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PATHS FOR CURRENT FLOW IN POLYCRYSTALLINE HIGH TEMPERATURE SUPERCONDUCTORS*

D. M. Kroeger, A. Goyal, and E. D. Specht

Metals and Ceramics Division, Oak Ridge National Laboratory
P.O. Box 2008, Oak Ridge, Tennessee 37831-6114

ABSTRACT

Determinations from x-ray and electron microdiffraction studies of the populations and geometrical arrangements of small angle grain boundaries in Bi-2223 and Bi-2212 conductors and Tl-1223 deposits suggest that current flow in these polycrystalline materials is percolative in character. Comparison of measured misorientation angle distributions to calculated distributions suggest that not only texture but also grain boundary energy is important in increasing the number of small angle grain boundaries.

INTRODUCTION

Long-range strongly-linked current flow in polycrystalline high temperature superconductors has been obtained in only a few materials. The most important of these technologically are Bi-2212 [1] and Bi-2223 [2] conductors, which at fields and temperatures below their irreversibility lines have excellent J_c vs H performance. Strongly-linked behavior has also been obtained in Tl-1223 deposits prepared by two-zone thallination [3], Y-124 multifilament conductor prepared by the oxidation-of-metallic-precursor process [4], and laser abated-123 films deposited on textured oxide buffer layers prepared by ion-beam-assisted deposition [5]. The microstructural factors which lead to such performance have been widely discussed. Of primary importance are the current transmission characteristics of individual grain boundaries and the spatial arrangements of strongly- linked grain boundaries which may lead to the presence of percolative paths of strongly-linked material.

Measurements of the dependence of grain boundary critical current on the grain boundary misorientation angle have been reported for several superconducting [6,7,8] compounds. For all of these compounds J_c(gb), the critical current density for current transmitted through a grain boundary, decreased strongly with increasing misorientation angle, suggesting that such behavior is universal.

Using electron and x-ray microdiffraction techniques, we have determined local textures and the distribution of grain boundary misorientation angles in high J_c Tl-1223 deposits, Bi-2223 powder-in-tube conductor, Bi-2212 melt-processed deposited conductor, and Y124 conductor prepared by the oxidation-of-metallic-precursor process. These measurements have been compared to calculations of the grain boundary misorientation distributions expected for specific macroscopic textures. The results suggest that in these materials current flow is percolative in character, and that grain boundary energy plays an important role in microstructure development.

Bi-2223 and Bi-2212 CONDUCTORS

Using electron backscatter diffraction the individual orientations of a large number of contiguous grains in a polycrystalline material can be determined. The silver sheath on one surface of a short segment of Bi-2223 powder-in-tube conductor was removed by etching. The conductor, prepared by American superconductor, had J_c = 20,000 A/cm^2 at 77 K in self-field. Electron back scatter diffraction patterns were obtained for all of the grains which could be distinguished in SEM in several small areas containing from 15 to 100 contiguous grains. From its diffraction pattern, the absolute orientation of each grain was determined relative to a reference coordinate system. The misorientations between adjacent grains were then calculated and expressed as an angle/axis pair, i.e., the angle through which one grain must be rotated about that axis to align it with the other grain. Depending on crystal symmetry there are a number of equivalent angle/axis pairs which describe a given misorientation. By convention the pair with the smallest rotation angle is used. Of 227 grain boundaries measured, more than 40% had misorientation angles smaller than 15 degrees and, therefore, may be expected to exhibit some degree of strongly-linked behavior [9]. Maps of misorientation angles over small areas indicate that this population of small angle boundaries is sufficient to provide percolative paths for strongly-linked currents. The analysis also indicates that ~8% of boundaries have misorientations which fit within the Brandon criterion [10] for ideal coincidence site lattice (CSL) boundaries which are predicted to have low energies and may also be strongly-linked. A similar analysis of a melt-processed Bi-2212 deposited conductor on silver provided by Dr. H. Kumakura of the National Research Institute for Metals in Tsukuba, Japan, indicated 33% of boundaries had misorientation angles less than 15 degrees and another 25% fit within the Brandon criterion for CSL boundaries.

EFFECT OF MACROSCOPIC TEXTURE ON THE MISORIENTATION ANGLE DISTRIBUTION

For interpreting such results it is useful to know the expected distribution of misorientation angles in a randomly oriented polycrystalline material and how the expected distribution is affected by macroscopic texture. These distributions have been determined from numerical simulations by a method in which the Euler angle description of the orientation of a crystal was used to generate a set of 200 randomly oriented grains [11]. The fraction of grain boundaries with misorientation angles less than a given angle is shown in Fig. 1, for randomly oriented polycrystals with cubic, tetragonal and orthorhombic structures. As expected, the concentration of grain boundaries with misorientation angles less than 15 degrees is small.

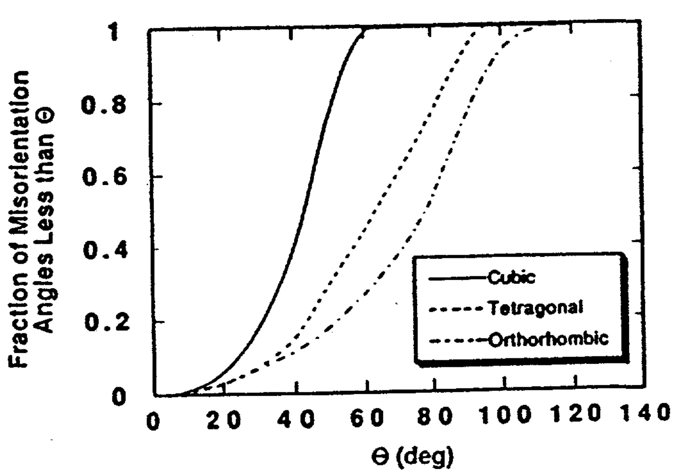

Fig. 1. Expected fraction of misorientation angles less than Θ as a function of Θ for randomly oriented cubic, tetragonal, and orthorhombic polycrystals

