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Grain-misorientation control of the critical current in high-j_c epitaxial $YBa_2Cu_3O_7/SrTiO_3$ films

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A systematic study is reported of physical factors that control the j_c values of high-j_c ($\geq 10^6$ A/cm^2 at $T=78$ K, $B=0$) epitaxial $YBa_2Cu_3O_7/SrTiO_3$ films. By x-ray analyses and transport j_c measurements of 15–20 microbridges produced on the same films, it is shown that the film microstructure is the most important factor controlling j_c. A direct correlation is found between j_c values and the grain misorientation angle Δ, and an exponential dependence $j_c(\Delta)$ is obtained as $j_c(78\text{ K}) = 7\times10^6 \exp(-\Delta/2.4\text{ deg})$ A/cm^2 for Δ in the range of 0.5–7.0 deg. A model of film critical current is developed in which the grain misorientation highly influences the quantity of weak links in a film.

I. INTRODUCTION

The low values of the critical current density, j_c, of high-temperature superconductivity (HTSC) materials are one of the main obstacles for their applications. High-j_c values ($j_c > 10^6$ A/cm^2, $T=77$ K, $B=0$), however, have been found in epitaxial films deposited on single-crystal substrates by pulsed laser deposition or magnetron sputtering.[1,2] But these values were demonstrated only on microbridges with lengths below 1 mm and often cannot be obtained on long current-carrying lines or even are not reproducible in a series of microbridges on one film surface. Long current-carrying lines, however, are an essential element of various electronic devices and j_c values $\geq 10^6$ A/cm^2 are necessary for their realization. Important examples of such devices are the multiturn input coil of superconducting quantum interference device magnetometers,[3] which has not been produced yet, or meander structures for ir radiation detectors. In this connection there is great interest to determine the main factors that control the value and spread of j_c of typical $YBa_2Cu_3O_7$ (YBCO) films.

Usually for the high-j_c films the j_c values and $j_c(B)$ dependences are described within the concept of flux creep in nongranular superconductors.[4] In this case the resistive state arises as a result of thermal activation of vortices from pinning centers with an activation energy U, reduced by the work of the Lorentz force, qj, on the vortex. For $U/kT \gg 1$ the possibility of vortex depinning, which is proportional to $\exp[-(U-qj)/kT]$ increases sharply at $qj \approx U$. This leads to a critical current density value proportional to U. This means that U is the main factor controlling j_c and a clear correlation between j_c and U must be observed.

To verify this correlation a detailed analysis of voltage-current characteristics (VCC) and $j_c(B)$ dependences for epitaxial YBCO/$SrTiO_3(100)$ films, produced by pulsed laser deposition, has been carried out in preceding work.[5] The films under investigation had superconducting transition temperatures, T_c (zero-resistance values), of 89–91 K but with a large spread of the critical current densities j_c (78 K, $B=0$) in the range of 4×10^5 to 6×10^6 A/cm^2. The U values have been deduced from a comparison of the exponential part of the voltage current characteristic, $E\sim\exp(-j/j_1)$, and the theoretical VCC,[5] describing the crossover between thermally assisted flux-flow, flux-creep, and flux-flow regimes as

$$U/kT = \frac{j_f}{j_1} + \ln C \ ,$$

where j_f is the current density value, at which the fit of the exponential part of the VCC reaches the flux-flow level of the electrical field $E_f \equiv \rho_f j$, with $\rho_f = \rho(100\text{ K})B/B_{c2}$. The absence of a direct correlation between j_c and U has been clearly shown in contrast to the creep model: the $U(T=78\text{ K}, B=10\text{ mT})$ values of about 0.35 eV are practically the same for all films. This fact led us to the suggestion of a model of nonuniform critical current, I_c, distribution, where the high-density critical current, $j_c^p(U)$, flows only through some part αS of the geometrical cross section S of a film. In this model the total critical current is given by

$$I_c \equiv j_c S = \alpha j_c^P(U) + (1-\alpha) S j_c^J(B) \; . \tag{1}$$

In Eq. (1), j_c^J is the part of the critical current density, which is sensitive to weak magnetic field in contrast to j_c^P. It flows through another area of the film cross section $(1-\alpha)S$. In this approach α is the important factor controlling j_c and obviously depending strongly on the microstructure of a film. For the best films $\alpha \approx 1$, and $j_c \approx j_c^P(U)$ limited by $U = U(T)$ is $\simeq 6\times 10^6$ A/cm^2 at $T = 78$ K, $B = 0$. The importance of the film microstructure is confirmed by numerous investigations showing large depressions in j_c at any deviation from perfect epitaxial growth, arising from bad film-substrate lattice matching or other nonoptimal growth conditions. A direct correlation between j_c and the angular distribution width, Δ, of film grains has been pointed out for epitaxial YBCO films on monocrystal substrates.[6] Also data of transport properties across single grain boundaries[7] demonstrate the large influence of the neighboring grains misorientation angle on critical current values.

