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LONG LASER-PULSE METHOD OF PRODUCING Y-Ba-Cu-O SUPERCONDUCTING FILMS

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The quality of superconducting thin films evaporated from $YBa_2Cu_3O_7$ targets has been improved using a long-pulse (ms) Nd-glass laser with 50 J/pulse energy. The film produced on $SrTiO_3$, held at 690 C, and post-annealed at 880 C for about three hours began the transition to superconductivity at 85 K and had zero resistance at 79 K. The observed variation in film thickness corresponded to diffuse emission from the target, and no significant variation in composition as a function of angle of emission up to 40° with respect to surface normal was seen.

Most studies on laser deposition of high T_c superconducting films have employed repetitive irradiation of a superconducting target with short (ns duration) pulses, principally from excimer lasers[1]. The peak power densities are in the range 10^7-10^8 W/cm^2 and the deposition rate is kept at a few angstroms per pulse. The angular distribution of the material emitted by this technique is reported to be non-diffuse [2], with a functional form as peaked as $\cos^8\theta$. Moreover, the variation in composition with angle is appreciable.

Recently, we have demonstrated that low-power-density pulses from a Nd-glass laser operated in the conventional mode (non-Q-switched) can produce superconducting films of Y-Ba-Cu-O by vacuum vaporization[3]. The present work was undertaken to determine whether the film quality can be improved by varying deposition parameters such as substrate temperature and post-annealing conditions. The angular distribution of the evaporated species and the variation of the composition with angle were also measured.

The apparatus shown in Fig. 1 has been described in detail previously. In brief, a 50 J Nd-glass laser with a wavelength of 1.06 μm produces approximately triangular pulses with baseline widths of 0.5 to 1.75 ms. The beam is focused to a spot about 2.5 mm in diameter on the target, which is a pellet of sintered $YBa_2Cu_3O_7$ of approximately 75% of theoretical density. The substrate is located in a vacuum chamber about 5 cm from the target and is heated by radiation from a hot tungsten filament. A small doser provides oxygen to the substrate at an equivalent pressure about 1000 times larger than the background pressure of the vacuum chamber (which is about 10^{-7} Torr). A 300 W rf discharge is generated at the end of the doser containing oxygen at a pressure of 200 mTorr. This arrangement is intended to produce excited oxygen molecules and atoms, which are more reactive than ground-state species and better able to replenish oxygen lost in the vaporization process.

The films were deposited on polished and cleaned $SrTiO_3$ substrates. Fig. 2 shows the resistance vs. temperature for three films with different substrate temperatures and post annealing conditions. For a substrate temperature of 540 C and a post-annealing time of about 15 minutes at 850 C, the onset of superconductivity was about 65 K and zero resistance was achieved at about 42 K. With additional 2 hours annealing, these temperatures were raised to 78 and 61 K, respectively. Prolonging the annealing time beyond

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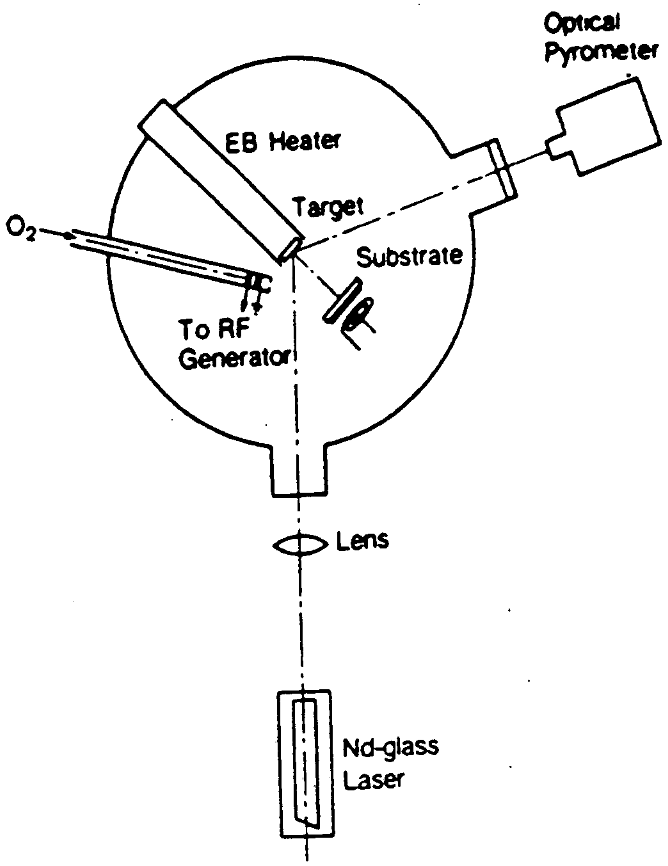

Fig. 1. Schematic diagram of the deposition system. The RF generator is employed to dissociate O_2 in the doser.

