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Low-field intragranular irreversibility line of $Bi_2Sr_2Ca_2Cu_3O_{10}$ high-temperature superconductors determined by intergranular critical-current measurements

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The low-field intragranular irreversibility line of $Bi_2Sr_2Ca_2Cu_3O_{10}$ high-temperature superconductors was obtained from transport critical-current measurements performed on polycrystalline bulk sintered samples with a pronounced granular character. For such samples, the intra- and intergranular critical-current densities are very different, the latter being largely mediated by tunneling. The method is based on the high sensitivity of the intergranular critical current to the local magnetic field at the intergrain contacts. The value of this field is strongly influenced by the grain magnetization and therefore depends on cooling conditions. The irreversibility temperature was determined as the temperature above which the voltage-current characteristics measured with the sample cooled in applied-field and in zero-field-cooling conditions become identical. For the investigated samples, an exponential decrease of the irreversibility field with increasing temperature was observed. A description of the irreversibility line in terms of thermally assisted flux flow is given.

I. INTRODUCTION

The appearance of an "irreversibility line" $T^*(H)$ in high-temperature superconductors (HTSC's), i.e., a line separating in the (H,T) plane the region where the material shows reversible magnetization from that in which the magnetization depends on the previous path in this plane, was reported by Müller, Takashige, and Bednorz[1] for $La_{1.85}Ba_{0.15}Cu_2O_4$. In this case the irreversibility line has the form

$$1-T^*(H)/T_c \propto H^{2/3} , \tag{1}$$

where T^* is the irreversibility temperature and T_c is the bulk critical temperature in zero applied magnetic field. Such a relation resembles known phenomena in spin-glass physics and was interpreted in terms of the superconducting glass state,[1-3] assuming the existence of intragranular weak links.

On the other hand, Yeshurun and Malozemoff[4] derived for $YBa_2Cu_3O_7$ single crystals a similar $T^*(H)$ variation in the classical flux-pinning picture, considering the limitation of the critical-current density by giant thermally activated flux creep. The investigation of the $T^*(H)$ dependence for various HTSC's can give information concerning the actual limiting factor of the critical-current density in these materials.

It was reported[5,6] that in the case of $Bi_2Sr_2Ca_2Cu_3O_{10}$ (Bi-2:2:2:3) HTSC's the $T^*(H)$ dependence clearly deviates from Eq. (1), following an exponential decrease of the irreversibility field with increasing temperature. De Rango *et al.*[5] interpreted this behavior taking into account the proximity effect in the layered structure of Bi-2:2:2:3 HTSC's. The irreversibility field would be the field value for which the induced superconductivity in the normal regions of the unit cell [the regions between the $(CuO_2)_3$ blocks] breaks down. However, it is not yet clear if these regions act as normal or insulating barriers. Moreover, finite critical-current densities were recently measured in the magnetic-field–temperature domain where this compound should exhibit bidimensional superconductivity.[7]

In our opinion, the existence of various $T^*(H)$ forms encourages an explanation in the flux-pinning picture, where the irreversibility line is expected to be strongly dependent on the temperature variation of the mean pinning-energy barrier. The latter may change significantly with the pinning mechanism details. In the present work, the low-field $T^*(H)$ line of Bi-2:2:2:3 HTSC's was determined using ceramic samples and transport critical-current measurements. A description of the irreversibility line in terms of thermally activated flux motion at low driving forces is given.

II. SAMPLE PREPARATION AND CHARACTERIZATION

Samples of nominal composition $Bi_{2.8}Pb_{1.2}Sr_4Ca_4Cu_{7.2}O_{10}$ were prepared by the conventional ceramic method with a final long-time sintering (550 h) in air, at 845 °C. The x-ray powder diffraction diagram indicated a large amount of high-T_c Bi-2:2:2:3 phase and the low-T_c Bi-2:2:1:2 compound as the second major phase. Scanning-electron-microscopy (SEM) and electron-diffraction studies revealed flat crystallites of mean dimensions $5\times5\times1$ μm^3 (sluggish growth along the c axis), having a small degree of preferential orientation with the (a,b) plane perpendicular to the direction of compression before the final sintering. The measured samples show zero electrical resistance at $\simeq 104$ K and $T_c^{\text{midpoint}} = 109.4$ K (Fig. 1). The multiphase nature of our samples and a relatively low mass density (3.7 g/cm^3) enhance their granular character. The transport critical-current density is rather low (20–50 A/cm^2 in zero field and at liquid-nitrogen temperature) and decreases strongly in applied field (Fig. 2). Using the Bean model,[8] the

