

## Anomalous magnetization in single-crystal $Tl_2Ba_2CuO_6$ : Evidence of dimensional crossover

F. Zuo, S. Khizroev, and G. C. Alexandrakis  
*Physics Department, University of Miami, Coral Gables, Florida 33124*

V. N. Kopylov  
*Institute of Solid State Physics, 142432 Chernogolovka, Russia*  
 (Received 26 April 1995)

Magnetization as a function of field parallel to the  $c$  axis shows an anomalous secondary peak for a single-crystal  $Tl_2Ba_2CuO_6$ . The peak disappears above a crossover temperature  $T_{cr}$ , which is correlated with a crossover in the irreversibility line  $H_{rev}(T)$ . For  $T > T_{cr}$ ,  $H_{rev}(T) \propto (1 - T/T_c)^n$  with  $n = 3/2$ ; for  $T < T_{cr}$ ,  $H_{rev}(T)$  increases with  $1/T$  exponentially, an indication of melting of two-dimensional (2D) vortices. Combined with magnetic-relaxation measurements at  $T < T_{cr}$ , we suggest the anomalous magnetization in this system is due to dimensional crossover from 3D to 2D vortex structure.

The experimental observation of anomalous magnetization  $M(H)$ , which shows an increasing critical current with increasing field, in the field parallel to the  $c$ -axis direction has been first reported in single crystals of  $YBa_2Cu_3O_{7-x}$  (Refs. 1 and 2) and  $Tl_2Ba_2CuO_6$ .<sup>3</sup> A similar effect has been observed in many high-quality single crystals such as  $La_{2-x}Sr_xCuO_{4-y}$ ,<sup>4</sup>  $Bi_2Sr_2CaCu_2O_8$ ,<sup>5,6</sup>  $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ ,<sup>7</sup>  $YBa_2Cu_4O_8$ ,<sup>8</sup> and Ge/Pb superlattices.<sup>9</sup> The enhancement of the critical current has been initially explained in terms of pinning by oxygen-deficient sites. Models based on collective pinning,<sup>10</sup> effects of surface barrier,<sup>3,11</sup> lattice matching between vortex and defect structure,<sup>5</sup> and dimensional crossover have been proposed.<sup>6</sup> The mechanism giving rise to the anomalous effect often referred to as "fishtail" remains controversial.<sup>12</sup>

To understand the origin of the anomalous magnetization, especially in the highly anisotropic system like  $Tl_2Ba_2CuO_6$ , we have performed extensive magnetic hysteresis measurements, combined with measurements of an irreversibility line on the same sample. We report a strong correlation found between the fishtail magnetization and the irreversibility line. The anomalous magnetization disappears above a crossover temperature  $T_{cr}$ , which separates two different regimes on the irreversibility field line  $H_{rev}(T)$ . For  $T > T_{cr}$ ,  $H_{rev} \propto (1 - T/T_c)^n$  with  $n = 3/2$ ; for  $T < T_{cr}$ ,  $H_{rev}$  increases with  $1/T$  exponentially, an indication of two-dimensional (2D) vortex melting. These results combined with the magnetic relaxation measurements at low temperatures provide clear evidence of dimensional crossover as the mechanism for the anomalous fishtail magnetization in the  $Tl_2Ba_2CuO_6$  system.

Single crystals of  $Tl_2Ba_2CuO_6$  were grown using a solid state self-flux method.<sup>3</sup> Several crystals were used in the measurements with average dimensions of  $1 \times 0.5 \times 0.1$  mm. Extensive measurements were made on a crystal with  $T_c = 92$  K. The transition width measured at 1 G was about 2 K. Measurements were performed using a Quantum Design magnetometer with low-field options. A typical hysteresis loop was measured after the sample was zero-field cooled (ZFC) to a set temperature and the magnetization was measured with the superconducting magnet in the persistent

mode. Magnetization relaxation measurements were performed by measuring the magnetization as a function of time in a constant magnetic field after the sample was ZFC from above  $T_c$  to a given temperature. After degaussing and magnet resetting (quenching), the remanent field was typically 5–10 mG. Samples were placed with the field parallel to the  $c$  axis.

