## Coupling between splay deformations and density modulations in splay-bend phases of bent colloidal rods

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Using a grand-canonical Landau–de Gennes theory for colloidal suspensions of bent (banana-shaped) rods, we investigate how spatial deformations in the nematic director field affect the local density of twist-bend and splay-bend nematic phases. The grand-canonical character of the theory naturally relates the local density to the local nematic order parameter S. In the splay-bend phase, we find S and hence the local density to modulate periodically along one spatial direction. As a consequence the splay-bend phase has the key symmetries of a smectic rather than a nematic phase. By contrast we find that S and hence the local density do not vary in space in the twist-bend phase, which is therefore a proper nematic phase. The theoretically predicted one-dimensional density modulations in splay-bend phases are in agreement with recent simulations.

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Dispersions of rodlike colloidal particles can spontaneously order into nematic liquid-crystal phases upon compressing a dilute isotropic fluid phase [1,2]. The nematic bulk phase is a homogeneous fluid that exhibits long-range orientational order of the long axes of the rods, which on average align along a so-called nematic director  $\hat{\mathbf{n}}$  that is uniform in space. An important characteristic of the nematic phase is its up-down symmetry, which identifies  $\hat{\mathbf{n}}$  with  $-\hat{\mathbf{n}}$ , to be contrasted with polar order of for instance a magnet. The simplest nematic phase does not only feature up-down symmetry but also azimuthal symmetry about  $\hat{\mathbf{n}}$ . This uniaxial symmetry is broken in the case of a biaxial nematic phase, which features not only ordering of the long axes of the rods along  $\hat{\mathbf{n}}$  but also of their short axes along a direction  $\hat{\mathbf{m}} \perp \hat{\mathbf{n}}$ , with  $\pm \hat{\mathbf{m}}$  equivalent [1]. This Letter, however, concerns the interesting and intricate case in which the azimuthal symmetry about  $\hat{\mathbf{n}}$  is broken by a *polar* ordering of the short axes of the rods in a preferred direction  $\mathbf{P} \perp \hat{\mathbf{n}}$ , with nonequivalent **P** and  $-\mathbf{P}$ . This type of ordering is well known to be strongly coupled, via the so-called bend flexoelectric effect [3], to spontaneous bend deformations of the nematic director field, such that  $\hat{\mathbf{n}}(\mathbf{r})$  is no longer a spatial constant if  $\mathbf{P} \neq \mathbf{0}$  [4]. As a pure bend deformation cannot uniformly fill three-dimensional (3D) space, Meyer [5] and later independently Dozov [6] argued that these local bend deformations should be accompanied by either a twist or a splay deformation. In the former case they theoretically postulated the stabilization of a spatially modulated twist-bend nematic  $(N_{\rm TB})$  phase, that displays a heliconical structure of  $\hat{\bf n}({\bf r})$  with bend and twist deformations in the molecular orientation. In

the latter case, they predicted a spatially modulated splaybend nematic ( $N_{\rm SB}$ ) phase that is characterized by alternating layers of splay and bend.

After its theoretical prediction, the  $N_{\text{TB}}$  phase has actually been observed experimentally in several molecular systems [7-14], and has meanwhile become well established as a new type of orientationally ordered fluid phase. By contrast, the  $N_{\rm SB}$  phase has never been observed in the absence of a strong external constraint, such as an electric field [15–17], a planar surface anchoring [18], or a topological constraint [19]. Rather, a huge variety of smectic phases was found either in systems of thermotropic bent-core mesogens [4,20,21] and in systems of bent silica rods [22,23]. Therefore, many doubts subsist on the existence and true nature of the  $N_{\rm SB}$ phase. Recently, however, simulations of systems of hard bent spherocylinders revealed that  $N_{\text{TB}}$  and  $N_{\text{SB}}$  phases can be thermodynamically stable in bulk, provided the smectic (Sm) phase is destabilized either by polydispersity in particle length or by curvature in the particle shape [24]. Inspired by these simulation results, a stable  $N_{\rm SB}$  phase has experimentally been sought for and actually observed, for the first time in a lyotropic system of smoothly curved colloidal rods [25] and later in polydisperse bent silica rods [26]. Recently, simulations and a Maier-Saupe theory showed that the (alleged)  $N_{\rm SB}$  phases of monodisperse curved rods display (weak) density modulations [27], which cannot be described by current Oseen-Frank and Landau-de Gennes type theories. The existing theoretical descriptions [5,6,28] are direct extensions of Oseen-Frank theory for the elastic deformations of  $\hat{\mathbf{n}}(\mathbf{r})$ ; they only account for the *direction* of the ordering and ignore the *degree* of ordering that is characterised by the scalar nematic order parameter S. Landau-de Gennes (LdG) theories for thermotropic liquid crystals do account for S but lack any coupling with density  $\rho$  [29–33].

