

High-quality Y-Ba-Cu-O Thin Films by PLD – Ready for Market Applications

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Abstract—Large-area pulsed laser deposition (PLD) has reached a state in terms of film quality and reproducibility which makes possible now real market applications of PLD-YBa₂Cu₃O_{7-δ} (YBCO) thin films on both sides of R-plane sapphire substrates as HTSC devices in satellite and mobile communication systems. Bandpass filters optimized from PLD-YBCO thin films presently fulfill the requirements of the main national companies which are active in future communication techniques.

A relatively simple PLD arrangement with fixed laser plume and rotating substrate, with an offset between the laser plume and the center of the substrate is employed to deposit laterally homogeneous 3-inch diameter Ag-doped YBCO thin films. With the experience of more than 1,000 double-sided 3-inch diam. films a high degree of homogeneity and reproducibility of j_c and R_s is reached. The extension up to 8-inch substrate diameter will increase the productivity of the flexible PLD technique considerably.

Index Terms—High-temperature superconducting thin films, microwave devices, pulsed laser deposition, YBa₂Cu₃O_{7-δ}.

I. INTRODUCTION

High- T_c superconducting (HTSC) Y₁Ba₂Cu₃O_{7-δ} (YBCO) thin films on low dielectric loss substrates are suitable candidates for applications as passive microwave devices in future communication systems. There is a huge number of activities in many countries to develop microwave devices using HTSC thin films, because in this field real market applications of HTSC subsystems seem to be possible in the nearest future [1], [2].

Devices like microwave stripline filters for satellite communication systems require HTSC thin films on both

sides of large-area single crystal wafers. Therefore, to support the breakthrough of HTSC microwave devices to real market applications, highly reproducible and well established HTSC deposition techniques are necessary. Only a few processes worldwide as for example thermal coevaporation up to now fulfill these quality requirements for large-area HTSC thin films.

The first attempts to deposit YBa₂Cu₃O_{7-δ} (YBCO) on 3-inch diameter substrates using PLD was reported as early as three years after discovery of the YBCO high- T_c superconductor [3]. For a nomenclature of the different arrangements of large area PLD see the reviews [4], [5]. A highly reproducible PLD process for large-area 3-inch diameter and double-sided YBCO films on sapphire substrates for microwave applications is developed and continuously improved at University of Leipzig [6]–[10]. The electrical and microwave performance of these PLD-YBCO films [8] is comparable to high-quality films deposited by other techniques as e.g. thermal coevaporation.

This paper describes the state of the art of the development of a highly reproducible large-area PLD technique for HTSC YBCO thin films on sapphire wafers by comparing maps of the critical current density, and presenting detailed structural and electrical film data. Some special growth features of PLD-YBCO-films are demonstrated by selected TEM cross sections.

II. EXPERIMENT

YBCO:Ag thin films and CeO₂ buffer layers are deposited by PLD, using a KrF excimer laser, on both sides of 3-inch diameter R-plane sapphire wafers of 430 μm thickness [6]–[8]. Despite the simple deposition principle of PLD there is a huge number of difficult to handle influences which may affect the quality of the deposited films. For example, accurate control of the laser pulse energy and of the substrate temperature, homogeneous target ablation, and the cleaning of the deposited laser entrance window are extremely important for a highly reproducible PLD process.

According to the nomenclature proposed by Greer [4, 5] a simple "off axis" PLD technique is used for deposition of CeO₂, YBCO, and gold. In the "off axis" approach the center of the rotating substrate is offset a fixed amount from the center of the ablation plume. The Ag-content of the PLD-YBCO target was optimized with respect to the critical current density (j_c) and microwave surface resistance (R_s), and the mechanical film properties. High quality films have

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been deposited using sintered YBCO targets with 3 to 5 weight-% Ag. Compared to Ag-free PLD-YBCO films, Ag doping of YBCO leads to reduced surface roughness and slightly lower R_s , and improved homogeneity of R_s [9], [10]. From SNMS depth profiles we have estimated the Ag-content in the PLD-YBCO:Ag films to be only about 0.02 % despite the much higher Ag-content of 4 % of the used PLD-target.

