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Phys. Rev. B **91**, 104524 — Published 30 March 2015

DOI: [10.1103/PhysRevB.91.104524](https://doi.org/10.1103/PhysRevB.91.104524)

Doping-dependent critical Cooper-pair momentum in thin, underdoped cuprate films

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(Dated: February 12, 2015)

We apply a low-field (< 100 G) technique to measure the critical pair momentum p_c in thin, underdoped films of $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ reflecting a wide range of hole doping. We observe that $p_c \propto \hbar/\xi$ scales with T_c and therefore superfluid density $n_s(T \rightarrow 0)$ in our two-dimensional cuprate films. This relationship was famously predicted by a universal model of the cuprates with a *doping-independent* superconducting gap, but has not been observed by high field ($> 10^4$ G) measurements of the coherence length ξ because high-field cuprate phenomena obscure the simple scaling we observe at much lower fields.

The phenomenology of underdoped, hole-doped cuprates is puzzling due to the observation of a multitude of characteristic spectroscopic energies, none of which appears to scale with the superconducting critical temperature T_c . On the other hand, there is significant evidence[1],[2] that T_c is regulated by superfluid density n_s . Previous work[3],[4] with thin (thickness $d < 5$ unit cells) cuprate films shows that T_c is consistent with an observed Kosterlitz-Thouless vortex-driven thermal phase transition and scales with $n_s(T \rightarrow 0)$ as expected for a 2D quantum critical point at $n_s \rightarrow 0$.

Angle-Resolved PhotoEmission Spectroscopy (ARPES) reveals an anisotropic gap structure in the cuprate superconductors, traditionally considered in a two-gap picture. Plotted against the d -wave angular dependence $|\cos(k_x a) - \cos(k_y a)|/2$, the spectroscopic gap rises linearly in directions away from the node at (π, π) and bends sharply upward near the antinodal directions $(\pm\pi, 0)$ and $(0, \pm\pi)$. The gap magnitude near the node is seen to vanish for temperatures above the superconducting T_c (for all but the lowest-doped samples), and a linear fit to the nodal gap[5] gives a value Δ_0 . The antinodal gap $\Delta^* > \Delta_0$ vanishes above the pseudogap temperature T_c^* . It appears from data on underdoped samples[6] that $\Delta^* \approx 2k_B T_c^*$ across the cuprates, rather like a dirty-limit BCS gap. As the superconducting T_c diminishes with underdoping, T_c^* and the pseudogap Δ^* are seen to rise.

On the overdoped side of the "superconducting dome" where the pseudogap is significantly depressed, Δ_0 scales roughly with T_c , leading to its identification as the superconducting gap. The situation is complicated on the underdoped side of the dome, where Δ_0 remains *constant* as the pseudogap emerges and T_c falls. The latest high-resolution Laser ARPES reveals that another gap opens at very low doping[7] (hole concentration $p < 0.075$), with a nonzero magnitude at the node. From these disparate data, a single spectroscopic energy that scales with T_c is not obvious. Based on the available ARPES data, Lee and Wen (L&W) used a doping-independent d -wave gap in their generalized underdoped cuprate model[8] and concluded that the superconducting coherence length ξ is inversely proportional to T_c , not the energy gap. Our two-coil experiments measure $p_c \propto \hbar/\xi$ directly, and thus make for a natural test of the theory.

Thin cuprate films were prepared via Pulsed Laser Deposition (PLD) using a 248-nm krypton-fluorine excimer laser. The laser was operated at a rate of 30 Hz with an en-

ergy density of 2.4 J/cm^2 at the target. Growth proceeded in an atmosphere of flowing oxygen at 300 mTorr. The $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (Ca-YBCO) films were grown on commercially manufactured strontium titanate (STO-001) substrates heated to 760°C , and the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) on lanthanum aluminum oxide (LAO-100) at 735°C .

