

**Supporting Information for:**

**Alignment of Helical Nanofilaments**  
**on the Surfaces of Various Self-Assembled**  
**Monolayers**

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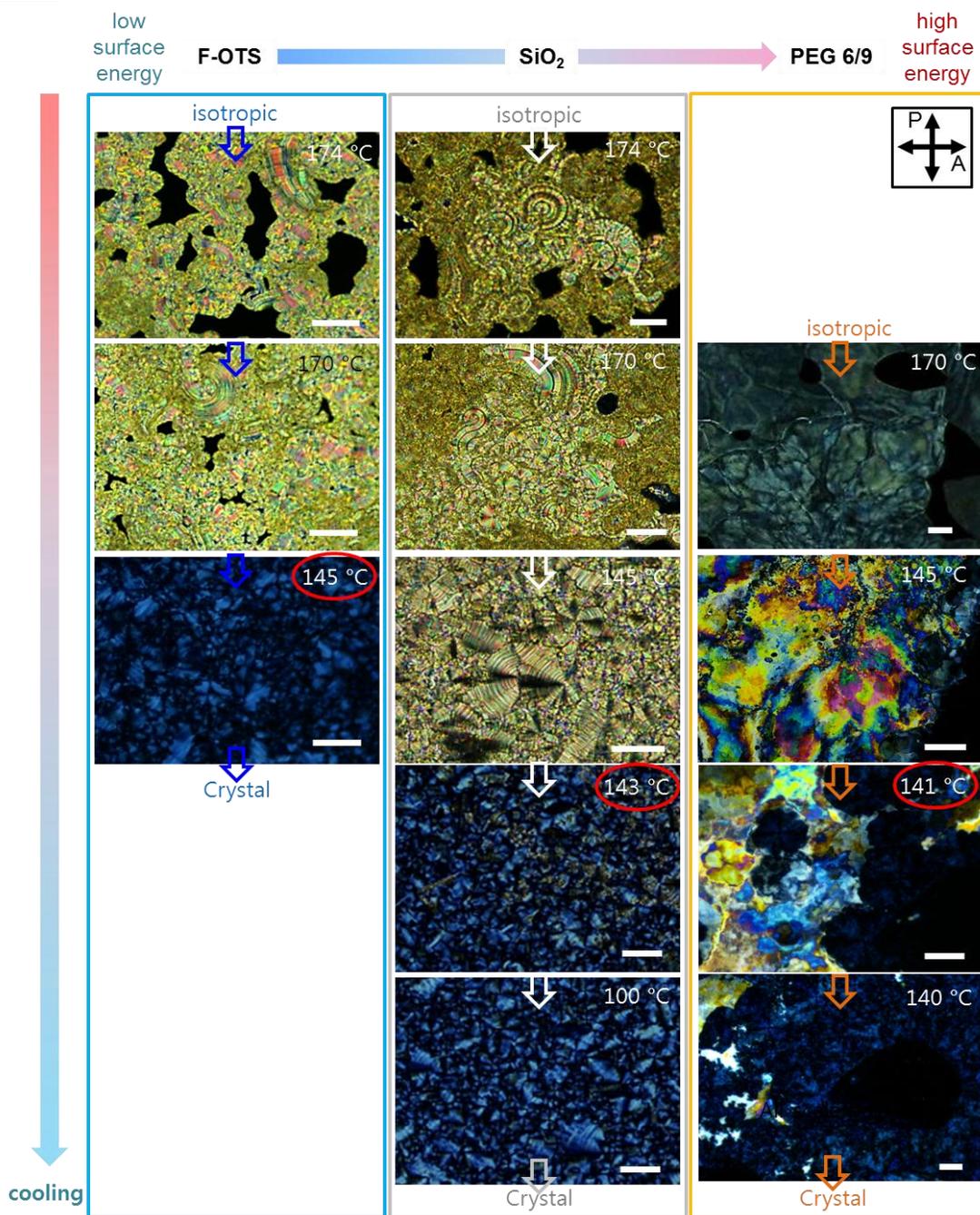
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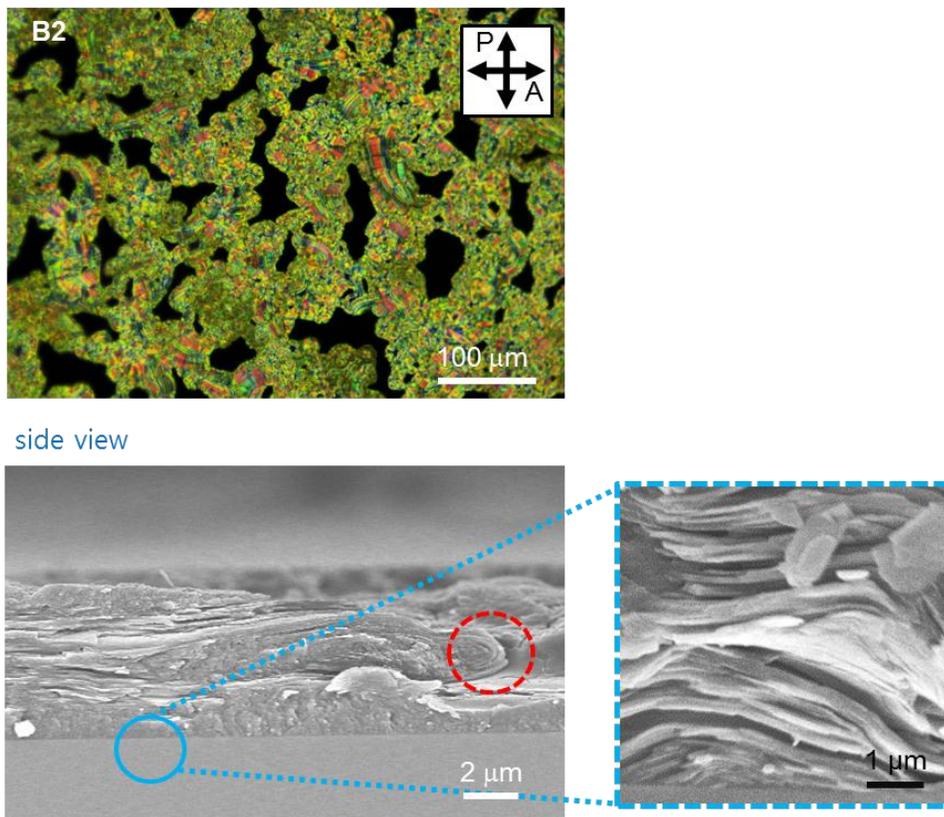
**Figure S1.**



