

## Supplementary Information for Frustration between two- and three-dimensional smectic ordering leads to a biaxial nematic phase

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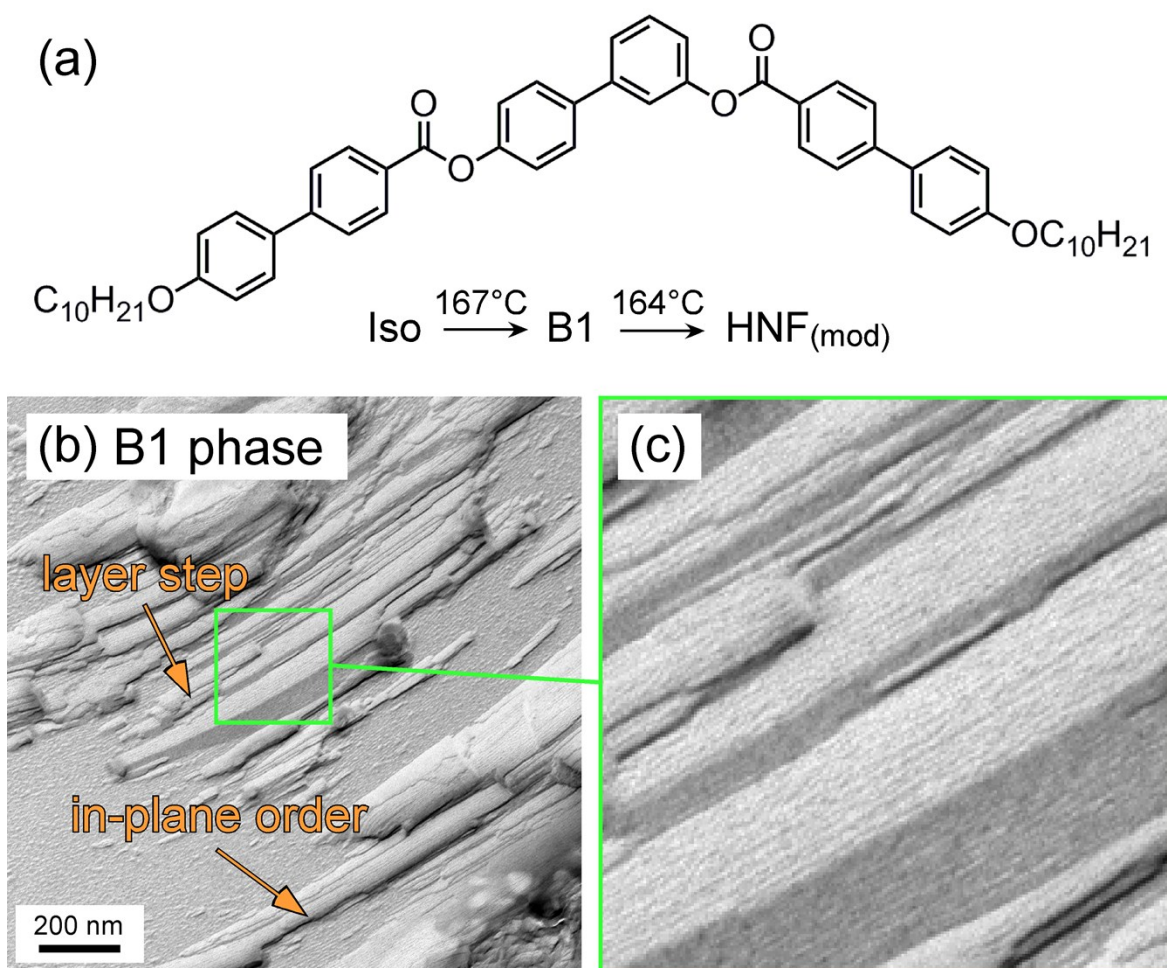


Figure S1: FFTEM images showing the character of fracture surfaces of a lamellar smectic phase with in-plane modulation of a bent-core mesogen. (a) Chemical structure and phase sequence of a homologue of biphenyl-3,4'-diyl bis-(4'-alkoxybiphenyl-4-carboxylate) [1]. (b) FFTEM image of the smectic B1 banana phase, showing layer steps and layers with, periodic ordering in two-dimensions, magnified in (c).

