

Supplementary Information

Metastability in pixelation patterns of coexisting fluid lipid bilayer phases supported by e-beam patterned substrates

Maria O. Ogunyankin and Marjorie L. Longo

Submitted to Soft Matter

Derivation of Equation 3

The diffusion driven dissolution of the domain pattern of total perimeter, P_t , via a concentration gradient in mole fraction ($C_i - C_b$) of domain lipids at the L_o - L_d interface can be described by equation (1):

$$\frac{dC_b}{dt} = \frac{DP_t}{A_{array}r} (C_i - C_b) \quad (1)$$

Equation (1) assumes the length over which diffusion is taking place is equal to the radius, r , of a single circular domain pixel in a quiescent media¹ and that the bulk area is approximated by the projected area of each array, A_{array} .

Assuming the domain lipid concentration at a flat (not curved) domain interface is the same as the bulk, the Gibbs-Thompson boundary condition gives $(C_i - C_b) = C_b v/r$ for a curved domain interface where $v = 2\sigma A_m C_b / RT$; v is the capillary length, σ is the line tension of the domain interface, and A_m is the area per mole of domain lipid.²

In addition, because the domain pattern is elongated in shape, we estimate the total area as $A_t = P_t r$ such that $P_t = A_t / r$.

These assumptions applied to Equation (1) give equation (2):

$$\frac{dC_b}{dt} = \frac{DC_b v A_t}{A_{array} r^3} \quad (2)$$

Assuming mole fraction and area fraction are closely related then $\frac{dC_b}{dt}$ can be replaced by

$-\frac{dA_f}{dt}$ and by definition for the domain pattern area fraction, $\frac{A_t}{A_{array}} = A_f$, then Equation (2) can

be approximated by Equation (3):

