Non-uniform glassy electronic phases from competing local orders

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We study non-uniform states and possible glassiness triggered by a competition between distinct local orders in disorder free systems. Both in Ginzburg-Landau theories and in simple field theories, such inhomogeneous states arise from negative gradient terms between the competing order parameters. We discuss applications of these ideas to a variety of strongly correlated systems.

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I. INTRODUCTION.

Accumulated experimental evidence strongly suggests that in many correlated electronic systems, different types of ordering phenomena compete and coexist over a wide range of tunable parameters. The most ubiquitous such cohabitation is between magnetic and superconducting orders. Itinerant antiferromagnetism (AFM) coexists with superconductivity in the 115 heavy fermion series (CeMIn5, where M = Co, Ir, or In). In UPt3, superconductivity emerges at TC ≈ 0.5K from a strongly correlated heavy electron state with small moment AFM below 6K. In some of the high-Tc cuprates charge density order coexists with spin density order (“stripes” and quantum critical behavior). Recent measurements indicate that in URu2Si2 there is a proliferation of competing phases under an applied magnetic field. Experiments also suggest multiple phases in the skutterudite superconductor PrOs4Sb12, and a number of other materials. Two trends are common to these experimental findings. First, the coexistence of different orders is often inhomogeneous. Second, this coexistence is frequently most pronounced near a Quantum Critical Point (QCP), where the transition temperature for one of the order parameters vanishes.

Additionally, dynamics of compounds with inhomogeneous coexistence of distinct orders is often glassy. In some systems, such as manganites, the glassy behavior is, most likely, due to disorder upon doping the system. In others, including cuprates, the glassiness may be self-generated (not simply due to doping disorder), and arise out of competing interactions at different length scales. The question remains, however, whether inhomogeneous and/or glassy behavior can arise out of a theory with local interactions and no disorder. In this article we address this question for a class of Ginzburg-Landau theories with competing order parameters. A comprehensive survey of classical systems with frustration and no disorder that display glassy behavior and proliferation of inhomogeneous ground states, can be found in Refs. 19, 20.

We study a minimal Ginzburg-Landau (GL) theory which includes amplitude-gradient coupling between two distinct local orders, and find the conditions for resultant inhomogeneous phases. A related interesting work examining gradient coupling in GL theories appeared slightly after the initial dissemination of our results. Extensions of the GL gradient couplings considered here are found in some studies of the supersolid transition. We show that, for a range of parameters described below, our theory maps onto an effective model that is likely to exhibit glassiness. Whether a particular system does or does not show glassy behavior upon cooling depends on the rate of temperature change and other dynamical variables that are not part of our equilibrium analysis. However, our approach allows us to conclude whether a glassy phase is possible and likely to occur. In this we follow the established approaches in the field.

The mapping that strongly suggests glassiness in our approach is to a Brazovskii-like model for one of the order parameters. The Brazovskii model for a single component order parameter is defined by a GL functional of the form

$$\mathcal{F} = \frac{V}{(2\pi)^d} \int d^d k |\frac{r_0}{2} + D(|\vec{k}| - q)^2||\Phi_k|^2 + ..., \quad (1)$$

in momentum ($k$) space with $V$ the volume of the system. In Eq. (1), the ellipses denote cubic, quartic, and higher order terms in the order parameter field $\Phi$. As the mass term, $r_0$ changes sign, the transition to a broken symmetry state $\Phi \neq 0$ involves the appearance of structures characterized by a finite wavenumber on a shell of radius $q > 0$. Structures that satisfy definite commensurability relations amongst the wavenumbers are most preferred. In Ref. 25, Brazovskii found that large phase space available for fluctuations around the minimizing shell alters the character of the transition to the ordered state once the fluctuations are accounted for, and suggested that it becomes first order. Thermal fluctuations renormalize the cubic terms of the GL theory. More recent replica calculations showed that the model has extensive configurational entropy, indicating proliferation of modulated low-energy states, and strongly suggesting slow dynamics and glassiness under generic experimental conditions. Once again, these replica calculations only establish that glassiness is a plausible and likely alternative to the first order transition into a uniformly modulated phase. Whether a finite temperature Brazovskii
transition does or does not transpire before the system undergoes a dynamical arrest (the glass transition outlined below) depends on microscopic details of the model. The known theoretical techniques (SCSA, DMFT, and others) do not enable the proof of a glassy phase. These methods only enable us to determine whether a glassy phase is possible.8,18,26,28

Below we find the mapping of systems with competing orders to Brazovskii type models. This mapping allows us to (i) Find resultant inhomogeneous phases in the GL analysis; (ii) Include fluctuations via a self-consistent field theory to establish that one of the two scenarios is realized: (a) the critical temperature for the onset of non-uniform states is suppressed to zero, suggesting that these states are more likely to be observed near a QCP; or, alternatively, (b) fluctuations lead to a low temperature Brazovskii transition; (iii) Appeal to existing replica calculation results to confirm the extensive configurational entropy associated with these incommensurate structures in disorder free systems with competing local orders, which strongly suggests slow dynamics and glassiness. Finally, we comment on possible realizations of our model and applicability of the results to itinerant electronic systems.

