

Analytical description of the Saturn-ring defect in nematic colloids

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We derive an analytical formula for the Saturn-ring configuration around a small colloidal particle suspended in nematic liquid crystal. In particular we obtain an explicit expression for the ring radius and its dependence on the anchoring energy. We work within Landau–de Gennes theory: Nematic alignment is described by a tensorial order parameter. For nematic colloids this model had previously been used exclusively to perform numerical computations. Our method demonstrates that the tensorial theory can also be used to obtain analytical results, suggesting a different approach to the understanding of nematic colloidal interactions.

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I. INTRODUCTION

Nematic liquid crystals are characterized by their long-range orientational order: Molecules tend to align in a common direction. When colloidal particles are immersed in nematic material, they induce elastic distortions of the nematic alignment, thus creating topological defects and anisotropic interactions. These effects yield a wide range of possible applications and have attracted considerable attention over the past years. The fascinating self-assembly phenomena first described in [1] can generate an impressive variety of structures, with promising optical applications [2–12]. The sensitivity of nematic liquid crystals to inclusion of foreign bodies can also be used to build new biological sensors [13–16]. Other applications include topological memory devices [11,17], particle sorting [18], or levitation [19]. Given the universal nature of topological defects and their theoretical importance in condensed matter physics, the possibility, offered by nematic colloids, of experiments in topology [20,21] is also of high interest.

It is of crucial importance to understand precisely the nature of the interactions in nematic colloids. Many theoretical results have been obtained, using either a director field (Oseen-Frank theory) or a tensorial order parameter (Landau–de Gennes theory) to describe nematic alignment. They all predict that the interaction is very sensitive to the characteristics of the single particles: size, surface anchoring, and shape. Within the director field approach, an electrostatic analogy allows for analytical calculations. Two types of particles can thus be distinguished, depending on the symmetry of the distortions they induce—dipolar or quadrupolar [22–24]—enforcing in turn different kinds of anisotropic interactions [25–30]. Within the more complex tensorial model, analytical results have not seemed possible so far, but numerical computations [4,6,31–34] lead to similar descriptions, which are also in good agreement with experimental studies [4,6,30,33–35].

In this paper we focus on the distortion of nematic order created by a spherical particle with homeotropic anchoring.

This is a subject already well covered [22–25,32,36], but our approach, based on the Landau–de Gennes tensorial theory, is different and yields more accurate results. Previous studies of the spherical colloid reach a qualitative agreement on the nature of the nematic configuration: for large spheres, a dipolar configuration with a single point defect, while small particles produce a Saturn ring in a field with quadrupolar symmetry. However, these previous works do not provide a concrete analytical description of the ring itself and hence disagree in their computation of the ring’s radius. While all previous works employing the Landau–de Gennes model relied exclusively on numerical computations, we are able to obtain analytical expressions when the particle radius is small (compared to the nematic coherence length), by explicitly solving the limiting differential equation. Hence we provide an alternative description of the Saturn-ring defect observed in quadrupolar configurations [37–41]. There are several advantages to our method. First, by using tensors rather than director fields we do not need to renormalize the energy around the ring defect, which is identified via an eigenvalue exchange mechanism. Second, since we derive an exact solution of the limiting equations we obtain explicit values for the ring radius and its dependence on surface anchoring strength, assuming small particle size. Moreover, our approach demonstrates the possibility of using the tensorial model directly to obtain analytical results, opening a promising way for the theoretical description of colloidal interactions.

II. MODEL AND DISCUSSION

We consider a spherical particle of radius r_p surrounded by nematic material. The tensorial order parameter $\mathbf{Q} = Q_{ij}(\mathbf{r})$ describing the nematic alignment is a symmetric traceless 3×3 matrix. The eigenvector associated with its largest eigenvalue can be interpreted as a director (the average direction of local nematic alignment) while other eigenvectors account for biaxiality. Within the one-elastic-constant approximation, the Landau-de Gennes free energy reads

$$\mathcal{F}[\mathbf{Q}] = \int_{r>r_p} \left[\frac{L}{2} \partial_k Q_{ij} \partial_k Q_{ij} + f_b(\mathbf{Q}) \right] d^3\mathbf{r} + \mathcal{F}_s[\mathbf{Q}], \quad (1)$$

