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Superfluid density in cuprate high-T_c superconductors: A new paradigm

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We show that the Uemura relation, namely T_c proportional to superfluid density ρ_s, is not generally satisfied in high-T_c superconductors. Instead, a modified plot of T_c/Δ_0 versus ρ_s where Δ_0 is the maximum *d*-wave gap at $T=0$, exhibits universal features that point to an alternative interpretation of the underlying physics. In the underdoped region this plot exhibits the canonical negative curvature expected when a ground-state correlation competes with superconductivity (SC) by opening up a gap (the pseudogap) in the normal-state density of states. Moreover we observe simple linear relations between $T_c\rho_s$, the electronic entropy at T_c, and the critical impurity concentration for suppressing SC. These relations confirm that the pseudogap persists to $T=0$, strongly modifying the SC ground state.

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While there is as yet no agreed theory for high-T_c superconductors (HTS's) several systematic trends in their phenomenology have been discerned. One of these is the universal relation $T_c = \text{const} \times \rho_s(0)$ between the superconducting (SC) transition temperature T_c and the superfluid density, often referred to as the Uemura relation.[1,2] The superfluid density $\rho_s(T) \equiv \lambda_{ab}(T)^{-2} = \mu_0 e^2 n_s/m^*$, where μ_0 is the vacuum permeability, e the electronic charge, n_s is the density of superconducting electrons, m^* is their effective mass, and λ_{ab} is the in-plane London penetration depth. $\rho_s(T)$ is a measure of the phase stiffness of the condensate[3] and plays a major role in governing the irreversibility field.[4] A quasilinear relation between T_c and ρ_s seems to be broadly applicable to underdoped cuprates irrespective of their structural type and maximum transition temperature $T_{c,\max}$. As a consequence, the Uemura relation has been extremely influential and is invoked, for example, in support of Bose-Einstein condensation of real-space pairs,[2,5] precursor pairing models,[3,6] holon condensation in a spin-charge separation model[7] and is generally discussed as a test of theoretical models.[8]

Since Uemura's pioneering work[1,2] we have obtained new data which suggest a fundamentally different relation for the doping-dependent trends of $\rho_s(0)$ in HTS's. (We do not consider the wider class of materials discussed by Uemura.[2]) To set the scene we show in Fig. 1 values of T_c plotted versus $\rho_s(0)$ for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ (Y-123), $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212), and $La_{2-x}Sr_xCuO_4$ (La-214). The $\rho_s(0)$ data for Y-123 are from muon spin relaxation (μSR) studies,[9] for Bi-2212 were obtained from field-dependent thermodynamic measurements,[10] while the La-214 data are from ac susceptibility and μSR studies.[11] The doping level, expressed as p holes/planar Cu, is just the magnitude of x in $La_{2-x}Sr_xCuO_4$ and, for Y-123 and Bi-2212, is conveniently determined[12] from the approximate empirical relationship $T_c = T_{c,\max}[1 - 82.6(p-0.16)^2]$. In each case maximum T_c occurs at *optimal doping* $p_{\text{opt}} \approx 0.16$, while the maximum superfluid density occurs at *critical doping*[9,11] $p_{\text{crit}} \approx 0.19$. The open symbols in the figure denote $\frac{1}{8}$, optimal and critical doping.

Figure 1 reveals several important features. First, the range of initial slopes shows there is no single universal Uemura line that applies to all HTS cuprates. Second, the dotted line for Y-123 and La-214 shows that $T_c \propto \rho_s(0)$ is not sustained, and instead all our data exhibit a negative curvature from the lowest doping to p_{crit}. In particular, T_c changes little between $p \approx 0.125$ and p_{crit} while ρ_s nearly doubles. It is the origin of this negative curvature below p_{crit} that the present work addresses. Within this general trend, each dataset exhibits deviations below $\frac{1}{8}$th doping which may be associated with incipient stripe instabilities. These anomalies, and a paucity of data points, possibly gave the impression of linearity in earlier data. The behavior for $p > p_{\text{crit}}$ seems more complex since ρ_s falls anomalously[9] in Y-123 and T1-1212 but shows little change in Bi-2212 and La-214.[11] Leaving aside the overdoped region, it is clear from Fig. 1 that the Uemura relation is not universal and, further the assumption of linearity is questionable and may conceal important systematic trends.

