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# Long-term nonlogarithmic magnetic relaxation in single-crystal $YBa_2Cu_3O_7$ superconductors

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The time dependence of the irreversible superconductive magnetization (flux creep) has been studied in a high-$J_c$, proton-irradiated $YBa_2Cu_3O_{7-x}$ single crystal for long periods, up to $3.5\times10^5$ s. A nonlogarithmic decay of the magnetization $M$ with time was observed. This deviation from the conventional logarithmic time dependence was not an approach of the system to the equilibrium state. The decay fits extremely well to the relation $M(t)=M_0/[1+(\mu kT/U_0)\times\ln(t/t_{\rm eff})]^{1/\mu}$ from vortex-glass theory or to the empirical expression $M(t)=M_0+a(T)\ln[\ln(t/t_{\rm eff})]$. Another possible explanation is based on the theory of thermally activated flux creep, wherein the magnitude of the pinning-energy barrier $U_0$ is a function of current density. From the latter we obtain an exponential dependence of $U_0$ on current density for $J$ near its critical value.

Among the most fascinating and widely studied features of high-temperature superconductors (HTSC) are their magnetic properties, both "static" and time dependent. In the vortex state, a rapid relaxation of the irreversible magnetization is generally observed,[1-3] with many researchers[4-7] reporting a logarithmic time decay of magnetization $M$, with $M(t)\sim\ln(t)$. In recent studies, however, we have found significant departures from a precise logarithmic dependence, under conditions that rule out any explanation based on a final approach to the equilibrium magnetization. The current study was performed using a well-characterized single crystal of $YBa_2Cu_3O_7$ that was irradiated with protons, creating many flux-pinning defects and a high critical current density $J_c$. The use of a single crystal with $\mathbf{H}\|c$ axis eliminated questions of crystalline anisotropy that arise with polycrystalline samples and we made certain that the sample was fully in the critical state during the measurements. These are studies of long-term magnetic relaxation in the archetypical HTSC, $YBa_2Cu_3O_7$. Our results are well described by an "interpolation formula" from vortex-glass theory,[8] which has recently provided an explanation[9] for the widely observed plateau in the flux-creep rate $d[\ln(M)]/d[\ln(t)]$ as a function of temperature $T$. The present results further support the vortex-glass model of HTSC's, although some questions remain. An alternate description of comparable accuracy is provided by a suitably generalized, conventional flux-creep theory.

A 98-$\mu$g $YBa_2Cu_3O_7$ single crystal of $1.15\times0.226\times0.045$ mm$^3$ was used in the experiments. Details of the sample preparation have been reported previously.[10] The sample, initially with $T_c=92.5$ K, transition width $\delta T_c<0.5$ K, and *relatively* reversible magnetization, was irradiated with 3-MeV protons[11] to a fluence of $2\times10^{16}$ ions/cm$^2$. The resulting defect density corresponds with a broad maximum in $J_c$ versus proton fluence, with $T_c$ reduced to 91 K. Flux-creep measurements were made in a superconducting quantum interference device (SQUID) -based magnetometer (Quantum Design, Inc.) with controls to prevent any overshoot of the applied field. The sample was cooled to the desired temperature in zero field and then field $H$ was applied parallel to the crystalline $c$ axis. Since $J_c$ was very large ($>10^7$ A/cm$^2$ at 5 K), we insured that the entire sample was in the critical state (field penetration to the center of the sample) by cycling around the hysteresis loop first to $-5$ T, then increasing $H$ to the measurement field of $+1$ T.

The magnetization $M(t)$ was observed for times up to $3.5\times10^5$ s ($\sim$4 days) over a wide range of temperatures. Figure 1, a plot of $M(t)$ vs $\ln(t)$, illustrates the results for $H=1$ T at 30 K. It is apparent that for a sufficiently short observation interval, the dependence appears logarithmic in time. However, the creep-rate slope $dM/d[\ln(t)]$ decreases significantly with time as $M$ and $J_c$ decrease, from 58 G in the first hour of measurement to 41 G after $\sim(2-3)\times10^5$ s. The important features are that the curvature begins very early, continues throughout the entire measurement, and occurs at all temperatures. This study complements the work of Xu *et al.*,[12] who pointed out the experimental complication of nonlinearities at higher temperatures and brought renewed attention to the question of how the effective pinning potential depends on current density. The nonlogarithmic time dependence of $M(t)$ is more apparent in our study, however, where the observation times were up to 20 times longer.

