Pattern Design for Directed Self-Assembly of Block Copolymers by Computational Evolutionary Strategy

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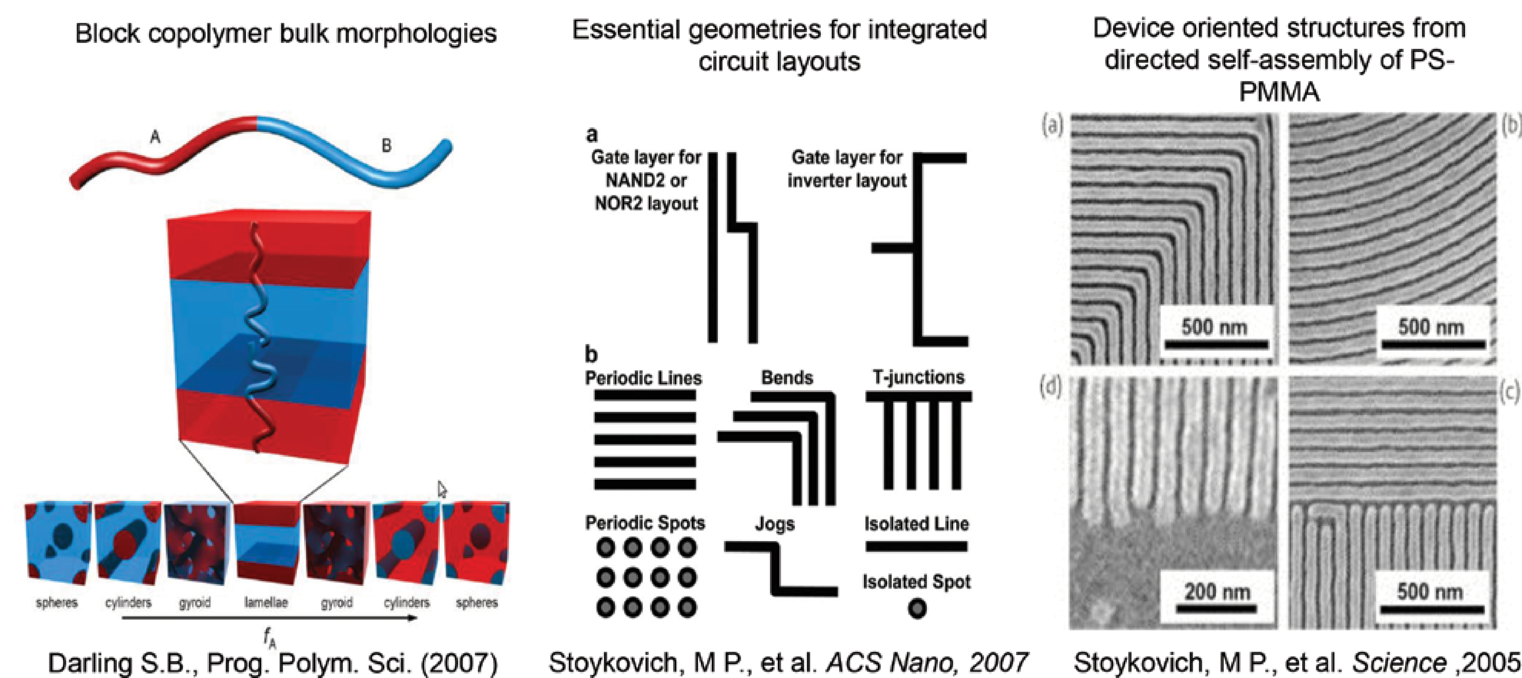
Introduction

- Directed self-assembly of block copolymers is a promising technique for fabrication of ultra-small feature dimensions
- Device oriented structures require assembly into non-bulk morphologies which are guided by external fields
- Design of external guiding pattern and choosing appropriate processing conditions is non-trivial as pattern may not have one-to-one correspondence with the target morphology
- Traditionally used trial-and-error methods become intractable with the increase in number of design variables



Darling S.B., Prog. Polym. Sci. (2007) Stoykovich, M P., et al. *ACS Nano*, 2007 Stoykovich, M P., et al. *Science* ,2005

Scope of Molecular Simulations

- Simulations help to better understand the underlying phenomena taking place during the self assembly process
- Complete 3D information about the morphology (difficult to get through experiments)

