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Flux trapping in transport measurements of $YBa_2Cu_3O_{7-x}$ superconductors

A fingerprint of intragrain properties

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A study of flux trapping in polycrystalline $YBa_2Cu_3O_{7-x}$ was made through measurements of transport J_c versus H characteristics. It was shown that most of the trapped flux which provokes hysteresis in such experiments comes from the superconducting grains, and not from persistent loops in the weak link network. The lower critical field of the grains and the intragrain critical current were calculated for oxygenated and partially deoxygenated samples, showing good coincidence with the commonly accepted values. Further evidence of the intragranular character of the flux trapping resulted from the study of the time variation of the voltage associated with flux creep, from which good estimates of the effective pinning energy of the grains were obtained.

1. Introduction

Low field hysteresis in the transport properties such as critical current density and magnetoresistance is now a very well-known phenomenon in high-T_c superconductors (see, for example, [1–3]). The generally accepted explanation was given by Evetts and Glowacki [1] at an early date when they stated that magnetic flux is trapped by the superconducting grains – or by persistent loops in the weak link network (WLN) – when the field is increased to a certain value, and the effect of this trapped flux at the intergrain junctions provokes the observed hysteresis of the transport properties in a decreasing field. Although most of the authors assume that the main contribution of the trapped flux comes from the superconducting grains [2–5,7], others [6] believe that persistent loops in the WLN are entirely responsible for such trapping.

A third mechanism for explaining the hysteresis shown by the $J_c(H)$ curve has been proposed [7] in which the differences between the increasing field and decreasing field curves are due, besides to the intragranular flux trapping, to the difference between the pinning forces relevant to each case, i.e., between the grain surface and grain volume pinning forces, respectively.

Although the measurements of $J_c(H)$ in rotating fields reported early in ref. [1] supplied evidence of the instability of the WLN as a flux trapping element, we give here further reasons to believe that the superconducting grains and not the persistent loops in the WLN are the source of most of the flux trapping relevant to transport measurements. On that basis, estimates of some intragranular properties such as the lower critical field of the grains H_{c1g}, intragrain critical current density J_{cg} and effective pinning energy U, were obtained through the analysis of different transport experiments. The results match very well with the values reported in the literature commonly measured by more conventional techniques.

2. Experimental

Samples were prepared by the standard ceramic technique using high purity Y_2O_3, $BaCO_3$ and CuO reagents. After mixing, powders were heated subsequently at 900°C, 920°C and 940°C in air for 16 h, with the corresponding milling in a rotatory agate mortar between the treatments. Compact disks of 13 mm in diameter and about 1 mm height were pressed at 300 MPa, sintered at 950°C for 16 h and cooled down slowly (1°C/min) to room temperature in an oxygen flow of 28 liters per hour. Densities ranging between 5.6–5.7 g/cm^3 (88–90% of the theoretical one) were obtained, with an average grain size of 10 μm. Several bars were cut from different disks (0.5×0.8×1.2 mm^3 in size) and the value of the critical current density at liquid nitrogen (LN) temperature in zero field, obtained using the four probe technique with silver paint contacts and taking 10^{-5} V/cm as critical current criterium, was $J_c(0)=400$ A/cm^2 with excellent repeatability. We denote this final product as sample I.

Some of the bars obtained as mentioned were partially deoxygenated by applying 400°C for one min in air. As a consequence, the critical current density measured under the same conditions as described above was lowered to 90 A/cm^2. We denote this as sample II.

The following experiments were performed for both types of samples:

(a) $J_c(H', H_0)$ versus H_0 curves.

An external field H_0 was applied perpendicular to the current flow along the longest axis of the samples and then decreased to a certain value H' at which the critical current density was measured. The values $H'=0$, $H'=400$ A/m (5 Oe) were selected. The samples were either zero field cooled (ZFC) of field cooled (FC) for each set of measurements and were sinked into LN directly. The entire process was performed for each value of H_0 previously erasing the magnetic history of precedent measurements by taking the sample outside the dewar for a few seconds in order to exceed the critical temperature. No degradation effects due to thermal cycling were detected when checked at the end of the measurement.

(b) V versus t curves.

A field of 8×10^3 A/m (100 Oe) was applied to the ceramic in ZFC conditions. Then, the field was removed and, after applying a transport current through the sample in order to put it in a dissipative state, the voltage between the inner probes of the four-probe arrangement was measured at intervals of 30 s for a total period of several minutes.

