

## SUPPORTING INFORMATION

### Ordering Transitions in Nematic Liquid Crystals Induced by Vesicles Captured through Ligand-Receptor Interactions

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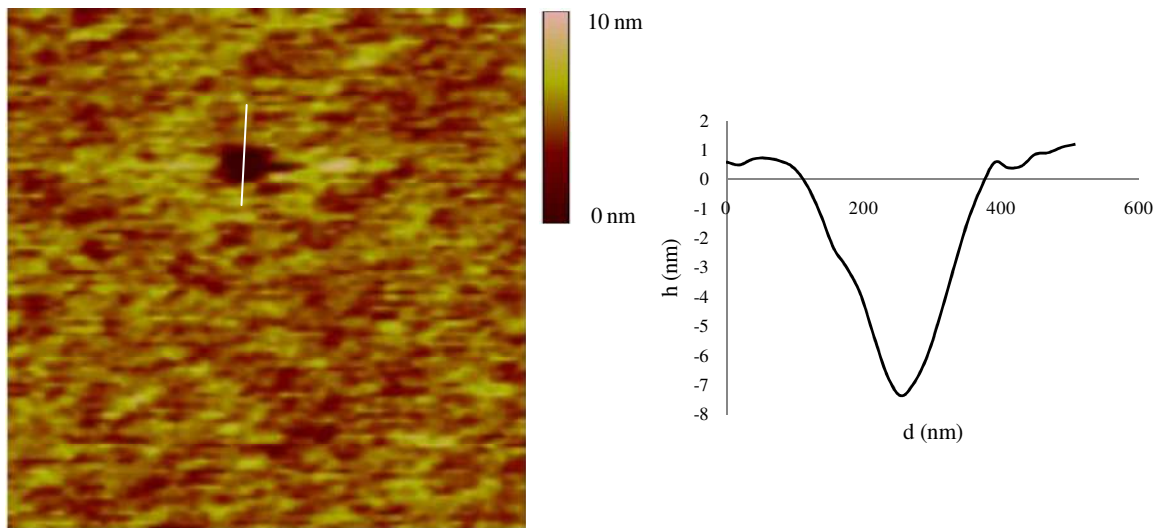


Figure S1. Topographic AFM image (tapping mode) of an avidin-functionalized surface under TBS buffer. The image size is  $2 \times 2 \mu\text{m}$ . The line in the micrograph lies across a region of the sample that was “scratched” using the AFM tip in order to estimate the thickness of the layer on the surface. This depth is in agreement with the ellipsometric thickness of avidin on the surface.

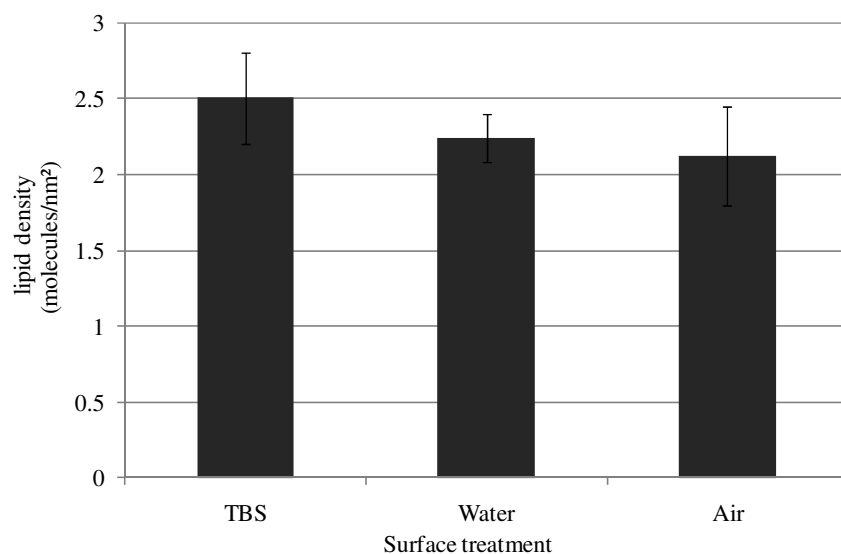


Figure S2. Plot of density of phospholipid captured on avidin-functionalized surfaces incubated with biotinylated vesicles (0.2 mM, 1 mol % biotin-DOPE), after sequential surface treatments, as measured by fluorimetric intensity.