In this Letter we investigate the nematic nature of the  $N_{\rm SB}$  phase displayed in colloidal systems, by employing a

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recently introduced grand-canonical LdG theory [34] applied to lyotropic curved rods [35], that does involve a coupling between *S* and  $\rho$ . We show that *S* and therefore also  $\rho$  varies periodically along one spatial direction in the *N*<sub>SB</sub> phase, such that the *N*<sub>SB</sub> phase has actually the symmetries of a smectic rather than a nematic phase. This is not the case, however, for the *N*<sub>TB</sub> phase, for which we find that *S* does not vary in space, and is therefore a proper nematic phase.

We follow Ref. [35] and extend the Oseen-Frank theory of Selinger and co-workers [28] to a mesoscopic LdG theory where the director  $\hat{\mathbf{n}}$  is replaced by the standard secondrank, symmetric, and traceless tensor  $\mathbf{Q}(\mathbf{r})$  with Cartesian components  $Q_{\alpha\beta}(\mathbf{r})$  for  $\alpha, \beta = x, y, z$ . The eigenvector of Q, corresponding to the maximum modulus of a nondegenerate eigenvalue, defines the nematic director  $\hat{\mathbf{n}}$  of the system [1]. A vector field  $\mathbf{P}(\mathbf{r})$  with Cartesian coordinates  $P_{\gamma}(\mathbf{r})$  for  $\gamma = x, y, z$  describes the polar order in a direction perpendicular to **n**. In order to describe lyotropic systems that become ordered with increasing density, we follow Ref. [34] and set up a Landau expansion for which we use the chemical-potential-dependent grand potential  $\Omega(\mu)$  rather than the temperature-dependent Helmholtz (or Gibbs) free energy F(T). More specifically, for a system of hard bent rods modeled as curved or kinked rods of contour length L and diameter D, at chemical potential  $\mu$  in a macroscopic volume V, and at fixed temperature T, we write the excess-overisotropic LdG grand potential as  $\Delta \Omega(\mathbf{Q}, \mathbf{P}) = \int_{V} d\mathbf{r} (\Delta \omega_b + \omega_b)$  $\omega_{eP}$ ), where  $\Delta \omega_b$  denotes a bulk term

$$\beta B_2 \Delta \omega_b = \frac{2}{3} a \beta (\mu^* - \mu) Q_{\alpha\beta} Q_{\beta\alpha} - \frac{4}{3} b \, Q_{\alpha\beta} Q_{\beta\lambda} Q_{\lambda\alpha} + \frac{4}{9} d \, Q_{\alpha\beta} Q_{\beta\alpha} Q_{\lambda\rho} Q_{\rho\lambda}, \qquad (1)$$

and  $\omega_{eP}$  an elastic-polar term

$$\beta B_2 \omega_{eP} = \frac{2}{9} l_1 (\partial_\alpha Q_{\beta\lambda}) (\partial_\alpha Q_{\beta\lambda}) + \frac{2}{9} l_2 (\partial_\alpha Q_{\alpha\lambda}) (\partial_\beta Q_{\beta\lambda}) + e_2 P_\alpha \left( \delta_{\alpha\beta} + \frac{2}{S_0} Q_{\alpha\beta} \right) P_\beta + e_4 P_\alpha P_\alpha P_\beta P_\beta - \lambda P_\alpha (\partial_\beta Q_{\alpha\beta}) + \kappa (\partial_\alpha P_\beta) (\partial_\alpha P_\beta).$$
(2)