3. Results and discussion

3.1. $J(H',H_0)$ versus H_0 curves

3.1.1. $H'=0$

The upper curve in fig. 1 corresponds to the $J_c(0, H_0)$ versus H_0 characteristic for sample I in ZFC conditions. For a maximum applied field lower than the one labelled H_{1cg} in the figure. $J(0, H_0)$ is a constant. This situation corresponds to a reversible section of the $J_c(H)$ curve. For $H_0>H_{1cg}$, irreversibility takes place: when the external field is completely removed the trapped flux acts on the junctions between superconducting regions, decreasing the maximum critical current through them. There are two possible contributions to flux trapping effects: the flux pinned after penetration into the grains and the flux trapped by persistent loops in the WLN.

If both contribution were present to a similar degree, there should exist two regions in the $J_c(0, H_0)$ versus H_0 characteristic associated with flux trapping: a low field one corresponding to the irreversibility produced by the flux trapped in the WLN loops, and a higher field one corresponding to the flux trapped by the grains. However, a smooth single step was detected within our experimental resolution, with the decrease in $J_c(0, H_0)$ values starting at 1.6×10^3 A/m (20 Oe).

Since the upper limit range of H_0 shown in fig. 1 (12×10^3 A/m or 150 Oe) exceeds the value commonly accepted in the literature for the first field penetration into the grains at LN-temperature [8,9] the detected step should correspond to the first flux

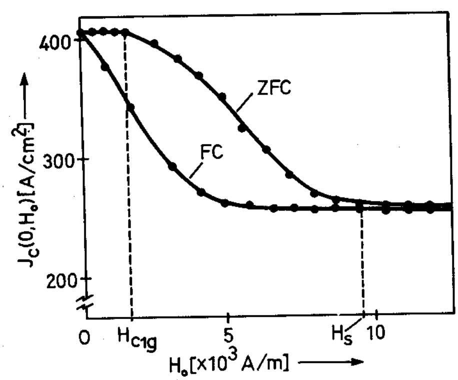

Fig. 1. $J_c(0, H_0)$ vs. H_0 characteristic for sample I.

trapped by the grains, and not by the WLN loops. Another possibility is to have the WLN contribution overlapped with the single step observed, which would imply persistent loops with a very high capacity to trap flux, but the values of the pinning energy reported below are strong evidence against this picture. It is quite unlikely, on the other hand, to have such highly hierarchic WLN in which a well-defined number of loops are "strong flux trappers" and the rest are not.

From the above discussion, we can interpret $H_{c1g}=1.6\times10^3$ A/m (20 Oe) as a lower limit for the first critical field of the superconducting grains. To reach this conclusion, it is important to make clear in what degree demagnetization effects increase the effective field "felt" by the grains with respect to the applied field. Senoussi et al. [10] have pointed out that, in dense ceramics, the effect of the macroscopic demagnetization factor of the sample is much more important than that corresponding to the grains. A simple calculation made for the specific geometry of our experiment [11] gives negligible demagnetization effects, so here the corresponding correction is not necessary.

As seen in fig. 1, there is a field labelled H_s above which the lowering of $J_c(0, H_0)$ saturates for the ZFC curve, indicating that no further flux is trapped once the field is removed. It is also important to notice the coincidence of the ZFC curve and the FC one for values higher than H_s.

In order to interpret H_s physically, let us assume here the validity of Bean's model. Following this, the flux trapping capacity of a grain is saturated if the maximal external applied field reaches a value $H_{c1g}+2H^*$ where H^* is the so-called "full penetration field" of the superconducting grains. That assertion is easily understood with the help of fig. 2. In fig. 2(a) is shown the field profile into a grain for an applied external field $H=H_{c1g}+2H^*$. In figs. 2(b), (c) and (d) can be seen the profile evolution when the external field H is decreased to $H=H_{c1g}+H^*$, $H=H_{c1g}$ and $H=0$ (where $J_c(0, H_0)$ is measured) respectively. For low field experiments, if $H_0>H_{c1g}+2H^*$, the trapped flux profile in $H=0$ is the same as that depicted in fig. 2(d), corresponding to the plateau observed in fig. 1 for $H>H_s$.

