Electronic Supplementary Information:

Liquid-Liquid Crystalline Phase Separation in Biological Filamentous Colloids: Nucleation, Growth and Order-Order Transitions of Cholesteric Tactoids

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1. Onsager Theory

Here are reported the definitions used in the main text and the series expansion for large α. For more details see reference[1].

$$\sigma_1(f) = \int f(\Omega)\log\left(4\pi f(\Omega)\right)d\Omega \tag{1}$$

$$\sigma_2(f) = \frac{4}{\pi}\int |\sin(\Theta)| f(\Omega) f(\Omega')\, d\Omega d\Omega' \tag{2}$$

When the angle distribution is the Onsager trial function $f_\alpha(\Theta) = \frac{\alpha}{4\pi}\sinh\alpha\cosh(\alpha\cos\Theta)$ the value of coexistence are obtained by solving

$$\begin{cases} \sigma_1'(\alpha) + \phi_n\sigma_2'(\alpha) = 0 \\ \phi_i + \phi_i^2 = \phi_n + \phi_n^2\sigma_2(\alpha) \\ \log\phi_i + 2\phi_i = \log\phi_n + \sigma_1(\alpha) + 2\phi_n\sigma_2(\alpha). \end{cases} \tag{3}$$

where the apex implies the derivative of $\sigma_{1,2}$ with respect to α. The order parameter becomes

$$S(\alpha) = \frac{\alpha^2 - 3\alpha\coth(\alpha) + 3}{\alpha^2}. \tag{4}$$

For $\alpha \to \infty$ the above terms can be expanded into series

$$\sigma_1(\alpha) \sim \log\alpha - 1, \tag{5}$$

$$\sigma_2(\alpha) \sim \frac{4}{\sqrt{\pi\alpha}}\left(1 - \frac{30}{32\alpha} + \frac{210}{1024\alpha^2} + O(\alpha^{-3})\right), \tag{6}$$

$$\alpha \sim \frac{4}{\pi}\phi^2 - \frac{45}{8} - \frac{11625}{2048}\frac{\pi}{\phi^2} + O(\phi^{-3}), \tag{7}$$

Eq. (7) can be substituted in Eq. (5) and (6) and consequently in the osmotic pressure and chemical potential to obtain the expressions used in the main text. The boundaries of the Onsager theory and the approximation obtained by the asymptotic approach are reported in the following table.

	Onsager	Approximation
ϕ_i	3.3399	3.3639
ϕ_n	4.4858	4.6837
ϕ_n/ϕ_i	1.343	1.392
α	18.584	22.306

Fig S1. shows the bifurcation diagram of the Onsager theory, that is the position of the minima and maxima of the free energy (Eq. (1) of the Main Text) for increasing concentration ϕ and the relative order parameter S. The green line represents the stable equilibrium points, the blue line the meta-stable equilibrium points and the red line the unstable ones. For $\phi \approx 3.65$ a saddle-node bifurcation appears, from which a meta-stable and unstable branch emerge. The unstable branch connects with the anisotropic state (S = 0) at ϕ = 4, as already discussed by Kayser and Raveché.[2] The metastable branch becomes stable for $\phi > 3.68$. A more in-depth discussion can be found in Reference.[3]