The aim of this study, which is a continuation of previous work,[5,8] is to elucidate the correlation and type of interrelation between structural and transport characteristics of YBCO/SrTiO$_3$ (100) epitaxial films. We first show the dependences of j_c, α, and j_c^J on the misorientation angle Δ and then demonstrate that Δ is the most important parameter controlling j_c.

II. EXPERIMENTAL PROCEDURES

The epitaxial c-axis-oriented Y-Ba-Cu-O films of 0.1–0.3 μm thickness have been prepared by pulsed laser deposition[8] on commercial substrates of SrTiO$_3$ (100). Each sample represented a large-square film (~ 2 cm^2), where one part of the surface was used for structural analyses and on another part 15–20 microbridges were fabricated by a photolithographic process and by ion etching for current measurements. Contacts were made using evaporated gold.

The film structure, thickness, and composition were analyzed by x-ray diffraction, Rutherford backscattering spectroscopy (RBS), and ion channeling. Especially, rocking curves determining the angular distribution of domains in films were measured with the detector set for the detection of the (005) YBCO line.

The temperature dependence of the resistivity, $\rho(T)$, and the critical current measurements were carried out on bridges having a width of 20 μm and length in the range of 200–1000 μm applying the four-probe technique. An external magnetic field up to 10 mT transversal to the film plane was produced by a coil. All critical current measurements of this work were performed at $T = 78$ K.

An electrical-field criterion of $E(j_c) = 10$ μV/cm was used for j_c determination. For the j_c range under investigation ($j_c = 3\times 10^5$–5×10^6 A/cm^2) this criterion corresponds to resistivity levels, $\rho \equiv E/j$, of 3.0×10^{-11}–1.6×10^{-12} Ω cm, where the VCC are sufficiently exponential, $E \sim \exp(j/j_1)$.[5] Due to the large factor of the exponent at $j \approx j_c$ ($j_c/j_1 \approx 50$ for all samples) the range induced deviations in j_c determination in the whole j_c range are not larger than 5%.

For a detailed study of j_c as a function of structural properties described, e.g., by the misorientation angle of crystalline grains, Δ, it is necessary to vary Δ in a reasonable range, i.e., over a few degrees. This is difficult to achieve on high-quality single-crystalline substrates without degrading other film properties like T_c because significant changes of the growth parameters leading to the desired structural changes normally are accompanied by T_c reduction.[9]

Therefore, in this study we used (100) SrTiO$_3$ substrates consisting of low-angle misoriented blocks. The angular distribution of these blocks delivered the desired variation of structure in terms of Δ without affecting T_c because on each single block perfect film growth has been observed. In addition, such substrates may gain importance in practical research and applications. As a reference point, we used an YBCO film on a standard single-crystalline substrate.

III. RESULTS AND DISCUSSION

The films applied in our investigations were c axis textured ($c\perp$ substrate surface). As an example, we show in Fig. 1 an x-ray-diffraction diagram revealing only $(00l)$ lines. The average composition of the films was studied by RBS. It is close to typical for films with 1:2:3 structure. For all films the temperature dependence of the resistivity, $\rho(T)$, was linear for $T > 120$ K with a characteristic slope $\gamma = \rho(300\text{ K})/\rho(100\text{ K})$ of 2.5–3.1 and resistivities $\rho(100\text{ K})$ of typically 100 $\mu\Omega$ cm. For the best films $\rho(100\text{ K})$ was 80 $\mu\Omega$ cm and $\gamma = 3.1$. Zero-resistance temperatures T_c were in the range of 89.7–90.5 K with $\rho(T_c) = 10^{-2}$ $\rho(100\text{ K})$. The critical current-density values j_c (78 K, $B = 0$) revealed a large spread both between different samples and between different bridges patterned on the same film. The j_c variations were in the range for about 3×10^5–5.3×10^6 A/cm^2 with no change in T_c.