**Fig S1.** DRLM results investigating the overall thermal transitions of NOBOW upon cooling (5 °C/min) under three different interfacial conditions that treated with SAMS, F-OTS(blue), SiO<sub>2</sub>(grey) and PEG6/9 (orange). There is a specific correlation between interfacial resistance and thermal behavior: the more surface anchoring gives, the higher resistance for the molecules arouse saddle-splay distortion. Consequently, it leads to the phase-delay in both iso-to-B2 and B2-to-B4 (red circles) transitions. All scale bars: 100 μm.

**Figure S2.**

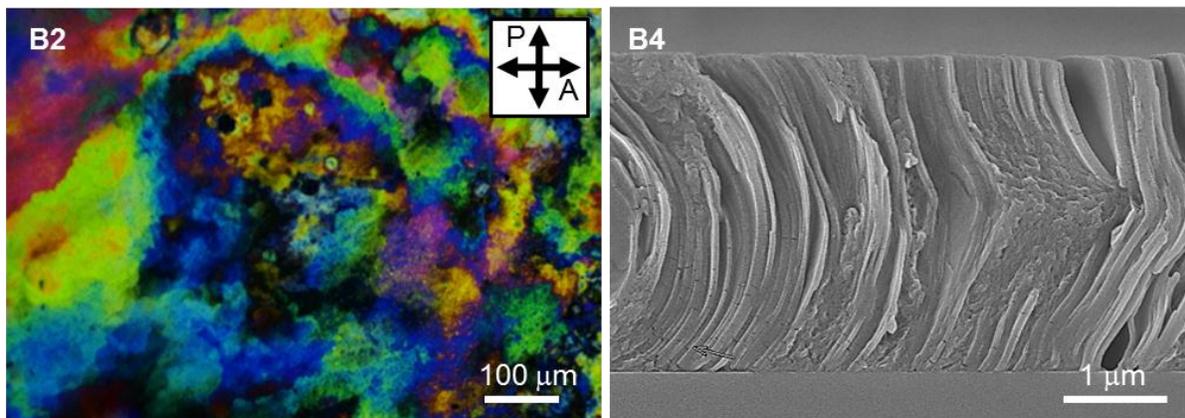
**F-OTS coated substrate**



**Fig S2.** On the F-OTS treated surface, the molecules are perpendicularly aligned at the LC/substrate, so that most of B2 smectic layers are preferentially oriented parallel to the F-OTS/LC interface. In the middle of sample, the film looks more like bulk state, so an elastic positive contribution can come into play to form the topological line defects (red circle) as the surface effect decrease weaker in the distance. These  $1/2$  disclination-like defects locally generate the small anisotropic optical domains representing alternative arrangements of  $\text{SmC}_S\text{P}_A$  layers that are randomly oriented in the middle of LC film.

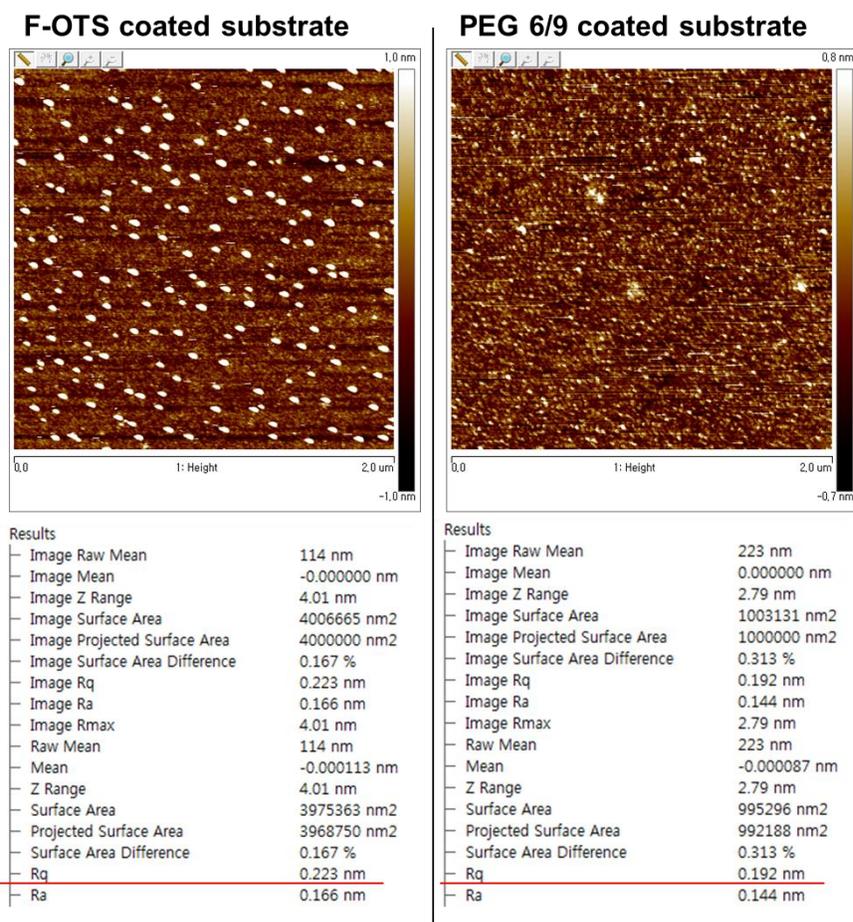
**Figure S3.**

### PEG6/9 coated substrates



**Fig S3.** Vertically aligned smectic layers are obtained between two planar aligned substrates with PEG6/9 moieties. DRLM images of B2 phase on PEG6/9 (above) showed very unique crystal-like textures instead of typical stripe-patterns of B2 phase. This can be understood by SEM cross-sectioned images (below) showing the chevron-like smectic layers originated from highly-packed planar alignment of the molecules due to high surface anchoring at the LC/substrate interfaces.

Figure S4.



**Fig S4.** Before testing these substrates with NOBOW molecule, the quality of SAMs is examined by direct imaging with AFM, showing low surface roughness. Surface topographies of the SAMs were observed under ambient conditions with an AFM (Multimode-N3: Bruker AXS Pte Ltd). Based on AFM investigations, each substrate (molecule-phobic and -philic) is prepared to control the orientation of B2 phase that might govern the B4, HNFs.