Patterns in triblock copolymers

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11ième biennalle française SMAI

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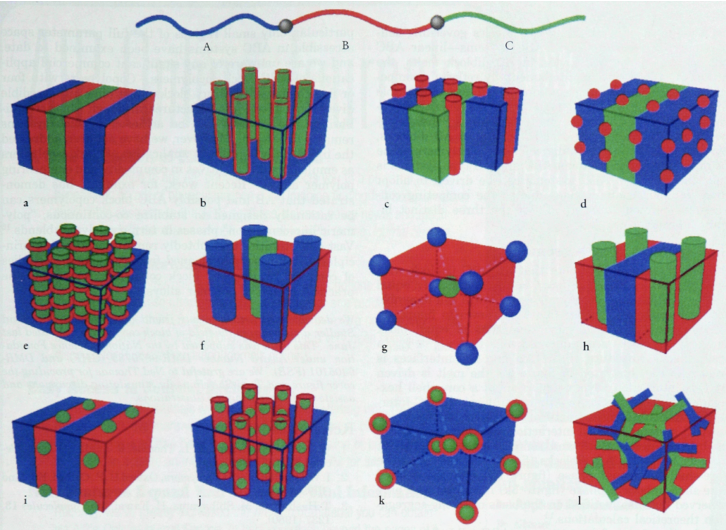
Triblock copolymers

There are three repelling monomer strands which are bonded together



Bates-Fredrickson; Zheng-Wang 1995

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- Vector order parameter, $u = (u_0, u_1, u_2)$, $u_i = \chi_{\Omega^i}$, with $u_0 + u_1 + u_2 = 1$ on $\mathbb{T}^d$.
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- We will consider dilute configurations, with $m_1, m_2 \ll m_0$, as in Choksi-Peletier with $0 < \eta \ll 1$, $m = M\eta^d$, strong nonlocal interaction $\gamma \sim \eta^{-3}$; $\gamma \sim [\eta^2 |\ln \eta|]^{-1}$, $d = 3, 2$.

The Isoperimetric Problem

First, consider the local term, but for $\Omega^i \subset \mathbb{R}^d$, $i = 1, 2$, $\Omega^0 = \mathbb{R}^d \setminus (\Omega^1 \cup \Omega^2)$, with $m_1 = |\Omega^1|$, $m_2 = |\Omega^2|$ given.

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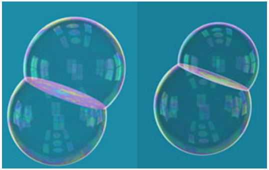
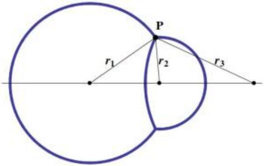
Case of equal surface tensions, $\sigma_{ij} = 1$, $\forall i, j$. For all $m_1, m_2 > 0$, the minimizer is a double bubble with equal contact angles at the junctions.





Left: J. Sullivan, nsf.gov. Right: Chong Wang

each chamber being bounded by spherical (circular, in 2D) patches.

- $n = 2$: Alfaro, Brock, Foisy, Hodges, & Zimba (1993);
- $n = 3$: Hutchings, Morgan, Ritoré, & Ros(2000).

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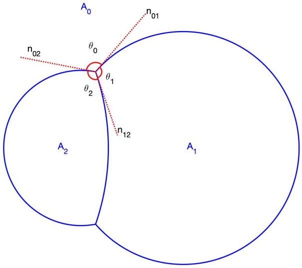
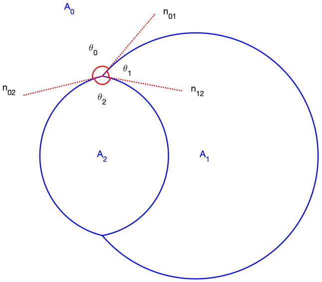
Minimizers are double bubbles (Lawlor) but with unequal angles:





The circular arcs meet at triple junctions according to Young's Law (see also Mullins, Bronsard-Reitich):

$$\sum_{i \neq j} \sigma_{ij}\, n_{ij} = 0, \text{ equivalently } \frac{\sin\theta_1}{\sigma_{02}} = \frac{\sin\theta_2}{\sigma_{01}} = \frac{\sin\theta_0}{\sigma_{12}}$$

where n_{ij} is the normal vectors to the arc separating phases i and j.