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where $L > 0$ is an elastic constant. The bulk free energy density $f_b(\mathbf{Q})$ is usually taken in the form

$$f_b(\mathbf{Q}) = \frac{a(T)}{2} Q_{ij} Q_{ij} - \frac{b}{3} Q_{ij} Q_{jk} Q_{ki} + \frac{c}{4} (Q_{ij} Q_{ij})^2.$$

After a proper rescaling, the coefficients are of order one: The effective elastic constant is then of the order of ξ_0^2 , where we use the coherence length $\xi_0 = \sqrt{27cL/b^2}$ (see, e.g., [42] for this nondimensionalization). The surface anchoring term \mathcal{F}_s in (1) enforces homeotropic anchoring at the particle surface

$$\mathcal{F}_s[\mathbf{Q}] = W \int_{r=r_p} (Q_{ij} - Q_{ij}^h)(Q_{ij} - Q_{ij}^h) d^2s, \quad (2)$$

where homeotropic alignment is described by a radial uniaxial tensor

$$Q_{ij}^h = s_* \left(\hat{r}_i \hat{r}_j - \frac{1}{3} \delta_{ij} \right), \quad \hat{r} = \frac{\mathbf{r}}{r}.$$

(The scalar order parameter $s_* > 0$ is chosen to minimize the bulk density.) Far away from the particle, the alignment is uniform:

$$Q_{ij}(\mathbf{r}) \approx Q_{ij}^\infty \quad \text{as } r \rightarrow \infty, \quad (3)$$

where

$$Q_{ij}^\infty = s_* (\hat{z}_i \hat{z}_j - \frac{1}{3} \delta_{ij}), \quad \hat{z} = (0, 0, 1).$$

The equilibrium equations corresponding to minimization of the free energy functional (1) are

$$\begin{aligned} L \nabla^2 Q_{ij} &= g_{ij}(\mathbf{Q}), \\ L \hat{r}_k \partial_k Q_{ij} &= W (Q_{ij} - Q_{ij}^h) \quad \text{for } r = r_p. \end{aligned}$$

Here the nonlinear term g_{ij} is the traceless part of the tensor $\partial f / \partial Q_{ij}$.

Since we want to investigate the effect of a small-size particle, we rewrite the equilibrium equations in terms of the rescaled variable \tilde{r} defined by $\mathbf{r} = r_p \tilde{r}$, obtaining

$$\begin{aligned} \tilde{\nabla}^2 Q_{ij} &= \frac{r_p^2}{L} g_{ij}(\mathbf{Q}), \\ \hat{r}_k \tilde{\partial}_k Q_{ij} &= \frac{r_p W}{L} (Q_{ij} - Q_{ij}^h) \quad \text{for } \tilde{r} = 1. \end{aligned}$$

Introducing an effective anchoring coefficient w , we assume that

$$r_p^2 \ll L, \quad \frac{r_p W}{L} = w \quad (4)$$

and deduce that any equilibrium configuration Q_{ij} must satisfy

$$Q_{ij} \approx Q_{ij}^0,$$

where \mathbf{Q}^0 is the unique solution of

$$\tilde{\nabla}^2 Q_{ij}^0 = 0, \quad \hat{r}_k \tilde{\partial}_k Q_{ij}^0 = w (Q_{ij}^0 - Q_{ij}^h) \quad \text{for } \tilde{r} = 1,$$

satisfying the far-field condition (3). By the approximation $Q_{ij} \approx Q_{ij}^0$, we mean that the maximum difference between the two solutions collapses to zero as the ratio $r_p^2/L \rightarrow 0$; a mathematical justification of this approximation is given in

