# Long-time magnetic relaxation in a detwinned  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  single crystal

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The time-dependent magnetic relaxation was measured in a full-field-penetrated YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7— $\delta$ </sub> detwinned crystal for long times up to  $5 \times 10^5$  sec. The relaxation data exhibit a power-law time dependence of magnetization over a wide range of temperature and magnetic field with the field oriented both parallel and perpendicular to the  $c$  axis of the crystal. The results are consistent with a flux-creep model assuming logarithmic current dependence of the flux-creep activation energy.

## INTRODUCTION

Since the discovery of the large magnetic relaxation in high-temperature superconductors,<sup>1</sup> a great amount of research on flux creep has been conducted based on thermally assisted hopping of the flux lines.<sup>2-5</sup> The experiments demonstrated a logarithmic time dependence of the magnetization in a time period of  $10^3$  sec, i.e.,  $M \sim$ lnt, where M is the magnetization at time t. The experimental results could be well described using classical phenomenological theories, which were developed in the 1960's for interpreting the relaxation behavior in conven tional superconductors.<sup>6-8</sup> In these theories, two assumptions were used to obtain the logarithmic relaxation with a form of  $dM/d$  lnt  $\sim k_B T/U_0$ , where  $U_0$  is the energy barrier. One assumption was the low-temperature limit,  $U_0 / k_B T \gg 1$ , which showed that the thermal activation induced the flux lines to move in bundles and jump over the energy barrier at a rate governed by  $\exp(-U_0/k_B T)$ . In the second assumption, the application of a magnetic field to a superconductor in a mixed state led to a gradient in the density of the flux lines. Then the critical state could be reached whenever the driving force was balanced with the local pinning force. In the critical state,  $J \sim J_c$  and a linear approximation of the energy barrier was obtained,  $U(J) = U_0(1 - J/J_c)$ . Extensions of the critical state to include field dependence were successful in explaining why the flux creep could be observed only by introducing an external field.<sup>5</sup>

Xu et  $al$ <sup>9</sup> reported a nonlogarithmic time decay of the magnetization in a c-axis-oriented YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> powder. As the temperature increased, they observed a deviation from a logarithmic time dependence. The explanation for this departure could be that the temperatures used in the experiments were too high for the application of the classical theory. Another argument was that the assumption of a critical state, that is, a linear current dependence of the activation energy, was an approximation valid only in a short-time period. Soon after this publication,<sup>9</sup> Zeldov et al.<sup>10</sup> reported a logarithm current-density dependence of the activation energy from transport measurements in epitaxial films of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  under magnetic fields. More recently, Ma-<br>lev *et al*,<sup>11</sup> and McHenry *et al*,<sup>12</sup> pointed out the same ley et al.<sup>11'</sup> and McHenry et al.<sup>12</sup> pointed out the same dependence of the activation energy on current density in grain-aligned  $YBa_2Cu_3O_{7-\delta}$  and  $La_{1.86}Sr_{0.14}CuO_4$  single crystals determined by magnetic measurements. Now it is generally accepted that the experiments of both magnetic and transport properties are performed well below the critical states,  $J \ll J_c$ . At low currents  $J \ll J_c$ , the vortex lattice is in a metastable state. Transitions between metastable states are due to thermal activation through a free-energy barrier whose characteristic scale is  $U(J)$ . If  $J \rightarrow J_c$ , then  $U(J) \rightarrow 0$ . The classical activation theories are applicable only under limited conditions (e.g., short time). The nature of the flux-creep phenomenon is due to weak randomly distributed defects. The dependence of the activation barrier  $U$  on the current J is shown to be logarithmic,  $U(J) \sim \ln J$ . A logarithmic current dependence for  $U(J)$  in high-temperature superconductors should be manifested in magnetic relaxation experiments.

