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Electronic supporting information (ESI)

Planar Confined Water Organisation in Lipid Bilayer Stacks of Phosphatidylcholine and Phosphatidylethanolamine

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Content

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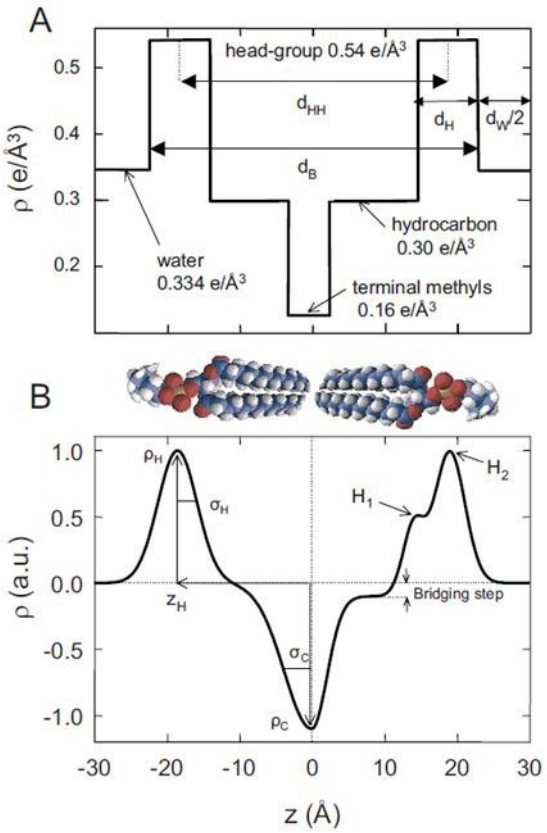

Fig. S1 Common bilayer electron density profile (EDP) models. (A) A strip model and (B) two different Gaussian component models. The figure has been taken with permission from reference [1].

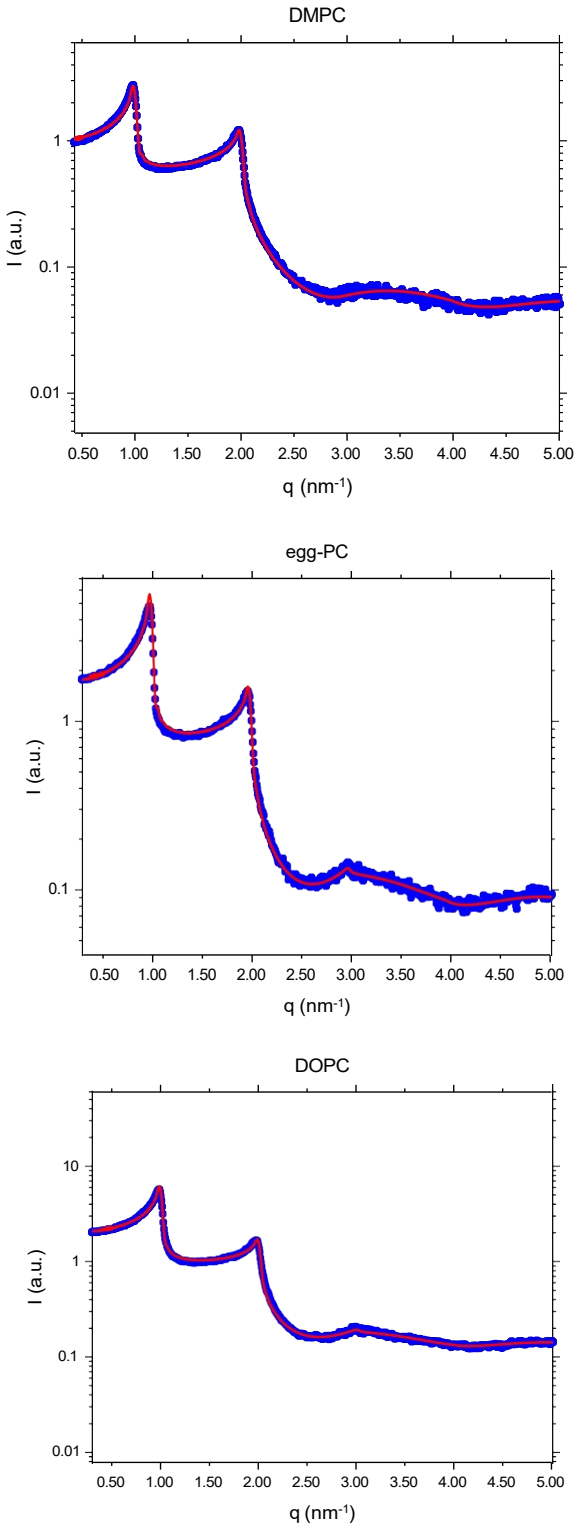

Fig. S2 Global fitting of SAXS data of DMPC, egg-PC and DOPC at 30 °C (shown from top to bottom). Solid red lines give best fits.

