Vortex fluctuations and interlayer coupling in cuprates

Y. Y. Xue, Y. Cao, Q. Xiong,* F. Chen, and C. W. Chu
Department of Physics and Texas Center for Superconductivity, University of Houston, Houston, Texas 77204-5932
(Received 8 June 1995)

The reversible magnetization (M) curves of $HgBa_2Ca_{m-1}Cu_mO_{2m+2+\delta}$ ($m=1$, 2, and 3) at different H cross at a temperature T^* and $M=M^*$ as suggested by various two-dimensional (2D) vortex-fluctuation models. However, the obtained M^*/T^* was found to be independent of either m or the sheet density $1/s$ of the CuO_2 blocks, where s is the size of the unit cell along the c axis, in contrast to the predictions of the models. A survey of published data shows a similar situation, i.e., M^*/T^* in either Bi- or Ti-based compounds is independent on s. This observation suggests that the coupling between neighboring CuO_2 blocks may be non-negligible in its effects on the magnetization, and that modifications to the existing 2D fluctuation models need to be made.

It was observed[1] that all magnetization $M(T)$ curves in cuprates at different $H \gg H_{cr}$ cross at a temperature T^* and $M=M^*$, where M and H_{cr} are the reversible magnetization with H parallel to the c axis and a crossover field, respectively. Immediately after its discovery, this field-independent magnetization was taken as evidence for two-dimensional (2D) vortex fluctuations. Experimental data further demonstrated that M^* and T^* serve as fundamental scales for M and T in the fluctuation regime. The data in a Bi2:2:1:2 sample[2] show that an additional term of $-TM^*/T^*$ in the expression $\partial M/\partial \ln H=\phi_0/[32\pi^2\lambda_{ab}^2(T)]$ of the London model is needed to account for the vortex-fluctuation effects, where ϕ_0 and λ_{ab} are the flux quantum and the penetration depth in the a,b plane, respectively. The scaling fit of M in Bi2:2:2:3 (Ref. 3) demonstrates that the ratio M/M^* is the proper dimensionless variable in the proposed scaling expression. It was also observed that the specific heat of Bi2:2:1:2 at various fields peaks at T^*.[4]

These observations led to much theoretical activity. The effects of vortex-configuration entropy were considered by Bulaevskii *et al.* (the BLK model).[5] There, a vortex line is treated as a stack of 2D vortex pancakes with an average separation of s, and is modeled as 2D classical particles of size $\alpha\pi\xi_{ab}^2$, held through Josephson coupling, where ξ_{ab} and α are the coherence length in the a,b plane and a numerical parameter ~ 1, respectively. Decoupling of the pancakes occurs above a crossover field H_{cr}, and leads to an additional configuration-entropy term in the vortex free energy F, which is proportional to the density $1/s$ of the 2D pancake sheets. As a result, the derivative

$$\left.\frac{\partial M}{\partial \ln H}\right|_{B\gg B_{cr}} = -\partial F/\partial B|_{B\gg B_{cr}} = \left.\frac{\partial M}{\partial \ln H}\right|_{B\ll B_{cr}} + k_B T/(\phi_0 s)$$

is ~ 0 at a temperature $T^* \sim -\phi_0 s(\partial M/\partial \ln H)|_{B\ll B_{cr}} < T_c$, and the magnetization at T^* is $M^*=(k_B T^*/\phi_0 s)\ln(\eta\alpha/\sqrt{e})$, where η is a parameter related to the energy of the vortex cores and $e=2.718...$.[6] In the critical fluctuation region, the fluctuation of order-parameter amplitude will be the dominant factor instead of the configuration entropy. This case has been considered by Tesanovic *et al.*[3] (TXBLS model). An approximate differentiation of their proposed free energy leads to a scaling expression of $M/M^*=\frac{1}{2}[1-\tau-h+\sqrt{(1-\tau-h)^2+4h}]$ with $\tau=(T-T^*)/(T_c-T^*)$, $h=H/H_{c2}(T^*)$, and $M^*=-k_B T^*/\phi_0 s$ where k_B is the Boltzman constant. Therefore, $M=M^*$ at $\tau=0$ will again be field independent. This scaling function agrees with the experimental data reasonably well if the experimentally determined M^* and T^* are used.[3,7] However, all of the measured $-M^*/T^*$ are smaller than the predicted value of $k_B/\phi_0 s$, if the s were taken as the size of an effective unit cell in the c direction, i.e., being the lattice constant if the cell is primitive, or half of that if the cell is body centered. It was assumed in previous investigations that either the superconducting volume fraction $f \ll 1$ for nearly all of the measured samples or $\ln(\eta\alpha/e^{1/2}) \ll 1$ in the BLK model. However, the assumed impurities have never been revealed by x-ray/neutron diffraction investigations.