According to the standard Anderson-Kim theory,[13,14] the evolution of magnetization can be described by the equation $dM/dt\propto e^{-U_0/kT}\sinh(J/J')$. Here, $U_0$ is the

 

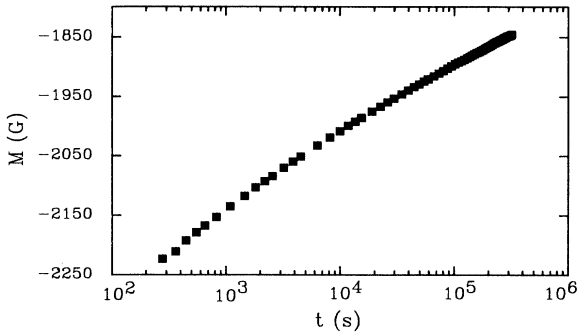


FIG. 1. Semilogarithmic plot of the magnetization $M$ vs time for single-crystal $YBa_2Cu_3O_7$ at $T=30$ K, with applied field $\mathbf{H}\|c$ and $H=1$ T. Curvature is apparent everywhere. For consistency, the same data are analyzed in the following figures, but results at other temperatures were very similar.

height of the pinning-energy barrier and $J'$, which has dimensions of current density, is a function of temperature $T$ and flux density $B$. The solutions of this equation are

$$M(t)=\begin{cases} M_0+(kT/U_0)\ln(t/t_0) \text{ for } J\gg J' \text{ or } t\ll t^*, & \text{(1a)} \\ M_{eq}+M'e^{-t/t^*} \text{ for } J\ll J' \text{ or } t>t^*, & \text{(1b)} \end{cases}$$

where the microscopic oscillation period $t_0$ is conventionally $\sim 10^{-10}$-$10^{-12}$ s. In our experiments, the largest relative change of $M$ was less than 25% (at $T=60$ K). Since $M\propto J$, the relative change in $J/J'$ of no more than 25% indicates that our observation times are too short to observe a crossover from Eq. (1a) to Eq. (1b). It is interesting, however, that a fit to the exponential form Eq. (1b) gives the impression of a good fit. For example, Fig. 2, a plot of $\ln(M/M_{eq}-1)$ vs $t$, appears to describe the data well. The problem is that this analysis yields unphysical values for the equilibrium magnetization $M_{eq}$, e.g., $-1800$ and $-500$ G at 30 and 60 K, respectively. In contrast, hysteresis measurements of $M$ show that $M_{eq}$ ranges from a few tens of gauss at 30 K to a few gauss at 60 K, *much* smaller than the irreversible magnetization. The true equilibrium magnetization is orders of magnitude smaller than the fitted values, and so some other mechanism is operative in these studies. Alternate

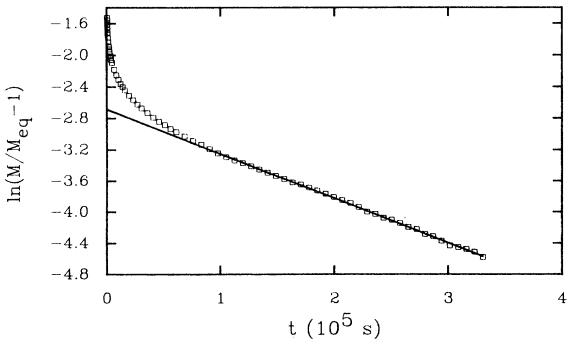


FIG. 2. Exponential decay: a plot of $\ln(M/M_{eq}-1)$ vs $t$. The deduced equilibrium magnetization $M_{eq}\approx -1800$ G is unphysically large.

forms[15] for the time dependence of $M$, including a "stretched exponential" $\sim\exp[-(t/\tau)^{\alpha}]$ and power law $\sim t^{\beta}$, also gave unsatisfactory results.

A more tanable explanation is the vortex-glass model of Fisher,[8] who argued that the single barrier height assumed by Anderson and Kim[13,14] is not appropriate in these high-$T_c$ materials. Instead, the current decay obeys an interpolation formula

$$M(t)=M_0/[1+(\mu kT/U_0)\ln(t/t_{\text{eff}})]^{1/\mu}, \quad (2)$$

where $\mu\leq 1$ is a temperature-independent exponent, $J_{c0}$ is the fluctuationless critical current density corresponding to $M_0$, and $t_{\text{eff}}$ is a macroscopic, effective time scale. This relation comes from a vortex-pinning potential that depends on the instantaneous current density $J$ as $U(J)=U_0[(J_0/J)^{\mu}-1]$. The vortex-glass model is based on an assumption that there exists a truly superconducting ordered phase at low temperature.

