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Transport relaxation and intragranular flux creep in polycrystalline $YBa_2Cu_3O_{7-x}$

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The pinning energies (U_0) of granular $YBa_2Cu_3O_{7-x}$ samples in the presence of a transport current were estimated by measuring the voltage relaxation in the classical four-probe arrangement under a range of different measuring conditions and using different criteria to calculate U_0. The resulting values agree with the ones determined from the magnetization decay, performed on the same samples through vibrational sample magnetometry, and do not show a dramatic dependence on the measuring conditions. Although some basic questions are still open, these results suggest that the transport relaxation technique may constitute an alternative for the estimation of the pinning energy in granular superconductors.

1. Introduction

It is quite well established that the measurement of the time decay of the magnetization can be used to estimate the pinning potential U_0 for high-temperature superconductors [1–4], though some authors [5,6] argue that the obtainment of U_0 through this method is not so straightforward as is generally accepted. Most of these experiments consist of the direct measurement of the magnetization decay, while only a few authors have measured the relaxation of the intergranular effective field associated with the magnetization of the grains in the presence of a transport current as an alternative way of estimating the pinning energy [7–9].

In this work we present relaxation measurements of polycrystalline $YBa_2Cu_3O_{7-x}$ samples in the presence of a transport current, following the method described in refs. [7–9], in order to observe the effect of different measuring conditions on the values of U_0. On the other hand, two different criteria for the derivation of U_0 from the data are examined. Finally, the pinning energies resulting from the transport relaxation experiments are compared with those derived from the magnetization decay obtained through Vibrating Sample Magnetometry (VSM).

2. Transport relaxation and intragranular creep

In order to clarify the foundations of our measurements, let us consider the following experiment. A granular superconductor bar is cooled down below T_c in zero-field cooling conditions (ZFC). An external magnetic field applied perpendicular to the long axis of the sample is increased to a value H_m (higher than the first critical field of the superconducting grains) and then decreased to zero. Assuming that the superconducting grains follows some critical state model, there remains an effective field H_{gj} at the intergrain junctions associated with the remanent magnetization of the grains [10,11]. If a transport current I is then applied along the long axis of the bar, provided $I_{cg} \gg I \gg I_{ct}(H_{gj})$ (where I_{cg} and $I_{ct}(H_{gj})$ are, respectively, the intragranular critical current and the transport critical current at $H=H_{gj}$), it is clear that the voltage dissipation, V, measured in a four-probe arrangement, will be a function of H_{gj} and then of M_r (see fig. 1).

Let us assume that M_r decays as [12]

$$M_r = M_{r0}\left[1 - \frac{kT}{U_0}\ln\left(1 + \frac{t}{\tau}\right)\right], \tag{1}$$

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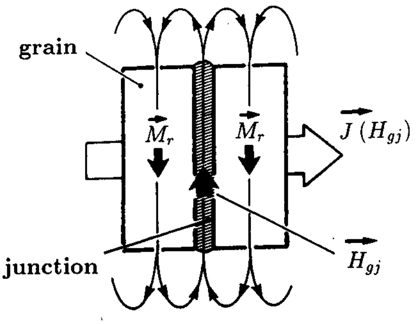

Fig. 1. Simplified view of the relation between the remanent intragrain magnetization, M_r, and the intergrain field, H_{gj}.

where M_{ro} is the remanent magnetization when the external field was taken to zero, U_0 is the pinning energy, τ is the characteristic time relaxation of a flux bundle (of the order of 10^{-6}–10^{-12} s) and k is Boltzmann's constant.

We will make two basic assumptions to establish the relation between the decay of the magnetization of the grains and the voltage decay in our experiment. In the first place, we will accept that $H_{gj}=GM_r$ where G is a geometrical factor depending on the morphology of the sample [11]. In the second place, we will assume that V is proportional to the average H_{gj}.