Vinokur, Feigel'man, and Geshkenbein<sup>13</sup> proposed a theoretical model on the assumption of an activation energy U that grows logarithmically with decreasing current J:  $U = U_0 \ln(J_c/J)$ . They developed an exact solution for flux creep in type-II superconductors. The solution described the spatiotemporary evolution of the self-organized critical state in high-temperature superconductors and predicted a power-law time dependence of the magnetization. From the numerical analysis by Schnack and Griessen, $14$  this power-law time decay can only be observed in the fully penetrated state, which occurs when the magnetic Aux expands into the sample and flux fronts from opposite sides of the sample meet at the center.

Liu et  $al$ .<sup>15</sup> observed a power-law decay of the magne tization in a LuBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> single crystal. This observation is consistent with the predictions of Vinokur, Feigel'man, and Geshkenbein. The experimental results showed that the effective pinning energy was temperature independent and decreased logarithmically with the increase of the applied field. However, the work was only concentrated on the field parallel to the c axis of the crystal.

Considering the large anisotropy present in the hightemperature superconductors, the magnetic measurements in both field orientations provide an important approach to fully characterize the physical behavior of the magnetization in the mixed state such as flux creep, flux pinning, and thermally activated flux motion. Because of the difficulty in the magnetic measurements for the field parallel to the ab plane of the high-temperature superconducting single crystals, the reported experiments on magnetic time relaxation were limited to the  $H \| c$ configuration.

In this paper, we present a detailed systematic study of the magnetic relaxation in a well-characterized detwinned single crystal of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  for two configurations: applied field parallel to the c axis and perpendicular to the  $c$  axis. The relaxation experiments show the power-law dependence of the magnetization in the temperature range  $5-55$  K at fields up to 5.5 T over a long-time period of  $5 \times 10^5$  sec. The results can be successfully analyzed with the model proposed by Vinokur, Feigel'man, and Geshkenbein.<sup>13</sup> With this analysis, the values of activation energy at different fields are determined.

#### THEORETICAL BACKGROUND

The classical theories of thermally activated flux creep were developed by Anderson,<sup>6</sup> Beasley *et al.*,<sup>7</sup> and Campbell *et al.*,<sup>8</sup> and are used frequently to analyze the relaxation data of high-temperature superconductors. In these theories, it is assumed that the magnetic measurements are performed in the region of current  $J_c-J \ll J_c$  (near the critical state) where  $J_c$  is the critical current density. Therefore, the thermally activated energy barrier  $U$ is assumed to vary linearly with current density J. This leads immediately to the well-known result  $J(t)=J_c[1-(k_BT/U_0)\ln(t/t_0)]$  or

$$
U(J) = U_0(1 - J/J_c) \tag{1}
$$

The relaxation rate for the magnetization in the critical state is defined as  $S = (1/M_0)(dM/d \ln t)$  and can be described by the Anderson formula

$$
S = k_B T / U_0 \t\t(2)
$$

where  $M_0$  is initial value of the magnetization  $M(t)$  and  $U_0$  is the characteristic energy barrier in the critical state. Therefore, at low temperatures,  $S \sim T$ , and the activation energy underlying the theories is temperature independent. Extensive studies of flux creep on the hightemperature superconductors such as  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub>$ ,  $Bi_2Sr_2CaCu_2O_8$ , and  $La_{2-x}Sr_xCuO_{4-\delta}$  systems were carried out by a number of research groups.<sup>16</sup> The result indicated that the magnetization decays logarithmically with time, which was what should be expected based on classical thermally activated flux-creep theory. However, most experiments were limited to a time period of  $10<sup>3</sup>$ sec, which might not be sufficient for these hightemperature superconducting materials.

