Crossover effects in the temperature dependence of the critical current in $YBa_2Cu_3O_{7-\delta}$

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Temperature dependence of the critical current I_c was investigated in thin films and ceramics of $YBa_2Cu_3O_{7-\delta}$ (YBCO) over a temperature range 10–90 K and in magnetic fields up to 700 G. The measurements were performed using superconducting rings in a persistent mode. A scanning axial Hall probe was used to record the profile of the magnetic field across the ring, generated by the persistent current at the critical level. The magnitude of I_c was inferred from the magnitude of the persistent current's self-field in the center of the ring. This technique eliminated the contribution of normal currents to the measured value of I_c and allowed us to distinguish between depairing and depinning critical currents. The results revealed the crossover between an Ambegaokar-Baratoff-like temperature dependence of the critical current to a Ginzburg-Landau dependence. The crossover was observed for both depairing and depinning critical currents in *c*-axis-oriented YBCO thin films during reduction of T_c from 91 down to 87 K and in a ceramic YBCO upon application of small magnetic fields. The experimental data imply the presence of superconducting microdomains of an effective diameter of about 30–40 Å, coupled by Josephson tunnel junctions, inside the grains and in the intergrain microbridges of YBCO, in agreement with the Clem's model for $I_c(T)$ in strongly coupled granular superconductors [Clem *et al.*, Phys. Rev. B **35**, 6637 (1987)]. The size of these domains decreases with a decreasing oxygen content and with an increasing applied magnetic field. [S0163-1829(96)08121-0]

I. INTRODUCTION

It is well known that the supercurrent density in a superconductor is limited by depairing and magnetic flux-depinning processes.[1,2] In the former case, the magnitude of the critical current (depairing critical current) is determined by pair breaking and subsequent suppression of the order parameter of the superconductor. From the London theory, the depairing critical current density equals the current density at which the kinetic energy of the charge carriers equals the condensation energy. In the latter case, the magnitude of the critical current (depinning critical current) is determined by magnetic flux movement and pinning forces. If the flux lines move in response to the Lorentz force exerted by the current, a voltage is induced in the superconductor, according to the Maxwell equations. The superconductor can support the current if the flux lines are held in place by pinning forces (and also by interactions with the rest of the flux-line lattice).

The measurements of temperature dependence of the critical current $I_c(T)$ in YBCO thin films, single crystals, and ceramics have been carried out in order to distinguish between different models of the supercurrent limiting mechanism. The models employed in order to explain the behavior of the intragrain $I_c(T)$ were those of Tinkham[3] (which is based on the Anderson-Kim flux-creep critical-state model) and of Ginzburg and Landau.[4] The intergrain $I_c(T)$ was interpreted using theories developed by Ambegaokar and Baratoff,[5] DeGennes,[6] and Likharev.[7] Currently, there is much less experimental data for $I_c(T)$ in YBCO over a wide temperature range than for $I_c(B)$ measured at fixed temperature. The reason for this are the experimental difficulties (like heating effects) in measuring high critical currents at low temperatures on bulk samples, exhibited by standard four-probe *I*-*V* and ac-magnetic induction methods. Therefore, most measurements of $I_c(T)$ over a temperature range 4–90 K were done on YBCO thin films and on thin-film YBCO/YBCO junctions. Regarding the intragrain critical current, the experimental data for YBCO thin films revealed three different dependencies of I_c on temperature, which are characterized by a ''convex'' curvature[8,9] (characteristic of Ambegaokar-Baratoff-type temperature dependence), a quasilinear behavior,[8–12] and ''concave'' curvature (Ginzburg-Landau-like dependence).[8,9,12–16] The data of Mannhart *et al.*[8] showed the transition from a quasilinear temperature dependence of I_c at zero-magnetic field to a concave temperature dependence upon application of high magnetic fields of 0.5–1.0 T. The crossover from quasilinear to concave temperature dependence was also seen by Jones *et al.*[12] upon reduction of the oxygen content. In most cases, Tinkham's model[3] for a flux-creep-limited $I_c(B,T)$ has been applied in order to explain a convex- and a quasilinear-like temperature dependence of I_c. Concave behavior of $I_c(T)$ was in general interpreted as due to the presence of an intragrain SNS weak-link structure. Regarding the intergrain critical current, the available experimental data showed a convex temperature dependence of I_c across a grain boundary of thin-film bicrystals of YBCO (Ref. 8) and a concave one across YBCO/YBCO thin-film junctions containing a $YBa_2(Cu_{1-x}Co_x)O_{7-\delta}$ normal layer.[17] In the former case, it has been suggested that the results are consistent with Likharev's model[7] which predicted $I_c(T)$ for dirty *SNS* junctions. However, the possibility that $I_c(T)$ can be described by Ambegaokar-Baratoff's theory[5] for *SIS* junctions has not been ruled out. In the latter case, it has been claimed that the conventional proximity effect of De Gennes,[6] modified by pair-breaking scattering, predicts the observed $I_c(T)$. The available data for $I_c(T)$ in YBCO single crystals are those of Gough *et al.*,[18] who measured the transport $I_c(T)$ over a temperature range 4–70 K on YBCO single-crystal rings using

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ac hysteresis loops, and those of Thompson *et al.*[19] (in a field of 1 T parallel to the c axis) and Abulafia *et al.*,[20] who measured the magnetization $I_c(T)$ over a temperature range 3–77 K. The results show a concave curvature of $I_c(T)$.

The experimental fact that both the intragrain and intergrain critical currents exhibit similar dependencies on temperature, and a variety of theories that have been applied to interpret the same behavior of $I_c(T)$, imply a lack of consensus about supercurrent limiting mechanism in YBCO for both intragrain and intergrain transport currents.

We therefore decided to investigate the temperature dependence of the critical current in c-axis-oriented thin films of YBCO characterized by various superconducting transition temperatures, and in ceramic YBCO as a function of applied magnetic field. The experiments were performed using a nonstandard technique, which was applied earlier to study critical currents in ceramic YBCO (Ref. 21) and decays of persistent currents in melt-textured c-axis-oriented YBCO (Ref. 22). This technique is based on measurements (with a scanning Hall probe) of the profile of the magnetic field generated by a self-supporting (persistent) current flowing in a ring. The magnitude of the maximum (critical) persistent current was inferred from the magnitude of the current's self-magnetic field measured at the ring's center. The method allowed us to eliminate unwanted contributions of normal currents to the measured temperature dependence of the critical currents and to distinguish between depairing and depinning critical currents. The results revealed the crossover from Ambegaokar-Baratoff-like (AB) to Ginzburg-Landau (GL) temperature dependence of I_c for both intragrain and intergrain currents. The experimental data were interpreted using Clem's theory[23] of the Ambegaokar-Baratoff to Ginzburg-Landau crossover effects on $I_c(T)$ of granular conventional superconductors, and Kresin's theory[24] of gaplessness in high-temperature superconductors.

