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C-Axis Oriented Growth of Magnetron Sputtered YBaCuO Thin Films on MgO Substrates

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Abstract. Highly textured YBaCuO thin films were sputtered on MgO (100)-oriented single crystal substrates at ambient temperature followed by an anneal in oxygen for 1 h at temperatures up to 920°C. X-ray diffractograms of the highly textured films indicate an orientation of the c-axis of the YBaCuO lattice perpendicular to the substrate surface. There are strong indications that the oriented c-axis growth is due to a CuO self-flux effect. Auger measurements reveal a copper diffusion profile into the substrate down to a depth of more than 400 nm.

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After the discovery of high-T_c superconductivity in LaBaCuO by Bednorz and Müller [1] and with the subsequent development of the ceramic superconductor YBaCuO for operation above liquid nitrogen temperature by Chu et al. [2], an immense initiative was begun, particularly in the field of high-T_c superconducting thin films.

In order to apply these thin films to e.g. superconducting electronics [3,4], the electrical film properties have to be optimized with regard to a high transition temperature as well as a high critical current. Furthermore, the lithographic treatment and multilayer techniques require flat surfaces and simple structuring possibilities without degradation of the film properties.

Here we report on the properties of YBaCuO thin films, sputtered on MgO substrates at ambient temperature with a subsequent high-temperature treatment in pure oxygen. These films were investigated with X-ray diffractometry (XRD), scanning Auger electron microscopy (SAM) and scanning electron microscopy (SEM) in order to obtain information about the growth mechanism on MgO substrates.

1. Preparation and Measurements

The YBaCuO thin films were deposited by rf magnetron sputtering from a stoichiometric sinter target in a turbo-pumped vacuum chamber. The background pressure before sputtering was better than 10^{-3} Pa. The sinter target was prepared from the oxides using a standard technique described elsewhere [5]. The target was mounted on a water-cooled cathode, providing a good thermal contact. This was found to be important in order to minimize the temperature increase of the substrate during the sputtering process by heat irradiation, thus enabling a structuring of the thin films using a lift-off technique with conventional photoresist. The films were deposited at a typical rf power level of 150 W at a frequency of 27 MHz and a resulting dc self-bias voltage of 60 - 90 V. The argon pressure was between 0.2 and 5 Pa and the deposition rate 0.5 - 2 Å/s.

(100)-oriented MgO single crystals were used as substrates. They were kept at ambient temperature during deposition. Finally, the films were loaded with oxygen in a furnace at 1 bar oxygen pressure. Since all films immediately annealed at 920°C show a polycrystalline structure, a prior treatment at lower temperatures was found to be necessary for highly oriented growth. The temperature was therefore increased from room temperature to 650°C over a period of 1 h. It was kept at 650°C for 2 h and then increased to 750°C over 30 min. After annealing at this temperature for 1 h, the temperature was decreased to room temperature at a rate of about 150°C/h. The films were stored in a dry atmosphere for several days and then finally annealed at 920°C [5].

The X-ray diffractograms were recorded in a commercially available diffractometer using a Cu K_α source. For the analysis of the surface region we used a PHI 600 SAM (scanning Auger microscopy) system with facilities for scanning electron microscopy (SEM), the lateral resolution achieved being 19 nm, and Ar^+ depth profiling.

2. Results and Discussion

Scanning Auger measurements indicate that the film's toplayer (light grey regions in Fig.1) is Cu deficient, the Y:Ba:Cu ratio being about 1:2:1 [6]. Furthermore, the MgO substrate (dark grey) is covered with a significant amount of Cu. A linescan on the edge of the sample fractured under atmospheric conditions provides us with information about the penetration depth of Cu into the MgO substrate. Figure 2 shows the SEM image of this freshly prepared edge. The linescan has been recorded on the line between the two black arrows: The toplayer is 400 nm thick (left-hand side of the edge) as confirmed by additional depth profiles [6] and the Cu linescan. The Cu linescan was recorded from the top layer (0μm) down to 1.6 μm (Fig.3). The following experimental conditions were used: primary beam energy: 15 keV, primary beam current: 0.25 nA, 160 points/line, analysis time per point (τ)=12s, energy resolution of the cylindrical mirror analyser of 1.2% and additional point analysis down to 1 μm using τ = 40 s. Under these conditions the detection limit for Cu<0.5% (atomic con-

