Electronic Supplementary Material (ESI) for Soft Matter.

Modeling the Dynamics of Phospholipids in the Fluid Phase of Liposomes

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ELECTRONIC SUPPLEMENTARY INFORMATION

Neutron Spin Echo Spectroscopy Basics

Neutron Spin Echo (NSE) spectroscopy detects the sum of coherent and incoherent scattering. Typically, the coherent part dominates which leads to[S1]

$$\frac{S(Q,t)}{S(Q)} = \frac{\sigma_{\mathrm{coh}} S(Q,t)_{\mathrm{coh}} - (1/3)\sigma_{inc} S(Q,t)_{\mathrm{inc}}}{\sigma_{\mathrm{coh}} S(Q)_{\mathrm{coh}} - (1/3)\sigma_{\mathrm{inc}} S(Q)_{\mathrm{inc}}} \approx \frac{S(Q,t)_{\mathrm{coh}}}{S(Q)_{\mathrm{coh}}} \quad \text{(S1)}$$

where σ_{coh} and σ_{inc} are the coherent and incoherent scattering cross-sections, respectively. The coherent and the incoherent intermediate scattering functions are represented by $S(Q,t)_{\mathrm{coh}}$ and $S(Q,t)_{\mathrm{inc}}$, respectively.

The time dependent mean-squared displacement of randomly diffusing, infinitely small and massless particles can be written as $(\langle (r_i(0) - r_i(t))^2 \rangle = 6\,D\,t)$.[S2] Assuming validity of the Gaussian approximation in the calculation of the time dependent incoherent dynamic structure factor, $S(Q,t)_{\mathrm{inc}}^{\mathrm{tr}}$, we arrive at the well-known expression[S2]

$$S(Q,t)_{\mathrm{inc}}^{\mathrm{tr}} = N \exp(-Q^2 D_{\mathrm{eff}} t) \quad \text{(S2)}$$

Equation S2 implicitly includes N particles randomly diffusion independently of each other.

In a next step, we exploit the fact that the pair-correlation function can be separated into inter- and intermolecular contributions. In case of dilute solutions which implies uncorrelated motion, the intermolecular contributions can be neglected.[S1,S2] Since we assume non-interacting, infinitely small and massless particles the intramolecular interactions can be ignored.[S1,S2]. In this or in the more general case, of objects with finite dimensions, center of mass diffusion and a separation ansatz, e.g., to separate rotational motion, can be utilized to derive the coherent dynamic structure factor, $S(Q,t)^{\mathrm{tr}}_{\mathrm{coh}}$,

$$S(Q,t)^{\mathrm{tr}}_{\mathrm{coh}} \propto N \exp(-Q^2 D_{\mathrm{eff}} t) \tag{S3}$$

Eqs. S2 and S3 illustrate the more general principle that the coherent dynamic structure factor contains pair- and self-correlation contributions. In those cases, when objects move independently the self-correlation part in $S(Q,t)_{\mathrm{coh}}$ has the same form as in the respective part in $S(Q,t)_{\mathrm{inc}}$.

