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Discontinuity in the low-field magnetization of single-crystal $Tl_2Ba_2CuO_6$ with $H\|ab$

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The zero-field-cooled magnetization $M(H,T)$ of a single crystal of $Tl_2Ba_2CuO_6$ (T_c=82.5 K) has been measured with the field applied parallel to the CuO_2 planes. For field sweeps carried out below T=50 K and temperature sweeps carried out in applied fields $H \geqslant 25$ Oe, a large jump in magnetization is observed at $H=H_d$, close to the field of first flux penetration. The data provide strong evidence that $Tl_2Ba_2CuO_6$ behaves as a weak-Josephson-coupled layered superconductor, the discontinuity arising from the destruction of Josephson screening currents between the planes at a field close to the lower critical field H_{c1}. The value of H_d is an order of magnitude lower than the predicted surface-barrier penetration field.

Because of their layered structure and large anisotropy, it has been commonly proposed that high-T_c superconductors can be viewed as a stack of CuO_2 planes weakly coupled along the c axis by the Josephson interaction. When a magnetic field is applied parallel to the superconducting layers, unconventional behavior of the magnetization is predicted to occur[1,2] due to the weak screening effects of the Josephson currents between the planes. Above H_{c1}, the Josephson screening currents are destroyed and magnetic flux can penetrate the sample completely between the ab planes, thus creating a so-called "magnetically transparent" state. Experimental evidence for such a state has been reported[3] recently in single crystals of the electron-doped compound $Nd_{2-x}Ce_xCuO_4$ (NCCO) where the magnetization along the ab planes collapsed to zero at, or close to, the field of first flux penetration. We have performed detailed measurements of $M(H,T)$ in this field orientation on a $Tl_2Ba_2CuO_6$ (Tl-2201) single crystal and report a similar large discontinuity in magnetization to that observed in NCCO. The field at which the discontinuity occurs agrees well with the theoretical estimate of H_{c1} for a Josephson-coupled layered system with $H\|ab$ (Ref. 4) but is an order of magnitude lower than the expected surface-barrier penetration field.[5] This result suggests that surface barriers play a minor role for high-T_c cuprates in this field orientation.

A batch of Tl-2201 single crystals was grown by a solid-state self-flux method similar to that described elsewhere.[6] The data reported here are for a $600\times600\times10\ \mu m^3$ crystal which was annealed under 5% hydrogen in argon at 690 K for two days to obtain the high T_c. The superconducting transition was sharp with an onset at 82.5 K and a transition width (10–90 %) $\Delta T=2$ K with $H=2$ Oe. The magnetization measurements were performed using a Cryogenic Consultants low-field superconducting quantum interference device susceptometer with a scan length of 5 cm. The crystal was zero-field-cooled ($\leqslant 0.02$ Oe) through the transition to a set temperature, then either the field or temperature were swept with the magnetic field applied parallel to the ab plane. After every field or temperature sweep, the sample was warmed through the transition, then recooled in zero field. Several sweeps were also performed with $H\|c$ to determine the anisotropy of the penetration fields. The demagnetization factor was negligible for $H\|ab$ and for $H\|c$ was calculated from the initial slope of the $M(H)$ curves to be $1/(1-N_c)=15$.

Figure 1(a) shows two $M(H)$ curves obtained at 7 K with $H\|ab$ using forward and reverse field sweeps from 0 to 100 Oe, with sweep 2 following immediately after sweep 1. On the first sweep, full screening persists until H_d=60 Oe, at which point there is a sharp drop in M, signaling a large, spontaneous penetration of flux. The jump is first order to within 0.1 Oe (the resolution of the susceptometer) and the size of the discontinuity is equivalent to 60% of the external flux density. As the field is increased further, the magnetization slowly decreases. The absence of a further rise in $M(H)$

