Inhomogeneous states in two-dimensional frustrated phase separation.

C. Ortix, J. Lorennana, and C. Di Castro

1 Institute-Lorentz for Theoretical Physics, Universiteit Leiden, P.O. Box 9506, 2300 RA Leiden, The Netherlands.  
2 SMC-INFM-CNR and Dipartimento di Fisica, Universit`a di Roma “La Sapienza”, P. Aldo Moro 2, 00185 Roma, Italy.  
3 ISC-CNR, Via dei Taurini 19, 00185 Roma, Italy.  

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We derive the phase diagram of a paradigmatic model of Coulomb frustrated phase separation in two-dimensional systems with negative short-range electronic compressibility. We consider the system subject either to the truly three-dimensional long-range Coulomb interaction (LRC) and to a two-dimensional LRC with logarithmic-like behavior. In both cases we find that the transition from the homogeneous phase to the inhomogeneous phase is generically first-order except for a critical point. Close to the critical point, inhomogeneities arrange in a triangular lattice with a subsequent first-order topological transition to stripe-like objects by lowering the Coulomb frustration. A proliferation of inhomogeneities which have inside smaller inhomogeneities is expected near all the transition lines in systems embedded in the three-dimensional LRC alone.

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I. INTRODUCTION

Domain pattern formation is a beautiful example of cooperative behavior in complex systems with competing interactions on different length scales. In electronic systems, this idea has gained momentum due to theoretical and experimental studies in materials like cuprates and manganites. Indeed it has become clear that strong electron correlations generally produce a tendency towards phase separation in electronic-rich and electron-poor regions. Although the long-range part of the Coulomb interaction (LRC) spoils phase separation (PS) as a thermodynamic phenomenon, the frustrated tendency towards charge segregation is still important and gives rise to inhomogeneous states where the charge is segregated over some characteristic distance but the average density is a fixed constant in order to avoid a diverging Coulomb cost in the thermodynamic limit.

From the experimental point of view, a prominent emergent tool in recent years has been scanning tunneling microscopy (STM). In the high temperature superconductors context, STM experiments in cuprates have revealed a phase segregation on the scale of \(\sim 20\) lattice constants reconcilable with a Coulomb frustrated phase separation (FPS) between an underdoped pseudogap phase and a superconducting phase at higher dopings. Coexistence of insulating and metallic regions on the scale of tens to thousands of nanometers have been reported also in thin films of colossal magnetoresistance manganites. Noticeably percolation of the metallic regions is closely correlated to abrupt changes in transport suggesting that FPS is at the heart of the colossal magnetoresistance behavior. More microscopic configurations with stripe patterns have been observed in cuprates, nickelates, and manganites. Even more, the discovery of pronounced anisotropies in transport measurements in ruthenates and in GaAs heterostructures at weak magnetic fields are in line with the proposal of exotic electronic liquid phases analogue to the intermediate order phases of liquid crystals. Evidence for mesoscopic electronic phase separation have been recently presented in organics, graphene, and pnictides.

The generality of this phenomena calls for simple models which neglect the microscopic details of the specific system rather capturing its general properties. The tendency towards PS is then caused by the appearance of anomalies in the electronic contribution to the free energy of the system \(f_e\), as a function of the density \(n\). Two kind of anomalies have been identified as the most relevant ones for phase separation in electronic systems. The first anomaly consists of a range of densities where the compressibility is negative while the second one corresponds to a single point where the inverse of the electronic compressibility has a Dirac-delta-like negative divergence due to a crossing of the free energies of the two competing phases. The two anomalies can be labeled by the exponent \(\gamma\) characterizing the behavior of the electronic free energy around a reference density \(n_e\), i.e., \(f_e = \alpha|n - n_e|^{\gamma}\) with \(\alpha < 0\) and \(\gamma = 2\) (negative compressibility region) or \(\gamma = 1\) (cusp behavior). Both anomalies appear often in model computations of electronic systems, for example Refs. correspond to \(\gamma = 2\) while Refs. correspond to \(\gamma = 1\).

