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Supplemental Information

GM1 Softens POPC Membranes and Induces the Formation of Micron-Sized Domains

Nico Fricke and Rumiana Dimova

SUPPORTING INFORMATION

Text S1. Assessing the membrane tension in vesicles exposed to electrodeformation

The force density f arising from the accumulation of electric charge at both interfaces of the membrane acts as a local pressure on the membrane in addition to the pressure difference Δp between the interior and exterior of the vesicles as described by the Young-Laplace equation. At the poles (pol) and at the equator (equ), the force balance between pressure and tension has the form $2M_{\mathrm{pol}}\Sigma = \Delta p + f_{\mathrm{pol}}$ and $2M_{\mathrm{equ}}\Sigma = \Delta p + f_{\mathrm{equ}}$, where M_{pol} and M_{equ} are the mean curvatures of the membrane at the poles and equator, respectively, and f_{pol} and f_{equ} are the respective force densities. By eliminating the osmotic pressure from these equations, one can obtains for the tension Σ of the vesicle

$$\Sigma = \frac{f_{\mathrm{pol}} - f_{\mathrm{equ}}}{2(M_{\mathrm{pol}} - M_{\mathrm{equ}})}$$

Full derivation of the force densities f_{pol} and f_{equ} can be found in Ref. (1), Appendix B (Ref. 30 in the main text). Here, we use the same notations. The resulting force densities for each angle θ along the vesicle are the superposition of the radial Maxwell stresses directed to the exterior, the bilayer and the interior of the vesicle, denoted as 1, 2 and 3 in the indices, respectively, at the exterior (ex) and interior (in) interface:

$$f(\theta) = [T_{1rr}(r_{\mathrm{ex}},\theta) - T_{2rr}(r_{\mathrm{ex}},\theta)] + [T_{2rr}(r_{\mathrm{in}},\theta) - T_{3rr}(r_{\mathrm{in}},\theta)]$$

where r_{ex} and r_{in}describe the outer and inner radius of the vesicle.

Following equations 84 and 85 in Ref. (1), the radial components of the stresses that may cause a deformation at the exterior interface are:

$$\begin{aligned} T_{1rr}(r_{\mathrm{ex}},\theta) &= \frac{1}{4}\epsilon_1 E_0^2[|\alpha_{1,\mathrm{ex}}|^2\cos^2\theta - |\gamma_{\mathrm{ex}}|^2\sin^2\theta] \\ T_{2rr}(r_{\mathrm{ex}},\theta) &= \frac{1}{4}\epsilon_2 E_0^2[|\alpha_{2,\mathrm{ex}}|^2\cos^2\theta - |\gamma_{\mathrm{ex}}|^2\sin^2\theta] \end{aligned}$$

and at the interior interface (following equations 88 and 89 in Ref. (1):

$$\begin{aligned} T_{2rr}(r_{\mathrm{in}},\theta) &= \frac{1}{4}\epsilon_2 E_0^2[|\alpha_{2,\mathrm{in}}|^2\cos^2\theta - |\gamma_{\mathrm{in}}|^2\sin^2\theta] \\ T_{3rr}(r_{\mathrm{in}},\theta) &= \frac{1}{4}\epsilon_3 E_0^2[|\alpha_{3,\mathrm{in}}|^2\cos^2\theta - |\gamma_{\mathrm{in}}|^2\sin^2\theta] \end{aligned}$$

Here the amplitudes at the exterior (equations 65, 66 and 68 in Ref. (1)) and at the interior interface (equations 70, 71 and 73 Ref. (1)) are:

$$\begin{aligned} \alpha_{1,\mathrm{ex}} &= \beta_1\alpha_{2,\mathrm{ex}} & \alpha_{2,in} &= 9/D \\ \alpha_{2,\mathrm{ex}} &= 3[(1+2\beta_3) + 2(1-\beta_3)\frac{r_{\mathrm{in}}^3}{r_{\mathrm{ex}}^3}]/D & \alpha_{3,in} &= \beta_3\alpha_{2,in} \\ \gamma_{\mathrm{ex}} &= -3\left[(1+2\beta_3) - (1-\beta_3)\frac{r_{\mathrm{in}}^3}{r_{\mathrm{ex}}^3}\right]/D & \gamma_{in} &= -\alpha_{3,in} \end{aligned}$$

with the denominator

$$D = (2-\beta_1)(1+2\beta_3) - 2(1-\beta_1)(1-\beta_3)\frac{r_{\mathrm{in}}^3}{r_{\mathrm{ex}}^3} \qquad ,$$

the complex-value electric parameters

$$\beta_1 = \frac{\sigma_2 - i\omega\epsilon_2}{\sigma_1 - i\omega\epsilon_1}, \quad \beta_3 = \frac{\sigma_2 - i\omega\epsilon_2}{\sigma_3 - i\omega\epsilon_3}$$

