Surface Densities Prewet a Near Critical Membrane Supplementary Material

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19 Overview

In section 1 we provide details on the Landau Theory introduced in the main text. We first derive 20 the Landau coefficients used in our expression of the tether free energy, f_{tether} . Next we detail the 21 derivation of the surface free energy, f_{surf} expressed as a function of three 'surface' variables ϕ_0, ψ, ρ . 22 We then examine the behavior of f_{surf} near the membrane critical point, showing enhancement 23 of prewetting through the diverging effective surface enhancement and surface chemical potential 24 as criticality is approached. We then derive a relationship between membrane's susceptibility and 25 the enhancement of prewetting. In Section 2 we provide the supplemental figures referenced in the 26 main text. Here we discuss the effect of finite tether-length used in our simulations, properties 27 of our bulk lattice polymers system and alternative polymer mixtures, expand on the three-phase 28 coexistence region, and discuss membrane- and bulk-dominated surface densities 29

³⁰ 1 Landau Theory

Our mean-field theory starts with the Landau Functional shown in Equation 1 of the main text:

$$\mathcal{L} = \mathcal{L}_{2D} + \mathcal{L}_{3D}$$

$$\mathcal{L}_{2D} = \int d^2 \vec{x} f_{2D}(\psi, \rho, \phi_0)$$

$$\mathcal{L}_{3D} = \int d^2 \vec{x} dz \frac{1}{2} \nabla \phi^2 + f_{3D}(\phi)$$

$$f_{2D}(\psi, \rho, \phi_0) = \underbrace{\frac{t_{mem}}{2} \psi^2 + \frac{u_{mem}}{4!} \psi^4 - \lambda_{\psi} \psi}_{f_{membrane}}$$

$$-\underbrace{\frac{5}{6} \rho + \frac{3}{2} \rho^2 - \frac{3}{3!} \rho^3 + \frac{2}{4!} \rho^4 - \frac{1}{4} - \lambda_{\rho} \rho}_{f_{tether}}$$

$$-\underbrace{h_{\psi} \rho \psi - h_{\phi} \rho \phi_0}_{f_{int}}$$
(1)

where f_{3D} is the free energy of a system displaying a conventional condensation transition:

$$f_{3D} = \frac{t_{bulk}}{2}\phi^2 + \frac{u_{bulk}}{4!}\phi^4 - \mu_{bulk}\phi$$
(2)

³¹ Our goal is to minimize \mathcal{L} over order parameters ρ, ψ and order parameter profile $\phi(z)$. Two ³² derivatives $\frac{\partial \mathcal{L}}{\partial \psi} = \frac{\partial \mathcal{L}}{\partial \rho} = 0$ minimize the free energy over purely 'surface' terms. A functional ³³ derivative $\frac{\delta \mathcal{L}}{\delta \phi(z)} = 0$ minimizes the energy over forms of the density profile $\phi(z)$.

³⁴ 1.1 Derivation of f_{tether}

The free energy of the tethers in our model is expressed as a Landau free energy, a function of powers of the order parameter ρ and Landau coefficients a,b,c,d,e

$$f_{tether}(\rho) = a\rho + \frac{b}{2}\rho^2 + \frac{c}{3!}\rho^3 + \frac{d}{4!}\rho^4 + e - \lambda_{\rho}\rho$$

The tethers are a non-interacting 2-dimensional lattice gas. The free energy of a 2D gas may be written as $f_{tether} = \rho \log \rho$. We approximate this by expanding f_{tether} in powers of ρ at ρ_{\star}

$$\rho \log \rho|_{\rho_{\star}} = (\rho - \rho_{\star}) \left(1 + \log \rho_{\star}\right) + \frac{(\rho - \rho_{\star})^2}{2\rho_{\star}} - \frac{(\rho - \rho_{\star})^3}{6\rho_{\star}^2} + \frac{(\rho - \rho_{\star})^4}{12\rho_{\star}^3}$$
$$f_{tether}(\rho, \rho_{\star}) = \frac{-3\rho_{\star}^4 - 12\rho_{\star}^4 \log \rho_{\star}}{12\rho_{\star}^3} + \frac{\rho \left(-10\rho_{\star}^3 + 12\rho_{\star}^3 \log \rho_{\star}\right)}{12\rho_{\star}^3} + \frac{3\rho^2}{2\rho_{\star}} - \frac{\rho^3}{2\rho_{\star}^2} + \frac{\rho^4}{12\rho_{\star}^3} - \lambda_{\rho}\rho$$