To explore the M^* dependence on s, we used the facts that the unit-cell size of Hg1:2:$m-1$:m increases by a factor of ~ 2 when m varies from 1 to 3, and assuming that both $\ln(\eta\alpha/e^{1/2})$ and f may not change significantly and systematically with m. In such a case, the proposed models can be meaningfully verified by a relative measurement. We will argue that the variation of f in our samples is indeed only $\pm 20\%$ or smaller based on the phase purity, the T_c distribution and, especially, the measured $n\lambda_{ab}^2$, where n is the normal-state carrier concentration. Several cross-checks also suggest that our experimental resolution in M^* is $\sim 20\%$. However, the observed M^*/T^* is nearly the same for the three compounds, in contrast with the model predictions. A survey on the published data reveals again that there is no systematic s dependence of M^*/T^* in Tl-, Bi-, and Hg-based compounds.

The samples used are ceramic disks. Hg1:2:0:1 was synthesized at ambient pressure; Hg1:2:1:2 and Hg1:2:2:3 were made in a 3 kbar piston-cylinder high-pressure cell. Details on the sample synthesis and annealing have been published before.[8] The normal-state carrier-concentration n was deduced from the thermopower $S(290\ \mathrm{K})$ at room temperature based on the proposed universal $S(290\ \mathrm{K})$-n correlation, which has been demonstrated before[9] and verified in Hg1:2:0:1 using iodometric titration and bond-valence

summation.[8] S was measured using a homemade instrument.[8] The magnetization was measured in a 5 T commercial superconducting quantum interference device magnetometer. The reversible magnetization M_{ceramic} of our ceramic samples at H_{ceramic} was converted into the magnetization M of a corresponding single crystal with $H\|c$ as $M=2M_{\text{ceramic}}$ and $H=H_{\text{ceramic}}e^{1/2}$. These expressions are the results of averaging over randomly oriented grains in highly anisotropic layer superconductors[10] and have been verified experimentally.[11] The paramagnetic background was subtracted by extrapolating a fit to the Curie-Weiss law well above T_c. The possible ferromagnetic impurities were checked by the M-H curves at a few K above T_c.

Both x-ray and neutron diffraction studies[12] show that the impurity phases in all the samples are at a few percent level. However, it has been shown that well-dispersed impurities up to 10% may escape detection of x-ray/neutron diffraction.[13] There is also an ongoing dispute about the Hg stoichiometry in Hg-based compounds: when most diffraction studies suggest a fully occupied Hg site, our chemistry analysis consistently shows a 30% Hg deficiency for all Hg1:2:$m-1$:m. Several possibilities such as Hg/Cu mixing or incorporation of CO_3^{-2} in the Hg site[14] have been proposed. In the most extreme cases, f will either be $\sim$0.70 if all the Hg deficiency is related to some invisible impurity phases, or $\sim$0.95 based on the x-ray/neutron diffraction. Scanning electron microscopy, energy-dispersive x-ray imaging, and Ramam spectrum have been used to verify the phase purity. The data suggest a full Hg occupation, and probably $f\sim0.70$ in all the three compounds. Nevertheless, $f=0.85\pm0.15$ was assumed here to cover both scenarios. It should be noted that an m-independent f, such as is suggested by the similarity in the x-ray/neutron data and the cation stoichiometry of Hg1:2:$m-1$:m, will not affect the s dependence of M^* even if $f\neq1$.