and the conductivities $\sigma_{1,2,3}$, the dielectric permittivities $\epsilon_{1,2,3}$ and the circular electric frequency ω.
At the poles ($\theta = 90°$) and equator ($\theta = 0°$), the force densities are then defined as:

$$f_{\mathrm{pol}} = \frac{1}{4}E_0^2\left[(\epsilon_1|\beta_1|^2 - \epsilon_2)\left|\alpha_{2,\mathrm{ex}}\right|^2 + (\epsilon_2 - \epsilon_3|\beta_3|^2)\left|\alpha_{2,\mathrm{in}}\right|^2\right]$$

$$f_{\mathrm{equ}} = -\frac{1}{4}E_0^2[(\epsilon_1 - \epsilon_2)|\gamma_{\mathrm{ex}}|^2 + (\epsilon_2 - \epsilon_3)|\gamma_{\mathrm{in}}|^2]$$

The terms in the square brackets are invariable during the experiment leading to the following dependence for the membrane tension

$$\Sigma = \frac{const\ E_0^2}{(M_{\mathrm{pol}} - M_{\mathrm{equ}})}$$

where $const$ is a dimensional constant. Then, from the logarithmic expression for the relative area change in Eq. 1 in the main text, one obtains the simplified expression in Eq. 2 in the main text.

An approximation for working at "small" field frequencies ω and low conductivities of the solutions $\sigma_{1,3} \gg \sigma_2$ and $\left|\beta_{1,3}\right| \ll 1 - (r_{in}/r_{ex})^3$ leads to:

$$\alpha_{2,\mathrm{in}} \to \frac{9}{2\left(1 - \frac{r_{\mathrm{in}}^3}{r_{\mathrm{ex}}^3}\right)}, \quad \alpha_{2,\mathrm{ex}} \to \alpha_{2,\mathrm{in}} - 3, \quad \gamma_{\mathrm{in}} \to 0, \qquad \gamma_{\mathrm{ex}} \to -\frac{3}{2}$$

yielding for the forces

$$f_{\mathrm{pol}} \to \frac{1}{4}\epsilon_2 E_0^2\left(\frac{27}{1 - \frac{r_{\mathrm{in}}^3}{r_{\mathrm{ex}}^3}} - 9\right) \text{ and } f_{\mathrm{equ}} \to -\frac{9}{16}\epsilon_1 E_0^2$$

From these simplified expressions, the actual membrane tension during electrodeformation can be also assessed.

Figure S1

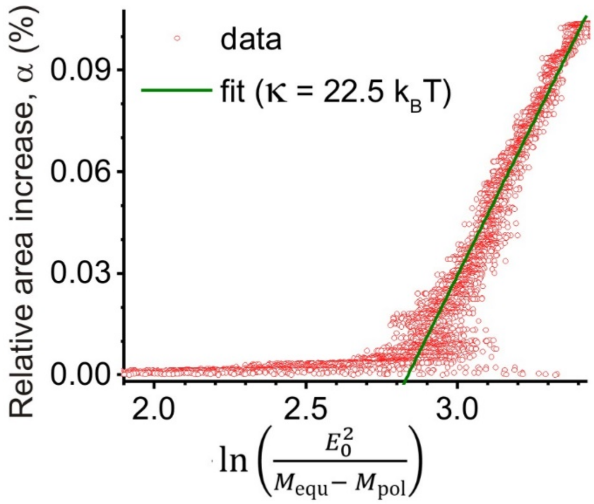

Figure S1. Change in the area of a POPC vesicle at 40 °C as a function of applied electric field strength (in V/m units) as measured in an electrodeformation experiment (the curvatures were measured in units 1/m). From the slope of the data, one obtains the bending rigidity following Eq. 2 in the main text.

Figure S2

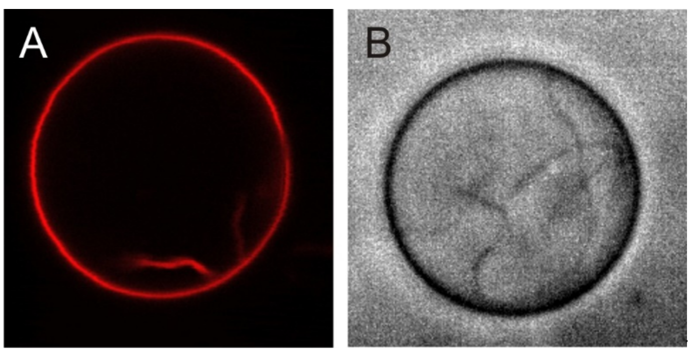

Figure S2. Images of vesicles with internal structures observed at room temperature for GM1 fractions of (A) 5.3 mol% (confocal cross section,) and (B) 10 mol% (phase contrast). The vesicle diameters are approximately (A) 35 μm and (B) 25 μm.