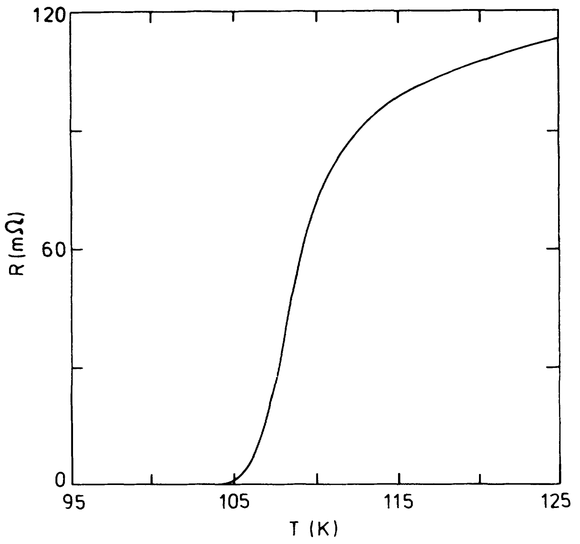

FIG. 1. Typical resistive-superconducting transition of the investigated samples in zero applied magnetic field.

hysteresis in the dc magnetization curve at liquid-nitrogen temperature (Fig. 3) gives relatively high intragranular critical-current densities in low applied fields ($\simeq 2\times10^4$ A/cm^2 at 200 Oe).

III. DETERMINATION OF THE IRREVERSIBILITY LINE

The irreversibility line of Bi-2:2:2:3 HTSC's was determined in Ref. 6 from zero-field-cooling (ZFC) and field-cooling (FC) susceptibility measurements. For an applied field H, the temperature-dependent ZFC and FC susceptibility curves converge at $T = T^*(H)$, yielding a point on the $T^*(H)$ line. However, the two susceptibility curves approach each other somehow asymptotically, and the determination of the intersection point introduces appreciable errors. At constant temperature the irreversibility field can also be obtained as the field value where the ZFC magnetic moment of the sample equals its Meissner signal (i.e., the FC magnetic moment, Fig. 3). Besides the above impediment, the time dependence of the magnetization and the difficulty of maintaining the apparatus zero point for a long time in classical dc magnetization measurements create new problems.

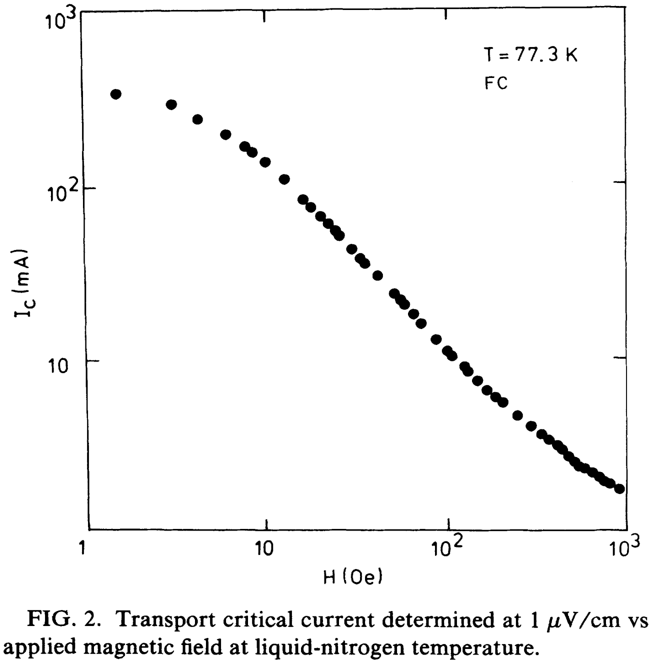

FIG. 2. Transport critical current determined at 1 μV/cm vs applied magnetic field at liquid-nitrogen temperature.