Film thickness was controlled by measuring the pulses-per-unit-cell for a thick film using atomic force microscopy. STO substrates were prepared with three unit cells of non-superconducting $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (PBCO) deposited on the bare STO. Ca-YBCO was deposited one unit cell at a time, with a 90 second rest between growth layers. Bi-2212 was deposited one half unit-cell at a time directly to bare LAO, with an 80 second delay between layers. All film samples were then capped with ten unit cells of PBCO to protect against atmospheric degradation. After deposition, the Ca-YBCO films were annealed at 450°C in oxygen pressures ranging from 1 to 650 Torr to control the oxygen doping. The Bi-2212 films were annealed at 700°C in pressures of 75 to 100 Torr. The hole concentration is not measured, so the primary measure of doping is the superfluid density $n_s(T \rightarrow 0)$.

Thin film characterization was conducted in a liquid helium cryostat capable of 1.4 K. Superfluid density was measured in a typical two-coil experiment[9],[10], using a small 10 kHz drive current and a correspondingly small drive field ($< 20 \text{ mG}$). Throughout the paper we shall refer to the quantity $1/\lambda^2(T)$, proportional to the three dimensional density of supercurrent carriers, as the "superfluid density". The areal superfluid density $n_s d$ is proportional to $1/\Lambda$, where $\Lambda \equiv 2\lambda^2/d$ is the two-dimensional penetration depth called the Pearl length. The two-coil experiment measures $1/\Lambda$ directly. Values of $1/\lambda^2$ therefore contain any uncertainty in the film thickness.

Figures 1a, 1b, and 1c give the superfluid density as a function of temperature for the nine films in this study. We analyzed the data in the manner of Hetel et al.[3], and the scaling of the extrapolated zero-temperature superfluid density $1/\lambda^2(0)$ with the transition temperature in Figure 1d follows that of Ref. [3], as well as Hinton et al.[4], so we conclude that the present films are comparable.

The samples were transferred to a second apparatus that was specially constructed to enable the high applied magnetic fields from the drive coil that are needed to explore the non-linear film response[11]. The samples were cooled to about

