Mechanisms to splay-bend nematic phases

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While twist-bend nematic phases have been extensively studied, the experimental observation of two dimensional, oscillating splay-bend phases is recent. We consider two theoretical models that have been used to explain the formation of twist-bend phases—flexoelectricity and bond orientational order—as mechanisms to induce splay-bend phases. Flexoelectricity is a viable mechanism, and splay and bend flexoelectric couplings can lead to splay-bend phases with different modulations. We show that while bond orientational order circumvents the need for higher order terms in the free energy, the important role of nematic symmetry and phase chirality rules it out as a basic mechanism.

I. INTRODUCTION

Liquid crystalline materials show a rich variety of structures and phases. Indeed even if we focus on the smectic or cholesteric mesophases, there are a nearly unlimited variety of structures and motifs. On the other hand, achiral nematic phases, the backbone of the display industry, the workhorse of experiment, and the most well understood, have only a few variants (it has not escaped our attention that their simplicity is the key to their value as devices). Indeed, only a handful of distinct nematic phases have been found, and the space of possible configurations is highly restricted for achiral molecules. It is well known that achiral rodlike and discotic molecules form uniaxial nematics and also biaxial nematics [1–3]. Over the past few decades, the study of bent core molecules has led to the discovery of a nematic phase in which the director field of achiral molecules follows an oblique helicoid, maintaining a constant oblique angle with a helical axis [4–8]. The texture is splay-free, having only twist and bend distortions. This new phase, the twist-bend phase, has attracted attention due to its unusual properties: a spontaneously chiral phase is formed out of achiral molecules [9,10]. Additionally, experiments show three times larger bend flexoelectric coefficients in bent core molecules than the typical value in rodlike liquid crystals [11,12]. A schematic of this phase is shown in Fig. 1.

With this phase as the backdrop, it is natural to contemplate additional nematic phases that show only twist and splay, or only splay and bend deformations. In this paper we consider both bond orientational order and flexoelectricity as effects that can stabilize "splay-bend" phases, also shown in the schematic in Fig. 1. Although flexoelectricity has been considered before, we show that different forms of the splay and bend couplings can give us two distinct splay-bend phases with different modulations [13,14]. The paper is organized as follows. In Sec. II, we consider bond orientational order and find that nematic symmetry and phase chirality make bond

order an unlikely mechanism for splay-bend. Next, in Sec. III we consider flexoelectric effects and look at the splay and bend flexoelectric couplings that could give rise to splay-bend phases with different modulations. In Sec. IV, we look at the two different "splay" phases that have been addressed in the literature—splay-bend [13] and splay nematic phases [14]—and show that these are related to each other by an exchange of the bend and splay deformations.

The mechanism behind the emergence of the twist-bend and splay-bend phases remains debated. Initial work argued that a purely elastic instability, resulting from negative bend elastic constants, could explain the emergence of both these phases [13,15,16]. However, this leads to a free energy unbounded from below: higher order and degree terms are necessary to find stable extrema. More recent theoretical work shows that a linear coupling between polar order and the deformations of the nematic director can give effective elastic constants, which can then be driven negative with changing temperature [17,18]. Bend flexoelectric couplings have been used to explain twist-bend phases, and a combination of both bend and splay flexoelectricity to explain splay-bend phases. Recent work shows that combinations of flexoelectricity and intrinsic chirality also predict yet unseen, but related, modulated phases [19].

Another mechanism that does not require higher order terms does exist for the twist-bend texture [20] but requires chiral bond order: upon cooling, nematic liquid crystals can give rise to a liquid crystalline phase with nematic order and hexatic order in the plane perpendicular to it [21]. If the hexatic order is itself chiral, then the twist-bend texture is stable. Such a mechanism would predict the emergence of twist-bend and splay-bend phases without the need for stabilizing arbitrary higher order terms, but pushes the problem on to find a mechanism for spontaneous achiral symmetry breaking in the case of achiral molecules. In this paper we will employ the tools of symmetry to study the phase behavior of a system with spontaneous nematic and polar order. The polarization is a vector which might be associated with molecular dipoles or some other geometric aspect of the mesogens. As in [22], we will not study the entire electromagnetic response of this

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complex material, but rather we will try to understand the allowed ground states through the Goldstone-mode/Landau-theory paradigm. In particular, couplings that are allowed by symmetry are, generically, nonzero. In the absence of a deeper symmetry we will assume that these couplings arise through an effective theory of the mesophases.