The sample homogeneity was checked by the distribution of T_c, which was measured both magnetically and resistively. The onset T_c of the optimally doped samples were 134, 127, and 97 K for $m=3$, 2, and 1, respectively. These values of T_c are consistent with the published data and suggest that there is no severe intergrowth. All samples have a sharp transition ($\Delta T_c\sim1$–2 K) as shown by the field-cooled magnetization at 5 G. The ΔT_c is smaller than the measured T_c-T^*(2–4 K) and, therefore, should not affect the obtained M^* significantly. All samples also have a flat plateau with a variation less than a few percent in the field-cooled magnetization below the transition region. It is known that the T_c of cuprates changes continuously with both n and defect density. A significantly doping unhomogeneity cannot be accommodated with this flat plateau.

To measure the relative change of f with m, we propose using the measured $n\lambda_{ab}^2(T/T_c)\propto nm_e/n_s$ at the same reduced temperature T/T_c, where m_e and n_s are the effective mass and the concentration of the Cooper pairs, respectively. It is observed[10,15] that the value of $n\lambda_{ab}^2(0)$ is the same around the optimum doping level for cuprates as different as Y1:2:3 and Tl2:2:0:1, when $\lambda_{ab}(0)$ is directly measured from the μSR depolarization rate and n is deduced from S(290 K). We chose to deduce $\lambda_{ab}^2(T)$ from the M above the irreversible line by the Hao-Clem model[6] $f/\lambda_{ab}^2=(32/\beta)\pi^2(\partial M/\partial \ln H)/\phi_0$ with $\beta\sim0.77$ and fit the data as $f/\lambda_{ab}^2=n_f[1-(T/T_c)^k]$ with both $2\leqslant k\leqslant4$ and n_f being fitting parameters. It should be noted that the actual $1/\lambda_{ab}^2$ may not follow this T dependence below $T_c/2$ and usually $1/\lambda_{ab}^2(0)>n_f$. However, it seems to be reasonable to assume that both the T dependence and the value of n_f/n are the same for all optimally doped Hg-1:2:$m-1$:m. In fact, a survey shows that n_f/n is 2.8×10^{-12} and 2.5×10^{-12} hole^{-1} cm for the well studied Y1:2:3 with $1/\sqrt{n_f}\sim1400$ Å (Ref. 16) and Bi2:1:1:2 with $1/\sqrt{n_f}\sim1700$ Å,[1,5] respectively, if the universal optimum doping level $n\sim0.16$ holes/CuO_2 (Ref. 9) is used. Although the data scattering is much larger for other Bi-, Tl-, and Hg-based cuprates around the optimum doping level, 2×10^{-12} hole^{-1} cm$\leqslant n_f/n\leqslant4\times10^{-12}$ hole^{-1} cm holds for most reported data. S(290 K), and the n deduced from it, is not sensitive to f, while $n_f\propto(\partial M/\partial \ln H)$ is proportional to it. Therefore, a change of f can be estimated based on the ratio n_f/n, although the absolute value of f has to be determined in other ways.

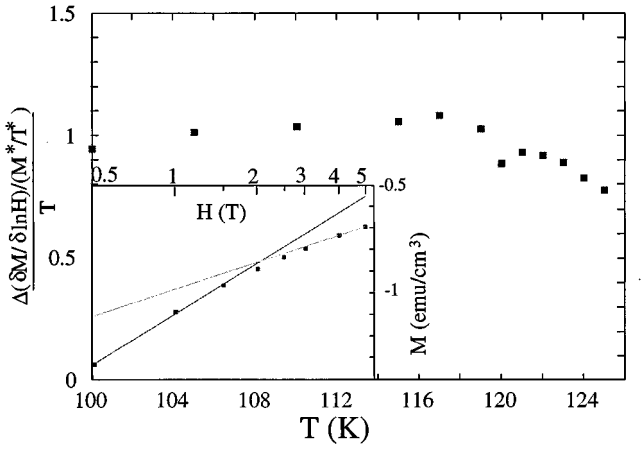

FIG. 1. $\Delta(\partial M/\partial \ln H)T^*/(TM^*)$ for an optimally doped Hg-1:2:2:3 sample with $T_c\sim133$ K. Inset: M vs H at 123 K; the lines are least-square linear fits.