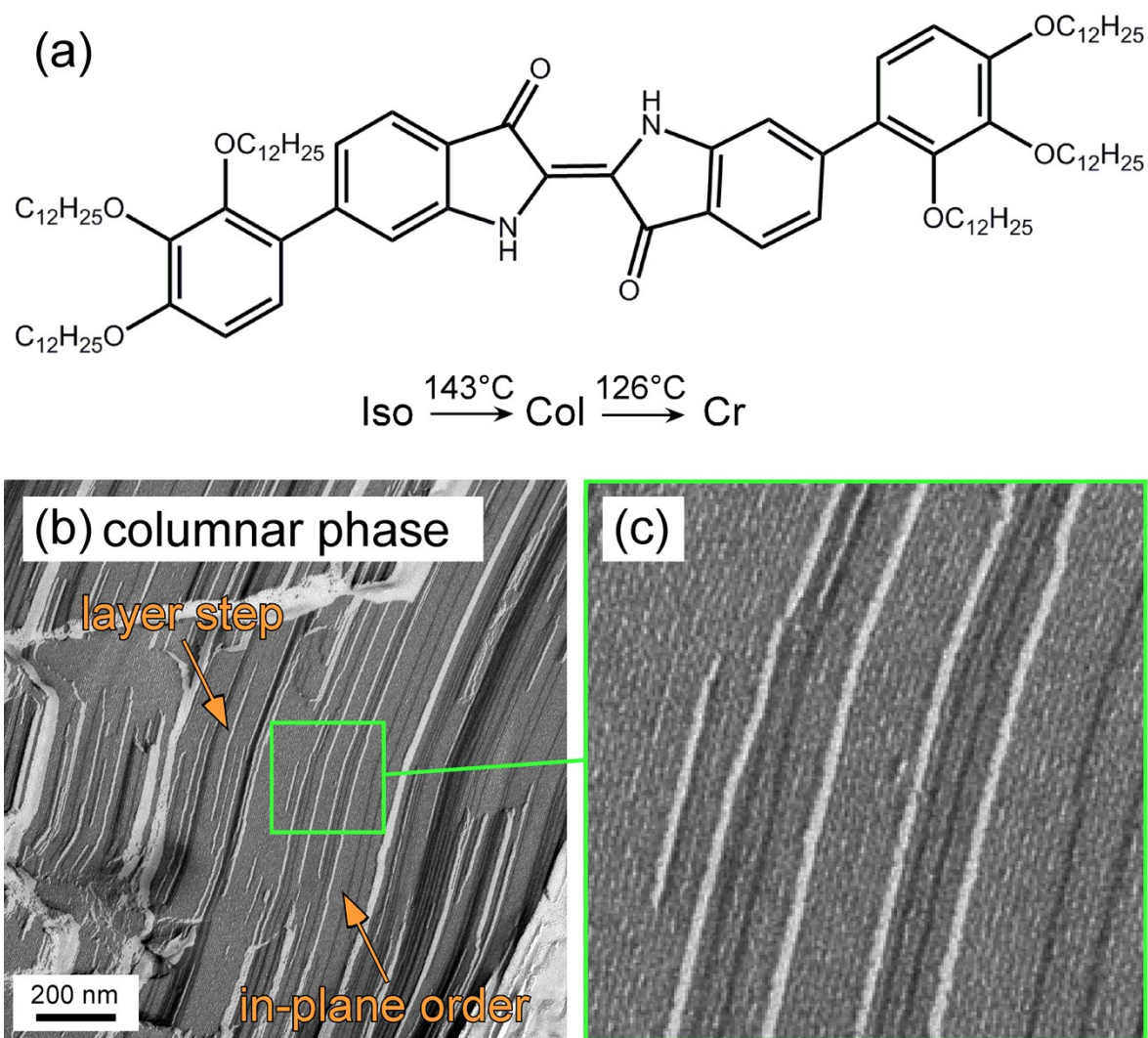


Figure S2: FFTEM images showing the character of fracture surfaces of a well-ordered columnar phase. (a) Chemical structure and phase sequence of phasmidic indigo derivative with 2,3,4-tridodecyloxyphenyl substituents [2]. (b) FFTEM image of the columnar phase, showing two-dimensional, periodic ordering, magnified in (c). The columnar phase shows long-range ordered, periodic structures in two spatial dimensions.

