Observation of \( J_c \) oscillations in bitextured YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) films

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A series of in-plane bitextured YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) thin films were obtained on yttria-stabilized ZrO\(_2\) substrates with various ratios of [100] and [110] grains. Their critical-current-density properties were measured in an applied magnetic field of up to 4 T. A diffraction pattern in \( J_c(H) - H \) is observed at high magnetic fields in thin-film samples. This diffraction pattern is due to the weak-link character of 45° grain boundaries. The volume-pinning-force densities were calculated, with two separate mechanisms governing thin-film \( J_c \) behavior in the magnetic fields identified. In the range of \( H < 0.5 \) T, weak-link properties of 45° grain boundary dominated. At fields above 1 T, flux flow along strongly coupled grains was determined to be the main mechanism of dissipation.

I. INTRODUCTION

The study of the electrical and magnetic properties of high-\( T_c \) cuprate superconductors is very interesting from an application as well as basic-science point of view. It was discovered by many authors that in bulk YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) (YBCO) material, the critical current \( I_c \) showed an oscillatory behavior at low magnetic fields. This was attributed to grain boundaries (GB's) in the samples and the resulting Josephson behavior.\(^1\)\(^-\)\(^3\) It was also found that in bulk and single crystal YBCO samples, there existed both intergrain junctions and intragrain junctions.\(^4\)\(^-\)\(^5\) Intragrain junctions are considered to be the main hindrance in the application of these high-\( T_c \) superconductors as they cause a steep drop of \( I_c \) in the presence of even a very small magnetic field. There is an abundance of experimental evidence to prove that GB's behave as Josephson junctions (JJ). In the case of bulk material, the field dependence of \( I_c \) can be explained by a model of JJ arrays with random orientations and grain sizes.\(^5\)\(^-\)\(^7\)

The situation looks better for the case of thin films. The GB's weak link property has actually been utilized in the fabrication of devices such as SQUID's. Ivanov et al.\(^8\) demonstrated a dc SQUID which was fabricated from artificial GB's on bicrystalline yttria-stabilized ZrO\(_2\) (YSZ). It is well known that GB's are very sensitive to magnetic fields. However, in previous studies, the applied magnetic field is only limited to a small range and is typically less than 100 Oe.\(^9\)\(^-\)\(^10\) This is because the classical JJ theory predicted \( J_c \) oscillation only when \( H < H_{c1} \). Higher fields are rarely studied for thin film. One of the few studies of \( J_c \) behavior of thin films at high magnetic fields was recently reported by Daumling et al. of IBM.\(^11\) But they have mainly concentrated on the \( I_c-\)vs-\( H \) hysteretic aspect.

In this paper we would like to report a study of the \( J_c \) characteristics of bitextured YBCO thin films (thickness 0.2 \( \mu m \)) in the presence of a strong magnetic field. Fields of up to 5 T were used in this investigation. For the first time, a diffraction pattern was observed near 0.1 T for thin film samples. It was also found that the diffraction oscillation period \( \mu_0H_0 \) agreed reasonably well with a model which considered vortex distribution in the mixed state.\(^12\) We believe that these oscillatory features are directly related to the 45° GB's. Besides these oscillations, a very broad and smooth peak at \( H > 4 \) T is also identified. The mechanism of flux flow is used to explain this phenomenon.

II. EXPERIMENTAL

The samples used in this study were bitextured YBCO thin films. By bitexture it is meant that the grains in these polycrystalline films have only two allowed orientations. This is a unique feature which is found so far only for YBCO on YSZ (100).\(^13\) The YBCO grains are typically 0.1–0.3 \( \mu m \) in size. They are all c-axis perpendicular, i.e., YBCO[001]||YSZ[001]. However, the YBCO axis can either be parallel to the YSZ a axis (11.9% lattice mismatch) or at 45° to the YSZ a axis (5.0% mismatch). Thus the GB's between adjacent grains are either 0° or 45° misoriented in the a-b plane of YBCO. We have previously shown that such bitexture films showed an interesting decrease in \( J_c \) as a function of the texture ratio \( \eta \) (Ref. 14)

\[
\eta = \frac{[100]}{([100] + [110])},
\]

where [100] stands for the percentage of YBCO grains with an axis parallel to YSZ, [100] and [110] is the percentage of YBCO grains with an axis at 45° to YSZ. Detailed descriptions of \( \eta \) measurement can be found in Ref. 14. In brief, by x-ray pole figure measurement (\( \Phi \) scans) of the (108) diffraction peak, \( \eta \) can be obtained by comparing the peak intensities along the \( \langle 110 \rangle \) and \( \langle 100 \rangle \) substrate directions. It was also reported in Ref. 14 that the value of \( \eta \) could be controlled by the deposition rate. We have also successfully modeled the \( J_c \) behavior of such films by assuming a random network of 0° and 45° GB's.
The bitextured YBCO films were deposited by standard pulsed laser deposition (PLD) technique on single crystal YSZ(100) substrates. By carefully controlling the laser fluence and keeping all the other deposition conditions constant, we successfully obtained a series of samples with relatively constant $T_c$ values but with texture ratio $\eta$ ranging from 0.0 to 0.5. Details of the films growth, electrical properties, and critical current characteristics can be found in Ref. 14. Table I shows a summary of the properties of the samples used in this study.