Shown in Fig. 1 is an overlay of magnetic hysteresis loops measured at low temperatures ( $T = 32, 38,$  and  $42$  K). Clearly, two peaks are observed in the field ascending branch. The first peak at  $H_p$  corresponds to the Meissner effect, followed by rapid penetration of vortices for  $H > H_p$ ; the magnetization  $M$  reaches a local minimum in magnitude at  $H_{min}$ ;  $M$  increases anomalously with increasing field for  $H > H_{min}$  and the second peak is reached at  $H_{max}$ .  $M$  decreases to a negative and nonzero value at high field  $H > H_{max}$ . On the descending branch, a mirror image of

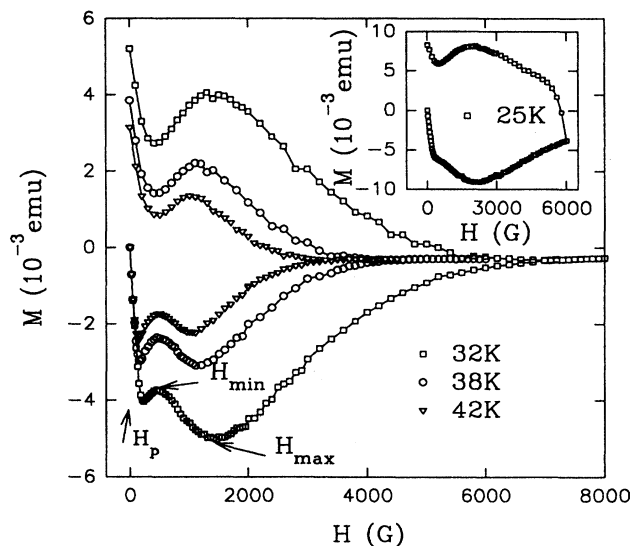


FIG. 1. An overlay of magnetic hysteresis loops at low temperatures  $T = 32, 38,$  and  $42$  K. The inset is a similar hysteresis loop at  $T = 25$  K.

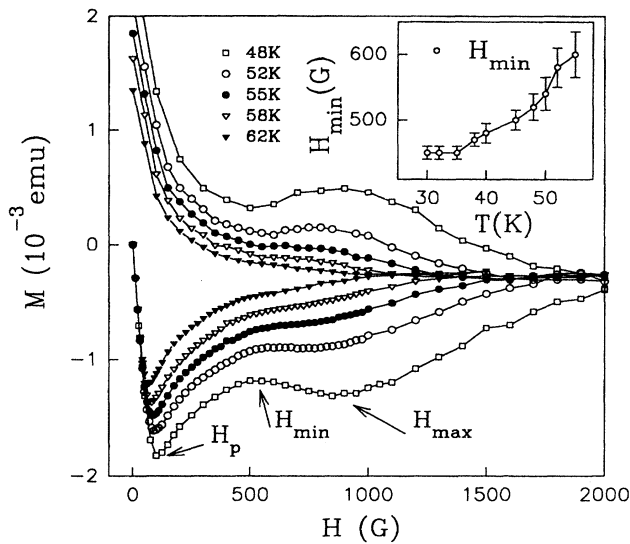


FIG. 2. An overlay of magnetic hysteresis loops at high temperatures  $T=48, 52, 55, 58$  and  $62$  K. The inset is a plot of  $H_{\min}(T)$  versus  $T$ .

the second peak is obtained at  $H_{\max}$ .  $M$  dips to a minimum value at  $H_{\min}$  and rises to a remanent value at  $H=0$ . With increasing temperature, both  $H_{\max}$  and the width of hysteresis loop defined by  $\Delta M=M_+-M_-$  at  $H_{\max}$  decrease. However,  $H_{\min}$  is almost a constant ( $\sim 450$  G) in this temperature range.

At lower temperatures, a similar magnetization curve is observed, as shown in the inset of Fig. 1. A change in slope in  $M(H)$  is observed for  $H>H_p$ , followed by a much broader secondary peak. On the descending branch, the second peak and the dip are again clearly visible.

