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# Laser ablated YBCO thin films - relations between structural and electrical properties

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**Abstract**

YBCO thin films were prepared by laser ablation with a KrF-excimer laser (248 nm) on (100) $SrTiO_3$-substrates. The aim of this work was to get smooth YBCO films with good superconducting properties for use in single-layer or multilayer Josephson elements. To study the relations between electrical and structural properties the influence of ablation parameters like oxygen partial pressure ($p_{O2}$), substrate temperature ($T_S$) and target to substrate distance (d) were investigated. To analyse the films X-ray diffraction, Rutherford backscattering (RBS) and Scanning electron microscopy (SEM) were used.We found a strong reproducible correlation between the position of the samples in the $p_{O2}$-$T_S$-diagram and their structural and electrical properties like c-axis orientation (FWHM of best samples < 0,15°), c-axis length, outgrowth density, critical temperature ($T_{Coff}$ for R=0) and current density at 77 K. The surface morphology strong correlates to the investigated parameters. In a parameter region of $p_{O2}$ ~ 70 Pa, $T_S$ ~ 720°C and d=4 cm we found outgrowth free smooth films with good superconducting properties. The results on single films were used to produce multilayer systems of YBCO-$SrTiO_3$-YBCO on (100) $SrTiO_3$-substrates.

## 1. Introduction

During the last years the pulsed laser deposition of high-$T_c$ thin films has become a well established technology. Considering the host of parameters influencing the PLD process like laser wavelength, pulse duration, energy density at the target, spot size, pulse repetition rate, target-to-substrate distance, oxygen partial pressure and substrate temperature there are many contributions dealing with the optimization of some of these parameters reviewed e.g. in [1].

The aim of this work was to investigate the correlation between typical ablation parameters like substrate temperature $T_S$, oxygen partial pressure $p_{O2}$ and the target-to-substrate distance d on one side and the electrical and structural properties of the films on the other side. For some laser ablated films outgrowths or hollows are known. In this paper we give a field of parameters corresponding to the surface morphology as well as to the electrical and structural properties of the YBCO films thus contributing to a better understanding of the growth mechanism of these films.

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## 2. Experimental details

To prepare YBCO thin films we use a KrF-excimer laser (248 nm, Lambda Physics 305i). The laser beam with a repetition rate of 5 Hz was directed into a special built vacuum chamber and focused to a spot size of 1,5•4 mm² using a quartz lense. The $SrTiO_3$-substrates were mounted on a stainless steel heater block using silver paint for good thermal contact. The substrate temperature is stabilized within a range of of 1-2 K during ablation time and within a range of 4 K over the 5•10 mm² substrate surface.

The vacuum chamber basic pressure is in the $10^{-4}$Pa range. Before starting the ablation process the chamber was filled with oxygen to the ablation partial value $p_{O2}$. After deposition the chamber was filled with oxygen to atmospheric pressure and the films were cooled down to room temperature with a rate of 50 K/min without any intermediate stop.

To find optimal conditions for the ablation process to get smooth YBCO films with good superconducting properties and to understand the film growth process the following parameters were used: a laser beam energy of 800 mJ and an energy density on the target of 4,3 J/cm². The substrate temperature was varied from 640°C to 760°C, the $O_2$ partial pressure was varied from 10 Pa to 150 Pa. To establish the

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influence of the geometry two target-to-substrate distances of 4 cm and 6 cm were used.
Electrical characterization of the films was done by four probe technique. To measure the current density, bridges of 6 μm length and 3-6 μm width were patterned by direct ion beam etching using a photoresist mask, $Ar^+$ ions with an energy of 500 eV and an ion current density of 1 mA/cm$^2$. To avoid film degradation during the milling process the samples were cooled with liquid nitrogen.
To characterize the surface morphology series of SEM pictures were taken. X-ray diffraction investigations and RBS analysis gave evidence on the crystal structure.

