

## Supporting Information

### Surface Pressure-Induced Inter-diffused Structure Evidenced by Neutron Reflectometry in Cellulose Acetate /Polybutadiene Langmuir films

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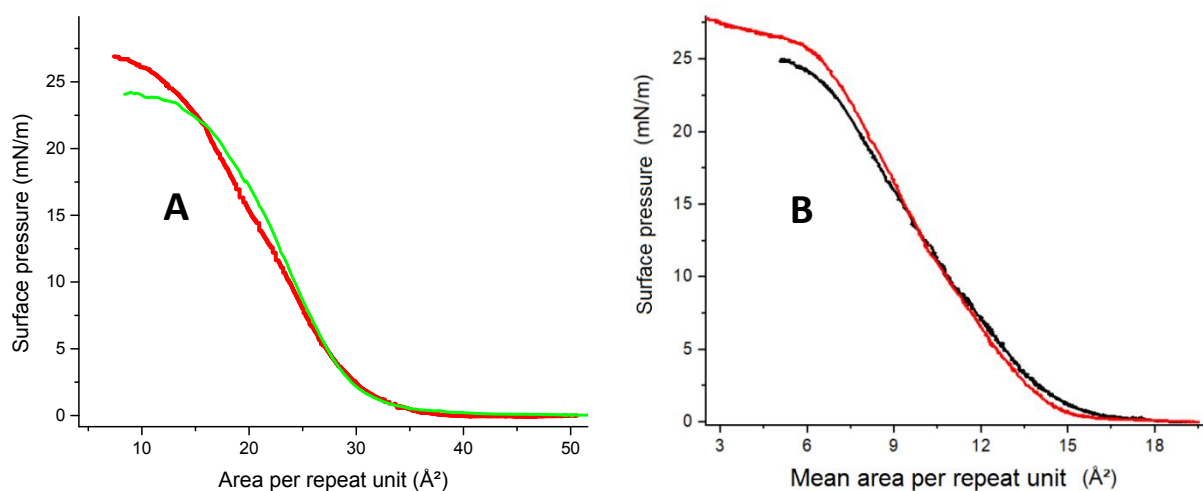
<sup>‡</sup> Synchrotron SOLEIL