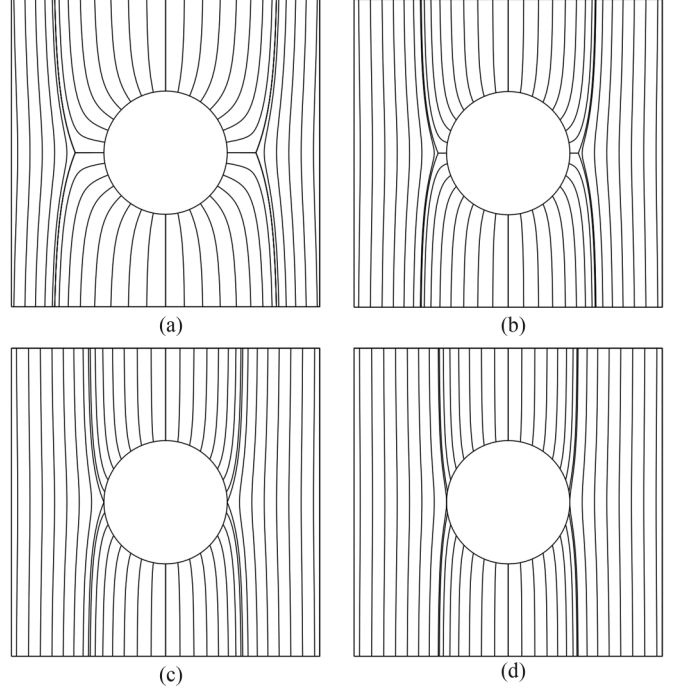


FIG. 1. Direction field \mathbf{n}^0 (eigenvector associated with the largest eigenvalue of \mathbf{Q}^0) as explicitly computed below (7) for different values of the effective anchoring coefficient: (a) $w = \infty$, (b) $w = 3$, (c) $w = 1.732 \approx \sqrt{3}$, and (d) $w = 1$.

[43]. It turns out that \mathbf{Q}^0 has a simple analytical expression given by

$$\mathbf{Q}^0 = \frac{w}{3+w} \frac{1}{\tilde{r}^3} \mathbf{Q}^h + \left(1 - \frac{w}{1+w} \frac{1}{\tilde{r}} \right) \mathbf{Q}^\infty, \quad (5)$$

as can be checked by a direct calculation.

We observe immediately that the tensor field \mathbf{Q}^0 has no singularity, at least not in the sense of Oseen-Frank theory. In fact, at nearly all points \mathbf{Q}^0 is biaxial, so an Oseen-Frank director cannot even be defined. The Saturn ring defect appears instead as an eigenvalue exchange [32], at points where eigenvalues of \mathbf{Q}^0 cross and the principal eigenvector is discontinuous. Thus, the Saturn ring singularity is only apparent when following the direction field \mathbf{n}^0 represented by the normalized principal eigenvector field of \mathbf{Q}^0 . The eigenvector field \mathbf{n}^0 is illustrated for several values of effective anchoring coefficient in Fig. 1 and clearly shows a singular Saturn ring.

Explicit expressions are easily derived for the eigenvalues $\lambda_1(\tilde{r}) \geq \lambda_2(\tilde{r}) \geq \lambda_3(\tilde{r})$ of \mathbf{Q}^0 . To substitute for the nematic director field (which is undefined), we consider a normalized eigenvector $\mathbf{n}^0(\tilde{r})$ associated with principal eigenvalue λ_1 . This is well defined, except at points where $\lambda_1 = \lambda_2$, across which the principal eigenvector \mathbf{n}^0 jumps from the radial to the vertical direction. Such singularity happens exactly at a circle in the horizontal plane: This is the Saturn-ring defect (see [43] for details). The radius of the ring (in units of r_p) is the unique positive solution ρ_w of

$$(\rho_w)^3 - \frac{w}{1+w} (\rho_w)^2 - \frac{w}{3+w} = 0. \quad (6)$$

It has an explicit expression, represented in Fig. 2. As the effective anchoring coefficient w decreases from $+\infty$ to $\sqrt{3}$,

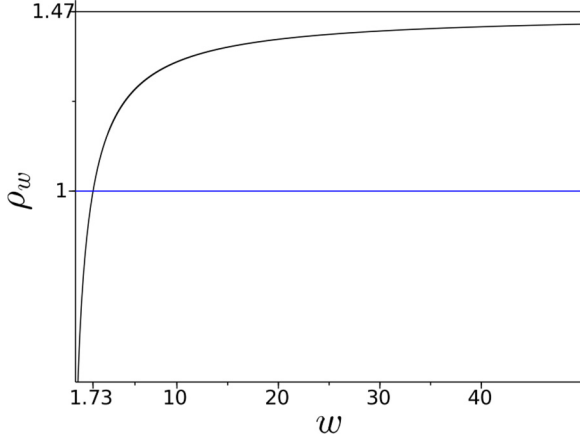


FIG. 2. Ring radius dependence on the effective anchoring coefficient (6). Strong anchoring yields $\rho_\infty \approx 1.47$. The surface ring $\rho_w = 1$ corresponds to $w = \sqrt{3} \approx 1.73$.

