A single theory to charge order, pseudo and superconducting gap, critical temperature and pairing interaction of cuprate superconductors

E. V. L. de Mello$^1$

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

We have developed a complete theory to cuprate superconductors that calculates all their most important properties: the pseudogap (PG) $\Delta_{PG}$, the local superconducting amplitudes $\Delta_{SC}(r_i)$, the critical temperature $T_c$ and charge ordering (CO). Distinct CO are simulated by the Cahn-Hilliard differential equation with a free energy potential $V_{GL}$, that produces alternating small charge modulations. The tiny charge oscillations favor charge fluctuations that induce atomic fluctuations that mediate the SC pair interaction proportional to the $V_{GL}$ amplitude. The local SC amplitude and phase $\theta_i$ are connected by Josephson coupling $E_J(r_{ij})$ and the SC long-range order transition occurs when $\langle E_J \rangle \sim k_B T_c$. The combined wavelength $\lambda_{CO}$, $\Delta_{PG}$, $\langle \Delta_{SC} \rangle$ and $T_c$ calculations are in good agreement with a variety of experiments.

A great deal of effort has been devoted to investigating the different energy scales of high-temperature superconductors, their hole-doping dependence and, most importantly, their interconnections. These might provide a clue to the pairing strength and a key to the superconducting (SC) mechanism. Under this program Raman scattering identified vibration modes along the nodal direction ($B_{2g}$) with energy $\Delta_n(p)$ that follows closely $T_c(p)$ and a second vibration mode measured along the antinode ($B_{1g}$), identified with $\Delta_{PG}(p)$ because it correlates well with the PG temperature $T^*(p)$. Other experiments like specific heat, angle-resolved photon emission (ARPES), scanning tunneling microscopy (STM) and submicron Josephson junction tunneling identified also $\Delta_{PG}(p)$ but measured another gap function ($\Delta_\sigma(p)$) that increases slowly in the underdoped region. In the overdoped region, $\Delta_\sigma(p)$ stays close to the PG and decreases rapidly beyond $p \sim 0.20$. The connection and the role of these three energy scales in the SC state is the purpose of this letter.

To reveal this connection is of fundamental importance to consider also the hole of the ubiquitous spontaneous symmetry breaking or anomalous incommensurate charge-ordered (CO) phase.$^{10}$ In particular, it was verified that the CO wavelength is correlated with the distance between the Fermi arcs tips, establishing an intriguing connection between CO in real space and the PG in $k$-space on Bi$_2$Sr$_2$La$_2$O$_6+\delta$ (B2201). Many other experiments measure some kind of instability near $T^*(p)$, for instance, polar Kerr effect and optical polarization rotation. On the other hand, inhomogeneous magnetic-field response to muon spin rotation ($\mu$-SR), STM and measurements of charge density wave (CDW) or CO by x-ray or REXS have maximum signals near $p = 0.12$ and do not follow the increasing trend of $T^*$ when $p \to 0$, probably because of the vanishing of the available charge. However, all these observations may be regarded as distinct manifestations of an intrinsic mesoscopic electronic phase separation with onset transition near $T^*(p)$, and this is one pillar of our theory.

Nanoscale electronic phase separations are predicted theoretically by many different microscopic models, mostly based on the Hubbard Hamiltonian, like for instance Refs.~[11,28–31]. These rigorous calculations are important to endorse the phenomenon of electronic phase separation on high correlated systems like cuprates, but they neither reproduce the small variations of $\lambda_{CO}(p)$ nor the very fine charge modulations $\Delta p \approx 10^{-2–3}$, like in YBa$_2$Cu$_3$O$_{6+\delta}$ (Y123). Another important point is that charge density modulations are unambiguously present in the entire system at low temperature and even above $T_c$ according to STM data and not in puddles occupying a volume fraction. This last point appears to be in conflict with the finite CO correlation lengths, but it is because the very weak nature of these electronic modulations and their strong fluctuations.

