

ELECTRONIC SUPPLEMENTARY INFORMATION

**Pinacyanol Chloride Forms Mesoscopic H- and
J-Aggregates in Aqueous Solution – A Spectroscopic and Cryo-
Transmission Electron Microscopy Study**

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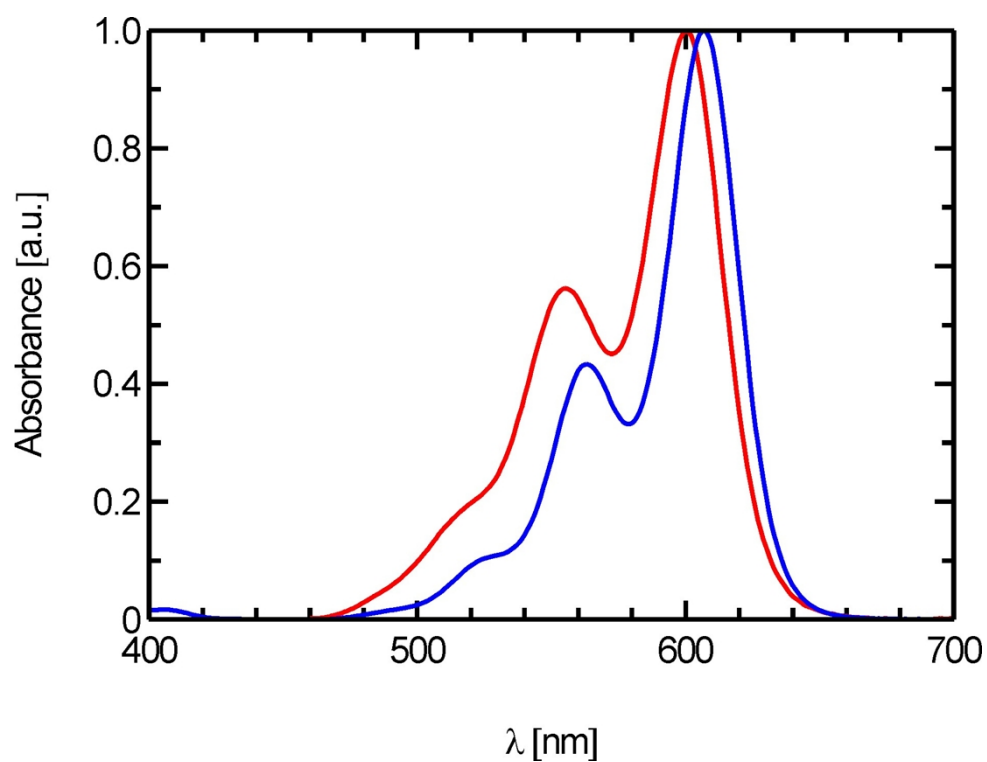


Fig. S1 Normalized absorption spectra of 1.0×10^{-5} M PCYN-Cl solutions in water (red) and ethanol (blue).

