Giant Flux Creep and Irreversibility in an Y-Ba-Cu-O Crystal: An Alternative to the Superconducting-Glass Model

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We report strong, anisotropic magnetic relaxation of the field-cooled and zero-field-cooled magnetization along the principal axes of an Y-Ba-Cu-O single crystal and interpret it with a thermally activated flux-creep model. A simple scaling argument shows that high thermal activation causes magnetic irreversibilities and critical currents to drop below the threshold of detectability at a reduced temperature difference $1-t$ proportional to $H^{2/3}$, a power frequently observed in experiment and in particular in our crystal.

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Many studies[1-6] of the new high-T_c ceramic superconductors have reported metastability of the zero-field-cooled (zfc) susceptibility and time-logarithmic relaxation of the remanent magnetization. Although such logarithmic decays are known to characterize flux creep in type-II superconductors,[7-9] it has been argued[1,10] that in the new materials, the relaxation reflects glassy features resulting from random weakly linked superconducting grains.[11] A key feature not found in conventional bulk superconductors was an "irreversibility" or "quasi de Almeida-Thouless" line $1-t\propto H^{2/3}$ where magnetic irreversibility set in; this suggested an analogy to spin-glasses. (Here t is the reduced temperature T/T_c and H is magnetic field.) The claimed reversible nature of the field-cooled (fc) susceptibility was also consistent with this idea. Early estimates indicated that the effective grain size of the superconducting glass might be smaller than the metallographically observed grain size, perhaps because of decoupling at twin boundaries.[12] At the same time, direct flux-line decoration[13] in crystals and the observation of conventional hysteresis loops interpretable in the Bean critical-state model[6,14] pointed towards a conventional flux-pinning picture.

To address these issues we investigate a better characterized system than the granular materials reported on so far. Here we report a systematic study of the temperature-dependent magnetic relaxation in an Y-Ba-Cu-O crystal. This extends the initial report by Worthington and co-workers[15] of logarithmic relaxation in a crystal. We find strong relaxation in both the zfc and fc susceptibility, in support of a flux-pinning picture. In most cases relaxation is logarithmic in time, but more complex non-logarithmic decays are observed in the high-temperature limit. The relaxation is anisotropic and faster for field parallel to the c axis (perpendicular to the Cu-O planes). We also find an irreversibility line in the crystal which is consistent with $1-t\propto H^{2/3}$. We show how these new results are interpreted naturally in the framework of a flux-pinning model, though the low trapping energies derived lead to some unusual consequences for critical currents and magnetic irreversibility, providing an alternative to the superconducting-glass model, at least in crystals. (The situation is less clear in ceramics where there is good evidence for Josephson weak links at grain boundaries.[16])

The $400\times400\times100$-μm^3 fully oxygenated crystal was prepared by Dinger *et al.*[14] as described earlier; the orthorhombic c axis (perpendicular to the Cu-O planes) is along the shortest edge. The magnetic measurements have been performed on a commercial SHE SQUID magnetometer with conventional procedures to be described in more detail elsewhere. Standard zfc and fc measurements show an overlap, or reversible regime, near T_c. Measuring in fields H ranging from 100 Oe to 40 kOe and using criteria as in Ref. 1, we thus identify an irreversibility line (Fig. 1) which fits the relation $1-t=(4.6\times10^{-4}\ G^{-2/3})H^{2/3}$ for $H\parallel c$. While such lines have been observed in ceramics, this is the first report of such behavior in a crystal.

Figure 2 shows the summary of the fc and zfc logarithmic relaxation rates $dM/d\ln t$ determined at an applied field of 1 kOe for both orientations. As mentioned above, deviation from logarithmic relaxation was ob-

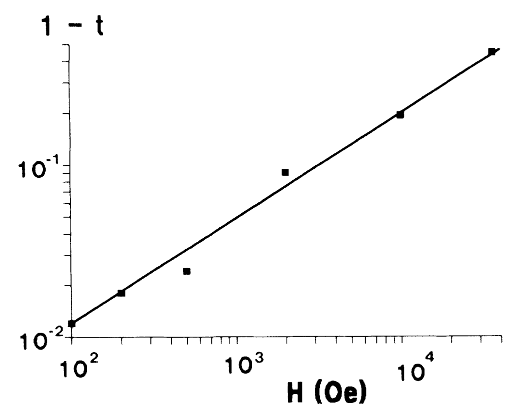

FIG. 1. Irreversibility line for Y-Ba-Cu-O single crystal for $H\parallel c$.

