Universality classes for Coulomb frustrated phase separation

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Abstract

We identify two “universality” classes in the Coulomb frustrated phase separation phenomenon. They correspond to two different kind of electronic compressibility anomalies often encountered in strongly correlated electronic systems. We discuss differences and similarities of their corresponding phase diagrams in two- and three-dimensional systems.

Key words: Non-Fermi-liquid ground states; electron phase diagrams; nanoscale phase separation

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1. Introduction

A large variety of systems with phase separation (PS) tendencies subject to long-range forces self-organize in domain patterns \cite{1,2,3,4}. Recently, advances in local probe techniques have revealed mixed states in materials like cuprates and manganites \cite{5,6} rekindling the study of this phenomenon in strongly correlated electronic systems. Indeed it has become clear that strong electron correlations generally produce a tendency towards PS \cite{7,8,9,10,11} which, however, is frustrated by the long-range part of the Coulomb interaction (LRC). This leads to the formation of inhomogeneities with a typical size determined by the competition among long range forces and surface energy effects.

Since domains have often mesoscopic scales of several lattice constant, one can perform a general analysis of the frustrated phase separation (FPS) mechanism which neglects the microscopic details of each specific system while capturing its general properties. Tendency towards PS is then recognized by the presence of anomalies in the electronic contribution to the free energy of the system \cite{12}. The anomalies found in a large variety of strongly correlated electronic models can be classified in two kinds corresponding to a short-range negative compressibility density region \cite{13,14,15,16} or a Dirac-delta-like negative divergence of the compressibility due to the crossing of the free energies of two distinct phases \cite{17,18}. Both situations can be captured by expanding the short range part of the electronic free energy density \( f_e \) of the system around a reference density \( n_c \) as \( f_e = \alpha |n - n_c|^\gamma \). Here \( \alpha < 0 \) encodes the tendency towards PS where \( \gamma = 1 \) corresponds to a compressibility divergence and \( \gamma = 2 \) to a negative compressibility region. Higher order terms are essential to analyze FPS from the limit of strong frustration down to the limit of zero frustration. As a minimal model we take a contribution to the free energy of the form \( \beta (n - n_c)^2 \) that for \( \beta > 0 \) provide a symmetric double-well form of the short range free energy.

When LRC effects can be considered as a weak perturbation upon the ordinary PS mechanism, one can achieve a universal picture of the FPS \cite{12}. On the contrary, in the strongly frustrated regime, the two short-range compressibility anomalies give rise to two different behaviors. The aim of this work is to review the foremost features of their corresponding phase diagrams for two- \((D = 2)\) and three-dimensional \((D = 3)\) systems embedded in the three-dimensional long-range Coulomb interaction.

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2. Universal behavior: the weak frustration regime

In the $\gamma = 2$ case, FPS can be analyzed by means of a paradigmatic $\phi^4$ model augmented with the long-range Coulomb interaction. Pattern formation within this model (or closely related variants) has been considered in a variety of systems [21] including mixtures of block copolymers [4], charged colloids in polymeric solutions [22] and electronic systems [3, 19]. The corresponding Hamiltonian reads:
\[
\mathcal{H} = \int dx \left[ \phi^2(x) - 1 \right]^2 + |\nabla \phi(x)|^2 + \frac{Q^2}{2} \times \int dx \int dx' \frac{\left| \phi(x) - \phi(x') - \phi - \bar{\phi} \right|}{|x - x'|} (1)
\]
where the classical scalar field $\phi$ represents the local charge density with $\bar{\phi}$ its average density. A rigid background ensures charge neutrality. The model Eq. (1) can be reached by measuring the electronic free energy density $f_\epsilon$ in unit of the barrier height $\alpha^2/(4\beta)$, densities in units such that the double-well minima are at $\phi = \pm 1$, and distances in unit of the bare correlation length $\xi$. This leads to a renormalized Coulomb coupling $Q^2 = 2e^2\xi^{D-1}/(\varepsilon_0\alpha)$ with $e$ being the electronic charge and $\varepsilon_0$ a dielectric constant due to external degrees of freedom.

