Magnetic properties of YBaCuO and BiSrCaCuO crystals: a comparative study of flux creep and irreversibility*

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We report strong, anisotropic magnetic relaxation for the zero-field-cooled magnetization of a BiSrCaCuO crystal measured with field $H = 1$ kOe parallel and perpendicular to the c axis. The relaxation rate increases with temperature, peaks at $\simeq 15$ K and drops to zero at $\simeq 30$ K, well below the transition temperature $T_c \simeq 84$ K. We interpret the low temperature data with a thermally activated flux creep model and derive a pinning energy $U_0^{\parallel} \simeq 8 \times 10^{-3}$ eV. These results show that for Bi-oxides the magnetization is already reversible at moderate temperatures. Similar conclusions are derived from a.c. susceptibility measurements which show an unusually strong frequency dependence of the irreversible temperature T_{irr} for Bi-oxides. These results are compared with those obtained for YBaCuO crystals in which the pinning forces are an order of magnitude larger, the reversible regime is limited to higher temperatures and the frequency dependence of T_{irr} is weaker.

Keywords: flux creep; anisotropic magnetic relaxation; irreversibility

In recent articles[1-5] we explored the strong magnetic relaxation observed in YBaCuO crystals and its implication for irreversibility, critical fields and critical currents. The picture which has emerged from these studies is that of a type II superconductor with unusually weak flux pinning. The combination of the low pinning energy and the unprecedented high temperature results in strong ('giant') flux creep. At higher temperatures and fields, above the 'irreversibility temperature' T_{irr}, thermal activation overcomes flux pinning barriers allowing free flux motion and hence a drop of J_c towards zero, reversible magnetization and broadening of the resistivity—temperature curves.

An important question which was left open in our previous studies is the origin of the pinning centres and, in particular, the role played by twin boundaries in flux trapping. In this article we address this question by a comparative study of twinned and twin-free Cu-oxide crystals. In the following sections we briefly review the experimental data for the YBaCuO crystals and then present new data for a relatively twin-free BiSrCaCuO crystal. The Bi crystal exhibits even stronger relaxation effects, smaller pinning forces and a wider reversible regime, at least for $H \| c$. These results suggest that twin boundaries contribute to flux trapping, although we cannot rule out the alternative possibility that chemical or structural differences between the YBaCuO and BiSrCaCuO systems are responsible.

Experimental techniques

Details of sample preparation are given in References 6, 7 and 8 for $Y_1Ba_2Cu_3O_{7-\delta}$ and in Reference 9 for $Bi_{2.15}Sr_{1.6}Ca_1Cu_2O_{8+\delta}$.

Magnetic relaxation measurements have been performed on a commercial SHE SQUID magnetometer using a standard procedure: the sample is cooled in zero field from well above the transition temperature T_c. At temperature T a field H is applied and the zero-field-cooled (zfc) magnetization M is measured as a function of time, typically for 1 h.

The in-phase and out-of-phase a.c. susceptibility χ' and χ'' are measured as a function of frequency (10^2–10^8 Hz) and at applied d.c. fields (0–1.4 T) oriented parallel to the crystal's c-axis. The a.c. field is also along this axis. Details of the measurements technique are given elsewhere[10]. A susceptibility anomaly defines an 'irreversibility line' in the H–T plane. A d.c. irreversibility line is defined by the temperature above which zero-field-cooled and field-cooled magnetization merge within experimental error.

