Supplementary material for "Dynamic simulations of multicomponent lipid membranes over long length and time scales"

Supplement A. NUMERICAL DETAILS OF SIMULATION METHOD

Our overdamped model H for simulations of phase separation in a model membrane is given by

$$(\partial_t + \mathbf{v}\cdot\nabla)\phi(\mathbf{r},t) = M\nabla^2 \frac{\delta F}{\delta\phi(\mathbf{r},t)} + \theta(\mathbf{r},t) \tag{A1}$$

$$v_i(\mathbf{r},t) \;=\; \int d^2r'\, T_{ij}(\mathbf{r}-\mathbf{r}') \left[\frac{\delta F}{\delta\phi(\mathbf{r}',t)}\nabla'_j\phi(\mathbf{r}',t) + \zeta_j(\mathbf{r}',t)\right]. \tag{A2}$$

where the continuum Fourier transform of T_{ij} is [1, 2]

$$T_{ij}(\mathbf{q}) = \int d^2r\, T_{ij}(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}} = \frac{1}{\eta_m(q^2+q/L_{sd})}\left(\delta_{ij} - \frac{q_i q_j}{q^2}\right) \tag{A3}$$

where the integral is over all space.

We follow Koga and Kawasaki [3] in using the fast Fourier transforms (FFT) as an efficient way to numerically evolve these dynamics. We modify their approach by using the Oseen tensor appropriate for the quasi-2D hydrodynamic environment as well as stochastic thermal forces. An $\mathcal{L}\times\mathcal{L}$ periodic geometry is assumed; the dynamics of the Fourier modes $\phi_{\mathbf{q}} = \int_0^{\mathcal{L}}\int_0^{\mathcal{L}} d^2r\,\phi(\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}}$ follow from Eq. A2:

$$\partial_t\phi_{\mathbf{q}}(t) + \{\mathbf{v}\cdot\nabla\phi(\mathbf{r},t)\}_{\mathbf{q}} = -Mq^2\left\{\frac{\delta F}{\delta\phi(\mathbf{r},t)}\right\}_{\mathbf{q}} + \theta_{\mathbf{q}} \tag{A4}$$

$$v_{\mathbf{q},i}(t) = T_{ij}(\mathbf{q})\left\{\frac{\delta F}{\delta\phi(\mathbf{r},t)}\nabla_j\phi(\mathbf{r},t) + \zeta_j\right\}_{\mathbf{q}} \tag{A5}$$

$$\langle\theta_{\mathbf{q}}(t)\theta^*_{\mathbf{q}'}(t')\rangle = 2k_BTMq^2\mathcal{L}^2\delta_{\mathbf{q},\mathbf{q}'}\delta(t-t') \tag{A6}$$

$$\langle\zeta_{\mathbf{q},i}(t)\zeta^*_{\mathbf{q}',j}(t')\rangle = 2k_BT\mathcal{L}^2\eta_m(q^2+q/L_{sd})\delta_{ij}\delta_{\mathbf{q},\mathbf{q}'}\delta(t-t'). \tag{A7}$$

where $\{f(\mathbf{r})\}_{\mathbf{q}}$ is the Fourier transform of $f(\mathbf{r})$ and * indicates complex conjugation. The variance of the Langevin forces θ and ζ are set by the fluctuation-dissipation theorem [4, 5].

These equations are solved numerically by truncating to $N\times N$ Fourier modes $\mathbf{q} = (m,n)2\pi/\mathcal{L}$ with $-N/2 < m,n \leq N/2$ (corresponding to a real space discretization size $\ell = \mathcal{L}/N$). $\{\ldots\}_{\mathbf{q}}$ terms are evaluated in a hybrid real-space / Fourier-space fashion, handling real-space derivatives and convolutions in $\mathbf{q}$-space, local real-space operations in $\mathbf{r}$-space and moving between the two representations via the FFT.

