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Investigation of the microstructure of a high-J_c bulk Y–Ba–Cu–O superconductor

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Abstract. In order to reveal the microstructural features of the recently developed high-J_c bulk YBCO material with $T_c = 91.2$ K, J_c (77 K, 2 T) = 2.38×10^4 A cm^{-2}, large-area TEM specimens containing several grains were successfully prepared from this material by a new technique. The material has a strong (001) texture. Many grain boundaries are clean (001) small-angle twist boundaries showing large-area moiré fringes in the micrographs which may be an essential cause of the improved critical current.

1. Introduction

Bulk sintered YBCO materials usually have a relatively low critical current density J_c which decreases rapidly at low magnetic fields [1, 2]. This is generally considered to be caused by the grain boundaries which act like Josephson weak links [3–5]. However, some groups have reported that critical current densities are improved to a certain extent in melt-textured grown (MTG) samples [6, 7] and quenched and melted grown (QMG) samples [8]. Recently, Ren *et al* [9] have achieved an even higher J_c for bulk material in a strong magnetic field: J_c (77 K, 2 T) = 2.38×10^4 A cm^{-2} [9]. Therefore, it is of great interest to learn more about the real microstructure of such a high-J_c well textured bulk superconductor. In the present work, we successfully prepared large-area transparent specimens containing several grains from this high-J_c material by a new technique [10] for transmission electron microscopy (TEM) investigation and revealed some interesting microstructural features.

2. Experimental details

Highly textured bulk $YBa_2Cu_3O_{7-x}$ superconductors with $T_c = 91.2$ K and J_c(77 K, 2 T) = 2.38×10^4 A cm^{-2} are synthesized by a special melt growth process. A sheet sample of dimensions 1.5 mm × 20 mm × 60 mm sintered from superconducting $YBa_2Cu_3O_y$ powder was introduced into a specially designed furnace to go through a

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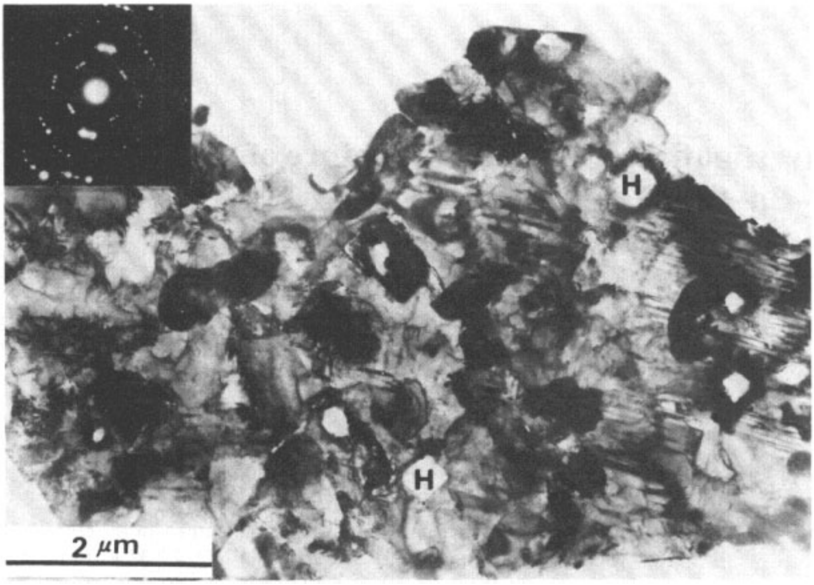

Figure 1. Bright-field (001 beam) image of a large transparent area of the high-J_c YBCO superconductor. The inset is the electron diffraction pattern.

circuit consisting of pre-heating at 1000–1150 °C, cooling at a rate of 0.5–5 °C h^{-1} for 12 h at 925–980 °C, cooling at a rate of 1 °C min^{-1} to room temperature and annealing in flowing O_2 at 400–600 °C for 100 h. X-ray diffraction data show that the grains of the 9 : Bu : Cu = 1 : 2 : 3 phase are preferentially aligned with the *c* axes parallel to each other. The peaks of the 2 : 1 : 1 phase can also be seen. Specimens with dimensions of 0.07–0.3 mm by 0.3–1 mm by 10 mm were cut from the sheet along the cleavage plane, which is also the *a–b* plane, for current density measurements. Silver deposition contacts were used. The critical currents were measured by the standard DC four-terminal method with a voltage criterion of 1 μV cm^{-1}. The transport current was always taken parallel to the *a–b* plane and normal to the magnet fields. Details of the preparation and characteristics of this material have been described in [9].

