Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information for

Systematic Analysis of Curvature-Dependent Lipid Dynamics in a Stochastic 3D Membrane Model

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Content:

1) Supplementary Information (for Appendix)

Define $p(a,b)$ with $a,b \in \{1,-1\}$ as the probability that two randomly selected spins have states a and b. Normalization yields $p(1,1)+2p(1,-1)+p(-1,-1)=1$

Calculation of $<S_iS_j>$ for 1D Ising model

$$<S_iS_j> \ = p(1,1) - 2p(1,-1) + p(-1,-1) = 1 \ - 4\,p(1,-1) = 1 \ - 4\frac{N_A}{N}\frac{N_B}{N-1} = 1 - \frac{N}{(N-1)}(1-M^2)$$

Note that the factor N-1 takes these correlations into account.

This can be rewritten as

$$<S_iS_j> \ = 1 - \frac{1}{\left(1-\frac{1}{N}\right)}(1-M^2) \approx 1 - \left(1+\frac{1}{N}\right)(1-M^2) = \ = M^2\left(1+\frac{1}{N}\right) - \frac{1}{N}$$

The correlation is reflected by the 1/N-terms.

Calculation of $<S_iS_jS_kS_l>$ **for 1D Ising model**

$$<S_iS_jS_kS_l> \ = p(1,1,1,1) - 4p(1,1,1,-1) + 6p(1,1,-1,-1) - 4p(1,1,-1,-1) + p(-1,-1,-1,-1)$$

$$= 1 - 8p(1,1,1,-1) - 8p(1,-1,-1,-1)$$

$$= \ 1 - \frac{8}{N(N-1)(N-2)(N-3)}N_AN_B[(N_A-1)(N_A-2) + (N_B-1)(N_B-2)]$$

$$= 1 - \frac{2(1-M^2)N}{(N-1)(N-2)(N-3)}\left[\frac{N(1+M)-2}{2}\,\frac{N(1+M)-4}{2} + \frac{N(1-M)-2}{2}\,\frac{N(1-M)-4}{2}\right]$$

$$= 1 - \frac{(1-M^2)N}{(N-1)(N-2)(N-3)}[N^2M^2 + (N-2)(N-4)]$$

$$= M^4\frac{N^3}{(N-1)(N-2)(N-3)} - M^2\frac{N^3 - N(N-2)(N-4)}{(N-1)(N-2)(N-3)} + 1 - \frac{N(N-4)}{(N-1)(N-3)}$$

$$\approx M^4\frac{N^3}{N^3-6N^2} - M^2\frac{6N^2-8N}{N^3-6N^2} \approx M^4\left(1+\frac{6}{N}\right) - \frac{6M^2}{N} + O\left(\frac{1}{N^2}\right)$$

Calculation of $<E^2>_M$ **for 1D Ising model**

$$<E^2>_M = J^2\left[N + 2N<S_iS_k>_M \ + (N^2-3N)\right]<S_iS_jS_kS_l>_M \,]$$

$$\approx [N + \ 2NM^2 + (N^2-3N)[M^4\left(1+\frac{6}{N}\right) - \frac{6M^2}{N}]]$$

$$\approx M^4(N^2+3N) - 4M^2N + N$$

Calculation of $<E^2>_M - <E>_M^2$ **for 1D Ising model**

$$< E^2 >_M - < E >_M^2 = J^2 \ [(N^2+3N)M^4 - 4M^2N + N \ - (N+1)^2M^4 + 2M^2(N+1) - 1]$$

$$\approx J^2 \ [NM^4 - 2M^2N + N]$$

when all extensive terms are taken into account

Calculation of $< E >_M$ for Q neighbors

$$< E >_M = -JN\binom{Q}{2} < S_iS_j >_M = -JN\binom{Q}{2}\left[M^2\left(1+\frac{1}{N}\right) - \frac{1}{N}\right] = -J\binom{Q}{2}[M^2(N+1) - 1]$$

$$\Rightarrow < E >_M^2 = J^2\binom{Q}{2}^2[N^2M^4 + 2NM^4 - 2NM^2] + O(N^0)$$

Calculation of $< E >_M - < E >_M^2$ for Q neighbors

$$< E^2 >_M = \left[\frac{NQ}{2} < S_1^2S_2^2 > \ + NQ(Q-1) < S_1S_2^2S_3 > + \left[\left(\frac{NQ}{2}\right)^2 - NQ\left(Q - \frac{1}{2}\right)\right] < S_1S_2S_3S_4 > \right]$$

$$= \left[\frac{NQ}{2} + NQ(Q-1)\left[M^2\left(1+\frac{1}{N}\right) - \frac{1}{N}\right] + \left[\left(\frac{NQ}{2}\right)^2 - NQ\left(Q-\frac{1}{2}\right)\right]\left[M^4\left(1+\frac{6}{N}\right) - \frac{6M^2}{N}\right]\right]$$