A number of $j_c(B)$ measurements were carried out for

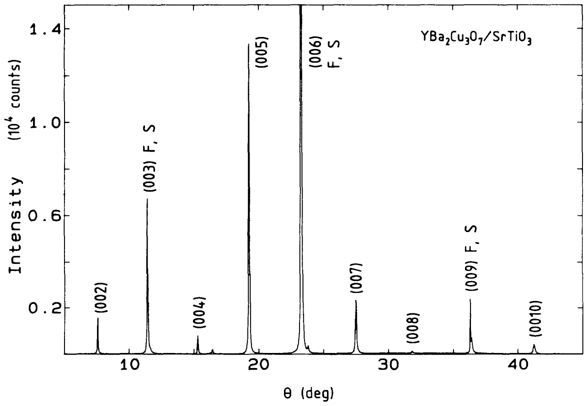

FIG. 1. X-ray diffraction diagram of an Y-Ba-Cu-O film deposited by laser ablation on (100) SrTiO$_3$.

bridges having different j_c values, and typical relationships for $j_c(B)$ are given in Fig. 2. In general, the $j_c(B)$ curves consisted of some decreasing part at small fields $B < 2$ mT, and then, for $B > 10$ mT, j_c were practically independent of B. In our I_c model the $j_c(10\text{ mT})$ value was the first term of Eq. (1) and the difference $j_c - j_c(10\text{ mT})$ corresponded to the field-dependent part of the critical current. Equation (1) predicts an increase of this part of j_c for $\alpha \rightarrow 0$, i.e., for low $-j_c$ bridges, and Fig. 2 clearly shows this relation. The films having the highest j_c values above 3×10^6 A/cm^2 were practically insensitive to low magnetic fields, but for $j_c \le 10^6$ A/cm^2 films a decrease of j_c was clearly observed for $B < 2$ mT. For j_c values below 4×10^5 A/cm^2 the $j_c(B)$ characteristics changed drastically: the critical current becomes very sensitive to the magnetic field, and j_c drops by more than one order of magnitude, but an independent on the field part of j_c still existed for $B > 2$ mT. Such bridges are sensitive to the earth's magnetic field; to reduce its influence, the film plane was oriented parallel to this field direction before j_c measurements.

The $j_c(B)$ dependences of our low-j_c samples are similar to those observed before in polycrystalline films[10] and bulk material.[11] It has been shown that the magnetic properties of polycrystals can be understood on the basis of a model consisting of high superconducting grains connected by "Josephson weak links"[12] The nature of these links (*SIS*,*SNS*,*SS'S*,...) is not yet clear, but it has been shown that, for a variety of barriers between grains the Josephson relation, $j \sim \sin\phi$ holds. An isolated thin-film grain boundary revealed the characteristics of a resistively shunted Josephson junction (7). Our data of Fig. 2 show the crossover between $j_c(B)$ characteristics of high-j_c epitaxial and low-j_c films (which are polycrystalline as we show later), and it is reasonable to assume the same physical mechanism for the j_c sensitivity to low magnetic fields, i.e., Josephson-like behavior, as discussed before. An experimental indication for the rise in granularity connected to such a behavior is the hysteretic behavior of $j_c(B)$. The $j_c(B)$ curves were measured after zero-field cooling in increasing magnetic field. The high-j_c curves are reversible, but the low-j_c ones reveal hysteresis as shown in Fig. 2 by the reverse j_c track after initial sweeping up to $B = 10$ mT. The reverse track shows a 30% increase of j_c at $B = 1$ mT but is limited at $B = 0.1$ mT, with a value 10% lower than the initial one. This type of hysteresis is known for polycrystals and related to a compensation of the external magnetic field by the shielding fields of the intergrain currents. This leads to an increase of the intergrain Josephson weak-link critical current.[13]

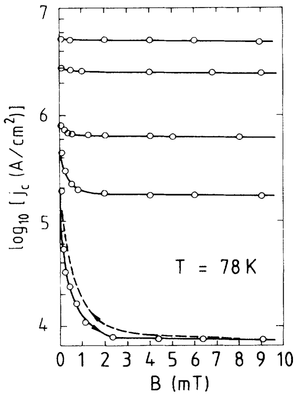

FIG. 2. Critical current density j_c vs transversal magnetic field for bridges with different $j_c(B=0)$ values. $T_c = \text{const} \approx 90$ K. The high-j_c curves are reversible, while hysteresis is observed in low-j_c films as indicated by the dashed line in the lower curve.