II. EXPERIMENTAL PROCEDURE

A. Sample preparation

Two types of YBCO materials were used in these studies: oriented epitaxial thin films (~100–300 nm thick) and bulk ceramics [YBCO and YBCO/Ag (2 wt %) composite]. YBCO thin films were deposited on (100) $SrTiO_3$ and $LaAlO_3$ (10×10 mm) substrates by two methods: off-axis rf magnetron sputtering and laser ablation from stoichiometric $YBa_2Cu_3O_{7-\delta}$ targets of 99.999% purity. The magnetron sputtering was performed using 50-W rf power at a total pressure of 100–120 mTorr in 1:1, 1.5:1, and 5:1 oxygen-argon mixtures. The substrate temperatures were kept between 650 °C and 730 °C for $LaAlO_3$ and between 700 °C and 800 °C for $SrTiO_3$. The deposition temperatures correspond to those which are required to achieve less than 20% of a-axis growth (alignment) in the c-axis-oriented YBCO films according to studies done by Jeschke *et al.*[25] Laser-ablated YBCO thin films were deposited at 800 °C and 300-mTorr oxygen pressure. Our measurements of a-axis alignment versus c-axis alignment using x-ray diffractometry indicated the presence of less than 5% of a-axis alignment in most thin films. T_c of the obtained c-axis-oriented, twinned thin films was kept within a range between 83 and 91 K. A standard photolithography technique was applied before etching thin-film rings of outer and inner diameters of 8.5 and 5.0 mm, respectively.

YBCO bulk ceramics were prepared by the standard solid-state reaction method from high-purity (99.999%) compounds. Calcining of YBCO was performed at 925 °C for 24 h in air or oxygen, followed by single or double sintering in oxygen at 920 °C–930 °C. For $YBa_2Cu_3O_{7-\delta}$/Ag (2 wt %) composite, silver was added during a single sintering process. By varying the number of sintering processes and the type of gas during calcining we produced ceramics of low and high intergrain critical current density and the same intragrain T_c of the order of 90–91 K. Ceramic YBCO rings were prepared in a manner described in Ref. 21. The outer and inner diameters of the rings were 15–16 mm and 6 mm, respectively. Two narrow constrictions were cut along diameter of the 3.0–3.5 mm thick rings, forming bridges of cross-sectional area of 3.0–3.5 mm^2.

B. Critical current measurement procedure

In order to measure the temperature dependence of the critical current in YBCO thin films and the intergrain critical current in YBCO ceramics over a temperature range 10–90 K, we used an unconventional technique, which has been developed to investigate intergrain junctions and flux pinning in YBCO ceramics (Ref. 21). The magnitude of the critical current was inferred from the magnitude of the axial component of the magnetic field generated by the maximum self-supporting supercurrent circulating in a ring-shaped sample. The persistent current was induced in the ring by applying and subsequently switching off the external magnetic field generated by a solenoid along the axis of the ring (c axis). The profile of the current's self-magnetic field was recorded with a scanning Hall probe at a constant distance from the sample. Figure 1(a) shows the profiles of the magnetic field due to persistent currents of various magnitudes, flowing in a zero-field-cooled YBCO thin-film ring. The profiles have a very symmetrical bell-like shape. It should be pointed out here that the axial component of the persistent current's self-field has a single maximum in the ring's center, whereas a field due to magnetic vortices trapped in the ring's bulk exhibits a profile with two maxima above the ring's bulk and a minimum in the ring's center. It is, therefore, possible to distinguish the field produced by trapped vortices from that due to a circulating persistent current. The experiments were designed to minimize contribution of trapped vortices to the self-field of the persistent current. Persistent currents were generated in the ring, starting at low applied magnetic fields [Fig. 1(a)]. This allowed us to detect any changes in the symmetric bell-shaped profile of the current's self-field caused by the trapped vortices. The experimental data for YBCO thin-film rings show that the symmetric profile is preserved for persistent currents of magnitude up to the saturation (critical) value. For YBCO ceramic rings trapping of intergrain vortices was minimized by using a special geometry, i.e., by cutting two notches along the ring's diameter (Ref. 21). The magnitude of the critical current was inferred from the maximum magnitude of the current's self-field at the ring's center using the Biot-Savart equation. Figure 1(b) presents the magnitude of the current's self-field, measured in the center of the zero-field-cooled (ZFC) YBCO thin-film