Frustrated phase separation for $\gamma = 1$ is more easily described by adding an auxiliary field $s$, equivalent to an Hubbard-Stratonovich variable, linearly coupled to the charge. It can be taken as a soft [22] or a conventional Ising spin ($s = \pm 1$) with the sign distinguishing the two competing phases. In the remainder we will refer to the latter case which is more straightforward to analyze. Domain walls of the Ising order parameter are sharp by construction with a surface tension $\sigma \propto 2J$ where $J$ indicates the Ising coupling. Then, in the continuum limit, one obtains the following Hamiltonian:
\[
\mathcal{H} = \sum \int dx \left[ \phi(x) - s(x) \right]^2 + \frac{Q^2}{2} \times \int dx \int dx' \frac{\left| \phi(x) - \phi(x') - \phi - \bar{\phi} \right|}{|x - x'|} (2)
\]
As before, we measure energy densities in units of $\alpha^2/(4\beta)$, the minima of the double well are at $\phi = \pm 1$ and lengths are measured in units of $\xi \equiv 4\sigma\beta/\alpha^2$ which represents the analogue of the bare correlation length in the present model. More precisely it indicates the size that inhomogeneities should have for the total interface energy to be of the same order as the phase separation energy density gain $\alpha^2/(4\beta)$. As for the model Eq. (1), the phase diagram is spanned by the dimensionless average density $\bar{\phi}$ and the renormalized Coulomb coupling $Q^2 = \alpha^2\xi^{D-1}/(\varepsilon_0\alpha)$.

In the absence of the Coulomb interaction, both models are subject to ordinary PS in the density range $|\bar{\phi}| < 1$ as ruled by the Maxwell construction (MC). Hence, the mixed state consists of macroscopic domains with local densities $\phi = \pm 1$. For $Q \neq 0$, PS is undermined as a thermodynamic phenomenon since the LRC energy cost grows faster than the volume in the thermodynamic limit. Thus charged domains at a mesoscopic scale appear. Their typical size $l_d$ is determined by the competition between the LRC cost $\sim Q^2l_d^{-1}$ and the surface energy density $\sim l_d^{-1}$. These terms are optimized whenever the inhomogeneities get a typical size $l_d^2 \sim 1/Q^2$.

Another important length scale is the screening length of the Coulomb interaction that controls the relaxation of the electronic charge inside the domains. Both the models Eqs. (1), (2), have a characteristic screening length defined by $l_s^2 \sim 1/Q^2$. It thus follows that in the weak frustration regime $Q \ll 1$, one obtains the hierarchy of length scales (in units of $\xi$): $l_s \gg l_d \gg 1$ that gives ground for a unified treatment of the FPS mechanism.

The strong separation between the typical size of the domains and the typical interface width $\sim 1$ allows to consider the smooth interface of the model Eq. (1) as sharp with a surface tension defined as the excess energy of an isolated interface [23]. Moreover, since the effect of LRC represents a small perturbation upon ordinary PS, inhomogeneities will appear with local densities near $\phi = \pm 1$. Then, FPS can be analyzed by expanding quadratically the double-well potential of Eq. (1) around its minima thus leading to a practical equivalence among the two FPS models.

A good approximation in the weakly frustrated regime lies in assuming a uniform density approximation (UDA) in which the local charge density is assumed constant $\bar{\phi} = \bar{\phi}$ for the $\gamma = 2$ case in 3D systems. Comparison with exact results shows that the UDA gives a very accurate description. From Fig. 1 one sees that droplet-like domains are the stable morphologies on entering in the inhomogeneous region. As $\bar{\phi}$ approaches the origin at fixed $Q$ a topological transition to rod-like
structures and subsequently to layered structures occurs. A similar behavior is also expected in 2D systems.

Our computations at weak coupling are variational so we cannot exclude more complicated phases including elongation of domains and "fingerling" as in classical systems [1]. Naturally the ordered phases will be very sensitive to quenched disorder. Also in the absence of quenched disorder the ground state may be hard to reach on a cooling experiment leading to a glass state.[19]

3. Universality classes: the strong frustration regime

By increasing the renormalized Coulomb coupling in Eqs. (1), (2), inhomogeneous states with local densities close to the reference density emerge. In this case the behavior of the two FPS models is radically different leading to two "universality" classes.

First, we consider the $\gamma = 2$ model Eq. (1). We restrict to 3D systems but similar ideas apply also to the 2D case. By computing the static response to an external field in momentum space, we get the charge susceptibility at $\mathbf{k} \neq 0$ as:

$$\chi(\mathbf{k}) = \left[ k^2 + \frac{2\pi Q^2}{k^2} - 2 + 6\phi^2 \right]^{-1}.$$

