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Optimization of YBCO thin films grown on MgO for microwave applications

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Summary

$YBa_2Cu_3O_{7-x}$ thin films have been grown on (100) MgO substrates by the Pulsed Laser Deposition (PLD) technique. Films properties of relevance for microwave applications were studied for different substrate preparation and deposition parameters. It is reported on the influence of composition deviations, substrate surface preparation and deposition parameters on Tc (R=0), Jc (77 K), the crystalline texture and the surface morphology of the films. An improvement of the PLD technique aimed at the optimizing of HTS thin film growth for electronics and microwave systems is proposed.

1. Introduction

The fabrication of reliable, high quality high-Tc superconducting (HTS) thin films on large surface areas is a prerequisite for the microwave application field. Although remarkable achievements have been reported in literature, mostly with the $YBa_2Cu_3O_{7-x}$ (YBCO) compound, in terms of both superconducting properties of films (1) and performances of passive microwave demonstrators (2,3), numerous problems still have to be overcome at each step of the fabrication process up to the final components. These steps include film preparation and substrate related problems, film patterning, packaging and cryogenic aspects, etc...

The present study is concerned with the first of these steps and is directed towards the optimization of (YBCO) film growth on (100) MgO substrate for passive microwave devices applications. The PLD growth equipments and characterization techniques used are briefly described ; the influence of substrate preparation and film deposition conditions, namely : film composition, growth temperature and oxygen pressure, on the superconducting (Tc (R=0), Jc (T)) and physical properties (texture, surface morphology) of the films is outlined. On the basis of these investigations, general trends for optimized film growth are given and future developments of the PLD technique in view of achieving well controlled, high quality films on large surface areas are proposed.

2. Experimental

YBCO films were grown by the PLD technique (4) using two equipments described elsewhere (5,6). Laser ablation of high density (> 0.9), stoichiometric YBCO targets was made using a 248 nm KrF laser (Lambda Physik LPX 200) in one equipment (multi-target system, noted 1) and a 308 nm laser (SOPRA 510) in the other one (single target system, noted 2). In both cases, pulse fluence and duration were 1-2 J/cm2 and 30 ns respectively for a repetition rate of 2 Hz. Substrate-target distances were set at 5 and 6 cm respectively. Growth was made under pure oxygen gas ; immediately after growth, the oxygen pressure in the chamber was increased to 300 mbar and temperature was decreased at a rate of typically 10 °C/min.

The crystalline texture of the films was analyzed by conventional X-ray diffraction and precession techniques (7). Scanning electron microscopy (SEM) and Rutherford back scattering spectroscopy (RBS) were applied to study film surface morphology and cationic composition. Substrate-film interface and the roughness of substrate surface were studied by high resolution transmission electron microscopy (HRTEM) and atomic force microscopy (AFM) respectively. R(T) curves were measured by the classical four probe technique and Jc(T) was measured along channels 50 μm x 200 μm in size with a 10 μV/cm criterion.

3. Results and discussion

3.1. *Compositional effects*

Deviations from optimal cationic composition have been shown to adversely affect the superconducting and morphological properties of films (8). This was clearly confirmed in this study as exemplified for Tc (R=0) in fig.1. This feature limits film deposition by the PLD technique to surface areas of a

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few square centimeters. For larger areas, improved PLD equipments as discussed in the following may be necessary.

was not a major parameter in this study. Main results, discussed below, are shown in figs. 4 to 6.

