Phospholipid monolayers form a plethora of liquid-crystalline phases, in which the molecules pack into hexagonal and pseudo-hexagonal lattices, and the tail groups tilt to accommodate the mismatch in projected area between the headgroup and the close-packed chains (1). Complicating this packing, natural lipids such as 1,2-dipalmitoyl-sn-glycero-3-sn-phosphocholine (r-DPPC) have an exclusively r-enantiomer chiral carbon. This induces a chiral orientational ordering in liquid-condensed (LC) domains that persists over tens of microns (Fig. 1). The chiral twist demands that r-DPPC, and hence the chain tilt, rotate from the domain center to the periphery, which is incompatible with a regular lattice (2). One solution is to localize this required twist to defects. In r-DPPC monolayers, this frustration between tilt and twist leads to “tilt gradient lines” across which tilt rapidly changes orientation, (yellow dotted lines in Fig. 1a). These discontinuous changes in tilt direction allow the chiral precession, while maintaining a constant nearest-neighbor-directed tilt orientation. We combine surface pressure-area isotherms (Fig. 1b) with fluorescence imaging to visualize the monolayer organization. For DPPC, the domains spiral counter-clockwise, which is a mesoscopic manifestation of molecular chirality. We have found that shearing these domains using a microbutton rheometer gives a different viscoelastic response depending on the rotation direction. Fig. 1c shows that domains (e.g. highlighted green example) are compacted when microbutton is CC-torqued (C-shear), but extended when C-torqued (CC-shear). O represents monolayer morphology before shear. The limiting rotational strain, $\Delta \theta$ is greater for the same ~ 440 nN/m torque applied in the C direction than the CC direction, reflecting the chirality of the rheology at the 100 µm length scale in response to molecular chirality (2).
As additional components are added to DPPC (3, 4), the monolayer morphology changes in interesting ways. In Figure 2, we show the domain structure of a 3:1 mixture of DPPC with hexadecanol, which is similar to the composition of the clinical lung surfactant Exosurf. To this, we add 1.5 mol% cholesterol, which is representative of the composition of the lung surfactant Infasurf, and to native lung surfactant. At low surface pressure, the domains lose circular symmetry, suggesting molecular orientation within the domains is heterogeneous. As the surface pressure increases, small dendritic fingers grow out of the domains, suggestive of a changing chemical composition. The domains are chiral as for pure DPPC. These ~ 50 µm domains are of the same order of magnitude as the radii of the alveoli in the lung. The coupling of interfacial curvature to monolayer morphology and dynamics is unexplored and is the focus of this research.

Fig. 3 shows how we measure the shear viscoelasticity of multiphase, multicomponent monolayers. A 25 µm nickel covered microbutton (bottom right) is torqued by controlled electromagnets to oscillate at a controlled torque and frequency in the monolayer film (6). By analyzing the amplitude and phase lag of the microbutton, the real and imaginary parts of the viscoelastic response can be determined. The microbutton size maximizes the Boussinesq number, $Bo = \eta_s P / \eta A$, in which $\eta_s$ is the surface viscosity, $\eta$ is the bulk fluid viscosity, $P$ is the perimeter and $A$ is the surface area of the microbutton probe. For large Bo, the surface viscosity dominates the drag on the microbutton and allows for accurate measurement of the surface viscoelasticity; the smaller the microbutton, the more sensitive the microbutton rheometer is to the surface viscosity (4).