TEM specimens parallel to the *a–b* plane of the 1 : 2 : 3 phase were prepared from this highly textured sample by the so-called 'back-protection mechanical cleaving' technique described in [10]. Large transparent areas of dozens of square micrometres can be obtained without special difficulties. The specimens were examined in a JEM 200CX transmission electron microscope and an H-800 transmission electron microscope.

3. Results and discussion

A typical large transparent area containing several grains from such a high J_c sample is shown in figure 1. The selected-area diffraction (SAD) pattern indicates that this piece of sample is mainly composed of several $YBa_2Cu_3O_7$ grains with *c* axes parallel to each other and normal to the surface of TEM specimens. The dimensions of most grains are about 1–10 μm. Figure 1 also shows that (110) microtwins are widespread in such material

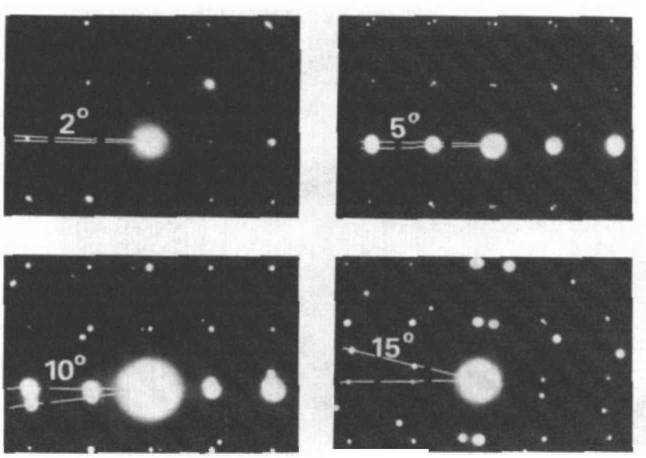

Figure 2. SAD patterns taken in four typical tilt grain boundaries with misorientation angles of 2°, 5°, 10° and 15°.

with a spacing ranged from 20 to 130 nm. This value is near that found in high-J_c single-crystal epitaxial YBCO films [11].

Second-phase particles, Y_2BaCuO_5 and CuO, are distributed in this sample with average distance of several micrometres between each other. The 2 : 1 : 1 phase is easily trapped inside the 1 : 2 : 3 phase owing to the peritectic reaction [12] and so the holes with the symbol H in figure 1 may be the 2 : 1 : 1 phase which dropped off during cleaving.

Most of the misorientation angles of the neighbouring grains rotated around *c* axes are smaller than 15°. In figure 2, the SAD patterns of four typical tilt grain boundaries with misorientation angles of 2°, 5°, 10° and 15° are given. Energy-dispersive x-ray analysis data show that there are no precipitates in the grain boundaries.

Of particular interest are the extensive moiré patterns observed in this high-J_c material. As discussed by Amelinckx and Dekeyser [13], moiré fringes result when

(i) two thin layers of the same crystalline material are superposed with a small orientation difference or

(ii) two thin layers with a slight difference in lattice parameter are superposed in parallel orientation.

We have carefully examined the SAD patterns for a large amount of observed moiré fringes and found that all of them are classified in the case (i), i.e. they are small-angle twist (001) boundaries between two thin $YBa_2Cu_3O_7$ grains. Figure 3 shows such an example. We see that the fringe spacing D is about 7.2 nm, which fits the relationship

$$d/D = 2\sin(\theta/2)$$

for misorientation [13] quite well. Here d is the lattice parameter in the a or b direction with $d = 0.38$ nm and θ is the misorientation angle between the neighbouring grains of value 3° measured from the two sets of SAD spots shown in the upper right corner. This is a typical twist grain boundary nearly parallel to the (001) plane. The common appearance of this type of moiré pattern in our samples demonstrates that many grain boundaries in this high-J_c bulk material are clean small-angle twist boundaries on the

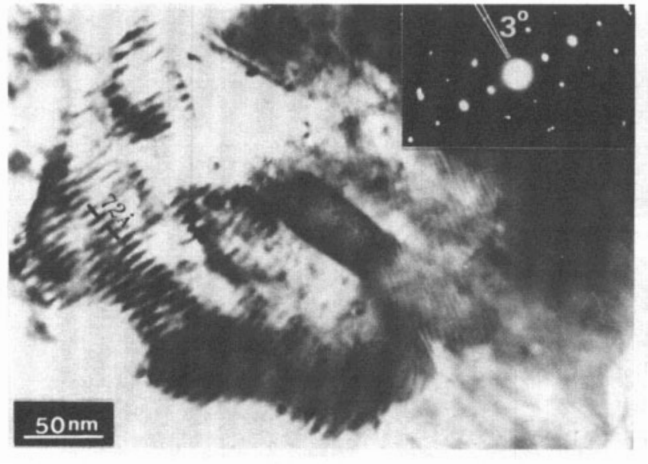

Figure 3. Typical bright-field (001 beam) image of a moiré pattern.

