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Large-area double-side pulsed laser deposition of $YBa_2Cu_3O_{7-x}$ thin films on 3-in. sapphire wafers

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A pulsed laser deposition (PLD) technique for $YBa_2Cu_3O_{7-x}$ (YBCO) thin films with CeO_2 buffer layers and gold contact films on both sides of 3-in. diameter sapphire wafers, which are applied for microwave strip-line filters, is described and some results of structural and compositional characterization are given. This large-area multilayer PLD technique allows for a homogeneous and reproducible YBCO deposition on both wafer sides with inductively measured critical current densities of $3\times10^6-5\times10^6$ A/cm^2 at 77 K with a YBCO thickness of 350–500 nm. The results indicate that PLD seems to have unique capabilities for fast deposition of high-quality large area oxide multilayers. © *1996 American Institute of Physics.* [S0003-6951(96)02123-7]

Simple and fast deposition techniques for large area high-T_c superconducting (HTSC) thin films are necessary for the realization of HTSC devices, for example in microwave applications. Devices like microwave strip-line filters for satellite communication systems require HTSC thin films on both sides of single-crystal wafers. Therefore, scaling up of established HTSC deposition techniques like pulsed laser deposition (PLD) and sputtering to substrate diameters of at least 3 in. is of relevance. The first attempts to deposit $YBa_2Cu_3O_{7-x}$ (YBCO) on 3-in. diameter substrates using PLD was reported as early as three years after discovery of the YBCO high-T_c superconductor.[1] Large-area PLD of two-sided YBCO thin films on 2-in. diameter $LaAlO_3$ wafers has been performed using a rotating substrate and an offset of the fixed laser plume respective to the substrate.[2] PLD of YBCO on 3-in. diameter $LaAlO_3$ using a large rotating target of 90 mm diameter which is rastered by the laser beam results in an excellent homogeneity of superconducting properties.[3] For a nomenclature of the different arrangements of large area PLD see the reviews.[4,5] Up to now, most work on large area PLD has been done using $LaAlO_3(100)$ wafers as substrate material due to its chemical inertness and good lattice match with YBCO.[1–5] However, $LaAlO_3$ has a twin structure which disappears above 500 °C, shows a relatively high dielectric constant, is fragile and expensive. Sapphire has lower dielectric constant and loss tangent and is available as large twin-free single-crystal wafers. Due to the chemical reactivity and the larger lattice mismatch of *r*-plane sapphire the deposition of a buffer layer such as CeO_2 is necessary prior to YBCO deposition. More conventional techniques like sputtering[6] and thermal coevaporation with sequential oxidation[7] have succeeded in the deposition of CeO_2 buffer layers and YBCO thin films on sapphire wafers. Nevertheless, some sputtering techniques suffer from a low deposition rate and nonstoichiometric transfer from the target to the substrate.

Recently, a relatively simple and fast large-area PLD technique for *in situ* deposition of yttrium stabilized zirconia buffer layers and YBCO thin films using small targets of 18 mm diameter was proposed, realized, and successfully tested.[8] Presently, this PLD equipment is used for the double-side deposition of YBCO thin films with CeO_2 buffer layers and gold contact films on 430 μm thick 3-in. sapphire wafers. This letter briefly describes the optimized large area PLD technique and reports on results of compositional and structural characterization. In contrast to other deposition techniques, very high critical current densities of up to 5×10^6 A/cm^2 at 77 K are achieved for 300–500 nm thick YBCO films, routinely with large area PLD techniques.[9,10]

According to the nomenclature proposed by Greer[4,5] a simple "off axis" PLD technique is used for deposition of YBCO and gold and a "rotational/translational" PLD approach is applied for the CeO_2 buffer layers. In the off axis approach the center of the rotating substrate is offset a fixed amount from the center of the ablation plume. An offset of about 30 mm is used with a target-substrate distance of 90 mm. With the "rotational/translational" approach in addition a controlled substrate translation during deposition is utilized in order to improve homogeneity of film properties.

Figure 1 shows schematically the applied PLD arrange-

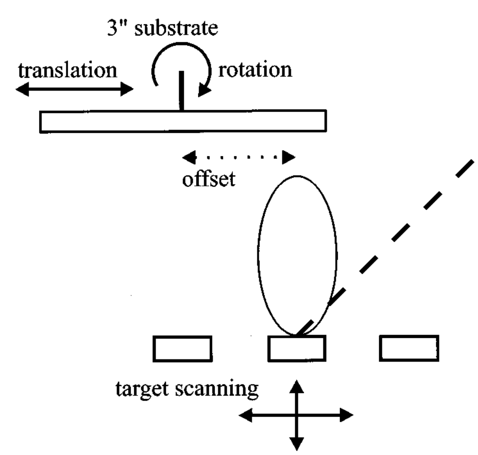

FIG. 1. Diagram of large area PLD arrangement.