Our experimental data are described very well by Eq. (2) which we write as an inverse power relation

$$M(t)^{-\mu}=M_0^{-\mu}+c(T)\ln(t), \quad (3)$$

where $\mu$, $M_0$, and $c(T)$ are treated as fitting parameters. An example is shown in Fig. 3, a plot of $M^{-\mu}$ at 30 K as a function of $\ln(t)$. From least-squares fits to this expression, we obtain values for $\mu$ ($H=1$ T) at various temperatures, which are shown in the inset of Fig. 3. Some values lie near the theoretical bound $\mu\leq 1$; others, however, significantly exceed this bound, e.g., near 30 K. A further problem is that $\mu$ appers to decrease almost linearly with temperature above 30 K and tends toward zero near $T_c$. Within the vortex-glass framework, we have no explanation for either the temperature dependence of $\mu$ or for the values $>1$.

In an alternate formalism based on collective pinning,

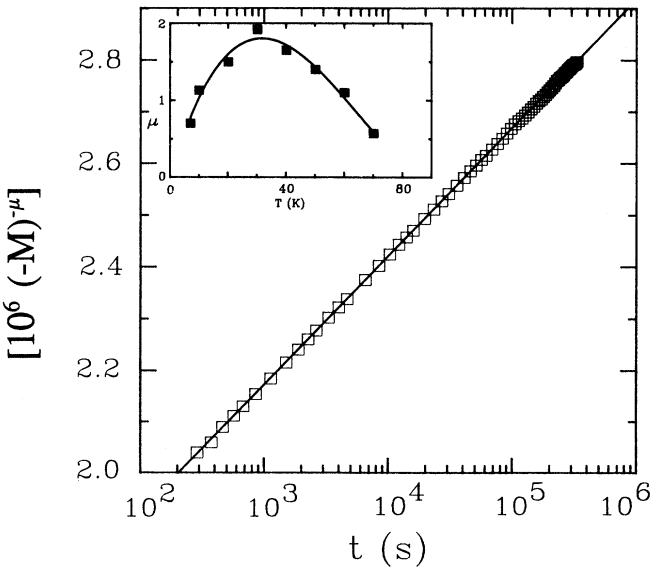


FIG. 3. Interpolation-formula analysis: semilogarithmic plot of $[-M(t)]^{-\mu}$ vs $t$, with $\mu$ as a fitting parameter. Inset: exponent $\mu$ vs $T$, for $\mathbf{H}\|c$ axis and $H=1$ T.

Feigel'man and co-workers[16,17] obtain similar expressions for $M(t)$ where, however, the exponent $\mu$ depends on $B$ and $T$. For the three-dimensional case in a weak field and low-temperature limit, they obtain $\mu=0.15$. For intermediate fields where collective creep is dominated by small flux bundles (size much less than the London penetration depth $\lambda_L$), then $\mu=\frac{3}{2}$. This is its maximum value and $\mu$ can be expected to decrease for still higher fields and temperatures, with the exact values depending on the dimensionality and relative scale of the system. Qualitatively, this is very similar to the dependence shown in the inset to Fig. 3. Furthermore, the maximum at 30 K lies roughly near the theoretical maximum value of $\frac{3}{2}$. Additional studies are underway to delineate more fully the field dependence of the nonlogarithmic relaxation. Preliminary results indicate that $\mu$ increases as $H$ increases, passes through a maximum, and decreases at still higher fields. This behavior is very similar to that predicted theoretically.[16,17]

Another perspective on the nonlogarithmic time dependence of magnetization can be obtained from the refined Anderson-Kim theory by Beasley, Labusch, and Webb (BLW).[18] From the BLW equation, $\partial\phi/\partial t=-2\pi R\times D(R,t)$, we have

$$\partial M/\partial t=D(R,t)/2\pi R\,, \tag{4}$$

where $\phi$ is the flux through the sample and $R$ is its radius. Here, $D$ is the flux-flow density which has the functional form[18]

$$D(R,t)=-(\nabla B/|\nabla B|)B\omega v_0\exp[-U(B,|\nabla B|)/k_BT]\,, \tag{5}$$

where $\omega$ is the average distance by which a flux bundle moves in a thermally activated jump; $v_0\propto 1/t_0$ conventionally lies[19,20] in the range $10^9$–$10^{12}$ $s^{-1}$; $U=U_0-|F|VX$, where $F$ is the Lorentz driving force and $X$ is the pinning length. This assumes that the driving force is large, as in this case.

