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Flux expulsion and penetration in superconducting $YBa_2Cu_3O_{7-\delta}$

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A large difference in the rate of flux flow in $YBa_2Cu_3O_{7-\delta}$ superconductors is observed between flux expulsion (field cooling then removing the field) and flux penetration (zero-field cooling then switching on a field). The mean activation energy and the width of its distribution for flux expulsion are 14 and 28 meV and for flux penetration are 34 and 67 meV. The smaller activation barrier for flux expulsion relative to flux penetration can be explained quantitatively in terms of flux pinning at the surface image potential and is also consistent with a model of randomly coupled superconducting grains.

Hard superconductors show time-dependent changes in their magnetization (diamagnetic susceptibility) under a magnetic field gradient. These changes, typically logarithmic in time,[1] can be modeled as thermally activated fluxon motions over a distribution of pinning barriers where the apparent activation energy increases as equilibrium is reached.[2] The driving force is the Lorentz force associated with the field gradient. Müller, Takashige, and Bednorz[3] stirred renewed interest in flux creep when their report of large time-dependent effects in $La_2(Sr)CuO_3$ reached the wide community now attracted to the field of high-T_c superconductivity. They interpreted the flux creep as evidence of a superconducting-glass (SCG) state with a random local-order parameter associated with the phase difference between the superconducting wave functions in adjacent coherent superconducting regions.[4] Moreover, they[3] identified the ergodic limit on the SCG H-T phase diagram as a de Almeida–Thouless glass transition beyond which the system is fluidlike, i.e., the time scale for relaxation is very short on a laboratory scale.

Critical to the understanding of the time-dependent effects, whether as conventional flux creep or as manifestations of an SCG state, is an appreciation of the preparation of the initial magnetization states. The initial magnetization can be set by zero-field cooling (ZFC) then suddenly switching the field on, $M_{ZFC}(T,t_0)$, or by field cooling (FC) then suddenly removing the field, $M_r(T,t_0)$. (Both of these magnetizations are also functions of the field involved but that dependence will not be explicitly expressed because its effects are not discussed here.) In the former case the time dependence describes flux penetration, in the latter case, flux expulsion. It is of interest to compare these two processes. It is known that in the absence of an applied field, the coherent superconducting region in Y 1:2:3 is the macroscopic specimen, whether it is a single crystal or a Josephson-coupled polycrystalline sample.[5] However, in the presence of a relatively weak field, not only are the grains of a polycrystal decoupled ($H_d < 100$ Oe),[5] but the coherent superconducting regions may become much smaller than the grain size.[3] Our flux-creep experiments were done at fields of 1 kOe and greater and, therefore, probe the intragranular pinning processes, not those of the weak links between the grains.

We have studied flux flow in a variety of polycrystalline high-T_c superconductors[6,7] and observed a significant difference between flux expulsion and penetration in several of these. Here we report the results for $YBa_2Cu_3O_{7-\delta}$ (Y 1:2:3). Flux expulsion is found to be a much easier process, penetration more difficult, in terms of the activation energies derived. In Y 1:2:3 flux expulsion is characterized by a mean activation energy of 14 meV, flux penetration by 34 meV. It is not possible to interpret this difference in terms of a simple flux-creep model in which the same pinning barriers are encountered by fluxons entering or leaving the material. We show that it is possible to explain the data quantitatively by extending the simple flux-creep model to include surface-pinning effects.[8]

Ceramic samples of Y 1:2:3 were prepared by conventional powder processing from high-purity oxides and carbonates, calcining at 1000 °C in air and annealing at 975 °C for 5 h followed by a slow cooling in flowing oxygen. Grain sizes range from 5 to 50 μm and the samples are of a single, orthorhombic phase. The samples show $T_c = 92$ K by resistivity and susceptibility measurements and ΔT_c (90%–10% resistivity) of 3 K or less is obtained. Magnetization measurements were carried out on a commercial SQUID magnetometer and on a vibrating sample magnetometer. Both instruments give equivalent results but only the latter allows measurements of times $t - t_0 < 2$ min.