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TABLE I. Properties of YBCO thin films on 10×10 mm^2 *r*-plane sapphire substrates with CeO_2 buffer layer in dependence on position at the 3-in. diameter substrate holder.

Position	T_c (K)	ΔT_c (K)	j_c(77 K) (A/cm^2)	Epitaxy (%)
Center	89.5	0.4	4.3×10^6	91
Edge	88.5	0.5	4.2×10^6	96

ment. A LAMBDA PHYSIK LPX 305*i* excimer laser is operating at 248 nm wavelength by pulse energies up to 1.2 J. Polycrystalline laser targets with diameter of only 18 mm are used for the PLD on the much larger 76 mm substrates. A hot isostatic pressed YBCO target reduces the surface roughness of the YBCO films. The rotating substrate is radiatively heated by a fixed heater element of KANTHAL wire fixed by ceramic tubes. An additional heater element is applied to the front side at the edge of the wafer in order to improve the homogeneity of substrate temperature which is very essential for homogeneous superconducting properties. The lifetime of the heater element is about 3 months with daily use. Standard PLD process parameters are applied for the YBCO deposition. The deposition rates averaged over the whole 3-in. substrate were 2.5 nm/min for CeO_2, and about 15 nm/min for YBCO and gold, when using a laser fluence of 2 J/cm^2 for CeO_2 and YBCO, and 5 J/cm^2 for gold, respectively. The laser spot size for YBCO deposition was about 5×1 mm^2.

The stoichiometry and superconducting properties of a YBCO film were tested using multiple 10×10 mm^2 sapphire samples simultaneously held in the 3-in. diameter substrate holder, as listed in Table I. Table I demonstrates the large potential of PLD for scaling up to technological important substrate diameter. The superconducting properties are measured by an inductive method without lateral structuring using a calculation of the current density distribution in the YBCO thin film.[11] The degree of epitaxy[12] as given in Table I is defined as the percentage of the epitaxial area of YBCO film relating to the total illumination area, which was about 0.02 mm^2 in our Raman experiments.

During double sided deposition on optically transparent 3-in. diameter sapphire wafers, the heater power was decreased during deposition of the first YBCO film in order to oppose the added absorption of the heater radiation by the

TABLE II. Un-normalized XRD 2Θ-ω scan intensities (10^3 cps) of selected peaks of YBCO, CeO_2, and the sapphire substrate in dependence on the radial position A (center) to E (edge) and the side of the 3-in. diameter wafer (1 or 2) of sample G 367.

Position	YBCO (003)	YBCO (005)	YBCO (006)	CeO_2 (200)	Al_2O_3 (024)
A1	39.8	72.8	44.7	37.1	30.2
B1	38.0	68.7	40.5	35.5	27.5
C1	38.1	51.9	37.3	35.2	29.7
D1	39.1	61.1	38.9	37.5	27.9
E1	41.9	82.9	37.9	43.5	29.3
A2	37.0	108.4	32.2	40.9	26.3
B2	35.1	114.7	32.7	44.5	28.9
C2	35.8	92.1	32.0	40.2	28.0
D2	35.2	100.9	32.1	39.8	25.7
E2	41.4	118.6	36.2	51.9	30.5

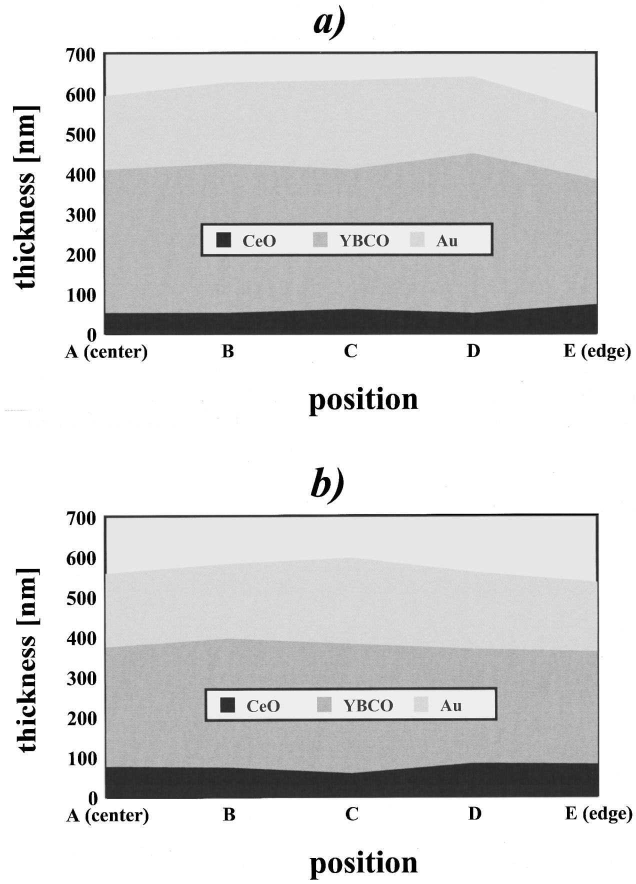

FIG. 2. Typical homogeneity of film thickness of Au, YBCO, and CeO_2 on side 1 (a) and side 2 (b) on the 3-in. sapphire wafer G 367, respectively.

