

## Grain misorientation and properties of HTSC thin films

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### Abstract

The transport properties of high-temperature superconductor (HTSC) thin films are closely related to the crystallographic growth quality. Three examples are presented for such relationships. In YBaCuO films on SrTiO<sub>3</sub> the critical current density decreases exponentially with increasing mosaic spread. For BiSrCaCuO-films (2212-phase) it is shown that the superior in-plane growth quality on SrTiO<sub>3</sub> results in higher  $j_c$ -values. ( $7 \cdot 10^5$  A/cm<sup>2</sup>) as compared to those on MgO ( $6 \cdot 10^4$  A/cm<sup>2</sup>). In films of different growth orientation (e.g. a-axis films) the specially needed deposition conditions also influence the properties.

### 1. Introduction

A fast characterization of the growth quality of HTSC thin films is an important prerequisite in an effective optimization process of the deposition parameters. If unique relationships between growth quality and transport properties are established, elaborate and time consuming preparations like, e.g., a structuring of the films for transport measurements can be avoided if the growth quality does not meet the requirements. X-ray diffraction and channeling experiments combined with Rutherford backscattering (RBS) are suitable methods for epitaxial growth characterization. Mosaic spreads obtained from rocking curves, the in-plane distribution of grains deduced from  $\phi$ -scans, or the minimum yield value,  $\chi_{\min}$ , in channeling measurements are parameters which can be combined with transport properties. Moreover, the mass selectivity of RBS allows the investigation of separate sublattices like, e.g., defect structures in the oxygen sublattice. In the following, three examples of growth quality and pro-

perties in HTSC thin films are reported.

### 2. Examples

A narrow *mosaic spread*,  $\Delta$ , as a necessary condition for high critical current densities,  $j_c$ , has been postulated before [1]. Here a quantitative relationship between  $\Delta$  and  $j_c$  is presented for YBaCuO films deposited onto SrTiO<sub>3</sub> substrates by laser ablation. The application of polycrystalline substrates with a high degree of [100] texture allowed the preparation of films with grain boundaries of up to 8° without degrading  $T_c$  (~90 K). The mosaic distributions of a film and the substrate are shown in Fig. 1 for two cross sections of the incident x-ray beam as an example. The distribution of grains in the film results as an envelope of the sharp slightly misaligned distribution of the grains in the substrate. Bridges over different  $\Delta$  regions have been structured on the films. An assignment of  $j_c$ -values measured on these bridges to the corresponding  $\Delta$  values yields an exponential decrease of  $j_c$  with increasing  $\Delta$ . This relationship is demonstrated in Fig. 2. Included are recalculated points of previous  $j_c$  measure-

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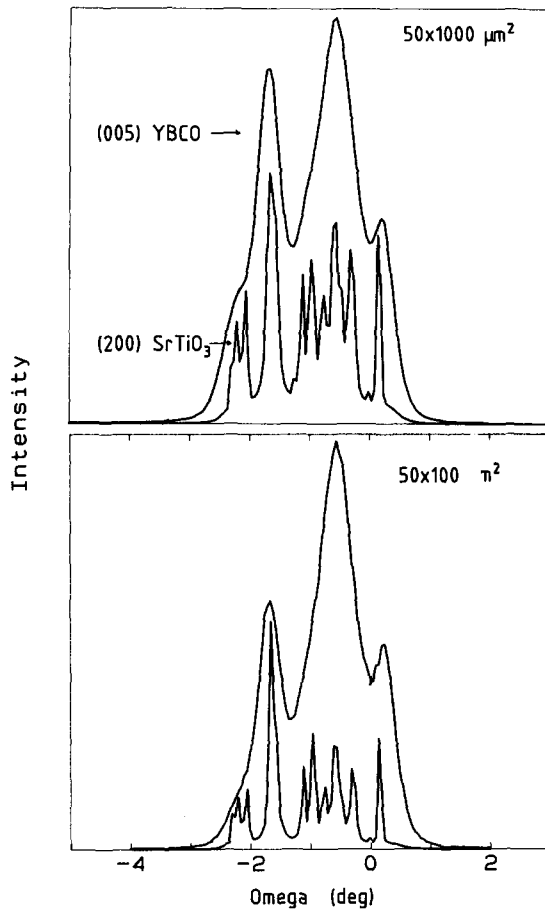