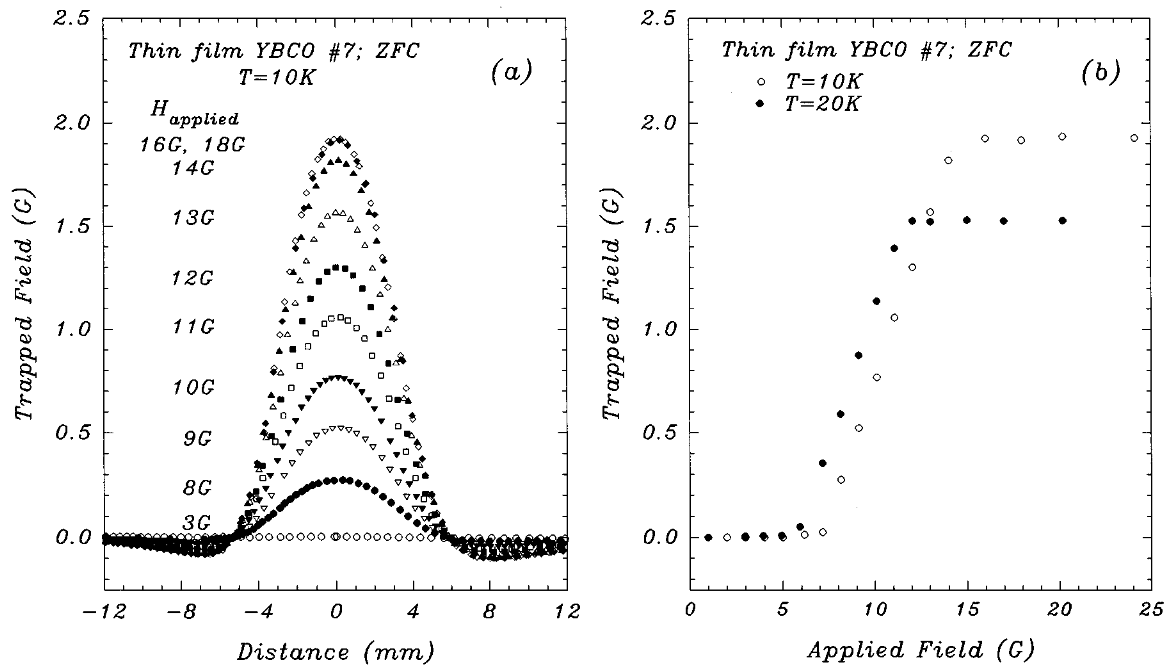

FIG. 1. (a) The profiles of the axial component of the magnetic self-field generated by persistent (self-supporting) currents in zero-field-cooled YBCO thin-film ring at 10 K. Persistent currents of various magnitudes were induced by applying and subsequently switching off an external magnetic field. The critical persistent current is reached when the current's self-field saturates. The field was measured with a scanning axial Hall probe at a distance of 2 mm from the surface of the film. Distances +4.25, −4.25 mm and +2.5, −2.5 mm mark the outer and inner edges of the ring, respectively. (b) Dependence of the persistent current's self-field measured at the ring's center on the applied magnetic field at 10 and 20 K. The saturation value of the self-field is proportional to the magnitude of the critical current at a given temperature.

ring, as a function of applied magnetic field. The saturation value of the field is proportional to the critical current at zero field and at a given temperature. The measurement of the temperature dependence of the critical current in a magnetic field was performed by using a field-cooling procedure at a given magnetic field H_0 (applied along the ring's axis) down to the temperature of the measurement. At this temperature the profile of the axial component of the Meissner field (which represents expulsion of magnetic flux out of the ring in the presence of external magnetic field H_0) was measured with a scanning Hall probe. An additional external magnetic field ΔH was then applied and subsequently reduced to zero, in order to induce a circulating persistent current at H_0. Similarly to the zero-field-cooling case, the current's critical value was reached by applying a higher ΔH. The critical current at H_0 is proportional to the current's self-field in the ring's center, which equals the difference between the field seen by the Hall probe and the corresponding Meissner field (see Ref. 21 for details of this procedure). The experimental setup used in these studies was equipped with a Hall probe (of sensitive area 0.4 mm^2 and ± 2 mG in sensitivity) which was scanned (over a scanning distance of 27 mm) along the ring's diameter at a constant height of 2 mm above the ring. The probe measured the axial component of the magnetic field (parallel to the ring's axis). An external magnetic field of up to a maximum of 750 G was generated along the ring's axis by a nonsuperconducting solenoid. The sample temperature was controlled over a range 10–90 K with GaAlAs diode and Pt resistance thermometers, and an inductionless heater. The system was able to record not only the magnitude of the critical current I_c but also the time decay of the current at various temperatures and magnetic fields. The measurement of I_c using the self-supporting supercurrent allowed us to prevent adding normal currents (which may flow through parts of the sample of very low resistance) to the measured value of I_c. Contribution of normal currents to I_c especially close to T_c cannot be avoided in standard methods such as I-V four-probe and ac-induction techniques. On the other hand, contactless detection of I_c allowed elimination of heating effects which are frequently encountered by standard methods at low temperatures in the presence of large currents. These experimental conditions ensured reliable measurement of the temperature dependence of I_c. By recording the time decay of the persistent current's self-field from its critical level, we were able to distinguish between a depinning critical current (decay observed) and a depairing critical current (no decay). Such a possibility has not been provided by any other critical current measurement method to date.

III. EXPERIMENTAL RESULTS

We measured the temperature dependence of the critical current (over a 10–90 K range) in about 20 c-axis-oriented YBCO thin films (manufactured in various laboratories) and in a few YBCO ceramic samples. The experimental data for selected YBCO thin films, which exhibit the most typical temperature dependence of I_c, are presented in Fig. 2. Critical currents in each sample were normalized to their values at 10 K. Thin film of the highest T_c of 90–91 K is characterized by Ambegaokar-Baratoff-like (AB) convex tempera-

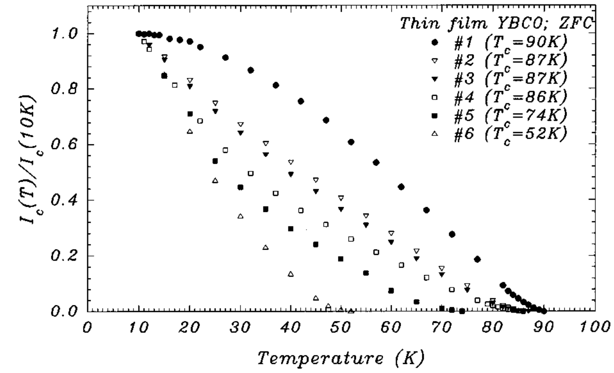

FIG. 2. Temperature dependence of the critical current in c-axis-oriented YBCO thin films with T_c over a range 52–90 K. The critical current is normalized to its value at 10 K. The results show a transition from an Ambegaokar-Baratoff-like (AB) temperature dependence to a Ginzburg-Landau-like (GL) one upon reduction of T_c. The AB→GL crossover is also seen in the film of T_c=90 K at a temperature of about 80–82 K. T_c is a temperature at which the level of the critical persistent current is less than about 5 mA (which corresponds to the noise level of a Hall probe). The critical current level at 10 K was within the range 2–45 A for various samples.

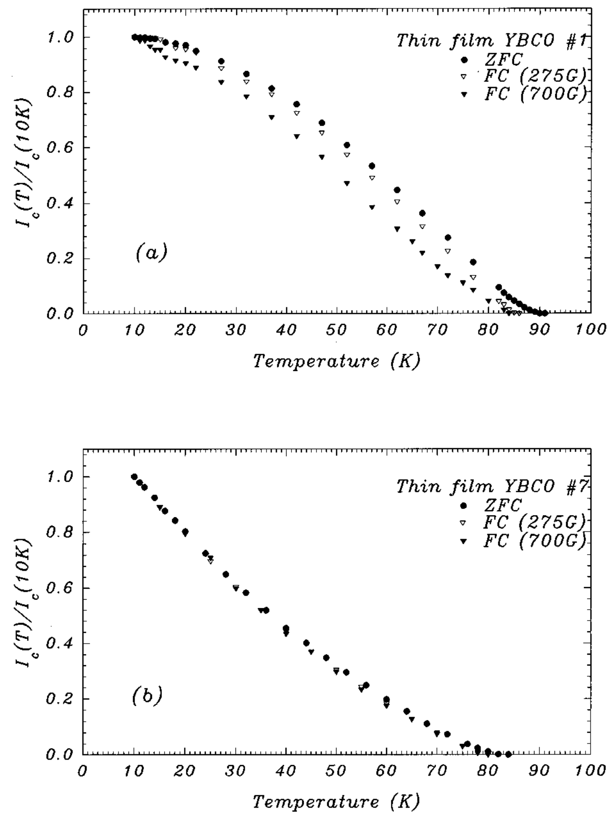

FIG. 3. Temperature dependence of the depairing critical current measured in c-axis-oriented YBCO thin films in magnetic fields up to 700 G. (a) Thin film of T_c=90 K which is characterized in the ZFC case by the Ambegaokar-Baratoff-like $I_c(T)$ below 80–82 K and by the Ginzburg-Landau-like $I_c(T)$ above this temperature. A magnetic field of 700 G removes the low temperature plateau of $I_c(T)$ and reduces the AB→GL crossover temperature. (b) Thin film of $T_c \simeq 83$ K characterized by the Ginzburg-Landau-like $(T-T_c)^{3/2}$ temperature dependence of $I_c(T)$. A magnetic field of 700 G does not have any effect on the shape of $I_c(T)$.

