

Confinement effect on the interaction between colloidal particles in a nematic liquid crystal: An analytical study

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Motivated by a recent experimental study on the interaction between colloidal particles in a confined nematic liquid crystal [M. Vilfan, N. Osterman, M. Čopič, M. Ravnik, S. Žumer, J. Kotar, D. Babič, and I. Poberaj, *Phys. Rev. Lett.* **101**, 237801 (2008)], we discuss in an analytical manner how the interaction potential U between spherical colloidal particles in a confined nematic cell behaves as a function of the interparticle distance r . We show that the short-range potential follows a power law $U(r) \sim r^{-5}$ as expected from the quadrupolar nature of the interaction, while the long-range potential is dominated by an exponential function $U(r) \sim \sqrt{d}/r \exp(-2\pi r/d)$, where d is the cell thickness. These two regimes are interchanged at $r/d \approx 0.8$. This behavior of $U(r)$ is in a good semiquantitative agreement with the experimental finding.

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I. INTRODUCTION

Liquid-crystal colloids have been attracting growing interest as a novel composite material based on a liquid crystal for the last decade [1–6]. One of the intriguing properties of liquid-crystal colloids is that colloidal particles can interact via the elastic distortions of the host liquid crystal induced by anchoring on the particle surfaces. Such interactions can lead to a various superstructures of colloidal particles specific to liquid-crystal colloids, including linear chains [2,7], two-dimensional crystalline structures [5,8], and cellular structures [9].

It has been shown by an analytical argument that spherical particles with weak surface anchoring, both normal and planar, exhibit a long-range quadrupolar interaction whose potential is proportional to r^{-5} [10–14], with r being the interparticle distance, and that the interaction between particles carrying a satellite hedgehog defect in the case of strong normal surface anchoring is of dipolar type; the potential is proportional to r^{-3} [12–16]. Even the possibility of Coulomb-type potential ($\sim r^{-1}$) has been discussed [13–15].

The first experimental measurement of the colloidal interaction in a nematic liquid crystal was carried out by Poulin *et al.* [17]. They observed how a colloidal particle moves under the influence of the distortion-mediated interaction and determined the interaction force by equating it with a viscous force. Later, Yada and co-workers [18] performed a more direct experimental evaluation of the interaction force by optical tweezers. In those two studies, dipolar interaction between particles carrying a hedgehog with strong normal surface anchoring was measured. Several experimental studies [19–24] have been devoted to a direct measurement of the interaction force or potential between colloidal particles in a liquid crystal.

In most of those experimental and theoretical studies, the host liquid crystal was considered as an infinite medium. In real systems, however, liquid crystals has to be confined in a container, or a cell, and confinement may affect the interaction between colloidal particles. Nevertheless, little attention has been paid to the effect of confinement, and, to our knowledge, the only theoretical study of the interaction between colloidal particles in a confined nematic liquid crystal was carried out by one of the authors [25,26]. It was found that the interaction in a confined cell can be regarded as that between infinite array of “mirror images” of particles (recently the method of mirror images was used also in Ref. [27] to discuss the interaction between a particle and a single wall). Concerning experiments, no previous studies had put emphasis on the effect of confinement until a very recent study focusing on the confinement effect was carried out by Vilfan *et al.* [28]. They used particles with planar surface anchoring and found by careful experiments that when the interparticle distance is on the order of or larger than the cell thickness, the interaction potential decays exponentially, rather than algebraically as expected in theories dealing with an infinite medium. They also confirmed this exponential behavior using numerical calculations.

In our previous study [25,26], attention was paid to the form of the interaction potential profiles when the interparticle distance is on the order of the cell thickness and the long-distance regime of the interaction was not discussed in detail. Motivated by the experimental study by Vilfan *et al.* [28], in this paper, we consider, in an analytical manner, the effect of confinement on the long-distance behavior of the interaction potential between colloidal particles in a nematic liquid crystal.