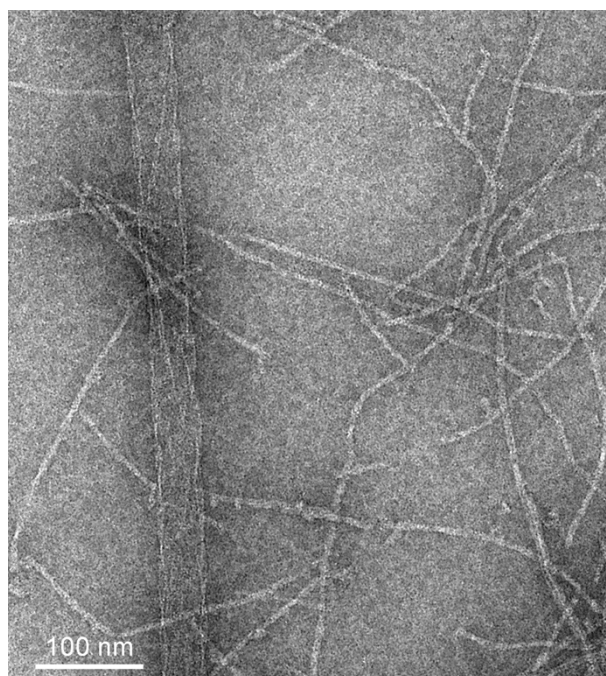


Fig. S2 Negative-stain TEM of a 0.7 mM PCYN-Cl solution taken after shearing the solution for 1.5 minutes in a Couette flow cell at 2000 rpm. In addition to ~ 6 nm thin tubular H-aggregates a wide tape-like J-aggregate can be seen.