II. BOND ORIENTATIONAL ORDER

Previous work shows how hexatic bond order in a chiral liquid crystal can give rise to a "conical phase" with the same texture as the twist-bend phase, while circumventing the need for higher order terms [20]. We consider now whether this is a viable mechanism to induce the splay-bend phase. Consider a nematic system with bond orientational order in the plane perpendicular to the nematic director. For our purposes, it is sufficient to consider the general case without specifying the number of nearest neighbors.

The fluctuations in the nematic director, $\mathbf{n}$, are given by the Frank free energy density,

$$f_{\mathbf{n}} = \frac{K_1}{2}[\mathbf{n}(\nabla \cdot \mathbf{n})]^2 + \frac{K_2}{2}[\mathbf{n} \cdot (\nabla \times \mathbf{n})]^2 + \frac{K_3}{2}[(\mathbf{n} \cdot \nabla)\mathbf{n}]^2, \tag{1}$$

where K_1, K_2, and K_3 are the splay, twist, and bend elastic constants, respectively. Here and throughout we require that these elastic constants are positive. Apart from the contributions to the free energy from modulations in the director field, we want to account for interactions between the director and the bond angle. The bond angle, Φ, quantifies the bond order in the system. The definition of Φ depends on the definition of the nematic director field [20]: in order to measure the sense of an angle in three-space we need a screw direction. Using the nematic director requires us to incorporate the nematic symmetry into the definition of Φ. In particular, it follows the nematic symmetry, and $\Phi \to -\Phi$ under the transformation $\mathbf{n} \to -\mathbf{n}$. We expect that the bond order contribution to the free energy density has a term that penalizes any sharp changes in Φ, and a term that captures the interaction between Φ and $\mathbf{n}$.

Since we require that the overall nematic symmetry is preserved in the free energy density, any term that represents the interaction between the bond angle and the nematic director must have an even power of Φ and $\mathbf{n}$ together. This means, for a term linear in $\nabla\Phi$, the interaction term must have an odd power in $\mathbf{n}$.

The twist-bend phase has a chiral structure, and so a chiral interaction term is expected. In order to construct the interaction term then, we want a vector with an odd number of derivatives to account for chirality, and an odd power of $\mathbf{n}$ to preserve nematic symmetry. The lowest order term that satisfies these constraints is $\mathbf{n} \cdot \nabla\Phi$. Considering this term,

$$f_{\Phi} = \frac{K_A}{2}(\nabla\Phi)^2 - K_A q_0(\mathbf{n} \cdot \nabla\Phi), \tag{2}$$

where the full free energy density is $f = f_{\mathbf{n}} + f_{\Phi}$ and f_{Φ} is the contribution to the free energy density from bond orientational order. The total free energy can be minimized to determine the parameters of the phase and the bond angle,
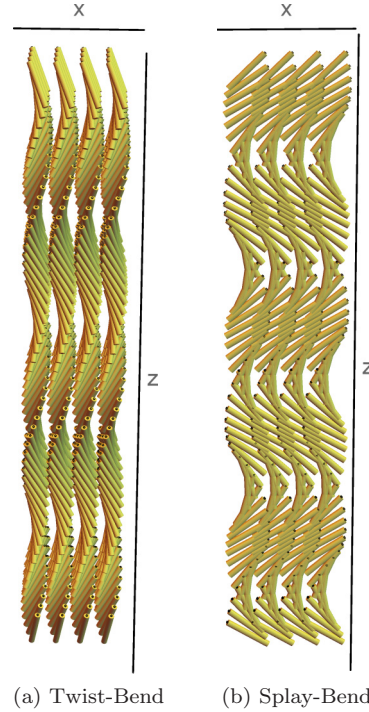

(a) Twist-Bend (b) Splay-Bend

FIG. 1. The figure shows (a) twist-bend and (b) splay-bend structures. The twist-bend structure has molecules rotating about the z direction while maintaining a constant angle with it. In the splay-bend texture, molecules oscillate along the z direction in two dimensions.