Figure 1 shows the magnetization before relaxation (i.e., $t = t_0$) as a function of temperature after FC $M_{FC}(T)$ (this is the Meissner state), and upon removing the applied field after FC, $M_r(T)$ (this is the remanent state), as well as upon sudden application of a field after ZFC, $M_{ZFC}(T)$. The Meissner state is the stable state if field cooling is done slowly enough. No relaxation occurs from this state. Relaxation has been reported from the Meissner state,[9] but such observations imply nonequilibrium cooling in a field to reach this state. The states $M_r(T)$ and $M_{ZFC}(T)$ are inherently unstable because of the field gradient established at the surface of the superconductor by the sudden change in applied field that defines the initial time t_0. For $M_r(T,t_0)$ there is trapped flux inside and

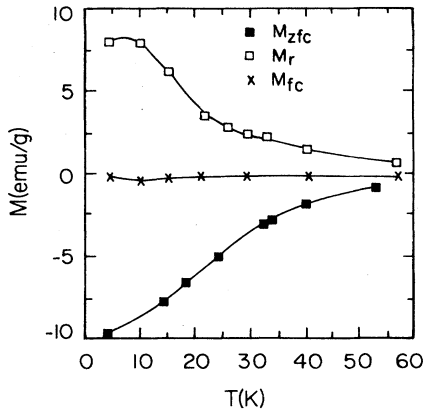

FIG. 1. Magnetization as a function of temperature for $YBa_2Cu_3O_{7-\delta}$ polycrystalline superconductor prepared in different ways. FC is field cooled in 1 kOe and measured at T. M_r is field cooled then measured 15 sec after turning off the field. ZFC is zero-field cooled then measured 15 sec after turning on a field of kOE.

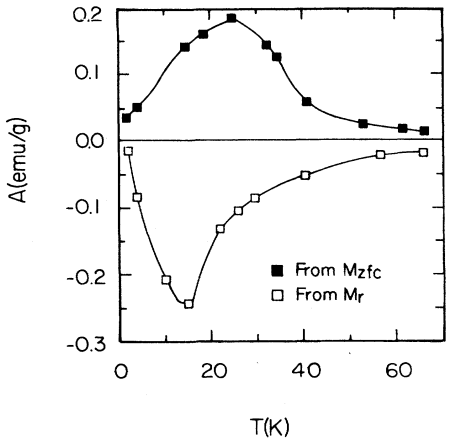

FIG. 2. Temperature dependence of the relaxation rate $A(H,T)=dM(t)/d\ln(t/t_0)$ for polycrystalline $YBa_2Cu_3O_{7-\delta}$ after preparation in the FC in 1 kOe then $H=0$ state and in the ZFC then $H=1$-kOe state.

$H=0$ outside the superconductor, whereas for $M_{ZFC}(T, t_0)$ B is small ($B<H_a$ the applied field, as a result of diamagnetism) but not zero inside and $H=H_a$ outside. The final state ($t=\infty$) for decay of $M_r(T,t)$ is $M=0$ which, because $H=0$, is also $B=0$. The final state for $M_{ZFC}(T,t)$ is the Meissner state.

Logarithmic decay of $M_{ZFC}(T,t)$ in doped La_2CuO (Refs. 3 and 10) and in Y 1:2:3 (Ref. 11) has been reported to follow the relation

$$M_{ZFC}(T,t)=M(t_0)-A\ln(t/t_0)\,, \tag{1}$$

as was observed in conventional hard superconductors by Kim, Hempstead, and Strnad. The decay rate[6,7,11] $A(H,T)$ vanishes at $T=0$, shows a rounded peak, and returns to zero for $T<T_c$.

We have measured the relaxation of both $M_{ZFC}(T,t)$ and $M_r(T,t)$ and found that the relaxation is described by Eq. (1) over several decades[6,7] but that the relaxation is *linear* in t below approximately 120 sec. This is the first report of linear time dependence in superconducting flux creep and is important to the proper determination of pinning energy distributions. Linear time dependence is observed in the relaxation of vortices in neutron stars,[12] where it is also recognized to be critical to a full understanding of the pinning barrier energies. Focusing first on the dominant logarithmic decay, we show in Fig. 2 the decay rates $A(T)=dM/d\ln(t/t_0)$ for Y 1:2:3 from the remanent and ZFC states. Although the *net* decay in both cases is comparable, it is remarkable that the *rate* of decay for M_r (flux expulsion) peaks at lower temperatures (13 K) than does that for the M_{ZFC} (flux penetration) (24 K). Our ZFC relaxation rate lies between the parallel and perpendicular single-crystal ZFC relaxation data of Yeshurun and Malozemoff[9] on Y 1:2:3. We now proceed to analyze these data to extract the respective distributions of activation energies and pre-exponential factors.