Figure 1.  $\omega$ -scans of YBaCuO films [(005)-line] on polycrystalline highly textured SrTiO<sub>3</sub> substrates [(200)-line] with two different cross sections of the incident x-ray beam.

ments over single artificial grain boundaries [2] which roughly agree with our findings. Further details of the measurements have been reported before [3].

In addition to the breadth of mosaic distributions also the *in-plane orientation* of grains in a film considerably influences the properties. Such orientations can be examined by TEM measurements or non-destructively by  $\phi$ -scans in x-ray experiments. In the latter measurements signals are detected from lattice planes inclined to the sample surface as a function of the rotation angle  $\phi$  through the surface normal. The intensity distribution of such scans in addition to the symmetry around the rotation axis reflects the orientation

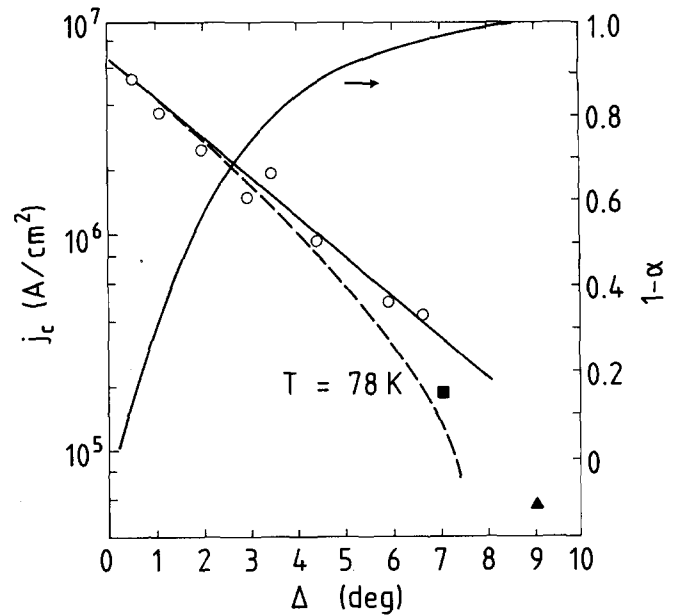


Figure 2. Critical current density,  $j_c$ , vs mosaic spread,  $\Delta$ , for YBaCuO films on SrTiO<sub>3</sub>.  $1-\alpha$  (right scale) yields the part of the current through weak links.

and abundance of grains in the film plane. As an example for the in-plane growth we discuss results for BiSrCaCuO films (2212 phase) which were prepared by sputtering employing the inverted cylindrical magnetron [4] and subsequent heating. The films were deposited on (100)-oriented SrTiO<sub>3</sub> and MgO substrates. In Fig. 3  $\phi$ -scans are shown of the substrate and a film on SrTiO<sub>3</sub>. The scan of the film reveals only four maxima corresponding to the symmetry of the rotation axis indicating a complete alignment in the film plane. The maxima of the film are shifted by 45° with respect to those of the substrate. This means that lattice matching between film and substrate is achieved via the diagonal of the (100) plane in the substrate. A similar rotation of the film unit cell with respect to the substrate axes has been detected for YBaCuO films on Zr(Y)O<sub>2</sub> substrates by channeling measurements [5]. Corresponding  $\phi$ -scans for BiSrCaCuO films on MgO are displayed in Fig. 4. In addition to [100] BSCCO || [100] MgO orientations here also grains of the film aligned [100] || [110] and [100] || [230]