The equation for $D$ is[18]

$$\frac{\partial D}{\partial t}=\pm\frac{D}{kT}\left(\frac{\partial U}{\partial|\nabla B|}\right)\times\left[\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial(rD)}{\partial r}\right)-\left(\frac{\partial(rD)}{r\partial r}\right)\left(\frac{\partial\ln|\nabla B|}{\partial r}\right)\right].$$

In conventional superconductors, $k_BT\ll U_0$. Thus, the change of $|\nabla B|$ ($|\nabla B|\propto J$) is so small during the magnetic relaxation measurement that it is a good approximation to neglect the time dependence of $\partial U/\partial|\nabla B|$ and $\partial\ln|\nabla B|/\partial r$. In that limit, the solution yields $D\propto 1/t$, which leads to a logarithmic evolution of the magnetic relaxation. However, the shielding current in HTSC materials changes quickly in the first few hours. Hence, the time dependence of $J$ is not negligible and we should expect to see a nonlogarithmic decay of $M$, which can provide information on the dependence of $U_0$ on $J$.

We have found an empirical "double logarithmic" relation which describes the data extremely well:

$$M(t)=M_0+a(T)\ln[\ln(t/t_{\rm eff})]\,, \tag{6}$$

where $M_0$, $a(T)$, and $t_{\rm eff}$ are fitting parameters. Figure 4 shows this expression fitted to the experimental data. In general, the "goodness-of-fit" parameters $R^2$ exceed 0.999 96 in the linear regressions, nearly identical to those obtained with the interpolation formula, Eq. (3). Our determinations of $t_{\rm eff}$ lie near $10^{-3}$ s for data at temperatures of 20 K and higher. These values are orders of magnitude larger than the microscopic oscillation period $t_0$, as has been recently cited and calculated.[17]

If we assume that the deviation from logarithmic decay is totally due to the dependence of $U_0$ on $J$, we can get the functional form of $U$ rather straightforwardly from Eqs. (4)–(6) as follows:

$$U/kT=[\ln(B\omega v_0/2\pi R)+\ln(t_{\rm eff})-\ln(a)+J_0/J_T]+\exp[-(J-J_0)/J_T]-J/J_T\,.$$

Here, $J_T=a/\Lambda$ ($\Lambda=R/30$) is a factor which relates $M$ and $J$ by $M=-\Lambda J$, with $J$ in units of ($A/cm^2$) and $M$ in gauss. Assuming $M_0\approx\Lambda J_0$ and considering that $U(J_0)\sim 0$, we have

$$-\ln(t_{\rm eff})=1+\ln(B\omega v_0/2\pi R)-\ln(a)\,.$$

Then, we have

$$U/kT=-1+[(J_0-J)/J_T]+\exp[-(J-J_0)/J_T]\,. \tag{7}$$

Over the entire temperature of 7–70 K, we find that the ratio $J_0/J_T$ is 4–5 and almost constant.

Previous work has sought to determine the current-density dependence of $U_0$. For example, Eq. (7) differs from the result of Zeldov *et al.*,[21] who deduced from transport measurements that $U(T,B,J)=U_J(T,B)\times\ln(J_0/J)$. However, our experiments show when $J$ is near to $J_0$, $U$ is exponentially dependent on $J$. Using the value of $M_0$ extrapolated from Eq. (6), $J_0$ should be near $8.6\times10^6$ $A/cm^2$ at $T=30$ K. The difference in dependencies deduced for $U(J)$ may arise from a difference in experimental regime. In the present study, the magnetic-field history insured that the sample was always close to the critical state; on the other hand, the relative current density $J$ was typically smaller in transport measurements.[21] This distinction does not apply, however, to the magnetic-flux-creep study of Maley *et al.*[22] on aligned powders of $YBa_2Cu_3O_7$, who deduced for $U(J)$ a depen-

