

Electronic Supplementary Information

1 Measurement of the helical twist power

To measure the Helical Twist Power (HTP) of the cholesteric mixtures studied in the main text, I prepared a Cano wedge sample as follows. Two glass plates were cleaned using the same protocol than in the main text. 1g of PVA (poly[vinyl alcohol], $M_w = 85000-124000$, 87–89% hydrolyzed, Sigma Aldrich) was dissolved in 50 cm³ of water and 3.22 cm³ of ethanol, and drops of the resultant solution were filtered and spin-coated at 2000 rpm during 20 s on the glass plates, which were subsequently baked at 110 °C during 1 h and rubbed with a rotating velvet cylinder. The wedge sample was assembled from the treated glass plates by separating them with a calibrated tungstene wire of diameter 50 μm (Goodfellow) on one side of the sample and nothing on the other side of the sample. The plates were fastened using epoxy glue on all sides of the sample, leaving only two holes for the filling. The rubbing direction was orthogonal to the wedge direction x . Because of residual stresses, the thickness profile $h(x)$ of the wedge is not perfectly linear and was therefore accurately mapped with a spectrometer as described in the main text. Finally, this sample was filled by capillarity with a mixture of 5CB and a mass fraction $C = 0.96$ wt% of R811.

As reviewed by Oswald and Pieranski¹, defects appear at positions x_n such that $h_n \equiv h(x_n) = P(2n - 1)/4$, with P the cholesteric pitch and n the integer defect number. I measured experimentally the defect positions x_n using the same reference point as for the thickness calibration curve $h(x)$, and deduced the defect thicknesses h_n . Plotting h_n against n and performing a linear fit gives a line crossing the abscissa axis at 0.5, with a slope of $P/2$. The results of this measurement and fit was $P = 9.02 \pm 0.03 \mu\text{m}$ at $T - T_{NI} = 5^\circ\text{C}$, from which we can deduce $\text{HTP} \equiv 1/(PC) = 0.1155 \pm 0.0004 \text{ wt}\%^{-1} \mu\text{m}^{-1}$.

2 Additional proofs for the case of small transverse wavevectors

2.1 Eigenvalue equation

Let us first calculate the actions of the dissipation operators (as defined in the main text) in the limit $\mathbf{k}_\perp \rightarrow 0$:

$$\Gamma^{\text{SB}} \equiv \gamma_1 \left(1 - \mu \left[\partial_Z^2 \right] \left[\partial_Z^4 \right]_{\text{D,N}}^{-1} \left[\partial_Z^2 \right] \right), \quad (1)$$

$$\Gamma^{\text{TB}} \equiv \gamma_1 \left(-\mu \left[\partial_Z \right] \left[\partial_Z^2 \right]_{\text{D}}^{-1} \left[\partial_Z \right] \right), \quad (2)$$

with $Z \equiv 2z/h$ and $\mu \equiv \alpha_2^2 / (\gamma_1 \eta_c)$ as in the main text.

To calculate the expression of $\Gamma^{\text{TB}} u$ for any function u , let us solve the differential equation $v''(Z) = u'(Z)$ with the Dirichlet Boundary Conditions (BCs) $v(\pm 1) = 0$. By integrating two times this differential equation and using the BCs to find the integration constants, one finds the following solution:

$$v(Z) = \left[\int_{-1}^Z u(Z') dZ' \right] - \langle u \rangle (1 + Z), \quad (3)$$

with the average operator $\langle u \rangle$ defined as in the main text. One deduces from this solution the identity $v'(Z) = u(Z) - \langle u \rangle$ and the following expression for the action of Γ^{TB} :

$$\Gamma^{\text{TB}} u = \gamma_1^* \left(u + \frac{\mu}{1 - \mu} \langle u \rangle \right) \quad (4)$$

with $\gamma_1^* \equiv \gamma_1 (1 - \mu)$.