ture dependence below approximately 82 K (with a plateau between 10 and 20 K and a linear regime between 55 and 80 K) and Ginzburg-Landau-like (GL) concave temperature dependence above 80 K.

In thin films of T_c reduced by only 2°–4° (i.e., in a 86–88 K range), the Ambegaokar-Baratoff to Ginzburg-Landau (AB→GL) crossover temperature shifts down to about 60–70 K extending the Ginzburg-Landau tail down to low temperatures by about 10°–20°. In this case the temperature dependence of I_c below the crossover temperature exhibits a quasilinear behavior for most samples. The crossover temperature decreases down to about 40 K in thin films of $T_c \simeq 84$–85 K. Ginzburg-Landau-like temperature dependence $(T-T_c)^{3/2}$ over a full (10–90 K) temperature range was observed in most thin films of T_c below approximately 84 K. The exponent α in the Ginzburg-Landau expression $(T-T_c)^{\alpha}$ was determined to be 1.5±0.1, which was based on the measurements of $I_c(T)$ in all thin films. AB→GL crossover effects were found to be independent of the magnitude of the critical current density and depairing or depinning character of the critical current. The magnitude of the ''apparent'' critical current density at 10 K (defined as the ratio of the critical current to the ring's cross-sectional area) in various films was within a range 5×10^5–10^7 A/cm^2 for depinning critical currents. An ''apparent'' critical current density of 5–6×10^5 A/cm^2 at 10 K for thin films which do not exhibit any decay of the current from its critical value, is within the lower range of the ''apparent'' depinning critical current density. This is because depairing critical currents may flow through narrow filaments in the sample.

We investigated the influence of external magnetic field up to 700 G on the temperature dependence of the critical current in two thin films, one with the Ambegaokar-Baratoff-like temperature dependence of I_c and the other with a predominant Ginzburg-Landau-like behavior. These results are plotted in Fig. 3. No decay of the current from its critical value was detected by the Hall probe in both films, suggesting a depairing nature of the critical current. A 10–20 K plateau in $I_c(T)$, which is visible in the first film [Fig. 3(a)], disappears after application of a magnetic field of 700 G. This suggests a magnetic-field-induced decrease of the AB→GL crossover temperature. However, the Ginzburg-Landau-like temperature dependence of I_c, which characterizes the second film, is little affected by this magnetic field [Fig. 3(b)]. The experiment implies that a magnetic field much higher than 750 G (which was the maximum field available in the experimental setup) is required in order to change the temperature dependence of the critical current in c-axis-oriented thin film from an Ambegaokar-Baratoff to a Ginzburg-Landau-like one. We therefore decided to test the effect of a magnetic field on intergrain critical currents in YBCO ceramic samples. Previous studies of intergrain critical currents over a temperature range of 65–90 K (Ref. 21) revealed that small magnetic fields up to 30 G cause suppression of the critical current's magnitude and changes in the Ambegaokar-Baratoff character of $I_c(T)$ curve measured at zero magnetic field. Extension of these studies to a 10–90 K temperature range allowed us to produce the results shown in Fig. 4. Two YBCO ceramic rings characterized by depairing

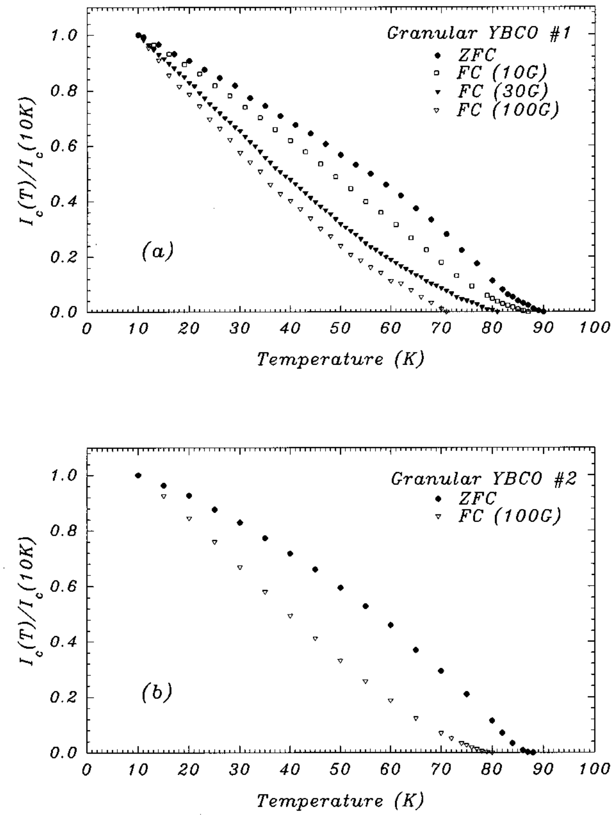

FIG. 4. Temperature dependence of the intergrain critical current measured in a granular (ceramic) YBCO in magnetic fields up to 100 G for depairing critical currents (a) and depinning critical currents (b). In both cases, a gradual transition from an Ambegaokar-Baratoff- to a Ginzburg-Landau-like behavior can be seen.