the defect ring shrinks from $\rho_\infty \approx 1.47$ to $\rho_{\sqrt{3}} = 1$, where it becomes an equatorial surface ring. When $w < \sqrt{3}$ the anchoring is too weak to enforce a defect: In that case, $\rho_w < 1$ so that the defect ring is virtually inside the particle. Note that in practice, quite strong anchoring would be needed to actually observe the Saturn ring: Taking $L = 10^{-11}$ N as in [32] and a particle radius $r_p = 10$ nm, we find $w = 10^3 W$, so that for $W = 10^{-3}$ J m⁻² our solution exhibits no Saturn ring, while for $W = 10^{-2}$ J m⁻² the Saturn ring radius is $1.34r_p$. Previous studies of the Saturn ring have produced a wide range of candidates for the ring radius, using these specific values for L, W ; the radii obtained were $1.15r_p$ [32], $1.10r_p$ [24], $1.08r_p$ [25], $1.2r_p$ [23], and $1.25r_p$ [22]. These discrepancies might be due to the different values of r_p and w considered there, as well as different means of approximating the actual solutions. With our approach we obtain the explicit dependence on w , for small particle radius r_p .

Away from the defect ring, the director field $\mathbf{n}^0(\vec{r})$ is well defined, as the eigendirection associated with the largest eigenvalue λ_1 , and has an explicit analytical expression. It is obviously symmetric under reflection with respect to the horizontal plane, so we only need to give its formula in the upper half space: In spherical coordinates $(\vec{r}, \theta, \varphi)$, for $0 < \theta < \pi/2$ it holds that

$$\mathbf{n}^0 = \sqrt{\frac{1-\mu}{2}} \hat{\rho} + \sqrt{\frac{1+\mu}{2}} \hat{z}, \quad (7)$$

where $\hat{\rho} = (\cos \varphi, \sin \varphi, 0)$ and $\mu = \mu(\vec{r}, \theta)$ is given by

$$\mu = \frac{\alpha(1 - 2 \sin^2 \theta) + \beta}{\sqrt{\alpha^2 + \beta^2 + 2\alpha\beta(1 - 2 \sin^2 \theta)}},$$

with

$$\alpha = \frac{w}{3+w} \frac{1}{\tilde{r}^3}, \quad \beta = 1 - \frac{w}{1+w} \frac{1}{\tilde{r}}.$$

The director field \mathbf{n}^0 is represented in Fig. 1 for different values of w . It is worth noting that from (7) we deduce the expansion

$$\mathbf{n}^0 = \hat{z} + \frac{w}{3+w} \frac{1}{\tilde{r}^5} (\tilde{x}\tilde{z}, \tilde{y}\tilde{z}, 0) + O\left(\frac{1}{\tilde{r}^4}\right) \quad \text{as } \tilde{r} \rightarrow \infty,$$

so that we recover explicitly the quadrupolar far-field behavior predicted from the electrostatic analogy [44].

III. CONCLUSION

In conclusion, we have presented a method showing the stability of the Saturn ring defect around a very small particle. We obtain the explicit value of the ring radius (on which previous works did not agree) as well as its dependence on the anchoring energy. In particular, measuring the ring radius gives an estimation of the anchoring coefficient. Note that, in practice, our approximation is only valid if the particle is much smaller than the coherence length ξ_0 , so it only holds for very small particles; for larger particles, the r^{-1} far-field behavior of our solution (5) might no longer be valid. Typical values of ξ_0 considered in theoretical studies are of the order of 100 nm, which corresponds to 4-methoxybenzylidene-4'-n-butylaniline [42] or 4-pentyl-4'-cyanobiphenyl [6] liquid crystals (in other nematic systems ξ_0 could be of a different order). Our results are also restricted by the anchoring coefficient, which has to be quite strong ($W = 10^{-2}$ J m⁻² as, e.g., in [32]) to observe the Saturn ring. However, we believe that a similar analytical approach could be of use in a wider range of situations. For instance, larger particles could be studied by introducing a virtual shell around the particle where nonlinear effects cannot be neglected, as in [26]. More generally, while Landau-de Gennes theory was previously used only numerically in the study of nematic colloids, the present paper demonstrates that explicit analytical calculations are also possible. Therefore, we hope that similar techniques could be applied successfully to improve the theoretical understanding of nematic colloidal interactions.

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