PHYSICAL REVIEW B VOLUME 40, NUMBER 7 1 SEPTEMBER 1989

Magnetic relaxation and critical current density limited by flux creep in $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$ ($T_c = 115$ K) and $YBa_2Cu_3O_{7-x}$ ($T_c = 92$ K)

H. S. Lessure, S. Simizu, and S. G. Sankar
Department of Metallurgical Engineering and Materials Science and Mellon Institute, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213

(Received 23 March 1989)

We have investigated the magnetization behavior of $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$ (Bi 2:2:2:3) and $YBa_2Cu_3O_{7-x}$ (Y-Ba-Cu-O) as a function of field, temperature, and time. We find a logarithmic time decay in both zero-field-cooled magnetization and remanent magnetization, consistent with the Anderson-Kim flux-creep model with activation energies of $U_0/k = 700$ K (0.06 eV) for Bi 2:2:2:3 and $U_0/k = 1700$ K (0.15 eV) for Y-Ba-Cu-O in an applied field of 1 kOe. Measured activation energies remain essentially temperature independent for T/T_c less than 0.6 while $J_c(T)$ decays rapidly as a result of the relatively small U_0.

The study of the magnetic behavior of superconductors has traditionally played a central role in understanding superconductivity. Such studies also have important implications for technical applications of superconductivity such as superconducting magnets. All high-T_c oxide superconductors show strong metastability well below T_c in their magnetic behavior. Although there remains a considerable degree of controversy concerning the role of "frustration" caused by weak incoherent coupling among superconducting components in the case of polycrystalline samples and single crystals with defects,[1-3] the highly metastable magnetic behavior of high-T_c oxide superconductors may be understood in terms of a classical critical-state theory and flux-creep model. Yeshurun and Malozemoff showed in their study of a single crystal of Y-Ba-Cu-O (Ref. 2) that the irreversible magnetization, slow decay of the zero-field-cooled magnetization M_{ZFC}, and H-T irreversibility line can all be explained by the Anderson-Kim flux-creep model.[4-6] They reported an anisotropic pinning energy of the order of 0.1 eV. Similar measurements are now being made on some other important oxide superconductors, namely, $Ba_{0.6}K_{0.4}BiO_3$,[7] $Tl_2Ca_2Ba_2Cu_3O_x$,[8] and $Bi_2Sr_{1.6}Ca_1Cu_2O_x$.[9] A basic understanding of the origin and nature of this magnetic decay has important implications upon critical current density and resistive dissipation in the mixed state.[10-12]

We have prepared polycrystalline samples of single-phase ($T_c = 115$ K) $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$.[13,14] To explore the underlying factors affecting J_c, we studied the characteristics of magnetic decay for our material as well as Y-Ba-Cu-O for comparison.

Samples of Y-Ba-Cu-O were prepared by solid-state reaction of pressed pellets of oxides and carbonates following conventional techniques.[15] Samples of the Pb-doped Bi 2:2:2:3 were prepared in a manner similar to Endo, Koyama, and Kawai[16] by reacting reagent grades of Bi_2O_3, $PbCO_3$, $SrCO_3$, and $CaCO_3$, and high-purity CuO in the ratio of 1.6:0.4:1.6:2.0:2.8, respectively. All samples studied were $1.5 \times 3 \times 8$ mm^3 bars having about 50% theoretical density.

Time-dependent magnetization measurements were made using a modified Princeton Applied Research vibrating-sample magnetometer with a custom-built power supply which could be ramped to ~1 kOe in approximately 10 sec. To acquire magnetic relaxation data, the sample was first heated above T_c. It was then cooled in zero field to the desired temperature. After thermal equilibration, the field was ramped up to its desired holding point to begin taking M_{ZFC} data. To measure remanent magnetization M_{rem} the field was ramped back to zero. The origin of time is taken to be the end of the 10-sec field ramp. The first data point was taken at $t = 10$ sec. To find the bulk current density from the magnetic measurements, one needs to subtract the effect of surface currents which give rise to the uniform diamagnetism M_{eq} in the equilibrium state. To estimate this effect, we measured the magnetization $M_{FC}(H,T)$ of the sample at the temperature T cooled slowly in a constant field H.