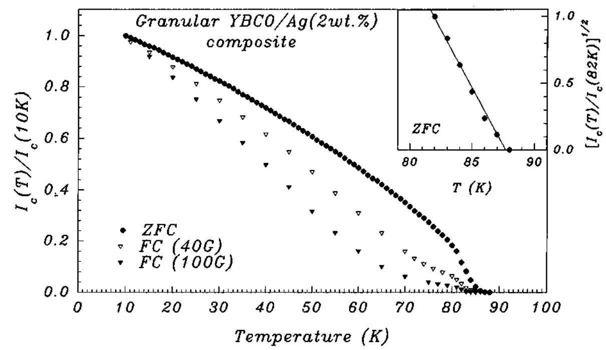

FIG. 5. Temperature dependence of the intergrain critical current in a granular YBCO/Ag (2 wt %) composite. In the ZFC case, $I_c(T)$ reveals an Ambegaokar-Baratoff-like behavior below 80 K, and a DeGennes-type behavior above 80 K with $I_c(T) \propto (T-T_c)^2$ shown in the inset. The applied magnetic field causes almost complete transformation of $I_c(T)$ into a Ginzburg-Landau-like one.

and depinning intergrain critical currents were used in the studies. In both of them $I_c(T)$ curves indicate a gradual decrease of the AB→GL crossover temperature with an increasing applied magnetic field. The crossover to Ginzburg-Landau behaviour was also investigated in YBCO/Ag (2 wt %) ceramic composite containing intergranular silver. The temperature dependence of the intergrain critical current in this composite under zero-field-cooling conditions (Fig. 5) manifests Ambegaokar-Baratoff-like behavior below 80 K and DeGennes (*SNS* junctions) behavior above this temperature. The temperature dependence of I_c in the DeGennes regime[6] is $(T-T_c)^\alpha$ with α=2.0±0.1 (see the inset in Fig. 5). An external magnetic field of 100 G is sufficient to cause almost complete conversion to a Ginzburg-Landau-like regime, and to screen contributions due to both SIS and SNS junctions.

IV. DISCUSSION

The experimental results for pure $YBa_2Cu_3O_{7-\delta}$ show the presence of both Ambegaokar-Baratoff-like (AB) and Ginzburg-Landau-like (GL) temperature dependences of the critical current in c-axis-oriented thin films and in randomly oriented granular ceramics (Figs. 6–8). The crossover from the AB to GL temperature dependence of I_c can be induced by either reduction in T_c [Fig. 6(b)] or by increasing the external magnetic field (Fig. 8). These features are independent of the type of transport critical current (depairing or depinning) and the magnitude of the critical current density (for the depinning critical current), suggesting that the type of flux-pinning defect does not affect the temperature dependence of the critical current. The shift of the AB→GL crossover temperature down to lower temperatures with decreasing T_c [Figs. 2 and 6(b)] implies its dependence on oxygen deficiency δ. According to Jorgensen *et al.*,[26] $YBa_2Cu_3O_{7-\delta}$ of $T_c \simeq 80$ K is characterized by oxygen deficiency δ=0.20–0.25. This means that for $\delta \geqslant 0.20$–0.25, the temperature dependence of I_c is mostly that of the Ginzburg-Landau-like $(T-T_c)^{3/2}$. Additional support of this statement is provided by the data of Jones *et al.*,[12] who studied the effect of oxygen deficiency on the transport properties of c-axis-oriented YBCO thin films. The temperature dependence of I_c in their best film (with δ=0.1) is quasilinear at low temperatures and a concave close to T_c [Fig. 3(a) in Ref. 12]. This temperature dependence corresponds to an intermediate stage (Fig. 2) of the transition from AB- to GL-type behavior of $I_c(T)$. Jones *et al.* removed oxygen from YBCO by sequential isobaric annealings at 550 °C under reduced partial pressure of O_2. A change of δ from 0.1 to 0.2 leads to Ginzburg-Landau-like temperature dependence of I_c. Our analysis of their data at δ=0.2 revealed that I_c is proportional to $(T-T_c)^\alpha$ with α=1.4–1.5. A decrease in the AB→GL crossover temperature can also be stimulated by applying external magnetic field. However, a magnetic field of 700 G, which we used in the experiments on thin films [Fig. 3(a)], did not produce a complete transformation of $I_c(T)$ from Ambegaokar-Baratoff to Ginzburg-Landau behavior. We have to refer, therefore, to an early work of Mannhart *et al.*[8] on c-axis-oriented thin films. The measurement of magnetic field dependence of $I_c(T)$ was performed on a thin film for which the zero field $I_c(T)$ exhibits a quasilinear behavior at low temperature and a pronounced concave tail close to T_c (Fig. 3 in Ref. 8). A magnetic field of the order of 0.5–1.1 T transforms the temperature dependence observed by Mannhart *et al.* into a Ginzburg-Landau-like $I_c(T)$. Our calculations based on their data revealed that $I_c \propto (T-T_c)^\alpha$ with α=1.5±0.1. When the temperature dependence of the critical current at zero magnetic field is already of a Ginzburg-

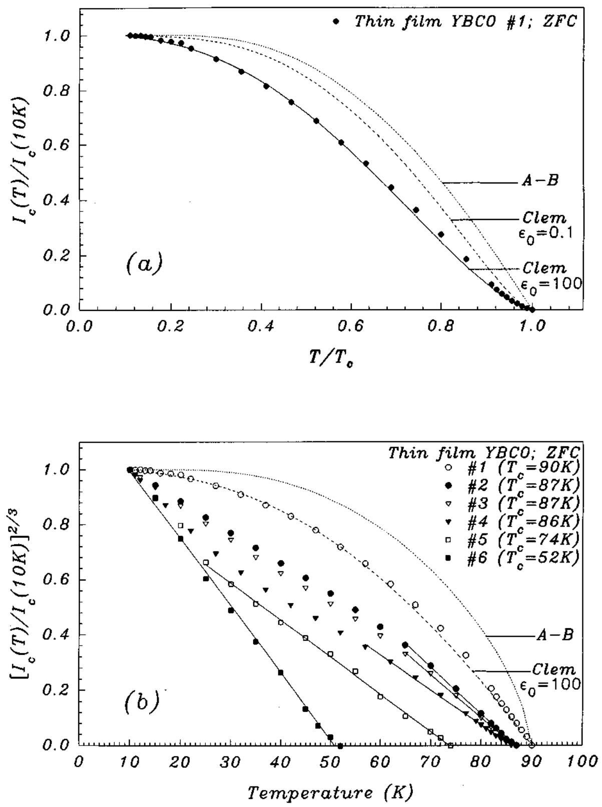

FIG. 6. (a) Comparison of the experimental data for $I_c(T)$ in c-axis-oriented YBCO thin film no. 1 with T_c=90 K (solid symbols) with the calculated ones from the Ambegaokar-Baratoff theory for Josephson tunnel junctions (Ref. 5), which are represented by the curve A-B, and from the Clem's model (Ref. 23) of Ambegaokar-Baratoff to Ginzburg-Landau crossover effects in granular weakly coupled (ϵ_0=0.1) and strongly coupled (ϵ_0=100) superconductors. The best fit to the experimental data was obtained for the case of $I_c(T)$ in strongly coupled granular superconductors (Clem, ϵ_0=100) with a temperature of the crossover from the Ambegaokar-Baratoff behavior at low temperatures to the Ginzburg-Landau behavior at high temperatures, at 80–82 K. (b) Temperature dependence of the critical current in c-axis-oriented YBCO thin films plotted for the results shown in Fig. 2 as $[I_c(T)/I_c(10\ \mathrm{K})]^{2/3}$ vs temperature. This allowed us to identify the Ginzburg-Landau portions of $I_c(T)$ (solid lines). All the experimental results are enclosed by two curves: by the Ambegaokar-Baratoff (AB) $I_c(T)$ or by Clem's form of $I_c(T)$ (AB→GL with ϵ_0=100) on the high-T_c side and by the Ginzburg-Landau $(T-T_c)^{3/2}$ form of $I_c(T)$ on the low-T_c side. A gradual expansion of the Ginzburg-Landau $(T-T_c)^{3/2}$ tail to low temperatures can be seen upon reduction of T_c.

