Supporting Information
Computer Simulation Studies on Passive Recruitment Dynamics of Lipids Induced by the Adsorption of Charged Nanoparticles

Yang Li*

1 The equilibrium configuration of the DSPC/DOPG lipid bilayer

The biomembrane model in the simulations consists of two kinds of lipid species: distearoyl-phosphatidylcholine (DSPC), which is a neutral-charged phospholipid; and dioleoyl-phosphatidylglycerol (DOPG), a negatively charged phospholipid. The lipid bilayer contains 7200 lipids with a 5:1 molar mixture of DSPC and DOPG which were uniformly mixed together. After energy minimization, equilibration runs have been performed for 40 ns with a time step of 40 fs. The final equilibrium configuration, shown in Figure 1, is used as the starting state for the next simulation with nanoparticles participation.

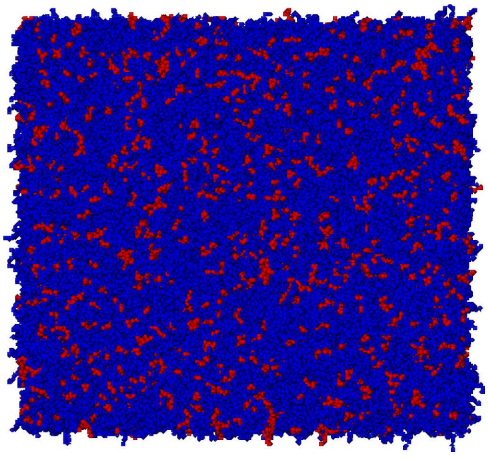

Figure 1: The equilibrium configuration of the DSPC/DOPG lipid bilayer. Red lipids stand for DOPG molecules and blue lipids stand for DSPCs.

2 Free lateral diffusion of individual DOPGs

For verifying the adequacy of MARTINI for simulation of this mixed bilayer, a pure DSPC/DOPG membrane was built; and plots of the flow patterns in absence of nanoparticle were obtained as a negative control for the simulations in presence of charged nanoparticle. A long simulation of the DSPC/DOPG membrane has been performed for 120 ns at the equilibration stage. A series of displacement changes of DOPGs are shown as

*Department of Biomedical Engineering, Tianjin Medical University, Tianjin 300070, P. R. China

vectors in external figure file named SI-2.gif, where the direction of the vector denotes the direction of the motion of a corresponding DOPG molecule, and the length of the vector, the displacement of the motion of a DOPG. The sampling scheme of plots of the flow patterns in SI-2.gif is shown in Figure 2. Shown in SI-2.gif, it is found that the in-plane motion trajectory of charged lipids yields a type of collective dynamics in spite of some local disorder flows in the plots which validates the effectiveness of the MARTINI coarse-grained force field for reproducing the lateral diffusion of lipids in the membrane.

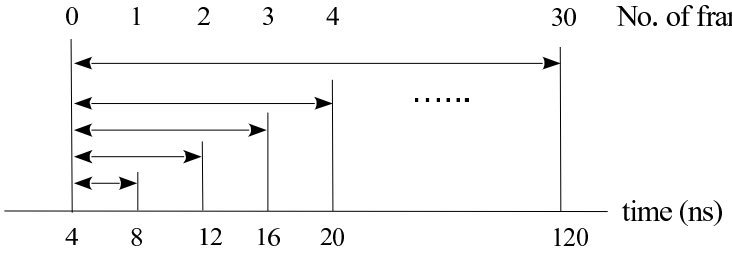

Figure 2: The sampling scheme of plots of the flow patterns. The configuration starts at the time of 4ns in the trajectory, which is regarded as the initial state t0, and ends at the time of 120ns. For frames in the external animation file of SI-2.gif, the sampling time intervals with respect to t0 increase at the rate of 4ns (e.g. the starting time of 4ns is t0, the first frame is taken at the time of 4ns, the second frame is taken at the time of 8ns, the third frame is taken at the time of 12ns, ..., and so on).

3 Umbrella sampling windows

Umbrella sampling approach was employed to obtain the potential of mean force (PMF) as a energetic function of moving a lipid cluster along the normal direction of the membrane to the water. A series of separate biased simulations were performed for 6 ns per simulation window in which the headgroups of lipids were restrained to a given distance from the center of the bilayer by a harmonic restraint on the z-coordinate only. Taking the system of 10 pulled lipids for example, several force constant values were tested for the justification of the force constant value used in the umbrella sampling simulations. The sampling windows were fixed at the distance of 5.045nm shown in Figure 3. Among these force constants, the force constant of 2500 kJ mol^{-1} nm^{-2} was used with a spacing of 0.1 nm between the centers of the biasing potentials to ensure the overlapping of the z histograms between adjacent umbrella sampling windows .