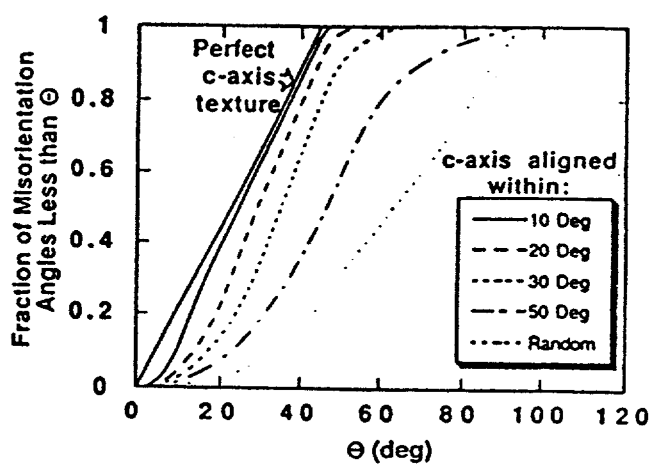

Fig. 2. Expected fraction of misorientation angles less than Θ as a function of Θ for a tetragonal polycrystal with varying degrees of c-axis alignment.

Figure 2 indicates the effect of macroscopic texture on the expected misorientation distribution. Macroscopic texture is simulated by generating the set of grains from an appropriately restricted range of values of the Euler angles. In Figure 2(a) c-axis texture is simulated by restricting the orientation of only the c-axis, the a-axis orientations being completely random. The population of small angle boundaries increases rapidly as the c-axis orientation is restricted, but even for perfect c-axis alignment only 33% of boundaries are expected to have misorientations under 15 degrees. The populations of small angle and CSL boundaries may be enhanced by the effects of grain boundary energy. This is suggested by the statistics reported above for Bi-2223 and Bi-2212. For example, the curve in Figure 2(a) for c-axes aligned to within 10 degrees (approximately the FWHM of the c-axis rocking curve for the Bi-2223 specimen) indicates that in the absence of grain boundary energy effects about 25% of boundaries would have misorientations under 15 degrees, compared to 40% as determined from EBSD.

The effect on misorientation angle distribution of partial in-plane alignment in c-axis aligned materials has also been calculated [11]. Even a modest degree of in-plane texture results in a large increase in the number of small angle boundaries.

Tl-1223 DEPOSITS

The population of small angle grain boundaries is not alone a good indicator of J_c. Tl-1223 deposits often have very high populations of small angle grain boundaries associated with a colony microstructure [12]. The grains within a colony have strong in-plane alignment, so that most grain boundaries within a colony have small misorientations. In these deposits 80% to 90% of grain boundaries may be small angle. However, colony orientations, defined as the average a-axis orientation within a colony, are random. Long-range current flow is limited by the properties, i.e., the misorientations, of grain boundaries at colony intersections. The misorientation angle distribution for grain boundaries at a colony intersection is determined largely by the misorientation of the two colonies, although grain boundary energy may bias the distribution toward small angle or CSL boundaries. Although intra-colony currents are strongly-linked, long-range current flow, we have proposed [13], utilizes a percolative network of strongly-linked (small angle or CSL) boundaries at colony intersections. The concentration of strongly-linked boundaries at colony intersections is lower than in the sample as a whole, and consequently inter-colony J_c is smaller than intra-colony J_c.

The number of small angle grain boundaries at colony intersections should increase with the spread in orientation of grains within a colony and with any tendency for adjacent colonies to have similar orientations as might result from a local or global bias of colony orientation. The FWHM of peaks in XRD azimuthal scans tends to be ~10-20 degrees, indicative of a similar spread in a-axis orientation. Since Tl-1223 is tetragonal the maximum misorientation angle between c-axis aligned grains is 45 degrees. If colonies are randomly oriented and each irregularly shaped colony intersects several other colonies, it is probable that a significant fraction of intersection area will have overlapping grain orientation distributions. The critical current of a lattice of randomly oriented colonies having a population of small angle boundaries at colony intersections derived from the overlap of the distributions of grain orientations within a colony was simulated [14] using the limiting path method [15]. The model predicts significantly higher in-field J_c for a colony microstructure than for a c-axis textured deposit with random a-axis orientations.

That current flow is percolative in character has important implications which have not yet been explored. For example, the "percolation threshold" for the density of strongly-linked boundaries, below which long-range J_c would be zero, depends upon, among other geometrical factors, the dimensionality of current flow. The number of percolative options increases with the number of nearest neighbors and thus with the dimensionality of the system. The Tl-1223 deposits which we have analyzed have very good c-axis alignment and are only a few grains thick. As a consequence, current flow is two-dimensional. Powder-in-tube Bi-2223 conductors have relatively broad c-axis rocking curves and many misaligned grains which may make current flow more three-dimensional. J_c is also expected to depend on conductor geometry. For example, as conductor width is reduced, percolative options, and consequently J_c, are decreased. This effect has been demonstrated experimentally in Tl-1223 deposits.

It is also possible that percolation effects are important in the design of multifilamentary Bi-2223 conductors. As filaments are made thinner and c-axis alignment is improved, current flow may become more two-dimensional, causing an increase in the percolation threshold for strongly-linked grain boundaries. Narrow filaments which are comparable in width to the Bi-2223 grain size would have fewer percolative options.

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