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**TRANSPORT CRITICAL CURRENTS AND GRAIN BOUNDARY COUPLING
IN HIGH- T_c SUPERCONDUCTORS***

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Transport Critical Currents and Grain Boundary Coupling in High- T_c Superconductors*

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Abstract

We propose that the transport critical current density for high- T_c thin films, bicrystals and bulk ceramics is determined by the magnetic field penetration into the grain boundaries; the grain orientation may not be an important factor. The parameter $(\lambda_L/\lambda_J)^2$ can characterize the strength of the grain boundary coupling, which depends mainly on the crystal coherence and connectivity at the boundary area, but not on the crystal orientation.

It has been observed that the grain boundaries in bulk samples of high- T_c superconductors exhibit so-called weak-link effect as a result of short coherence length, which severely limits transport J_c at very low fields of a few hundred Gauss (1-7). To study the transport current behavior in association with the weak-link effect at grain boundaries, researchers have focused on the correlation between the transport J_c and grain boundary structure and orientation.

Dimos et al. (8) developed a unique method to study the angle dependence of the critical current density across the grain boundaries, J_{gb} . By epitaxial growing of 123 thin films on the bicrystals of $SrTiO_3$ substrates, they were able to make films with single grain boundaries. These boundaries are oriented in different angles characterized by θ . They found that, near 4.2 K, the J_{gb}/J_g (J_g is the J_c measured within a single grain) is reduced 50 times as θ increases from 0° to 35° . Based on these results, they concluded that the strong angular dependence of J_{gb} is associated with the distortion of the vortices by the grain boundary dislocations. Their conclusion also clearly implies that materials with high-angle grain boundaries are not capable of carrying high-transport J_c .

However, Babcock et al. have recently reported (9), that the weak-link behavior is not observed in bicrystals with high-angle grain boundaries. In the report by Babcock et al. the J_{gb} of five bicrystals with $\theta = 3^\circ, 14^\circ, 22^\circ, 38^\circ$, and 90° was measured up to 7 T. Surprisingly, all these crystals exhibited critical current density behavior controlled by flux-pinning (i.e., weak-link effects were not observed). These striking experimental data clearly indicate that the high-angle grain boundaries are not necessarily connected with the Josephson junction weak links.

We propose that the major influence on the transport J_c is not the orientation of the grain boundaries but rather the degree to which the grain boundaries can be penetrated by the external applied field. In this paper, we use the physical picture of the Josephson junction to explain the flux pinning-dominated transport J_c in $\text{YBa}_2\text{Cu}_3\text{O}_x$ samples with high-angle grain boundaries.

One can assume that each grain boundary in $\text{YBa}_2\text{Cu}_3\text{O}_x$ is a Josephson junction with a junction length (grain size) L and a junction thickness t . By definition, the Josephson junction current density, J , is associated with the phase difference, ϕ , of the pair wave functions on both sides of the junction (10),

$$J = J_c \sin \phi, \quad (1)$$

where J_c is the maximum zero-voltage current density across the junction. As an external magnetic field, H , is applied, a phase gradient is created, which is given by

$$\partial\phi/\partial x = 2e(t+2\lambda_L)B/h, \quad (2)$$

where λ_L is the London penetration depth, and B is the magnetic induction. By integrating Eq. (2), we have

$$\phi(x) = 2e(t+2\lambda_L)By/h + C, \quad (3)$$

where C is the integration constant. Substituting Eq. (3) into Eq. (1) yields

$$J = J_c \sin[2e(t+2\lambda)B/h + C]. \quad (4)$$

It has been shown that J should decay in an oscillating manner as the applied field increases, which can be expressed as

$$J = J_c |\sin(\pi H/H_0)| / \pi H/H_0, \quad (5)$$

where H_0 is a characteristic field, J_c goes to zero at $H = H_0$, and

$$H_0 = \Phi_0 / (t + 2\lambda_L)L, \quad (6)$$

where Φ_0 is the flux quantum. For polycrystalline samples, H_0 is not well defined. However, H_0 has been approximated as the field at which the transport J_c is reduced to half of the $J_c(0)$ (i.e., the zero-field J_c), which has been estimated to be ~ 25 Gauss for $\text{YBa}_2\text{Cu}_3\text{O}_x$ (7).

