Supplemental Material: Bifurcation analysis of twisted liquid crystal bilayers

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This Supplemental Material (SM) explicitly derives the matrices $\mathbf{Q}^{(r)}$, $\mathbf{H}_{c,s}^{(r)}$ and $\mathbf{F}_{c,s}^{(r)}$ (for r = 1, 2), that are appearing in the analytical solution of Section 5 of the main article. We first expand the governing equations of the main text (A5.5)¹

$$k_1^{(r)}(\Delta n_{2,22} + \Delta n_{3,32}) + k_2^{(r)}(\Delta n_{2,33} - \Delta n_{3,23}) + k_3^{(r)}\Delta n_{2,11} = 0,$$
(1)

$$k_{1}^{(r)}(\Delta n_{2,23} + \Delta n_{3,33}) + k_{2}^{(r)}(\Delta n_{3,22} - \Delta n_{2,32}) + k_{3}^{(r)}\Delta n_{3,11} + \frac{C_{\perp}^{(r)}}{C_{b}^{(r)}}d_{0}\left[C_{\perp}^{(r)}\frac{d_{0}}{\varepsilon_{0}}\Delta n_{3} + \Delta\varphi_{,1}\right] = 0, \quad (2)$$

$$\Delta\varphi_{,11} + \Delta\varphi_{,22} + \Delta\varphi_{,33} + \frac{C_{\perp}^{(r)}}{C_b^{(r)}} \left[\Delta\varphi_{,11} + C_{\perp}^{(r)} \frac{d_0}{\varepsilon_0} \Delta n_{3,1} \right] = 0.$$
(3)

Substituting the eigenmode (A5.8) to the above, we obtain

$$k_1^{(r)}(\omega_2^2 \Delta \mathcal{N}_2^{(r)} + \omega_2 \mathcal{N}_{3,3}^{(r)}) - k_2^{(r)}(\Delta \mathcal{N}_{2,33}^{(r)} + \omega_2 \Delta \mathcal{N}_{3,3}^{(r)}) + k_3^{(r)}\omega_1^2 \Delta \mathcal{N}_2^{(r)} = 0,$$

$$k_1^{(r)}(\omega_2 \Delta \mathcal{N}_{2,3}^{(r)} + \Delta \mathcal{N}_{3,33}^{(r)}) -$$

$$(4)$$

$$k_{2}^{(r)}(\omega_{2}^{2}\Delta\mathcal{N}_{3}^{(r)} + \omega_{2}\Delta\mathcal{N}_{2,3}^{(r)}) - k_{3}^{(r)}\omega_{1}^{2}\Delta\mathcal{N}_{3}^{(r)} + C^{(r)}d_{0}\left[C_{\perp}^{(r)}\frac{d_{0}}{\varepsilon_{0}}\Delta\mathcal{N}_{3}^{(r)} + \omega_{1}\Delta\Phi^{(r)}\right] = 0,$$
(5)

$$-\omega_1^2 \Delta \Phi^{(r)} - \omega_2^2 \Delta \Phi^{(r)} + \Delta \Phi^{(r)}_{,33} - C^{(r)} \left[\omega_1^2 \Delta \Phi^{(r)} + C_{\perp}^{(r)} \frac{d_0}{\varepsilon_0} \omega_1 \Delta \mathcal{N}_3^{(r)} \right] = 0, \tag{6}$$

where $C^{(r)} = C_{\perp}^{(r)}/C_b^{(r)}$. One can show that the above set of ordinary differential equations admit a general solution of the form (A5.9). The above, upon substitution of (A5.9) yields a set of three homogeneous algebraic equations (A5.10), where $\mathbf{Q}^{(r)}$ is given by

$$\mathbf{Q}^{(r)} \equiv \begin{bmatrix} k_1^{(r)}\omega_2^2 + k_3^{(r)}\omega_1^2 - k_2^{(r)}\rho^2 & (k_1^{(r)} - k_2^{(r)})\omega_2\rho & 0\\ (k_1^{(r)} - k_2^{(r)})\omega_2\rho & -k_2^{(r)}\omega_2^2 - k_3^{(r)}\omega_1^2 + k_1^{(r)}\rho^2 + C^{(r)}C_{\perp}^{(r)}d_0^2/\varepsilon_0 & C^{(r)}d_0\omega_1\\ 0 & C^{(r)}C_{\perp}^{(r)}d_0\omega_1/\varepsilon_0 & (1+C^{(r)})\omega_1^2 + \omega_2^2 - \rho^2 \end{bmatrix}.$$
(7)

To obtain a non-trivial solution of (A5.10), the condition det[$\mathbf{Q}^{(r)}$] = 0 must be satisfied. This condition leads to a characteristic bi-cubic polynomial in ρ given by (A5.11). Subsequently, the eigenmodes $\Delta \hat{\mathcal{N}}_1$ and $\Delta \hat{\mathcal{N}}_2$ are given in (A5.12).

