Deposition of YBCO Thin Films over Large Areas by a 90° Off-Axis Sputtering Technique

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***Abstract*—We report the deposition of YBCO thin films with uniform thickness (<±5% variation), and stoichiometric composition (<2.3% deviation from the target stoichiometry) over an 8" diameter area, by a single target 90° off-axis sputtering technique using a 3" diameter target and optimized substrate rotation. A 3" diameter resistive heater was used with optimized rotation in two consecutive deposition runs to study the variation in film properties of small test samples deposited over an 8" diameter area. Due to the inherent temperature non-uniformity of the heater, all the samples could not be deposited under optimum growth conditions. However, all the films displayed a consistently high transition temperature (T_C > 87.5°K) and critical current density ($J_{C\ 77K} > 1 \times 10^6$ A/cm^2, $J_{C\ 4.2K} > 1 \times 10^7$ A/cm^2) over an 8" diameter area. Considering only the better optimized samples the films displayed high critical current densities ($J_{C\ 77K} > 3 \times 10^6$ A/cm^2, $J_{C\ 4.2K} > 3 \times 10^7$ A/cm^2). The variations in the film properties have been correlated to the variations in crystalline quality, amount of c-axis grains and substrate temperature during deposition.**

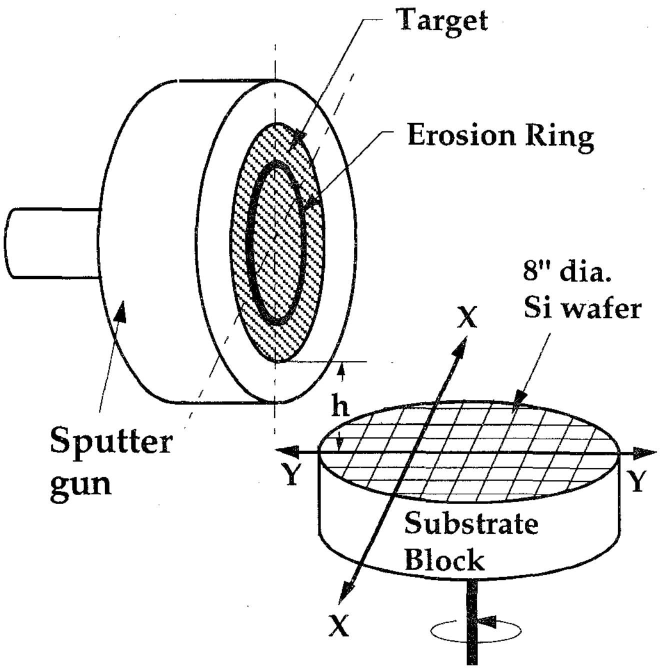

Fig. 1. Schematic of the 90° off-axis sputtering geometry showing the various reference axes on the substrate

I. INTRODUCTION

High T_c superconducting $YBa_2Cu_3O_7$ (YBCO) thin films are already finding applications in planar microwave devices [1], SNS Josephson junctions [2], and SQUIDs [3]. In most of these applications, large area uniform deposition of high quality films is critical. For instance, multichip modules and complete integrated microwave devices such as receivers and signal processing systems require high quality YBCO thin films to be grown on large area (>2 inch diameter) substrates. Furthermore, large area deposition is important for efficient and industrial scale manufacture.

High quality YBCO thin films have been grown *in-situ* over areas ranging from 2 inches in diameter to 9 inches in diameter by various techniques including co-evaporation [4], laser ablation [5], sputtering [6] and metal-organic chemical vapor deposition (MOCVD) [7], [8]. Among the various techniques, 90° off-axis magnetron sputtering is especially well suited for large area deposition because it is a very reproducible and easily controllable technique [9], [10]. This technique has been used for the deposition of high quality heterostructure and multilayer films over 3 in. diameter areas [11]. However, the growth rate is relatively slow and acceptable uniformity has been demonstrated only over 3 inch diameter areas, which is still small for many device applications. We have used this technique in conjunction with larger diameter sputtering target (3 in. diameter) and optimized substrate rotation to deposit YBCO thin films over an 8" diameter area.

