Supporting Information

MECHANICAL PROPERTIES OF DPPC-POPE MIXED LANGMUIR MONOLAYERS

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Figure S1. Isotherms of POPC monolayers for different times after spreading on the air-water interface.



Figure S2. BAM images obtained at the indicated surface pressures for different DPPC molar fractions; the horizontal length of each image corresponds to 292 μm.



Figure S3. Surface pressure at which the compressibility modulus E of each monolayer is discontinuous (IIk, circles) and surface pressure at the onset of the liquid expanded phase as observed with BAM (IIe, triangles). The grey line is a linear fit of the experimental data.



Figure S4. Expanded area to condensed area ratio as a function of the molar fraction of DPPC at $\Pi = 15$, 25, 30 and 35 mN/m. The solid line represents the theoretical prediction according to the lever rule (equation 4) using the parameters provided by the isotherms and indicated in the figure.



Figure S5. Gibbs excess free energy for different surface pressures as a function of the monolayer composition.

IMAGE ANALYSIS TO DETERMINE THE DOMAIN AREA FROM BAM IMAGES

We used ImageJ software,^{1,2} for the analysis of the BAM images in order to obtain the values of the condensed area. We first applied a band pass filter to improve the uniformity of the illumination over the images. The gray level is set to a limit value in order to visualize all the domains. The higher gray levels are considered to be black (LE phase)... The images are then converted to binary images with white LC phase. These binary images were used to evaluate the fraction of condensed phase area. Figure S6 show examples with images taken in different monolayers for $\chi_{DPPC} = 0.90$ and a surface pressure *ca*. 15 mN/m.



Figure S6. BAM images near the percolation threshold (left pictures), and analysis using ImageJ (right pictures). Thea condensed area fraction x is 0.85 ± 0.017 ; the standard deviation is estimated from 3 to 5 analyzed images for monolayers at the same pressure and the same χ_{DPPC} .

The percolation threshold was determined as the moment when most of the domains are in contact. Then the condensed area was taken from those images (Figure S7).



Figure S7. Untreated BAM images from monolayers in a non-percolation regime (left) and a percolated monolayer (right).

References

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- (2) Schneider, C. A.; Rasband, W. S.; Eliceiri, K. W. NIH Image to ImageJ: 25 Years of Image Analysis. *Nat. Methods* **2012**, *9* (7), 671.