It is well established that for $p < p_{\text{crit}}$ the normal-state (NS) density of states (DOS) $N(E)$ is depleted due to the presence of the pseudogap.[13] The pseudogap energy scale E_g rises with progressive underdoping and the loss of spectral weight strongly modifies all NS and SC properties. These effects persist to high temperatures and should not be confused with precursor effects which occur near T_c.[14] For example, the ''lost entropy'' associated with the pseudogap is not even partially recovered to at least 300 K nor the ''lost susceptibility'' to at least 400 K.[10] Many interpretations of the pseudogap have been advanced including various forms of precursor pairing[3,6] or, as we have argued, independent correlations which modify the ground state.[13,15] Here we show that the general features of the Uemura plot can be explained by such a competing pseudogap and we find a linear relation between $\rho_s(0)$ and the electronic entropy at T_c which helps confirm this scenario.

In Fig. 2 we compare the doping dependence of $\lambda_0^{-2} T_c$ with that of the electronic entropy $S(T_c)/R$ for Y-123, Bi-2212, and La-214.[10,16] Here R is the gas constant. With the exception of overdoped La-214 both quantities agree very well across the entire doping range. This correlation is remarkable because it demonstrates a link between a true $T=0$ ground-state property $\rho_s(0)$ and a finite-temperature

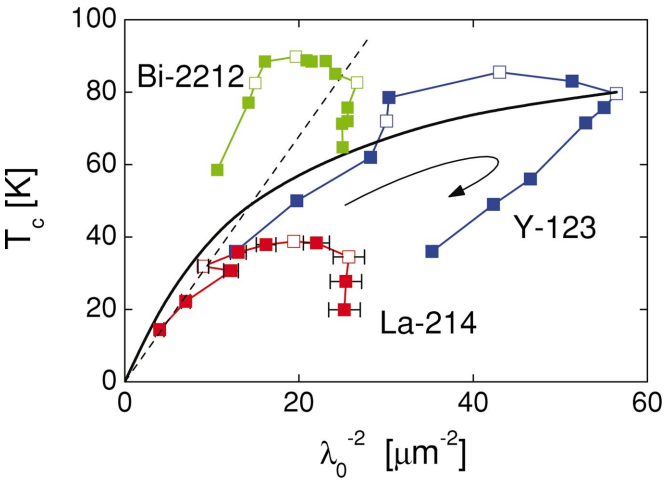

FIG. 1. (Color) T_c plotted as a function of superfluid density $\lambda_0{}^{-2}$ for La-214, Bi-2212, and Y-123. The arrow indicates increasing doping. Open data points denote 1/8th, optimal and critical doping. The dashed straight line shows the Uemura relation $T_c \propto \rho_s(0)$ and the solid curve is the calculated behavior when a competing NS correlation opens up a gap in the DOS.

normal-state thermodynamic property $S(T_c)/T_c$, whose magnitude we know is strongly suppressed by the normal-state pseudogap[10,13] We conclude then that the superfluid density is also suppressed by the pseudogap and our goal now is to calculate its quantitative effect on T_c and $\rho_s(0)$.

Assuming the NS gap to be non-states-conserving with triangular energy dependence pinned to the Fermi level E_F as indicated by specific-heat measurements,[17,10] we have solved standard weak-coupling *d*-wave BCS expressions using the normal-state DOS:

$$N(E) = N_0 |E - E_F| / E_g(p) \quad \text{for } |E - E_F| < E_g(p),$$
$$= N_0 \quad \text{for } |E - E_F| > E_g(p). \tag{1}$$

