Magnetic Penetration Depth and Flux Dynamics in Single-Crystal Bi₂Sr₂CaCu₂O_{8+ δ}

D. R. Harshman, R. N. Kleiman, ^(a) M. Inui, ^(b) and G. P. Espinosa AT&T Bell Laboratories, Murray Hill, New Jersey 07974

D. B. Mitzi^(c) and A. Kapitulnil

Department of Applied Physics, Stanford University, Stanford, California 94305

T. $Pfiz^{(d)}$ and D. Ll. Williams

University of British Columbia, Vancouver, British Columbia, Canada V6T 2A6

(Received 2 January 1991)

The muon-spin-relaxation technique has been used to study vortex dynamics in single-phase superconducting single crystals of Bi₂Sr₂CaCu₂O₈₊₈ ($T_c \approx 90$ K). The data indicate motional narrowing of the internal field distribution due to vortex motion (on a time scale comparable to th field-dependent lattice transition is also observed at $T_x \sim 30$ K, as evidenced by the onset of an asymline shape below T_x . Narrowing arising from disordering of the vortices along [001] is also discussed with reference to its effect on the measured penetration depth.

PACS numbers: 74.30.Ci, 74.60.Ge, 74.70.Vy, 76.75.+i

A number of experimental studies of vortex motion in both YBa₂Cu₃O₇-_{δ} and Bi₂Sr₂CaCu₂O_{8+ δ} have been conducted, spawning a variety of interpretations ranging from activated [1-3] to critical [4-7] behavior. At present, it is unclear whether the various views are mutually exclusive since the external driving forces employed the problem. In contrast to conventional techniques vary greatly and the theories consider different aspects of muon spin rotation $(\mu^+$ SR) provides a microscopic measure (under near equilibrium conditions) of the dynamics, where the flux lines are subject only to thermal excitations. Here we present the first μ^+ SR investigation of the fluctuating vortex state in the high- T_c superconductor $Bi₂Sr₂CaCu₂O_{8+\delta}$.

The measurements were performed on single crystals of unannealed material having exact cation stoichiometry, $Bi_{2.1}Sr_{1.94}Ca_{0.88}Cu_{2.07}O_{8+\delta}$ (with $T_c \approx 90$ K and ± 0.03 stoichiometric error). The crystals (about fifty in total), each with approximate dimensions $3 \times 3 \times 0.1$ mm³ (the shortest dimension being parallel to the crystgallographic c axis), were grown by a technique described elsewhere [8]. They were assembled into a flat mosaic sample with their c axes aligned, and mounted on a 99.999% pure aluminum plate. Time-differential μ ⁺SR [9] was employed (in transverse-field geometry), where an external magnetic field, H_{ext} , is applied perpendicular to the incident μ^+ spin polarization. The experiments were conducted on field cooling with H_{ext} applied along the beam momentum, and parallel to the c axis of the mosaic sample. As detailed in previous work [10-12], the penetration depth $\lambda(T)$ is determined from the relaxation of the μ^+ spin polarization, with H_{ext} chosen so that the vortex lattice spacing l is small compared to λ . Given the small lattice spacing *l* is small compared to λ . Given the small coherent length ξ (~10 Å), and the associated large H_{c2} coherent length ξ (\sim 10 Å), and the associated large H_{c2}
(\sim 440 kG), the London picture $(H_{c1} \ll H_{ext} \leq H_{c2}/4)$ is satisfied for the fields (3, 4, and 15 kG) used here. Under such conditions, the μ^+ -spin-relaxation rate is expected to be independent of H_{ext} , and directly related to the second momentum of the field distribution, $\langle (\Delta B)^2 \rangle$. For

a perfect triangular vortex lattice, $\langle (\Delta B)^2 \rangle$ is related to λ via the equation [13]

$$
\langle (\Delta B)^2 \rangle = \langle B^2 \rangle - \langle B \rangle^2 = 0.00371 \phi_0^2 / \lambda^4 , \qquad (1)
$$

where ϕ_0 (=2.068×10⁻⁷ Gcm²) is the magnetic-flux quantum. Implicit in Eq. (1), of course, are the assumptions that the vortices are static and form ines parallel to 1' H_{ext} . In the present case, however, these conditions are not satisfied, due to thermal fluctuations and disordering.

