

Supporting Information on

Lateral Diffusion of Lipids in DMPG Membrane across the Anomalous Melting Regime: Effects of NaCl

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Dynamic light scattering (DLS)

The intensity autocorrelation function, $g^2(\tau)$, is associated to the first order autocorrelation function of the electric field, $g^1(\tau)$, by the Siegert relation¹,

$$g^2(\tau) = A \left[g^1(\tau) \right]^2 + 1 \quad (\text{S1})$$

Here, A is the spatial coherence factor. For an ideal monodisperse vesicles system, $g^1(\tau)$ function decays as a single exponential. However, for practical case, there is a polydispersity in the system, for which $g^1(\tau)$ can be written as

$$g^1(\tau) = \int_0^\infty G(\Gamma_{\text{DLS}}) \exp(-\Gamma_{\text{DLS}}\tau) d\Gamma_{\text{DLS}}$$

where $G(\Gamma_{\text{DLS}})$ represents the relative weight factor for relaxation rate Γ_{DLS} . For narrow polydispersity, the above expression can be simplified to the well-known cumulant expansion²

$$g^1(\tau) = \exp \left[-\overline{\Gamma_{\text{DLS}}}\tau + \frac{\mu_2\tau^2}{2} \right] \quad (\text{S2})$$

where first and second cumulant, $\overline{\Gamma_{\text{DLS}}}$ and μ_2 are the mean decay constant and variance, respectively. The ratio of the variance to the square of the mean is a measure of the polydispersity in the diffusion coefficient or hydrodynamic size and is represented by the polydispersity index (PDI).

CONTIN³ analysis, which employs inversion of the Laplace integral equation providing full distribution in the size, was also used to describe the observed DLS data for DMPG vesicles in absence and presence of 100 mM NaCl. It is found to describe the data well for both the vesicles at all the measured temperatures. Intensity-weighted distributions of hydrodynamic diameter as obtained from CONTIN analysis for DMPG vesicles with no salt and DMPG vesicles in presence of 100 mM NaCl are shown in Fig. S1.