## 3. Results and discussion

Electrical measurements of the samples gave for the best ones a $T_{Coff}$ > 90 K with a transition width of 0,5 K and a current density $j_C(77\ K) \geq 3 \cdot 10^6 A/cm^2$. These values correspond to data from literature. Many authors demonstrated the possibility to grow high quality YBCO films on different substrates [2,3]. Figures 1a and 1b show the influences of the oxygen partial pressure $p_{O2}$ and the substrate temperature $T_S$ on the $T_{Coff}$ values for films made at target-to-substrate distances of 4 cm and 6 cm respectively. For both geometries we find the highest $T_{Coff}$ for partial pressures of about 70 Pa and substrate temperatures of 720°C. The valleys in the plot for d=6 cm are reproducible experimental fact. They may be related to the temperature dependent misfit between the film and the substrate, but there is no clear explanation up to now.
Further investigations have shown, that the region of highest $T_{Coff}$ values does not inevitably correspond to the best morphological and structural properties.
Series of SEM pictures taken from all samples in these $p_{O2}$-$T_S$-field have been presented in a previous paper [4]. For both geometries we found similar surface morphology, clearly depending on the $O_2$ partial pressures used and the substrate temperatures. However for the different d-values the surface roughnesses were similar for different positions in the $p_{O2}$-$T_S$-diagram. A common tendency is outgrowth in all temperature regions for high $O_2$ partial pressures. For low partial pressures the surfaces show hollows which seem to be of regular geometrical form for higher substrate temperatures. For well defined $p_{O2}$-$T_S$-values we found a region of smooth film growth without outgrowth and hollows. Comparing the data for the two distances, we found the best surface morphology for d=6 cm at an

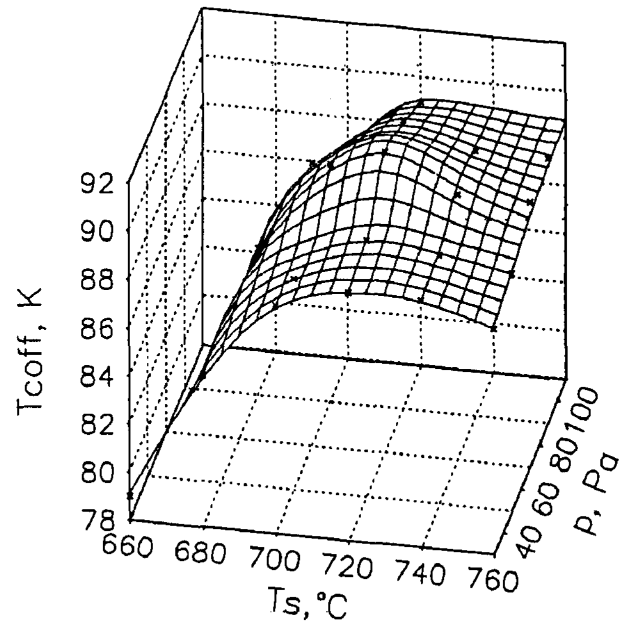


Fig. 1b $T_{Coff}$ as a function of $p_{O2}$ and $T_S$ for a target-to-substrat distance d=4 cm

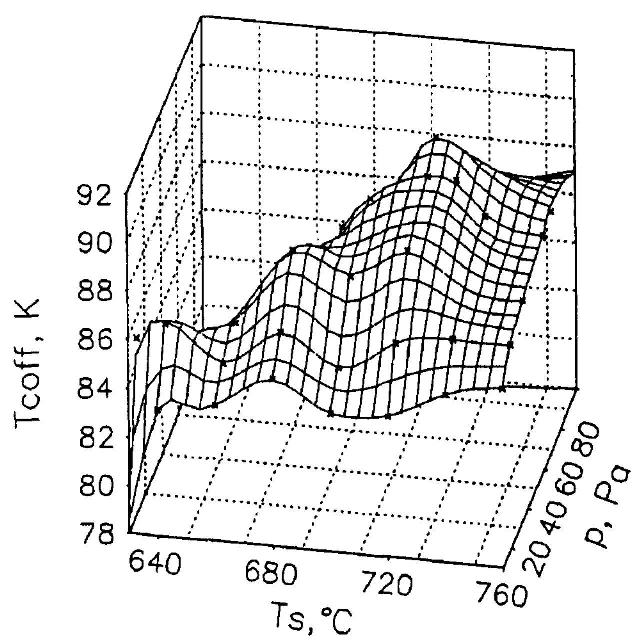


Fig.1a $T_{Coff}$ as a function of $p_{O2}$ and $T_S$ for a target-to-substrat distance d=6 cm

$O_2$ partial pressure of about 30 Pa and a substrate temperature of about 720°C. For the 4 cm target-to-substrate distance the region of smooth surfaces shifts to higher $p_{O2}$ values. We found the smoothest films in the same temperature region but at about 70 Pa. This means that for d=4 cm the parameters to get outgrowth free films suitable for multilayer applications correspond to the highest $T_{Coff}$ values. Fig. 2 shows the influence of the variation of the

