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Relaxation of the transport critical current in deoxygenated $YBa_2Cu_3O_{7-\delta}$

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Abstract

We present a study of the transport critical current relaxation for a set of carefully deoxygenated $YBa_2Cu_3O_{7-\delta}$ polycrystalline samples, in such a way that the oxygen losses mostly come from the intergrain region, while the grains remain essentially unchanged. The dependence of the relaxation rates on the maximum magnetic fields applied to the sample, in the remanent state, is consistent with the predictions of a phenomenological model, in which the thermally activated relaxation of the intragrain magnetization provokes a time decay of the intergrain local fields, thus producing a temporal increase of the transport critical current. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Critical currents; Polycrystals; $YBa_2Cu_3O_{7-\delta}$; Flux creep

1. Introduction

The study of the relaxation of the magnetization in the high-T_c superconducting oxides is a frequent and useful approach to characterize the vortex dynamics (particularly flux creep effects), and the role of different types of structural defects as pinning centers. However, the time evolution of the transport critical current I_c, which is closely connected with the flux creep of the intragrain magnetization in polycrystals, has received by itself much less attention [1,2]. It is commonly accepted that the relaxation of the flux trapped in the grains changes the effective magnetic field at the weak links connecting them, thus influencing the transport current through the sample [3,4]. A systematic study of the time variation of I_c can be conducted by measuring the voltage at the critical current, after applying a given magnetic history to a properly chosen set of samples. Recently, we found that the transport critical current in polycrystalline high-T_cs increases logarithmically in time, while the relaxation rate dependence on the maximum applied field H_m shows a broad maximum at low fields [5]. Although these results were reasonably described by a phenomenological model in that work, further analysis is needed to understand the peculiar behavior of relaxation rate with the magnetic history. In the present paper, we study the time evolution of the transport critical current I_c, in a set of carefully deoxygenated

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polycrystalline samples of the system $YBa_2Cu_3O_{7-\delta}$, in such a way that the oxygen losses mostly come from the intergrain region, while the grains remain essentially unchanged. Our measurements were performed using a PI controller, which allows us to collect thousands of values of I_c per hour. The aim is to characterize the I_c relaxation behavior, and its relation with the magnetic history, in samples with the same microstructure but with weak-link networks of different quality, looking for a comprehensive description of this process in bulk samples.

2. Experimental

$YBa_2Cu_3O_{7-\delta}$ samples were obtained by a standard solid-state reaction method. Stoichiometric amounts of high purity Y_2O_3, $BaCO_3$ and CuO were well mixed and pressed into pellets. The samples were sintered at 950°C for 16 h and slowly cooled (1°C/min) down to room temperature in an oxygen flow. Three intermediate treatments at 900°C, 920°C and 940°C for 16 h each were performed, followed by fine crushing. A bar of approximately $10 \times 1 \times 1$ mm^3 was cut from one of the pellets.

The sample in as-sintered conditions will be referred as A_1. Once the transport measurements were performed, the A_1 bar was introduced for 5 min in a long tubular furnace, previously stabilized at 300°C in air, (i.e., placed in the temperature plateau of the open furnace) and then quenched to room temperature. The resulting sample is denoted by A_2. After a new set of measurements, the A_2 bar was annealed in air at 300°C for 25 min as described above and quenched to room temperature; this new sample is called A_3. The selection of this temperature and of the short annealing times is discussed below. Isothermal thermogravimetric (TG) measurements at 300°C in air were performed with a heating rate of 10°C/min, until the preset temperature is reached.

The resistive transition was measured by the four probe method using silver painted contacts. A bias ac current of 1 mA at a frequency of 80 Hz was applied through the outer pads, while the voltage across the inner ones was measured with a lock-in amplifier with a maximum noise of 50 nV. The temperature was dynamically controlled with a resolution of ± 0.02 K.

The critical measurements were performed with a special PI pulsed-current set-up [6]. A sequence of alternate current pulses were injected to the sample in such a way that the PI system dynamically adjusted the height of the pulses in order to keep a constant dissipation of 1 μV (taken as the voltage criterion to define I_c). The voltage was sampled only at the plateau of each current pulse. In this way, I_c at any applied magnetic field, temperature or time was automatically measured just by reading the corresponding bias current. The magnetic field, generated by a long copper solenoid, was oriented perpendicular to the applied current. The maximum field used was $H_m = 500$ Oe with an error of ± 0.3 Oe.

The I_c measurements were performed for different H_m values after the complete removal of the applied field. Once any previous magnetic history of the sample was erased by increasing the temperature up to $1.3T_c$ at zero field, the temperature was stabilized to the chosen measurement value (85 K). The magnetic field was then increased at a rate if 20 Oe/s to the desired value of H_m and suppressed at the same rate. Special care was devoted to guarantee a true zero-field cooling of the sample, within the limitation imposed by the lack of shielding for the Earth's magnetic field. Then, the transport critical current was measured every second during 1 h, resulting in a $I_c(H_m, t)$ vs t curve. The process was repeated for different values of H_m.