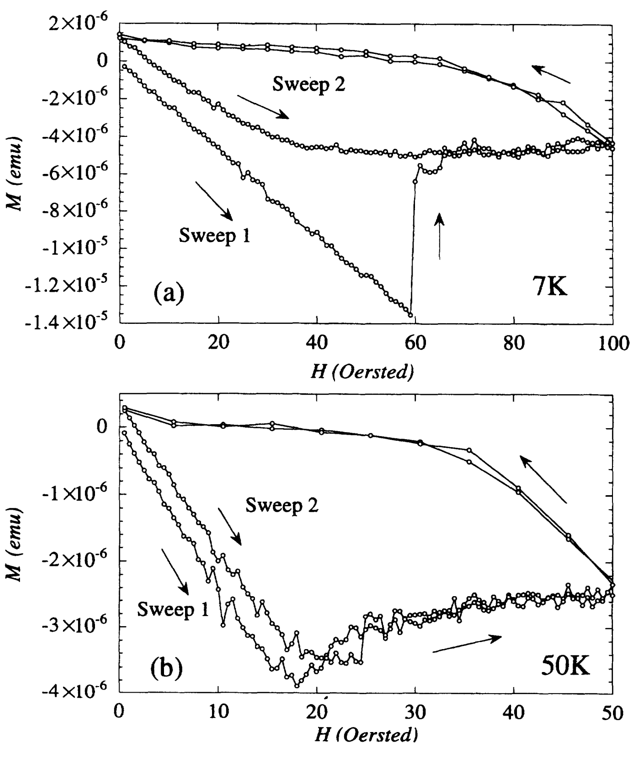

FIG. 1. (a) Magnetization versus field at 7 K, sweep 2 following immediately after sweep 1. (b) Magnetization versus field at 50 K.

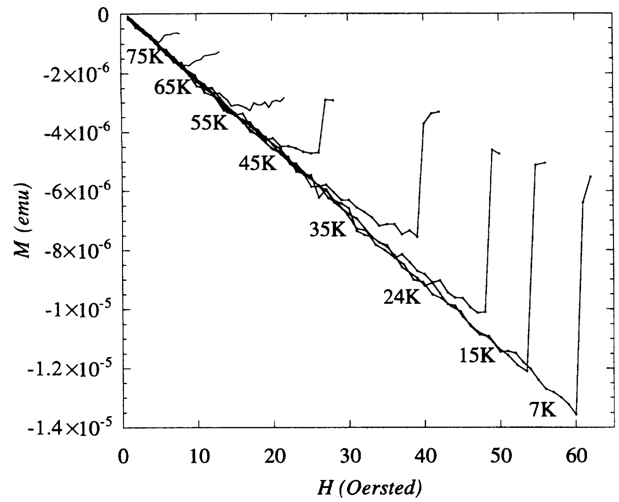

FIG. 2. Initial field sweeps (sweep 1) at 7, 15, 24, 35, 45, 55, 65, and 75 K. The data are included just beyond the deviation from the Meissner state for clarity.

beyond H_d precludes any contribution from transverse magnetization $M_\perp$, due to a slight misalignment of the ab planes with respect to the applied field.[3,7,8] When the crystal was purposely tilted in the magnetic field, the discontinuity was still observed, though M began to rise for $H \geqslant H_d$, showing the expected contribution from $M_\perp$. The reverse leg of sweep 1 shows typical hysteretic behavior and varies smoothly with decreasing field with a small positive offset at 0 Oe. The flux density on the forward leg of the second sweep varies as H^2 for the entire field range measured, in agreement with Bean critical-state behavior[9] for hard type-II superconductors and coincides exactly with sweep 1 for $H \geqslant H_d$. There was no Meissner region on the second sweep. The decreasing leg of sweep 2 also coincides with that of the first. Subsequent sweeps coincided perfectly with sweep 2. Figure 1(b) shows a similar field sweep at 50 K, this time up to 50 Oe. The discontinuity is now absent, though there is still a sharp signature of increased flux penetration on the initial sweep at around 18 Oe. Again, the two curves coincide beyond this critical point and on the descending branch of each sweep. Unlike the 7 K data, however, the initial magnetization curves for both sweeps are parallel for $H \leqslant 18$ Oe. To illustrate the evolution of the discontinuity with temperature, the low-field $M(H)$ data for all initial field sweeps (i.e., sweep 1 only) are plotted in Fig. 2. As T increases, H_d decreases and the size of the discontinuity shrinks until eventually it vanishes around $T=45$ K. For 15 K$\leqslant T \leqslant$45 K, a small deviation from linearity develops at $H=H_a$ below the discontinuity (most probably due to edge effects or thermal activation).