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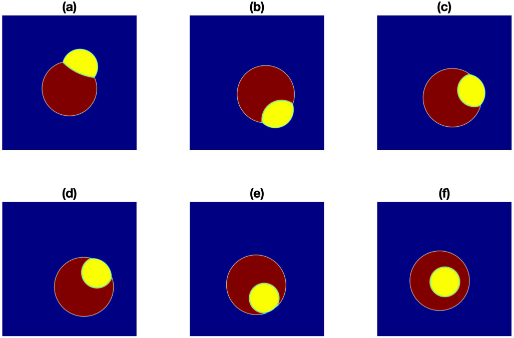

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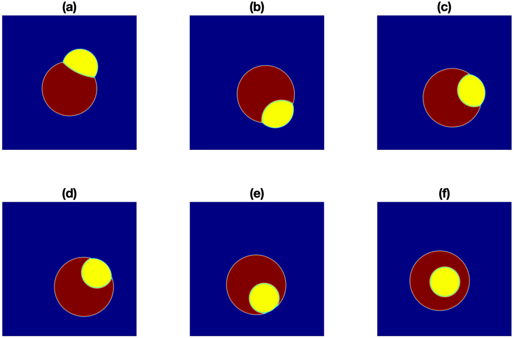
- The minimizer is degenerate since the location of the inner disk is free. In fact, the inner disk can even be tangential to the boundary of the outer disk.



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Construction of solutions of the Euler-Lagrange equations via Lyapunov-Schmidt. [Equal surface tensions $\sigma_{ij} = 1$]

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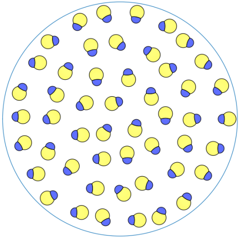

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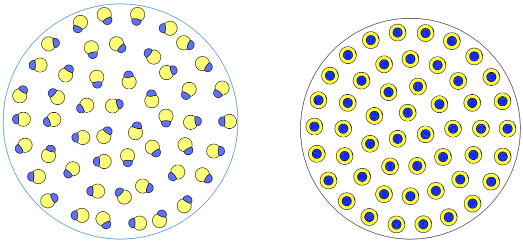

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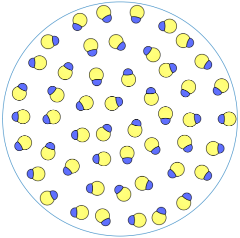

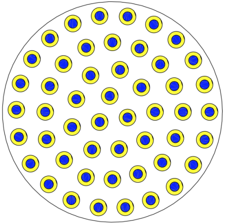

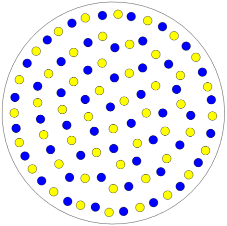

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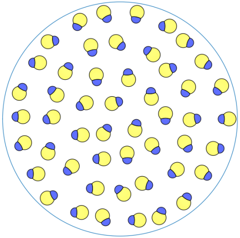

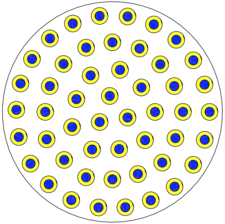

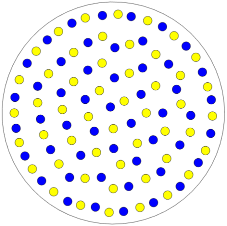

Question: what do global minimizers in the 2D torus T^2 look like for dilute regimes?

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- The energy rescales to:

$$E_\eta(v_\eta) := \sum_{i=0,1,2} \eta \int_{T^2} \beta_i |\nabla v_\eta^i| + \frac{1}{2|\ln \eta|} \sum_{i,j=1,2} \Gamma_{ij} \int_{T^2} \int_{T^2} v_\eta^i(x)\, G(x-y)\, v_\eta^j(y)\, dx\, dy,$$

  where $\beta_i \geq 0$ encode the surface tensions σ_{ij}.
  We look for minimizers as $\eta \to 0$.

