Droplet Phases in Non-local Ginzburg-Landau Models with Coulomb Repulsion in Two Dimensions

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Received: 14 May 2009 / Accepted: 4 May 2010
Published online: 29 July 2010 – © Springer-Verlag 2010

Abstract: We establish the behavior of the energy of minimizers of non-local Ginzburg-Landau energies with Coulomb repulsion in two space dimensions near the onset of multi-droplet patterns. Under suitable scaling of the background charge density with vanishing surface tension the non-local Ginzburg-Landau energy becomes asymptotically equivalent to a sharp interface energy with screened Coulomb interaction. Near the onset the minimizers of the sharp interface energy consist of nearly identical circular droplets of small size separated by large distances. In the limit the droplets become uniformly distributed throughout the domain. The precise asymptotic limits of the bifurcation threshold, the minimal energy, the droplet radii, and the droplet density are obtained.

1. Introduction

Spatial patterns are often a result of the competition between thermodynamic forces operating on different length scales. When short-range attractive interactions are present in a system, phase separation phenomena can be observed, resulting in aggregation of particles or formation of droplets of new phase, which evolve into macroscopically large domains via coarsening or nucleation and growth (see e.g., [1]). This process, however, can be frustrated in the presence of long-range repulsive forces. As the droplets grow, the contribution of the long-range interaction may overcome the short-range forces, whereby suppressing further growth. This mechanism was identified in many energy-driven pattern forming systems of different physical nature, such as various types of ferromagnetic systems, type-I superconductors, Langmuir layers, multiple polymer systems, etc., just to name a few [2–11]. Remarkably, these systems often exhibit very similar pattern formation behaviors [10,12].

One important class of systems with competing interactions are systems in which the long-range repulsive forces are of Coulomb type (for an overview, see [13,14] and references therein). The nature of the Coulombic forces may be very different from
system to system. For example, these forces may arise when particles undergoing phase separation carry net electric charge \[15–18\], or they may be a consequence of entropic effects associated with chain conformations in polymer systems \[19–23\]. Coulomb interactions may also arise indirectly as a result of diffusion-mediated processes \[4,24,25\]. All this makes systems with repulsive Coulombic interactions a ubiquitous example of pattern forming systems.

Studies of systems with competing short-range attractive interactions and long-range repulsive Coulomb interactions go back to the work of Ohta and Kawasaki, who proposed a non-local extension of the Ginzburg-Landau energy in the context of diblock copolymer systems \[19\]. Even though its validity for diblock copolymer systems may be questioned \[21,26–28\], the Ohta-Kawasaki model is applicable to a great number of physical problems of different origin \[14\]. On the other hand, mathematically the Ohta-Kawasaki model presents a paradigm of energy-driven pattern forming systems which has been receiving a growing degree of attention \[9,29–37\].

The Ohta-Kawasaki energy is a functional of the form \[13,14,19,24,38\]:

\[
\mathcal{E}[u] = \int_{\Omega} \left( \frac{\varepsilon^2}{2} |\nabla u|^2 + W(u) \right) dx + \frac{1}{2} \int_{\Omega} \int_{\Omega} (u(x) - \bar{u}) G_0(x, y)(u(y) - \bar{u}) dx dy. 
\] (1.1)

Here, \( u : \Omega \to \mathbb{R} \) is a scalar quantity denoting the “order parameter” in a bounded domain \( \Omega \subset \mathbb{R}^d \). Different terms of the energy are as follows: the first term penalizes spatial variations of \( u \) on the scales shorter than \( \varepsilon \), the second term, in which \( W \) is a symmetric double-well potential drives local phase separation towards the minima of \( W \) at \( u = \pm 1 \), and the last term is the long-range interaction, whose Coulombic nature comes from the fact that the kernel \( G_0 \) solves the Neumann problem for

\[
- \Delta G_0(x, y) = \delta(x - y) - \frac{1}{|\Omega|}, \quad \int_{\Omega} G_0(x, y) dx = 0, 
\] (1.2)

where \( \Delta \) is the Laplacian in \( x \) and \( \delta(x) \) is the Dirac delta-function. The parameter \( \bar{u} \) denotes the prescribed uniform background charge, and the overall “charge neutrality” is ensured via the constraint

\[
\frac{1}{|\Omega|} \int_{\Omega} u dx = \bar{u}. 
\] (1.3)

It is important to note that the kernel \( G_0 \) solves (1.2) in the space of the same dimensionality as the order parameter \( u \) (not to be confused with the case in which the kernel solves Laplace’s equation in the space of higher spatial dimensionality, as is common in many other systems with competing interactions, see e.g. \[7,16\]).

The parameter \( \varepsilon > 0 \) in (1.1) determines both the scale of the short-range interaction and the magnitude of the interfacial energy between the regions with different values of \( u \) when \( \varepsilon \) is sufficiently small. In fact, it is known that no patterns can form in the system if \( \varepsilon \) is sufficiently large \[13,14,39\]. On the other hand, when \( \varepsilon \ll 1 \), the first term in the functional \( \mathcal{E} \) becomes a singular perturbation, giving rise to “domain structures” (see Fig. 1), which are of particular physical interest. These patterns consist of extended regions in which \( u \) is close to one of the minima of the potential \( W \), separated by narrow domain walls. In this situation one can reduce the energy functional appearing in (1.1)