Constant-field temperature sweeps were also taken for a range of fields up to 65 Oe. Figure 3(a) shows the susceptibility data $[\chi(T)=M(T)/H]$ for a temperature sweep carried out at 20 Oe. As T is increased from 7 K, the signal remains constant corresponding to full flux exclusion $[\chi=\chi(0)]$, then at 30 K, vortices start to penetrate slowly into the sample (due to penetration at the edges of the crystal). At $T=T_a$, $\chi(T)$ increases sharply and linearly with temperature up to $T=T_b$, at which point it rises more slowly to zero as T approaches T_c. The behavior of $\chi(T)$ at $H=20$ Oe [Fig. 3(a)] is very similar to that observed[10,11] in $YBa_2Cu_3O_7$ crystals with $H \| ab$ and can be explained for this system within an extended Bean critical-state model.[10] In this model, T_a corresponds to the temperature at which H_{c1} becomes lower than the applied magnetic field. M increases linearly with temperature above T_a as flux fronts advance from the edges of the crystal and the critical current J_c and the slope dB/dx gradually decrease as T increases. Finally at T_b, the flux fronts meet at the center and at higher temperatures, the flux profile flattens out, giving a more gradual though still linear increase of M to zero as T approaches T_c.

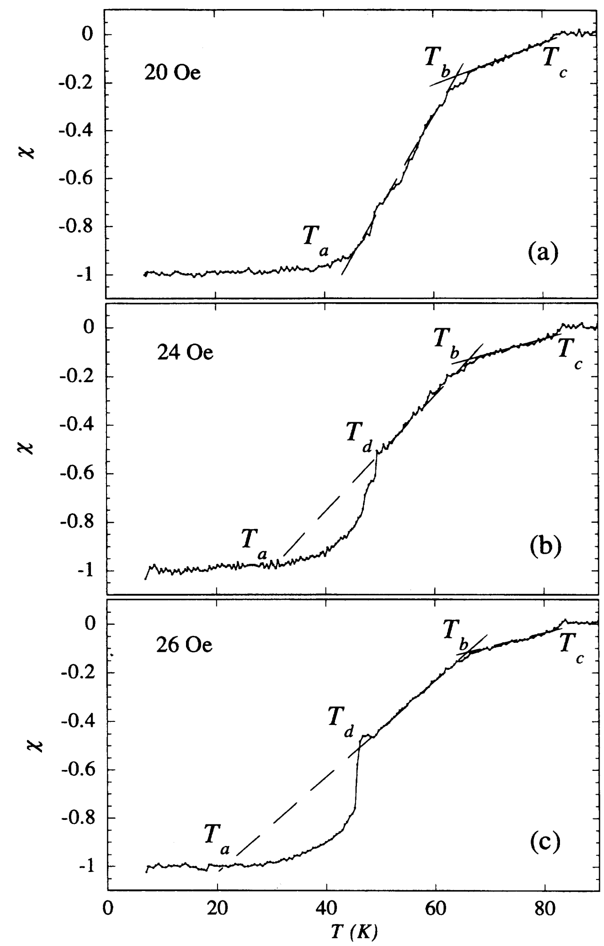

FIG. 3. $\chi(T)$ for $H=$(a) 20, (b) 24, and (c) 26 Oe. The temperatures T_a, T_b, T_c, and T_d are explained in the text.

At higher fields, a new feature appears in $\chi(T)$, as shown in Fig. 3(b) for $H=24$ Oe. Unlike the linear variation from T_a to T_b seen in Fig. 3(a), χ initially increases at a much quicker rate and only starts the linear-T variation at a higher temperature T_d. The behavior of $\chi(T)$ from T_d to T_c is then qualitatively similar to that seen for $H \leqslant 20$ Oe. If we extrapolate the linear variation observed from T_b to T_d down to $\chi(0)$, the intercept can be considered as the value of T_a at

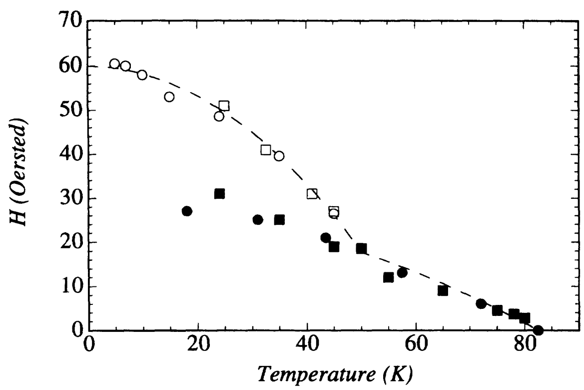

FIG. 4. Values for $H_a(T)$ (closed) and $H_d(T)$ (open) derived from $M(H)$ (circles) and $M(T)$ (squares) sweeps. The line is provided as a guide only.