$$\frac{dA_f}{dt} \propto -\frac{DC_b v A_f}{r^3} \quad (3)$$

Supporting Figures

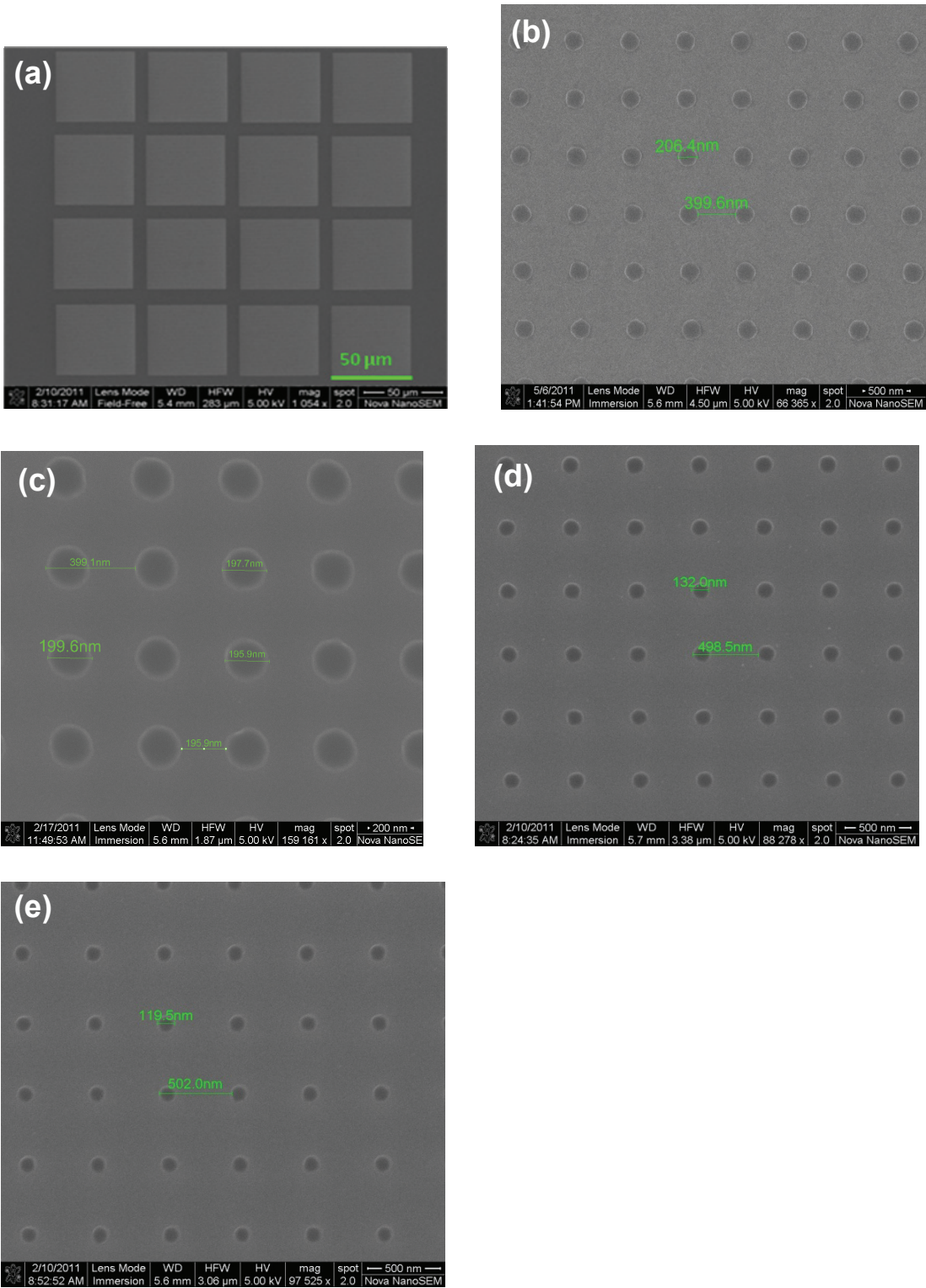


Figure S1. SEM images of e-beam patterned substrates. (a) Side-by-side arrangement of 16 lattice arrays each with spacing between the bumps of 375 nm and bump projected radius of 65 nm. (b-c) 100 nm projected radius bumps with spacing of (b) 400 nm and (c) 200 nm. (d-e) Spacing of 375 nm and projected radius of (d) 65 nm and (e) 60 nm

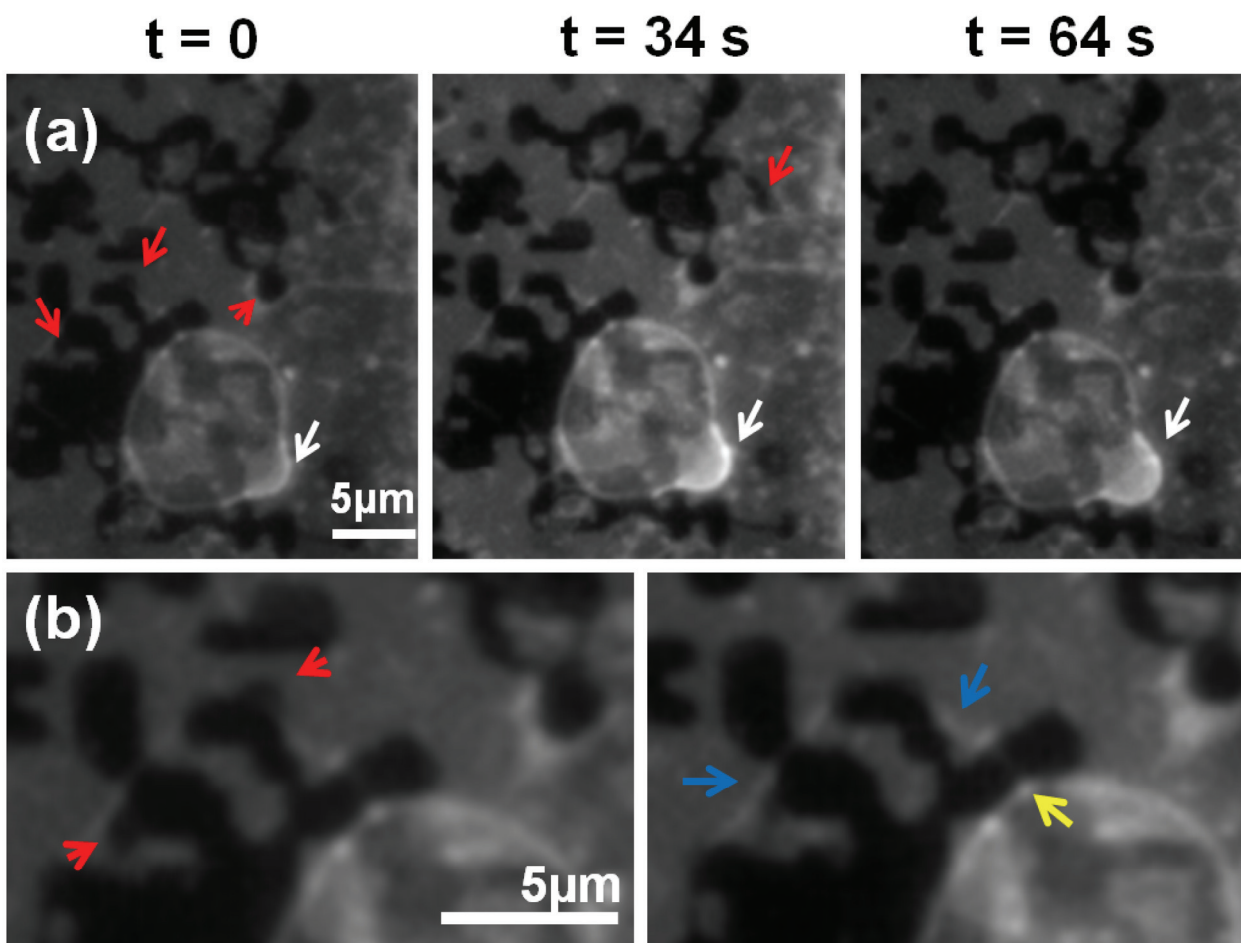


Figure S2. Giant vesicle attached to lipid tubule. (a) White arrows show giant vesicle growth while red arrows point to sections of the domain pattern that simultaneously disappear. (b) Close up – blue arrows pointing to a lipid tubule that appears to be taking up the domain lipids at the positions of the red arrows and is attached to the giant vesicle at the yellow arrow. Underlying lattice – 375 nm lattice spacing of 65 nm bumps.