The f/λ_{ab}^2 of three optimally doped Hg-1:2:$m-1$:m samples was deduced below T^*. The data can be fitted rather well into $f/\lambda_{ab}^2=n_f[1-(T/T_{c1})^k]$ above $T_c/2$ with the same $k=3.5$. The parameter n_f is 28, 38, and 52 μm^{-2} for $m=1$, 2, and 3, respectively. The corresponding n are 1.19, 1.59, and 1.86×10^{21} holes/cm^3 based on the measured S(290 K). Therefore, the ratio $n_f/n\sim(2.5\pm0.2)\times10^{-12}$ hole^{-1} cm is nearly the same for different m. A change in the fitting parameter k will increase the uncertainty in n_f. However, the conclusion that f is independent of m will not be affected within $\pm20\%$ if $2\leqslant k\leqslant4$. Therefore, we believe that the superconducting volume fraction is not a significant factor in the measured M^*/T^* vs s, even if the actual value of f deviated from 1.

A clear crossing of various $M(T)$ curves is seen in all samples. It should be noted that the position of the crossing point changes slightly with H with a spread $\sim$0.5 K. Similar behavior has been noted before.[7] To estimate our experimental resolution of M^*, the obtained M was compared with the predictions of the BLK and TXBLS models. The M vs ln$H$ plot at 123 K for an optimally doped Hg1:2:2:3 sample with $T_c=133$ K is shown in Fig. 1 (inset). A change of slope can

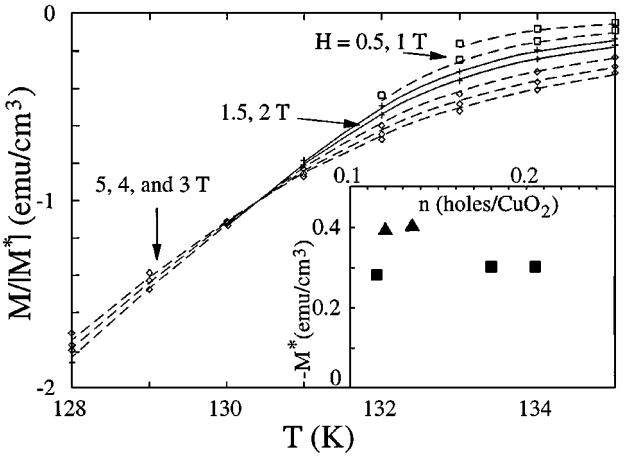

FIG. 2. 2D-scaling fit for the same sample above $H_{c2}/3$, with $T_c=133.6$ K and $dH_{c2}/dT=2.1$ T/K; symbols: data; lines: fits. Inset: $-2M^*_{\text{ceramic}}$ vs n for ■: Hg-1:2:0:1 and △: Hg-1:2:2:3.

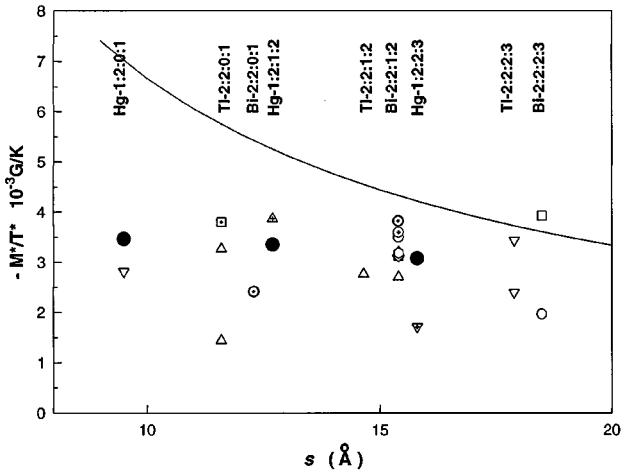

FIG. 3. M^*/T^* vs s. ●: This work; ⬭: Ref. 1; ☆: Ref. 5; ○: Ref. 7; □: Ref. 10; ▽: Ref. 11; △: Ref. 18; ⊗: Ref. 22; ⊞: Ref. 23; ⧋: Ref. 24; ⧩: Ref. 25; ⊙: Ref. 26; –: calculated from the models of Refs. 3 and 5.

