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Dependence of flux-creep activation energy upon current density in grain-aligned $YBa_2Cu_3O_{7-x}$

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We have investigated magnetic relaxation on a grain-aligned sample of $YBa_2Cu_3O_{7-x}$ powder embedded in epoxy with the magnetic field along the c axis. Through a thermal-cycling procedure and through variations in the measuring temperature over the range 10–30 K, we manage to commence magnetic relaxation from a range of values of the magnetization M or current density J. Analysis of relaxation measurements at a magnetic field of 1.0 T over the temperature range 10–30 K reveals a nonlinear dependence of the effective activation energy U_e on M or J that is consistent with the logarithmic dependence obtained by Zeldov *et al.* from transport measurements. This demonstration of a nonlinear dependence of $U_e(M)$ is capable of resolving anomalies in the magnitudes and temperature dependence of flux-creep activation energies obtained from magnetic-relaxation studies.

INTRODUCTION

The phenomenon of the rapid decay of the magnetization of all of the high-temperature superconducting (HTS) compounds is well established and the subject of intensive study.[1-3] Higher operating temperatures combined with small activation energies related to the short coherence lengths and large anisotropy are widely believed to account for the magnitude of various flux-creep effects. In addition to magnetic relaxation, resistive transitions in high magnetic fields have been attributed to thermally activated flux motion.[4,5] Both types of experiment have been successfully analyzed with the use of a standard phenomenological model first proposed for superconductors by Anderson[6] and further refined by Beasley, Labusch, and Webb (BLW).[7] Recent extensions of the phenomenology have been made by several authors.[8-10] Unfortunately, activation energies extracted from such analyses vary widely for a given compound, with much higher values obtained from resistance studies (0.1–1 eV for Y-Ba-Cu-O) (Ref. 5) than from magnetic relaxation experiments (10–100 meV for Y-Ba-Cu-O) (Ref. 1) at comparable temperatures and fields. One major difference in the conditions under which these two experiments are performed is the current density J relative to its critical state value J_c. In a typical magnetic relaxation measurement the sample is cooled in zero field, the magnetic field is then applied, and the time dependence of the magnetization M is measured. Of necessity, the magnetization begins its decay from the critical state $J=J_c$, where the effective barrier to flux motion is zero. As flux motion proceeds, the system creeps away from the critical state and the effective barrier U_e increases monotonically with time, giving the characteristic logarithmic time dependence of $M(t)$. By contrast, the resistive measurements are generally performed under the conditions where $J \ll J_c$. BLW first pointed out that the linear approximation $U_e = U_0 - JBV_c a$, where the effective barrier is reduced by the Lorentz stress JB multiplied by a correlation volume V_c and a hop distance a, is a poor approximation for a realistic potential when $J \cong J_c$. In general, $U_e(J)$ is a nonlinear function. The linear approximation (LA) results in[10] $M(t) = M_0 - A\ln(1+t/\tau)$, where $A \equiv dM/d\ln(t) \propto kT(J_c/U_0)$ and τ is a characteristic time $\ll 1$ s. For a more general nonlinear form for $U_e(J)$,[7]

$$A = kT(dU_e/\nabla B)^{-1} \propto kT(dU_e/dJ)^{-1} \propto kT(dU_e/dM)^{-1}.$$

The use of the LA expressions has led to the widespread practice of inferring an activation energy $U_0^* = kTM_0/A$ from the logarithmic creep rate A, where M_0 is the initial magnetization, assumed to be close to its critical-state value.[1,11] Xu *et al.*[12] have recently pointed out that $U_0^*(T)$ obtained in this manner is quite generally a monotonically increasing function of temperature for magnetic fields sufficiently large enough to ensure that the entire sample is well into the critical state. The anomalous $U_0^*(T)$ dependence may be understood as a result of a nonlinear $U_e(J)$, characteristic of most physically reasonable potentials. Since $dM/d\ln t$ is generally proportional to dU_e/dM or dU_e/dJ, the algorithm for U_0^* actually gives the intercept of the tangent line to $U_e(M)$ at the instantaneous value of M as shown in Fig. 1.[12] Increasing temperatures then move the operating point for the creep measurements to larger values of U_e and smaller values of M, causing the intercept to increase monotonically. This model presumes that any explicit temperature dependence of $U_e(M)$ is weak and should thus be valid for $T \ll T_c$.