Review of YBaCuO results

Magnetic relaxation

Figure 1 exhibits typical time dependences of several magnetic isotherms[4], after applying a field $H = 0.6$ kOe

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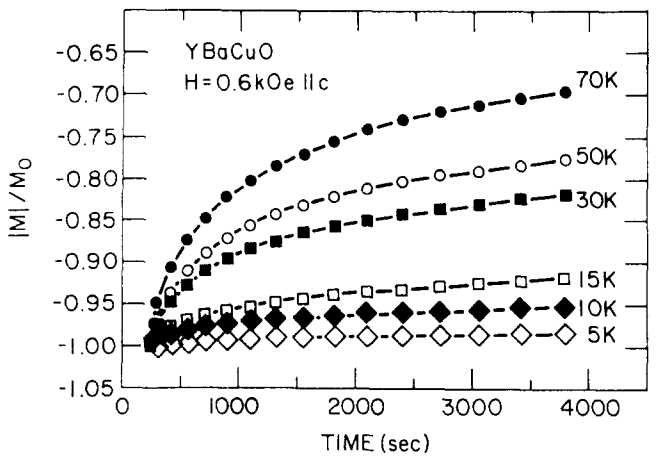

Figure 1 Decay of the normalized magnetization as a function of time for a YBaCuO crystal. The field $H = 0.6$ kOe, parallel to c, is applied after cooling the sample in zero field (from Reference 4)

parallel to c. The measured magnetic values M are normalized to $|M_0|$, the first measured data point. (This point is taken at $t_0 = 200$ s after field application). This figure demonstrates that flux is penetrating the sample even at the lowest temperature. Note the large size of the effect, e.g., at 70 K we observe $\simeq 30\%$ change in M during the first hour. For most of the temperature regime studied here the relaxation of the magnetization is linear with the logarithm of the time. However, strong deviations from linearity are observed in high-temperature high-field measurements. In this limit we take the relaxation rate to be the slope $dM/d\ln t$ at the end of the measurement time window, assuming that this reflects better the flux creep in a critical state.

Figure 2 summarizes the 0.6 kOe *zfc* decays for $H \| c$ by displaying the absolute relaxation rate $S \equiv dM/d\ln t$ as a function of temperature. Qualitatively, the relaxation rate increases with T, peaks at $\simeq 25$ K and decreases slowly to zero close to T_c.

The field dependence of the logarithmic relaxation rates S at 6 K is studied in Reference 3. We find that S increases initially as H^3 beyond a threshold at a field H_{c1} which we identify as the lower critical field, the onset for flux

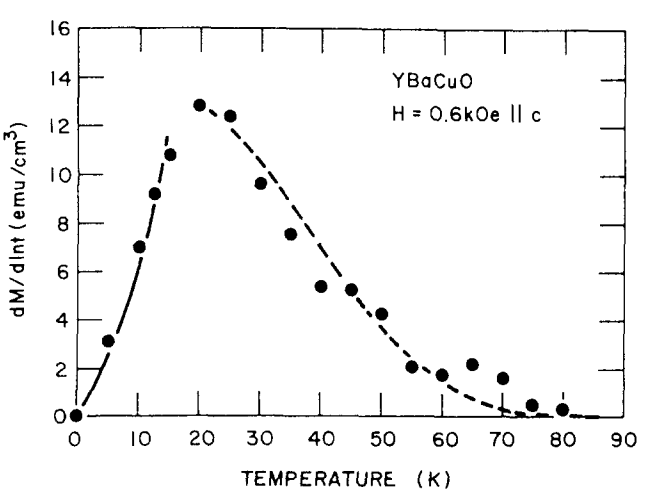

Figure 2 Relaxation rate of the zero-field-cooled magnetization as a function of temperature for field 0.6 kOe parallel to the orthorhombic c-axis of a YBaCuO crystal. Solid line is a fit to Equation 1 with $U_0(0) = 0.015$ eV. Broken line is a guide for the eye for $H > H^*(T)$ (from Reference 4)

penetration and irreversibilities. At higher fields, S reaches a maximum and then slows down. We find $H_{c1} \simeq 900$ Oe for $H \| c$. Because of demagnetization, the 0.6 kOe data in *Figure 2* are in fact above H_{c1}.