For this DOS $T_c \to 0$ as $E_g \to 2.397 k_B T_c^0$, where k_B is Boltzmann's constant and $T_c^0 = T_c(E_g = 0)$. Figure 3 shows T_c plotted as a function of $\zeta = E_g/(2.397 k_B T_c^0)$. The depression in T_c is slow at first and more rapid as $\zeta \to 1$. The superfluid density may be calculated from[18]

$$\rho_s = 1/\Omega \sum [2(\partial \varepsilon_k / \partial k_x)^2 \partial f(\Gamma_k)/\partial \Gamma_k - (\partial^2 \varepsilon_k / \partial k_x^2)$$
$$\times (\varepsilon_k / \Gamma_k) \tanh(\Gamma_k / 2 k_B T)], \tag{2}$$

where the sum is over momentum vectors **k**, x is the direction of superfluid current, Ω is the energy cutoff, $f(E)$ is the Fermi function, ε_k is the quasiparticle energy dispersion, and $\Gamma_k = \sqrt{(\varepsilon_k^2 + \Delta_k^2)}$. We assume this triangular NS DOS and solve the BCS gap equation to obtain $\Delta_k(T)$ and hence $\rho_s(T)$ from Eq. (2). Such a Fermi-liquid approach is open to criticism but seems to be valid at lower temperatures where the nodal regions dominate and quasiparticles have long lifetimes. $\rho_s(\zeta)$, plotted as a function of ζ in Fig. 3, exhibits an initial rapid fall which slows as E_g grows and $\zeta \to 1$. From Fig. 3 we construct an Uemura plot $T_c(\zeta)$ versus

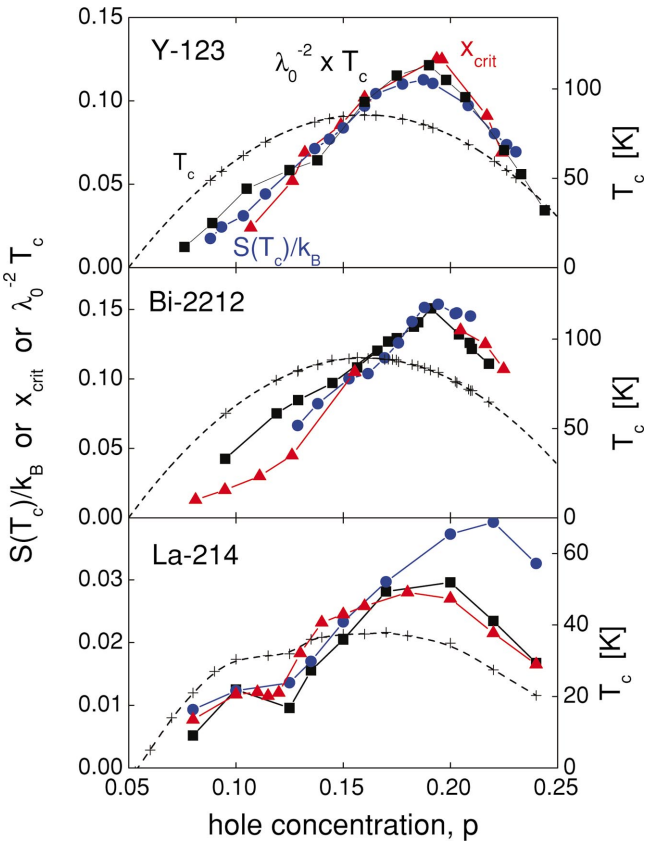

FIG. 2. (Color) The doping dependence of $T_c(p)$ (dotted curves) the electronic entropy $S(T_c)/k_B$ (blue circles), and of $T_c \lambda_0{}^{-2}$ (black squares) for pure Y-123, Bi-2212, and La-214. Also shown is the critical density x_{crit} of Zn (or of Co for Bi-2212) for suppressing SC (red triangles).

$\rho_s(\zeta)$ shown by the solid curve in Fig. 1. This has an implicit dependence on E_g and exhibits a canonical negative curvature.

As noted, the data show a similar trend, but with some differences. Specifically, the data deviate from this curve between p_{crit} and p_{opt} where the observed T_c rises to a maximum. This occurs because the doping-dependent SC gap Δ_0 is itself increasing here. This problem, that T_c is governed

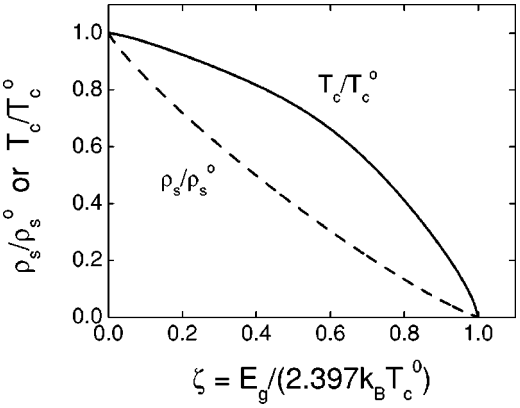

FIG. 3. The calculated normalized reduction in superfluid density and T_c arising from a "triangular" gap in the NS DOS. E_g is the temperature-independent NS gap.