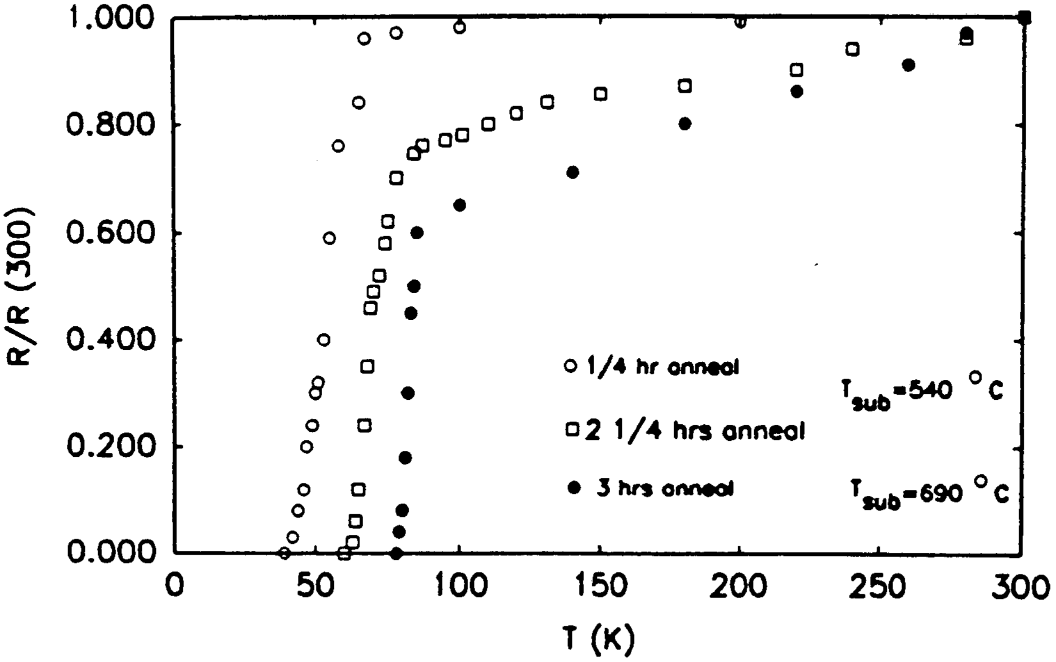

Fig. 2. Resistance vs temperature of films deposited on $SrTiO_3$ substrates for different substrate temperatures and annealing times.

three hours had no appreciable effect on the transitions. However, raising the substrate temperature during deposition to 690 C and annealing for three hours at 880 C raised these temperatures to 85 K and 79 K, respectively. The improvement of the quality of the film as a result of increasing the substrate temperature suggests that inter-diffusion of the metal species in the film during deposition plays an important role in obtaining desirable structure and/or stoichiometry.

The dependences of thickness and composition on the angle of emission were obtained by mounting small pieces of silicon in an arc with radius of curvature of 5 cm and with its center on the laser beam spot on the target. The Si surfaces were partially masked by a photoresist. After the removal of the photoresist, the thickness of the films were measured by a profilometer. Fig. 3 shows the thickness variation as a function of angle for three different laser pulse widths, 0.5, 1.0 and 1.75 ms. The variation in thickness clearly suggests a cosine angular distribution for all three laser pulse widths. The films were also examined by RBS for the 1 ms pulse duration experiment. Fig. 4 shows no significant change in composition for angles up to about 40° with respect to the target-normal

It is generally believed that the high film quality obtained by the short (ns) pulse technique is the result of the low deposition rate (10 angstroms per pulse at most). The high mobility of the deposited materials is presumably responsible for establishing the desired structure and stoichiometry. Long-pulse (ms) vaporization provides approximately 1000 angstroms per pulse at 50 J/pulse. However, due to the fact that the pulse is 100,000 times longer, the flux of materials arriving at the substrate is two orders of magnitude lower than in the ns pulse technique.