When grain boundaries act as Josephson junctions, they can be uniformly penetrated by the external field. The Josephson penetration depth is given by

$$\lambda_J^2 = h / 2eJ_c \mu (2\lambda_L + t). \quad (7)$$

The coupling strength of the grain boundaries is related by the ratio of the London penetration depth to the Josephson penetration depth, $F = (\lambda_L/\lambda_J)^2$, as suggested by Hylton and Beasley in a laminar model (11). In the strong-coupling limit (7) where the barrier disappears, Eq. (7) can be written as

$$F = (\lambda_L/\lambda_J)^2 = J_{gb}/J_g. \quad (8)$$

Thus, one can see that grain boundaries exhibit weak coupling when $(\lambda_L/\lambda_J)^2 \ll 1$, while grain boundaries are strongly coupled for $(\lambda_L/\lambda_J)^2 \gg 1$.

The key point here is that the occurrence of the Josephson junction is dependent solely on the ratio of the London penetration depth to the Josephson penetration depth; it is not directly related to the crystal orientation of the grains. This point is illustrated in Figure 1.

Figure 1a shows schematically a low-angle grain boundary with the applied field in the c axis. Because the London penetration depth along the ab plane is the same on both sides of the grains, we expect an evenly distributed field profile as indicated in Fig. 1a. If the grain boundary is weakly coupled, as has been observed for sintered samples, the boundary area is uniformly penetrated by the applied field, and we have $(\lambda_L/\lambda_J)^2 \ll 1$. In contrast, the grain boundary is strongly coupled, as in the case of melt-textured or flux-grown bicrystals, high crystal coherency at the boundary makes it difficult for the field to penetrate the grain boundary. Thus, $(\lambda_L/\lambda_J)^2 \gg 1$ can hold for a strongly coupled grain boundary. In either case, the coupling strength has nothing to do with grain orientation. In reality, weak-link effects have been observed in magnetically aligned samples with low-angle boundaries (12), as represented in Fig. 1a. A similar situation is represented in Fig. 1b for c axis-oriented grains, but with an angle θ . The field penetration is symmetrical on both sides of the boundary, since the values of λ_{ab} is the same in both grains. Again, both weak and strong coupling could exist for the grain orientation presented in Fig. 1b. The field penetration becomes uneven as both grains are misoriented by 90° as shown in Fig. 1c. According to previous measurements (13), $\lambda_{ab} \sim 1500 \text{ \AA}$ and $\lambda_c \sim 5000 \text{ \AA}$. Thus, for the configuration shown in Fig. 1c, the field penetration on the

right side (H || c) is more severe than that on the left side (H || ab). An important point to be noted here is that the same situation shown in Fig. 1a and Fig. 1b can occur for the orientation shown in Fig. 1c depending up on the grain boundary contact.

In previous studies, the path for the transport of electrons at the grain boundaries was overemphasized and failed to explain satisfactorily the complex nature of the superconducting critical currents. In reality, although the superconductor is highly anisotropic, the current flow will redistribute across the high-angle grain boundaries if strongly coupled. On the other hand, weak-link behavior can still exist at low-angle grain boundaries when weakly coupled. The transport J_c will be severely reduced only when the grain boundaries are penetrated by the applied field. We therefore conclude that the high supercurrent transport is determined, to a large degree, by the resistance of the grain boundary area to the penetration of the applied field but not by the "smooth path" of the electrons.

It has been well reported that transport J_c exhibits flux pinning-dominated behavior in melt-processed samples, including flux-grown bicrystals (9), zone-melted samples and melt-textured samples (14,15). High-angle grain boundaries have been well observed in these samples, providing clear evidence for strongly-coupled high-angle grain boundaries.