To calculate the expression of $\Gamma^{\text{SB}} u$ for any function u , let us solve the differential equation $v''''(Z) = u''(Z)$ with the Dirichlet BCs $v(\pm 1) = 0$ and Neumann BCs $v'(\pm 1) = 0$. By integrating four times this differential equation and using the BCs to find the integration constants, one finds the following solution:

$$v(Z) = \left[\int_{-1}^Z \int_{-1}^{Z'} u(Z'') dZ'' dZ' \right] - \frac{(1+Z)^2}{2} [\langle u \rangle + (Z-2)\langle Z'u \rangle], \quad (5)$$

One deduces from this solution the identity $v''(Z) = u(Z) - \langle u \rangle - 3Z\langle Z'u \rangle$ and the following expression for the action of Γ^{SB} :

$$\Gamma^{\text{SB}} u = \gamma_1^* \left(u + \frac{\mu}{1 - \mu} [\langle u \rangle + 3Z\langle Z'u \rangle] \right). \quad (6)$$

By taking the limit $|\mathbf{k}_\perp| \rightarrow 0$ in eqn (22) of the main text and using eqn (4,6), one finds the following system of equations:

$$\begin{pmatrix} \partial_Z^2 + \phi_\tau^2 & 2\phi_q \partial_Z \\ -2\phi_q \partial_Z & \partial_Z^2 + \phi_\tau^2 \end{pmatrix} \begin{pmatrix} m^{\text{SB}} \\ n^{\text{TB}} \end{pmatrix} = -\frac{\phi_\tau^2 \mu}{1 - \mu} \begin{pmatrix} \langle \tilde{n}^{\text{SB}} \rangle + 3Z\langle Z'\tilde{n}^{\text{SB}} \rangle \\ \langle \tilde{n}^{\text{TB}} \rangle \end{pmatrix}, \quad (7)$$

with $\phi_q \equiv (qh/2)(K_2/K_3)$ and $\phi_\tau \equiv (h/2)\sqrt{\gamma_1^* f/K_3}$ as in the main text. One can eliminate the right-hand-side of eqn (7) by defining the following auxiliaries variables:

$$m^{\text{SB}} = \tilde{n}^{\text{SB}} + \frac{\mu}{1 - \mu} \left(\langle \tilde{n}^{\text{SB}} \rangle + 3Z\langle Z'\tilde{n}^{\text{SB}} \rangle \right), \quad (8)$$

$$m^{\text{TB}} = \tilde{n}^{\text{TB}} + \frac{\mu}{1 - \mu} \left(\langle \tilde{n}^{\text{TB}} \rangle + \frac{6\phi_q}{\phi_\tau^2} \langle Z'\tilde{n}^{\text{SB}} \rangle \right), \quad (9)$$

from which one deduces:

$$\begin{pmatrix} \partial_Z^2 + \phi_\tau^2 & 2\phi_q \partial_Z \\ -2\phi_q \partial_Z & \partial_Z^2 + \phi_\tau^2 \end{pmatrix} \begin{pmatrix} m^{\text{SB}} \\ m^{\text{TB}} \end{pmatrix} = \mathbf{0}. \quad (10)$$

Finally, one can find \tilde{n}^{SB} and \tilde{n}^{TB} as functions of m^{SB} and m^{TB} by noticing that:

$$\langle m^{\text{SB}} \rangle = \frac{\langle \tilde{n}^{\text{SB}} \rangle}{1 - \mu}, \quad (11)$$

$$\langle Z'm^{\text{SB}} \rangle = \frac{\langle Z'\tilde{n}^{\text{SB}} \rangle}{1 - \mu}, \quad (12)$$

$$\langle m^{\text{TB}} \rangle = \frac{1}{1 - \mu} \left(\langle \tilde{n}^{\text{TB}} \rangle + \frac{6\mu\phi_q}{\phi_\tau^2} \langle Z'\tilde{n}^{\text{SB}} \rangle \right), \quad (13)$$

from which one deduces with eqn (8,9):

$$\tilde{n}^{\text{SB}} = m^{\text{SB}} - \mu \left[\langle m^{\text{SB}} \rangle + 3Z \langle Z' m^{\text{SB}} \rangle \right] \quad (14)$$

$$\tilde{n}^{\text{TB}} = m^{\text{TB}} - \mu \left[\langle m^{\text{TB}} \rangle + \frac{6\phi_q(1-\mu)}{\phi_\tau^2} \langle Z' m^{\text{SB}} \rangle \right] \quad (15)$$

Eqn (10,14,15) are the same than the ones given at the beginning of Sec. 3.2.2 in the main text, and therefore conclude this proof.

