# Temperature and field dependence of magnetic relaxation in a $Bi_2Sr_2CaCu_2O_x$ single crystal

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We measured the magnetic relaxation in a  $Bi_2Sr_2CaCu_2O_x$  single crystal at a wide range of temperatures (6-25 K) and field strengths (100-4000 G). We found that the relaxation rate  $(dM/d \ln t)$  is highly temperature and field dependent. By expanding Kim's model, we developed an expression for  $dM/d \ln t$  as a function of field that we used to interpret the rise and fall of the magnetic relaxation in both the  $H < H^*$  and  $H > H^*$  regions.

#### INTRODUCTION

Strong magnetic relaxation has been observed in all high- $T_c$  superconductors.<sup>1-5</sup> This phenomenon raises fundamental questions about the nature of the mixed state at high fields  $(H > H_{c1})$ , where properties such as magnetic irreversibility, vortex lattice behavior, flux pinning, and critical current density are found to be essentially different from those of conventional superconductors.

Muller *et al.*<sup>1</sup> attributed the large magnetic relaxation to the glassy characteristics of weakly lined superconducting grains in the samples. Later, however, it was found that single crystals of high- $T_c$  superconductors exhibit similar magnetic relaxation.<sup>2,3</sup>

Based on both magnetic and transport measurements, a flux-creep model was then proposed to explain the large magnetic relaxation.<sup>2,6,7</sup> These studies indicated that the magnetization decays logarithmically in a wide range of temperature and field strength. The studies also reported that the motion of the flux lines is thermally activated. The activation energies of high- $T_c$  superconductors were estimated to be about 100 times lower than those of conventional superconductors. More important, an irreversibility line was found below the  $H_{c2}(T)$  boundary in the H-T phase diagram, which shows that high- $T_c$  superconductors have a distinctly different mixed state from that of conventional superconductors.

In contrast with the flux-creep model, Fisher proposed the so-called vortex-glass superconductivity model and demonstrated that an equilibrium phase boundary exists in the H-T plane.<sup>8</sup> Crossing this boundary, the flux lines transform into a new state: vortex-glass, in which a true superconducting state is present. Koch *et al.* later reported experimental results in epitaxial YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> thin films. Their data support the vortex-glass model and indicate a phase transition from vortex liquid to vortex glass.<sup>9</sup>

In this paper we present magnetic relaxation data for a  $Bi_2Sr_2CaCu_2O_x$  single crystal. We develop an expression for the magnetic relaxation rate  $dM/d \ln t$  and interpret the rise and fall of the magnetic relaxation in different range of applied field. We discuss the relationships be-

tween the relaxation rates and the temperature and the applied field.

# **EXPERIMENTAL PROCEDURE**

A single crystal was grown with the flux method.<sup>10</sup> The quality of the crystal was examined by x-ray diffraction, resistivity, and magnetization measurements. The x-ray-diffraction results showed a single  $Bi_2Sr_2CaCu_2O_x$  phase. A sharp superconducting transition at 86 K was observed in both the resistivity and magnetization experiments. The magnetization data were taken by using commercial SQUID magnetometer over a wide range of temperatures (6-25 K) and applied fields (100-4000 G). The sample was first cooled in zero magnetic field to a desired temperature T below the transition temperature  $T_c$ . A magnetic field H was then applied and the magnetization M of the sample was measured as a function of time t. The initial data point of the magnetization was taken at t=180 s. The direction of the applied field was normal to the a-b plane of the single crystal.

### RESULTS

Figure 1 shows magnetization versus time data plots at different field for a constant temperature of 8 K. As can be seen in the figure, the magnetization exhibits small relaxation rates at low fields (0-1000 G), which gradually increase with increasing field and reach a maximum near 1500 G. The relaxation rate then decreases with increasing field thereafter. Figure 1 shows that considerable magnetic relaxation is still present at the highest field (5000 G).

In Fig. 2 we plot the relaxation rate  $(dM/d \ln t)$  as a function of field for different temperatures indicated. It should be noted that the  $dM/d \ln t$  values are the slope of the M versus  $\ln t$  curves shown in Fig. 1, which are taken between  $\ln t = 7$  and  $\ln t = 9$ , since the relaxation rate becomes constant at a given field in this time interval. Again, we note, the relaxation rate increases with the increasing field at low-field regions. Figure 2 shows a peak shift of the  $dM/d \ln t$  values at different temperatures.

