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Scaling of the superfluid density in strongly underdoped $YBa_2Cu_3O_{6+y}$: Evidence for a Josephson phase

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Recent measurements on extremely underdoped $YBa_2Cu_3O_{6+y}$ [Phys. Rev. Lett. **99**, 237003 (2007)] have allowed the critical temperature T_c, superfluid density $\rho_{s0} \equiv \rho_s(T \ll T_c)$, and dc conductivity $\sigma_{dc}(T \gtrsim T_c)$ to be determined for a series of electronic dopings for $T_c \simeq 3-17$ K. The general scaling relation $\rho_{s0}/8 \simeq 4.4\sigma_{dc}T_c$ is observed, extending the validity of both the *ab*-plane and *c*-axis scaling an order of magnitude and creating a region of overlap. This suggests that strongly underdoped materials may constitute a Josephson phase; as the electronic doping is increased a more uniform superconducting state emerges.

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Since the discovery of superconductivity at elevated temperatures in the cuprate materials, there has been a concerted effort to identify empirical scaling relations in the hope of providing insights to the mechanism for superconductivity in these materials. While the nature of the superconductivity remains elusive, it is now generally accepted that in the optimally doped compounds the superconducting energy gap has a *d*-wave symmetry. Much of the current research in the cuprate materials focuses on the underdoped compounds where the development of a pseudogap[1] in the antinodal region in the normal state at a characteristic temperature T^* leads to the formation of Fermi arcs (or pockets) centered around the nodal regions.[2,3] There is currently a considerable amount of debate as to whether or not the pseudogap represents preformed pairs that simply lack the coherence required for superconductivity[3] or if superconductivity originates in the Fermi arcs (pockets) and the pseudogap reflects some alternative ground state that competes with superconductivity.[4–6] An important scaling relation in the underdoped cuprates is the Uemura relation,[7] which notes that the superfluid density at low temperature (ρ_{s0}) is proportional to the critical temperature (T_c). The superfluid density is defined here as $\rho_{s0} \equiv 1/\lambda_0^2$, where $\lambda_0 = \lambda(T \ll T_c)$ is the effective penetration depth. Alternatively, ρ_{s0} is also referred to as the superfluid stiffness where $\rho_{s0} = 4\pi n_{s0}e^2/m^*c^2$, where n_{s0} is the density of the superconducting carriers and m^* is an effective mass. The Uemura relation works well over much of the underdoped region but does not apply in optimal and overdoped materials.[8] In addition, as the phase diagram for $YBa_2Cu_3O_{6+y}$ has been extended to the strongly underdoped regime,[9] the scaling of ρ_{s0} is observed to change from a linear to power-law relation.[10–13]

In this work we demonstrate that the strongly underdoped data for $YBa_2Cu_3O_{6+y}$ is in fact consistent with a more general scaling relation $\rho_{s0}/8 \simeq 4.4\sigma_{dc}T_c$, where σ_{dc} is the dc conductivity measured at $T \gtrsim T_c$ (note that in this representation both σ_{dc} and T_c are shown in units of cm^{-1} so that the constant is dimensionless and ρ_{s0} has the units of cm^{-2}).[14–16] In the cuprates this scaling relation is valid for the copper-oxygen planes, as well as along the poorly conducting *c* axis. The underdoped $YBa_2Cu_3O_{6+y}$ data extend the validity of both the *ab*-plane and *c*-axis scaling by almost an order of magnitude and also provides a previously unavailable region of overlap between the *a*-*b* planes and the *c* axis. The possibility of a continuous evolution of the in-plane behavior from a Josephson phase in the strongly underdoped materials to a more uniform superconducting state in systems with higher electronic dopings is considered.

In general, the study of the extremely underdoped region of the phase diagram for the cuprate materials has been complicated by the cation disorder accompanied by broad transition widths. An advantage of $YBa_2Cu_3O_{6+y}$ is that the hole doping can be tuned in a reversible way by controlling the amount of oxygen in the copper-oxygen chains. At room temperature, the dopant oxygens in the chains are mobile and gradually order into chain structures, removing electrons from the copper-oxygen planes and increasing the hole doping and T_c. Recent advances in the synthesis of $YBa_2Cu_3O_{6.333}$ (Ref. 17) allows the electronic doping in the copper-oxygen planes to be tuned continuously in a single sample with no change in the cation disorder and relatively sharp superconducting transitions.[18] Through annealing the chain order can be controlled and T_c may be increased from $\simeq 3$ K to as high as 17 K and then relaxed back to $\simeq 3$ K; this is a remarkable achievement in a material with a maximum $T_c \simeq 93$ K. In between periods of annealing, microwave surface impedance techniques have been employed to determine T_c and the *ab*-plane values of ρ_{s0} and $\sigma_{dc}(T \gtrsim T_c)$ for a series of 39 dopings.[13] The *c* axis properties have been determined in a separate experiment for a series of 13 dopings.[19]

