Superfluid Density in a Highly Underdoped YBa$_2$Cu$_3$O$_{6+y}$ Superconductor

D. M. Bruun, W. A. Huttema, P. J. Turner, S. Özcan, B. Morgan, Ruiying Liang, W. N. Hardy, and D. A. Bonn

1Department of Physics, Simon Fraser University, Burnaby, BC, V5A 1S6, Canada
2Cavendish Laboratory, Madingley Road, Cambridge, CB3 0HE, United Kingdom
3Department of Physics and Astronomy, University of British Columbia, Vancouver, BC, V6T 1Z1, Canada

(Dated: October 31, 2018)

The superfluid density $\rho_s(T)$ is measured at 2.64 GHz in highly underdoped YBa$_2$Cu$_3$O$_{6+y}$, at 37 dopings with $T_c$ between 3 K and 17 K. Within limits set by the transition width $\Delta T_c \approx 0.4$ K, $\rho_s(T)$ shows no evidence of critical fluctuations as $T \to T_c$ with a mean-field-like transition and no indication of vortex unbinding. Instead, we propose that $\rho_s$ displays the behaviour expected for a quantum phase transition in the $(3+1)$-dimensional $XY$ universality class, with $\rho_s \propto (p-p_c), T_c \propto (p-p_c)^{1/2}$ and $\rho_s(T) \propto (T_c - T)^{1}$ as $T \to T_c$.

PACS numbers: 74.72.Bk, 74.25.Nf, 74.25.Bt, 74.25.Ha

Current research on high temperature superconductivity focuses on the underdoped cuprates, in a region of the phase diagram where $d$-wave superconductivity gives way to antiferromagnetism [1]. One proposal for this regime is that at temperatures up to about 100 K above $T_c$, superconductivity persists locally, with long-range phase coherence suppressed by fluctuations in the phase of the superconducting order parameter [2] [3] [4] [5] [6] [7] [8]. Early results showing a linear relation between $T_c$ and the superfluid density $\rho_s(T = 0)$ [9] provided the original motivation for this point of view, suggesting that $T_c$ is low in underdoped materials because the phase stiffness is low. Further support for this idea has come from measurements showing a finite phase stiffness above $T_c$ at terahertz frequencies [10], and from experiments that appear to detect the phase-slip voltage of thermally diffusing vortices in the normal state [11]. If the physics of the underdoped cuprates is indeed that of a fluctuating $d$-wave superconductor there should be a regime where quantum fluctuations come into play as $T_c$ falls to zero with decreasing doping. Here we test this idea with a detailed study of the doping dependence of the superfluid density in the vicinity of the critical doping for superconductivity.

High homogeneity of $T_c$ is particularly difficult to achieve in the underdoped cuprates, where the control parameter is chemical doping and the materials are well away from plateaus or turning points in $T_c(y)$. The YBa$_2$Cu$_3$O$_{6+y}$ system has two advantages in this doping range: with careful work, there can be sufficient control of doping homogeneity to produce samples with sharp superconducting transitions [12] [13]; and the process of CuO-chain ordering can be harnessed to provide continuous tunability of the carrier density in a single sample, with no change in cation disorder [14]. This is possible because the loosely held chain oxygen atoms in these high quality samples remain mobile at room temperature and gradual ordering into CuO-chain structures slowly pulls electrons from the CuO$_2$ planes, smoothly increasing hole doping over time [15] [16]. For this experiment, single crystals of YBa$_2$Cu$_3$O$_{6+y}$ were grown in barium zirconate crucibles and have high purity and low defect levels, with cation disorder at the 10$^{-4}$ level [17]. A crystal 0.3 mm thick was cut and polished with Al$_2$O$_3$ abrasive into an ellipsoid of revolution about the sample $c$-axis, 0.35 mm in diameter. The oxygen content of the ellipsoid was adjusted to O$_{6.33}$ by annealing at 914°C in flowing oxygen, followed by a homogenization anneal in a sealed quartz ampoule at 570°C and a quench to 0°C. At this point the sample was non-superconducting. After allowing chain oxygen order to develop at room temperature for three weeks, $T_c$ was 3 K. The sample was then further annealed at room temperature for six weeks under hydrostatic pressure of ~ 30 kbar, raising $T_c$ to 17 K. The sample was cooled to $-5$°C, removed from the pressure cell, and then stored at $-10$°C to prevent the oxygen order from relaxing. All further manipulation of the ellipsoid was carried out in a refrigerated glove box at temperatures less than $-5$°C. In between measurements of surface impedance, periods of controlled in-situ annealing at room temperature and ambient pressure were used to generate a sequence of 36 dopings as $T_c$ relaxed back to 3 K. We emphasize that no oxygen entered or left the sample during these annealing steps. Subsequent reannealing under hydrostatic pressure, for a further six weeks, returned $T_c$ to the starting value of 17 K, where the sample was remeasured to demonstrate the reversibility of the technique.