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FIG. 1. Magnetization vs time at a given temperature T=8 K for a Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> single crystal at the various field indicated. The field, parallel to c, is applied after cooling the sample in zero field.

Although some  $dM/d \ln t$  peaks are not seen in the figure because of the limited data points, we can distinguish a clear peak shifting associated with the temperature change. Figure 2(a) shows the relaxation versus applied field data at 6, 8, and 10 K. The relaxation rate at 8 K initially increases with the increasing field in the low-field region. It reaches a maximum value near 1500 G and



FIG. 2. Magnetic relaxation rate  $dM/d \ln t$  vs field at various temperatures. The solid lines are a guide for the eye.

then gradually decreases. The field,  $H_p$ , at which the  $dM/d \ln t$  experiences the peak also shifts towards the low fields with the increasing temperature. As shown in Fig. 2(a), the  $dM/d \ln t$  peak takes place at near 1000 G as the temperature is increased to 10 K. However, the  $H_p$  value at 6 K cannot be estimated well in this experiment because of the limited data points. As indicated in Fig. 2(b), the  $dM/d \ln t$  peak may have shifted to much lower field regions (below 500 G) when the temperature increases to above 15 K. The absolute values of  $dM/d \ln t$  at different fields are also significantly suppressed by increasing the temperature. As shown in Fig. 2(b), the  $dM/d \ln t$  value at 1000 G drops from 150 emu/cm<sup>3</sup> to zero as the temperature increases from 8 to 25 K.

### DISCUSSION

Yeshurun et al.<sup>2,3,11</sup> explain the magnetic relaxation based on Anderson's classic flux-creep model.<sup>12</sup> The model assumes certain pinning mechanisms caused by inhomogeneities in the materials. An Abrikosov vortex sitting in a potential pinning well with a height of  $U_0$  may be activated an hop out of the well as a result of thermal excitation. Such motion of the flux lines results in magnetic relaxation and reduction of the critical current density,  $J_c$ . Yeshurun et al. extended Bean's critical state model and qualitatively described the magnetic relaxation in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> and Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> single crystals.

According to Bean's model,<sup>13</sup> the local critical current  $J_c(T, H_i)$  is assumed to be a constant,  $J_c(T)$ , independent of the local field  $H_i$ . This model was later modified by Kim<sup>14</sup> can be written as

$$J_c(T,H_i) = \frac{J_c(T)}{1 + H_i / H_0(T)} , \qquad (1)$$

where  $H_0$  is a material parameter with magnetic field dimension which can be determined experimentally. In the studies by Yeshurun *et al.*,<sup>3,11</sup> the  $H_0$  value is assumed to be comparable to  $H_{c1}$  and is much lower than  $H_i$ . They extended Eq. (1) and obtained

$$J_{c}(T,H_{i}) = J_{c}(T)(H_{0}/H_{i})^{n} .$$
<sup>(2)</sup>

For n = 1, Eq. (2) can be derived from the Kim model [Eq. (1)] with condition  $H_i \gg H_0$ . For n = 0, Eq. (2) becomes the expression of Bean model. Using this assumption, Yeshurun *et al.* derived an expression for the magnetic relaxation in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> single crystals.

However, the condition  $H_i \gg H_0$  is not always satisfied as we indicate later. Moreover, Watson suggested that  $H_0$  can be much greater than  $H_i$  for conventional superconductors.<sup>15</sup> With these considerations, we expand Kim's model Eq. (1) for  $H_i \ll H_0$  and obtain

$$J_{c}(T,H_{i}) = J_{c}(T)(1 - H_{i}/H_{0}) .$$
(3)