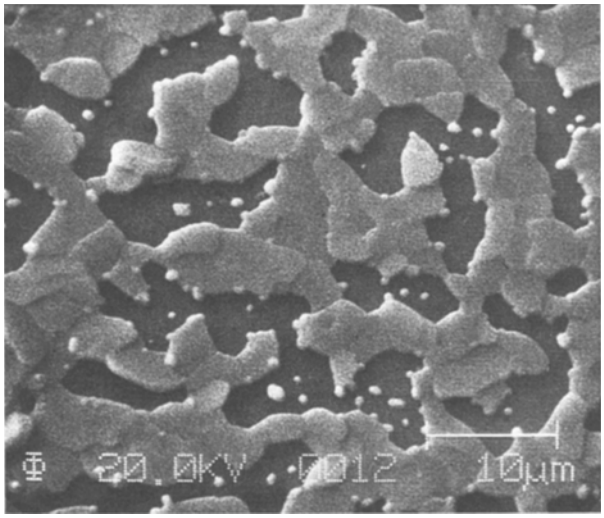

Fig.1. SEM picture (magnification 2000x) of a percolative YBaCuO thin film

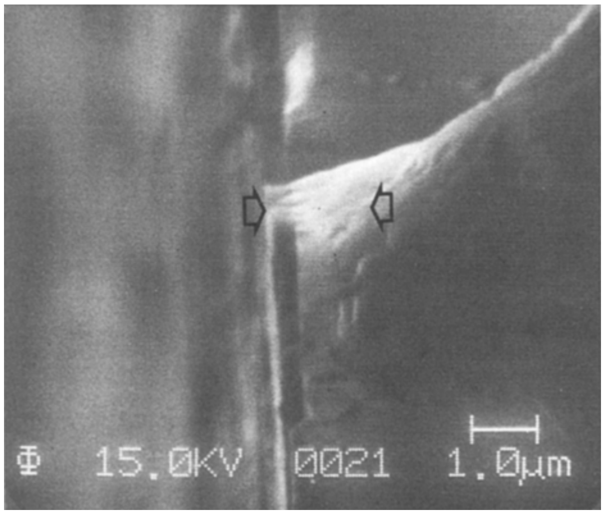

Fig.2. SEM picture (magnification 10000x) of a fractured MgO substrate with a YBaCuO film on top

