

Figure S1. Aminodextran, myoglobin and α -synuclein do not induce the collapse of L_d domains in liquid-liquid phase separated GUVs containing 10 mol.% CL. Images are taken approximately 40 min. after the addition of protein/polymer. Rh-DPPE (red) labels L_d domains and NBD-PE (green) labels the L_o domains. Scale bars in the bottom right of the images are 50 µm.

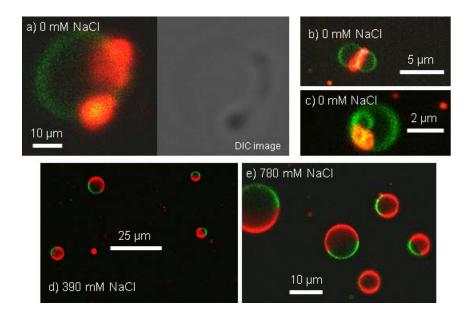


Figure S2. Domain morphology transitions investigated at different ionic strengths: (a-c) at 0 mM NaCl, collapse of the L_d domains (red) is still observed. Part (a) shows a confocal fluorescence image and a DIC image of the same GUV. In (b) collapsed domains on adjacent vesicles are observed to adhere to one another. Note that (d, e) at 390 mM and 780 mM NaCl GUVs are unperturbed after incubation for 30 min. in the presence of 17 µm cyt *c*.

Supplementary Video captions:

Video S1. Collapse of CL-containing domains induced by cyt *c*. 10% CL phase separated GUVs labeled with Rh-DPPE (red) and NBD-DPPE (green) in the presence of Alexa Fluor 568 – labeled cyt *c*. The 16 s video clip represents 2 min. in real time. The L_d domain (red) of the GUV in the center of the image is seen to collapse over time. Beading of the CL-containing domain of the GUV in the top left corner begins midway through the video clip accompanied by a sudden decrease in vesicle size. No morphology changes are observed in the GUV in the bottom right corner during the video.

Video S2. Final stages of L_d domain collapse of a GUV. 10% CL phase separated GUVs labeled with Rh-DPPE (red) and NBD-DPPE (green) in the presence of Alexa Fluor 568 – labeled cyt *c*. The 16 s video clip represents 2 min. in real time. The focal plane through the GUV is varied during the time course of the video.

Video S3. Morphology changes in a CL-containing, phase separated GUV induced by PAMAM generation 3 dendritic polymers. The GUV is labeled with Rh-DPPE (red) and

NBD-DPPE (green). Folding of the L_d domain can be seen; these folds bud off into small vesicular structures localized in the lumen of the GUV. The 31 s video clip represents 2 min. in real time.