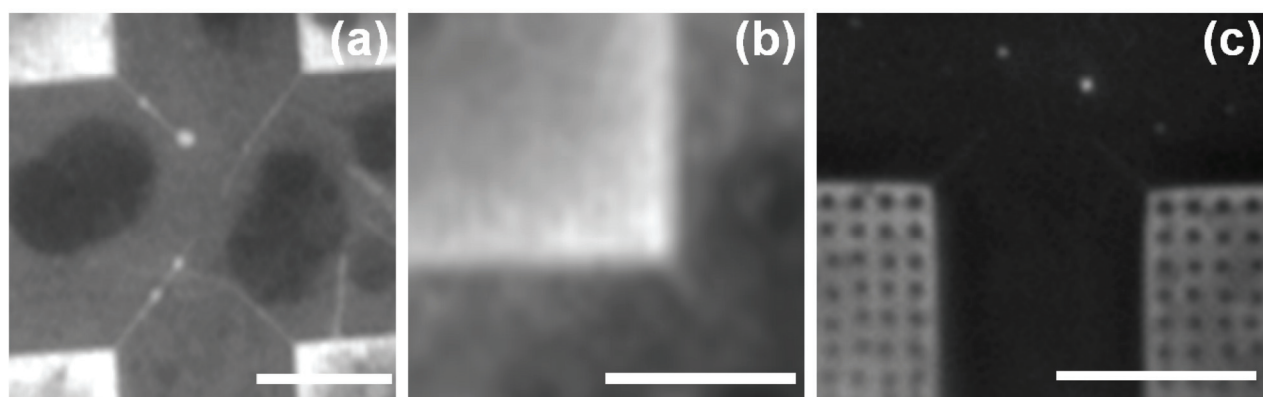


Figure S3. Lipid tubules at corners of arrays. (a) Where four arrays are spaced closely, the tubules radiate toward each other. (b) Shows that each tubule is shorter for the 200 nm lattice spacing. (c) Tubules radiate from the corners - lipid multilayer is only DOPC. Note, the dark appearance of the bumps is due to an interference contrast effect present with the thicker PMMA layer used for this array of 500 nm projected radius bumps. Scale bar a) and b) 5 μm , c) 10 μm .

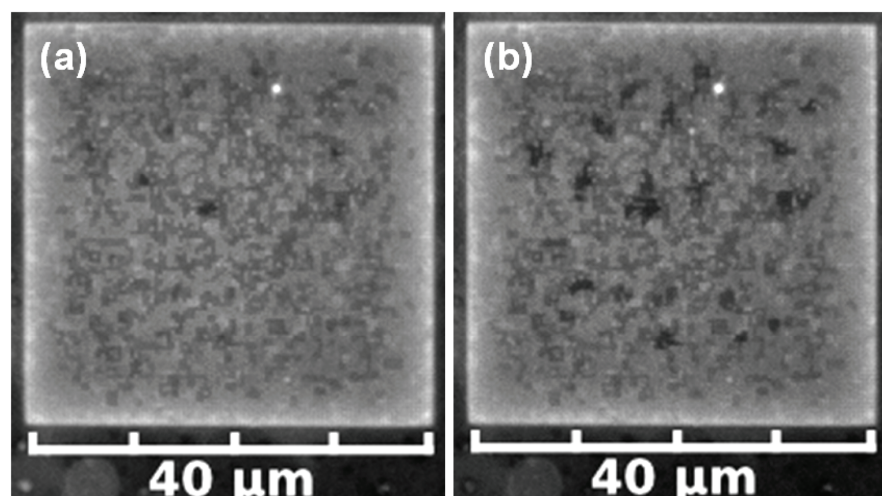


Figure S4. Overlay of Figure 2b (time = zero) with (a) Figure 7a (21.2 °C) and (b) Figure 7a (6.5 °C). The darkest portions of the pattern correspond to locations of exact pattern regeneration.

References

1. E.L Cussler, in Diffusion: Mass Transfer in Fluid Systems (Cambridge Series in Chemical Engineering), Cambridge University Press, Cambridge, 3rd edition, 1997.
2. J. H. Yao, K. R. Elder, H. Guo, and M. Grant, *Phys. Rev. B*, 1993, **47**, 14110-14125.