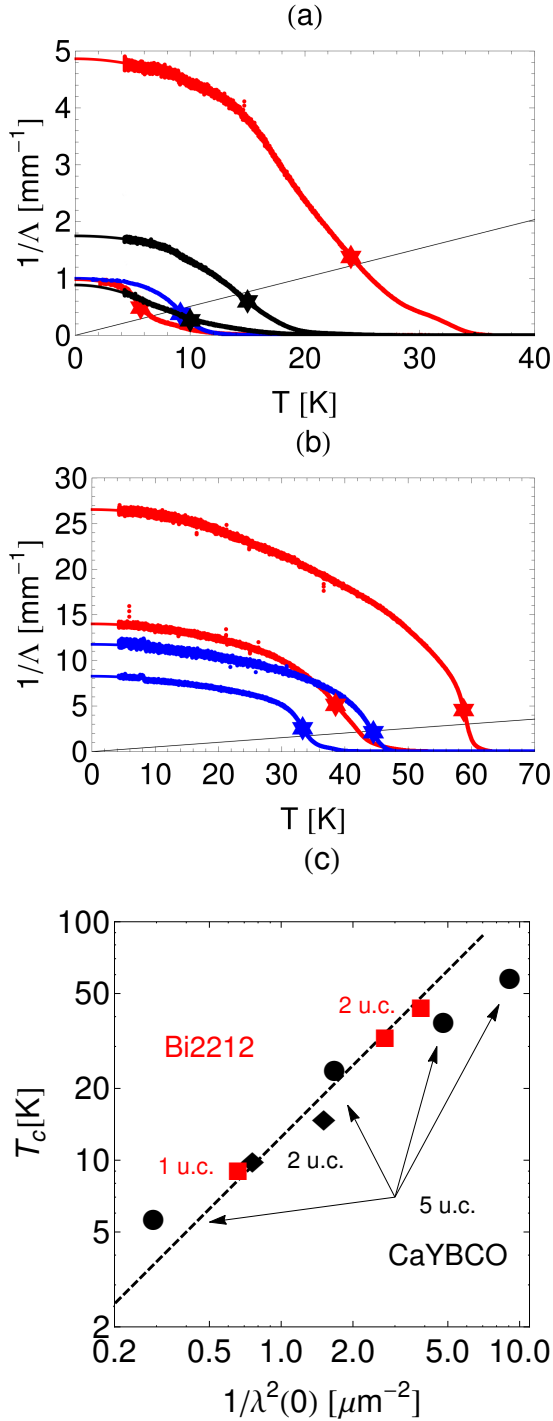


FIG. 1. Areal superfluid density $1/\Lambda(T)$ measured for the nine films in this study: five-unit-cell Ca-YBCO (red: a,b), two-unit-cell Ca-YBCO (black: a), one-unit-cell Bi2212 (blue: a), and two-unit-cell Bi2212 (blue: b). We define T_c to be the temperature at which the real part of the conductivity, σ_1 (not shown) has a peak, as indicated by the asterisks on the curves. The Kosterlitz-Thouless (KT) construction for T_c , namely the intersection of the line with $1/\Lambda(T)$, yields essentially the same results as appropriate for 2D superconductors. T_c shows linear scaling with superfluid density, as expected for quantum fluctuations in a 2D superconductor (c). Dashed line is the 2D scaling reported by Hetel et al. [3].

1.4 K and measurements begun. Our superconducting drive coil is designed so that the maximum amplitude B_0 of the applied magnetic field is perpendicular to the film surface at the center of the film. B_0 is smoothly varied from 0-40 Gauss at constant temperature to produce the mutual inductance traces presented in Figure 2. The normalization reveals the distinct peak feature in the out-of-phase mutual inductance common to all of the films, regardless of their thickness and doping, and this peak tracks the field at which the in-phase mutual inductance rises to midway between the normal and superconducting state values. We identify this value of B_0 as B_{NL} , the field at which the induced supercurrent density in the film reaches its critical value and vortex-antivortex pairs unbind as the metastable Meissner state breaks down. This interpretation of the data was verified qualitatively and quantitatively from measurements on thin Nb and MoGe films[11].

The critical pair momentum p_c is computed from the low-temperature values of B_{NL} by a calculation of Lemberger and Ahmed[12] using an idealized version of our experimental configuration. We report values of p_c for our cuprate films using this result, assuming that the measured B_{NL} coincides with the applied field at which the Meissner state becomes unstable, and vortex-antivortex pairs unbind *en masse*[13]. B_{NL} is essentially flat at low temperatures, decreasing by only about 10% between 1.4 K and 8 K, consistent with calculations of $p_c(T)$ in s-wave[14] and d-wave superconductors[15] and in sharp contrast to the measured vortex lattice-melting field[16], which decreases rapidly at low temperature.

Figures 1a and b show the measured areal superfluid densities, $\propto 1/\Lambda$, of the Ca-YBCO and Bi-2212 films in this study. Some films show a downturn consistent with Kosterlitz-Thouless (KT) theory for 2D superconductors, while others have broader transitions. T_c , indicated by asterisks in Fig. 1a and b, is defined as the temperature where the real conductivity, σ_1 , has a peak. An alternative definition, the intersection of the KT line with the measured $1/\Lambda$, yields essentially the same T_c 's, again consistent with KT theory. Importantly, the scaling of T_c with superfluid density, $\propto 1/\lambda^2(0)$, is linear and in quantitative agreement with data of Hetel et al. on similar films, supporting the notion that these films are two-dimensional superconductors with a thickness equal to that of the film, not that of a single copper-oxide bilayer, and that quantum fluctuations play an important role in them. We note that our single- and double-unit-cell Bi2212 films, which are considerably thinner than those studied by Yong et al.[17], do not show the linear suppression of superfluid with temperature observed by those authors in samples with $T_c < 50$ K.

Figure 2 shows normalized mutual inductance vs applied field at 1.4 K for all of our films. The applied field is normalized by a field, B_{NL} , defined as the field at the peaks in imaginary mutual inductance, as was done in benchmarking measurements on MoGe and Nb films [11]. The curves are similar enough to suggest that the underlying phenomenology is essentially the same. Deviations, like the green curves in Fig. 2 for a Ca-YBCO film with $T_c = 42$ K, likely arise from minor flaws in these very thin films, occasional poor grain

boundaries, for example. For comparison, Fig. 3 shows that cuprate films are quite similar to Nb and amorphous MoGe films, so we extract the critical Cooper pair momentum using the same model.

