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Evidence for homogeneous superconducting grains in high-T_c oxides

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We present direct and magnetization critical current data on ceramic $YBa_2Cu_3O_{7-\delta}$ which indicate that the "homogeneous superconducting regions" are much larger than the penetration depth and in fact are grain size. Thus, the current-limiting Josephson junctions in the high-T_c oxide ceramics must be *inter*granular rather than *intra*granular. In light of these findings, we believe that a high density of twin boundaries probably *enhances* the intragranular critical current.

The recently discovered high-T_c oxides have perovskite-based nominally layered structures.[1] While the presence of Cu-O chains in the orthorhombic structure of the 90-K superconductors (for example, $YBa_2Cu_3O_{7-\delta}$ may be crucial to the high transition temperature (T_c),[2] these materials generally behave like anisotropic uniaxial (layered) superconductors.[3] Most research has centered on ceramic pellets; average grain sizes vary widely, but 10 μm is a typical value.[4] The ceramics have very low critical current densities (J_c's), orders of magnitude lower than those seen in epitaxial films[5] or crystals.[3] These low J_c's are generally recognized as due to Josephson weak links, and are a major obstacle to most potential applications of the ceramics.[6]

Granular superconductors have often been modeled as random arrays of small "homogeneous superconducting regions" weakly coupled by Josephson junctions (we shall use junctions to refer to generalized weak links).[7] Such a system becomes weakly random (glassy) at a field $H_{c1}^{*}=\Phi_0/2S$, where Φ_0 is the flux quantum, 2×10^{-7} Oe cm^2, and S is the projected area normal to the field of an average loop of connected superconducting regions.[8] Some observed low-field magnetic properties of the high-T_c oxides have been interpreted in terms of this superconductive glass picture.[9,10] In order for the glass model to be appropriate, however, the size of the superconducting regions must be less than the temperature-dependent bulk penetration depth $\lambda(T)$, so that the magnetic response is that of clusters of loops rather than of individual superconducting regions.[8] But $\lambda(0)$ for the high-T_c oxides has been estimated to be <0.2 μm,[11] much less than the typical sizes of either ceramic grains or single crystals. In order to provide a microscopic basis for the glass picture, Duetscher and Müller recently proposed that Josephson junctions occur at (110) and (001) twin boundaries, which form within grains on a length scale <0.1 μm.[12] They suggest that elimination of the twin boundaries is necessary for the attainment of high J_c's.

If, on the other hand, the grains are homogeneous superconductors, i.e., only *inter*granular junctions are important, the observed magnetic properties suggest a characteristic field H_d at which a grain coupling-decoupling transition occurs due to the collapse of intergranular supercurrents.[13] In this case, the twin density is irrelevant for the intergranular (bulk) J_c of existing ceramics, but most likely *enhances* the observed intragranular and single crystal J_c's.[6] The weak links in the ceramic high-T_c oxides have frequently been assumed to be intergranular, but this assumption has not been critically tested against the twin-boundary hypothesis.

In this paper, we report direct and magnetization J_c results on single-phase ceramic $YBa_2Cu_3O_{7-\delta}$ which indicate that the grains are homogeneous superconductors. In particular, the strong temperature dependence of the length scale inferred from $J_c(H,T)$, as well as the observation that flux trapped in the samples during low-field cycles does not affect J_c, both imply interjunction homogeneous regions larger than the penetration depth. This conclusion is independent of any particular estimate for the penetration length. In fact, the magnitudes of the magnetization J_c at 5 and 76 K are consistent with grain-sized (or larger) homogeneous regions. Finally, given homogeneous grains, we briefly consider some likely strategies for achieving high-J_c ceramic materials.

Details of sample preparation and characterization and experimental techniques are given elsewhere.[4,13] We emphasize here that our ceramic samples are comparable to those in the literature—single-phase, ~70% dense, 10-μm diameter grains, resistivities of ~1 mΩ cm at room temperature, and a sharp transition above 90 K.