When LRC effects can be considered as a weak perturbation upon the PS mechanism, one can achieve a universal picture of the FPS phenomenon. Antithetically, upon strong frustrating effects, the two short-range compressibility anomalies give rise to two different universality classes. Within the \(\gamma = 1\) universality class, different studies in 3D systems and in 2D systems have enlightened the key role of the system dimensionality in the FPS mechanism. Recently, the full phase diagram for the \(\gamma = 2\) class has been characterized in Ref. both in isotropic and strongly anisotropic 3D systems.
systems. The aim of this work is to investigate the properties of the FPS phase diagram for \( \gamma = 2 \) at strong frustration in 2D systems. This is an important generalization as many interesting strongly correlated systems are layered as cuprates, nickelates, iron oxypnictides, superconductors, or are truly two dimensional as the two-dimensional electron gas in heterostructures, graphene, etc. Relevant for cuprates, manganites and nickelates is how charge density wave instabilities of a uniform phase evolve into the very anharmonic structures experimentally observed.

We will consider two versions of the Coulomb interaction labeled by an effective dimensionality \( d \). The \( d = 3 \) case corresponds to the usual three dimensional Coulomb interaction and applies to the physical systems above mentioned (layered systems, heterostructures, graphene, etc.). The \( d = 2 \) case, instead, corresponds to a fictitious logarithmic Coulomb interaction. The effective dimensionality \( d \) regards only the interaction and should not be confused with the dimensionality of the system which is 2D. Alternatively the 2D system with logarithmic interaction can be considered as an anisotropic 3D system subject to the conventional Coulomb interaction where modulations in one “hard” direction are forbidden. This anisotropy can originate in the underling crystal structure and the case of only one hard direction (as opposed to two hard directions considered in Ref. 36) corresponds to the conventional Coulomb interaction where elongated domains have been reported.

II. PHASE DIAGRAMS IN 2D SYSTEMS

Our starting point is a paradigmatic model of Coulomb frustrated phase separation in systems with a short-range negative compressibility density region that is defined by the following free energy:

\[
F = \int dx \left[ \frac{e^2}{2\varepsilon_0} \frac{|n(x) - n_0|^2}{G^{-1}(|x - x'|)} + \frac{e^2}{2\varepsilon_0} \frac{|n(x) - n^c|^2}{G^{-1}(|x - x'|)} + \alpha |n(x) - n^c|^2 + \beta |n(x) - n^c|^4 \right]
\]

Here the gradient term models the surface energy of smooth interfaces and is parametrized by the stiffness constant \( e \) whereas \( \alpha < 0 \) is proportional to the inverse short-range compressibility. The inclusion of a fourth order term with \( \beta > 0 \) provides a symmetric double-well form of the short-range part of the free energy with minima at \( n = n_c \pm [\alpha/(2\beta)]^{1/2} \). In general, \( \alpha \) will depend on external parameters like the pressure. It can be also taken as temperature-dependent, as in Landau theory, in which case the model becomes a mean-field description of a temperature-driven transition to an inhomogeneous state. In addition, \( \varepsilon_0 \) indicates a static dielectric constant due to external degrees of freedom and \( \Pi \) is the average density. A rigid background ensures charge neutrality. Finally \( G(|x - x'|) \) is a positive-definite LRC kernel that in the \( d = 2 \) case has a logarithmic-like behavior whereas for \( d = 3 \), \( G^{-1}(|x - x'|) = |x - x'| \).

Since the model Eq. (1) has several parameters, it is convenient, in order to study the phase diagram, to measure all lengths in unit of the bare correlation length \( \xi = \sqrt{2\pi/\alpha} \), densities in units of \([\alpha/(2\beta)]^{1/2}\) and energy densities in terms of the barrier height \( f_0 = \alpha^2/(4\beta) \). Then one reaches a dimensionless form consisting, apart from an irrelevant constant, of a \( \phi \) model augmented with a long-range Coulomb interaction. The corresponding Hamiltonian is defined by:

\[
\mathcal{H} = \int dx \left[ \phi^2(x) - 1 \right]^2 + \nabla \phi(x)^2 + \frac{Q^2}{2} \times \int dx \int dx' \frac{\phi(x) - \bar{\phi}}{G^{-1}(|x - x'|)} \phi(x') - \bar{\phi}
\]

where, \( Q^2 \) is dimensionless renormalized frustration parameter of the long-range interaction given by:

\[
Q^2 = \frac{\xi^2}{\varepsilon_0} \frac{\xi^{4-d}}{|\alpha|}
\]

and the classical scalar field \( \phi \) represents the dimensionless local charge density with average density \( \bar{\phi} \). The effect of frustration can also be measured by the parameter \( Q^2/(5-d) \) introduced in Refs. 35,12,13. It can be obtained from the following dimensional analysis. Taking \( \phi \sim 1 \), the Coulomb energy density per domain can be estimated making the integrals in a volume of order \( l_d^d \) as \( Q^2/(5-d) \) and \( l_d \) indicating the typical size of the inhomogeneities measured in unit of \( \xi \). Within a sharp interfaces approach, the surface energy density goes like \( 1/l_d \). Both quantities are optimized whenever \( l_d \sim 1/Q^2/(5-d) \). Then \( Q^2/(5-d) \) takes the meaning of ratio between the additional energy cost induced by frustration and the typical PS energy gain.

