Correlation between Superfluid Density and T_C of Underdoped $YBa_2Cu_3O_{6+x}$ Near the Superconductor-Insulator Transition

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We report measurements of the *ab*-plane superfluid density n_s (magnetic penetration depth λ) of heavily underdoped films of $YBa_2Cu_3O_{6+x}$, 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 $T_C \propto n_s^{1/2.3\pm0.4}$. 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 $YBa_2Cu_3O_{6+x}$ (YBCO) films between two $PrBa_2Cu_3O_{6+x}$ (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 σ_1 was well defined, if broad, whereas in previous attempts the peak in σ_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 $x \leq 0.5$.

From our mutual inductance measurements at 50 kHz we determine the sheet conductance $\sigma d_{\rm film} = \sigma_1 d_{\rm film} - i\sigma_2 d_{\rm film}$ of the film. From the imaginary part, $\sigma_2 d_{\rm film}$, we extract the *ab*-plane penetration depth $\lambda^{-2} = \sigma_2 \mu_0 \omega$. The superfluid density, n_s, is proportional to λ^{-2}. Details are given in Refs. [16,17]. The width of the fluctuation peak in σ_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 μ-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 μ-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

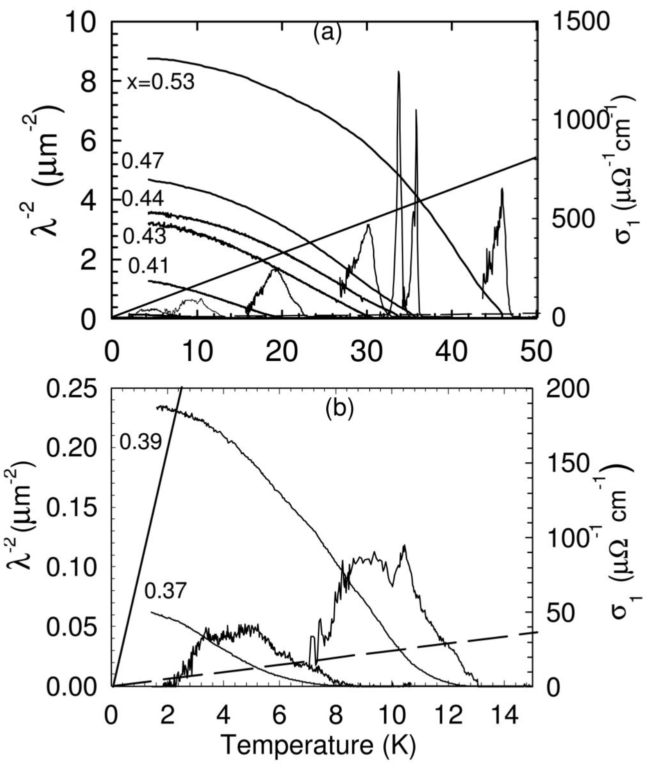

FIG. 1. (a) Superfluid density $n_s \propto \lambda^{-2}$ and real conductivity σ_1 as functions of T for $YBa_2Cu_3O_{6+x}$ films with nominal oxygen contents: $x = 0.53$, 0.47, 0.43, 0.42, and 0.41; (b) Close-up of 0–15 K region for films with $x = 0.39$ and 0.37. Solid lines are KTB lines for independent bilayers ($d =$ 11.7 Å), dashed lines are KTB lines for bilayers, coupled throughout film thickness ($d = 40 \times 11.7$ Å).

σ_1 peak. For purposes of determining $\lambda^{-2}(0)$ vs T_C, we consider these transitions sufficiently narrow because at the lowest temperatures σ_1 is undetectably small. Optimally doped films, not shown in the figure, had $T_C =$ 90 K, and $\lambda^{-2}(0) \approx 25\ \mu\text{m}^{-2} \times \lambda^{-2}(T)$ 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:

$$kT_{2D} = \frac{\Phi_0^2}{8\pi\mu_0} \frac{d}{\lambda^2(T_{2D})} \tag{1}$$

where Φ_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 in 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 σ_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,