At higher temperatures, the magnetization curve changes drastically with increasing  $T$ . Plotted in Fig. 2 is an overlay of magnetic hysteresis loops at  $T=48, 52, 55, 58$ , and  $62$  K. At  $T=48$  K, the magnetization at the second peak has smaller magnitude than that of the first peak, in contrast to its low temperature counterpart. At  $T=52, 55$ , and  $58$  K, the second peak evolves into a shoulderlike shape. At  $T=62$  K, the second peak has disappeared completely. The inset shows the temperature dependence of  $H_{\min}$ , which increases with increasing  $T$  toward the crossover temperature ( $\sim 60$  K) and saturates at lower temperatures. At  $T=55$  K,  $H_{\min}$  is about 600 G. It is noted that the error bar in  $H_{\min}$  increases near 60 K as the local minimum becomes less well defined.

Shown in Fig. 3 is a plot of  $\Delta M$  at  $H_{\max}$  as a function of temperature in a semilog scale.  $\Delta M$  decreases with increasing temperature almost exponentially at low temperatures ( $T<50$  K). For temperature above 55 K, we have used  $\Delta M$  as the width in the hysteresis loop at about 700 G, due to the difficulty in identifying the second peak. Clearly, there is a crossover at about 60 K in the temperature dependence of  $\Delta M(T)$ . The solid line is a fit to  $\Delta M(T)=\Delta M_0 \exp(-T/T_0)$  with  $T_0 \approx 10$  K. Plotted in the inset of Fig. 3 is a semilog plot of the second peak field  $H_{\max}$  as a function of  $T$ . Again, an exponential dependence is observed, the line is a fit to  $H_{\max}(T)=H_0 \exp(-T/T_1)$  with  $T_1 \approx 20$  K.

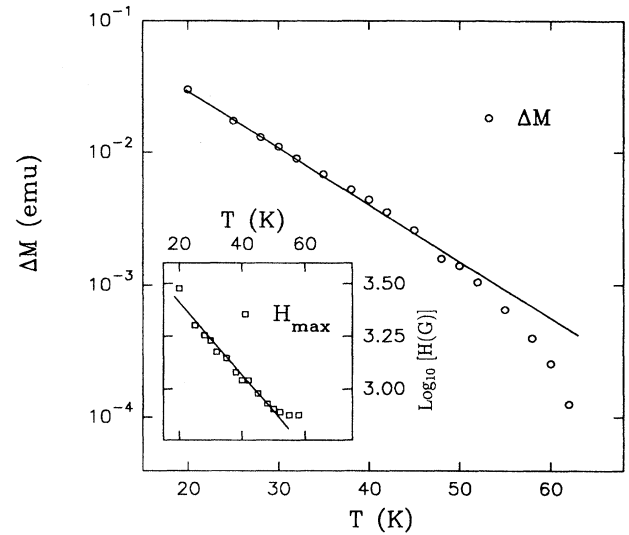


FIG. 3.  $\Delta M$  versus  $T$  in a semilog scale. The inset is a plot of  $\log_{10} H_{\max}$  versus  $T$ .

To correlate the anomalous magnetization with other physical quantities, we have performed extensive magnetic measurements to determine the irreversibility field  $H_{\text{rev}}$  as a function of  $T$ . Using the criterion where  $M$  in both ascending and descending direction coincides with each other, we have obtained an irreversible field  $H_{\text{rev}}(T)$ , as shown in Fig. 4. The open circles are the experimental results plotted in the semilog scale. The dashed and dotted lines are theoretical models. With decreasing temperature,  $H_{\text{rev}}(T)$  increases rapidly. The temperature dependence shows a crossover from negative curvature to positive curvature in the semilog scale. The crossover temperature of about 60 K is the same as the temperature where the anomalous magnetization disappears.

To probe the vortex dynamics of the fishtail magnetization, detailed magnetic relaxation measurements have been performed at different fields and temperatures ( $T<60$  K).