<sup>&</sup> Laboratoire Léon Brillouin

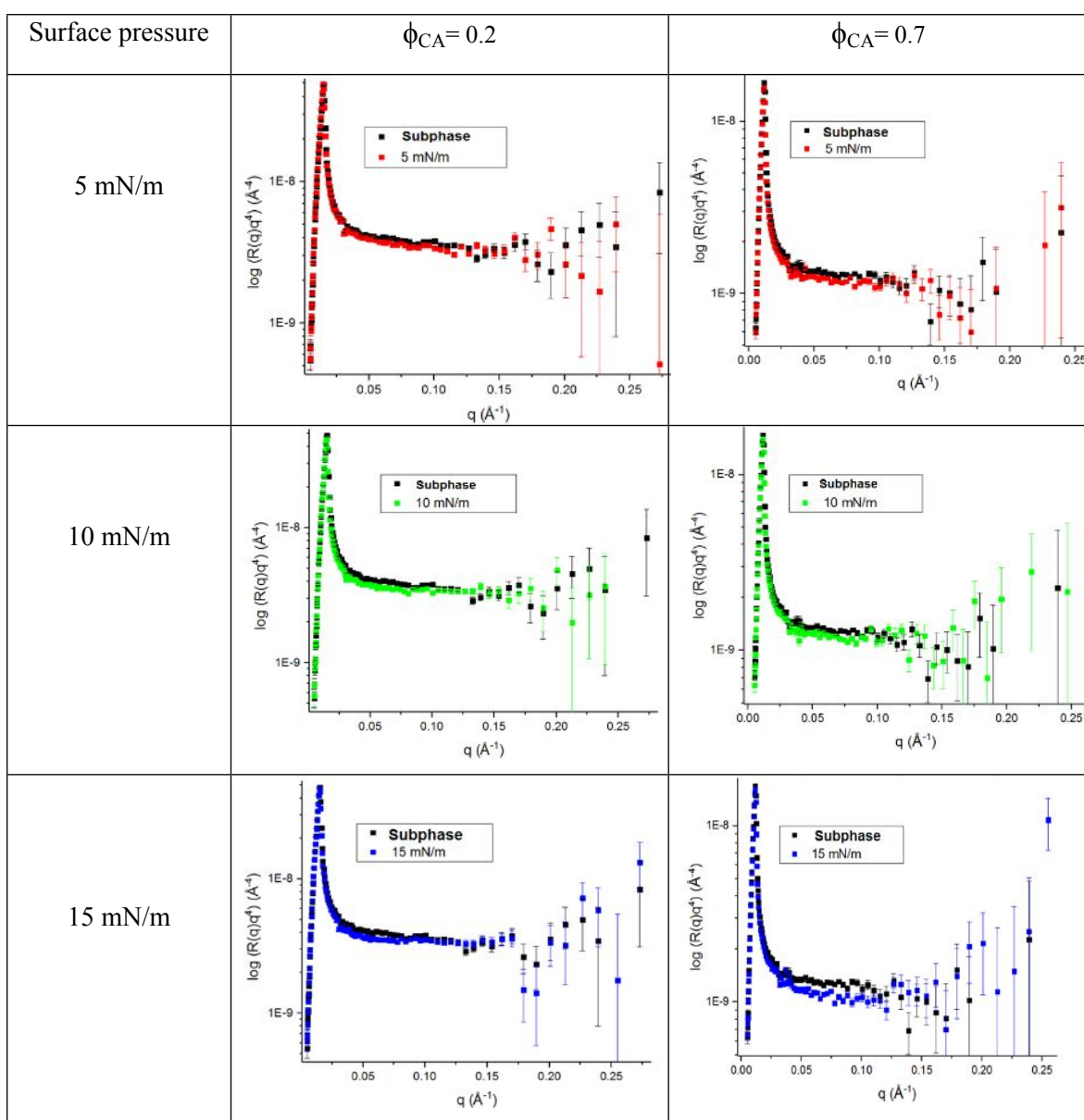
<sup>‡</sup> Institut Laue-Langevin

<sup>‡</sup> Institut des NanoSciences de Paris

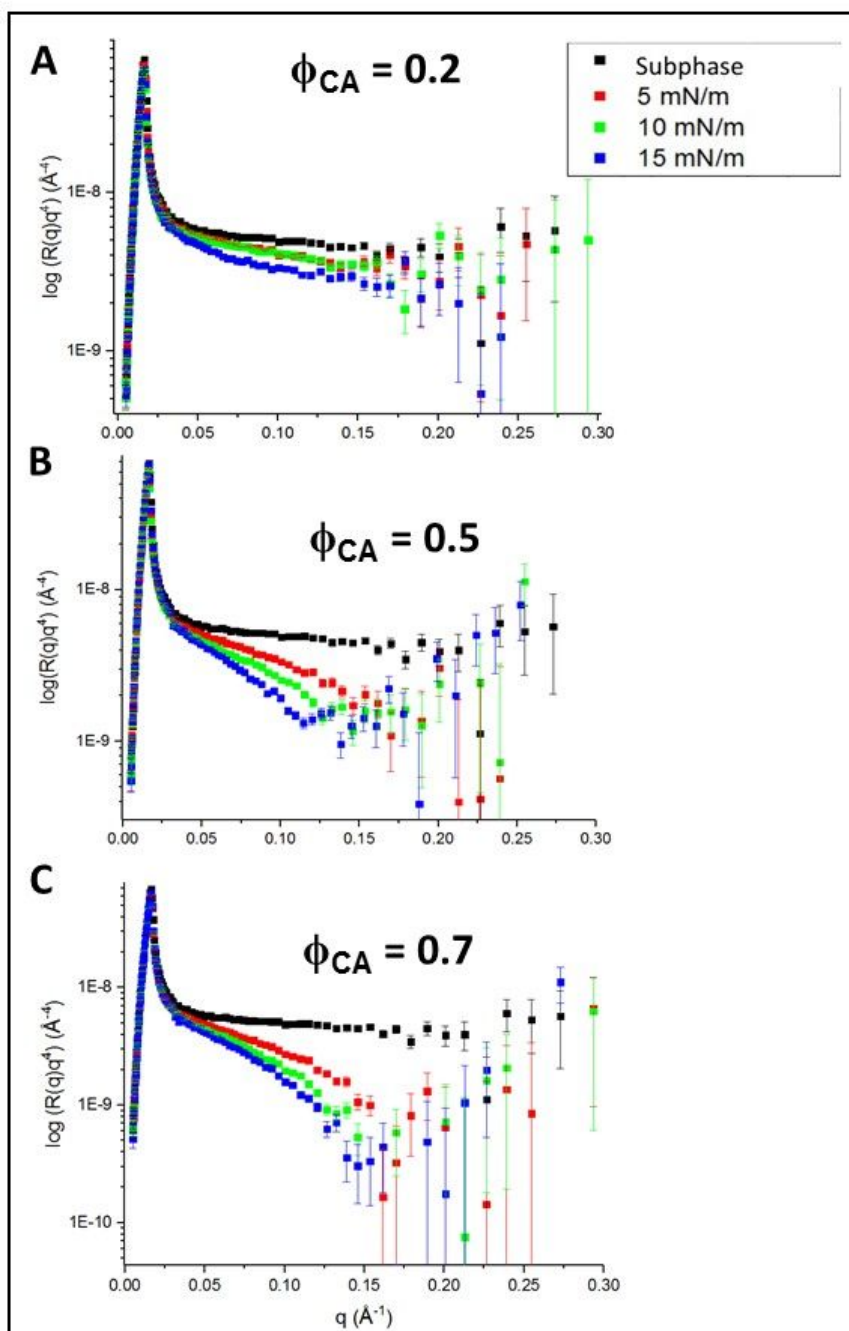
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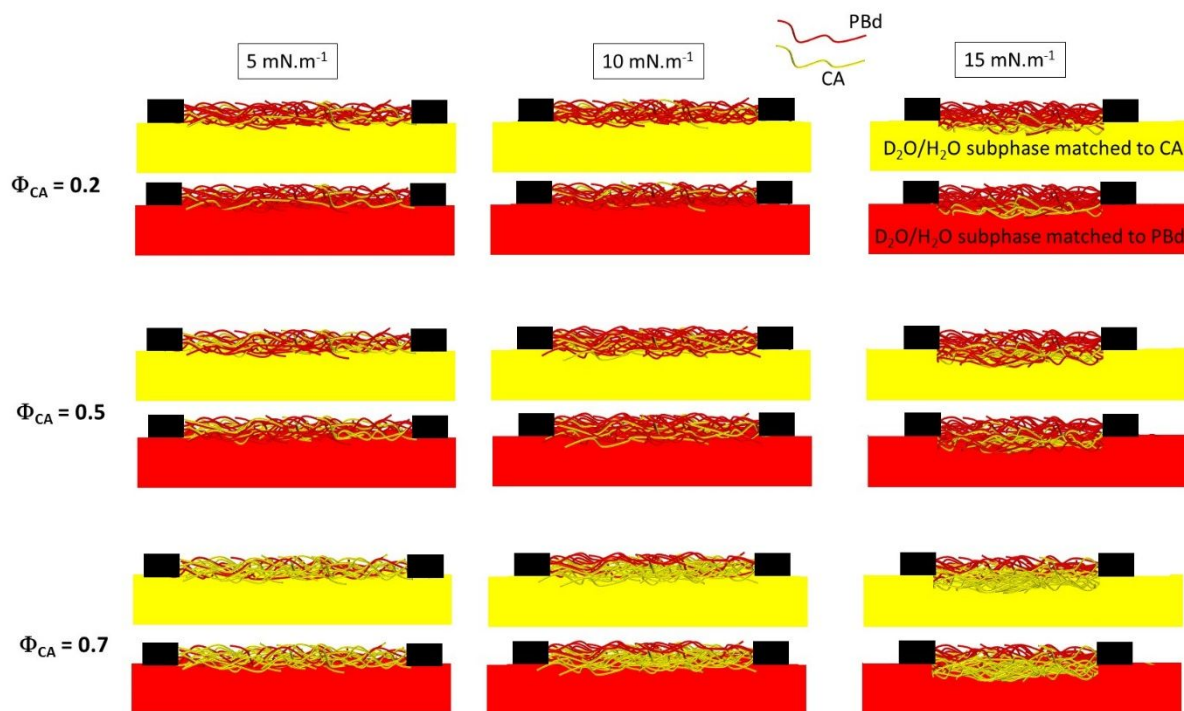
**Figure S1.** (A) Compression isotherms of CA monolayers over pure water. The two curves illustrate the reproducibility of the isotherms. (B) Compression isotherms of hydrogenated (black curve) and deuterated (red curve) PB monolayers at the air-water interface.