These observations suggest that the time-dependent nonlinear Cahn-Hilliard (CH) differential equation is appropriate to cuprates because the different structures and the amplitude of the charge oscillation may be tuned slowly up to reproduce the experimental results. This is done using appropriated parameters in the CH equation and stopping the simulation time $n \cdot \delta t$ when a given charge configuration is achieved. It reproduces the measured CO weak modulations $\lambda_{CO}(p)$ and other forms of alternating hole-rich and hole poor regions on 100% volume fraction.

The CH equation is solved by a stable and fast finite difference scheme with free boundary conditions. The conserved order parameter $u$ is associated with local electronic density $p(r,t) = Au(r,t) + p$, where A controls the charge amplitude of oscillation. It is 0.0005 to the very weak $\Delta p \approx 10^{-2–3}$ variations around $p$, like what was measured in YBa$_2$Cu$_3$O$_{6+\delta}$ (Y123). Such weak charge modulation masked CDW in this system for many years and it is probably the reason to the very few charge inhomogeneities observations in the overdoped regime.

The method has also the great advantage to concomitantly provide the free energy that yields a connection between the SC interaction and the charge modulations. The CH equation is based on the Ginzburg-Landau (GL) free energy expansion in terms of the phase separation conserved order parameter $u(r,n \cdot \delta t)$, function of position and $n$ simulation time step $\delta t$:

$$f(u) = \frac{1}{2} \varepsilon |\nabla u|^2 + V_{GL}(u,T),$$

where $V_{GL}(u,T) = -\alpha |T_{PS} - T|u^2/2 + B^2u^4/4 + ...$ is a double-well potential that characterizes the rise of charge oscillations below the temperature $T_{CO}$ that is taken, according to the above, around $T^*(p)$. In general $\alpha$ and $B = 1$ and $\varepsilon$ is
Figure 1(a) shows a typical CH simulation (Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi2212) for $p = 0.16$) with a checkerboard pattern of $\lambda_{CO} \approx 6\xi_0$ where $\xi_0$ is the lattice parameters. $\Delta p \approx 10^{-2}$, and in Fig. 1(b) the $V_{GL}(r)$ map that originates this specific charge structure. We discussed already these simulations in detail and here we want to focus mainly on the SC interaction promoted by $V_{GL}(r)$ that has a double role: i- it generates non-uniform charge patterns like the checkerboard modulation displayed in Fig. 1(a) that affect the ionic electronic clouds. The small atomic oscillatory displacements are verified by several neutron and x-ray scattering experiments. ii- The hole density oscillations $\Delta p$ are very small and fluctuates strongly what is transmitted to the atomic electronic clouds that transmit back to the holes, generating a hole-hole lattice mediated interaction.

In Fig. 1(c) we plot $V_{GL}(x)$ along the $x$-direction together with some localized holes represented by the black filled circles. The planar Cu atoms are represented schematically on the top of Fig. 1(c) by the filled blue circles slightly displaced to (from) the hole-rich/poor) domains. When the temperature drops below $T^*$ the $V_{GL}(r)$ modulations increases as shown schematically in the inset of Fig. 1(b) favoring alternating charge domains like those of Fig. 1(a). These domains are large compared with $\xi_0$ and if the $V_{GL}(r)$ modulations are high enough, the holes may move around or oscillate in the domains what induce also fluctuations on the nearby Cu atoms that, like a mirror, interact back with the other holes in the same domain.

This process leads to our main assumption; the SC local hole-hole pairing interaction is proportional to the spatial average $\langle V_{GL}(p, 0) \rangle = \sum_i N V_{GL}(r_i, p, 0)/N$, where the sum is over all the planar sites. The indirect role of the lattice in the SC interaction is confirmed by the relatively large isotope effect on the onset of superconductivity below $T^*$ and also on $T_c$.