2.2 Dispersion relations

Let us now deduce the dispersion relation associated with the eigenvalues of eqn (10). As explained in the main text, the modes can be splitted into even-odd and odd-even profiles. Let us examine each case separately.

2.2.1 n^{SB} is even and n^{TB} is odd.

Even-odd solutions of eqn (10) have the following general form:

$$\tilde{n}^{\text{SB}}(Z) = A_+ \cos(Z\phi_+) + A_- \cos(Z\phi_-) \quad (16)$$

$$\tilde{n}^{\text{TB}}(Z) = B_+ \sin(Z\phi_+) + B_- \sin(Z\phi_-) \quad (17)$$

with $\phi_\pm \equiv \sqrt{\phi_q^2 + \phi_\tau^2} \pm \phi_q$. From eqn (10), one finds that the coefficients of this solution must fulfill the identity $A_\pm = \pm B_\pm$. From the Dirichlet BCs $\mathbf{n}(Z = \pm 1) = \mathbf{0}$ and eqn (14,15), one also deduce the following relations:

$$A_+ \cos(\phi_+) + A_- \cos(\phi_-) = \mu [A_+ \text{sinc}(\phi_+) + A_- \text{sinc}(\phi_-)] \quad (18)$$

$$B_+ \sin(\phi_+) + B_- \sin(\phi_-) = 0 \quad (19)$$

using the identity $\langle \cos(Z'\phi) \rangle = \text{sinc} \phi$, valid for arbitrary ϕ . Using the identity $A_\pm = \mp B_\pm$ mentioned just above, one can eliminate the coefficients B_\pm from the previous system of equation:

$$\begin{pmatrix} \cos(\phi_+) - \mu \text{sinc}(\phi_+) & \cos(\phi_-) - \mu \text{sinc}(\phi_-) \\ -\sin(\phi_+) & \sin(\phi_-) \end{pmatrix} \begin{pmatrix} A_+ \\ A_- \end{pmatrix} = \mathbf{0}. \quad (20)$$

The determinant of the previous system must be zero in order to get non-trivial solutions, and therefore constitutes the dispersion relation of the eigenvalue ϕ_τ^2 :

$$\sin(2\phi_s) = \mu [\sin(\phi_+) \text{sinc}(\phi_-) + \sin(\phi_-) \text{sinc}(\phi_+)], \quad (21)$$

with $\phi_s \equiv \sqrt{\phi_q^2 + \phi_\tau^2}$. Using elementary trigonometric identities, one can transform this dispersion relation under the same form than the one given in the main text:

$$R_1(\phi_s)R_2(\phi_s) = \frac{3\mu\phi_q^2}{\phi_\tau^2} R_q(\phi_s), \quad (22)$$

with:

$$R_1(\phi_s) \equiv \cos \phi_s - \mu \text{sinc} \phi_s, \quad (23)$$

$$R_2(\phi_s) \equiv \text{sinc} \phi_s, \quad (24)$$

$$R_q(\phi_s) \equiv \frac{\text{sinc}^2 \phi_s - \text{sinc}^2 \phi_q}{3} \quad (25)$$

2.2.2 n^{SB} is odd and n^{TB} is even.