The internal field distribution $\eta(B)$, obtained via Fourier transform of the 3- and 4-kG data, is shown for selected temperatures in Fig. 1. Note the evolution of the line shape with temperatures above and below $T_x \sim 30$ K: For $T_x \lesssim T \leq T_c$, a rather narrow and symmetric field distribution is observed, while for $T \lesssim T_{x}$, the line shape

FIG. 1. Fourier spectra (peak height normalized to 1 at 76 K) for (unannealed) $Bi_{2,1}Sr_{1,94}Ca_{0.88}Cu_{2,07}O_{8+6}$ at (a) 3 and (b) 4 kG (shown for selected temperatures). The time-domain lata were smoothly truncated using a Gaussian function $\exp(-\sigma^2 t^2)$ with $\sigma = 0.25 \mu s^{-1}$, prior to transformation.

becomes broadened and more asymmetric. To model $\eta(B)$, we employ a back-to-back cutoff exponential [12] of the form

$$
\eta(B) = \begin{cases} \frac{1}{B_1 + B_2} \exp\left(+\frac{B - B_c}{B_1}\right), & B \leq B_c, \\ \frac{1}{B_1 + B_2} \exp\left(-\frac{B - B_c}{B_2}\right), & B > B_c, \end{cases}
$$
(2)

where B_c is the peak frequency. Unique fits were obtained using the Fourier transform of Eq. (2), multiplied by a Gaussian function representing the nuclear dipole contribution. For '15 kG, we were able to impose the additional condition of $\langle B \rangle = B_0$ on Eq. (2) in order to reduce the number of free parameters $(B_c$ was still allowed to vary).

The fitted values for $\langle (\Delta B)^2 \rangle$, $\langle B \rangle - B_0$, and $\langle (\Delta B)^3 \rangle$ (inset shows $B_c - B_0$), for $H_{ext} = 3$ and 4 kG, are shown as a function of temperature in Fig. 2. The data show a diamagnetic shift in $\langle B \rangle - B_0$ [Fig. 2(b)], including a pronounced minimum at $T_x \sim 30$ K. A related response is also seen in both $\langle (\Delta B)^2 \rangle$ and $\langle (\Delta B)^3 \rangle$ [Figs. 2(a) and 2(c), respectivelyl, with $\langle (\Delta B)^2 \rangle$ exhibiting behavior strikingly different from that observed for $YBa_2Cu_3O_{7-\delta}$ strikingly different from that observed for γ Ba₂Cu₃O₇ – δ
[11,12]. Recall that YBa₂Cu₃O₇ – δ was found to be con-
sistent with s-wave pairing, with a penetration depth well
described by the two-fluid sistent with s-wave pairing, with a penetration depth well described by the two-fluid model [14],

$$
\lambda(T) = \lambda(0)[1 - (T/T_c)^4]^{-1/2},\tag{3}
$$

represented by the dashed curves. The fitted results for $\langle (\Delta B)^2 \rangle$, corresponding to the 15-kG data, are shown as a function of temperature in Fig. 3. In contrast to the lower-field data, $\langle (\Delta B)^2 \rangle$ for 15 kG appears relatively independent of temperature for $T \lesssim T_c/2$, deviating only slightly from two-fluid behavior (dashed curve, $\beta = 0$) nearer T_c .

