Field dependence of magnetization and magnetic relaxation in $(Bi,Pb)_2Sr_2CaCu_2O_8$ crystals

Lu Zhang, J. Z. Liu, M. D. Lan, P. Klavins, and R. N. Shelton
Department of Physics, University of California, Davis, California 95616
(Received 15 April 1991)

The hysteresis loops and magnetic relaxation in $(Bi,Pb)_2Sr_2CaCu_2O_8$ crystals were measured at fields up to 5.5 T. The relaxation rate $dM/d\ \ln t$ exhibits a strong field dependence in the temperature range 6–20 K. By expanding Bean's model of the critical current density to include the field dependence of the flux-creep effect, we developed expressions for both M and $dM/d\ \ln t$ as a function of the field. These expressions were used to fit our experimental data of field-dependent magnetization and relaxation rate.

INTRODUCTION

Giant magnetic relaxation has been observed in the high-T_c superconductor since 1987.[1] This phenomenon has generated much interest in the nature of the mixed state including magnetic irreversibility, vortex-lattice behavior, flux pinning, and transport properties. Simultaneous studies of the magnetic hysteresis loop and magnetic relaxation on single crystals may provide an important approach to understand the physical behavior of the critical state, flux creep, and thermally activated flux motion. These studies can also yield basic physical parameters, such as the lower critical field H_{c1}, the thermal-activated energy barrier U_0, and the critical current density J_{c0}. Magnetic relaxation experiments of strongly layered superconductors such as $Bi_2Sr_2CaCu_2O_8$ and $Tl_2Ba_2CaCu_2O_8$ crystals have been performed by several research groups. Yeshurun *et al.*[2,3] measured the magnetic relaxation on both $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ crystals and presented a flux-creep model in which they assumed that the critical current density J_c has a power-law dependence on the applied magnetic field, H^{-n}. They successfully described their field-dependent magnetic relaxation measurements in $YBa_2Cu_3O_7$ with $n=1$.[4] However, the relaxation data for $Bi_2Sr_2CaCu_2O_8$ were not explained by their model for $n>0$.[3] Shi *et al.*[5] used a different approach to analyze their experimental results of the relaxation rate, assuming the critical current density to have a linear field dependence in the form $1-H/H_0$. These authors adequately interpreted the fall of the relaxation rate when $H>H^*$, but failed to explain the saturation of the relaxation rate at high field. In this work we give a detailed procedure for the preparation of high quality $(Bi,Pb)_2Sr_2CaCu_2O_8$ crystals with transition temperatures of 92–96 K. Experimental data of hysteresis loop and magnetic relaxation on these crystals are also presented. Finally, the field dependence of both the magnetization and magnetic relaxation rate are interpreted with an expanded flux-creep model.

SAMPLE PREPARATION

The superconducting crystals $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ were grown by a flux method.[6] A nonstochiometric mixture of Bi_2O_3, Sr_2O_3, $CaCO_3$, CuO, and PbO with atomic ratios of [Bi]:[Pb]:[Sr]:[Ca]:[Cu]=1.7:0.3:2:2:3 were used in the preparation of $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystals. The mixture was placed in a 50-cm^3 Pt crucible, heated up to 1050 °C at a rate of 50°C/h, held for 1 h at this temperature, and slowly cooled at a rate of 1–2 °C/h to 800 °C, followed by furnace cooling to room temperature. Thin, micalike crystals with a dark color and well-developed faces were obtained. The crystals had dimensions 1–3 mm in the plane of the sheet and were approximately 50–100 μm thick. Compared with the other high-T_c superconducting crystals such as $YBa_2Cu_3O_{7-\delta}$ and $Nd_{2-x}Ce_xCuO_{4-\delta}$, the surface morphology of the crystals is very different, with $Bi_2Sr_2CaCu_2O_{8-\delta}$ having typically cleaved faces, and layered growth features are observed.