We express  $\Delta \omega_b$  and  $\omega_{eP}$  in units of  $\beta^{-1} = k_B T$  with  $k_B$ the Boltzmann constant, and in units of the second virial coefficient  $B_2 = \pi L^2 D/4$  of long needles in the isotropic (I) fluid phase, which is a convenient unit of volume for systems of rodlike particles. We use the Einstein summation convention for repeated indices, and indicate the Landau phenomenological parameters by a, b, d, l<sub>1</sub>, l<sub>2</sub>, e<sub>2</sub>, S<sub>0</sub>, e<sub>4</sub>,  $\lambda$ , and  $\kappa$ . We note that only the quadratic term  $Q_{\alpha\beta}Q_{\beta\alpha}$  has a  $\mu$ -dependent prefactor that changes sign at the nematic spinodal  $\mu^*$  and drives the phase transitions. Stability of the grand-potential expansion in the dilute limit requires a > 0and  $e_2 > 0$ , while stability with respect to an unlimited growth of **Q**, **P**, and  $\partial_{\alpha}P_{\beta}$  requires that d > 0,  $e_4 > 0$ , and  $\kappa > 0$ , respectively. The sign of the *b* term determines the nature of the *I*-*N* transition, rodlike for b > 0 and platelike for b < 0. The coefficients  $2/S_0$  and  $\lambda$  represent the **Q-P** coupling and the bend flexoelectric  $\mathbf{P}$ - $\nabla \mathbf{Q}$  coupling, respectively. In order to describe a favored polarization perpendicular to the nematic director, leading to a bend flexoelectric effect, we set  $S_0 > 0$ . Finally the coefficients  $l_1$  and  $l_2$  are related to the Oseen-Frank elastic constants [36,37] through the relations  $\beta B_2 K_{11} = \beta B_2 K_{33} = (2l_1 + l_2)S^2$  and  $\beta B_2 K_{22} = 2l_1S^2$ . As shown in Ref. [35], a complete mapping exists between our LdG theory and the Oseen-Frank theory of Selinger and co-workers [28].

Minimization of the grand potential  $\Delta\Omega(\mathbf{Q}, \mathbf{P})$  with respect to  $\mathbf{Q}$  at  $\mathbf{P} \equiv \mathbf{0}$ ,  $\nabla \mathbf{Q} \equiv \mathbf{0}$ , and  $\nabla \mathbf{P} \equiv \mathbf{0}$ , gives a first-order *I-N* transition at  $\beta \mu_{IN} \equiv \beta \mu^* - b^2/(4ad)$  [34,35]. The nematic order parameter equals  $S_I(\mu) = 0$  for  $\mu \leq \mu_{IN}$ , and

$$S_N(\mu) = \frac{3b}{8d} \left( 1 + \sqrt{1 - \frac{32ad\beta(\mu^* - \mu)}{9b^2}} \right)$$
(3)

for  $\mu > \mu_{IN}$ . In order to describe the "Onsager"-type *I-N* phase transition of uniaxial hard needles [2,38] we follow Ref. [34] and set a = 1.436, b = 5.851, d = 3.693, and  $\beta\mu^* = 6.855$ . As shown in Ref. [35], the uniaxial *N* phase becomes unstable with respect to an  $N_{\text{TB}}$  or  $N_{\text{SB}}$  phase at a critical  $\mu$  where the renormalized bend elastic constant  $K_{33}^{\text{eff}} = 0$ . The  $N_{\text{TB}}$  phase is described by a chiral nematic director and polarization vector