The M_{FC} versus temperature curves shown in Fig. 1 allow for the determination of $T_c = 115$ K in our Bi 2:2:2:3 sample. In a field of 20 G, we find a transition width of ~5 K which is similar to that found for the Tl 2:2:2:3 compound.[17] M_{FC} is shown in Fig. 2 as a function of field.

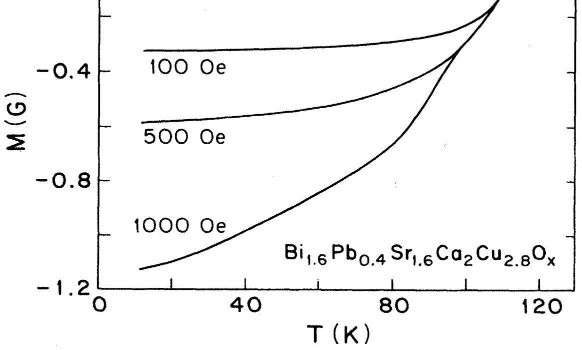

FIG. 1. Field-cooled magnetization M_{FC} of $Bi_{1.6}Pb_{0.4}Sr_{1.6}$-$Ca_2Cu_{2.8}O_x$ as a function of temperature.

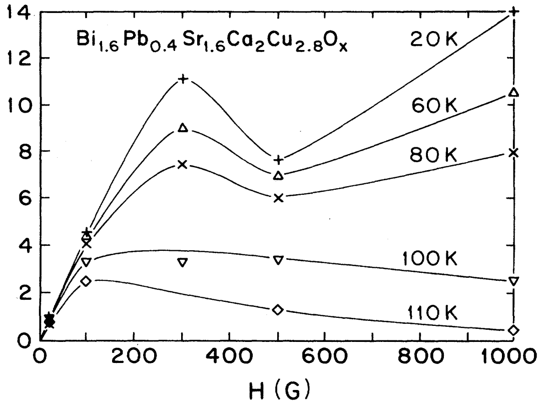

FIG. 2. M_{FC} as a function of applied field.

It clearly shows a strong deviation from equilibrium at lower temperatures. Therefore, the values derived from these curves for M_{eq} must be taken as overestimates at lower temperatures although this correction to M_{ZFC} is less than 30% for $T < 40$ K.

Figure 3 shows typical zero-field-cooled magnetic relaxation at various temperatures for the Pb-doped Bi 2:2:2:3 sample upon application of a 1-kOe field. The relaxation is found to be logarithmic in time after an initial steep nonlogarithmic decay. Similar results were obtained for M_{rem}. In the logarithmic region, magnetization is described by

$$M(t) = M_0 + S\ln(t) . \qquad (1)$$

Although alternative explanations such as a superconducting glass model[3] or wide distribution of activation energies[8] may be invoked for the logarithmic flux decay, we find the conventional flux-creep model to be satisfactory.

In the Anderson-Kim flux-creep model,[18,19] the thermally activated flux-creep rate may be conveniently expressed in terms of the magnetization. For a cylindrical sample of radius r carrying a current density $J_c(B,T)$ in the critical state, Beasley, Labusch, and Webb[20] found when $kT/U_0 \ll 1$:

$$S = dM/d\ln(t) = (J_c r/3c)(kT/U_0) , \qquad (2)$$

where U_0 is the flux pinning energy and c is the speed of light. A correction term which arises from the field dependence of U_0 and J_c may be neglected in our case. Since $4\pi M$ is significantly smaller than B, the variation of B in the material is not significant compared with the applied field.

The low-temperature dependence of S observed in Fig. 4 may be easily understood from the explicit temperature dependence in Eq. (2). The decrease in S at higher temperatures apparently reflects the rapid drop in J_c. Rather small absolute values of S in our experiments may be taken to be primarily the result of the small grain size in our polycrystalline specimens estimated to be $\sim 10\ \mu$m.

In order to relate the measured magnetization with critical current, one has for a cylinder of radius r, in the framework of the critical state model,[21]

$$-(M_0 - M_{eq}) = (J_c r/3c) . \qquad (3)$$

M_{eq} is subtracted from M_0 to account for the surface current.