3. Results

The isothermal TG measurements at 300°C in air exhibit an initial flat region (including the heating interval), followed by a mass loss section which begins ~30 min after the present temperature is attained. This loss gradually increases with time, until the curve levels off smoothly at an equilibrium value. A_1-type samples annealed in air for times inside the mass-loss interval exhibit severe changes in their resistivity curves. Fig. 1 shows the resistive transitions for samples A_1, A_2

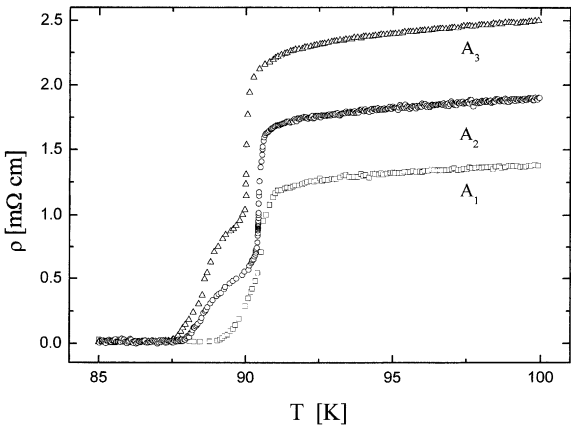

Fig. 1. Resistive transitions for $YBa_2Cu_3O_{7-\delta}$ samples after heat treatments. A_1: sintered at 950°C in oxygen flow and slowly cooled; A_2: A_1 + 5 min at 300°C for 25 min in air and quenched to room temperature.

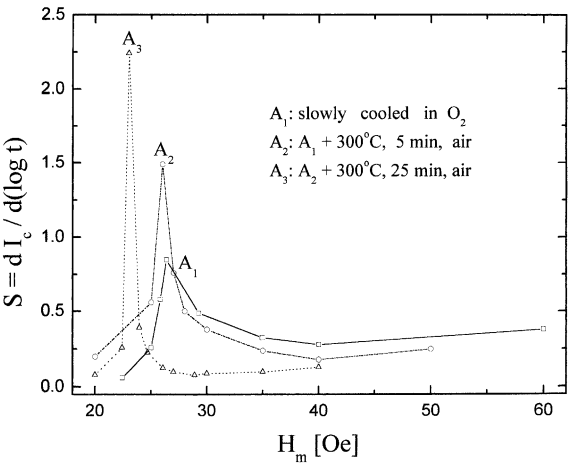

Fig. 2. Relaxation rates $S = dI_c/d(\log t)$ for the $YBa_2Cu_3O_{7-\delta}$ samples under study as a function of the maximum applied field H_m after complete removal. The lines are a guide the eyes.

and A_3. As can be verified, the normal-state resistivity regularly increases and the transitions become broader from A_1 to A_3. The onset critical temperature, T_{co}, taken as the maximum of the derivative of the $\rho(T)$ curves, is the same for A_1 and A_2 within the error of the measurements (90.76 and 90.75 K, respectively), while the zero-resistivity temperature, $T_R = 0$, is clearly lower for A_2 (87.23 K) when compared to A_1 (88.66 K). For A_3, both T_{co} and $T_R = 0$ slightly diminish (90.13 and 87.19 K, respectively) relative to A_1 and A_2.

The relaxation curves for all the samples ($I_c(H_m, t)$ vs $\log t$), measured at 85 K for some of the different applied values of H_m, follow the same behavior as the one of samples measured in Ref. [5]. We verified, through hysteresis measurements of I_c at low magnetic fields (known as "flux trapping curve" [5,7,8]), that the first critical field of the grains at 85 K in our samples is lower than 20 Oe being the same for samples A_1 and A_2 and slightly smaller for sample A_3; so, in all these cases the grains were penetrated to different extents by the external field. As in Ref. [5], the linearity of the curves is remarkable, and the slope, $S = dI_c/d(\log t)$, gives the corresponding relaxation rates. Samples A_2 and A_3 have absolute values of I_c for a given H_m smaller than A_1.

Fig. 2 shows the dependence of S with H_m for the three studies samples. All of them shows a maximum, which becomes more intense and narrow from A_1 and A_2 (26 Oe), while it is slightly displaced to a lower H_m value (23 Oe) for A_3. The high field region of the curves stabilizes at a level, which diminishes from A_1 to A_3, i.e., the reverse behavior of the intensity of the corresponding maxima.