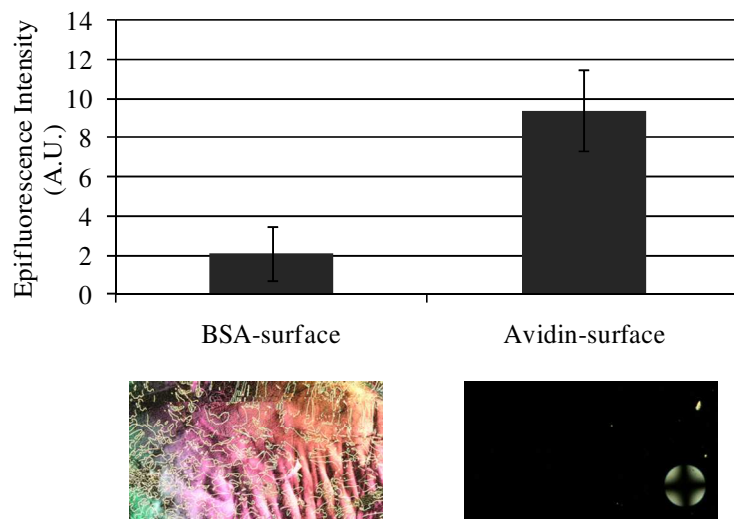


Figure S3. Plot of epifluorescence intensity of functionalized surfaces measured following incubation of biotinylated vesicles. The optical appearances of 5CB in contact with the surfaces are shown below the chart.

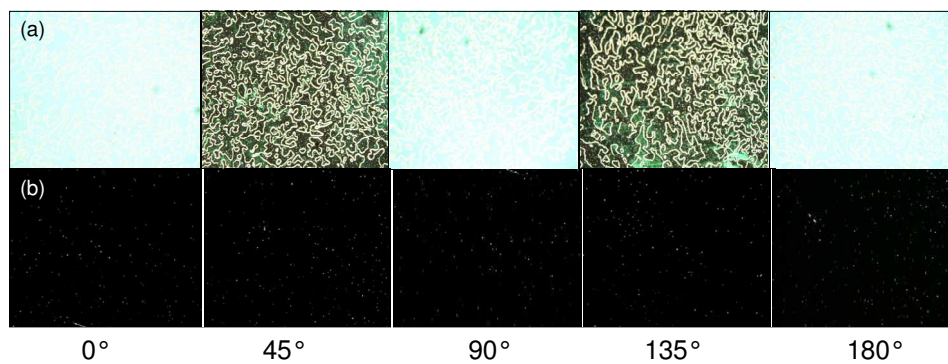


Figure S4. Optical images (crossed polars) of nematic 5CB sandwiched within optical cells with avidin-functionalized surfaces that were incubated against (a) biotin-free and (b) biotinylated vesicles, as a function of the angle of rotation of the cell.

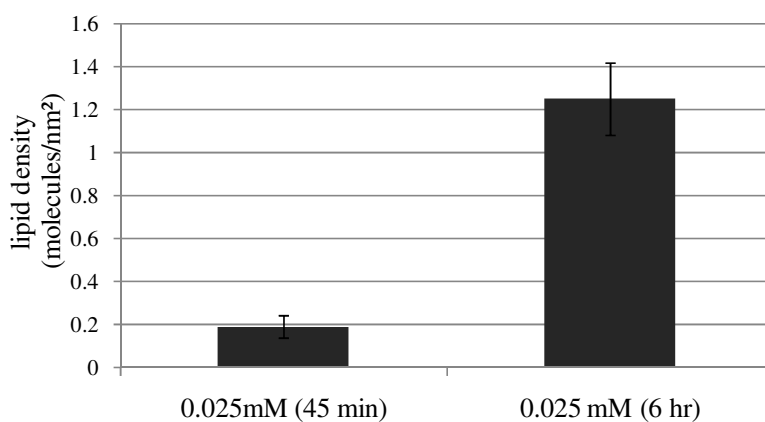


Figure S5. Plot of surface density of phospholipid after incubation with a vesicle solution (1 mol % biotin-DOPE) for different durations, as measured by fluorimetric intensity.