Concentration Theorem (Alama-B-Lu-Wang)

Let $v_\eta = \eta^{-2}\chi_{\Omega_\eta}$, $\Omega_\eta = (\Omega_\eta^1, \Omega_\eta^2)$ be minimizers of E_η. Then, there exists a subsequence $\eta \to 0$,

- $N \in \mathbb{N}$ and distinct points $\xi_1, \ldots, \xi_N \in \mathbb{T}^2$;
- points $x_{\eta,k} \in T^2$, with $x_{\eta,k} \to \xi_k$, $k = 1, \ldots, N$;
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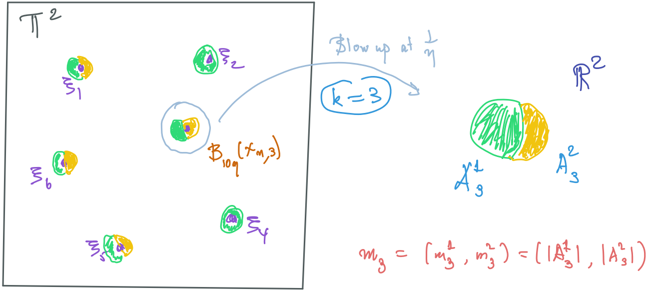

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- The blow-up components A_k minimize weighted perimeter $P_{\mathbb{R}^2}(A_0, A_1, A_2)$, in $\mathbb{R}^2$, among clusters with mass $(m^1_k, m^2_k) = (|A^1_k|, |A^2_k|)$.

Concentration Theorem (Alama-B-Lu-Wang)

Let $v_\eta = \eta^{-2}\chi_{\Omega_\eta}$, $\Omega_\eta = (\Omega^1_\eta, \Omega^2_\eta)$ be minimizers of E_η. Then, there exists a subsequence $\eta \to 0$,

- $N \in \mathbb{N}$ and distinct points $\xi_1, \dots, \xi_N \in \mathbb{T}^2$;
- points $x_{\eta,k} \in T^2$, with $x_{\eta,k} \to \xi_k$, $k = 1, \dots, N$;
- finite perimeter clusters $A_k = (A^1_k, A^2_k)$, $A^0_k = (A^1_k \cup A^2_k)^c$, in $\mathbb{R}^2$, $k = 1, \dots, N$, with
$$\left| \Omega_\eta \triangle \bigcup_{k=1}^{N} (\eta A_k + x_{\eta,k}) \right| \longrightarrow 0,$$
- The blow-up components A_k minimize weighted perimeter $P_{\mathbb{R}^2}(A_0, A_1, A_2)$, in $\mathbb{R}^2$, among clusters with mass $(m^1_k, m^2_k) = (|A^1_k|, |A^2_k|)$.
- The energy decomposes as
$$E_\eta(v_\eta) = \sum_{k=1}^{N} e_0(m_k) + O(|\ln \eta|^{-1}),$$
where $e_0(m_k) = \inf \left\{ E_0(A) \,|\, |A^1| = m^1, |A^2| = m^2 \right\}$, and
$$E_0(A) := P_{\mathbb{R}^2}(A^0, A^1, A^2) + \frac{1}{4\pi} \sum_{i,j=1,2} \Gamma_{ij} m^i\, m^j,$$

Remarks

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Once $m_k = (m_k^1, m_k^2)$ are known, the minimizers are as discussed earlier. However, the interaction term Γ_{12} plays a role in choosing m_k^i...

The role of Γ_{12}

Assume equal surface tensions $\sigma_{ij} = 1$. The isoperimetric problem prefers double bubbles, provided both $m^1, m^2 > 0$.

Proposition: $\exists$ explicit $\Gamma_{12}^* = \Gamma_{12}^*(\Gamma_{11}, \Gamma_{22})$ so that when $\Gamma_{12} > \Gamma_{12}^*$ and $M^i \geq 4m_*^i$, $i = 1, 2$, the minimizer has only single bubbles (disks).

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Luo-Ren-Wei: rectangular lattices for certain parameters.

Double bubbles?

To have double bubbles, the interaction $\Gamma_{12} = 0$ or very small.

Proposition

1. *Given* $\Gamma_{ii} > 0$*, for all* $M^i < \min\{m_*^i, \pi\Gamma_{ii}^{-2/3}\}$ *there exists* $\overline{\Gamma}_{12}(\Gamma_{ii}, M^i) > 0$ *so that if* $\Gamma_{12} < \overline{\Gamma}_{12}$*, then minimizers consist of exactly one double-bubble.*

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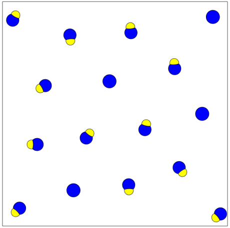

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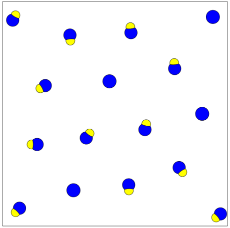
3. *If there are single-bubbles, they must be of the same species and all have the same mass.*



When there is coexistence of single and double-bubbles, all of the single bubbles must be of the same phase. In some sense, coexistence occurs in a minimizer when there is a large enough excess of one phase compared with the other.