Although no microscopy data indicate the essential structural differences between strongly and weakly coupled grain boundaries in terms of field penetration, certain mechanisms can be ruled out in terms of the formation of grain boundaries through different processing methods. An important point which may have been ignored in transport studies is that the grain boundaries must be treated differently in liquid-processed and sintered samples. It is reasonable to consider the high-angle grain boundaries to be the

following three types: (a) amorphous layer (AGB); (b) transition structure (TGB), and (c) perfect grains up to the boundary (PGB) (16). When there exists a layer with no well-defined lattice structure relating to the grains, and the properties of the boundary is independent of the difference of the orientation, it may be considered as AGB. However, if the grain boundary is structurally related to the adjacent grains and the properties of the layer depend on the difference of the orientation, TGB may be an appropriate term to describe the boundary. The term PGB describes the distinct interface between two perfect crystals without any structural distortion at the edge of the crystals.

Amorphous-type grain boundaries are likely to be formed in sintered samples. The bulk sample is formed through solid-state diffusion at near 950° C. The sintering process through solid-state diffusion is slow, and grains are randomly oriented. As a result of sintering, the final product is always porous and grain boundaries are poorly connected both electrically and mechanically. Grain boundaries thus possess a high degree of disorder and are glassy in nature. This type of boundary can be penetrated easily by the applied field even for low-angle boundaries; thus, $F \ll 1$.

In contrast, grains are formed in a fundamentally different way in the melt-texturing process. According to the phase diagram of $\text{YBa}_2\text{Cu}_3\text{O}_x$, peritectic and eutectic reactions occur during partial melting (17-19). The diffusion is much more rapid as a result of the high kinetic energy of the atoms. More important, the growing process is significantly longer (3 - 5 days) than the sintering time (less than 10 h), so that the grain boundaries can migrate and lower the free energy of the system. In particular, the motion of the grain boundaries results in a decrease in the surface area, which leads to the crystal growth in a certain orientation with respect to the temperature gradient. Naturally, low-angle and low energy boundaries are preferable in

terms of energy requirement, and texturing in large regions is established. Since melt-growth is a long and natural process, the grain boundaries tend to be types PGB and TGB (20). The crystal coherence and connectivity at grain boundaries (including high-angle boundaries) are thus much higher for melt-processed samples than for sintered samples. Indeed, it has been observed in high-resolution transmission electron microscopy experiments that many lattice facets exist at the grain boundaries of thin films and melt-processed samples, while no such structural feature has been observed in the sintered form (21). This is an important structural difference, since the high crystal coherence can be achieved by facet structure at grain boundary of the thin film and melt-processed materials. Such types of grain boundaries are strongly coupled ($F \gg 1$) and are not easily penetrated by the applied field. Note that the strong coupling applies to high-angle boundary as well since field penetration does not concern crystal orientation for well-joint boundaries.

The degree of field penetration can be examined by magnetization measurements. It has been shown (22) that the peak position in the flux creep-rate ($dM/d\ln t$) versus field (or temperature) plot corresponds to the full penetration field, H^* . Figure 2 shows the field penetration, H^* , versus temperature for a sintered and a melt-textured sample with high-angle grain boundaries. Note that the melt-texture sample has large domains separated by high-angle grain boundaries and the field is applied randomly to the sample. As can be seen in the figure, the sintered sample has considerably lower H^* values than does the melt-textured sample. It is clear that H^* is closely related to the field penetration at weakly linked areas and is an indirect measure of the coupling parameter, F . As calculated by T. L. Hylton and M. R. Beasley (11), $F = 0.00063$ and 8.6 for a ceramic sample and a single crystal, respectively. Magnetization measurements provide strong evidence

for the difference of field penetration in weakly and strongly coupled samples, which proves that the coupling strength is mainly related to the connectivity, rather than to the orientation of the grain boundaries.

In summary, the transport J_c is significantly reduced only when the grain boundaries are penetrated by the applied field and $(\lambda_L/\lambda_J)^2 \ll 1$. The degree to which a boundary area is penetrated by the applied field is largely related to the crystal coherence and connectivity at the boundary area (or, in other words, the boundary structure), but is not dependent upon the orientation.

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Figure captions

Fig. 1 Schematics showing the field penetration at grain boundaries with different orientation.

Fig. 2 Full field penetration, H^* , versus temperature for a sintered sample and a melt-textured sample of $\text{YBa}_2\text{Cu}_3\text{O}_x$. Note that the sample sizes of both samples are approximately the same ($\sim 2\text{mm}$).

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