The changes of PMF profiles are relevant to the number of pulled lipids. Four umbrella sampling simulations were performed in which the pulled lipid cluster includes one lipid, 4 lipids, 10 lipids, and 20 lipids respectively. Overlapping of the z histograms between adjacent umbrella sampling windows for different pulled lipids clusters are shown as follow.

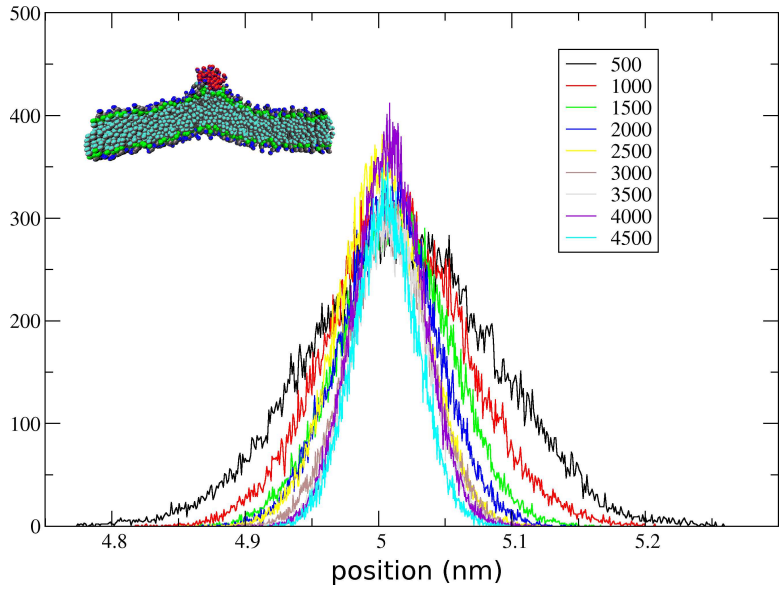

Figure 3: Umbrella sampling windows of force constant of 500, 1000, 1500, 2000, 2500, 3000, 3500, 4000, 4500 kJ mol^{-1} nm^{-2} at the distance of 5.045nm.

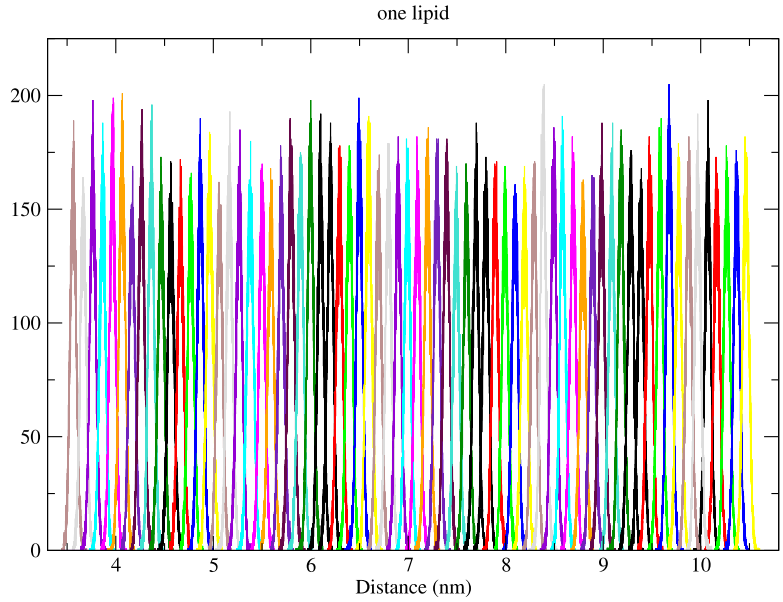

Figure 4: Overlapping of the z histograms between adjacent umbrella sampling windows for the PMF of one pulled lipid.