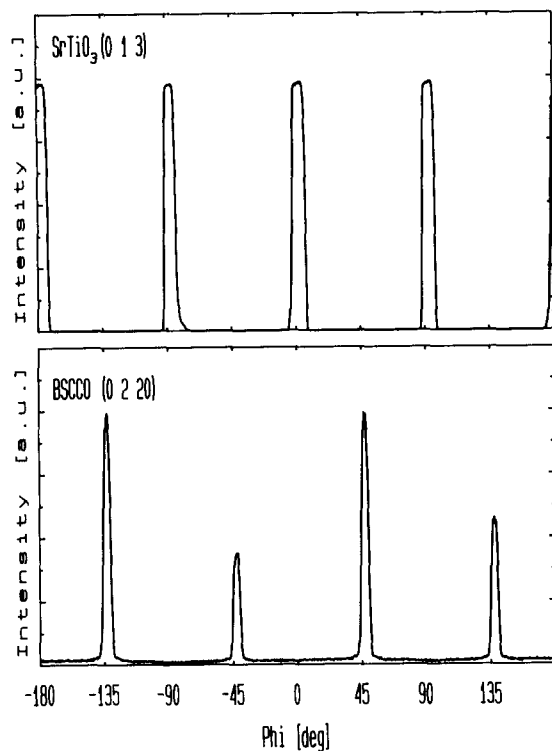


Figure 3.  $\phi$ -scans from film and substrate for BiSrCaCuO on SrTiO<sub>3</sub>. Four peaks in the diagram of the film correspond to the symmetry of the rotation axis and indicate epitaxy in the film plane.

with respect to MgO are observed. This results in additional grain boundaries which influence the transport properties. The superconducting transition temperatures  $T_{c0}$  (resistively / inductively) are 80/76 K on MgO and 88/82 K on SrTiO<sub>3</sub>. A striking difference is observed for  $j_c$  (4.2 K) with values more than one order of magnitude larger on SrTiO<sub>3</sub> ( $7 \cdot 10^5$  A/cm<sup>2</sup> against  $6 \cdot 10^4$  A/cm<sup>2</sup> on MgO). Surprisingly the mosaic spreads on MgO ( $0.3^\circ$ ) are narrower than on SrTiO<sub>3</sub> ( $0.5^\circ$ ). This underlines the importance of in-plane alignment for the transport properties.

The properties of films with specific growth directions different from c-axis orientation are influenced due to the anisotropy of the "123" structure and by the special deposition conditions for the specific growth achievement. E.g., in *a*-axis films, which in comparison to c-axis films grow at lower substrate tempera-

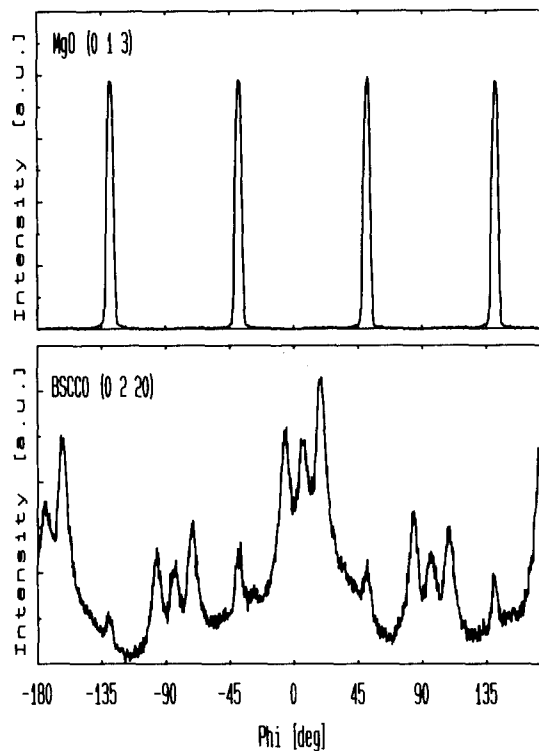


Figure 4:  $\phi$ -scans from film and substrate for BiSrCaCuO on MgO. Various orientations of grains in the film plane produce additional grain boundaries which influence the properties.

