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Transport properties of low angle grain boundaries in $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films at high magnetic fields

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The critical current density of grain boundaries and grains in $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films has been measured in magnetic fields up to 10 T. As a function of temperature the critical current densities across 8° [001]-tilt boundaries display a maximum at ≈15–20 K for $0.04 \leqslant x \leqslant 0.2$ and fields of several teslas. Opposite to the behavior of large angle grain boundaries, calcium doping is found not to enhance the critical current densities of low angle grain boundaries ($\theta \leqslant 8°$). © *2006 American Institute of Physics*. [DOI: 10.1063/1.2190460]

The decrease of the critical current density (J_c) across grain boundaries in high temperature superconductors as a function of the boundary angle[1] is the major difficulty to be solved for the realization of competitive cables from these materials. The most promising technology, coated conductors,[2–7] reduces the deleterious effects of large angle grain boundaries on J_c by aligning the grains to a very few degrees along all major crystal axes. In addition, the critical current density of high-T_c cables may also be increased by using grains with large surface areas[8,9] or by doping the grain boundaries.[10–15] As was shown, the critical current density of [001]-tilt grain boundaries in $YBa_2Cu_3O_{7-\delta}$ can be substantially increased at small magnetic fields by partially substituting Y^{3+} by Ca^{2+}.[10] To clarify whether doping also enhances the critical current density in large fields, as required for the use of coated conductors in magnet applications, we performed systematic studies of the transport properties of low angle grain boundaries in fields up to 10 T.

The films were grown by pulsed laser deposition from polycrystalline $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ targets onto single crystalline and bicrystalline $SrTiO_3$ substrates. After deposition at 760 °C in 0.25 mbar of oxygen the samples were cooled within 1 h to 400 °C in an oxygen atmosphere of 0.4 bar and, after holding this temperature for 20 min, to room temperature. The samples were patterned by photolithography and wet etching into standard four-point configurations with 20 μm wide tracks straddling the grain boundaries and into 6 μm wide tracks located in the grains. The heights and the widths of the tracks were determined by scanning force microscopy. The critical currents were derived from the current voltage characteristics, using a criterion of 10 μV. The current densities (J) were obtained from the ratios of the critical currents and the cross sectional areas of the tracks.

In magnetic fields of several teslas, the doped grain boundaries reveal an unusual behavior: By warming the samples from 4.2 to 20 K the critical current density increases. This is illustrated in Fig. 1. The graph displays the J-V characteristics of a $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{7-\delta}$ film grown on a $SrTiO_3$ bicrystal with an 8° [001]-tilt boundary. This zero field cooled measurement was done in a magnetic field of 10 T, which was applied in the boundary plane, parallel to the c axis. As shown by Fig. 1, $J_c(20\text{ K})=0.31\text{ MA/cm}^2$ exceeds $J_c(4.2\text{ K})=0.21\text{ MA/cm}^2$ by a factor of 1.5. This phenomenon was reproducibly observed on all boundary bridges in the sample and was reproduced by other samples.

To analyze the origin of this effect, we studied a set of doped $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films (x=0, 0.04, 0.06, 0.10, 0.20, and 0.30) with 8° [001]-tilt boundaries. Figure 2 displays representative results of these experiments. While the undoped $YBa_2Cu_3O_{7-\delta}$ samples show a standard $J_c(T)$ dependence in high magnetic fields, an addition of 4% calcium causes a depression of J_c at low temperatures. With increasing calcium content, a gradual improvement of J_c is achieved. The critical current density of the sample doped with 30% calcium finally exceeds the J_c of the $YBa_2Cu_3O_{7-\delta}$ sample for $T<15$ K. Maxima of the $J_c(T)$ dependence occurring at ≈15–20 K are observed for $0.04 \leqslant x \leqslant 0.20$.