In a critical state we have

$$-dH_i/dx = 4\pi J_c(H_i)/c , \qquad (4)$$

where c is the speed of light (we use Gaussian units

throughout the paper). If we consider a slab of thickness D with the field parallel to the plane of the slab and assume  $H_{c1}$  negligible, we can develop an expression for the local field,  $H_i$ ,

$$H_i(x) = H_0 - (H_0 - H) \exp(x / x_0) , \qquad (5)$$

where  $x_0 = cH_0/4\pi J_c(T)$  and H is the applied magnetic field. The average magnetic induction is given by

$$\langle B \rangle = \frac{2}{D} \int_0^{D/2} H_i(x) dx \quad . \tag{6}$$

Substituting Eq. (5) into Eq. (6) and considering appropriate boundary conditions, we obtain

$$\langle B \rangle = -H_0 \frac{2x_0}{D} \ln \left[ 1 - \frac{H}{H_0} \right] - \frac{2x_0}{D} H, \quad H \le H^*, \quad (7a)$$
  
 $\langle B \rangle = H_0 + \frac{2x_0}{D} \left[ \exp \left[ \frac{D}{2x_0} \right] - 1 \right] (H - H_0),$ 

where  $H^* = H_0[1 - \exp(-D/2x_0)]$ , which is the field required for the flux to first completely penetrate the sample. According to Campbell and Evetts<sup>16</sup>

$$J_{c}(T) = J_{c0} [1 - (kT/U_{0})\ln(t/t_{0})], \qquad (8)$$

 $H \geq H^*$ ,

(7b)

where  $J_{c0}$  is the critical current density when the thermal disturbance is not present,  $1/t_0$  is the attempt frequency for the flux lines to jump over the pinning well, and  $U_0$  is the activation energy for the motion of the flux lines. Substituting Eq. (8) into Eq. (7) and knowing that the magnetization  $4\pi M$  is given by  $\langle B \rangle - H$ , we take the derivative of the magnetization with respect to time to the first-order approximation in  $kT/U_0$ . We obtain<sup>17</sup>

$$4\pi \frac{dM}{d\ln t} = \begin{cases} \alpha (H^2/H_0)(kT/U_0), & H \le H^*, \\ \beta (-H+H_0)(kT/U_0), & H \ge H^*, \end{cases}$$
(9a)

where  $\alpha = x_0 / D$  and

$$\beta = (2x_0/D) \{ 1 + [(D/2x_0) - 1] \exp(D/2x_0) \} .$$

It should be pointed out that the induction  $\langle B \rangle$ .  $d\langle B \rangle/dH$ , and  $dM/d \ln t$  [Eq. (9)] are all continuous at  $H = H^*$ . However, we noticed that  $d^2M/dH^2$ ,  $d^2M/(d \ln t)^2$ , and  $d^2M/dHd \ln t$  are discontinuous at  $H = H^*$ . From Eq. (9a) we can see that in the low-field region with  $H < \dot{H}^*$ ,  $dM/d \ln t$  increases with  $H^2$ . By fitting the experimental data of  $dM/d \ln t$  taken at 8 K with Eq. (9a), we find the results are quite reasonable [see the solid line (a) in Fig. 3]. As indicated in Eq. (9b),  $dM/d \ln t$  should decrease linearly with the applied field. This is confirmed by fitting  $dM/d \ln t$  data in the higherfield region  $(H > H^*)$  [see the solid line (b) in Fig. 3]. We have obtained the parameters  $J_c$ ,  $H_0$ , and  $U_0$  through fitting the experimental data to Eq. (9), which are  $10^5$ A/cm<sup>2</sup>, 9000 G, and 5 meV, respectively. These values quite reasonably agree with the previously reported results in the Bi-Sr-Ca-Cu-O system.<sup>18</sup>

Based on the Bean model  $J_c(T,H_i) = J_c(T)$ , we can



FIG. 3.  $dM/d \ln t$  vs field at 8 K for a Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> single crystal. The solid lines are the fit to Eq. (9a) (line *a*) and Eq. (9b) (line *b*).