Odd-even solutions of eqn (10) have the following general form:

$$\tilde{n}^{\text{SB}}(Z) = A_+ \sin(Z\phi_+) + A_- \sin(Z\phi_-) \quad (26)$$

$$\tilde{n}^{\text{TB}}(Z) = B_+ \cos(Z\phi_+) + B_- \cos(Z\phi_-) \quad (27)$$

with ϕ_\pm defined as in the previous subsection. From eqn (10), one finds that the coefficients of this solution must fulfill the identity $A_\pm = \mp B_\pm$. From this identity, the Dirichlet BCs $\mathbf{n}(Z = \pm 1) = \mathbf{0}$ and eqn (14,15), one deduce the following relations:

$$\sum_{\sigma=\pm} A_\sigma \left[\sin(\phi_\sigma) - 3\mu \frac{\text{sinc}(\phi_\sigma) - \cos(\phi_\sigma)}{\phi_\sigma} \right] = 0, \quad (28)$$

$$\sum_{\sigma=\pm} A_\sigma \left\{ \sigma [\cos(\phi_\sigma) - \mu \text{sinc}(\phi_\sigma)] + \frac{2(1-\mu)\phi_q}{\phi_\tau^2} \sin(\phi_\sigma) \right\} = 0, \quad (29)$$

using the identities $\langle \cos(Z'\phi) \rangle = \text{sinc} \phi$ and $\langle Z' \sin(Z'\phi) \rangle = (\text{sinc} \phi - \cos \phi)/\phi$, valid for arbitrary ϕ . Similar to the previous subsection, the next step is to impose that the determinant of the system just above is zero. After a long but straightforward calculation based on the following relations:

$$\sin(\phi_+) \sin(\phi_-) = \sin^2 \phi_s - \sin^2 \phi_q, \quad (30)$$

$$\cos(\phi_+) \cos(\phi_-) = \cos^2 \phi_s - \sin^2 \phi_q, \quad (31)$$

$$\frac{\sin(\phi_+) \cos(\phi_-)}{\phi_-} - \frac{\sin(\phi_-) \cos(\phi_+)}{\phi_+} \quad (32)$$

$$= \frac{2\phi_s \phi_q}{\phi_\tau^2} [\text{sinc} 2\phi_s + \text{sinc} 2\phi_q],$$

$$\frac{\sin(\phi_+) \cos(\phi_-)}{\phi_+^2} + \frac{\sin(\phi_-) \cos(\phi_+)}{\phi_-^2} \quad (33)$$

$$= \frac{2\phi_s}{\phi_\tau^4} \left[(\phi_s^2 + \phi_\tau^2) \text{sinc} 2\phi_s - \phi_q^2 \text{sinc} 2\phi_q \right],$$

one finds the dispersion relation under the form given in the main text:

$$R_1(\phi_s)R_2(\phi_s) = \frac{3\mu\phi_q^2}{\phi_\tau^2} R_q(\phi_s), \quad (34)$$

with:

$$R_1(\phi_s) \equiv \cos \phi_s - \mu \operatorname{sinc} \phi_s, \quad (35)$$

$$R_2(\phi_s) \equiv \operatorname{sinc} \phi_s - \mu T(\phi_s), \quad (36)$$

$$R_q(\phi_s) \equiv \left[\frac{2}{3} + \frac{\mu}{\phi_\tau^2} - \frac{4(1-\mu)\phi_q^2}{\phi_\tau^4} \right] \left[\operatorname{sinc}^2 \phi_q - \operatorname{sinc}^2 \phi_s \right] + \frac{2(2-\mu)}{\phi_\tau^2} \left[\operatorname{sinc} 2\phi_s - \operatorname{sinc} 2\phi_q \right] + \operatorname{sinc}^2 \phi_s \quad (37)$$

$$+ \frac{T(\phi_s)}{3} \left[\cos \phi_s + \left(\frac{4(1-\mu)\phi_s^2}{\phi_\tau^2} - \mu \right) \operatorname{sinc} \phi_s \right],$$

$$T(\phi_s) = 3 \frac{\operatorname{sinc} \phi_s - \cos \phi_s}{\phi_s^2}. \quad (38)$$