The second assumption can be understood in the following terms. If the material is modeled as a parallel array of S–I–S junctions, we can expect an inverse proportionality between the critical current and the normal state resistance from Ambegaokar–Baratoff's formula at constant temperature [13,14]. So, if a constant current $I \geq I_c(H_{gj})$ is applied through the sample, the dissipation will be inversely proportional to I_c. On the other hand, provided the condition $0 \ll H_{gj} \ll H_0$ (where $H_0=\phi_0/A$, A being the area of the junction), we can roughly approximate the experimental dependance of I_c on H_{gj} by $I_c \propto 1/H_{gj}$ [15]. Then, we get $V \propto H_{gj}$.

Hence, we can derive from eq. (1) the following expression for the voltage relaxation in the presence of a transport current $I \geq I_c(H_{gj})$

$$V(t)=V_0\left[1-\frac{kT}{U_0}\ln\left(1+\frac{t}{\tau}\right)\right], \tag{2}$$

where V_0 is the voltage at $t=0$ (i.e., when the external field is decreased to zero). Then, we can adopt the following formula for the calculation of U_0 from the experimental data:

$$U_0=kT\left[\ln\left(\frac{t_b}{\tau}\right)-\frac{V(t_b)}{[\mathrm{d}V(t)/\mathrm{d}\ln t]t_b}\right], \tag{3}$$

where $V(t_b)$ is the voltage measured at a certain instant $t=t_b$. This formula is similar to the one proposed by Hagen and Griessen [3] to calculate U_0 from the magnetization decay, provided $V(t)$ and $V(t_b)$ are substituted by $M(t)$ and $M(t_b)$, respectively.

Matthews et al. [7] experimentally found two different regimes in the $V(t)$ decay. The second one (which took place beyond $t \approx 200$ s in their case) is much slower, so they defined a "zero-level voltage", V', as the asymptote of the $V(t)$ curve. Then, the authors substituted V_0 by $V''=V_0-V'$ in formula (2) for their fits. In most of our experiments a slower-decay behavior might be defined beyond approximately 700 s, so a modification of formula (2) according to the criterium of Matthews et al. will be applied in some of the calculations, in order to compare with the results presented in ref. [7]. We will call "criterium 1" the fittings based on formula (2) and "criterium 2" the ones based on the modified formula proposed in ref. [7].

It should be mentioned that some authors [16] interpret the voltage dissipation observed in quite similar experiments as a measure of the number of excited intergranular weak links instead of as a fingerprint of the intragranular creep. The work of D. López et al. [17], however, strongly supports the latter interpretation: passing a transport current through the sample in the four-probe arrangement, they performed measurements of $V(t)$ after applying increasing steps of external field. The voltage decay was observed only when $H > H_{c1g}$, i.e., when the flux lines began to penetrate the superconducting grains.

3. Experimental

Our experiments were performed at liquid nitrogen temperature on two $YBa_2Cu_3O_{7-x}$ ceramic samples, A and B, obtained through the standard solid state reaction technique described elsewhere [9], only differentiated by the milling regimes before sintering. A and B were shaped as bars of dimensions $1\times0.4\times10$ mm^3, and their critical current densities measured through the four-probe technique at zero applied field were 300 and 260 A/cm^2, respectively. The average first critical fields for the superconducting grains (H_{c1g}) were 3.2 kA/m (40 Oe) and 2.4 kA/m (30 Oe), respectively; while the Bean's full penetration fields of the grains (H^*) were 4 kA/m (50 Oe) and 3.6 kA/m (45 Oe), respectively. These parameters were measured using a transport technique described in ref. [9] and checked through VSM. The most probable values of H_0 were determined to be 0.95 kA/m (12 Oe) and 1.1 kA/m (14 Oe) for samples A and B, respectively, by the fitting of the increasing-field dependence of the transport critical current density as described in ref. [18]. The average values of H_{gj}, on the other hand, oscillated in the range 240–477 A/m (3–6 Oe) as implied from the fitting of the hysteretic $J_c(H)$ dependences [11]. Then, we can reasonably accept the condition $0 \ll H_{gj} \ll H_0$ to be fulfilled in our experiments.