also develop an expression for  $dM/d \ln t$  with the same approach indicated earlier, which can be written as

$$-\frac{dM}{2h^*} = \begin{cases} \frac{H^2}{2h^*} \frac{kT}{U_0}, & H \le h^* \end{cases},$$
(10a)

$$4\pi \frac{dM}{d \ln t} = \begin{cases} 2\pi & -6 \\ \frac{h^*}{2} \frac{kT}{U_0}, & h \ge h^* \end{cases},$$
(10b)

where  $h^*$  is the field for the flux to first penetrate the sample and  $h^* = 2\pi D J_c(T)/c$ . It should be noted that although the physical meanings of  $H^*$  and  $h^*$  are identical, they have different expressions due to employment of different critical state models.<sup>19</sup> For  $H < h^*$ , the same field of  $dM/d \ln t$  ( $\sim H^2$ ) is obtained indicating that the Bean model is applicable in the low-field region. However,  $dM/d \ln t$  becomes independent of field in the region  $H > h^*$ , which disagrees with our experimental data. This results from the fact that the Bean model assumes a constant critical current density which is also independent of magnetic field. Again, we checked the continuity of Eq. (10) and found that the induction  $\langle B \rangle$ ,  $d \langle B \rangle / dH$ , and  $dM/d \ln t$  [Eq. (9)] are all continuous while the second-order derivatives of magnetization such as  $d^2M/dH^2$ ,  $d^2M/(d \ln t)^2$ , and  $d^2M/dHd \ln t$  are not continuous at  $H = H^*$ .

As we indicated before, Yeshurun *et al.* developed an expression for  $dM/d \ln t$  based on the extended Bean model

$$J_c(T,H_i) = J_c(H_0/H_i)^n$$

[Eq. (2)], which well described magnetic relaxation behavior in the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> with n = 1. By fitting the experimental data with Eq. (9), we have estimated the  $H_0$  value to be 9000 G. This value is much greater than the  $H_{c1}$  value in Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub>. Thus, the assumption of Yeshurun *et al.* is that  $H_i \gg H_0$  may not be appropriate for the Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> system. This conclusion implies that Eq. (3) provides a better physical foundation for de-

veloping the field dependence of  $dM/d \ln t$  for  $Bi_2Sr_2CaCu_2O_x$ .

Furthermore, by taking n=0 in Eq. (2) the model developed by Yeshurun *et al.* gives an  $H^2$  dependence of  $dM/d \ln t$  in a Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> single crystal in the  $H < H^*$  region. However, their model is incapable of describing the magnetic relaxation in the  $H > H^*$  region (n=0 results in a constant  $dM/d \ln t$  in the  $H > H^*$  region). Also, since n=0 implies the original Bean model, it thus allows a quite different expression for  $dM/d \ln t$  to be developed as we indicated in Eq. (10). In contrast, our model [Eq. (9)], based on the condition  $H_i \ll H_0$ , is able to fit both the low- and high-field magnetic relaxation well.

From Eq. (9) and Fig. 3 we can assume that the  $H^*$  value should be roughly the field at which  $dM/d \ln t$  experiences a peak  $H_p$ . Therefore,  $H_p$  is directly related to the temperature dependence of  $H^*$ . As we indicated earlier,

$$H^* = H_0[1 - \exp(-D/2x_0)]$$
.

The term  $[1-\exp(-D/2x_0)]$  {= $[1-\exp(-2\pi DJ_c/cH_0)]$ } contains  $H_0$  and  $J_c$  which are both decreasing functions of the temperature. In the low-temperature range (~10 K), where  $J_c$  is large, the condition  $D/2x_0 \ll 1$  is well satisfied and the full penetration field can be written as  $H^* = 2\pi DJ_c(T)/c$  (see Ref. 19). From

this relation,  $H^*$  is directly related to the temperature dependence of the critical current density  $J_c$ . As temperature increases,  $J_c$  is reduced, and thus  $H^*$  shifts to lower field. However, it is difficult to predict the specific temperature dependence of  $H^*$  based on the present experimental data.

#### CONCLUSIONS

We measured the magnetic relaxation in a  $Bi_2Sr_2CaCu_2O_x$  single crystal at various temperatures and applied fields. We observed that the relaxation rate rises and falls with field and that the field  $(H_p)$  at which the maximum dM/d lnt takes place is highly dependent on temperature. By expanding Kim's model we developed an expression for the field dependence of dM/d lnt. Our model agrees excellently with the experimental data and gives some new physical interpretations for the rise and fall of the magnetic relaxation.