It is worth noting that, as pointed out by Chen et al. [12], the usual Bean's model formalisms assume

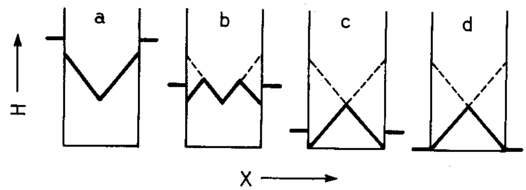

Fig. 2. Evolution of the field profile of a superconducting grain when the external field is decreased from an initial value $H=H_{c1g}+2H^*$ (a), to $H=H_{c1g}+H^*$ (b), $H=H_{c1g}$ (c) and $H=0$ (d).

$H_{c1}=0$, but this does not apply in our case.

From the above discussion, it is clear that for the grains with highest capacity to trap flux

$$H_s=H_{c1g}+2H^*\,, \qquad (1)$$

so, from the data given in fig. 1 we find $H^*=4\times10^3$ A/m (50 Oe). If we now assume the validity of Bean's result [13]

$$H^*=J_c a\,, \qquad (2)$$

and we consider the average half width of our grains, a, to be of the order of 5 μm, we can estimate the intragrain critical current density at LN-temperature of our material as $J_{cg}=8\times10^4$ A/cm^2, in good agreement with the results of Küpfer et al. [14] for polycrystalline $YBa_2Cu_3O_{7-x}$ in zero external field using AC susceptibility techniques and for monocrystals measured under the same conditions in [15] using a vibrating sample magnetometer, which strongly confirms that the grains are the main flux trapping elements in our case.

The flux trapping saturation observed in the FC curve depicted in fig. 1 for $H>\frac{1}{2}(H_{c1g}+H_s)$ strongly confirms our interpretation of H_s. In fact, following eq. (1), $\frac{1}{2}(H_{c1g}+H_s)=H_{c1g}+H^*$, so we expect that, for $H_0>\frac{1}{2}(H_{c1g}+H_s)$, the flux trapped in FC conditions when H is completely removed coincide with the profile shown in fig. 2(d), i.e. saturation begins. This also explain the overlapping of the ZFC and FC characteristics for $H_0>H_s$.

A partial deoxygenation of samples can bring further insight in our investigation, by changing the properties of both the WLN and the grains to a different extent, as we will see.

Figure 3 shows the $J_c(0, H_0)$ versus H_0 curves for sample II. The saturation of the FC curve for

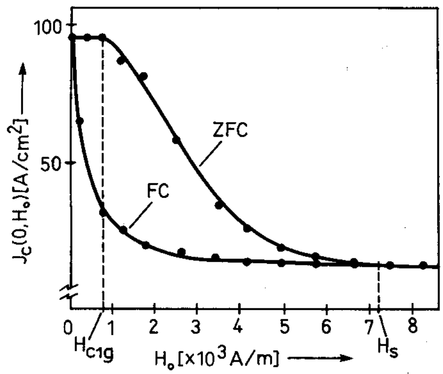

Fig. 3. $J_c(0, H_0)$ vs. H_0 characteristics of sample II.

$H_0 > \frac{1}{2}(H_{c1g}+H_s)$ and the overlapping of the ZFC and FC characteristics for $H_0 > H_s$ are observed again. The estimated values of H_{c1g} and J_{cg} are, respectively, 0.8×10^3 A/m (10 Oe) and 5.6×10^4 A/cm^2. Compared to the ones corresponding to the fully oxygenated sample, both values have decreased about 30% after deoxygenation. However, the absolute value of the transport critical current density at zero applied field, $J_c(0)=90$ A/cm^2, was decreased almost 80%, which clearly indicates that the deoxygenation process has produced much stronger effects on the WLN than on the superconducting grains. On the other hand, the identical decrease in H_{c1g} and J_{cg} after deoxygenation suggests that the oxygen vacancies have not created an important number of new pinning centers or increased their depths, coherent with our flux creep measurements, as we will see. New pinning centers generated by deoxygenation would have either increased J_{cg}, or decreased it in a smaller measure than H_{c1g}. One possibility is, however, that the density of the new centers would only produce a real increase of the pinning force only if higher values of the external field are used. In such a case, Abrikosov's lattice parameter would decrease, allowing more flux lines to be pinned by the extra pinning centers. In fact, Daeumling et al. [15], have stated that minor deoxygenation introduces new pinning centers in $YBa_2Cu_3O_{7-x}$ single crystals. A deeper study on the subject will be published elsewhere.

