Why the superfluid density tracks $T_c$ in cuprate superconductors?

E. V. L. de Mello

1 Instituto de Física, Universidade Federal Fluminense, 24210-346 Niterói, RJ, Brazil

One of the first finding concerning the superconducting (SC) density $n_{sc}$ in cuprates was the small magnitudes that revealed the importance of phase fluctuations. More recently, measurements in a variety of overdoped cuprates indicate that it is also much smaller than expected from BCS theories and falls smoothly to zero as doping is increased. We explain these observations by an electronic phase separation theory with a Ginzburg-Landau potential $V_{GL}$ that produces alternating charge domains whose fluctuations lead to localized SC order parameters that are connected by Josephson coupling $E_J$. The average $\langle E_J(p, T) \rangle$ is proportional to the local superfluid phase stiffness $\rho_{sc} \propto n_{sc}$. The fraction of condensed carriers decreases in the overdoped region due to the weakening of $V_{GL}$. The results agreed with $\rho_{sc}(p)$ vs. $T_c(p)$ and the Drude-like peak measurements.

Almost thirty years ago Uemura et al. performed a series of $\mu$-SR experiments and found a universal linear scaling between $T_c(p)$ and the zero temperature superfluid density $n_{sc}(p, 0)$ at low $p$. The proportionality law was not confirmed by subsequent experiments that measured a slightly parabolic behavior, but all measurements agreed with the saturation of $n_{sc}(p, 0)$ at or beyond optimum doping. Another general observation was the low superfluid densities with one to two orders of magnitude less than of conventional BCS superconductors. The small magnitudes of $n_{sc}$ are indicative that phase fluctuations play a significant role in the physics of cuprates. One possible implication is that Cooper pair formation occurs at some onset temperature, but long-range phase coherence does not occur until the temperature is lowered to $T_c$. In fact, many experiments have measured persistent SC correlations and diamagnetic responses at temperatures well above $T_c$.

Recently Božović et al. found a similar linear scaling relation on overdoped La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) films but with a negative slope, in close agreement with earlier works on Ti$_2$Ba$_2$CuO$_{6+\delta}$(T12201). Putting all experiments together, the dominant picture is an almost linear increase of $n_{sc}(p, 0)$ in the underdoped region that saturates right after optimum doping and decreases with $T_c(p)$ going down to zero in the far overdoped samples. This decreasing of $n_{sc}(p, 0)$ after saturation was not expected because doping brings more charges to the CuO planes what, in principle would increase the number of Cooper pairs in a standard BCS framework of superconductivity. A subsequent experiment found that a significant fraction of the carriers remains uncondensed in a wide Drude-like peak as $T \to 0$, while $n_{sc}(p, 0)$ remains proportional to $T_c(p)$ and vanishes in the limit of superconductivity. This experiment shows that overdoped superconductors behave like a two fluids system and helped to understand the puzzle of the “missing” carriers.

After all these years there are not a wide accepted theory to the superfluid densities in cuprates. Most likely because the ubiquitous presence of incommensurate charge ordering (CO) on cuprates introduced a degree of complexity difficult to be incorporated in any realistic theory. On the other hand, overdoped materials have been believed to be well described by Fermi liquid theory and to have a SC state with conventional BCS-like properties. While a model considering pair-breaking due to impurity scattering in a BCS like $d$-wave superconductor reproduced well the $T_c(p)$ vs. $n_{sc}(p, 0)$, it is inconsistent with the recent observation of a wide Drude peak. A subsequent work reconciled the observed broad residual Drude peak with the behavior of $n_{sc}(p, 0)$ within the Born limit. However, an infinite number of weak scatterer center (Born limit) is not reasonable considering that the scattering rate of the $T \to 0$ residual Drude is about the same of the normal state.

In this letter we provide a unified explanation to the old results, the suppression in superfluid densities in the overdoped experiment and to the residual Drude contribution. The calculations follow the method of the preceding paper and in earlier works which starting point is the simulations of charge instabilities like stripes, incommensurate charge order (CO) or charge density waves (CDW) within the framework forelectron: 17–21. It is important to emphasize that some of these experiments provide strong indications that the pseudogap (PG) is correlated to the charge instabilities and this was confirmed by specific calculations. Based on these results, we apply the method to the overdoped regions where CO is more difficult to be detected.