For the present work, we measured the temperature dependence of B_{NL} for $T < 8$ K and found that it falls with negligible slope, (consistent with calculations of $p_c(T)$ in s-wave[14] and d-wave superconductors[15]), until the experiment reaches liquid helium temperature. Therefore, we attribute the drop in $B_{NL}(T)$ beginning near 4.2 K to heating of the film as vortices are produced (Figure 4). As seen in the figure, the change in superfluid density over this range is insufficient to account for this drop. Since error due to heating is only a 10% effect and values of p_c in the present work were measured below 4.2 K to minimize heating, we are confident that our measurements of p_c are the zero-temperature values.

Finally, Figure 5 is the major result of the present study: we observe that the critical pair momentum scales with the measured T_c in thin, severely underdoped cuprate films. For an s-wave BCS superconductor, this is expected because T_c gives a measure of the energy gap, and depairing[18] occurs when the applied vector potential un-gaps the quasiparticle spectrum. In the cuprates, however, the gap is seemingly doping-independent, so it is not obvious that p_c *should* scale with T_c . Nonetheless, taking p_c as inversely proportional to L&W's coherence length we expect to see $p_c \propto T_c$ as observed in our low T_c data.

The latest high-magnetic field measurements[19] of $B_{c2} \propto 1/\xi^2$ affirm a non-monotonic increase of B_{c2} with T_c , rather than the simple quadratic dependence that follows from the L&W model. Ref. [19] gives $\xi \approx 15 - 40$ Å for the $T_c > 20$ K samples studied. For those of our films that were only slightly underdoped, our non-linear technique yields comparable values, with $\xi \approx 20 - 30$ Å. Severely underdoped films, on the other hand, have much larger values of ξ , and consequently much lower values of the critical momentum p_c . It is these films that match the Lee and Wen theory while high-field data do not. Given that spin order and charge order have been directly observed in conjunction with the vortex lattice[20],[21],[22] and the vortex radius itself is seen to be field-dependent[?], it is not surprising that the L&W theory does not fit the B_{c2} data. The framework of L&W simply does not include such physics, which appear to become important at *much* higher applied magnetic fields than our experiment.

In summary, we performed an inductive measurement of the critical pair momentum in nine thin, underdoped, hole-doped cuprate films. T_c is proportional the zero-temperature superfluid density $n_s(T \rightarrow 0)$, consistent with two-dimensional quantum critical behavior. The critical pair momentum is observed to scale with T_c and thus $n_s(T \rightarrow 0)$, directly confirming the theory of Lee and Wen[8], which assumes a doping-independent superconducting gap consistent with the latest ARPES studies, where no evidence of a diminishing gap as $T_c \rightarrow 0$ is observed.

TABLE I. B_{NL} data for the nine experimental samples.

Sample	Unit-cells	T_c [K]	B_{NL} [G]
Ca-YBCO	2	8.3	0.24
Ca-YBCO	2	14	0.45
Ca-YBCO	5	6.6	0.11
Ca-YBCO	5	25	1.1
Ca-YBCO	5	42	7.6
Ca-YBCO	5	59	16
Bi2212	1	9.9	0.066
Bi2212	2	34	4.1
Bi2212	2	44	5.1

ACKNOWLEDGMENTS

The authors wish to thank Mohit Randeria and Sumilan Banerjee for their interest and input. This research was supported by DOE-Basic Energy Sciences through Grant No. FG02-08ER46533.