be seen around a $B_{\text{cr}}\sim 2$ T. This slope change would correspond to a 2D-3D crossover in the BLK model, and

$$\Delta(\partial M/\partial\ \ln H)=\left.\frac{\partial M}{\partial\ \ln H}\right|_{B\gg B_{\text{cr}}}-\left.\frac{\partial M}{\partial\ \ln H}\right|_{B\gg B_{\text{cr}}}=TM^*/T^*$$

is predicted. The measured

$$\Delta(\partial M/\partial\ \ln H)T^*/(TM^*)=\left[\left.\frac{\partial M}{\partial\ \ln H}\right|_{B\sim 5\ \text{T}}-\left.\frac{\partial M}{\partial\ \ln H}\right|_{B\sim 0.5\ \text{T}}\right]T^*/(TM^*)$$

is shown in Fig. 1. Over a broad temperature region from 100 to 120 K, the ratio is $\sim 1\pm 0.1$ as required by the BLK model. This ratio drops with further increase of T, probably due to the fact that the lowest field used (0.5 T) is not low enough above 120 K. Similar results were observed for all other samples with the $\Delta(\partial M/\partial\ln H)T^*/(TM^*)$ varying from 0.8 to 1.1. This ratio, which compares the directly measured M^* and the M^* deduced from the slope change, is a cross-check of our data acquisition/deduction procedure. The self-consistency suggests that our experimental resolution in M^* is ±20% or better.

The scaling expression in the TXBLS model was also used to fit the data above $H_{c2}(T)/3$. The data (symbols) and the scaling fit (lines) are shown in Fig. 2 for the optimally doped Hg1:2:2:3 sample. The agreement is good and the used parameters ($T_c=133.6$ and $dH_{c2}/dT=2.1$ T/K) are reasonable. Such a fit is rather sensitive to the background subtraction, and the good agreement further demonstrates the reliability of our measurement.

The same measurement was carried out for several Hg1:2:0:1 and Hg1:2:2:3 samples at different doping levels. The obtained M^* is shown in Fig. 2 (inset) as the function of n in the units of holes/CuO_2. Within our experimental resolution, M^* is independent of doping, again in agreement with the fluctuation models.

The measured $-M^*/T^*$ is shown in Fig. 3 as a function of s for Hg-1:2:m−1:m with $1\leqslant m\leqslant 3$. To our surprise, the proposed linear dependence $-M^*/T^*\propto 1/s$ does not exist. Instead, $-M^*/T^*\sim(3.2\pm 0.2)\times 10^{-3}$ G/K^0 appears to be independent of both m and s. This is in strong contrast with the expectation based on the BLK/TXBLS models.

