**Intermediate spin-charge order in the cuprates**

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**Abstract.** On the basis of a rigorous constraint we show that the doped cuprates either exhibit electronic phase-separation, or balance right at the phase-separation threshold. We suggest that the pseudo-gap phase can be identified with the state that is nano-scale phase-separated in magnetic and non-magnetic phases with different charge carrier concentrations. Long-range Coulomb interaction should drive the nano-scale phase-separated state into a self-organized ordered phase, even though such ordering is most likely frustrated by chemical disorder. Identifying this nano-scale self-organization should clarify a number of confusing issues related to the origin of the superconductivity in the cuprates.

1. Introduction

High-temperature superconductivity (HTSC) in the cuprates is known to occur at charge carrier concentrations separating magnetic and non-magnetic phases. When charge carriers are injected into magnetic background, they have natural tendency to destroy it. The magnetic background, in turn, tends to expel them, or, when complete expulsion is not possible, to assemble the charge carriers together so that they destroy less magnetic correlations. This signifies the tendency towards charge inhomogeneity [1-5]. Such a tendency is opposed mainly by the increase in the kinetic energy of the charge carriers, and by the Coulomb repulsion between charge carriers if phase-separation occurs at low temperatures where ionic diffusion is not possible and electronic phase-separation is not compensated by ionic motions. Thus electronic phase-separation, if present, should be limited to nano-scale along at least one spatial direction due to the long-range Coulomb interaction. As we argue below it is possible that the presence of inhomogeneities or the proximity to the phase-separation threshold is crucial for the onset of superconductivity in the cuprates. The purpose of this work is to put forward a simple, and hence reliable, quantitative criterion for the formation of electronic inhomogeneity in the cuprates and other materials with similar phase diagram, and discuss possible implications to the HTSC phenomenon.

Reliable experimental proofs of bulk electronic phase-separation in cuprates are limited. When spin and charge modulations are static and periodic, they can be well characterized by neutron scattering and soft x-ray scattering. The most extreme, and well-studied, case of static and periodic electronic inhomogeneity in the cuprates is the non-superconducting phase in \(\text{La}_{2-x-y}\text{Nd}_x\text{(Sr,Ba)}_y\text{CuO}_4\) with \(y = 0.125 (= 1/8)\) with spin [6, 7] and charge [8] modulation. This phase is frequently referred to as the stripe phase even though the actual dimensionality of its spin and charge modulation pattern is still under debate [9-12]. However, in the case of disordered and possibly slowly fluctuating electronic...

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inhomogeneities, it is much more difficult to determine their nature by experiments. For the superconducting phases, recent observations by the scanning tunneling spectroscopy (STS) convincingly demonstrated static spatial electronic inhomogeneities, most probably pinned by chemical disorder, at least at the surface of the cuprates [13-16], while phonon dispersions suggest phase-separation in the bulk [17, 18].

Given the experimental limitations, one may choose to attack the problem theoretically, and, indeed, numerous attempts to justify or disprove electronic inhomogeneities in cuprates have been made so far [1-3, 19-38], most of them in the framework of simplified theoretical models with short range interactions, such as the \( t-J \) model or Hubbard model. For the values of model parameters relevant to cuprates, the answers to the questions of phase-separation differ from a study to study and depend on the model and the approximation involved. However the overall impression projected by these studies is that the phase-separation is a subtle business in the models with the short-range interactions, but the inclusion of the full-range Coulomb interaction would certainly suppress it. In fact, if, in the absence of Coulomb interaction, the system is balancing near the threshold of phase-separation, then the inclusion of the full-range Coulomb interaction would not only eliminate the possibility of phase-separation, but would also strongly suppress any local fluctuation towards the phase-separated state. In this work, however, we show that the situation vis-a-vis real cuprates is not as unfavorable to phase-separation as that in the above mentioned models. The detailed version of our analysis can be found in Ref. 39.