4. Discussion

4.1. Phenomenological model

In this section, we outline how to generate I_c curves as a function of the *effective* magnetic field, H_{eff}, at the intergrain junctions (i.e. not the *applied* field) for networks of different quality in their connectivity. This allow us a more clear interpretation of the I_c relaxation measurements in our samples, based on the phenomenological model reported in Ref. [5].

A widely accepted model to describe the behavior of the transport critical current I_c in a polycrystalline high-T_c superconductor was early proposed by Peterson and Ekin [9]. The key idea is to consider the intergrain junctions embedded into an effective magnetic field H_{eff} resulting from the superposition of the external applied field and the one associated to the magnetization of the neighboring superconducting grains, which depends on

the magnetic history and time. Altshuler et al. [8] and Müller and Matthews [10] further extended the model, allowing the estimation of I_c at a given field after *any* magnetic history, assuming that the grains follow the Bean's critical state model [11]. The resulting I_c is then a function of the maximum applied field, H_m the temperature, T, and the time elapsed since the magnetic field was suppressed, t, and can be written in its normalized form [5], as:

$$I_c(H_m,T,t) = I_c(T)\left|\frac{\mathrm{Sen}(\pi H_{\mathrm{eff}}(H_m,T,t)/H_0(T))}{\pi H_{\mathrm{eff}}(H_m,T,t)/H_0(T)}\right| \quad \text{if } H_{\mathrm{eff}} \leqslant H_0/2 \tag{1.1}$$

$$I_c(H_m,T,t) = I_c(T)\frac{H_0(T)}{\pi H_{\mathrm{eff}}(H_m,T,t)} \quad \text{if } H_{\mathrm{eff}} > H_0/2 \tag{1.2}$$

where H_0 is the effective field value at which the first minimum appears in the I_c vs H "Fraunhofer" pattern of an *average* Josephson junction of the sample [12]; $I_c(T)$ is the temperature-dependent transport critical current at zero field.

Depending on the magnetic history of the sample, H_{eff} is described by different expressions [5,8,10], corresponding to different profiles of intragrain currents, according to the Bean's model. In this way, H_{eff} is expressed in terms of the full penetration field of the grains, $H_g^*(T,t)$, which allows us to introduce the time dependence in Eqs. (1.1) and (1.2). Taking into account that the original Anderson–Kim model cannot properly account for a various features of flux creep in high-T_c superconductors, we use the so called *interpolation formula*, after Blatter et al. [13], for the time dependence of $H_g^*(T,t)$:

$$H_g^*(T,t) = H_g^*(0)\left(1+\mu\frac{K_B T}{U_0}\ln\left(\frac{t}{\tau}\right)\right)^{-1/\mu} \tag{1.3}$$

where $H_g^*(0)$ is the average full penetration field of the grains at the initial instant and $T=0$, k_B is the Boltzmann's constant, U_0 is the average pinning energy of the grains, τ is a characteristic time of the order of 10^{-6} s [14] and μ ranges from 1/7 to 7/4, depending on the dimensionality of the pinning and creep regimes [15,16].

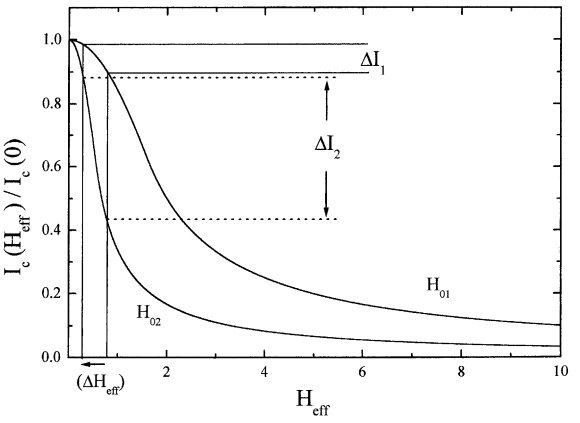

Fig. 3. Normalized $I_c(H_{\mathrm{eff}})/I_c(0)$ curves generated using Eqs. (1.1) and (1.2) (see text), for two weak-link networks with different characteristic fields H_0 for an average junction ($H_{01} > H_{02}$). The corresponding variations in the critical current ΔI_1 and ΔH_{eff}, are indicated for each curve.

Fig. 3 shows two $I_c(H_{\mathrm{eff}})/I_c(0)$ curves generated using expressions 1.1 and 1.2 (for a constant temperature and a fixed time) with $H_{01} > H_{02}$, which will be used later on in the interpretation of our experimental results. We have $U_0 = 0.6$ eV, $H_g^*(0) = 90$ Oe and $\mu = 1$ [5] in Eq. (1.3). A steeper decrease at low fields is obtained for the "weaker" network, represented by H_{02}, while the curve levels off more rapidly than that for H_{01} at higher H_{eff}. The introduction of a parameter as H_0 is particularly convenient, because it allows to characterize the quality of the weak-link network as a whole.