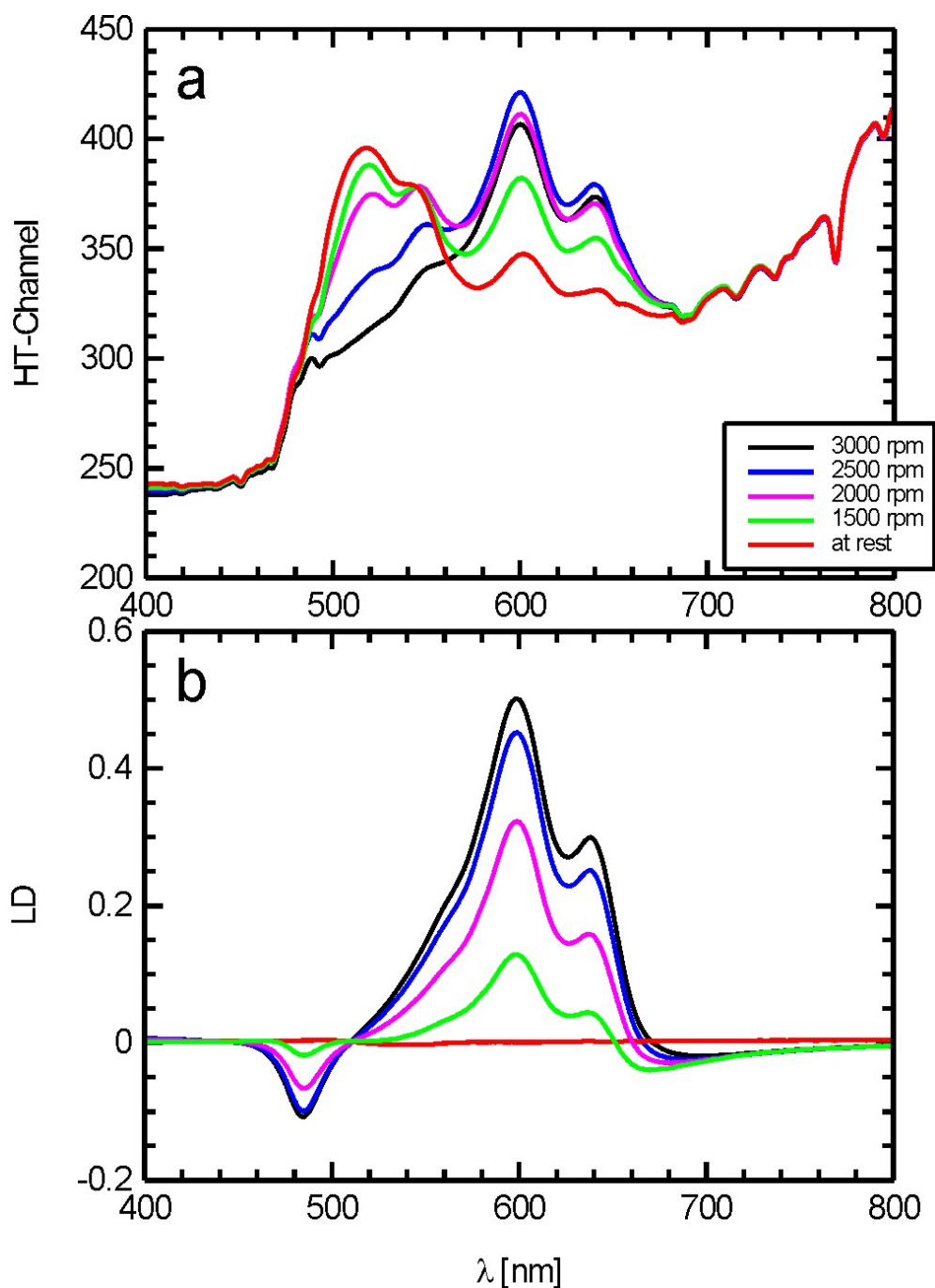


Fig. S3 Structure transformation of a 1-day-old 0.7 mM PCYN-Cl solution induced by shearing in a Couette flow cell. (a) For increasing rotation speed the HT-channel of the spectropolarimeter monitors a dramatic change of absorbance under shear. Strong peaks at ~ 600 and ~ 640 nm evolve at the expense of the 508 nm band. (b) The LD spectra collected in parallel reveal a negative band at ~ 485 nm and positive bands at 597 and 639 nm, respectively. The shape of LD-spectra remains unaffected by the rotation speed, while the intensity of all bands increases before saturation is reached at 3000 rpm.