In short, to define the important variables and parameters, our approach is based on the time-dependent Cahn-Hilliard (CH) nonlinear differential equation via a Ginzburg-Landau (GL) free energy expansion in terms of a diffusive or phase separation order parameter $u(r, t)$ that evolves in time $t$. As discussed above, at $T \sim T^*$, the GL free energy potential $V_{GL}(r, t)$ starts to segregate the charge wave functions in superlattices formed by alternating hole-rich and hole-poor domains, with different CO wavelength $\lambda_{CO}$ and structures. The non-uniform charge distribution interacts with the Cu atoms electronic clouds and may induce local SC pairing interactions. This fundamental point is discussed in some detail in Ref. [21].

To calculate $T_c(p)$ we have performed Bogoliubov-deGennes calculations on various CO density maps what yields local amplitudes $\Delta_d(r)$ with the same wavelength $\lambda_{CO}$. The localized $\Delta_d(r)$ is in agreement with SC coherence lengths $\xi$ typically smaller than average $\lambda_{CO}$, what implies that the charge domains may behave as mesoscopic SC grains. This gives rise to Josephson coupling between the distinct SC regions with energy $E_J(r_{ij})$ that is the lattice version of the local superfluid density $E_{J}(r_{ij})$ that is the lattice version of the local superfluid density $\rho_{sc}$. We have already explained that for two $d$-wave superconductors junction is
where $R_n(p)$ are taken to be proportional to the $T \lesssim T_c$ normal-state in-plane resistivity $\rho_{ab}(p)$ obtained from typical $\rho_{ab}(p,T) \times T$ curves for Bi2212, LSCO and Y123. The values for Bi2201 are the same used in Ref. [24]. The proportionality constant between $R_n$ and $\rho_{ab}$ is found matching the optimal $T_c(p = 0.16)$. The same constant is used between all $\rho_{ab}(p)$ and $R_n(p)$ and we list some $R_n(p)$ in Table I. The spatial average $\langle \Delta_d(p,T) \rangle \equiv \sum_i \Delta_d(r_i,p,T)/N$ contains all the planar sites.

We evaluate $\langle E_1(p,T) \rangle$ for the Bi$_{2-\delta}$Pb$_\delta$Sr$_{2-y}$La$_y$CuO$_{6+\delta}$ (Bi2201), LSCO, Bi$_2$Sr$_2$CaCu$_2$O$_{6+\delta}$ (Bi2212) and YBa$_2$Cu$_3$O$_{6+\delta}$ (Y123) systems and show the plots in Fig. 1(a-d). They contain the two important quantities to this work: The low temperature superfluid phase stiffness $\rho_{sc}(p,0)$ (or $\langle E_1(p,0) \rangle$) that is read directly at $T = 0$ K and $T_c(p)$ that is obtained from the intersection between the Josephson coupling $\langle E_1(p,T) \rangle$ and the thermal disorder energy $k_B T$. The results for $T_c(p)$ and $\rho_{sc}(p,0)$ for four underdoped cuprates are listed in Table I and plotted in Fig. 2. We also draw the Uemura line for reference that shows that our points are closer to a parabola than a straight line in better agreement with the LSCO and Y123 results. We also plot a slightly overdoped Ti2201 with $T_c(p)$ that is close to the optimal points.