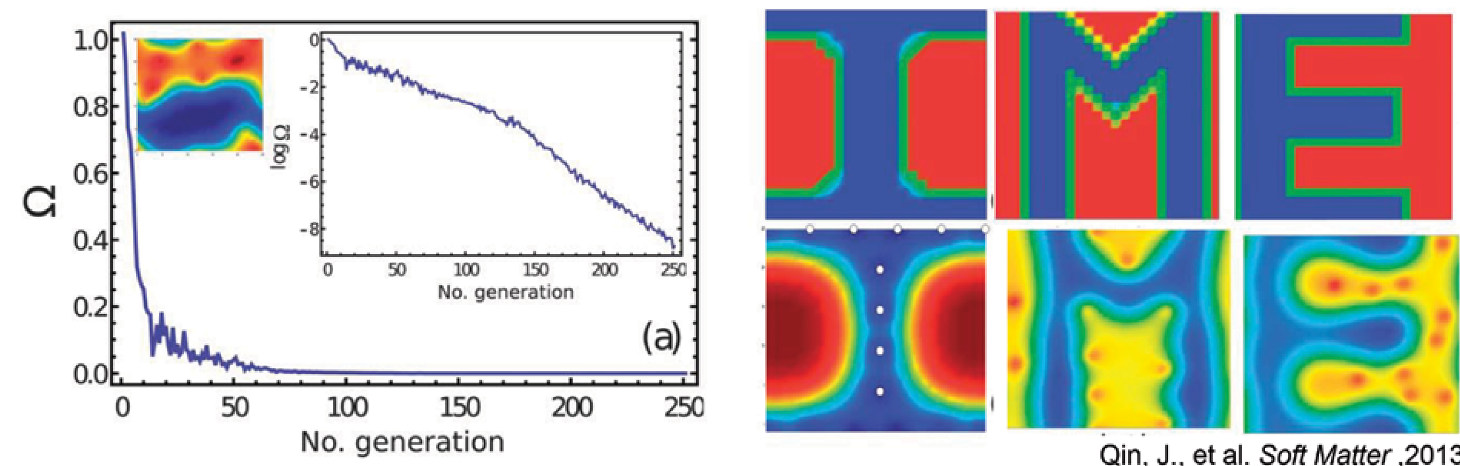
Method

- Essential physics of block copolymer system is represented in the mathematical description.
- Efficient numerical scheme enables access to large length and time scales.
- Equilibrium morphology minimizes the total system free energy:

$$\frac{H}{k_B T} = \frac{3}{2}\sum_{i=1}^{n}\sum_{s=1}^{N-1}\frac{[\mathbf{r}_i(s+1)-\mathbf{r}_i(s)]^2}{b^2} + \sqrt{\bar{N}}\int_V \frac{d\mathbf{r}}{R_e^3}\left[\chi N \phi_A \phi_B + \frac{\kappa N}{2}(\phi_A+\phi_B-1)^2\right] + H_{ext}$$

Optimizing Block Copolymer Self-Assembly by Evolutionary Computing

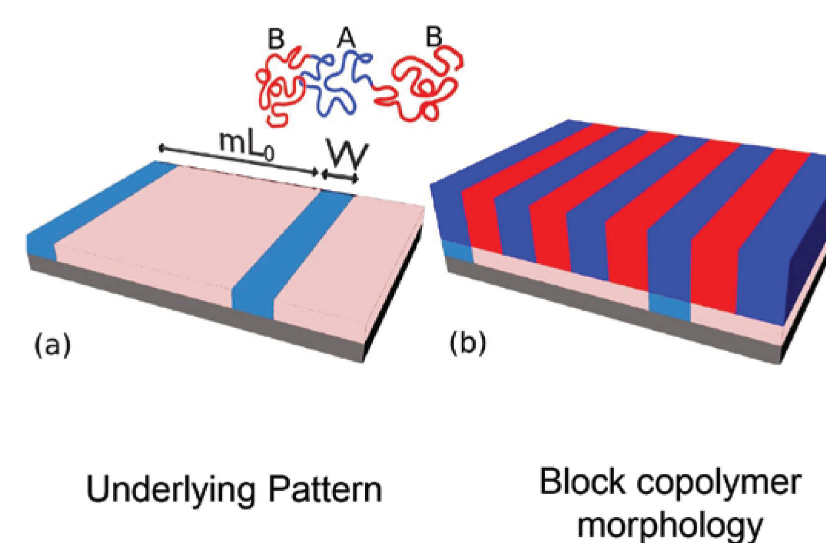
- Morphologies relevant to semiconductor devices require external guiding fields, e.g. chemical patterns on substrate
- Finding the optimal chemical pattern corresponding to a desired morphology is challenging
- Traditional trial-and-error and random search methods become intractable with increase of independent variables