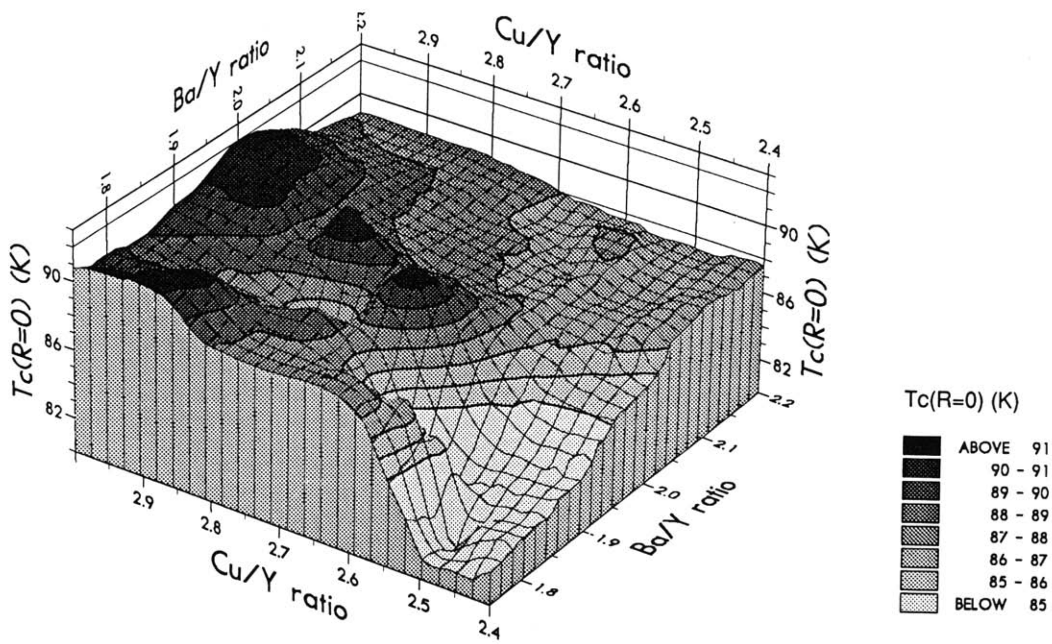

Figure 1. Tc (R=0) versus (Ba/Y) and (Cu/Y) reduced contents in films grown at T,P values of 750 °C and 0.2 mbar respectively.

3.2. Substrate related effects

MgO substrates are extensively used, especially for the preparation of microwave passive demonstrators. Despite its well known advantages, this material has its own limitations due to i) grown-in imperfections as inclusions and holes and ii) its surface sensitivity to chemical agents and moisture. A severe limitation of MgO substrates is in the frequent formation of a disordered film texture where grains are rotated about their common c axis perpendicular to the substrate surface. This texture, often specified by the surface coverage of grains having their a, b directions rotated at a 45° angle off the [100], [010] axes of the substrate (c ⊥ 45° grains, fig.2), is known to be associated with large increases of the surface resistance at microwave frequencies (8). It was found in this study that the onset of this mosaic texture was primarily linked to the preparation of the substrate surface and that it could be totally prevented. Other deviations from the c ⊥ 0° ideal texture are due to substrate imperfections as shown in fig.3.

3.3. P,T diagramme

Despite the different characteristics of PLD equipments 1 & 2, the characteristics of the films did not significantly differ, thus indicating that the deposition rate (five times larger in equipment 1)

The analysis of figs. 4 & 5 suggests that best quality films are obtained along a ridge in the P,T diagramme, which is approximately parallel to the YBCO thermodynamic stability limit. It may be noted that best quality films 300 nm thick exhibit Tc (R=0) and Jc (77 K) values of 92 K and 4×10^6 A/cm^2 respectively.

The achievement of perfect film surfaces is a prerequisite for device applications. Therefore, surface roughness due to pinholes, outgrowths, droplets etc... should be minimized and eventually eliminated. Micrographs in fig. 6 show that i) surface morphology critically depends on the P,T parameters and ii) niches in the P,T diagramme yield high quality surfaces.

4. Concluding remarks

The most significant results of this work may be summarized in four main points.