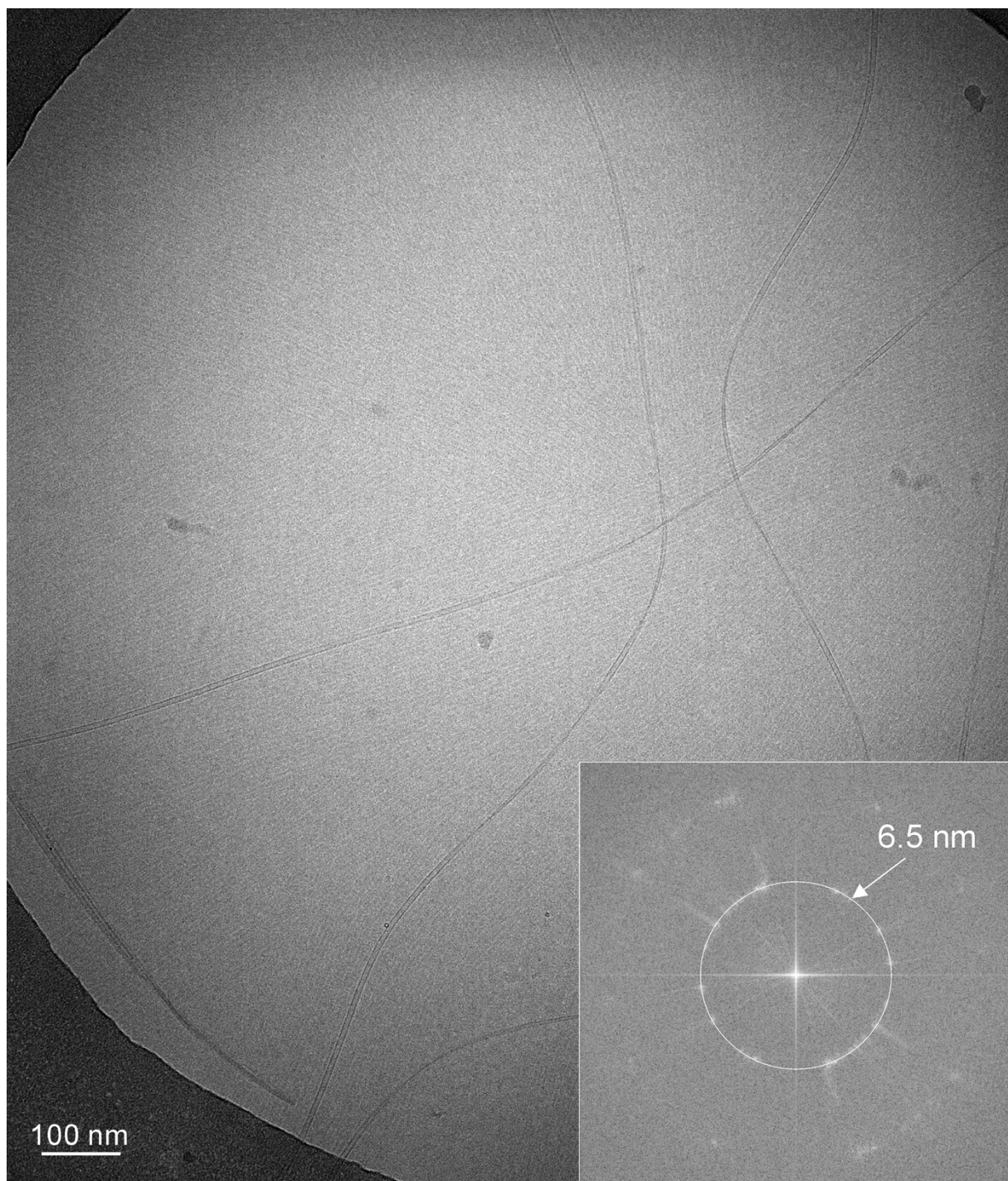


Fig. S4 Cryo-TEM micrograph of a 1.0 mM PCYN-Cl solution taken 14 days after dilution from a 10 mM sample. Few J-aggregates and a background of stripes can be seen. The stripes forming the background pattern are uniformly oriented in separate domains. The Fourier transform (inset) gives for each domain sharp spots that are arranged on a uniform circle corresponding to a repetition period of stripes of 6.5 nm. Due to the high packing order second-order reflections are also visible.

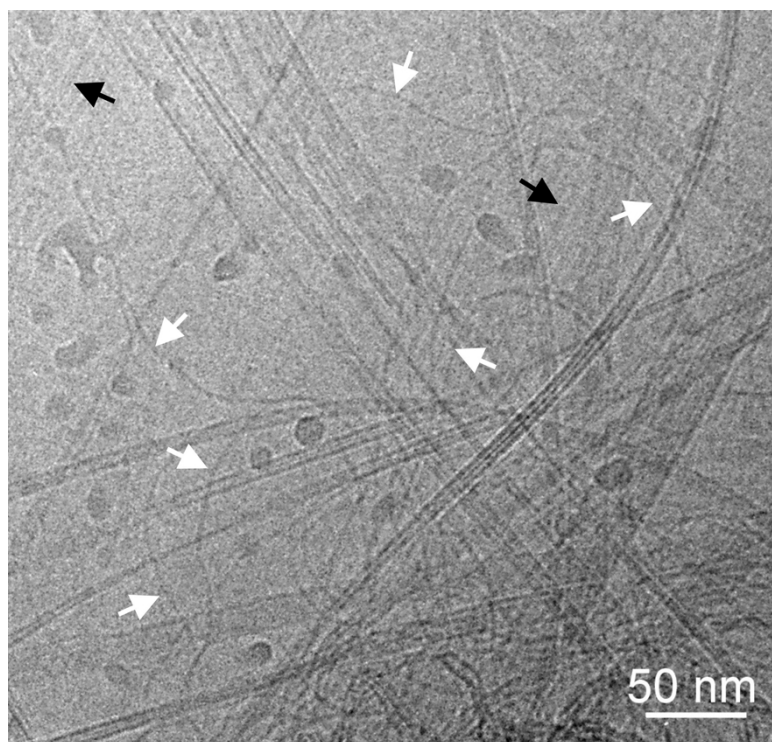


Fig. S5 Cryo-TEM micrograph of a 1.0 mM PCYN-Cl solution taken 4 days after dilution from a 10 mM sample. The micrograph shows thread-like fibres (white arrows) of ~ 2.5 nm diameter among tubular H-aggregates (black arrows) and typical J-aggregate fibrils.