Setting $\rho_{\star} = 1$, as used in all calculations here, we obtain the free energy of the tethers and values of all Landau coefficients:

$$f_{tether}(\rho, \rho_{\star} = 1) = -\frac{1}{4} - \frac{5}{6}\rho + \frac{3}{2}\rho^2 - \frac{3}{3!}\rho^3 + \frac{2}{4!}\rho^4 - \lambda_{\rho}\rho$$

³⁵ This Taylor series provides an analytically tractable approximation to the ideal gas free energy of

the tethers near $\rho = 1$. However, because its first derivative is finite at $\rho = 0$, some of our physical

phases spill over into negative values of ρ . As such we interpret ρ more as an order parameter than as a more rigorously defined tether density.

1.2Derivation of surface free-energy 39

Contributions from the bulk are a function of $\phi(z)$. To simplify calculations we express the bulk 40 energy as a function of initial and final values of ϕ , $\phi(z=0) = \phi_0$ and $\phi(z=\infty) = \phi_{\infty}$. these

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solely determine the behaviour of the density profile. We write $f_{3D}(\phi) - f_{3D}(\phi_{\infty})$ as $f_{3D}^{\phi - \phi_{\infty}}$. First we perform the functional derivative, $\frac{\delta \mathcal{L}_{bulk}}{\delta \phi(z)}$

$$\mathcal{L}_{bulk}(\phi(z)) = \int_0^\infty dz \frac{1}{2} (\nabla \phi)^2 + f_{3D}^{\phi - \phi_\infty}$$
$$\delta \mathcal{L}_{bulk}[\phi(z)] = \int_0^\infty dz \nabla \phi \nabla \delta \phi + \frac{\partial f_{3D}^{\phi - \phi_\infty}}{\partial \phi} \delta \phi$$
$$\frac{\delta \mathcal{L}_{bulk}[\phi(z)]}{\delta \phi(z)} = \frac{\partial f_{3D}^{\phi - \phi_\infty}}{\partial \phi} - \nabla^2 \phi = 0$$
(3)

The last equality serves as a second order ODE for $\phi(z)$ which we solve given boundary conditions at $z = 0, \infty$, making use of mathematical tools developed in [1].

$$\frac{\delta \mathcal{L}_{bulk}}{\delta \phi(z)} = 0$$

$$\frac{\partial^2 \phi}{\partial z^2} = \frac{\partial f_{3D}^{\phi - \phi_{\infty}}}{\partial \phi}$$

$$\frac{\partial \phi}{\partial z} \frac{\partial^2 \phi}{\partial z^2} = \frac{\partial \phi}{\partial z} \frac{\partial f_{3D}^{\phi - \phi_{\infty}}}{\partial \phi}$$

$$\frac{1}{2} \frac{\partial}{\partial z} \left(\frac{\partial \phi}{\partial z}\right)^2 = \frac{\partial}{\partial z} f_{3D}^{\phi - \phi_{\infty}}$$

$$\frac{1}{2} \int_{z=0}^{z=\infty} dz \frac{\partial}{\partial z} \left(\frac{\partial \phi}{\partial z}\right)^2 = \int_{z=0}^{z=\infty} dz f_{3D}^{\phi - \phi_{\infty}}$$

$$\frac{1}{2} \left(\left(\frac{\partial \phi}{\partial z}\right)_{z=\infty}^2 - \left(\frac{\partial \phi}{\partial z}\right)_{z=0}^2\right) = f_{3D}^{\phi - \phi_{\infty}}(\phi_{\infty}) - f_{3D}^{\phi - \phi_{\infty}}(\phi_{0})$$

$$\frac{1}{2} \left(\frac{\partial \phi}{\partial z}\right)_{z=0}^2 = f_{3D}^{\phi - \phi_{\infty}}(\phi_{0})$$

$$\frac{\partial \phi}{\partial z} = \pm \sqrt{2f_{3D}^{\phi - \phi_{\infty}}(\phi_{0})} \qquad (4)$$