The behavior of the $T_c(p)$ vs. $\rho_{sc}(p,0)$ may be understood by examining the Josephson energy curves $\langle E_1(p,T) \rangle$ vs. $T$ plotted in Figs. 1(a-d). In the underdoped region, $\langle \Delta_d(p,T) \rangle$ is almost constant at low $T$ and vanishes much above $T_c(p)$. This implies that $\langle E_1(p,T) \rangle$ vs. $T$ are slowly decreasing approximate straight lines (see Figs. 1(a-d)) establishing a direct relation between $\langle E_1(p,0) \rangle = \rho_{sc}(p,0)$ and $\langle E_1(p,T_c) \rangle = k_B T_c(p)$ that is not strictly linear but is close to Uemura’s originated proposal (see Fig. 2). When $p$ gets near the optimum doping $p_{opt}$, $T^*(p)$ and $\langle \Delta_d(p,T) \rangle$ diminish with $T$ and $\langle E_1(p,T_c) \rangle$ vs. $T$ decrease faster and some series saturates near $p \geq 0.13$. On the other hand, the Bi2201 series have very steep $T^*(p)$ vs. $p$ and their $\langle E_1(p,T_c) \rangle$ are almost horizontal lines and does not saturate up the optimal doping. Thus, whenever $T^*(p)$ becomes closer to $T_c(p)$, $\langle E_1(p,T_c) \rangle$ will have steeper slopes and $\rho_{sc}(p,0)$ vs. $T_c(p)$ saturates and may decrease with $p$. This behavior was observed in the cuprates studied by Uemura et al.

Table I. Selected values of $T_c(p)$ estimated from the Josephson couplings given in Fig. 1 for the four cuprate families. The second lines give the low temperature superfluid densities $\rho_{sc}(0)$. The third lines give the normal resitivity just above $T_c(p)$ that enters in Eq. (1) and are proportional to the experimental measured values of $\rho_{ab}$ just above $T_c$. $\Delta_{GL}(p,0)$ (or $\langle E_1(p,0) \rangle$) and $B$ from $R_n = B \times \rho_{ab}$ are the only two adjustable parameters of each series and are in bold blue.

| $p$ (LSCO) | 0.06 | 0.08 | 0.10 | 0.12 | 0.14 | 0.16 |
|-----------|------|------|------|------|------|------|
| $V_{GL}$ (meV) | 318 | 294 | 275 | 267 | 248 | 234 |
| $\langle \Delta_{GL}(0K) \rangle$ (meV) | 18.7 | 16.2 | 18.1 | 18.3 | 17.5 | 16.8 |
| $T_c$ (K) | 6.7 | 19.5 | 29.4 | 35.4 | 39.8 | 41.9 |
| $\rho_{sc}(0)(K)$ | 7.0 | 25.0 | 38.5 | 44.5 | 60.6 | 90.1 |
| $R_n$ (m$\Omega$cm$^{-1}$) | 0.790 | 0.281 | 0.175 | 0.127 | 0.092 | 0.067 |

From the above arguments, we expected in the overdoped region a different behavior because $T^*(p)$ diminishes and becomes closer to $T_c(p)$ and, concomitantly the SC maximum gap $\Delta_0(p)$ goes down to zero. In fact, measurements on three overdoped Ti2201 compounds revealed a decreasing straight line with lower slope than Uemura’s line. A more complete set of data was taken recently on overdoped LSCO compounds by Božović et al. and found a similar linear behavior of the old Ti2201 data. They developed a technique to grow homogeneous overdoped films with less than 1% variations in $T^*$.
FIG. 2. The values of $T_c(p)$ and $\rho_{sc}(p,0)$ derived directly from the plots of Fig. 1 and listed in Table I. The curves are not linear at low $p$ as measured by Ref. [11] but close to experimental results of Ref. [3]. A TI2201 slightly overdoped sample with $T_c = 84$ K is included. We also draw the linear Uemura proposal and the arrow points to the direction of increase $p$.

To deal with their measurements, we extend our calculations of $\rho_{sc}(p,0)$ to the overdoped LSCO films. Like for the underdoped systems, $\langle V_{GL}(p, T) \rangle$ has no dimension and we need to multiply it by a constant to define the attractive pairing potential $\langle V_{GL}(p, T = 0) \rangle$ in eV units. We adjust $\langle V_{GL}(p_0 = 0.16, T = 0) \rangle$ to reproduce the measured SC gap $\Delta_{sc}(p_0 = 0.16) = 0.16)$, and all the others $\langle V_{GL}(p, 0) \rangle$ follow without additional parameters.

FIG. 3. The calculations of $\langle \Delta_d(T) \rangle$ and $\rho_{sc}(T)$ for overdoped LSCO. (a) $\langle \Delta_d(T) \rangle$ as function of $T$ for several compounds. Notice that it remains finite above $T_c$. (b) The average Josephson energy $\langle E_J(p,T) \rangle/k_B$ as function of $T$ uses the same resistivity measured in Ref. [14]. The intersections with $T$ yield $T_c(p)$.