In the copper-oxygen planes, the maximum electronic doping yields a $T_c = 17.4$ K, $\sigma_{dc} = 5400\ \Omega^{-1}\,cm^{-1}$, and $\lambda_0 = 3870$ Å; for the minimum electronic doping $T_c \simeq 3$ K while the dc conductivity is roughly half of its previous value $\sigma_{dc} = 3020\ \Omega^{-1}\,cm^{-1}$ and the penetration depth has increased dramatically to $\lambda_0 \simeq 0.24\ \mu m$. When σ_{dc} is discussed in terms of a sheet resistance $R_\square = \rho_{dc}/d$ (where the interbilayer separation is $d = 11.8$ Å) then for the maximum electronic doping $R_\square = 3.2\ k\Omega$ (per sheet), while the minimum electronic doping yields $R_\square = 5.6\ k\Omega$, which is remarkably close to the threshold for the superconductor-insulator transition[20] observed to occur close to $R_\square = h/4e^2 \simeq 6.9\ k\Omega$. The underdoped materials are extremely anisotropic, with $(\lambda_{0,c}/\lambda_{0,ab})^2 \simeq 10^4$ over much of the doping range. Along the

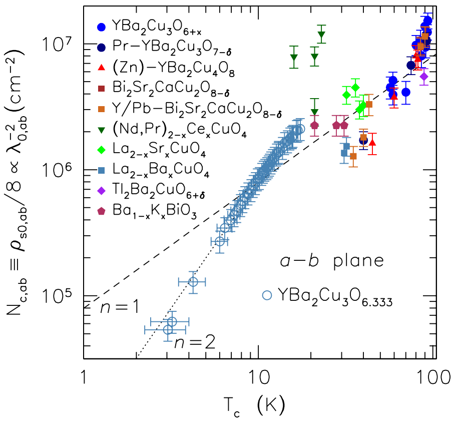

FIG. 1. (Color online) The log-log plot of the spectral weight of the superfluid density $N_c \equiv \rho_{s0}/8$ vs T_c for the *a-b* planes for a variety electron- and hole-doped cuprates as well as the microwave data for a single crystal of strongly underdoped $YBa_2Cu_3O_{6.333}$ for 39 different electronic dopings, yielding a range of $T_c \simeq 3-17$ K. The functional form $\rho_{s0} \propto T_c^n$ is considered for $n=1$ (dashed line) and $n=2$ (dotted line).

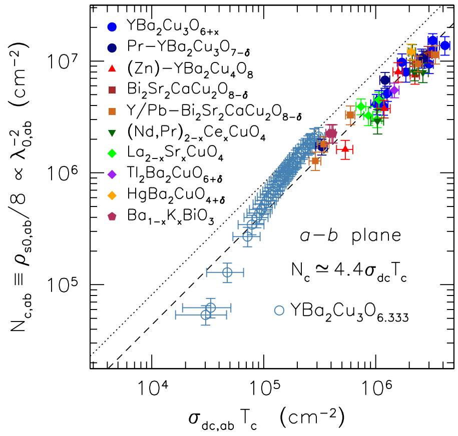

FIG. 2. (Color online) The log-log plot of the spectral weight of the superfluid density $N_c \equiv \rho_{s0}/8$ vs $\sigma_{dc}T_c$ for the *a-b* planes axis for a variety electron- and hole-doped cuprates, including the strongly underdoped $YBa_2Cu_3O_{6.333}$ material. The dotted line is the expected result for a BCS superconductor in the weak-coupling limit where the normal-state scattering rate is much larger than the isotropic gap [$N_c = \rho_{s0}/8 \simeq 8.1\sigma_{dc}T_c$ (Ref. 15)]; the dashed line is the observed scaling ($N_c = \rho_{s0}/8 \simeq 4.4\sigma_{dc}T_c$).

poorly conducting *c* axis, for the maximum electronic doping $T_c = 16.9$ K, $\sigma_{dc} = 0.67$ Ω^{-1} cm^{-1}, and $\lambda_0 = 40$ μm, while for the minimum $T_c = 3.9$ K, $\sigma_{dc} = 0.52$ Ω^{-1} cm^{-1}, and $\lambda_0 = 137$ μm.