Finally we substitute this into our original functional and rewrite as an integration over values

of ϕ instead of z to define the bulk contribution, Δf_{bulk}

$$\Delta f_{bulk}(\phi(z)) = \int_0^\infty dz \frac{1}{2} (\nabla \phi)^2 + f_{3D}^{\phi - \phi_\infty}$$

$$= \int_0^\infty dz (\nabla \phi)^2$$

$$= \int_{\phi_0}^{\phi_\infty} dz \frac{d\phi}{dz} \frac{dz}{d\phi} (\nabla \phi)^2$$

$$= \int_{\phi_0}^{\phi_\infty} d\phi \nabla \phi$$

$$\Delta f_{bulk}(\phi_0, \phi_\infty) = \int_{\phi_0}^{\phi_\infty} d\phi \sqrt{(2(f_{\phi_0} - f_{\phi_\infty}))}$$
(5)

The sign of $\frac{\partial \phi}{\partial z}$ depends on whether ϕ_0 is above or below ϕ_∞ : polymer density always moves towards the bulk density ϕ_∞ . An expression for the gradient may be obtained by substituting in the expression for $f_{3D}^{\phi-\phi_\infty}(\phi)$ and $u_{bulk} = \frac{1}{6} \left(6t_{bulk} \phi_\infty + \phi_\infty^3 \right)$:

$$\frac{\partial\phi}{\partial z} = (\phi - \phi_{\infty}) \frac{\sqrt{(12t_{bulk} + \phi^2 + 2\phi\phi_{\infty} + 3\phi_{\infty}^2)}}{2\sqrt{6}} \tag{6}$$

Writing Δf_{bulk} as a function of ϕ_0 and ϕ_∞ now allows minimization of the systems Landau free energy over values of ϕ_0, ρ , and ψ . After solutions are obtained we compute the energy of each of these solutions. This can be expressed as

$$f_{surf} = \Delta f_{bulk}(\phi_0, \phi_\infty) + f_{2D}(\phi_0, \psi, \rho)$$

$$= \int_{\phi_0}^{\phi_\infty} d\phi \sqrt{2(f_{\phi_0} - f_{\phi_\infty})} + f_{2D}$$

$$f_{surf} = \int_{\phi_0}^{\phi_\infty} d\phi \left\{ \nabla \phi - \frac{\partial f_{2D}}{\partial \phi} \right\} + \frac{\partial f_{2D}}{\partial \phi} |_{\phi_\infty}$$
(7)

where the final equation is the is the integration between the curves of $\nabla \phi$ and $-\frac{\partial f_{2D}}{\partial \phi_0}$ shown in Figure 5B of the main text.

45 1.3 Enhancement of prewetting regime near membrane critical point

In our Monte-Carlo simulations and numerical calculations, we find a large expansion of the prewetting region as the membrane is brought near to its critical point. We now aim to show using a simplified Landau theory how this enhancement arises. First we simplify the free energies of membrane and tether components, and obtain expressions for minimal values of ψ and ρ to quadratic order:

$$f_{2D} = \frac{t_{mem}}{2}\psi^2 - \lambda_{\psi}\psi - \frac{1}{4} - \frac{5}{6}\rho + \frac{3}{2}\rho^2 - \lambda_{\rho}\rho - h_{\psi}\psi\rho - h_{\phi}\phi_0\rho$$

$$\frac{\partial f_{2D}}{\partial \psi} = \frac{\partial f_{2D}}{\partial \rho} = 0$$

$$\psi_{min} = -\frac{-5h_{\psi} - 6(h_{\psi}\lambda_{\rho} - 3\lambda_{\psi} - h_{\phi}h_{\psi}\phi_{0})}{6(3t_{mem} - h_{\psi}^{2})}$$

$$\rho_{min} = -\frac{5t_{mem} - 6(t_{mem}\lambda_{\rho} + h_{\phi}\lambda_{\psi} + t_{mem}h_{\phi}\phi_{0})}{6(h_{\psi}^{2} - 3t_{mem})}$$
(8)

