PHYSICAL REVIEW B, VOLUME 65, 052507

Electrical transport properties of $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ thin film [001] tilt grain boundaries

J. Hänisch,* A. Attenberger, B. Holzapfel, and L. Schultz
IFW Dresden, PO Box 270016, D-01171 Dresden, Germany

(Received 17 September 2001; published 11 January 2002)

Epitaxial thin films of $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ have been prepared by pulsed laser deposition on $SrTiO_3$ bicrystal substrates with symmetrical [001] tilt grain boundaries of 8°, 16°, 24°, 26°, 30°, and 36.8° misorientation angle. The intergrain and intragrain properties of the critical current density and the current-voltage characteristics have been investigated at various temperatures and in magnetic fields up to 7.5 T. The 8° grain boundary exhibits flux creep behavior; it does not constitute a weak link. In contrast, grain boundaries with larger angles exhibit weak link behavior. The critical current density falls exponentially with increasing misorientation angle.

DOI: 10.1103/PhysRevB.65.052507 PACS number(s): 74.60.Jg, 74.72.Hs, 74.76.Bz

I. INTRODUCTION

Since the early days of high-T_c superconductivity it has been known that the critical current is strongly limited by grain boundaries constituting weak links,[1] such as in $YBa_2Cu_3O_7$ (YBCO) (Ref. 2) or $Bi_2Sr_2Ca_1Cu_2O_{8+\delta}$ (Bi2212).[3] Whereas the fabrication of superconducting tapes from $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) via the powder-in-tube technique is quite well developed, the growth of Bi2223 single crystals has been achieved only recently,[4] and the deposition of high quality films is far more complicated than in the case of YBCO or Bi2212. As a result, only a few bicrystal experiments have been performed on Bi2223 until now.[5] Because the critical current density in Bi2223 tapes is relatively good, in spite of their rather complicated microstructure, the question of whether grain boundaries (GB's) in Bi2223 constitute weak links or do not limit the critical current density, has been a controversial issue until now. Here we present a detailed study of electrical transport properties of [001] tilt grain boundaries covering the range from low angle GB's to large angle GB's.

II. EXPERIMENTAL

Using a stoichiometric target, epitaxial Bi2223 films were deposited via pulsed laser deposition on $SrTiO_3$ (STO) bicrystals with symmetric [001] tilt grain boundaries. A KrF excimer laser was used with $\lambda = 248$ nm and a repetition rate of 2 Hz. The deposition rate was 1 Å/s and the resulting film thickness between 100 nm and 150 nm. The deposition temperature was 995 °C in a 2.5 mbar O_2/O_3 atmosphere. To improve the superconducting properties of the films, they were post annealed *in situ* at 880 °C for 3 h and at 830 °C for 21 h in 150 mbar O_2. The temperatures given here are the heater temperatures; the substrate surface temperature, measured using a pyrometer, is up to 150 °C lower depending on the oxygen pressure. The quality of the films was checked by inductive T_c measurements and x-ray diffraction. Bridges of 20 μm width with a length of 0.634 mm were patterned over the grain boundary as well as within the grains using photolithography and wet chemical etching [using ethylenediaminetetraacetic acid (EDTA)]. Resistivity and current-voltage characteristics were measured in magnetic fields up to 7.5 T in a split coil magnet using a four-point geometry.

The critical current was determined with an electric field criterion of 10 μV/cm, calculated along the bridge length.

III. RESULTS AND DISCUSSION

After deposition, the films are epitaxial and already superconducting but show a reduced T_c of about 60 K due to extreme oxygen underdoping. To improve the oxygen doping and hence T_c, the films are post annealed *in situ*. After annealing, inductive T_c values up to 98 K can be achieved.

