# Supporting Information

## Condensing Effect of Palmitic Acid on DPPC in Mixed Langmuir Monolayers

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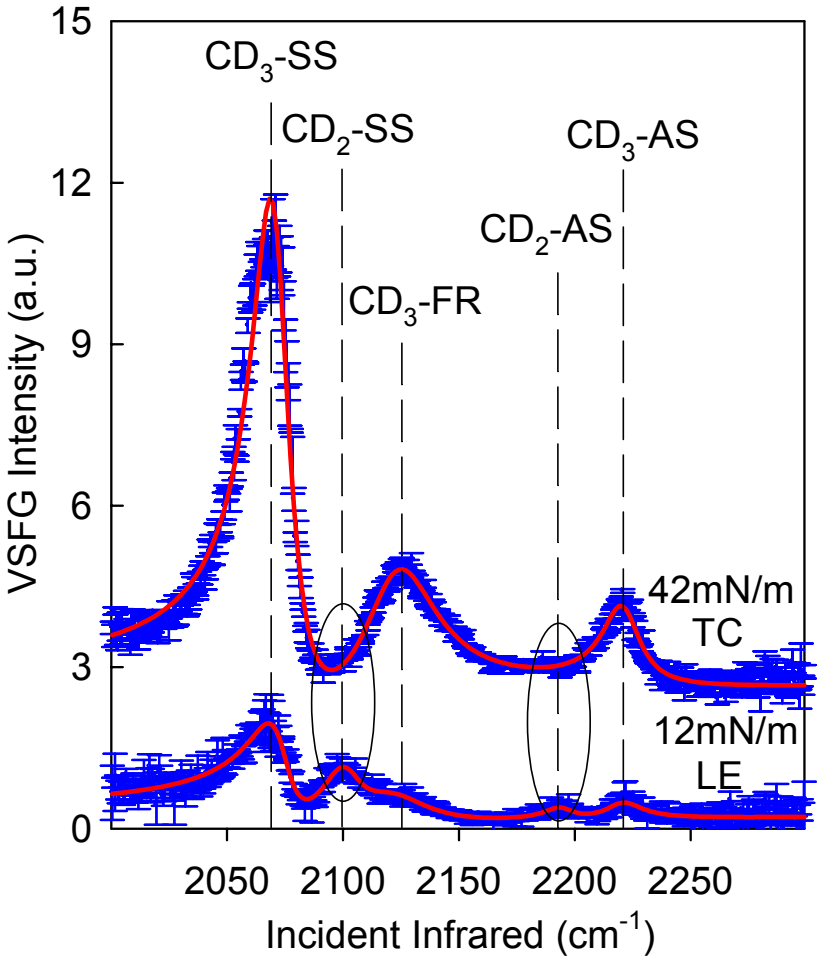


**Figure S1.** The ssp VSFG spectra (with error bars, ± standard deviation) of the DPPC-*d62* monolayer in the C-D stretching region at different surface pressures. Solid red curves are spectral fits. Dashed vertical lines reveal the spectral assignments: SS, symmetric stretch; FR, Fermi resonance; AS, asymmetric stretch. The spectral regions within the ellipses show the presence and absence of $CD_2$ stretches.