$$\hat{\mathbf{n}}_{\text{TB}}(z) = [\sin\theta\cos(qz), \sin\theta\sin(qz), \cos\theta],$$
$$\mathbf{P}_{\text{TB}}(z) = P(z)[\sin(qz), -\cos(qz), 0],$$
(4)

with q and  $\theta$  the variational wave number and tilt angle, respectively, and P(z) the variational magnitude of the polarization. The  $N_{\rm SB}$  phase is described by an achiral nematic director and polarization vector

$$\hat{\mathbf{n}}_{\text{SB}}(z) = [\sin \phi(z), 0, \cos \phi(z)],$$
$$\mathbf{P}_{\text{SB}}(z) = P(z) \cos(qz)[-\cos \phi(z), 0, \sin \phi(z)], \quad (5)$$

where  $\phi(z) = \theta \sin(qz)$ . The system either stabilizes an  $N_{\text{TB}}$  phase, if  $l_1$  and  $l_2$  are such that  $K_{11} > 2K_{22}$ , or an  $N_{\text{SB}}$  phase, if  $l_1$  and  $l_2$  are such that  $K_{11} < 2K_{22}$  [35]. The N- $N_{\text{TB}}$  as well as N- $N_{\text{SB}}$  phase transitions are continuous and proceed in both cases without any jump in  $S(\mu)$ .

We employ the thermodynamic identity  $\partial(\Omega/V)/\partial \mu|_{V,T} = -\rho$  to perform the conversion between chemical potential  $\mu$  and number density  $\rho$ . To this end, we introduce the grand-potential density  $\omega_I$  of the isotropic *I* state and define  $\omega \equiv \omega_I + \Delta \omega_b$ , where  $\Delta \omega_b$  is the excess bulk grand-potential density (1). From  $\partial(B_2\omega)/\partial\mu = -c$ , with  $c \equiv B_2\rho$  the dimensionless particle concentration, we find

$$c(\mu) = c_I(\mu) + aS^2(\mu).$$
 (6)

The particle concentration of the *I* phase,  $c_I(\mu) = -\partial(B_2\omega_I)/\partial\mu$ , can be calculated within Onsager theory, by using an isotropic distribution function, such that  $\beta\mu(c_I) = \log(c_I/4\pi) + 2c_I$  [38]. By inverting this relation we obtain  $c_I(\mu)$  straightforwardly. Together with Eq. (6), we can determine *c*, for every value of  $\mu$  for which  $S(\mu)$  is known. We observe that Eq. (6) not only allows us to convert a dependence on the chemical potential into a dependence on the scalar order parameter *S* and the particle concentration *c*. In this Letter we employ this coupling to obtain insight into the structure of the N<sub>SB</sub> phase.

In contrast to earlier works [6,28-33,35] we consider here a *z*-dependent nematic order parameter S(z) in the definition of

the tensorial order parameter  $Q_{\alpha\beta}(z) = S(z)[3n_{\alpha}(z)n_{\beta}(z)/2 - \delta_{\alpha\beta}/2]$ , as well as a z-dependent magnitude P(z) of the polarization vectors  $\mathbf{P}_{\text{TB}}$  and  $\mathbf{P}_{\text{SB}}$  in Eqs. (4) and (5). In order to study the relative stability of the  $N_{\text{TB}}$  and  $N_{\text{SB}}$  phases, we insert  $\hat{\mathbf{n}}_{\text{TB}}$  and  $\hat{\mathbf{n}}_{\text{SB}}$ , respectively, into  $\mathbf{Q}$ . After insertion of  $\mathbf{Q}$  and  $\mathbf{P}$  into  $\Delta\Omega(\mathbf{Q}, \mathbf{P})$ , we perform, at fixed chemical potential  $\mu$ , a full grand-potential minimization. To this end we numerically solve the system of Euler-Lagrange equations

$$\frac{\delta \Delta \Omega}{\delta S(z)} = \partial_z \frac{\delta \Delta \Omega}{\delta(\partial_z S(z))}, \quad \frac{\delta \Delta \Omega}{\delta P(z)} = \partial_z \frac{\delta \Delta \Omega}{\delta(\partial_z P(z))}, \quad (7)$$