$H=24$ Oe, as defined in Fig. 3(a). Thus it appears that a new screening process becomes effective at higher H and lower T which prevents flux from entering the sample at $T=T_a$. At still higher fields, this low-T feature becomes more pronounced, as shown in Fig. 3(c) for $H=26$ Oe, and χ has a discontinuity at $T_d=45$ K, in quantitative agreement with the $M(H)$ data where $T=45$ K is the highest temperature at which the discontinuity is observed. As the field is increased above $H=26$ Oe, the data show essentially the same features as in Fig. 3(c). Above $H=60$ Oe, the discontinuity is no longer observed, again in agreement with the field sweep data.

The various values for $T_a(H)$ and $T_d(H)$ have been plotted in Fig. 4 together with values for $H_d(T)$ and $H_a(T)$ obtained from the $M(H)$ data, where H_a is defined as the field at which the magnetization deviates from linearity (Fig. 2). It is apparent from the combined $M(H,T)$ data that there are two distinct regions of the $H(T)$ phase diagram which correspond to different modes of flux penetration into the sample. At low temperatures [in the $M(H)$ sweeps] and at high fields [in the $M(T)$ sweeps], bulk flux penetration occurs discontinuously (neglecting edge effects). At high temperatures and low fields, the behavior is reminiscent of critical state behavior of strongly-pinned type-II superconductors. Furthermore, critical state behavior is always observed for $H \geqslant H_d$.

In the previous reports,[3,7,8] the field of first flux penetration was assumed to be governed by Bean-Livingston type surface-barrier effects which accounted for the almost-zero magnetization in the reverse sweep of the hysteresis curve. In type-II superconductors,[12] first flux penetration may occur at a field higher than H_{c1} due to the interaction of vortices with their mirror image at the specimen surface. The surface-barrier penetration field derived for Josephson-coupled superconductors with $H \| ab$ is[5]

$$H_{\text{pen}}=\phi_0/4\pi\lambda_{ab}\gamma s, \tag{1}$$

where ϕ_0 is the flux quantum, λ_{ab} the penetration depth for currents flowing in the ab planes, γ the anisotropy, and s the interlayer spacing, approximately 12 Å. This expression generally gives values of γ much greater than those found in resistivity measurements. For NCCO,[3] Zuo *et al.* obtained $\gamma \approx 400$ from Eq. (1), whilst the resistivity anisotropy in NCCO has been found[13] to be $\rho_c/\rho_{ab} \leqslant 1000$, leading to a value for $\gamma=\sqrt{\rho_c/\rho_{ab}} \leqslant 30$. From measurements of the penetration fields in the two field directions (i.e., $H \| ab$ and $H \| c$), we found $\gamma=25-30$ for the Tl-2201 sample, in good agreement with values of γ obtained from resistivity measurements[14,15] and from the anisotropy in H_{c2}.[16,17] Using Eq. (1), this value of γ would give a derived value of $H_{\text{pen}} \approx 400$ Oe, an order of magnitude higher than observed. Furthermore, if Bean-Livingston surface barriers are important in this case, H_{pen} should be of the order $H_c(\xi_c/d)$, where ξ_c is the coherence length along the c axis.[5] From specific-heat measurements,[18] $H_c \approx 6000$ Oe for nearly-optimum doped Tl-2201, and given $H_{\text{pen}} \approx 60$ Oe, we then obtain an unphysically small value of $\xi_c \approx 0.1$ Å.

Thus, in both NCCO and Tl-2201, the penetration field appears to be too small to be accounted for by surface-barrier effects. However, it is thought that H_{pen} may be strongly reduced from its value given in Eq. (1) in the presence of surface defects. We carried out $M(H)$ measurements on several other Tl-2201 crystals and for each sample, first flux penetration was manifested as either a discontinuity or a sharp peak in $M(H)$ at 70 Oe$\pm$30 Oe at $T=10$ K. These values agree well with the value of H_d for the sample reported here and with the data of Zuo *et al.*[7] for other Tl-2201 samples. Thus it appears more probable that the penetration field is an intrinsic parameter of the system, rather than a defect-controlled surface-barrier effect.