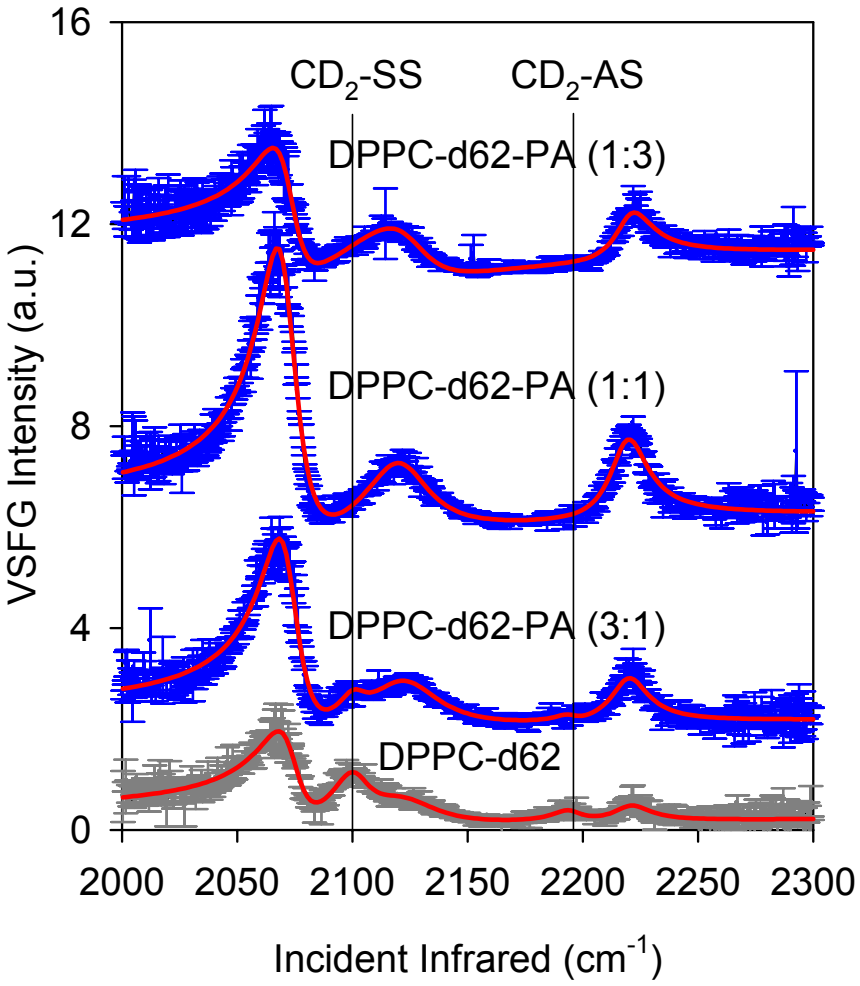


**Figure S2.** Surface pressure at 12 mN/m: ssp VSFG spectra (with error bars, ± standard deviation) of DPPC-*d62*–PA mixed monolayers in the C-D stretching region with three different molar ratios of DPPC-*d62* to PA. Solid red curves are spectral fits. Vertical lines indicate peak positions of the $CD_2$-SS and the $CD_2$-AS. Gray data points are the VSFG data of DPPC-*d62* at 12 mN/m.

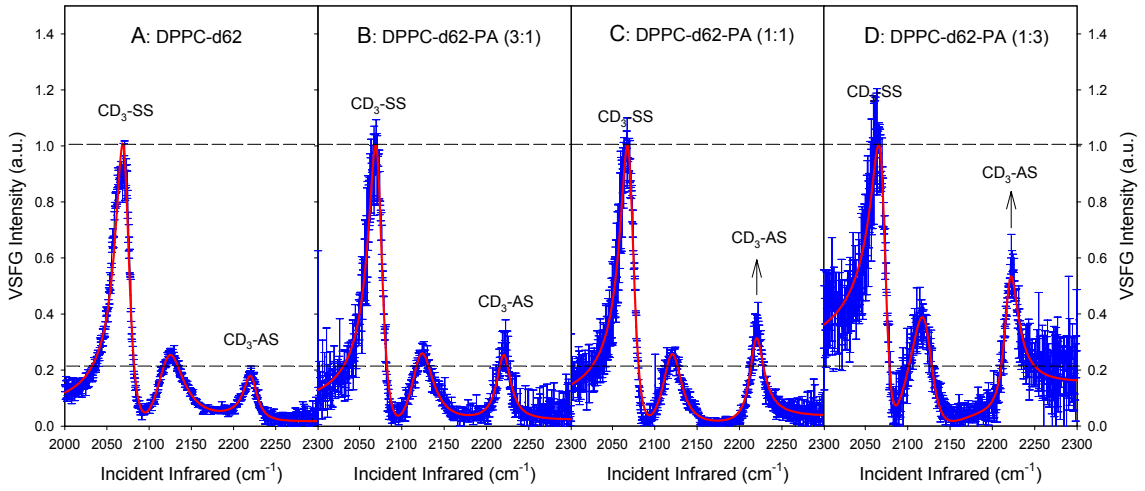


**Figure S3.** Surface pressure at 42 mN/m: ssp VSFG spectra (with error bars, ± standard deviation) of DPPC-*d62*–PA mixed monolayers in the C-D stretching region with three different molar ratios of DPPC-*d62* to PA. Solid red curves are spectral fits. Dashed horizontal lines demonstrate the variation of the relative peak height ratio between $CD_3$-SS and $CD_3$-AS. (Spectra are scaled so that the $CD_3$-SS peaks have an intensity of unity. The seemingly significant baseline increase and error bar increases in Figure D are due to spectral rescaling.)