The transport relaxation measurements were performed as follows. After ZFC to liquid nitrogen temperature, an external field was applied perpendicular to the long axis of the sample and increased to a certain H_m. After a few seconds the field was taken to zero, and a current $I > I_c(H_{gj})$ applied in the direction of the long axis of the bar trough two silver paint contacts near the edges. Then, the voltage between the inner contacts was measured as a function of time. This experiment was performed on samples A and B for different values of I (at constant H_m) and different values of H_m (at constant I). Between experiments, the magnetic history was erased by taking the sample to $T > T_c$. As in the case of Mota et al. [19], no differences in the decay rate were observed for different waiting times at H_m or for different rates of increasing or decreasing the external field after the first 60 s from turning it off.

Figure 2 shows a typical voltage relaxation curve on sample *A* with $H_m = 12.7$ kA/m (160 Oe) and a transport current of 0.98 $I_c(0)$. A reasonable logarithmic behavior was observed.

In the case of the VSM relaxation experiments, the magnetic field was applied in the same conditions as for the transport experiments. Once taken to zero, the magnetization was measured as a function of time. This sequence of steps was performed on samples A and B for different values of H_m. Between experiments, the samples were warmed up to $T > T_c$.

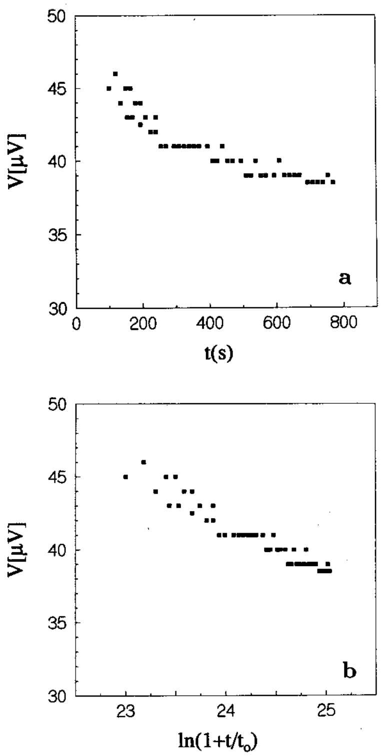

Fig. 2. Linear (a) and logarithmic (b) plots of the voltage decay measured on sample A with $H_m = 12.7$ kA/m (160 Oe) and $I = 0.98\,I_c(0)$.

4. Results and discussion

4.1. Relaxation at different transport currents (constant H_m)

Figures 3(a) and (b) present the pinning energies calculated for samples A and B, respectively, for different transport currents ($H_m = 12.7$ kA/m) through criteria C1 (open squares) and C2 (black squares). We used $\tau = 10^{-8}$ s and $t_b = 120$ s for the calculations. As demonstrated in ref. [3], the choice of τ does not strongly influence the calculation of U_0. It should be noticed that, in the cases when $I \geq I_c(0)$, we inserted $V_{oc} = V_0 - V(I_i)$ instead of V_0 in formula (3), where $V(I_i)$ is the dissipation at the current I_i taken from

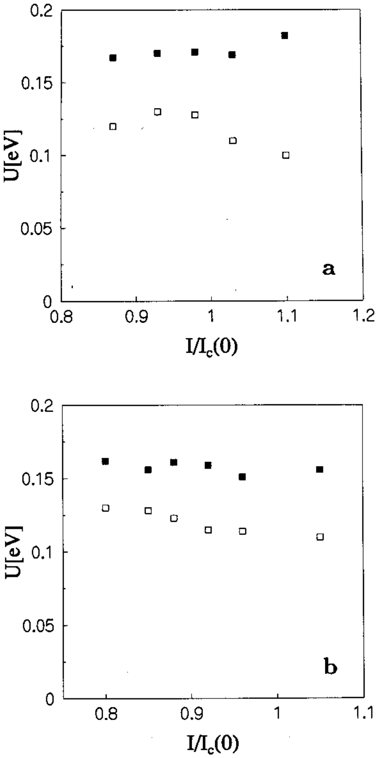

Fig. 3. Pinning energies calculated for samples A (a) and B (b) for different transport currents with $H_m = 12.7$ kA/m (160 Oe). The values calculated following criteria 1 and 2 (see text) are represented by open and black squares, respectively.