To explain the logarithmic relaxation in superconductors[11,12], Anderson[13] suggested a flux creep model in which flux lines in the critical state hop over potential barriers U_0 due to thermal activation. To interpret the present results we extended this model to include field dependence[3]. We calculated the magnetization of a slab of thickness D with field in the slab plane within an extended Bean model[14,15], assuming that the critical current J_c is inversely proportional to a power n of the local field B and approximating B by a discontinuous vertical drop to zero when B is less than H_{c1}. The explicit time dependence of the critical current[16,17] was inserted into the expressions for the magnetization and the relaxation rate was calculated[3]. The model predicts relaxation setting in above the lower critical field H_{c1} according to

$$dM/d\ln t = \frac{1}{4\pi}\frac{2}{C_0 D}\frac{n+1}{n+2}\left[H^{n+2} - H_{c1}^{n+2}\right]\frac{kT}{U_0},$$

$$H_{c1} \leqslant H \leqslant H^* \qquad (1)$$

$$dM/d\ln t = \frac{1}{4\pi}\frac{1}{2(n+1)}\frac{C_0 D}{2} H^{-n}\frac{kT}{U_0}, \quad H \gg H^* \qquad (2)$$

where C_0 is the value of $C \equiv (4\pi/10)(n+1)J_{c1}H_{c1}^n$ in the absence of thermal activation and $H^* \equiv (CD/2 + H_{c1}^{n+1})^{1/(n+1)}$ is the first field for which currents flow through the entire volume of the sample. Modified expressions for high temperatures (in the limit $kT/U_0 \gg 1$) are calculated elsewhere[18]

To understand the behaviour of *Figure 2* we first note that H^* and C_0 depend implicitly on J_c and therefore decrease with temperature. Thus, for a constant field H the validity range of Equation 1 (i.e., $H_{c1} \leqslant H \leqslant H^*$) is limited to low temperatures. In this limit $dM/d\ln t$ increases initially linearly with T, and then more rapidly because of the temperature dependence of C_0 and U_0. At higher temperatures H is no longer less than H^* and the behaviour of the relaxation rate, which is described now by Equation 2, is controlled by the implicit dropoff of C_0/U_0 with temperature. Though Equation 2 gives the essence of the physics in the high temperature limit, it is apparent from the non-logarithmic time dependence which is observed in this regime that the situation is more complex. This complexity is addressed in Reference 18 where we suggest a reasonable extension of the present model assuming a distribution of potential barriers[19]. Here, to avoid this complication, we focus on the prediction of Equation 1 which enables a reasonable estimation of the low-T pinning energies and allows a comparison between various high-T_c superconductors.

The various parameters in Equation 1 can be extracted independently. In particular, J_c is measured from the magnetic hysteresis loop[14] and H_{c1} is estimated from the field threshold of the relaxation measurements. D is the slab thickness for $H \perp c$ and the mean radius of the slab for $H \| c$. (In the low-field limit Equation 1 is valid for a cylinder of radius $R \equiv D$.) The solid line in *Figure 2* is a result of a fit[4] to Equation 2, which yields $U_0^{\|} \simeq 0.02$eV. A similar procedure for $H \perp c$ yields $U_0^{\perp} \simeq 0.15$eV. (The symbols $\|$ and $\perp$ refer to values for $H \| c$ and $H \perp c$,

respectively). These numbers are typical for several twinned YBaCuO crystals we have studied.

The field dependence of $dM/d\ln t$ is explained by Equations 1 and 2 in a similar way as above. The experimentally observed H^3 increase, as well as the threshold at H_{c1}, are apparent in Equation 1. The maximum in $S(H)$ is a result of the implicit decrease of S with H in Equation 2. A two parameter fit to Equation 1 yields an H_{c1} value ($\simeq$900 Oe) which is discussed in more detail elsewhere[3], and a similar U_0 value as above.