Vinokur, Feigel'man, and Geshkenbein<sup>13</sup> presented an analytic solution for the thermally activated flux creep in a one-dimensional superconductor from the logarithmic current dependence of the activation energy:

$$
U(J) = U_0 \ln(J_c/J) \tag{3}
$$

The logarithmic current-density dependence of the activation energy is in a good agreement with the magnetic tivation energy is in a good agreement with the magnetic<br>measurement by Maley et al.,<sup>11</sup> McHenry et al.,<sup>12</sup> and the transport measurement by Zeldov *et al.*<sup>10</sup> In the magnetic relaxation on grain-aligned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> powder embedded in epoxy with the magnetic field along the c embedded in epoxy with the magnetic field along the  $c$  axis, Maley *et al.*<sup>11</sup> obtained a logarithmic dependence of the activation at a magnetic field of <sup>1</sup> T over a temperature range of  $10-30$  K. Zeldov *et al.*<sup>10</sup> studied the voltage-current characteristics of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  epitaxia films at  $H \parallel c$  and found that the resistivity is thermally activated at low temperatures and the temperatureindependent activation energy  $U_0$  is a logarithmic function of current density.

Using a scaling form for the solution, Vinokur, Feigel'man, and Geshkenbein<sup>13</sup> finally obtained two formulas for the magnetization and its relaxation rate:

$$
\ln M = \text{const} - \frac{x_f(t)}{d} \frac{1}{\sigma} \ln \left( \frac{t}{\tau_0} \right), \quad t \ll t^* \tag{4}
$$

$$
\ln M = \text{const} - \frac{1}{\sigma} \ln \left( \frac{t}{\tau_0} \right), \quad t \gg t^* \tag{5}
$$

or

$$
S = \left| \frac{d \ln M}{d \ln t} \right| = \frac{x_f(t)}{d} \frac{1}{\sigma}, \quad t \ll t^* \tag{6}
$$

$$
S = \frac{1}{\sigma}, \quad t \ll t^* \tag{7}
$$

where d is the thickness of the sample,  $x_f(t)$  is the position of the flux front, and  $t^*$  is the time at which the sample is fully penetrated. From the model of Vinokur, Feigel'man, and Geshkenbein, some conditions and results are obtained:

(1) A power-law time dependence of the magnetic relaxation should be observed in high-temperature superconductors by taking into consideration the logarithmic current-dependent activation energy. These results are only applicable for fully penetrated samples. When applying a field, the flux gradient is pushed into the interior of the sample from both sides and flux fronts begin to meet in the center of the specimen at  $H^*$ . Above this field, the sample is in a fully penetrated state.

(2) For relaxation measurements carried out at low temperatures, the condition  $\sigma = U_0 / k_B T >> 1$  should be well satisfied. The relaxation rate is linearly temperature dependent, which results in a temperature-independent activation energy  $U_0$ .

(3) The relaxation rate should vanish when the temperature approaches zero, which indicates the relaxation is only thermally assisted.

(4) The theory is limited for the creep activation in a single vortex region, thus no field dependence of the relaxation is expected in a whole range of temperature and field.

(5) The assumption of logarithmic  $U(J)$  dependence is valid when the vortex motion is controlled by intrinsic pinning in a layered system with the field parallel to the layers, i.e.,  $H || ab$ .

These theoretical predictions emphasize the importance of measuring the magnetic relaxation over extended times. In order to fully understand the mechanism of flux creep due to logarithmic  $U(J)$  dependence, a systematic study of the magnetic relaxation in a wellcharacterized detwinned single crystal of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> is investigated for both  $H || c$  and  $H || ab$  configurations.

#### EXPERIMENTAL DETAILS

All experiments reported here have been performed on a detwinned  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  single crystal with fields oriented both parallel and perpendicular to the c axis. The sample was prepared using a self-flux growth and oxygen-annealing technique, then detwinned by a thermal-mechanical method.<sup>17</sup> The crystal's quality was confirmed by the superconducting transition and microstructural characterizations. Polarized light microscopy showed that the twin boundaries have been completely removed in this crystal. In addition, no twinning was identified on the crystal produced by this technique through detailed x-ray diffraction and specific-heat studthrough detailed x-ray diffraction and specific-heat stud<br>ies.<sup>18,19</sup> The transition temperature was 92.5 K with 10–90% transition width of 0.5 K measured by dc susceptibility. This crystal is platelet shape and has a dimension of  $2 \times 1.5 \times 0.05$  mm<sup>3</sup> with the c axis along the short dimension. The mass of the crystal is about <sup>1</sup> mg.

