Differential resistance and critical-current distribution in $YBa_2Cu_3O_{7-x}$ ceramics

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V-I curves of several Y-Ba-Cu-O samples have been measured in the temperature range 77–91 K in weak magnetic fields ($H < 150$ Oe). For the well prepared low-resistivity samples $R_f \equiv dV/dI$ reaches a value of about $0.1R_n$ (R_n is the normal resistance just above transition) in the linear *V-I* regime. This is consistent with a picture of decoupled superconducting grains and does not yield any direct information about the flux-flow effects. Because of self-field effects, critical-current density (J_c) obtained from transport (and magnetization) measurements does not provide clear insight on the nature of intergrain coupling. Information on the distribution of critical currents, obtained from d^2V/dI^2, supports the tunneling and/or proximity-effect coupling between the grains and seems to rule out large enhancements of J_c in sintered samples with nonoriented grains.

I. INTRODUCTION

Soon after the discovery of high-temperature superconductors (HTS) it was realized that sintered HTS samples behave essentially as two-phase systems (grains and intergranular junctions, respectively) and that the separation of the effects of these "phases" is not that simple.[1] Extensive ac susceptibility measurements[2–4] confirmed that conclusion and have shown that the superconducting properties of these phases are very different.[5] In particular, critical currents within the grains appeared to be practically the same as those of good monocrystals or epitaxial films[6] whereas the bulk (intergrain) critical currents were much lower. In spite of a large effort the exact nature of the reduction of critical current (J_c) at the grain boundaries has still not been clarified.

Recently, critical currents within a single grain and through the grain boundary have been measured.[7] Whereas the intragrain critical current appeared to be flux-creep limited, no clear-cut decision about the nature of the reduction of J_c at the grain boundary (tunneling or proximity-effect coupling) was reached. Simultaneously high-resolution electron microscopy[8] indicates that most of the grain boundaries are clean (as far as the metal content is concerned) whereas microwave experiments clearly indicate the Josephson's loops on the scale of neighboring grains.[9] Although the sensitivity of the intergrain critical currents to low magnetic fields supports the tunneling or proximity-effect coupling between the grains, more recently explanations of the critical-current reduction in terms of the flux creep[10] and flux flow[11] at the grain boundaries have been proposed. The extreme sensitivity of the bulk J_c to the magnetic field is also detrimental in understanding the nature of intergrain coupling because the self-field effects mask the actual $J_c(H)$ and $J_c(T)$ dependences, thus rendering the results obtained from transport[11] or magnetization measurements[12] useless for quantitative analysis.[13]

The studies of the voltage-current (*V-I*) curves and of the associated differential resistance ($R_f = dV/dI$) proved very useful in understanding the mechanism of dissipation in conventional type-II superconductors. Therefore, similar studies of sintered and monocyrstalline HTS samples may also be very useful. Following previous results for sintered HTS samples[11,14,15] we report here the main results of our systematic investigation of *V-I* curves and R_f in sintered $YBa_2Cu_3O_{7-x}$ samples.

II. EXPERIMENTAL

The ceramic samples were prepared in the usual way.[1] According to x-ray diffraction all the samples were orthorhombic monophase. The majority of samples had a rather low resistivity at 95 K ($\rho_n < 500$ $\mu\Omega$ cm) and rather high resistivity ratio ($\rho_{300K}/\rho_n > 2.5$). Their "zero-field" values of J_c at 78 K ranged from 100 to 250 A/cm^2 (1 μV/cm criterion) and showed no systematic dependence on their normal-state properties. The samples with high initial ρ_n (>1 mΩ cm) as well as those which deteriorated during repeated thermal cycling (77–300 K) had lower values of J_c (<100 A/cm^2).