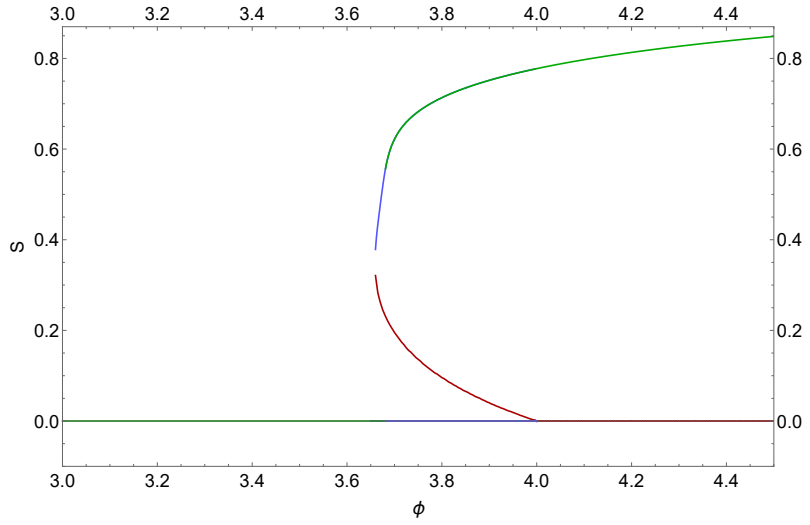

Fig. S 1: Order parameter S and concentration ϕ for stable (green line), meta-stable (blue line) and unstable (red line) equilibrium points for the Onsager free energy, as of Eq. (1) of the main text.

2. Growth Phase

In the figure S2, we calculated the partial energy contribution of anisotropy $\Delta F_W = \frac{F_W - \gamma S}{F_W}$, as a function of the anchoring strength ω; where F_W comes from Eq. (6) of the main text, and $S = 6^{2/3}\pi^{1/3}V^{2/3}$ is the surface energy of a sphere of volume V.

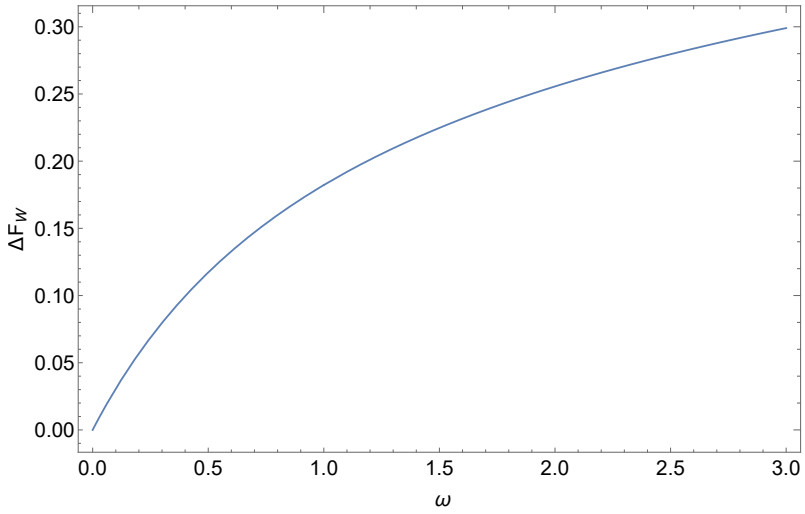

Fig. S 2: Energy contribution of anisotropy ΔF_W for homogenous tactoids with anchoring strength ω.

It is true that for high anchoring strength, this contribute becomes more and more important, around 30% for ω = 3. However, in our case ω < 1.5, the largest part of the total energy is given by the surface and not by the anisotropy. For larger volumes, this contribute will become less important.

3. Discussion

The fitted parameters used in Figure 5 of the main text are:

	K (μm)	ν	q (μm)
BLG	262	2.4	-118
C-CNC	122	3.5	-83
S-CNC	654	-12.6	-22

Table S 1: Fitted parameters of Eq. (14) of the main text for each system.