The purpose of the present investigation is to demonstrate the explicit dependence of U_e upon the instantaneous value of J or $M - M_{eq} \propto Jd$, where M_{eq} is the equilibrium magnetization and d is an average grain diameter. We have employed two methods to attempt to determine $U_e(M)$. The first is a thermal cycling sequence that allows us to commence magnetic-relaxation measurements far removed from the critical state, i.e., $M \ll M_c$. The

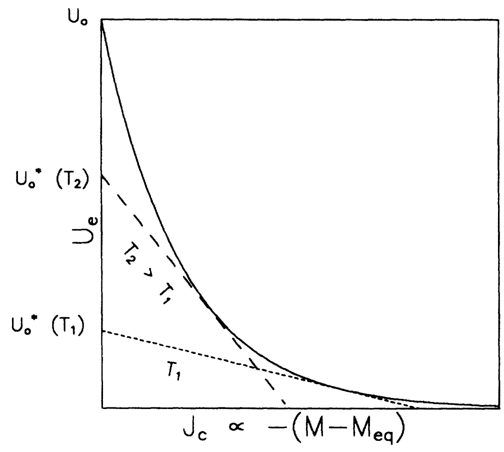

FIG. 1. Schematic representation showing an example of an arbitrary nonlinear effective potential U_e, plotted as a function of $M-M_{eq} \propto J_c$. Two tangent lines marked by temperatures T_1 and $T_2 > T_1$ are drawn to show the construction used to obtain values of U_0^* from measurements of the logarithmic creep rate.

sample is first cooled in zero field to an initial temperature $T_i > T_m$, where T_m is the measurement temperature; the field is then applied and the temperature is then lowered to T_m. Utilizing the lower J_c and faster relaxation rate at T_i allows us to begin relaxation measurements at different values of M. We have also employed a method of representing the relaxation measurements, described below, that directly displays $U_e(M)$. Recently Zeldov *et al.*[13] observed a nonlinear functional form for $U_e(J)$ from an analysis of resistive transitions in a magnetic field at different current densities. Our measurements show a similar dependence of $U_e(M)$ from magnetic-relaxation measurements.

EXPERIMENTAL DETAILS

The measurements reported here have been performed on a sample of magnetically aligned $YBa_2Cu_3O_{7-x}$ powder embedded in an epoxy matrix. The magnetic field is aligned parallel to the c axis, ±5° as determined by x-ray diffraction and confirmed by NMR measurements.

Magnetic susceptibility measurements showed a sharp transition at $T_c \sim 92$ K. The particle size distribution was determined by sedigraphy and showed an average diameter ~ 12 μm. Magnetic relaxation measurements were performed with a Quantum Design superconducting quantum interference device (SQUID) magnetometer operating with a 3-cm scan length to minimize field inhomogeneity effects. Extensive tests were performed to ascertain the effect of scan length on the measurements. Comparison of the 3-cm scan length results with measurements on the same sample under identical conditions in a vibrating sample magnetometer showed agreement within experimental error. Tests for effects of decay of the applied field were performed by comparing results for different ramping sequences and found to be insignificant. Values of M_{eq} used in the analysis were taken from the average of field increasing and field decreasing magnetizations for full hysteresis loops taken out to 5.0 T.