The *V-I* curves were measured while passing half-sinusoidal-current pulses through the sample. The duration of the pulses (typically 1 s) was selected in such a way that the heating of the sample was negligible. The 40 ms acquisition rate was consistently employed in the system for automatic recording and processing of the data. The heating of a sample was minimized by the low resistance (a few milliohms), high-temperature-treated silver-paint contacts, and immersion of the samples in appropriate cryogenic baths (liquid nitrogen, liquid oxygen, and their mixtures). Since the transport critical current of sintered HTS samples is self-field limited[16] and thus depends on the shape and cross section of the sample, all our samples had practically the same cross section ($\simeq 1.5$ mm^2) and distance between the voltage contacts (7 mm).

III. RESULTS AND DISCUSSION

The V-I curves and their first and second derivatives for two samples of very different quality, J_c (78 K)$=220$ A/cm^2 and 80 A/cm^2 for samples $S1$ and $S2$, respectively, are shown in Fig. 1. These curves are qualatively the same as those observed in conventional type-II superconductors but are also similar to those of bulk normal metal–superconductor composites[17] and of some variable thickness microbridges.[18]

For a conventional type-II superconductor[19] a nonlinear increase of voltage (dissipation) just above J_c is associated with depinning of the vortices, whereas a linear variation at higher currents reflects their viscous flow (flux flow). The slope of a linear part of a V-I curve (R_f) is then independent of the actual J_c value but depends sensitively on the magnetic field and temperature ($T > 0.5T_c$). Furthermore, V-I curves (thus R_f) should depend strongly on the direction of the magnetic field with respect to that of the current through the sample.

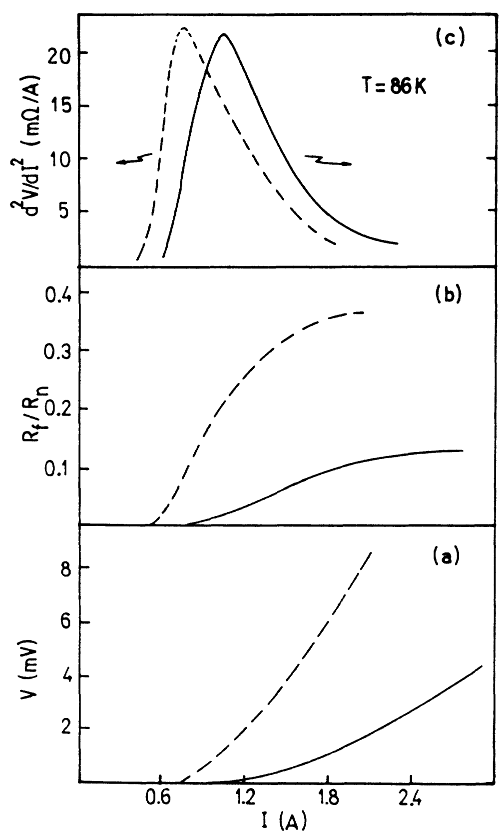

FIG. 1. I-V characteristcs (a), its first derivative normalized to resistance just above T_c (b), and second derivative (c) for two $YBa_2Cu_3O_{7-x}$ samples of different quality; (sample $S1$—solid line, sample $S2$—dashed line).

Our present and previous results[11,14,15] on sintered Y-Ba-Cu-O samples are at variance with the above observations. V-I curves are insensitive to the direction of the magnetic field; R_f is sample dependent (Fig. 1) and hardly depends on temperature ($65 < T < 90$ K) and magnetic field ($B < 5$ T at 77 K, Ref. 14). Therefore at low fields the flux-flow model does not seem to provide an adequate explanation of V-I curves of sintered Y-Ba-Cu-O samples. However, at higher fields ($B > 5$ T) a strong increase of R_f with B has been observed.[14] The results for 77 K indicate $[R_n^{-1}(dR_f/dB) \sim B_{c2}(0)^{-1}]$ a higher critical field of about 50 T. This is a lower bound for $B_{c2}(0)$ since at such high temperature R_f should increase faster than linearly with B. Thus, the flux flow (within the grains) dominates the dissipation for $B > 5$ T but does not explain the magnitude and behavior of R_f at lower fields.