II. GINZBURG-LANDAU THEORY: INSTABILITY OF UNIFORM COEXISTENCE.

To empirically account for competing orders, we analyze the Ginzburg-Landau (GL) functional with two order parameters, Φ1 and Φ2, which will choose to be real and scalar without loss of generality. We remark that our very general GL approach applies to various types of order parameters. Of course, the symmetry, number of components of the order parameters etc. changes. Nevertheless, the conclusions are generally much the same. The uniform part of the free energy is \( F_0 = \int dx F_0 \) where

\[
F_0 = \frac{r_1}{2} |\Phi_1|^2 + \frac{r_2}{2} |\Phi_2|^2 + \frac{t}{2} |\Phi_1|^2 |\Phi_2|^2 + \frac{u}{4} |\Phi_1|^4 + \frac{u}{4} |\Phi_2|^4. \quad (2)
\]

In the spirit of the GL theory, \( r_{1,2} = a_{1,2}(T - T_{1,2}) \), with \( T_i \) the mean field transition temperatures. All other coefficients are taken to be temperature-independent. The quadratic coupling of the order parameters is allowed for all symmetries. We consider competing orders, \( t > 0 \), so that the uniform coexistence region (\( \Phi_1 \neq 0, \Phi_2 \neq 0 \)) occurs only below the lower of the transition temperatures, and for \( u > t_r^2 \). In that case, the values of the fields minimizing the free energy are \( \Phi_1^2 = (r_2 t - r_1 u)/(u - t^2) \), and \( \Phi_2^2 = (r_1 t - r_2)/(u - t^2) \). In disorder free systems, the only alternative to the uniform coexistence is phase separation unless non-trivial gradient terms are present.26 Therefore we include the inhomogeneous contribution to the free energy, \( F_q = \int dx F_q(x) \), where

\[
F_q = \sum_i |\nabla \Phi_i|^2 - \sum_{i,j} g_{ij} |\Phi_i|^2 |\nabla \Phi_j|^2 + \sum_i p_i |\nabla^2 \Phi_i|^2. \quad (3)
\]

Here, we included general symmetry allowed low order gradient terms. To flesh out the quintessential physics in what follows, we set \( g_{11} = g_{22} = g_{21} = 0, g_{12} > 0 \), and \( p_1 = 0 \). This is the essential aspect of the model that allows us to investigate the appearance of the inhomogeneous states. The coupling of the form \(-g_{12} |\Phi_1|^2 |\nabla \Phi_2|^2\) implies that in the effective theory for the order parameter \( \Phi_2 \) the coefficient of the gradient term, \( 1 - g_{12} |\Phi_1|^2 \), may become negative, making the transition of the Brazovskii type. We now investigate when this is possible.

With \( F(x) = F_0 + F_q \), the order parameter profiles satisfy the Euler-Lagrange equations, \( [\nabla \cdot (\partial F/\partial(\nabla \Phi_i))] = (\partial F/\partial \Phi_i) \). By constructing inhomogeneous variational states whose free energy is lower than the minimum amongst all possible uniform configurations, we prove that the uniform solution is unstable towards the appearance of inhomogeneities. We study the phase diagram of the model assuming that the mean field transition temperatures \( T_i \) can be tuned by an external parameter, \( x \) (pressure, doping, magnetic field etc.), as shown in Fig. 4 with \( T_1(x) \) monotonically decreasing, and \( T_2(x) \) monotonically increasing. That is,

\[
T_1 = T_{1(0)} - a_1 x, \\
T_2 = T_{2(0)} + a_2 x, \quad (4)
\]

with \( T_{1,2}^{(0)} \) and \( a_{1,2} \) positive constants.

We first concentrate on the region \( T_1 > T_2 \). Upon lowering the temperature, the first transition is into the uniform state with \( \Phi_2 = 0 \) and \( \Phi_1(x) = -r_1 \). Consequently, below the transition \( T_q = T_1 - 1/(g_{12} a_1) \) the coefficient of the \( |\nabla \Phi_2|^2 \) term becomes negative indicating the tendency towards the development of an inhomogeneous \( \Phi_2 \) phase. The structure of this modulation depends on the difference \( T_q - T_2 \). If this difference is sufficiently large, it is disadvantageous to create non-vanishing bulk average of \( \Phi_2 \). Local “bubbles” of the order may appear upon lowering \( T \), but their study is not our focus in the present work.