Irreversibility line

The unusual large values of kT_c/U_0 in high T_c oxides, as compared to more conventional type II superconductors, have important implications. In particular, the effective pinning force drops unusually quickly at high temperatures and therefore a relatively wide reversible regime is expected. The irreversibility line, which separates the reversible and irreversible regimes in the H–T phase diagram, is usually determined from conventional zero-field-cooled/field-cooled magnetic measurements[1,20]. Another, more sensitive, technique is based on measurements of the loss signal χ'' of the a.c. susceptibility. The loss signal has a sharp peak at a temperature T_{irr} where magnetic irreversibilities set in[2,5]. In conventional superconductors T_{irr} is indistinguishable from T_c. In YBaCuO, however, U_0 drops to small values well below T_c and the loss peak appears at $T_{irr} < T_c$. To find an equation for the irreversibility line, we use the criterion[5] that J_c reaches a critical value J_m. In the a.c. experiment we take[5] J_m to be $c\delta h/2\pi D$ where δh is the amplitude of the a.c. field. In the d.c. experiment J_m is determined by the experimental sensitivity. J_c is in turn related to the flux hopping attempt time t_0 and measurement time t by[17,18]

$$J_c = J_{c0}\frac{kT}{U_0}\frac{t_0}{2t}e^{U_0/kT} \tag{3}$$

which is simply the low-J limit for the Arrhenius relation for flux hopping. Next we take U_0 to be $p'(H_c^2/8\pi)a_0^2\xi$ at high temperature and fields; ξ is the coherence length and $a_0 = (\phi_0/B)^{1/2}$ is the Abrikosov lattice spacing. This means that at this limit the activation volume for flux hopping is limited in two dimensions by a_0. This expression for U_0 represents a crossover from the field-independent formula $U_0(0) = p(H_{c0}^2/8\pi)\xi_0^3$ which we take to describe our low-temperature low-field relaxation data. The constants p and p' are assumed to parameterize the specific defect pinning strength. Combining these expressions and using the Ginsburg–Landau temperature dependence of $H_c \propto (1-t)$ and $\xi = \xi_0(1-t)^{-1/2}$, we can solve for $1-t$ to obtain the irreversibility line formula

$$1 - t = \left[\frac{pBkT\xi_0^2}{p'U_0(0)\phi_0}\ln(c'f_0/f)\right]^{2/3} \tag{4}$$

where now $t \equiv T_{irr}/T_c$ and $c' = 2J_m U_0/J_{c0}kT$. Since the implicit field and temperature dependence of $\ln c'$ is only a weak correction, this equation gives the observed $B^{2/3}$ dependence of the irreversibility line[1,5,20].

Equation 4 also predicts that T_{irr} decreases with decreasing frequency; this frequency dependence was also verified experimentally[2,5]. The experimentally observed drop in T_{irr} with temperature is physically plausible because at longer measurement times there is more time for flux lines to relax to an equilibrium configuration, and thus reversibility can be achieved at lower temperatures. The size of the effect is controlled mainly by $p\xi_0^2/p'U_0(0)$.

New results for BiSrCaCuO

Magnetic relaxation

Figure 3 exhibits typical time dependences of several magnetic isotherms of a BiSrCaCuO crystal with $T_c \simeq$ 84 K, after applying a field $H = 1$ kOe parallel to c. As in *Figure 1*, the measured magnetic values M are normalized to $|M_0|$, the first measured data point. Here flux penetration is much more dramatic than in YBaCuO. At 20 K, the initial magnetization M_0 already decreases to 30% (!) of its value during the first hour of measurement. At higher temperatures flux presumably flows into the sample at a rate which is beyond detectability in the time scale of the present experiment. This is demonstrated further in *Figure 4* where we summarize the temperature dependence of the relaxation rate for this sample. The low temperature behaviour in *Figure 4* is similar to that