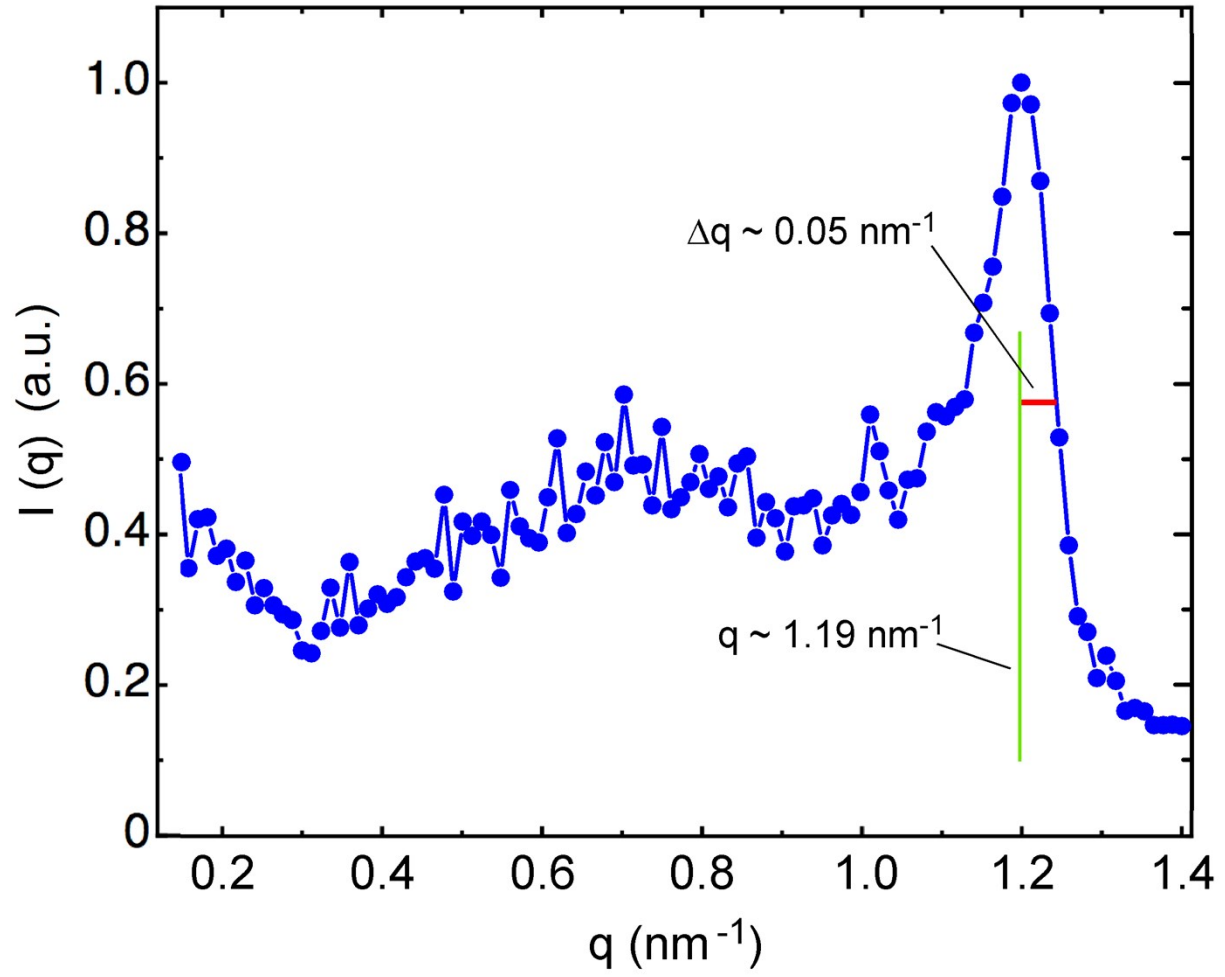


Figure S3: Radial intensity profile in wave vector  $q$  of the Fourier transform scattering pattern of Figure 3e. The peak at about  $1.19 \text{ nm}^{-1}$  corresponds to an average FFTEM stripe spacing of around  $5 \text{ nm}$ .

