

Supporting Information for

**Membrane Poration, Wrinkling, and Compression: Deformations of Lipid Vesicles  
Induced by Amphiphilic Janus Nanoparticles**

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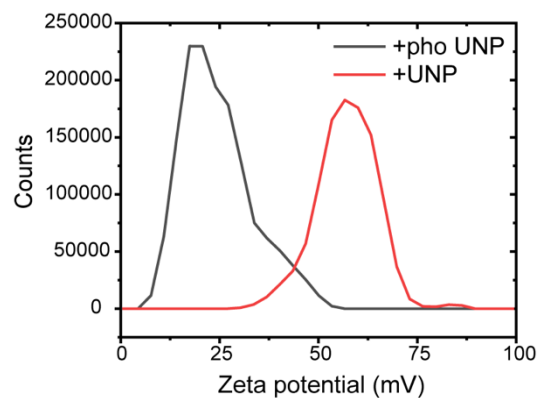
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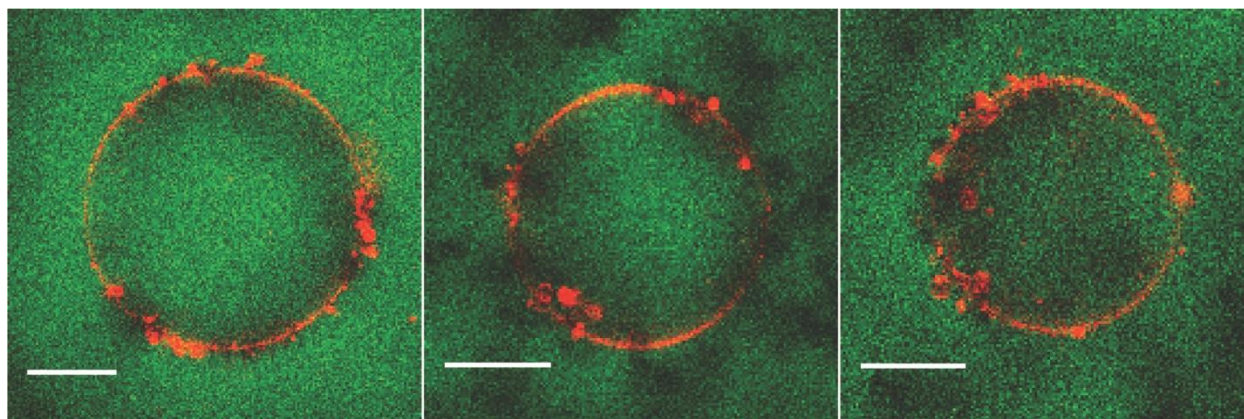
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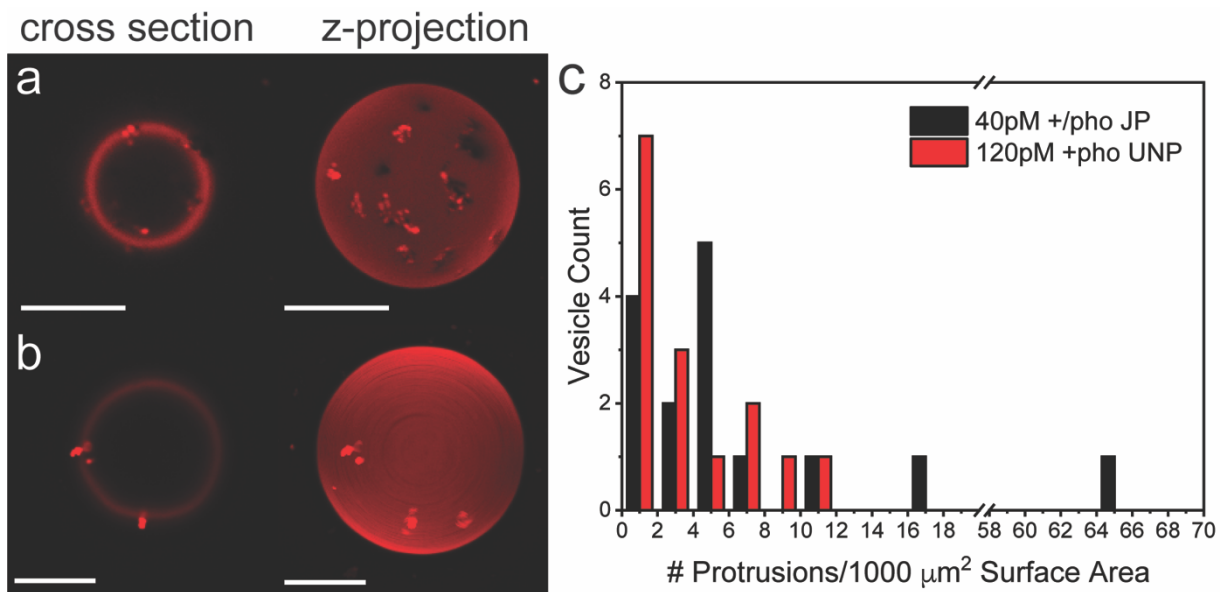
Movies S1-3