Alternatively one can model sintered HTS samples as a composite material consisting of superconducting particles (T_c) embedded in a nonsuperconducting matrix. Such systems may undergo a transition into a "coherent state" when the intergrain coupling energy overcomes the thermal fluctuations.[20] This state manifests itself by zero resistance at some temperature $T_{cs} < T_c$ which depends sensitively on the applied field and current flowing through the sample. In HTS these dependences are interrelated because of the self-field effects. Whatever is the nature of intergrain coupling, a sufficiently high magnetic field and/or current should effectively decouple the superconducting grains, and thus show up the resistance of the integranular material. Indeed the magnitudes of R_f observed by us (Fig. 1) and others[11,14] as well as the insensitivity of R_f to the magnitude of magnetic field or current (up to fairly high values) support that view.

A highly simplified picture of a sintered HTS sample in a decoupled state is that of an assembly of superconducting spheres embedded in a nonsuperconducting matrix with resistivity ρ_n. The effective resistivity of such a medium depends on the packing fraction of spheres[21] and for rather dense packing appropriate to well prepared sintered samples one obtains $\rho_{\text{eff}} \sim 0.1\rho_n$, which agrees well with measured ρ_f (R_f) values. Allowing for a less dense packing (such as in poorly prepared samples) and/or the bad quality of some grains one may obtain larger $\rho_{\text{eff}} = \rho_f$ as was indeed observed in our experiments (Fig. 1). Although the results for R_f of well prepared sintered samples can be interpreted in terms of the superconducting grains with average diameter d separated by the normal layer of thickness 2λ (λ is the magnetic-field penetration depth) if $d > 10\lambda$ (which is often the case for sintered samples), the best interpretation for $R_f < R_n$ is that even at currents several times larger than J_c a large fraction of grains remains superconducting. This was confirmed in our experiments at temperatures rather close to T_c. In those cases R_f remained constant up to about $10J_c$ when the resistive transition of grains occurred. We note, however, that $R_f < R_n$ does not exclude tunneling or proximity-effect coupling between the grains. Indeed a temperature- and magnetic-field-independent $R_f < R_n$ was also observed in conventional normal metal–superconductor bulk composites.[17]

According to the above interpretation, the insensitivity of R_f to low magnetic fields reflects the low magnetoresistance of ρ_{eff} (consistent with the results for sintered Y-Ba-Cu-O samples in a normal state), whereas the independence of R_f on temperature (65–90 K) probably indicates a weak temperature dependence of ρ_{eff} in that temperature range.

The nature of dissipation in nonlinear parts of V-I curves is of particular interest. For conventional type-II superconductors it is usually attributed to the flux creep and an initially exponential variation of V with I or H (if I is kept constant) is expected.[19] In sintered Y-Ba-Cu-O samples a nonlinearity of V-I curves extends over a very broad range of I ($\Delta I > 2I_c$ for $T > 77$ K) but no exponential variation of V with I or H (constant I) in any significant range of I or H has been observed in our experiments. Instead, all our V-I curves exhibited an initial $V \sim I^n$ variation, consistent with the earlier reports.[11,15] At 77 K the exponents n ranged from 4 to 15 but no correlation between J_c and n could have been established (the largest J_c had sample with $n=10$).

