Crossover from 2D to 3D behavior in the superfluid density of thin \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) films

Yuri L. Zuev\(^a,1\), John A. Skinta\(^a\), Mun-Seog Kim\(^b\), Thomas R. Lemberger\(^a,\ast\), E. Wertz\(^c\), K. Wu\(^c\), Q. Li\(^c\)

\(^a\) Department of Physics, The Ohio State University, Columbus, OH 43210-1117, USA
\(^b\) Division of Electromagnetic Metrology, Korea Research Institute of Standards and Science, P.O. Box 102, Yuseong, Daejeon 305-600, Republic of Korea
\(^c\) Department of Physics, Pennsylvania State University, University Park, PA 16802, USA

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Abstract

Ultrathin underdoped films of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) (YBCO) exhibit a prominent and abrupt downturn in \( ab \)-plane superfluid density \( n_S(T) \) at a temperature consistent with a 2D vortex-pair unbinding transition. In this paper, we show that this characteristic feature of 2D superconductors diminishes with increasing film thickness consistent with theory, provided one uses the full film thickness rather than a copper-oxide bilayer thickness to calculate \( T_C \). In thick films, there is no evidence for 3D-XY fluctuations preceding the 2D downturn, as would be expected in a quasi-2D superconductor.

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1. Introduction

It has been proposed and widely accepted that cuprate superconductors should be viewed as stacks of weakly coupled two-dimensional copper-oxide sheets, \( \text{i.e.,} \) as quasi-2D superconductors. From this viewpoint, the temperature, \( T^* \), where the pseudogap appears may be the mean-field superconducting transition temperature, \( T_{\text{CO}} \), and 2D thermal fluctuations in the phase of the order parameter suppress the onset of phase coherence down to the measured \( T_C \) \([1\text{–}3]\). This model also accounts for the approximately linear scaling of \( T_C \) with superfluid density, \( n_S(0) \), in moderately underdoped samples, as reported by Uemura et al. \([4]\).

Striking new results require reconsideration of this viewpoint. Strongly underdoped \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) crystals and thick films \([5\text{–}7]\) show that \( T_C \) is actually sublinear in \( n_S \). Only in ultrathin films, which are two-dimensional by construction, does the 2D linear proportionality between \( T_C \) and \( n_S(0) \) appear \([8]\). Importantly, ultrathin films also exhibit the abrupt downturn in \( n_S(T) \) expected for 2D superconductors, showing that the peculiarities of cuprates do not preclude the usual vortex-pair unbinding transition.

The issue then arises as to how the transition is made between 2D ultrathin and “quasi-2D” thick samples. In this paper, we examine the superfluid densities of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) films of various thicknesses and doping levels. We find that the drop diminishes smoothly as film thickness increases, consistent with theory provided the film is considered as a single 2D entity, \( \text{i.e.,} T_C \) is calculated using the full film thickness, rather than the thickness of a copper-oxide bilayer.
The transition temperature of a 2D superconductor with thickness $d$ is given by the well-known relationship [9]:

$$\lambda_d^{-1}(T_{\text{KT}}) = \frac{8\pi\mu_0}{\phi_0} k_B T_{\text{KT}} = \frac{T_{\text{KT}}}{9.8 \, \text{mm} \, \text{K}},$$

(1)

where $\lambda_d^{-1} = d\lambda^{-2} \propto \eta_{3d}$ is the 2D magnetic penetration depth, and $\Phi_0$ is the flux quantum. In effect, Eq. (1) predicts a transition when the thermal energy, $k_B T$, equals the superconducting condensation energy in a characteristic volume $2\pi \xi^2 d$. Measurements of $\eta_{3d}$ in 2D films of conventional $s$-wave superconductors validate this equation [10,11]. For our films, it is useful to define two temperatures from Eq. (1). The lower, which we call $T_{2D}$, is the hypothetical transition temperature of a single CuO$_2$ bilayer, calculated from Eq. (1) using $d = 1.17$ nm, the thickness of a bilayer. If bilayers were decoupled, then $\eta_{3d}$ would drop discontinuously to zero at $T_{2D}$. Assuming that bilayers are coupled, but only weakly, then $T_{2D}$ should mark the beginning of a 3D-XY critical region rather than a discontinuity. In this case, for $T > T_{2D}$ and for a perfectly homogeneous sample, $\eta_{3d}(T)$ should arc downward proportional to $(T_C - T)^{2/3}$. The higher temperature, which we call $T_{2D}$, is the transition temperature of the film taken as a single 2D entity, calculated from Eq. (1) with $d = \text{film thickness}$. $T_{2D}$ is the upper limit on $T_C$ set by the film thickness. At this temperature, typical thermally excited vortex loops associated with 3D-XY critical fluctuations would be as big as or bigger than the film thickness. Their intersection with the film would look like a vortex–antivortex pair. In effect, fluctuations become 2D.