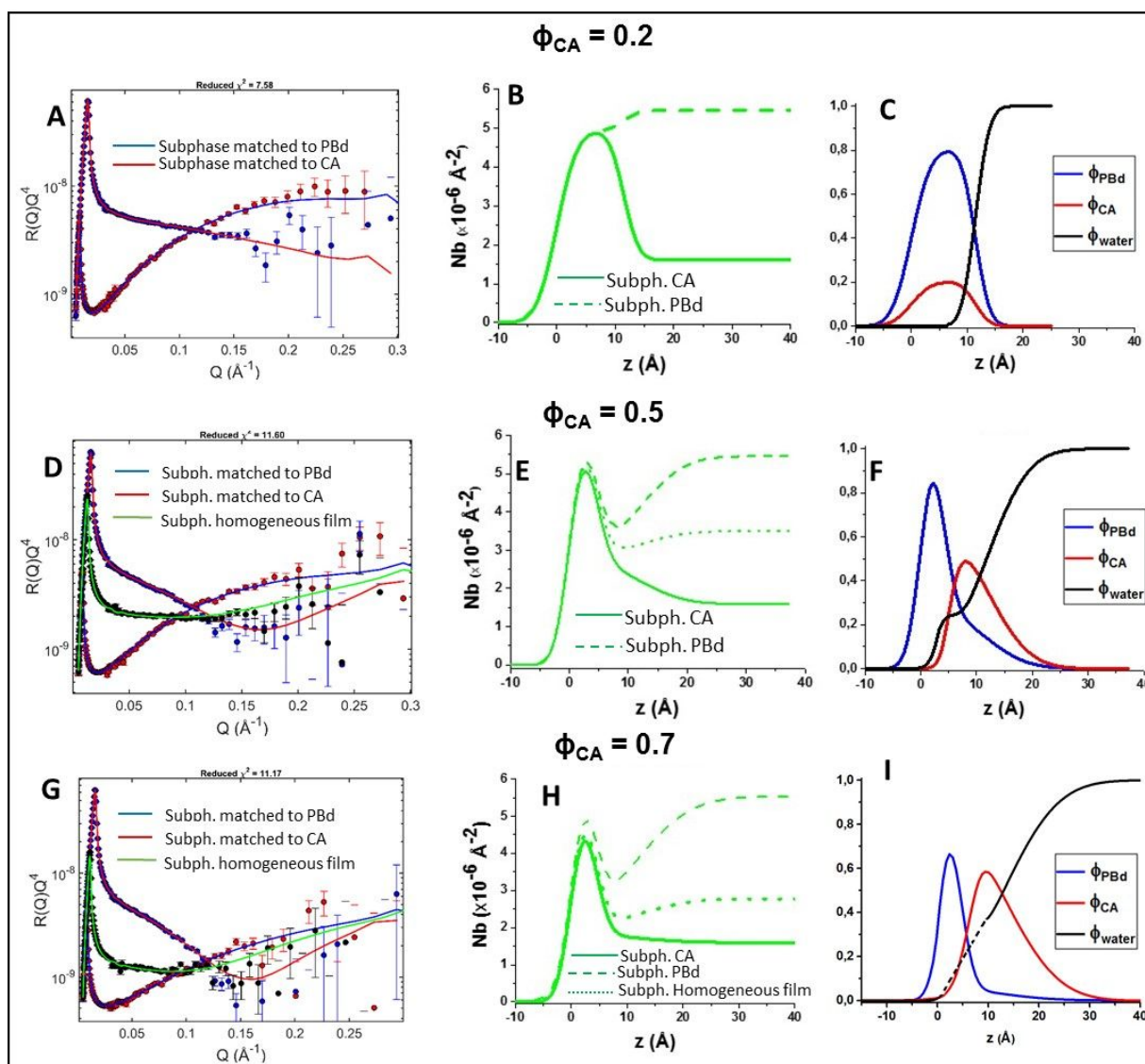
**Figure S2.** Neutron reflectivity curves for  $\phi_{CA} = 0.2$  and  $0.7$  CA/PBd Langmuir films at 5, 10 and 15 mN/m on  $D_2O/H_2O$  subphases matched to the homogeneous film. The subphase reflectivity is presented as a reference (black curves).