RESULTS AND DISCUSSION

Figure 2 shows the magnetic relaxation data taken at 20 K and at a field of 1.0 T. The various sets of data are labeled with the value of T initial at which the magnetic field was applied before cooling to 20 K and beginning the measurements. The data labeled 20 K show a ln$t$ behavior over two decades with some bending over at later times. A determination of the logarithmic rate for all of the temperatures studied shows A decreasing monotonically with temperature for these nonthermally cycled runs. Deriving $U_0^*(T)$ in the standard way produces a $U_0^*(T)$ that increases monotonically with temperature as described above. The other sets of data show the result of beginning the relaxation from successively smaller values of the initial magnetization. It is evident that modest reductions in M_0 result in substantial reduction in the relaxation rates. As pointed out by Sun *et al.*[14] this could have beneficial consequences for the operation of superconducting devices.

In order to display the explicit dependence of U_e on M,J, it proves useful to avoid *a priori* assumptions about $U_e(M)$ and to work with the rate equation for thermally activated motion of flux:[7]

$$\frac{d(M-M_{eq})}{dt} = \frac{B\omega a}{\pi d} e^{-U_e/kT} , \qquad (1)$$

where ω is the attempt frequency, a the hop distance, and d the average grain diameter for our powdered samples. This expression leads to

$$U_e/k = -T\ln|dM/dt| + T\ln(B\omega a/\pi d) . \qquad (2)$$

Figure 3(a) shows the result of plotting the data for 20 K in the form $T\ln dM/dt$ vs $M-M_{eq}$, which should be equivalent to within an additive constant to U_e/k vs $M-M_{eq}$. The data set labeled 1 was not thermally cycled, but zero-field cooled to 20 K, followed by ramping

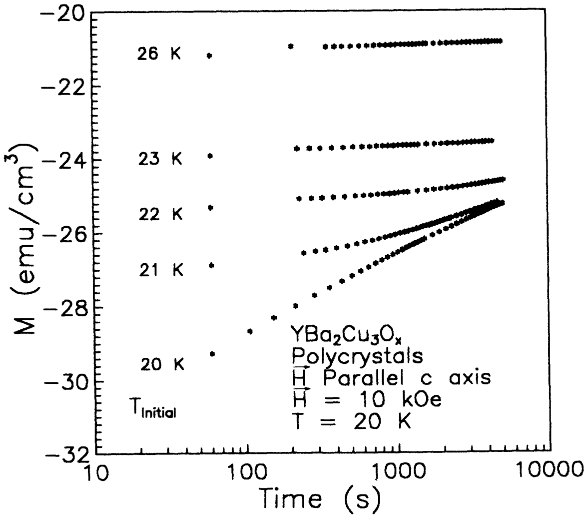

FIG. 2. Plots of magnetization vs time (on a logarithmic scale) for $T=20$ K and $H=10$ kOe. Several sets of data are marked by T-initial values at which the magnetic field was applied before cooling to the measuring temperature of 20 K.