The interrelation between both parts of the critical current, j_c^P and j_c^J, is presented in Fig. 3(a), where the relative fraction of the Josephson-like current $[j_c - j_c(10\text{ mT})]/j_c$ is plotted versus the total j_c value. For the best films this fraction does not exceed a value of about 0.03, but for bridges with $j_c < 10^6$ A/cm^2 quickly amounts to 1. This behavior is in accordance with our model [Eq. (1)] and led us to use the following relations for both parts of j_c:

$$j_c - j_c(10\text{ mT}) \equiv (1-\alpha) j_c^J , \quad j_c(10\text{ mT}) = \alpha j_c^P . \qquad (2)$$

Because U does not depend on j_c, as was shown in Ref. 5, a constant $j_c^P(U)$ value of 6×10^6 A/cm^2 for $T = 78$ K and $B = 0$ may be accepted, which is in accordance with data measured in the best films $[\alpha = 1, j_c \simeq j_c^P(U)]$.[6,14] Then, from the $j_c(B)$ curve and relations (2), the $\alpha(j_c)$ and $j_c^J(j_c)$ dependences may be obtained separately [Figs. 3(b) and 3(c)]. The $\alpha(j_c)$ relationship shows that α is the main factor controlling the critical current of high-j_c films, i.e., α is proportional to j_c and the contribution of the Josephson-like current is small [the dashed line in Fig. 3(b) corresponds to the $j_c^J = 0$ case]. For low-j_c films, $j_c \le 5\times 10^5$ A/cm^2, the critical current is carried via weak links mainly, and α approaches zero for $j_c < 2\times 10^5$ A/cm^2.

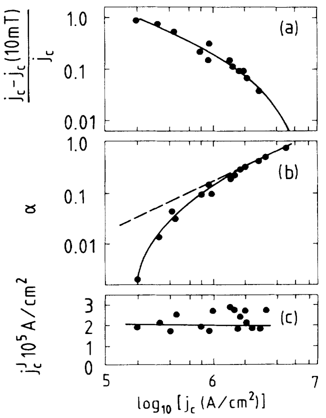

FIG. 3. Fraction of critical current flowing through weak links (a); fraction of geometrical film cross section, carrying high-density critical current $j_c^P \simeq 6\times 10^6$ A/cm^2 (b); density of critical current of weak links (c) as a function of the averaged critical current density $j_c \equiv I_c/S$ at $B = 0$. Hard lines—simulation by Eq. (3). The dotted line of (b) corresponds to the $j_c^J = 0$ approximation.

The $j_c^J(j_c)$ dependence [Fig. 3(c)] unexpectedly shows an almost constant j_c^J value of about 2×10^5 A/cm^2 for all films. This means that Eq. (1) may approximately be written as

$$j_c(\alpha)=\alpha(6\times10^6)+(1-\alpha)(2\times10^5)\ (\mathrm{A/cm^2})\ . \quad (3)$$

The $j_c(\alpha)$ relationship given by Eq. (3) is shown in Fig. 3(b), and $(1-\alpha)j_c^J/j_c=f(j_c)$ in Fig. 3(a). The solid lines are in good agreement with experimental points. This result demonstrates the applicability of the model of Eq. (1) for j_c description, and α as a main factor, which controls j_c [Eq. (3)]. The physical interpretation of α suggests the existence of uninterrupted channels in the film volume able to carry a current limited by U only. This channel quantity α obviously depends strongly on film grain boundary structure, which may be influenced by their misorientation angle Δ.

In this work a relationship between α and Δ was obtained for Δ in the range of 0.5°–7° by using (100) $SrTiO_3$ substrates, which consisted of low-size blocks misoriented at small angles. Such substrates allowed a variation of the film grain structure at optimum deposition conditions without affecting T_c.