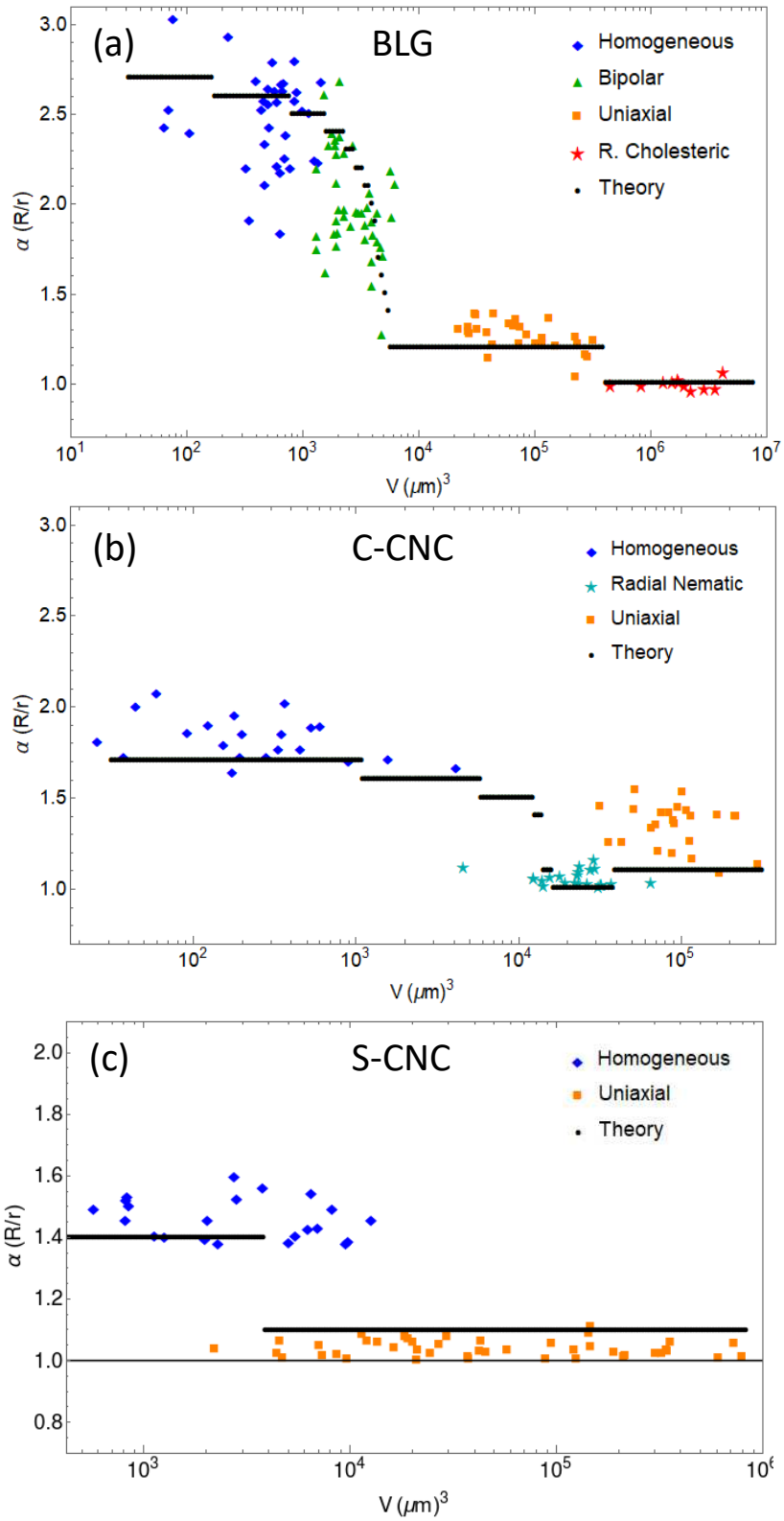

Fig. S 3: Aspect ratio (α) of the tactoids as a function of their volume (V). Experimental points in colors, theory in black for: (**a**) BLG, adapted from Bagnani *et al.*;[4] (**b**) carboxylated cellulose nanocrystals and (**c**) sulfurated cellulose nanocrystals, adapted from Bagnani *et al.*[5]

References

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[4] Massimo Bagnani, Paride Azzari, Salvatore Assenza, and Raffaele Mezzenga. Six-fold director field configuration in amyloid nematic and cholesteric phases. *Scientific reports*, 9(1):1–9, 2019.

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