This simplification of f_{2D} excludes fourth order terms in ψ and ρ is only reasonable for $t_{mem} > t_{c,mem}$. Substituting ψ_{min}, ρ_{min} into the original f_{surf} we obtain the surface free energy in terms of ϕ_0 :

$$f_{surf}(\phi_0, \psi, \rho) = \Delta f_{bulk}(\phi_0, \phi_\infty) - \frac{t_{mem}h_{\phi}^2}{2(3t_{mem} - h_{\psi}^2)}\phi_0^2 - \frac{h_{\phi}\left(t_{mem}\left(5 + 6\lambda_{\rho}\right) + 6h_{\psi}\lambda_{\psi}\right)}{6(3t_{mem} - h_{\psi}^2)}\phi_0 - \frac{t_{mem}\left(79 + 60\lambda_{\rho} + 36\lambda_{\rho}^2\right) + 6\left(-3h_{\psi}^2 + 2h_{\psi}\left(5 + 6\lambda_{\rho}\right)\lambda_{\psi} + 18\lambda_{\psi}^2\right)}{72(3t_{mem} - h_{\psi}^2)}$$
(9)

Here second order couplings in ϕ_0 emerge after the minimizing over the membrane and tether degrees of freedom. Each of these terms has a denominator, $3t_{mem} - h_{\psi}^2$ which defines the membrane critical point as $t_{mem} = h_{\psi}^2/3$. The surface enhancement (prefactor to the ϕ_0^2 term) is proportional to $(J_{c,mem} - J_{mem})^{-1}$ which is consistent with the scaling of the Ising model susceptibility exponent $\gamma = 1$ in mean-field.

⁵⁶ 1.4 Membrane susceptibility and prewetting

The susceptibility-mediated enhancement of prewetting can be seen directly by examining higherorder interactions between membrane and tether components. We split \mathcal{L}_{2D} into non-interacting membrane and tether contributions and interaction terms mixing all components:

$$\mathcal{L}_{2D} = \int d^2 \vec{x} \{ f_{mem}(\psi) + f_{tether}(\rho) + f_{int}(\psi, \rho, \phi_0) \}$$
$$f_{int}(\psi, \rho, \phi_0) = -h_{\psi}\rho\psi - h_{\phi}\rho\phi_0$$
(10)

We focus on the interactions and define $\mathcal{L}_{int} = \int d^2 \vec{x} f_{int}(\psi, \rho, \phi_0)$. We calculate \mathcal{L}_{int} as a sum of cumulants:

$$\mathcal{L}_{int} \approx \mathcal{L}_{int}^{0} - \langle \mathcal{L}_{int} \rangle - \frac{1}{2} \left(\langle \mathcal{L}_{int}^{2} \rangle - \langle \mathcal{L}_{int} \rangle^{2} \right) - \dots$$

$$\langle \mathcal{L}_{int} \rangle = \int d^{2} \vec{x} h_{\psi} \psi \rho + h_{\phi} \phi_{0} \rho$$

$$\langle \mathcal{L}_{int}^{2} \rangle = \int d^{2} \vec{x} \rho (h_{\psi} \psi + h_{\phi} \phi_{0}) \int d^{2} \vec{x'} \rho' (h_{\psi} \psi' + h_{\phi} \phi_{0})$$

$$\langle \mathcal{L}_{int} \rangle^{2} = \left(\int d^{2} \vec{x} h_{\psi} \psi \rho + h_{\phi} \phi_{0} \rho \right)^{2}$$
(11)

We assume $t_{mem} > t_{c,mem}$ and the membrane is at it's critical composition, $\psi = 0$. We assume we are far from the prewetting critical point and neglect second order terms in ϕ_0

$$\langle \mathcal{L}_{int} \rangle = \langle \rho \rangle (h_{\phi} \langle \phi_0 \rangle + h_{\psi} \langle \psi \rangle)$$

$$\langle \mathcal{L}_{int}^2 \rangle - \langle \mathcal{L}_{int} \rangle^2 = h_{\psi}^2 \langle \rho \rangle^2 \int d^2 \vec{x} \psi_x \int d^2 \vec{x'} \psi_{x'} - \left(\int d^2 \vec{x} h_{\psi} \psi_x \rho_x \right)^2$$