The charge susceptibility has a maximum at the characteristic finite wavevector $k_0 = [2\pi Q^2]^{1/4}$ and diverges approaching the Gaussian instability line [dotted line in Fig. 1] $Q_g = Q_c(1 - 3\phi^2)$ where $Q_c = 1/\sqrt{2\pi}$. This indicates an instability towards a sinusoidal charge density wave (SCDW) with direction chosen by spontaneous symmetry breaking. At small $Q$ the Gaussian transition line predicts inhomogeneities within the spinodal region $|\phi| < 1/\sqrt{3}$. This contrast with ordinary PS at $Q = 0$ which implies a mixed state in the global density region $|\phi| < 1$. The situation has been recently clarified in Ref. [21] where it has been shown that inclusion of non-Gaussian terms results in a first-order phase transition preempting the second-order Gaussian instability line except for the critical point (CP) $(\phi, Q) = (0, Q_c)$. The mixed region smoothly connects with the macroscopically phase separated state.

Away but close to the CP, the transition is weakly first-order with more complicated morphologies. Approaching the first-order line from above, inhomogeneities are predicted to form a BCC lattice with subsequent topological transitions to a planar hexagonal lattice of rods and layered structures [see Fig. 1]. One then finds the same topology as in the weak coupling regime, but now inhomogeneities have smooth interfaces in between. They continuously evolve into sharply defined droplets, rods and layers (disks and stripes in 2D) as $Q \to 0$ with a proliferation of higher order harmonics.

Next, we analyze the $\gamma = 1$ universality class. Fig. 2 shows the phase diagrams for two- and three-dimensional systems respectively for striped and layered structures which describe a smectic elec-
tronic liquid phase that possesses orientational order and breaks the translational symmetry only in one direction. As for the previous model other morphologies will be competitive away from $\phi = 0$. Indeed at weak coupling, as mentioned above, the two phases diagrams are identical. Here, however, we are interested in the behavior approaching $\phi = 0$ where the considered morphology is the most stable one.

In 3D systems, one finds a maximum frustration degree above which only uniform phases are allowed. This is antithetical to the behavior of 2D systems where the transition lines diverge logarithmically at $\phi = 0$ and thus the system always breaks into domains no matter how strong the frustrating effects are.

The difference between 2D and 3D results can be traced back to the different role of screening for different dimensionalities. In three dimensions, the charge density decays exponentially from the domain interfaces on the scale of the Thomas-Fermi screening length whereas in 2D systems it decays as a power law. Phase separation energy gain stems from the region where the electronic density is significantly different from its average value \cite{24}. In two-dimensions this is fulfilled everywhere in the domains, even far from the boundaries. This allows for domains with any typical size, even exponentially larger than the screening length. On the contrary, in three-dimensions the systems gain PS energy in a region of width $l_s$ around the interface. Regions far from the boundary produce an exponentially small energy gain and thus the system adjust itself to eliminate them. As a consequence a maximum size rule is generally valid that says that inhomogeneities cannot have all linear dimensions much larger than the three-dimensional screening length $l_{s,3D}$. This allows for arbitrary large inhomogeneities in 2D systems since one of the dimension is already smaller than the $l_{s,3D}$.

4. Conclusions

In this work we reviewed the main features of frustrated phase separation in charged systems considering two kind of short-range compressibility anomalies often encountered in strongly correlated electronic systems. The effect of long-range forces can be measured by introducing a dimensionless renormalized Coulomb coupling $Q$ which is a measure of the amount of frustration. Frustration tends to reduce the range of density where a mixed state appears hence stabilizing the homogeneous phase at densities where ordinary PS would occur. This situation is in accord with thermodynamic measurements \cite{29} of the uniform two-dimensional electron gas.

In the weak frustration regime, the FPS mechanism is not sensitive to the particular compressibility anomaly and a unified treatment can be reached. A series a morphological transitions is generally expected resembling the situation found in other classical systems \cite{1,14}. In this limit the phase diagram can be safely determined by means of a simple uniform density approximation\cite{21,25,26,27,28}.

On the contrary, at strong frustration, two different universality classes arise. In systems with a negative electronic compressibility region ($\gamma = 2$) a critical value of the frustration exist $Q_c$ for both $D = 2, 3$. Close to $Q_c$ soft inhomogeneities appear. They continuously evolve into the sharply defined structures of the weak frustration regime.

For systems with a cusp singularity in the electronic compressibility ($\gamma = 1$) the situation is in accord with thermodynamic measurements\cite{26,27,28} of the uniform two-dimensional electron gas. According to the maximum size rule, domains cannot have all linear dimensions much larger than the screening length. Therefore mesoscopic domains are generally expected in systems with small compressibility as bad metals, systems close to metal-insulator transitions and systems with very anisotropic electronic properties as indeed found.

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