Which mechanism causes this effect? To answer this question, we note that the magnetic field at the boundary differs from the applied magnetic field due to flux focusing.[16,17] In the experiments, the samples were cooled in zero magnetic field (zfc). When the sample is superconducting and the field is increased, vortices enter first the grain boundary and then the grains. Flux is trapped close to the boundary and the field is focused into the boundary region. As a consequence, the flux density in the boundary exceeds the applied field by the flux focusing factor.[16] Since strong pinning results in a high flux density in the interface, it is expected that at low temperatures, where the pinning is strongest, the flux density in the boundary is largest. Since J_c

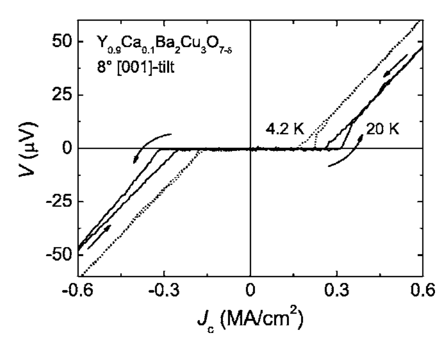

FIG. 1. Current density-voltage characteristics of a $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{7-\delta}$ film grown on a $SrTiO_3$ bicrystal with an 8° [001]-tilt grain boundary. The characteristics were measured at 4.2 and at 20 K in a magnetic field of 10 T (zero field cooled) applied in the boundary plane, parallel to the c axis of the film.

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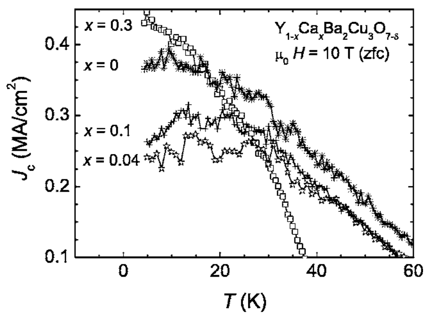

FIG. 2. Critical current density measured for several doped $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films grown on $SrTiO_3$ bicrystals with 8° [001]-tilt grain boundaries as a function of temperature. The data were taken at 10 T (zero field cooled), with the field being applied parallel to the c axis.

decreases with increasing flux density, it is possible that J_c becomes smaller with decreasing temperature.

To test this idea, we measured the $J_c(T)$ dependence of the bicrystals at 10 T in the field cooled state. If the magnetic field is applied in the normal state, a homogeneous flux distribution is achieved, which is conserved upon cooling through T_c. Thus, if flux focusing was the main reason for the maximum in the $J_c(T)$ dependence, the maximum is not expected to be present in the field cooled experiment. Figure 3 shows the $J_c(T)$ dependencies of doped and undoped samples with 8° grain boundaries at 10 T for both cooling modes. The maximum in the $J_c(T)$ dependence vanishes indeed in the field cooled experiment as expected. Therefore we conclude that the maximum is caused by flux focusing.

Why does calcium doping enhance the pinning in the grain regions adjacent to the grain boundary? According to most models describing the transport properties of grain boundaries in high-T_c superconductors, depletion layers with a reduced density of mobile carriers are formed near the boundary due to band bending or due to defects in the oxygen lattice. In these depletion layers the order parameter is

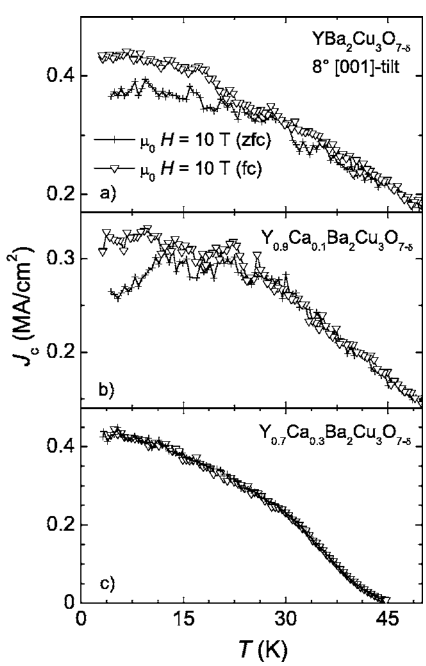

FIG. 3. Critical current density measured as a function of temperature for several doped $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films with 8° [001]-tilt boundaries. The data were taken at 10 T, zero field cooled (zfc) and field cooled (fc).