Combining Eqs. (2) and (3) gives the following expression for S:

$$S = dM/d\ln(t) = -(M_0 - M_{eq})(kT/U_0) . \qquad (4)$$

Thus, using Eqs. (3) and (4), we may obtain the critical current density $J_c(H,T)$ and the pinning energy $U_0(H,T)$ from our measurements of $M_0(H,T)$, $M_{eq}(H,T)$, and $S(H,T)$.

In Fig. 5, the activation energies U_0 of Y-Ba-Cu-O and Pb-doped Bi 2:2:2:3 are plotted against temperature. The value obtained for our polycrystalline Y-Ba-Cu-O sample is very close to values reported in single-crystal measurements as discussed below. It is remarkable that these activation energies remain essentially temperature independent for T/T_c less than 0.6 for Bi 2:2:2:3 and 0.9 for Y-Ba-Cu-O while $J_c(T)$ decays rapidly (see Fig. 6 and Refs. 12, 22, and 23). The drop in U_0 in Bi 2:2:2:3 well below T_c may be related with flux lattice melting well below T_c

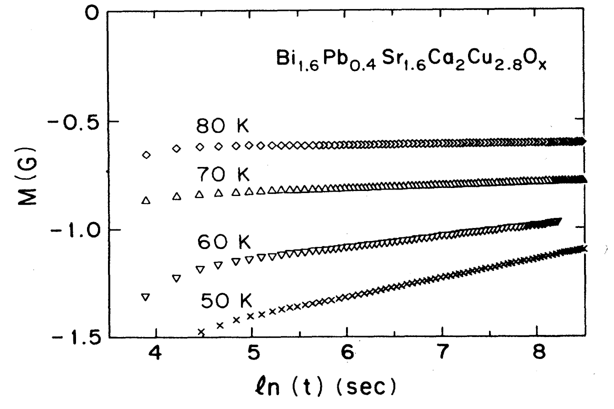

FIG. 3. Typical logarithmic decay of M_{ZFC} in $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$.

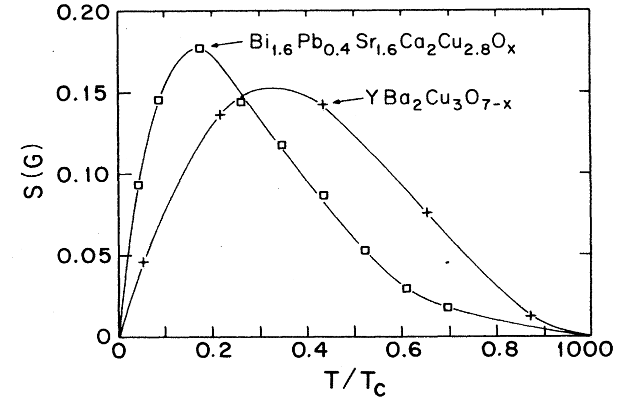

FIG. 4. Temperature dependence of relaxation parameter S for $YBa_2Cu_3O_{7-x}$ and $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$.

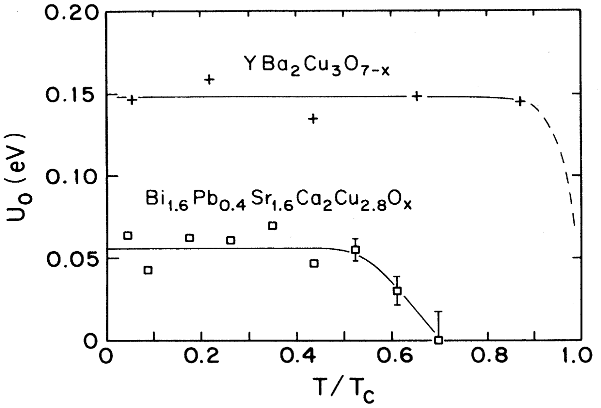

FIG. 5. Temperature dependence of the pinning energies U_0 for $YBa_2Cu_3O_{7-x}$ and $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$. Lines are shown to guide the eye.