2. Condition of phase separation

We will start our discussion with an idealized, and possibly imaginary, phase diagram shown in Fig. 1. Here we consider only homogeneous phases, and suppress phase-separation as well as quantum and thermal fluctuations. The transition can be of any order, or merely a crossover. The line in Fig. 1 represents the condition (\( T \) and \( x \)) at which the free energies of the two homogeneous phases are equal in the mean-field approximation. In reality phase-separation may take place, and such a phase line as shown in Fig. 1 may not be observed. However, the usefulness of such a theoretical construct is well known for the liquid-solid phase diagram, known as the \( T_0 \) line [40]. In the context of cuprates, the line in Fig. 1 separates a homogeneous insulating phase with antiferromagnetic (AFM) correlations from the homogeneous metallic phase without AFM correlations. Therefore, it extends significantly beyond the experimentally observed region of the static AFM order.

![Fig. 1. Temperature-vs.-concentration phase diagram for imaginary homogeneous phases when phase-separation is inhibited even when the system prefers to phase-separate (see text). The solid line can represent a phase transition of any order or a crossover.](image)

The total free energy \( F_{\text{tot}} \) of a homogeneous charge compensated cuprate per one CuO\(_2\) in-plane unit can be decomposed as

\[
F_{\text{tot}}(x) = F_0(x) + F_\eta(x)
\]  

(1)
where \( x \) is a dimensionless charge carrier concentration, \( F_d(x) \) is the free energy of a non-magnetic phase, and \( F_\eta(x) \) is the energy associated with the onset of AFM correlations. The system becomes unstable towards phase separation, when the curvature of \( F_{\text{tot}} \) (defined as \( \frac{1}{2} \frac{d^2 F_{\text{tot}}(x)}{dx^2} \)) reaches a finite negative value outweighing the positive curvature of the additional energy contribution, which appears in the phase separated state, and which we assume to originate mainly from the Coulomb repulsion between uncompensated charges. In order to formalize this condition we introduce the free energy curvature of non-magnetic state, \( K_0 \equiv \frac{1}{2} \frac{d^2 F_{\text{tot}}}{dx^2} \), which is assumed to be positive everywhere; the \textit{negative} curvature of the AFM free energy, \( K_\eta \equiv \frac{1}{2} \frac{d^2 F_\eta}{dx^2} \); and the positive curvature associated with the Coulomb repulsion energy \( \Delta F(\Delta x) \) between uncompensated charges having concentration \( \Delta x \), \( K_{\text{Coul}} \equiv \frac{1}{2} \frac{d^2 \Delta F}{d\Delta x^2} \). The condition for the instability of the homogeneous state then becomes:

\[
K_\eta \geq K_0 + K_{\text{Coul}}. \tag{2}
\]

We estimate the right-hand-side of inequality (\( K_{\text{tot}} \)) assuming [39] that the non-magnetic phase has a character of Fermi-liquid [41], and that the onset of phase-separation proceeds according to the \textit{lasagna} scenario (layered phase separation). This gives

\[
K_0 = \frac{\pi \hbar^2 (1 + f_0^s)}{2 m^* a^2}, \tag{3}
\]

where \( a \) is the lattice period in CuO\(_2\) planes, \( m^* \) is the effective mass and \( f_0^s \) the Landau Fermi-liquid parameter [41]; and

\[
K_{\text{Coul}} = \frac{2 e^2 R_s}{\varepsilon a}, \tag{4}
\]

where \( e \) is the charge of an electron, \( R_s \) is the distance between the CuO\(_2\) planes, and \( \varepsilon \) is the dielectric constant. We assume [39] \( a = 4 \text{ Å}, f_0^s = 6, m^* = 4 m_e \) (\( m_e \) is a free electron mass), \( R_s = 6 \text{ Å}, \varepsilon = 30 \), thus obtaining \( K_0 = 1.35 \text{ eV} \) and \( K_{\text{Coul}} = 0.57 \text{ eV} \).

