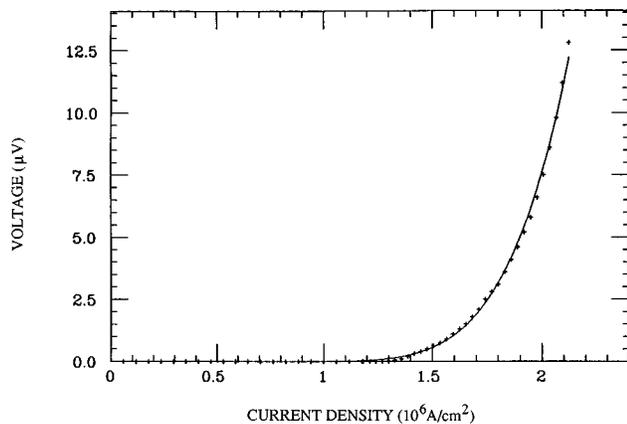


FIG. 5. Voltage-current characteristics of the film shown in Fig. 2. The solid line is a fitting curve with the quantum nucleation mechanism of vortex rings.

value is about  $0.6$ – $0.7 \mu\Omega$  cm/K for  $\text{YBa}_2\text{Cu}_3\text{O}_x$  films. In our measurements of more than 50  $\text{YBa}_2\text{Cu}_3\text{O}_x$  films deposited in air ambient, however, a unique aspect was that none of the films exhibited a linear temperature dependent normal state resistivity or, in other words, the  $\alpha$  in Eq. (1) was not a constant. It increased with decreasing temperature. One example is shown in Fig. 4 with solid line, where the  $\alpha$  is 0.47 at 240 K, 0.53 at 180 K, and 0.65 at 120 K. We do not attribute this behavior to the contribution of off-plane resistivities or secondary semiconductor phase, since the off-plane resistivities and semiconductor phase always have negative  $\alpha$  values at the temperatures before transition, and because x-ray diffraction indicated that our samples were  $c$ -axis normal to the film plane and no secondary phase exist. Although we did not detect any  $a$ -axis material or secondary phase by x-ray diffraction (Fig. 2) or by SQUID magnetometry, we do not totally rule out the possibility of a small amount secondary phase presented in our samples. We believe further investigation of the nonlinear resistivity behavior is worthwhile from both the theoretical and applied points of view.

In conclusion, high quality  $\text{YBa}_2\text{Cu}_3\text{O}_x$  superconducting films have been deposited in a partial pressure of air using PLD. The optimal deposition parameters are  $T_s$  about 730–760 °C,  $P_{\text{air}}$  about 580–590 mTorr, and  $D_{t-s}$  about 4–5 cm. The transition temperatures of these films are  $2^\circ$ – $3^\circ$  higher than films deposited in the same system but using pure oxygen. Both have similar critical current densities at 77 K. An unusual and unique aspect of nonconstant  $\alpha$  values is reported and discussed. Further investigation is needed to fully understand the transport properties in the normal state regime of these films.

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