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Existence of pearled patterns in the planar functionalized Cahn–Hilliard equation

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Abstract

The functionalized Cahn–Hilliard (FCH) equation supports planar and circular bilayer interfaces as equilibria which may lose their stability through the pearling bifurcation: a periodic, high-frequency, in-plane modulation of the bilayer thickness. In two spatial dimensions we employ spatial dynamics and a center manifold reduction to reduce the FCH equation to an 8th order ODE system. A normal form analysis and a fixed-point-theorem argument show that the reduced system admits a degenerate 1:1 resonant normal form, from which we deduce that the onset of the pearling bifurcation coincides with the creation of a two-parameter family of pearled equilibria which are periodic in the in-plane direction and exponentially localized in the transverse direction.

Keywords: Functionalized Cahn–Hilliard; Pearled bilayer; Spatial dynamics; Normal form; Singular perturbation

1. The functionalized Cahn–Hilliard equation

Amphiphilic materials are typically small molecules which contain both hydrophilic and hydrophobic components. This class of materials includes surfactants, lipids, and block copolymers. Their propensity to spontaneously assemble network morphologies has drawn scientific attention for more than a century, [1]. While amphiphilic materials are ubiquitous in organic settings, where lipid bilayers form cell membranes and many organelles, their widespread use

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as charge separators in energy conversion devices is more recent. Network morphologies must be distinguished from single layer interfaces that are typical of binary metals and other purely hydrophobic blends. While single layer interfaces separate a phase A from a phase B, network morphologies are comprised of thin regions of a phase B which interpenetrate, and typically percolate through, a domain dominated by phase A. The Cahn–Hilliard free energy, proposed in 1958, [5], has been very successfully employed as a model of single layer morphology in hydrophobic blends, and its gradient flows accurately describe their evolution. Models of amphiphilic mixtures, such as [24] and [12], have been proposed. The functionalized Cahn–Hilliard free energy, [19,10,6], is a special case of these earlier models that supports stable network morphologies including co-dimension one bilayers and co-dimension two pores as well as pearled morphologies and defects such as end-caps and junctions. Rigorous results for the FCH free energy include the existence of bilayer structures, [7], and an analysis of their bifurcation structure, [14], in particular the pearling bifurcation which initiates changes in the co-dimension of the underlying morphology, and is commonly observed in amphiphilic polymer blends, see [4,27]. The goal of this paper is to rigorously establish the existence of pearled bilayers, as modulations to stationary bilayers, in the planar FCH equation.

Amphiphilic mixtures, such as emulsions formed by adding a minority fraction of an oil and soap mixture to water, form network morphologies due to the tendency of the surfactant phase, e.g. soap, to enhance the formation of interfaces. To model the network formation, the authors of [24] and [12] were motivated by small-angle X-ray scattering (SAXS) data to include a higher-order term in the usual Cahn–Hilliard expansion for the free energy. Viewing the mixture as a binary phase, where $u \in H^2(\Omega)$ denotes the volume fraction of surfactant contained within the bounded material domain $\Omega \subset \mathbb{R}^3$, they proposed a free energy of the form

$$\mathcal{F}(u) := \int_\Omega f(u) + \epsilon^2 A(u)|\nabla u|^2 + \epsilon^2 B(u)\Delta u + C(u)(\epsilon^2\Delta u)^2\, dx, \tag{1.1}$$

where for well-posedness $C > 0$ and the dimensionless parameter $\varepsilon \ll 1$ dictates the ratio of the interfacial width to a characteristic size of Ω. Assuming zero-flux boundary conditions, integration by parts on the $A(u)$ term permits a re-writing of the energy in the completed-square form

$$\mathcal{F}(u) = \int_\Omega C(u)\left(\epsilon^2\Delta u - \frac{\overline{A} - B}{2C}\right)^2 + f(u) - \frac{(\overline{A} - B)^2}{4C(u)}\, dx, \tag{1.2}$$

where $\overline{A}$ is a primitive of A. To simplify the form we replace $C(u)$ with $\frac{1}{2}$, relabel the potential within the squared term by $W'(u)$, and scale the potential outside the squared term as $\delta P(u)$ with $\delta \ll 1$, yielding

$$\mathcal{F}(u) = \int_\Omega \frac{1}{2}\left(\epsilon^2\Delta u - W'(u)\right)^2 + \delta P(u)\, dx. \tag{1.3}$$

The first term is the square of the variational derivative of a Cahn–Hilliard type free energy $\mathcal{E}(u) := \int_\Omega (\frac{\varepsilon^2}{2}|\nabla u|^2 + W(u))dx$, and the strongly degenerate case $\delta = 0$, has the special property