A similar power-law variation of V was also observed in composite superconducting wires and was related to the distribution of critical currents $f(I_c')$ within the wire.[22] [The distribution of critical currents is obtained[23] from the second derivative of the V-I curve, $f(I_c') \sim d^2V/dI^2$.] In particular, lower n corresponded to wider distribution, which in turn was observed in samples with very irregular superconducting filaments.[22]

Considering the microstructure of ceramic HTS it seems clear that a broad distribution of critical currents should exist in these samples. The results for d^2V/dI^2 (numerical derivatives over three successive data points on the V-I curve) shown in Fig. 1 support that viewpoint. Whereas in composite superconductors the width of distribution ($\Delta I_c'$) was always less than I_c,[22] for our samples at 78 K $\Delta I_c' > 2I_c$. Furthermore, since the samples with lower I_c had higher $\Delta I_c'/I_c$ ratio, the values of the highest possible critical current were quite similar for all our samples. Thus from our results for $f(I_c')$ we may conclude that $J_c > 1000$ A/cm^2 at 77 K is quite unlikely to be achieved in sintered Y-Ba-Cu-O samples prepared in a conventional way. This conclusion is in accord with the reported J_c data. Furthermore, the shape of $f(I_c')$ is usually asymmetric with the maximum closer to the measured I_c (Fig. 1), which further limits the prospects of achieving technologically relevant critical-current densities in nontextured sintered HTS.

As a more direct check of the analogy between the composite superconductors and sintered HTS we studied the effects of the sample deterioration on $f(I_c')$ and the exponent n for some of our samples. As illustrated in Fig. 2 the distribution is broadened and the exponent n decreased in the deteriorated sample. As expected, the deterioration introduces considerably lower I_c' than in the fresh sample but has less effect on the maximum of $f(I_c')$ and on its high I_c' part (tail). Thus high critical currents can only be obtained in samples in which practically all intergrain links are strong. At the same time the measured J_c does not give a proper description of the overall quality of the sintered sample (it reflects the weakest intergrain links in a given sample) which in turn may explain the absence of correlation between the normal state properties and J_c in sintered Y-Ba-Cu-O samples. Therefore the d^2V/dI^2 versus I variation seems to be the most powerful diagnostic tool for the evaluation of the quality of sintered HTS samples and/or of the progress made in their improvement.

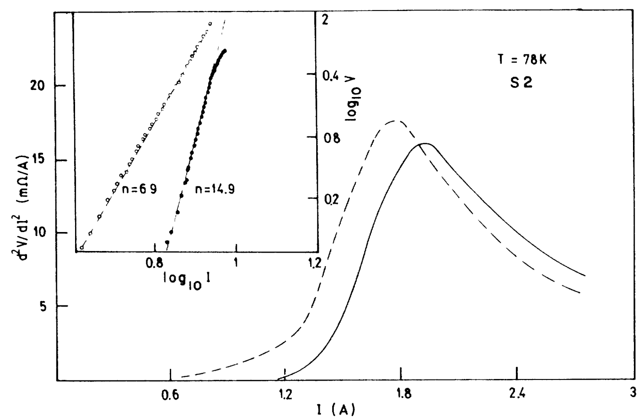

FIG. 2. Second derivative of V-I curve (critical-current distribution) for the same sample before (solid line) and after the degradation (dashed line). The inset: log-log plot of I-V data for the same sample proving $V \sim I^n$ variation.

We also studied the effects of magnetic field and temperature on $f(I_c')$. As illustrated in Fig. 3, already rather low magnetic fields decrease I_c strongly and thus shift the distribution and its maximum to lower current values. Since the high-current tail of $f(I_c')$ is less affected by the magnetic field, the distribution is strongly broadened and the exponent n accordingly reduced. However, the variation of n with H illustrated in the inset of Fig. 3 is very different from that observed in composite superconductors.[22] Whereas in Y-Ba-Cu-O n decreases very rapidly in small fields but tends asymptotically to $n=1$ for fields larger than the lower critical field of the grains,[12] in com-

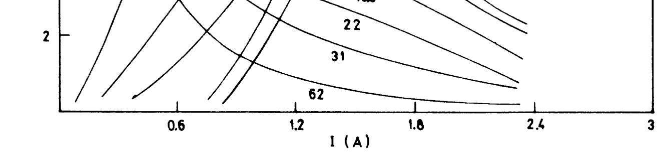

FIG. 3. Dependence of the critical-current distribution and (inset) power-law-fit exponent ($V \sim I^n$) on the applied magnetic field (in Oe).