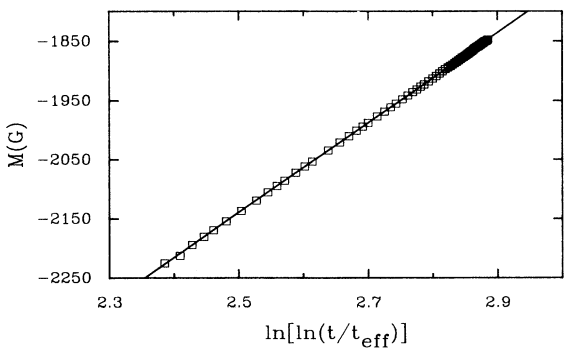


FIG. 4. $M$ vs $\ln[\ln(t/t_{\rm eff})]$, where $t_{\rm eff}$ is an adjustable parameter. Solid line shows a regression to Eq. (6) of text.

dence similar to that of Zeldov. A corresponding analysis of our experimental data gave results similar in appearance and magnitude,[22] but only for $T \leq 15$ K. Given the observed temperature dependence of $\mu$, it is not surprising that this analysis works over a limited temperature range only. In flux-creep annealing studies similar to those of Maley *et al.*, we obtain a *consistent* dependence of $U$ on $J$ that was not apparent in the former work. These results will be reported elsewhere.

Both the interpolation formula from vortex-glass theory and the empirical double-logarithm expression describe the experimental results for single-crystal $YBa_2Cu_3O_7$ quite well. Very recently, Svedlindh *et al.*[23] drew similar conclusions from their study of $Bi_{2.2}Sr_{1.7}Ca_1Cu_2O_8$ materials. Further work is underway to test the applicability of these expressions to other nonequilibrium properties, such as the scaling behavior of the flux pinning.[24]

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[1]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
[2]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
[3]M. Tuominen, A. M. Goldman, and M. L. Mecartney, Phys. Rev. B **37**, 548 (1988).
[4]A. C. Mota *et al.*, Phys. Rev. B **36**, 4011 (1987).
[5]Y. Yeshurun, A. P. Malozemoff, and F. Holtzberg, J. Appl. Phys. **64**, 5797 (1988).
[6]E. L. Venturini *et al.*, Appl. Phys. Lett. **56**, 2456 (1990).
[7]Shigemi Kohiki *et al.*, Appl. Phys. Lett. **56**, 298 (1990).
[8]M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989).
[9]A. P. Malozemoff and M. P. A. Fisher, Phys. Rev. B **42**, 6784 (1990).
[10]D. L. Kaiser, F. Holtzberg, M. F. Chisholm, and T. K. Worthington, J. Cryst. Growth **85**, 593 (1987).
[11]L. Civale *et al.*, Phys. Rev. Lett. **65**, 1164 (1990).
[12]Youwen Xu *et al.*, Phys. Rev. B **40**, 10882 (1989).
[13]P. W. Anderson, Phys. Rev. Lett. **9**, 309 (1962).
[14]P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).
[15]A. P. Malozemoff, in *Physical Properties of High Temperature Superconductors* I, edited by D. M. Ginsberg (World Scientific, Singapore, 1989), p. 71ff.
[16]M. V. Feigel'man *et al.*, Phys. Rev. Lett. **63**, 2303 (1989).
[17]M. V. Feigel'man, V. B. Geshkenbein, and V. M. Vinokur, Phys. Rev. B **43**, 6263 (1991).
[18]M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. B **181**, 682 (1969).
[19]P. H. Kes and J. van den Berg, in *Studies of High Temperature Superconductors,* edited by A. V. Narlikar (Nova Science, New York, in press), Vol. 5.
[20]A. P. Malozemoff *et al.*, *Strong Correlations and Superconductivity,* edited by H. Fukuyama, S. Maekawa, and A. P. Malozemoff (Springer, Heidelberg, 1989), p. 349.
[21]E. Zeldov *et al.*, Appl. Phys. Lett. **56**, 680 (1990).
[22]M. P. Maley *et al.*, Phys. Rev. B **42**, 2639 (1990).
[23]P. Svedlindh, C. Rossel, K. Niskanen, P. Nordling, P. Nordblad, L. Lundgren, and G. V. Chandrasekhar (unpublished).
[24]L. Civale, J. R. Thompson, D. K. Christen, and M. McElfresh (unpublished).