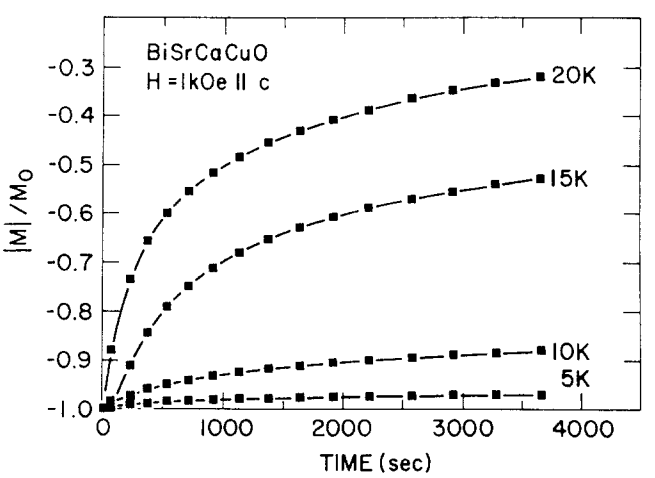

Figure 3 Decay of the normalized magnetization as a function of time for a BiSrCaCuO crystal. The field $H = 1$ kOe, parallel to c, is applied after cooling the sample in zero field

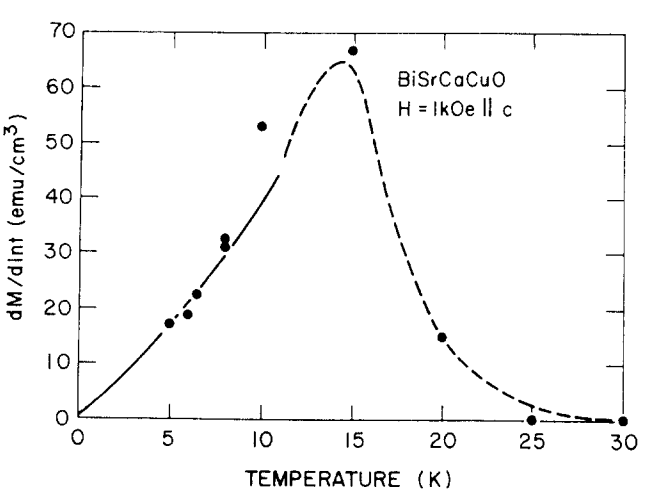

Figure 4 Relaxation rate of the zero-field-cooled magnetization as a function of temperature for field 1 kOe parallel to the orthorhombic c-axis of a BiSrCaCuO crystal. Solid line is a fit to Equation 1 with $U_0(0) = 8 \times 10^{-4}$ eV. Broken line is a guide to the eye

described in *Figure 2*. However, the field dependence of the relaxation rate[21] follows more closely an H^2 behaviour (rather than the H^3 found for Y crystals) with a threshold at $\simeq 80$ Oe. The solid line in *Figure 4* is a result of a fit to Equation 1 with $n=0$, $J_c = 2 \times 10^5$ A cm^{-2} (derived from our hysteresis loop measurements), $H_{c1} = 80$ Oe and $U_0 \simeq 8 \times 10^{-3}$ eV. Since this small U_0 value corresponds to only 10 K, it is not surprising that the relaxation is completed and equilibrium is achieved well below T_c.

For H$\perp c$ the measured relaxation rates in comparable fields and temperatures are smaller by two orders of magnitude. A quantitative analysis of this result is still in progress.

Irreversibility line

From the discussion of the irreversibility line and from Equation 4 it is apparent that the extremely small $U_0^{\parallel}$ observed in the Bi crystal should affect $T_{irr}(f, B)$ in a very pronounced way. In *Figure 5* we compare the effect of frequency on T_{irr} in YBaCuO and in BiSrCaCuO. Indeed, T_{irr} in the Bi-oxide is much more frequency dependent, suggesting that U_0 for the Bi sample is much smaller, in qualitative agreement with the magnetic relaxation data.

Similarly, in *Figure 6* we compare the effect of field on T_{irr} in YBaCuO and in BiSrCaCuO as determined from d.c. field-cooled and zero-field-cooled experiments. It is apparent from this figure that T_{irr} in the Bi-oxide is pushed to lower temperatures, exhibiting a wider reversible regime. A more quantitative analysis is in progress.