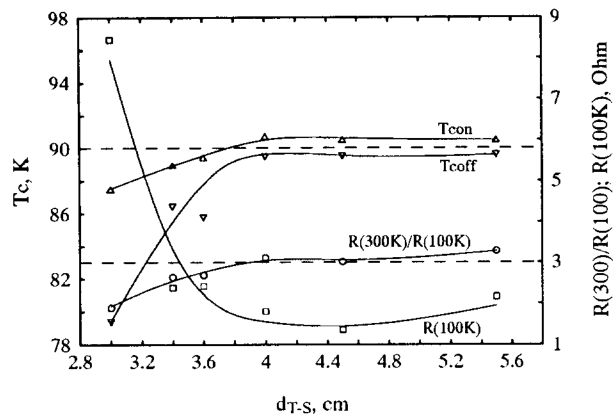


Fig. 2 Ablation of YBCO-films at different target-to-substrate distances

target-to-substrate distance for films made at the optimized ablation parameters of $T_S$=720°C and $p_{O2}$=70 Pa on the critical temperature, the film resistance at 100 K and the R(300K)/R(100K) ratio. We got the best parameters for distances greater than 4 cm. For smaller distances the critical temperatures decrease, the transition widths to the superconducting state become broader and the resistances change to higher values.

To clarify the connection of the paramaters discussed above to the crystalline structure of the films, X-ray and RBS measurements were made. Fig. 3a and 3b show the comparision of the $\chi_{min}$ from RBS channeling experiments for films, made at three different substrate temperatures, varying the oxygen partial pressures to the data obtained from X-ray measurements (FWHM and c-axis length) and the critical current densities.

For each geometry the $\chi_{min}$ values and the FWHM-values are in good correspondence, the lowest