In Sec. II, we present the calculation of the interaction potential in a confined nematic liquid crystal. We deal with a spherical particle with weak surface anchoring, around which the director field exhibits quadrupolar symmetry. In Sec. III, we discuss in detail how the interaction potential behaves

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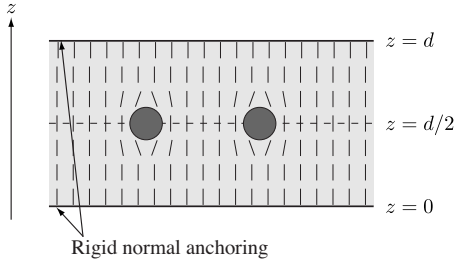


FIG. 1. Geometry of our nematic cell containing two colloidal particles. Rigid normal anchoring is assumed at the cell surfaces $z=0$ and d . In Sec. III, we assume that the centers of the particles are located at the midplane $z=d/2$. Here weak planar anchoring on the particle surfaces is assumed.

with the variation of the interparticle distance. Section IV concludes this paper.

II. CALCULATION OF THE INTERACTION ENERGY

We consider a nematic liquid-crystal cell with thickness d that contains two spherical colloidal particles. At the cell surfaces, we impose rigid normal anchoring, so that the nematic liquid crystal is aligned perpendicular to the cell surfaces. We take the z axis perpendicular to the cell and the cell surfaces are located at $z=0$ and d . The geometry of our cell is illustrated in Fig. 1.

Before proceeding further we comment that there are two ways of a general calculation of the interaction potential between colloidal particles in a uniformly aligned nematic liquid crystal. One is a method developed by Lev *et al.* [12,13], in which the director profile and the resultant interaction energy are determined by minimizing the total free energy of the system: the sum of the Frank elastic energy and the surface anchoring energy of the colloidal particles. One of the advantages of their method is that when the surface anchoring is weak enough, the interaction potential can be determined for particles with arbitrary shape and surface anchoring profiles. However, to facilitate the calculation, their method implicitly assumes that the director field is defined in the whole system, even inside the colloidal particles. The “extrapolation” of the director field inside the particles is carried out, so that the linearized elasticity [Eq. (1) below] is satisfied inside the particle.

Recently Pergamenschik and Uzunova [14,15,27] proposed a different method in which the “auxiliary” director field inside the particles need not be taken into account. They introduced spheres surrounding the particles, outside which the director distortions are small enough to allow linearized elasticity of the director. They showed that once the director profiles at the surface of those spheres are known, the interaction potential can be determined with the aid of the Green’s functions (from the electrostatic analogy, they referred to the director distribution at the sphere surfaces as “elastic charge density”). However, they did not give any prescriptions to determine the elastic charge density for given particles.

In view of the advantage that the interaction potential can be determined when the properties of the colloidal particles

are given, hereafter we use the method of Lev and *et al.* [12,13], which was later adopted by one of the present authors [26] for a nematic cell with finite thickness.

For the calculations to be analytically tractable, we assume that the distortion of the orientational profile of the host nematic liquid crystal is small enough and no defects are present. Therefore a Frank description in terms of the director \mathbf{n} can be used and the distortion of \mathbf{n} is small enough, so that it is described as $\mathbf{n}=(n_x, n_y, 1)$ with $|n_x|, |n_y| \ll 1$. This treatment can be justified if we consider colloidal particles whose surfaces impose sufficiently weak anchoring (as we shall see below, we can treat both normal and planar anchorings so long as weak anchoring is assumed). Then it is sufficient to retain up to second-order terms in n_x and n_y in the Frank elastic energy. We also adopt the one-constant approximation and the Frank elastic energy in terms of n_x and n_y is thus given by

$$F_b = \frac{1}{2}K \int d^2\mathbf{r}_\perp \int_0^d dz \{(\nabla n_x(\mathbf{r}))^2 + (\nabla n_y(\mathbf{r}))^2\}, \quad (1)$$

where K is the elastic constant and $\mathbf{r}_\perp=(x, y)$.

Next we write the surface energy due to the colloidal particles in the Rapini-Papoular form [29] as

$$F_s = \sum_{p=1}^2 \oint_{\Omega_p} d^2S W(s) [\mathbf{v}(s) \cdot \mathbf{n}(s)]^2. \quad (2)$$

Here $p=1, 2$ labels the two particles and Ω_p represents the surface of the p th particle. s denotes the point on the surface and the anchoring strength at point s is given by $W(s)$. The unit surface normal at s is denoted by $\mathbf{v}(s)$. In the case of planar anchoring $W(s) > 0$ and vice versa for normal anchoring. As noted above, we assume sufficiently weak surface anchoring, so that $WR_0/K \ll 1$, in which R_0 is the characteristic size of the particles.