The double-sided films are deposited subsequently at substrate temperature around 760°C. The deposition rate averaged over the whole 3-inch substrate was optimized to about 0.3 nm/min for CeO₂, and about 10 nm/min for YBCO and gold, when using a laser fluency of 2 J/cm² for CeO₂ and YBCO, and 5 J/cm² for gold, respectively. All parameters of the large-area PLD process are controlled by a sophisticated PC-software and a log file is saved for each deposition run. During deposition of about one thousand 3-inch diameter sapphire wafers a high degree of film quality and reproducibility is reached. In order to further improve the efficiency of the PLD process as a series deposition method a 8-inch diameter substrate heater was developed which is shown in Fig. 1. This 8-inch diam. substrate heater will allow a simultaneous PLD of three 3-inch diam. YBCO-films.

X-ray analysis of YBCO-films was performed at Max-Planck-Institute for Microstructure Physics in Halle/Saale using a PHILIPS X'Pert diffractometer. Critical current density is routinely scanned by the side sensitive and inductive method [11]. Microwave surface resistance was measured with dielectric sapphire resonator at 5.8 GHz and 8.5 GHz in Leipzig and Wuppertal, respectively, and scaled to 10 GHz (see [10]). R_s mapping was carried out in Karlsruhe at 145 GHz (see [10]). TEM was carried out with a PHILIPS CM 30 T microscope equipped with an EDX analyzer at the National Center for HREM, The Netherlands.

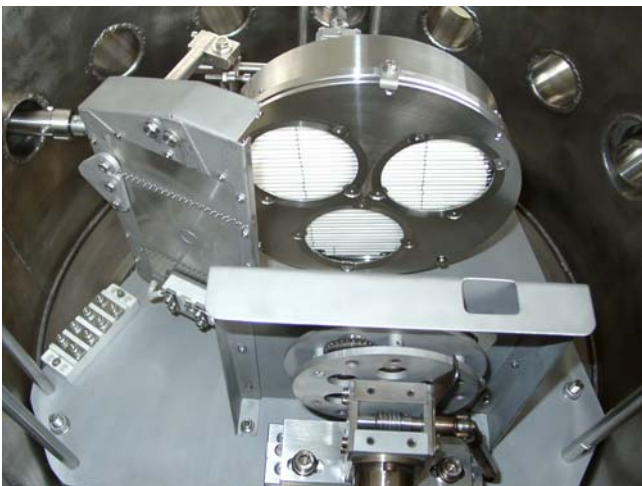


Fig. 1. New PLD chamber with 8-inch diam. substrate heater (top) which will allow simultaneous deposition of three 3-inch diam. films and target manipulator for three planar targets (bottom). Design by D. Natusch.

III. REPRODUCIBILITY OF LARGE-AREA PLD

Fig. 2 demonstrates the obtained lateral homogeneity and reproducibility of the critical current density of the double-sided 3-inch diam. YBCO films on sapphire. In more detail,

Fig. 2 shows the j_c -scans of 10 consecutively deposited double-sided YBCO films with sample No. S 100 to S 110. High critical current densities of 4 to 5.5 MA/cm² at 77 K with a YBCO thickness of about 250 nm have been obtained routinely by the described large area PLD technique. The variation of j_c from sample to sample is of nearly the same quantity as for one and the same 3-inch diam. YBCO film.