The unique behavior exhibited by the lower-field (3 and 4 kG) data clearly indicates two distinct temperature regimes, $T \lesssim T_x$ and $T_x \lesssim T \leq T_c$. The observed evolution of $\eta(B)$ with temperature, coupled with the marked departure of $\langle (\Delta B)^2 \rangle$ from Eq. (3) [Fig. 2(a)], suggests a motional narrowing of the internal field distribution arising from vortex motion, which "freezes" out with decreasing temperature. This idea is further supported by $\langle (\Delta B)^3 \rangle$ [Fig. 2(c)], showing a persisting near-symmetric line shape above T_x , which becomes more asymmetric below T_x . The evolution from a symmetric to asymmetric line shape is certainly not first order, since all three moments still exhibit a significant temperature dependence below T_x . However, in the absence of a detailed microscopic theory of the transition dynamics, the distinction between a second-order (or Kosterlitz-Thouless [15]) phase transition and the sharp crossover of the thermal activation model (to be discussed) is difficult. The minimum observed in $\langle B \rangle - B_0$ at T_x [Fig. 2(b)] does not follow directly from a simple model of vortex freezing

FIG. 2. Fitted values for (a) $\langle (\Delta B)^2 \rangle$, (b) $\langle B \rangle - B_0$ (where B_0 is measured above T_c), and (c) $\langle (\Delta B)^3 \rangle$ (inset shows $B_c - B_0$) vs temperature in single-crystal $Bi_{2,1}Sr_{1,94}Ca_{0.88}Cu_{2.07}$ - $O_{8+\delta}$ (unannealed), for $H_{ext} = 3$ and 4 kG (open and solid symbols, respectively). The solid lines are a fit of a temperaturedependent phenomenological function incorporating our crossover model (Ref. [Ig]), while the dashed curves represent the two-fluid behavior of Eq. (3).

or localization; normally, one expects that $\langle B \rangle = B_0$ $+4\pi M(1-N)$, where M is the sample magnetization and N the demagnetization factor. Since no magnetic transitions have been observed below T_c for this system, it is natural to attribute the anomalous behavior in $\langle B \rangle$ to changes in the flux lattice. Given this, we are presented with two possibilities: One possibility, specific to μ ⁺SR,

FIG. 3. The second moment, $\langle (\Delta B)^2 \rangle$, plotted as a function of temperature for (unannealed) single-crystal $Bi_{2,1}Sr_{1,94}Ca_{0.88}$ - $Cu_{2.07}O_{8+\delta}$, for H_{ext} =15 kG. The solid curve corresponds to the two-fluid behavior of Eq. (3), multiplied by the Debye-Waller factor, discussed in the text, for $\beta = 0.25$. The dashed curves represent the same, in the static $(\beta = 0)$ and "nonlocal" $(\beta = 1, \text{Ref. } [6])$ limits.

suggests an enhanced correlation between the stopped muons and the vortex cores. This is plausible if we, for example, assume that the muons preferentially trap at existing pinning centers. The second possibility, although generally ruled out by dc magnetization (where the sample is moved in and out of a homogeneous field) and NMR studies of this system, is a change in the bulk magnetization of the flux lattice (magnetization measurements [16] where the sample is held stationary and cooled in ^a constant magnetic field—as is the case for μ ⁺SR— have not yet been completed). In higher fields (e.g., 15 kG), the intervortex interactions begin to dominate, restricting the independent motion (see Fig. 3) of individual "vortex disks" of the Lawrence-Doniach (LD) model [17]. In the LD model, the lattice coherence between layers is expected to decrease with increasing field [17], producing a situation where the density fluctuation of vortices is suppressed without forming a 3D lattice [a trend opposite to that observed [18] in low-field (i.e., \lesssim 100 G) decoration experiments. An indication that this picture may be appropriate is the comparatively reduced asymmetry in $\eta(B)$. Whether the LD model is in fact consistent with a reduced asymmetry, however, remains an open theoretical question. The solid curve in Fig. 3 represents the best fit of the two-fluid model incorporating a Debye-Wailer-like vibration of the vortices, represented by $exp(-2\beta(|u|^2)/l^2)$, where $\langle |u|^2 \rangle$ is the mean-square displacement from equilibrium [6]. The excellent agreement with the 15-kG data may indicate that the vortex motion is indeed dominated (at this field) by the intervortex interactions. Given the uncertainties involved, however, it is impossible to make a quantitative comparison.