In the treatment of this surface energy, we make a gradient expansion of the director field $\mathbf{n}(s)$ around a point $\mathbf{r}^{(p)}$ which specifies the position of the p th particle. In the case of spherical particles, it is reasonable from the symmetry to choose the center as $\mathbf{r}^{(p)}$. When the particle radius is R_0 and the anchoring strength W is uniform, the surface free energy (2) becomes

$$F_s \simeq \sum_{p=1}^2 \sum_{l=x,y} \mathcal{A}_{z,l}^{(p)} n_l(\mathbf{r}^{(p)}), \quad (3)$$

in which

$$\mathcal{A}_{z,l}^{(p)} = 2\Gamma \frac{\partial^2}{\partial r_z^{(p)} \partial r_l^{(p)}} \quad (4)$$

with $\Gamma=4\pi WR_0^4/15$. In Eq. (4), we have retained the lowest-order relevant term in the gradients (zeroth- and first-order terms are absent). For this truncation to be valid, the characteristic size of the particle R_0 must be much smaller than the characteristic length of the deformation of the host nematic liquid crystal. Otherwise, higher-order terms in the gradients should be retained in $\mathcal{A}_{z,l}^{(p)}$. Higher-order terms in the gradients can influence the short-distance behavior of the interac-

tion potential. However, as shown in the Appendix, higher-order terms give only a minor contribution in the long-distance behavior.

In the cases of particles with general shape or nonuniform surface anchoring, the operator $\mathcal{A}_{z_l}^{(p)}$ is modified. How to construct $\mathcal{A}_{z_l}^{(p)}$ for general particles can be found in Ref. [26].

The total free energy of the system F in the presence of particles can be written as the sum of the Frank elastic energy F_b [Eq. (1)] and the surface energy F_s [Eq. (3)]. The distortion profile of the nematic liquid crystal is the one that minimizes the total free energy and thus satisfies

$$\frac{\delta(F_b + F_s)}{\delta n_{x,y}} = 0. \quad (5)$$

Substituting the profile minimizing the total free energy into $F = F_b + F_s$, we obtain the total free energy in the presence of colloidal particles. The result is formally written as $F = U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) + \sum_{p=1}^2 F_p(\mathbf{r}^{(p)})$, where $U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)})$ is the interaction energy between two particles and $F_p(\mathbf{r}^{(p)})$ is the self-energy of one particle p . Here we do not repeat the arguments for the separation of the total free energy into the interaction and the self-energy parts. We also just mention that the self-energy can be regarded as the interaction between a particle and two confining surfaces, and that $F_p(\mathbf{r}^{(p)})$ is minimized when $r_z^{(p)} = d/2$ [26]. We note that a recent experiment [30] reported the levitation of a colloidal particle in a nematic cell attributable to the self-energy in the sense mentioned above, although the experimental setup, in which the confining surfaces adopt planar alignment and the particle carries a hedgehog defect, is different from our theoretical one.

The interaction potential $U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)})$ is explicitly given by [26]

$$U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) = -\frac{1}{4\pi K} \sum_{l=x,y} \mathcal{A}_{z_l}^{(1)} \mathcal{A}_{z_l}^{(2)} [h_1(|\mathbf{r}_\perp^{(12)}|, r_z^{(1)} - r_z^{(2)}) - h_1(|\mathbf{r}_\perp^{(12)}|, r_z^{(1)} + r_z^{(2)})], \quad (6)$$

where $\mathbf{r}_\perp^{(pp')} = \mathbf{r}_\perp^{(p)} - \mathbf{r}_\perp^{(p')}$ and we have defined

$$h_1(\xi, \eta) = \frac{1}{\sqrt{\xi^2 + \eta^2}} + \sum_{m=1}^{\infty} \left\{ \frac{1}{\sqrt{\xi^2 + (\eta - 2md)^2}} + \frac{1}{\sqrt{\xi^2 + (\eta + 2md)^2}} - \frac{1}{md} \right\}. \quad (7)$$

In the following we discuss how $U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)})$ behaves with the variation of the interparticle distance.

III. DISCUSSION

As noted above, the self-energy of a particle is minimized when it is located at $z=d/2$. Therefore, we restrict our following discussion to the cases where two particles are located at $z=d/2$, that is, $r_z^{(1)} = r_z^{(2)} = d/2$. In those cases, the distance between the centers of the particles is equal to $|\mathbf{r}_\perp^{(12)}|$.