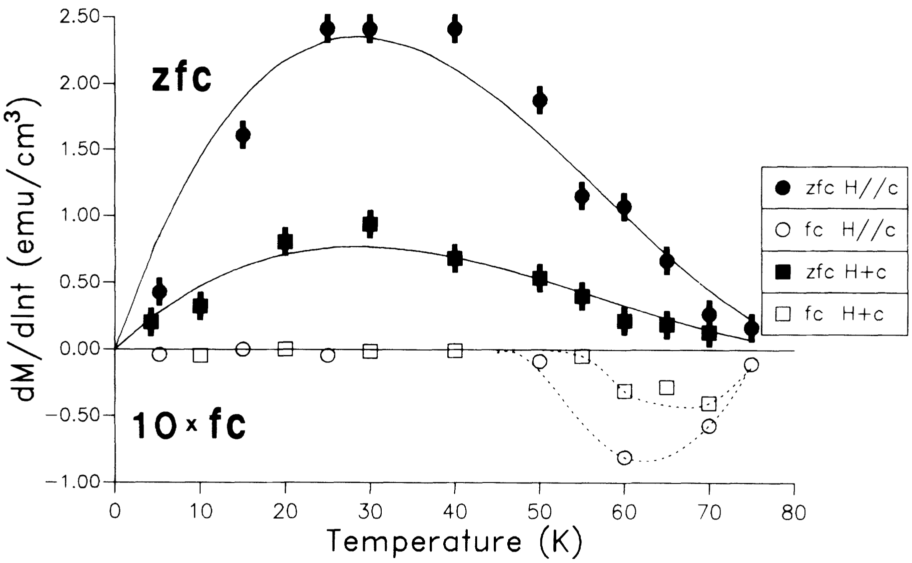

FIG. 2. Relaxation rate of field-cooled (fc) and zero-field–cooled (zfc) magnetization with logarithm of time, as a function of temperature for fields parallel and perpendicular to the orthorhombic c axis of an Y-Ba-Cu-O crystal. Solid lines are fits of zfc data with Eqs. (1) and (2) with $m=2$; see text. Dotted lines are guides for the eye for the fc data.

served at high temperatures, and the values in Fig. 2 represent only the logarithmic region, where presumably the critical state is well established. All four curves in Fig. 2 exhibit a common feature, namely that the relaxation rate increases with temperature, reaches a maximum, and then drops. The opposite signs for the zfc and fc values denote flux penetration and expulsion, respectively. The fc data are multiplied by a factor of 10. Nevertheless relative to the size of the starting fc signal, the relaxation is substantial—for example, increasing by 8% at 60 K during the first hour. The fact that the absolute size of the relaxation is smaller in the fc case may explain why the effect was not detected in earlier work.

To interpret the relaxation results we use a classical flux-creep model.[7-9] Consider thermal activation of flux lines over a barrier with activation energy U, in a critical state with magnetic induction gradient $\nabla B = 4\pi \mathbf{J}_c/c$, where $\mathbf{J}_c$ is the critical current and c is the velocity of light, both in centimeter-gram-second units. Then the magnetization relaxation of a cylinder of radius r is given by[8]

$$dM/d\ln t = (rJ_c/3c)(kT/U_0), \qquad (1)$$

where U_0 is the activation energy in the absence of a gradient. We use this formula for our $H\parallel c$ zfc data, taking $r=0.02$ cm, half the platelet dimension. (Corrections due to field dependence will be treated elsewhere.)

We develop a corresponding formula for a slab of thickness a with field in the slab plane:

$$dM/d\ln t = (aJ_c/4c)(kT/U_0). \qquad (2)$$

We use this formula for our zfc data with $H\perp c$, taking $a=0.01$ cm, the platelet thickness. We also assume that demagnetization shifts the effective applied field scale by the usual factor $1/(1-N)$ but does not affect the magnitude of the relaxation.