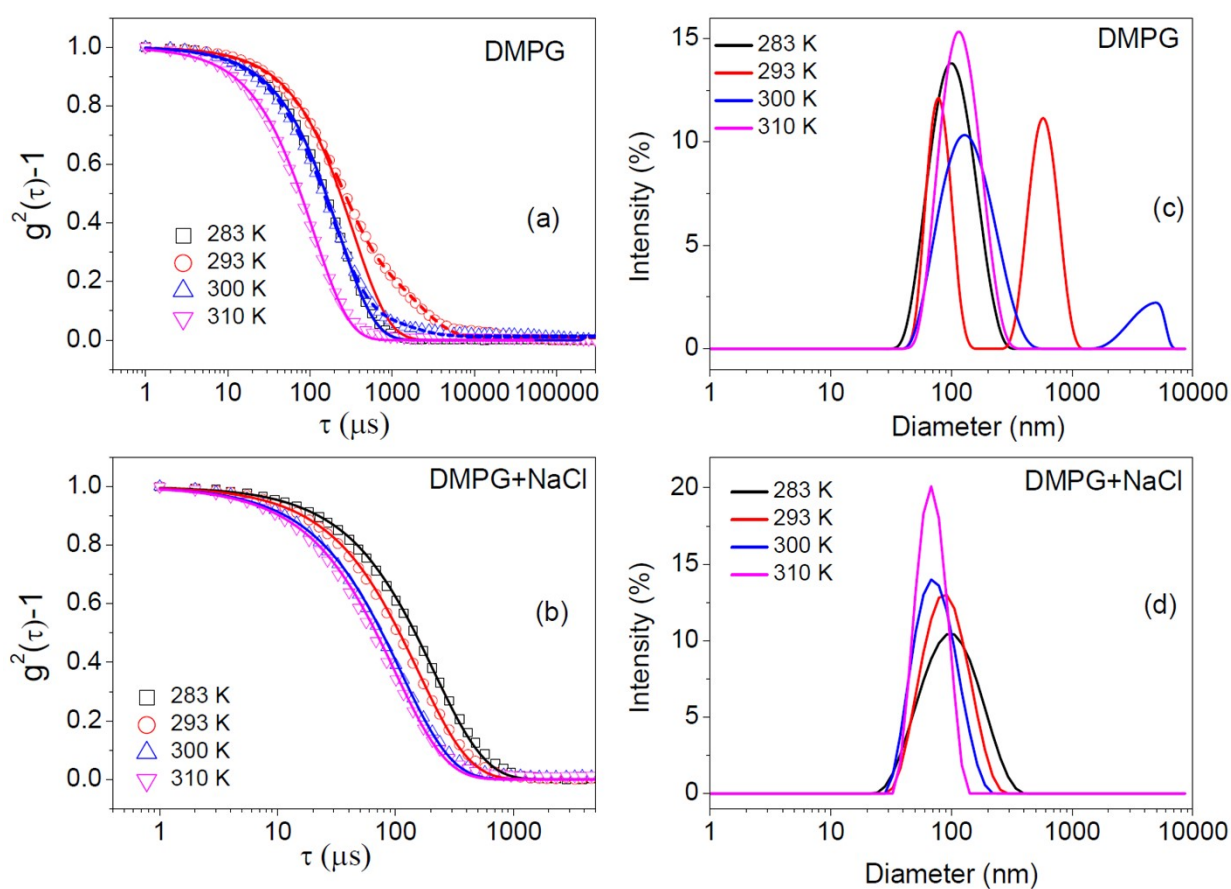


Fig. S1 Intensity autocorrelation function for (a) DMPG vesicles (b) DMPG vesicles with 100 mM NaCl at different temperatures. Solid and dashed lines are the fits as per cumulant and CONTIN analysis, respectively. It is evident that cumulant analysis could describe all the observed data except DMPG vesicles in the intermediate phase (at 293 K and 300 K), where CONTIN analysis describes data well. Size distributions of (c) DMPG vesicles (d) DMPG vesicles with 100 mM NaCl as obtained from CONTIN analysis.

Small Angle Neutron Scattering

Small angle neutron scattering (SANS) measurements have been carried out on DMPG vesicles in D₂O using the SANS diffractometer at Dhruva, BARC, India, and the observed data are shown in Fig. S2. The SANS data shows a Q^{-2} dependence in the low Q region and the absence of a Bragg peak, suggesting that the structure of the aggregates is unilamellar vesicles.

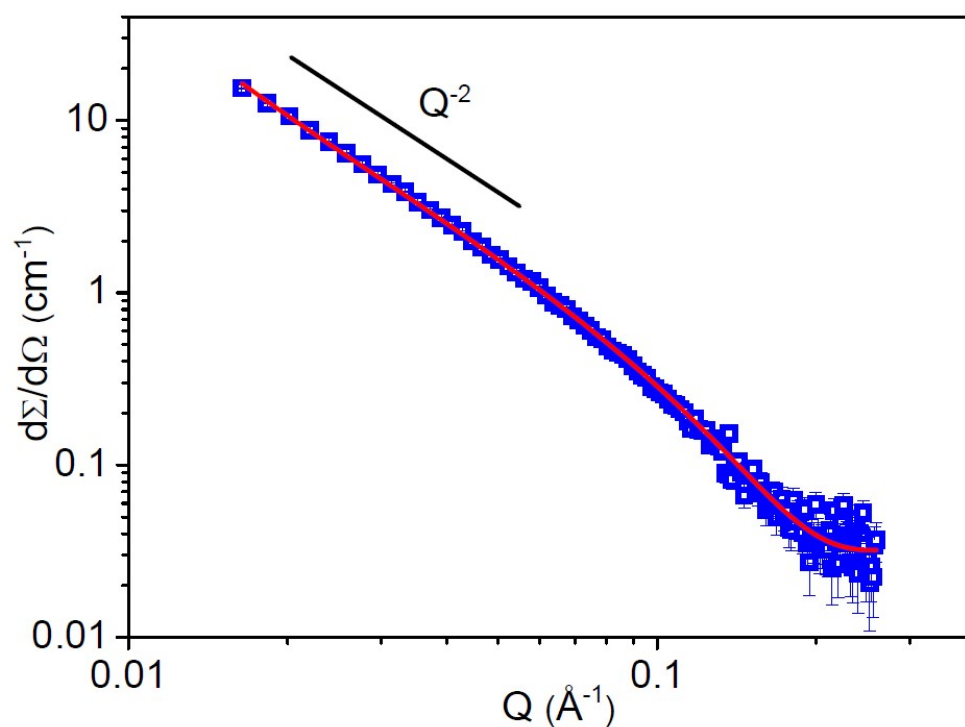


Fig. S2 SANS data for DMPG vesicles in D₂O at 310 K. It is evident that at low Q , the data shows a Q^{-2} behavior and the absence of a Bragg peak.