A. Short-distance regime

In the case of small interparticle distance, the first term of Eq. (7) in $h_1(|\mathbf{r}_\perp^{(12)}|, r_z^{(1)} - r_z^{(2)})$ dominates. In other words, the

cell thickness d is not relevant and the terms involving d in Eq. (7) can be neglected. Therefore, the interaction potential $U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)})$ in the short-distance regime becomes

$$U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) \simeq -\frac{\Gamma^2}{\pi K} \frac{\partial}{\partial r_z^{(1)}} \frac{\partial}{\partial r_z^{(2)}} \left(\frac{\partial}{\partial r_x^{(1)}} \frac{\partial}{\partial r_x^{(2)}} + \frac{\partial}{\partial r_y^{(1)}} \frac{\partial}{\partial r_y^{(2)}} \right) \times \left| \frac{1}{|\mathbf{r}^{(1)} - \mathbf{r}^{(2)}|} \right|_{r_z^{(1)}=r_z^{(2)}=d/2} = \frac{9\Gamma^2}{\pi K} \frac{1}{|\mathbf{r}_\perp^{(12)}|^5}. \quad (8)$$

In the case of spherical particles with weak surface anchoring, quadrupolar interaction whose potential is proportional to the fifth power of the interparticle distance has already been derived in an infinite medium [10–13]. In our case, when the interparticle distance is small enough, the effect of confinement is negligible because the confining walls are far away from the two particles as compared with their distance. Therefore it is quite natural that the quadrupolar nature of the interaction is reproduced in this short-distance regime. We also note that the interaction is repulsive. It should be mentioned that when higher-order terms in the gradients are retained in the operator $\mathcal{A}_{z_l}^{(p)}$, a contribution whose power in $|\mathbf{r}_\perp^{(12)}|$ is different from -5 can exist, for example, $|\mathbf{r}_\perp^{(12)}|^{-7}$.

B. Long-distance regime

Before discussing the long-distance regime, we recall that $h_1(\xi, \eta)$ defined in Eq. (7) can be rewritten as

$$h_1(\xi, \eta) = \frac{2}{d} \sum_{m=1}^{\infty} K_0(m\pi\xi/d) \cos(m\pi\eta/d) - \frac{1}{d} \left(\gamma + \ln \frac{\xi}{4d} \right). \quad (9)$$

Here K_0 is a modified Bessel function and γ is Euler's constant. See the Appendix of Ref. [25] or Ref. [31] for the derivation of Eq. (9). The last term in Eq. (9) is unimportant because it exactly cancels out in $h_1(|\mathbf{r}_\perp^{(12)}|, r_z^{(1)} - r_z^{(2)}) - h_1(|\mathbf{r}_\perp^{(12)}|, r_z^{(1)} + r_z^{(2)})$ in $U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)})$. Using Eq. (9), one can rewrite the interaction potential $U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)})$, or Eq. (6), as

$$U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) = -\frac{\Gamma^2}{\pi K} \frac{2}{d} \sum_{m=1}^{\infty} \left[\sum_{l=x,y} \frac{\partial^2}{\partial r_l^{(1)} \partial r_l^{(2)}} K_0 \left(\frac{m\pi |\mathbf{r}_\perp^{(12)}|}{d} \right) \right] \times \frac{\partial^2}{\partial r_z^{(1)} \partial r_z^{(2)}} \left(\cos \frac{m\pi(r_z^{(1)} - r_z^{(2)})}{d} - \cos \frac{m\pi(r_z^{(1)} + r_z^{(2)})}{d} \right) \Bigg|_{r_z^{(1)}=r_z^{(2)}=d/2} = \frac{\Gamma^2}{\pi K} \frac{16\pi^2}{d^3} \sum_{m'=1}^{\infty} m'^2 \Delta_\perp K_0 \left(\frac{2m'\pi |\mathbf{r}_\perp^{(12)}|}{d} \right). \quad (10)$$

Here we have defined a two-dimensional Laplacian $\Delta_\perp \equiv \sum_{l=x,y} \frac{\partial^2}{\partial (r_l^{(1)})^2} = \sum_{l=x,y} \frac{\partial^2}{\partial (r_l^{(2)})^2}$. It should be noticed that only the contributions from even $m (=2m')$ survive and the contributions from odd m vanish.

