Phase Separation and the Dual Nature of the Electronic Structure in Cuprates

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The dual nature of the electronic structure of stripes in La$_{2-x}$Sr$_x$CuO$_4$ was characterized by many experiments. We present here an attempt to characterize this dual behavior based on the Cahn-Hilliard theory of a phase separation transition which is assumed to occur at the high pseudogap temperature. The resulting inhomogeneous low doping system is formed of hole-rich (metallic like) regions embedded in a hole-poor (insulator like). This inhomogeneous configuration is analyzed by a new method within the Bogoliubov-deGennes superconducting theory. This approach describes well the electronic nodal-antinodal dichotomy and parts of the phase diagram.

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The high critical temperature superconductors (HTSC) represents today one of the greatest challenges of condensed matter physics. It is likely that the main difficulty to understand the physical properties of HTSC is due to the fact that some families seem to have a high inhomogeneous electronic structure while others families appear to be more homogeneous or, at least, without some gross inhomogeneity.

An important technique to study the HTSC electronic structure is provided by angle resolved photoemission (ARPES) experiments. With the improvement of the energy and momentum resolution in recent works [1, 2, 4, 5], it was possible to distinguish a two component ARPES spectra. Moreover, the values of $\Delta_0$ decreases with doping but the quasiparticle spectral weight near the nodal directions, at the Fermi level, increases, showing the distinct behavior of these two aspects of the electronic structure [1, 2, 3, 4].

A complementary technique to ARPES is provided by scanning tunneling microscopy (STM) since it probes the differential conductance or $\Delta_N$ directly on the surface of the compound. Recent STM data have revealed a patchwork of (nanoscale) local spatial variations in the density of states which is used to measure the local superconducting gap $\Delta_N$ [5, 6]. With this technique, it was also possible to distinguish two distinct behavior: well defined coherent and ill-defined incoherent peaks depending on the spectra location on a Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (B2212) surface [7, 8].

These and others unusual features of cuprates led to theoretical proposals that phase separation is essential to understand their physics [9, 10, 11]. Also, tunneling experiments using superconductor insulator superconductor (SIS) with different insulator layers have shown distinct sets of energy scales and have also led to the idea that the richness of the phase diagram as function of doping is due to the charge inhomogeneity in the Cu-O planes [12].

In this paper we take the large pseudogap temperature of the HTSC phase diagram [13, 14] as the phase separation temperature $T_{ps}(p)$. To obtain quantitative results on the charge separation as a function of the temperature, we apply the Cahn-Hilliard (CH) theory [20] to this transition. It yields the phase separation patterns found in many HTSC and it provides also an interpretation to the energy $E_g$ common to all cuprates [15], as the potential barrier between the two equilibrium hole-rich and hole-poor phases. Since $T_{ps}(p)$ increases as the average doping level $p$ decreases, the differences between high and low local densities are enhanced in low doping compounds. With the charge structure that comes out of these calculations in a $N \times N$ cluster, we use the BdG method, with the local chemical potential associated with this variable charge structure, to calculate the superconducting properties. This is a novel approach to the charge inhomogeneities in the BdG context. Several interesting
features comes out of this procedure which allows us to study the insulator, metallic and superconducting phases by quantitative calculations that compares well with the experimental results.

The CH non-linear differential equation which describes the time evolution process of a phase separation process, at a temperature $T$ below the phase separation transition at $T_{ps}(p)$, can be written as a time ($t$) derivative\cite{14, 20}:

$$
\frac{\partial u}{\partial t} = -M \nabla^2 (\varepsilon^2 \nabla^2 u + A^2(T) u - B^2 u^3). \tag{1}
$$