Since the discontinuity occurs at the field of first flux penetration (at low temperatures), it is tempting to suggest that H_d corresponds to the lower critical field of the crystal. In conventional Abrikosov theory, neglecting intervortex interactions, $M(H) \propto [\ln(H_{c1}/(H-H_{c1}))]^{-2}$ for $H \geqslant H_{c1}$, in complete contrast to the large jump observed here. Theoretical predictions[1,2] have been made, however, for such unconventional behavior for Josephson-coupled layered superconductors when the field is applied parallel to the ab planes. Above H_{c1}, the Josephson screening currents are destroyed and magnetic flux can penetrate the sample completely. Zuo *et al.*[3] reported evidence of such a transparent state in two single crystals of NCCO with $H \| ab$, in which the magnetization was zero for the entire reverse field sweep beyond the discontinuity at H_d. Other measurements of $M(H)$ in this field orientation on $Bi_2Sr_2CaCu_2O_8$ (Bi-2212) (Ref. 8) and Tl-2201 (Ref. 7) have shown peaks in the magnetization at low fields but no discontinuity. The magnetization behavior observed here confirms that a similar process of flux penetration is taking place in the electron- and hole-doped single-layered compounds and strongly suggests that the peak in $M(H)$ observed in Bi-2212 is of similar origin. Indeed, intrinsic Josephson effects have been measured directly for Bi-2212.[19]

The lower critical field H_{c1} for a Josephson-coupled layered superconductor with $H \| ab$ is[4]

$$H_{c1}=\phi_0/4\pi\lambda_{ab}\lambda_c[\ln(\lambda_{ab}/d)+1.12]. \tag{2}$$

Using $\gamma=25-30$ and $H_{c1}=H_d=60$ Oe, we obtain estimates for the penetration depths, $\lambda_{ab}(0)=720\pm30$ Å and $\lambda_c(0)=2.00\pm0.15$ μm. Given $H_c(0) \approx 6000$ Oe,[18] we are also able to determine values for the Ginzburg-Landau parameters $\kappa_{ab}=2\sqrt{2}\pi H_c\lambda_{ab}\lambda_c/\phi_0\approx 380$ and $\kappa_c=2\sqrt{2}\pi H_c\lambda_{ab}\lambda_{ab}/\phi_0\approx 14$. Finally, we obtain values for the coherence lengths $\xi_{ab}\approx 50$ Å and $\xi_c\approx 2$ Å. Note that ξ_c is less than the interlayer spacing as expected for the layered cuprates.

Using our assumption that H_d corresponds to H_{c1} for this system, one can explain the field-dependent penetration of vortices as follows. At low temperatures, the Josephson currents between the planes provide perfect screening until $H_d=H_{c1}$. Then, at H_d, the Josephson currents are quickly destroyed and the vortices penetrate the sample freely until their separation is of the order of the equilibrium penetration depths, i.e., λ_c along the layers and λ_{ab} across them, at which point intervortex interactions becomes significant. Once bulk penetration has taken place, the motion of the vortices within the sample is then governed by critical state behavior, though the origin of the pinning is as yet unknown. The discontinuity is not observed on the second field sweeps since vortices are already present within the junctions (shown by the small offset of 0 Oe after the initial field sweep) and thus the Josephson screening currents between the layers are suppressed. This also explains the lack of a Meissner region on the forward leg of the second sweep. The magnetization on the reverse sweeps is suppressed because pinned flux in the crystal suppresses the Josephson currents. Assuming that, just above H_{c1}, the vortices form a regular triangular flux lattice, defined by the centers of closely-packed ellipses with major and minor axes, λ_c and λ_{ab}, respectively, we can calculate the size of the flux jump ΔB expected at $H=H_d$. This is given by $\Delta B=\phi_0/2\sqrt{3}\lambda_c\lambda_{ab}$, i.e., 42 Oe for $\lambda_{ab}(0)=720$ Å and $\lambda_c(0)=2.0$ μm. This is in excellent agreement with the measured value of 36 Oe (60% of $H_d=60$ Oe) and provides confirmation of the physical picture described above.

