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MESOSCOPIC MORPHOLOGY AND SCALING LAW OF STATIONARY INTERFACIAL PATTERNS FOR REACTION DIFFUSION SYSTEMS

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ABSTRACT. A scaling law for stationary interfacial patterns is considered. When there is a micro-scopic constraint in the system like block copolymer, the resulting steady states display a variety of patterns from lamellar to labyrinthine ones. The goal is to determine the characteristic domain size in terms of interfacial thickness and the nonlocal interaction length. The result is consistent with the experiments.

The morphology of *final* patterns in phase transition is usually a simple one: only one phase dominates the whole domain (non-conserved) or it is decomposed into simple subdomains (conserved) after the coarsening process. This is due to the tendency to minimize the area of interface. However, if there is a microscopic constraint to the system, the final pattern becomes much richer and has in general a variety of morphologies from lamellar to labyrinthine patterns. Block copolymer is one of such materials where two monomers (say, A and B) are connected at some point (constraint), and this is responsible for the formation of very fine and complicated structures depending on the ratio of composite monomers in the process of micro-phase separation [4, 2, 16]. Locally monomers move in a random way and tend to segregate each other (bistability), but connectivity does not allow them to form a large domain consisting of one monomer (nonlocality) only. One of the important issues is to clarify how the domain size L (i.e., characteristic wave length) depends on the polymerization index N (i.e., the chain-length of block copolymer). Experimentally it is confirmed that L is proportional to $N^{2/3}$ [3]. One of the goals of this short note is to outline how we can set up a mathematical framework to justify this scaling law. It should be noted that N is usually quite large and, hence, the domain size lies in between micro- and macro-scales. Such intermediate scale is called the *mesoscopic*

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one. Ohta and Kawasaki [11] proposed the following model system for non-conserved case to describe.

$$\begin{cases} u_t = \varepsilon^2 \Delta u + f(u,v) & \text{in } \ \Omega, \\ 0 = \Delta v + D^{-1} g(u,v), & \\ \dfrac{\partial u}{\partial n} = 0 = \dfrac{\partial v}{\partial n} & \text{on } \ \partial\Omega. \end{cases} \tag{1}$$

where u is the *order parameter* (say, ranging from -1 to 1) indicating A-rich (≈ -1) or B-rich (≈ 1) phase, v represents the nonlocal effect due to connectivity, $\varepsilon(\ll 1)$ corresponds to the interfacial thickness, $D(\gg 1)$ is proportional to N^2, $f(u,v)$ is a cubic nonlinearity (typically of the form $u - u^3 - v$) and Ω is a smooth domain in $\mathbb{R}^n$. Possibly, many other phenomena could be described by models similar to (1), since the basic mechanism creating a variety of patterns is due to the competition between local dynamics and nonlocal effect. In fact similar patterns are observed in liquid crystal, magnetic thin film, and so on. A naive approach to find nontrivial patterns of (1) is to consider the limiting case of either $\varepsilon \downarrow 0$ or $D \uparrow \infty$. In the latter case it is known (see [5, 6]) that the resulting equations become a scalar equation with a constraint of integral type and it is unlikely to have a stable complicated pattern for such a system, since there are no stable multi-layered solutions even in 1D case [7]. On the other hand we know very little about the former case in higher space dimensions, since it has been regarded to be extremely difficult to find the first approximate stationary solutions in the limit of $\varepsilon \downarrow 0$. Our main interest is the behavior of the asymptotic configuration of the interface Γ^ε (0-level contour of the order parameter u) and its dependency on ε and D. It turns out that, under several technical conditions, generically there are no smooth interfacial patterns up to $\varepsilon = 0$.

THEOREM 1. (Nonexistence of Smooth Limiting Interface [9]**).**

(a) (*Disk Symmetry.*) *Suppose that* (1) *with* $n = 2$ *has an* ε*-family of matched asymptotic solutions with a simple closed smooth interface* Γ^ε *up to* $\varepsilon = 0$ *and that the first order matching condition holds* (*see Lemmas 3.4.1 and 3.4.2 of* [9]). *Then* Γ^0 *must be a circle.*

(b) (*Non-existence.*) *Moreover under the assumption that a uniqueness result for the reduced elliptic problem* (*the first outer approximation of* (1)) *holds outside of the limiting interface* Γ^0 (*see Hypothesis 4.1 of* [9]), *then* (a) *implies that the reduced elliptic problem has no solutions for generic domains* Ω, *and hence there does not exist an associated* ε*-family of matched asymptotic solutions.*

