Supplementary Information (SI)

Orientational Order Induced Mode Switching at Coupled Interfaces of Nematic-Isotropic Free Bilayer

Vinod Babasaheb Vanarse[1], Neeru Yadav[1], Srijita De[2], Abir Ghosh[3], Tapas Kumar Mandal[1,2,4], Dipankar Bandyopadhyay[1,2,4 @]

[1]Department of Chemical Engineering, Indian Institute of Technology, Guwahati 781039, Assam, India.
[2]Centre for Nanotechnology, Indian Institute of Technology, Guwahati 781039, Assam, India.
[3]Department of Chemical Engineering and Technology, Indian Institute of Technology (BHU) Varanasi, Varanasi 221005, India
[4]Jyoti and Bhupat Mehta School of Health Sciences and Technology, Indian Institute of Technology, Guwahati 781039, Assam, India

[@]Email address for correspondence: dipban@iitg.ac.in

Appendix A

Mass conservation and equation of motion

Momentum balance equation for layer 1:

$$\rho_1\left(\frac{D\boldsymbol{v}}{Dt}\right) = \boldsymbol{\nabla}\cdot(-p_1\boldsymbol{I}) + \boldsymbol{\nabla}\cdot\boldsymbol{\tau} \tag{A1}$$

where,

$$\boldsymbol{\tau} = \mu_1(\boldsymbol{\nabla u} + (\boldsymbol{\nabla u})^{\mathrm{T}})$$

After applying the scaling rules the x and z direction momentum balance equations are respectively modified to,

$$\frac{\partial P_1}{\partial x} = \mu_1\left(\frac{\partial^2 u_1}{\partial z^2}\right) \tag{A2}$$

$$\frac{\partial P_1}{\partial z} = 0 \tag{A3}$$

NLC description: The couple balance or the angular momentum balance equation for NLC

$$\lambda\boldsymbol{n} - \frac{\partial E_f}{\partial \boldsymbol{n}} + \boldsymbol{\nabla}\cdot\left(\frac{\partial E_f}{\partial \boldsymbol{\nabla n}}\right) + \boldsymbol{G} + \boldsymbol{g} = 0 \tag{A4}$$

As there is no external field, therefore $\boldsymbol{G} = 0$ and $\boldsymbol{g} = -\lambda_1\boldsymbol{N} - \lambda_2(\boldsymbol{e}\cdot\boldsymbol{n})$ where, $\boldsymbol{N}$ is the rotation vector and can be expressed as, $\boldsymbol{N} = \dot{\boldsymbol{n}} - \boldsymbol{\omega}\cdot\boldsymbol{n}$, whereas, $\boldsymbol{\omega}$ is the spin tensor, $\boldsymbol{\omega} = 0.5(\boldsymbol{\nabla u} - \boldsymbol{\nabla u}^{\mathrm{T}})$ and $\boldsymbol{e}$ is the strain tensor, which is $\boldsymbol{e} = 0.5(\boldsymbol{\nabla u} + \boldsymbol{\nabla u}^{\mathrm{T}})$.

After expanding the x and z direction components and applying scaling laws the final angular momentum balance equation reads as follows,

$$-K_f\left(\frac{\partial^2\theta}{\partial z^2}\right)=0 \tag{A5}$$

Momentum balance equation for NLC-layer 2:

$$\rho_2\left(\frac{D\boldsymbol{v}}{Dt}\right)=\boldsymbol{\nabla}\cdot(-p_2\boldsymbol{I})+\boldsymbol{\nabla}\cdot\boldsymbol{\tau}^{\mathrm{V}}+\boldsymbol{\nabla}\cdot\boldsymbol{\tau}^{\mathrm{E}} \tag{A6}$$