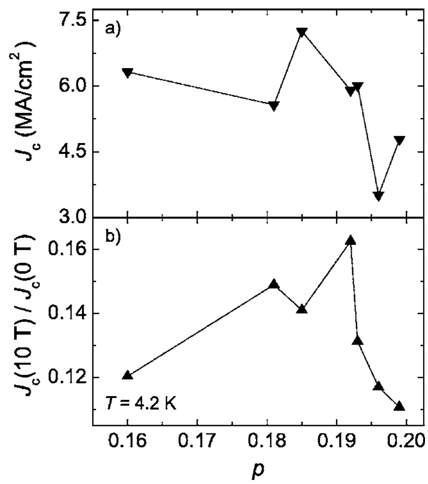

FIG. 4. Critical current density measured at 4.2 K and 10 T as a function of the number of holes per planar copper (p) in $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films (intragrain measurements).

depressed and pinning is correspondingly reduced. Calcium doping enhances the carrier density, resulting in a better doped superconductor close to the grain boundary and improved flux focusing. In $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ films with $x=0.3$ the large calcium concentration reduces the intragrain J_c. In these films the pinning appears to be weaker. This is also evident from Fig. 3(c), which shows that the zero field cooled and the field cooled J_c across an 8° grain boundary are identical for $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$. Thus, the maximum is expected to be most pronounced for moderately doped films.

It has been pointed out that the doping concentration causing the largest pinning is not the one that leads to a maximum T_c.[18,19] This effect is ascribed to the closing of the pseudogap at a concentration of 0.19 holes per planar copper (p).[18,19] According to that model the condensation energy and hence both the flux pinning and the critical current density are largest at $p=0.19$, which is in the overdoped part of the phase diagram. Are our data consistent with that model? Figure 4 shows the critical current density of the grains of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ at 10 T as a function of holes per planar copper. The hole concentration was estimated via T_c using the empiric equation $T_c=T_{c,\max}[1-82.6(p-0.16)^2]$.[20] As shown by Fig. 4(a), doping does not cause a clear peak of J_c at $p=0.19$, 4.2 K, and 10 T. But intriguingly, if $J_c(10\text{ T})$ is normalized to $J_c(0\text{ T})$, as plotted in Fig. 4(b), a clear maximum is found at $p=0.19$. The reason why the maximum

TABLE I. Critical current densities of $YBa_2Cu_3O_{7-\delta}$ and $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ films at 0 and 10 T measured at 4.2 K (zero field cooled). For some misorientation angles several bridges were measured on a chip. In this case the data of a characteristic bridge are given.

	$YBa_2Cu_3O_{7-\delta}$		$Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$	
θ	$J_c(0\text{ T})$ $\left(\frac{A}{cm^2}\right)$	$J_c(10\text{ T})$ (zfc) $\left(\frac{A}{cm^2}\right)$	$J_c(0\text{ T})$ $\left(\frac{A}{cm^2}\right)$	$J_c(10\text{ T})$ (zfc) $\left(\frac{A}{cm^2}\right)$
0°	5.3×10^7	6.3×10^6	3.0×10^7	3.5×10^6
4°	1.8×10^7	4.2×10^6	2.1×10^7	3.9×10^6
8°	8.7×10^6	3.5×10^5	7.1×10^6	4.1×10^5
16°	5.5×10^5	1.1×10^4	2.3×10^6	6.4×10^4
24°	8.73×10^5	4.6×10^3	1.12×10^6	2.07×10^4

appears in $J_c(10\ \mathrm{T})/J_c(0\ \mathrm{T})$ but not in $J_c(10\ \mathrm{T})$ remains to be revealed.

Does calcium doping increase the critical current density of the grain boundary in large magnetic fields? The results of our studies are listed in Table I. A clear enhancement of J_c is found for the 16° (by a factor of 4 in 0 T and 6 in 10 T) and 24° (by a factor of 1.3 in 0 T and 4.5 in 10 T) boundaries investigated. Boundaries with 4° or 8° misorientation do not show a significant improvement.

In summary, calcium doping was found not to enhance the critical current density of low angle grain boundaries ($\theta \leqslant 8°$) in large magnetic fields. The $J_c(T)$ dependencies of 8° grain boundaries of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films ($0.04 \leqslant x \leqslant 0.20$) are characterized by a maximum at $\approx 15-20$ K in magnetic fields of 10 T. We conclude that this maximum is caused by doping dependent flux focusing.

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