We also note that the modified Bessel function $K_0(x)$ for large x behaves as

$$K_0(x) \approx e^{-x} \sqrt{\frac{\pi}{2x}} \quad (11)$$

and decays fast for $x \rightarrow \infty$. Therefore, for large interparticle distance, the term with $m'=1$ dominates in Eq. (10), and thus

$$U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) \approx \frac{\Gamma^2}{\pi K} \frac{16\pi^2}{d^3} \Delta_{\perp} K_0\left(\frac{2\pi|\mathbf{r}_{\perp}^{(12)}|}{d}\right). \quad (12)$$

Using Eq. (11), we can further simplify Eq. (12) to yield

$$U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) \approx \frac{\Gamma^2}{\pi K} \frac{32\pi^4}{d^5} \sqrt{\frac{d}{|\mathbf{r}_{\perp}^{(12)}|}} \exp\left(-\frac{2\pi|\mathbf{r}_{\perp}^{(12)}|}{d}\right). \quad (13)$$

In interpreting their experimental results, Vilfan *et al.* [28] argued, using electrostatic analogy, that the interaction potential can be written in the form of $K_0(2\pi|\mathbf{r}_{\perp}^{(12)}|/d)$ (in our notation) and compared their results using its asymptotic form $\exp(-2\pi|\mathbf{r}_{\perp}^{(12)}|/d)$. Our analytical argument indicates that the interparticle dependence of the interaction potential is given by $\Delta_{\perp} K_0(2\pi|\mathbf{r}_{\perp}^{(12)}|/d)$ for large interparticle distance and that in the previous argument the two-dimensional Laplacian Δ_{\perp} was missing.

C. Behavior of the interaction potential for all distances

Here we discuss the behavior of the interaction potential for all distances. For this purpose, we use the second equality of Eq. (10). However, the infinite sum there cannot be evaluated analytically or numerically. Therefore in our numerical calculation presented below, the infinite sum $\sum_{m'=1}^{\infty}$ is replaced with $\sum_{m'=1}^{100}$ and we make use of MAPLE 11.02. We have checked that no observable differences can be found between the sums $\sum_{m'=1}^{100}$ and $\sum_{m'=1}^{200}$, and therefore the contributions from the terms with $m' > 100$ can be safely neglected. In the following we abbreviate $|\mathbf{r}_{\perp}^{(12)}|$ as r .

In Fig. 2(a), we plot the rescaled interaction potential $U(r)/(\Gamma^2/\pi K d^5)$ as a function of the interparticle distance rescaled by the cell thickness, i.e., r/d , for $0.05 \leq r/d \leq 5$. We find from Fig. 2(a) that, for $r/d \geq 0.8$, the interaction potential [solid red line, Eq. (10)] is indistinguishable from that in the long-distance regime [dashed green line, Eq. (12) with $\Delta_{\perp} K_0(2\pi r/d)$, or dashed-dotted black line, Eq. (13), with an exponential form]. On the other hand the interaction potential falls on the power-law behavior r^{-5} [dotted blue line; see Eq. (8)] for $r/d \leq 0.8$. Figure 2(a) also demonstrates that Eqs. (12) and (13) show almost the same behavior, at least in the range of r/d plotted there.

To see the transient behavior around $r/d \approx 0.8$ more closely, in Fig. 2(b) we give the same but magnified plot for $0.6 \leq r/d \leq 1$. We see from Fig. 2(b) that the interaction potential can be expressed by the short-range power-law behavior fairly well for $r/d \leq 0.7$ and by the long-distance regime [Eq. (12) or Eq. (13)] for $r/d \geq 0.9$. However, if one is interested not in the absolute value of the interaction potential but in the logarithm of it as in Fig. 2(a), we can safely say that the interaction potential is expressed by the power-law behavior ($\sim r^{-5}$) and the long-distance behavior [Eq. (12) or Eq. (13)] interchanged at $r/d \approx 0.8$.