the corresponding I–V curve at zero field. This refinement, however, did not change too much the calculated pinning energies (less than 5 percent above the unrectified value for the highest currents used).

The values of U_0 shown in fig. 3 match well with the ones commonly reported in the literature from the magnetization decays for monocrystals (see, for example, ref. [3]). In particular, the ones calculated using criterion 2 (black squares) are closer to the pinning energies reported in ref. [7]. Criterion 1 (open squares) gave systematically lower values of U_0.

Taking into account the relatively large error in the determination of U_0 (estimated to be typically ± 0.02 eV), the pinning energy seems to be quite independent of the applied current, as reported in ref. [7]. At first sight, this would not be expected, since the transport current flowing through the grains could disturb the "pure magnetometric decay" of the grain magnetization by modifying the intragranular magnetic field profile, thus provoking variations in the decay rate not described by formula (1). However, the applied transport currents are typically two orders of magnitude weaker than the intragranular shielding currents responsible for such profiles [9], so their effect can be neglected.

4.2. Relaxation for different maximum applied fields (constant I)

Figures 4(a) and (b) present the pinning energies calculated for samples A and B, respectively, corresponding to different values of H_m at $I = 1.05\ I_c(0)$ in the case of A and $I = 1.2\ I_c(0)$ in the case of B. The squares correspond to the U_0 derived from the transport decay through criterion 1 with $\tau = 10^{-8}$ s and $t_b = 120$ s. The circles correspond to the U_0 calcu-

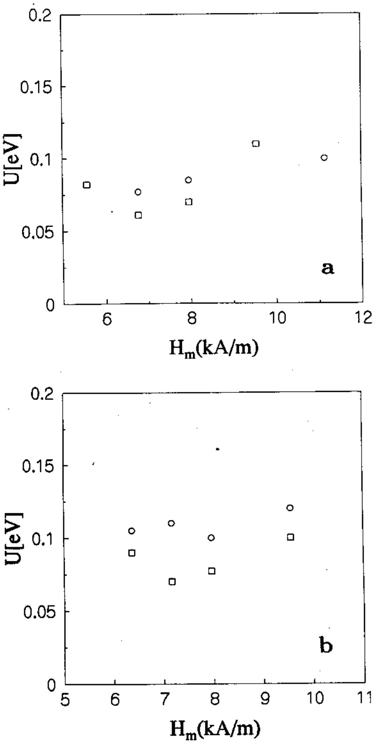

Fig. 4. Pinning energies calculated for sample A (a) and B (b) for different values of H_m based on transport relaxation (squares) and magnetic moment relaxation (circles) using criterion 1 (see text). $I = 1.05\ I_c(0)$ and $I = 1.2\ I_c(0)$ were used in the transport relaxation experiments for A and B, respectively.

lated from the magnetization decay recorded by VSM using the conventional expressions given in ref. [3] with similar values of τ and t_b. We have assumed in the VSM measurements that the magnetization came basically from the field trapped in the grains, although it is clear that some field trapped by the persistent loops of the weak link network overlaps. The similitude between our decay rates and the ones reported for single crystals [3] strongly supports our assumption.

Within our experimental errors (roughly similar to the ones reported for the pinning energies in fig. 3) a good coincidence was found between the U_0s derived from magnetic and transport decay experiments.