$$\Rightarrow < E^2 >_M - < E >_M^2 = NM^2\left[Q(Q-1) - \frac{3Q^2}{2} + 2\binom{Q}{2}^2\right] + NM^4\left[\frac{NQ^2}{4} - Q^2 + \frac{Q}{2} + \frac{6Q^2}{4} - \frac{NQ^2}{4} - \frac{Q^2}{2}\right]$$

$$= -NM^2Q + NM^4\frac{Q}{2}$$

2) Supplementary Figure Legends

Figure S1: Supporting data for the numerical model. (A) Hexagonal packing of lipids with SP = 1 yields a planar membrane. **(B)** Hexagonal packing of lipids with SP = 0.8 yields a positively curved membrane. **(C)** Graphical representation of the model. **(D)** Calculation of surface curvature. From left to right, a 3D surface, a polyhedral approximation of the surface, and the calculation of the surface curvature are shown.

Figure S2: Supporting data for curvature-dependence of lipid enrichment. (A) Temporal evolution of Lipid 'A' concentration on wedge-shaped rigid surface for $\kappa = 27\ k_BT$. **(B)** Relative enrichment of lipid 'A' on wedge-shaped rigid surface using ($\kappa = 27\ k_BT; K_v = 0\ k_BT$). Note a weak accumulation of lipid 'A' on the wedges and a slight decrease of accumulation upon temperature increase. **(C)** Lipid aggregates do not show any curvature dependency for ($\kappa = 0\ k_BT; K_v = 2\ k_BT$). With increasing temperature, the lipid clusters diffuse. **(D)** Simulation of freely fluctuating membranes for a vesicle with a diameter of 300 nm composed of two lipid species with SP>1 and SP<1, respectively. From top to bottom, simulations of ($\kappa = 27\ k_BT$), and ($\kappa = 0\ k_BT$) are shown. For each condition, the relative concentration of lipid 'A' (left, top), local membrane curvature (left, bottom), and the correlation of these two parameters (right) are depicted. See also **Movie S1**.

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3) Supplementary Movie Legends

Movie S1: Temporal evolution of curvature-dependent lipid enrichment, as shown in Fig. S2D. Temporal evolution of Lipid 'A' concentration on fluctuating membrane composed of two lipid species with SP<1 and SP>1 at a molar ratio of 1:1 using default parameter setting ($\kappa = 27\ k_BT; K_v = 2\ k_BT; T = 300\ K$).

