PHYSICAL REVIEW B VOLUME 37, NUMBER 16 1 JUNE 1988

Exponential H and T decay of the critical current density in $YBa_2Cu_3O_{7-\delta}$ single crystals

S. Senoussi, M. Ousséna, G. Collin, and I. A. Campbell
Laboratoire de Physique des Solides, Université Paris-Sud, Bâtiment 510, 91405 Orsay, France

(Received 13 January 1988)

We report magnetic measurements on single crystals of $YBa_2Cu_3O_{7-\delta}$. The magnetic critical current density in the Cu-O basal planes (1.5×10^6 A/cm^2 at 4.2 K) decreases exponentially with temperature as well as with field for $T \gtrsim 50$ K. This is ascribed to current tunneling through micro-Josephson-junctions. The behavior is radically different from that associated with macrojunctions typical of "granular" samples. It is argued that the anisotropy and the T-H anomalous behavior of J_c are connected with the T dependence and the anisotropy of both the coherence length and the electron mean free path.

"Granular" $YBa_2Cu_3O_{7-\delta}$ and $La_{1-x}Sr_xCuO_4$ ceramics prepared in various metallurgical conditions exhibit a tiny (M-H) magnetic cycle around the origin.[1,2] The latter is generally characterized by a first threshold field $H_{c_1}^w$ (5 to 30 G at 4 K) marking the onset of a regime of "weak" irreversibilities followed by a second threshold field $H_{c_2}^w$ beyond which the curve again becomes reversible and linear. The reversible regime holds up to a third critical field $H_{c_1}^g$ of order 500–600 G above which strong irreversibilities take place.

Resistance measurements performed on the same samples showed that the transport critical current density $J_{c,t}$ was closely related to the low-H cycle and obeyed a Silsbee-like criterion of the form

$$H_{c_1}^w \lesssim 2\pi J_{c,t}/RC \leq H_{c_2}^w ,$$

where R is the sample radius (1 to 5 mm).

The above magnetotransport data generally depended strongly on the macrostructure of the sample and were ascribed to the barriers (hereafter referred to as macro-weak-links) connecting the grain network. The field interval $(H_{c_1}^w, H_{c_2}^w)$ seems to represent a gradual transition from a coherent to a percolative superconducting state. On the other hand, the high-H behavior $(H \gg H_{c_2}^w)$ was found to be governed by the size as well as the internal microstructure of the individual grains acting independently.[1,2]

To get more insight into the coherent superconducting state and avoid the complications from the macro-weak-links we have carried out an extensive investigation of the hysteresis cycle, and the associated critical current density J_c in several (~ 10) single crystals as a function of T, H, and θ, the angular direction of H with respect to the basal Cu—O plane. The results depend strongly on θ but, in this paper, we will concentrate mainly on the case $\theta = \pi/2$ (i.e., H perpendicular to CuO plane). The data differ in many aspects radically from the granular case. At first, no low-H cycle was detected (within ~ 0.5 G) whether H was in or out of the basal plane. Secondly, both J_c and M exhibit an exponential decay over a wide range of H and T. However, at the lowest temperatures ($T \approx 4.2$ K) j_c as deduced from the critical state model[3] for $H \gg H_{c_1}^g$ within the decoupled grains and in the present monocrystals are comparable (few 10^6 A/cm^2).[4] The same is true for H_{c_1} (600 G within 100 G in the two cases at 5 K). That the apparent H_{c_1} in granular and in single crystals have comparable values suggests strongly that the surface barriers opposing the entry of flux within the particles is vanishingly small.