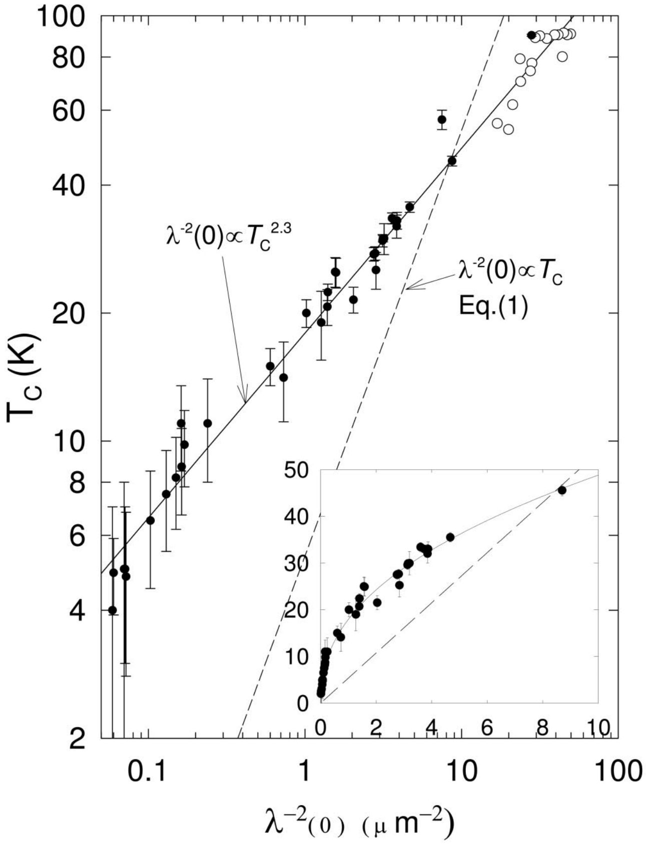

FIG. 2. Plot of $T = 0$ superfluid density vs T_C in log-log (main graph) and linear-linear (inset) plots. The best power-law fit for $T_C < 50$ K is $\lambda^{-2}(0) \propto T_C^{2.3}$, as shown by the solid line. Vertical error bars are full widths of σ_1 peaks. The straight dashed lines show the T_C from quasi-2D thermal phase fluctuations, Eq. (1), $d = 1.17$ nm. Open circles show original μSR data of Uemura *et al.* (see, e.g., [14]), converted according to $\sigma = (2700\ \text{Å}/\mu s^{1/2}/\lambda)^2$ [21].

but with a different degree of Cu-O chain disorder [and hence different T_C and $\lambda^{-2}(0)$], fall on the same curve as samples with different oxygen contents. It is clear that T_C is not proportional to $n_s(0)$. Instead, we find $\lambda^{-2}(0) \propto T_C^{2.3\pm0.4}$ for samples with $T_C < 50$ K. Above 50 K same power law is also in fairly good agreement with the data. The straight dashed line in both the main graph and inset, Fig. 2, shows the prediction for T_C from quasi-2D thermal phase fluctuations, Eq. (1), with $d = 1.17$ nm. For most films T_C is significantly larger than this upper limit. This result is important because it points to a limited role of classical thermal phase fluctuations in suppressing T_C of underdoped cuprates, as discussed below.

Values of $\lambda^{-2}(0)$ in underdoped YBCO films are about 5 times smaller than are seen in the cleanest YBCO crystals with the same T_C. In this sense YBCO films are more similar to other cuprates, e.g., BiSrCaCuO and LaSrCuO, than are YBCO crystals. In fact YBCO crystals at optimal doping possess the highest superfluid density of all hole-doped cuprates. Nevertheless, recently Liang and coworkers found $\lambda^{-2} \propto T_C^{1.6}$ in YBCO crystals [20], i.e., also power-law relationship with power greater than 1.

Our central result is the finding that $\lambda^{-2}(0) \propto T_C^{2.3\pm0.4}$ 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 μSR YBCO data of Uemura *et al.* [14]. The muon spin relaxation rate $\sigma = [2700\ (\text{Å}/\mu s^{1/2})/\lambda]^2$, 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. μSR measurements by Keren *et al.* on $Ca_xLa_{1-x}Ba_{1.75-x}La_{0.25+x}Cu_3O_y$ system down to $T_C \approx 8$ 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] $\omega_{ps}^2(0) = 120$ Ohm/cm K $T_C\sigma_{dc}(T_C^+)$, at least down to $T_C \approx 10$ K, as long as the dc conductivities of severely underdoped YBCO films just above T_C are approximately proportional to T_C, [24]. Here the condensate plasma frequency $\omega_{ps}^2 \approx 0.025\lambda^{-2}$ [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 $\omega_{ps}^2 = 6.25 \times 10^7$ cm^{-2}. Conductivity $\sigma_{dc} = 10^4$ Ohm^{-1} cm^{-1}, and hence $\omega_{ps}^2/\sigma_{dc}T_C = 69$ 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 superfuid 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 ρ/d in film) becomes comparable to a quantum resistance, $R_Q = h/4e^2 \approx 6$ kΩ. In s-wave MoGe films with sheet resistances up to 900 Ω quantum phase fluctuations do not have an observable effect on $\lambda^{-2}(0)$ [28]. For severely underdoped YBCO with $T_C \approx 10$ 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 Ω, well below R_Q. The sheet resistance of a single CuO_2 bilayer is about 8 kΩ, 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: $n_s(0) \propto T_C^{2.3}$. 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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