The optically-determined values of $\rho_{s0}/8$ in the *a-b* planes are shown as a function of T_c in Fig. 1 for a variety of single-layer and double-layer cuprates.[15] The quantity $\rho_{s0}/8$ is also referred to as the spectral weight of the condensate, N_c. The spectral weight is defined as

$$N(\omega_c, T) = \int_{0^+}^{\omega_c} \sigma_1(\omega, T) d\omega,$$

which is simply the area under the conductivity curve over a given interval. The spectral weight of the condensate is $N_c = N(\omega, T \simeq T_c) - N(\omega, T \ll T_c)$ where the cutoff-frequency is chosen so that N_c has converged. The superfluid density is precisely $\rho_{s0} \equiv 8N_c$; this transfer of spectral weight is also known as the Ferrell-Glover-Tinkham sum rule.[21] While some of the optical data fall close to the $\rho_{s0} \propto T_c$ (dashed) line in Fig. 1, there is a great deal of scatter.[15] In the optimal and overdoped materials, there is a clear departure from the linear relation, as has been previously noted in other works.[8] The in-plane values of $\rho_{s0}/8$ determined by microwave techniques for the strongly underdoped $YBa_2Cu_3O_{6.333}$ material are shown and display a clear $\rho_{s0} \propto T_c^2$ behavior.[10–13] As Fig. 1 illustrates, there is no simple scaling relation between ρ_{s0} and T_c that is valid over the entire range of electronic dopings.

The values for $\rho_{s0}/8$ in the copper-oxygen planes in Fig. 1 have been replotted as a function of $\sigma_{dc}T_c$ in Fig. 2 [in the optical and microwave measurements, $\sigma_{dc} \equiv \sigma_1(\omega \to 0)$ at $T \gtrsim T_c$]. The dashed line in Fig. 2 is the best fit to the data, $N_c = \rho_{s0}/8 \simeq 4.4\sigma_{dc}T_c$, while the dotted line is the calculated result $N_c = \rho_{s0}/8 \simeq 8.1\sigma_{dc}T_c$ for a BCS superconductor where the normal-state scattering rate $1/\tau$ (taken at $T \gtrsim T_c$) is greater than the isotropic gap 2Δ in the weak-coupling limit;[15] this is equivalent to the so-called "dirty-limit" condition that $l \lesssim \xi_0$, where $l = v_F\tau$ is the mean free path and $\xi_0 = \hbar v_F/\pi\Delta_0$ is the coherence length (v_F is the Fermi velocity). The importance of this result lies in the observation that for $T \ll T_c$ there is always a dramatic suppression of the low frequency optical conductivity.[22] This "missing area" under the conductivity curve upon entering the superconducting state is a consequence of the transfer of normal-state spectral weight into the condensate; the fact that this transfer may be observed at all is due to the self-consistent condition that $1/\tau \gtrsim 2\Delta_0$ (Ref. 23). From this argument it is a relatively straightforward matter to construct a geometric scaling relation based on the transfer of spectral weight that yields the observed $\rho_{s0}/8 \propto \sigma_{dc}T_c$ dependence.[15] This is an important result in that it allows statements to be made about the nature of the superconductivity, and negates arguments based on the assumption that $1/\tau \ll 2\Delta$ ($2\Delta_0$ in a *d*-wave system) that assert that the scaling relation only contains information about the normal state. An interesting trend in the scaling of the microwave data is an increase in the slope at lower dopings; however, if the last three points are neglected (dopings for which $T_c \lesssim 6$ K), then this trend becomes less noticeable. Overall, the in-plane microwave data for $YBa_2Cu_3O_{6.333}$ agrees quite well with the optically determined results for other underdoped materials and provides a region of substantial overlap, illustrating that microwave techniques provide a complementary method for the determination of ρ_{s0} and σ_{dc} (this is especially useful when ρ_{s0} may be too small to be determined accurately using optical techniques). In addition, the data extend the validity of the scaling within the copper-oxygen planes by nearly an order of magnitude. In comparison, the Uemura scaling shown in Fig. 1 constitutes is only about half a cycle in Fig. 2.

The values for $\rho_{s0,\alpha}$ are plotted against $\sigma_{dc,\alpha}T_c$ in Fig. 3, where α denotes either the *a-b* plane or *c* axis direction (the regions encompassed by the microwave data are denoted by

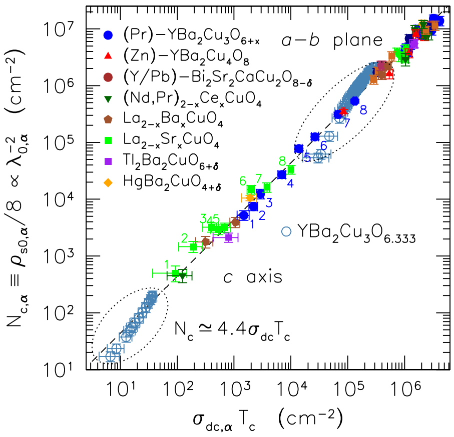

FIG. 3. (Color online) The log-log plot of the spectral weight of the superfluid density $N_{c,\alpha} \equiv \rho_{s0,\alpha}/8$ vs $\sigma_{dc,\alpha}T_c$, where α denotes either the *a*-*b* plane or the *c*-axis direction, for a variety of electron- and hole-doped cuprates. The dashed line corresponds to $\rho_{s0,\alpha}/8 \simeq 4.4\sigma_{dc,\alpha}T_c$. The *a*-*b* plane data for strongly-underdoped $YBa_2Cu_3O_{6.333}$ sample merges smoothly with the existing optical data and provides a substantial region of overlap with the *c* axis results. In addition the scaling along the *c* axis has been extended by nearly an order of magnitude. The numbers next to the *c*-axis data for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+y}$ correspond to different chemical compositions (electronic doping) (Refs. 14–16).