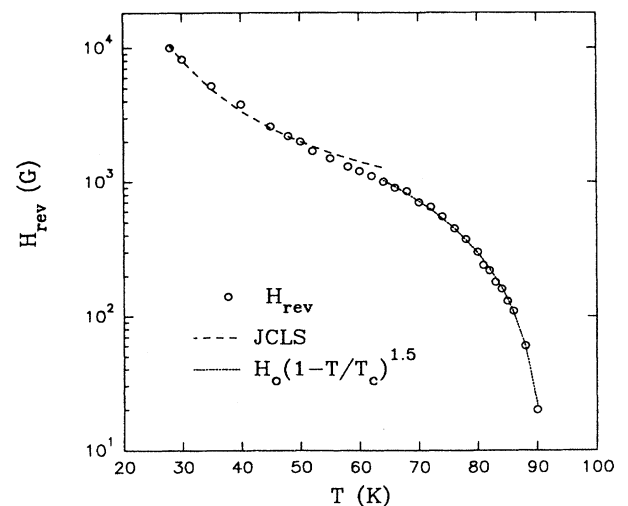


FIG. 4. A plot of irreversibility field  $H_{\text{rev}}(T)$  as a function of  $T$  in a semilog scale. The dashed and dotted lines are theoretical fits.

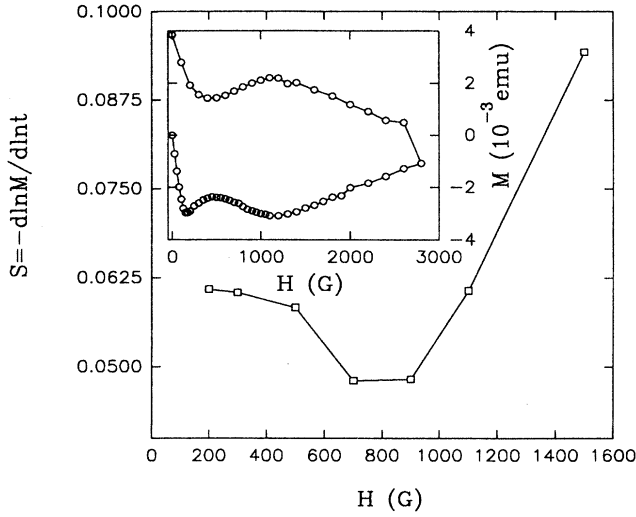


FIG. 5. The normalized relaxation rate  $S$  versus  $H$  at  $T = 38$  K. The inset is a plot of hysteresis loop at the same temperature.

$M(t)$  does not decay linearly with  $\ln t$  as in conventional superconductors, rather it can be well fit by the collective pinning theory<sup>13</sup>  $M(t) = M_0 [1 + (\mu k_B T / U) \ln(t/t_0)]^{-1/\mu}$ , where  $\mu$  is an exponent defined in  $U(j) \propto j^{-\mu}$ ,  $U$  is the pinning barrier,  $t$  is the measuring time, and  $1/t_0$  is the attempt frequency. Shown in Fig. 5 is a plot of the normalized relaxation rate for different fields at  $T = 38$  K. The normalized relaxation rate is defined by  $S = -d \ln M(t) / d \ln t = T/U + \mu T \ln(t/t_0)$ . The inset displays the fishtail magnetization at the corresponding temperature of 38 K.

The normalized relaxation rate  $S$  decreases with increasing field for  $H < H_{\max}$ , above which  $S$  increases sharply with increasing field. An approximate correspondence between the minimum in  $S$  and the secondary peak in  $M(H)$  is observed. The exponent  $\mu$  extrapolated by fitting the collective pinning theory is about 2 at  $H_{\min}$ , and  $\mu$  decreases to about 0.5 at  $H_{\max}$ . It is also noticed that for a similar initial magnetization  $M(H)$  on both sides of the second peak,  $S$  is much larger when  $H > H_{\max}$  than when  $H < H_{\max}$ .