The calculation of \( K_\eta \) may appear to be the most difficult part in assessing the inequality (2), since it is not clear how the destruction of the AFM correlations proceeds in cuprates with increased doping: it may pass through a magnetic Fermi-liquid with difficult-to-compute magnetic energy, or through a more exotic magnetic phases such as spin glass or resonating valence bond liquid [42]. In fact, however, in order to estimate \( K_\eta \), it is only necessary to know the value of the critical charge carrier concentration \( x_{c0} \), where AFM correlations disappear from the system, and the rather easily accessible value of magnetic energy at zero doping \( F_{\eta0} = -F_\eta(0) \). With the two above parameters, we were able to show rigorously [39] that

\[
K_\eta \geq \frac{F_{\eta0}}{x_{c0}} \approx \frac{J}{2 x_{c0}^2}, \tag{5}
\]

where \( J \) is the exchange coupling constant, which we assume \( J = 125 \text{ meV} \). The equality in constraint (8) is realized in the case of quadratic dependence of \( F_\eta(x) \) on \( (x_{c0} - x) \).

3. Example of a second order phase transition

Leaving the rigorous proof of constraint (5) to Ref. 39, here, we would like to show how one can arrive to the right-hand side of the above constraint through a crude approximation in the framework
of Landau theory of second-order phase transitions. One can start from the Landau expansion of the free energy in powers of AFM order parameter (staggered local magnetization), of which absolute value is denoted as $\eta$:

$$ F_\eta = A\eta^2 + B\eta^4 $$

(6)

where $A$ and $B$ are two expansion coefficients. Near the critical temperature of phase transition $T_C$, the coefficient $A$ can be parameterized as $A = \alpha (T - T_C)$, where $\alpha$ is a positive constant, and $T$ is temperature. At $T < T_C$, the minimum of the free energy is reached at $\eta = \sqrt{\alpha (T_C - T)/2B}$, resulting in $F_\eta = -\alpha^2 (T_C - T)^2/4B^2$. Near the critical concentration $x_{c0}$, we neglect the dependence of $\alpha^2/4B$ on $x$, and assume the linear dependence of $T_C$ on $x$: $T_C = \lambda (x_{c0} - x)$, where $\lambda$ is a slope parameter. Note that any alternative power law dependence of $T_C$ on $x_{c0}$ would be more favorable to phase-separation [39]. Thus we obtain

$$ F_\eta = -K_\eta \left[x_c(T) - x\right]^2, $$

(7)

where $K_\eta = \lambda^2 \alpha^2/4B$ and $x_c(T) = x_{c0} - T/\lambda$. The right-hand side of Eq.(7) can be estimated by further assuming (see Fig. 1) that $\lambda \approx T_{c0}/x_{c0}$, where $T_{c0}$ is the critical temperature at $x = 0$. If one now applies the Landau expansion all the way up to $x = 0$, then one obtains $F_{\eta0} = \frac{\alpha^2}{4B} T_{c0}^2$, and thus

$$ K_\eta = \frac{F_{\eta0}}{x_{c0}^2} $$

(8)

in agreement with constraint (5).

4. Chances of phase-separation

In our further estimates, we will use the minimum value of $K_\eta$ given by the right-hand side of the constraint (5). Figure 2 combines all estimates from Section 3 as a function of a still unknown value of $x_{c0}$. The shaded areas in that plot indicate the region of uncertainty by the factor of 2 in the value of $K_{\eta0} + K_{\text{Coul}}$ and the factor of 2 increase above the minimum value of $K_\eta$ as follows from the constraint (5).