Table S1: Lattice spacing and form factor values of DMPE.

		Fourier Coefficients F(h)/F(1)			
Temperature (°C)	**d-spacing (nm)**	**F(1)/F(1)**	**F(2)/F(1)**	**F(3)/F(3)**	**F(4)/F(1)**
57	4.935	-1.000	-0.227	+0.220	-0.276
60	4.902	-1.000	-0.231	+0.217	-0.268
62	4.87	-1.000	-0.244	+0.222	-0.259
65	4.839	-1.000	-0.243	+0.223	-0.255
67	4.815	-1.000	-0.234	+0.223	-0.262
70	4.785	-1.000	-0.240	+0.221	-0.261
72	4.765	-1.000	-0.237	+0.221	-0.254
75	4.736	-1.000	-0.232	+0.211	-0.230
80	4.69	-1.000	-0.240	+0.211	-0.248

Table S2: Structural parameters of DMPC, egg-PC and DOPC (30 °C) and DMPE (60 °C). Parameters are compared to literature values given in brackets.

Parameter	**DMPC (30 °C)**	**DOPC (30 °C)**	**Egg-PC (30 °C)**	**DMPE (60 °C)**
***d*-spacing (nm)**	6.27 (6.27 [2])	6.27 (6.31 [2])	6.35 (6.63 [2])	4.90 (4.83 [3])
d_{HH} **(nm)** (head to head)	3.44 (3.60 [2])	3.63 (3.69 [2])	3.66 (3.69 [2])	3.61 (3.39 [3])
d_{LZ} **(nm)** (bilayer thickness by Luzzati)	3.49 (3.69 [2])	3.53 (3.59 [2])	3.56 (3.63 [2])	3.49 (3.41 [3])
A_L **(nm^2)**	0.619 (0.596 [2])	0.723 (0.725 [2])	0.694 (0.694 [2])	0.582 (0.596 [3])
n_w **total** (per lipid)	24.1 (25.6 [2])	33.0 (32.8 [2])	31.8 (34.7 [2])	12.2 (11 [4])

Derivation of the parameter D_{H2}

D_{H2} can be derived from setting the **water volume in the headgroup regions** equal, using the excluded headgroup volume expression and Luzzati ansatz[5], respectively:

$$A_L = \frac{2V_L}{d_{Lz}} = \frac{2V_L}{d_{HH}+2(D_{H2}- D_{H1})} \quad (1)$$

$$D_H A_L - V_H = A_L(D_H - D_{H2}) \quad (2)$$

Inserting (1) in (2):

$$\Rightarrow \frac{2D_H V_L}{d_{HH}+2(D_{H2}- D_{H1})} - V_H = \frac{2V_L(D_H- D_{H2})}{d_{HH}+2(D_{H2}- D_{H1})}$$

$$\Rightarrow V = \frac{D_{H2}2V_L}{d_{HH}+2(D_{H2}- D_{H1})}$$

$$\Rightarrow V_H d_{HH} + 2V_H D_{H2} - 2V_H D_{H1} = 2D_{H2}V_L$$

$$\Rightarrow V_H(d_{HH} - 2D_{H1}) = 2D_{H2}(V_L - V_H)$$

$$\Rightarrow \boldsymbol{D_{H2} = \frac{V_H(d_{HH}- 2D_{H1})}{2(V_L-V_H)}}$$

Alternatively, one may set the **area per lipid expressions** equal that derive from McIntosh[6] and Luzzati[5] ideas, respectively:

$$A_L = \frac{2(V_L - V_H)}{d_{HH} - 2D_{H1}} = \frac{2V_L}{d_{HH} + 2(D_{H2} - D_{H1})}$$

$$\Rightarrow 2V_L d_{HH} + 4V_L D_{H2} - 4V_L D_{H1} - 2V_H d_{HH} - 4V_H D_{H2} + 4V_H D_{H1} = 2V_L d_{HH} - 4V_L D_{H1}$$