where $u$ is the local order parameter. In a finite size scheme\cite{14}, it is associated with the local charge $p(i)$, i.e., the local variation from the average number of holes per copper atom $p$, defined at a site $i$ as $u(i) \equiv p(i) - p$ and $u(i) \approx 0$ above and near the $T_{ps}$, $\varepsilon = 0.01$ and $B = 1$ are fixed parameters, $A$ depends on the temperature $T$ and the ratio $A/B$ yields the two local equilibrium densities $p_{\pm}$, a low and a high density. $M$ is the mobility of the particles and it dictates the phase separation time scale. Thus, the degree of phase separation depends on how fast the system is quenched, what is an explanation why similar compounds may exhibit different degree of inhomogeneity. As the temperature goes down below $T_{ps}$, the two equilibrium order parameters (or densities) increases their differences and the energy barrier between them, $E_g$, also increases\cite{21}. $E_g = A^2(T)/B$ which is proportional to $(T_{ps} - T)^2$ and can be taken as the energy associated with the upper pseudogap temperature\cite{15}. In Fig. 1 we display the mapping of the order parameter for a $100 \times 100$ system with $p = 1/8$. The two equilibrium local densities for this system is $p_{-} = zero$ and $p_{+} = 0.24$. At the beginning, the phase separation process is quite symmetric, and could be the reason for the symmetric phases as stripes and checkerboard\cite{22} order. As it evolves in time, the systems tends towards a complete phase separation, with larger and less ordered stripes, as shown in Fig. 1.

Our main point here is that this phase separation process is the cause of the charge inhomogeneity found in many HTSC materials. Taking the large pseudogap, as the phase separation temperature $T_{ps}(p)$, it is possible to infer that a HTSC compound, depending on its doping level, evolves in different patterns as, for instance, in the way depicted in Fig. 1. Therefore underdoped compounds which, in general, have a very high $T_{ps}$, may phase separate into a complete bimodal charge distribution with larger charge stripes or phase domains (Fig. 1: or d)), while compounds with $p \approx 0.19$ which have very low values of $T_{ps}$, segregates in small and symmetric regions as shown in Fig. 1 or b). Above $T_{ps}$ or for compounds with $p > 0.19$ the systems are described by small doping variations around the mean value or a Gaussian distribution. This change in behavior near $p \approx 0.19$ has been detected in many experiments\cite{18}. More recently, STM data\cite{10, 11} detected the vanishing of the zero temperature pseudogap peaks (ZTPG) for compounds approaching 0.19. In the CH scenario, the origin of the ZTPG is the value of the energy barrier $E_g$ between the hole-poor and hole-rich phases\cite{14}. Since the charge domains and $E_g$ decrease with $p$, the ZTPG must behave in a similar fashion: decreasing in number and intensity and vanishing near $p = 0.19$, exactly as observed in recent STM results\cite{10, 11}.

Now, that we have derived how the charge inhomogeneity sets in a HTSC material below the high pseudogap temperature $T_{ps}$, we can study how the superconductivity develops in a charge inhomogeneous system, like in a stripe phase, as function of the temperature. With this aim, we feed into a local superconducting calculation with the BdG mean-field theory\cite{23, 22}, the charge or domains mappings derived from the CH solutions. As usual, the method starts with the extended Hubbard Hamiltonian

\begin{equation}
H = - \sum_{\langle ij \rangle \sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + \sum_{i\sigma} (\mu_i) n_{i\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \frac{V}{2} \sum_{\langle ij \rangle \sigma \sigma'} n_{i\sigma} n_{j\sigma'}, \tag{2}
\end{equation}

where $c_{i\sigma}^{\dagger} (c_{i\sigma})$ is the usual fermionic creation (annihilation) operators at site $x_i$, spin $\sigma \{\uparrow \downarrow \}$, and $n_{i\sigma} = c_{i\sigma}^{\dagger} c_{i\sigma}$. $t_{ij}$ is the hopping between site $i$ and $j$. In an attempt to model real systems, we have used hopping values up to $5^{th}$ neighbors derived from the ARPES data of YBCO\cite{24}. In their notation, the hopping parameters here are: \( t \equiv t_1 = 0.15 \text{eV}, t_2/t_1 = -0.70, t_3/t_1 = 0.25, t_4/t_1 = 0.08, t_5/t_1 = -0.08 \). $U = 1.1t$ is the on-site and

