Correlation between Superfluid Density and $T_C$ of Underdoped YBa$_2$Cu$_3$O$_{6+x}$
Near the Superconductor-Insulator Transition

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We report measurements of the $ab$-plane superfluid density $n_s$ (magnetic penetration depth $\lambda$) of heavily underdoped films of YBa$_2$Cu$_3$O$_{6+x}$, with $T_C$'s from 6 to 50 K. We find the characteristic length for vortex unbinding transition equal to the film thickness, suggesting strongly coupled CuO$_2$ layers. At the lowest dopings, $T_C$ is as much as 5 times larger than the upper limit set by the 2D Kosterlitz-Thouless-Berezinskii transition temperature calculated for individual CuO$_2$ bilayers. Our main finding is that $T_C$ is not proportional to $n_s(0)$; instead, we find $T_C \approx n_s^{1/2.3 \pm 0.4}$. This conflicts with a popular point of view that quasi-2D thermal phase fluctuations determine the transition temperature.

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The problem of high-$T_C$ superconductivity at severe underdoping is complicated by admixtures of different physics, intrinsic or extrinsic to superconductivity itself, e.g., stripes, pseudogap, metal-insulator transition. In particular, the relationship between the pseudogap and superconductivity is perhaps the central issue in the field [1]. There is now a wide variety of theories available that attempt to explain the coexistence of the pseudogap and superconductivity. At the mean-field level, most of them fail to account for the observed decrease of $T_C$ to zero with underdoping, and they appeal to thermal phase fluctuations to do the job [2–11]. This is reasonable if interlayer coupling in underdoped cuprates is weak enough that samples are quasi-2D, and $T_C$ is approximately the 2D-XY (or Kosterlitz-Thouless-Berezinskii, described below [12]) transition temperature. After all, this relationship holds, at least approximately, for cuprates that are moderately underdoped [13,14].

In some models [2–4] the pseudogap arises from quasi-2D phase fluctuations. Electron pairing occurs at the pseudogap temperature, which increases with underdoping, but phase fluctuations delay phase coherence to a much lower temperature, the observed transition temperature. In this framework, the measured $T_C$ is approximately the Kosterlitz-Thouless-Berezinskii (KTB) transition temperature for a single superconducting layer. Thus, $T_C \propto n_s(0)$. Therefore, the empirical Uemura proportionality between $T_C$ and $n_s(0)$ in underdoped cuprates finds a natural explanation.

In this Letter we show that $T_C$ is not proportional to superfluid density below optimal doping. In fact, $T_C$ is roughly proportional to $n_s(0)^{1/2}$ in underdoped samples, implying that phase fluctuations do not suppress $T_C$ as has been conjectured.

Experimental progress in this area is impeded by the difficulty of preparing homogeneous specimens at severe underdoping. The problem is that $T_C$ changes rapidly with $x$ near the superconductor-to-insulator transition, so a small oxygen composition variation across the sample results in a wide transition. We have made progress reducing transition widths to the point that the conclusions of this Letter are insensitive to them.

In an effort to improve oxygen homogeneity, we grew our YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) films between two PrBa$_2$Cu$_3$O$_{6+x}$ (PBCO) layers. Oxygen gradients near the substrate-film interface or near the free surface of the film should occur mostly inside the insulating PBCO layers. PBCO/YBCO/PBCO trilayers were deposited on (001) SrTiO$_3$ substrates by pulsed laser ablation with a Kr-F excimer laser (Lambdaphysik 305i, 248 nm wavelength, pulse energy 150 mJ). For the first PBCO layer, 10 unit cells thick, the substrate heater was at 820 °C and oxygen pressure was 140 mTorr. After deposition, this layer was fully oxidized at 500 °C for 10 minutes in 760 torr O$_2$. Then a 20 or 40 unit cell (235 or 470 Å, respectively) layer of YBCO and 20 or 40 unit cell cap layer of PBCO were deposited at 760 °C and 140 mTorr of O$_2$. PBCO films grown the same way were not superconducting.

After deposition, the films were annealed in situ for 12 to 24 hrs in 10 to 200 torr O$_2$ at 600 °C or 700 °C and then either quenched by dropping onto crumpled aluminum foil or cooled slowly with the heater turned off. It took about an hour to cool from 600 °C to under 200 °C, where oxygen exchange with ambient becomes negligibly slow. Films were $c$-axis oriented, as given by x-ray diffractometry. They had $T_C$'s down to 6 K, and even at this low $T_C$ the peak in $\sigma_1$ was well defined, if broad, whereas in previous attempts the peak in $\sigma_1$ spread down to $T = 0$.

Samples annealed for 12 hours showed lower $T_C$ and superfluid density than those annealed for 24 hours at the same (or even slightly lower) oxygen pressure and same temperature. From this we conclude that 12 hours is not enough to reach equilibrium with atmosphere. For the same anneal time, lower oxygen pressure results in lower $T_C$. From $T_C$ we infer oxygen content $x$ using a canonical
phase diagram (see, e.g., [15]). The films have $0.37 \leq x \leq 0.95$, with most films having $x \approx 0.5$.