Figure 1 shows a $\theta/2\theta$ scan of a typical film on a logarithmic scale. Aside from the substrate peaks with their K_β peaks, only the (00l) peaks of Bi2223 are observed. In particular, no peaks are seen arising from the Bi2212 phase. The full width at half maximum (FWHM) of the (0 0 12) rocking curve is 0.8° indicating a good c-axis texture of the films. Figure 2 shows the in-plane texture in the form of a {115} pole figure for the 16° sample consisting of four peaks from each orientation. The shift in position seen between the two upper peaks and again between the two lower peaks arises

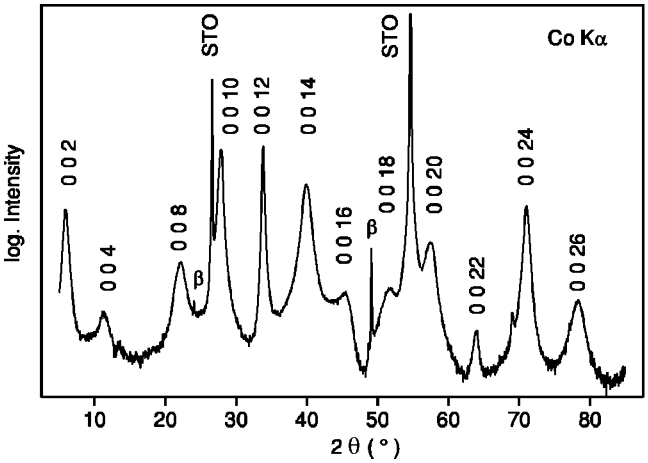

FIG. 1. X-ray diffraction pattern of a typical Bi2223 thin film. Only (00l) peaks of Bi2223 are observed, together with those of the substrate $SrTiO_3$ (STO).

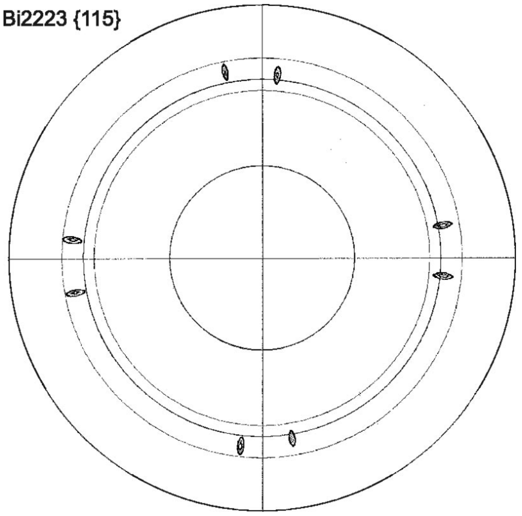

FIG. 2. {115} pole figure of the 16° grain boundary Bi2223 sample.

from a negligible twist of 1.5° in the substrate surface orientation around the GB normal. The FWHM of the peaks is 0.75°. The in-plane and out-of-plane textures are thus comparably good, but indicate the existence of low angle grain boundaries in the films.

The temperature dependences of the resistivity for bridges across the grain boundary and within the grain are very similar as seen in Fig. 3. For $T > 200$ K the temperature dependence is ohmic, the grain boundary producing a higher temperature coefficient. Typical resistivities at room temperature are 350 $\mu\Omega$ cm for grain boundaries and 250 $\mu\Omega$ cm for reference bridges. The residual resistivity defined as the intersection of the linear fit of the Ohmic part of the curve with zero temperature is always less than 25 $\mu\Omega$ cm. T_c^{onset} is 110 K, $T_c^{R=0}$ around 92 K. The relatively large transition width is due to the underdoping, which cannot be totally avoided even with annealing, as well as to the inhomogeneity of the oxygen distribution.

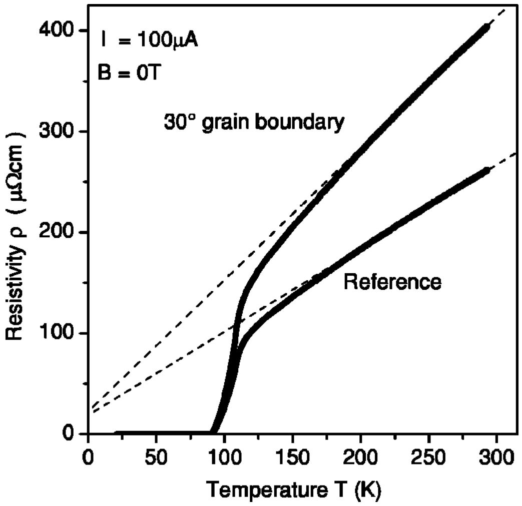

FIG. 3. Temperature dependence of the resistivity of a Bi2223 thin film across a grain boundary (here $\theta = 30°$) and within the grain.