The magnetic properties were measured on a superconducting quantum interference device (SQUID) magnetometer.<sup>20</sup> The scan length, the distance which a specimen travels through a set of detection coils, was set at 3.0 cm. This relatively short distance minimized the magnetic variation where the specimen travels. Variation in field at this setting was estimated to be  $< 0.05\%$ . An iterative regression mode was used to calculate the magnetization.

In order to measure the magnetic relaxation in the fully penetrated sample, we first determined the penetration field  $H^*$ , which can be taken to be roughly the field at which the peak in the  $M$  versus  $H$  curve occurs. The value of  $H^*$  is plotted in Fig. 1 as a function of temperature. At higher temperatures, the field easily penetrates into the sample from its sides and collapses at the center to form the fully penetrated state in a short time. Therefore, for al1 the relaxation measurements, the applied field was chosen to be greater than the penetration field,  $H > H^*$ , to insure a well-defined penetrated state for a field oriented both parallel and perpendicular to the  $c$  axis of the crystal.

All the magnetization measurements were made by first cooling the sample in zero field and then applying a field using the SQUID no-overshoot mode to begin the magnetic decay experiment on the virgin magnetization curve. However, there may be some residual field left in superconducting magnet (remnant field) even through the superconducting magnet is reset. To reduce the effect of



FIG. 1. The characteristic penetration field  $H^*$  as a function of temperature for a field applied both parallel and perpendicular to the c axis of a detwinned crystal of  $YBa_2Cu_3O_{7-s}$ , which is determined from the peak in the hysteresis loop.

the remnant field, the sample was first held 6—8 cm above the magnet and cooled down to a temperature well below the transition temperature to insure the sample was in the superconducting state under zero field. Then the sample was slowly moved downward to the desired position. Any small remnant field would not affect the sample since it was well below  $H_{c1}$ , the lower critical field. After the temperature stabilized, there was another 120-sec waiting time to allow the sample to reach a completely stable state. Then a magnetic field was carefully applied in a no-overshoot mode. The fluctuation (variation} of field in this mode was estimated to be less than 0.5 G.

Because of the anisotropy present in this material, the orientation of the crystallographic axes must be aligned carefully with respect to the applied field. The degree of orientation or the demagnetization factor can be obtained from the diamagnetic measurement by taking into account the full shielding effect.

### RESULTS AND DISCUSSION

The relaxation data were taken over a time up to  $5 \times 10^5$  sec at temperatures between 5 and 55 K and in the range of applied field of <sup>1</sup> and 5.5 T. At temperatures higher than 55 K, the relaxation appeared to be negligible within the measurement accuracy. The applied field was chosen to be larger than  $H^*$  to ensure complete flux penetration of the sample. A normal-state baseline was observed to be temperature independent from  $T_c$  to 150 K, which indicates the background was due to the sample holder and could be subtracted from the data. Figure 2 gives the resultant magnetic relaxation of a  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  detwinned crystal for the field oriented both parallel and perpendicular to the c-axis plane at  $T=5$  K. An experimental uncertainty of  $\pm 0.5\%$  was estimated from the measurement. From these data, it is apparent that for a sufficiently short-time interval, the dependence of the magnetization appears logarithmic in time [see inset of Fig. 3(a)]. However, notable curvature



FIG. 2. Magnetization vs time at temperature of 5 K and applied field of 5 T of a dewinned  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  crystal over a long-time period of  $5 \times 10^5$  sec (a) for field parallel to the c axis and (b) for field parallel to the ab plane of the crystal.