The superconducting transition temperatures (T_c's) were determined by dc susceptibility measurement on a commercial superconducting quantum interference device (SQUID) magnetometer.[7] Figure 1 shows the Meissner (FC), shielding (ZFC), and flux trapping (REM) curves for a single crystal of $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ in the temperature range of 10–110 K. The applied field was parallel to the c axis. The experimental curves indicate bulk superconductivity in the crystal with a supercon-

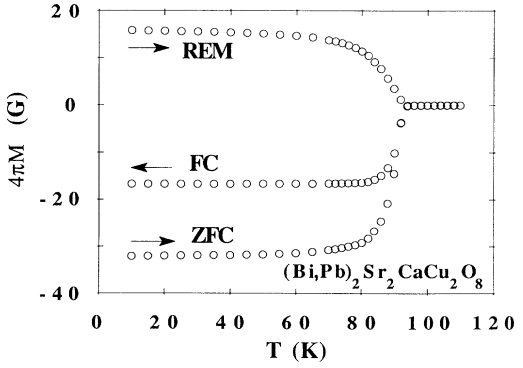

FIG. 1. Shielding (ZFC), Meissner (FC), and flux trapping (REM) measurements in a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal with $H\|c$ and $H_a=3$ Oe.

ducting transition temperature onset of about 96 K and a 10%–90% transition width of about 4 K. The Meissner fraction was 52%.

EXPERIMENTAL METHODS AND RESULTS

In order to study the magnetic flux creep on $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$, hysteresis loop and time relaxation measurements on two crystals were made on the SQUID magnetometer. Because of the large anisotropy present in this material (discussed elsewhere[8]), the direction of the c axis of the crystal must be aligned very carefully with respect to the applied field. In addition, the experimental data for $\mathbf{H}\|c$ had to be precisely corrected by taking into consideration the demagnetization effect. For example, for a large demagnetization factor (D between 0.9 and 0.95), a variation of 5% can cause a 50% change in the magnetization ($4\pi M$).

During magnetic hysteresis measurements, there was a 5-min waiting time after setting a new field, so that the magnetization inside the sample could reach a relatively stable state. The scan length of the specimen for the measurement of magnetization was 4 cm. Figure 2 presents two hysteresis loops of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal at 6 and 10 K for the applied field parallel to the c axis. The initial slope of $M(H)$ ($H<H_{c1}$) corresponds to almost 100% shielding and a demagnetization factor between 0.93 and 0.94. The value H^* corresponds to a maximum in the magnitude of the magnetization and shifts from 0.6 T at 6 K to 0.15 T at 15 K. At higher temperature the size of the hysteresis loop shrinks rapidly, which demonstrates that there is very weak flux pinning in this material.

The magnetic relaxation experiments were performed at temperatures between 6 and 30 K and in external magnetic fields in the range of 5×10^{-3} to 5 T. Before the start of all relaxation runs, the sample was first warmed to a temperature well above T_c and then zero-field cooled (ZFC) to the desired temperature. Brief M-vs-H measurements for $H<H_{c1}$ were always made to determined the demagnetization factor. A magnetic field was then applied, and the magnetization at fixed temperatures and fields was measured as a function of time over a period of 1.5 h. Figure 3 shows the M-vs-t curves for $T=10$ K and $H<H^*$ using a logarithmic time scale. The magnetization at each temperature and field is clearly linear on this type of plot. The value of H^* was determined to be 0.3 T, which is the same as obtained from the hysteresis loop data. The slopes of each curve gave the relaxation rates which correspond to each point in Fig. 4. The logarithmic flux relaxation rates at 8, 10, and 12 K for a range of applied fields 5×10^{-3} to 4 T are shown in Fig. 4. The relaxation rates initially increase with increasing field. Above a field H^*, a gradual decrease of the relaxation rate sets in, followed by saturation at very high fields. All the data exhibit the same characteristic rise and fall for the magnetic relaxation rates as the applied field increases. The relaxation rates (S) change significantly from 394.7 emu/cm^3 at 8 K to 63.5 emu/cm^3 at 12 K for $H_a=0.2$ T. The strong temperature dependence of the relaxation rates reflects that giant flux creep is induced by thermally activated flux motion in the $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ system.

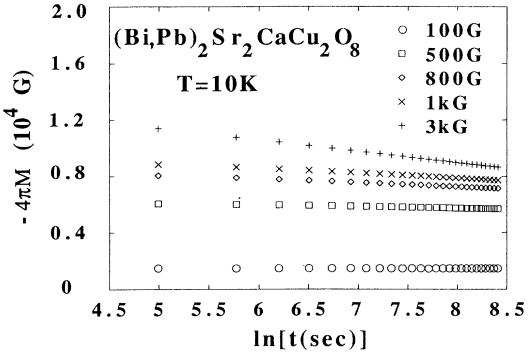

FIG. 3. Magnetization (ZFC) vs time at 10 K for fields parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal.