With this phenomenological potential we developed a particular type of Bogoliubov-deGennes (BdG) SC approach that converges self-consistently to the local chemical potential $\mu_i$ and the local $d$-wave amplitude $\Delta_d(r_i)$, keeping always the original CO structure fixed. This is done diagonalizing the BdG matrix with the Hubbard Hamiltonian with hopping parameters taken from ARPES and nearest neighbor attractive potential with temperature dependence from the GL method; $V_{GL}(p, T) = \langle V_{GL}(p, 0) \rangle (1 - T/T^*)^2$.

Notice that $V_{GL}(p, T)$ is defined as a function of the dimensionless phase separation order parameter $u(r, t)$ (Eq. 1) and needs to be multiplied by a dimensionless constant to be converted to energy units in the Hubbard Hamiltonian. This parameter is obtained making the low temperature CH-BdG calculations with the attractive potential $\langle V_{GL}(p, 0) \rangle$ yield the experimental optimal gap $(\Delta_d(p_{opt} = 0.16, 0)$). To other compounds, $\langle V_{GL}(p, 0) \rangle$ changes because the different $\lambda_{CO}(p)$ and is suffice to use the same constant, what gives only one adjustable parameter to all $\langle \Delta_d(p, T) \rangle$ of a given family.

These CH-BdG calculations on a charge density map like that of Fig. 1(a) yield local SC amplitudes $\Delta_d(r_i)$ inside each charge domains, in agreement with typical SC coherence length $\xi_{CO}$ smaller than typical $\lambda_{CO}$. The $\Delta_d(r_i)$ have the same modulations of the charges, what is a natural consequence of the simultaneous self-consistent approach on $\mu_i$ and $\Delta_d(r_i)$.

To extend this approach to the overdoped region we recall our pillar connecting the CO with the PG and that $T^*(p)$ vanishes only at $p = 0.27$, the end of the SC dome. This argument suggests that weak incommensurate charge modulations are also present, most likely with much weaker amplitudes, in the overdoped region. In fact, different types of inhomogeneities are observed in overdoped Bi-based families and in La-based materials that is possibly connected with charge instabilities. Electronic transport anisotropy in the CuO plane that decreases with temperature and doping, persisting up to at least $p \approx 0.22$ was recently measured and associated with a nematic phase. More recently, REXS experiments in strong overdoped Bi2201 observed CO peak signals similar to those of underdoped cuprates. They also measured a continued decrease of the CO vector $Q_{CO}$ versus doping similar to what is seen in overdoped compounds.

Taking these observations into consideration and the $\lambda_{CO}(p)$ data of La and Bi-based compounds we extend the calculation to the overdoped region. The values of $\langle V_{GL}(p, 0) \rangle$ are plotted in Fig. 2 and used again in the BdG calculations to derive the low temperature SC gaps $\langle \Delta_d(p, 0) \rangle$.
experiments and our calculations of finite \( \Delta \) tunneling SC domains and their boundaries that blocks the Cooper pairs (c) suggest that the system resistance just above the SC trans-

\[ \langle \Delta_a(p, T) \rangle \] for underdoped LSCO starting with the approximately constant \( \langle \Delta_0(p, T) \rangle \approx 17.5 \text{meV} \) that are close to the measured maximum gap \( \Delta_0(p) \) as shown in the inset of Fig. 3 b) Josephson energy \( \langle E_J(p, T) \rangle \) derived from the gaps shown in a). The intersections with \( k_B T \) yields the two sets of \( T_c(p) \). c) The same of a) for overdoped LSCO compounds. d) Josephson energy \( \langle E_J(p, T) \rangle \) derived from the gaps shown in c).

