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# Buffer layers of yttria stabilized zirconia for $YBa_2Cu_3O_7$ devices on silicon substrates

F. Goerke and M. Schilling

Universität Hamburg, Institut für Angewandte Physik, Jungiusstraße 11, D-2000 Hamburg 36, F.R.G.

**Abstract**

The preparation of high quality $YBa_2Cu_3O_7$ films on Si(100) is essential for devices on this important substrate material. Interdiffusion, lattice mismatch and different thermal expansion coefficients make the use of buffer layers necessary for the growth of $YBa_2Cu_3O_7$ on Si(100). Yttria stabilized zirconia (YSZ) has been proven to be suitable for this purpose. We investigate the growth of YSZ films on Si(100) substrates by pulsed laser deposition with a KrF excimer laser. The YSZ films are characterized by X-ray scattering, ellipsometry and scanning electron microscopy. On these buffer layers $YBa_2Cu_3O_7$ films are prepared in-situ by laser deposition. While the critical temperatures $T_{c,0}$ are above 85 K, the critical current densities $j_c$ are still degraded compared to the films on $SrTiO_3$ substrates. We report on the dependence of the superconducting and morphological properties of the $YBa_2Cu_3O_7$ films on the deposition parameters.

## 1. Introduction

The laser deposition of very smooth $YBa_2Cu_3O_7$ films on $SrTiO_3$ and MgO substrates is neccessary for the preparation of multilayer devices such as superconducting quantum interference devices (SQUIDs). The growth of almost particle free films was demonstrated with this deposition method.[1] DC-SQUIDs with artificial $PrBa_2Cu_3O_x$ barriers are prepared in a multilayer process.[2] We investigate the properties of these devices in dependence of the film quality. To avoid shorts in the insulator layers, the films have to be smooth with a low density of outgrowths and droplets. The deposition of $YBa_2Cu_3O_7$ films on Si substrates for device applications is of interest for the integration of superconducting and semiconducting devices on the same chip, for superconducting interconnects between semiconductor devices and for novel applications. The growth directly on Si is complicated by interdiffusion, different thermal expansion coefficients and lattice mismatch.[3] So buffer layers have to be deposited before the growth of high quality $YBa_2Cu_3O_7$ films. It was shown that yttria stabilized zirconia (YSZ) is well suited for this purpose.[4] $YBa_2Cu_3O_7$ films on Si with YSZ buffer layers have good superconducting properties. This is achieved with epitaxial YSZ films on Si(100) and epitaxy of $YBa_2Cu_3O_7$ on YSZ as well.[4] Sometimes, the appearance of cracks in the $YBa_2Cu_3O_7$ film is found, if the thickness exceeds 50 nm.[5] Here we study the dependence of the morphology of laser deposited YSZ films and of in-situ deposited $YBa_2Cu_3O_7$ films on the deposition temperatures and on the film thicknesses.

## 2. Experimental set-up

The $YBa_2Cu_3O_7$ targets for the laser ablation process are prepared by standard calcination methods in order to obtain polycrystalline ceramic pellets of densities above 85 % of the theoretical density of a single crystal. The YSZ targets are prepared by careful mixing and grinding of $ZrO_2$ powder with 10 mol% $Y_2O_3$. Afterwards these targets are sintered at 950 °C in air for 8 h for a higher mechanical stability. As will be discussed in the next section, the natural oxide of Si was not removed by wet etching before deposition of the buffer layer. For the ablation a KrF excimer laser LPX 301 iCC (248 nm wavelength, 25 ns pulse length) is used. For both materials an energy density of 2.3 $J/cm^2$ on the target surface is choosen at a pulse frequency of 5 Hz. The targets are not rotated. The ablation of YSZ is performed at an oxygen pressure of $3 \cdot 10^{-4}$ mbar, while $YBa_2Cu_3O_7$ is ablated in 0.4 mbar oxygen atmosphere. During deposition the Si substrates are mounted on a heater. The substrate temperature was measured by a thermocouple tightly clamped to the heater, so that the actual temperature at the surface of the substrate during deposition will be lower. For better reproducability the electrical heating power was calibrated to temperature. These substrate temperatures are given in the text and were varied between 745 °C and 820 °C for the growth of $YBa_2Cu_3O_7$ and between 690 °C and 850 °C for the deposition of YSZ. The distance between substrate and target was 54 mm for all films. At this distance the deposition rate was 0.09 nm/pulse for YSZ and 0.2 nm/pulse for $YBa_2Cu_3O_7$.