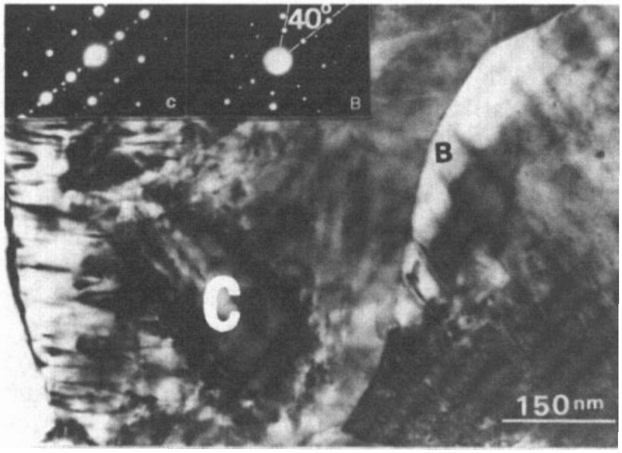

Figure 4. Bright-field (001 beam) image showing a large-angle tilt boundary and a small grain with the *c* axis parallel to the *a* or *b* axis of the large basal grain.

basal plane, which may favour the strong superconducting link and can offer a higher effective critical current consistent with some recent bicrystal results [14, 15].

Dimos *et al* [14] have measured the critical current density J_c^{gb} as a function of misorientation angle for three different grain boundary geometries in $YBa_2Cu_3O_7$ thin-film bicrystals and found a large and systematic drop in J_c^{gb} with increasing misorientation angle. Strong coupling is maintained for small-angle boundaries, where the misorientation angle is less than about 5° [14]. Babcock *et al* [15] have shown that some large-angle grain boundaries between flux-grown bicrystals can also be weak link free.

In our sample a few large-angle tilt boundaries were also found. Figure 4 is such an example. The misorientation angle measured from the SAD pattern shown on the right or from the directions of twins in neighbouring grains is about 40°. At C, we see a small

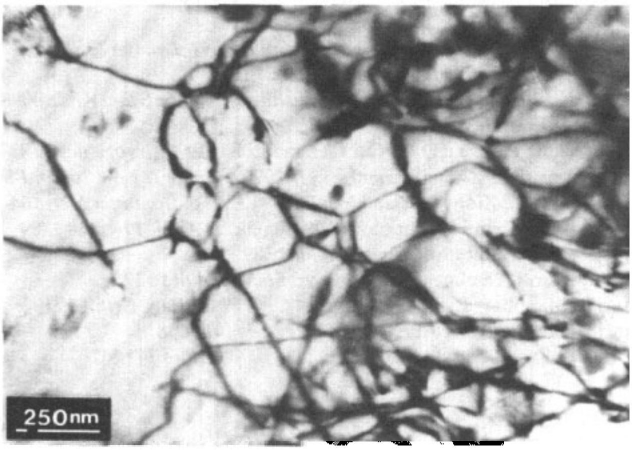

Figure 5. Bright-field image of dislocations in the high-J_c material.

grain with the *c* axis parallel to the *a* or *b* axis of the large basal grain, i.e. a 90°-angle boundary, which is identified with the SAD pattern in the upper left corner. In addition, dislocation substructure is often found. A typical one is shown in figure 5. These dislocations may act as flux-pinning centres and contribute to the enhancement of the critical current.

In summary, by means of a large-area TEM investigation we found that the grain boundaries present in high-J_c bulk YBCO are markedly different from those in the usual YBCO sintered ceramics. Pores, cracks and grain boundary precipitates are absent. A large number of the grain boundaries are coherent twins or clean small-angle twist boundaries nearly parallel to the basal plane. These microstructural features may be the reason for the partial elimination of the grain boundary weak links in the high-J_c material.

Acknowledgments

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