Φ, as a function of the Frank constants, the pitch (q_0), and the bond-angle stiffness (K_A). Since the interaction term is of lower order than the terms in the Frank elastic energy, the total free energy remains bounded from below. Indeed, extremizing over Φ we have $\nabla^2\Phi = q_0\nabla \cdot \mathbf{n}$. For the twist-bend texture $\mathbf{n}_{\text{tb}} = [\cos(qz)\cos\theta, \sin(qz)\cos\theta, \sin\theta]$ and we can only have $\nabla\Phi = v_0\hat{z}$, a constant vector along the z axis. Minimizing over the value of $\mathbf{v}_0$ and integrating over a period generates a term [20]

$$f_{\Phi} = -\frac{K_A q_0^2}{2}\sin^2\theta, \tag{3}$$

and the bond order acts as a magnetic aligning field, as studied half a century ago by Meyer [23], stabilizing the texture. Note that this mechanism is markedly different from the bold proposal of Dozov [13] that requires that Frank constants become negative. However, this mechanism for a twist-bend texture requires chirality, something absent from the observed systems.

Here, we drop chirality and attempt to build a bond-orientational order model that stabilizes a splay-bend texture. For splay-bend, we use the ansatz

$$\mathbf{n}_1 = \{\sin[\theta\sin(qz)], 0, \cos[\theta\sin(qz)]\}. \tag{4}$$

This describes an oscillating, two-dimensional structure that alternates between regions of splay and bend deformations. Here, q is the pitch of the phase and θ is the maximum angle to which the molecules tilt. The direction of modulation here
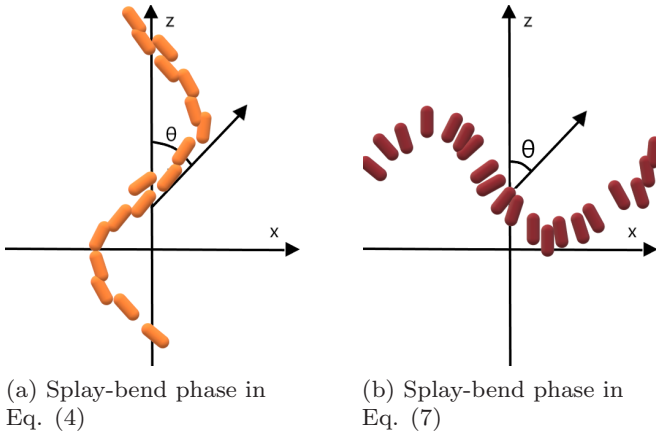
(a) Splay-bend phase in Eq. (4)

(b) Splay-bend phase in Eq. (7)

FIG. 2. Schematics of the splay-bend phase in the ansatz in (a) Eq. (4) and (b) Eq. (7) showing the maximum angle θ. The modulations are parallel to the average nematic director direction in the left schematic, and perpendicular to it in the right.

is parallel to the average nematic director field as shown in Fig. 2.

Using this to calculate the splay and bend free energy density contributions, and averaging over a period π/q, we find a free energy density

$$\bar{f}_{\mathbf{n}_1} = \frac{K_1 q^2}{8}[\theta^2 - \theta J_1(2\theta)] + \frac{K_3 q^2}{8}[\theta^2 + \theta J_1(2\theta)]. \quad (5)$$

Here, $J_\nu(z)$ are Bessel functions of the first kind. Using the properties of Bessel functions it is straightforward to check that when both K_1 and K_3 are positive, $\bar{f}_{\mathbf{n}_1} \geqslant 0$ and has only one minimum at $\theta = 0$, the uniaxial namatic. Since the splay-bend phase is achiral, an achiral coupling is necessary: $\tilde{f}_\Phi$. The symmetries that the new term must have are as follows: continuing to require that the nematic symmetry, $\mathbf{n} \rightarrow -\mathbf{n}$, is preserved, the interaction term must have an even power of Φ and $\mathbf{n}$ together. Further, since the texture is achiral, we assume that the interaction term must also be achiral and thus even in derivatives of fields. Thus a term linear in $\nabla\Phi$ requires a vector with an odd order of derivatives and an odd power of $\mathbf{n}$.