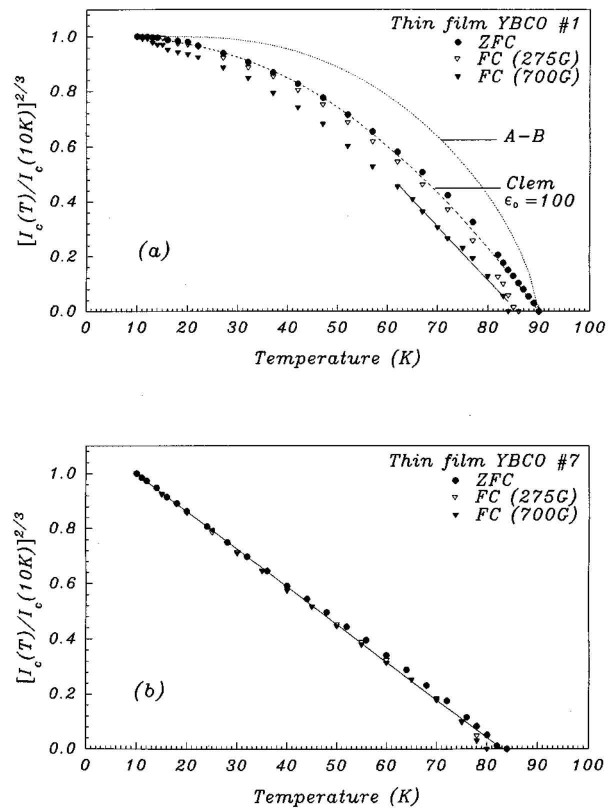

FIG. 7. Dependence of $[I_c(T)/I_c(10\ \mathrm{K})]^{2/3}$ on temperature plotted for the experimental results obtained for YBCO thin films no. 1 and no. 7 in magnetic fields of 275 and 700 G (Fig. 3). (a) Magnetic-field-induced expansion of the Ginzburg-Landau portion of $I_c(T)$ can be seen in zero-field-cooled YBCO thin film no. 1. For the ZFC case, the form of $I_c(T)$ agrees with Clem's model (ϵ_0=100). (b) The data reveal that the magnetic field does not affect the Ginzburg-Landau temperature dependence of $I_c(T)$ measured in zero-field-cooled YBCO thin film no. 7. Note that discrepancy between the experimental data and the Ginzburg-Landau solid lines very close to T_c is caused by the experimental difficulty in measuring the self-fields of very small persistent currents in the presence of much larger external magnetic fields.

Landau-like type, an external magnetic field does not change it. This can be seen in Figs. 3(b) and 7(b) for magnetic fields up to 700 G and in Fig. 6 of Ref. 14 for magnetic fields up to 12 T. In both cases the direction of magnetic field was perpendicular to the surface of c-axis-oriented thin films.

A magnetic field as low as 30 G is able to convert the Ambegaokar-Baratoff temperature dependence of the intergrain (weak-link) critical current to the Ginzburg-Landau-like one. The results of systematic measurements of AB→GL crossover in YBCO granular superconductors, presented in Figs. 4 and 8, are identical to those obtained for c-axis-oriented thin films. This implies that the AB→GL crossover effects are a characteristic property of a $YBa_2Cu_3O_{7-\delta}$ superconductor, independent of a crystallographic direction of the supercurrent's flow. On the other hand, similarities in $I_c(T)$ for both ceramic and thin films suggest that the intergrain connections (weak links) in ceramic YBCO have the form of narrow microbridges of intrinsic grainlike material.

An essential question that has to be answered here is ''what is the physical reason for the observed AB→GL crossover in the temperature dependence of the critical current?'' Clem *et al.*[23] stated that the Ambegaokar-Baratoff to Ginzburg-Landau crossover effects can be observed in conventional granular superconductors. The crossover from the Ambegaokar-Baratoff to the Ginzburg-Landau form of $I_c(T)$ occurs when the Josephson-coupling energy of an intergrain junction is approximately equal to the superconducting condensation energy of a grain. For YBCO thin-film and ceramic samples a gradual expansion of the Ginzburg-Landau tail $(T-T_c)^{3/2}$ in $I_c(T)$ to low temperatures is observed upon reduction of T_c or in an increasing applied magnetic field

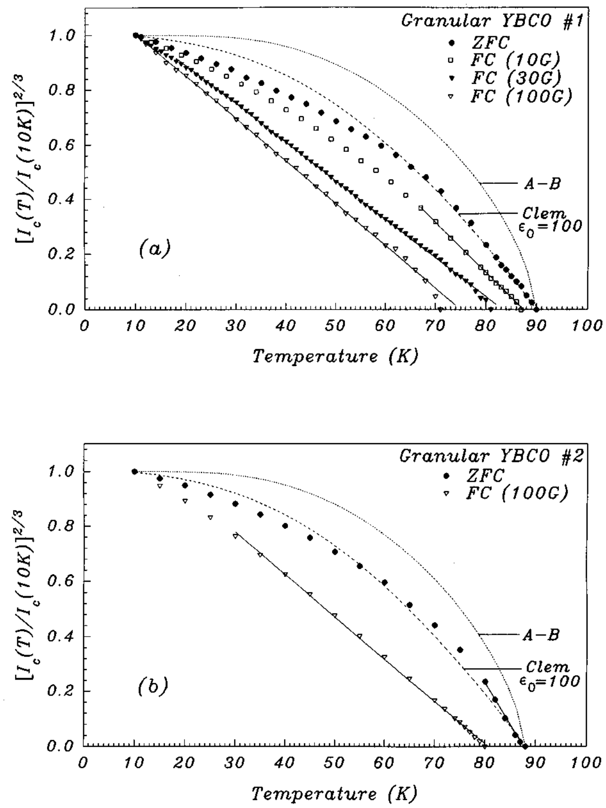

FIG. 8. $[I_c(T)/I_c(10\text{ K})]^{2/3}$ vs temperature plotted for the case of $I_c(T)$ measured in a granular YBCO (Fig. 4). A transition from the Ambegaokar-Baratoff form of $I_c(T)$ (dotted and dashed lines) to the Ginzburg-Landau one (solid lines) is observed for both depairing (a) and depinning (b) critical currents upon application of an increasing external magnetic field. Note that deviation of the experimental points from the solid lines very close to T_c is a result of experimental difficulties in measuring self-fields of very small persistent currents in the presence of much larger external magnetic fields.