The crystals are grown from flux ($BaCuO_2$+CuO) between 1000 °C and 880 °C. They were then reannealed in an oxygen flow at 900 °C and cooled down to 300 °C in three days. This yielded crystals in the shape of rectangular parallelepipeds with typical dimensions of order 0.5 to 1 mm in the basal (Cu—O) plane and 0.1 to 0.4 mm in the perpendicular direction ($\mathbf{H} \parallel c$ axis). The critical temperature as deduced from magnetic data varied from ~ 80 to 90 K and depended on the thermal treatment in a nonpredictable way, perhaps related to copper [Cu(1)] deficiencies.[5]

Figure 1 shows a set of hysteresis cycles at various temperatures ($T = 4.2$ to 31 K) in fields varying from ~ -30 to $+30$ kG. We note a global and very rapid fall off of the magnetization with increasing temperature whereas the variation of M with H is rather slow. As compared with ceramic samples $YBa_2Cu_3O_7$ and $La_{1-x}Sr_xCuO_4$ (Refs. 1 and 2) the most important features of the isotherms of Fig. 1 are as follows. (i) A very high value of the magnetization (~ 1000 against 50 to 100 emu/cm^3 for granular $YBa_2Cu_3O_7$ at 4.2 K). (ii) A second significant result is the persistence of some hysteretic effects at 4.2 and 7 K upon cycling the field from ~ -30 to $+30$ kG (this is not a trivial effect and implies that the critical current density is probably pessimistic and lower than that predicted by the critical state model of Bean.[3] (iii) As already mentioned, we find that the initial magnetization branch is remarkably linear and reversible up to ~ 550 G (after correction for the demagnetizing field).

The evolution of the magnetic loops at higher temperatures ($50 \leq T \leq 90$ K) is presented in Fig. 2. As can be seen the M vs H relationship is strikingly different from that corresponding to Fig. 1 for which $T \leq T_c/2$.

At first, we observe a marked depression[6] of the magnetic signal near $H = 0$. Second, as T approaches T_c (not shown) the depression disappears gradually in such a way that the curve becomes highly reversible near $H = 0$. However, this effect has been also observed in ceramics[1,2] but over a wider temperature range. It is a further signature that the surface hysteresis is vanishingly low[7] at high

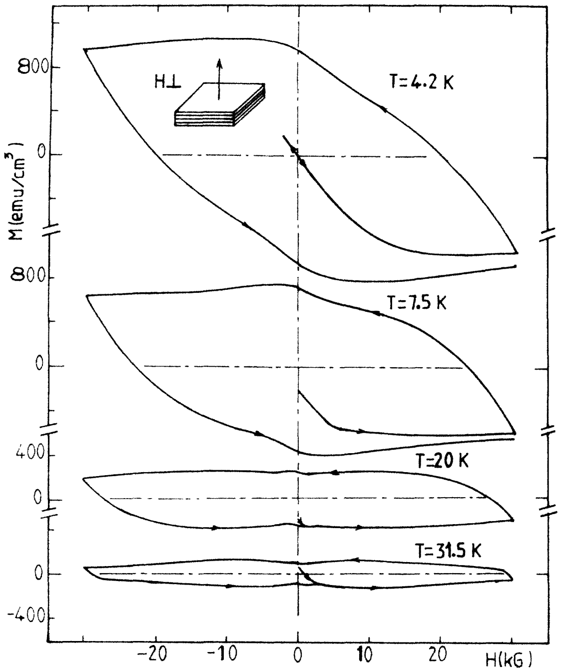

FIG. 1. The magnetization as a function of H for the indicated temperatures. The field is perpendicular to the basal planes.

enough temperature. Third, at sufficiently large field the curves (Fig. 2) become again reversible above some threshold (but ill defined) value $H^g(T)$. A critical field of that kind has already been observed both in $La_{1-x}Sr_xCuO_4$ and in $YBa_2Cu_3O_7$ ceramics but at temperatures notably nearer to the transition temperature T_c than in the present case.

According to the critical state model of Bean,[3] $H^g(T)$ is the field at which the pinning forces become vanishingly small or equivalently it is the field limit above which the critical current density J_c becomes immeasurably low. More generally, in this model, the relationship between J_c and M is given by the formula

$$J_c = 15[M_+(H) - M_-(H)]/\langle R\rangle , \qquad (1)$$

where $\langle R\rangle$ is the average radius of the single crystal assumed to be in a coherent or homogeneous superconducting state. $M_+(H)$ and $M_-(H)$ are the algebraic values of the magnetization at H on the ascending and descending branches, respectively, of the magnetic cycle (i.e., the distance between the two branches at H) in the field region $H \gg H_{c_1}$.