A 3 inch diameter YBCO target was mounted on a US' Gun II planar magnetron sputter source. Figure 1 shows the schematic of the 90° off-axis sputtering geometry that was employed and the reference axes along which the variation in film quality was measured. The X-X axis is the lateral axis along which the distance from the target is fixed. The Y-Y axis is the longitudinal axis along which the distance from the target increases. All the films were deposited at an operating pressure of 200 mTorr in a sputtering atmosphere consisting of 80% Ar and 20% O_2. An RF power of 180 W was applied to the target which generated a self bias voltage of -65 V approximately. The films for thickness and composition uniformity measurement were deposited at room temperature on 8 inch diameter Si wafers.

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II. THICKNESS AND COMPOSITION UNIFORMITY

Due to its geometry, the thickness distribution obtained on a stationary substrate in 90° off-axis sputtering is highly non-uniform. Substrate rotation has been known to improve the uniformity of deposition. However, in order to maximize the area with uniform thickness obtained by substrate rotation, the complete thickness distribution obtained on a stationary substrate needs to be determined. Therefore, YBCO thin films were deposited at room temperature on stationary 8" Si wafers. A grid consisting of 200 μm wide lines spaced every 0.25" was prepared on these films by photolithography and wet etching. The thickness was measured at every 0.25" on the film as the depth of the trench at that point, using an Alpha Step 500 Surface Profiler. The thickness of the films ranged from 1000Å to 6000Å. The error in the measured thickness values is less than ±50Å.

Fig. 2(a) and (b) show the surface and contour plots of the measured two dimensional thickness profile on a stationary 8" Si wafer, respectively. The thickness profile obtained along the X-X axis is symmetric about the Y-Y axis because the erosion ring is also symmetric about the same axis. However, the distribution along the Y-Y axis is asymmetric and non-uniform, as expected.

One can expect that substrate rotation about a point on the Y-Y axis would average out the non-uniformities in the distribution, leading to uniform thickness distribution over a large area. However, the optimum target distance from the center of substrate rotation needed to be determined in order to obtain the maximum area with uniform thickness. A computer simulation program was used for this purpose. The simulation uses the thickness profile data obtained on a stationary substrate (Fig. 2) in an azimuthal averaging algorithm and iteratively determines the optimum distance for a prespecified thickness uniformity (<±5% variation).

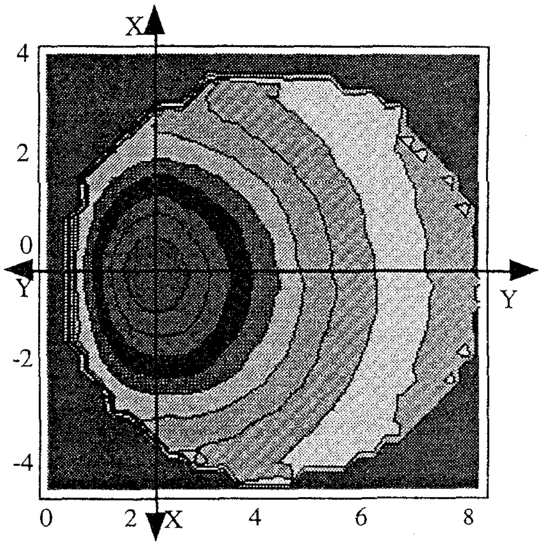

Fig. 2.(b) Contour plot (below) of the measured two dimensional thickness profile obtained from a 3" YBCO target on a stationary 8" Si wafer.

Having determined the optimum distance, we deposited YBCO thin films on substrates rotating at 12 rpm, with the target being held at the optimum distance from the center of rotation. The thickness distribution was measured in a similar manner and the variation in thickness was found to be less than ±5% over the 8" wafer. Fig. 3(a) and (b) show the thickness distribution obtained on a rotating substrate in comparison with that obtained on a stationary substrate along the X-X and Y-Y axis. Due to the averaging effect, the deposition rate obtained on a rotating substrate decreases to 172Å/hour from a maximum rate of 390Å/hour obtained on a stationary substrate.