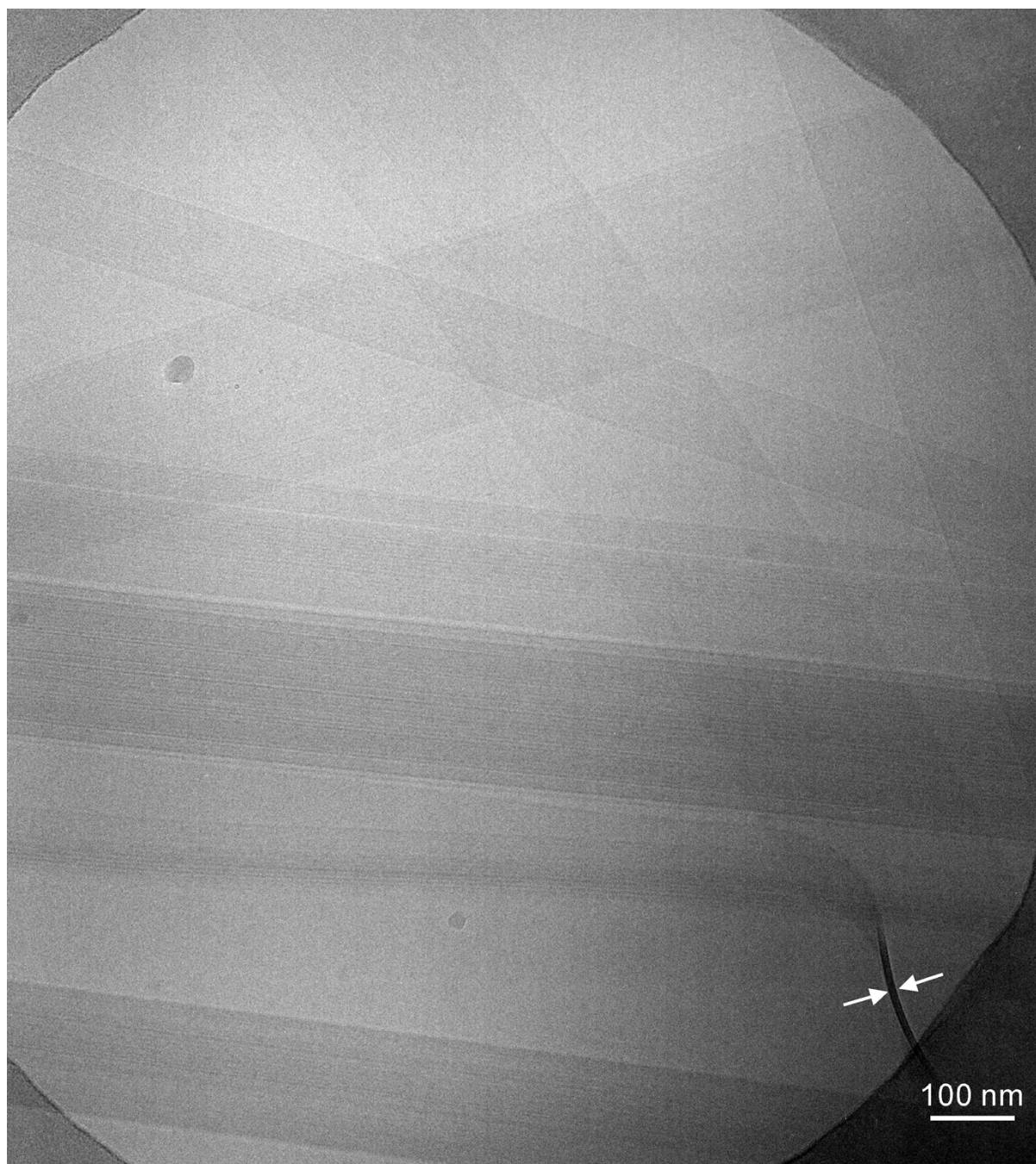


Fig. S6 Cryo-TEM micrograph of a 1.0 mM PCYN-Cl solution after dilution from a 10 mM sample taken after two months of storage. Broad tape-like aggregates dominate. The tape thickness estimated in edge-on orientation (arrows in the lower right corner) amounts to 9.0 ± 1.0 nm.