# Conditions for the textured growth of Y-Ba-Cu-O films on Zr foils

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# Abstract

 $Y_1Ba_2Cu_3O_y$  films were grown in situ on oxidized Zr foils by pulsed laser ablation. The substrate temperature and the oxygen partial pressure were varied from 500 °C to 800 °C and between 0.01 mbar and 1 mbar, respectively, in order to determine the optimum conditions for a c-textured growth. Particular attention was paid to the special features of metallic substrates as oxidation, surface roughness, and rolling texture. X-ray diffraction experiments and resistance measurements were performed in order to study the texture and the superconducting properties of the films. For a substrate temperature of 750 °C and an oxygen partial pressure of 0.3 mbar a superconducting transition temperature of 89 K (midpoint) was obtained.

# 1. Introduction

Since the discovery of high T<sub>c</sub> superconductors various techniques have been reported to produce high quality thin films of these materials [1]. In laser deposited  $Y_1Ba_2Cu_3O_v$  (YBCO) films on single crystalline substrates, for instance  $SrTiO_3$  and MgO, superconducting transition temperatures  $T_c$  above 90 K and critical current densities of more than 10  $^{6}$  A/cm<sup>2</sup> at 77 K are obtained [2,3,4]. To achieve these high critical current densities, a well aligned epitaxial growth of the YBCO grains with the c-axis perpendicular to the substrate surface is required. Nevertheless, also on amorphous substrates, like Si with amorphous  $SiO_2$  top layer, c-textured growth can be obtained [5]. The lack of an a/b-orientation as well as the interdiffusion between substrate and film impairs the superconducting properties of the films on Si. To reduce the diffusion, various materials are used as buffer layers. Among these, especially  $ZrO_2$  has been proved as a suitable diffusion barrier [6,7].

For this reason we deposited YBCO films directly on oxidized Zr foils by pulsed laser ablation. In this paper we report on the optimum growth conditions with respect to the oxygen partial pressure vs. temperature p(T) diagram. We show a correlation between the structure, the microstructure, and the superconducting properties. Moreover, the influence of the rolling texture and the surface roughness of the foils on the properties of the films is investigated.

# 2. Experimental procedures

For the film deposition a KrF excimer laser (Lambda Physik LPX 110i) was used. A repetition rate of 5 Hz, an energy density of about  $3 \text{ J/cm}^2$  and a target-to-substrate distance of 4 cm were taken. The Zr foils were oxidized in the ablation chamber during heating to the deposition temperature in an oxygen partial pressure of 0.3 mbar. At this, the typical substrate temperature of 750 °C was reached in about 5 min. The oxide barrier attained a thickness of about 2.5  $\mu {
m m}$ after this time. During film deposition, the ZrO<sub>2</sub> (monoclinic) intermediate layer grew further to a thickness of about 5  $\mu$ m. In order to reduce the surface roughness of the substrates and to remove possible surface contaminations, most of the substrates were polished using diamond paste with decreasing grain sizes from 10  $\mu$ m down to 0.25 μm.

### 3. Results and discussion

In Fig. 1 typical X-ray diffraction patterns (Co-K<sub> $\alpha$ </sub> radiation) of films deposited on unpolished (a) and polished (b) Zr foils are compared. The intensity ratios of the Zr and ZrO<sub>2</sub> reflections can be used as an indicator for the oxidation state of the foils. In Fig. 1 (a) the preferred appearance of the YBCO (001) reflections shows a pronounced c-texture of the film. The reduced intensity of the (110)/(103)/(013) reflection, briefly referred to as (110), demonstrates the



Fig. 1. X-ray diffraction patterns of YBCO films on unpolished (a) and polished (b) Zr foils.

existence of a small fraction of randomly oriented grains. Contrary to this, Fig. 1 (b) shows only the (001) reflections of YBCO indicating that the film on polished Zr is completely c-textured. Due to this improvement of the film texture, for the further investigations only polished Zr foils were used as substrates.

Besides the surface morphology of the foils also the substrate temperature and the oxygen partial pressure are crucial parameters for the growth of YBCO films. Fig. 2 shows the orientation of a series of YBCO thin films for different substrate temperatures T<sub>Sub</sub> and a fixed oxygen partial pressure of 0.3 mbar. The film texture can be described by comparing the X-ray intensities of the (110), (200), and (006) reflections. At substrate temperatures up to 550 °C the films grow without any preferred orientation. While at about 575 °C a predominant a-orientation is observed, c-textured growth appears with further increasing substrate temperature. Above 650 °C all films were found to be completely c-textured. In order to characterize the c-texture of these