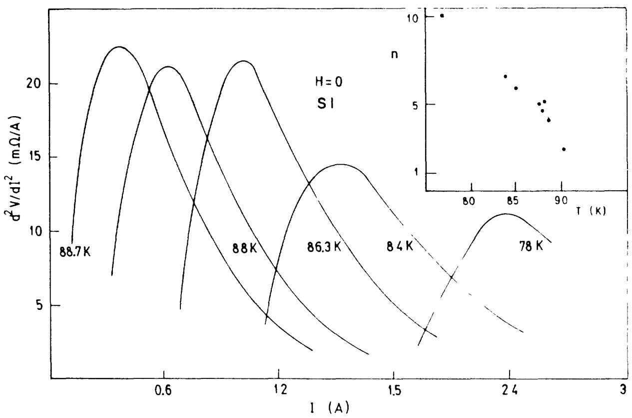

FIG. 4. Dependence of the critical-current distribution and (inset) power-law-fit exponent ($V \sim I^n$) on temperature.

posite superconductors with comparable n values[22] n is rather insensitive to field up to fields close to H_{c2} where it rapidly decreases to $n=1$. This difference probably reflects different origins of $f(I_c')$. The behavior of $f(I_c')$ and n in Y-Ba-Cu-O seems to support the tunneling or proximity-effect coupling between the grains (strong effect of low fields, but the absence of an upper critical field). We also observed the self-field effects on $f(I_c')$. In particular $f(I_c')$ was rather insensitive to the applied magnetic field for fields lower than the self-field (H_s) due to current flowing through the sample (Fig. 3). Because of this the actual $f(I_c')$ in true zero field cannot be deduced. Apparently the magnetic field causes redistribution of currents within the sample and these effects can in principle be investigated by means of $f(I_c')$.

The effects of temperature (77–90 K) on $f(I_c')$ are illustrated in Fig. 4. The increase of temperature shifts roughly linearly the maximum of $f(I_c')$ to lower currents but does not broaden the distribution as much as the magnetic field does (Fig. 3). In particular the part of $f(I_c')$ at lower currents becomes steeper on increasing temperature (Fig. 4) in contrast to what is observed on increasing the magnetic field (Fig. 3). We note, however, that this could be due to decreasing self-field (I_c decreases with increasing temperature) which makes a more detailed analysis of the temperature dependence of $f(I_c')$ more complicated. Typical variation of the exponent n with temperature is shown in the inset to Fig. 4. Whereas the initial linear decrease of n may be affected by the change in the self-field, a rapid reduction of n, approaching $n \sim 1$ close to T_c (T_{cs}), again supports the proximity effects and/or tunneling between the grains. A more detailed analysis of the effects of the magnetic field and temperature on $f(I_c')$ will be given elsewhere.

IV. CONCLUSION

Systematic investigation of V-I curves of sintered Y-Ba-Cu-O samples show that the superconducting grains in these samples can be effectively decoupled by the application of the magnetic field and/or current in excess of I_c. In either case a total magnetic field of the order of the lower critical field of the grains[12] that acts on the sample is sufficient to practically decouple the grains. In the decoupled state the differential resistivity ρ_f of the sample can be modeled as that of a system consisting of superconducting grains embedded in a matrix with the resistivity ρ_n (ρ_n is the resistivity of the sample just above T_c).