We may then list our possibilities for the lowest order term: one could consider interactions that involve the splay vector, $\mathbf{n}(\nabla \cdot \mathbf{n})$, but these do not follow the nematic symmetry. The same is true for interactions that involve the bend vector, $\mathbf{n} \times (\nabla \times \mathbf{n})$.

One possibility that has the required symmetries is $\nabla\Phi \cdot (\nabla \times \mathbf{n})$. In this case, the extremal equation for Φ is again $\nabla^2\Phi = 0$. Since $\nabla \times \mathbf{n}_1 = [q\theta \cos(qz) \sin(\theta \sin(qz)), 0, 0]$, we can consider the standard harmonic solutions of Laplace's equation for Φ. If $\nabla\Phi \cdot \nabla \times \mathbf{n}_1 \neq 0$ then Φ must depend on x. There is a solution linear in x which, when inserted and averaged over a z period, results in no coupling between the bond order and the director. Other solutions are of the form $\cosh(\alpha x_i) \cos(\alpha x_j)$ where $i \neq j$ and α is a constant. Since we would need $\partial_x \Phi \neq 0$, the only possible term that would not vanish upon spatial averaging would be of the form $\Phi = \cosh(\alpha x) \cos(\alpha z)$ (up to translations). Unfortunately, while surviving the z-averaging, a solution like this would lead to an unbounded free energy density. Whether it is possible to have defect walls between regions of bounded $\nabla\Phi$ is the topic of future work.

Finally, were we to consider an interaction term with higher order in derivatives than either the bend or splay vectors, we would generate an odd power of q higher than 2 in the free energy integrated over one pitch, requiring even higher order terms to assure stability. Since that was the *raison d'être* for considering this mechanism, we conclude that there are then no interaction terms that have the appropriate symmetries and a low enough order to give a nontrivial minimum for q and θ.

We conclude then that bond orientational order is not a simple mechanism that can give splay-bend phases. In order to get a splay-bend phase, a vector field, like the polarization vector, $\mathbf{P}$, is required [17]. Such a field plays the part of a vector that need not follow the nematic symmetry. Several of the interaction terms that are not available to us with the bond angle are then permitted by symmetry.

III. FLEXOELECTRICITY

Recall that the flexoelectric effect is a linear coupling between a polarization vector and director deformations. A coupling may be constructed with either the splay or bend vectors that, in turn, gives rise to an effective negative K_1 or K_3, respectively [17]. Such a coupling induces spontaneous splay or bend in the system. Previous work has shown how a negative effective K_3 can lead to both the twist-bend and splay-bend phases [13]. Similarly, a negative effective value of K_1 has been used to explain the observation of the splay nematic phase [14].

We look at both of these couplings independently. We consider the following ansatz for the polarization vector $\mathbf{P}$ and nematic director field $\mathbf{n}$ [14]:

$$\mathbf{n}_2 = \{\sin[\theta \sin(qx)], 0, \cos[\theta \sin(qx)]\}, \quad (6)$$

$$\mathbf{P} = \mathbf{n}_2 p \cos qx. \quad (7)$$

This ansatz is different from the one in Eq. (4), and the direction of modulation is perpendicular to the average direction of the nematic director field, as is shown in Fig. 2. We will show in the next section how the two splay-bend systems can be mapped on to each other. This form for the polarization, $\mathbf{P}$, breaks the nematic up/down symmetry. When averaging over the sample, modulations that are at different wavelengths than $2\pi/q$ will vanish and so we pick the dipole modulation accordingly. For a splay flexoelectric coupling, the free energy density is

$$f_{\text{splay}} = f_\mathbf{n} - \gamma \mathbf{P} \cdot [\mathbf{n}(\nabla \cdot \mathbf{n})] + \frac{b}{2}(\nabla \mathbf{P})^2 + \frac{t}{2}\mathbf{P}^2, \quad (8)$$

