## Nonlinear logarithmic time decay of magnetization in a single crystal of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub>

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Nonlinear logarithmic time decay of magnetization has been observed over a long measuring time period (t = 22 h) for a single crystal of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub>. At relatively low temperatures (T = 12 K) linear ln(t) decay of the magnetization is observed for field (= 1 kG) parallel to the c axis, which can be described well by the Anderson-Kim model. However, at high temperatures (T > 26 K) the magnetization exhibits nonlinear logarithmic time near the irreversibility line. Possible connectons between the experimental data and current theories are discussed.

Time dependence of magnetization has been observed in both conventional and high- $T_c$  superconductors. According to the Anderson-Kim flux-creep model, 1,2 the time decay of magnetization is logarithmic for conventional superconductors. Recent studies have shown that some nonlinear logarithmic time decay of magnetization occurs in certain temperature and field regions for a short measuring time (<3h).<sup>3,4</sup> This behavior reflects the complexity of the magnetic properties and is closely related to the so-called irreversibility line observed in high- $T_c$  superconductors.<sup>5</sup> Because of the existence of the irreversibility line in high- $T_c$  superconductors, the magnetic relaxation and flux dynamics could be fundamentally different from those of conventional superconductors.<sup>6</sup> Feigel'man et al.<sup>7,8</sup> proposed a collective-creep model

that predicts the nonlinear logarithmic decay of current density. The model is developed based on a concept of collective pinning.9 The long-range translational order of the flux-line lattice may be destroyed by crystal defects. But the short-range order still remains in a correlation volume  $V_c$ . The entire vortex system can still be strongly pinned, although the individual flux pinning is relatively weak. Therefore the volume  $V_c$  may be considered as one large pinning center (i.e., the flux lines can be effectively pinned by an ensemble of weak pins). The collective pinning behavior has been observed in some conventional superconducting systems such as amorphous Nb<sub>3</sub>Ge films. 10 However, no convincing experimental evidence has indicated such a pinning effect in high- $T_c$  superconducting oxides.

In this paper, we present magnetization (M) versus time (t) data for a single crystal of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub>. We have clearly observed nonlinear logarithmic time decay of magnetization over a long measuring time period (t=22 h) and wide temperature region (12-40 K). We discuss the nonlinear behavior of the magnetic relaxation based on collective-creep and Anderson-Kim flux-creep models.

The single crystal of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> was grown by using a so-called flux method reported previously.<sup>11</sup> The crystal was  $2 \times 1.5 \times 0.1$  mm<sup>3</sup>. The sample quality was examined by x-ray diffraction and magnetization measurements on a commercial Quantum Design SQUID magnetometer. The x-ray data showed that the crystal possessed a single phase and high purity. The zero-fieldcooled SQUID measurements at 20 G also showed that the single crystal exhibited a sharp transition at 86 K.

Measurements of magnetization versus time (M versus t) were performed in the following sequence. The sample was cooled to a temperature below  $T_c$  in a zero magnetic field. An external field of 1 kG was then applied to the crystal. According to our previous measurements, 12 at 1 kG, the sample was fully penetrated above 10 K. Our magnetization measurements in this study took place above 12 K, to ensure the full penetration of the field into the sample. The first magnetization data point was taken 2 min after the application of the field. Subsequent magnetization data were taken every 6.3 min for a total of 22 h. After the measurements were completed at a given temperature, the temperature of the sample was raised to 120 K for the purpose of completely removing the trapped flux lines inside the sample. The scan length was set at 30 mm. The field was monitored by a calibrated Hall probe and directly read by the computer. The temperature and applied field control accuracy was  $\pm 0.007$  K and less than  $\pm 0.001$  G over the 22-h time period.

In Fig. 1 we show *M* versus *t* data at 12, 20, 26, 30, 34, and 40 K for the direction of the field parallel to the c axis of the crystal ( $\mathbf{H}||c$ ). As shown in Fig. 1(a), at a relatively low temperature (T=12 K) the magnetization exhibits linear logarithmic time decay, particularly at the initial stage of relaxation. However, a small curvature develops as the temperature is increased to 20 K [Fig. 1(b)]. The magnetic relaxation exhibits nonlinear logarithmic decay at even higher temperatures; at 26 K [Fig. 1(c)], 30 K [Fig. 1(d)], 34 K [Fig. 1(e)], and 40 K [Fig. 1(f)]. It should be pointed out that, according to the previously reported results,<sup>5</sup> the irreversible temperature  $T^*(H)$  is near 30 K at 1 kG (see Fig. 2). This fact indicates that the nonlinear behavior is connected with the irreversible nature of the Bi-Sr-Ca-Cu-O superconducting system.