begins very early ( $\sim$  10<sup>4</sup> sec) and continue throughout the entire measurement (up to  $5 \times 10^5$  sec). As shown in Fig.  $3(a)$  in a semilogarithmic scale, the departure from linearity is approximately  $24\%$ , which is significant when compared with the  $40\%$  decrease of M in the time interval between  $10^4$  sec and  $5 \times 10^5$  sec. This reflects that the logarithmic time decay, i.e.,  $M \sim \ln t$ , based on the classical thermally activation theory, may not be applicable over a sufficiently long-time period. The same data in Fig. 2 are shown in Fig. 4 in a log-log plot where the solid line in the figure represents a linear fit between  $\ln M$  and  $\ln t$ . This linear fit is remarkably accurate over almost four decades in time. This indicates that our experimental results provide a good fit to the Vinokur model assuming a logarithmic current dependence of the activation energy, which allows the data analysis in the form suggested by Eq. (5).

From the power-law time dependence of the magnetization in our experiments,  $M = at^{-b}$  (both a, b are fitting parameters), one can obtain  $T \ln |dM/dt| \propto \ln M$  by a simple mathematic derivation. Since  $U_{\text{eff}} \propto T \ln |dM/dt|$  and  $M \propto J$ , a logarithmic current dependence of the activated energy barrier,  $U_{\text{eff}}(J) \propto \ln J$ , can be determined. This is inconsistent with the previous observations.<sup>11,12</sup> inconsistent with the previous observations.<sup>11,12</sup>

Figure 5 illustrates the experimental measurements at various temperatures and the same field applied both parallel and perpendicular to the c axis with the corresponding power-law fits indicated by the solid lines. Further scrutiny of this time dependence reveals that the quality of fitting is seen to deteriorate slightly for the relaxation data taken at high temperatures. Although the power-law fits become poor at high temperatures, it is found that the power-law fits are always better than the logarithmic fits for all the temperatures and applied fields. The temperature dependence of the logarithmic relaxation rate  $S = d \ln M / d \ln t = |dM/Md \ln t|$  is given in Fig. 6, which is obtained from the slope of each curve of Fig. 5. This is our central result and is used to compare with the theoretical solutions.<sup>13</sup> From the plot, the relaxation rate  $S$  is found to be linearly increasing with temperature up to 40 K, which is consistent with Eq. (5) or Eq. (7) in a fully penetrated state where the theory<sup>13</sup> presumes that the temperature is so low that  $\sigma = U_0 / k_B T \gg 1.$ 

By extrapolating linearly to zero temperature, a finite intercept was found. This indicates that the rate S does



FIG. 3. Magnetization as a function of time at 5 K and 5 T of a detwinned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> crystal on a semilog scale for field (a) parallel to the  $c$  axis and (b) parallel to the  $ab$  plane of the crystal. The solid line represents a linear fit. The figure indicates a significant departure from the linear fit on the logarithmic scale, which is expected from the classical flux-creep model. The inset of the figure exhibits a good fit over a short-time period ( $\sim 10^3$ ) sec).

not vanish when  $T\rightarrow 0$ , which corresponds to a nonthermal activation flux creep. In the thermally activated flux-creep theories, magnetic relaxation of type-II superconductors should disappear at zero temperature. For example, Eq. (5) of Vinokur's solution also shows that  $S=k_BT/U_0 \rightarrow 0$  as temperature approaches zero. Our results confirm the existence of a low-temperature magnetic relaxation and show that the  $T=0$  relaxation should be temperature independent. At sufficiently low temperatures where the thermal processes are frozen out, we could expect some process such as quantum tunneling of vortices to dominate the relaxation process so that  $S_{\text{tunneling}} >> S_{\text{thermal}}$ . Another reason to expect quantum tunneling of vortices is the small coherence length of high-temperature superconductors. However, our simple extrapolation may not be adequate to quantitatively establish the model of quantum tunneling of vortices. Direct measurements of magnetic relaxation at ultralow temperature rather than attempting extrapolations from the high-temperature range were carried out by severa groups.<sup>21,22</sup> Fruchter et al.<sup>23</sup> reported a significant relaxation in the low-temperature range of  $0.1-1$  K of an  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  crystal by a Hall measurement. Their



FIG. 4. Magnetization time relaxation of a detwinned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> crystal at  $T=5$  K and  $H=3$  T for field (a) parallel to the c axis and (b) parallel to the ab plane of the crystal in a log-log scale. The solid lines are power-law fits to Eq. (4) in the text.