where γ, b, and t are Landau coefficients. The coefficient t is determined by various factors such as electrostatic energy, entropy which make a positive contribution, and packing energy that depends on molecular shape which has a stabilizing effect. With all these contributing factors, t changes with temperature and drives the transition to a spontaneously polarized state [14]. Since the free energy is second order in $\mathbf{n}$, the effective period of its variation is π/q. Inserting the ansatz into the free energy density, and integrating over a period π/q,
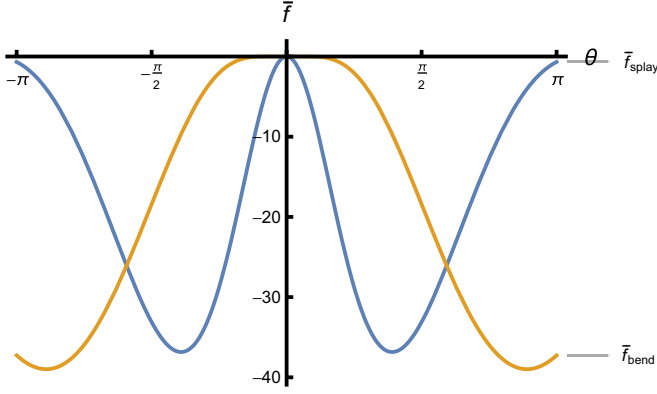
FIG. 3. Plots of $\bar{f}_{\text{splay}}$ and $\bar{f}_{\text{bend}}$ as a function of θ at the free energy minimizing values of q and p. The plots clearly show that both free energies have a minimum at a nontrivial value of θ, implying that the splay-bend phase is a possibility with both couplings. The parameter values are $K_1 = 1.5$, $K_3 = 2$, $\gamma = 40$, $b = 2$, and $t = 10$.

we find an average free energy density of

$$\bar{f}_{\text{splay}} = \frac{K_1}{8}q^2[\theta^2 + \theta J_1(2\theta)] + \frac{K_3}{8}q^2[\theta^2 - \theta J_1(2\theta)]$$
$$- \gamma pq \frac{J_1(\theta)}{\theta} + \frac{tp^2}{4} + \frac{b}{16}p^2q^2(3\theta^2 + 4). \quad (9)$$

This free energy can then be minimized with respect to p and q to obtain the following expressions at the free energy minimum:

$$q^2_{\text{splay}} = -\frac{4\sqrt{t}}{b(3\theta^2 + 4)}$$
$$\times \left\{ \sqrt{t} + \frac{2\sqrt{2}\gamma|J_1(|\theta|)|}{\sqrt{(K_1 + K_3)\theta^2 + \theta(K_1 - K_3)J_1(2\theta)}} \right\}, \quad (10)$$

$$p_{\text{splay}} = \frac{8\theta\gamma J_1(|\theta|)q_{\text{splay}}}{|\theta|\left(b(3\theta^2 + 4)q^2_{\text{splay}} + 4t\right)}. \quad (11)$$

Note that the radicand in (10) is non-negative ($\bar{f}_{\mathbf{n}_1} \geqslant 0$). In order to check the validity of these expressions, we plug in typical values of the different constants and take $\theta \sim 1$. Using $K_1 = 1$ pN, $K_3 = 10$ pN, $\gamma = 10^{-3}$ V, $b = 2 \times 10^{-18}$ V m^3/(A s), and $t = 8 \times 10^{-8}$ V m/(A s), we obtain $q = 0.1$ nm^{-1} and $p = 10^7$ (A s)/m^2. This is consistent with experiments where a nanometer range for pitch is observed [14]. Further, using the typical density of 1 g/cm^3, the value of the polarization density p translates to a molecular polarization of 10 debye, which is approximately the same as that of the molecule of RM734 seen to form splay-bend phases [14].

We substitute these expressions for p_{splay} and q_{splay} into the free energy and plot it as a function of the maximum angle θ in Fig. 3. As can be seen, there is a nontrivial minimum at a nonzero value of θ, so the splay-bend phase is stable in the case of a splay flexoelectric coupling.