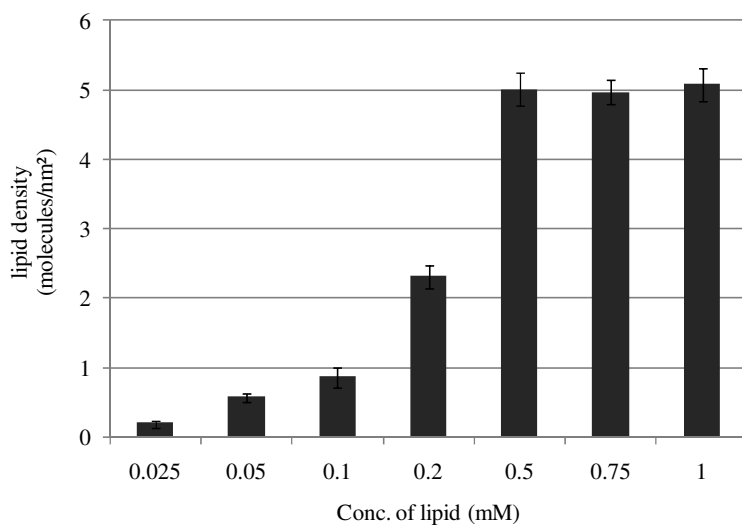


Figure S6. Plot of surface density of phospholipid measured following incubation of avidin-functionalized surface against solutions of biotinylated vesicles (1 mol % biotin-DOPE), as a function of phospholipid concentration, as measured by fluorimetric intensity.

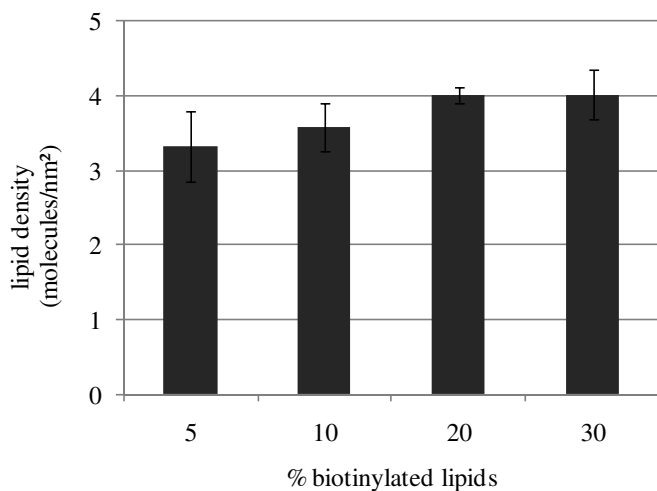


Figure S7. Plot of surface density of phospholipid measured following incubation with solutions of vesicles (0.1 mM total phospholipid concentration), as a function of mole fraction of biotinylated lipid in the vesicles, as measured by fluorimetric intensity.

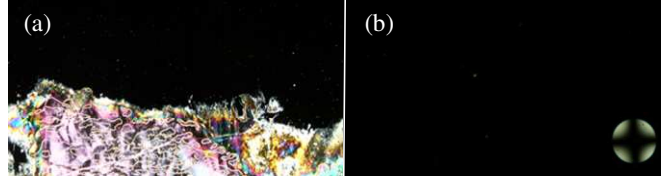


Figure S8. Optical images (crossed polars) of nematic 5CB sandwiched in optical cells with avidin-functionalized surfaces which were incubated against biotinylated vesicles (1 mol % biotin-DOPE) with total lipid concentration of (a) 0.025 mM of lipid and (b) 0.1 mM of lipid for 6 h. The surfaces were measured to possess surface densities of (a) 1.2 phospholipid molecules/nm<sup>2</sup> and (b) 1.8 phospholipid molecules/nm<sup>2</sup>.