Firstly, it is mandatory to achieve films with a quasi-stoichiometric formulation of the cationic species. This requirement has direct implications for the growth of films on large wafers (up to 10 cm in diameter) by the PLD technique as there is an angular distribution of metallic species in the plasma plume. The new PLD equipment (FLAME from Alcatel CIT) being installed at Alcatel Alsthom

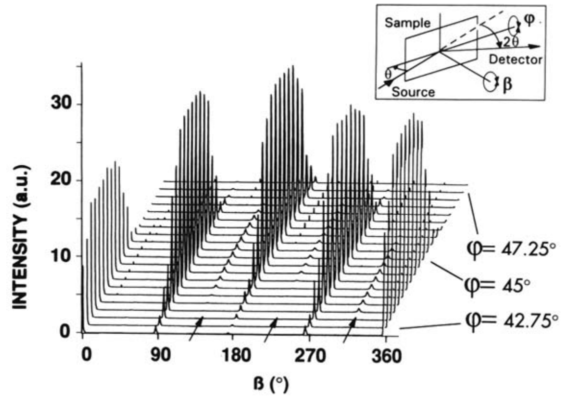

Figure 2. X-ray pole figure [103] ; diffraction peaks due to c ⊥ 45° grains are indicated by arrows.

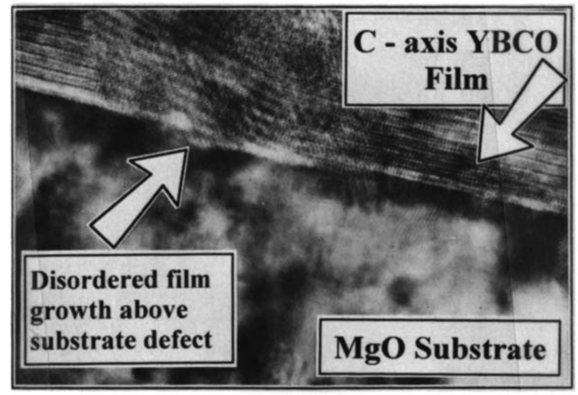

Figure 3. HRTEM cross-section micrograph of the MgO/YBCO layer interface. Observe the loss of the c ⊥ growth at a substrate surface defect.

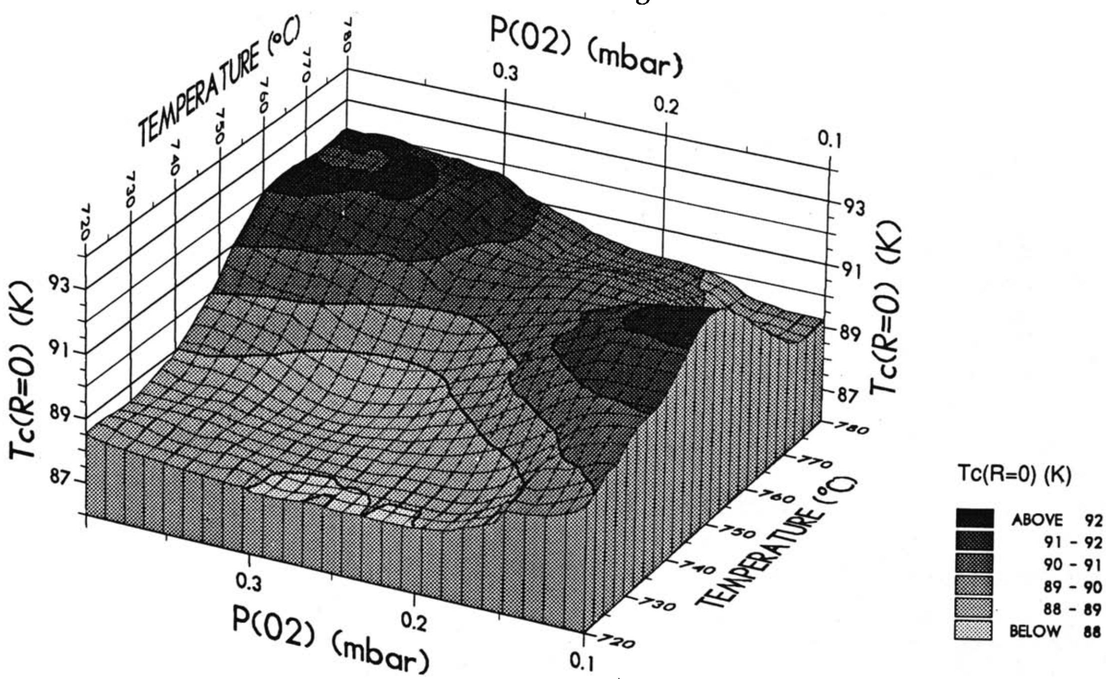

Figure 4. Tc (R=0) in the P,T diagramme. Films 300 nm thick were quasi stoichiometric.