In Fig. 4 rocking curves are presented of the (200) $SrTiO_3$ peak ($2\theta=46.32°$) and of the (005) YBCO line ($2\theta=38.50°$), which characterize correspondingly the angular distributions of the crystallite blocks in the substrate and of the *c*-axis-oriented YBCO grains. The multiblock structure of the substrate is visible with peak widths below 0.1 degree for one block and a total distribution width of about 3°. The YBCO film angular grain distribution reflects the substrate blocks orientation completely, having the same total width. For the parts of a film occupying one block, the rocking curve width may be estimated to a value of about 0.4°. For an estimation of a block size in comparison to a bridge length the curves of Fig. 4 are shown for reduced size of 50×100 μm^2 of the incident x-ray beam. There is no significant change of the width of the distribution, and it is clear that numerous blocks are contained on one bridge length.

To obtain a $j_c(\Delta)$ dependence the rocking curve was compared to a histogram of the j_c value distribution for the different microbridges, produced on one film. The results are shown in Fig. 5. Three typical kinds of rocking curves and corresponding histograms are presented. For the first case (a) (reference film on single crystal substrate) the film's angular grain distribution is narrow with a width Δ of 0.6° and the j_c distribution concentrates near $j_c\sim5\times10^6$ A/cm^2 with a small spread of 30%. For misoriented grains, film (b), the larger width Δ in the range of 2°–4° correlates with a j_c distribution shift to values in the range of $(1-3)\times10^6$ A/cm^2 and with an increasing j_c spread. To obtain the j_c-versus-Δ dependences the most probable j_c values from histograms were used in cases (a) and (b). Because the large-angle misoriented grains most seriously influenced the bridge critical current, the full widths of the corresponding rocking curves were used as Δ. Some films, of the third kind (c), have large misoriented grains with Δ values of about 6°–7°, in addition to the main distribution. The appearance of a tail in the angular distribution correlates with an essential increase of the width of the j_c distribution due to the presence of bridges with low-j_c values. We therefore assigned such low-j_c values to large misoriented grains.

The $j_c(\Delta)$ dependence (Fig. 6) obtained from the rocking curves and j_c histograms, shows an exponential de-

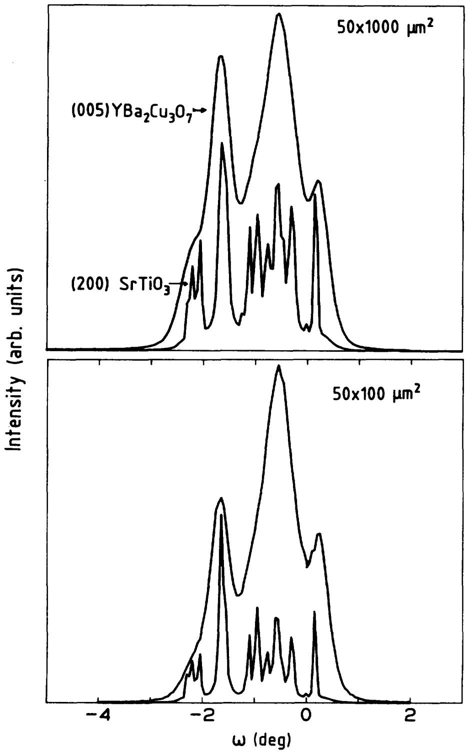

FIG. 4. Rocking curves (ω scans) of an Y-Ba-Cu-O film on $SrTiO_3$ substrate. The scans were taken for the (005) line of Y-Ba-Cu-O and (200) line of $SrTiO_3$. The curves are shown for different sizes of the incident x-ray beam.

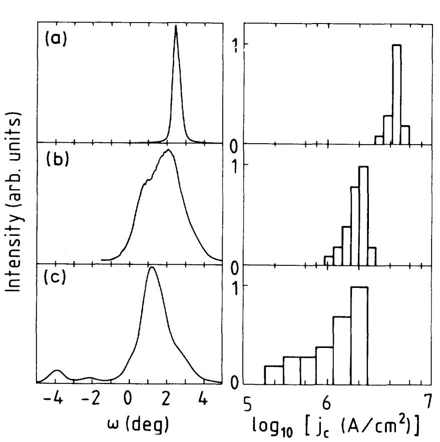

FIG. 5. Left: typical rocking curves of Y-Ba-Cu-O films produced on a single-crystal substrate (a) and on low-size misoriented blocks substrates (b and c). Right: corresponding histograms of the critical current density distributions.