The data points in Fig. 1 show the observed field dependence of J_c in a monotonically increasing field for a typical sample at 4, 30, and 74 K; the zero-field J_c's at these temperatures were 1380, 1130, and 305 A/cm^2 ($\pm10\%$), respectively. We interpret these data by first considering two large superconducting regions of identical material connected by a single square Josephson junction in a transverse magnetic field, for which the critical current is[14]

$$I_c(H,T)=b^2J_0(T)\,|\sin(\pi H/H_0)/(\pi H/H_0)|\ . \qquad (1)$$

$J_0(T)$ is the zero-field critical current density of the junction and b^2 is the junction cross section. H_0 is given by $H_0\simeq\Phi_0/2b\lambda(T)$ ($\propto\epsilon^{1/2}$ near T_c, where $\epsilon\equiv1-T/T_c$), if one ignores the junction thickness compared to $\lambda(T)$, and assumes that b is small compared to the Josephson penetration depth $\lambda_J(T)$. λ_J describes the penetration of a field into the junction itself:[14]

$$\begin{aligned}\lambda_J(T)&\simeq[c\Phi_0/16\pi^2J_0(T)\lambda(T)]^{1/2}\\&\simeq10^{-4}[J_0(T)\lambda(T)]^{-1/2}\ .\end{aligned} \qquad (2)$$

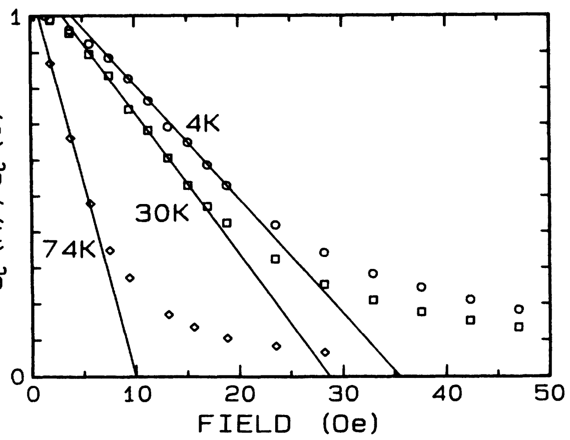

FIG. 1. Directly measured critical current density vs magnetic field at various temperatures for a typical ceramic sample of $YBa_2Cu_3O_{7-\delta}$. The behavior is as expected for a random array of Josephson junctions. The lines are extrapolations of the steepest portions of the data to zero current which provide a measure of the temperature-dependent field H_0 characteristic of an average junction in the array (see text).

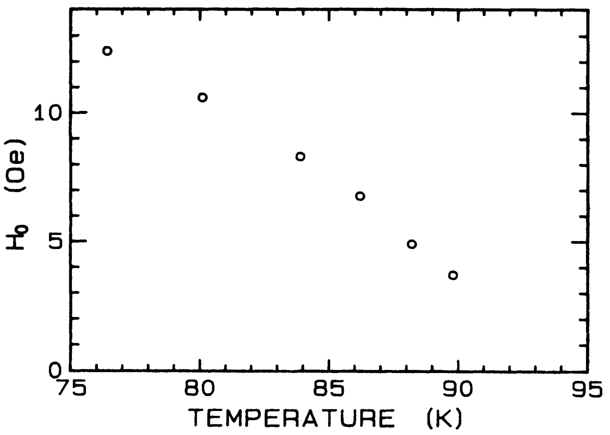

FIG. 2. Temperature dependence of the characteristic field H_0 (see Fig. 1) for typical ceramic $YBa_2Cu_3O_{7-\delta}$, above 76 K. The pronounced temperature dependence implies an average interjunction spacing large compared to the bulk penetration depth (see text).