Taking into account the estimates of the average H_{c1g} and H^* mentioned above for samples A and B, the H_m range was selected in such a way as to qualitatively explore different intragrain field profiles associated with the remanent magnetization at $t=0$. Figure 5 illustrates the "ideal" intragranular field profiles when the external field is decreased (after ZFC) from $H=H_m$ (thin lines) to $H=0$ (bold lines) following a Bean's model [20] conveniently modified to take into account the surface magnetization effects. In principle, the profile at $H=0$ can be associated with the remanent magnetization at $t=0$ in our relaxation experiments. Then, a profile of the kind shown in fig. 5(a) (bold line) is obtained at $t=0$ for sample A when 11.2 kA/m$\leq H_m$, and in sample B when 9.6 kA/m$\leq H_m$. A profile of the kind shown in fig. 5(b) (bold line) is obtained for A when 7.2 kA/m$\leq H_m \leq$11.2 kA/m and for B when 6 kA/m$\leq H_m \leq$9.6 kA/m; and a profile like the one shown in fig. 5(c) (bold line) exists for A when 3.2 kA/m$\leq H_m \leq$7.2 kA/m and for B when 2.4 kA/m$\leq H_m \leq$6 kA/m. Actually, the situation in our case is much more complex, since there is no single kind of intragrain profile within each H_m range given above, since H^* and H_{c1g} are average values for the granular system.

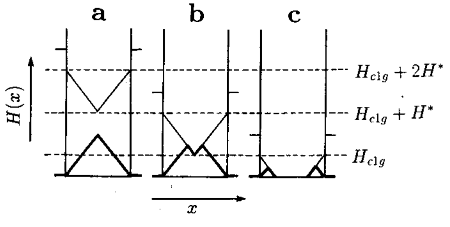

Fig. 5. Profiles of the intragranular field when the external field is taken from $H=H_m$ (fine lines) to $H=0$ (bold lines) in ZFC conditions. Illustrative situations when $H_{c1g}+2H^* \leq H_m$, $H_{c1g}+H^* \leq H_m \leq H_{c1g}+2H^*$ and $H_{c1g} \leq H_m \leq H_{c1g}+H^*$ are shown in (a), (b) and (c), respectively.

Following the calculations reported in ref. [21], only the initial "saturated" profiles of the type showed in fig. 5(a) at $t=0$ might be expected to give a logarithmic decay in the magnetization (i.e.; only our highest H_m values). However, we were able to fit formula (2) to the experiment in the whole range of H_m with reasonable quality. The Monte Carlo simulations of the magnetization decay for the profiles at $t=0$ illustrated in figs. 5(a) and (b) (bold lines) made in the spirit of ref. [21] would be illuminating from that point of view. We expect to report such calculations in the future.

5. Conclusions

Following the method proposed in refs. [7–9], we have estimated the pinning energy of granular $YBa_2Cu_3O_{7-x}$ ceramics in the presence of a transport current by measuring the voltage relaxation in the classical four-probe arrangement. Two different criteria for calculating U_0 were compared. Some of the experimental parameters (namely, I and H_m) were varied without detecting dramatic changes in the resulting pinning energies, though $H_m \geq 2H^*+H_{c1g}$ (H_m higher than 12 kA/m or 150 Oe in common ceramics) and $I \approx 0.9\ I_c(0)$ are recommended as practical conditions for the relaxation experiments, since they guarantee easily measurable dissipations with a near logarithmic decay, combined with zero voltage level at very long times. Finally, we presented the comparison between U_0 determined through transport and "magnetometric" relaxations, which coincide within 25 percent of the absolute values, approximately. It should be stressed, however, that these are gross estimations of the pinning energies of the superconducting grains. Factors such as the existence of different decay regimes in time, the inaccuracy of formula (1) in describing the magnetization decay even in the region $H_m \geq H_{c1g}+2H$ [5] and the weak link "switching" effects proposed in ref. [16] should be taken into account for a more precise determination of U_0.

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