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¹henceforth the letter A will be used to denote the corresponding equation number in the main article.

Then, it only remains to calculate the unknown amplitudes Ξ from the interface conditions (A5.6), (A5.7) and the boundary conditions (A4.3) and (A5.2). For that, we write the interface conditions $(A5.6)_1$ and $(A5.7)_1$ at $x_3 = 0$, such that

$$[[\Delta \mathcal{N}_2]] = 0 \implies \sum_{I=1}^3 (\xi_I^{c,(2)} \Delta \widehat{\mathcal{N}}_2^{I,(2)} - \xi_I^{c,(1)} \Delta \widehat{\mathcal{N}}_2^{I,(1)}) = 0, \tag{8}$$

$$[\![\Delta \mathcal{N}_3]\!] = 0 \implies \sum_{I=1}^3 (\xi_I^{s,(2)} \Delta \widehat{\mathcal{N}}_3^{I,(2)} - \xi_I^{s,(1)} \Delta \widehat{\mathcal{N}}_3^{I,(1)}) = 0, \tag{9}$$

$$[\![\Delta\Phi]\!] = 0 \implies \sum_{I=1}^{3} (\xi_{I}^{s,(2)} - \xi_{I}^{s,(1)}) = 0.$$
(10)

Similarly, the interface conditions $(A5.6)_2$ and $(A5.7)_3$ are now obtained explicitly as

$$\begin{bmatrix} \left[\mathcal{L}_{23kl}^{\nabla n \nabla n, (r)} \Delta n_{k,l} \right] \right] = 0 \implies \left[\left[k_{2}^{(r)} (\Delta n_{2,3} - \Delta n_{3,2}) \right] \right] = 0$$

$$\implies \sum_{I=1}^{3} \left[k_{2}^{(2)} \xi_{I}^{s,(2)} \left(\rho_{I}^{(2)} \Delta \widehat{\mathcal{N}}_{2}^{I,(2)} + \omega_{2} \Delta \widehat{\mathcal{N}}_{3}^{I,(2)} \right) - k_{2}^{(1)} \xi_{I}^{s,(1)} \left(\rho_{I}^{(1)} \Delta \widehat{\mathcal{N}}_{2}^{I,(1)} + \omega_{2} \Delta \widehat{\mathcal{N}}_{3}^{I,(1)} \right) \right] = 0, \quad (11)$$

$$\begin{bmatrix} \left[\mathcal{L}_{33kl}^{\nabla n \nabla n, (r)} \Delta n_{k,l} \right] \right] = 0 \implies \left[\left[k_{1}^{(r)} (\Delta n_{2,2} + \Delta n_{3,3}) \right] \right] = 0$$

$$\implies \sum_{I=1}^{3} \left[k_{1}^{(2)} \xi_{I}^{c,(2)} \left(\omega_{2} \Delta \widehat{\mathcal{N}}_{2}^{I,(2)} + \rho_{I}^{(2)} \Delta \widehat{\mathcal{N}}_{3}^{I,(2)} \right) - k_{1}^{(1)} \xi_{I}^{c,(1)} \left(\omega_{2} \Delta \widehat{\mathcal{N}}_{2}^{I,(1)} + \rho_{I}^{(1)} \Delta \widehat{\mathcal{N}}_{3}^{I,(1)} \right) \right] = 0, \quad (12)$$

$$\begin{bmatrix} \left[\mathcal{L}_{3j}^{n \nabla \varphi, (r)} \Delta n_{j} + \mathcal{L}_{3j}^{\nabla \varphi \nabla \varphi, (r)} \Delta \varphi_{,j} \right] \right] = 0 \implies \left[\left[\frac{\varepsilon_{0}}{C_{\perp}^{(r)}} \Delta \Phi_{,3} \right] \right] = 0$$

$$\implies \sum_{I=1}^{3} \left(\frac{\varepsilon_{0}}{C_{\perp}^{(2)}} \rho_{I}^{(2)} \xi_{I}^{c,(2)} - \frac{\varepsilon_{0}}{C_{\perp}^{(1)}} \rho_{I}^{(1)} \xi_{I}^{c,(1)} \right) = 0. \quad (13)$$