2.3 Theoretical expressions of the eigenvalues for the soft modes

Using the dispersion relations of the previous section, we can find approximate expressions for the eigenvalues of the soft modes associated with vanishingly small decay frequency when $\phi_q \rightarrow \pi/2$, i.e. when the sample approach the threshold of absolute destabilization of the unwound phase with a mass fraction of chiral molecules C equal to the critical fraction C_c . This calculation relies on a Taylor series expansion of the dispersion relation in the small parameter $\varepsilon \equiv 1 - (C/C_c)^2$ and a solution of the following form:

$$\phi_\tau^{(\alpha,0)} = \frac{\pi}{2} \sqrt{\frac{(1-\mu)\varepsilon}{1 - \frac{\mu}{2} [f_\alpha(\mu) + g_\alpha(\mu)(1-\varepsilon)]}} \quad (39)$$

The previous equation is the same than the one given in the main text, apart from the explicit use of the small parameter ε instead of C/C_c and the use of the rescaled eigenvalue $\phi_\tau = (\pi/2)\sqrt{\tau_h/\tau}$ instead of τ_h/τ .

The eigenvalue $\phi^{(0,0)}$ (resp., $\phi^{(1,0)}$) is associated with the even-odd (resp., odd-even) dispersion relation. The calculation of the Taylor series of these dispersion relations is a bit tedious and was therefore facilitated with extensive use of Mathematica. Since there are terms in $1/\phi_\tau^4$ and $1/\phi_\tau^2$ in the dispersion relations, one could expect ill-defined behaviour when $\varepsilon \rightarrow 0$. However, I found out that for both dispersion relations, terms of order $1/\varepsilon^2$, $1/\varepsilon$ and 1 cancels out, leading to well-defined solutions. Cancelling out the terms of order ε and ε^2 gives the following expressions of f_α and g_α :

$$g_0(\mu) = \frac{1}{2} + \frac{\mu}{8} \left[\frac{1}{1 - \frac{\mu}{2}} \right], \quad (40)$$

$$g_1(\mu) = v - \frac{\mu}{8} \left[\frac{1 - 4v - 3v^2}{1 - \frac{\mu}{2}(1-v)} \right], \quad (41)$$

$$f_0(\mu) = 1 - g_0(\mu), \quad (42)$$

$$f_1(\mu) = 1 - g_1(\mu) - v, \quad (43)$$

with $v \equiv 3/\pi^2$.

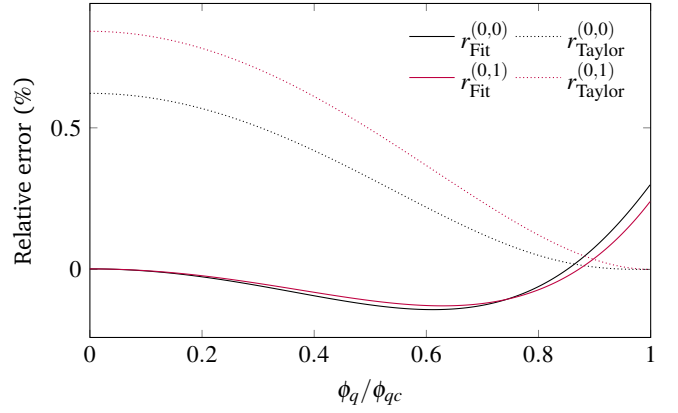


Fig. 1 Relative error between eqn (39) and the numerically calculated eigenvalues plotted against the rescaled mass fraction of R811, when defining the functions f_α and g_α either from the Taylor expansion of this section or the fitted functions from the main text.

Let us define $r_{\text{Taylor}}^{(\alpha,0)}$ as the relative error between the numerically calculated eigenvalue $\phi_\tau^{(\alpha,0)}$ and its approximation calculated with eqn (39–43). For completeness, let us also define $r_{\text{Fit}}^{(\alpha,0)}$ as the same quantity calculated with the fitted functions f_α and g_α introduced in the main text. These quantities are plotted as a function of C/C_c on Fig. 1. One may observe that the Taylor expansion formula gives, by construction, asymptotically good accuracy near $C = C_c$, but is not particularly good near $C = 0$. Conversely, using eqn (39) with the fitted functions f_α and g_α of the main text gives much better accuracy over the whole range of values for C .