Different Illustrations of the Results

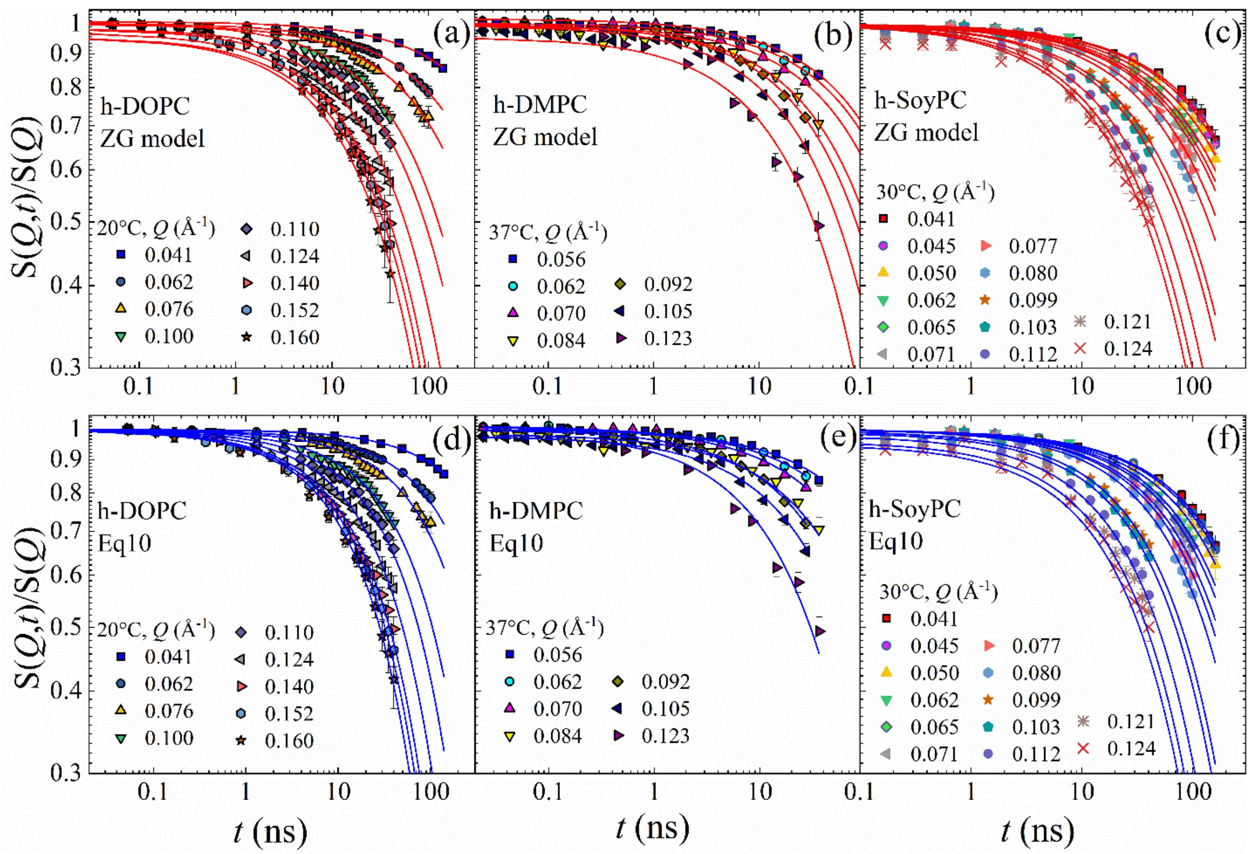

Figure S1: Log-log representations of the normalized intermediate scattering function, S(Q,t)/S(Q), as a function of Fourier time, t, for different Q's, for, (a,d) 5 % lipid mass fraction of protonated DOPC at 20 ℃, (b,e) 5 % lipid mass fraction of protonated DMPC at 37 ℃ and (c,f) the 5 % lipid mass fraction of protonated Soy-PC sample at 30 ℃, all in D_2O. The same data sets are analyzed by fits using the (a-c) Zilman-Granek model (ZG) (equation 5) and (d-f) the full model that starts from equation 3 and includes diffusion of liposomes and confined motion of lipid tails (equation 10). The error bars representing one standard deviation.

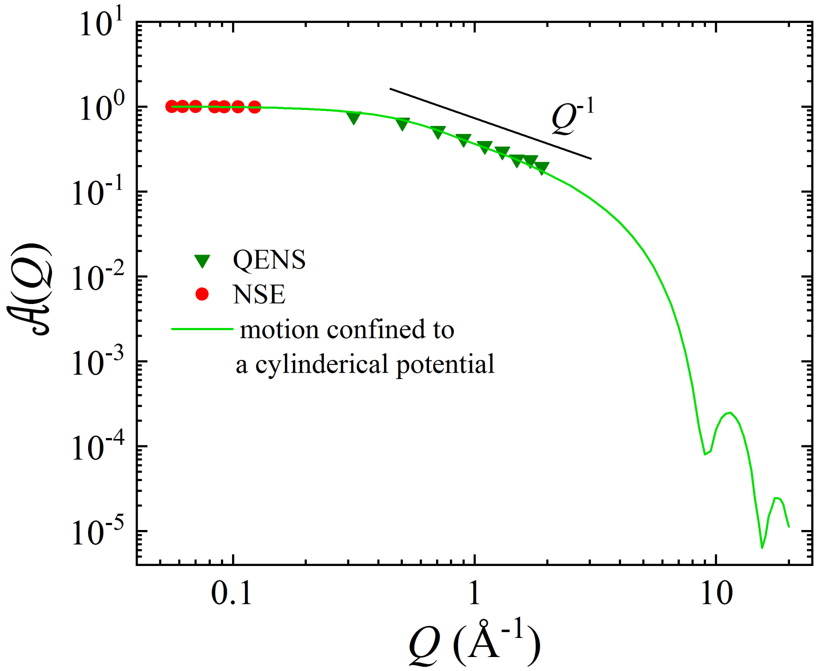

Figure S2: The $\mathcal{A}(Q)$ for h-DMPC obtained from NSE and QENS studies, over a broad Q-range. The data is modelled using $\mathcal{A}(Q)$ for a particle diffusion in a cylinder.

References:

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S2. J. S. Higgins and H. C. Benoit, *Polymers and Neutron Scattering*, Clarendon Press, Oxford New York, 1996.

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S4. G. J. Schneider, K. Nusser, S. Neueder, M. Brodeck, L. Willner, B. Farago, O. Holderer, W. J. Briels and D. Richter, Soft Matter, 2013, 9, 4336.

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