All the $J_c$'s were obtained by standard four-probe transport measurement crossing microbridges of 20 $\mu$m wide and 1 mm long with a criteria of 1 $\mu$V/cm. The microbridges were patterned by a cw Nd:YAG laser operating at the second harmonic wavelength. The magnetic field $H$ was applied perpendicular to the film surface (Cu-O plane) while the current flowed in the plane. In the measurements, the films were cooled in zero field to a designated temperature and the field was then increased monotonically to the desired value without overshooting.

### III. RESULTS AND DISCUSSION

The $J_c$ dependence on the $H$ field for films with different textures is shown in Fig. 1. The $J_c$ is normalized to the zero field value. It can be seen that for those films with low texture ratio, $J_c$ drops slowly. Figure 2 shows the field dependence of $H_{1/2}$ which is defined as the field at which the $J_c$ has dropped by one-half. It can be seen that $H_{1/2}$ is as high as 1.3 T for the films with low texture ratio. On the other hand, films with many 45° GB's, show $J_c$ values dropping very steeply. $H_{1/2}$ was only about 0.05 T for the most mixed textured sample. These results imply that high angle GB's are responsible for the fast decay of $J_c$ in small magnetic fields. Near the 45° GB, the superconducting state is very weak and a small magnetic field can have a significant effect in causing a superconducting to nonsuperconducting phase change. Hence, in the transport measurement, the current that passes through the grain and GB network is essentially limited by the 45° GB.

While a 0.05-T field caused the transport $J_c$ to decrease by one-half in our highly bitextured sample, $J_c$ was reported to drop by 2 orders of magnitude in bulk samples under the same field. This tremendous difference, we believe, is due to the intrinsic difference of the grain-grain coupling in thin films and in sintered bulk material. Similar observations have been noted by many groups. In general, the coupling of GB is much stronger in thin film samples.

It is also interesting to compare the transport intergranular $J_c$ to the intragranular $J_c$. The intragranular $J_c$'s, can be obtained in terms of magnetic $J_c$ ($J_{cm}$). It showed a different behavior versus the $H$ field. As mentioned in Ref. 14 we obtained the intragranular $J_c$'s by performing the magnetic moment measurement on the disc-shaped sample with $r=1.22$ mm and applying Bean's model: $J_{cm}=30M/R$, where $M$ is the macroscopically averaged magnetic moment per unit volume. The

![Field dependence of normalized transport $J_c$ for bitextured thin YBCO films at 35 K, the texture ratios $\eta$ are 0%, 13.6%, 41.0%, 50.0% from top to bottom.](image-url)
value of \( R \) is subject to discussion even if Bean’s model is applicable in the present situation. Nevertheless, this formula can be used to obtain the relative trend of \( J_{\text{em}} \). \( M \) can be measured as a function of \( H \). Hence, the field \( H_{1/2} \), at which \( J_{\text{em}} \) drops by one-half can be obtained for the series of samples. The results are also shown in Fig. 2. It is very clear that \( H_{1/2} \) does not have a strong dependence on the texture ratio. The obvious reason is that \( J_{\text{em}} \) measures the intragranular critical current density and the existence of high angle GB’s is not of concern. On the other hand, the transport \( J_c \) is strongly dependent on \( H \).

There is another important feature in Fig. 1 that needs to be pointed out. In samples with large texture ratios, we observed \( J_c \) oscillations in the high \( H \) field region \( \sim 0.5 \) T. As far as we know this is the first report of this kind of diffraction pattern in the high field range for thin films. Obviously the correlation of many 45° GB’s in the film is responsible for this observation.

The oscillatory behavior of \( J_c \) versus \( H \) is better seen by plotting the flux pinning force density \( F_p = |J_c \times H| \). Figure 3 shows a plot of \( F_p \) versus \( H \) for three samples. It can be seen that the high texture ratio samples show pronounced oscillations, while the more pure texture sample does not show such behavior. In Fig. 4 we show the log-log plots of \( J_c \) vs \( H \) for samples No. 9 and No. 10. It is seen that the curve for sample No. 9 has a striking resemblance to Ekin’s observation.\(^1\) First of all, \( J_c \) decays slowly initially, and then decreases rapidly. Following the steep drop, there is a plateau, and a second gradual \( J_c \) drop afterward. The only and very important difference is that the \( H \) field is shifted approximately 2 orders of magnitude higher.\(^1\)