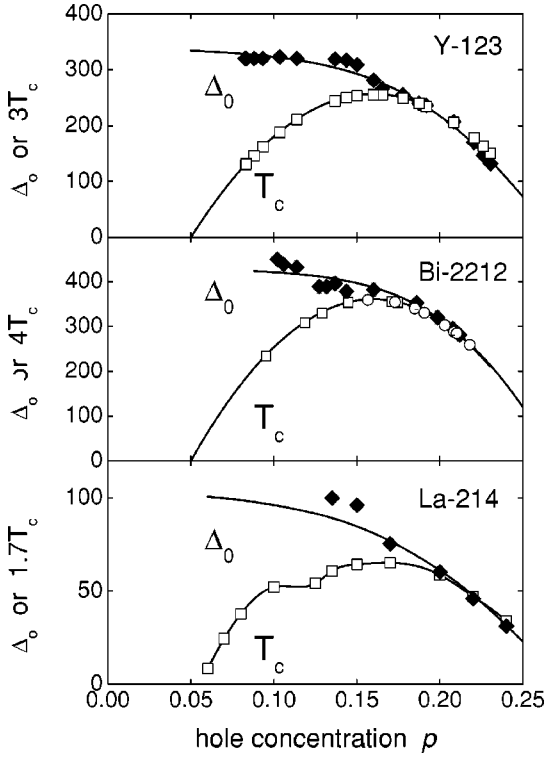

FIG. 4. The doping dependence of T_c (open squares) and Δ_0 (filled diamonds) for Y-123, Bi-2212, and La-214. The solid curves are fits as described in the text.

both by E_g and Δ_0 while $\rho_s(0)$ is controlled only by the magnitude of E_g, is resolved by employing the ratio T_c/Δ_0 which depends only on E_g. We thus recast the Uemura plot as T_c/Δ_0 versus $\rho_s(0)$. Note that, here, Δ_0 is the maximum *spectral gap*, and not the SC order parameter.[13] Our next goal, then, is to determine the ratio T_c/Δ_0.

Figure 4 shows the p dependence of T_c and Δ_0. For Y-123 and La-214 Δ_0 is found from specific-heat studies[10,16] using the low-T linear slope in the specific-heat coefficient $\gamma(T)$:

$$\gamma(T)-\gamma(0)=3.14\gamma_n k_B T/\Delta_0. \tag{3}$$

Here γ_n is the NS value of $\gamma(T)$. Δ_0 values for Bi-2212 in Fig. 4 are obtained from B_{1g} Raman scattering.[19] Figure 4 shows that for $p>p_{\rm crit}$ the ratio T_c/Δ_0 remains constant and only begins to decrease with the opening of the pseudogap as p falls below $p_{\rm crit}$. This is precisely what is predicted by our model calculation. To parametrize the data we fit Δ_0 to the form $\Delta_0(p)=\Delta_0(0)\tanh[\alpha(0.27-p)]$ (see solid curves in Fig. 4) and using this we present a modified Uemura plot in Fig. 5 of the form T_c/Δ_0 versus $\rho_s(0)$ for the three HTS materials.

The plots in Fig. 5 each display three regions. There is now good agreement between data and calculation (solid red curve) from about $\frac{1}{8}$th doping to critical doping. This confirms that it is the pseudogap that dominates T_c and $\rho_s(0)$ here. The second region $p>p_{\rm crit}$ appears to be nonuniversal as noted above. In the third region, $p<\frac{1}{8}$, $\rho_s(0)$ is higher than expected. We suggest this is due to the abrupt partial filling of the pseudogap, observed in this region from either the entropy[15] or the Knight shift,[20] and possibly associated

