

# Supporting Information: Creating gradients of amyloid fibrils from the liquid-liquid interface.

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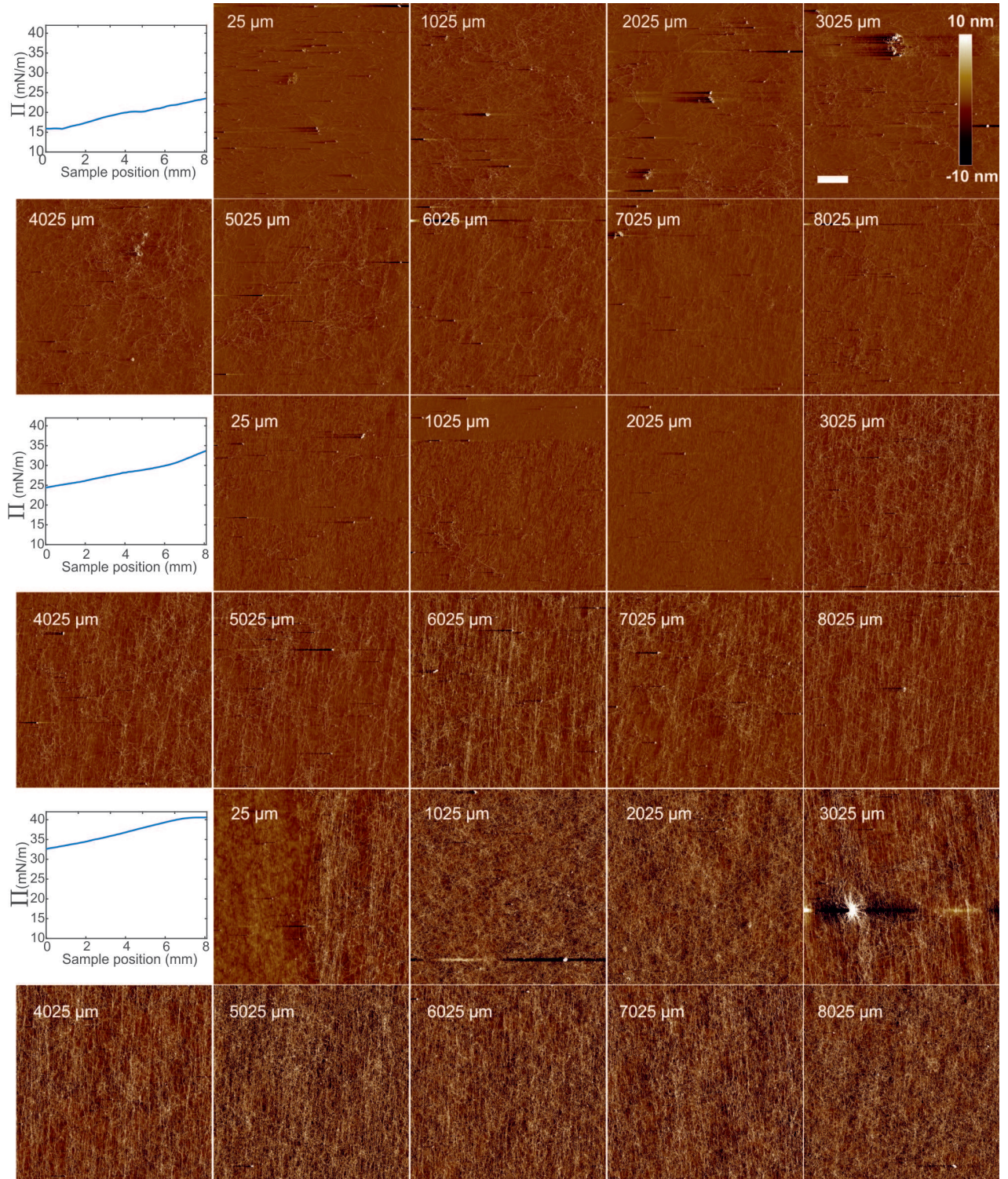


Figure S1: Complete AFM sets of the three samples displayed in main text, with their equivalent compression curves. Scale bar 5  $\mu\text{m}$ .