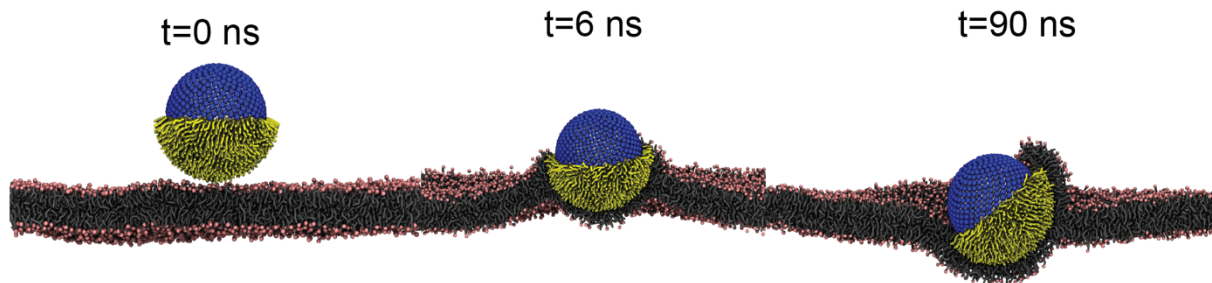
**Figure S1.** Zeta potential of +UNP and +pho UNPs suspended in 2 mM acetic acid (pH = 3.9). Negative shift in zeta potential indicates successful functionalization of particles with octadecylmethoxysilane.



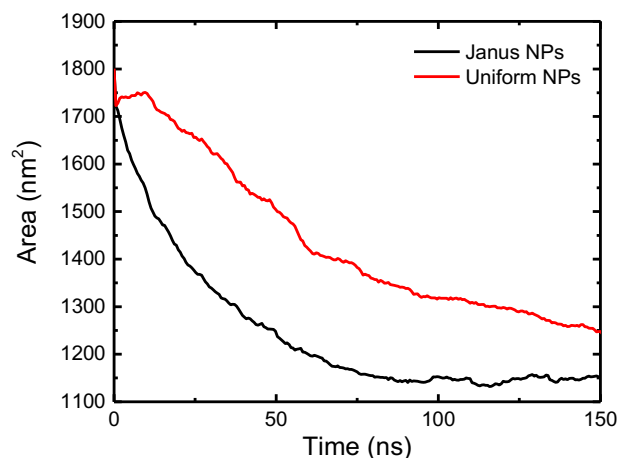
**Figure S2.** Colocalization of membrane protrusions and amphiphilic Janus nanoparticles on GUVs incubated with 40 pM +/pho JP. Scale bars: 20  $\mu$ m.