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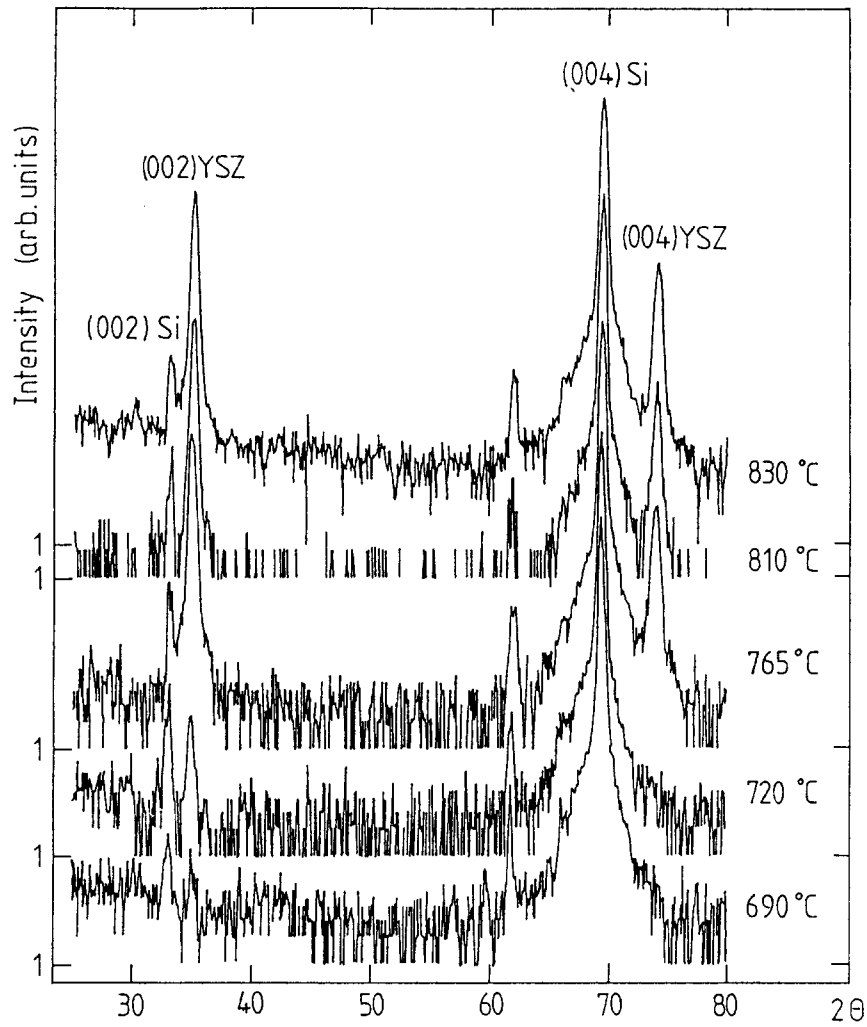


Figure 1: X-ray diffraction on YSZ films on Si(100) substrates. The $\Theta - 2\Theta$ scans were recorded on films deposited at different substrate temperatures. The thickness of all films is 75 nm.

After the in-situ deposition of both layers, the substrates are slowly cooled down to room temperature in 15 minutes at a pressure of 1000 mbar oxygen. This cooling process was optimized for $YBa_2Cu_3O_7$.[6]

## 3. Properties of YSZ films

After deposition the YSZ films are analyzed by ellipsometry and X-ray diffraction. The morphology is investigated by scanning electron microscopy (SEM). In Fig. 1 the $\Theta - 2\Theta$ scans of films deposited at different substrate temperatures are shown. Only (00*l*)-peaks of YSZ and Si are found indicating good orientation. The (00*l*) peaks of YSZ become more intense for higher substrate temperatures. While the (002) peak can be found for all films, the less intense (004) peak is only resolved for substrate temperatures above 765 °C. This indicates that the degree of orientation and epitaxy increases at higher substrate temperature. For a temperature of 810 °C we find optimum conditions for the growth of YSZ on Si(100) with a width of the (004)-peak of 0.7 °. For higher substrate temperatures the X-ray diffraction shows no significant change any more, but in SEM micrographs we find cracks all over the film surface. This may result from too large strain in these films. For the lower substrate temperatures the YSZ films are very smooth, so that no structures could be resolved in the SEM. With ellipsometric measurements the index of refraction n for a wavelength of 633 nm is determined. For films of the same thickness, we find no dependence of the index of refraction on the substrate temperature. But it depends on the thickness of the film. The value for single crystals of