(Figs. 6–8). The Ambegaokar-Baratoff-like dependence of $I_c(T)$ seems to be a characteristic property of YBCO samples of T_c above approximately 88 K and in the absence of an external magnetic field. The results presented in Figs. 6–8 imply, therefore, an AB→GL crossover in $I_c(T)$ of YBCO and a continuous decrease of the crossover temperature if the oxygen deficiency δ or the applied magnetic field are increased. According to Clem *et al.*[23] at the AB→GL crossover temperature in $I_c(T)$ of granular superconductors, the Ginzburg-Landau coherence length is of the order of the grain size. The observation of AB→GL crossover effects in YBCO, in combination with the short coherence length in this compound, indicate therefore the presence of granularity (superconducting microdomains coupled by Josephson tunnel junctions) on the level of a few nm. In thin films with highest $T_c \simeq 90$–91 K, the AB→GL crossover temperature is around 80–82 K. This means that the size of superconducting domains that exist in these films is equal to a coherence length at 80–82 K. Temperature dependence of upper critical field H_{c_2} in YBCO thin films in magnetic fields (up to 500 T) perpendicular to the c axis, was recently measured by Goettee *et al.*[27] These results allowed us to make a rough estimation of the coherence length ξ_{ab} at various temperatures using the relationship $H_{c_2}(T) = \Phi_0/2\pi\xi^2(T)$. ξ_{ab} varies from about 10 Å for a temperature range 8–30 K up to about 20 Å at 80 K and 30–40 Å at 83–84 K. Therefore, the maximum size of superconducting domains in samples with an optimum doping ($T_c \simeq 91$ K) and at zero magnetic field should vary between 30 and 40 Å. Oxygen deficiency and external magnetic field cause a decrease of size of these domains and a subsequent reduction of the AB→GL crossover temperature. In general, at low temperatures $I_c(T)$ is governed by interdomain Josephson tunnel junctions (Ambegaokar-Baratoff form). At higher temperatures $I_c(T)$ is affected by the ability of the supercurrent to suppress the gap parameter (Ginzburg-Landau form) when the coherence length is equal or larger than the size of superconducting domains.

Clem *et al.*[23] calculated the dimensionless parameter ϵ_0 which is proportional to the ratio of the Josephson coupling energy of a junction to the superconducting condensation energy of a grain, both energies being evaluated at zero temperature. For a weakly Josephson-coupled regime, $\epsilon_0 \ll 1$ and the temperature dependence of the critical current is that of the Ambegaokar-Baratoff form. For larger ϵ_0, approximately over a range 1–100, the Ambegaokar-Baratoff temperature dependence holds only at low temperatures, this behavior giving way to the Ginzburg-Landau $(T-T_c)^{3/2}$ temperature dependence above the crossover temperature. In the strongly Josephson-coupled regime ϵ_0 is very large, and current-induced gap suppression is dominant at all temperatures. In this case, the critical current does not obey the Ambegaokar-Baratoff behavior at any temperature. Instead the temperature dependence is governed by Ginzburg-Landau-like behavior at all temperatures. The coupling parameter ϵ_0 is given by the following formula:

$$\epsilon_0 = 2.93 \frac{\hbar k_B}{e^2 \gamma T_c \rho_n a_0^2}, \tag{1}$$

where γ is the Sommerfeld constant, ρ_n is the normal-state tunneling resistivity of a junction and a_0 is the effective grain diameter.

Reported values of γ for YBCO vary between 16 mJ/mol K^2 and 35 mJ/mol K^2 (0.16–0.35 mJ/cm^3 K^2).[28,29] We take the average $\gamma \simeq 0.26$ mJ/cm^3 K^2, and $a_0 = 40$ Å in YBCO with $T_c = 91$ K. In order to calculate ε_0,ρ_n of an intrinsic Josephson tunnel junction is needed. Such information was provided by current-voltage characteristics at 72 K of a 1-μm-wide Dayem microbridge patterned in a YBCO thin film (Ref. 11). The microbridge was twin-boundary free and had Josephson-like properties. A normal resistivity of the bridge was approximately 0.5 $\mu\Omega$ cm. We obtain $\varepsilon_0 = 92$ for YBCO with $T_c = 91$ K. For oxygen-deficient YBCO with $\delta = 0.20$–0.25, and $T_c \simeq 80$ K, the AB→GL crossover temperature drops down to 10–20 K implying that $a_0 \simeq \xi = 10$ Å. In this case YBCO is in a very strong-coupling regime with $\varepsilon_0 \simeq 1650$. The calculations of $I_c(T)$ for various coupling strengths, performed by Clem *et al.*[23] indicate the absence of a low-temperature plateau in the Ambegaokar-Baratoff part of $I_c(T)$ for $\varepsilon_0 \geq 1$. This is clearly seen in the temperature dependence of the critical current in the best YBCO samples.

Some evidence of ordered superstructures and phase separation in $YBa_2Cu_3O_{7-\delta}$ on a scale of a few nm has been provided by electron- and neutron-diffraction studies. Phase separation is more evident for oxygen-deficient samples with $\delta \lesssim 0.3$ (Refs. 30–33). However, studies by Beyers *et al.*[30] have indicated that 90 K-$T_c(\delta)$ plateau may correspond to two-phase regions. The presence of intragrain granularity in single crystal of YBCO has been suggested by Daeumling *et al.*[34] and Osofsky *et al.*[35] The granular behavior was attributed to clusters of oxygen defects, causing a separation of regions of oxygen-rich material by boundaries of oxygen-poor material. Recent scanning tunnelling spectroscopy measurements of YBCO single crystals,[36] which show a spatially varying energy gap over a few nm distances, may also indicate the presence of microdomains in YBCO. Recent measurements of the dynamic resistance of YBCO single crystals[37] suggest the presence of intra- and inter-unit-cell Josephson junctions. Electromigration experiments in YBCO thin films by Moeckly *et al.*[38] indicated that regions of strong oxygen disorder result in a network of superconducting filaments within the bulk film. Superconductivity across a grain boundary arises from an overlap of these filaments.[38,39] The same temperature dependence of the critical current in films and ceramics (Figs. 2 and 4) implies that intergranular transport in ceramics is limited by microbridges whose internal structure is similar to that of the grain itself, i.e., consisting of superconducting domains separated by Josephson tunnel junctions. If the size of these domains is less that the coherence length, the supercurrent will not ''see'' the junctions and the temperature dependence of the intergrain critical current changes from Ambegaokar-Baratoff type to Ginzburg-Landau one.