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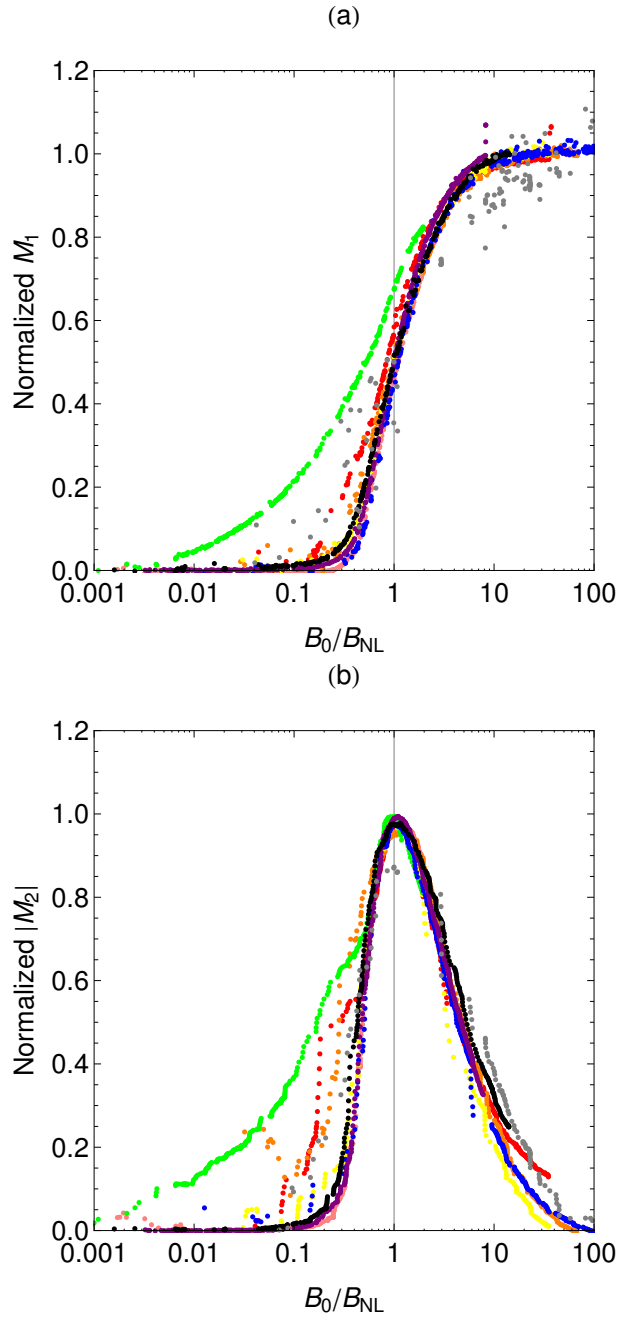


FIG. 2. In-phase (M_1) and out-of-phase (M_2) mutual inductances as functions of drive field at the center of the film (B_0) for all nine films in the study (at 10 kHz frequency). Normalization is as follows: $M_1 = \frac{M_1(B_0) - M_1(B_0 \rightarrow 0)}{M_0 - M_1(B_0 \rightarrow 0)}$, $M_2 = \frac{|M_2(B_0)|}{\max |M_2(B_0)|}$. Signal is phase-calibrated at low excitation field $B_0 \rightarrow 0$: $M_2 \rightarrow 0$. Initial position is measured from the normal-state response: $M_0 \equiv M_1(T > T_c)$.

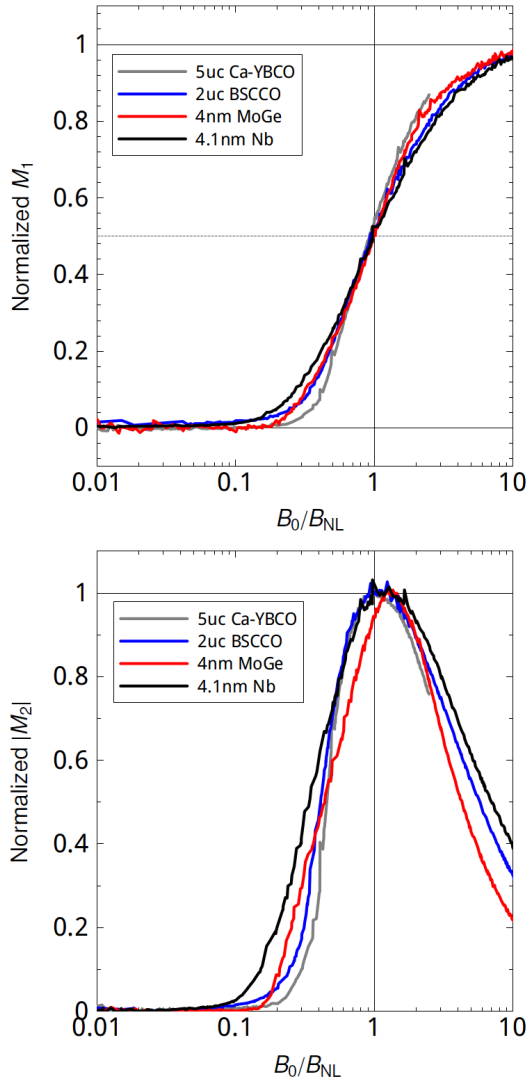


FIG. 3. Comparison of mutual inductance vs. applied field for a wide range of superconducting materials: two cuprates and two conventional BCS superconductors, showing the similar phenomenology. Normalization follows Fig. 2.