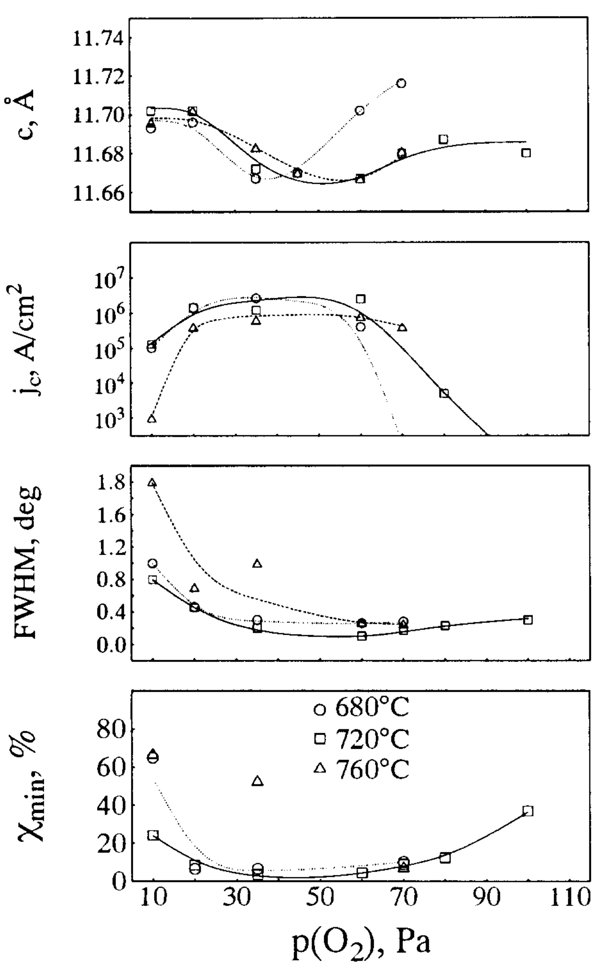


Fig. 3a $\chi_{min}$, FWHM, c-axis length and $j_c$ for d=6 cm

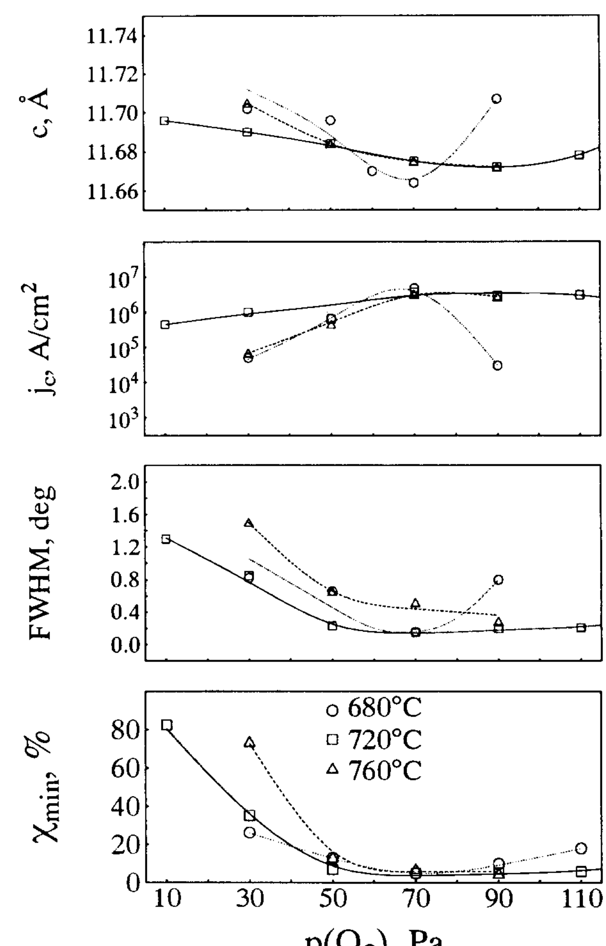


Fig. 3b $\chi_{min}$,FWHM, c-axis length and $j_c$ for d=4 cm

$\chi_{min}$ also give the lowest FWHM, whereby the best values are found for the optimal substrate temperature of 720°C. The critical current densities at 77 K are the highest for the samples having the shortest c-axis lengths. As these values also correspond to the X-ray and RBS-data, we conclude that the current densities are in the first place a result of the structural properties of the films.

Comparing now the curves for the two different geometries, we find a correspondence to the results concerning the surface morphologies discussed in the first part of this paper.

For the 6 cm distance we see the optimal values of the properties shown in Fig. 3 for samples made at oxygen partial pressures of about 35 Pa and thus for samples made at positions in the $p_{O2}$-$T_S$-diagram giving the smoothest outgrowth and hollow free surfaces. But as we also have seen in the first part for the 6 cm distance these smooth films have lower critical temperatures.

From the view point of the user it is necessary to combine the high current densities of the YBCO-films following from their structural data with high critical temperatures and narrow transition widths to the superconducting state.

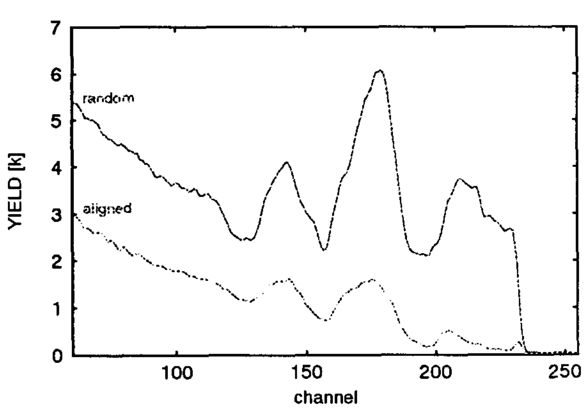


Fig. 4 RBS channeling for YBCO-$SrTiO_3$-YBCO on (100) $SrTiO_3$-substrates

That this is possible we see from the results of the samples made at a target-to-substrate distance of 4 cm. There the films having the highest $j_c$ values and the best X-ray and RBS data also have the highest critical temperatures. This means that by changing the geometry in the chamber the shift in the growth conditions enables us to find an optimal combination between the electrical and structural properties of the films to use them for Josephson contacts [5].

Further investigations were made to produce multilayer systems of YBCO - $SrTiO_3$ - YBCO on $SrTiO_3$-substrates. For ablating the YBCO the optimized parameters from this work were used. The insulating $SrTiO_3$ layers were made without interrupting the vacuum process at $T_S$=660°C and $P_{O2}$=30 Pa, whereby the growth parameters for the $SrTiO_3$ are not criticall. The growth of all three layers we monitor with the RBS channeling data shown in Fig. 4. The $\chi_{min}$ for this trilayer system, containing a 115 nm thick YBCO bottom layer, a 115 nm thick $SrTiO_3$ insulator and a 115 nm thick YBCO top layer, is 3,8%, a value corresponding to a very good alignment of the crystal axis of the samples. Also for a 530 nm thick $SrTiO_3$ layer we find a $\chi_{min}$=14% what means a quite good crystal orientation of the trilayer. The electrical parameters of the top- and the bottom layer are satisfying, both show critical temperatures up to 90 K. The trilayer systems show smooth surfaces and should be suitable for integrated Josephson junctions. Investigations on patterning devices by ion beam etching and on the insulating behaviour of the $SrTiO_3$ layer are in progress.

## 4. Conclusions

In this work we have shown the close connection between electrical and structural parameters for laser ablated YBCO thin films. Based on the relationships between the ablation parameters and the properties of the films we have given a field of parameters to combine good electrical properties with smooth film surfaces suitable for multilayer devices.

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