Similar time dependence of magnetization has been observed when the direction of the field is parallel to the ab plane of the crystal  $(\mathbf{H}||ab)$ , except that the temperature at which the magnetic relaxation becomes nonlinear increases to 34 K. As shown in Figs. 3(a) through 3(d), magnetic relaxation is almost linear at 26 and 30 K, while nonlinear behavior is observed at 34 and 40 K. These results clearly indicate that the irreversibility line for  $\mathbf{H} \parallel ab$  has shifted to a much higher temperature and field region, consistent with the anisotropy effect in a Bi-Sr-Ca-Cu-O system.

Feigel'man et al. considered that the thermal activation energy, U, has a power-law dependence on the current density:  $U(J) = U_0(J_c/J)^{\mu}$ , where  $U_0$  is the characteristic energy scale and  $\mu$  is a dimensionality and current-related parameter.<sup>7,8</sup> If the process is thermally activated, the hopping time of the flux lines  $t=t_0\exp(U/kT)$ . Combining the two expressions for U

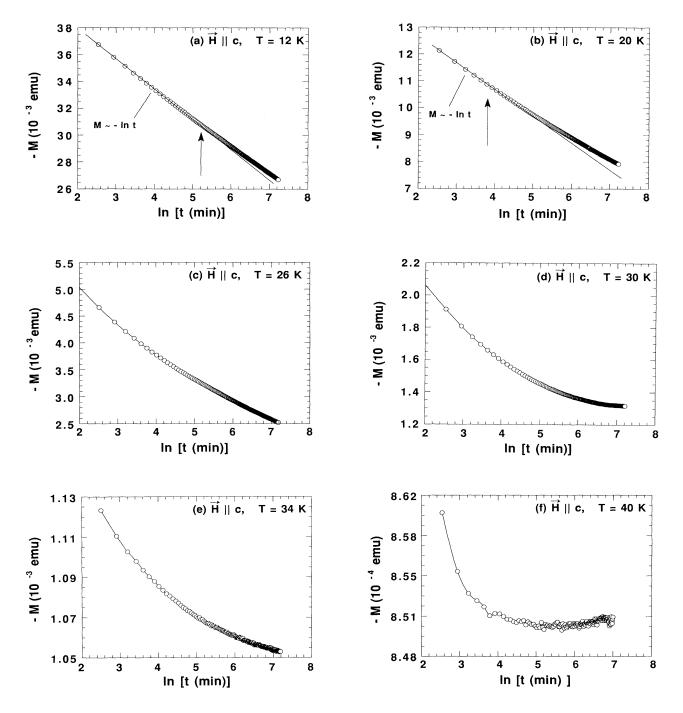


FIG. 1. Magnetization vs time at a given field H = 1000 G for a  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  single crystal at various temperatures indicated. The field, parallel to the c axis, is applied after cooling the sample in zero field. (a) T = 12 K, (b) T = 20 K, (c) T = 26 K, (d) T = 30 K, (e) T = 34 K, (f) T = 40 K. The solid lines are a guide for the eye.

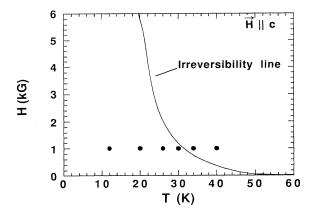


FIG. 2. The irreversibility line for  $\mathbf{H} || c$  (Ref. 5) in the H vs T phase diagram for the Bi-Sr-Ca-Cu-O system. The temperature and field regions in which the magnetic relaxation experiments were performed are indicated in the figure.

and t, we can write the time dependence of the current density as

$$J(t) = J_c [(T/U_0) \ln(t/t_0)]^{-1/\mu} . \tag{1}$$

According to the Anderson-Kim flux-creep model,

 $U(j) = U_c (1-J/J_c)$ , where  $U_c = \mu U_0$ . Solving the diffusion equation for flux motion in Refs. 1 and 2 with the condition U/kT >> 1, one can obtain a linear logarithmic decay of current density,

$$J(t) = J_c [1 - (T/U_c) \ln(t/t_0)]. \tag{2}$$

Thus, interpolation of Eq. (1) has the form

$$J(t) = J_c [1 + \mu (T/U_c) \ln(t/t_0)]^{-1/\mu}.$$
 (3)

The collective-creep model [Eq. (3)] has been claimed to offer a general description for current density decay of high- $T_c$  superconductors. The model states that the long-range order of the flux-line lattice is destroyed by random pinning centers such as oxygen vacancies in high- $T_c$  superconductors. According to this model, the short-range order still exists and results in collective pinning in a finite volume,  $V_c (= L_c R_c^2)$ , where  $L_c$  and  $R_c$  are the longitudinal and transverse pinning lengths, respectively. The size of the pinning volume is closely related to  $\mu$ , which will be discussed in detail later.