Table II. Properties of overdoped LSCO. The calculated quantities are shown in red. $\varepsilon(p)$ is the CH parameter that accounts for the decreasing PG strength that influences $\langle V_{GL}(p) \rangle$. $V_{GL}(p = 0.16) = 0.247$ eV exactly as used in Table I. $B = 8.0$ from $R_n = B \times \rho_{sc}(p = 0.16) = 8.0 \times 0.10) = 0.01391$ is the only adjustable parameter for the entire overdoped LSCO series and with $V_{GL}(p = 0.16)$ are in bold blue. The calculate $\langle \Delta_d(0K) \rangle$, $\rho_{sc}(0)$, $T_c$ and the resistivity $\rho_{sc}$ are in red. The experimental quantities measured in Ref. [14] are in parenthesis and in black color for comparison. The time of all overdoped simulations were with $t \sim 7000 t_n$.

| $p$ | 0.16 | 0.19 | 0.21 | 0.23 | 0.24 | 0.25 |
|-----|------|------|------|------|------|------|
| $\varepsilon(p)$ | 0.0133 | 0.01364 | 0.01379 | 0.0139 | 0.01391 | 0.01396 |
| $\langle V_{GL}(r) \rangle$ | 0.0997 | 0.0779 | 0.0593 | 0.0422 | 0.0358 | 0.0327 |
| $V_{GL}$ (meV) | 234 | 183 | 139 | 100 | 84 | 77 |
| $\langle \Delta_d(0K) \rangle$ (meV) | 16.9 | 12.15 | 7.54 | 3.19 | 1.47 | 0.33 |
| $\rho_{sc}(0)$(K) | 90.6 | 80.1 | 66.1 | 43.4 | 27.3 | 8.0 |
| $\rho_{sc}$(m$^2$/cm) | 0.10 | 0.081 | 0.059 | 0.038 | 0.028 | 0.016 |
| $\langle E_J(p,T) \rangle$ | (0.10) | (0.08) | (0.065) | (0.036) | (0.022) | (0.01) |
| $T_c$ (K) | 42.2 | 38.0 | 32.3 | 21.6 | 13.0 | 4.2 |
| (Estimate) | (41.5) | (37.8) | (31.1) | (21.2) | (13.0) | (7.9) |

because there is no Meissner effect without phase coherence. Our $\rho_{sc}(p,T)$, the lattice version of the superfluid density, is equal to $k_B T_c(p)$ at $T_c(p)$ and vanishes only when the superconducting fluctuations $(\Delta_J)$ vanishes, i.e., above $T_c(p)$. The derived $T_c(p) \times \rho_{sc}(p,0)$ from Fig. 3(b) are plotted in Fig. 4 together with the measurements of Božović et al. The agreement in the doping range $p = 0.16 - 0.24$ is almost exact. For $p = 0.24 - 0.27$ the discrepancy increases with $p$ and it is probably because the domains of charge instabilities decrease, the charge distribution becomes almost uniform, the SC amplitudes and the Josephson coupling vanish.

To show that the decreasing of $\rho_{sc}(p,0)$ in the overdoped regime is universal, we included three experimental measurements on TI2201 and three calculated Bi2212 points ($p = 0.16, 0.185$, and 0.22) based on STN data and resistivity measurements $R_n(p)$. Both TI2201 and Bi2212 points are larger than the LSCO values and were divided by 2.2 (that is their $T_c(p = 0.16)$ ratio) in order that all the $\langle E_J(p,T) \rangle$ vs. $T_c$ plots be compared in the same figure. These results indicate that the linear relation of overdoped LSCO is common to other cuprates. We draw also the Uemura line to show that $\rho_{sc}(p,0)$ for overdoped samples are generally smaller than the underdoped compounds with same $T_c(p)$ despite the factor of 2 to 3 in the doping level.