3.1.2. $H' \neq 0$

Following ref. [1], the flux trapped by the WLN during the field increase disappears when the external field H is reduced to zero due to a "phase slip" process. Taking this into account, we repeated the described process without removing completely the external field but choosing the value $H'=400$ A/m (5 Oe) just looking for the contribution of the flux trapped by the WLN. The inset in fig. 4 illustrates qualitatively the evolution of the hysteretic curves of the critical current densities when the external field is decreased from different maximum fields H_0. The field H' at which the measurement of $J_c(H', H_0)$ was performed is indicated. As in the case of fig. 1, only one step was detected in the $J_c(H', H_0)$ versus H_0 curve shown in fig. 4, and it must correspond to the trapping contribution of the superconducting grains. Furthermore, the values of H_{c1g} and H_s match well with the values extracted from fig. 1. Then, we can conclude that, even when $H'>0$, if some extra trapped flux comes from the WLN, it is very small compared with the contribution of the grains.

3.2. *V* versus *t* curves

Figure 5(a) shows the *V* versus *t* experimental values corresponding to sample I measured with an applied transport current $I=0.8I_c$ after removing an external field of 8×10^3 A/m (100 Oe).

As independently reported by Mathews et al. [4] and Decca et al. [5], this curve is a fingerprint of intragranular flux creep. In zero external field trapped flux is "released" from the grains through a flux

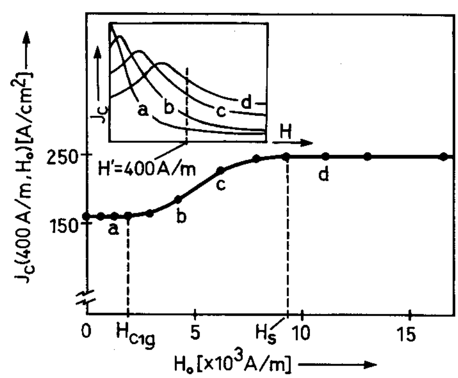

Fig. 4. $J_c(400$ A/m, $H_0)$ vs. H_0 characteristic of sample I. The inset shows four hypothetical decreasing field J_c vs. H curves labelled (a), (b), (c) and (d) which correspond to the identically labelled points of the $J_c(400$ A/m, $H_0)$ vs. H_0 curve.

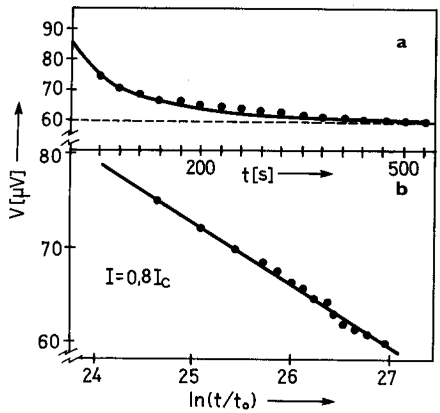

Fig. 5. V vs. t experimental values for sample I measured with $I=0.8I_c$ (a) and V vs. $\ln(t/t_0)$ plot (b) showing the straight line which fits formula (3).

creep process, so the effective magnetic flux field at the junctions decreases with time. If an adequate transport current is applied to the sample in order to produce a dissipation V, it will decrease in time following the law [4]:

$$V(t)=V''\{1-[kT/U]\ln(1+t/t_0)\}\,, \qquad (3)$$

where U is the effective pinning energy, $1/t_0$ an attempt frequency of the order of 10^9 s^{-1} [16] and V'' is the difference between the voltage at $t=0$ and the extrapolation beyond 500 s.

A V versus $\ln(t/t_o)$ plot of the experimental values shown in fig. 5(a) can be seen fig. 5(b) as well as the straight line fitted according to formula (3). If we take $V''=25$ μV by extrapolation, the computed value of U is 30 meV, which is near the lowest values reported in ref. [4] for different transport currents. Our low value of U is due to the relatively high current used ($0.8I_c$) in order to obtain a measurable dissipation.