4.2. Analysis of the experimental curves

The increase in the normal-state resistivity values for samples A_2 and A_3, along with the reduction in $T_R = 0$ and the constancy in T_{co} (in addition to decrease of the absolute value of I_c), indicate that the weak-link network is affected by the short-time annealing in air, while the grains remain unchanged. Since no decomposition of the YBCO matrix is observed, the mass loss must be due to oxygen release. Thus, we believe that the features of the resistivity curves for samples A_2 and A_3 are a signature of a very early stage of bulk deoxygenation (i.e., deoxygenation just at the junctions), which magnitude is beyond the sensitivity of our TG analyzer. The emergence of microcracks, due

to rapid warming and quenching, is possible. Both effects, oxygen deficiency and microcracks, lead to a degradation of the weak-link network. We note that, due to the low temperature and the short times of annealing, the original microstructure of A_1 was preserved after the two thermal treatments; so, no changes in I_c are expected from variations in magnetic flux distribution in the link network. Thus, the relaxation of I_c is governed by the one of the grains magnetization, acting on a weakened intergrain network. In the light of the considerations presented in the previous section, the features of the S vs H_m curves should be described through the I_c dependence on H_{eff} displayed in Fig. 3. The curve for $H_{01} > H_{02}$ is associated to the A_1 sample.

For sample A_3, in addition to the increase in the normal-state resistivity and a broader transition, there was also observed a slight reduction in T_{co}, indicating that there begins the removal of some oxygen from the grains, promoted by the increase in the extension of the heat treatment. It is interesting to note that $T_{R=0}$ diminishes to a lesser extent for A_3 in comparison with the corresponding change between A_1 and A_2, suggesting that the weakly bounded oxygen at the intergrain junctions was almost completely released after the 25 min annealing. We also included the analysis of the A_3 sample, because it allows us to follow the regularities in the transport properties.

Once the external field is removed, an H_{eff} will be acting on the weak links, represented by an average junction of characteristic field H_0. This value of H_{eff} depends on the H_m reached in the particular experiments, and on the superconducting properties of the system. According to the corresponding slope in Fig. 3, this magnetic history determines the rate at which I_c will relax. Since the $I_c(H_{eff})/I_c(0)$ vs H_{eff} dependence has an inflection point at low fields, the S vs H_m dependence will exhibit a maximum. This peak will be narrower for $H_{02} < H_{01}$, due to a more pronounced reduction in I_c for a weaker network. This is just the result obtained for sample A_2, as compared with A_1. It is worth to notice that, for both samples, a given value of H_m promotes the same H_{eff} since, as mentioned above, the oxygen content in the grains of these samples is essentially similar. Any possible difference between the H_{eff} values for which the inflection points in the $I_c(H_{eff})/I_c(0)$ curves occur are beyond the sensibility of our measurements. Furthermore, although the rate of change in H_{eff} due to the intragranular creep must be the same for both samples, the associated variation of the current in sample $A_2(\Delta I_2)$ near the inflection point, corresponding to a given reduction in H_{eff}, would be greater than that for sample $A_1(\Delta I_1)$, as indicated in Fig. 3; this implies $dI_2/d(\log t) > dI_1/d(\log t)$, resulting in a more intense peak for S in sample A_2, in agreement with Fig. 2.

As H_m increases, H_{eff} after field removal is higher than that for H_{02}. This means that the baseline of the S vs H_m curves at high fields should decrease as the intergrain transport properties are depleted, again in agreement with the behavior observed in our samples when going from A_1 to A_3.

In the case of sample A_3, in addition to the effects of the deoxygenation treatments, on the intergrain connections, there are also small changes in the grain properties, as reflected by the slight reduction observed in T_{c0}. Deoxygenation also reduces the value of the first critical field H_{clg}, as mentioned above. This implies that, for the same value of H_m, this sample will trap more flux than the A_1 and A_2 ones, thus giving a higher value of H_{eff} at the junctions. This means that the value of H_{eff} for the maximum relaxation rate in I_c (i.e., the inflection point in the corresponding $I_c(H_{eff})/I_c(0)$ curve) would be reached for a lower H_m in comparison A_1 and A_2, which is just the behavior detected, as shown in Fig. 3.

5. Conclusions

In this work, we have been able to tune, through careful deoxygenation treatments, the inter- and intragranular "quality" of a $YBa_2Cu_3O_{7-\delta}$ polycrystal, without disturbing its microstructure, and to study the temporal relaxation of the transport critical current on the resulting set of samples. The results are coherent with the predictions of a phenomenological model in which the flux creep of the intragrain magnetization produces the decay of the intergrain effective field and, thus, a time increase of the transport critical current.

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