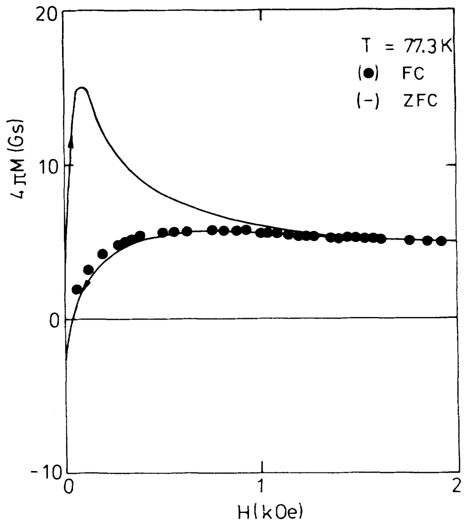

FIG. 3. Magnetization of the sample (in powder form) vs applied field in different cooling conditions at liquid-nitrogen temperature.

In this work we used a method in which the accuracy in the determination of the irreversibility temperature is improved. As shown below, when the irreversibility line is reached we have two curves which must be identical, these curves involving variables other than T or H. The proposed procedure is applicable for bulk sintered samples with a pronounced granular character. For such samples the transport critical current is limited by Josephson junctions located at grain boundaries, which are very sensitive to applied magnetic fields.[9] On the other hand, the intragranular critical-current density is relatively high, leading to appreciable differences between the grain magnetization in ZFC conditions and for increasing field (pinning to flux entry) and that appearing in FC conditions (paramagnetically trapped flux at pinning centers). At the same time, the local magnetic field at the intergrain contacts is directly related to the grain magnetization.[10,11] Taking into account the shape of the grains, implying a demagnetizing factor close to unity in the perpendicular field, the local field at the intergrain contacts will depend strongly on cooling conditions. The above differences vanish when the irreversibility line is reached, and in this way one can determine $T^*(H)$ as the temperature above which the voltage-current (V-I) characteristics obtained with the sample cooled in applied field and in ZFC conditions become identical (Figs. 4 and 5). The

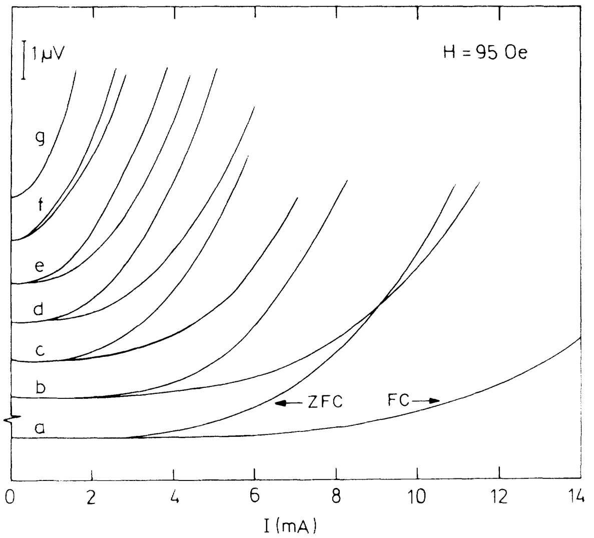

FIG. 4. Low-voltage-level *V-I* curves obtained with the sample cooled in applied field and in ZFC conditions for $H=95$ Oe: (a) $T=77.3$ K, (b) $T=81.35$ K, (c) $T=85.5$ K, (d) $T=88$ K, (e) $T=90.3$ K, (f) $T=93.25$ K, and (g) $T=96.55$ K.

degree of preferential crystallite orientation is not very important in this context, the irreversible magnetization having a large component parallel to the *c* axis whatever the field direction, except for angles very close to the (a,b) plane.[12]

In our experiments the FC and ZFC conditions were first realized at liquid-nitrogen temperature, in magnetic-field values up to 500 Oe generated by a copper solenoid. Then the sample was heated in steps of 0.5 K or less, and the temperature was stabilized with an accuracy better than 0.05 K within $\simeq 2$ min, using an Oxford Instruments precision temperature controller. The *V-I* curves were obtained in the geometry with the magnetic field perpendicular to the transport current and to the plane of preferential crystallite orientation, at a constant sweeping rate of $\simeq 1$ mA/sec. The distance between the potential leads was $\simeq 1$ cm. The samples were maintained in a helium-gas atmosphere, and in order to avoid heating due to the transport current, low-electrical-resistance contacts with a specific resistance of the order of 10^{-4} Ω cm^2 at room temperature were performed using high-temperature silver paint and adequate short-time diffusion heat treatments. In Figs. 4 and 5, a few pairs of *V-I* characteristics are presented. Repeated experiments give similar results, which indicates a negligible influence of flux jumps at high temperatures.