Elastic Scattering Intensity Scan

Elastic scattering intensity scan data were also measured on higher energy resolution HFBS spectrometer ($\Delta E=0.8$ μ eV) on 5 (w/w) % DMPG ULVs with and without 100 mM NaCl with a cooling rate of 0.25 K/min. Measurement time at each temperature was set for 2 min per data point. Observed elastic intensity scan data is shown in Fig. S

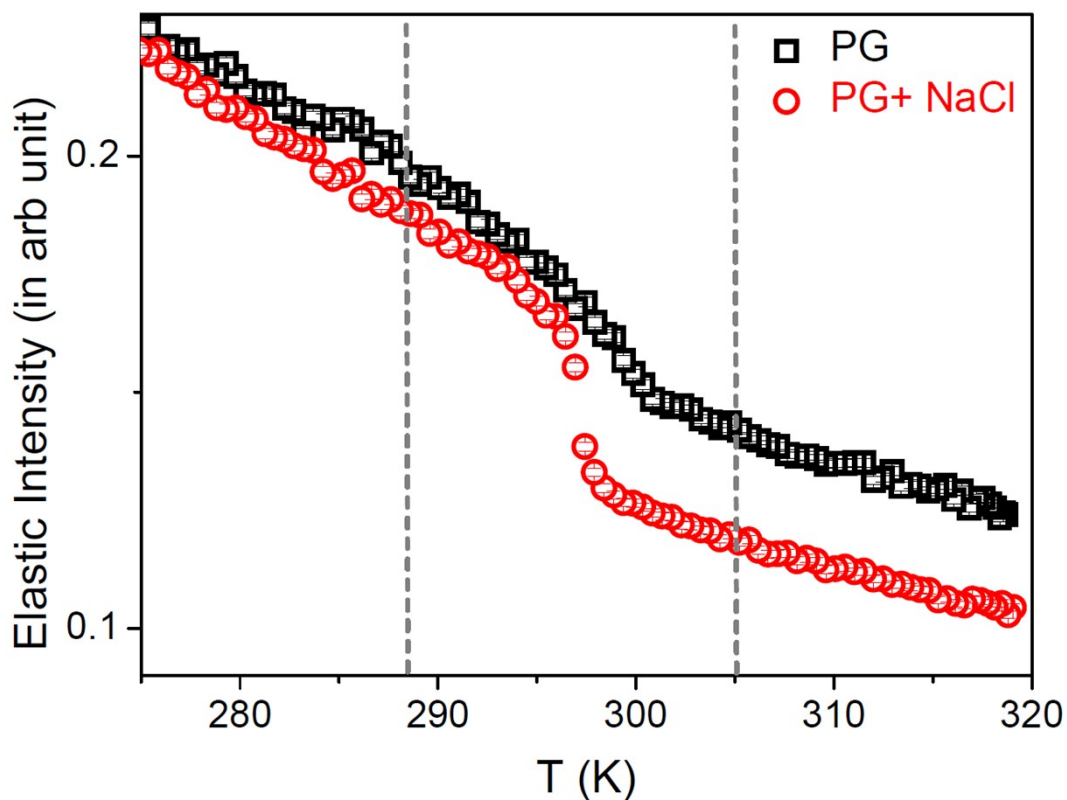


Fig. S3 Q -averaged elastic scattering intensity for 5 (w/w) % DMPG unilamellar vesicles with and without 100mM NaCl as observed using higher energy resolution HFBS spectrometer ($\Delta E=0.8 \mu\text{eV}$). The vertical short dashed lines represent the onset and endpoint of the phase transitions as obtained from DSC.

References

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