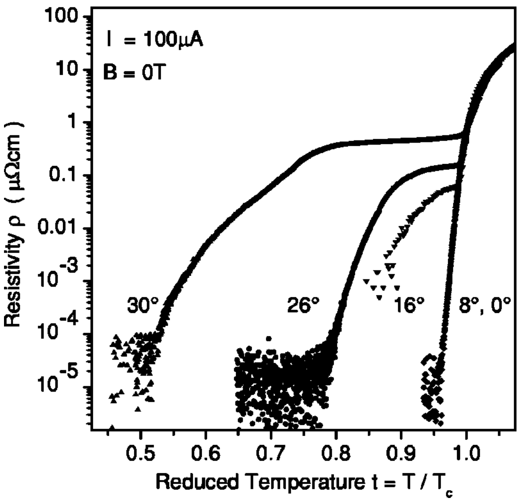

FIG. 4. $\rho(\mathrm{T})$ dependence near the transition temperature T_c for different grain boundaries. The extra structure seen for the large angle grain boundaries is due to TAPS.

Figure 4 shows the superconducting transition of the grain boundary bridges on a logarithmic plot. The sharp drop in resistance at T_c is the resistive transition of the superconducting films (0°). The broad extra structure for $T < T_c$, the so-called foot structure, can be explained by thermally activated phase slip (TAPS)[6,7] across weak links. The 8° grain boundary does not show this feature. The same difference between the 8° GB and the large angle GB's can be seen in the *I*-*V* curves. Whereas the 8° GB possesses power law *I*-*V* characteristics (the exponent of 17.5 at 26 K is relatively small) indicating that j_c is limited by the pinning properties in the grains (flux creep), the large angle grain boundaries show tunnel characteristics, hence do constitute weak links.

In Fig. 5, the temperature dependence of j_c is illustrated for grain boundaries with different misorientation angles and one reference bridge without grain boundary. The low j_c values of the large angle grain boundaries near T_c are an effect due to noise rounding of the *I*-*V* characteristics caused by TAPS. Additionally, a small residual magnetic field can-

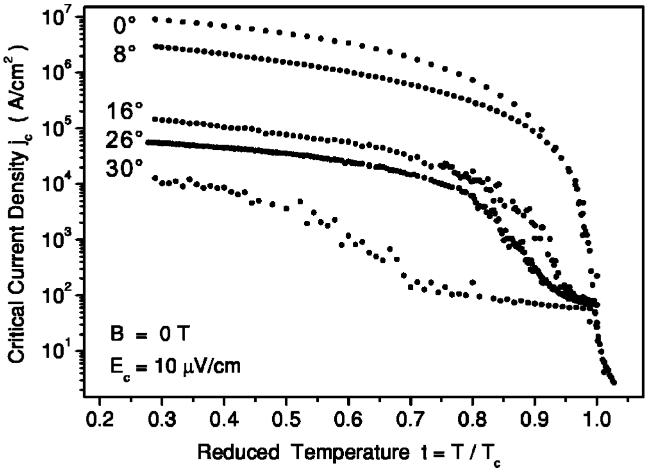

FIG. 5. Temperature dependence of the critical current density of various grain boundaries.

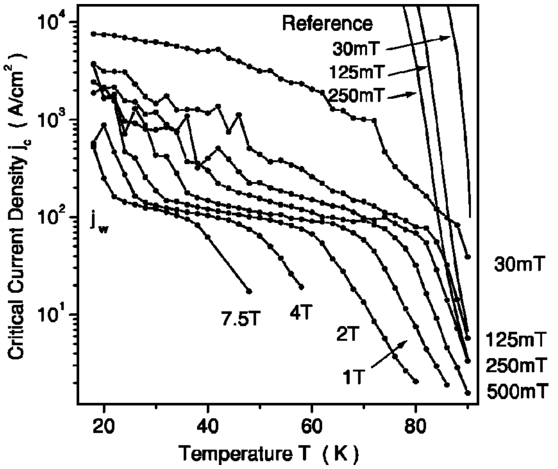

FIG. 6. Influence of the magnetic field on the critical current density of the 24° grain boundary.