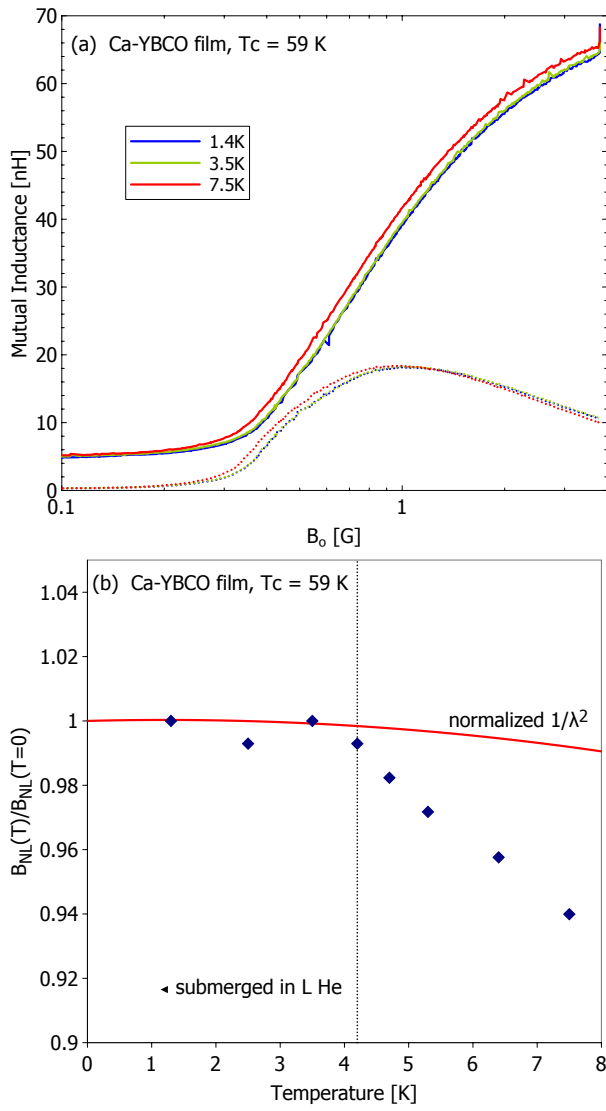


FIG. 4. a) Raw mutual inductance data for a single Ca-YBCO film at different temperatures. Below 4.2 K, the curves are virtually indistinguishable, while at $T/T_c = 0.13$, the curves are shifted slightly. b) Temperature dependence of the out-of-phase mutual inductance peak for the same Ca-YBCO film, showing negligible spread in B_{NL} error due to temperature. The superfluid density is effectively constant over the range of the plot, and thus small errors in B_{NL} will be linearly proportional to the error in p_c . Dashed line is liquid helium temperature. Repeated mutual inductance traces made on the same film are so consistent that changes in the out-of-phase peak position on the order of 1% or less can be resolved.

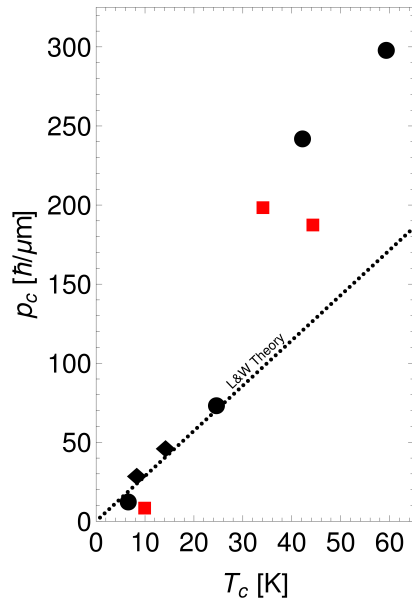


FIG. 5. Measured critical pair momentum p_c plotted against T_c . Plotting conventions follow Fig. 1c. Dashed line is relationship predicted by Ref. [8], with slope adjusted to fit the severely underdoped data. High T_c data, which is in line with high-field measurements, is shown for the contrast between slightly and severely underdoped films.