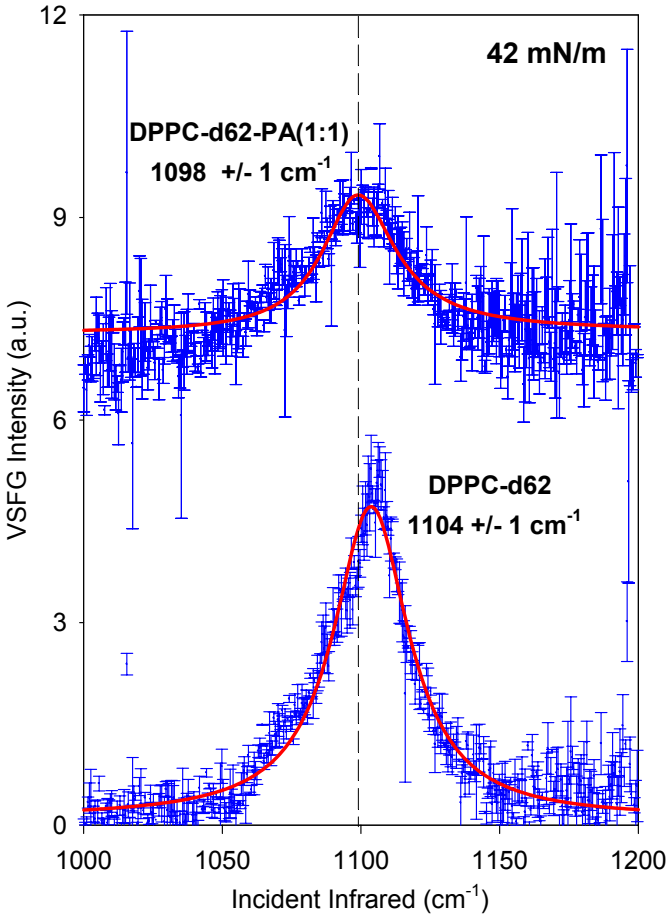


**Figure S4.** Surface pressure of 42 mN/m: ssp VSFG spectra (with error bars, ± standard deviation) in the $PO_2^-$ symmetric stretching region of DPPC-*d62*–PA mixed monolayer (1:1 molar ratio) and DPPC-*d62*. Solid red curves are spectral fits. Dashed vertical lines indicate the frequency red-shift.

**TABLE S1.** At 42 mN/m, chain tilt angle ($\alpha$) calculated based on different literature values of $r$ ($\beta_{aac}$ / $\beta_{ccc}$) and $\beta_{caa}/\beta_{aac}$ from the VSFG spectra of DPPC-*d62* and DPPC-*d62*–PA mixtures with different molar ratios.

| DPPC-*d62* and DPPC-*d62*–PA Mixtures | $\alpha$ ($r = 2.3$[1]; $\beta_{caa}/\beta_{aac} = 4.2$[2,3]) | $\alpha$ ($r = 3.4$[4]; $\beta_{caa}/\beta_{aac} = 1$[4]) |
|---|---|---|
| DPPC-*d62* | 25° ± 1° | 9° ± 1° |
| DPPC-*d62*–PA (3:1) | 24° ± 1° | 4° ± 3° |
| DPPC-*d62*–PA (1:1) | 22° ± 1° | 2.5° ± 1° |
| DPPC-*d62*–PA (1:3) | 21° ± 1° | 2° ± 2.5° |

* $\alpha = 41.5° - \theta$. (3:1), (1:1) and (1:3) are molar ratios.


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