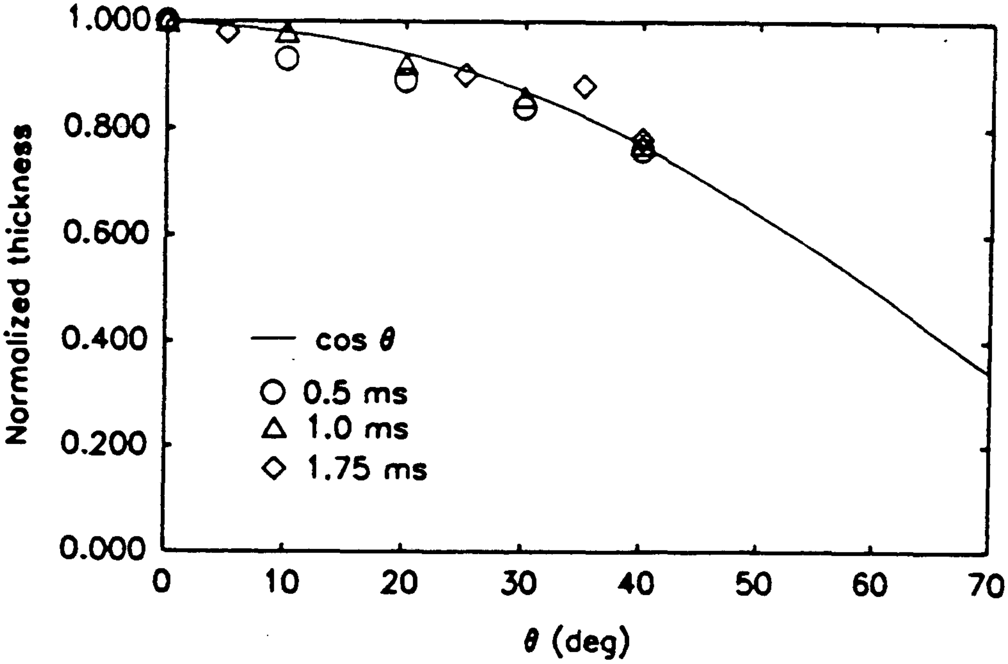

Fig. 3 Angular dependence of the deposited film thickness with respect to the target normal for different laser pulse widths.

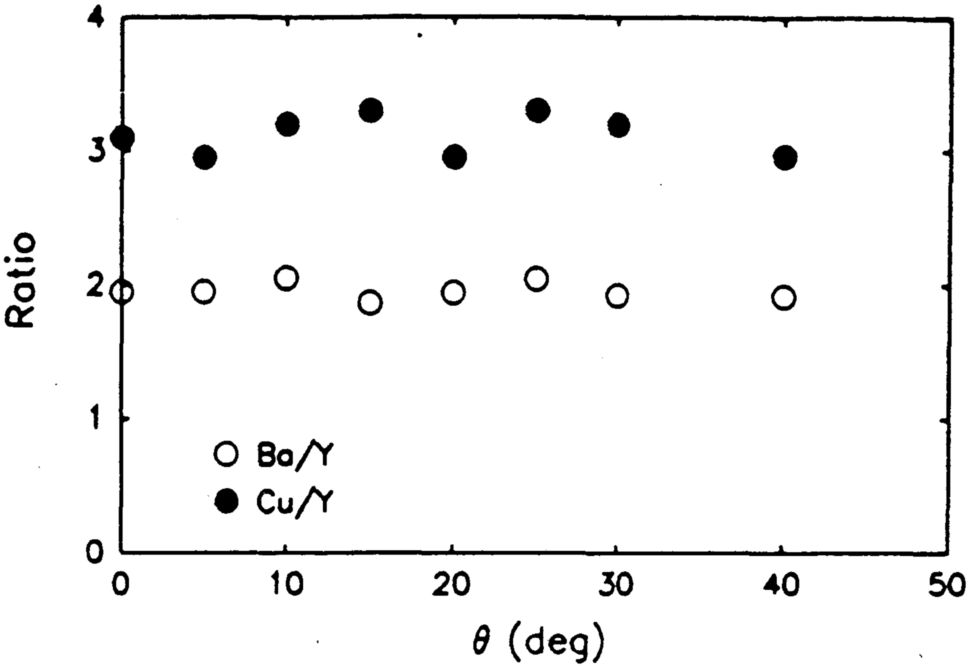

Fig. 4 Composition variation of the deposited film with angle for vaporization with pulses 1 ms long.

In summary, the deposition of high T_c superconducting films by long-pulse laser vaporization produced very promising results. The transition temperature of the films were improved over earlier work by increasing the substrate temperature from 540 to 690 C. The cosine angular distribution and the insignificant variation in composition with emission angle with respect to target-normal produces high quality films over areas much larger than achievable in the ns laser pulse technique.

The long-pulse laser vaporization approach may prove to be useful in device applications and for processes such as coating tapes and wires on a quasi-continuous time scale.

Acknowledgements

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