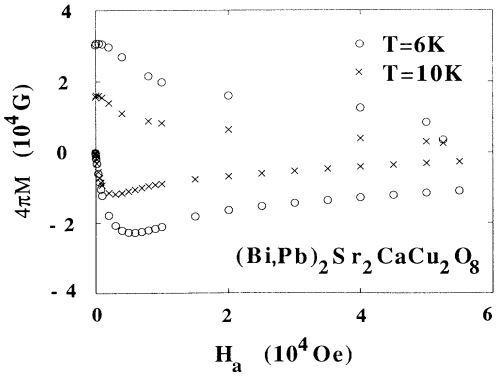

FIG. 2. Magnetic hysteresis loops for fields parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal.

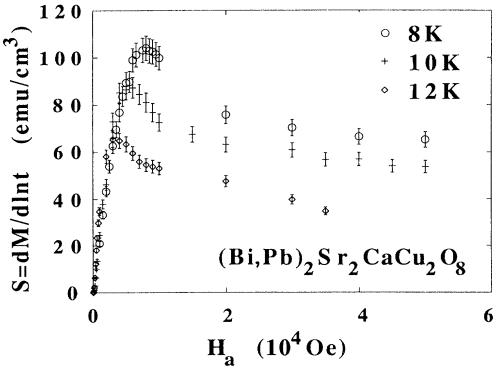

FIG. 4. Magnetic relaxation rate (ZFC) vs applied field for fields parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal.

DISCUSSION

The theory of thermally activated flux creep was first proposed by Anderson,[9] and lately, by Beasley, Labusch, and Webb[10] and Campbell and Evetts,[11] which were based on Bean's critical-state model.[12] In the critical-state model the application of a magnetic field $H > H_{c1}$ to a superconductor leads to a gradient in the density of flux lines. At zero temperature these flux lines can move only if the driving force $\mathbf{F}$ is larger than the local pinning force $\mathbf{F}_p$. The critical state can be reached whenever the two forces are balanced. In the critical state,

$$\nabla \times \mathbf{B} = \frac{4\pi}{c} \mathbf{J}_c(\mathbf{B}) , \tag{1}$$

where $\mathbf{B}$ is local field and $\mathbf{J}_c$ is the critical current density.

Bean assumed the critical current density J_c to be independent of the local field B. For a slab of thickness d in a field parallel to its surface, the magnetization $4\pi M$ was derived by Bean in practical units (J in A/cm^2, M and H in G, lengths in cm):

$$4\pi M = \begin{cases} \dfrac{H^2}{2H^*} - H, & H < H^* \quad (2a) \\ -\dfrac{H^*}{2}, & H > H^*, \quad (2b) \end{cases}$$

where $H^* = (\pi/5) J_c d$ is the field when the currents start to flow through the entire volume of the sample. The average magnetic-flux density $\langle B \rangle$ displays quadratic field dependence at low field ($H < H^*$). At high field ($H > H^*$) the magnetization $4\pi M$ is constant.

At finite temperature thermal energy may allow flux lines to jump from one pinning region to another. In the absence of any flux-density gradient, the jumps have the same probability of occurring in one direction as in the other; therefore, no net creep occurs. By introducing an external field, the thermal-activated flux lines jump more rapidly in the direction of the decreasing flux density. The resulting flux creep leads to a slow change in the trapped field as a function of time, and this time dependence is logarithmic in experiments on the oxide superconductors. Moreover, this creep can be observed only when the flux-density gradient is very near to the critical state.[13] According to Anderson's flux-creep theory,[9] the thermal activation induces the flux lines to move in bundles and jump over the pinning barrier at a rate governed by an Arrhenius expression: $\exp(-U/k_B T)$, where U is the effective thermal activation energy and k_B is the Boltzmann constant:

$$U = U_0 - |F| V X_0 = U_0 - 2X_0 V \frac{c}{4\pi} B \left| \frac{\partial B}{\partial x} \right| . \tag{3}$$

