Supporting Information:
Thermodynamics of charged nanoparticles adsorption on charge-neutral membranes: a simulation study

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1 SI_1: Force field parameters of the coarse-grained model[1]

The coarse-grained model includes four main types of interaction sites: polar (P), nonpolar (N), apolar (C), and charged (Q). For particles of type N and Q four subtypes are further distinguished to allow fine tuning of the Lennard-Jones (LJ) interactions, reflecting hydrogen bonding capabilities: 0 for no hydrogen bonding, d for hydrogen bonding donor, a for hydrogen bonding acceptor, and da for hydrogen bonding donor and acceptor. Nonbonded interactions between the CG particles are presented by the standard Lennard-Jones potential. For all particles the same effective LJ particle size of 0.47 nm is used. There are five levels of LJ interaction defined by the force field, ranging from attractive (I, $\epsilon = 5\,\text{kJ/mol}$), semi-attractive (II, $\epsilon = 4.2\,\text{kJ/mol}$), intermediate (III, $\epsilon = 3.4\,\text{kJ/mol}$), semi-repulsive(IV, $\epsilon = 2.6\,\text{kJ/mol}$), and repulsive (V, $\epsilon = 1.8\,\text{kJ/mol}$), details as Table 1.

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And potential energy functions of the coarse-grained force field can be subdivided into two parts: the non-bonded interactions including Lennard-Jones potential and Coulombic potential and the bonded interactions including bonded stretching and angle bending.

Lennard-Jones potential:

$$U_{\text{LJ}}(r) = 4\epsilon_{\text{ij}}[(\frac{\sigma_{\text{ij}}}{r})^{12} - (\frac{\sigma_{\text{ij}}}{r})^{6}]$$

with ϵ_{ij} representing the effective minimum distance of approach between two particles and σ_{ij} the strength of their interaction. The value of ϵ_{ij} is summarized in Table 1 and $\sigma = 0.47\,\text{nm}$ in the simulation.

Coulombic potential:

$$U_{\text{el}}(r) = \frac{q_i q_j}{4\pi\epsilon_0\epsilon_{\text{r}}r}$$

with relative dielectric constant $\epsilon_{\text{r}} = 20$ for explicit screening.

Bond stretching interactions are described by a weak harmonic potential:

$$V_{\text{bond}}(R) = \frac{1}{2}K_{\text{bond}}(R - R_{\text{bond}})^2$$

The force constant of the harmonic bonding potential is $K_{\text{bond}} = 1250\,\text{kJ}\,\text{mol}^{-1}\,\text{nm}^{-2}$ and the equilibrium distance $R_{\text{bond}} = 0.47\,\text{nm}$.

Angle bending interactions are also described by a weak harmonic potential of the cosine type:

$$V_{\text{angle}}(\theta) = \frac{1}{2}(\cos(\theta) - \cos(\theta_0))^2$$

The basic equilibrium bond angle $\theta_0 = 180°$, with a force constant of $K_{\text{angle}} = 25\,\text{kJ}\,\text{mol}^{-1}\,\text{rad}^{-2}$.

Table 1: Interaction Matrix

group	subtype	P	N				C	Q			
			0	d	a	da		0	d	a	da
P		I	IV	III	III	II	V	I	I	I	I
N	0	IV	III	III	III	III	III	III	III	III	III
	d	III	III	II	II	II	IV	III	III	II	II
	a	III	III	II	II	II	IV	III	II	III	II
	da	II	III	II	II	I	V	III	II	II	I
C		V	II	IV	IV	V	III	V	V	V	V
Q	0	I	III	III	III	III	V	III	III	III	II
	d	I	III	III	II	II	V	III	III	II	I
	a	I	III	II	III	II	V	III	II	III	I
	da	I	III	II	II	I	V	II	I	I	I

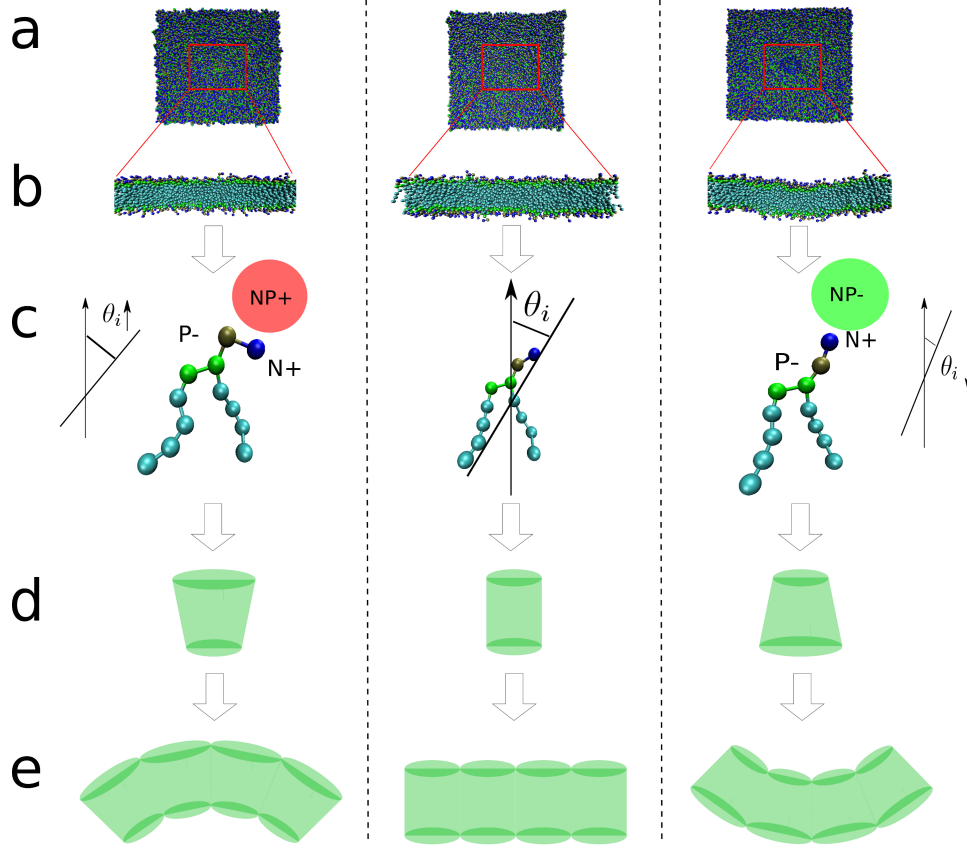# 2   SI_2: Structural changes of lipids induced by the adsorption of charged NPs