Fig. 2. Correlation between the substrate temperature and the film orientation, characterized by the intensity ratios I(110)/I (statistical), I(200)/I (a-orientation) and I(006)/I (c-orientation). I=I(110)+I(200)+I(006).

films in detail, rocking curves of the YBCO (006) reflection were determined. As can be followed in Fig. 3, a decrease of the half width of the rocking curve occurs with increasing temperature. The best value of 2.7<sup>0</sup> is reached at 775 °C. Additionally, the a/b-orientation within in the substrate plane was determined by four-circle diffractometry (Mo- $K_{\alpha}$  radiation). The (103)/(013) pole figures of films deposited on unpolished (a) and polished (b) Zr foils at 750  $^{0}$ C and 0.3 mbar O<sub>2</sub> are compared in Fig. 4. Corresponding to the annular maximum both films are c-textured, but the half width in radial direction is reduced for the film on polished Zr due to the improvement of the c-texture discussed



Fig. 3. Temperature dependence of the rocking curve full width at half maximum.



Fig. 4. (103)/(013) pole figures of YBCO films on unpolished (a) and polished (b) Zr foil.

above. Moreover, the appearance of four intensity peaks in toroidal direction in both pole figures reveals a partial a/b-orientation of the grains. The in-plane ordering was also directly observed by scanning tunneling microscopy [8].

In order to understand these results, detailed texture measurements of the Zr foil and the oxide layer were performed (not shown here). It was found that a rolling texture occurs in the sheets, which is described by two preferred orientations. As the  $ZrO_2$  layer also grows highly textured, we conclude that the rolling texture of the sheets, mediated by the oxide layer, leads to the a/b-alignment of the YBCO films.

The c lattice parameter, determined from

Fig. 5. Correlation of substrate temperature and lattice parameter c.

the (001) reflections of X-ray diffraction spectra, strongly depends on the substrate temperature for a fixed oxygen partial pressure of 0.3 mbar. In the temperature range from 700  $^{\circ}$ C up to 750  $^{\circ}$ C the length of the c axis gets less than 11.70 Å. Thus, in this range YBCO films with a low density of lattice defects and high oxygen content close to 7 can be prepared. Minimal lattice parameters of 11.68 Å are reached at 750  $^{\circ}$ C.

The superconducting properties, examined by a four-probe resistance measurement, improve with increasing substrate temperature up to  $750 \ ^{0}C$  (see Fig. 5). Our best films with a thickness of 600 nm, obtained at  $750 \ ^{0}C$ , are



Fig. 6. Dependence of  $T_c$  from the substrate temperature.

characterized by a  $T_c$  of 89 K (midpoint), a transition width of about 3 K, a resistance ratio R(300 K) / R(100 K) of 2.1 and a critical current density of about 10<sup>4</sup> A/cm<sup>2</sup> at 10 K.

#### 4. Summary

Summarizing the results discussed above, highly textured YBCO thin films can be prepared directly on oxidized Zr foils. The c-texture of the films strongly depends on the surface roughness of the foils and can be improved by polishing. The rolling texture of the Zr foils leads to a partial a-b alignment of the YBCO grains. The textures of Zr and YBCO are strongly correlated and mediated by the  $ZrO_2$  (monoclinic) layer. Similar correlations can also be expected for other rolled foils as was shown in the case of Ni [10].

All results about the structure, texture and the superconducting properties, also for different oxygen partial pressures, are summarized in the oxygen partial pressure vs. substrate temperature p(T) diagram (Fig. 7). The films grow com-



Fig. 7. Oxygen partial pressure vs. temperature p(T) diagram. The area of completely c-textured growth is surrounded. Lattice parameters c of less than 11.70 Å are reached in the hatched area,  $T_c$  values of more than 87 K (midpoint) are observed in the area hatched densely.

pletely c-textured in an expanded range near to the stability limit of the 123-phase. In contrast to this, on Si substrates with amorphous SiO<sub>2</sub> top layer a c-textured growth is only obtained along a special line in the p(T) diagram [5]. This indicates that the rolling texture of Zr foils not only leads to the partial a/b-orientation of the films, but also advances the c-textured growth. Only in a smaller part of the area of c-textured growth, lattice parameters c of less than 11.70 Å are obtained. Within a even smaller area high  $T_c$ values of more than 87 K (midpoint) are observed.

Under optimum growth conditions, a substrate temperature of 750  $^{0}$ C and an oxygen partial pressure of 0.3 mbar, a pronounced texture and a structure with a lattice parameter c of about 11.68 Å is reached. This leads to a superconducting transition temperature T<sub>c</sub> of 89 K (midpoint). Nevertheless, the critical current densities are reduced due to the granularity of the films. In order to improve the critical currents, further experiments are in progress.

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