Measurements of the $ab$-plane surface impedance $Z_s = R_s + iX_s$ were carried out at 2.64 GHz by cavity perturbation, using a sapphire hot-finger to position the sample at the $H$-field antinode of the TE$_{015}$ mode of a rutile dielectric resonator [18]. All data sets reported here were taken with the $c$-axis of the ellipsoid oriented along the microwave magnetic field $H_{\parallel}$ to induce $ab$-plane screening currents. The surface impedance of the sample has been obtained from the measured cavity response using the cavity perturbation formula $\Delta f_B(T) = f_B^0 - f_B(T) = \Gamma(R_s + i\Delta X_s)$, where $\Delta f_B(T)$ is the change in bandwidth of the TE$_{015}$ mode upon inserting the sample into the cavity, and $\Delta f_B(T)$ is the shift in resonant frequency upon warming the sample from base temperature to $T$. 

arXiv:cond-mat/0509223v3 [cond-mat.supr-con] 5 Dec 2007
distance is set in the usual way by matching the data to an accuracy of 2.5%. The absolute surface reactivity is empirically determined using a Pb–Sn replica sample. Γ is a scale factor that applies to the data set as a whole, where we expect the imaginary part of the microwave conductivity to be very small. This is seen more clearly in the microwave conductivity measurements made starting in the most ordered state (Tc ≈ 17 K) followed by controlled oxygen annealing in small steps down to Tc ≈ 3 K. At the end of the experiment the sample was reordered and measured again to verify reproducibility. Lines mark where the vortex-unbinding transition should occur for a 2D superconductor. The dashed line corresponds to ρs2D = h^2/d/(4k_BTc^2μ0λ^2) = (2/π)T in each CuO2 plane. The solid line shows ρs2D = (2/π)T in each CuO2 bilayer. ρs(T) instead passes smoothly through this region. While mean-field-like over most of the doping range, ρs(T) develops downwards curvature near Tc at the highest dopings, a possible indication of the onset of the 3D-XY critical fluctuations.

The superfluid density is given by ρs ≡ 1/λ^2 = ωμ0σ2. Figure 2 shows ρs(T) at 20 of the 37 dopings. The most striking feature of the data is the wide range of linear temperature dependence, extending from close to Tc down to T ≈ 4 K. Below 4 K ρs(T) crosses over to an accurately quadratic temperature dependence. Such behaviour is well established in YBa2Cu3O6+y at higher dopings and is consistent with d-wave superconductivity in the presence of a small density of pair-breaking defects [20, 21]. One unusual feature of the data is the nature of the thermal transitions, which, within limits set by ΔTc appear mean-field-like. At optimal doping YBa2Cu3O6+y is the most three dimensional cuprate, with λ^2(T → 0)/λab^2(T → 0) ≈ 50 [22]. Its critical behaviour has been firmly established to be in the 3D-XY universality class [23, 24, 25]. In the doping range explored in this paper, YBa2Cu3O6+y is highly anisotropic, with λ^2(T → 0)/λab^2(T → 0) ≈ 10,000 [14]. In these circumstances, one would anticipate fluctuations in adjacent layers to be uncorrelated, and a Kosterlitz–Thouless–Berezinsky (KTB) vortex unbinding transition [6, 26] should occur when the 2D phase stiffness in a layer of thickness d. ρs2D(T) = h^2/d/(4k_BTc^2μ0λ^2) falls to (2/π)T. This defines minimum superfluid densities for isolated planes and CuO2 bilayers, shown in Fig. 2 by the dashed and solid lines respectively. ρs(T) instead passes smoothly through these lines, with no indication of vortex unbinding. Surprisingly, this implies that fluctuations remain correlated over many unit cells in the c direction. Recent work on YBa2Cu3O6+y thin films supports this, showing that the KTB transition does occur but that the effective thickness for fluctuations is the film thickness [27]. At the highest dopings in our experiment, ρs(T) develops slight downward curvature near Tc, possibly indicating the emergence of 3D-XY criticality.
Previous studies of superfluid density in the underdoped cuprates have focused on the strong correlation between $T_c$ and $\rho_{s0} \equiv 1/\lambda^2(T \to 0)$ over the entire underdoped region in considerably more detail. The strong correlation between $T_c$ and $\rho_{s0}$ remains, but $\rho_{s0}$ is up to an order of magnitude larger than in the earlier work on thin films [28]. As in the thin film study, $\rho_{s0}$ falls continuously to zero on underdoping and varies approximately quadratically with $T_c$ at low doping. Recent experiments on the CuO$_2$ plane doping state in YBa$_2$Cu$_3$O$_{y+y'}$ have established a mapping between $T_c(y)$ and $p$, the hole concentration per planar Cu [13]. In Fig. 3 a linear fit to the $T_c(p)$ data from Ref. 13 has been used to determine $p$ and plot $\rho_{s0}(p)$. At higher doping $\rho_{s0}$ has a linear doping dependence. This behaviour appears to be very robust: an extrapolation of the linear fit in Fig. 3 passes within 5% of the ab-averaged superfluid density of Ortho-II YBa$_2$Cu$_3$O$_{6+y'}$ [22]. To the extent that the linear extrapolation of $T_c(p)$ holds, $\rho_{s0}(p)$ varies approximately quadratically close to the onset of superconductivity. However, as we will discuss below, this quadratic behaviour is difficult to understand theoretically. Later on we will present an alternative proposal in which it is $\rho_{s0}$, not $T_c$, whose linear doping dependence extends to the edge of the superconducting phase.