The last expression in Eq. (2) is in practical units (J_0 in A/cm^2, λ's in cm). For a cubic array of junctions, $J_c(H,T)=I_c(H,T)/a^2$, where a is the interjunction spacing. One expects the diffraction pattern implicit in Eq. (1) to be washed out for a random array, as we observe in Fig. 1, but the steep drop at low fields will remain. Details of an averaging procedure which preserves these features will be given elsewhere.[15] For our purposes here, it is sufficient to define H_0 from the data as the field at which the steep drop in J_c extrapolates to $J_c=0$, which gives $H_0=10$, 29, and 35.5 Oe at 4, 30, and 74 K, respectively.

For an array, the interpretation of H_0 must be generalized to include the possibility of large junctions $[b>\lambda_J(T)]$ or small spacings $[a<\lambda(T)]$. For small junctions and small spacings, the situation closest to a glass, we expect $H_0\approx\Phi_0/\pi ab$, independent of temperature. For the opposite case of large junctions and large spacings, $H_0\approx\Phi_0/\pi\lambda(T)\lambda_J(T)$ ($\propto\epsilon^{3/4}$ near T_c).[15] In fact, the observed $J_c(0,T)$ and the literature values for $\lambda(T)$ yield $b/\lambda_J(T)\sim 1$ in the investigated temperature range (below 90 K). The H_0 values between 76 and 90 K (Fig. 2) derived from the present data well fit $\epsilon^{3/4}$ with $T_c\equiv 93$ K. Although $\epsilon^{1/2}$ behavior above 90 K cannot be ruled out (the critical current became too small to measure reliably), Fig. 2 suggests that our ceramics are in the large-junction–large-spacing regime. Regardless of the exact exponent near T_c, the pronounced temperature dependence of H_0 over a wide range of temperatures indicates interjunction spacings large compared to λ, consistent with intergranular junctions.

A completely independent test of the interjunction spacing is possible from the critical current analog of remanent flux measurements. Because of the extreme sensitivity of J_c to field, one should easily see residual flux trapped within a penetration depth of the junctions after a field cycle. Experimentally, one ramps the field up to some value H and back to zero before measuring what we will call J_{c0}; subsequent data points can only be obtained at successively higher values of H. In Fig. 3, we show the results of such experiments on the sample of Fig. 1 at 4 and 30 K and the sample of Fig. 2 at 76 K (the data above 76 K are similar and are omitted for clarity). We observe no drop in J_{c0} for cycles up to above 100 Oe at any of these temperatures, despite signs of trapped flux in the sample from magnetization data.[13] This finding indicates that flux is not trapped within λ of the junctions, which therefore *must be separated by homogeneous regions larger than* λ. In fact, the magnitude and temperature dependence of the field for which J_{c0} begins to drop suggest an effect related to the bulk H_{c1}.[13,15] In a glassy system of junctions at twin boundaries, however, flux would

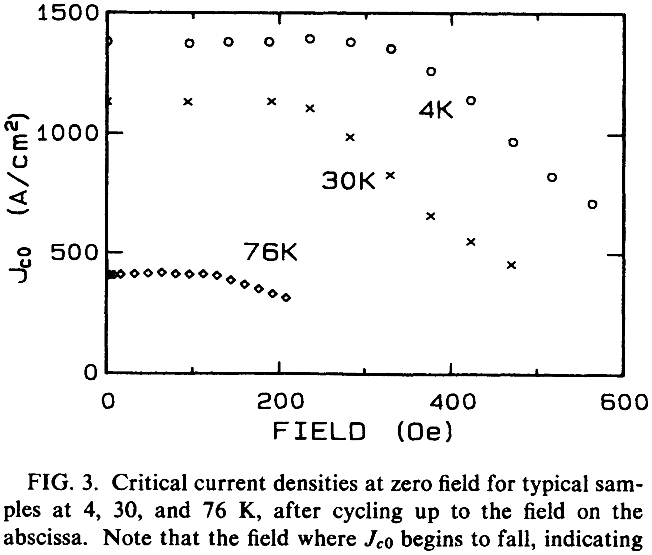

FIG. 3. Critical current densities at zero field for typical samples at 4, 30, and 76 K, after cycling up to the field on the abscissa. Note that the field where J_{c0} begins to fall, indicating trapped flux near the junctions, is much greater than H_0, which is always below about 20 Oe. This result shows that the current-limiting junctions are not at intragranular twin boundaries.