The data of Fig. 2, as well as those published earlier on high-T_c superconductors[6,9,11] extend well beyond the low-temperature regime where the relaxation rate is sometimes observed to be linear[1,2] in T. (Doped La_2CuO_3 does show this linearity at low temperatures.[10]) Peaks in relaxation spectra as a function of temperature are well described by first-order kinetic rate theory.[13–15] However, in order to account for the observed nonexponential time dependence, one may assume a probability distribution of relaxation times $P(\tau)$. The simplest assumption for this distribution is given by the Richter model[16] in which $P(\tau)=[\tau\ln(\tau_2/\tau_1)]^{-1}$ for $\tau_1<\tau<\tau_2$ and zero otherwise. (This is a box distribution in Q.) Thus the magnetization decays according to[17]

$$M(t)=M(t_0)-[M(t_0)-M(\infty)]\int_{\tau_0}^{\infty}P(Q)[1-\exp(t/\tau)]d\tau=M(t_0)-\Delta MG(t)\,, \tag{2}$$

where $\Delta M=M(t_0)-M(\infty)$ and $G(t)$ is the familiar exponential integral.

Kronmuller[18] has evaluated such exponential integrals in various limits and found for the short time limit

$$G(t)=t/[\tau_1\ln(\tau_2/\tau_1)],\quad t\ll\tau_1\,, \tag{3}$$

and at longer times

$$G(t)=C+D\ln(t/t_0),\quad \tau_1<t<\tau_2\,, \tag{4}$$

where $C=G(t_0)$, $D=[\ln(\tau_2/\tau_1)]^{-1}$, and Arrhenius expressions relate the τ_i to activation barriers Q_i: $\tau_i=\tau_{0i}\exp(Q_i/k_BT)$, $i=1,2$ as in Eqs. (3) and (4). From Eqs. (1) and (4),

$$A=\Delta M/\ln(\tau_2/\tau_1)\,, \tag{5}$$

or, if $\tau_{01}=\tau_{02}$,

$$A=k_BT\Delta M/(Q_2-Q_1)\,.$$

The magnetizations implicit in Eq. (5) can be either M_r or M_{ZFC} and the difference ΔM is approximately M_r and $M_{ZFC}-M_{FC}$, respectively.

A proper analysis of the data makes use of Eq. (3) to fit the regime linear in t ($t<120$ sec) *and* Eq. (4) to fit the logarithmic data. This procedure gives both τ_1 and τ_2 at every temperature. Arrhenius plots of τ_1 and τ_2 vs $1/T$ allow determination of the bounding activation energies Q_1 and Q_2 of the distribution as well as of the two pre-exponentials. This procedure is carried out for both $M_r(t)$ and $M_{ZFC}(t)$ sets of data. The results are shown in Fig. 3. Not only is the flux penetration process $M_{ZFC}(t)$ characterized by a much greater mean activation energy than flux expulsion (34 meV compared to 14 meV), it also shows a much broader distribution in this constant $P(Q)$ approximation (67 vs 28 meV). We measure similar activation-energy distributions in small single crystals which we have grown. Our values of Q_2 are 37% and 15%, respectively, of the geometric mean of the activation energies derived by Yeshurun and Malozemoff[9] in single crystals of $YBa_2Cu_3O_{7-\delta}$ using an expression similar to Eq. (5) and assumptions about the temperature dependence of J_c and about the shape of the coherent regions when $H\neq 0$. Those authors have recently revised the calculations of their activation energies which now agree with ours.

As relaxation proceeds, the system can be modeled as exhausting the low-activation-energy processes first. Thus, with time, only the larger values of Q remain and the process slows down. Anderson's model[2] of flux-creep expresses this situation with a time-dependent activation energy

$$dM/dt=-\alpha\exp[-(Q_0-b\,|M(t)|)/k_BT]\,, \qquad (6)$$

so that as $|M(t)|$ decreases, the effective activation energy approaches its maximum value Q_0. This maximum value should correspond to the upper limits of our distributions Q_2. If we fit our data with Anderson's expression, we get $Q_0=33$ and 67 MeV for the $M_r(t)$ and $M_{ZFC}(t)$ processes, respectively, in very good agreement with our results in Fig. 3. However, neither Anderson's model nor the simple flux-creep model can explain why the activation energy should differ for flux penetration and expulsion.

