

## Movie captions

**Movie 1:** Structural instability and self-healing of a tensionless planar bilayer, initially assembled from  $N_{ul} = 986$  red-green lipids in the compressed upper leaflet and  $N_{il} = 696$  purple-blue lipids in the stretched lower leaflet. The bilayer first bulges towards the upper leaflet, which then undergoes a structural instability by expelling about 100 red-green lipids that form a globular micelle in contact with the upper leaflet. After about 1000 ns, the red-green lipids start to undergo flip-flops along the contact line between micelle and bilayer towards the stretched lower leaflet. These flip-flops lead to a self-healing process that is completed after 1700 ns. The restored bilayer contains 93 red-green lipids in its lower leaflet and remains in a long-lived metastable state without flip-flops until the end of the simulations.

**Movie 2:** Structural instability and self-healing of a large nanovesicle with a diameter of 19 nm. The tensionless bilayer of the nanovesicle is initially assembled from  $N_{ol} = 2105$  red-green lipids in the compressed outer leaflet and  $N_{il} = 770$  purple-blue lipids in the stretched inner leaflet. The bilayer first undergoes pronounced shape fluctuations which have a ‘kinky’ appearance and then forms a cylindrical micelle from about 180 red-green lipids that are expelled from the outer leaflet. After about 1700 ns, the red-green lipids start to undergo flip-flops towards the stretched inner leaflet along the contact line between micelle and bilayer. These flip-flops drive a self-healing process that is completed after 2710 ns. The restored bilayer contains 111 red-green lipids in its inner leaflet and remains in a long-lived metastable state without flip-flops until the end of the simulations.

**Movie 3:** Structural instability and self-healing process of a small nanovesicle with a diameter of 13 nm. The tensionless bilayer of the nanovesicle is initially assembled from  $N_{ol} = 1041$  red-green lipids in the compressed outer leaflet and  $N_{il} = 276$  purple-blue lipids in the stretched inner leaflet. The bilayer first undergoes pronounced shape fluctuations which have a ‘kinky’ appearance and then forms a cylindrical micelle from about 120 red-green lipids that are expelled from the outer leaflet. After about 4700 ns, the red-green lipids start to undergo flip-flops towards the stretched inner leaflet along the contact line between micelle and bilayer. These flip-flops drive a self-healing process that is completed after 5800 ns. The restored bilayer contains 57 red-green lipids in its inner leaflet and remains in a long-lived metastable state without flip-flops until the end of the simulations.