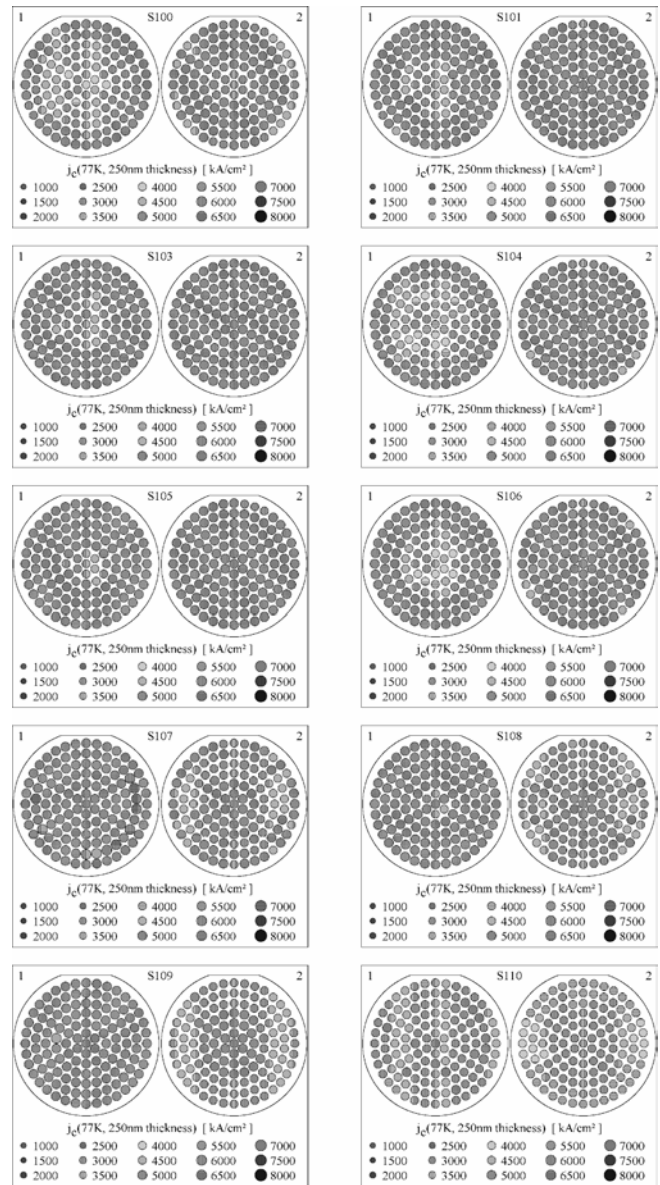


Fig. 2. Short term reproducibility of the large-area PLD-Process: j_c maps of 10 consecutively deposited double-sided 3-inch diam. YBCO:Ag thin films on R-plane sapphire. Each single j_c -mapping shows results for side one (left) and side two (right) of the same wafer separately.

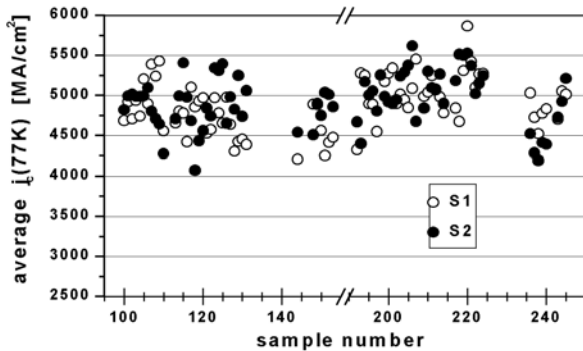


Fig. 3. Long-term reproducibility of the large-area PLD process: j_c values laterally averaged over the whole YBCO films at side one (open symbols) or side two (solid circles) of 76 double-sided 3-inch diam. sapphire wafers deposited within one year from 1999 to 2000. All of these wafers showed excellent performance as microwave filters.

Fig. 3 demonstrates the long-term reproducibility of the PLD process. 76 double sided 3-inch diam. YBCO films deposited within one year period all show very high average critical current density on both wafer sides well above 3.5 MA/cm^2 at 77 K and $B=0$. The remaining scattering of j_c values in Fig. 3 is due to added uncertainties of PLD process and inductive j_c measurement. Figures 2 and 3 are a proof for the good reproducibility of the large-area PLD, which is obtained after more than 5 years optimization work on the process. Indeed, C-band filters with very good bandpass performance which are suitable for satellite communication subsystems have been structured from PLD-YBCO:Ag thin films at Robert BOSCH GmbH [8]-[10]. Therefore, the PLD process is space qualified now to ensure the future use of PLD-YBCO films in space born communication technology [12].