Qin, J., et al. *Soft Matter* ,2013

- We used a simple 2D block copolymer model coupled with Covariance Matrix Adaptation Evolution Strategy (CMA-ES) to search for optimal arrangements of attractive spots which directs the desired morphology
- The objective function (Ω) which defines the similarity between a given morphology and a target morphology is minimized by smart search of the parameter space

- **Optimal chemical pattern for pattern interpolation**

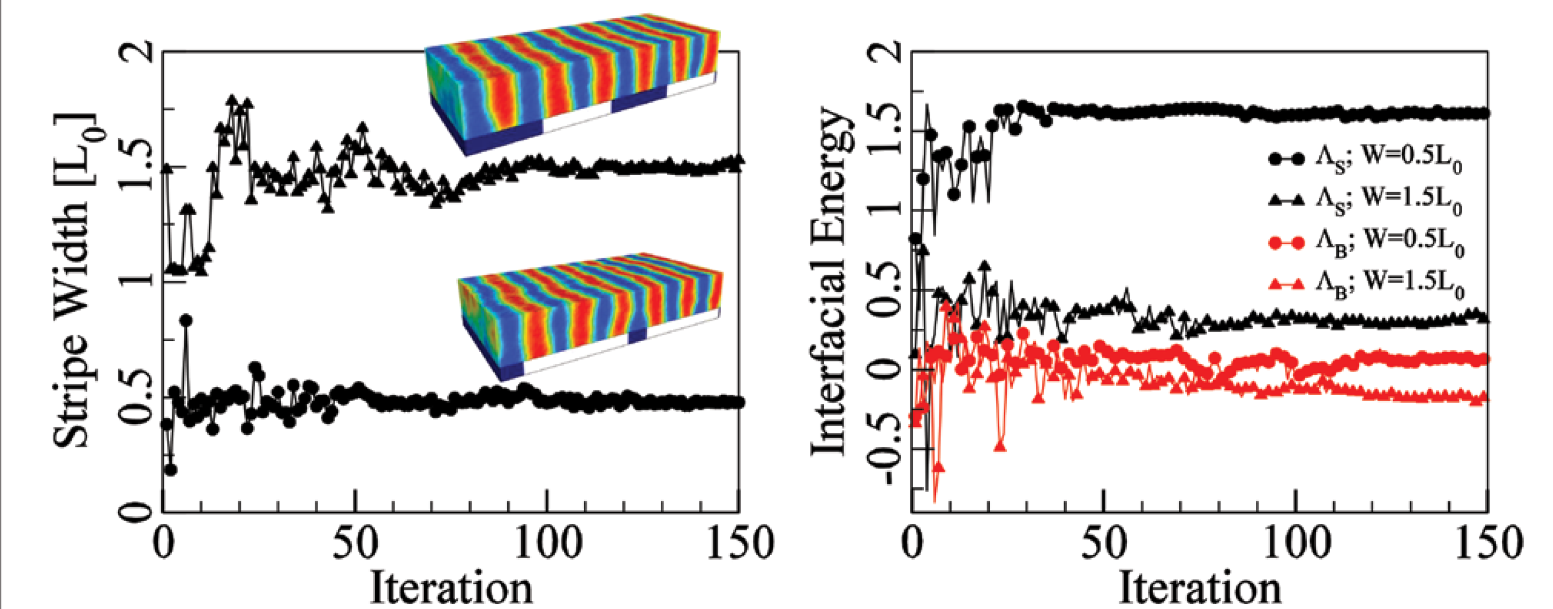


Underlying Pattern Block copolymer morphology

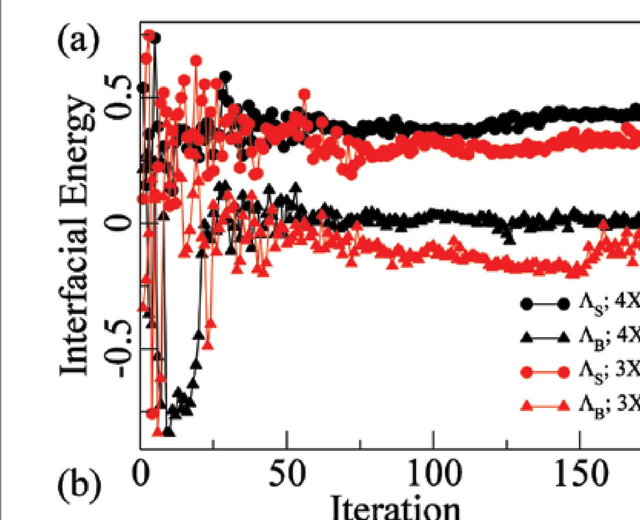
- Coarser chemical patterns could be printed using the conventional lithography and the resolution is further increased by assembling block copolymer

- CMA-ES coupled with more realistic 3D molecular simulations can find optimal pattern geometry and chemistry of the coarser pattern
- Variables to be Optimized
  - Geometry: Width of the guiding stripe