For both the truly long-range Coulomb interaction and the logarithmic-like interaction, the charge susceptibility in momentum space can be derived by computing the static response to an external field. At \( k \neq 0 \) one finds:

\[
\chi(k, d) = k^2 + \frac{2^{3-d} \pi Q^2}{|k|^{4-d}} - 2 + 6\bar{\phi} \]
represents the maximum frustration degree above which the system is in its homogeneous phase.

The Gaussian instability line Eq. (1) indicates an instability towards a sinusoidal charge density wave (SCDW) of period $2\pi/k_0$ and direction chosen by spontaneous symmetry breaking. Still, it cannot persist up to $Q \to 0$ since inhomogeneities are predicted only for average densities inside the “miscibility” gap$^{30} \mid \bar{\phi} \mid < 1/\sqrt{3}$. This is not in tune with the usual Maxwell construction that on the contrary predicts an inhomogeneous state for $\mid \bar{\phi} \mid < 1$. Analogously to 3D systems$^{29}$, indeed, we find that the inclusion of non-gaussian terms results in a first-order transition preempting the Gaussian instability except for the critical point (CP) $(\bar{\phi}, Q) = (0, Q_c)$ as we show below.

By restricting to periodic textures, the free energy density difference between a modulated phase and the homogeneous phase can be expressed in momentum space as:

$$\frac{\delta F}{V} = \sum_{\mathbf{G} \neq 0} \phi_{\mathbf{G}} \chi^{-1}(\mathbf{G}, d) \phi_{-\mathbf{G}} +$$

$$+4\bar{\phi} \sum_{\mathbf{G}_1, \mathbf{G}_2, \mathbf{G}_3 \neq 0} \phi_{\mathbf{G}_1} \phi_{\mathbf{G}_2} \phi_{\mathbf{G}_3} \delta_{\mathbf{G}_1 + \mathbf{G}_2 + \mathbf{G}_3, 0} +$$

$$+ \sum_{\mathbf{G}_1, \mathbf{G}_2, \mathbf{G}_3, \mathbf{G}_4 \neq 0} \phi_{\mathbf{G}_1} \phi_{\mathbf{G}_2} \phi_{\mathbf{G}_3} \phi_{\mathbf{G}_4} \delta_{\mathbf{G}_1 + \mathbf{G}_2 + \mathbf{G}_3 + \mathbf{G}_4, 0}$$

where the $\mathbf{G}$'s are the wavevectors of the reciprocal lattice and $V$ indicates its unit cell volume. The presence of the cubic term in Eq. (5) opens the way to first-order transitions except for the CP. Away but close to the CP, the transition will be weakly first-order. Hence it can be treated, analogously to the liquid-solid transition$^{25,38}$, by fixing the wavevectors to have an equal magnitude $|\mathbf{G}| = k_0$. In this case, to gain an energetic advantage from the cubic term, one needs triads of wavevectors forming an equilateral triangle so that the Kronecker delta is satisfied. In 2D systems this is only verified by the hexagonal reciprocal lattice that is defined by the set of six wavevectors $\mathbf{G}/k_0 = (\pm \cos \pi/3, \pm \sin \pi/3), (\pm 1, 0)$. The corresponding free energy density reads:

$$\frac{\delta F}{V} = \left[ -2 + 6\bar{\phi}^2 + 2 \left( \frac{Q}{Q_c} \right)^{15/19} \right] 6\phi_G^2 + 48\bar{\phi}\phi_G^3 + 90\phi_G^4$$

As a result, one finds that the first structure to become stable corresponds to droplet-like structures forming a triangular crystal of inhomogeneities. Fig. 1 (a) shows the corresponding charge modulation close to the critical point.