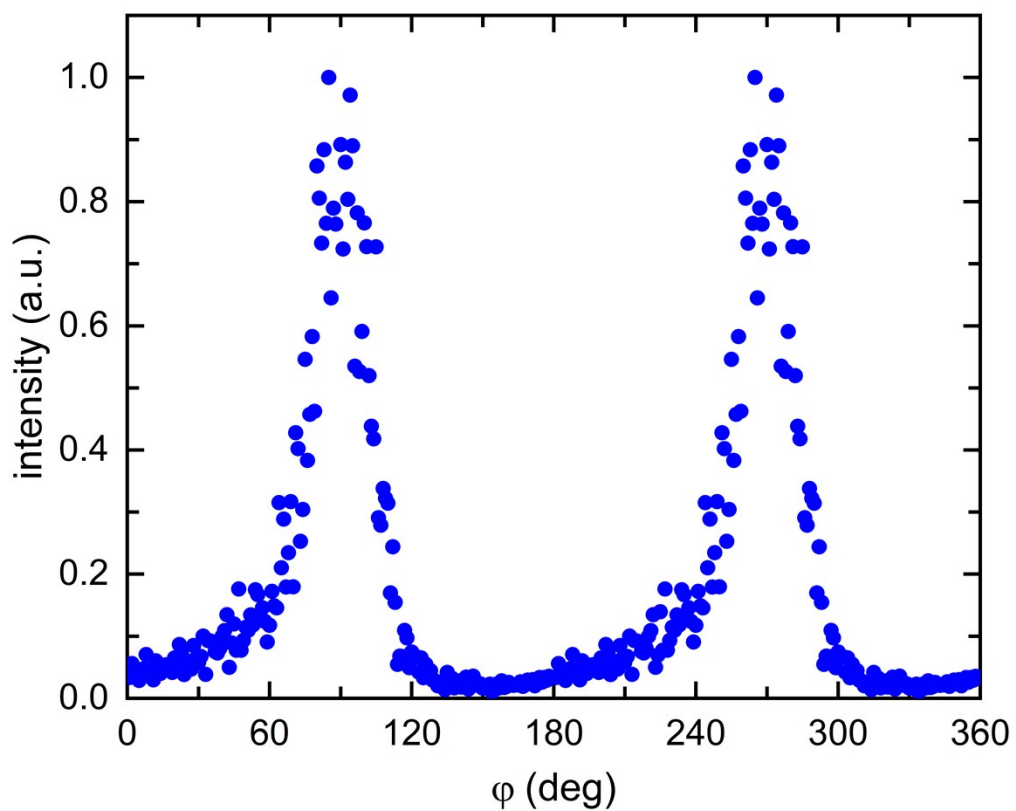


Figure S4: Intensity vs. azimuthal angle  $\phi$  derived from the Fourier transform scattering pattern shown in Figure 3e. The broad peaks reflect the orientational anisotropy of the in-plane smectic layering.

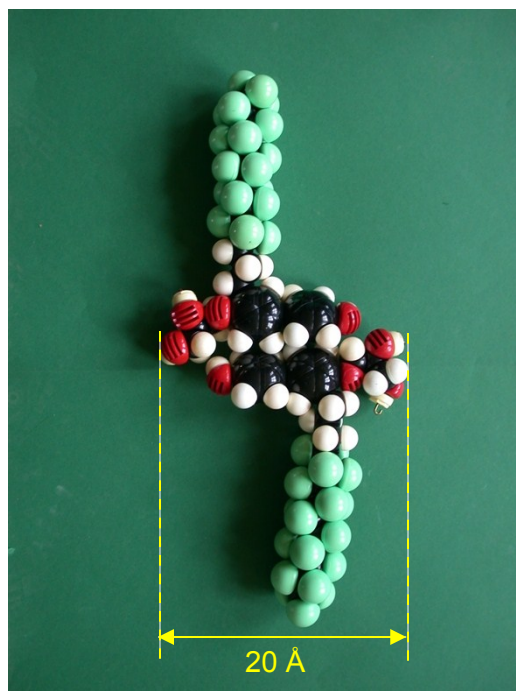


Figure S5. Space-filling molecular model showing two CT2 molecules. The rod-like molecular cores form an antiparallel, fully intercalated pair, giving an in-plane periodicity of  $d = 20 \text{ \AA}$ .

## References

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- [1] E. Tsai, J. M. Richardson, E. Korblova, M. Nakata, D. Chen, Y. Shen, R. Shao, N. A. Clark, and D. M. Walba, *Angew. Chem. Int. Ed.* **52**, 5254-5257 (2013).
  - [2] J. H. Porada and D. Blunk, *J. Mater. Chem.* **20**, 2956-2958 (2010).