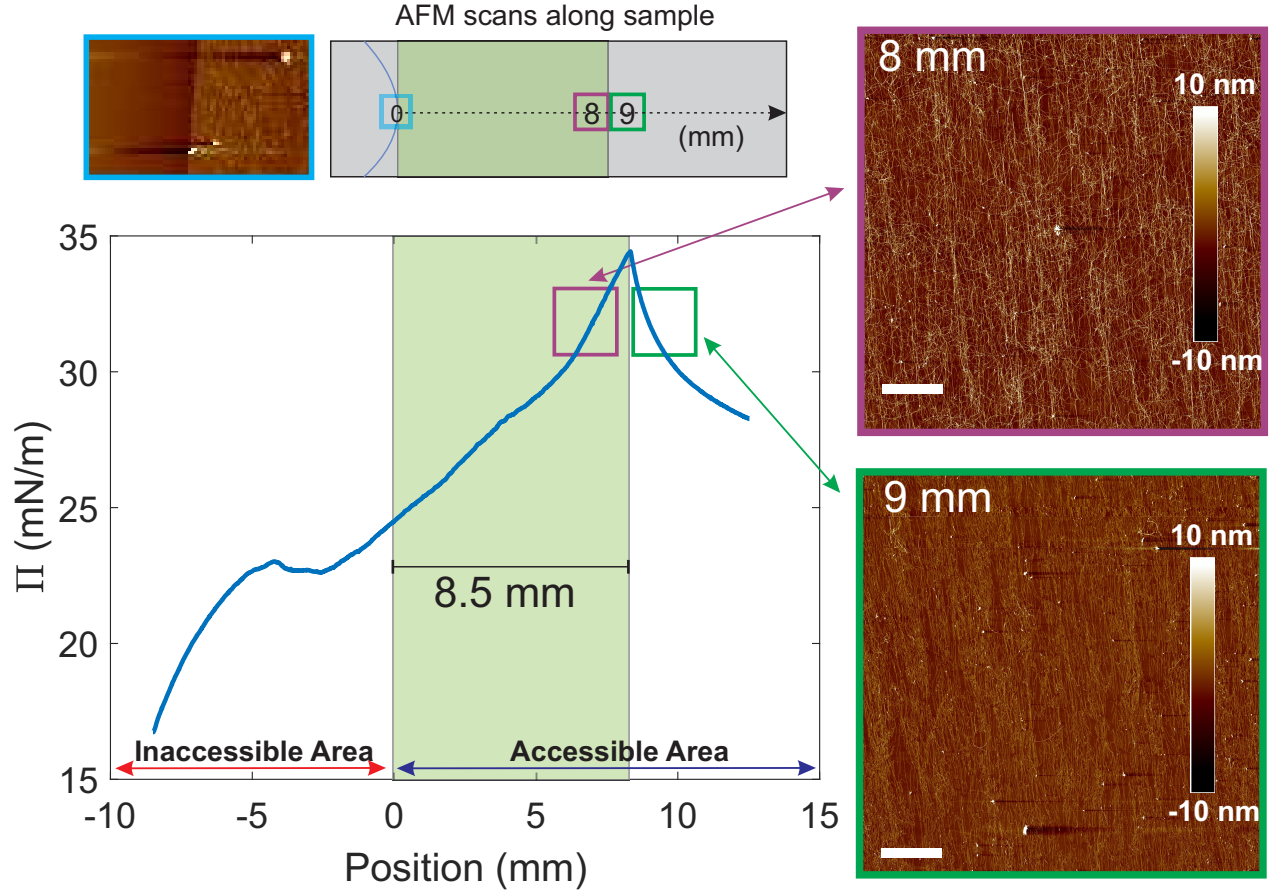


Figure S2: Schematic representing the methodology applied to determine the region of the compression curves corresponding to the AFM images. We create a reference point in the pressure curve, by suddenly opening the barriers of the Langmuir trough, forcing a drop in the surface pressure. Having placed a part of the substrate above the interface, we can observe by AFM the edge of the meniscus (blue box). We set this as position zero. From this point, an AFM scan is taken every millimeter and, when a sudden change in the fibril density is observed, the position can be correlated with the drop of surface pressure. The inaccessible area due to pinning can thus be identified. The region of interest, is indicated by the green box. This method is accurate to an order of  $\pm 0.5$  mm. Scale bar of the AFM images 5  $\mu\text{m}$ .