Table I summarizes main structural, superconducting and microwave properties of typical PLD - YBCO:Ag films. For details concerning R_s measurements see [10], [13]. The X-ray and Raman measurements have been done on selected wafers with slightly varying R_s values to prove recently obtained correlation to film microstructure [13], [14].

TABLE I

TYPICAL STRUCTURAL AND SUPERCONDUCTING PROPERTIES OF DOUBLE-SIDED 3-INCH DIAM. PLD-YBCO THIN FILMS ON SAPPHIRE SUBSTRATES

Property	Typical Value
YBCO-thickness d_{YBCO}	$(230 \pm 30) \text{ nm}$
c-axis lattice constant c	$(11.659 \pm 0.006) \text{ \AA}$
a-axis lattice constant a	$(3.829 \pm 0.003) \text{ \AA}$
orthorhombic splitting $(b-a)/a$	$(1.52 \pm 0.05) \%$
percentage of a-axis oriented grains	0 to 0.5 %
in-plane epitaxially oriented film area to total illuminated area in Raman spectroscopy	75 to 94 %
critical temperature T_c	87.5 to 90 K
critical current density j_c at 77 K and $B=0$	3.5 to 5.5 MA/cm^2
surface resistance R_s at 10 GHz and 77 K	500 to 600 $\mu\Omega$
microwave surface magnetic field B_s at 8.5 GHz and 77 K for $R_s + 20 \%$ [15]	8 to 10 mT
surface resistance R_s at 145 GHz and 77 K	40 to 60 m Ω

IV. TEM CROSS-SECTIONS OF PLD-YBCO FILMS

Two typical results of the cross sectional transmission electron microscopy (TEM) investigations of PLD-YBCO:Ag thin films on CeO_2 buffer layer on sapphire substrate are shown in Fig. 4 and 5. These figures are selected from a large number of TEM pictures taken at both wafer sides and at different radial positions of six wafers to ensure TEM results which are representative for the PLD YBCO films.

Usually, the interface of YBCO and the CeO_2 buffer layer seems to be atomically flat. At the substrate/ CeO_2 interface, occasionally an amorphous or polycrystalline reaction layer was found (see Fig. 4 and 5). Often a „wavy“ TEM contrast can be observed in the YBCO region with uniform stress distribution in the (001) planes (Fig. 4). Sometimes microcracks of the CeO_2 buffer layer facilitate the formation of interface precipitates which, however, do not introduce any considerable film surface roughness. Due to the very high film growth velocity in PLD the interface precipitates are overgrown after some monolayers by well c-axis textured YBCO:Ag as shown clearly in Fig. 4 and detailed in Fig. 5.

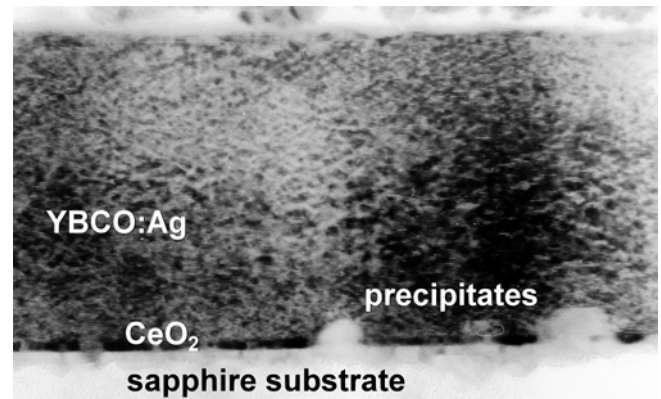


Fig. 4. TEM cross section of a typical PLD YBCO:Ag thin film with CeO_2 buffer layer on sapphire substrate with some interface precipitates overgrown by c-axis textured YBCO:Ag. To give an estimate for the length scale, the YBCO thickness is about 220 nm in this figure.