In the case of a bend flexoelectric coupling, the only term that changes is the interaction term with coupling γ. A bend flexoelectric coupling is of the form $\mathbf{P} \times [\mathbf{n} \times (\nabla \times \mathbf{n})]$. However, this is a vector. If the material were sandwiched

between two different plates separated in the direction perpendicular to director (the y axis), then a coupling of the form $\hat{y} \cdot (\mathbf{P} \times [\mathbf{n} \times (\nabla \times \mathbf{n})])$ is allowed. Further, a term of this form is a pseudoscalar, and the coupling constant γ also must be a pseudoscalar. The pseudoscalar character of γ may be because of chirality in molecular shape, or a chiral asymmetry in the anchoring conditions. The latter could be possible as a contributing factor in experiments that show the splay-bend phase with achiral molecules. The strength of this coupling depends on the strength of the anchoring conditions and is characterized by γ. This is likely to be a weak effect, and we expect that it penetrates into the bulk for liquid crystal films very thin in the $\hat{y}$ direction. In this case the average free energy density is

$$\bar{f}_{\text{bend}} = \frac{K_1}{8}q^2[\theta^2 + \theta J_1(2\theta)] + \frac{K_3}{8}q^2[\theta^2 - \theta J_1(2\theta)]$$
$$- \gamma pq \boldsymbol{H}_1(\theta) + \frac{tp^2}{4} + \frac{b}{16}p^2q^2(3\theta^2 + 4). \quad (12)$$

Here, $\boldsymbol{H}_\nu(z)$ is the Struve function of order ν. Repeating the same procedure as earlier, we plot the average free energy density in Fig. 3. As can be seen, a nontrivial minimum exists at a higher value of θ than for splay flexoelectricity. Thus, we conclude that the splay-bend phase given by the ansatz in Eq. (7) can be obtained by either splay flexoelectric coupling or a bend flexoelectric coupling along with a sample asymmetry, providing the direction $\hat{y}$.

IV. SPLAY-BEND AND SPLAY NEMATIC PHASES

Previous work on nematic phases with splay and bend modulations makes a distinction between the ansatz in Eq. (7), a "splay nematic phase," and the "splay-bend phase" in Eq. (4) [14]. In particular, the direction of the modulation is perpendicular to the director in Eq. (7), as opposed to along the director, as in Eq. (4). In the "splay-bend phase," the splay and bend contributions to the free energy density, integrated over a period π/q, are then

$$\bar{f}_{\mathbf{n}_2} = \frac{K_1 q^2}{8}[\theta^2 + \theta J_1(2\theta)] + \frac{K_3 q^2}{8}[\theta^2 - \theta J_1(2\theta)]. \quad (13)$$

As can be seen from a comparison of the above equation with Eq. (5), the splay and bend contributions have been interchanged. The two systems can be mapped on to each other by exchanging K_1 with K_3. The "splay nematic phase" and the "splay-bend phase" are closely related phases. This is expected since a rotation of the nematic director field by $\pi/2$, as would be required to turn $\mathbf{n}_1$ into $\mathbf{n}_2$, would turn splay deformations into bend deformations and bend into splay. If we consider the case where the polarization vector has its origins in shape asymmetry for bent-core molecules, a spontaneous bend in the system would be equivalent to a spontaneous splay in a similar system of wedge-shaped molecules, since the two shapes can be related to each other by a rotation of $\pi/2$. A mechanism that leads to a "splay-bend" phase of bent-core molecules would then induce a "splay nematic" phase in a system of wedge-shaped molecules.

We could have begun by using the ansatz in Eq. (4), and repeated the process outlined in Sec. III by inserting

the new ansatz into the free energies with the two different flexoelectric couplings. Minimizing with respect to q and p, we would find that the results in Sec. III are reversed, and the curves for $\bar{f}_{\mathrm{splay}}$ and $\bar{f}_{\mathrm{bend}}$ are interchanged in Fig. 3. Thus, both the splay-bend and "splay nematic" phases can be obtained with splay and bend flexoelectric couplings, and can be related to each other by an exchange of the bend and splay elastic constants.

V. CONCLUSIONS

We have demonstrated that, while bond orientational order is a mechanism that could circumpass the problem of an unbounded free energy, nematic symmetry and achirality of the splay-bend phase prevent it from explaining the formation of the splay-bend phase. Flexoelectricity provides a viable mechanism for introducing splay and bend modulations in nematic systems. Bend and splay flexoelectric couplings lead to effective elastic constants that stabilize splay and bend modulations. Both flexoelectric couplings can give rise to splay-bend phases with modulation in the average direction of the director field or modulations perpendicular to the average direction of the nematic director. These two modulations, treated previously in the literature as "splay-bend" and "splay nematic" phases, are related to each other by an exchange of the splay and bend elastic constants.

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