Upon minimizing Eq. (6) with respect to the wave amplitude $\phi_G$ and requiring $\delta F/V = 0$, one finds that the first-order transition line is given by:

$$Q_T = Q_c \left[ 1 - \frac{37}{15} \bar{\phi}^2 \right]^{(6-d)/4}.$$  

On entering in the inhomogeneous phase density region, the triangular crystal phase is expected to compete with a unidirectional SCDW in which the local charge is modulated along stripes [see Fig. 1(b)] whose free energy density reads:

$$\frac{\delta F}{V} = \left[ -2 + 6\bar{\phi}^2 + 2 \left( \frac{Q}{Q_c} \right)^{15/19} \right] 2\phi_G^2 + 6\phi_G^4$$

We find that at fixed frustration $Q$, stripes become stable close to $\bar{\phi} = 0$ [see Fig. 2]. This leads to a first-order morphological transition that partially restores the translation symmetry. In the frustration-density plane, the morphological transition line can be determined by equating the free energies densities Eqs. (5), (6) of the two phases minimized with respect to the wave amplitude $\phi_G$. We find:

$$Q_M = Q_c \left[ 1 - \frac{87}{19 - 6\sqrt{6}} \bar{\phi}^2 \right]^{(6-d)/4}.$$
By decreasing the frustration degree, the charge density $\phi$. In addition, $\sigma$.

FIG. 2: (Color online) Comparison for one recovers the surface tension of a interface approach where surface energy effects are taken at weak frustration. This is shown in Fig. 3 for diagram found in the strong frustration regime to per-

36 cases at the topological transition.

Although the transition lines are only asymptotically exact close to CP$^{36}$, we find the topology of the phase diagram found in the strong frustration regime to persist at weak frustration. This is shown in Fig. 3 for a $d = 2$ Coulomb interaction where we characterized the full phase diagram. In the weak frustration regime ($Q << 1$), we considered a Uniform Density Approximation (UDA) where we assumed domains of uniform density of one or the other phases separated by sharp interfaces.$^{11,14,35,37,39}$ The UDA is an accurate description in this regime since there is a sufficient separation among the typical size of the domains $l_d$ and the screening length $l_s$ that controls the charge relaxation inside the domains.$^{14}$ For $d = 2, 3$, indeed, the latter goes like$^{13}$:

$$l_s \sim \frac{1}{Q^{\frac{n-2}{2}}}$$

In addition, $l_d$ is much larger then the typical Gaussian instability wavelength $1/k_0$. Thus one can rely on a sharp interface approach where surface energy effects are taken into account by neglecting long-range effects and computing the excess energy of an isolated interface. Then one recovers the surface tension of a $\phi^4$ kind$^{36}$ $\sigma = 8/3$.

In the weak frustration regime, we find the same topological transitions (thick lines in Fig. 3) as in strong frustration but now the inhomogeneities form sharply defined circular droplets and stripes. We have also studied the crossover from weak to strong frustration minimizing a discretized version of the model Eq. (6) in the Wigner-Seitz Approximation. The corresponding transition lines are shown with squares and diamonds in Fig. 3. For droplet-like structure we assumed circular symmetry in order to reduce the effective dimensinality of the minimization procedure to one. The upper panel of Fig. 4 shows the corresponding charge density profile in the radial direction at $\bar{\sigma} = 0.5$ and different frustrations. By decreasing the frustration degree, the charge density modulation in the radial direction gets more unharmonic similarly to what we find for striped states at $\bar{\sigma} = 0$ (see lower panel of Fig. 4).

The features of the FPS phase diagram are dramatically different if the stiffness constant is made strongly anisotropic. More in detail, we assume that $c_{\parallel}/c_{\perp} > 1$ where $c_{\parallel}/c_{\perp}$ indicates the stiffness component in the “hard” (“soft”) direction of the 2D plane and focus on the limit where any charge modulation in the “hard” direction is completely forbidden, i.e. $c_{\parallel}/c_{\perp} \to \infty$. In a 2D system subject to the logarithmic-like $d = 2$ interaction, in view of the analogy discussed in Sec. 4 this would correspond to consider a 3D system with two “hard” directions. The corresponding phase diagram has been shown in Fig. 3 of Ref. 36. In that case it has been shown that this strong anisotropy allows for second- and first-order transitions transition lines joined by a tricritical point. We now show that this feature appears also in 2D systems subject to the ordinary $d = 3$ Coulomb interaction. Since the strong anisotropy allows only for unidirectional charge modulations, the cubic term of Eq. (5) identically vanishes by restricting to wavevectors of $k_0$ magnitude. Thus, to include non Gaussian terms, one has to retain at least two collinear wavevectors so that the Fourier decomposition of the order parameter reads:

$$\phi(\mathbf{x}) = \bar{\phi} + 2\phi_1 \cos(k_1 \cdot x) + 2\phi_2 \cos(k_2 \cdot x)$$