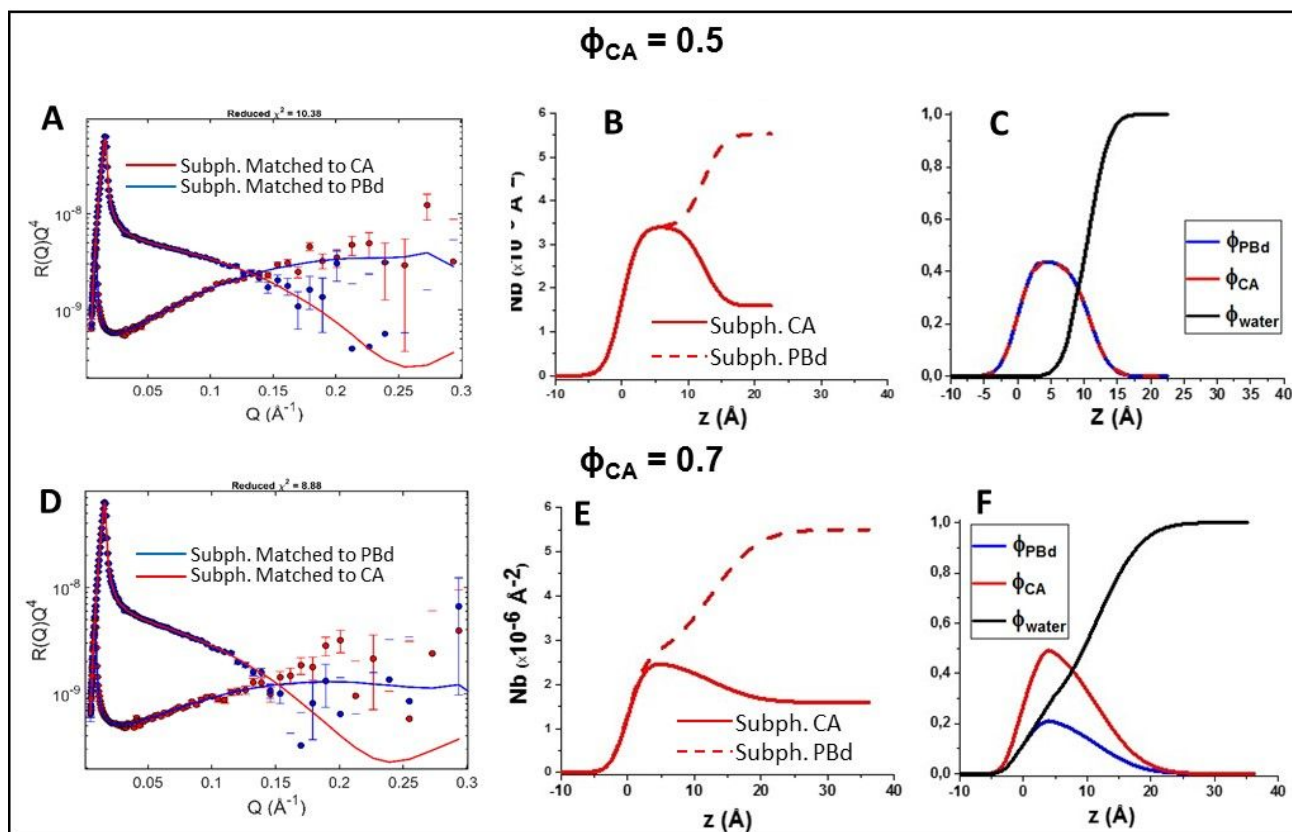
**Figure S3.** Neutron reflectivity curves for  $\phi_{CA} = 0.2$  (A), 0.5 (B) and 0.7 (C) CA/PBd Langmuir films on  $D_2O/H_2O$  subphases allowing contrast matched to PBd at 5, 10 and 15 mN/m. The subphase reflectivity curve is presented as a reference.



**Figure S4.** Schemes of the vertical structure of  $\phi_{CA} = 0.2, 0.5$  and  $0.7$  Langmuir films at 5, 10 and 15 mN/m, showing the strategy followed in neutron reflectivity measurements using contrast variation method through adequate isotopic substitution. Structures shown: red and yellow polymer chains correspond to PBd and CA, respectively. The water subphase is consisting of D<sub>2</sub>O/H<sub>2</sub>O mixtures allowing contrast-matching to either the PBd (red subphase) or the CA (yellow subphase).



**Figure S5.** Fit of the reflectivity curves at 10 mN/m using simultaneously two (for  $\phi_{CA}=0.2$ , A) or three subphases (for  $\phi_{CA}=0.5$  and  $0.7$ ; D and G respectively) allowing contrast matched to CA, homogeneous film or PBd. SLD profiles as a function of depth and CA, PBd and water volume fractions depth profiles at 10 mN/m for  $\phi_{CA} = 0.2$  (B and C),  $0.5$  (E and F) and  $0.7$  (H and I) CA/PBd Langmuir films. The water volume fraction profile in the graph I corresponding to the  $\phi_{CA} = 0.7$  film at 10 mN/m was difficult to be precisely determined near the air-layer interface, thus it has been drawn in dashed lines.



**Figure S6.** Fit of the reflectivity curves at 5 mN/m using simultaneously two subphases allowing contrast matched to CA and PBd for  $\phi_{CA}=0.5$  (A), 0.7 (D) CA/PBd mixed films. SLD profiles as a function of depth and CA, PBd and water volume fractions depth profiles for  $\phi_{CA} = 0.5$  (B and C) and 0.7 (E and F).