FIG. 5. Magnetization time relaxation of a  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$ detwinned crystal observed in a 3 T with field applied (a) parallel and (b) perpendicular to the  $c$  axis over the temperature range of 10-50 K. The solid lines represent power-law fits at different temperatures. However, the fitting curves start to deviate from the data slightly at high temperatures. Overall, the power-law fits are always better than the logarithmic fits for all the temperatures and fields.

10<sup>5</sup> 10<sup>6</sup> studies present evidence from the existence of quantum<br>10<sup>5</sup> 10<sup>6</sup> studies of vertices. However, the field density is aftunneling of vortices. However, the field dependence of the tunneling was not discussed. Our analysis shows that the  $T = 0$  relaxation is strongly field dependent.

> By subtracting the temperature-independent part of the flux creep, we can obtain a relaxation rate for only



FIG. 6. The temperature dependence of the fitting parameter  $S=d \ln M/d \ln t$  (relaxation rate) for  $H = 5$  T with field applied parallel to the  $c$  axis over the temperature range of  $5-55$  K. The relaxation rates are linear at low temperatures ( $T < 40$  K).



FIG. 7. The relaxation rate  $S$  vs temperature with the field applied (a) parallel and (b) perpendicular to the c axis at temperatures below 40 K. The relaxation rates show a linear temperature dependence at different fields, which yields a temperatureindependent effective activation energy  $S = k_B T/U_0$ . However, S is found to be a function of the applied field, which indicates that the efFective activation energy is field dependent.

the thermal activation  $S_{\text{thermal}}$ . Our result is shown in Fig. 7, which yields a thermally activated energy barrier  $U_0$  as a function of field.

In our experiments at various temperatures and fields, we did not observe any sharp transitions between the partial and full penetration regimes when the flux fronts from opposite sides of the sample collapses at the center  $[x_f(t)=d/2]$ . Recently, Schnack and Grissen<sup>14</sup> obtained the numerical results of magnetization from the differential equation and concluded that there is no kink, no sharp transition in the relaxation plot when the flux fronts merge at the center of the sample. This is consistent with our experimental results.

A notable feature of the thermally activated flux-creep rate is that S<sub>thermal</sub> has not only temperature dependence but field dependence as well as shown in Fig. 8. This has 'also been discussed in the papers of Zeldov et al.,<sup>10</sup> Liu et  $al$ ,<sup>15</sup> and Zhang et  $al$ .<sup>5</sup> The thermally activated activation energy  $U(T, H, J)$  can be generally written as

$$
U(T,H,J) = U(T,H) \ln(J_c/J)
$$
.

In the analysis of the experimental data, it was found that  $U(T, H)$  is almost temperature independent at low temperatures, which is consistent with the  $(1-T/T_c)^{\alpha}$ -type behavior predicted theoretically. The field dependence of  $U_0$  shown in Fig. 8 is seen to be nonlinear in H and appears to follow a  $ln H$  dependence (see inset of Fig. 8), again similar to the previous theoretical prediction.<sup>5</sup>

## **CONCLUSION**

We measured the magnetic relaxation on an  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  detwinned crystal over a long-time period of  $10^5$  sec. The power-law time dependence of the magnetization was obtained at various temperatures and fields with the field oriented both parallel and perpendicular to the  $c$  axis of the crystal. The measurements can



FIG. 8. Field dependence of effective activation energy for both field orientations. Inset indicates straight lines of  $U_0$  vs  $H$  in a semilog scale.

be well interpreted by the model of Vinokur, Feigel'man, and Geshkenbein, $^{13}$  which yields a thermally activated energy barrier as a function of field. The results of this study suggests that the theory could be extended to include field dependence and applied on both field orientations.

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