Charge Renormalization of Bilayer Elastic Properties

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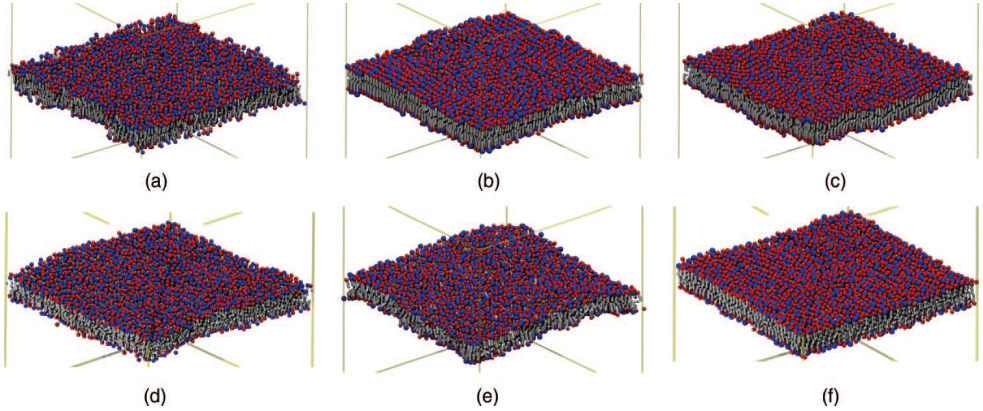
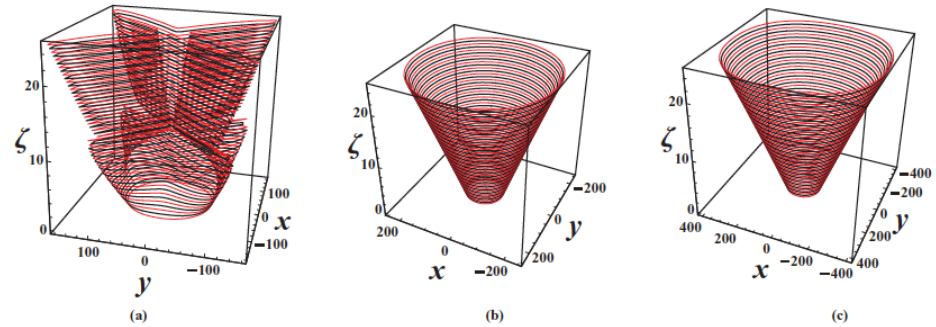
We have combined coarse-grained MD simulations and analytical arguments to show that the presence of charges on the lipid head groups can significantly renormalize the elastic properties of a lipid bilayer, that is, the bending rigidity, **κ** , bulk modulus, **λ** , and the Young modulus, **Y** . We find that **κ** , **λ** , and **Y** strongly depend on the local charge of the lipid head groups. Bilayers with larger head group charges, such as the 1 : 3 systems, are stiffer than those with lower charges on their head groups under the same conditions. MD simulations reveal a clear dependence of the elastic parameters on temperature, an effect that has been only partly addressed due to a high computational cost.



Snapshots of equilibrated bilayer with 4000 lipid molecules at T = 0.7 (top) and T = 0.6 (bottom) for the head group charge systems 1 : 1 (a) and (d), 1 : 2 (b) and (e), and 1 : 3 (c) and (f). For T = 0.7, one can observe onset of ordering of lipid tails in the 1 : 3 head group charge bilayer; however, the system is still liquid, but with very low lipid mobility. At T = 0.6, the tail ordering is apparent already in the 1 : 2 system. Tracking of a randomly selected chain shows that at this temperature both 1 : 2 and 1 : 3 head group charge systems are solid and the lipid molecules are locked in place during the entire duration of the simulation.



*Orientational dependence of the Young modulus, **Y** (**x** , **y**), on the relative strength of the Coulomb interaction ζ , for 1 : 1, 1 : 2, and 1 : 3 head group charges. In the 1 : 1 case, for weak electrostatic interactions the minimum-energy configuration is a close-packed hexagonal lattice, resulting in an isotropic Young's modulus reflected by the circular shape of the **Y** (**x** , **y**). As the strength of the electrostatic interaction is increased, the 1 : 1 system undergoes a structural transition into a square lattice, and the Young modulus develops a directional dependence (fan-like shape for large ζ). The 1 : 2 and 1 : 3 systems are hexagonal for all values of ζ, and their Young's moduli are always isotropic.*

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