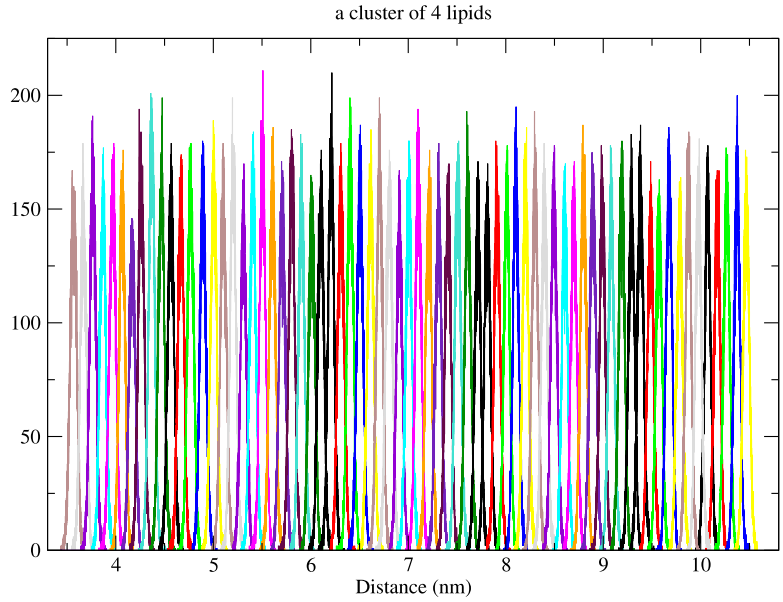

Figure 5: Overlapping of the z histograms between adjacent umbrella sampling windows for the PMF of 4 pulled lipids.

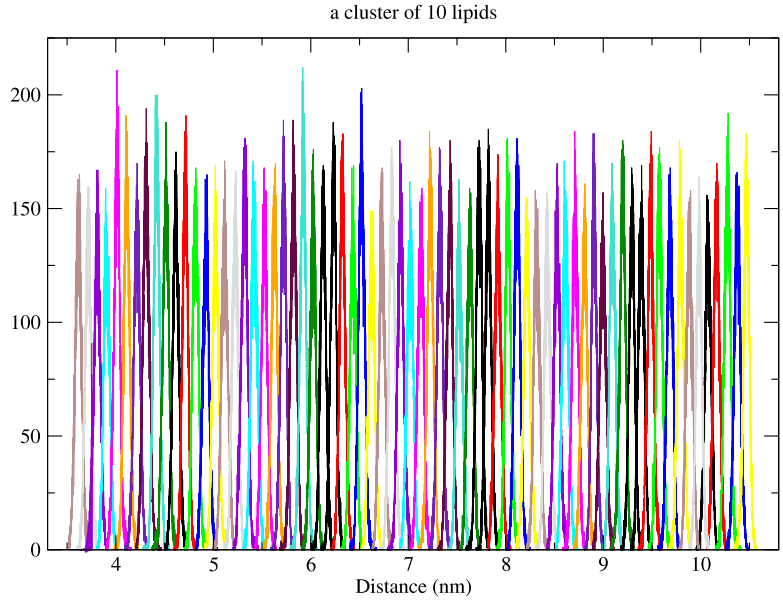

Figure 6: Overlapping of the z histograms between adjacent umbrella sampling windows for the PMF of 10 pulled lipids.

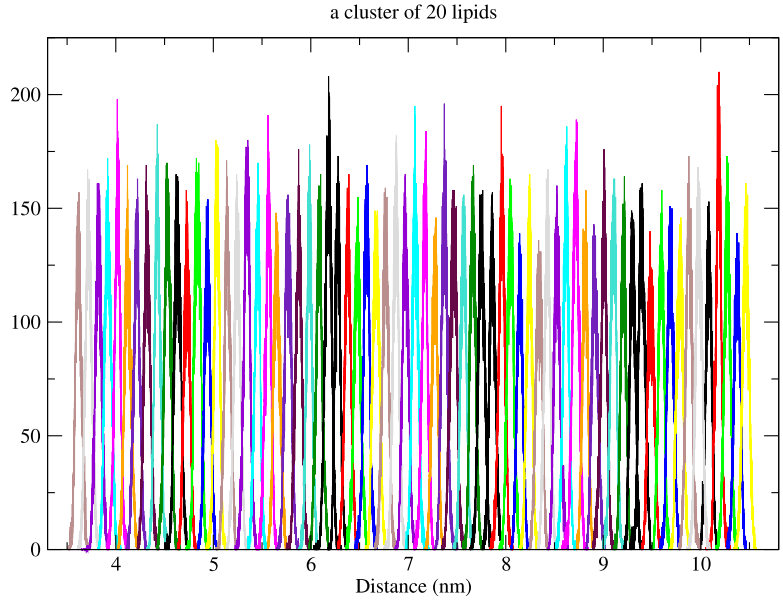

Figure 7: Overlapping of the z histograms between adjacent umbrella sampling windows for the PMF of 20 pulled lipids.