In this case \( T_c(p) \) is the long-range phase order temperature obtained by Josephson coupling between the phases \( \theta_i \) in the charge domains. We have explained previously that for a d-wave superconductor junction is sufficient to use the following s-wave analytical average Josephson coupling expression

\[ \langle E_J(p, T) \rangle = \frac{\pi \hbar \langle \Delta_a(p, T) \rangle}{2e^2 R_n(p)} \tanh \left[ \frac{\langle \Delta_a(p, T) \rangle}{2k_B T} \right], \]

where \( R_n(p) \) is taken to be proportional to the \( T \overset{\approx}{=} T_c \) normal-state in-plane resistivity \( \rho_{ab}(p) \) obtained from typical \( \rho_{ab}(p, T) \) vs. \( T \) curves. The proportionality constant between \( R_n \) and \( \rho_{ab} \) is found matching the optimal \( T_c \approx 42 \text{ K} \) or \( \Delta_0(p) \) plotted in the inset of Fig. 5 for both under and under-
doped \( La_{2-x}Sr_xCuO_4 \) (LSCO). The results are close to the measured ARPES nodal gaps \( \Delta_0(p) \) extrapolated to the antinodal direction, specific heat and STM measurements, indicating that \( \Delta_0(p) \) is a good candidate to the SC gap \( \Delta_{SC}(p) \).

Now, we have the ingredients to demonstrate the correlation between the PG and the CO through the derivation of \( \Delta_{PG}(p) \) for Y123 and Bi2201 using the measured \( \lambda_{CO} \) compiled in Ref. [10] and the \( \langle V_{GL}(p, 0) \rangle \) that we derived and plotted in Fig. 2. We equate \( \Delta_{PG}(p, 0) \) to the ground state energy of a shallow 2D well \( U = \langle V_{GL}(p, 0) \rangle \): 

\[ \Delta_{PG} = \frac{\hbar^2}{m\lambda_{CO}^2} \exp\left[-\frac{2\hbar^2}{m\lambda_{CO}^2}U\right]. \]

The results of five Y123 and four Bi2201 calculations are plotted in Fig. 3 together with the experimental data. The agreement near optimal doping is almost perfect and we emphasize that there is not any adjusted parameter in Eq. 2.
The results shown in Fig. 5 comprise all the CH-BdG calculations described previously in this paper yielding under and overdoped $T_c(p)$ for the LSCO case. The agreement with the experiments is almost perfect, it deviates only in the strong overdoped region where the PG vanishes, the system becomes almost uniform and the $\langle E_1(p, T) \rangle$ uncertainty is large. The $T_c(p)$ dome shape has a simple interpretation with Eq. 3: the competing contribution of $\langle \Delta_d(p, 0) \rangle$ that decreases steadily with $p$ and vanishes near $p = 0.27$ together with $T^*(p)$ (see inset of Fig. 5), and $R_n(p)$ that has an exponential decreasing behavior and diverges near $p = 0.05$.

Another novel interpretation that comes out of this approach is that the Cooper pairs acquire long-range order at $T \sim T_c$ and like a superfluid spreads over the charge modulation domains on the CuO planes. This superfluid uniformly the total charge density leading to a substantial decrease of the CO interference x-ray scattering signal. This was interpreted as due to the competition between the CO and the SC phase\textsuperscript{19,24}, but it is a consequence of the local Cooper pairs long-range SC transition, as it is schematically shown at the bottom of Fig. 1(c) (for $T > T_c$ and $T < T_c$).

We have shown how to calculate the pseudogap $\Delta_{PG}(p)$, the average SC gap $\langle \Delta_d(p) \rangle$ and $T_c(p)$ of different cuprates in very good agreement with experiments. To deal with many important properties discussed in the paper and reproduce accurately several quantitative results, our approach is essentially phenomenological but should provide clear guidelines to any fundamental theoretical calculation on cuprates. Another advantage of our phenomenological theory is to reveal in a simple way the connection between the most fundamental energy scales and the relation between distinct properties like, for instance, the PG and the SC interaction.

The method is general and simple to be used in any problem involving superconductivity with CDW or any other type of charge instability that, otherwise do not have a simple theoretical approach. Under this program, we will soon present calculations on the correlation between the superfluid density $\rho_s(0)$ and $T_c(p)$, the interpretation of high magnetic field quantum oscillations experiments, proximity effects, and other challenging problems of cuprates. Eq. 3 also points the way to combine materials to produce larger values of $T_c$ what is important to technological applications.

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* Corresponding author: evandro@if.uff.br
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