for many combinations of fixed wave number q and tilt angle  $\theta$ , from which we identify the grand-potential minimum of the  $N_{\text{TB}}$  phase characterized by  $q_{\text{TB}}(\mu)$ ,  $\theta_{\text{TB}}(\mu)$ ,  $S_{\text{TB}}(\mu; z)$ , and  $P_{\text{TB}}(\mu; z)$ , and that of the  $N_{\text{SB}}$  phase at  $q_{\text{SB}}(\mu)$ ,  $\theta_{\text{SB}}(\mu)$ ,  $S_{\text{SB}}(\mu; z)$ , and  $P_{\text{SB}}(\mu; z)$ .

We start by considering the coefficients  $l_1 = 0.165L^2$ ,  $l_2 =$  $0.427L^2$ ,  $e_2 = 1$ ,  $S_0 = 0.99$ ,  $e_4 = 0.5$ ,  $\kappa = 0.3L^2$ , and  $\lambda =$ 0.18L. It follows that  $K_{11}/K_{22} = 2.294$  and that, as shown in Fig. S2(a) in the Supplemental Material (SM) [39], a secondorder N-N<sub>TB</sub> phase transition takes place at  $\beta \mu = 5.663 \equiv$  $\beta \mu_{NN_{\text{TB}}}$ . We plot  $S_{\text{TB}}$  and  $P_{\text{TB}}$  as a function of  $z \in [0, 2\pi/q_{\text{TB}}]$ for several values of the chemical potential  $\mu \equiv \mu_{NN_{TB}} + \Delta \mu$ in Figs. S4(b) and S4(b), respectively. We clearly find that  $S_{\text{TB}}$ and  $P_{\text{TB}}$  are constant along z for every  $\mu$ . As a consequence, the corresponding density profile  $c_{\text{TB}}$  is also a spatial constant. Interestingly, we show in the SM and in Fig. S5 [39] that the extremal solutions  $q_{\text{TB}}(\mu)$ ,  $\theta_{\text{TB}}(\mu)$ ,  $S_{\text{TB}}(\mu)$ ,  $P_{\text{TB}}(\mu)$ , and  $c_{\text{TB}}(\mu)$  exactly coincide with those obtained when S and P are assumed z independent in Q and  $P_{TB}$ , respectively. In particular we observe from Fig. S5(a) that  $S_{\text{TB}}(\mu)$ coincides with  $S_N(\mu)$  of Eq. (3). We conclude that spatial twist and bend modulations do not affect the scalar nematic order.

Subsequently, we reset the elasticity parameters to  $l_1 =$  $0.1L^2$  and  $l_2 = 0.0427L^2$  such that  $K_{11}/K_{22} = 1.214$  and, as shown in Fig. S2(b), a second-order N-N<sub>SB</sub> phase transition takes place at  $\beta \mu = 5.354 \equiv \beta \mu_{NN_{SB}}$ . In Fig. 1(a) we plot for several chemical potentials  $\mu \equiv \mu_{NN_{SB}} + \Delta \mu$  the (shifted) scalar nematic order parameter  $S_{SB} - S_N$  as a function of  $z \in [0, 2\pi/q_{\rm SB}]$ . The periodic modulation of  $S_{\rm SB}$  is manifest, with an amplitude that vanishes at  $\Delta \mu = 0$  and increases to 0.0075 at  $\Delta \mu = 0.5k_BT$  (see also SM [39]). The inset shows the minimum and maximum of  $S_{SB}(z)$  as a function of  $\mu$ , the difference being small but clearly discernible. Note that the wavelength of the modulation of the scalar order parameter equals half of the pitch  $2\pi/q_{\rm SB}$  of the nematic director. This can be explained by the fact that one period of a nematic director modulation corresponds to two alternating bend and splay domains with opposite polar order. Figure 1(b) shows the corresponding (shifted) density profiles  $c_{SB}(z) - c_N$ , obtained from Eq. (6) with  $c_N$  the density of the metastable N phase. Figure 1(b) clearly reveals spatial density modulations with the same wavelength as  $S_{SB}(z)$  and an amplitude that grows from zero at  $\Delta \mu = 0$  to a few percent of  $c_N$  at  $\Delta \mu = 0.5k_BT$ ; the inset shows the minimum and the maximum of the density profile as a function of  $\mu$ , which confirms the density variations on the order of a few percent. From the periodic modulation of  $c_{SB}$  along z, it follows that the  $N_{SB}$  phase has all