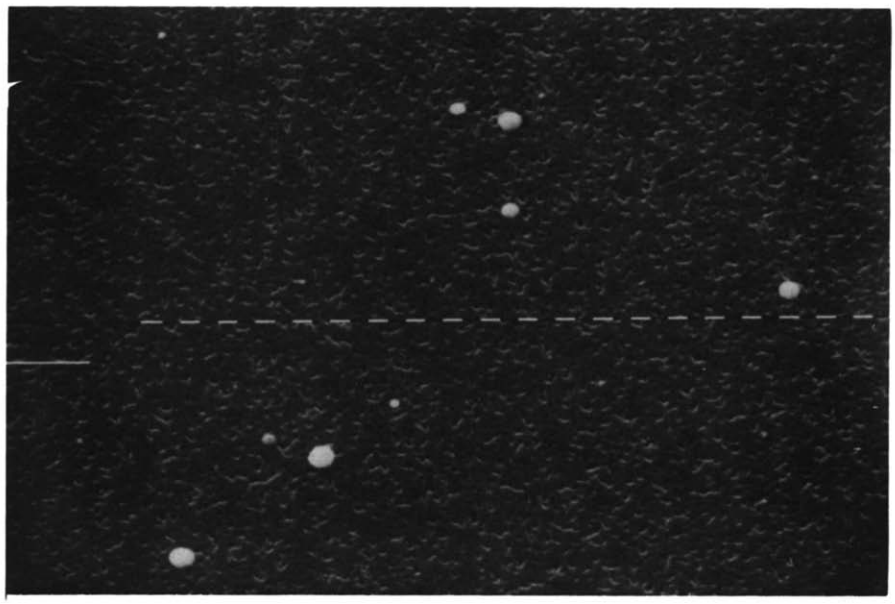

Figure 2: Morphology of a 100 nm thick $YBa_2Cu_3O_7$ film on a 75 nm thick YSZ buffer layer on Si(100). The $YBa_2Cu_3O_7$ film has $T_{c,0}$ = 85.6 K and a resistance ratio R(300 K)/R(100 K) = 3.0. The marker is 1 $\mu$m long.

n = 2.16 [7] we find for a film thickness of 75 nm. For thicker films the index of refraction increases up to n = 2.7 for a film thickness of 320 nm. The interpretation of this increase is complicated by the increasing thickness of the oxide layer between the silicon surface and the YSZ film due to the high mobility of oxygen ions in the YSZ films.[8] The thickness of this oxide layer depends on the time the substrate remains at high temperature in the oxygen atmosphere. Hence, for thicker YSZ films the oxide layer should also be thicker and can influence the ellipsometric measurement. This will be investigated in more detail.

## 4. Properties of $YBa_2Cu_3O_7$ films

For the deposition of $YBa_2Cu_3O_7$ on the YSZ buffer layers on Si(100) we chose the optimum thickness of the YSZ layer of 75 nm at a deposition temperature of 810 °C. The optimization of the $YBa_2Cu_3O_7$ deposition was first made for the substrate temperature with $YBa_2Cu_3O_7$ films of 100 nm thickness. Then, in a second step, the film thickness was increased at the optimum substrate temperature of 765 °C. In Fig. 2, the surface of a typical $YBa_2Cu_3O_7$ film is shown. We find almost no outgrowth particles, but droplets of densities in the $10^5$ $cm^{-2}$ range can be found. The underlying roughness of the film is due to the inherent growth mechanism of $YBa_2Cu_3O_7$ and is attributed to growth spirals.[9] The holes in the film may be due to slight deviations in the Cu content of $YBa_2Cu_3O_7$.[10] In the SEM we find no cracks in any of these films for thicknesses up to 300 nm after one temperature cycle to T = 30 K. The observation that cracks appear after a few days could not be verified.[8] For films of 200 nm we found no cracks after 4 days. This investigation will be extended to longer times and more temperature cycles. The electrical characterization is made by resistance measurements in dependence on the temperature. These data are depicted in Fig. 3. We find that the critical temperature $T_{c,0}$ is always around 85 K for substrate temperatures in the range between 745 °C and 790 °C. For higher substrate temperatures, the films become semiconducting most probably because of strong diffusion of Si into the superconductor. The highest resistance ratio of R(300K) / R(100K) = 3.1 we find for a substrate temperature of 790 °C. Towards lower substrate temperatures the ratio decreases, indicating a deterioration in the epitaxial orientation. For increased $YBa_2Cu_3O_7$ film thickness an increase of the resistance ratio is observed, reaching a ratio of 3.6 for a thickness of 300 nm. A possible reason for this is, that the mismatch of lattice constants in the bilayer causes strain in the $YBa_2Cu_3O_7$ film. These strained films may have different characteristics, as was also observed by Fork et al..[4] The highest critical temperature $T_{c,0}$ = 86.7 K was observed for a 150 nm thick film which has a resistivity of 280 $\mu\Omega$cm at 100 K. For increasing film thickness almost no variation of the critical temperature, but an increase of the resistivity is found, yielding for a 300 nm thick $YBa_2Cu_3O_7$ film a critical temperature of $T_{c,0}$ = 85.7 K and a resistivity of 570 $\mu\Omega$cm at 100 K.