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- <sup>1</sup>K. A. Muller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. 58, 1143 (1987).
- <sup>2</sup>Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
- <sup>3</sup>Y. Yeshurun, A. P. Malozemoff, T. K. Worthington, R. M. Yandrofski, L. Krusin-Elbaum, F. H. Holtzberg, T. R. Dinger, and G. V. Chandrashekhar, Cryogenics **29**, 258 (1989).
- <sup>4</sup>T. T. M. Palstra, B. Batlogg, R. B. van Dover, L. F. Schneemeyer, and J. V. Waszczak, Appl. Phys. Lett. 54, 763 (1989); T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. 61, 1662 (1988).
- <sup>5</sup>J. P. Rice, D. M. Ginsberg, M. W. Rabin, K. G. Vandervoort, G. W. Crabtree, and H. Claus, Phys. Rev. B **41**, 6532 (1990).
- <sup>6</sup>M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).
- <sup>7</sup>C. W. Hagen and R. Griessen, Phys. Rev. Lett. **62**, 2857 (1989).
- <sup>8</sup>M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989).
- <sup>9</sup>R. H. Koch, V. Foglietti, W. J. Gallagher, G. Koren, A. Gupta, and M. P. A. Fisher, Phys. Rev. Lett. 63, 1511 (1989).
- <sup>10</sup>Donglu Shi, Ming Tang, Y. C. Chang, P. Z. Jiang, K. Vandevoot, B. Malecki, and D. J. Lam, Appl. Phys. Lett. 54, 2358 (1989).
- <sup>11</sup>Y. Yeshurun, A. P. Malozemoff, F. H. Holtzberg, and T. R. Dinger, Phys. Rev. B 38, 11 828 (1988).
- <sup>12</sup>P. W. Anderson, Phys. Rev. Lett. 9, 309 (1962); M. R. Beasley,

R. Labusch, and W. W. Webb, Phys. Rev. 181, 682 (1969).

- <sup>13</sup>C. P. Bean, Phys. Rev. Lett. 8, 250 (1962); Rev. Mod. Phys. 36, 31 (1964).
- <sup>14</sup>Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. Lett. 9, 306 (1962); Phys. Rev. 129, 528 (1963).
- <sup>15</sup>J. H. P. Watson, J. Appl. Phys. **39**, 3406 (1968).
- <sup>16</sup>A. M. Cambell and J. E. Evetts, Adv. Phys. 21, 199 (1972).
  <sup>17</sup>It should be noted that the original form of Eq. (9a) is

$$4\pi \frac{dM}{d \ln t} = H_0 \frac{2x_0}{D} \left[ -\ln \left[ 1 - \frac{H}{H_0} \right] - \frac{H}{H_0} \right] \frac{kT}{U_0}, \quad H \le H^*$$
(9a')

As we assume  $H/H_0 \ll 1$ , the term  $\ln(1-H/H_0)$  can be expanded to be  $-H/H_0 - (H/H_0)^2/2$ . Thus, Eq. (9a') is simplified and can be written as Eq. (9a).

- <sup>18</sup>Y. Yeshurun, A. P. Malozemoff, T. K. Worthington, R. M. Yandrofski, L. Krusin-Elbaum, F. H. Holtzberg, T. R. Dinger, and G. V. Chandrashekhar, Cryogenics **29**, 258 (1989).
- <sup>19</sup>We show  $H^* = h^*$  as the condition  $H_0 \gg 2\pi J_c(T)D/c$ is satisfied. As we indicated in the paper,  $H^* = H_0[1 - \exp(-D/2x_0)]$ . The condition  $H_0$  $\gg 2\pi J_c(T)D/c$  is equivalent to  $D/2x_0 \ll 1$  where  $x_0 = cH_0/4\pi J_c(T)$ . The term  $\exp(-D/2x_0)$  can be expanded to be  $(1 - D/2x_0)$  as we take the first-order approximation in  $D/2x_0$  if  $D/2x_0 \ll 1$ . Thus,  $H^*$  can be written as  $H^* = H_0 D/2x_0 = 2\pi J_c(T)D/c = h^*$ .