  - Surface Chemistry: Strength of Stripe (Λ_S) and Background (Λ_B)

Optimal Pattern Interpolation Conditions



- For both 3X and 4X density multiplication, two stripe width regimes for optimal assembly: 0.5L_0 guiding and 1.5L_0 guiding
- Stronger pattern strength and more neutral background needed for 0.5L_0 guiding

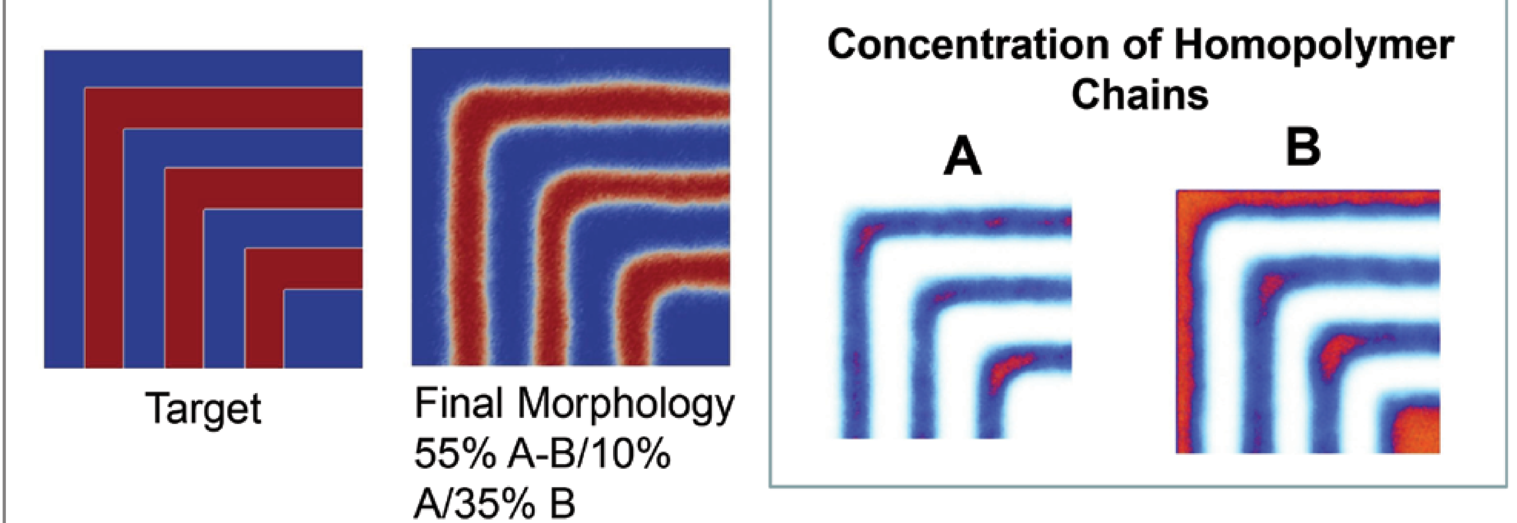


- For 1.5L0 guiding, stronger pattern is predicted in 4X case
- Background is more neutral in 4X density multiplication than 3X
- More fluctuations in 3X parameters suggest wider operational window

Khaira, G.S. et al. *ACS Macro Letters*, 2014

Optimal Blends for Perpendicular Bends

- Optimal block copolymer blend composition was searched simultaneously with the pattern variables
- The target was designed artificially with no interface
- Hompolymers concentrate at the bends to stabilize the high local curvature



Target Final Morphology 55% A-B/10% A/35% B

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