In order to make the connection with the slow dynamics and Brazovskii transition, we study the onset of the periodically modulated phase of the form \( \Phi_2(x) = \Theta_2 \cos(q x) \). Of the numerous contending low (free) energy configurations, we will focus on analytically tractable modulated structures; we do so in order to obtain stringent variational bounds that we are able to extremize, and based on the original analysis that showed the single modulation structures are most advantageous.21 In the regime \( T_q \geq T_2 \) minimization of the GL functional with respect to both \( q \) and \( \Theta_2 \) gives the transition temperature

\[
T_{c2} = T_q - (g_{12} a_1)^{-1} \left[ \sqrt{z^2 + \frac{2 \Theta_2}{g_{12}} + 2 p a_2 (T_q - T_2)} - z \right],
\]

\[
z \equiv \frac{p a_2 - t p a_1}{g_{12} a_1} \quad (5)
\]

to the phase \( \Theta_2 \neq 0 \) with modulations at a finite wave
FIG. 1: The phase diagram obtained from the Ginzburg-Landau expansion. The lines $T_{1,2}$ denote the bare mean field transition temperatures as a function of a tuning parameter $z$. $T_0$ is defined in text. An inhomogeneous phase appears below $T_{c2}$. Double line denotes the first order transition.

vector,

$$q = \sqrt{\frac{g_{12}a_1(T_1 - T_{c2})}{2p_2}}. \tag{6}$$

In the regime $T_q < T_{1,2}$ the first transition, at $T_q < T$, occurs into a spatially homogeneous phase. We next investigate the phase diagram for the more general variational ansatz $\Phi_2^{var}(x) = \Phi_2 + \Theta_2 \cos(q_1 \cdot x)$. Introduction of spatial modulations reduces the condensation energy and therefore is unfavorable, unless compensated by a significant gain due to the negative gradient term. As a result we find a (generically first order, but dependent on the magnitude of the coefficients in the GL expansion) transition from the homogeneous to modulated, with a finite $q$, phase at low $T$. In Fig. 1, we show the phase diagram of Eqs. 2-3 for $t = 0$. Of course, since we allowed only for the restricted variational states in the above analysis, our bounds are more potent for the global free energy minima - $\Phi_2$ is strictly inhomogeneous for all $T < T_{c2}(x)$; unrestricted inhomogeneous states (not bound to the form of $\Phi_2^{var}$) may extend to temperatures somewhat higher than $T_{c2}(x)$.

III. SELF-CONSISTENT FIELD THEORY FOR COMPETING ORDER PARAMETERS.

To improve on the GL analysis and incorporate the effect of fluctuations self-consistently, we generalize our model to $n$-component vector fields and utilize a large $n$ expansion. As well known, the $n = \infty$ limit is equivalent to the spherical model describing single component (scalar) particles. The physical engine for the inhomogeneities is, as in preceding section, the amplitude gradient coupling which drives non-uniformities in $\Phi_2$ once $\Phi_1$ is finite. For a finite $\Phi_1(x) = \Phi_1$, the effective free energy for $\Phi_2$ is

$$F_{\text{eff};2} = \int \frac{d^dk}{(2\pi)^d} \left[ \frac{T_2}{2} + \frac{u}{4} \int \frac{d^dk_1}{(2\pi)^d} \int \frac{d^dk_2}{(2\pi)^d} \int \frac{d^dk_3}{(2\pi)^d} \right]$$

$$\times \Phi_2(k_1)\Phi_2(-k_1) + u \int \frac{d^dk_1}{(2\pi)^d} \int \frac{d^dk_2}{(2\pi)^d} \int \frac{d^dk_3}{(2\pi)^d}$$

$$\times \Phi_2(k_1)\Phi_2(k_2)\Phi_2(k_3)\Phi^*_2(-k_1 - k_2 - k_3), \tag{7}$$

where $d$ is the dimensionality of the system. The bare inverse Green’s functions are given by $G_0^{-1} = [\tau_2/2 + t/2 + (1 - g_{12}\Phi_1^2)k^2 + pk^4]$. Incorporating fluctuations self-consistently, we have $G^{-1} = [\tau_2/2 + (1 - g_{12}\Phi_1^2)k^2 + pk^4]$, where, by the Dyson equation, $\tau_2/2 = r_2/2 + t/2 + \Sigma$. To lowest order in $1/n$, the self-energy is given by $\Sigma = \int \frac{d^4k}{(2\pi)^4} G(k)$, see Ref. [32]. This leads to a self-consistency equation for $\tau_2$. Similar self-consistency equations appear for $\Phi_1$; before the transition to an ordered $\Phi_2$ state, $\Phi_2 = -r_1$. A phase transition to an ordered state $\Phi_2 \neq 0$ occurs when the Green’s function acquires a pole on the real $k$ axis. If the pole is at $k_{\text{min}} = 0$, the transition is to a uniform phase of $\Phi_2$; if the pole first appears for $k_{\text{min}} \neq 0$, the transition is into a modulated phase.