To further verify the observed s independence of M^*, a survey of the published data on Bi-, Tl-, and Hg-based cuprates was made. Although our relative measurement of M^*/T^* vs m should be valid even if $f\ll 1$, the absolute value of f is important when data from different groups are compared. A different approach has been proposed: that the vortex fluctuations follow the BLK/TXBLS models exactly, therefore, $f=-M^*/[k_BT^*/(\phi_0 s)]$. This reasoning led to a much smaller $f=0.41$ for a Hg1:2:0:1 sample with $T_c\sim 95$ K.[11] The small Meissner signal of this sample was used to support this approach.[11] However, the $\lambda_{ab}(T)\propto(\partial M/\partial\ln H/f\ )^{-1/2}$, or the n_f so obtained, will be unusually small compared with both the λ_{ab} measured by μSR in a Hg1:2:0:1 sample with a similar $T_c\sim 97$ K (Ref. 18) and the deduced n_f in other optimally doped cuprates with comparable T_c. For example, the obtained $\lambda_{ab}(T)$ at 60 K is ~1400 Å in Ref. 11, but is ~1900 Å ($\sigma\sim 2.0\ \mu\text{s}^{-1}$) from the μSR measurement.[18] Similarly, the extrapolated $1/\sqrt{n_f}$ is ~1170 Å in Ref. 11, too small compared with those of ~1400–1500 Å in Y1:2:3,[16] 1500–1700 Å in Bi2:2:1:2,[1,2,5] and 1300 Å in Hg1:2:2:3.[17] The observation suggests that the actual f may be closer to 1, and the small Meissner signal might only be the result of vortex pinning. Therefore, raw data in the literature were directly quoted based on the claimed high phase purity in the used samples. One exception is the Tl2:2:2:3 samples in Ref. 11, where the sample phase purity is only 65% as determined by x-ray diffraction. Despite a moderate spread, which is expected without a case-by-case study, the collected $-M^*/T^*$ appears again to be s independent (Fig. 3). A least-square fit leads to $-M^*/T^*=(3.18\pm 0.7)\times 10^{-3}+[1\times 10^{-3}/s(\text{Å})]$ G/K. The obtained slope of 10^{-3} G/K Å and its statistical uncertainty ($\pm 6\times 10^{-3}$ G/K Å) are insignificant (leads to a variation smaller than $\sim 10^{-4}$ G/K from Hg1:2:0:1 to Bi2:2:2:3) as compared to the s-independent term of $\sim 3\times 10^{-3}$ G/K. The actual trend is expected to be more clear since some of the exceptional data points (all of which have

a smaller $-M^*/T^*$ at $1–2\times10^{-3}$ G/K level) might have true phase-purity problems as suggested by their unusually small $1/\lambda_{ab}^2$ and synthesis methods.

We would argue that the data scattering is relatively insignificant compared with the expected s dependence. In fact, all published data in the well studied Bi2:2:1:2 are within a $\pm20\%$ band (Fig. 3). All one-CuO_2-layer cuprates have a measured $-M^*/T^*$ between 2.5×10^{-3} and 4×10^{-3} G/K except one, a Tl2:2:0:1 single crystal in Ref. 19, which has an even lower M^*/T^* (Fig. 3). The average data spreading ($\pm7\times10^{-4}$ G/K^0) is much smaller than the expected increase of M^*/T^* ($>2\times10^{-3}$ G/K) caused by the s difference between the one-CuO_2-layer compounds and the bi-CuO_2-layer compounds. The related n_f/n was also deduced when the data were available, with no systematic s dependence being observed. Therefore, it would be difficult to attribute the discrepancy to either accidental data scattering or change of f, which would require $f<0.5$ for all measured one-CuO_2-layer cuprates but $f\sim1$ in most tri-CuO_2-layer compounds.

The linear relationship between M^* and the pancake-sheet density $1/s$ should be more or less model independent in the 2D scenario.[1,3,4] Both the vortex entropy and the M^* caused by it should depend on the pancake sheet density, although the choice of s is strongly dependent on our understanding of the interlayer coupling. In all of the above investigations, s was taken as the size of an effective unit cell in the c direction. In other words, it is assumed that the coupling across the charge reservoir is negligible, but the coupling between adjacent CuO_2 layers is as strong as the intralayer coupling. This assumption, although supported by many experimental observations, seems to be in contrast with the observed M^*/T^* vs s.

A possible alternative is to assume that the vortex decoupling occurs between all adjacent CuO_2 layers, i.e., the sheet density$=m/s$. However, this possibility is not supported by the data either. A two times variation in $-M^*/T^*$ will again be expected among the compounds listed in Fig. 3, with the average sheet separation $s/m=11.6$ and 6.2 Å for Tl2:2:0:1 and Bi2:2:2:3, respectively.