Though we have written Eqs. A1-A2 as two separate equations, they only represent one dynamical equation, as the velocity field is set by the composition by Eq. A2. Substituting Eq. A2 into Eq. A1 yields a single Langevin equation for ϕ, but the coefficient of the thermal noise ζ depends on ϕ through the $\nabla\phi$ term of Eq. A1. This so-called "multiplicative noise" [4] should be treated via the Stratonovich interpretation, as it approximates a thermal force with a finite correlation time (set by the neglected fluid inertia) [4, 6]. We use a semi-implicit Stratonovich integrator, which requires the nonconstant coefficient of the Langevin force to be averaged over its value at ϕ and an auxiliary value $\tilde\phi$ [7]; the linear (but potentially most unstable) q^4 term is treated implicitly, and the nonlinear parts explicitly, as in semi-implicit solvers for the Cahn-Hilliard equation [8]. Our scheme is:

$$\phi_{\mathbf{q}}(t+\Delta t) = \phi_{\mathbf{q}}(t) - \frac{1}{1+M\gamma q^4\Delta t}\left(\Delta t\left\{\mathbf{v}^{\text{det}}\cdot\nabla\phi(t) + \mathbf{v}^{\text{therm}}\cdot(\nabla\phi(t)+\nabla\tilde\phi(t))/2 - M\nabla^2(r\phi(t)-u\phi(t)^3)\right\}_{\mathbf{q}} - \Theta_{\mathbf{q}}\right) \tag{A8}$$

$$v^{\text{det}}_{\mathbf{q},i}(t) = T_{ij}(\mathbf{q})\left\{\frac{\delta F}{\delta\phi}\nabla_j\phi\right\}_{\mathbf{q}} \tag{A9}$$

$$v^{\text{therm}}_{\mathbf{q},i}(t) = T_{ij}(\mathbf{q})\left(\frac{1}{\Delta t}Z_{\mathbf{q},j}\right) \tag{A10}$$

$$\tilde\phi_{\mathbf{q}}(t) = \phi_{\mathbf{q}}(t) + \Theta_{\mathbf{q}} - \Delta t\left\{\mathbf{v}^{\text{therm}}\cdot\nabla\phi(t)\right\}_{\mathbf{q}} \tag{A11}$$

$$\langle\Theta_{\mathbf{q}}\Theta^*_{\mathbf{q}}\rangle = 2k_BTMq^2\mathcal{L}^2\Delta t \tag{A12}$$

$$\langle Z_{\mathbf{q},i}Z^*_{\mathbf{q},i}\rangle = 2k_BT\mathcal{L}^2\eta_m(q^2+q/L_{sd})\Delta t \tag{A13}$$

Since $\phi(\mathbf{r})$ is a real field, we know that not all modes $\phi_{\mathbf{q}}$ are independent: $\phi^*_{\mathbf{q}} = \phi_{-\mathbf{q}}$. This means that (assuming N is even), the modes $(m,n) = (0,0), (N/2,0), (0,N/2)$, and $(N/2,N/2)$ are guaranteed to be real. We choose these to be four of the required N^2 dynamical variables. The other independent modes are chosen to be (m,n) for $-N/2 < m < N/2$ and $0 < n < N/2$, $(m,0)$ for $0 < m < N/2$, $(m,N/2)$ for $0 < m < N/2$, and $(N/2,n)$ for $0 < n < N/2$, as in [9]. The real and imaginary parts of each of these modes are both independent dynamic variables. The remaining modes are determined by the complex conjugates of the evolved modes. We also note that because the dynamics conserves total concentration, the mode $(0,0)$ must remain constant, and is not evolved.

The random thermal forces $\theta(\mathbf{r},t)$ and $\zeta_j(\mathbf{r},t)$ are also required to be real, which affects their Fourier transforms, and therefore the variance of the integrals $\Theta_{\mathbf{q}}(\Delta t)$ and $Z_{\mathbf{q},j}(\Delta t)$. We know from the variance of $\theta_{\mathbf{q}}(t)$ (in the main paper) that $\langle \Theta_{\mathbf{q}}(\Delta t)\Theta^*_{\mathbf{q}}(\Delta t)\rangle = 2TMq^2\mathcal{L}^2\Delta t$. If we write $\Theta_{\mathbf{q}} = f + ig$, we see that for the explicitly real modes $(m,n) = (0,0)$, $(N/2,0)$, $(0,N/2)$, and $(N/2,N/2)$ where $g = 0$, $\langle |f|^2\rangle = 2TMq^2\mathcal{L}^2\Delta t$, but for complex modes, f and g are selected from a distribution with variance $\langle |f|^2\rangle = \langle |g|^2\rangle = TMq^2\mathcal{L}^2\Delta t$. The variance of $Z_{\mathbf{q},i}$ is exactly analogous, but with $\langle Z_{\mathbf{q},i} Z_{\mathbf{q},j}\rangle = 2T\mathcal{L}^2\eta_m(q^2 + q/L_{sd})\delta_{ij}\Delta t$.

The method of Eqs. A8-A13 allows stable time steps to be chosen that are nearly two orders of magnitude larger than those allowed by a simple explicit scheme, comparable to the gains seen in [8].

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