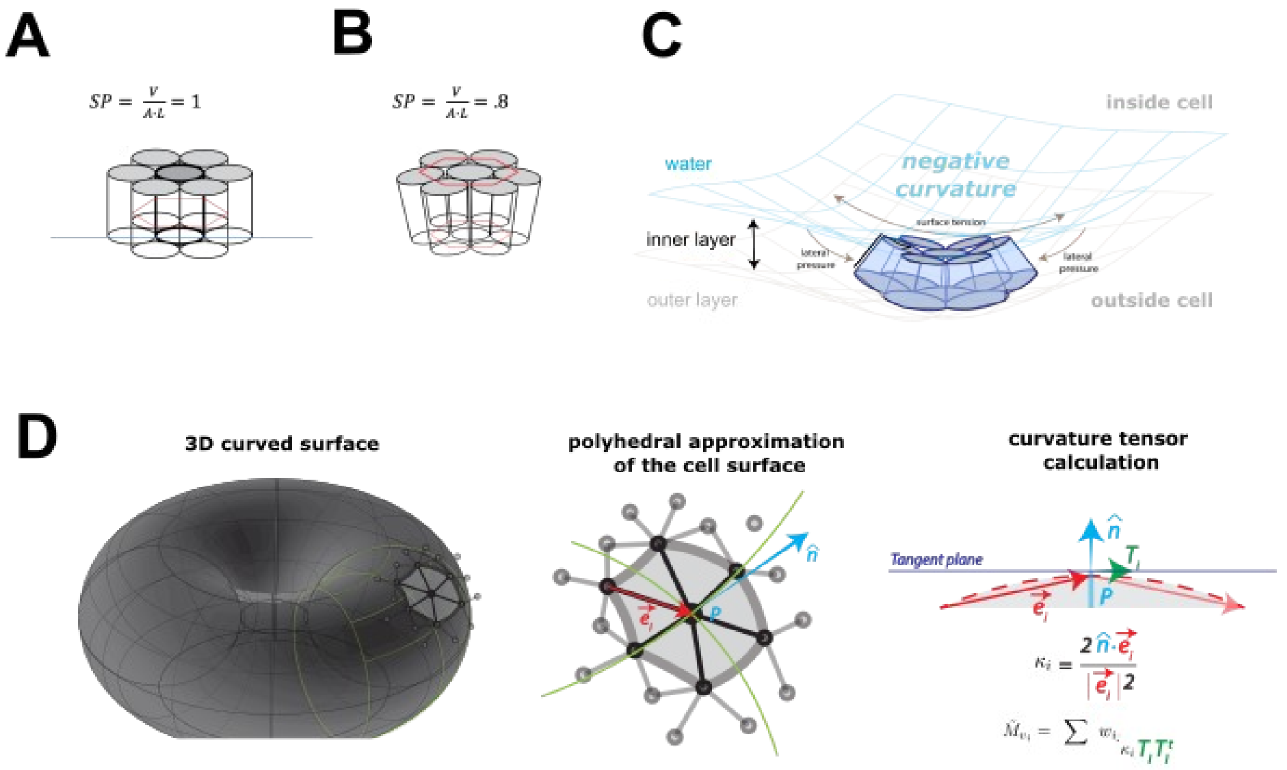

Fig. S1

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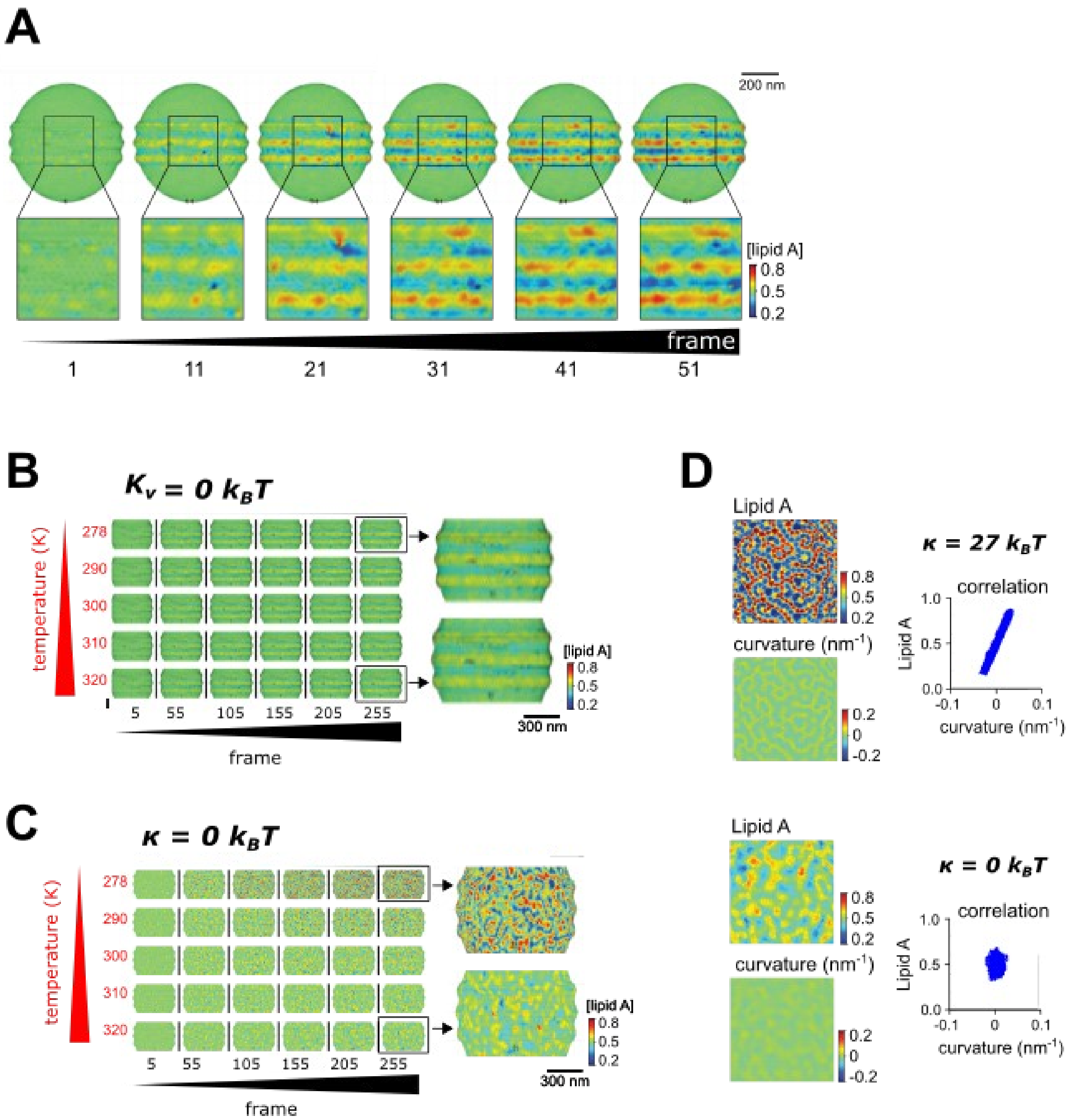

Fig. S2

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