Finding the value of $x_{c0}$ now becomes the final piece of a puzzle associated with the phase-separation condition (2). This parameter is difficult to pinpoint, but this difficulty should certainly be less significant than the difficulty of describing the entire decay of AFM correlations in cuprates through possibly exotic magnetic phases. The constraint (5) implies that the faster AFM correlations decay, the stronger is the tendency towards phase separation. Helpful for an approximate analysis is the fact that the inverse quadratic dependence of the constraint (5) on $x_{c0}$ is very steep in the range of interest, and, therefore, a relatively crude placement of the value of $x_{c0}$ would still result in a useful insight into the chances of phase-separation in cuprates.

Here we examine several possibilities.

1) Identification of $x_{c0}$ with the AFM phase boundary

This implies $x_{c0} = x_{AFM} = 0.02$ for La$_{2-x}$Sr$_x$CuO$_4$ and $x_{c0} \approx 0.06$ for YBa$_2$Cu$_3$O$_{6+\delta}$. However, Fig. 2 indicates that the left-hand side in inequality (2) would be two orders of magnitude greater than the right-hand side for $x_{c0} = 0.02$, and by a factor of ten for $x_{c0} = 0.06$. This means that the formation of inhomogeneities on the both sides of $x_{c0}$ would be unavoidable, whereas the AFM phase is observed below these concentrations. Therefore this choice is likely to be incorrect.

2) $x_{c0} \approx 0.18$

This is where in our estimates the left- and the right-hand-side of condition (2) are equal to each other. This concentration is also close to the value for which the quantum criticality is suggested by
some estimates [43] and experiments [44]. However, by the nature of constraint (5), we underestimate the left-hand-side of (2). Thus this value of $x_{c0}$ would still make a good case for an unstable homogeneous state.

3) $x_{c0} \approx 0.3$

Here it is more likely than not the homogeneous state remains stable, but the chances of phase separation are still significant, and, in any case, the fluctuations of charge carrier density should be large.

![Image](image_url)

**Fig. 2.** Estimates for negative $K_\eta$ and positive ($K_\eta + K_{Coul}$) contributions to energy curvature per one in-plane Cu. Negative curvature is plotted as a function of an unknown critical concentration $x_{c0}$. Solid lines represent the estimates by formulas (8), (3) and (4) with the numbers given in the text. Shaded areas around the lines cover the regions of the factor-of-two uncertainty for the above estimates.

Recent data by Wakimoto et al. [45] on frequency-integrated intensity of inelastic neutron magnetic scattering from La$_{2-x}$Sr$_x$CuO$_4$ and La$_{2-x}$Ba$_x$CuO$_4$ indicate that AFM correlations are still present at $x = 0.3$ but reduced compared to $x = 0$ by at least one order of magnitude. This finding may appear to indicate $x_{c0} \approx 0.3$, whereas similar results for YBa$_2$Cu$_3$O$_{6+x}$ reported earlier by Bourges [46] suggest a smaller value, close to $x_{c0} \approx 0.2$, but with a smaller range of energy integration. However, the value of $x_{c0}$ must be smaller than these values because of the possibility that what experiments are observing is the result of phase-separation into two phases: magnetic phase with $x < x_{c0}$ and non-magnetic one with $x > x_{c0}$. The disappearance of magnetic signal would then mean that the relative volume of magnetic phase approaches zero once the average concentration approaches 0.2 or 0.3.

In Ref. 39 we proposed to further constrain $x_{c0}$ using the phenomenology of an atypical cuprate family La$_2$CuO$_4+\delta$, where intercalated oxygen ions are mobile at sufficiently high temperatures, and, as a result, large-scale phase-separation is possible and had, indeed been observed. We further employed a reasonable assumption [39] that the value of $x_{c0}$ should limit the spinodal (locally unstable) range of charge carrier concentrations both in the case of mobile and frozen dopant ions. This idea is implemented in Fig. 3 on the basis of the experimental results of Ref. 47. The straight line in this figure extrapolates the boundary of unstable region from the higher temperatures, where the intercalated oxygen ions are mobile to the lowers temperatures, where these ions are frozen. Given that $x \approx 2\delta$ (or smaller [48]), the above extrapolation suggests that $x_{c0} \approx 0.13$, which, according to Fig. 2 corresponds to the region of likely phase-separation.