From Fig. 3, it is seen that the magnetic critical current density at the lowest temperature (4.2 K) is very high and nearly field independent in the ($T \leq 40$ K). The high J_c value reported here is consistent with previous data both for single[8] and polycrystalline materials[1,2] (using the radius grain).

The next important property which can be deduced from Fig. 3 is the strong dependence of J_c upon the applied field for $T \gtrsim 50$ K. At first we observe that J_c goes through a maximum at some field H_m of order 3 kG at 50 K. Such a maximum could at first sight suggest that the pinning forces fixing the vortex lines pass also through a maximum near $H = H_m$. However, it is to be recalled that the critical state predictions strictly apply in the limit $|H| \gg H_{c_1}$. So, as for the magnetization curves it is likely that as $|H|$ is decreased towards H_{c_1} a growing frac-

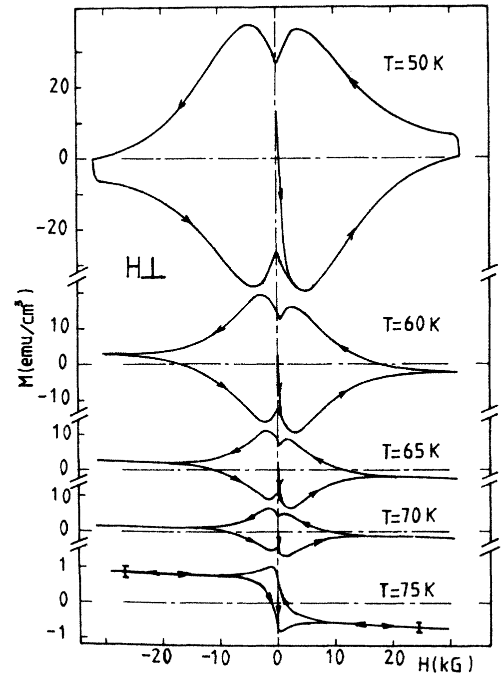

FIG. 2. A set of hysteresis cycles at high temperatures with H perpendicular to the basal planes. The inset is an expansion of the cycle near $T_c \approx 86$ K.

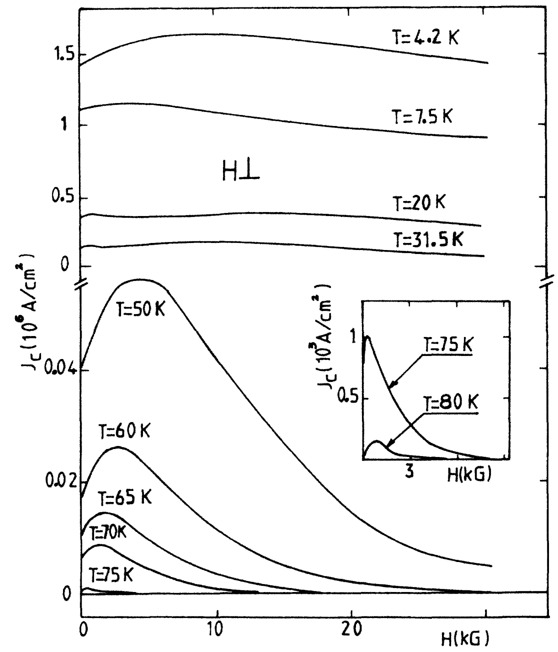

FIG. 3. The critical current density deduced from magnetization data. The inset is an expansion of the curves near $T_c \approx 86$ K. Note that J_c appears to vanish at $H = 0$ for $T \gtrsim 80$ K. This is because the M-H curve is completely reversible near $H = 0$ (see Ref. 7).

tion of the magnetic flux trapped at high field is expelled from the sample and this phenomena is gradually enhanced as T approaches T_c.

Considering the high-field behavior ($H > H_m$) of the critical current density we find that (see inset to Fig. 4) the J_c vs H law follows an exponential behavior with a good precision.

