Magnetic relaxation and intrinsic pinning in a single crystal of $Bi_2Sr_2CaCu_2O_x$

S. Sengupta, Donglu Shi, Zuning Wang, and M. E. Smith Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

S. Salem-Sugui, Jr.

Instituto de Fisica, Caixa Postale 6165, Universidade Estadual de Campinas 13081-Campinas, São Paulo, Brazil

P. J. McGinn

Department of Electrical Engineering, University of Notre Dame, Notre Dame, Indiana 46556 (Received 15 June 1992; revised manuscript received 17 September 1992)

Magnetic-relaxation experiments were performed on $Bi_2Sr_2CaCu_2O_x$ single crystals with the direction of the field parallel to the *ab* plane. Based on the relaxation data, we have obtained relationships between the activation energy U and the current density j by an approach we developed previously. We found that the activation energy has a logarithmic dependence on j in a wide regime of driving force. It has been reported that CuO_2 planes in high- T_c superconductors can act as strong intrinsic pinning centers and that the relation $U \sim U_0 \ln(j_c/j)$ may describe such a pinning mechanism. Our experimental results have shown good agreement with such a physical model of intrinsic flux pinning.

INTRODUCTION

In the presence of a magnetic field larger than the lower critical field, H_{c1} , the magnetic properties of a type-II superconductor are determined by the static and dynamic properties of vortices. These vortices are pinned by various defects known as pinning centers, resulting in a nonuniform vortex density. As the magnetic pressure is balanced by the pinning force, the system enters a socalled Bean critical state.¹ This critical state is metastable and will relax to achieve a uniform density of vortices as a result of thermal activation. This relaxation of magnetization, which is also known as flux creep, has been observed in type-II superconductors.²⁻¹⁰ Most of the previous studies have focused on relaxation measurements for H parallel to the c axis, in which intrinsic pinning may not dominate. Our study focused on the U-j relationship for layered superconductors with strong intrinsic pinning as the field is applied parallel to the layers. In particular we studied $Bi_2Sr_2CaCu_2O_x$ single crystals. Our motivation was twofold. First, as has been reported, the coherence length for high- T_c superconductors is very small ($\xi_c \sim 1-2$ Å); hence, the vortices formed for H parallel to the ab plane will also be small (since the core diameter is proportional to coherence length). These vortices will then be easily trapped between the copper oxide planes.¹¹⁻¹³ Second, according to the previously proposed intrinsic pinning model,^{11,12} for H||ab plane, $Bi_2Sr_2CaCu_2O_x$ should have a strong intrinsic pinning effect. This motivated us to study the magnetic relaxation of $Bi_2Sr_2CaCu_2O_x$ single crystal for H||ab plane. In this paper we report the results of long-time (22 h) isothermal magnetic relaxation in $Bi_2Sr_2CaCu_2O_x$ single crystals and discuss the U-j characteristics related to intrinsic pinning.

EXPERIMENTAL DETAILS

High-quality $Bi_2Sr_2CaCu_2O_x$ single crystals were grown using self-flux method. The crystals were characterized using x-ray-diffraction, resistivity, and magnetization measurements which have been previously reported.¹⁴ Magnetic-relaxation measurements were performed with a Quantum Design superconducting quantum interference device (SQUID) magnetometer. The samples were first zero-field cooled to a desired temperature below T_c . The magnetic field was then applied, parallel to the *ab* plane of the single crystal. The initial magnetization was recorded 120 sec after stabilization of the field. The travel length of the sample in each scan was 3 cm to avoid any field inhomogeneity.

RESULTS AND DISCUSSION

Anderson and Kim first explained magnetic relaxation by a thermally activated flux-creep model.¹⁵ They assumed that the flux motion is thermally activated and

<u>47</u> 5414

MAGNETIC RELAXATION AND INTRINSIC PINNING IN A . . .

that the rate with which these flux bundles jump over the pinning barriers can be described by an Arrhenius-type expression

$$v = v_0 \exp\{-U(j)/kT\}$$
, (1)

where v_0 is an attempt frequency and U(j) is the effective activation energy.