The term $\boldsymbol{\tau}^{\mathrm{V}}$ in Eq. (6) is associated with the viscous stress tensor for the NLC film,

$$\boldsymbol{\tau}^{\mathrm{V}}=\alpha_1\boldsymbol{e}{:}\boldsymbol{nnnn}+\alpha_2\boldsymbol{nN}+\alpha_3\boldsymbol{nN}+\alpha_4\boldsymbol{e}+\alpha_5\boldsymbol{nn}\cdot\boldsymbol{e}+\alpha_6\boldsymbol{e}\cdot\boldsymbol{nn}$$

where, $\boldsymbol{e_{ij}}=\frac{1}{2}\left(\frac{\partial\boldsymbol{u}_i}{\partial x_j}+\frac{\partial\boldsymbol{u}_j}{\partial x_i}\right)$, $\boldsymbol{N_i}=\frac{D\boldsymbol{n}_i}{Dt}-\boldsymbol{w}_{ik}\boldsymbol{n}_k$ and $\boldsymbol{w_{ij}}=\frac{1}{2}\left(\frac{\partial\boldsymbol{u}_i}{\partial x_j}-\frac{\partial\boldsymbol{u}_j}{\partial x_i}\right)$, respectively.

The viscous stress tensor is expressed as $\boldsymbol{\tau}^{\mathrm{V}}=\begin{bmatrix}\boldsymbol{\tau}_{xx} & \boldsymbol{\tau}_{xz}\\ \boldsymbol{\tau}_{zx} & \boldsymbol{\tau}_{zz}\end{bmatrix}$, where the matrix components are

$$\boldsymbol{\tau}_{xx}=a_1\frac{\partial u}{\partial x}+a_2\frac{\partial u}{\partial z}+a_3\frac{\partial w}{\partial x}+a_4\frac{\partial w}{\partial z}+a_5\frac{D\theta}{Dt} \tag{A7}$$

$$\boldsymbol{\tau}_{xz}=b_1\frac{\partial u}{\partial x}+b_2\frac{\partial u}{\partial z}+b_3\frac{\partial w}{\partial x}+b_4\frac{\partial w}{\partial z}+b_5\frac{D\theta}{Dt} \tag{A8}$$

$$\boldsymbol{\tau}_{zx}=c_1\frac{\partial u}{\partial x}+c_2\frac{\partial u}{\partial z}+c_3\frac{\partial w}{\partial x}+c_4\frac{\partial w}{\partial z}+c_5\frac{D\theta}{Dt} \tag{A9}$$

$$\boldsymbol{\tau}_{zz}=d_1\frac{\partial u}{\partial x}+d_2\frac{\partial u}{\partial z}+d_3\frac{\partial w}{\partial x}+d_4\frac{\partial w}{\partial z}+d_5\frac{D\theta}{Dt} \tag{A10}$$

The variables a_i, b_i, c_i and d_i utilized in Eq's. (A7–A10) are given below:

$$a_1=[\alpha_1\sin^4\theta+\ \alpha_4+\ (\alpha_5+\alpha_6)\sin^2\theta], \tag{A11}$$

$$a_2=\sin\theta\cos\theta\left[\alpha_1\sin\theta\cos\theta+\frac{1}{2}\ (\alpha_5+\alpha_6-\alpha_2-\alpha_3)\right], \tag{A12}$$

$$a_3=\sin\theta\cos\theta\left[\alpha_1\sin\theta\cos\theta+\frac{1}{2}\ (\alpha_5+\alpha_6+\alpha_2+\alpha_3)\right], \tag{A13}$$

$$a_4=[\alpha_1\sin^2\theta\cos^2\theta], \tag{A14}$$

$$a_5=[\alpha_1+\alpha_3]\sin\theta\cos\theta, \tag{A15}$$

$$b_1=\sin\theta\cos\theta[\alpha_1\sin^2\theta+\ \alpha_5], \tag{A16}$$

$$b_2=\left[\alpha_1\sin^2\theta\cos^2\theta+\ \frac{\alpha_4}{2}+\frac{\cos^2\theta}{2}(\alpha_5-\alpha_2)+\frac{\sin^2\theta}{2}(\alpha_6+\alpha_3)\right], \tag{A17}$$

$$b_3 = \left[\alpha_1 \sin^2\theta \cos^2\theta + \frac{\alpha_4}{2} + \frac{\cos^2\theta}{2}(\alpha_5 + \alpha_2) + \frac{\sin^2\theta}{2}(\alpha_6 - \alpha_3)\right], \tag{A18}$$