To quantify the low-field behavior, we adopt an ap-

proximation in which individual vortices are represented by the function $W(r, T)$ defining the time-averaged (over a few muon lifetimes) probability of finding a vortex at position r , given the initial position $r=0$. This is not exact, but appropriate in the limit of significant diffusive or vibrational motion during the observation time set by τ_u . From this, the local magnetic field $h(r, T)$, in the presence of vortex motion, can be approximated by solving the modified London equation [14],

$$
\mathbf{h}(\mathbf{r},T) + \lambda^2 \mathbf{\nabla} \times (\mathbf{\nabla} \times \mathbf{h}) = \hat{\mathbf{e}}_z \phi_0 \sum_{[\mathbf{r}_i]} W(\mathbf{r} - \mathbf{r}_i, T) , \qquad (4)
$$

where the $[r_i]$ are the initial positions of the vortices at time $t = 0$ and $\lambda(T)$ is expected to diverge according to Eq. (3) . For vortex displacements comparable to l , the width of $W(r, T)$ becomes large, whereas if the motion is constrained (as in the case of pinning), then $W(r, T)$ has a narrow peak near the origin. We can thus reproduce the motional narrowing of $\langle (\Delta B)^2 \rangle$ with this method. A complete analysis will be published separately [19]. For this work, however, we present only order-of-magnitude arguments in order to extract the energy scale of the crossover between the two temperature regimes, above and below T_x . The simplest approach is to assume activation dynamics, and define T_x to be the temperature where the inverse of the experimentally available time window (of order τ_{μ}^{-1}) becomes comparable with the vortex escape (depinning) rate, $\tau^{-1} = \tau_0^{-1} \exp(-E/kT)$. Equating the two rates at $T = T_x$ and using an attempt Equating the two rates at $T = T_x$ and using an attempt

frequency of $\tau_0^{-1} \sim 1 \times 10^{10}$ s⁻¹ from Ref. [2] yields activation (or crossover) energies of \sim 200 and \sim 300 K for 3 and 4 kG, respectively, in reasonable agreement with resistivity measurements [1].

The nature of the low-temperature vortex state is an important factor when determining the intrinsic penetration depth: For a triangular lattice of rigid rods aligned parallel to H_{ext}, $\lambda(0)$ is directly related to $\langle (\Delta B)^2 \rangle$ via Eq. (1), whereas if the flux lines are tilted with respect to H_{ext} , as in the case of flux entanglement [20], or the disordered vortex disks of the LD model [17], the measured line shape would be narrower. While complications arising from motional narrowing and possible μ^+ vortex correlation effects are reduced in high fields (due to intervortex interactions), narrowing due to vortex disordering along [001] becomes increasingly important. Ignoring this effect results in an artificially high estimation of the basal-plane penetration depth, $\lambda_{ab}^{eff}(0) \approx 4200$ Å, for the 15-kG data of Fig. 3 [assuming a Gaussian line shape gives $\lambda_{ab}^{\text{eff}}(0) \approx 4700$ Ål, which is significantly greater than the 1400 Å determined earlier for $YBa_2Cu_3O_7$ [11,12]. Theoretical estimates [19] of the narrowing associated with c -axis disorder, however, set the intrinsic value, $\lambda_{ab}(0)$, at least 20%-30% lower. In any case, it is clear that a simple scaling [21] of the Gaussian width parameter with T_c does not hold.

In summary, we have probed the vortex dynamics in single-crystal (unannealed) $Bi_2Sr_2CaCu_2O_{8+\delta}$ using

 μ ⁺SR. Lower-field (3 and 4 kG) data indicate motional narrowing associated with vortex motion. A field-dependent localization transition is also observed at $T_x \sim 30$ K, as evidenced by the onset of an asymmetric line shape and anomalous rise in $\langle B \rangle - B_0$ below T_x . Although the data are consistent with thermally activated depinning (incorporating a time crossover), the possibility of a second-order or Kosterlitz-Thouless transition cannot be ruled out. In high fields (15 kG) where the intervortex interactions restrict the motion of individual vortices, the familiar s-wave signature is nearly recovered. The observed line shape, however, is considerably more symmetric and independent of temperature than that found at lower fields, possibly suggesting a more disordered lattice. By correcting for the narrowing effects of c -axis vortex disorder, we estimate an intrinsic basal-plane penetration depth at least $20\% - 30\%$ lower than the effective value, $\lambda_{ab}^{\text{eff}}(0) \approx 4200$ Å. Finally, our ability to observe the anomalous behavior at the lower fields (3 and 4 kG) clearly indicates that most (if not all) of the vortices are potentially mobile.