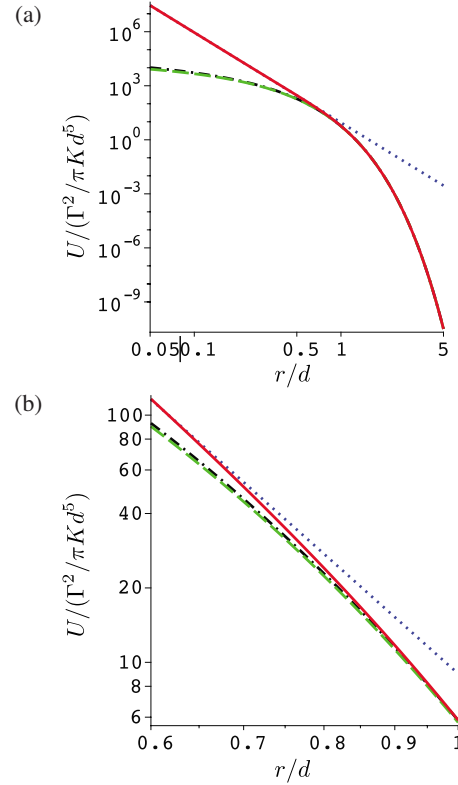


FIG. 2. (Color online) Log-log plots of the rescaled interaction potential $U(r)/(\Gamma^2/\pi K d^5)$ as a function of the rescaled interparticle distance r/d for the range (a) $0.05 \leq r/d \leq 5$ and (b) $0.6 \leq r/d \leq 1$. Solid red lines represent the full interaction potential calculated using Eq. (10) with replacing the infinite sum with $\sum_{m'=1}^{100}$. Dotted straight blue lines are the power-law behavior given in Eq. (8). Dashed green lines correspond to Eq. (12), the long-distance result with $\Delta_{\perp} K_0(2\pi r/d)$. Black dashed-dotted lines represent the exponential behavior in Eq. (13).

In the experiments of Vilfan *et al.* [28], the short-range power-law behavior is found for $r \leq 0.9d$, in good agreement with our theoretical finding. We also note that, for the same interparticle distance r , Fig. 2(a) clearly indicates that the interaction potential becomes larger for larger cell thickness d . This tendency can be found in a clear manner in the experiments and numerical calculations presented in Ref. [28].

To conclude this section, we make a more quantitative comparison of our results with experimental ones. In the experiments of Ref. [28], the absolute values of the interparticle potential are given for $d = 1.5 \times 2R_0$ and $1.8 \times 2R_0$. From Fig. 2 of Ref [28], we find that, for $d = 1.5 \times 2R_0$, $U(r \approx 2R_0)/U(r \approx 2.7 \times 2R_0) = U(r \approx 0.67d)/U(r \approx 1.8d) \approx 10^4$. From our theoretical results, we have

$$\frac{U(r \approx 0.67d)}{U(r \approx 1.8d)} \approx 2 \times 10^3. \quad (14)$$

As mentioned below, for our calculations to be analytically tractable, we have made a number of assumptions that may not conform to the experimental situations. With this in

mind, we can say that the agreement between the experimental findings and our result (14) is satisfactory.

D. Some further remarks on our theoretical approach

Here we recall several assumptions made for our calculations to be performed analytically. We assumed perfect normal anchoring at the cell surfaces and real surfaces with finite anchoring strength, even when the anchoring is sufficiently strong, might yield a small quantitative difference.

Concerning colloidal particles, we made the assumption of weak surface anchoring and the smallness of colloidal particles as compared to the cell thickness d . The former guarantees that the elastic distortion of the host nematic liquid crystals is small enough to enable one to expand the Frank energy and retain terms up to second order in n_x and n_y [see Eq. (1)]. The latter allows a gradient expansion of the surface energy and retaining the lowest-order relevant term in the gradients [Eqs. (3) and (4)].