These formulas are valid in the region above the lower critical field H_{c1} where the Bean critical state[9] can be established. For $H\perp c$, H_{c1}, estimated[15] to be about 500 Oe, is adequately below our 1-kOe measurement field. For $H\parallel c$, H_{c1} has been estimated at anywhere from 0.6 (Krusin-Elbaum *et al.*[17]) to 5 kOe.[15] We shall also address this issue in a later publication, based upon field-dependent relaxation data which indicate something close to the lower value. With our demagnetization factor $N=0.7$ in this orientation, our 1-kOe applied field corresponds to 3.3 kOe, well above H_{c1}.

Interpretation of the fc data is complex because it requires a detailed knowledge of the metastability as a function of temperature through the critical region. We remark only that observation of fc metastability supports our recent interpretation of the field-dependent Meissner effect in terms of flux pinning.[18] We focus next on the zfc data.

Equations (1) and (2) describe the qualitative trends of Fig. 2 very well. The rise and fall with temperature can be interpreted in terms of a competition between the explicit linear T term and the implicit dropoff in J_c to zero at the critical temperature. (U_0 is also temperature dependent but usually more weakly; see below.) Equations (1) and (2) also suggest the origin of the relaxation anisotropy observed in Fig. 1. With larger J_c values for $H\parallel c$,[14] and with $r > a$ for our sample geometry, Eqs. (1) and (2) imply that $dM/d\ln t$ will be larger for $H\parallel c$ as observed. Since higher J_c implies stronger pinning, one might have naively thought that relaxation would be weaker in this geometry. The opposite is the case because in the critical state, higher J_c implies a larger flux gradient, which drives the flux lines to hop faster.

A quantitative treatment of the experimental data is hampered by a lack of direct temperature-dependent transport J_c data in crystals. We therefore use the phenomenological scaling form $J_c = J_c(0)(1-t)^n$, with the exponent n typically ranging from 1 to $\frac{5}{2}$ in experiments. We also take U_0 proportional to $(1-t)^{1/2}$, as will be shown below. We thus expect $J_c/U_0 \propto (1-t)^m$ with $m = n - \frac{1}{2}$. Substitution of this expression into Eqs. (1) and (2) gives the solid lines in Fig. 2, with $m=2$ and $U_0 = 0.6$ eV for $H \parallel c$ and 0.1 eV for $H \perp c$. If we consider the many approximations involved, the U_0 values should be trusted with only a factor-of-2 accuracy: Of most concern are the different effective applied fields and the field dependence of the critical state. Nevertheless these numbers, the first extracted for the new superconductors as far as we are aware, are of great interest in the following respects: First, the fact that $U_{0,\parallel} > U_{0,\perp}$ is suggestive of the role of twin boundaries whose planes contain the c axis and which would therefore act as pinning centers most effectively for $H \parallel c$. Second, Anderson and Kim[7] have suggested that U_0 should scale as $H_c^2\xi^3/8\pi$, where H_c is the thermodynamic critical field and ξ is the superconducting coherence length. For conventional type-II materials this usually comes out to several electronvolts, in agreement with earlier experiments.[8,9] For Y-Ba-Cu-O we take $\xi^3 = \xi_{ab}^2\xi_c$ with published parameters[15] and obtain $U_0 \simeq 0.15$ eV at low temperatures, which is between the two experimental determinations.

This order-of-magnitude lower U_0, coupled with the almost order-of-magnitude higher T_c's, in Y-Ba-Cu-O as compared to conventional superconductors leads to the observed "giant" flux creep and has important implications. Consider the role of thermal activation in the determination of critical current density, with use of the relation[9]

$$J_c = J_{c0}[1 - (kT/U_0)\ln(Bd\Omega/E_c)], \qquad (3)$$

where J_{c0} is the critical current in absence of thermal activation, B the magnetic induction, d a distance between pinning centers, Ω an oscillation frequency of a flux line in a pinning well, and E_c a minimum measurement voltage per meter. For typical parameters,[9] the logarithm is about 30, but since kT_c/U_0 is of order 10^{-3} in conventional low-temperature superconductors, the thermal-activation term is a negligible 3% correction. By contrast, for Y-Ba-Cu-O, kT_c/U_0 is of order 0.05, so that the thermal activation can be of order unity! Thus, through Eq. (3), critical-current measurements and even magnetic measurements of $H_{c2}(T)$ may be strongly affected by thermally activated flux creep. In particular, the creep term in Eq. (3) could contribute to the large temperature dependence of the measured critical current. These topics require more detailed quantitative treatment and will be addressed elsewhere.