$$b_4 = \sin\theta\cos\theta[\alpha_1 \cos^2\theta + \alpha_6], \tag{A19}$$

$$b_5 = [\alpha_2 \cos^2\theta - \alpha_3 \sin^2\theta], \tag{A20}$$

$$c_1 = \sin\theta\cos\theta[\alpha_1 \sin^2\theta + \alpha_6], \tag{A21}$$

$$c_2 = \left[\alpha_1 \sin^2\theta \cos^2\theta + \frac{\alpha_4}{2} + \frac{\cos^2\theta}{2}(\alpha_6 - \alpha_3) + \frac{\sin^2\theta}{2}(\alpha_2 + \alpha_5)\right], \tag{A22}$$

$$c_3 = \left[\alpha_1 \sin^2\theta \cos^2\theta + \frac{\alpha_4}{2} + \frac{\cos^2\theta}{2}(\alpha_3 + \alpha_6) + \frac{\sin^2\theta}{2}(\alpha_5 - \alpha_2)\right], \tag{A23}$$

$$c_4 = \sin\theta\cos\theta[\alpha_1 \cos^2\theta + \alpha_5], \tag{A24}$$

$$c_5 = [\alpha_3 \cos^2\theta - \alpha_2 \sin^2\theta], \tag{A25}$$

$$d_1 = [\alpha_1 \sin^2\theta \cos^2\theta], \tag{A26}$$

$$d_2 = \sin\theta\cos\theta\left[\alpha_1 \cos^2\theta + \frac{1}{2}(\alpha_5 + \alpha_6 + \alpha_2 + \alpha_3)\right], \tag{A27}$$

$$d_3 = \sin\theta\cos\theta\left[\alpha_1 \cos^2\theta + \frac{1}{2}(\alpha_5 + \alpha_6 - \alpha_2 - \alpha_3)\right], \tag{A28}$$

$$d_4 = [\alpha_1 \cos^4\theta + \alpha_4 + (\alpha_5 + \alpha_6)\cos^2\theta], \tag{A29}$$

$$d_5 = [\alpha_2 \cos^2\theta - \alpha_3 \sin^2\theta]. \tag{A30}$$

Similarly, the elastic (Ericksen) stress tensor for the NLC film $\boldsymbol{\tau}^{\mathrm{E}}$ describes as,

$$\boldsymbol{\tau}^{\mathrm{E}} = -\left(\frac{\partial E_f}{\partial \nabla \boldsymbol{n}}\right) \cdot (\nabla \boldsymbol{n})^{\mathrm{T}} \tag{A31}$$

where, $E_f = \frac{K_f}{2}[(\nabla \cdot \boldsymbol{n})^2 + (\boldsymbol{n} \times \nabla \times \boldsymbol{n})^2] = \frac{K_f}{2}[(\nabla \boldsymbol{n}) : (\nabla \boldsymbol{n})^{\mathrm{T}}]$, therefore on simplification, Eq. (11) takes the form as follows,

$$\boldsymbol{\tau}^{\mathrm{E}} = -K_f[(\nabla \boldsymbol{n}) \cdot (\nabla \boldsymbol{n})^{\mathrm{T}}] = -K_f \begin{bmatrix} \left(\frac{\partial\theta}{\partial x}\right)^2 & \left(\frac{\partial\theta}{\partial x}\right)\left(\frac{\partial\theta}{\partial z}\right) \\ \left(\frac{\partial\theta}{\partial z}\right)\left(\frac{\partial\theta}{\partial x}\right) & \left(\frac{\partial\theta}{\partial z}\right)^2 \end{bmatrix} \tag{A32}$$

After considering the scaling parameters, and performing all the simplifications, final forms of momentum balance equations for the x and z directions take the following forms,

$$\frac{\partial}{\partial x}\left(P_2 + \frac{K_f}{2}\left(\frac{\partial\theta}{\partial z}\right)^2\right) = \frac{\partial}{\partial z}\left(c_2 \frac{\partial u_2}{\partial z}\right) \tag{A33}$$

$$\frac{\partial}{\partial z}\left(P_2 + \frac{K_f}{2}\left(\frac{\partial\theta}{\partial z}\right)^2\right) = 0 \tag{A34}$$

Appendix B

Boundary conditions and thin film evolution equations:

The integration of the momentum Eq. (A2) gives,

$$\mu_1\left(\frac{\partial u_1}{\partial z}\right)=\frac{\partial P_1}{\partial x}z+C_1 \tag{B35}$$

$$\mu_1 u_1=\frac{\partial P_1}{\partial x}z^2+C_1 z+C_2 \tag{B36}$$

where, $C_1,\ C_2$ are integration constants.