One can extract U(j) from the relaxation of magnetization using the relation developed by Maley *et al.*,¹⁶ which can be written as a

$$U(j)/k = -T \ln |dM/dt| + T \ln(Bv\omega/\pi d) , \qquad (2)$$

where d is the thickness of the sample, M is the magnetization, and ω is the average distance that a flux bundle can hop. In the approach of Maley *et al.*, one first calculates $T \ln |dM/dt|$ for various temperatures at a given field. Then by adjusting the constant $C = \ln(Bv\omega/\pi d)$, one obtains the U vs j relationship. For obtaining the best fit, one uses a scaling function g(t), where $t = T/T_x$ is the reduced temperature and T_x is a temperature characteristic of the system.¹⁷

We have previously developed an alternative and equivalent way of extracting U(j) from magnetic-relaxation experiments.^{18,19} In this approach one starts

with the standard rate equation

$$dj/dt = A \exp\{-U(j)/kT\}.$$
(3)

The effective activation energy U(j) can be expanded about some current density, j_0 , at time t_0 as

$$U(j) = U(j_0) + [\partial U/\partial j]_0(j - j_0) + \frac{1}{2} [\partial^2 U/\partial j^2]_0(j - j_0)^2 + \cdots \approx U(j_0) + \alpha(j - j_0) + (\beta/2)(j - j_0)^2 .$$
(4)

Substituting Eq. (4) in Eq. (3) and assuming that the preexponential factor A is constant in the range of temperature considered, one obtains

$$j(t) = j_0 + (kT/\alpha) \ln(t/t_0) - (k^2 T^2 \beta / 2\alpha^3) \ln^2(t/t_0) .$$
 (5)

Equation (5) can be rewritten using Bean's model as

$$M(t) = M_0 + a \ln(t/t_0) + b \ln^2(t/t_0) .$$
(6)

One can determine the constants a and b from relaxation experiments and use them to calculate α and β . Since $M - M_{eq} \propto j$ and M_{eq} (M_{eq} is the equilibrium magnetization) is observed negligibly small compared with M, we have studied U vs M instead of U vs j (which are equivalent).



FIG. 1. Magnetization vs lnt at a field of 0.1 T applied parallel to the *ab* plane of a Bi₂Sr₂CaCu₂O_x single crystal: (a) T = 26 K, (b) T = 30 K, (c) T = 34 K, and (d) T = 40 K. The curved lines are the fits to $a \ln(t/t_0) + b \ln^2(t/t_0)$.

It has been reported that when the vortex motion is controlled by the intrinsic pinning in a layered superconducting system with the field parallel to the layers, the effective activation barrier grows logarithmically with decreasing current

$$U(j) = U_0 \ln(j_c/j) , \qquad (7)$$

where $U_0 = -kT/(d \ln M/d \ln t)$ gives the proper scaling to the pinning energy.²⁰

Figure 1 shows the decay of magnetization for a $Bi_2Sr_2CaCu_2O_x$ single crystal with H ||ab plane at temperatures 26, 30, 34, and 40 K. As the temperature increases the nonlinearity of magnetization becomes more pronounced, particularly near the irreversibility line. These magnetization curves are well fitted with Eq. (6) as shown in Fig. 1, indicating the physical relevance of the nonlinear relationship between U and j [Eq. (4)].

According to Eq. (4), we can define the change in activation energy as magnetization has decayed from M_0 to M. Thus, we have

$$\Delta U = U(M) - U(M_0)$$

= $\alpha (j - j_0) + (\beta/2)(j - j_0)^2$. (8)

Figure 2 shows ΔU vs M at various temperatures. One can see that for the temperatures 26, 30, 34, and 40 K, ΔU varies logarithmically, a result that is in good agree-

ment with Eq. (7). However, the relationships shown in Fig. 2 are all obtained at a given temperature where the variation of j is small. We studied the U-j relationship in a much wider current range and determined the validity of Eq. (7) as j has significantly decayed from M = -0.00174 emu to M = -0.000359 emu at T = 26 K.