#### Diffusion Coefficient of Vesicles

$$D = \frac{k_B T}{6\pi\eta r} = \frac{1.38 \cdot 10^{-23} \text{ kg s}^{-2} \text{ K}^{-1} \cdot 298 \text{ K}}{6\pi \cdot 8.94 \cdot 10^{-4} \text{ kg s}^{-1} \text{ m}^{-1} \cdot 65 \cdot 10^{-9} \text{ m}} = 3.76 \cdot 10^{-12} \text{ m}^2/\text{s}$$

#### Calculation of osmotic pressure exerted on vesicles

To achieve mechanical equilibrium, the osmotic pressure difference across the interface must to be balanced by the membrane tension.

$$\Delta P \cdot \pi r^2 = \sigma \cdot 2\pi r t$$

Where  $\Delta P$  is the pressure difference,  $\sigma$  is the stress and  $r$  and  $t$  are the radius and thickness of the membrane vesicles respectively. The stress can be expressed as

$$\sigma = k \cdot \alpha$$

Where  $k$  is the Young's stretching modulus and  $\alpha$  is the strain ( $\alpha = \frac{\Delta A}{A}$ ).  $k$  is related to  $K$ , the area compressibility elastic modulus, by the following relation

$$K = k \cdot t$$

Combining the above equations, we get

$$\Delta P = \frac{2K\alpha}{r}$$

The rupture tension  $\tau_c$  is related to  $K$  by

$$\tau_c = K \cdot \alpha_c$$

Measured osmotic pressure of 50 mM Tris buffer was found to be  $2.6 \cdot 10^5 \text{ N/m}^2$ .<sup>1</sup> Therefore, the DOPC membrane tension generated at this osmotic pressure is

$$\tau_c = \frac{\Delta P \cdot r}{2} \approx 8.5 \text{ mN/m}$$

The membrane lysis tension of DOPC membrane is reported as  $9.9 \text{ mN/m}$ .<sup>2</sup> Hence, the osmotic pressure across the membrane can exert a sufficiently high tension to induce rupturing.

Calculation of surface density in hexagonal close packing, where the packing density is 0.907.

$$\text{Area occupied per vesicle} = \pi r^2 \cdot \frac{1}{0.907} \approx 1.5 \cdot 10^{-14} \text{ m}^2$$

Assuming that each phospholipid molecule occupies an area of  $0.70 \text{ nm}^2$ ,

$$\text{Number of phospholipid molecules per vesicle} = \frac{8\pi r^2}{0.7} \approx 151000$$

$$\text{Surface density for hexagonal close packed} = \frac{151000}{1.5 \cdot 10^{-14} m^2} \approx 10.1 \text{ molecules/nm}^2$$

Interpretation of LC ordering transition on surfaces incubated with vesicles of varying mole fraction of biotinylated lipid

Low composition of 0.1 mol % biotinylated phospholipids in vesicles led to a low interfacial concentration of ~0.3 phospholipid molecules/nm<sup>2</sup>, which gave planar anchoring of 5CB. This is supported by the birefringence value of the 5CB film in contact with this surface (0.099 ± 0.01). In contrast, vesicles with high biotin loading at 5 mol% gave rise to a uniform homeotropic anchoring due to the relatively high interfacial concentration of ~3.3 phospholipid molecules/nm<sup>2</sup>. Incubation against vesicles of intermediate biotin loading at 1 mol % gave a mixture of planar and homeotropic orientations. The trend and discontinuous transition observed in Figure 6(d) is similar to that shown previously.

References:

- (1) Patton, J. N.; Palmer, A. F. *Biomacromolecules* **2005**, *6*, 414-424.
- (2) Olbrich, K.; Rawicz, W.; Needham, D.; Evans, E. *Biophysical Journal* **2000**, *79*, 321-327.