Boundary condition at $z=0$, $u_1=w_1=0$ (no slip and impermeable boundary conditions) lead to $C_2=0$. Utilizing the boundary condition at $z=h_1$, (tangential stress balance) lead to

$$c_2\left(\frac{\partial u_2}{\partial z}\right)=\mu_1\frac{\partial u_1}{\partial z} \tag{B37}$$

where,

$$c_2=\alpha_1\sin^2\theta\cos^2\theta+\frac{\alpha_4}{2}+\frac{\cos^2\theta}{2}(\alpha_6-\alpha_3)+\frac{\sin^2\theta}{2}(\alpha_2+\alpha_5)$$

Applying tangential boundary condition, which leads to,

$$C_1=\frac{\partial P_{2LC}}{\partial x}(h_1-h_2)-\frac{\partial P_1}{\partial x}h_1 \tag{B38}$$

where,

$$P_{2LC=}P_2+\frac{K_f}{2}\left(\frac{\partial\theta}{\partial z}\right)^2$$

On inserting the obtained integration constants, we obtain the velocity u_1 as,

$$(u_1)_{h_1}=\frac{1}{\mu_1}\left[\frac{\partial P_{2LC}}{\partial x}(h_1-h_2)z+\frac{\partial P_1}{\partial x}\left(\frac{z^2}{2}-h_1 z\right)\right] \tag{B39}$$

where, $(u_1)_{h_1}$is the u_1 solved at h_1 height.

Differentiating Eq. (19) with respect to z leads to,

$$\left(\frac{\partial u_1}{\partial z}\right)_{h_1}=\frac{1}{\mu_1}\left[\frac{\partial P_{2LC}}{\partial x}(h_1-h_2)+\frac{\partial P_1}{\partial x}(z-h_1)\right] \tag{B40}$$

The momentum balance equation for layer-2 for the x and z directions, respectively

$$\frac{\partial}{\partial x}(P_{2LC})=\frac{\partial}{\partial z}\left(c_2\frac{\partial u_2}{\partial z}\right) \tag{B41}$$

$$\frac{\partial}{\partial z}(P_{2LC})=0 \tag{B42}$$

The integration of these momentum equations gives,

$$c_2\left(\frac{\partial u_2}{\partial z}\right) = \frac{\partial P_{2LC}}{\partial x} z + C_3 \tag{B43}$$

with p_{2LC} is constant in the z-direction.

Boundary condition at $z = h_2$, (tangential stress balance) suggests that $\frac{\partial u_2}{\partial z} = 0$, which gives $C_3 = \frac{\partial P_{2LC}}{\partial x} h_2$ and on simplification it turns out to be,

$$\left(\frac{\partial u_2}{\partial z}\right)_{h_2} = \frac{\partial P_{2LC}}{\partial x}\left(\frac{z - h_2}{c_2}\right) \tag{B44}$$

The thin film equation for layer 1 is described as,

$$\frac{\partial h_1}{\partial t} + \frac{\partial}{\partial x}\int_0^{h_1}\left(\frac{\partial u_1}{\partial z}\right)_{h_1}(h_1 - z)dz = 0 \tag{B45}$$