The composition of 500Å thick YBCO films deposited at room temperature—on a rotating substrate with the target held at an optimum distance from the center of rotation—was measured by Rutherford Backscattering Spectroscopy (RBS).

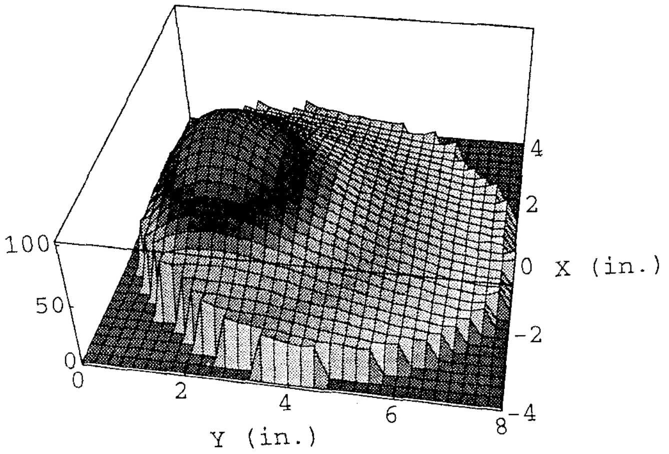

Fig. 2(a). Surface plot of the measured two dimensional thickness profile obtained from a 3" YBCO target on a stationary 8" Si wafer. The thickness is normalized on a scale of 0 to 100% .

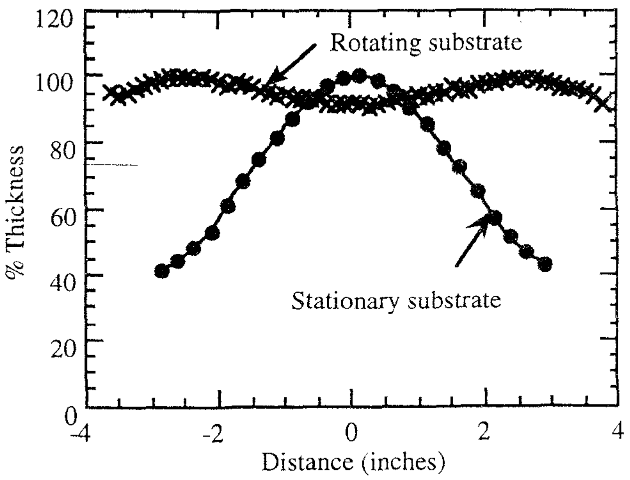

Fig. 3(a). Comparison of the thickness profiles obtained on a stationary and rotating substrate along the X-X axis .

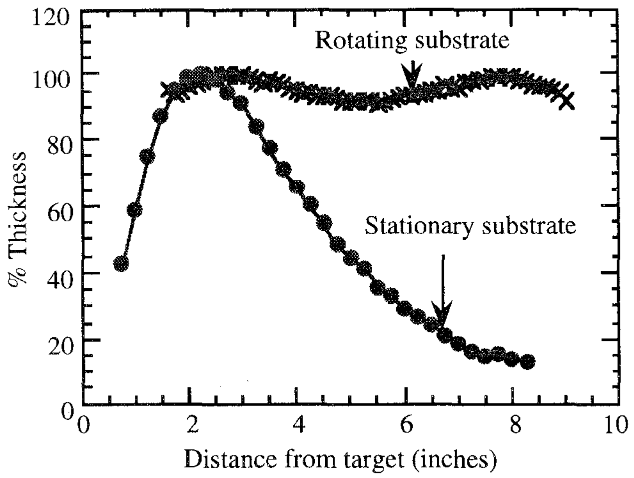

Fig. 3(b). Comparison of the thickness profiles obtained on a stationary and rotating substrate along the Y-Y axis .