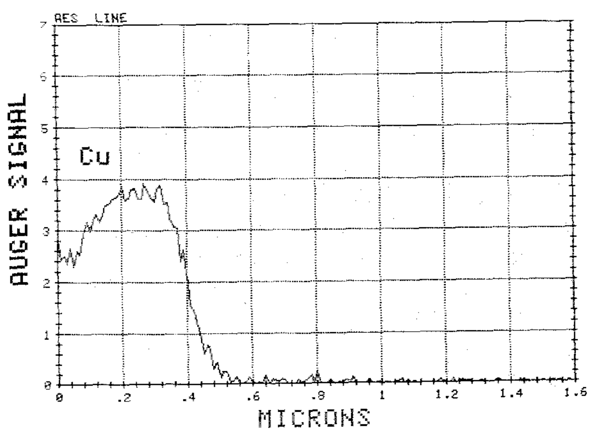

Fig.3. Auger Cu linescan, recorded between the two black arrows in Fig.2

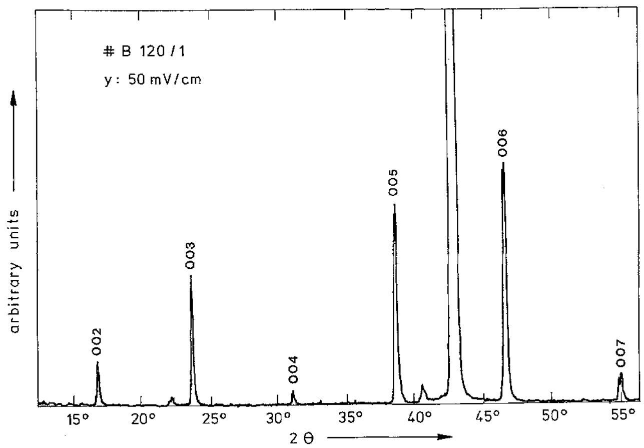

Fig.4. X-ray diffractogram of the percolative YBaCuO thin film of Fig.1, exhibiting c-axis oriented growth

centration) [7] and the lateral resolution for Auger analysis is between 50 and 100 nm. From the linescan it has been determined that the depth of the penetration of Cu extends 400 nm below the Cu-Ba-Cu-oxide/MgO interface and the bare MgO substrate. Additional evidence from Auger depth profiles recorded for various points at the sample surface will be published in a forthcoming article [6] and reveal a homogeneously diffused Cu layer. The linescan in Fig.3 also shows Cu to be homogeneously distributed in the top layer. The drop in Cu signal intensity around 0 μm is due to an edge effect [8] (Fig.2).

After the deposition at room temperature, all YBaCuO films are amorphous, as shown from XRD measurements. During the anneal YBaCuO will start to grow from nucleation centers. Metallic Mg plays an important role as nucleus for the crystallization in the subsequent annealing procedure [9]. Due to the high surface mobility caused by CuO diffusion in the substrate and homogeneous CuO coverage of the substrate [6], the film may even show percolation-type growth, as shown in Fig.1. This film exhibits a $T_{c,end}$ of 62 K. Growth of large single crystalline grains on a MgO substrate is visible. The X-ray diffractogram of this film reveals c-axis oriented growth (Fig.4). This orientation can be achieved in the case of MgO; However, epitaxial growth of the YBaCuO film (a_0 = 3.9Å) on the MgO substrate (a_0 = 4.2Å) is impossible.

In conclusion, we have shown that non-epitaxial c-axis oriented growth of cold sputtered YBaCuO films on MgO substrates can be achieved in a subsequent annealing step at 920°C, in which CuO diffusion into the MgO is a crucial factor. This phenomenon is similar to the self-flux growth of large single crystals of YBaCuO in CuO [10]; however, the anneal temperature is lower, i.e., less than or equal to 920°C.

References

1. J.G. Bednorz, K.A. Müller: Z. Phys. B64, 189 (1986)
2. M.K. Wu, J. Ashburn, C.J. Torng, P.H. Hor, R.L. Meng, L. Gao, Z.J. Huang, Y.Q. Wang, C.W. Chu: Phys. Rev. Lett. **58**, 908 (1987)
3. B. Häuser, M. Diegel, H. Rogalla: Appl. Phys. Lett. 52, 844 (1988)
4. R.H. Koch, C.P. Umbach, G.J. Clark, P. Chaudhari, R.B. Laibowitz: Appl. Phys. Lett. **51**, 200 (1987)
5. B. Häuser, H. Rogalla: In *Novel Superconductivity*, ed. by S.A. Wolf, V.Z. Kresin (Plenum, New York 1987), p.951
6. B. Häuser, E.G. Keim, H. Rogalla, A. van Silfhout (to be published)
7. Detectability limits for Auger analysis in the PHI. Techn. Bulletin no. T8401 (5/5/84), Physical Electronics Industries, a division of the Perkin-Elmer Corp.
8. M.M. El Gombati, M. Prutton, B. Lamb, C.C. Tuppen: Surf. Interface Anal. **11**, 251 (1988)
9. J. Halbritter, H.-J. Hedbabny, H. Rogalla: Mater. Lett. 5, 337 (1987)
10. S. Hosoya, M. Sato, H. Katayama-Yoshida: Symp. on "High Temperature Superconductors. Structure and Microstructure", Bad Nauheim, FRG (April, 1988)

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