the enclosed regions). As previously shown in Fig. 2, the results merge smoothly with existing in-plane data and extend the results so that there is now a significant overlap with the *c* axis data. Furthermore, the results for $YBa_2Cu_3O_{6.333}$ along the *c* axis extend the overall scaling by a further order of magnitude so that the general scaling relation is now valid over nearly six orders of magnitude. As previously observed for the in-plane results, the last two points of the most strongly underdoped data ($T_c \lesssim 6$ K) along the *c* axis may also fall slightly below the scaling line. It has been previously noted that the scaling observed in the *a*-*b* planes and along the *c* axis is the same; $N_c \equiv \rho_{s0}/8 \simeq 4.4\sigma_{dc}T_c$ (Ref. 14). This is surprising given that the normal-state transport in the planes is coherent and the superconductivity is accompanied by the formation of an energy gap with *d*-wave symmetry,[24,25] while the normal-state transport perpendicular to the copper-oxygen layers along the *c* axis is governed by hopping and the superconductivity is due to the Josephson effect.[26,27] It has been previously demonstrated[15,16] that a BCS superconductor in the weak-coupling limit yields the same linear scaling relation for both the dirty limit (*a*-*b* planes) as well as for tunneling between the planes due to the Josephson effect (*c* axis); $\rho_{s0}/8 \simeq 8.1\sigma_{dc}T_c$ (note that the calculated constant is somewhat larger than the experimentally observed value). Until this point, it was never possible to reduce the electronic doping in the copper-oxygen planes to the extent that there was any region of overlap between the *ab* planes an the *c* axis data. However, as Fig. 3 demonstrates, the electronic doping in $YBa_2Cu_3O_{6.333}$ has been strongly reduced, extending the in-plane results by nearly an order of magnitude and creating a substantial region of overlap with the *c*-axis data. This suggests that in the most strongly underdoped case the superconductivity in the copper-oxygen planes may be inhomogeneous and dominated by Josephson effects (a "Josephson phase"), evolving with increasing doping into a more uniform superconductor.

Such a progression is not difficult to envision. It is generally accepted that the underdoped cuprates are electronically inhomogeneous,[28–31] and some materials even display static charge- and spin-stripe order in which the material is segregated into hole-rich and hole-poor regions;[32,33] such a scenario has also been described in terms of frustrated phase separation.[34] We speculate that the strongly underdoped materials are electronically segregated into superconducting hole-rich regions and hole-poor regions that form a poorly conducting barrier region. If the "granularity" of such a system is fine enough, then the superconducting regions will be linked through the Josephson effect,[35] in essence forming a Josephson phase,[36] and in fact it has recently been demonstrated that the scaling relation observed here can be derived for a two-dimensional Josephson array.[37] Within this framework the doping level may in principle be reduced to such an extent that Josephson coupling between the superconducting regions is no longer possible. In practice, this would correspond to doping levels below ~0.05 holes per planar copper atom. At this critical doping one would also expect that the sheet resistance would approach $h/4e^2 \simeq 6.9$ kΩ, close to the value of $R_\square \simeq 5.6$ kΩ observed at the lowest doping. However, as the electronic doping is increased the material becomes more homogeneous, the size of the superconducting regions increases and the "granularity" is reduced, allowing the system to revert to a more conventional behavior. It should be noted that in this "large-grain" picture, the Uemura relation is expected to be recovered,[36] suggesting that this is a reasonable description of the moderately underdoped region.

In summary the superfluid density in strongly underdoped $YBa_2Cu_3O_{6.333}$ is found to follow the general scaling relation $\rho_{s0}/8 \simeq 4.4\sigma_{dc}T_c$, extending the validity of the scaling in the copper-oxygen planes and the *c* axis by nearly an order of magnitude and providing a region of substantial overlap between the *a*-*b* plane and *c* axis data. We speculate that in-plane response in the strongly underdoped region is electronically inhomogeneous and that the superconductivity in this region may constitute a Josephson phase. However, as the doping is increased a more homogeneous electronic state emerges and the superconductivity becomes more uniform.

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