$$= h_{\psi}^2 \langle \rho \rangle^2 \int d^2 \vec{x} \int d^2 \vec{x'} \langle \psi_x \psi_{x-x'} \rangle - h_{\psi}^2 \langle \rho \rangle^2 \langle \psi_x \rangle^2$$

$$= h_{\psi}^2 \langle \rho \rangle^2 \int d^2 \vec{x} \langle \psi_x^2 \rangle - \langle \psi_x \rangle^2$$

$$= h_{\psi}^2 \langle \rho \rangle^2 \int d^2 \vec{x} G(\vec{x})$$

$$= h_{\psi}^2 \langle \rho \rangle^2 \int dr G(r)$$

$$= h_{\psi}^2 \langle \rho \rangle^2 \chi_{\psi}$$
(12)

⁵⁷ Here χ_{ψ} is the susceptibility of the membrane and G(r) is the two-point correlation function. The ⁵⁸ integral over all space of G(r) is χ_{ψ} . Here we have assumed that the membrane order parameter is ⁵⁹ translationally invariant and that G(r) is spherically symmetric.

$$\mathcal{L}_{int} \approx \mathcal{L}_{int}^{0} - \langle \rho \rangle \left(h_{\phi} \langle \phi_{0} \rangle + h_{\psi} \langle \psi \rangle \right) - \frac{h_{\psi}^{2} \langle \rho \rangle^{2}}{2} \chi_{\psi}$$
(13)

At the critical composition, $\psi = 0$ and critical temperature, $\chi_{\psi} \to \infty$, implying that interactions become very favorable even with negligible contributions from other terms. Substituting \mathcal{L}_{int} into our original Landau functional, we obtain a new free energy function, neglecting fourth order terms:

$$f_{2D} = -\frac{5}{6}\rho + \frac{3}{2}\rho^2 - \frac{1}{4} - \lambda_\rho \rho - h_\phi \phi_0 \rho - \frac{h_\psi^2 \rho^2}{2} \chi_\psi$$
(14)

Minimizing over ψ and ρ and substituting back in the free energy, as in equation 9 above, we express f_{2d} just in terms of surface polymer density ϕ_0

$$f_{2D} = -\frac{h_{\phi}^2}{2(h_{\psi}^2\chi_{\psi} - 3)}\phi_0^2 + \frac{h_{\phi}(5 + 6\lambda_{\rho})}{6(h_{\psi}^2\chi_{\psi} - 3)}\phi_0 + \frac{-18h_{\psi}^2\chi_{\psi} + 36\lambda_{\rho}^2 + 60\lambda_{\rho} + 79}{72(h_{\psi}^2\chi_{\psi} - 3)}$$
(15)

This is similar to Equation 9, where the denominator vanishes at the 'shifted' membrane critical point $\chi_{\psi} = \frac{3}{h_{\psi}^2}$. This differs from the calculation above where the divergence near the critical point is stronger, as the Ising susceptibility exponent $\gamma = 7/4$ instead of 1. A full treatment of our model beyond mean-field would likely expand prewetting boundaries beyond the values we present here.

⁶⁴ 2 Supplementary Figures

65 2.1 Effect of tether length in monte-carlo simulations

Our Monte Carlo Simulations implement tethers with a finite three-dimensional length while the 66 tethers in our landau theory lack a three-dimensional component. We examined the effect of tether 67 length with simulations varying tether length from 1-10 mononmers S1. As expected, prevet 68 phases are stable at higher bulk temperatures as tether length is increased. The form of the 69 polymer density profile is influenced by tether length. The density profile has a maximum at short 70 but non-zero distances from the wall (see Main text Figure 2D, Figure 3C, S1). The location of 71 this maximum extends further from the wall as tether length is increased. This is in contrast to 72 conventional wetting and prewetting where the density profile is monotonic with distance from the 73 membrane. 74 The non-standard behavior of the density profile is explained by the entropic repulsion expe-

The non-standard behavior of the density profile is explained by the entropic repulsion expereferenced by polymers near a surface. Along the length of the tether, the membrane- and tethermediated interactions between polymers are the same. However there are more interactions available

⁷⁸ to polymers at finite distance from the surface. With a non-zero tether length, polymers prefer to

⁷⁹ concentrate their density away from the surface while still within the tether length.