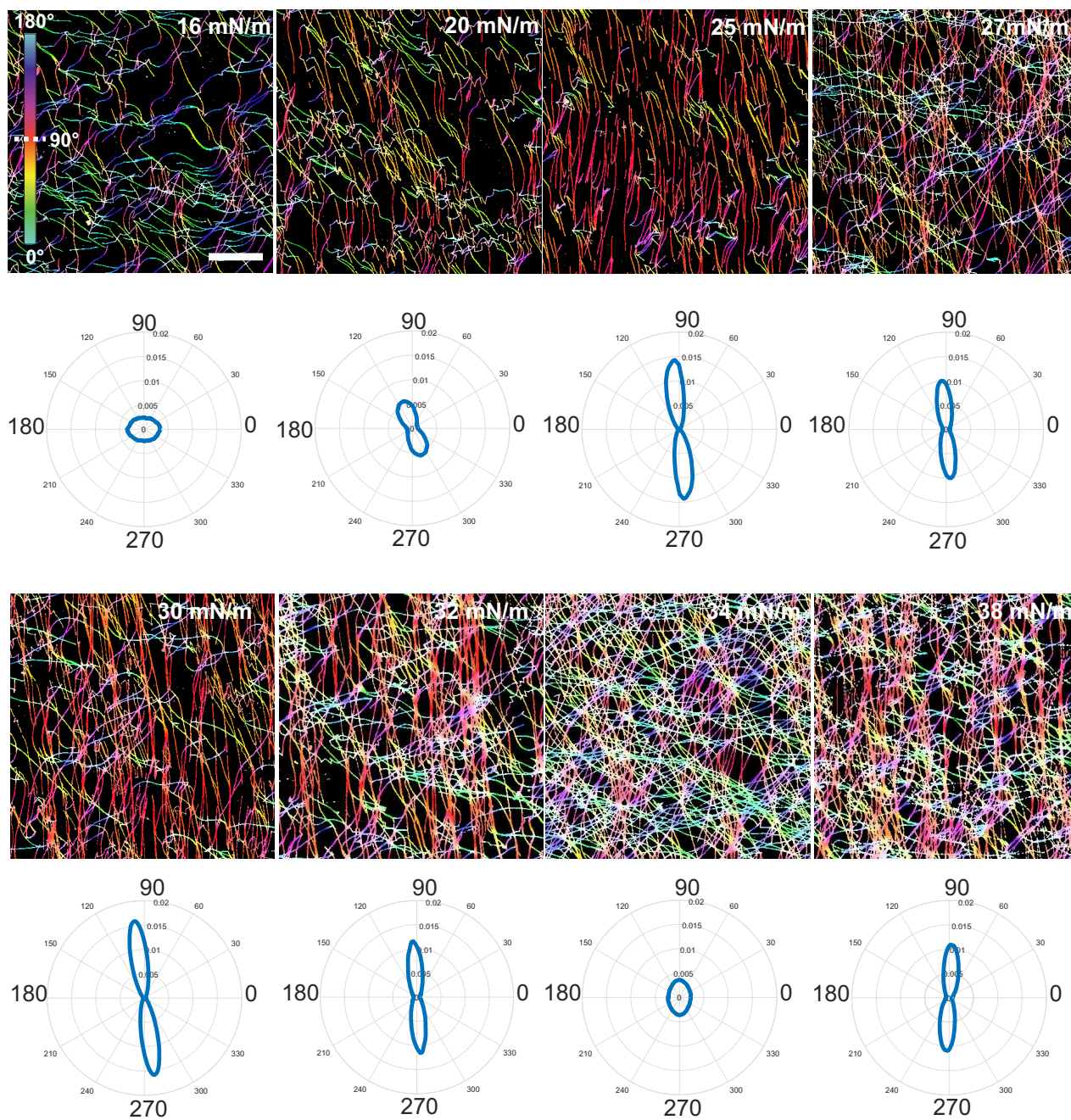


Figure S3: Further examples of the orientation distribution according to sample pressure. Scale bar 1  $\mu\text{m}$ .