Equations (8), (12) and (13) are now re-written in the following matrix form:

$$\mathbf{H}_{c}^{(2)}\boldsymbol{\xi}^{c,(2)} = \mathbf{H}_{c}^{(1)}\boldsymbol{\xi}^{c,(1)},\tag{14}$$

where

$$\mathbf{H}_{c}^{(r)} \equiv \begin{bmatrix} \Delta \widehat{\mathcal{N}}_{2}^{1,(r)} & \Delta \widehat{\mathcal{N}}_{2}^{2,(r)} & \Delta \widehat{\mathcal{N}}_{2}^{3,(r)} \\ k_{1}^{(r)} \left(\omega_{2} \Delta \widehat{\mathcal{N}}_{2}^{1,(r)} + \rho_{1}^{(r)} \Delta \widehat{\mathcal{N}}_{3}^{1,(r)} \right) & k_{1}^{(r)} \left(\omega_{2} \Delta \widehat{\mathcal{N}}_{2}^{2,(r)} + \rho_{2}^{(r)} \Delta \widehat{\mathcal{N}}_{3}^{2,(r)} \right) & k_{1}^{(r)} \left(\omega_{2} \Delta \widehat{\mathcal{N}}_{2}^{3,(r)} + \rho_{3}^{(r)} \Delta \widehat{\mathcal{N}}_{3}^{3,(r)} \right) \\ \varepsilon_{0} \rho_{1}^{(r)} / C_{\perp}^{(r)} & \varepsilon_{0} \rho_{2}^{(r)} / C_{\perp}^{(r)} & \varepsilon_{0} \rho_{3}^{(r)} / C_{\perp}^{(r)} \end{bmatrix}$$

with r = 1, 2. Similarly, (9 - 11) are now expressed as

$$\mathbf{H}_{s}^{(2)}\boldsymbol{\xi}^{s,(2)} = \mathbf{H}_{s}^{(1)}\boldsymbol{\xi}^{s,(1)},\tag{15}$$

where

$$\mathbf{H}_{s}^{(r)} \equiv \begin{bmatrix} \Delta \hat{\mathcal{N}}_{3}^{1,(r)} & \Delta \hat{\mathcal{N}}_{3}^{2,(r)} & \Delta \hat{\mathcal{N}}_{3}^{3,(r)} \\ 1 & 1 & 1 \\ k_{2}^{(r)} \left(\rho_{1}^{(r)} \Delta \widehat{\mathcal{N}}_{2}^{1,(r)} + \omega_{2} \Delta \widehat{\mathcal{N}}_{3}^{1,(r)} \right) & k_{2}^{(r)} \left(\rho_{2}^{(r)} \Delta \widehat{\mathcal{N}}_{2}^{2,(r)} + \omega_{2} \Delta \widehat{\mathcal{N}}_{3}^{2,(r)} \right) & k_{2}^{(r)} \left(\rho_{3}^{(r)} \Delta \widehat{\mathcal{N}}_{2}^{3,(r)} + \omega_{2} \Delta \widehat{\mathcal{N}}_{3}^{3,(r)} \right) \end{bmatrix}.$$

The boundary conditions $(A4.3)_3$ and $(A5.2)_3$ lead to

$$\Delta \mathcal{N}_2 = 0 \quad \Longrightarrow \quad \sum_{I=1}^3 \{\xi_I^{s,(2)} \sinh(\rho_I^{(2)} \ell_3^{(2)}) + \xi_I^{c,(2)} \cosh(\rho_I^{(2)} \ell_3^{(2)})\} \Delta \widehat{\mathcal{N}}_2^{I,(2)} = 0, \tag{16}$$

$$\Delta \mathcal{N}_3 = 0 \quad \Longrightarrow \quad \sum_{I=1}^3 \{\xi_I^{s,(2)} \cosh(\rho_I^{(2)} \ell_3^{(2)}) + \xi_I^{c,(2)} \sinh(\rho_I^{(2)} \ell_3^{(2)})\} \Delta \widehat{\mathcal{N}}_3^{I,(2)} = 0, \tag{17}$$

$$\Delta \Phi = 0 \quad \Longrightarrow \quad \sum_{I=1}^{3} \{\xi_{I}^{s,(2)} \cosh(\rho_{I}^{(2)} \ell_{3}^{(2)}) + \xi_{I}^{c,(2)} \sinh(\rho_{I}^{(2)} \ell_{3}^{(2)})\} = 0.$$
(18)