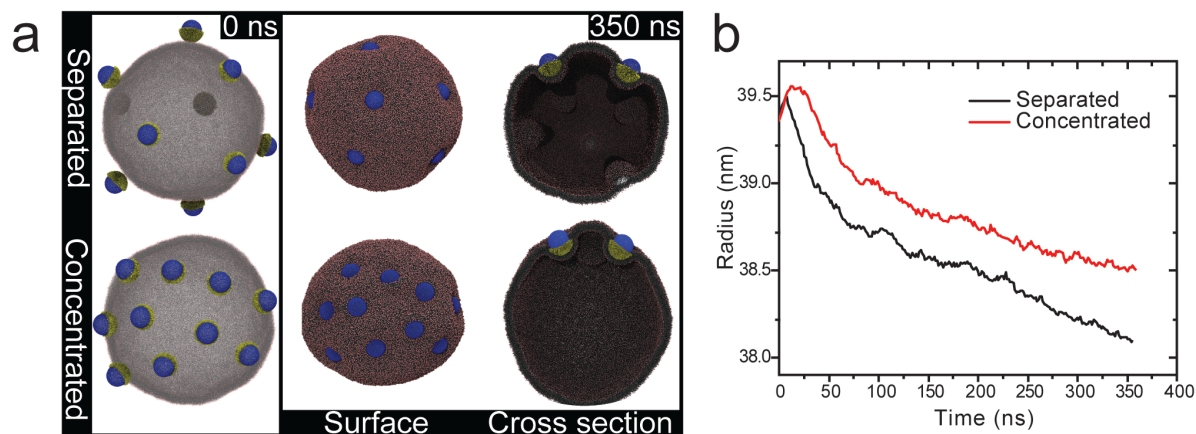
**Figure S3.** Cross section and z-projection of vesicles incubated with (a) 40 pM +/pho JP and (b) 120 pM +pho UNP. (c) Number of membrane protrusions normalized by vesicle surface area (N=15 vesicles). Scale bars: 15 μm.



**Figure S4.** Interaction between a Janus nanoparticle with a membrane of increased size of (42×42) nm<sup>2</sup>.



**Figure S5.** Area of the lipid membrane changes as a function of time when interacting with Janus nanoparticles and uniform nanoparticles as shown in Figure 4c,d. The membrane area is defined as projection area in the xy plane parallel to the membrane.



**Figure S6.** Molecular simulation results showing the effect of spatial distribution of Janus nanoparticles on a lipid vesicle. (a) A total of ten Janus nanoparticles are distributed either uniformly over the entire vesicle (“separated”) or concentrated on one side of the vesicle (“concentrated”). (b) Changes in the vesicle radius as a function of simulation time for the two cases.

**Movie S1.** Z-stack fluorescence confocal images showing GUVs labeled with RhB-DOPE (shown in red) and 40 pM +/-pho JPs. To make the metal-coated non-fluorescent +/-pho JPs visible, carboxyfluorescein (shown in green) was added in the aqueous solution such that the +/-pho JPs appear as dark spots under the fluorescent background. Scale bars: 10  $\mu\text{m}$  .

**Movie S2.** Confocal images of a collapsing giant vesicle (red) incubated with carboxyfluorescein dye (green) and 40 pM +/-pho JPs. +/-pho JPs appear dark due to blocking of carboxyfluorescein emission. Particles concentrate in the center of the collapsed vesicle.  $t = 0$  is set when +/-pho JPs were added to the vesicles. Scale bars: 20  $\mu\text{m}$ .

**Movie S3.** Z-projection of a giant vesicle labeled with RhB-DOPE (gray) incubated with 40 pM +/-pho JPs collapsing into a lipid bilayer. After collapsing, a bright particle-lipid complex remains in the middle of the newly formed planar lipid bilayer.  $t = 0$  is set when +/-pho JPs were added to the vesicles. Scale bar: 30  $\mu\text{m}$ .