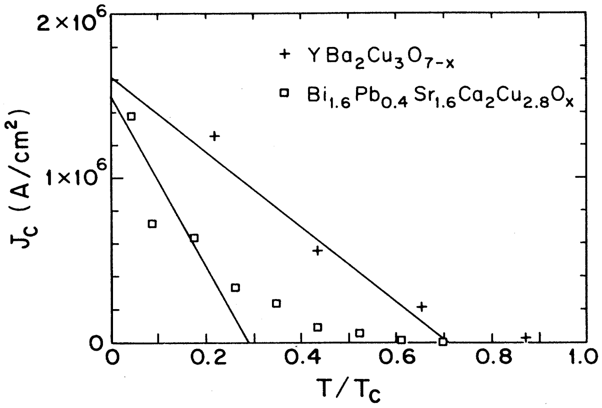

FIG. 6. Temperature dependence of the critical current density J_c for $YBa_2Cu_3O_{7-x}$ and $Bi_{1.6}Pb_{0.4}Sr_{1.6}Ca_2Cu_{2.8}O_x$. Lines represent the linear relationship given in Eq. (5).

found for the related $T_c = 85$ K superconductor $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_8$ (Bi 2:2:1:2).[24] Insofar as $U_0(T)$ reflects the barrier to flux motion, the $U_0(T)$ plots may be taken to be consistent with flux melting for $T > 0.9\ T_c$ in Y-Ba-Cu-O and suggest such behavior may be possible for $T < 0.7\ T_c$ in Bi 2:2:2:3.

The temperature dependence of the critical current density J_c is given by the following expression:[25]

$$J_c = J_{c0}[1-(kT/U_0)\ln(v_0/v)]\,, \tag{5}$$

where J_{c0} is the critical current density in the absence of thermal activation given by

$$J_{c0} = U_0/BVx\,. \tag{6}$$

In Eqs. (5) and (6), x is the effective size of the pinning site, V is the effective volume of the flux bundle which can creep, and v and v_0 are the creep velocities in the presence and absence of pinning, respectively. From the lower-temperature data in Fig. 6, one obtains $\ln(v_0/v) = 21$ for Bi, and 26 for Y-Ba-Cu-O using our values for U_0 obtained above. One might estimate this logarithmic term as follows. The creep velocity v can be given approximately by

$$v \approx (d/M_0)dM/dt = (kT/U_0)(d/t)\,, \tag{7}$$

where d is the distance that the flux travels leaving the sample. One may take d to be the grain size under the assumption that the grains are decoupled. Taking $kT/U_0 = 10^{-2}$ and $t = 10^3$ sec as the other representative values in our experiments yields $v = 10^{-8}$ cm/sec. If we take v_0 to be 1-10^6 cm/sec (Ref. 25) we have $\ln(v_0/v) = 18$-32. This is in surprisingly good agreement with the experimental data. Although the above estimation is crude, there do not seem to be any physically reasonable choices for v and v_0 leading to $\ln(v_0/v)$ outside this range.

From Eqs. (5) and (6), we note that increasing pinning site density (V^{-1}) is effective only to increase J_{c0}. To avoid excessive decay of J_c due to thermal activation, U_0/k must be at least 2 orders of magnitude higher than the temperature at which the superconductor is used. While this does not pose any severe problem for conventional superconductors whose T_c is generally much smaller than U_0/k, this could seriously limit the useful temperature range of the high-T_c superconductors.

Moreover, in the presence of external fields, flux creep gives rise to resistive dissipation for type-II superconductors in the mixed state, even for current densities less than J_c.[10,11] Palstra *et al.*[11] studied this dissipative effect in Bi 2:2:1:2 finding the following relationship for the resistivity:

$$\rho = \rho_0 \exp(-U_0/kT)\,, \tag{8}$$

with activation energies $U_0/k = 3000$ K for $H(\perp c) = 1$ kOe and $U_0/k = 900$ K for $H(\parallel c) = 1$ kOe. Rather surprisingly, they found that ρ_0 is a constant independent of temperature and field. The very good fit they obtained implies that the distribution of activation energies is quite narrow, perhaps within a few percent.