The crossover in  $H_{\text{rev}}(T)$  has been discussed recently in the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  system<sup>14</sup> as evidence of dimensional crossover. For Josephson-coupled layered superconductors (JCLS) with moderate anisotropy, the mean-squared thermal vortex fluctuation displacement  $\langle u^2 \rangle$  for  $B \gg \phi_0 / \lambda_J^2$  ( $\lambda_J = \gamma s$ ), is given<sup>15,16</sup> by  $\langle u^2 \rangle = (8\pi\lambda_{ab}^2 k_B T / \phi_0 s B) \ln[B\lambda_J^2 / \phi_0]$ , where  $\gamma = \lambda_c / \lambda_{ab}$  is the anisotropy constant, and  $s$  is the separation between the superconducting layers. Using the Lindermann melting criterion  $\langle u^2 \rangle \approx c_L^2 a^2 = c_L^2 \phi_0 / B$ , where  $a$  is the intervortex distance and  $c_L$  is the Lindermann number, the melting line has been obtained<sup>14</sup>

$$B_m \approx \frac{\phi_0}{\lambda_J^2} \exp\left(\frac{\phi_0^2 c_L^2 s}{8\pi\lambda_{ab}^2 k_B T}\right). \quad (1)$$

At low temperatures, the exponent in Eq. (1) is dominated by the  $1/T$  term. Using  $\lambda_{ab} \approx \lambda_0 = 2000$  Å, we obtain  $c_L \approx 0.17$

and  $\gamma \approx 190$ , in agreement with the limits  $0.1 \geq c_L \geq 0.4$ . The fit to the JCLS model [Eq. (1)] is shown as the dashed line in Fig. 4.

At high temperatures and for  $B \ll B_{\text{cr}}$ , 3D-like vortex fluctuation is expected with the melting line given by  $B_m(T) \approx B_0(T_c/T - 1)^n$  with  $n = 2$  for pure thermal fluctuation. In our case, we find  $n = 3/2$ . The fit is shown as the dotted line in Fig. 4. The almost universal temperature dependence of the irreversibility line with the exponent  $n = 3/2$  has been mostly attributed to a depinning energy.<sup>17</sup> Recently, the same  $3/2$  exponent has been successfully fit with a quantum fluctuation model. By combining both quantum fluctuation and thermal fluctuation, the melting line is given by a universal function<sup>18</sup>  $B_m(\theta) = 4\theta^2 / (1 + \sqrt{1 + 4Q\theta})^2$ , where  $\theta \propto (1 - T/T_c)$  and  $Q$  measures the relative strength of quantum fluctuation. The universal function gives the same quality of fit as that of the power-law with  $n = 3/2$  within our experimental errors.

Vortex dimensional crossover as suggested from the irreversibility line has been indeed observed by microscopic probes such as neutron-scattering measurements<sup>19</sup> and muon-spin rotation measurements in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ .<sup>20</sup> The crossover field from 3D to 2D as seen from the change of line shape is about 500 G at low temperatures. Using  $\gamma = 190$  obtained for  $\text{Tl}_2\text{Ba}_2\text{CuO}_6$ , the crossover field can be estimated  $B_{\text{cr}} \approx 4\phi_0 / \lambda_J^2 \approx 1$  kG, which corresponds to the boundary separating the different regimes of  $H_{\text{rev}}(T)$ . At high temperatures, magnetic vortices encounter 3D melting first;<sup>21</sup> at low temperatures ( $T < 60$  K) and high fields ( $H > B_{\text{cr}}$ ), vortices assume 2D structure and 2D melting is crossed first as  $H$  is increased. Below the melting line, 2D vortex structure is expected. Consequently, the fishtail magnetization is in the 2D vortex regime.

To describe the motion of 2D vortices, one has to consider intra- and interlayer interaction between vortices. When the interaction between the vortices becomes stronger than the single-vortex pinning energy  $U_p$ , collective motion will dominate. In the 2D collective pinning theory, the pinning energy as function of current has been derived when the hopping distance  $u(j)$  is much smaller than the lattice constant  $a$  (Ref. 22) with  $U(j) \approx U_1(j_c/j)^\mu$ , where  $U_1 \approx (\epsilon_0^2 / U_p)(\xi/a_0)^2$  is the pinning energy at the critical state with  $\epsilon_0 = \phi_0^2 d / (4\pi\lambda)^2$ ,  $\xi$  is the coherence length and  $j_c$  is the critical current for the vortex bundle. The current decays with time according to  $j(t) = j_c [U_1 / T \ln(t/t_0)]^{1/\mu}$ . The exponent  $\mu$  changes from  $7/4$  for large  $j_c$  to  $13/16$  for intermediate  $j_c$ .<sup>23</sup> For small  $j_c$ , corresponding to a large hopping distance  $u(j) \gg a$ ,  $\mu$  is  $1/2$ .<sup>24</sup> However, the crossover between these critical exponents remains to be solved.