In previous studies, j_c was generally defined as the current density in the absence of flux creep. Ideally, to obtain a complete U-j curve at constant temperature and field, one has to study long-time relaxation. Since the relaxation is approximately logarithmic, one would require years to get a considerable portion of the U-j curve. Alternatively, we can construct a considerable portion of the U-*i* curve by studying the isothermal magnetic relaxation at a constant field for various temperatures or, equivalently, by studying the magnetic relaxation at constant temperature for various fields. In these cases we change the critical current density j_c . We now construct the U-j curve for the highest j_c (T = 26 K). Then we calculate the change in U, due to the change in j, achieved by varying temperature. Next we add a constant to the ΔU values for the lower j_c . Mathematically one can write

$$U(j/j_{c0}) = U(j/j_{c1}) + C(j_{c0}, j_{c1}) , \qquad (9)$$



FIG. 2. ΔU vs magnetization *M* plots for (a) T = 26 K, (b) T = 30 K, (c) T = 34 K, and (d) T = 40 K. The lines are fit to $\ln(j)$.

where C is a constant depending on j_{c0} and j_{c1} . As we have shown previously,²¹ Eq. (9) can be used if and only if ΔU varies linearly with $\ln j$. As we have already shown in Fig. 2, ΔU varies linearly with $\ln j$, and hence one can use Eq. (9) to construct the U-j curve for T = 26 K.

In Fig. 3 we have plotted U vs M for T = 26 K using Eq. (9). Again a logarithmic dependence of U on $M(\sim j)$ was also observed in this plot, indicating the validity of Eq. (7) in a wide current-density regime. As can be seen in Fig. 3, the U value obtained by this approach is 0.05 eV at 26 K and 0.1 T at the lowest driving force. This value is comparable with what has been previously reported by Inui, Littlewood, and Coppersmith²² and Harshman *et al.*²³ using the resistivity and muon-spin-relaxation methods, respectively.

A strong intrinsic pinning mechanism has been previously reported by Tachiki and Takahashi.¹¹ Considering the layered crystal structure and superconducting characteristics of high- T_c oxides, they proposed that the vortices can be strongly pinned by the CuO₂ layers. As the order parameter varies considerably from the CuO₂ conducting planes to the CuO chains, a modulation of the order parameter must exist in all two-dimensional high- T_c systems. If one assumes that the CuO₂ planes are principally responsible for the occurrence of superconductivity, the order parameter is the highest where the carrier density is also high. The order parameter gradually decreases to a minimum as it approaches the center between the CuO_2 planes. As the flux lines enter the superconductor with the direction of the field parallel to the ab plane, the regions where the order parameter is low (i.e., between the superconducting CuO₂ planes) will be penetrated by the external field. In this situation, the vortices are highly stabilized between the layers, and thus are strongly pinned by the CuO₂ planes. This physical picture is illustrated in Fig. 4(a). Assuming that the xand y axes parallel to the a and b axes in CuO_2 plane and z axis is parallel to the c axis, one can write the periodic spatial variation of the order parameter as¹¹

$$\Psi_0(z) = \Psi_1 + \Psi_2 \cos[2\pi z/a_c] , \qquad (10)$$



FIG. 3. Effective activation energy U vs magnetization M plots for a $Bi_2Sr_2CaCu_2O_x$ single crystal with the H||ab plane at T=26 K and H=0.1 T.



FIG. 4. Schematic diagram showing (a) vortices trapped between the CuO_2 layers where the superconducting parameter is suppressed, and (b) the "breakdown of the FLL lattice."

where Ψ_1 and Ψ_2 are positive parameters and Ψ_1 is larger than Ψ_2 . Here a_c is the period of the modulation of order parameter. In the presence of a vortex whose axis is parallel to the y axis passing through the point x = y = 0and $z = z_0$, the spatial variation of the order parameter can be written as¹¹

$$\Psi(r) = \Psi_0(z) \tanh\{(x/\xi_{ab})^2 + [(z-z_0)/\xi_c]^2\}^{1/2}.$$
 (11)