In the experiments of Ref. [28], the diameter of the particle is on the order of the cell thickness, which does not conform to our assumption of small particle size (in Ref. [28], nothing was mentioned about the anchoring strength of the particle surfaces, but in their simulations strong surface anchoring was employed). Nevertheless, as we have seen in Sec. III C, we find some semiquantitative agreement of our theory with the experiments. One might wonder why we can observe such a semiquantitative agreement in spite of the difference between the experimental situation and the assumptions made in our theory. As noted in Sec. II, when the particle is not sufficiently small, we may have to retain higher-order terms in the gradients in the operator $\mathcal{A}_{z_l}^{(p)}$ [Eq. (4)]; our theoretical results presented above should be regarded as giving a leading-order contribution lowest order in the gradients. Vilfan *et al.* [28] found in their experiments that for $d=1.5 \times 2R_0$, the short-range interaction potential obeys a power law $U \sim r^{-\beta}$, but $\beta=5.4$ not 5 as expected theoretically. This difference might be a manifestation of the presence of higher-order contributions in the gradients. On the other hand, as shown in the Appendix, higher-order terms in the gradients in $\mathcal{A}_{z_l}^{(p)}$ yields only a minor contribution in the long-range interaction potential. Therefore, our result in the long-distance regime given in Sec. III B can be safely applied even when particles are not necessarily small.

Concerning the anchoring strength, we note here that in Ref. [13] Lev *et al.* introduced the concept of “coat,” outside of which the elastic distortions are small enough to allow an expansion of the Frank elastic energy and retaining only up to second order in n_x and n_y as mentioned above. In the case of weak surface anchoring, the concept of the coat need not be introduced, because the integration of the surface anchoring energy density [Eq. (2)] can be carried out at the actual particle surfaces. On the other hand, when the anchoring is strong enough for topological defect(s) such as a hedgehog, a Saturn ring, or boojums to accompany the particle, the coat should be taken large enough to cover the defects with strong elastic distortions inside. In the case of planar surface anchoring where the orientational profile outside the coat has a quadrupolar symmetry, one can safely use our analytical scheme to evaluate the interaction potential.

We also note that the assumption of weak surface anchoring leads to the conclusion that particles with normal anchoring [$W < 0$ in Eq. (2)] give the same resultant interaction potential as those with planar anchoring ($W > 0$). That is, particles with small interparticle distance experience quadrupolar interaction irrespective of the type of surface anchoring. The quadrupolar nature of the interaction for small interparticle distance will remain valid even when the anchoring is not weak, so long as the planar anchoring is assumed. However, in the case of normal anchoring, a spherical particle with large anchoring strength induces the dipolar orientational profile around it due to a satellite hedgehog defect accompanying the particle [2,4,16]. In other words, the shape of the coat loses one of the mirror symmetry planes [13]. In that case, a quadrupolar picture of the interaction is no longer valid, and therefore a different treatment of the interaction potential will be necessary. Therefore, when particles with normal anchoring are used, a special care is required if one wants to compare their experimental results with ours.

Here we comment on how the absolute value of the interaction potential depends on the characteristic parameters of the particles. In the case of weak surface anchoring and small particle size, the interaction potential is proportional to $\Gamma^2 \propto W^2 R_0^8$, where the parameter Γ was defined just after Eq. (4), W is the surface anchoring strength, and R_0 is the particle radius. Although the dependence of the interaction potential on the particle radius might be obtained by experiments or simulations, one has to keep in mind that the above R_0^8 dependence was derived in the case of sufficiently small R_0 and weak surface anchoring. When the particle is not small as compared to the cell thickness, or surface anchoring should be regarded as strong, one must resort to the above-mentioned coat description and/or retain higher-order gradients in $\mathcal{A}_{z_l}^{(p)}$. However, how to determine the operator $\mathcal{A}_{z_l}^{(p)}$ in the coat description is not a trivial problem. What one could do is to compare the far-field behavior of the orientation profiles of the “original” particle with strong anchoring and that of a coat with specific values of W and R_0 . It may be a difficult task, but once the coat is specified, one can determine the operator $\mathcal{A}_{z_l}^{(p)}$ and compare our theory with experiments in a direct and quantitative manner.

IV. CONCLUSION

We carried our an analytical study as to how confinement by two parallel surfaces affects the interaction potential between spherical colloidal particles in a nematic liquid crystal. Our study was motivated by a recent experiment, together with a simulation study, which found an exponential decay of the interparticle potential in a confined nematic liquid crystal [28]. We found an analytical formula on the interparticle potential in a nematic cell whose confining surfaces impose rigid normal anchoring. Our potential can be regarded as consisting of two regimes: in a short-distance regime with $r \leq 0.8d$, where r is the interparticle distance and d is the cell thickness, and the potential obeys a power law $U(r) \sim r^{-5}$. On the other hand, in a long-distance regime with $r \geq 0.8d$, it behaves as $U(r) \sim \Delta_{\perp} K_0 (2\pi r/d)$ or

$U(r) \sim \sqrt{d/r} \exp(-2\pi r/d)$ with K_0 being a modified Bessel function.