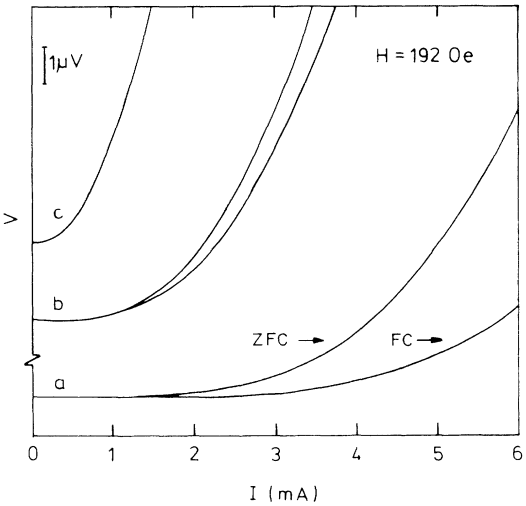

FIG. 5. Low-voltage-level *V-I* curves obtained in ZFC and FC conditions for $H=192$ Oe: (a) $T=77.3$ K, (b) $T=86.05$ K, and (c) $T=91.3$ K.

IV. RESULTS AND DISCUSSION

The low-field irreversibility line of our Bi-2:2:2:3 HTSC's can be fitted by the expression

$$(T_c - T^*)/T^* = C\ln(H/H_0)\ , \tag{2}$$

with $H_0=16.5$ Oe and $C=8\times 10^{-2}$ (Fig. 6). While significantly different from Eq. (1), we show below that such a relation can be obtained in the flux-pinning picture.[13-16]

The electric field E generated by thermally activated flux creep inside the grains is given by

$$E=2dvB\exp\left[-\frac{U}{k_BT}\right]\sinh\left[\frac{BJVr}{k_BT}\right]\ , \tag{3}$$

where J is the intragranular current density related to the irreversible magnetic properties of the grains, U a mean pinning energy barrier for a flux bundle of volume V, r the range of the pinning potential, v an attempt frequency, d the hopping range, and B the magnetic induction. Although the magnetic phenomena should be most suitably described in terms of magnetic diffusion,[14] we adopt-

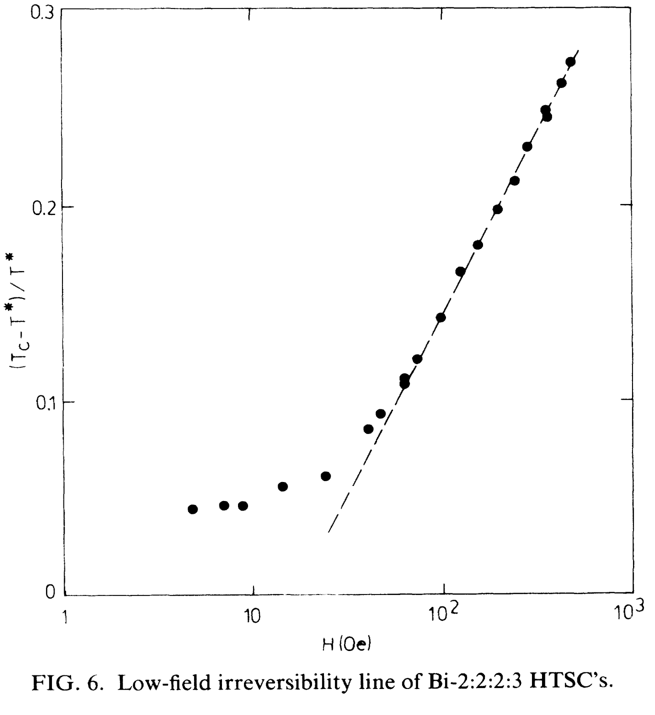

FIG. 6. Low-field irreversibility line of Bi-2:2:2:3 HTSC's.