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1}
\caption{(Color online) The mapping of the order parameter during the process of phase separation in a compound with average doping $p = 0.12$, starting with variations around $u = 6$. Panel a) is for $t=400$ time steps, b) $t=800$, c) $t=1000$ and d) $t=4000$.} \label{fig:1}
\end{figure}
\[ V = -1.1t \] is the nearest neighbor phenomenological interactions. \( \mu_i \) is the local variable chemical potential which reproduces the charge inhomogeneity solutions shown in Fig. 1, according the CH results. The BdG equations are:

\[
\begin{pmatrix}
  K & \Delta \\
  \Delta^* & -K^*
\end{pmatrix}
\begin{pmatrix}
  u_n(x_i) \\
  v_n(x_i)
\end{pmatrix}
= E_n
\begin{pmatrix}
  u_n(x_i) \\
  v_n(x_i)
\end{pmatrix}
\]  
(3)

These equations are solved self-consistently in clusters from 14 \times 14 to 24 \times 24 sites for the positive eigenvalues \( E_n \) or quasiparticle excitations and the eigenvectors or charge amplitudes \( u_n(x_i) \) and \( v_n(x_i) \), together with the pairing amplitudes \[ 23 \]

\[
\Delta_U(x_i) = -U \sum_n u_n(x_i)v_n^*(x_i) \tanh \frac{E_n}{2k_BT},
\]  
(4)

\[
\Delta_{\delta}(x_i) = -\frac{V}{2} \sum_n |u_n(x_i)v_n^*(x_i+\delta) + v_n^*(x_i)u_n(x_i+\delta)| \tanh \frac{E_n}{2k_BT},
\]  
(5)

and the hole density is given by

\[
p(x_i) = 1 - 2 \sum_n |[u_n(x_i)]^2 f_n + [v_n(x_i)]^2 (1 - f_n)|,
\]  
(6)

where \( f_n \) is the Fermi function.

The major difference from previous calculations is in the way that the disorder is taken into account. Ghosal et al.\[24\] used an impurity potential defined by a random variable between the limits \([-V, V]\). Here, as mentioned above, we feed into the initial conditions the non-constant local charge, as those shown in Fig. 1, derived from the CH solutions. With this fixed charge mapping in a \( N \times N \) cluster, we let the chemical potential \( \mu \) change, self-consistently, until it yields these initial charge domains.

Fig. 2 shows the superconducting d-wave gap \( \Delta(i, T) \) at each site \( i \) of two 14 \times 21 clusters and their temperature evolution. Panel a) represents an underdoped compound of \( p = 0.05 \) with total phase separation into a bimodal charge distribution made of charge variable stripes of 14 sites each, and in a less symmetric geometry, as taken from Fig. 1. The values of the local doping level \( p(i) \) are shown on the top of each panel. \( p = 0 \) is the light color and \( p = 0.24 \) is the dark color phase depicted in the Fig. 1. Panel b) represents a compound of average doping of \( p = 0.12 \), made of values of \( p(i) = 0.0, 0.12 \) and 0.24 similar to Fig. 1. The gaps are calculated by the BdG equations and the chemical potential \( \mu(i) \) evolves self-consistently at each site \( i \), until it yields the initial conditions on the local densities, which are held fixed. We have verified that changing the stripes configuration in the clusters yields the same type of gap structure and temperature dependence.