It is interesting that the pinning energy $U_0 \approx 0.15$ eV we obtained for polycrystalline Y-Ba-Cu-O agrees fairly well with the single-crystal values (0.1-0.6 eV).[2] This suggests that the energy barrier in both cases has the same general physical origin. Phenomenologically, $U_0 \sim (H_c^2/8\pi)\xi_{ab}^2\xi_c \approx 0.15$ eV for Y-Ba-Cu-O (Ref. 2) in close agreement with observed values. Thus, the relatively small value of U_0 for high-T_c superconductors may be reflecting the very short coherence lengths. In fact, measurements reported in the literature indicate $U_0 \approx 0.08$ eV for $Ba_{0.6}K_{0.4}BiO_3$ (Ref. 7) and $U_0 \approx 0.33$ eV for $Tl_2Ca_2Ba_2Cu_3O_x$.[8] Recently, direct observation of the vortex lattice in Y-Ba-Cu-O (Ref. 26) has suggested that pinning may be associated with both twin boundaries and some higher density pinning sites, spaced no more than 100 nm apart, whose origin is undetermined, but which appear to dominate the pinning.

In conclusion, we have found that the flux decay in polycrystalline Bi 2:2:2:3 and Y-Ba-Cu-O is consistent with a conventional flux-creep model. However, the relatively small pinning energies in these materials cause a dramatic decay of their critical current densities, especially for Bi 2:2:2:3 whose pinning energy is only 0.06 eV. Pinning in these materials may be limited by the short coherence lengths.

This work was supported by National Science Foundation Grant No. MSM-8814441.

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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] C. Rossel, Y. Maeno, and I. Morgenstern, Phys. Rev. Lett. **62**, 681 (1989).

[4] Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. Lett. **9**, 306 (1962).

[5] P. W. Anderson, Phys. Rev. Lett. **9**, 309 (1962).

[6] Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).

[7] M. E. McHenry, M. P. Maley, G. H. Kwei, and J. D. Thompson, Phys. Rev. B **39**, 7339 (1989).

[8] M. E. McHenry, M. P. Maley, E. L. Venturini, and D. L. Ginley, Phys. Rev. B **39**, 4784 (1989).

[9] B. D. Biggs, M. N. Kunchur, J. J. Lin, S. J. Poon, T. R. Askew, R. B. Flippen, M. A. Subramanian, J. Gopalakrishnan, and A. W. Sleight, Phys. Rev. B **39**, 7309 (1989).

[10] M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).

[11] T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1662 (1988).

[12] J. Mannhart, P. Chaudhari, D. Dimos, C. C. Tsuei, and T. R. McGuire, Phys. Rev. Lett. **61**, 2476 (1988).

[13] S. M. Green, C. Jiang, Yu Mei, H. L. Luo, and C. Politis, Phys. Rev. B **38**, 5016 (1988).

[14] S. G. Sankar, H. S. Lessure, and S. Simizu, in *Proceedings of the Indo-U.S.-Japan Workshop on Electronic Materials, Pune, India, 1989* [Ferroelectrics (to be published)].

[15] J. D. Jorgensen, B. W. Veal, W. K. Kwok, G. W. Crabtree, A. Umezawa, L. J. Nowicki, and A. P. Paulikas, Phys. Rev. B **36**, 5731 (1987).

[16] U. Endo, S. Koyama, and T. Kawai, Jpn. J. Appl. Phys. **27**, L1476 (1988).

[17] A. M. Hermann, Z. Z. Sheng, D. C. Vier, S. Schultz, and S. B. Oseroff, Phys. Rev. B **37**, 9742 (1988).

[18] P. W. Anderson, Phys. Rev. Lett. **9**, 309 (1962).

[19] P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).

[20] M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682 (1969).

[21] C. P. Bean, Rev. Mod. Phys. **36**, 31 (1964).

[22] T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. **58**, 2687 (1987).

[23] A. J. Panson, G. R. Wagner, A. I. Braginski, J. R. Gavaler, M. A. Janocko, H. C. Pohl, and J. Talvacchio, Appl. Phys. Lett. **50**, 1104 (1987).

[24] P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. L. Bishop, Phys. Rev. Lett. **61**, 1666 (1988).

[25] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), pp. 176-7.

[26] G. J. Dolan, G. V. Chandrashekhar, T. R. Dinger, C. Feild, and F. Holtzberg, Phys. Rev. Lett. **62**, 827 (1989).