The disappearance of the discontinuity at higher temperatures may also be explained in terms of the Josephson effect. It is well known that thermal fluctuations can destroy the phase coherence across a single junction, and thus the Josephson currents, once the temperature becomes comparable to the Josephson coupling energy, i.e., $k_BT_J\approx\hbar I_0/2e$ where I_0 is the Josephson current. For temperatures $T\geqslant T_J$, the Josephson effect is no longer observed. Indeed, as is shown in Fig. 1(b), at temperatures above 45 K (when the discontinuity is absent), screening currents are as effective on the second forward sweep as on the primary sweep, suggesting that the effect caused by the Josephson interaction has been suppressed. The data of Zuo *et al.*[3] also showed evidence for the thermal smearing of the Josephson interaction below T_c.

In conclusion, we have investigated the low-field magnetization $M(H,T)$ of a single crystal of Tl-2201 and observed a large discontinuity close to the field of first flux penetration. There is strong evidence that this discontinuity arises from the destruction of Josephson screening currents between the CuO_2 planes at the lower critical field of the multilayer system.[4] Vortices penetrate the sample along the layers to form a periodic lattice whose dynamics are then governed by critical state behavior. In the presence of trapped flux or at higher temperatures, it seems that the Josephson currents are suppressed and more conventional (irreversible) $M(H)$ curves, with lower values of H_{c1}, are obtained. This provides further evidence that high-T_c cuprates can be described as Josephson-coupled layered superconductors. The peaks observed in the low-field magnetization of other high-T_c samples[3,8] may correspond to a range of critical fields due to a spread of Josephson coupling strengths between the layers. Finally, the value of the penetration field is an order of magnitude lower than the expected Bean-Livingston surface-barrier penetration field and may reflect a minor role for surface barriers in a Josephson-coupled layered superconductor when the field is aligned along the layers.

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[1] P. H. Kes, J. Aarts, V. M. Vinokur, and C. J. van der Beek, Phys. Rev. Lett. **64**, 1063 (1990).

[2] S. Theodorakis, Phys. Rev. B **42**, 10 172 (1990).

[3] F. Zuo, S. Khizroev, Xiuguang Jiang, J. L. Peng, and R. L. Greene, Phys. Rev. Lett. **72**, 1746 (1994).

[4] J. R. Clem, M. W. Coffey, and Z. Hao, Phys. Rev. B **44**, 2732 (1991).

[5] A. Buzdin and D. Feinberg, Phys. Lett. A **165**, 281 (1992).

[6] R. S. Liu, S. D. Hughes, R. J. Angel, T. P. Hackwell, A. P. Mackenzie, and P. P. Edwards, Physica C **198**, 203 (1992).

[7] F. Zuo, S. Khizroev, S. Voss, and A. M. Hermann, Phys. Rev. B **49**, 9252 (1994).

[8] N. Nakamura, G. D. Gu, and N. Koshizuka, Phys. Rev. Lett. **71**, 915 (1993).

[9] C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962).

[10] L. Krusin-Elbaum, A. P. Malozemoff, D. C. Cronemeyer, F. Holtzberg, J. R. Clem, and Z. Hao, J. Appl. Phys. **67**, 4670 (1990).

[11] L. Krusin-Elbaum, A. P. Malozemoff, Y. Yeshurun, D. C. Cronemeyer, and F. Holtzberg, Phys. Rev. B **39**, 2936 (1989).

[12] C. P. Bean and J. D. Livingston, Phys. Rev. Lett. **12**, 14 (1964).

[13] M. A. Crusellas, J. Fontcuberta, S. Piol, T. Grenet, and J. Beille, Physica C **180**, 313 (1991).

[14] H. M. Duan, R. M. Yandrofski, T. S. Kaplan, B. Dlugosch, J. H. Wang, and A. M. Hermann, Physica C **185-189**, 1283 (1991).

[15] T. Manako, Y. Kubo, and Y. Shimakawa, Phys. Rev. B **46**, 11 019 (1992).

[16] A. Carrington, A. P. Mackenzie, D. C. Sinclair, and J. R. Cooper, Phys. Rev. B **49**, 13 243 (1994).

[17] M. Hasegawa, Y. Matsushita, Y. Iye, F. Sakai, and H. Takei, in *Proceedings of M^2S-HTSC-IV Conference*, special issue of Physica C (to be published).

[18] J. M. Wade and J. W. Loram (private communication).

[19] R. Kleiner and P. Müller, Phys. Rev. B **49**, 1327 (1994).