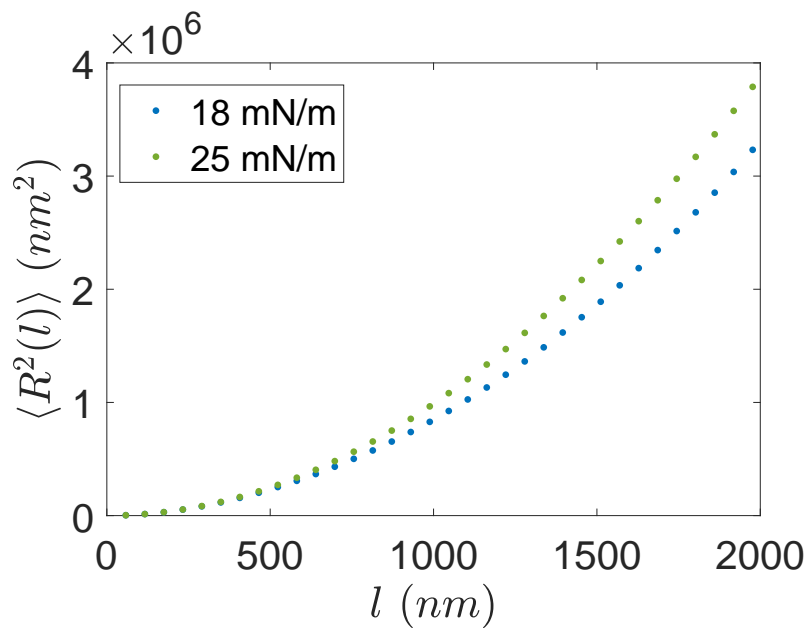


Figure S4: Mean-squared end-to-end distance of the fibrils represented in Figure 3 of main text, which increases with the pressure.