FIG. 1. The excess scalar nematic order parameter  $S_{\rm SB} - S_N$ (a) and the excess particle concentration  $c_{\rm SB} - c_N$  (b) in the splaybend phase  $N_{\rm SB}$  as a function of position  $z \in [0, 2\pi/q_{\rm SB}]$ , for several values of the chemical-potential excess  $\Delta \mu \equiv \mu - \mu_{NN_{\rm SB}}$  above the N- $N_{\rm SB}$  transition, at wave numbers  $q = q_{\rm SB}$  of the director modulations [see Eq. (5)] given by the orange curve in (c). The period average  $\bar{S}_{\rm SB}$  and  $\bar{P}_{\rm SB}$  of the nematic order parameter  $S_{\rm SB}(z)$  and the polarization  $P_{\rm SB}(z)$  are presented by the dots in (c), which are indistinguishable from the corresponding solid curves that follow from the assumption of a spatial constant  $S_{\rm SB}$  and  $P_{\rm SB}$ . The violet curve in (c) represents the opening angle  $\theta_{\rm SB}$ . The expansion coefficients are  $l_1 = 0.1L^2$ ,  $l_2 = 0.0427L^2$ ,  $e_2 = 1$ ,  $S_0 = 0.99$ ,  $e_4 = 0.5$ ,  $\kappa = 0.3L^2$ , and  $\lambda = 0.18L$ . The insets of (a) and (b) show the  $\mu$  dependence of the minima and maxima of the modulating profiles  $S_{\rm SB}(z)$  and  $c_{\rm SB}(z)$ .

the symmetries of a smectic (or lamellar) phase. However, in contrast to ordinary smectic phases with periods on the order of the particle length L, the wavelength  $\pi/q_{\rm SB}$  of the density modulation in the splay-bend phase is much larger than L. This becomes manifest from the orange curve in Fig. 1(c), which presents the  $\mu$  dependence of the wave number  $q_{\rm SB}$ , which is infinitesimally small at the  $N-N_{SB}$  transition and grows to typically  $|q_{\rm SB}L| \simeq 0.2$  in the splay-bend phase; this corresponds to density modulations with wavelengths of the order  $\pi/q_{\rm SB} \simeq 15L$  well in the splay-bend phase and a diverging wavelength upon approach of the transition to the N phase. Figure 1(c) also shows the tilt angle  $\theta_{SB}$  and the period average of the profiles of the nematic order parameter  $S_{SB}(z)$  and the polarization  $P_{SB}(z)$ ; Fig. S7 of the SM [39] shows  $P_{SB}(z)$ in the  $N_{\rm SB}$  phase, which also exhibits spatial modulations. The infinitesimally small  $q_{\rm SB}$ ,  $\theta_{\rm SB}$ , and  $P_{\rm SB}(z)$  at the N-N<sub>SB</sub> transition are in line with the continuous character of the  $N-N_{SB}$  transition. Additionally, Fig. 1(c) shows that the period average of the profiles  $S_{SB}(\mu; z)$ ,  $P_{SB}(\mu; z)$ , and  $c_{SB}(\mu; z)$  as well as  $q_{SB}$  and  $\theta_{SB}$  (see SM [39]) actually coincide with the solutions  $S_{SB}(\mu)$ ,  $P_{SB}(\mu)$ ,  $c_{SB}(\mu)$ , and  $q_{SB}(\mu)$  and  $\theta_{SB}(\mu)$ , that are obtained when S and P are assumed z independent in  $\mathbf{Q}$ and **P**<sub>SB</sub>. In particular the period average of  $S_{SB}(\mu; z)$  is very close to  $S_N(\mu)$  of Eq. (3), as can be seen in Fig. 1(c), where their tiny difference is only discernible at the largest  $\mu$ 's and stems from the small- $\theta$  approximation of the grand-potential density [35].