The very low superfluid density, its continuous variation with doping, and the importance of 3D-XY fluctuations near optimal doping [23, 24, 25] together suggest that close to the critical doping, $p_c$, the transition out of the superconducting state may be controlled by fluctuations near a quantum critical point. In the scaling theory of a quantum phase transition, physical properties are related to a single, divergent correlation length, $\xi \propto |x|^{-\nu}$, where $x = (p - p_c)$. Scaling analysis of the $XY$ model predicts $T_c \propto \xi^{-z} \propto |x|^{\nu z}$ and $\rho_{s0} \propto \xi^{-(d-2+z)} \propto |x|^{\nu (d-2+z)}$, where $z$ is the dynamical critical exponent $[6, 29, 30]$. Together, these relations imply $T_c \propto \rho_{s0}^{\nu z/(d-2+z)}$, independent of $\nu$. In $d = 2$ this requires $T_c \propto \rho_{s0}$, whereas our experiments show $T_c \propto \rho_{s0}^{1/2}$. In $d = 3$, $T_c \propto \rho_{s0}^{2/(1+z)}$ and is consistent with our observations if $z = 1$, the so-called $(3+1)$D-XY universality class [8]. $D = 4$ is the upper critical dimension of the $XY$ model, so mean-field critical behaviour ($\nu = 1/2$) should follow. However, this choice of $\nu$ is not compatible with the doping dependences of $T_c$ and $\rho_{s0}$ shown in Fig. 3: scaling arguments predict $T_c \propto x^{1/2}$ and $\rho_{s0} \propto x$ whereas the analysis shown in Fig. 3 has $T_c \propto x$ and therefore $\rho_{s0} \propto x^{2}$. This difficulty may well stem from our determination of doping in Fig. 3 which is based on a linear fit and extrapolation of the $T_c(p)$ data from Ref. 13. The sparseness of the $T_c(p)$ data, along with the lack of an anchor point as $T_c \to 0$, allow a different interpretation in the low doping regime. In Fig. 3 all data with $T_c \geq 8$ K follow an accurately linear doping dependence of $\rho_{s0}$, which seems to extrapolate well to much higher dopings. The
robustness of this form leads us to put forward the following suggestion as a means of understanding the data. We propose that $\rho_{s0}(p)$ is, in actual fact, proportional to $(p - p_c)$ in the low doping range and use this linear mapping to determine $T_c(p)$. The results of this analysis are shown in Fig. 4. The mapping leaves $T_c(p)$ consistent with the data of Ref. [13] the effect is simply to refine the $T_c(p)$ curve in the vicinity of the critical doping. $T_c(p)$ now grows initially as $(p - p_c)^{1/2}$, in accord with scaling arguments, before crossing over to a linear doping dependence at higher dopings. Over a substantial doping range this yields the well-known result that $T_c$ scales approximately with $\rho_{s0}$. In addition, our proposed interpretation gives a plausible explanation of another aspect of the data. At the lowest dopings, the fluctuation peaks in $\sigma(T)$ broaden considerably, as shown in Figs. 4 and 5. In the presence of small, macroscopic variations in oxygen concentration, such broadening would be a natural consequence of a steepening of $T_c(p)$ on the approach to $p_c$. Future experiments, including those of the sort carried out in Ref. [13], will be needed to confirm our conjecture. However, the proposed scenario has several compelling features, not least of which is a substantial simplification of our theoretical picture of the transition from nonsuperconductor to superconductor in the underdoped cuprates.