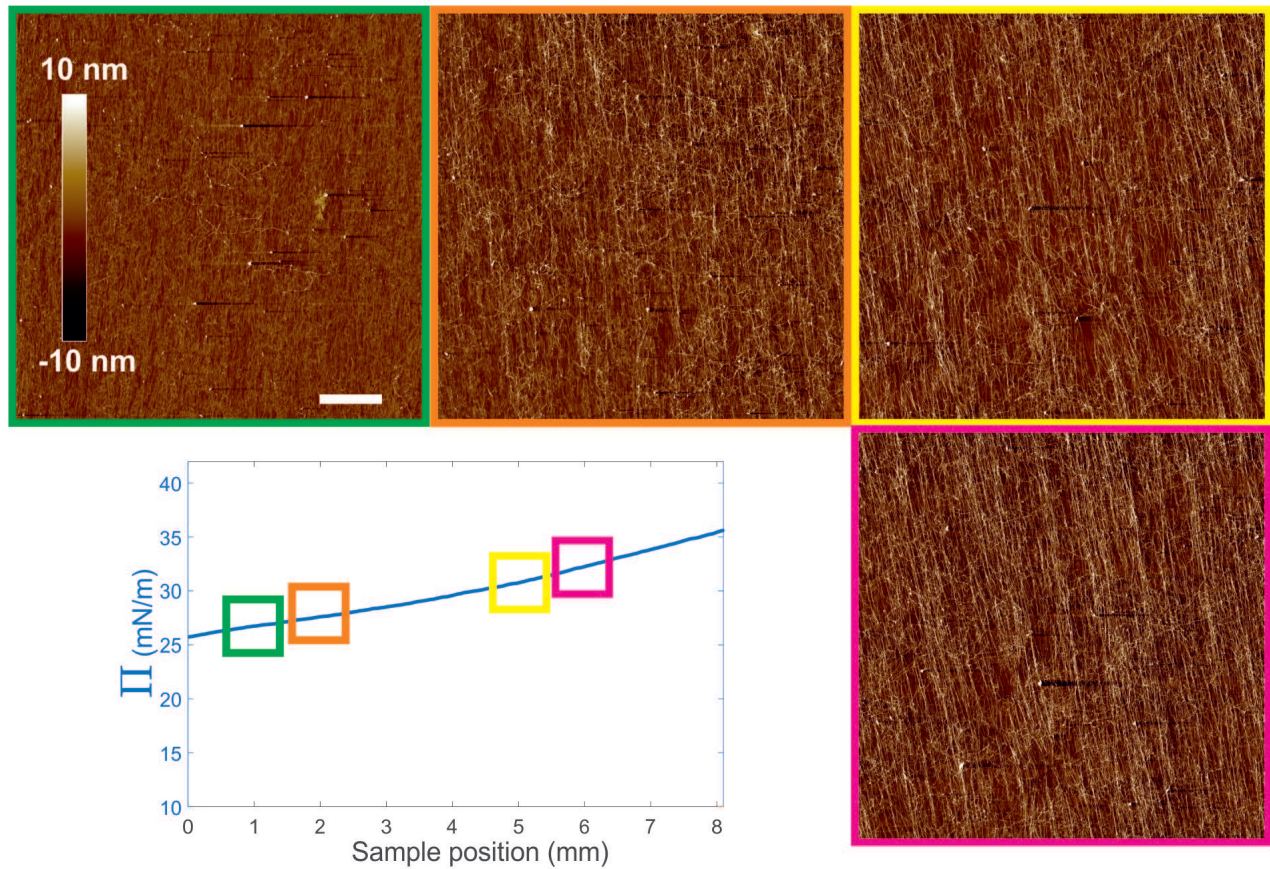


Figure S5: Supporting pressure curve and AFM images displaying the transitions from monolayer to bilayer and bilayer to multilayer  $27.2 \pm 0.5$  and  $31.8 \pm 0.8$  mN/m, respectively. Scale bar  $5 \mu m$ .

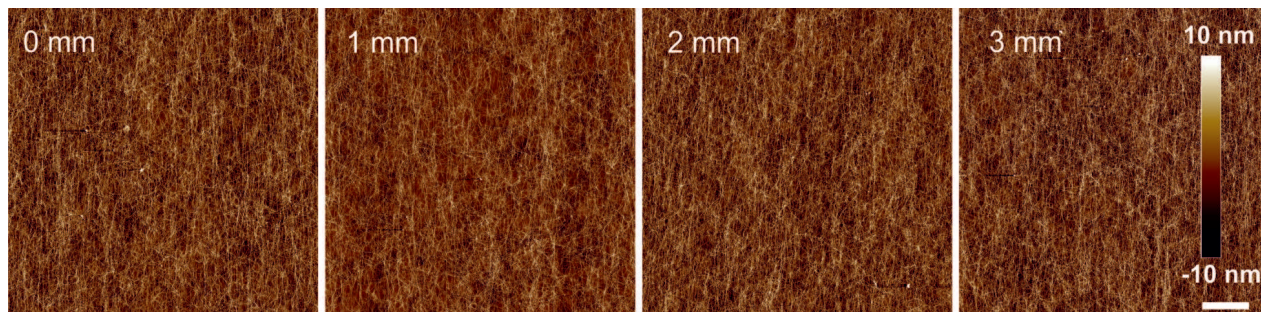


Figure S6: AFM images from a sample produced by holding the pressure constant at 30 mN/m. Scale bar 5  $\mu\text{m}$ .

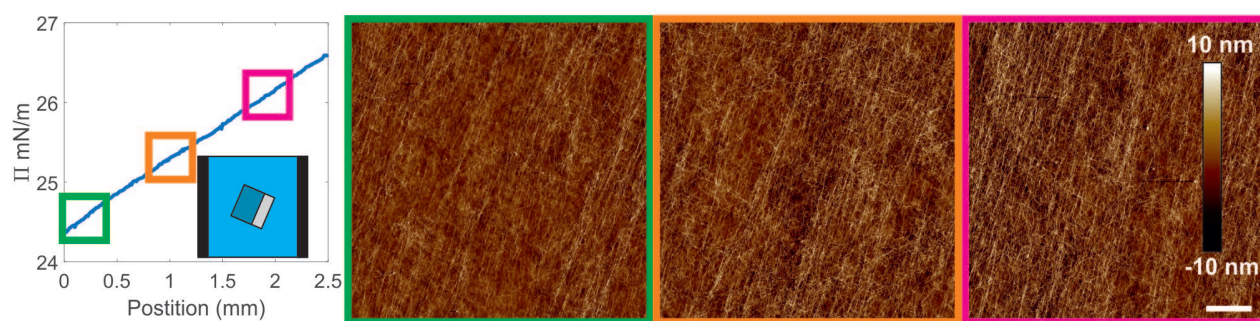


Figure S7: AFM images of a sample held at an angle to the Delrin barriers with corresponding compression curve. Transition of monolayer to bilayer at  $24.9 \pm 0.6$  mN/m. Scale bar 5  $\mu\text{m}$ .