In the next section, we will see that all films show an abrupt downturn in $\eta_{3d}$ at $T_{2D}$. None of them shows evidence for 3D-XY critical behavior between $T_{2D}$ and $T_{2D}$. This behavior seems to conflict with experimental support for strong thermal fluctuations effects in YBCO provided by a critical region about 5 K wide in the superfluid density [12–14] and other properties of very clean YBCO crystals [15,16]. We argue that there is something anomalous about these results, however, since they are apparently very sensitive to disorder, and are thus not supported by data on slightly less clean YBCO crystals, or on YBCO films [17]. Moreover, strongly underdoped crystals also do not show the wide critical region seen near optimal doping [6].

2. Experiment

This study reports superfluid density measurements on four samples. Films A, B, and C were optimally doped. They were grown epitaxially by pulsed laser deposition (PLD) on NdGaO$_3$ substrates, as detailed elsewhere [18]. They are fully oxygenated. Each 1 cm × 1 cm film consists of buffer layers of semiconducting Pr$_{0.7}$Y$_{0.3}$Ba$_2$Cu$_3$O$_{7-\delta}$ that are 12 unit cells thick above and 8 unit cells thick below the YBCO film. The underlayer lessens the strain of substrate lattice mismatch on the YBCO, while the capping layer protects the YBCO film from damage during handling. Films A, B, and C are nominally 4, 8, and 10 unit cells thick, however, the top and bottom YBCO layers may not be perfectly smooth or homogeneous. Our conclusions are insensitive to this uncertainty. Film D was strongly underdoped. It was grown by PLD on a SrTiO$_3$ substrate at 760 °C, then annealed at 600 °C in a low pressure of oxygen. There were buffer layers of insulating PrBa$_2$Cu$_3$O$_{7-\delta}$ above and below the 40-unit-cell thick YBCO film.

We measured the effective $ab$-plane penetration depth $\lambda_d^{-1}(T) = d_{\text{film}}/\lambda^2$ with a two-coil mutual inductance technique [19]. Each film was centered between two coils roughly 2 mm in diameter and 1 mm long. In a typical measurement, the sample was cooled to 4.2 K, and a current of roughly 100 µA was driven at 50 kHz through the coil pressed against the back of the substrate. In this geometry, the induced $ac$ electric field in the film was parallel to the plane of the film and had azimuthal symmetry. It was very nearly uniform through the films because film thicknesses were much less than $\lambda$. Thus, the conductivities of all layers in the film were in parallel, and the measurement yielded the sheet conductivity of the film: $\sigma(\omega, T)d_{\text{film}} = \sigma_1(\omega, T)d_{\text{film}} - \sigma_2(\omega, T)d_{\text{film}}$. When the liquid helium level dropped below the sample, $T$ slowly increased, and the voltage induced in the secondary coil, which was pressed against the film, was measured continuously. $\lambda_d^{-1}(T)$ was obtained with an accuracy of about 3% from $\sigma_{2d_{\text{film}}}$ by using the relation:

$$\lambda_d^{-1}(T) \equiv \frac{\mu_0 \sigma_2(T) d_{\text{film}}}{2 \sigma_{2d_{\text{film}}}},$$

(2)

where $\mu_0$ is the permeability of vacuum. Because $\sigma_{2d_{\text{film}}}$ is the directly measured quantity, uncertainty in $d_{\text{film}}$ enters only in calculating the 3D penetration depth, $\lambda_d^{-1}(T)$, or the 2D penetration depth of a single CuO$_2$ bilayer from $\lambda_d^{-1}(T)$.