The temperature variation of the critical current density J_c at some fixed fields (2.25, 15 kG) is depicted in Fig. 4 in a semilog scale. The most striking feature of the figure is the nearly exponential drop of J_c over a wide range of temperature except near T_c where J_c obeys a power law.

It turns out that there are essentially two possible origins for the anomalous behavior of J_c as a function of H and T. These are either the conventional flux pinning flux creep mechanism of Abrikosov's vortices or the less conventional tunneling process[9-11] across some microjunctions or micro-weak-link within the single crystal. Both phenomena could lead to new effects because of the high temperatures involved in the new superconductors which enhance the role of thermally activated phenomena and because of the very short coherence length which favors tunneling effects. What is important here is that our present data seem to allow distinguishing between the two mechanisms. The pinning force is generally exceedingly complicated to handle but it is generally accepted that in the case where the defects have dimension of order ξ the pinning force per unit volume acting on the vortex lattice is of the form

$$F_c = \mu_0 \xi H_c^2 \left[1 - \frac{B}{B_{c_2}} \right] \Big/ \phi_0 - k_B T \ln \left[\frac{v_c}{v_0} \right] \Big/ x_p V_p \,. \qquad (2)$$

Here X_p is the effective volume of the flux "bundle" X_p ($\sim \xi$) is some characteristic length relating the force and the energy, H_c is the thermodynamic field, and v_c/v_0 is a reduced drift velocity[12] above which the displacement of the vortex lines becomes appreciable on the experimental time scale.

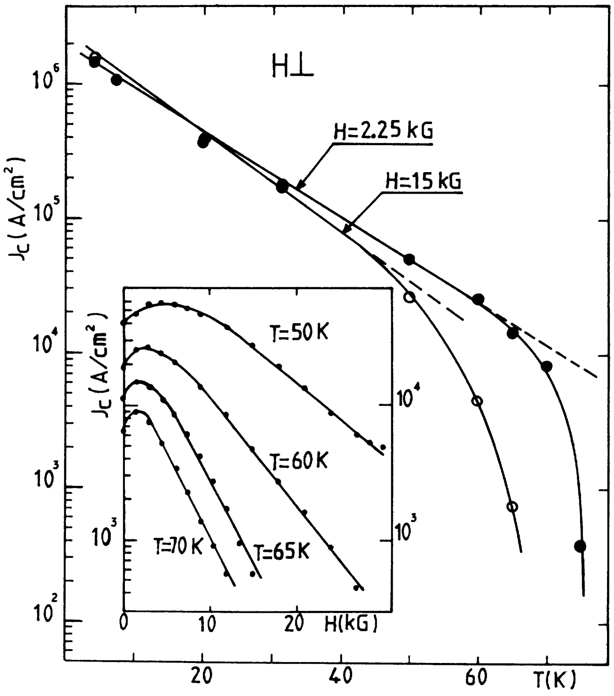

FIG. 4. Temperature variation of J_c plotted in a semilog scale. The inset represents J_c as a function of H also in a semilog scale. The deviation from linearity near $H = 0$ is probably an artifact because the condition $H \gg H_{c_1}$ is not satisfied (Ref. 7). The lines are guides for the eyes.

It can easily be shown by inspecting formula (2) that j_c should follow a power law with both T and H, in contradiction with the present data. It is to be emphasized however that the depinning force could be radically modified for a number of very typical reasons.

(i) The size of the vortex core is comparable with that of dislocation cores (which is generally of the order of two to four Burgers vectors) and other extended defects. This together with the unusually high-temperature values might favor considerably flux creep phenomena through displacement of dislocations, as already suggested by Frank for conventional superconductors and other avalanch phenomena.[12]

(ii) It has been reported recently[13,14] that some kind of order-disorder transition involving either twin boundary or oxygen vacancies (or both) takes place in $YBa_2Cu_3O_{7-\delta}$ around 239 K.

Obviously, this should imply that the associated activation energies must be of the same order as $k_B T_c$ and would therefore favor considerably "Frank"-like depinning mechanisms. Nevertheless, there is not yet any definite evidence for such phenomena.