From our mutual inductance measurements at 50 kHz we determine the sheet conductance $\sigma d_{\text{film}} = \sigma_1 d_{\text{film}} - i \sigma_2 d_{\text{film}}$ of the film. From the imaginary part, $\sigma_2 d_{\text{film}}$, we extract the $ab$-plane penetration depth $\lambda^{-2} = \sigma_2 \mu_0 a_0$. The superfluid density, $n_s$, is proportional to $\lambda^{-2}$. Details are given in Refs. [16,17]. The width of the fluctuation peak in $\sigma_1$ at $T_C$ is partly intrinsic and partly due to a spread in $T_C$’s in the sample. Thus, the width sets an upper limit on film inhomogeneity. Our best samples have peak widths of $\approx 2$ K, while others have several Kelvin wide peaks.

All measurements for this study were performed in doubly $\mu$-metal shielded cryostats to minimize the effect of the Earth’s magnetic field. For comparison, one data set was taken in both shielded and unshielded cryostats. The latter showed slightly lower $T_C$, therefore the $\mu$-metal shields were an important precaution.

We start with an overall look at the superfluid density of severely underdoped YBCO films, Fig. 1. This figure shows a representative series of severely underdoped films, 40 unit cells thick, with $T_C$’s from 6 to 46 K. The narrowest transitions are about 2 K wide, as given by the width of the $\sigma_1$ peak. For purposes of determining $\lambda^{-2}(0)$ vs $T_C$, we consider these transitions sufficiently narrow because at the lowest temperatures $\sigma_1$ is undetectably small. Optimally doped films, not shown in the figure, had $T_C = 90$ K, and $\lambda^{-2}(0) = 25 \mu m^{-2} \times \lambda^{-2}(T)$ was quadratic in $T$ at low $T$. These results are consistent with the films being disordered $d$-wave superconductors. Assuming that the scattering rate does not change significantly with underdoping, and that the superconducting gap is the pseudogap, then the underdoped films are actually cleaner than optimally doped films in the sense that the scattering rate is a smaller fraction of the superconducting gap energy.

In quasi-2D layered superconductors, an important temperature is the temperature where a 2D phase transition would occur in individual layers if they were uncoupled. This is the well-known Kosterlitz-Thouless-Berezinskii transition that is mediated by thermally generated topological excitations (vortex-antivortex pairs), which unbind at the transition. In a single CuO$_2$ layer it occurs at temperature $T_{2D}$ which is related to the measured magnetic penetration depth:

$$kT_{2D} = \frac{\Phi_0^2}{8\pi\mu_0} \frac{d}{\lambda^2(T_{2D})}$$

where $\Phi_0$ is the flux quantum and $d = 11.7$ Å is the thickness of 1 unit cell (note that if we use the film thickness in the above equation, we get an upper limit on the temperature at which thermal phase fluctuations must become important). The superfluid density drops discontinuously to zero, in theory, for a 2D superconductor. In cuprates, we expect (weak) interlayer coupling to soften the discontinuity to a continuous rapid downturn.

In Fig. 1, intersections of the solid straight lines with the measured $\lambda^{-2}(T)$ curves give $T_{2D}$, as dictated by Eq. (1). Quite obviously, $\lambda^{-2}(T)$ does not vanish only a little above that temperature. In fact, for the two most severely underdoped films, the observed $T_C$’s are 5 times larger than $T_{2D}$. On the other hand, intersections of the dashed lines with $\lambda^{-2}(T)$ give the 2D transition temperatures predicted by using the full film thickness in Eq. (1). The so-derived $T_{2D}$’s closely match the positions of the peaks in $\sigma_1$. We conclude that the characteristic length for the KTB transition is the film thickness and not a single unit cell thickness; severely underdoped films are not quasi-2D insofar as thermal phase fluctuations are concerned. Present results on many films augment our earlier report on the $T_C = 34$ K film [18].

We now turn to the central result of this Letter, which is the comparison of our data with the famous Uemura plot [13,14,19]. Figure 2 shows $T_C$ vs extrapolated values $\lambda^{-2}(0)$ in log-log and linear-linear (inset) scale. Figure 2 includes data on many more samples than are shown in Fig. 1. Within some noise, all of them fall on the same curve, irrespective of annealing procedure or transition width. In particular, samples with same oxygen content,
Fig. 2. Plot of $T = 0$ superfluid density vs $T_C$ in log-log (main graph) and linear-linear (inset) plots. The best power-law fit for $T_C < 50$ K is $\lambda^{-2}(0) \propto T_C^{2.3}$, as shown by the solid line. Vertical error bars are full widths of $\sigma_1$ peaks. The straight dashed lines show the $T_C$ from quasi-2D thermal phase fluctuations, Eq. (1), $d = 1.17$ nm. Open circles show original $\mu$SR data of Uemura et al. (see, e.g., [14]), converted according to $\sigma = (2700 \, \mu \Omega \, \text{s}^{1/2} / \lambda^2)$. Here the condensate plasma frequency $\omega_p^2$ = 120 Ohm/cm K $T_C$ is proportional to the superfluid density $\sigma_{dc}$.