3. Results and discussion

3.1. Very thin optimally doped films

Fig. 1 shows $\lambda_d^{-1}(T)$ vs. $T$ (thick curves) measured for the three thin, optimally doped YBCO films. The value of $\lambda_d^{-2}(0)$ for the 10-unit-cell-thick film is what we routinely observe in thick YBCO films made by pulsed laser deposition. $\lambda_d^{-2}(0)$ decreases for thicknesses less than 10 unit cells, for reasons yet to be determined. $T_{\text{C}}$, denoted by vertical dotted lines, is defined as the center of the peak in $\sigma_1(50 \, \text{kHz}, T)$. The two open circles represent $T_{2D}$ and $T_{2D}$ calculated from Eq. (1) with $d = 1$ unit cell and $d = d_{\text{film}}$, respectively.

The dashed curves in Fig. 1 are quadratic, (i.e., $A \cdot BT^2$) fits to $\lambda_d^{-1}(T)$ just below $T_{2D}$, where fluctuations should be relatively weak. The quadratics approximate mean-field behavior and give us an idea of where the mean-field transition would occur. They also highlight the downturn in $\lambda_d^{-1}(T)$. The downturns are consistent with a 2D transition at $T_{2D}$. We see that for the thinnest film the mean-field transition temperature is only about 15 K above
Insets enlarge the transition regions. Dashed curves are quadratic fits to $k_{\perp}$ peaks in $r$ YBCO films A, B, and C (thick solid lines). Vertical dotted lines locate the $T_c$.

$3.2.\text{Strongly underdoped film}$

Fig. 2 shows $\lambda_{\perp}^{-1}(T)$ and $\sigma_1(T)$ measured at 50 kHz for film D. From its $T_C$ of 34 K, we estimate its oxygen stoichiometry to be $O_{6.4}$. The peak in $\sigma_1$ is due to fluctuations; the peak value of $\sigma_1$ is more than $10^5$ larger than the film’s dc conductivity above $T_c$. The width of the peak, about 2 K, is an upper limit on the inhomogeneity in $T_c$. In the vicinity of $T_2D \approx 22$ K the downward curvature of $\lambda_{\perp}^{-1}(T)$ is decreasing. The curvature is positive between 29 and 32 K before finally dropping rapidly at $T_{2D} \approx 33.7$ K. The quadratic fit to data between 15 and 22 K, i.e. just below $T_{2D}$, actually extrapolates to a temperature lower than $T_C$. Consistent with the optimally doped films, the ultimate downturn in $\lambda_{\perp}^{-1}(T)$ looks like a 2D transition with a characteristic thickness equal to the film thickness. Thermal phase fluctuations as understood from the literature cannot account for positive curvature between $T_{2D}$ and $T_{2D}$.

$T_{2D}$: Sample inhomogeneity is irrelevant because whatever the inhomogeneity, it does not extend below about 32 K, as indicated by the lower edge of the peak in $\sigma_1$. Our conclusion is that a vortex-pair unbinding transition occurs in this 40-unit-cell thick film, as does in ultrathin films. Associated thermal fluctuations suppress $T_C$ by only a few Kelvins. This is inconsistent with the pseudogap temperature being the mean-field superconducting transition. The relative importance of thermal and quantum fluctuations vs. single-particle excitations in the $T$-dependence of $\lambda_{\perp}^{-1}(T)$ remains to be clarified.

$4.\text{Conclusion}$

In conclusion, we have made high precision, low frequency measurements of $\lambda_{\perp}^{-1}(T)$ and $\sigma_1(T)$ in YBCO films 4–40 unit cells thick. The central result of this paper is that the 2D transition persists in films of all thicknesses, and that associated fluctuations suppress the transition by only a few degrees. A 3D-XY critical region is expected in thicker films, but is not observed. The reason is unclear.

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$\text{References}$