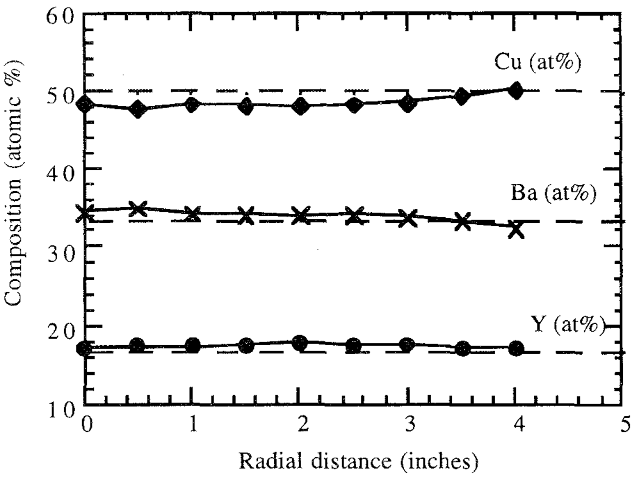

Fig. 4. The composition variation obtained on a rotating substrate in comparison with the target composition (shown in broken lines).

Fig. 4 shows the composition distribution obtained over a 4" radial distance. The target composition is shown in broken lines. These films displayed a maximum variation in composition of less than 2.5% over an 8" diameter area. The maximum deviation from the target stoichiometry was observed to be +1.2% in Y, +1.5% in Ba and -2.2% in Cu, in contrast to a maximum deviation of -1.8% in Y, -2.4% in Ba, and -3.4% in Cu obtained on a stationary substrate. The percentages mentioned here refer to the sum of Y, Ba and Cu content as 100%.

III. STRUCTURAL AND SUPERCONDUCTING PROPERTIES

In order to determine the variation in crystalline quality, texture, superconducting properties, and surface resistance (R_s) over an 8" diameter area, crystalline YBCO films were deposited at high temperatures (725°C - 735°C) on $LaAlO_3$ substrates mounted on a 3" diameter US Inc. resistive heater. To overcome the limitation imposed by the heater size, we used a rotation scheme, as shown in Fig. 5. The heater was mounted on a rotating arm that was continuously oscillated during the deposition at a speed of 12 rpm. Each oscillation consisted of a complete circular motion in one direction followed by a circular motion in the reverse direction. Thus, the samples were exposed to all the regions at the same radial distance from the center of rotation. Two deposition runs were carried out with the heater mounted at different radii from the center of rotation such that the entire 8" diameter area was studied, as shown in Fig. 5. All the films were deposited at an operating pressure of 200 mTorr (60% Ar / 40% O_2). 4000Å thick YBCO films were deposited for studying the variation in crystalline structure and film properties (T_c, J_c and R_s).

One drawback of the rotation scheme described above is that there is considerable variation in the deposition temperature for different samples made in the same run. The samples mounted closer to the center of the heater had a higher deposition temperature while the samples mounted away from the center had a lower deposition temperature. Therefore, while the growth conditions were optimized for samples at the center of the heater, the samples away from the center of the heater were not deposited under optimum conditions.

The crystalline structure of 4000Å thick YBCO films deposited on $LaAlO_3$ substrates at 735°C was analyzed using a Siemens D5000 four circle X-ray Diffractometer. The variation in crystalline quality and amount of c-axis oriented