A more natural interpretation of the present data can probably be found in the glassy picture[9-11] in which J_c would be governed by microstructural defects such as dislocations (the core of which is several lattice spacings large) behaving as internal "micro" superconductor-normal-metal-superconductor junctions. Then, it turns out that J_c would depend on the temperature as $J(T)\exp[-(T/T_0)^n]$ where $J(T)$ obeys a power law the exact form of which depends on the ratio l/ξ_N of the electron mean free path to the coherence length within the junction. The exponent factor n also depends on that ratio whereas the temperature factor T_0 is proportional to the Fermi velocity V_F. On the other hand, J_c is expected to vary exponentially[15] with the product $D(T)H$ where $D(T)$ is the diffusion length. It is to be emphasized that $D(T)$ (which depends on l) is probably strongly anisotropic and temperature dependent as is the resistivity ρ.

This is radically different from the case of more usual type-II superconductors for which the residual resistivity dominates totally the temperature-dependent term for $T < T_c$. It is equally probable that the anisotropy of V_F must be reflected in the exponential factor T_0. These anisotropic effects could explain the enormous T dependence of the anisotropy[8] of J_c.

In conclusion, we have carried out detailed investigations and analysis of the magnetic behavior of $YBa_2Cu_3O_7$ single crystals and argued that these are governed mainly by intragrain microjunctions. However, we are aware that there is probably no net distinction[16] between the latter and pinned Abrikosov's vortices: the apparent difference would depend on the experimental conditions and the techniques used. The matter is perhaps somewhat different for the macrojunction in granular systems the lateral dimensions of which should be very large compared to $\xi(T)$ except very near to T_c or at very high field[17] where most of the macrojunctions separating the grains are broken.

---

[1] S. Senoussi, M. Oussena, M. Ribault, and G. Collin, Phys. Rev. B **36**, 4003 (1987).

[2] S. Senoussi, M. Oussena, and S. Hadjoudj, J. Appl. Phys. (to be published).

[3] W. A. Fietz and W. W. Webb, Phys. Rev. **178**, 657 (1969).

[4] A. Umezawa, G. W. Crabtree, J. Z. Liu, T. J. Moran, S. K. Malik, L. H. Nunez, W. L. Kwok, and C. H. Sowers (unpublished).

[5] G. Collin, A. C. Audier, and R. Comes, J. Phys. (Paris) **49**, 383 (1988).

[6] The relationship $f(M^+,M^-)$ of Eq. (1) assumes that J_c within a radial section of the sample is constant in magnitude and direction and assumes $H \gg H_{c_1}$. Equation (1) assumes also that the sizes of the structural defects are small enough not to break the sample in several independent grains. Otherwise J_c deduced from Eq. (1) would be lower than the optimum value.

[7] A. M. Campbell, in *Proceedings of the Eighteenth International Conference on Low Temperature Physics (LT-18)* [Jpn. J. Appl. Phys. **26**, Supl. 26-3, 1115 (1987)].

[8] T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. **58**, 2687 (1987).

[9] G. Deutscher and K. A. Müller, Phys. Rev. Lett. **59**, 1745 (1987).

[10] M. Oussena, S. Senoussi, and G. Collin, Europhys. Lett. **4**, 625 (1987).

[11] C. Giovannella, G. Collin, P. Raoult, and I. A. Campbell, Europhys. Lett. **4**, 104 (1987).

[12] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966), pp. 90-91.

[13] R. N. Bhargava, S. P. Herko, and N. Osborne, Phys. Rev. Lett. **59**, 786 (1987).

[14] M. T. Béal-Monod, J. Phys. (Paris) **49**, 103 (1988).

[15] T. Y. Hsiang and D. K. Finnemore, Phys. Rev. B **22**, 154 (1980).

[16] P. Martinoli, Phys. Rev. B **17**, 1175 (1978).

[17] O. Laborde, J. L. Tholence, P. Lejay, A. Sulpice, R. Tournier, J. J. Capponi, C. Michel, and J. Provost, Solid State Commun. **36**, 877 (1987).