The experimental observation of lowest relaxation rate  $S(H)$  at the second peak field supports strongly the picture of 2D collective motion. With increasing field, the vortex bundle has to overcome an increasingly larger pinning barrier. The large barrier decreases the relaxation rate, combined with the decreasing critical current, a peak in the measured ( $t \approx 100$  s) magnetization is quite expected. The exponent  $\mu$  obtained experimentally changing from about 2 to  $1/2$  is in an agreement with the predicted values.<sup>23</sup> The exponential temperature dependence of the critical current ( $\propto \Delta M$ ) at the peak field and  $H_{\max}(T)$  is consistent with the collective

pinning theory.<sup>13</sup> Using  $T_0 \approx U_c / \ln(t/t_0)$  and  $\ln(t/t_0) \approx 10^{10}$ , we find a reasonable barrier height  $U_c$  of order of  $\sim 100$  K.

Thus, the anomalous magnetization in  $\text{Ti}_2\text{Ba}_2\text{CuO}_6$  observed at low temperature provides clear evidence of dimensional crossover. At low field, the Josephson or magnetic interaction between 2D vortices in different layers is strong enough to form 3D vortex lines. Vortex penetration and relaxation are governed by 3D dynamics. At the crossover field, in this case we identify it to be  $H_{\min}$ , strong in-plane interaction dominates the interlayer coupling and vortices are quasi-2D like. The almost constant value of  $H_{\min}$  at low temperature ( $T < 40$  K) demonstrates the nature of crossover. The peak effect in magnetization is due to collective pinning of 2D vortices. The disappearance of the anomalous magnetization above the crossover temperature is due to the fact that the melting field of 3D vortex solid is below the crossover field. The difference between the crossover field obtained from  $M(H)$  at low  $T$  and the 1 kG value from  $H_{\text{rev}}(T)$  may be due to the temperature-dependent  $\gamma$ . The increasing  $H_{\min}$  with increasing  $T$  suggests that  $\gamma$  decreases as  $T$  increases, contrary to a recent model in which  $\gamma$  increases with increasing  $T$ .<sup>25</sup>

A possible alternative model for the observed anomalous magnetization is that there are oxygen-deficient low- $T_c$  phases in the crystal studied, as suggested initially for the fishtail in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ .<sup>1,2</sup> In this model, the oxygen-deficient sites are driven normal under high field ( $\sim H_{c2}$ ),

thus becoming strong pinning centers. The exponential temperature dependence observed in  $\Delta M(T)$  could be consistent with a log-normal distribution of  $T_c$ . However, recent studies on high-quality single crystals<sup>8,10-12</sup> point to a more intrinsic mechanism responsible for the fishtail magnetization. In the case here, the extremely small field at the second peak compared to  $H_{c2}$  and its exponential temperature dependence rule against this possibility. If the oxygen-deficiency model were correct, one would also expect the peak field to be smaller in lower  $T_c$  samples. A recent study on a crystal with  $T_c = 34$  K shows a considerably larger peak field at the same reduced temperature,<sup>7</sup> contrary to the secondary phase model. Transport measurement on the same crystal ( $T_c = 92$  K) indicates a very high quality of this material.<sup>26</sup>

In summary, magnetic hysteresis measurements on a single crystal of  $\text{Ti}_2\text{Ba}_2\text{CuO}_6$  show an anomalous fishtail-type magnetization at low temperatures ( $T < 60$  K). At high temperatures ( $T > 60$  K), the fishtail magnetization disappears. The crossover temperature corresponds to a crossover in the temperature dependence of the irreversibility line  $H_{\text{rev}}(T)$ . Combined with the magnetic relaxation measurements, we suggest the anomalous magnetization at low  $T$  is a result of dimensional crossover in the vortex structure.