$$\Rightarrow 4V_L D_{H2} - 4V_H D_{H2} = 2V_H d_{HH} - 4V_H D_{H1}$$

$$\Rightarrow 2D_{H2}(V_L - V_H) = V_H(d_{HH} - 2D_{H1})$$

$$\Rightarrow \boldsymbol{D_{H2} = \frac{V_H(d_{HH}- 2D_{H1})}{2(V_L-V_H)}}$$

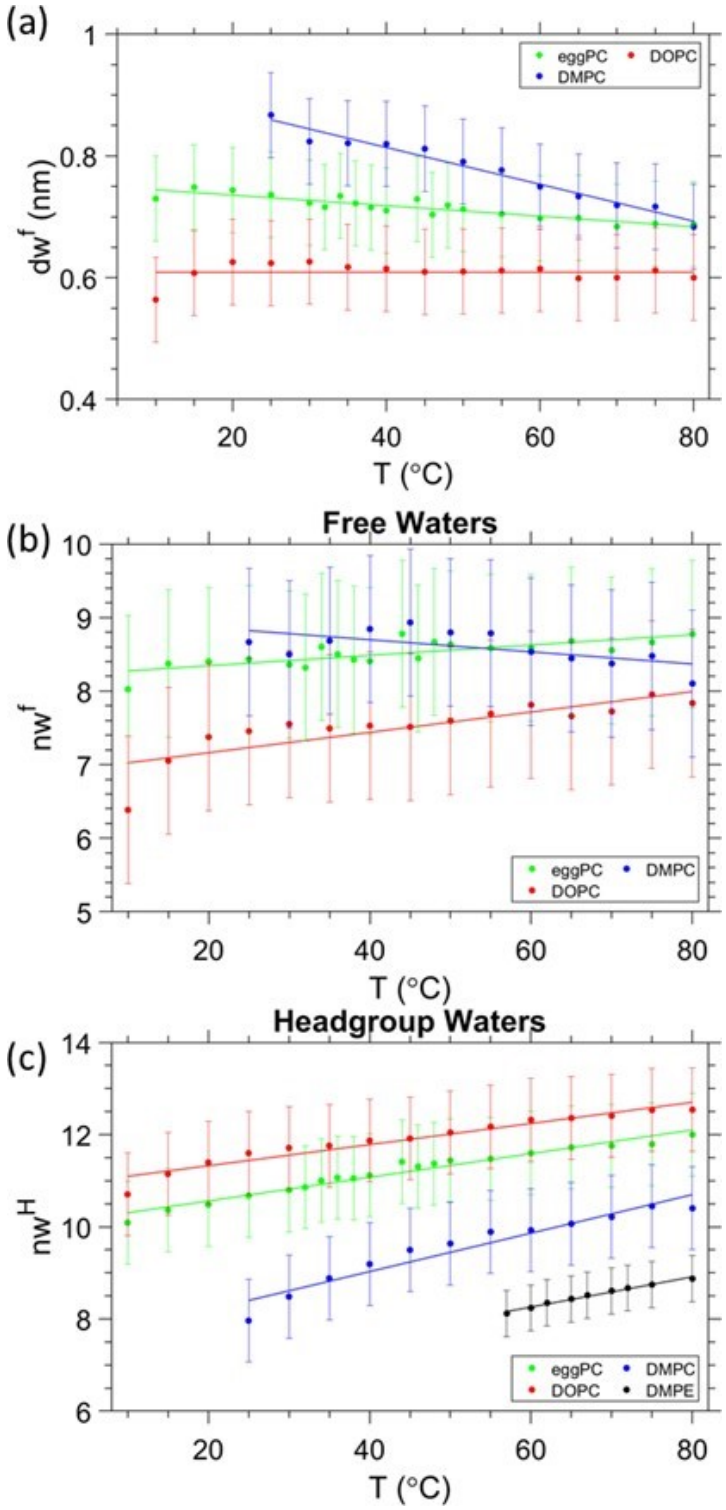

Fig. S3 Refined membrane hydration. a) The refined free water layer thickness for the PCs is presented, using the linear regression of $D_H\,(T)$ obtained from DOPC and DMPE data, respectively. b) Refined number of waters in the headgroup region, and c) the refined number of water molecules in the free water layer are shown. Note, the perturbed water layer remains unchanged, when applying $D_H\,(T)$ instead of D_H = constant.

References

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