Since the time dependence of the current density describes a decay of the metastable state, the magnetization time decay should follow the same law as the time decay of the current density. Thus, Eqs. (1), (2), and (3) can be written as

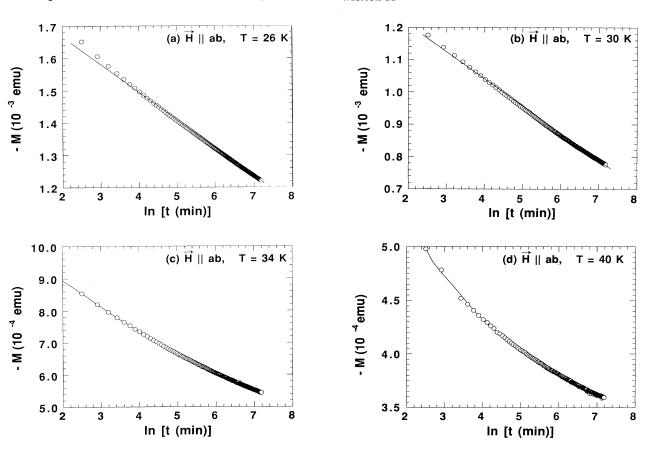


FIG. 3. Magnetization vs time at a given field H=1000 G for a  $Bi_2Sr_2CaCu_2O_x$  single crystal at various temperatures indicated. The field, parallel to the *ab* plane of the crystal, is applied after cooling the sample in zero field. (a) T=26 K, (b) T=30 K, (c) T=34 K, (d) T=40 K. The solid lines are a guide for the eye.

$$M(t) = M_0[(T/U_0)\ln(t/t_0)]^{-1/\mu}, \qquad (4)$$

$$M(t) = M_0[1 - (T/U_c)\ln(t/t_0)],$$
 (5)

$$M(t) = M_0 [1 + (T/U_c) \ln(t/t_0)]^{-1/\mu}, \qquad (6)$$

where  $M_0$  is the magnetization when there is no fluxcreep effect. It should be noted that, for conventional superconductors, the activation energy  $U_c$  is on the order of 1 eV. If we assume T = 10 K, then  $T/U_c \sim 10^{-3}$ . For measuring time of  $10^4$  sec,  $\ln(t/t_0) \sim 30$ . This implies that  $(T/U_c) \ln(t/t_0) \ll 1$ . Thus, Eq. (6) can be expanded and written as Eq. (5), which is the expression for flux creep in the Anderson-Kim model.

Equation (5) may hold true for high- $T_c$  superconductors in the low-temperature region as long as the condition  $(T/U_c)\ln(t/t_0) \ll 1$  is satisfied. However, high- $T_c$ superconductors have considerable lower  $U_c$  values compared to conventional ones, especially in a Bi-Sr-Ca-Cu-O system. Indeed, previous (5) have shown that the  $U_c$ value in a single crystal of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> is on the order of 10 meV at 10 K, which gives  $(T/U)\ln(t/t_0) \le 1$ . Here, we see that the magnetization time decay may not be precisely described by Eq. (5). Obviously, the condition  $(T/U_c)\ln(t/t_0) \ll 1$  becomes worse in the hightemperature region as  $U_c$  is severely reduced. Moreover, the linear logarithmic decay [Eq. (5)] cannot hold for large t.

In the collective-creep model, the pinning strength is characterized by the length  $L_c$  when  $R_c$  is smaller than the lattice constant, a.  $L_c = \pi [\phi_0^4 \xi^2 / (2\pi)^4 a^2 W \lambda^4]^{1/3}$ , where  $\phi_0$  is the flux quanta,  $\xi$  is the coherence length, Wis the mean square value of pinning force generated by defects, and  $\lambda$  is the penetration depth. In the case of  $\xi < L_c < a$ , and a relatively high current density  $[J_c > J > J_1$ , where  $J_1 = J_c (L_c / a)^{7/5}$ ],  $\mu$  is 0.14. For an intermediate current density  $(L_c \ge a \text{ and } J \le J_1), \mu = 1.5,$ which is on the same order of magnitude as obtained in our experiments. For a relatively low current density region  $[J_1 > J > J_2$ , where  $J_2 = J_1(a/\lambda)^2$ ,  $\mu$  is predicted to be 0.77. Thus, the nonlinearity of magnetic relaxation is characterized by  $\mu$ , which depends solely on the magnitude of current decay.