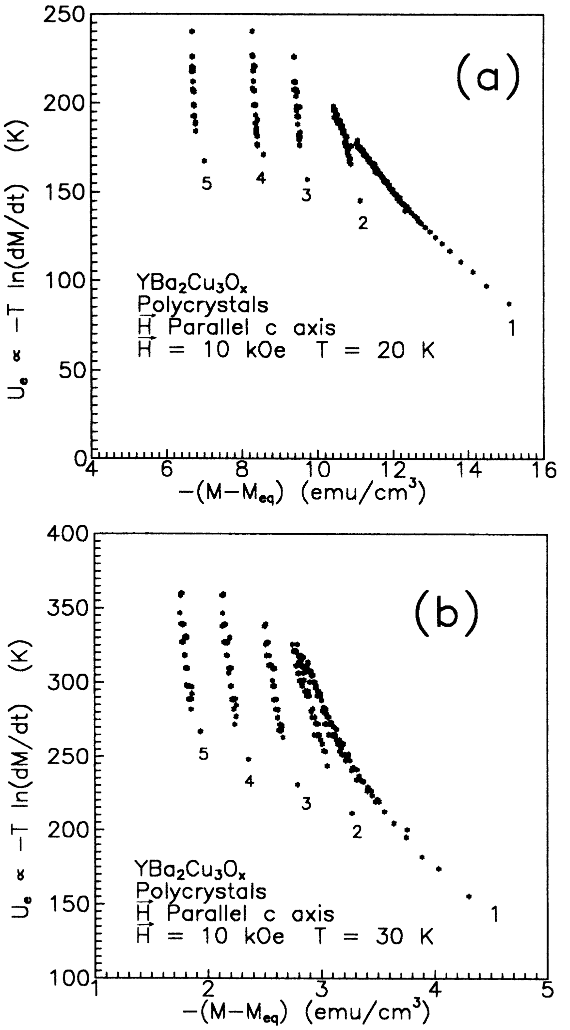

FIG. 3. Plots of $T\ln(dM/dt) \propto U_e$ plotted vs $M - M_{eq}$. The data sets marked 1 were zero-field cooled to the measuring temperature (a) 20 and (b) 30 K before the magnetic field was applied. The data sets 2–5 were thermally cycled as described in the text.

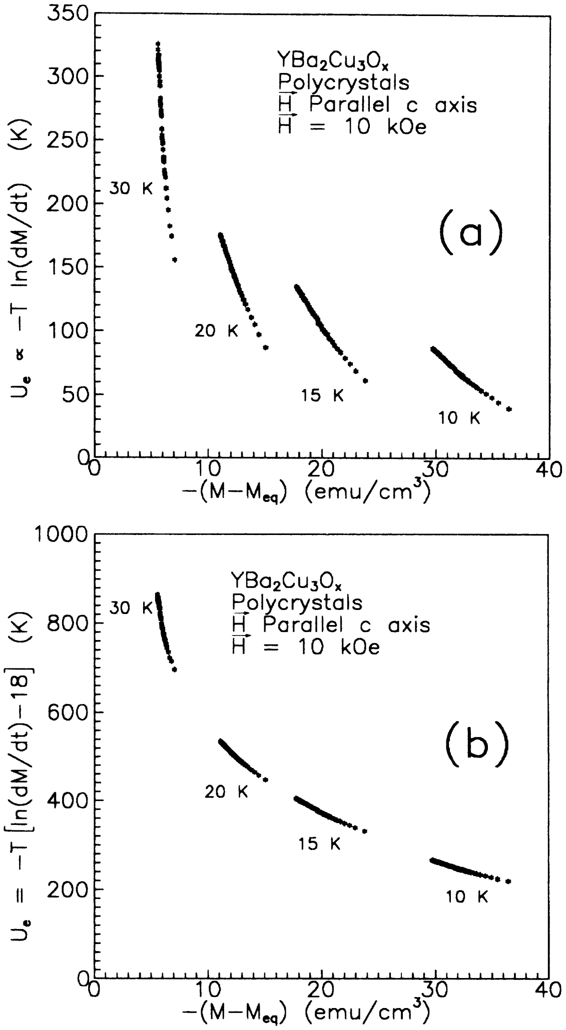

FIG. 4. (a) Plots of nonthermally cycled data: $T\ln(dM/dt)$ vs $M - M_{eq}$ for temperatures 10, 15, 20, and 30 K. (b) Plots of the same data shown in (a) with 18 T added to each data set where T is the temperature. As discussed in the text, this represents U_e vs $M - M_{eq}$.

the field to 1 T; and the measurements were commenced after ~1 min. The decay exhibits considerable curvature as M decreases, with an increasing slope dU_e/dM. The data set 2 was commenced at a smaller starting value of M and begins with a steeper slope that eventually flattens to match the slope of set 1. The remaining data sets 3–5, begun at successively smaller values of M, continue to relax with steeper dU_e/dM dependence. Because $M - M_{eq}$ is not varying appreciably during these runs, we believe that the thermal-cycling procedure has trapped a non-steady-state flux distribution that slowly relaxes with time. The behavior of set 2 then represents the eventual convergence to the true steady-state relaxation rate. Figure 3(b) shows a similar systematic evolution for $U_e(M - M_{eq})$ for relaxation rates at 30 K. The curvature in data sets 1 and 2 is even more pronounced.