After inserting, solving and on simplification Eq. (A25) leads to following form,

$$\frac{\partial h_1}{\partial t} - \frac{\partial^2 P_{2\mathrm{LC}}}{\partial x^2}\left(\frac{h_1^2 h_2 - h_1^3}{2\mu_1}\right) - \frac{\partial^2 P_1}{\partial x^2}\left(\frac{h_1^3}{2\mu_1}\right) - \frac{\partial P_1}{\partial x}\left(\frac{h_1^2}{\mu_1}\frac{\partial h_1}{\partial x}\right) + \frac{\partial P_{2LC}}{\partial x}\left(\frac{\partial h_1}{\partial x}\left(\frac{3h_1^2 - 2h_1 h_2}{2\mu_1}\right) - \frac{\partial h_2}{\partial x}\left(\frac{h_1^2}{2\mu_1}\right)\right) = 0 \tag{B46}$$

Similarly, the thin film equation for layer 2 reads as follows,

$$\frac{\partial(h_2 - h_1)}{\partial t} + \frac{\partial}{\partial x}\left[\left(\int_{h_1}^{h_2}\left(\frac{\partial u_2}{\partial z}\right)_{h_2}(h_2 - z)dz\right) + (h_2 - h_1)(u_2)_{h_1}\right] = 0 \tag{B47}$$

At $z = h_1$: $(u_2)_{h_1} = (u_1)_{h_1}$

On integrating the Eq. (A27), it takes the form as follows,

$$\begin{aligned}\frac{\partial h_2}{\partial t} &- \frac{\partial^2 P_1}{\partial x^2}\left(\frac{h_1^2 h_2 - h_1^3}{2\mu_1} + \frac{h_1^3}{3\mu_1}\right) \\ &- \frac{\partial^2 P_{2\mathrm{LC}}}{\partial x^2}\left(\frac{h_1^2 h_2 - h_1^3}{2\mu_1} + \frac{h_1^3 - 2h_1^2 h_2 + h_2^2 h_1}{\mu_1} + \left(\frac{h_1 - h_2}{\theta_1 - \theta_2}\right)^3 \frac{I_1}{\mu_2}\right) \\ &+ \frac{\partial P_{2\mathrm{LC}}}{\partial x}\frac{\partial}{\partial x}\left(-\left(\frac{h_1 - h_2}{\theta_1 - \theta_2}\right)^3 \frac{I_1}{\mu_2} - \frac{h_1^3 - 2h_1^2 h_2 + h_2^2 h_1}{\mu_1} \right. \\ &\left. + \frac{h_1^2}{2\mu_1}(h_1 - h_2)\right) + \frac{\partial P_1}{\partial x}\frac{\partial}{\partial x}\left(\frac{-h_1^2 h_2 + h_1^3}{2\mu_1} - \frac{h_1^3}{3\mu_1}\right) = 0\end{aligned} \tag{B48}$$

where, $I_1 = \int_{\theta_1}^{\theta_2} \frac{(\theta-\theta_2)^2}{(K_1+K_2\sin^2\theta+K_3\sin^4\theta)} d\theta$; $K_1 = \frac{\alpha_4-\alpha_3+\alpha_6}{\alpha_4}$; $K_2 = \frac{2\alpha_1+\alpha_2+\alpha_5+\alpha_3-\alpha_6}{\alpha_4}$; $K_3 = -\frac{2\alpha_1}{\alpha_4}$; and $\theta = \theta_2 + \frac{\theta_2-\theta_1}{h_2-h_1}(z-h_2)$. The NLC bulk viscosity is defined by $\mu_2 = 0.5\alpha_4$, where, α_4 is the fourth Leslie coefficient.

Appendix C

***Micro-thin* film linear stability analysis**

The total pressure at NW interface is $P_1 = p_1 - \Pi_1$ and Π_1 is the disjoining pressure at the liquid-NLC interface. Similarly, the total pressure at NI surface is $P_2 = p_2 - \Pi_2$ and Π_2 is the disjoining pressure at the NI surface. The disjoining pressure is defined in terms of Gibbs free energy $\left(\Pi = -\frac{\partial(\Delta G_{\mathrm{E}}^{\mathrm{LW}})}{\partial \mathrm{h}}\right)$ as follows,

$$\Delta G_{\mathrm{E}}^{\mathrm{LW}} = \frac{-A_{11} + A_{1s} - A_{2s} + A_{12}}{12\pi h_1^2} + \frac{-A_{22} + A_{12}}{12\pi h_3^2} + \frac{A_{2s} - A_{12}}{12\pi(h_1 + h_3)^2} + \frac{K_f \theta^2}{2h_3} \tag{C49}$$