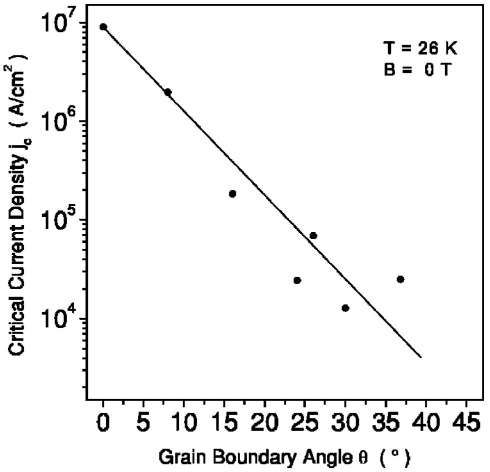

FIG. 7. Dependence of j_c on misorientation angle showing an exponential decay with grain boundary angle.

not completely be excluded, so small fields trapped in the grain boundary may also contribute to this structure. To overcome this noise rounding, the critical current is determined better by the intersection of the tangent $(dV/dI)_{\max}$ with zero voltage (not shown here), as is usual for tunnel characteristics. Then the temperature dependence of j_c can reveal which sort of contact the GB's behave as. For superconductor-insulator-superconductor (SIS) contacts, $j_c(T)$ should follow the Ambegaokar-Baratoff formula,[8,9] which could not be successfully fitted to these results. The quadratic temperature dependence of j_c near T_c together with the excess current j_e in the I-V characteristics and the linear temperature dependence of the quasiparticle resistance ρ_n indicates superconductor-normal-superconductor (SNS) behavior. For low temperatures the difference between the two methods of determining j_c is smaller than the geometrical errors.

Figure 6 shows the influence of a magnetic field on j_c. Near T_c, the critical current density is limited by the pinning properties of the two electrodes; at lower temperatures by the weak-link properties of the grain boundary. The "weak link current density" j_w separating the regions of weak link behavior and flux creep behavior is about 100 A/cm^2. It falls slightly with increasing grain boundary angle. The temperature range of the j_w plateau for a certain field is larger the higher the grain boundary angle. In this temperature range and in magnetic fields up to 7.5 T, which are quite high for tunnel junctions, the large angle grain boundaries show an ohmic characteristic. The magnetic field distribution is uniform for high magnetic fields in SNS junctions[10] and the measured voltage cannot be explained with the movement of Josephson vortices, but in terms of a "simple" resistor. Hence, the plateau at j_w illustrates the temperature dependence of ρ_n in magnetic fields.

In Fig. 7, the angular dependence of j_c is shown for $T=26$ K and $B=0$ T. All these values lie within the range of literature values[11] for YBCO at 4.2 K.

It is clear to see that Bi2223 also shows the exponential angle dependence seen in YBCO and other high-T_c superconductors:

$$j_c(\theta)=j_c(0)\exp(-\theta/\theta_0),$$

where in this case $j_c(0)=9\times10^6$ A/cm^2 and $\theta_0=5°$ at 26 K.

Although the 8° grain boundary does not constitute a weak link in Bi2223, its critical current density is reduced compared to that without GB. This reduction in j_c can be explained as due to a reduction in cross section caused by stress-induced nonsuperconducting regions around the dislocations forming the GB.[12] A reduction in T_c in the grain boundary layer as reported for a 7° [100] tilt grain boundary in Bi2212 (Ref. 13) was not observed for this grain boundary.

IV. CONCLUSION

Bicrystal experiments on epitaxial Bi2223 thin films clearly show that the large exponential depression of j_c with increasing grain boundary angle as seen in other high-T_c superconducting materials also holds for Bi2223. The 8° grain boundary does not constitute a weak link, in contrast to grain boundaries with larger angles. There is no reason to consider Bi2223 as a special high-T_c superconductor in respect to grain boundaries.

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*Email address: j.haenisch@ifw-dresden.de

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