When $[1 - g_{12}\Phi_1^2] > 0$ the minimum of $G^{-1}$ is always at $k = 0$, and both $\Phi_1$ and $\Phi_2$ may exhibit uniform orders. On the other hand, if $[1 - g_{12}\Phi_1^2] < 0$, the minimum for the $\Phi_2$ inverse Green’s function, $G^{-1}(k)$ occurs at $k_{\text{min}} = -[1 - g_{12}\Phi_1^2]/(2p_2)$ leading to a real axis pole when $\tau_2 = \tau_2_{\text{min}} = [1 - g_{12}\Phi_1^2]/(2p_2)$. The quartic $G^{-1}$ has two pairs of conjugate poles in the $k$ plane which lie on a circle of radius $\rho = (\tau_2/2p_2)^{1/4}$. The finite real component of the poles means that the correlation function $\langle \Phi_2(x)\Phi_2(y) \rangle$ exhibits sinusoidal modulations in addition to exponential decay. The modulation and correlation lengths are given, respectively, by

$$l_2 = 4\pi[\sqrt{\tau_2}/2 + (1 - g_{12}\Phi_1^2)/2]^{-1/2},$$

$$\xi_2 = 2[\sqrt{\tau_2}/2 - (1 - g_{12}\Phi_1^2)/2]^{-1/2}, \tag{8}$$

with $\Phi_1$ the uniform competing order field.

Irrespective of the spatial dimensionality, whenever $[1 - g_{12}\Phi_1^2] < 0$, as $\tau_2 \to \tau_2_{\text{min}}$ the self-energy diverges as $\Sigma \sim (\tau_2 - \tau_2_{\text{min}})^{-1/2}$. The phase transition which would occur (at the mean field level) when $\tau_2 = \tau_2_{\text{min}}$ is thwarted by the divergence of the self energy due to fluctuations. This implies that $T_2 = 0$ similar to systems with competing long range interactions. However, finite $n$ corrections (especially for $n = 1$), may make the transition temperature finite. In this case, this low temperature transition for $\Phi_2$ is of Brazovskii type, with a shell of minimizing modes.

A similar analysis holds for competing local orders in large $n$ quantum systems by extending. For bosonic fields, after a summation over Matsubara frequencies, the $\Phi_2$ correlator is

$$G_2(k) = \frac{\frac{1}{\tau_2} + n_B \left( \sqrt{\frac{1}{\tau_2} + (1 - g_{12}\Phi_1^2)k^2 + pk^4}/(k_B T) \right)}{\sqrt{\frac{1}{\tau_2} + (1 - g_{12}\Phi_1^2)k^2 + pk^4}},$$
with $n_B(x) = [\exp(x) - 1]^{-1}$. Order, at large $n$, is still inhibited in the quantum rendition of our system, although the divergence of the self-energy in this case is less severe than for its classical counterpart. In the bosonic system, due to integration over imaginary time, the $\Phi_2$ self energy diverges as $-\ln |\mathbf{T}_2 - \mathbf{T}_{2\text{ min}}|$ when $\mathbf{T}_2 \to \mathbf{T}_{2\text{ min}}$, whereas in the classical system it diverges as $|\mathbf{T}_2 - \mathbf{T}_{2\text{ min}}|^{-1/2}$.

The extensive value of $S_c$ implies that $N_m \propto e^V$, and strongly suggests glassiness for $T < T_K$34. The condition for possible glassiness formulated in Refs. 18,28 is that ratio of the coherence length to the modulation scale exceeds a number of order two. As seen from Eq. (5) in our model at low temperatures $\xi_2/l_2 \geq 2$ satisfying this condition. Once again, the realization of the glassy phase depends on the details of dynamics in a particular measurement, but extensive entropy makes such an outcome likely.