We performed additional verification of this process by introducing different kinds of Josephson junctions into the microbridges. Temperature dependence of the intergrain critical current of zero-field-cooled YBCO/Ag (2 wt %) ceramic composite (Fig. 5) indicates that the intergrain current is controlled by Josephson SIS tunnel junctions, [characterized by the Ambegaokar-Baratoff form of $I_c(T)$] up to 80 K, and by Josephson SNS proximity junctions (with $I_c(T) \simeq (T - T_c)^2$] above 80 K. This could happen if oxygen-depleted superconducting layers (of $T_c \simeq 80$ K) cross the microbridges. Increasing the applied magnetic field seems to introduce disorder (reduce the size of superconducting domains) in the same manner as increasing oxygen deficiency does. Figure 5 shows that an almost complete transformation of the intergrain $I_c(T)$ into Ginzburg-Landau form occurs in a magnetic field of 100 G. This suggests that in the case of superconducting domains coupled by SNS proximity junctions, the crossover from DeGennes-like $(T - T_c)^2$ temperature dependence of I_c to Ginzburg-Landau-like one may also be observed. A microbridge-type model was proposed very early by Larbalestier *et al.*[40] in order to explain the field-independent residual intergrain critical current in ceramic high temperature superconductors. Halbritter[41] stated that these conclusions may be valid not only for intergrain weak links but also for intragrain ones which are formed by twin boundaries. Moeckly *et al.*[38] suggested that rf Josephson data and nonideal I-V characteristics of most tilt boundary weak links favor microbridge limited transport.

Interpreting critical current $I_c(T)$ data requires discussion of the microscopic mechanism by which the microdomain size is being reduced. Both oxygen depletion and external magnetic field penetrating through insulating layers can suppress superconductivity through pair-breaking interactions. Oxygen deficiency leads to appearance of Cu^{2+} magnetic moments on CuO_2 planes[42] and CuO chains,[43] as shown by nuclear-spin relaxation data. Pair breaking in conventional superconductors caused by magnetic impurities and by external magnetic field has been analyzed by Maki[44] and Skalski *et al.*[45] These external perturbations break the time-reversal symmetry of the electron system and lead to gapless superconductivity. The gapless situation can also be induced by a different mechanism: the spatial variation of the order parameter.

In Kresin's model[24] of gaplessness in high-temperature superconductors, YBCO contains two subsystems, i.e., CuO_2 planes and CuO chains. The planes are intrinsically superconducting, whereas the superconducting state in the chains is induced by charge transfer via an intrinsic proximity effect. Magnetic impurities (Cu^{2+} magnetic moments) act like pairbreakers and suppress the induced gap on the chains so that at an oxygen content around 6.9 YBCO is already gapless. The gaplessness can be extended without a noticeable shift in T_c.

We believe that the oxygen vacancies and the associated Cu^{2+} magnetic moments already produce spatial inhomogeneities (domains on the scale of a lattice parameter) in YBCO of oxygen content of 6.95. These inhomogeneities would be similar to those suggested by Phillips[46] and Phillips *et al.*[28] An external magnetic field easily penetrates the inhomogeneities and reduce the size of superconducting domains through pair-breaking interactions.

V. CONCLUSIONS

We investigated the temperature dependence of the critical current in thin film and ceramic YBCO. The crossover from Ambegaokar-Baratoff to Ginzburg-Landau temperature dependence of I_c was observed, in agreement with the Clem's model[23] of $I_c(T)$ in strongly coupled granular superconductors. All the experimental $I_c(T)$ data are enclosed by two theoretical curves: the Ambegaokar-Baratoff curve on the high-T_c side and the Ginzburg-Landau one at low T_c. The crossover effects are observed for both intragrain, intergrain, depairing, and depinning critical currents. The temperature dependence of the critical current and the AB→GL crossover are independent of the type of flux-pinning defect and the crystallographic direction of the supercurrent's flow. On the basis of the Clem's model, the AB→GL crossover implies the presence of superconducting microdomains of 30–40 Å in size, coupled by Josephson tunnel junctions in the best YBCO samples (of $T_c \simeq 90$–91 K determined from the condition $I_c = 0$; no self-supporting current at T_c). The size of these microdomains decreases with increasing oxygen deficiency and with increasing applied magnetic field. We believe that the oxygen vacancies, the associated Cu^{2+} magnetic moments, and the external magnetic field (as a secondary effect) could break up YBCO into small microdomains through pair-breaking interactions. Intrinsic spatial inhomogeneities are responsible for nodal gap regions in $\vec{r}$ space according to Phillips.[46] It should be pointed out here that the presence of Ginzburg-Landau regime in $I_c(T)$ could suggest spatial homogeneity, since in this case the supercurrent does not ''see'' intrinsic junctions and other intrinsic defects. This is especially important for research on YBCO single crystals. The measurement of $I_c(T)$ in zero-field-cooled YBCO single crystals of $T_c \simeq 91$–92 K by either the transport or magnetization methods (Refs. 18, 20) shows a Ginzburg-Landau-like behavior of $I_c(T)$ with $I_c(T)=0$ between 70 and 77 K. This confirms that these single crystals are well oxygenated on their surface and poorly oxygenated in the bulk, and that $I_c(T)=0$ above 70–77 K is not necessarily caused by the magnetic flux lattice melting.

Another important conclusion, that should be addressed here, concerns superconducting YBCO ceramics. YBCO ceramics were considered dirty or ill-defined by ''single-crystal'' researchers. The temperature dependence of the intergrain critical current illustrates that intergrain connections in YBCO ceramics have a form of clean microbridges (or thin wirelike bridges) with microdomain structure the same as that of the grain. These microbridges can be easily penetrated by an external magnetic field. Therefore, the AB→GL crossover in the intergrain $I_c(T)$ can be induced by weak magnetic fields. For the zero-field-cooling case, the low temperature part of the $I_c(T)$ in pure YBCO ceramics and thin films is governed by an Ambegaokar-Baratoff regime, characteristic of SIS tunnel junctions. *SNS* normal metallic junctions appear only in YBCO/Ag composites. Therefore, this implies that superconducting microdomains are coupled by tunnel junctions and not by proximity (normal) junctions.

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