where $|G_2| = 2|G_1| = 2k_0$. By minimizing with respect to the periodicity of the charge density modulation and assuming $\phi_2 < \phi_1$, that can be checked a posteriori, one obtains the following Landau free energy expansion in terms of $\phi_1$:

$$\frac{\delta F_{11}}{V} = r\phi_1^2 + u_4\phi_1^4 + u_6\phi_1^6$$

(8)
where the quadratic term coefficient

\[ r = 4 \left( \frac{Q}{Q_c} \right)^{4/3} - \left( \frac{Q_g}{Q_c} \right)^{4/3} \]

vanishes along the Gaussian instability line whereas \( u_4 = 6 - \frac{54}{5} (Q_c/Q)^{4/3} \) and \( u_4 \) is a positive constant. It is simple to notice that this is the canonical form of the Landau free energy expansion around a tricritical point that is determined by the vanishing of \( u_4 \) along the Gaussian instability and is therefore given by \( (\bar{\phi}, Q) = (1/(2\sqrt{3}), (3/4)^{3/4}) \). The finite density window of the second order line is important because it allows for a second order quantum critical point (QCP) as discussed in Ref.8. Furthermore as parameters are changed (density, frustration) an evolution similar to the one displayed in Fig. 4 will occur. This establishes a connection between the Gaussian instabilities predicted in models of the cuprates and the probably anharmonic stripe patterns observed.

Apart from small shifts of the transition lines, the effective dimensionality of the Coulomb interaction does not change the features of the 2D phase diagram both in absence and in presence of strong anisotropies. This is reported in Fig. 5 where we show the phase diagram of an anisotropic 2D system subject to the truly \( d = 3 \) Coulomb interaction. An important difference, however, will appear close to the transition lines as discussed below.

### III. Discussion and Conclusions

The topology of the FPS phase diagrams in 2D systems has a strong similarity to the 3D case of Ref.36 except that a crystal of drops appears in addition in 3D. Thus one can state that the system dimensionality \( D \) and the effective dimensionality of the long-range Coulomb interaction \( d \) do not qualitatively affect the FPS phenomenology in the \( \gamma = 2 \) universality class. This is not quite the end of the story since the transition from the homogeneous to the inhomogeneous phase and the topological transitions are first-order-like. A cusp singularity in the minimal free energy of the system will be naturally produced as indicated by arrows in Fig. 2. This drives the system towards a new stage of FPS tendency which now, however, is governed by the \( \gamma = 1 \) universality class and a new renormalized frustration constant \( Q' \) which depends on the distance from the critical point and diverges at the critical point. At this second stage of domain pattern formation, a dramatic difference between 3D and 2D \( d = 3 \) systems will arise. Indeed in 3D systems, for \( \gamma = 1 \), there is a critical value of \( Q' \) above which the second stage of pattern formation is suppressed while in a 2D, \( d = 3 \) system the second stage always occurs no matter how big is \( Q' \). For the case of 2D \( d = 2 \) systems, a critical frustration for \( Q' \) exists since, as mentioned in Sec. III it can be viewed as an anisotropic 3D, \( d = 3 \) system.
The stabilization of inhomogeneities which have inside smaller inhomogeneities represents a relevant new effect to be considered in the 2D, $\gamma = 2$ phase diagram. These states will consist of stripes of droplets alternating with stripes of stripes (homogeneous stripes) and will appear in a narrow but still finite window near the first-order morphological (homogeneous-inhomogeneous) transition.

In conclusion, we have derived the Coulomb frustrated phase separation phase diagram in 2D systems with a short-range negative compressibility subject either to a fictitious two-dimensional logarithmic-like Coulomb interaction and the truly three-dimensional Coulomb interaction. Similarly to three-dimensional systems, we find that the transition from the homogeneous phase to the inhomogeneous phase is always first-order except for a CP. Close to the CP, inhomogeneities are predicted to form a triangular lattice with a subsequent transition to striped states. The transition lines continuously evolve into the weak frustration limit. In 2D systems subject to the three-dimensional Coulomb interaction both first order and second order transition lines are found separated by a tricritical point. A proliferation of inhomogeneities is naturally expected near the first order transition lines. The second order transition lines make a QCP when crossed by the change of one parameter, like doping considered in Ref. [8]. Then one can expect very harmonic charge modulations close to the QCP which evolve into more anharmonic modulations as the distance from the QCP increases.

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