A natural explanation for the different rates of flux penetration and expulsion can be found in terms of the flux-creep model if a surface image-potential pinning is included. Bean and Livingston[8] show that a surface barrier exists to flux penetration in the presence of an applied field but that flux expulsion in $H=0$ is unimpeded by a barrier. The surface barrier exists because of the combined effects of the attractive image potential seen by a fluxon as it approaches a plane surface,

$$-(\phi_0/4\pi\lambda)^2K_0(2x/\lambda)\,, \qquad (7)$$

and the repulsive potential a fluxon sees near a surface when a field of the same sense as the fluxon is applied,

$$(\phi_0/4\pi)He^{-x/\lambda}\,. \qquad (8)$$

Here ϕ_0 is the flux quantum, λ the penetration depth, H the applied field, x the distance into the superconductor from the surface, and K_0 a Bessel function of the second kind. Because K_0 diverges as x approaches zero, exact solutions for the barrier height cannot be obtained. However, we can estimate the potential barrier by the height of its maximum, which occurs at $x_0=\phi_0/(4\pi\lambda H)$, relative to the interior potential far from the surface. The energy per unit length of a fluxon at the potential maximum is

$$E_0=(\phi_0/4\pi\lambda)^2[\log_{10}(\lambda/\epsilon)-K_0(2x_0/\lambda)] +(\phi_0/4\pi)He^{-x_0/\lambda}\,. \qquad (9)$$

For $H=1$ kOe, $x_0=8\times10^{-7}$ cm, and E_0 is determined essentially by the field-dependent term

$$E_0\approx 1.6\times10^{-5}\ \text{erg/cm}\,.$$

The activation-energy difference we observe between flux penetration and expulsion is approximately $\Delta Q=0.04$ eV. These units can be misleading because atomic processes are not involved. The activation-energy difference per unit volume is $\Delta Q=3.25\times10^8$ ergs/cm^3, and the difference in the number of fluxons per unit area for M_{ZFC} and M_r processes is of order $1\ \text{G}/\phi_0\approx 5\times10^6\ \text{cm}^{-2}$. Thus the energy difference per unit length of fluxon is measured to be $\Delta E=6.5\times10^{-5}$ erg/cm, in reasonable agreement with the estimated barrier height E_0. The surface-pinning model indicates that the image-potential barrier to flux penetration vanishes at high fields. We have observed[17] that the activation-energy difference also decreases with increasing applied field approximately as H^{-2}. A more detailed calculation should explicitly include the flux distribution inside the material. Such a treatment is beyond

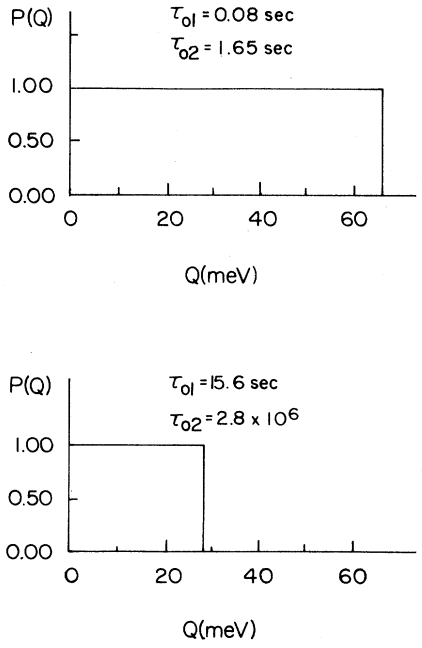

FIG. 3. Activation-energy distributions and pre-exponential factors for the two different preparations [above: $M_{ZFC}(t)$; below: $M_r(t)$] determined from linear and logarithmic time dependences of magnetization with use of Eqs. (3) and (4).

the scope of this article.

Our data also admit of an explanation in terms of a random array of weakly coupled superconducting grains. The difference in magnetization between initial and final states is approximately the same for flux penetration and expulsion. In the flux-creep model the driving energies are also the same for these two processes. However, in a granular superconductor the presence of an applied field increases the energy of the local configuration. The vector potential of the applied field causes a spatially varying phase difference A_{ij} between adjacent coherent superconducting regions.[19] The energy increases as a result of decreased coupling J_{ij} (flipped spins in the spin-glass analogy) between these regions:

$$\langle H \rangle = -J_{ij}\cos(\Delta\phi_{ij} - A_{ij}) , \quad (10)$$

where $\Delta\phi_{ij}$ is the zero-field phase difference between adjacent superconducting regions and $\Delta\phi_{ij}=0$ in the ground state. The elevated energy of the final state for flux penetration ($H\neq 0$) reduces the driving force for that process compared to that for flux expulsion. The presence of the field in flux penetration breaks the symmetry between the two processes. The reduced dirving force for flux penetration effectively enhances the activation energy extracted from the data in the same way that a reduced field-gradient results in an effective increase in activation energy.[20] However, the origin of the remanent moment remains a problem for the superconducting glass picture, as does the direct observation of a relatively conventional flux lattice in somewhat lower fields.

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