Figure S1: Effect of Tether Length A) Density profiles of simulations at various tether lengths. Simulations with longer tethers have a broader maximum further from the membrane. B) Distance of maximum polymer Density plotted against tether length

80 2.2 Simulations of bulk polymer mixtures

81 Our 3D lattice polymer simulations phase-separate into coexisting dense and dilute phases in-

⁸² lieu of membrane and tethers. Here, as in the main text, red and blue polymers of equal length

⁸³ (n=20 monomer units) interact attractively with one another and like polymers repel each other.

Polymers phase-separate through increasing J_{bulk} (See Figure S2). Higher polymer concentrations

⁸⁵ phase-separate at lower J_{bulk} .



Figure S2: Monte-Carlo Simulations of Bulk Lattice Polymers A) Lattice polymer simulations show phase coexistence. Simulations at $0.25k_BT$ have a homogeneous polymer density.B Simulations at strong polymer coupling $1.0k_BT$, bottom, see coexisting dense and dilute phases.

⁸⁶ 2.3 Alternative bulk polymer mixtures

⁸⁷ We also ran simulations with different bulk polymer mixtures and observed similar phases to those ⁸⁸ in our symmetric mix of red and blue polymers (See Figure S3). We simulated a unary polymer ⁸⁹ mixture with strong nearest-neighbor interactions, an asymmetric mixture of polymers (n=5 and ⁹⁰ n=20), and a mixture of two short (n=5) polymers. We observe one,two, and three surface phase-⁹¹ coexistence in all of these mixtures. The classical theories of wetting and prewetting only require a ⁹² bulk systems that phase separates, and a surface with affinity for one of these phases. Diverse bulk ⁹³ systems ought display these phases, so long as they can phase-separate in the absence of a surface.



Figure S3: Several Bulk Polymer Mixtures Produce Similar Results. A) Single surfacephase system with a single polymer species (left), two polymers of unequal length (middle), and short polymers (right) B)Surface phase coexistence in mixtures as above C) Three surface phase coexistence

⁹⁴ 2.4 Two phase coexistence regions bound three-phase coexistence regions on ⁹⁵ the surface phase diagram

Our landau theory predicts three two-phase coexistence regions bounding the three-phase coex-96 istence region (main text Figure 7A). We examined our Landau theory in these regions, Figure 97 S4A. Shifting the surface composition eliminates one of the three stable solutions. After finding a 98 parameter regime in simulations demonstrating three-phase coexistence we shifted membrane and 99 tether compositions to access the two phase coexistence regions S4. Decreasing membrane order 100 at constant tether density eliminates the prewet dry phase (l_d Dry- l_o Prewet). Likewise increas-101 ing membrane order with a slight increase in tether density shows l_o -Dry l_o -Prewet Coexistence. 102 Finally, removing tethers eliminates the polymer rich phase, displaying a l_o -Dry l_d -Dry membrane. 103



Figure S4: **Two-phase coexistence near three-phase coexistence A**) Gradient construction within each two phase region depicted in Figure 7A. **B**) Two-phase coexistence regions observed in simulation

2.5 Dependence of three-phase coexistence region on bulk and membrane tem peratures

The three-phase coexistence region requires a phase-separated membrane and polymers that prevet 106 in the absence of membrane interactions. We calculated the phase diagram in our Landau theory 107 over membrane and bulk couplings in Figure S5A. Three-phase coexistence occurs at low membrane 108 and bulk temperatures, and the parameter regime shrinks as either temperature is increased. We 109 plotted the surface phase-diagram at several points within this phase diagrams in Figure S5B. 110 Decreasing J_{mem} shrinks the l_o Dry $-l_d$ -Dry coexistence region and respective edge of the three 111 phase region. Decreasing J_{bulk} shrinks the l_o Dry- l_o Prewet coexistence region. Outside of the 112 parameter regime where three-phases coexist, two-phases coexist. The character of these phases 113 depends on whether the dominant force is membrane or bulk. 114



Figure S5: **Bulk and Membrane Coupling govern 3-phase coexistence A**) Phase diagram over membrane and bulk couplings showing one, two, and three phase regions (yellow, blue, and red). Three phase coexistence generally requires strong membrane and bulk interactions, but extends to weaker J_{bulk} near the membrane critical point. **B**) Surface phase diagrams at various membrane and bulk couplings. Weak membrane couplings collapse the purple (l_0 -dry, l_d -dry) region, top to bottom. Weak bulk couplings shrink the green (l_o -prewet, l_o -dry) edge, left to right.