The above equations can be expressed in matrix form as

$$\mathbf{F}_{c}^{(2)}\boldsymbol{\xi}^{c,(2)} = \mathbf{F}_{s}^{(2)}\boldsymbol{\xi}^{s,(2)},\tag{19}$$

where

$$\mathbf{F}_{c}^{(2)} \equiv \begin{bmatrix} \cosh(\rho_{1}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{2}^{1,(2)} & \cosh(\rho_{2}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{2}^{2,(2)} & \cosh(\rho_{3}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{2}^{3,(2)} \\ \sinh(\rho_{1}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{3}^{1,(2)} & \sinh(\rho_{2}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{3}^{2,(2)} & \sinh(\rho_{3}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{3}^{3,(2)} \\ \sinh(\rho_{1}^{(2)}\ell_{3}^{(2)}) & \sinh(\rho_{2}^{(2)}\ell_{3}^{(2)}) & \sinh(\rho_{3}^{(2)}\ell_{3}^{(2)}) \end{bmatrix},$$

and

$$\mathbf{F}_{s}^{(2)} \equiv \begin{bmatrix} -\sinh(\rho_{1}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{2}^{1,(2)} & -\sinh(\rho_{2}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{2}^{2,(2)} & -\sinh(\rho_{3}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{2}^{3,(2)} \\ -\cosh(\rho_{1}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{3}^{1,(2)} & -\cosh(\rho_{2}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{3}^{2,(2)} & -\cosh(\rho_{3}^{(2)}\ell_{3}^{(2)})\Delta\widehat{\mathcal{N}}_{3}^{3,(2)} \\ -\cosh(\rho_{1}^{(2)}\ell_{3}^{(2)}) & -\cosh(\rho_{2}^{(2)}\ell_{3}^{(2)}) & -\cosh(\rho_{3}^{(2)}\ell_{3}^{(2)}) \end{bmatrix}.$$

Similarly, from $(A4.3)_1$ and $(A5.2)_1$, we obtain

$$\mathbf{F}_{c}^{(1)}\boldsymbol{\xi}^{c,(1)} = \mathbf{F}_{s}^{(1)}\boldsymbol{\xi}^{s,(1)},\tag{20}$$

where the matrices $\mathbf{F}_{c}^{(1)}$ and $\mathbf{F}_{s}^{(1)}$ are given by

$$\mathbf{F}_{c}^{(1)} \equiv \begin{bmatrix} \cosh(\rho_{1}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{2}^{1,(1)} & \cosh(\rho_{2}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{2}^{2,(1)} & \cosh(\rho_{3}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{2}^{3,(1)} \\ -\sinh(\rho_{1}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{3}^{1,(1)} & -\sinh(\rho_{2}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{3}^{2,(1)} & -\sinh(\rho_{3}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{3}^{3,(1)} \\ -\sinh(\rho_{1}^{(1)}\ell_{3}^{(1)}) & -\sinh(\rho_{2}^{(1)}\ell_{3}^{(1)}) & -\sinh(\rho_{3}^{(1)}\ell_{3}^{(1)}) \end{bmatrix}$$

 $\quad \text{and} \quad$

$$\mathbf{F}_{s}^{(1)} \equiv \begin{bmatrix} \sinh(\rho_{1}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{2}^{1,(1)} & \sinh(\rho_{2}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{2}^{2,(1)} & \sinh(\rho_{3}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{2}^{3,(1)} \\ -\cosh(\rho_{1}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{3}^{1,(1)} & -\cosh(\rho_{2}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{3}^{2,(1)} & -\cosh(\rho_{3}^{(1)}\ell_{3}^{(1)})\Delta\widehat{\mathcal{N}}_{3}^{3,(1)} \\ -\cosh(\rho_{1}^{(1)}\ell_{3}^{(1)}) & -\cosh(\rho_{2}^{(1)}\ell_{3}^{(1)}) & -\cosh(\rho_{3}^{(1)}\ell_{3}^{(1)}) \end{bmatrix}$$

Finally, from (14), (15), (19) and (20) we obtain the set of 12 homogeneous algebraic equations, which is put in the compact form

$$\underbrace{\begin{bmatrix} -\mathbf{H}_{c}^{(1)} & \mathbf{0} & \mathbf{H}_{c}^{(2)} & \mathbf{0} \\ \mathbf{0} & -\mathbf{H}_{s}^{(1)} & \mathbf{0} & \mathbf{H}_{s}^{(2)} \\ \mathbf{0} & \mathbf{0} & \mathbf{F}_{c}^{(2)} & -\mathbf{F}_{s}^{(2)} \\ \mathbf{F}_{c}^{(1)} & -\mathbf{F}_{s}^{(1)} & \mathbf{0} & \mathbf{0} \end{bmatrix}}_{\mathbf{M}(\omega_{1},\omega_{2},d_{0})} \underbrace{\begin{bmatrix} \boldsymbol{\xi}^{c,(1)} \\ \boldsymbol{\xi}^{s,(1)} \\ \boldsymbol{\xi}^{c,(2)} \\ \boldsymbol{\xi}^{s,(2)} \\ \boldsymbol{\xi}^{s,(2)}$$

In this expression, \mathbf{M} is the matrix defined in equation (A5.13) in the main text.