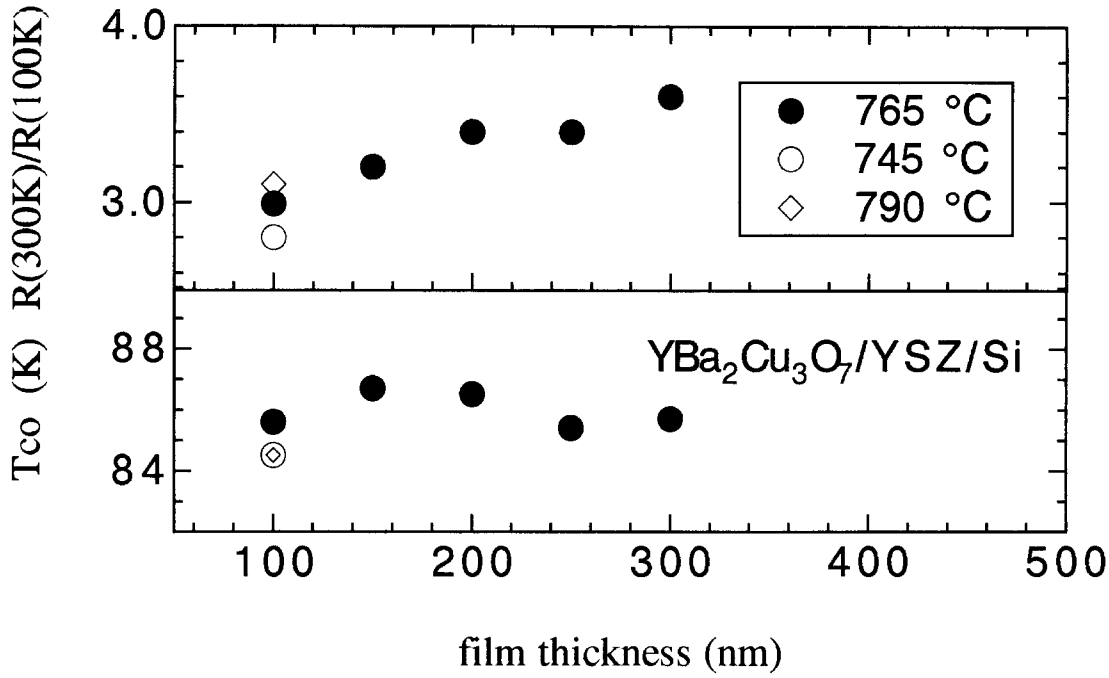


Figure 3: Superconducting properties of $YBa_2Cu_3O_7$ films on 75 nm YSZ buffer layers on Si(100). The dependence on the thickness of the $YBa_2Cu_3O_7$ film is shown. The variation with substrate temperature is also given for a fixed film thickness of 100 nm.

## 5. Conclusions

$YBa_2Cu_3O_7$ films with good superconducting properties can be prepared on Si(100) substrates with YSZ buffer layers by in situ laser deposition. We obtain critical temperatures up to $T_{c,0} = 86.7$ K and critical current densities $j_c(60K) \geq 10^4$ A/cm$^2$. An enhancement of the degree of epitaxy should be possible by removing the oxide layer on the Si substrate before the buffer layer is deposited. The analysis of the film properties will be extended to Raman measurements. The films will be further optimized with respect to the higher critical current densities required for applications such as DC-SQUIDs.

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