Our central result is the finding that $\lambda^{-2}(0) \propto T_C^{2.3}$ for heavily underdoped YBCO films. This result disagrees with expectations based on the idea of cuprates as quasi-2D layered superconductors and with the phenomenological proportionality, $\lambda^{-2}(0) \propto T_C$, implied by the Uemura plot. Regarding the latter, it is worth noting that most of the data in the original Uemura plot come from samples that are not as severely underdoped as the samples presented here. For reference, the open circles in Fig. 2 show original $\mu$SR YBCO data of Uemura et al. [14]. The muon spin relaxation rate $\sigma = [2700 \, \mu \Omega \, \text{s}^{1/2} / \lambda]^2$, according to [21]. While there is a region of linearity between $T_C$ and $\lambda^{-2}(0)$, the open circles are in line with our data. $\mu$SR measurements by Keren et al. on Ca$_x$La$_{1-x}$Ba$_{2.75}$-La$_{0.25+}$Cu$_3$O$_y$ system down to $T_C = 8$ K show approximately linear relationship between $T_C$ and the muon spin rotation rate, which is proportional to the superfluid density [22]. It is unclear to us why their findings disagree with ours. Perhaps underdoped cuprates do not follow a universal behavior. On the other hand, our data are consistent with the phenomenological universal scaling proposed by Homes et al. [23], $\omega_p^2 = 0.025 \lambda^{-2}$ [23]. Putting numbers in, note that penetration depth for, e.g., optimally doped YBCO used by Homes et al. was 150 nm, while in our films it is 200 nm, corresponding to $\omega_p^2 = 6.25 \times 10^7$ cm$^{-2}$. Conductivity $\sigma_{dc} = 10^8$ Ohm$^{-1}$ cm$^{-1}$, and hence $\omega_p^2 / \sigma_{dc} T_C = 69$ Ohm/cm K, as must be for our low values of superfluid density.

A secondary result is that $T_C$ is not limited by quasi-2D thermal phase fluctuations, as they are understood from simulations of Josephson-coupled superconducting grains. Coupling between CuO$_2$ planes is apparently strong enough to make fluctuations effectively 3D and therefore relatively unimportant up to temperatures a few K below $T_C$.

Why does $\lambda^{-2}(0)$ decrease so rapidly with underdoping? If the pseudogap is due to an order parameter unrelated to superconductivity, like the $d$-density wave order parameter of Chakravarty et al. [25] then one can appeal to disorder (scattering) and a decreasing gap, $\Delta \propto T_C$, to account for the reduction in $\lambda^{-2}(0)$ with underdoping. If the pseudogap is the superconducting gap, then the films become cleaner with underdoping, and scattering cannot be the explanation. It is possible that percolation of some sort is important. If superconductivity is confined to localized regions, like malformed stripes, then the measured superfluid density may be determined by coupling between regions. One
can try to invoke quantum phase fluctuations (zero-point motion of the superfluid) to account for a rapid suppression of the superfluid density with underdoping. It has been shown theoretically in Josephson junction arrays [26,27], that the superfluid density (phase stiffness) should decrease rapidly when shunt resistance (sheet resistance $\rho/d$ in film) becomes comparable to a quantum resistance, $R_Q = h/4e^2 \approx 6 \, k \Omega$. In $s$-wave MoGe films with sheet resistances up to $900 \, \Omega$ quantum phase fluctuations do not have an observable effect on $\lambda^{-2}(0)$ [28]. For severely underdoped YBCO with $T_C = 10 \, K$, the $ab$-plane resistivity is about $1 \, m\Omega \times cm$ (see, e.g., [24]). The sheet resistance of the entire film would be about 200 $\Omega$, well below $R_Q$. The sheet resistance of a single CuO$_2$ bilayer is about 8 $k \Omega$, but it is hard to see why quantum phase fluctuations would be quasi-2D when thermal phase fluctuations are not. Finally, it has been speculated that electronic charge is renormalized to zero away from $d$-wave nodes in underdoped cuprates [29,30]. In the end, currently there is no reliable model that we are aware of, that predicts or explains our finding.

In conclusion, we find that the picture of quasi-2D thermal phase fluctuations in thin YBCO films does not account for the suppression of $T_C$ with underdoping. The transition temperature is significantly higher than suggested by Kosterlitz-Thouless-Berezinskii model of independent CuO$_2$ bilayers. The CuO$_2$ planes must be coupled together through the entire film thickness. At severe underdoping $T_C$ and superfluid density, $n_s(0) \propto \lambda^{-2}(0)$ are related by power law: $n_s(0) \propto T_C^{3/2}$. This disagrees with a popular conjecture in the field, that low values of the superfluid density in underdoped cuprates set the lowest energy scale and thus determine $T_C$. In other words, the suppression of $T_C$ from optimal values in underdoped cuprates cannot come (solely) from fluctuations of the phase of the order parameter.

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