Figure 1: Structural changes of lipids induced by the adsorption of charged NPs. The first column shows the changes of the membrane due to the adsorption of cationic NPs of $\rho_e = +3.0\,\mathrm{e/nm^2}$. The second column shows the situation of the pure lipid bilayer. The third column shows the changes of the membrane due to the adsorption of anionic NPs of $\rho_e = -3.0\,\mathrm{e/nm^2}$. Panels (a) shows the top view of the membrane, similar to Figure 2(a) and (c) in the manuscript. Panel (b) shows the side view of the marked region of the membrane in the panel (a). Panel (c) show the change of tilt angle (θ_i) of single lipid molecule in the panel (b). Panel (d) show the shape scheme of single lipid molecule, where the changed area of head group would re-shape lipids. The change of the area of the head group can be found in Figure 2(b) and (d) in the manuscript. Panel (e) show that the change of lipids could affects the rearrangement of the lipid assembly[2].

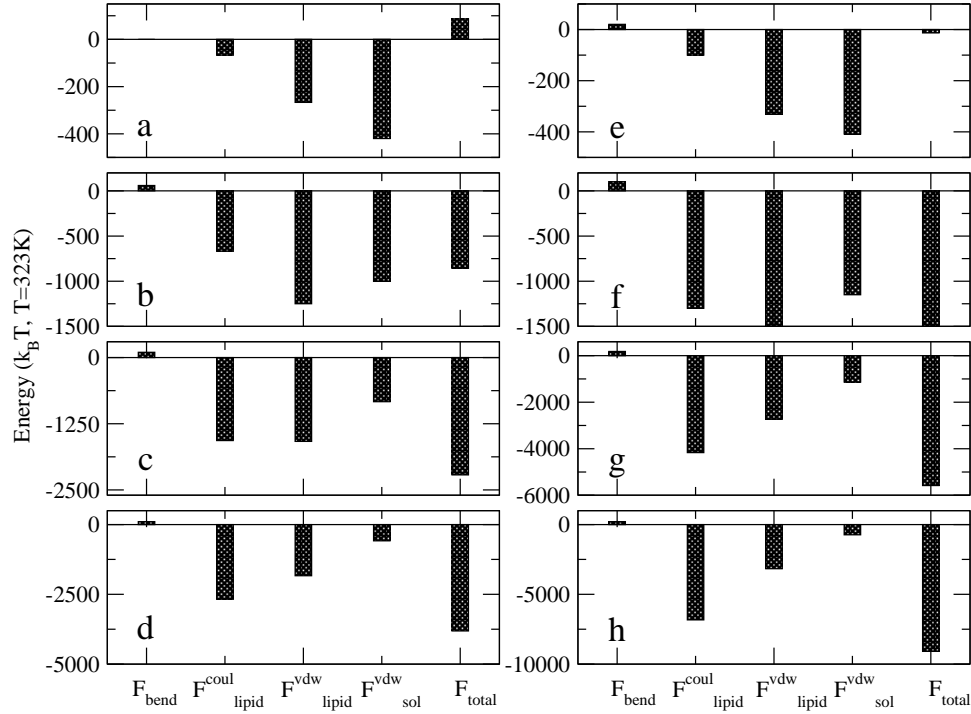# 3   SI_3: System energy contributions of different components in the interactions

Figure 2: System energy contributions of different components in the interactions. Panels (a)–(d) show the energy contributions of positively charged NPs of $\rho_e = +3.0, +6.0, +9.1, +12.1\,\mathrm{e/nm^2}$; Panels (e)–(h) show the energy contributions of negatively charged NPs of $\rho_e = -3.0, -6.0, -9.1, -12.1\,\mathrm{e/nm^2}$.

References

[1] S. J. Marrink, A. H. de Vries, and A. E. Mark. Coarse grained model for semi-quantitative lipid simulations. *Journal of Physical Chemistry B*, 108(2):750–760, 2004.

[2] Ira R. Cooke and Markus Deserno. Coupling between lipid shape and membrane curvature. *Biophysical Journal*, 91(2):487–495, July 2006.