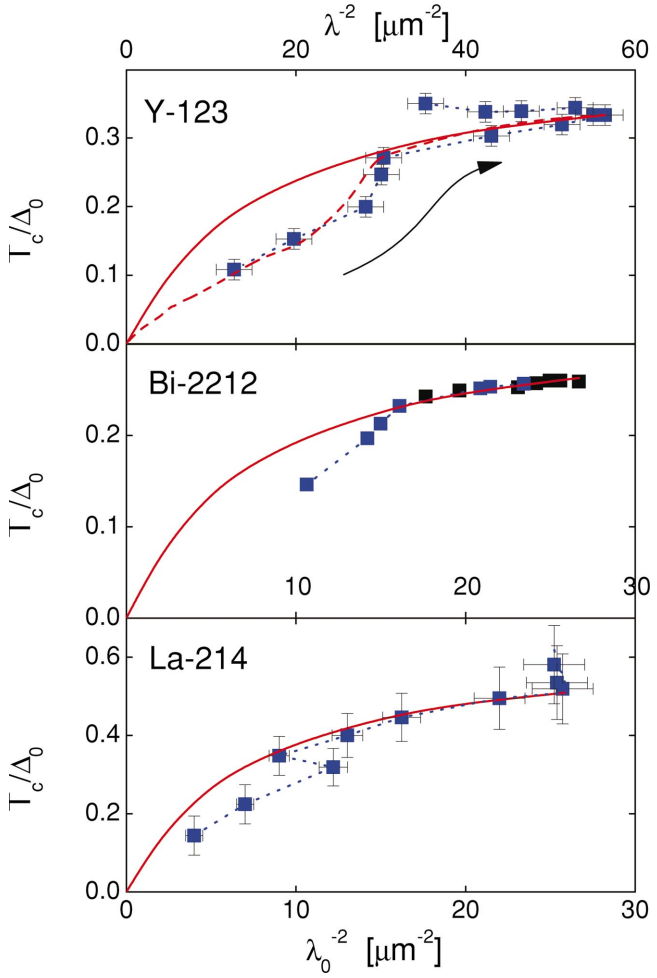

FIG. 5. (Color) Modified Uemura plots in the form of T_c/Δ_0 plotted as a function of λ_0^{-2}. The red solid curves are the calculated behavior within the pseudogap model. The dashed red curve shows the gap-filling model calculation. The arrow indicates increasing doping.

with incipient stripe instabilities breaking up the pseudogap state. Singer and Imai[21] have also recently drawn attention to this gap filling. To illustrate that this can account for the detailed behavior of $\rho_s(0)$ we have repeated our calculation using a triangular gap, $N(E)=N_1+(N_0-N_1)|E-E_F|/E_g$ with a finite DOS N_1 at E_F. We take N_1 to increase linearly with doping then fall abruptly to zero at $p\approx 0.12$, as observed.[15] The result, shown by the dashed red curve in Fig. 5(a), follows the data well enough to illustrate that the canonical pseudogap behavior is probably present for all $p<p_{\rm crit}$, including $p<\frac{1}{8}$.

We return now to the empirical relation, shown in Fig. 2, between λ_0^{-2} and $S(T_c)/T_c$. This can be understood if $\rho_s(0)$ is expressed in terms of Fermi-surface parameters using[22]

$$\rho_s(0)=\mu_0 e^2\langle \nu_x^2 N(E)\rangle, \tag{4}$$

where ν_x is the Fermi velocity projected in the supercurrent direction and the average is taken over an energy shell $E_F\pm\Delta_0$. As $\partial\varepsilon_k/\partial k$ is rather constant near the Fermi surface[23] ν_x^2 may be shifted outside the brackets giving $\rho_s(0)\propto\langle N(E)\rangle\propto(S/T)_{T_c}$, where $(S/T)_{T_c}$ is the average of γ from