The high degree of low temperature entropy can be made rigorous. In all large $n$ (and several Ising) systems29, the extensive configurational entropy found at higher temperatures by replica calculations is supplemented by a ground state degeneracy scaling as the surface area of the system ($S_{\text{ground}} \propto q^{d-1}V^{(d-1)/d}$)28 in $d$ spatial dimensions. By explicit construction, these systems can be shown to possess a multitude of zero energy domain walls26. These low temperature excitations going hand in hand with a multiple metastable low energy states. Numerical simulations of single component systems in similar classical models of liquids also report exceptionally sluggish dynamics34,37 with strong indications of glassiness36. Thus, the non-uniform structures arising in our model of competing order parameters naturally exhibit slow dynamics and is likely to become glassy.

Summarizing, the field theoretical analysis accounting for fluctuations around the inhomogeneous minimizing structure extends the GL picture and strongly suggests the phase diagram shown in Fig. 2. For the low $n$ systems of relevance, the low temperature first order Brazovskii transition can be pre-empted by a transition into a glass.
V. RELEVANCE TO ELECTRONIC SYSTEMS.

We showed that, when there is a competition between two order parameters of different origin, and when a general symmetry allowed gradient-amplitude coupling in a local theory is negative (even if of moderate magnitude), the coexistence of two orders is inhomogeneous, and, generally, either the dynamics of the system is slow or a first order Brazovskii transition occurs. Crucially, even though we start with a local theory, the inhomogeneous coexistence leads to a low-energy theory of the same class as considered in models of self-generated glassiness due to competing length scales of interaction, although the origin of the phenomenon is very different.

Moreover, the transition temperatures for both order parameters are suppressed compared to the mean field value. We emphasize that the gradient-amplitude coupling is required to stabilize an inhomogeneous state: in its absence only uniform or phase-separated configurations are thermodynamically stable, as has been shown for stripe orders. Therefore in competing coexisting phases, $T_c$ is lower than in other parts of the phase diagram, structure factor measurements will indicate non-uniform order, and dynamical measurements likely display slow dynamics. The natural question to ask is what systems offer the best chance for realization of the model considered above.

Cuprates provide one obvious example of such competing orders when static low temperature spin and charge density waves (stripes) are inhibited in the presence of superconducting order. In these materials, STM measurements indicate incommensurate coexistence of superconductivity and a pseudo-gap state at nanoscale level; however, the dynamics in this situation is strongly energy dependent, which suggests that the mapping on a simple GL theory with temperature-independent coefficients is insufficient. At least in one example the scaling form of the dielectric function in the glassy state goes smoothly to quantum critical scaling as the glass transition temperature tends to zero.

Heavy fermion systems provide perhaps the best chance for observing the phenomena described here. In materials of the 115 family proximity or coexistence of antiferromagnetic and superconducting phases is now well established, and experiments indicate an inhomogeneous coexistence of the two orders in a magnetic field. Moreover, there is strong evidence that Cd and Hg dopants create antiferromagnetic regions in their vicinity, suggesting that the system is on the border of inhomogeneous coexistence of two orders. The Neel temperature drops precipitously if superconducting transition occurs first. No dynamical measurements have yet been carried out in the relevant regime of the phase diagram, but it would be interesting to see if, for example, in CeRhIn$_3$ under pressure the spin dynamics as determine by NMR shows signatures of slowing down or freezing at low temperatures.

In several systems the inhomogeneous coexistence was proposed in the presence of coupling terms that exist only under special circumstances. A particularly relevant example are manganites where the coupling due to deviation from half-filling that promotes the inhomogeneous coexistence of the magnetic and charge orders was proposed recently based on considerations similar to ours. As mentioned above, in these materials glassiness may emerge due to bona fide disorder, and not be self-generated. Non-trivial couplings appear in some of the multiferroic materials, e.g., spiral magnets such as RMnO$_3$ with $R=$ Tb, Ho, Dy.

It is important to note that, if we extend the treatment to include external parameters such as strain and field to act as a massive (i.e. with fluctuations towards order but no symmetry breaking since the quadratic coefficient in the GL expansion remains positive) "competing orders" within the GL framework, the resulting inhomogeneous state only occurs for moderately large coupling. One candidate for such a scenario is MnSi, where the low energy theory exhibiting these features has recently been put forward on the basis of Dzyaloshinskii-Moriya couplings.

In conclusion, we believe that many of the observed low temperature transitions, inhomogeneities, and slow dynamics/glassiness found in strongly correlated electronic systems are a natural consequence of competing local orders. As we illustrated, competing local orders may trigger inhomogeneities with likely first order transition or possible glassiness. In our calculations, the proliferation of incommensurate ground and metastable states is the common origin of both the dramatic lowering of the transition temperature or viable first order Brazovskii transition and possible glassy dynamics.

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