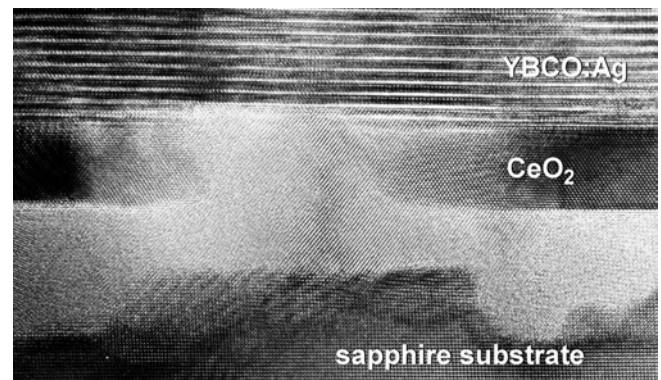


Fig. 5. High resolution TEM cross section of the interface region of a typical PLD-YBCO thin film showing a break of the buffer layer which nevertheless influences the perfect YBCO growth only for about two monolayers. At the CeO_2 / sapphire interface a crystalline interdiffusion layer occurs. The length scale is given by the c-axis lattice constant of 11.66 \AA in this figure.

In this way, the PLD YBCO films on sapphire substrates show minor microstructure imperfectness as for example

stress modulation and interface defects. Nevertheless, the PLD YBCO films show state of the art electrical and microwave properties.

V. CONCLUSIONS

In conclusion, a PLD technique is presented which allows the fully reproducible double-sided coating of 3-inch diameter sapphire wafers by thin Au/YBCO/CeO₂ multilayers with laterally homogeneous critical current densities of 3.5 to 5 MA/cm² at 77 K with about 230 nm film thickness. It is a well known fact that YBCO thickness on sapphire substrates is limited to about 250 – 300 nm due to the differences in thermal expansion coefficients to prevent microcracking of YBCO. The lateral variation of j_c within one and the same 3-inch diameter YBCO-film is of the same quantity as the j_c -variation from wafer to wafer for consecutively deposited samples (Fig. 2 and 3). Laterally homogeneous maps of microwave surface resistance R_s of about 40 mΩ at 145 GHz and 77 K have been measured for selected PLD-YBCO films (Table I). Homogeneous PLD-YBCO films show lateral variations of R_s in the range of the statistical fluctuation of the measurement. However, up to now the variation of microwave surface resistance from wafer to wafer is higher [10], [13]. The R_s at 8.5 GHz and 77 K determined in the center position of the YBCO:Ag films remains constant at about 370 μΩ up to a microwave surface magnetic field of about 10 mT (compare Table I). Some correlation of R_s and film microstructure is given in [14].

By long term process optimization and by extensive electrical, microwave, and microstructure investigations ([9]-[11], [13], and Fig. 4, 5, and Table 1) a very reproducible and stable PLD process for double-sided YBCO:Ag thin films on R-plane sapphire wafers with CeO₂ buffer layer could be established. The PLD-YBCO:Ag films are suitable for the envisaged microwave applications in future communication systems of Robert BOSCH GmbH Stuttgart, Germany. The flexible PLD-technique seems to be advantageous compared to other deposition techniques, particularly if more complicated multilayer systems as for example YBCO / SrTiO₃ films for use as electrically tunable microwave filters have to be deposited.

At present, even highly reproducible deposition processes result in YBCO thin films with microstructure defects. According to the about 4 times lower microwave surface resistance of very pure YBCO single crystals compared to thin films [16], there seems to be a further great potential for improvement of the microwave performance of YBCO thin films. However, the presented results show the principal suitability of PLD as a series deposition technique for complicated multielement multilayers for advanced applications in the microwave communication technique of the future.

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