Before concluding this paper we mention that, although our present study was restricted to the case of normal anchoring on the cell surfaces, we can consider the case of planar anchoring whose preferred direction is fixed to one direction [26]. In that case the interparticle potential depends not only on the interparticle distance $|r_{\perp}^{(12)}|$ but also on the direction of $|r_{\perp}^{(12)}|$ with respect to the easy direction at the cell surfaces. Therefore we can expect a richer behavior of the interaction potential.

So far as we know, Ref. [28] is the only experimental and simulation study focusing on the effect of confinement on the interparticle potential in a nematic liquid crystal. Much remains to be investigated in this subject and we hope that the present study will stimulate further experimental or simulation study in this direction.

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APPENDIX

In this appendix, we consider how the higher-order terms in the gradients in the operator $\mathcal{A}_{zl}^{(p)}$ contribute to the interaction potential $U(r)$ in the long-distance regime. From the procedures developed in Refs. [12,13,25,26], in the case of spherical particles, we find that the operator $\mathcal{A}_{zl}^{(p)}$ is given, up to the second lowest order in the gradients, by

$$\mathcal{A}_{zl}^{(p)} = 2\Gamma \frac{\partial^2}{\partial r_z^{(p)} \partial r_l^{(p)}} + \delta_{zlm\nu\rho\lambda} \frac{\partial^4}{\partial r_z^{(p)} \partial r_{\mu}^{(p)} \partial r_{\nu}^{(p)} \partial r_{\rho}^{(p)} \partial r_{\lambda}^{(p)}} \quad (\text{A1})$$

instead of Eq. (4). Here

$$\delta_{zlm\nu\rho\lambda} = \frac{1}{12} \oint_{\Omega_p} d^2S W(s) (s - \mathbf{r}^{(p)})_{\mu} (s - \mathbf{r}^{(p)})_{\nu} (s - \mathbf{r}^{(p)})_{\rho} \times (s - \mathbf{r}^{(p)})_{\lambda} \nu_z(s) \nu_l(s). \quad (\text{A2})$$

Here Ω_p denotes the surface of the p th particle. The integral is taken over Ω_p and d^2S is the surface element. In Eq. (A1), summations over repeated indices from x to z are implied. After some calculations, the explicit form of $\mathcal{A}_{zl}^{(p)}$ for $l=x$ and y is given by

$$\mathcal{A}_{zl}^{(p)} = 2\Gamma \frac{\partial^2}{\partial r_z^{(p)} \partial r_l^{(p)}} + \frac{R_0^2}{7} \Gamma \times \left\{ \frac{\partial^4}{\partial (r_z^{(p)})^3 \partial r_l^{(p)}} + \frac{\partial^4}{\partial r_z^{(p)} \partial (r_l^{(p)})^3} + \frac{\partial^4}{\partial r_z^{(p)} \partial r_l^{(p)} (r_k^{(p)})^2} \right\}, \quad (\text{A3})$$

where R_0 is the particle radius. In the last term of Eq. (A3), $k \neq z, l$.

After some tedious but straightforward calculations along the same line as in Sec. III B with the aid of Eq. (11), we find that the asymptotic long-range form of the interaction is written, instead of Eq. (13), as

$$U(\mathbf{r}^{(1)}, \mathbf{r}^{(2)}) \simeq \frac{\Gamma^2}{\pi K} \frac{32\pi^4}{d^5} \sqrt{\frac{d}{r}} \exp\left(-\frac{2\pi r}{d}\right) \times \left[1 + \frac{R_0^2}{28r^2} + \mathcal{O}\left(\frac{R_0^2 d}{r^3}\right) \right], \quad (\text{A4})$$

where $r \equiv |r_{\perp}^{(12)}|$ as in Sec. III C. Equation (A4) indicates that the ratio of the contribution of next-lowest-order terms in the gradients to that of lowest-order terms becomes $R_0^2/28r^2$. The interparticle distance r is always larger than the particle diameter $2R_0$, and with the factor of $1/28$, we can safely say that the higher-order contributions in the operator $\mathcal{A}_{zl}^{(p)}$ yield only a minor effect on the long-distance behavior of the interaction potential.

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