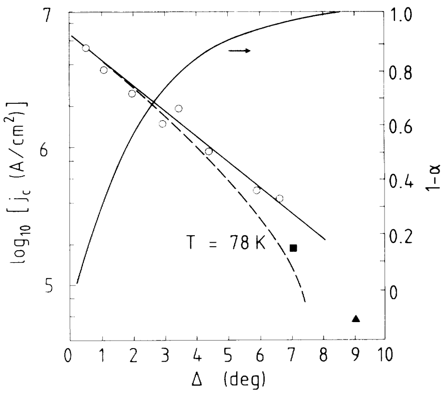

FIG. 6. Critical current density measured in zero magnetic field (left scale) and fraction of cross section occupied by weak links (right scale) vs angle of film grain misorientation. Dotted line: critical current density in external magnetic field of 10 mT. Black square and triangle: single-grain boundary critical current from Ref. 7 for laser-ablated and e-gun-evaporated films, respectively.

crease of j_c with increasing misorientation angle Δ. The exponential relationship can be described by

$$j_c(\Delta)=j_c(0)\exp[-\Delta(\text{deg})/2.4] \tag{4}$$

with $j_c(0)=7\times10^6$ A/cm^2, which is close to the value of j_c^P. A clear depression of j_c is observed for misorientions $\Delta>2°$; we estimate a misorientation threshold value of about 1°. Using the $\alpha(j_c)$ relation [Fig. 3(b)] we obtained a dependence of the weak-link quantity on the misorientation angle, $(1-\alpha)=f(\Delta)$, which is shown in Fig. 6. For $\Delta>4°$ more than 80% of a film cross section is occupied by weak links, carrying an essential part of the critical current at $B=0$ only. Correspondingly, the $j_c(\Delta)$ dependence for $B=10$ mT (dashed line in Fig. 6) reveals a sharper decrease of j_c for $\Delta>5°$.

For misorientation angles $\Delta\geq7°$ all critical current flows through weak links, and the j_c value of 2×10^5 A/cm^2 may be compared well with the data of Ref. 7 for the critical current of a single grain boundary of misoriented YBCO grains. The data for a laser-ablated film in Ref. 7, however, were given for a misorientation angle of 24°. For a direct comparison with our laser-ablated films, we have recalculated the $\Delta=24°$ results to a misorientation angle of 7° by using the following relationships: $j_c(\Delta=7°)/j_c(\Delta=24°)\approx10$ (Fig. 11 of Ref. 7) and $j_c(T=4.2\text{ K})/j_c(78\text{ K})\approx5$. This results in a j_c value of 2×10^5 A/cm^2 at 78 K. This j_c point is included in Fig. 6 and agrees with our data. The j_c value for a $\Delta=9°$ boundary and e-gun-evaporated films from Ref. 7 is slightly lower than extrapolated from our findings.

To estimate the homogeneity of the j_c^J current flow we must compare the Josephson penetration depth λ_J with the grain size of our films, which is supposed to be a characteristic dimension of weak links. A homogeneous current flows in weak links if their width is smaller than $2\lambda_J$. For λ_J we have

$$2\lambda_J\approx(\hbar/2e\mu_0 j_c^J\lambda)^{1/2}\ . \tag{5}$$

λ is the London penetration depth, and μ_0 is the magnetic permeability. For a magnetic field aligned with the c axis of a film, the λ value at 78 K is about 3×10^{-5} cm. Then, from Eq. (5) using j_c^J of 2×10^5 A/cm^2 we find $2\lambda_J\simeq1$ μm. This is comparable with the grain size of our films, and a nearly homogeneous current flow may be expected. On the other hand, the width of a weak-link loop may be obtained from the $j_c(B)$ data of Fig. 2. A characteristic field H_1 of about 15 Oe for j_c depression leads to a width value $\sim\phi_0/2H_1\lambda\approx2$ μm (ϕ_0 is the flux quantum), which is of the same order of magnitude as above.

In conclusion, we have shown the possibility of describing j_c values and $j_c(B)$ dependences in an approach, summarized in Eq. (1), where the main parameter is $\alpha=\alpha(\Delta)$. $(1-\alpha)$ is given in Fig. 6 as a function of grain misorientation angle Δ. This model naturally includes the bulk material case, where $\alpha=0$ and S means only some part of geometrical cross section having a nonzero $j_c(B=0)$ value. From the viewpoint of applications, the reported data show the origin for j_c decrease in long current-carrying lines and underline the necessity of high-quality epitaxial films for large-scale devices.

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