tures one observes higher resistivities, temperature activated conductivity and depressed  $T_c$  values. Though a considerable property improvement is obtained in a template deposition process (metallic conductivity behaviour,  $T_c = 84$  K) [6], they are still degraded in comparison to perfect c-axis films. The high resistivity ( $\sim 800 \mu\Omega\text{cm}$ ) results from the anisotropy of the structure and the in-plane orientation of the grains - two perpendicular directions of the c-axis in the film plane are equally probable on cubic substrates. The lower  $T_c$  values, however, are not yet understood. In preliminary channeling investigations of the oxygen sublattice employing the  $^{16}\text{O}(\alpha,\alpha)^{16}\text{O}$  resonance at an energy of 3.05 MeV of the incident particles in c- and a-axis oriented films we observed larger  $\chi_{\text{min}}$  values, i.e., more disorder in a-axis films. In films of both orientations, however, i.e. also in c-axis films,

the  $\chi_{\min}$  values were appreciably larger than expected from theoretical values obtained by Monte-Carlo simulation calculations. This indicates that the  $T_c$  values ( $>90$  K in c-axis films) are not directly related to disorder in the oxygen sublattice as integrally described by  $\chi_{\min}$ . Moreover, in recently prepared a-axis films of high growth quality,  $\chi_{\min}$  in the oxygen sublattice measured at 80 K was only 12%, i.e. better than the value of about 15% determined in best c-axis films. Random and aligned backscattering spectra of the high quality a-axis films are displayed in Fig. 5 as an example.  $\chi_{\min}$  in

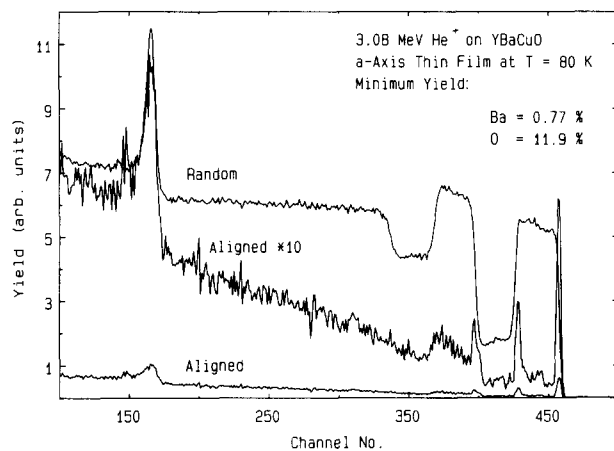


Figure 5: Random and [100] aligned backscattering spectra of a high quality YBaCuO/SrTiO<sub>3</sub> a-axis film measured at 80 K with 3.08 MeV He<sup>+</sup> ions. The <sup>16</sup>O( $\alpha,\alpha$ )<sup>16</sup>O resonance allows a reliable detection of the oxygen in the film. The aligned spectrum is also shown in 10 fold magnification.

the Ba sublattice is 0.8%. Very narrow rocking curves with  $\Delta < 0.08^\circ$  are in agreement with the good channeling data. Nevertheless, the  $T_c$  values of these a-axis films are still degraded. Monte-Carlo simulations for a perfect structure in a-axis direction at 80 K yield  $\chi_{\min}$  values of about 7%, i.e. considerably lower than our observations. We therefore speculate that degraded  $T_c$  values still may be due to dis-

order in the oxygen sublattice with a special arrangement on lattice positions (superstructures) which, e.g., have been observed in irradiated films with depressed  $T_c$  values [7]. Such defect structures, however, need further clarification.

### 3. Conclusion

The examples presented show that some properties of HTSC thin films like  $j_c$  can be related to the growth morphology like the in or out of plane distribution of grains measured in simple  $\phi$ - or  $\omega$ -scans. More elaborate investigations are required if properties like  $T_c$  in a-axis oriented films are influenced by structural distortions on an atomic scale, e.g., lattice disorder in the oxygen sublattice.

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