In summary, at the critical doping for superconductivity $\rho_{s0}$ becomes nonzero and appears to grow linearly with doping, at a rate that remains constant up to much higher doping. Fluctuations in the low doping range are three dimensional, with no indication of the physics of 2D vortex unbinding, and with critical exponents characteristic of $(3 + 1)$-D-XY universality. Quantum fluctuations appear to control $T_c$ in the critical region, with $T_c \propto (p - p_c)^{1/2}$, but become less effective at depleting $\rho_s$ away from $p_c$, allowing $T_c(p)$ to cross over to a linear doping dependence.

We acknowledge useful discussions with A. J. Berlinsky, M. J. Case, S. Chakravarty, J. Cooper, J. C. Davis, J. S. Dodge, M. Franz, I. F. Herbut, S. Kivelson, J. E. Souier and Z. Tešanović. This work was funded by the National Science and Engineering Research Council of Canada and the Canadian Institute for Advanced Research.

FIG. 5: (color online). Fluctuation peaks in $\sigma(T)$ at 2.64 GHz as doping is varied. Peak width is approximately constant in the higher doping range and then broadens considerably below $T_c \approx 8$ K, consistent with a steepening of $T_c(p)$ in that range.

[1] J. Orenstein and A. J. Millis, Science 288, 468 (2000).
[2] V. J. Emery and S. A. Kivelson, Nature 374, 434 (1995).
[3] M. Franz and Z. Tešanović, Phys. Rev. Lett. 87, 257003 (2001); M. Franz, Z. Tešanović, and O. Vařek, Phys. Rev. B 66, 054555 (2002).
[4] I. F. Herbut, Phys. Rev. Lett. 88, 047006 (2002).
[5] I. F. Herbut, Phys. Rev. B 66, 094504 (2002).
[6] I. F. Herbut and M. J. Case, Phys. Rev. B 70, 094516 (2004).
[7] I. F. Herbut, Phys. Rev. Lett. 94, 237001 (2005).
[8] M. Franz and A. P. Iyengar, Phys. Rev. Lett. 96, 047007 (2006).
[9] Y. J. Uemura et al., Phys. Rev. Lett. 62, 2317 (1989).
[10] J. Corson et al., Nature 398, 221 (1999).
[11] Y. Wang et al., Science 299, 86 (2003).
[12] R. Liang et al., Physica C 383, 1 (2002).
[13] R. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. B 73, 180505 (2006).
[14] A. Hosseini et al., Phys. Rev. Lett. 93, 107003 (2004).
[15] J. Zaanen et al., Phys. Rev. Lett. 60, 2685 (1988).
[16] B. W. Veal et al., Phys. Rev. B 42, 6305 (1990).
[17] R. Liang, D. A. Bonn, and W. N. Hardy, Physica C 304, 105 (1998).
[18] A. W. Huttema et al., Rev. Sci. Instrum. 77, 023901 (2006).
[19] D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B 43, 130 (1991).
[20] D. A. Bonn et al., Phys. Rev. B 50, 4051 (1994).
[21] P. J. Hirschfeld and N. Goldenfeld, Phys. Rev. B 48, R4219 (1993); P. J. Hirschfeld, W. O. Putikka, and D. J. Scalapino, Phys. Rev. B 50, 10250 (1994).
[22] T. Perez-Barnea et al., Phys. Rev. B 69, 184513 (2004).
[23] S. Kamal et al., Phys. Rev. Lett. 73, 1845 (1994).
[24] A. Junod et al., Physica B 280, 214 (2000).
[25] C. Meingast et al., Phys. Rev. Lett. 86, 1606 (2001).
[26] L. B. Ioffe and A. J. Millis, J. Phys. Chem. Solids 63, 2259 (2002).
[27] Y. Zuev et al., unpublished, cond-mat/0407113; I. Hetel, T. R. Lemberger, and M. Randeria, Nature Phys. 3, 700 (2007).
[28] Y. Zuev, M. S. Kim, and T. R. Lemberger, Phys. Rev. Lett. 95, 137002 (2005).
[29] A. Kopp and S. Chakravarty, Nature Phys. 1, 53 (2005).
[30] S. Sachdev, Quantum Phase Transitions (Cambridge University Press, 1999).