Next we consider the field and temperature dependence of U_0. While many detailed theories have been developed,[9,19] there is no clear justification for these models yet in the new superconductors. Therefore we adopt a rather general scaling approach to obtain possible behaviors. For example, near T_c and for sufficiently low applied field B, we use the Anderson-Kim form $U_0 = H_c^2\xi^3/8\pi$ with the clean limit Ginzburg-Landau formulas, $H_c = 1.73H_{c0}(1-t)$ and $\xi = 0.74\xi_0(1-t)^{-1/2}$, where $t = T/T_c$ is the reduced temperature. This gives $U_0 = 1.21H_{c0}^2\xi_0^3(1-t)^{1/2}/8\pi$. When the flux lattice spacing $a_0 = 1.075(\Phi_0/B)^{1/2}$ becomes significantly smaller than λ, we expect, particularly in high-κ materials, a crossover to a new kind of pinning behavior due to collective effects. This crossover occurs somewhere between the lower and the upper critical fields, for example, at about $H = 0.2H_{c2}$ according to recent critical-current measurements[19] on Y-Ba-Cu-O. This corresponds to $a_0 = f\xi$, with f about 6. Above this field the energy per flux line is spread out fairly evenly through the entire cell of the Abrikosov flux lattice, and U_0 becomes limited in two dimensions by a_0. Along the third dimension the minimum possible characteristic length is ξ.[20] Thus we expect U_0 to scale as $H_c^2a_0^2\xi/8\pi f^2$, where the $1/f^2$ factor comes from the requirement of continuity of U_0 at the crossover. Substituting the expressions for H_c and a_0, we find

$$U_0 = 2.56H_{c0}^2\Phi_0\xi_0(1-t)^{3/2}/8\pi f^2B .$$

This simple scaling argument which predicts that U_0 decreases with temperature leads to the conclusion that thermal activation causes J_c to drop below the measurement threshold above a critical temperature. By substituting U_0 into Eq. (3), we find the condition for zero J_c:

$$1 - t = \left[\frac{8\pi f^2BkT_c\ln(Bd\Omega/E_c)}{2.56H_{c0}^2\Phi_0\xi_0}\right]^{2/3}, \qquad (4)$$

or, with typical parameters[15] ($H_{c0} = 2.7$ T, $\xi = 4$ Å), $1 - t \simeq 8\times10^{-4}B^{2/3}$, where the field B (which is approximately H near T_c) is in gauss.

While only an order-of-magnitude estimate, this result offers a simple explanation for several phenomena in Y-Ba-Cu-O, particularly the "quasi de Almeida-Thouless" or irreversibility line[1,4,5,11] $1 - t \propto H^{2/3}$, where magnetic hysteresis sets in. As mentioned earlier, this effect had been perhaps the principal argument for the invoking of novel glassy theories.[1,10] But since magnetic hysteresis is well known to correlate with J_c in the critical-state model,[9] Eq. (4) also gives a new derivation of this irreversibility line in terms of a classic flux-pinning picture. Remarkably, the seemingly ubiquitous $\frac{2}{3}$ power appears again,[20] though here from completely different physical arguments than in the spin-glass case. Our measurements of the irreversibility line for this crystal, giving about $1 - t \simeq (4.6\times10^{-4}\ \mathrm{G}^{-2/3})B^{2/3}$, are in a factor-of-two agreement with Eq. (4), which is remarkable if we consider the approximations in the pinning theory.

In summary we see that the flux-pinning picture is capable of explaining key magnetic and critical-current phenomena in the new superconductors, but clearly old guidelines about the magnitudes of various parameters must be modified. The unusually small coherence length leads to an unusually low pinning energy and thus to an unusual degree of thermal activation. A good deal of data, as well as prospects for applications, need to be reevaluated in this light.

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[20]Another possible choice for the third dimension is a_0. In this case we expect $U_0 = H_c^2 a_0^3 / 8\pi f^2$, and this leads to

$$U_0 = 3.72 H_{c0}^2 \Phi_0^{3/2} (1-t)^2 / 8\pi f^2 B^{3/2}.$$

The condition for $J_c = 0$ in Eq. (3) corresponds now to $1 - t \propto B^{3/4}$. We are indebted to P. H. Kes for this comment.