¹¹⁵ 2.6 Surface densities may be dominated by bulk or membrane forces

Near $J_{c,mem}$, two surface phases coexist at very low J_{bulk} ; at high J_{bulk} surface phases may also 116 coexist at low J_{mem} (main text Figure 6A). We explored our Landau theory in these two limits 117 to determine the character of these phases Figure S6. Order parameter profiles show that while 118 the bulk-dominated phase is similar to that of classical prevetting [1] the membrane dominated 119 phases only have marginal enrichment relative to the bulk equilibrium density ϕ_{∞} as with a classical 120 'surface transition' [2]. This is similar to Nakanishi and Fisher's description of wetting and surface 121 transitions [3], where surface transitions and prewetting are revealed to be the same transition 122 viewed from different regions of a higher-dimensional phase diagram. Biological systems likely 123 utilize these forces in a variety ways where membrane and bulk systems both have a role.



Figure S6: Bulk and Membrane Dominated Surface densities A) Density profiles for surface densities dominated by bulk (purple) and membrane (green) forces. B) Gradient construction for bulk and membrane dominated systems. A near-critical membrane allows surface coexistence when the gradient term is steep, but only over a narrow range of ϕ_0 values

¹²⁴ 2.7 Membrane composition and tether density alter surface polymer density

Our simulations show a strong dependence on membrane coupling and tether-bulk coupling. We explored the effect of surface composition, performing simulations varying tether density (see Figure S7A) and membrane composition (see Figure S7B) along with bulk coupling. We find that as tether density is increased, the density of polymers on the surface increases. As membrane order decreases

- towards the membrane critical point (membrane order = 0, or M =0), the density of surface
- 130 polymers similarly increases. As membrane order further decreases, polymer density increases as tethers relegated to smaller regions on the membrane, effectively increasing their density



Figure S7: Surface composition alters polymer density at the membrane: A) Varying tether density and bulk coupling increases surface polymer density. High densities, dark blue; low densities, light blue. B) Decreasing membrane order increases surface polymer density

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Figure	Tether Density	Membrane Order	$J_{bulk}, k_B T$	J_{mem}, T_c	J_{tether}	μ_{bulk}
S1A/B	0.25	0.8	0.4	1.1	1.0	-4.5
S2A, One Phase	N/A	N/A	0.25	N/A	N/A	0.06
S2A, Coexistence	N/A	N/A	1.0	N/A	N/A	0.06
S3A Single	0.094	0.5	0.55	2.0	1.0	-4.5
S3A, Unequal	0.0625	0.5	1.62	2.0	1.0	-3.0
S3A, Short	0.0625	0.5	3.31	2.0	1.0	-3.5
S3B, Single	0.094	0.5	0.55	1.0	1.0	-4.5
S3B, Unequal	0.0625	0.5	1.62	1.0	1.0	-3.0
S3B, Short	0.0625	0.5	3.31	1.05	1.0	-3.5
S3C, Single	0.094	0.5	0.55	0.9	1.0	-4.5
S3C, Unequal	0.0625	0.5	1.64	0.0	1.0	-3.0
S3C, Short	0.0625	0.5	3.31	0.9	1.0	-3.5
$S4B, l_o - Prewet/l_d Dry$	0.08	0.2	0.5	0.5	1.0	-4.5
S4B, $l_o - Dry/l_o - Prewet$	0.14	1	0.5	0.5	1.0	-4.5
S4B, $l_d - Dry/l_o - Dry$	0.02	0.5	0.5	0.5	1.0	-4.5

Table S1: Simulation parameters used in supplemental figures

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