Another possibility is an accidental correlation of $\ln(\eta\alpha/e^{1/2})=s$ in the BLK model of $-M^*=k_BT^*\ln(\eta\alpha/e^{1/2})/(\phi_0 s)$. This would require an unusual threefold increase in the parameter α, which characterizes the size of the normal core, when s decreases from ~18 to 9 Å. In addition, $M(H)$ is H independent also in the critical region ($>H_{c2}/3$), where the configuration entropy and α do not play significant roles.[5,3]

Our observations suggest that either the true nature of the vortex fluctuation in cuprates or the concept of vortex decoupling between adjacent CuO_2 blocks around T^* need to be reconsidered. There is a possibility that the vortex coupling across the charge reservoir cannot be ignored in the analysis of magnetization. In fact, a similar crossing with a smaller $-M^*/T^*\sim1\times10^{-3}$ G/K has been observed in Y1:2:3,[19,20] even though data[21] suggest the fluctuation in this compound is 3D.

In summary, a $1/s$ independent M^*/T^* have been observed in the cuprates with different charge reservoirs and different unit-cell sizes along the c axis. The observation calls for modifications to the proposed 2D vortex-fluctuation models.

The authors thank Professor L. Vant-Hull, Dr. J. Clayhold, Dr. Z. D. Hao, and Dr. D. M. Frenkel for many helpful discussions. This work is supported in part by NSF Grant No. DMR 91-22043, USAFOSR Grant No. F49620-93-1-0310 by BMDO, EPRI RP-8066-04, the State of Texas through the Texas Center for Superconductivity at the University of Houston, and the T. L. L. Temple Foundation.

*Present address: Physics Department, University of Arkansas, Fayetteville, AR 72701.

[1] P. H. Kes *et al.*, Phys. Rev. Lett. **67**, 2383 (1991).
[2] V. G. Kogan *et al.*, Phys. Rev. Lett. **70**, 1870 (1993).
[3] Z. Tesanovic *et al.*, Phys. Rev. Lett. **69**, 3563 (1992).
[4] A. Junod *et al.*, Physica C **229**, 209 (1994).
[5] L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).
[6] Z. Hao and J. R. Clem, Phys. Rev. Lett. **67**, 2371 (1991).
[7] Q. Li *et al.*, Phys. Rev. B **46**, 3195 (1992); Q. Li *et al.*, Phys. Rev. **46**, 5857 (1992); Q. Li *et al.*, Phys. Rev. B **48**, 9877 (1993).
[8] Q. Xiong *et al.*, Phys. Rev. B **50**, 10 346 (1994); Q. Xiong *et al.*, Physica C **231**, 233 (1994); Z. H. He *et al.*, Physica C (to be published).
[9] S. D. Obertelli *et al.*, Phys. Rev. B **46**, 14 928 (1992).
[10] J. H. Cho *et al.*, Physica C **212**, 419 (1993).
[11] J. R. Thompson *et al.*, Phys. Rev. B **48**, 14 031 (1993).
[12] Q. Huang *et al.* (unpublished).
[13] L. E. Dolhert and N. D. Spencer, Meter. Lett. **9**, 537 (1990).
[14] M. Marezio *et al.* (unpublished).
[15] Ch. Niedermayer *et al.*, Phys. Rev. Lett. **72**, 2502 (1994).
[16] For example, L. K. Krusin-Elbaum *et al.*, Phys. Rev. Lett. **62**, 217 (1989).
[17] A. Schilling *et al.*, in *Proceedings of M^{2}S HTSC IV*, Grenoble, 1994 (Elsevier Science, New York, 1994).
[18] Y. J. Uemura *et al.* (unpublished).
[19] G. Triscone *et al.*, Physica C **224**, 263 (1994).
[20] U. Welp *et al.*, Phys. Rev. Lett. **67**, 3180 (1991).
[21] For example, S. Kamal *et al.*, Phys. Rev. Lett. **73**, 1845 (1994).
[22] K. Kadowaki, Physica C **185-189**, 2249 (1991).
[23] F. Zuo *et al.*, Phys. Rev. B **47**, 8327 (1993).
[24] Z. J. Huang *et al.*, Physica C **228**, 211 (1994).
[25] M.-K. Bae *et al.*, Physica C **228**, 195 (1994).
[26] R. Jin, H. R. Ott, and A. Schilling, Physica C **235-240**, 1923 (1994).