We acknowledge many useful discussions with Dr. Peter H. Kes, Dr. Myron B. Salamon, Dr. Yu Fang, Dr. Stewart Barnes, and Dr. Joshua Cohn. This work was supported by a general research grant from the University of Miami (F.Z.).

- 
- <sup>1</sup>M. Daeumling, J. M. Seuntjens, and D. C. Larbalestier, *Nature* **346**, 332 (1990); J. L. Vargas and D. C. Larbalestier, *Appl. Phys. Lett.* **60**, 1741 (1992).
- <sup>2</sup>M. S. Ososky *et al.*, *Phys. Rev. B* **45**, 4916 (1992); J. G. Ossandon *et al.*, *ibid.* **45**, 12 534 (1992).
- <sup>3</sup>V. N. Kopylov *et al.*, *Physica C* **170**, 291 (1990).
- <sup>4</sup>T. Kimura *et al.*, *Physica C* **192**, 247 (1992).
- <sup>5</sup>G. Yang *et al.*, *Phys. Rev. B* **48**, 4054 (1993).
- <sup>6</sup>T. Tamega *et al.*, *Physica C* **213**, 33 (1993); A. K. Pradhan *et al.*, *Phys. Rev. B* **49**, 12 984 (1994).
- <sup>7</sup>F. Zuo *et al.*, *Phys. Rev. B* **49**, 12 326 (1994); F. Zuo *et al.*, *J. Appl. Phys.* **76**, 6953 (1994).
- <sup>8</sup>Ming Xu *et al.*, *Phys. Rev. B* **48**, 10 630 (1993).
- <sup>9</sup>D. Neernick *et al.*, *Phys. Rev. Lett.* **67**, 2577 (1991).
- <sup>10</sup>L. Krusin-Elbaum *et al.*, *Phys. Rev. Lett.* **69**, 2280 (1992).
- <sup>11</sup>N. Chikumoto *et al.*, *Phys. Rev. Lett.* **69**, 1260 (1992).
- <sup>12</sup>A. A. Zhukov *et al.* (unpublished).
- <sup>13</sup>M. V. Feigel'man, V. B. Geshkenbein, and V. M. Vinokur, *Phys. Rev. B* **43**, 6263 (1991); M. V. Feigel'man *et al.*, *Phys. Rev. Lett.* **20**, 2303 (1989).
- <sup>14</sup>A. Schilling *et al.*, *Phys. Rev. Lett.* **71**, 1899 (1993).
- <sup>15</sup>E. H. Brandt, *Phys. Rev. Lett.* **63**, 1106 (1989); A. Houghton, R. A. Pelcovits, and A. Sudbo, *Phys. Rev. B* **40**, 6763 (1989).
- <sup>16</sup>A. L. Glazman and A. E. Koshelev, *Phys. Rev. B* **43**, 2835 (1991).
- <sup>17</sup>C. C. Almasan *et al.*, *Phys. Rev. Lett.* **69**, 3812 (1992).
- <sup>18</sup>Gianni Blatter and B. Ivlev, *Phys. Rev. Lett.* **70**, 2621 (1993).
- <sup>19</sup>R. Cubbit *et al.*, *Nature* **365**, 407 (1993).
- <sup>20</sup>S. L. Lee *et al.*, *Phys. Rev. Lett.* **71**, 3862 (1993).
- <sup>21</sup>Y. M. Wan, S. E. Hebboul, and J. C. Garland, *Phys. Rev. Lett.* **72**, 3867 (1994).
- <sup>22</sup>M. V. Feigel'man, V. B. Geshkenbein, and A. I. Larkin, *Physica C* **167**, 177 (1990); V. M. Vinokur, P. H. Kes, and A. E. Koshelev, *ibid.* **168**, 29 (1990).
- <sup>23</sup>The correct values of  $\mu$  are obtained from a private communication with Dr. P. H. Kes.
- <sup>24</sup>Thomas Nattermann, *Phys. Rev. Lett.* **64**, 2454 (1990).
- <sup>25</sup>L. L. Daemen *et al.*, *Phys. Rev. Lett.* **70**, 1167 (1993).
- <sup>26</sup>Fong Yu *et al.* (unpublished).