growing film. To meet the stability conditions of the YBCO phase diagram, at first the CeO_2 buffer layers on both wafer sides must be deposited by an oxygen partial pressure of about 5×10^{-4} mbar. The gold contact and protection films of about 200 nm thickness should be deposited immediately after cooling down of the two YBCO films in order to achieve low specific contact resistance of less than 10^{-6} Ω cm^2. Table II compares XRD 2Θ-ω scan intensities of selected peaks at five different radial positions of a double side Au/YBCO/CeO_2 film system on 430 μm thick 3-in. diameter *r*-plane sapphire wafer deposited by *in situ* PLD. The results of Table II have been obtained by a SIEMENS KRISTALLOFLEX diffractometer with position sensitive x-ray detector. No additional normalization of the XRD intensities was performed. The YBCO (003) and YBCO (006) diffraction peak intensities of each side deviate only by 10%–20%. The intensity of the (100)- and (200)-reflections of *a*-axis oriented grains of the YBCO films on both wafer sides is estimated to be less than 1% of the intensity of the *c*-axis reflections.

Homogeneity of thickness and composition is routinely monitored by secondary neutrals mass spectrometry (SNMS). Contrary to secondary ion mass spectrometry (SIMS) the analysis of sputtered neutrals by SNMS allows for a more accurate determination of elemental concentrations (Ref. 13 and references therein). As expected from the homogeneity of superconducting properties listed in Table I, no differences of elemental concentration and film thickness, respectively, were obtained for samples at the center and

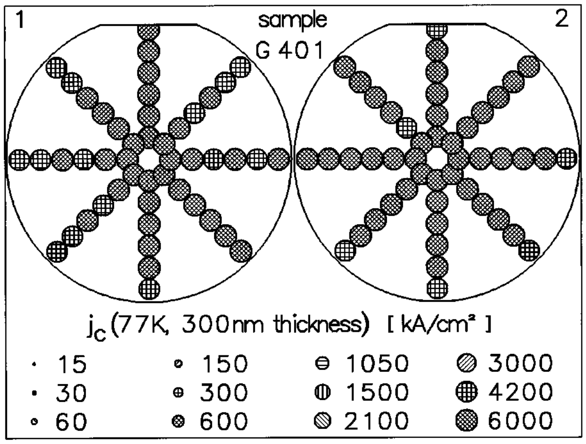

FIG. 3. Homogeneity of the critical current density at 77 K on both sides of a 3-in. HTSC wafer.

edge position on the 3-in. diameter substrate holder by SNMS depth profiling within the measuring accuracy of less than 10% (compare Table I and Ref. 14). Figure 2 shows the thickness homogeneity under routine deposition conditions extracted from SNMS concentration depth profiles at five radial points of each substrate side. For determination of sputtering rates and film thickness the depth of the sputtering craters of the SNMS measurement is measured by a Dektak surface profiler.

Figure 3 demonstrates the obtained homogeneity of the critical current density at 77 K on both sides of the 3-in. sapphire wafer with *in situ* deposited double side CeO_2/YBCO/Au film system. For the side selective j_c measurement of double-sided 3-in. diameter wafers a six-coil arrangement was designed by modification of the above-mentioned two-coil technique.[11] YBCO films with very high critical current densities of $3\times10^6-5\times10^6$ A/cm^2 at 77 K with a thickness of 350–500 nm have been routinely obtained by the described large area PLD technique. Sometimes microcracks have been observed in YBCO films on *r*-cut sapphire due to the difference in thermal expansion. However, the area density and crystallographic appearance varies considerably across a given surface and from sample to sample (for details see Ref. 15). Therefore, further investigation of microcracking and its influence on electrical performance is in progress.

In conclusion, a PLD technique is presented which allows the reproducible double-side coating of 3-in. sapphire wafers by thin Au/YBCO/CeO_2 multilayers with critical current densities up to 5×10^6 A/cm^2 by 77 K. Microwave *C*-band strip-line filters with a bandwidth dependent insertion loss of 0.2–0.6 dB have been structured routinely from these PLD HTSC wafers by Robert Bosch GmbH Stuttgart, Germany. The application of these planar HTSC strip-line filters was demonstrated by a two-channel input multiplexer for satellite communication systems. From these results, we are hopeful for the further development of the HTSC device technology.

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