We are grateful to C. Ballard and K. Hoyle for technical assistance. We also thank G. Aeppli, P. B. Littlewood, A. T. Fiory, S. N. Coppersmith, M. A. Schlüter, C. A. Murray, B. Batlogg, and D. R. Nelson for many helpful discussions. The work at TRIUMF is supported in part by a grant from NSERC and, through TRIUMF, by NRC of Canada.

- Present address: Los Alarnos National Laboratory, Los Alamos, NM 87545. '
- Present address: IBM T. 3. Watson Research Center, Yorktown Heights, NY 10598.
- Present address: University of Stuttgart, D-7000 Stuttgart 80, Germany.
- [1] T. T. M. Palstra et al., Phys. Rev. Lett. 61, 1662 (1988); T. T. M. Palstra et al., Phys. Rev. B 41, 6621 (1990), and references therein.
- [2] A. P. Malozemoff et al., Phys. Rev. B 38, 7203 (1988); J. van den Berg et al., Supercond. Sci. Technol. 1, 249 (1989).
- [3] M. Inui et al., Phys. Rev. Lett. 63, 2421 (1989); M. V. Feigelman et al., Phys. Rev. Lett. 63, 2303 (1989).
- [4] P. L. Gammel et al., Phys. Rev. Lett. 61, 1666 (1988).
- [5] R. H. Koch et al., Phys. Rev. Lett. 63, 1511 (1989).
- [6] A. Houghton et al., Phys. Rev. B 40, 6763 (1989); A. Sudbø (private communication).
- [7] M. P. A. Fisher, Phys. Rev. Lett. 62, 1415 (1989).
- [8] D. B. Mitzi et al., Phys. Rev. B 41, 6564 (1990).
- [9] A. Schenck, Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics (Hilger, Bristol, 1985).
- [10] G. Aeppli et al., Phys. Rev. B 35, 7129 (1987); W. J. Kossler et al., Phys. Rev. B 35, 7133 (1987); F. N. Gygax et al., Europhys. Lett. 4, 473 (1987); P. Birrer et al., Physica (Amsterdam) 158C, 230 (1989).
- [11] D. R. Harshman et al., Phys. Rev. B 36, 2386 (1987).
- [12] D. R. Harshman et al., Phys. Rev. B 39, 851 (1989).
- [13] E. H. Brandt, Phys. Rev. B 37, 2349 (1988).
- [14] See, for example, M. Tinkham, Introduction to Superconductivity (Krieger, Malabar, FL, 1980), Chap. 5.
- [15]J. K. Kosterlitz and D. J. Thouless, J. Phys. C 6, 1181 (1973);J. M. Kosterlitz, J. Phys. C 7, 1046 (1974).
- [16] B. Batlogg (private communication).
- [17] W. E. Lawrence and S. Doniach, in Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, 1970, edited by E. Kanda (Keigaku, Tokyo, 1971), p. 361; S. Doniach, in High Temperature Su perconductivity, edited by K. S. Bedell et al. (Addison-Wesley, Redwood City, 1990), p. 405.
- [18] D. G. Grier et al. (to be published).
- [19] M. Inui and D. R. Harshman (unpublished).
- [20] D. R. Nelson, Phys. Rev. Lett. 60, 1973 (1988); D. R. Nelson and H. S. Seung, Phys. Rev. B 39, 9153 (1989).
- [21] Y. J. Uemura et al., Phys. Rev. B 38, 909 (1988); Y. J. Uemura et al., Phys. Rev. Lett. 62, 2317 (1989).

 (a) Also at Cornell University, Ithaca, NY 14853.