The existence of intergrain couplings with very different strengths suggests a broad distribution of critical currents within the sintered sample. This distribution can be deduced from a nonlinear part of V-I curve in analogy with the technique developed for composite superconducting wires and ribbons.[22] The critical-current distributions of our samples obtained in that manner appear to be very broad ($\Delta I_c > 2I_c$ at 77 K) but with only a narrow tail extending to currents several times larger than I_c. From these results we estimate that critical current densities exceeding 1000 A/cm^2 are unlikely to be achieved for the sintered samples without texture. The critical-current distributions can, however, be used as a sensitive diagnostic tool of the quality of samples and may as well help the understanding of the nature of intergrain coupling in sintered HTS. Our results for the effects of the magnetic field and temperature on the critical-current distributions support the proximity-effect and/or tunneling origin of the intergrain coupling.[14,20] In that respect, comparative studies of V-I characteristics in single crystals and grain-oriented high-J_c sintered samples would be of particular interest.

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[1]E. Babić, Ž. Marohnić, M. Prester, and N. Brničević, Philos. Mag. Lett. **56**, 91 (1987).

[2]R. B. Goldfarb, A. F. Clark, A. I. Braginski, and A. J. Panson, Cryogenics **64**, 79 (1987).

[3]E. Babić, Ž. Marohnić, Dj. Drobac, M. Prester, and N. Brničević, Int. J. Mod. Phys. **B1**, 987 (1987).

[4]H. Küpfer, I. Apfelstedt, W. Schauer, T. Wolf, and H. Wühl, Physica **153-155C**, 367 (1988).

[5]E. Babić, Ž. Marohnić, Dj. Drobac, M. Prester, and N. Brničević, Physica **153-155C**, 1511 (1988).

[6]P. Chaudhary, R. H. Koch, R. B. Laibowitz, R. R. McGuire, and R. J. Gambino, Phys. Rev. Lett. **58**, 2684 (1987).

[7]J. Mannhart, P. Chaudhary, D. Dimos, C. C. Tsuei, and R. R. McGuire, Phys. Rev. Lett. **61**, 2476 (1988).

[8]H. W. Zanderbergen, R. Gronsky, and G. Thomas, Physica **153-155C**, 1002 (1988).

[9]G. B. Donaldson, Cryogenics **28**, 668 (1988), and references therein.

[10]M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).

[11]R. Meisels, S. Bungre, and A. D. Caplin, J. Less-Common

Met. **151**, 83 (1989).

[12]E. Babić, Dj. Drobac, J. Horvat, Ž. Marohnić, and M. Prester, J. Less-Common Met. **151**, 89 (1989).

[13]E. Babić, M. Prester, Ž Marohnić, T. Car, N. Biškup, and S. A. Siddiqi, Solid State Commun. **72**, 753 (1989).

[14]J. W. Ekin, A. I. Braginski, A. J. Panson, M. A. Janocko, D. W. Capone, M. J. Zaluzec, B. Flandermeyer, O. F. de Lima, M. Hong, J. Kwo, and M. S. Liou, J. Appl. Phys. **62**, 4821 (1988).

[15]J. E. Evetts and B. A. Glowacki, Cryogenics **28**, 641 (1988).

[16]R. B. Stepens, Cryogenics **29**, 399 (1989).

[17]P. England, F. Goldie, and A. D. Caplin, J. Phys. F **17**, 447 (1987).

[18]K. K. Licharev, O. V. Snigirev, and E. S. Soldatov, in *17th International Conference on Low Temperature Physics (Contributed papers),* edited by U. Eckern *et al.* (Elsevier, Amsterdam, 1984), p. 915.

[19]V. B. Kim and M. J. Stephen, in *Superconductivity,* edited by R. D. Parks (Marcel Decker, New York, 1969), p. 1107.

[20]J. R. Clem, Physica **153-155C**, 50 (1988), and references therein.

[21]J. C. Maxwell, *A Treatise on Electricity and Magnetism* (Oxford University, New York, 1904), p. 441.

[22]W. H. Varnes and D. C. Larbalestier, Appl. Phys. Lett. **48**, 1403 (1986).

[23]J. Baixeras and G. Fournet, J. Phys. Chem. Solids **28**, 1541 (1967).