In Eq. (3), U_0 is the effective height of the energy barrier for the thermally activated motion of a flux bundle, V is the flux bundle volume, X_0 is the pinning length or the effective width of the energy barrier, and F is the driving force on the flux lines. The flux-creep equation in the $k_B T/U_0 << 1$ limiting case is then given by[14]

$$\frac{\partial B}{\partial t} = -X_0 w_0 e^{-U_0/k_B T} \frac{\partial}{\partial x} \times \left[\left[B - X_0 \left| \frac{\partial B}{\partial x} \right| \right] \exp\left[c_1 B \left| \frac{\partial B}{\partial x} \right| \right] \right] , \tag{4}$$

where $c_1 = (c/4\pi) 2X_0 V/k_B T$. Campbell and Evetts[11] obtained a solution for the above equation in the long-time approximation as

$$J_c = J_{c0} \left[1 - \frac{k_B T}{U_0} \ln\left[\frac{t}{t_0} \right] \right] , \tag{5}$$

where J_{c0} is the critical current density in the absence of thermally activated flux creep and $1/t_0$ is a characteristic attempt frequency for flux hopping over pinning barriers. The current decays logarithmically with time, and the value of $k_B T/U_0$ can be obtained from the rate of decay.

Substituting Eq. (5) into Eqs. (2a) and (2b), we obtain the relaxation rate $d4\pi M/d \ln t$ for both $H < H^*$ and $H > H^*$:

$$\frac{d4\pi M}{d \ln t} = \begin{cases} \dfrac{H^2}{2H_0} \dfrac{k_B T}{U_0}, & H < H^* \quad (6a) \\ \dfrac{H_0}{2} \dfrac{k_B T}{U_0}, & H > H^* \quad (6b) \end{cases}$$

where $H_0 = (\pi/5) J_{c0} d$ is H^* at zero temperature.

Figures 5 and 6 show both the $4\pi M$ and $d4\pi M/d \ln t$ of zero-field-cooled magnetization as a function of field (corrected for demagnetization) for the field parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal. The fit H^* is about 0.2 T at 6 K, which is smaller than the maximum magnitude of the demagnetization in the experiment. The experimental points at $H < H^*_{\text{fit}}$ are fit well by Eq. (2a). At high fields $H > H^*_{\text{fit}}$, however, the experimental data present a prominent field dependence for $4\pi M$ and

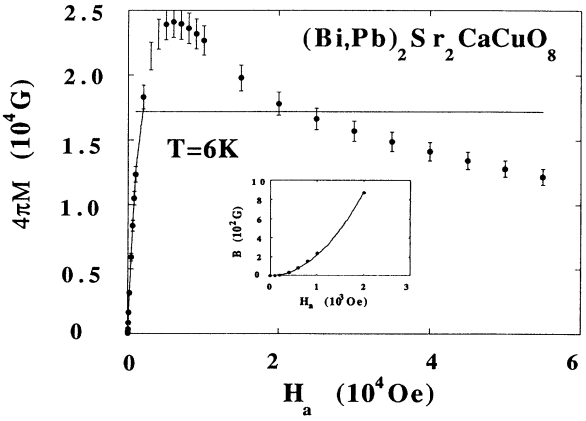

FIG. 5. Partial magnetic hysteresis loop for field parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal. Inset: magnetic flux density vs field for $H < H^*$. Solid line is a fit with Eqs. (2a) and (2b).

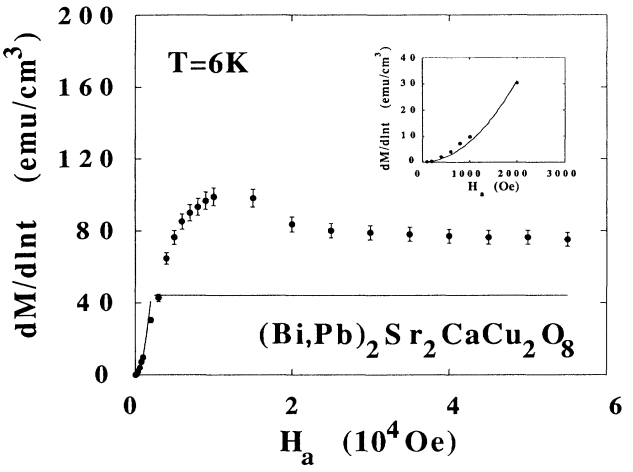

FIG. 6. Magnetic relaxation rate vs field for fields parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal. Inset: relaxation rate for $H<H^*$. Solid line is a fit with Eqs. (6a) and (6b).