0 to T_c. Thus the p dependence of $\rho_s(0)T_c$ is the same as that of $S(T_c)$, as demonstrated in Fig. 2. Taking $\nu_F^2=2\nu_x^2$ where ν_F is the Fermi velocity, we have in detail

$$\lambda_0^{-2}T_c=3\mu_0 e^2\nu_F^2/(\pi^2 k_B 2V_a)S(T_c)/R, \quad (5)$$

where V_a is the volume of the unit atomic cell. For Y-123 Fig. 2 shows $T_c\lambda_0{}^{-2}=(3.7\times10^{16})[S(T_c)/R]$ in mks units which, using Eq. (5), yields $\nu_F=1.35\times10^5$ m/s, in good agreement with other estimates.[8] Using the relation $\xi_0=\hbar\nu_F/\pi\Delta_0$ and taking for Δ_0 the angular average $\Delta_0/\sqrt{2}$ with $\Delta_0=270$ K at $p=p_{\rm crit}$ (Ref. 10) one finds $\xi_0=1.7$ nm, as observed.[24]

Alternatively, the electronic entropy may be calculated from[13]

$$S(T)=-2k_B\int [f\ln f+(1-f)\ln(1-f)]N(E)dE, \quad (6)$$

where $f(E/k_BT)$ is the Fermi function. Using the NS DOS in Eq. (1) we find that $S(T_c)/T_c$ has almost the same dependence on $\zeta\equiv E_g/(2.397k_BT_c^0)$ as does $\rho_s(0)$ from Eq. (2), in agreement with our empirical results in Fig. 2.

Another important observation shown in Fig. 2 is the numerical agreement between $S(T_c)/R$ and the critical concentration $x_{\rm crit}$ of impurity scatterer needed to just suppress superconductivity. Here the impurity is Zn for La-214 and Y-123 (Refs. 25 and 26) and Co for Bi-2212 (Ref. 27). For a *d*-wave order parameter, and under the assumption that Zn is a unitary scatterer, $x_{\rm crit}=1.3\langle N(E)\rangle\Delta_0$ (Refs. 24 and 25). Since $\langle N(E)\rangle\Delta_0$ is the pair density, this implies that SC is destroyed when the density of unitary scatterers approximately equals the SC pair density, i.e., each scatterer breaks one pair.[22] We therefore conclude that $S(T_c)/R$ equals the density of pairs at $T=0$ in the Zn-free material, as expected if the pseudogap reflects a loss of normal-state spectral weight. If on the other hand T_c and $S(T_c)$ were limited by thermally induced SC fluctuations then $S(T_c)/R$ is much less than the density of pairs at $T=0$ and would not equal $x_{\rm crit}$.[22] The results of Fig. 2 are thus summarized by

$$\lambda_0{}^{-2}T_c\propto S(T_c)/R=x_{\rm crit}. \quad (7)$$

A central conclusion here is that the doping-dependent magnitude of ρ_s not only correlates with, but may also be estimated *numerically* from S/T_c. This ties the p dependence of ρ_s closely to that of the pseudogap and, as noted, provides a link between a ground-state SC property $\rho_s(0)$ and a finite-temperature normal-state property $S(T_c)$. Such a link naturally arises from the presence of a competing normal-state correlation which persists to $T=0$. In the precursor pairing model of Levin and co-workers[6] a two-gap ansatz of the form

$$\Delta(\mathbf{k})^2=\Delta'(\mathbf{k})^2+E_g(\mathbf{k})^2 \quad (8)$$

is employed (see also Ref. 13). While E_g in this model may be relatively T independent above T_c, its magnitude below T_c falls to zero as $T\to0$. In such a case $S(T_c)/T_c$ is diminished by precursor pairing but $\rho_s(T=0)$ is not. Thus, within their model, there is no reason for the observed direct correspondence between these two quantities. We believe that this link between ρ_s and $S(T_c)/T_c$ implies that E_g remains finite at $T=0$, i.e., the pseudogap coexists with SC even at $T=0$.

In summary we have shown that the Uemura relation is not universal in HTS cuprates. Moreover, we observe simple numerical relations between $T_c\rho_s$, $S(T_c)$, and $x_{\rm crit}$ which place strong constraints on the nature of the pseudogap. A Fermi-surface model of the pseudogap which successfully accounts for these correlations predicts a sublinear dependence of T_c versus ρ_s over the entire underdoped regime which agrees better with experiment than the usually assumed linear dependence. With the opening of the pseudogap at $p_{\rm crit}$ the superfluid density ρ_s falls rapidly at first while T_c/Δ_0 falls at first slowly—the canonical variation expected if pseudogap correlations coexist with superconducting correlations in the ground state.

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