Gathering the results from Figs. 2 and 3 and from the Table I and II, we can see that an overdoped compound with $T_c \approx 20$ K has $\rho_{sc} = 25$ K and hole carriers $p = 0.08$, while an overdoped sample with the same $T_c \approx 20$ K has the same $\rho_{sc} = 43$ K but with much more carriers $p = 0.23$. Thus for a variation of almost 200% in hole density $p$, $\rho_{sc}$ is increased by only 72% and this behavior of under/overdoped materials is a common feature. The reason is the weakening of the SC pair interaction potential $V_{GL}$ with $p$ in the overdoped as schematic.
ically shown in Fig. 3. Inspecting Figs. 2 and 4 we can see that the maximum superfluid density occurs at optimal doping where it is likely that nearly 100% of the carriers are in the SC state. Taking this as reference, we can estimate the low temperature superfluid fraction of any overdoped compound by the ratio \((V_{GL}(p))/V_{GL}(0.16))\). Some values of \(V_{GL}(p)\) are in Table II from which we obtain a way to estimate the superfluid fraction \(f_{sc}(p) = \langle \rho_{sc}(p) \rangle/\langle \rho_{sc}(0) \rangle\) and the agreement is reasonable.

The calculated SC fraction \(f\) plotted vs. \(T_c\) we find a parabolic behavior as \(T_c \rightarrow 0\).

**FIG. 4.** The theoretical values of \(\rho_{sc}(p, 0)\) and \(T_c(p)\) with the experimental results of Ref. [14]. We included also three points from Bi2212 (\(p = 0.16, 0.185,\) and 0.22) using the STM data and the TI2201 results from Ref. [2] that, to be plotted together with LSCO, were divided by 2.2, their ratio of maximum \(T_c\). We draw the Uemura line just for comparison. The inset shows the SC fraction \(f_{sc}\) (empty red circles) and uncondensed carriers \(f_u = 1 - f_{sc}\) (filled red circles) together with the experimental results in empty and filled blue squares respectively.

This scenario was confirmed by recent THz optical conductivity combined with kHz range mutual inductance measurements on overdoped LSCO films. They observed that the free carriers increases as verified by the Drude peak dependence on \(p\) at low temperatures while \(\rho_{sc}\) has an opposite behavior. In the inset of Fig. 4 we plot their spectral weight of the superfluid and uncondensed carriers data in empty and filled blue squares respectively, normalized by their sum. The calculated SC fraction \(f_{sc}\) (empty red circles) and uncondensed carriers \(f_u = 1 - f_{sc}\) (filled red circles) are also plotted and the agreement is reasonable.

In their comments, they pointed out that phase separation models could explain their data if the SC regions would be embedded in a quite large normal volume fraction. For the film with \(T_c = 7\) K, nearly 95% had to be in the normal state, what was against the uniformity of \(T_c\) in their films. However, our CH-BdG phase separation approach for LSCO produces CDW-like charge modulations on the entire system as explained here and before. The increase of normal carriers is due to decreasing amplitude of \(V_{GL}(p)\) in 100% of the system as shown in the plots for \(p = 0.016, 0.21\) and 0.25 shown in Fig. 1(c) of Ref. [41]. The \(V_{GL}(p)\) decreases smoothly with doping diminishes also the values of the SC order parameter amplitude \(\Delta_d(p)\) (see Table II) and consequently the condensed carrier density, as schematically shown in Fig. 5.

**FIG. 5.** Low temperature schematic distribution of the holes and Copper pairs in the presence of the \(V_{GL}(x)\) potential, represented here along the \(x\)-direction. In the underdoped region most holes form SC pairs. In the overdoped region the modulation amplitude decreases letting most holes be free carriers and the superfluid density goes down.

We conclude pointing out that all the measured superfluid density as function of \(T_c\) are interpreted in a unified way by the \(V_{GL}(p)\) potential that promotes the charge instabilities and concomitantly, the SC interation. The decreasing of \(V_{GL}(p)\), that is correlated with the PG, reduces the CO constraint and favors free particles at the same time that the superfluid density is reduced, like schematically shown in Fig. 5. Together with the quantitative calculations of Fig. 4 we show why the superfluid density decreases in the charge abundant overdoped regime.

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