ed the analog formalism[16] using the above $E(J)$ relation, which, in the limit of small driving forces $(BJVr < k_B T)$, becomes

$$E = \frac{2d\nu B^2 JVr}{k_B T} \exp\left[-\frac{U}{k_B T} \right] . \tag{4}$$

In order to simplify the discussion, we assumed a single barrier height. Equation (4) can be related to the reversibility transition by imposing that the resistivity due to thermally assisted flux flow,[14] $\rho = E/J$, which is strongly temperature dependent, takes a critical value ρ_c that allows the equilibrium mixed state for the grains to be reached in the time scale of the experiment.

The flux-pinning mechanism in Bi-2:2:2:3 HTSC's, as well as in all HTSC's, is not yet known in detail. However, the absence of twin boundaries and the low stacking-fault density in our samples[17] justify considering, in the geometry with the applied field perpendicular to the (a,b) plane, pinning by voids, treated as point defects. In the low-field limit, the collective pinning theory[18] gives

$$U(B,T) = U_0(1 - T/T_c) , \tag{5}$$

with $U_0 = \text{const}$. The fact that U does not depend on the field seems natural for isolated vortices. For defects with the relevant dimension smaller than the Ginzburg-Landau coherence length $\xi(T)$, $r \simeq \xi$. Also, in the amorphous limit,[19] $V \simeq a_0^2 L_c$, where the vortex-lattice parameter $a_0 \simeq (\phi_0/B)^{1/2}$ (with ϕ_0 the magnetic-flux quantum), and L_c is a coherence length along the flux line. The layered structure of Bi-2:2:2:3 HTSC's implies a very small L_c value, probably of the order of the distance between the $(CuO_2)_3$ blocks. With these considerations, taking the hopping range $d \simeq \xi(T) = \xi_0(1 - T/T_c)^{-1/2}$, Eq. (4) leads to a $T^*(H)$ relation of the form

$$(T_c - T^*)/T^* = \frac{k_B T_c}{U_0} \ln \left[\frac{H}{H_0(T^*)} \right] , \tag{6}$$

where

$$H_0(T^*) = \frac{\rho_c k_B T^*(1 - T^*/T_c)}{2\mu_0 \phi_0 \nu L_c \xi_0^2} . \tag{7}$$

Equation (6) is a good agreement with the experimentally determined $T^*(H)$ line [Fig. 6, Eq. (2)]. The deviation close to T_c appears as a result of the decrease of $H_0(T^*)$ as $T^* \to T_c$. Below $\simeq 100$ K, the temperature dependence of the logarithm can be neglected, and Eqs. (2) and (6) give $U_0 = k_B T_c / C = 120$ meV. Equation (5) thus leads to $U(T^*)/k_B T^* \lesssim 1$ in the considered temperature range, which represents a sufficient condition for the low-driving-force regime to apply. Considering typical values, $\xi_0 \simeq 10$ Å, $\nu \sim 10^{11}$ Hz, and $L_c \simeq 10$ Å, for $H_0 = 16.5$ Oe (1313 A/m) and $T^*(1 - T^*/T_c) \simeq 10$ Eq. (7) gives $\rho_c \sim 10^{-12}$ $\Omega\,m$, which is a plausible value for the thermally assisted flux-flow resistivity.

V. CONCLUSION

The low-field intragranular irreversibility line of $Bi_2Sr_2Ca_2Cu_3O_{10}$ high-temperature superconductors was obtained using polycrystalline bulk sintered samples and transport critical-current measurements. The method is based on the high sensitivity of the transport critical current to the magnetic field at the intergrain contacts. The latter is directly related to the irreversible magnetic properties of the grains, and this allows one to determine the irreversibility temperature as the temperature above which the voltage-current characteristics measured with the sample cooled in applied field and in zero-field-cooling conditions become identical. For the samples investigated, the observed exponential decrease of the irreversibility field with increasing temperature was explained in terms of thermally assisted flux flow.

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