Figure S3

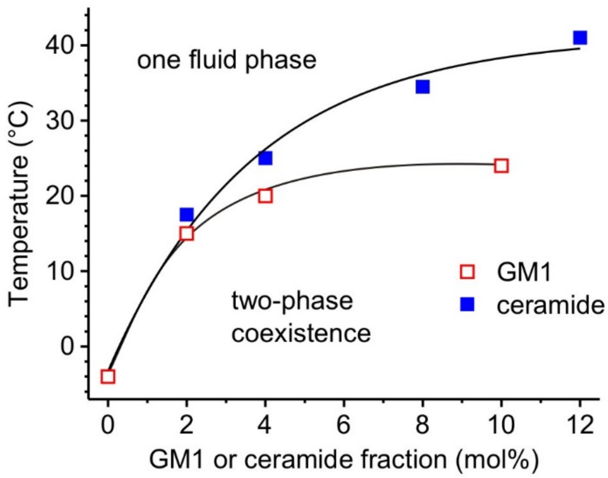

Figure S3. Partial phase diagram of POPC with palmitoyl ceramide (filled squares) or with GM1 (open squares). The data for GM1 is identical to that in Fig. 3A in the main text. The data for the POPC/palmitoyl ceramide system is from Ref. (2) (Ref. 54 in the main text). The solid curves are guides to the eye.

Figure S4

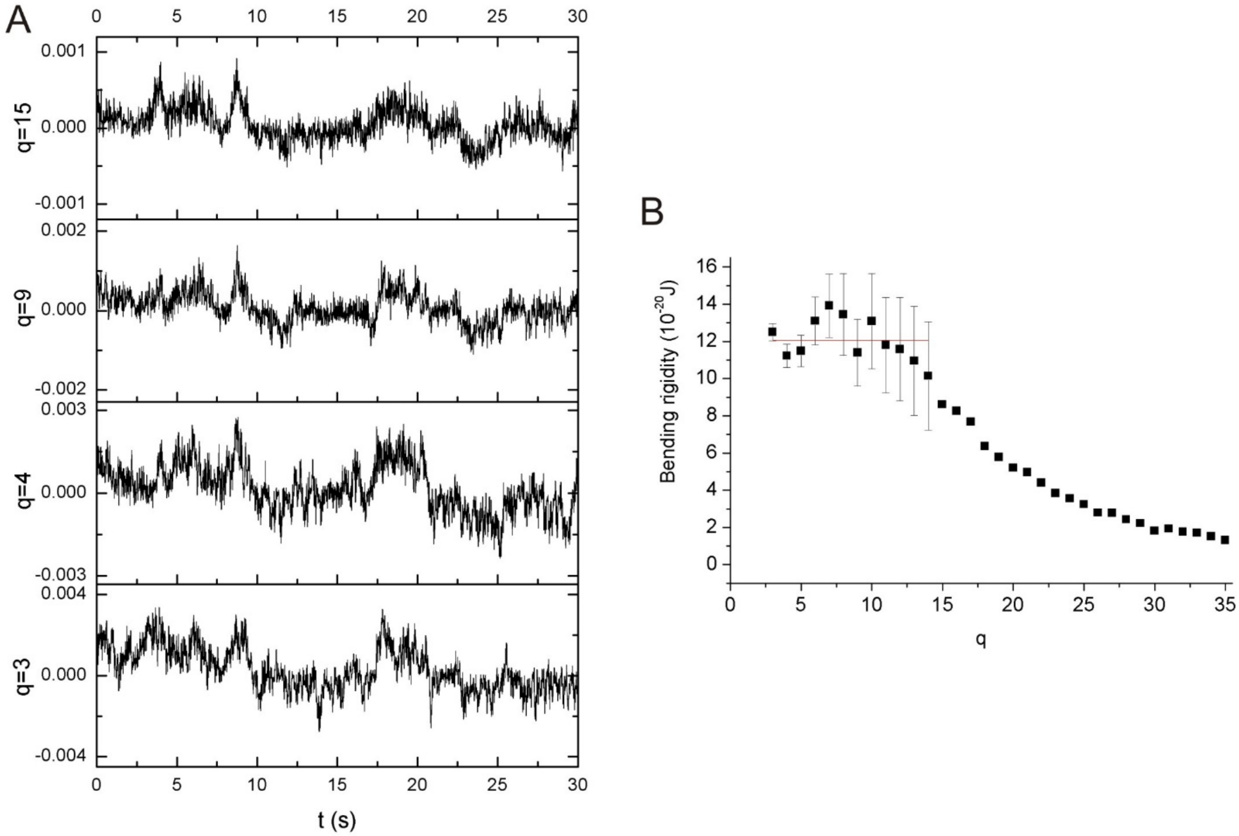

Figure S4. Example data for fluctuation analysis on vesicles with gel-like domains. The analysis is done following the approach in Ref. (3). The data was acquired on a vesicle with 8 mol% GM1 at room temperature. (A) Absolute values of the Fourier coefficients $|v_q|$ for several of the modes q with subtracted mean value. (B) Fit for the bending rigidity deduced for the same vesicle, $\kappa = 12.1 \pm 3.3 \times 10^{-20}$ J.

References

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