Since we only find variations in the local density of the  $N_{\rm SB}$  phase and not in that of the  $N_{\rm TB}$  phase, we investigate the possibility of a direct connection between density modulations and splay deformations. Referring to the SM [39] for more details, we first note that the  $S_{\rm SB}(z)$  profiles shown in Fig. 1(a) accurately fit the functional form  $S_{\rm SB}(z) = S_{\rm SB}^{\rm max} \cos[\theta_S \sin(q_{\rm SB}z)]$  with  $\theta_S$  a fit parameter. Next, for several values of  $\mu$ , we plot  $S_{\rm SB}(\nabla \cdot \hat{\mathbf{n}}_{\rm SB})$  (solid lines) and  $H(\nabla S_{\rm SB}) \cdot \hat{\mathbf{n}}_{\rm SB}$  (dashed lines) as a function of z in Fig. S10, with H a suitably chosen spatial constant that varies with  $\mu$  as shown in the inset of Fig. S10. We find that for each  $\mu$  the condition  $S_{\rm SB}(\nabla \cdot \hat{\mathbf{n}}_{\rm SB}) = H(\nabla S_{\rm SB}) \cdot \hat{\mathbf{n}}_{\rm SB}$  holds, from which we can derive

$$\nabla \cdot \hat{\mathbf{n}}_{\text{SB}}(z) = \frac{H}{2} \frac{\nabla c_{\text{SB}}(z)}{[c_{\text{SB}}(z) - c_I]} \cdot \hat{\mathbf{n}}_{\text{SB}}(z), \qquad (8)$$

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through Eq. (6). We observe that an expression similar to the one of Eq. (8) was found by de Gennes [40] and Taratuta and Meyer [41] for polymer nematics. It expresses the coupling between concentration gradients  $\nabla c_{\text{SB}}$  and splay deformations, given by  $\nabla \cdot \hat{\mathbf{n}}_{\text{SB}}$ , such that splay deformations cannot exist without density gradients.

To conclude, in this Letter we have employed the LdG theory introduced in Ref. [35] for lyotropic colloidal suspensions of bent rods to investigate the repercussions of spatial distortions in the nematic director field on the density of the twist-bend and splay-bend nematic phases. In contrast with the existing theories [6,28–33], our theory allows to analyze the spatial dependence of the nematic order parameter S and provides a natural coupling between S and the particle density  $\rho$ . We show that the  $N_{\rm SB}$  phase is characterized by a one-dimensional density modulation such that the originally predicted [5,6,28] N-N<sub>SB</sub> transition is strictly speaking a N-Sm transition. In the case of an  $N_{\text{TB}}$  phase, instead, we find that S and hence  $\rho$  are spatial constants such that this phase is a true nematic phase. Our findings are in agreement with simulations [24] and a Maier-Saupe theory [27]. Furthermore, the existence of a coupling between  $\nabla c$  and  $\nabla \cdot \hat{\mathbf{n}}$ , as shown in this Letter, lends strong support that the splay deformations in the nematic director field  $\hat{\mathbf{n}}$  are inherently coupled to density modulations.

Finally the theory presented in this Letter could be employed to verify the existence and the nematic nature of a recently experimentally observed splay nematic phase [32,42–47] and a recently postulated twist-splay-bend phase [27]. In addition, biaxiality and external constraints could be taken into account. While we do not expect biaxial order to change the smectic nature of  $N_{\rm SB}$  phase, it could influence the "shape" of the density modulations. Understanding the repercussions of external constraints on the local density of the bulk  $N_{\rm SB}$  phase, instead, could be very important to connect the findings presented in this Letter to those of Refs. [15–19].

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