Double Bubbles?

Open question: prove that minimizers must form all double bubbles, in some region of the parameter space (M^1, M^2, Γ). VIDEO

Core Shells

The problem of adjacency:

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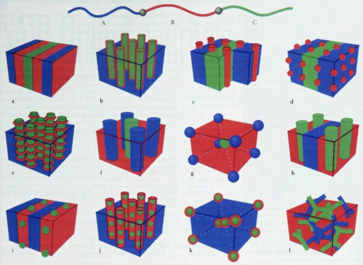

Core Shells

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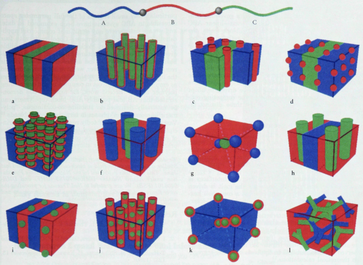
- Microscopically, an ABC-copolymer interposes a B-strand between the A, C monomers.
- Macroscopic patterns should penalize A-to-C transitions.
- Suggests degenerate case $\sigma_{02} = \sigma_{01} + \sigma_{12}$ more physically appropriate.



Minimizers with core shells

VIDEO

Resolving the degeneracy

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This is the term which will resolve the degeneracy in core-shells:

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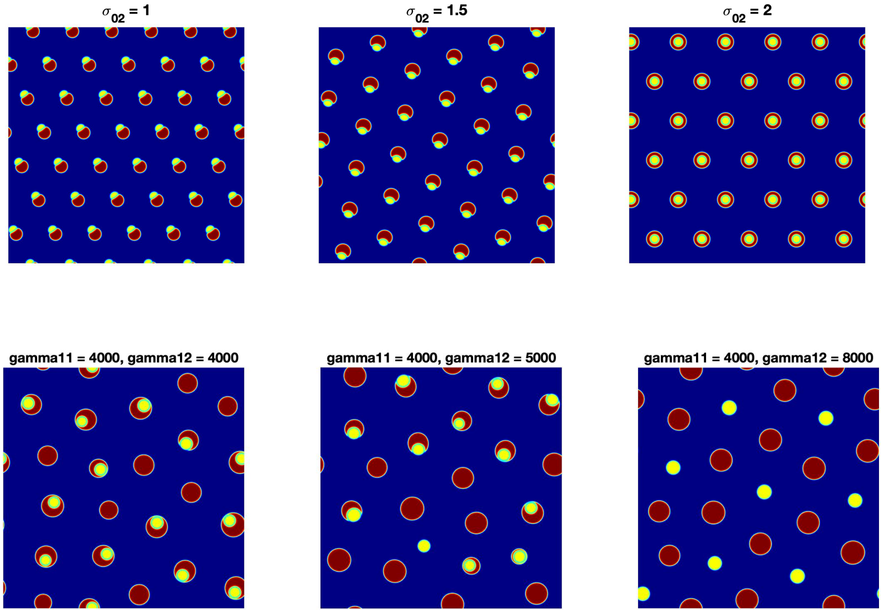
Proposition

In case $\sigma_{02} = \sigma_{01} + \sigma_{12}$, and $m = (m_1, m_2)$ with $m_1, m_2 > 0$.

(a) *If $\Gamma_{11} > \Gamma_{12}$, then the minimum in $f_0(m)$ is attained by a concentric core shell $A = \mathcal{C}_{m_2}^{m_1}$.*

(b) *If $\Gamma_{11} < \Gamma_{12}$, then the minimum in $f_0(m)$ is attained by a core shell $A = \mathcal{C}_{m_2}^{m_1}$ whose inner boundary circle is tangent to the outside circle.*



σ_{02} = 1
σ_{02} = 1.5
σ_{02} = 2
gamma11 = 4000, gamma12 = 4000
gamma11 = 4000, gamma12 = 5000
gamma11 = 4000, gamma12 = 8000