$d4\pi M/d\ln t$, which cannot be predicted by Eqs. (2b) and (6b). The substantial agreement with theory when $H<H^*$ implies the assumption of J_c independent of B in Bean's critical current model may be valid only for low fields. The effective energy barrier U_0 can be estimated to be on the order of 0.01 eV.

Using Campbell's flux-creep result [Eq. (5)], Yeshurun *et al.*[4] modified Bean's critical-current-density assumption to include the field dependence:

$$J_c=J_{c1}\left[\frac{H_{c1}}{B}\right]^n, \tag{7}$$

$$H+4\pi M=\frac{1}{(n+2)H_{c1}^n H_0}(H^{n+2}-H_{c1}^{n+2}),\quad H<H^*, \tag{8a}$$

$$\frac{d4\pi M}{d\ln t}=\frac{1}{(n+2)H_{c1}^n H_0}\frac{k_BT}{U_0}(H^{n+2}-H_{c1}^{n+2}),\quad H<H^*, \tag{8b}$$

and

$$4\pi M=-\frac{H_0}{2}\left[\frac{H_{c1}}{H}\right]^n,\quad H>H^*, \tag{9a}$$

$$\frac{d4\pi M}{d\ln t}=\frac{H_0}{2}\frac{k_BT}{U_0}\left[\frac{H_{c1}}{H}\right]^n,\quad H>H^*, \tag{9b}$$

where H_{c1} is the lower critical field. For $n=0$ the above equations give Bean's original result [Eqs. (2) and (6)]. Yeshurun *et al.* were able to explain well their relaxation data in a $YBa_2Cu_3O_{7-\delta}$ crystal using $n=1$, where $dM/d\ln t$ increases as H^3 for both $\mathbf{H}\|c$ and $\mathbf{H}\perp c$.[4] Qualitatively, the fall of $dM/d\ln t$ with field can be predicted by Eq. (8b), with the assumption $n>0$. Yeshurun *et al.*[3] also measured the relaxation of $Bi_2Sr_2CaCu_2O_{8-\delta}$ crystals. They obtained $U_0=8\times10^{-3}$ eV by fitting the relaxation rate with their model and assuming $n=0$, rather than $n=1$, as they emphasized in the paper. This is actually Bean's solution and cannot predict the field-dependent magnetization and relaxation rate at $H>H^*$ as mentioned above.

To interpret the present data, we first extend the flux-creep theory of Campbell and Evetts to include a field dependence of the critical current density, $J_c(T,H,t)$. Equation (4) can be solved when the sample is in the critical state with large applied field ($H\gg H^*$), where we expect the condition $k_BT/U_0\ll1$ to hold. The result obtained is the lowest-order solution in an expansion in the powers of k_BT/U_0. Since the relaxation rate is one order higher in this parameter than that of the magnetization, we obtained the second-order solution in $dM/d\ln t$, which is sufficiently accurate for our purpose. The justification is to keep the only special derivatives of B in the exponential term and suggest the exponential term to be a separate function of space and time variables (the detailed derivation is similar to that of Tinkham[13] and Beasley, Labusch, and Webb[10]). The solution of Eq. (4) is given as

$$J_c=J_{c0}\left[1-\frac{k_BT}{U_0}\ln\left[\frac{t}{t_0}\right]\right]\left[1-\frac{k_BT}{U_0}\ln\left[\frac{H}{H_{c1}}\right]\right]. \tag{10}$$

The $\ln H$ term can be also found in the paper by Campbell and Evetts. Furthermore, we can obtain both field- and temperature-dependent magnetization $M(T,H,t)$ by using the critical-state formula [Eq. (1)]. The relaxation rate $dM/d\ln t$ can be easily found by the simple derivative of M with respect to logarithmical time. The expressions for $4\pi M$ and $d4\pi M/d\ln t$ are given as follows:

$$4\pi M=\frac{H_0}{2}\left[1-\frac{k_BT}{U_0}\ln t-\frac{k_BT}{U_0}\ln H\right], \tag{11}$$

$$\frac{d4\pi M}{d\ln t}=\frac{H_0}{2}\frac{k_BT}{U_0}\left[1-\frac{k_BT}{U_0}\ln H\right], \tag{12}$$

where we only keep the first order of k_BT/U_0 in $4\pi M$ and second order of k_BT/U_0 in $d4\pi M/d\ln t$. Figures 7