Recherche has been conceived to meet this requirement. It permits the growth of films on wafers 50 mm in diameter with controlled cationic composition and thickness across the wafer. One originality of the FLAME machine is in the selection of the plume cone where the cationic composition of the target is readily transfered.

Secondly, the control of film texture on MgO substrates appears to primarily depend on the physico-chemical state of the substrate surface. By careful substrate preparation techniques and appropriate conditioning and handling procedures, it has been possible to virtually eliminate the formation of c ⊥ 45° grains as reported in other works (8).

Thirdly, optimal film quality appears to occur in the P,T diagramme along a line parallel to the YBCO thermodynamic stability line.

Fourthly, optimal film morphology, specified by a single texture and a smooth surface, may be achieved at relatively low growth temperature and oxygen pressure values.

The latter two observations indicate that high quality YBCO films may be grown at low deposition temperature and oxygen pressure values i.e. in a P,T range well below that explored in this study. This may be achieved by completing the PLD technique with a high density source of atomic oxygen as was shown in ref. 9.

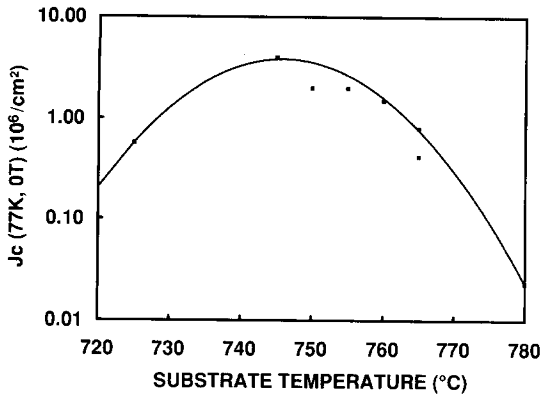

Figure 5. Jc (77 K) versus growth temperature T along the P = 0.2 mbar cut of the P,T diagramme.

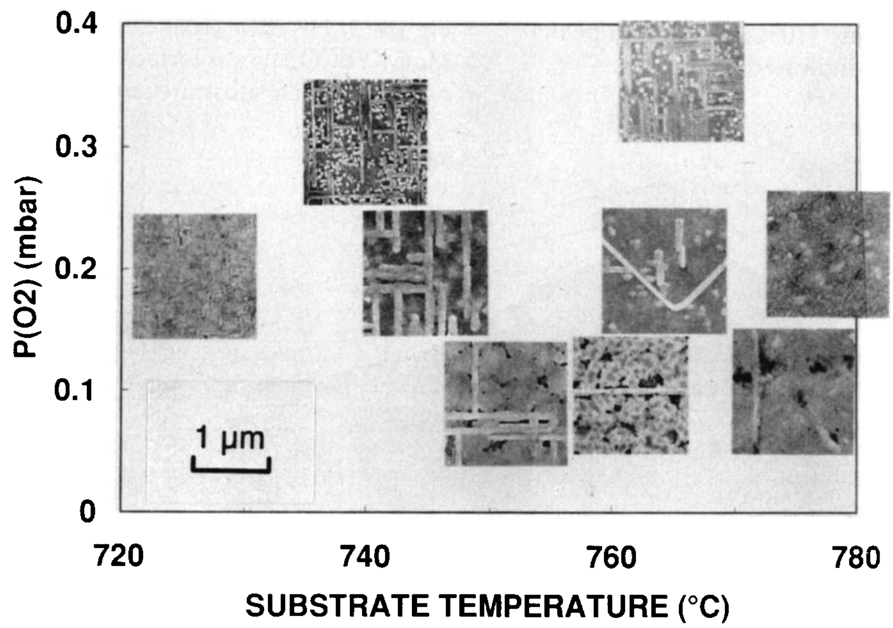

Figure 6. SEM micrographs of the surface of films 300 nm thick (equipment 2) versus P,T.

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