Discussion

Why is the flux pinning energy smaller in the Bi than in the Y material? One possibility is that the underlying material parameters H_c and ξ_0 in U_0 are smaller. Another possibility to explain the difference between the two systems is based on the absence of twin boundaries in BiSrCaCuO. The role played by twin boundaries in YBaCuO in pinning fluxons is still debated. Deutcher and Müller[22] argue that the small coherence length

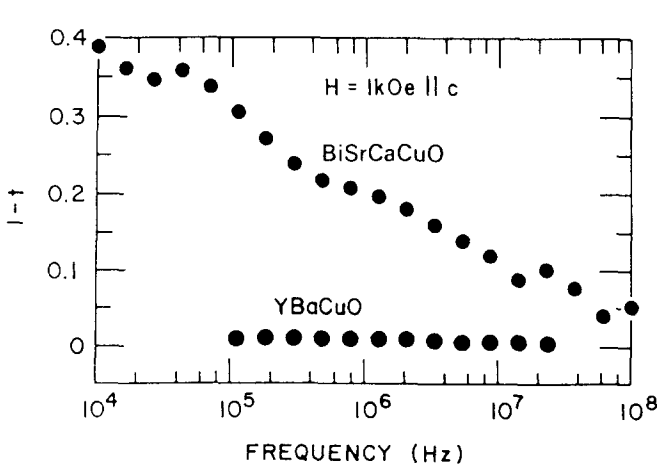

Figure 5 Frequency dependence of the irreversibility reduced temperature $1 - T_{irr}/T_c$ of a twinned YBaCuO crystal and a BiSrCaCuO crystal determined by a.c. susceptibility for an applied field of 1 kOe applied along the *c* axis.

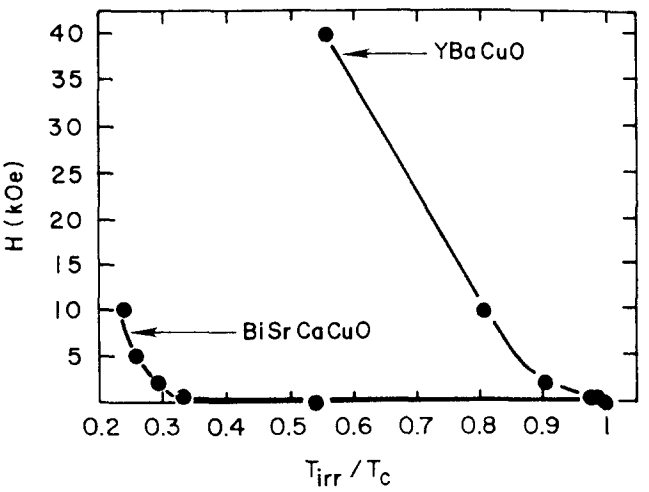

Figure 6 Field dependence of the reduced temperature $1 - T_{irr}/T_c$ of a twinned YBaCuO crystal and a BiSrCaCuO crystal determined by standard *zfc/fc* measurements with the field applied along the *c* axis

(possibly smaller than the typical interboundary distance) makes the boundaries more favourable for flux flow. Indirect support for this claim comes from relaxation[23] and critical current[24,25] measurements on untwinned ceramics and thin films. Kes[26], on the other hand, argues that twin boundaries, the most obvious defects in YBaCuO crystals, act as anisotropic pinning centres. This approach is consistent with the present comparative study of Bi and Y oxides.

In summary, our results indicate that 'giant flux creep' dominates the magnetic behaviour of BiSrCaCuO even more strongly than YBaCuO. The activation energy for flux hopping is an order of magnitude smaller in the Bi system, at least for $H \| c$. This implies a wider reversible regime as we find here, and offers a natural explanation for various recent experiments on Bi oxides, such as the 'melting' of the Abrikosov lattice at quite low temperatures[27] and the broader resistive transitions[28] in the presence of a field. Our comparative study suggests that the absence of twin boundaries in Bi oxides might be an important factor in the enhanced flux creep.

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