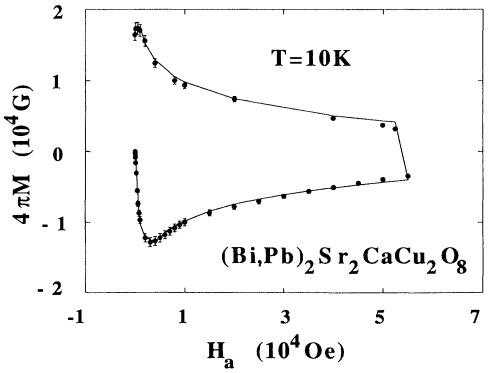

FIG. 7. Magnetic hysteresis loops for fields parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal at $H>H^*$. Solid line is a fit with Eq. (11).

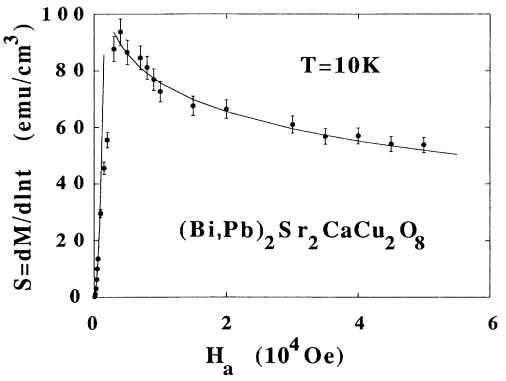

FIG. 8. Magnetic relaxation rate vs field for fields parallel to the c axis of a $(Bi,Pb)_2Sr_2CaCu_2O_{8-\delta}$ crystal at $H > H^*$. Solid line is a fit with Eq. (12).

and 8 show our experimental data and the fitting curves by using Eqs. (11) and (12) at $H > H^*$. The result of the fit yields a value for U_0 of about 0.01 eV, which is consistent with Bean's model for $H < H^*$.

CONCLUSION

We have measured the magnetic hysteresis loop and time relaxation on $(Bi,Pb)_2Sr_2CaCu_2O_8$ single crystals at various fields in low temperatures. We observed the large logarithmic magnetic relaxation, with the relaxation rate showing a peak as a function of the field. By consideration of the flux creep, we expanded Bean's model of the critical current density to include the field dependence and developed expressions of both M and $dM/d\ln t$. Our model agrees with the experimental data and gives an interpretation for the decrease of the amplitude of magnetization and relaxation rate over the entire range of the magnetic field.

ACKNOWLEDGMENTS

Research supported by the U.S. Department of Energy under Contract No. W-7405-ENG-48 to Lawrence Livermore National Laboratory and by the National Science Foundation under Grant No. DMR-90-21029.

[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]Y. Yeshurun, T. K. Worthington, R. M. Yandrofski, L. Krusin-Elbaum, F. H. Holtzberg, T. R. Dinger, and G. V. Chandrashekhar, Cryogenics **29**, 258 (1989).
[4]Y. Yeshurun, A. P. Malozemoff, F. H. Holtzberg, and T. R. Dinger, Phys. Rev. B **38**, 11 828 (1988).
[5]Donglu Shi, Ming Xu, A. Umezawa, and Ronald F. Fox, Phys. Rev. B **42**, 2062 (1990).
[6]J. Z. Liu, G. W. Crabtree, L. E. Rehn, Urs Geiser, D. A. Young, W. K. Kwok, P. M. Baldo, Jack M. Williams, and D. J. Lam, Phys. Lett. A **127**, 444 (1988).
[7]Quantum Design, Inc., San Diego, CA.
[8]L. Zhang, J. Z. Liu, M. D. Lan, P. Klavins, and R. N. Shelton, Bull. Am. Phys. Soc. **36**, 667 (1991).
[9]P. W. Anderson, Phys. Rev. Lett **9**, 309 (1962).
[10]M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682 (1969).
[11]A. M. Campbell and J. E. Evetts, Adv. Phys. **21**, 199 (1972).
[12]C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962).
[13]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).
[14]C. W. Hagen and R. Griessen, in *Studies of High Temperature Superconductors,* edited by A. V. Narliker (NOVA, Commack, NY, 1989), Vol. 3, pp. 159–195.