Here,

$$p_1 = p_0 - \gamma_2 h_{2xx} - \gamma_{12} h_{1xx} \text{ and}$$
$$\Pi_1 = -\frac{A_2}{6\pi {h_1}^3} + \frac{A_3}{6\pi {h_2}^3} \tag{C50}$$

where, $h_{2xx} = \frac{\partial^2 h_2}{\partial x^2}$. Similarly, p_2 reads as follows and terms $A_1 = A_{22} - A_{12}$, $A_2 = A_{11} + A_{S2} - A_{S1} - A_{12}$ and $A_3 = A_{S2} - A_{12}$, respectively.

$$p_2 = p_0 - K_f \left(\frac{\partial \theta}{\partial z}\right)^2 - \gamma_2 h_{2xx} - \Pi_2 \text{ and}$$
$$\Pi_2 = -\frac{A_1}{6\pi(h_2 - h_1)^3} + \frac{A_3}{6\pi h_2^3} + \frac{K_f \theta^2}{2(h_2 - h_1)^2} - \frac{K_f C_{LC} \theta}{(h_2 - h_1)} \tag{C51}$$

where, $C_{LC} = \frac{\Delta\theta}{h_3}$ is a constant and obtained from the angular momentum balance equation. The binary Hamaker constants are calculated using the surface tensions. As mentioned in the main manuscript, $A_{lm} = 12\pi {d_0}^2(\gamma_l + \gamma_m - \gamma_{lm})$, where, d_0(~0.157 nm) is the van der Waals cut-off distance between the two surfaces and $\gamma_{lm} = \gamma_l + \gamma_m - 2\sqrt{\gamma_l \gamma_m}$.

Thus, the double differentiation of Eq's. (C50, C51) lead to following forms,

$$P_{1xx} = -\gamma_2 h_{2xxxx} - \gamma_{12} h_{1xxxx} - \frac{A_2}{2\pi {h_1}^4} h_{1xx} + \frac{A_3}{2\pi {h_2}^4} h_{2xx} \tag{C52}$$

$$P_{2LCxx} = -\gamma_2 h_{2xxxx} - \frac{A_1}{2\pi(h_2 - h_1)^4}(h_{2xx} - h_{1xx}) + \frac{A_3}{2\pi {h_2}^4} h_{2xx} + \frac{K_f((\Delta\theta)^2 - (\theta)^2)}{(h_2 - h_1)^3}(h_{2xx} - h_{1xx}) \tag{C53}$$

Inserting above equations in kinematic conditions for the respective layer results in the following equations.

$$h_{1t} - P_{2LCxx}\left(\frac{h_1^2 h_2 - h_1^3}{2\mu_1}\right) - P_{1xx}\left(\frac{h_1^3}{2\mu_1}\right) = 0 \tag{C54}$$

$$h_{2t} - P_{1xx}\left(\frac{h_1^2 h_2 - h_1^3}{2\mu_1} + \frac{h_1^3}{3\mu_1}\right) - P_{2LCxx}\left(\frac{h_1^2 h_2 - h_1^3}{2\mu_1} + \frac{h_1^3 - 2h_1^2 h_2 + h_2^2 h_1}{\mu_1} + \left(\frac{h_1 - h_2}{\theta_1 - \theta_2}\right)^3 \frac{I_1}{\mu_2}\right) = 0 \tag{C55}$$

The introduction of normal modes in the above equations give,

$$\tilde{h}_1\,\omega + \tilde{h}_2 A' + \tilde{h}_1\,B' = 0 \tag{C56}$$

$$\tilde{h}_2\,\omega + \tilde{h}_2 C' + \tilde{h}_1\,D' = 0 \tag{C57}$$

Algebraic operation on Eq's (36, 37) gives the dispersion relation in the form given below.

$$\omega = \frac{-(B' + C') \pm \sqrt{(B' - C')^2 + 4A'D'}}{2} \tag{C58}$$

where, $A', B',\ C',\ D'$ are coefficients and given in **Section B(ii)** of the main manuscript.