Apparently, for special domains like rectangles and spheres, there are ε-families of interfacial patterns whose interfaces are smooth up to $\varepsilon = 0$. However, even if such non-generic patterns exist, they can not remain stable as solutions of (1). Namely we have

THEOREM 2. **(Instability Theorem** [10]. *Suppose that (1) with $n = 2$ has an ε-family of matched asymptotic solutions whose interface is smooth up to $\varepsilon = 0$. Then, it must become unstable for small ε.*

The above non-existence and instability theorems are not dissapoiting results. In fact, they suggest an important thing about the behavior of the interface as $\varepsilon \downarrow 0$. Namely, if some stationary pattern of (1) exists up to $\varepsilon = 0$, but does not have a smooth limiting interface, then the configuration of the interface must become fine and complicated as $\varepsilon \downarrow 0$. In order to understand the morphology of the complicated patterns, it seems necessary to apply an appropriate rescaling to blow up the degenerate situation. There are at least two ways for patterns to become complicated depending on the ratio of composite of two species.

(a) *Dot pattern.* If one of the species dominates the other, the minority one is surrounded by the other, and the configuration of the interface becomes circular-like. This is the dot case. Due to the connectivity, the diameter of the disk depends not only on ε but also on D and becomes smaller (resp. larger) when $\varepsilon \downarrow 0$ (resp. $D \uparrow \infty$). The issue is to determine the dependency of diameter of this unit cell on ε and D.

(b) *Snaky pattern.* On the other hand if the two species have almost the same ratio of composite, its morphology looks like a labyrinthine pattern which corresponds to the Snaky case. This could be created by a sequence of tip-splitting instabilities starting from a smooth interfacial pattern. In this case the characteristic width of snake should be determined by ε and D. For both cases, suppose that such patterns exist, by applying the *marginal stability criterion*, we have the following result in 2D case, which is consistent with the experiment.

MAIN THEOREM. *The characteristic domain size of stable patterns of* (1) *is proportional to* $(\varepsilon D)^{1/3}$.

Idea of proof. We sketch the main idea of the proof for the case of dot pattern. First we assume that it is a periodic pattern in 2D space and we restrict the problem to a unit cell (denoted by U) of it. Also we assume that both u and v satisfy the zero flux boundary conditions on ∂U. Now we rescale the diameter of U by ε^{α} $(0 < \alpha < 1)$. Here we assume that the dot pattern becomes finer and finer so that $\lim\{\text{diameter of } U\}/\varepsilon^{\alpha}$ = non-zero constant. The rescaled system takes the following form

$$\begin{cases} u_t = \tilde{\varepsilon}^2 \Delta u + f(u,v) & \text{in } \widetilde{U}, \\ 0 = \Delta v + D^{-1} \tilde{\varepsilon}^{\frac{2\alpha}{1-\alpha}} g(u,v), & \\ \dfrac{\partial u}{\partial n} = 0 = \dfrac{\partial v}{\partial n} & \text{on } \partial \widetilde{U}, \end{cases} \tag{2}$$

where $\tilde{\varepsilon} = \varepsilon^{1-\alpha}$ and $\widetilde{U}$ is the rescaled unit region of U (one has to be careful about the inverse of Laplacian when $g(u,v) = u$). There are two problems we have to consider:

(i) For which α and D does (2) have a layered solution with a smooth interface up to $\varepsilon = 0$?

(ii) Is the resulting solution obtained in (i) locally stable as a solution of (2)? Although the first problem (i) is not yet completely solved, the second stability criterion is sufficient for the selection of the unique exponent $1/3$ as a necessary condition. The extension of the idea of the SLEP method ([7, 8, 14]) to higher space dimensions is useful for the application of the marginal stability criterion.

In view of the scaling law in the Main Theorem, we see that an interfacial pattern of *any scale* could be observed as a stable solution to (1) if we take ε and D appropriately. As a special case, let D tend to ∞ at least with the order $O(1/\varepsilon)$, then it follows from the above Main Theorem that the resulting pattern should be $O(1)$. The Allen–Cahn equation with mass conservation is included into this case. Finally it should be noted that the exponent $1/3$ coincides with that of the fastest growing wave length of the perturbation to the planar front (see [14, 15]). This is not an accidental coincidence, since the snaky pattern should be created through the successive instabilities of tip-splitting type which could be regarded as instabilities of local planar fronts.

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