The elementary pinning force f_p can be estimated as

$$f_p = (H_{c2}/8\pi) \{ 2\pi a_c (\xi_{ab}/\xi_c)\eta \}$$

where η is a nondimensional quantity defined by

$$\eta = (4\Psi_2/\Psi_1 a_c^2) \int dx \, dz \{ 1 + (\Psi_2/\Psi_1) \cos[2\pi (z+z_0)/a_c] \}$$
$$\times \sin[2\pi (z+z_0)/a_c] \\\times \operatorname{sech}^2[\{ (x^2+y^2)^{1/2} \}/\xi_c]].$$

This is the pinning force that acts on one vortex at the boundary between weak and strong superconducting regions. The total pinning force can now be obtained by direct summation, that is,

$$F_p = nf_p \quad , \tag{12}$$

where n is the vortex density. Therefore

$$F_{p} = (H_{c}^{2} / 8\pi a_{c})(\xi_{ab} / \xi_{c})\eta\{(B / B_{0})[1 - B / H_{c2}]\}, \quad (13)$$

where $B_0 = \Phi_0 / 2\pi a_c^2$.

Recently Kes *et al.*²⁴ reported that for layered superconductors such as $Bi_2Sr_2CaCu_2O_x$, the concept of a flux-line lattice (FLL) breaks down when the field is parallel to the superconducting CuO₂ planes. This situation is schematically shown in Fig. 4(b). In that case, a description in terms of superconducting layers coupled by a two-dimensional (2D) Josephson junction is more appropriate than an anisotropic 3D Ginzburg-Landau model. In the 2D model, the order parameter is large in the CuO₂ planes and is nearly uniformly zero between the layers. However, the situation is different in the intrinsic pinning picture where the CuO₂ planes are not completely decoupled by the field. According to the Ginzburg-Landau theory for anisotropic superconductors, for the $H \parallel ab$ plane, the field penetrates in the form of a flux lattice consisting of isosceles triangles.²⁵ Farell et al.²⁶ have estimated the FLL unit cell to be 6.3 nm in the c direction and 300 nm in the direction perpendicular to c for B = 1 T. We have used a field of 0.1 T in our experiment. In this low field, the distance between the FLL planes is 19.9 nm. This distance is significantly larger than the separation between CuO₂ planes. This suggests that the Abrikosov lattice can still exist and hence the planes can act as intrinsic pinning centers. It should be pointed out that similar results have been reported earlier by Harshman et al.²³ By using muon-spin relaxation they have shown that at low fields (0.3 T and 0.4 T) the CuO₂ planes are still coupled and the Abrikosov lattice exists. However, at higher fields (~ 1.5 T), the intervortex interaction begins to dominate and leads to breakdown of a 3D lattice.

CONCLUSION

We have conducted long-time relaxation studies of $Bi_2Sr_2CaCu_2O_x$ single crystals for the H||ab| plane. We

have shown that Eq. (7) is valid in the range (26-40 K) of temperature considered. Our results indicate that the pinning in Bi₂Sr₂CaCu₂O_x single crystals for the H||ab plane is intrinsic to the system and that the CuO₂ planes can act as strong pinning centers. The decoupling of the CuO₂ plane may not occur at such low magnetic fields (0.1 T). Our findings are also in agreement with the previously reported results indicating CuO₂ layers are strong intrinsic pinning centers.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under Contract No. W-31-109-ENG-38 (D.S., S.S.S., Z.W., and M.S.). S.S. and P.M. acknowledge support from the Midwest Superconductivity Consortium (DOE Contract No. DE-FG02-90ER45427). S.S.S. acknowledges partial support from Fundação de Amparo a Pesquisa do Estado de São Paulo (FAPESP), Brazil.

