Supporting Text

Calculation of the Scattering Cross-Section

Form Factor. For specular data, we calculate the form factor $\tilde{\rho}(q_z) = \left|\int \rho(z) e^{iq_z z} dz\right|^2$ using the 1G-hybrid model:

$$\begin{aligned}\tilde{\rho}(q_z) &= \frac{2(\rho_{CH_2} - \rho_{H_2O}) \sin(q_z z_h) \cos(q_z \sigma_h)}{q_z(1 - (2/\pi q_z \sigma_h)^2)} \\ &+ 2\sqrt{2\pi} \rho_h \sigma_h cos(q_z z_h) \exp(-1/2 q_z^2 \sigma_h^2) \\ &+ 2\sqrt{2\pi} \rho_{CH_3} \sigma_{CH_3} cos(q_z z_{CH_3}) \exp(-1/2 q_z^2 \sigma_{CH_3}^2).\end{aligned}$$

[7]

Note that σ_h and σ_{CH_3} lump contributions not only from rms thermal fluctuations but also from the width of the transition between two neighboring regions.

For diffuse scattering data, we use the Fourier expansion

$$\tilde{\rho}(q_z) = \int_{D/2}^{D/2} [\rho(z) - \rho_w] e^{-iqz} dz = \sum_{h=-h_{\max}}^{h_{\max}} F_h \frac{\sin(qD/2 - \pi h)}{(qD/2 - \pi h)}.$$

[8]

Height:Height Correlation Function. To calculate the height:height correlation function $\langle z(\mathbf{0}) z(\mathbf{r}_\parallel)\rangle$, we take the Fourier transform of the Helfrich thermal fluctuation spectrum $\langle z(q_\parallel) z(q_\parallel)\rangle$ (see main text) and obtain

$$\langle z(\mathbf{0}) z(\mathbf{r}_\parallel)\rangle = \frac{k_B T}{2\pi\sqrt{\Delta}} \times \left[K_0(q_1 r_\parallel) - K_0(q_2 r_\parallel)\right],$$

[9]

where $\Delta = \gamma^2 - 4U''\kappa$. K_0 is the modified Bessel function of the second kind of order 0. For real arguments,

$$K_0(x)_{x \to 0} \approx Log2 - \gamma_E \; Log(x)$$

[10]

where γ_E is the Euler constant and $\lim_{x\to\infty} K_0(x) = 0$.

If $\Delta > 0$, q_1 and q_2 are real:

$$\begin{aligned} q_1 &= \sqrt{\left(\gamma - \sqrt{\Delta}\right)/2\kappa} \\ q_2 &= \sqrt{\left(\gamma + \sqrt{\Delta}\right)/2\kappa}. \end{aligned}$$

[11]

If $\Delta < 0$, they become complex:

$$\begin{aligned} q_1 &= \sqrt{(\gamma - i\sqrt{-\Delta})/2\kappa} \\ q_2 &= \sqrt{(\gamma + i\sqrt{-\Delta})/2\kappa}. \end{aligned}$$

[12]

For $|z| < 2$, following Abramovitz and Stegun (1), we use

$$K_0(z) = -\left[\log\left(\frac{z}{2}\right) + \gamma_E\right] I_0(z) + z_d + \frac{3}{8}z_d^2 + \frac{11}{216}z_d^3 + \frac{25}{6912}z_d^4 + ...$$

[13]

with $z_d = 1/4z^2$ and $I_0(z) = \sum_{k=0}^{\infty} z_d^k/(k!)^2$, where I_0 is the modified Bessel function of the first kind of order 0. For $|z| > 2$, we use

$$K_0(z) = \sqrt{\frac{\pi}{2z}}e^{-z}\left[1 - \frac{1}{8z} + \frac{9}{2}\frac{1}{(8z)^2} - \frac{75}{2}\frac{1}{(8z)^3} + \frac{600}{(8z)^4} + ...\right].$$

[14]

To calculate the scattered intensity, we insert the correlation function (Eq. **9**) together with the appropriate form factor (Eq. **8**) into the scattering cross-section (Eq. **3**). The sensitivity to the parameters of the model is illustrated in Fig. 1.

Scattering by the ripple phase. To test the effect of possible static height-fluctuations, we calculate the scattering by ripples schematized in Fig. 9 Inset. The Fourier expansion is

$$z(x) = \sum_{p=1}^{\infty} a_{2p+1} \sin\left[\frac{2\pi(2p+1)x}{\Lambda}\right],$$

[15]

with

$$a_{2p+1} = \frac{2\Lambda^2}{(2p+1)^2\pi^2\Lambda_1(\Lambda-\Lambda_1)}\sin\left[(2p+1)\pi\left(1-\frac{\Lambda_1}{\Lambda}\right)\right].$$

[16]

Here, x is a one-dimensional coordinate, and we obtain the height:height correlation function as a two-dimensional average over randomly oriented domains (powder diffraction diagram):

$$\langle z(0)z(r_\parallel)\rangle = \frac{1}{2}\sum_{p=1}^{\infty} a_{2p+1}^2 J_0\left[\frac{2\pi(2p+1)r_\parallel}{\Lambda}\right],$$

[17]

where J_0 is the Bessel function of the first kind of order 0. Inserting this correlation function in Eq. **3**, we can estimate the scattering by a ripple phase.

Although we do not know of publications reporting ripple phase in pure di-C_{18}-PC [DSPC], we can take order-of-magnitudes on related systems: di-C_{14}-PC [DMPC] (2), di-C_{16}-PC [DPPC] (3) and DSPC-DMPC mixtures (4). Typical values correspond to a wavelength Λ in the range of ($\sim\ 10^{-9}$–10^{-7}) m and an amplitude of the order of $0.5-10$ nm. Fig. 9 demonstrates that, on such examples, in this wavelength range our experiment has a sensitive detection level as low as ($0.1-0.2$ nm) in amplitude. A smoother undulation shape, resulting in a higher and narrower peak in Fourier space, would have been even easier to detect. To summarize, unless it had a very small wavelength (of order of 10^{-9}m, or less) (5), a static undulation like a ripple phase would certainly have been detected.

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