Figure 4(a) shows $T\ln dM/dt$ vs $M - M_{eq}$ plots taken at 10, 15, 20, and 30 K. Only the nonthermally cycled data sets are shown here. The principal effect of increasing temperature is to produce monotonically decreasing initial values of $M - M_{eq}$. As $M - M_{eq}$ decreases, the slope dU_e/dM becomes progressively steeper. Referring to Eq. (2), we see that a constant equal to $T\ln(B\omega a/\pi d)$ added to each of these data sets will produce values equal to U_e/k for each temperature. As discussed in the introduction, the explicit temperature dependence of $U_e(M)$ over the range 10–30 K should be insignificant compared with the variation brought about by changes in the range of M values sampled at each temperature. Then selection of a single constant multiplied by T for each temperature will cause all of the data to fall on the same smooth $U_e(M)$ curve. The result of adding 18 T to each of the sets is shown in Fig. 4(b). A good agreement to the matching of magnitude, slope, and curvature to a simple universal curve is evident from the figure. The fit is only qualitative and inclusion of the neglected temperature dependence for $U_e(M)$ would be required to provide better definition. The constant $18 = \ln(B\omega a/\pi d)$ implies a reasonable value for the product $\omega a \cong 25$ cm s^{-1}, consistent with, e.g., a hop distance ~100 Å and an attempt frequency of 2.5×10^7 Hz. With this selection of the constant each temperature set begins at a nearly constant value of $U_e/kT \sim 22$. This suggests that, during the first minute after the magnetic field has stabilized, the system undergoes a rapid decay away from the critical state until $U_e \gg kT$ is established, and further relaxation occurs at a moderate rate. As T increases, successively smaller values of $M - M_{\mathrm{eq}}$ are required to increase U_e to the required level. The nonlinear dependence of U_e on M,J is apparent from Fig. 4(b), and adequate fits can be made to the functional form $U_e = U_0 \ln(J_0/J)$ observed by Zeldov *et al.*[13] over the limited range of $M - M_{\mathrm{eq}}$ values accessed by this investigation. A somewhat less satisfactory fit could be made to the form $U_e \propto (M - M_{\mathrm{eq}})^{-\alpha}$ $(0.5 < \alpha < 1)$, suggested by Feigelman *et al.*[15] on the basis of a collective flux-creep model. Investigations of $U_e(M - M_{\mathrm{eq}})$ over a wider range of $M - M_{\mathrm{eq}}$ values will be required to define the correct functional dependence.

CONCLUSIONS

We have observed in magnetic relaxation measurements a nonlinear dependence of the activation energy for thermally activated flux motion on magnetization or current density. The functional form $U_e(M - M_{\mathrm{eq}})$ is consistent with the logarithmic dependence observed in transport measurements and provides a natural explanation for the anomalous temperature dependence of $U_0^*(T)$ obtained from fits of magnetic relaxation data to a logarithmic time dependence. We find that plotting of $T \ln dM/dT$ vs $M - M_{\mathrm{eq}}$ reveals directly $U_e(M - M_{\mathrm{eq}})$ and that varying the temperature allows a range of $M - M_{\mathrm{eq}}$ values to be accessed at temperatures $T \leq T_c/3$ where the explicit temperature dependence of $U_e(M - M_{\mathrm{eq}})$ is relatively insignificant. Attempts to use a thermal cycling technique to commence magnetic relaxation from states far removed from the critical state was less successful because of thermal history effects on the relaxation.

These results provide a possible resolution of the inconsistencies and anomalous temperature dependencies associated with determination of activation energies for flux motion in HTS compounds. The model of a nonlinear $U_e(J)$ is an alternative, not necessarily incompatible, to assuming a broad spectrum of activation energies.[10] The results suggest that the activation energies U_0^* obtained from the conventional analysis will depend strongly on the magnetization or current-density level.

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