penetrate the system for fields just above $H^*_{c1} \ll H_{c1}$, and should be preferentially trapped near the junctions.[9]

An indication of the actual size of the homogeneous superconducting regions comes from magnetization J_c data.[13,16] For a collection of randomly oriented spherical regions (diameter a) of a layered superconductor, each with the maximum possible current flowing in the basal plane, but with negligible current between regions, the net magnetization is $M \simeq \pi a J_c/640$ (in practical units). We measure typical remanent magnetizations of 45 emu/cc at 5 K and 1.4 emu/cc at 76 K, comparable to values in the literature.[16] Identifying the regions with the observed grains of 10 μm average diameter, $J_c \approx 10^7$ A/cm^2 at 5 K and 3×10^5 A/cm^2 at 76 K, which agree remarkably well with most crystalline and epitaxial film data.[3,5] Further support for grain-sized regions comes from reports that the magnetization scales with the average grain size.[16] Note that interpretation of the magnetization J_c in terms of twin-domain-sized regions would imply J_c's of $\sim 10^9$ A/cm^2, 2 orders of magnitude higher than have ever been seen in these materials. In fact, the values thus implied meet or exceed the calculated depairing limit on J_c, whereas in practice the limit is never approached due to imperfect pinning.[17]

In conclusion, our data indicate that ceramic $YBa_2Cu_3O_{7-\delta}$ consists of weakly coupled homogeneous superconducting grains with large intragranular critical current densities not limited by Josephson junctions. In this picture, the weakly coupled grains of a ceramic become decoupled in a magnetic field due to the collapse of the intergranular currents.[13] We believe that this "grain decoupling" transition is responsible for the anomalous low-field magnetic properties of the ceramics.[4,9,10,13,16]

The conclusions above must be interpreted, however, in the context of the layered crystal structure of the high-T_c oxides. The coherence length along the c axis is so short[12] that the coupling between layers must be weak regardless of any (001) twin boundaries.[18] The real issue then is whether currents *in the basal plane* are disrupted at the (110) twin boundaries; our data indicate that they are *not*. This conclusion is supported by reported observations in oriented films of Josephson junction characteristics along c, but not in the basal plane,[5] as well as by the reported temperature- and field-dependent single-crystal anisotropy.[3] Also, Esteve *et al.*,[19] using a point contact on ceramic Sr-doped La_2CuO_4, observed sharply defined Josephson junction characteristics indicative of one or at most several naturally occurring junctions in series *inside* the sample (their interpretation). Clearly, such sharp structure is possible only if the interjunction spacing in the ceramic is large enough to result in very few current paths near the probe. Therefore, the 0.1 μm^2 area deduced by these authors from their J_c vs H data cannot reflect the interjunction spacing of a fine-scale twin-boundary network, but most likely reflects junction size as discussed above.

Of course, a system of relatively large homogeneous regions is not as amenable to calculation as a system of very small regions, since one must allow for both shielding effects and variations of the order parameter within a given region.[20] On the other hand, the effort would be worthwhile precisely because the grain-boundary and twin-boundary pictures differ tremendously in the prospects for high J_c. With intergranular junctions, four important conditions for high-J_c ceramic materials are (i) oriented grains, (ii) minimal intergranular impurity phases or dislocations, (iii) low void density, and (iv) reasonably high intragrain J_c at liquid-nitrogen temperatures (see also Ref. 6). With regard to the last point, since the (110) twin boundaries do not form junctions, then their influence on J_c, if any, must be a positive one as pinning centers. The drop in the magnetization J_c with increasing temperature probably reflects either the ability of fluxoids to jump between twin boundaries or mobility of the boundaries themselves, although depairing[17] or the appearance of free mobile fluxoids near T_c (the Kosterlitz-Thouless state)[7] are also possibilities.

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