It is also interesting to note that this concentration, $x_{c0} \approx 0.13$, is close to the concentration for metal-insulator transition when superconductivity is suppressed by strong magnetic field [49]. Since the effect of such a strong magnetic field on the energy balance discussed here would not be insignificant these two cannot be the same, but if the magnetic field suppresses not only superconductivity but also nano-scale phase-separation, the coincidence of the two may be meaningful.
The fact that the best estimate of $x_{c0}$ happens to be above the composition limit of the AFM phase, $x_{AFM} = 0.06$, and below the composition limit for the pseudo-gap, $x_{PG} = 0.25$, implies that the pseudo-gap phase most likely is indeed the nano-scale phase-separated phase. As shown in Fig. 4, the real phase diagram of the system should show only the upper and lower limit lines, ending at $x_{10}$ and $x_{20}$ at $T = 0$. The line that ends at $x_{20}$ ($x_{20}$ line) should define the pseudo-gap temperature, $T_{PG}$. Note that in this construct the quantum critical point is not $x_{20}$ where $T_{PG}$ ends, but is $x_{c0}$, which is at the center of the superconducting dome. This is a very important point. The lower limit line could signify the observed metal-insulator transition. This could also be the phase boundary for the AFM phase in an ideal case, but chemical disorder can separate them as is clearly the case for LSCO.

5. Implications of the results

Our analysis indicates that a generic cuprate either phase-separates or balances right at the threshold of phase-separation. As discussed in the introduction, such a conclusion contradicts the impression projected by the theoretical studies of the models with only short-range interactions and without long-range Coulomb interaction. We have shown that Coulomb repulsion between charge carriers separated by 1 to 3 lattice periods helps the system to destroy AFM correlations faster, and thus favors, rather than opposes, phase-separation [39]. This result leads naturally to the conclusion that the pseudo-gap phase is made of the nano-scale mixture of two phases, one magnetic and the other metallic and non-magnetic. The identification of the pseudo-gap phase with the phase-separated region shown in Fig. 4
is consistent with the recent report by STM-STS that the distribution of the superconducting gap is still inhomogeneous even in the overdoped region [50,51].

We conclude that the quantum critical point is not the concentration where $T_{PG}$ extrapolates to $T = 0$, but is a point significantly lower in concentration, right in the middle of the superconducting dome in the phase diagram. However, the fluctuations associated with the “quantum critical” concentration are strongly modified and likely suppressed by nano-scale phase-separation. As a consequence of identifying the $x_{c0}$ line with the pseudo-gap temperature our result suggests that the nano-scale phase-separation is either necessary for or at least co-exists with superconductivity. The role of the quantum critical point may not to provide quantum fluctuations that may pair holes, but may be more complex, through the formation of a new state involving nano-scale phase-separation.

One strong possibility is that the long-range Coulomb interaction tends to drive the charge-separated regions into a well-defined charge ordering, perhaps the superlattice structure with an extended unit cell. The spin-charge stripe state is one of such possible states, but the stripe state apparently competes against superconductivity, so the state that is required for HTSC most likely is a different one, probably a state with strongly two-dimensional spin-charge order. In reality it is likely that such ordering is frustrated by chemical disorder, and develops only into a nano-scale medium-range order. Indeed our recent neutron scattering study suggests that the spin correlations in the underdoped cuprates are more complex than usually assumed, with spin-glass-like disorder and the possibility of nano-scale intermediate order which is distinct from the AFM order [52]. It is possible that the formation of such local spin-charge order is intimately connected to the occurrence of HTSC. Whereas these results are preliminary and require further study, it is likely that accurate knowledge of such ordering will lead to better understanding of the HTSC physics [53].

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