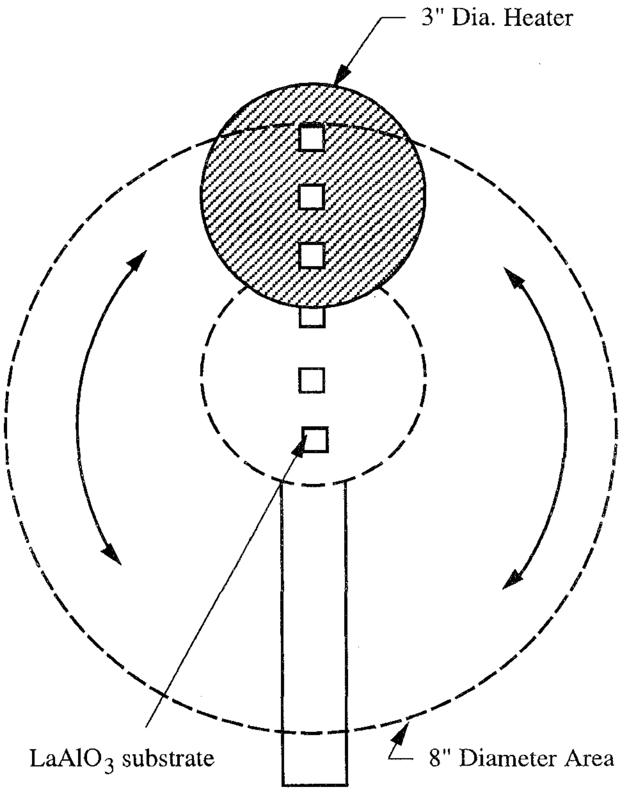

Fig. 5. Rotation scheme for depositing films at high temperature over an 8" diameter area

grains in the films was measured as a function of radial distance. The texture of the films was determined by ϕ-scans of the off-axis (102) and (104) peaks of the YBCO films. These peaks were observed at the positions corresponding to both c-axis grains and a-axis grains normal to the substrate. The average ratio of the intensities of the peaks at these two positions was used to determine the amount of c-axis grains in the films. The crystalline quality of the films was assessed by measuring the full width at half maximum (FWHM) of the rocking curve of the (005) peak of the c-axis oriented grains in the YBCO films.

Fig. 6 shows the variation in amount of c-axis grains and the FWHM of the (005) peak of the films as a function of radial distance. The films at 0.75" and 3.0" display a much higher percentage of c-axis grains (>95%) and narrower rocking curves (FWHM = 0.37°) compared to the samples deposited at other radial distances. These samples were mounted at the center of the heater, while the other samples were mounted away from the center of the heater during the two deposition runs, as shown in Fig. 6. Therefore, these two samples were deposited at optimum conditions and correspondingly displayed a higher amount of c-axis grains and were of better crystalline quality as compared to the samples at other radial distances from the center.

The resistivity of the films was measured in the Van der Pauw geometry as a function of temperature in a cryostat using a closed cycle helium refrigerator. The films deposited over an 8" diameter area consistently displayed a sharp (≤1K) superconducting transition with a T_c > 87.5K as shown in Fig. 7. The resistivity at room temperature of these films varied between 150 and 260μΩ-cm. The critical current density (J_c) of the films was calculated from the DC magnetization hysteresis loops using the Bean's formula. The magnetization hysteresis of the films was measured in an applied field range of -1 to +1 Tesla using a vibrating sample magnetometer. The J_c near zero field at 4.2K and 77K were consistently greater than 1 x 10^7 A/cm^2 and 1 x 10^6 A/cm^2 for the films deposited over an 8" diameter area, as shown in Fig. 7.

The abrupt changes seen in the variation of film properties such as T_c and J_c are not due to compositional variations, as the composition profile obtained on rotating substrates showed smooth changes (Fig. 4). These changes are due to variations in deposition temperature. The samples at 0.75" and 3.0" from the center of rotation were deposited under optimum conditions and as a result displayed higher J_c at 77K (> 3 x 10^6 A/cm^2) and 4.2K (> 3 x 10^7 A/cm^2), narrow rocking curves and a higher amount of c-axis grains compared to the samples at other radial distances. The J_c at 77K and the amount of c-axis grains display the same trend in variation over the 4" radial distance, as seen from Fig. 6 and Fig. 7. Thus, the variations in deposition temperature of the samples lead to variations in crystalline quality and amount of c-axis grains in the films, which in turn lead to abrupt variations in J_c. With a more uniform heating of the substrates, all the samples can be deposited under optimum conditions and the resulting films will be highly c-axis oriented with sharp rocking curves and have uniformly high critical current densities.