The results for the \( p = 0.05 \) sample with \( p_- = 0 \) and \( p_+ = 0.24 \) is shown in the top panel of Fig. 2. As one can see, the largest values of the zero temperature gap \( \Delta_0(i) \) is in the regions where \( p(i) = 0.24 \). This is a direct interpretation of the 1D metallic behavior and the corresponding high values of the leading edge shift on the Fermi surface\[2\] along the \((\pi, 0)\) and \((0, \pi)\) antinodal straight segments. As \( p \) increases, the size of hole-rich stripes increases, for instance, for \( p = 0.05 \) they are typically made of 3 lines and for \( p = 0.12 \) they are made of 6 lines. Thus, upon doping, the hole-rich stripes increases in number and size, changing the properties of the system from one dimensional to two dimensional character and enhances the overall metallic electronic behavior. Such changing with the average doping level was also verified by the ARPES data on many lightly doping samples\[2\] by the measurement of spectral weight at the antinodal and, the increase of the spectral weight along the \((\pm \pi, \pm \pi)\) nodal regions with \( p \).

We discuss now how \( T_c(p) \) and \( T^*(p) \) (the lower pseudogap) are estimated in our calculations. We see from Fig. 2 that, at zero temperature, there is a large variation of \( \Delta(i, T) \) throughout the samples. Hole-poor regions have \( \Delta(i, T) \approx 0 \) and hole-rich have finite values. As the temperature increases slightly, mostly local gaps at the hole-poor regions vanish, leaving only finite superconducting gaps at the hole rich regions. Thus the \( p = 0.05 \) sample, as concerns it resistivity, is an insulator at all finite temperatures, although it has a few metallic stripes which were detected by ARPES\[2\]. For the \( p = 0.12 \) compound, even the gaps at the hole-poor stripes remain up to \( T = 40K \). Thus, it is clear that it is the superconducting percolating temperature which, in agreement with previous work\[26\], is the superconducting critical temperature \( T_c \). In other words, above \( T_c \), the local superconducting gaps vanish at hole-poor regions.
and it prevents the superconducting regions to percolate, there are still some non-vanishing $\Delta(i, T)$ at the hole-rich or metallic regions but, since they occupy marginally less than 50% of the system size, it cannot hold a superconducting current. Thus, above $T_c$, the $p = 0.12$ sample has a metallic behavior. As it is also shown in Fig. 2, these local gaps decreases continuously as the temperature increases further and, for $p = 0.05$ and $p = 0.12$, they totally vanish at $T = 110$K and $T = 90$K respectively. This is the interpretation to the onset of the local superconducting or lower pseudogap temperature $T^*(p)$.

The maximum zero temperature $\Delta_0(i)$ as function of $p$ is shown in Fig. 3. $\Delta_0(i)$ scales reasonable well with the ARPES measurements of the leading edge shift[4] demonstrating that the maximum gap in a sample increases with the disorder. As already shown in Fig. 2, a system with density disorder in the form of stripes, enhances the local gap. Thus, we find that the maximum gap for a such inhomogeneous sample is larger than that of a homogeneous similar (same $p$), that is, the disorder enhances the local gaps at the hole-rich regions and at their borders. However, with the same coupling, the average gap in a disordered system is less than the gap of a homogeneous one, and the average values of $T_c$ is also enhanced. These results are shown, for s-wave calculations, in the inset of Fig. 3 and are in agreement with some recent calculations[27].

In summary we have used the CH phase separation approach to model the inhomogeneity found in LSCO. The derived charge stripe-like disorder describes well the dual nature of the underdoped electronic structure. The energy barrier $E_g$ between the two equilibrium densities furnishes an interpretation to the large pseudogap energy scale and the ZTPG measured on Bi2212 by STM. The spinodal separation provides also a mechanism of how the electronic structure of LSCO, as seen by ARPES, evolves with doping: the increase of the leading edge shift at the straight (1D) segments near the antinodal region and the increase of the spectral weight near the nodal region, with the concomitant developing of the metallic behavior. The disorder favor the clustering of metallic regions embedded in an insulator matrix at the low doping compounds. The site dependent local superconducting gaps at low and high doping regions provide also an interpretation to the onset of superconductivity or lower pseudogap $T^*(p)$, in agreement with the phase diagram and the nonconventional properties of HTSC.

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