2.4 Anomalous divergence of the pressure at small wavevector

Let us define $v_k \equiv \mathbf{v}_\perp \cdot \mathbf{e}_k$ and $n_k \equiv \mathbf{n}_\perp \cdot \mathbf{e}_k$, with $\mathbf{e}_k \equiv \mathbf{k}_\perp/|\mathbf{k}_\perp|$ as in the main text. Based on the equations of Sec. 3.1 and 3.2 in the main text and the incompressibility condition, one can express the pressure in terms of the relaxation rate of n_k as follows:

$$\left(\frac{i|\mathbf{k}_\perp|h}{2} \right) P = \left\{ \alpha_2 + \mathcal{H}^{\text{SB}} \left[\mathcal{H}^{\text{SB}} \right]_{\text{D,N}}^{-1} \mathcal{C}^{\text{SB}} \right\} (\partial_Z \partial_t n_k), \quad (44)$$

where we used the dimensionless coordinate $Z = 2z/h$ as in the main text. One immediately observe that if the right-hand-side of eqn (44) does not goes to zero when $\mathbf{k}_\perp \rightarrow \mathbf{0}$, the pressure will become singular. We therefore need to calculate and simplify the limit of the right-hand-side, which can be done analytically with the same method as in Sec 2.1 of this supplementary:

$$\lim_{\mathbf{k}_\perp \rightarrow \mathbf{0}} \left[\left(\frac{i|\mathbf{k}_\perp|h}{2} \right) P \right] = \alpha_2 [\langle u \rangle + 3Z \langle Z' u \rangle] \quad (45)$$

with $u \equiv \partial_Z' \partial_t n_k$. Let us calculate the right-hand-side of eqn (45) in the case of the fundamental eigenmode $\alpha = m = 0$, using the nomenclature of the main text. To simplify the calculation, let us assume that there is no chirality ($q = 0$), so that the eigenvalue ϕ_τ is the smallest positive solution of the dispersion relation $\cos \phi_\tau = \mu \operatorname{sinc} \phi_\tau$. For this particular eigenmode and using the results of Sec. 2.2.1 of this supplementary, we find the following profile for

the splay-bend component of the director field:

$$n^{\text{SB}} = A [\cos(\phi_\tau Z) - \mu \text{sinc } \phi_\tau] \quad (46)$$

Since $n_k = n^{\text{SB}} \exp[i\mathbf{k}_\perp \cdot \mathbf{r}_\perp - t/\tau]$, we can simplify eqn (45) using eqn (46) and the dispersion relation $\cos \phi_\tau = \mu \text{sinc } \phi_\tau$:

$$\lim_{\mathbf{k}_\perp \rightarrow \mathbf{0}} \left[\left(\frac{i|\mathbf{k}_\perp| h}{2} \right) P \right] = \frac{3\alpha_2 Z (1 - \mu) A \text{sinc } \phi_\tau}{\tau} \exp[i\mathbf{k}_\perp \cdot \mathbf{r}_\perp - t/\tau] \quad (47)$$

Since the right-hand-side of this equation is not zero, we conclude that the pressure becomes singular in the small wavevector limit. I emphasize that this result is not an artefact due to the calculation procedure, and was also observed numerically by solving the Stokes equation when the forcing term associated with director relaxation is a confined Fourier mode. As explained in the main text, preliminary calculations indicate that the only way to avoid this divergence is to take into account the compressibility of the fluid.

Notes and references

- 1 P. Oswald and P. Pieranski, *Nematic and Cholesteric Liquid Crystals: Concepts and Physical Properties Illustrated by Experiments*, CRC Press, Boca Raton, 2006.