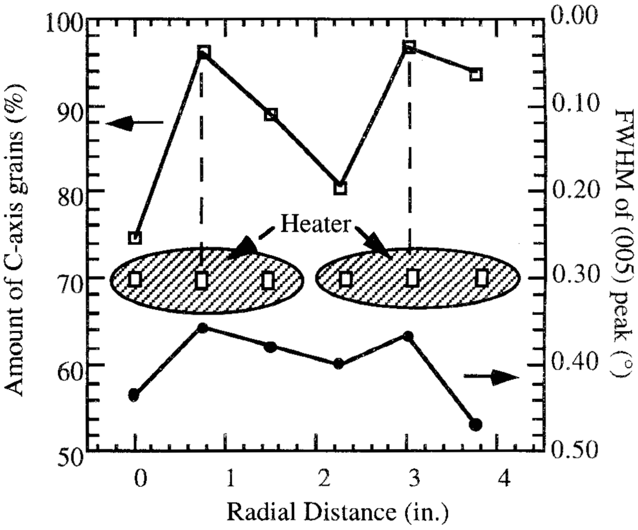

Fig. 6. Variation in amount of c-axis grains and rocking curve width of (005) peak of YBCO films as a function of radial distance

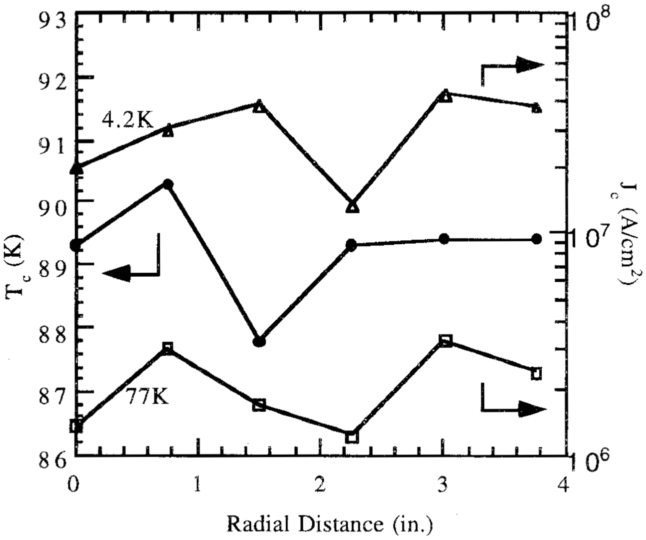

Fig. 7. The variation in T_c and J_c at 4.2K and 77K of 4000Å thick YBCO films over an 8" diameter area.

The samples for surface resistance studies were deposited on 1cm x 1cm $LaAlO_3$ substrate pairs mounted side by side on the heater. The microwave surface resistance (R_s) of the films was measured by the Parallel Plate Resonator (PPR) method [12]. The R_s of the films measured at 4.2K and 11.75GHz was found to vary between 50μΩ and 150μΩ over a 4" radial distance. No correlation could be observed between the variation in R_s and substrate temperature on the heater block.

IV. CONCLUSIONS

In summary, we have demonstrated the deposition of YBCO thin films over 8" diameter areas. The films deposited on rotating substrates displayed uniform thickness (< ±5% variation) and composition (< 2.3% deviation from the target stoichiometry) over the 8" diameter area. A consistently high transition temperature ($T_c > 87.5°K$) and critical current density ($J_{c\ 4.2K} > 1 \times 10^7\ A/cm^2$) were also observed for these films. The R_s of the films measured at 4.2K was found to vary between 50μΩ and 150μΩ. Due to the variation in substrate temperature on the heater block, the crystalline quality and the amount of c-axis grains in the YBCO films varied with radial distance. A corresponding variation was observed in the J_c of the films at 77K.

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