Figure 6 shows a plot similar to that given in fig. 5(b), but for sample II. Two transport currents were used: $I_1=0.8I_c$ (as in the case of sample I) and $I_2=0.4I_c$. The pinning energy calculated for sample II when I_1 was applied ($V''=45$ μV) was $U=35$ meV, so we can infer that the deoxygenation process did not increase the depths of the pinning centers, which is coherent with our first conclusion from the analysis of the $J_c(0, H_0)$ versus H_0 data for sample II. The pinning energy calculated when I_2 was applied

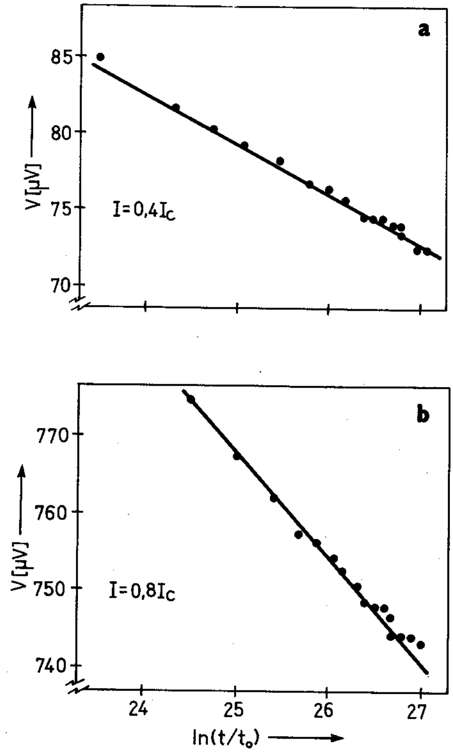

Fig. 6. V vs. $\ln(t/t_0)$ plots for the experimental values for sample II showing the straight line which fits formula (3) for $I=0.4I_c$ (a) and $I=0.8I_c$ (b).

($V''=18$ μV) was $U=70$ meV, in good agreement with the values reported for similar applied currents by ref. [4] and with the value of $U=65$ meV given by Griessen et al. [17] by reanalyzing the data given by Yeshurum and Malozemoff [18] for $YBa_2Cu_3O_{7-x}$ crystals with a small field applied parallel to the C-axis. This is strong evidence of the intragranular character of the pinning energy calculated and then, of the intragranular flux trapping in our experiment.

It is worth noting that, for both samples, two different creep rates were detected, the first one occurring within the first few seconds of the measurement and corresponding to a slightly lower pinning energy. The results presented here correspond to the second regime which is, in our opinion, the one relevant to our practical DC measurements.

Although it can be inferred from the evidence we have presented that the flux trapped by the grains is the cause of the fundamental features of the J_c versus H curves for decreasing fields, we believe, as Evetts and Glowacky do [1], that some moderate amount of flux is trapped by the WLN and is responsible of some minor effects such as the asymmetry of the returning J_c versus H characteristics, which was demonstrated in our case through a simple experiment. The full bell-shaped J_c versus H hysteretic curves (i.e. including a reversal of the external field after its complete removal coming from H_0) were obtained for both samples taking $H_0=2H_{c1g}$. The asymmetry of the characteristics obtained was defined as the percentual difference between the half-widths of the curves at the half-height. The asymmetry was 11% for sample I and only 5% for sample II. Taking into account that the deoxygenation process affects in a higher degree the WLN – as proved above – it is reasonable to say that less flux was trapped by the WLN in sample II and this is the cause of its more symmetrical J_c versus H characteristic for decreasing fields.

4. Conclusions

Evidence was given which points at the superconducting grains as the main flux trapping elements in transport measurements of $YBa_2Cu_3O_{7-x}$ ceramics. The flux trapped by persistent loops in the WLN was proved to influence the J_c versus H characteristics in a much lesser degree, basically provoking the asymmetry of the bell-shaped J_c versus H curves for decreasing fields.

Estimates of the lower critical field of the superconducting grains, intragrain critical current and effective pinning energy were calculated from our transport measurements. Although a systematic comparison with the values measured using more established techniques is needed, it is clear that our experiments provide reasonable estimates of some intragrain properties.

The evolution of such properties after the sample's deoxygenation evidenced that the oxygen removal essentially affected the intergranular material. The related analysis showed good coherence among H_{c1g}, J_{cg} and U for both types of samples.

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