As interpreted by Ekin et al., the magnetic field \( H_0 \), where the first precipitous \( J_c \) drop occurs (in our film, \( \mu_0H_0 \sim 0.1 \) T), is the field where Josephson weak links are decoupled. \( H_0 \) is given by

\[
\mu_0H_0 = \frac{\Phi_0}{Ld} = \frac{\Phi_0}{Ld} = 2.07 \times 10^{-7} \text{ G cm}^2,
\]

where \( L \) is the junction width, and in the classical model \( d = 2\lambda_{ab} + t \) where \( t \) is the junction thickness, and \( \lambda_{ab} \) the penetration depth in the \( ab \) plane. On setting \( L \) to be the average grain size, \( \langle L \rangle = 0.25 \) \( \mu \text{m} \) in our film, \( d \) turns out to be 83 nm. Apparently, this number is inconsistent with a \( \lambda_{ab} \) of about 200 to 300 nm in YBCO. This discrepancy could actually be expected if we closely examine the difference in the physical assumptions of the classical model and our experimental situation. The classical model is developed assuming that the superconductor is in the Meissner state with \( H < H_{c1} \), and no vortex exists in the sample. However, in the field regime of near 0.1 T in our situation, the high-\( T_c \) type-II superconductor is in the mixed state. There are flux lines penetrating the sample and are pinned by various pinning centers. The transport current can only flow in the vortex-free region. Therefore the vortex distribution in the neighbor-
ing grains that form the Josephson junction should be considered. A very recent model given by Bulaevskii et al.\textsuperscript{12} takes the vortex state into account. It is assumed that, the current flow near the grain surface region where it is vortex free. The thickness of this region is $z_f$, rather than the whole junction width $L$. They found that $z_f = \lambda_{ab}\cosh^{-1}(H/B)$, where $B$ is the induction in the mixed state beyond that vortex-free region. By calculating the oscillation period by using Eq. (1), $d$ turns out to be

$$d = t + 2\lambda_{ab}\tanh\frac{z_f}{\lambda_{ab}}. \tag{2}$$

In high magnetic fields of $H > H_c1$, $d$ approaches $t$, instead of the classical $2\lambda_{ab} + t$. At a first glance however, our result of $d = 83 \text{nm}$, does not fit this result either. But, it needs to be noted that in the calculation, the junction width $L$ is assumed to be the grain size. This assumption is appropriate for bulk samples, but may not hold well for thin films because the 0° GB are strongly coupled. Therefore the effective junction width could be larger than the individual grain size. Consequently, our result could actually be consistent with Bulaevskii’s prediction. Finally, an important point that needs to be noted is that in Bulaevskii’s\textsuperscript{12} calculations, $H$ is applied to the $ab$ plane, i.e., along the junction direction. In our experiment, $H$ is parallel to the $c$ axis. However, it is still applicable to our situation because in the microscopic picture, vortices are misaligned in different grains, especially in those higher mixture ratio films. Therefore, a component of field parallel to the 45° GB’s does exist even when $H$ is externally applied to the $c$ axis of the film. In the experiment we also observe that $J_c$ oscillation is relatively independent of whether $H$ field is parallel or perpendicular to $c$ axis.

The flux pinning force density $F_p$ is a useful parameter to describe the pinning mechanism in the sample. It is interesting to note that for all films $F_p(H)$ gradually increases and approaches saturation value near 3 T. For films with high texture ratios, there are diffraction peaks due to the weak link behavior of the 45° GB’s. We interpret the broad plateau near 3 T as the nonweak-link component in pinning. Ekin \textit{et al.} also observed this component in their study of bulk samples.\textsuperscript{1,16} It was explained that this component in intergranular transport current was due to the anisotropic flux flow characteristic of $J_c$. In our films, this component is present because even in high fields, some current paths still exist where the grains are strongly coupled to each other. These current paths are not limited by weak links. Instead, they are governed by the mechanism of flux flow. Actually, from our results, this flux flow component is dominant in films with low texture ratios of up to 21%. The results for sample No. 6 with the ratio of 13.6% in Fig. 3 is typical for a flux flow mechanism. In films having many 45° GB’s, the flux flow mechanism and JJ weak links are both important in determining the behavior of $J_c$ in a magnetic field. It can be noticed from Fig. 3 that these two mechanisms have fairly different field dependent features. The flux flow is seen to resemble a broad peak (it should approach zero at $H_c2$). The weak link diffraction oscillations show a narrow width.

In conclusion, we measured the transport critical current densities of bitextured films in the presence of high magnetic fields of up to 4 T. In films with high texture ratios we observed diffraction pattern due to weak link properties of 45° grain boundaries at $H$ near 0.1–0.5 T. Our results are consistent with Bulaevskii’s prediction about the oscillation period in high magnetic fields. The flux pinning force densities clearly show that there are two mechanisms governing the $J_c$ property in a magnetic field. One of them is the grain boundary weak link behavior and the other is flux flow in the path consisting of strongly coupled grains.

\section*{ACKNOWLEDGMENTS}

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\begin{thebibliography}{99}
\bibitem{5} R. B. Stephens, Cryogenics \textbf{29}, 399 (1989).
\bibitem{14} S. Y. Dong, D. H. Kim, and H. S. Kwok, to be published in Physica C (to be published).
\end{thebibliography}