It should be pointed out that the zero-field-cooled magnetization data can be strongly influenced by the experimental procedures in the SQUID measurement. Since the current decay occurs while the field is increasing, the magnetic relaxation is affected by the field ramping rates and by the field values at which the relaxation data are taken. For example, the decay process at 34 K within a few minutes can be in a current regime far away from that at 12 K, in which the magnetic relaxation is much slower. Thus,  $M_o$  in Eq. (6) cannot be determined experimentally and rapid change in the field rampingdependent current decay processes makes it difficult to connect the measured magnetization directly with the assumed current regime  $(J \ll J_c)$  in any flux-creep models. Because of the above reasons, we have experienced difficulties in fitting our magnetic relaxation data with Eq. (6) and obtaining meaningful  $M_0$  and  $t_0$ . According to Feigel'man *et al.*, 8 the linear decay of

magnetization [Eq. (5)] occurs at the initial relaxation stage when  $(J-J_c) \ll J_c$ . Collective creep takes place as the current density has significantly decayed from the critical state,  $J_c$ . This is schematically illustrated in Fig. 4. This phenomenon implies that the nonlinear behavior of magnetic relaxation should be observed in a relatively small current region. As shown in Fig. 1, the linear decay of the magnetic relaxation occurs at low temperatures (T = 12 - 20 K for H||c), indicating relatively large current densities. At high temperatures (T > 26 K), however, the current density decays into the collective- creep regime. Therefore, the entire M versus  $\ln(t)$  curves exhibit nonlinear behavior. We have found that the linear decay portion of the magnetic relaxation varies considerable at different temperatures. As shown in Fig. 1(a), at T=12 K, the magnetization time decay exhibits linear logarithmic time decay at the initial relaxation stage. A small curvature is developed at  $t = 120 \text{ min } (\ln t \approx 4.8)$ with lower decay rate. The time at which the curvature begins becomes shorter ( $t \approx 45 \text{ min}$ ,  $\ln t \approx 3.8$ ) as the temperature is increased to 20 K [Fig. 1(b)]. The magnetic relaxation exhibits nonlinear logarithmic decay at temperatures above 26 K. At relatively low temperatures (T=12 and 20 K), the current decay from the critical state is slow. It was measured in a finite time interval  $(\Delta t = 6.3 \text{ min})$  by the SQUID magnetometer. At high temperatures (T > 26 K), the current decay becomes much faster, so that the linear relaxation part cannot be observed. Therefore, the entire M versus lnt curves exhibit nonlinear decay.

We note that the measuring time scale is extremely important in observing the true magnetic relaxation behavior. For example, at 12 K, the linear decay portion extends to 120 min, which is longer than measuring times used in most of the reported studies. However, nonlinear decay will become pronounced as the measuring time is prolonged to a large scale (in this study, t = 22 h) at this temperature. It is also important to note that much longer linear magnetization decay should be expected at extremely low temperatures.

It should be noted that the collective-creep theory

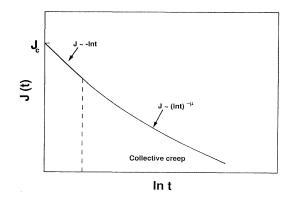


FIG. 4. Schematic expression for the time decay of current density in high- $T_c$  superconductors. Notice that in the collective- creep regime,  $\mu$  varies as a function of current. The decay rate of current density is dependent on temperature.

deals only with the situation when the field is parallel to the c axis of the single crystal. Completely different pinning mechanisms may exist when the field is parallel to the ab plane of the crystal (Fig. 3). New physical models must be developed to interpret the magnetic relaxation behavior for  $\mathbf{H} \| ab$ .

In summary, we have found a nonlinear logarithmic decay of magnetization in a single crystal of  $Bi_2Sr_2CaCu_2O_x$  over a long measuring time period (t=22 h). The nonlinear magnetic relaxation may be connected with the collective creep in the system. The

different degree of nonlinearity in the M versus  $\ln(t)$  curves may reflect the fact that  $\mu$  varies in the different current regimes as predicted by the collective-creep model:  $J(t) = J_c [(T/U_0) \ln(t/t_0)]^{-1/\mu}$ .

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