- ¹C. P. Bean, Phys. Rev. Lett. 8, 250 (1962).
- ²M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682 (1969).
- ³K. A. Muller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
- ⁴Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
- ⁵H. S. Lessure, S. Simizu, and S. G. Sankar, Phys. Rev. B **40**, 5165 (1989).
- ⁶Y. Xu, M. Suenaga, A. R. Modenbaugh, and D. O. Welch, Phys. Rev. B **40**, 10 882 (1989).
- ⁷D. Shi and M. Xu, Phys. Rev. B 44, 4548 (1991); D. Shi, M. Xu, A. Umezawa, and R. F. Fox, *ibid.* 42, 2062 (1990); D. Shi and S. Salem-Sugui, Jr., *ibid.* 44, 7647 (1991); S. Salem-Sugui, Jr. and Donglu-Shi, *ibid.* 46, 6618 (1992).
- ⁸S. Sengupta, P. McGinn, N. Zhu, and W. Chen, Physica C 171, 174 (1990).
- ⁹M. Turchinskaya, L. H. Bennett, L. J. Swartzendruber, A. Roitburd, C. K. Chiang, M. Hill, J. E. Blendell, and K. Sawano, *High Temperature Superconductors: Fundamental Properties and Novel Materials Processing*, edited by D. K. Christen, J. Narayan, and L. F. Schneemeyer, MRS Symposia Proceedings No. 169 (Materials Research Society, Pittsburgh, 1990), p. 931; U. Atzmony, R. D. Shull, C. K. Chiang, L. J. Swartzendruber, L. H. Bennett, and R. E. Watson, J. Appl. Phys. 63, 4179 (1988).
- ¹⁰M. D. Lan, J. Z. Liu, and R. N. Shelton, Phys. Rev. B 44, 2751 (1991).
- ¹¹M. Tachiki and S. Takahashi, Solid State Commun. **70**, 291 (1989).
- ¹²D. Fienberg and C. Villard, Phys. Rev. Lett. **65**, 919 (1990).
- ¹³D. H. Kim, K. E. Gray, R. T. Kampwirth, and D. M. McKay,

Phys. Rev. B 42, 6249 (1990).

- ¹⁴Donglu Shi, Ming Tang, Y. C. Chang, P. Z. Jiang, K. Vandervoort, B. Malecki, and D. J. Lam, Appl. Phys. Lett. 54, 2358 (1989).
- ¹⁵P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).
- ¹⁶M. P. Maley, J. O. Willis, H. Lessure, and M. E. McHenry, Phys. Rev. B 42, 2639 (1990).
- ¹⁷M. E. McHenry, S. Simizu, H. Lessure, M. P. Maley, Y. C. Coulter, I. Tanaka, and H. Kojima, Phys. Rev. B 44, 7614 (1991).
- ¹⁸B. M. Lairson, J. Z. Sun, T. H. Geballe, M. R. Beasley, and J. C. Bravman, Phys. Rev. B 43, 10405 (1991).
- ¹⁹S. Sengupta, Donglu Shi, S. Salem-Sugui, Jr., Zuning Wang, P. J. McGinn, and K. DeMoranville, J. Appl. Phys. **72**, 592 (1992).
- ²⁰V. M. Vinokur, M. V. Fiegel'man, and V. B. Geskenbein, Phys. Rev. Lett. **67**, 915 (1991).
- ²¹S. Sengupta, Donglu Shi, Zuning Wang, and M. E. Smith (unpublished).
- ²²M. Inui, P. B. Littlewood, and S. N. Coppersmith, Phys. Rev. Lett. **63**, 2421 (1989).
- ²³D. R. Harshman, R. N. Kleiman, N. Inui, G. P. Espinosa, D. B. Mitzi, A. Kapitulnik, T. Pfiz, and D. Ll. Williams, Phys. Rev. Lett. 67, 3152 (1991).
- ²⁴P. H. Kes, J. Aarts, V. M. Vinokur, and C. J. van der Beek, Phys. Rev. Lett. **64**, 1063 (1990).
- ²⁵P. H. Kes, Physica C 153-155, 1121 (1988).
- ²⁶D. E. Farell, S. Bonham, J. Foster, Y. C. Chang, P. Z. Jiang, K. G. Vandervoot, D. J. Lam, and V. G. Kogan, Phys. Rev. Lett. **63**, 782 (1989).



FIG. 4. Schematic diagram showing (a) vortices trapped between the CuO_2 layers where the superconducting parameter is suppressed, and (b) the "breakdown of the FLL lattice."