Colloidal interactions in a homeotropic nematic cell with different elastic constants

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We propose a theoretical description of the interaction mediated by a nematic-liquid-crystal host with different Frank elastic constants. A general expression for the energy of such an interaction between colloidal particles of arbitrary size and shape suspended in a homeotropic cell is obtained. In the cells of large thickness, the presented potential converges to that found previously for small particles in the nematic bulk. In general, our results confirm the validity of the one-constant approximation for weakly elastically anisotropic nematic liquid crystals. For nematics with a high splay-to-bend ratio we predict a larger range of the interaction. Using the dependence of this range on the elastic constants, we show that there exists a qualitative similarity between the interactions in a nematic and in a smectic-*A* phase. It manifests itself, in particular, in a decrease of the angle between a chain of quadrupole particles and the uniform far-field director across a nematic–smectic-*A* phase transition. We also demonstrate that the anisotropy of the elastic constants can lead to the formation of thermodynamically stable linear superstructures of asymmetric particles (elastic monopoles) with large, compared to usual dipole chains, interparticle distances.

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I. INTRODUCTION

Liquid-crystal colloids are of considerable interest because of their unusual properties arising from a specific type of interaction between colloidal particles [1]. Particles suspended in such a medium distort its orientational ordering. An overlap of the distortions, produced by different particles, gives rise to the effective colloidal interactions, which do not occur in usual isotropic hosts [2]. These anisotropic long-range interactions in nematic liquid crystals result in various superstructures such as linear [2,3] and inclined chains [4–6] of beads. One can observe a rich variety of two-dimensional crystals formed by colloidal particles suspended in a thin nematic cell or at a nematic-air interface [7–9]. These structures, as well as three-dimensional colloidal crystals [10], are very sensitive to external electromagnetic fields [10–12] and might be utilized as unique composite materials [13].

An understanding of the interactions, mediated by a liquid-crystal medium, is essential for controlling and predicting properties and behavior of composite materials based on colloidal systems of this type. The vast majority of approaches toward an analytical description of the elastic interactions in a nematic host are based on the so-called one-constant approximation, which assumes equal splay K_1, twist K_2, and bend K_3 elastic constants [14–18]. Despite minor differences, they all employ the fact that under such an assumption small director deformations are governed by the Laplace equation and therefore can be expanded in multipoles. This makes it possible to greatly simplify the problem via a representation of the particle by an effective pointlike source of the deformations. Some of these approaches [14,18] have been confirmed experimentally in both bulk [19–21] and confined [22,23] liquid crystals. However, those experiments were carried out with quite typical calamitic nematics, in which the elastic constants are indeed comparable.

At the same time, a growing interest in chromonic (with the twist constant being an order of magnitude smaller than the splay and bend [24]) and bent-core nematics (where the splay constant is a few times higher that the bend [25]) raises a question about the role of the elastic constants in the interactions mediated by a liquid-crystal host. It has to be admitted that other effects besides the elasticity (particularly flexoelectricity and possible chirality) can contribute to the interactions between colloids in such media. Those mechanisms lie beyond the scope of the present study, which is restricted to only the interactions arising from the elastic properties of the host. A similar problem was considered in [26], but the authors focused on the case of small particles embedded in an infinite nematic liquid crystal. In practice, though, the host medium must be always confined. Theoretical [18,27,28] as well as experimental [22,23] studies demonstrate that the presence of confining surfaces can significantly influence the interaction potential. Therefore, in this paper we propose a theory of the elastic interactions between colloidal particles of arbitrary shape and size suspended in a homeotropic nematic cell, a commonly used experimental setup consisting of a liquid crystal confined by two parallel planes with perpendicular alignment of the director.

II. EFFECTIVE FREE-ENERGY FUNCTIONAL

The bulk free energy of a nematic liquid crystal can be written in the well-known Frank form

$$F_{\text{bulk}} = \frac{1}{2}\int d\mathbf{r}\{K_1(\operatorname{div}\mathbf{n})^2 + K_2(\mathbf{n}\cdot\operatorname{rot}\mathbf{n})^2 + K_3(\mathbf{n}\times\operatorname{rot}\mathbf{n})^2\}, \tag{1}$$

where $\mathbf{n} = \mathbf{n}(\mathbf{r})$ is the director field and K_1, K_2, and K_3 are the splay, twist, and bend elastic constants, respectively. In order to simplify this functional the one-constant approximation $K_1 = K_2 = K_3 = K$ is commonly adopted. Under such an

assumption, the bulk energy reduces to

$$\tilde{F}_{\text{bulk}} = \frac{K}{2}\int d\mathbf{r}\{(\text{div}\,\mathbf{n})^2 + (\text{rot}\,\mathbf{n})^2\}. \quad (2)$$

Due to a preferred alignment of the director at the surface of a colloidal particle, the bulk energy has to be supplemented by a surface term. The latter is usually taken in the form of Rapini-Papoular energy

$$F_{\text{surf}} = \oint d\mathbf{s}\, W(\mathbf{s})[\boldsymbol{\nu}(\mathbf{s})\cdot\mathbf{n}(\mathbf{s})]^2, \quad (3)$$

where W is the anchoring constant and $\boldsymbol{\nu}$ is the outer normal at point $\mathbf{s}$ on the particle surface. As a result of competition between the bulk and surface energies, the director deviates from its homogeneous ground state $\mathbf{n}_0 = (0,0,1)$. When the deviations are small, i.e., $\mathbf{n}(\mathbf{r}) \approx (n_x, n_y, 1)$, one can further simplify the free energy (2) to the harmonic form

$$\tilde{F}^{\text{har}}_{\text{bulk}} = \frac{K}{2}\int d\mathbf{r} \sum_{\mu=x,y} (\nabla n_\mu \cdot \nabla n_\mu)^2. \quad (4)$$

The Euler-Lagrange equations arising from (4) are of Laplace type $\Delta n_\mu = 0$ and their solutions at large distances can be expanded in multipoles

$$n_\mu(\mathbf{r}) = \frac{\tilde{q}_\mu}{r} + \frac{\tilde{p}^\alpha_\mu r_\alpha}{r^3} + \frac{\tilde{Q}^{\alpha\beta}_\mu r_\alpha r_\beta}{r^5} + \cdots, \quad (5)$$

where α and β take values x, y, and z (summation over repeated greek indices is implied hereafter). Quantities $\tilde{q}_\mu$, $\tilde{p}^\alpha_\mu$, and $\tilde{Q}^{\alpha\beta}_\mu$, called, respectively, elastic monopole, dipole, and quadrupole moments, determine the director field far from the particle surface and, consequently, the long-range interactions between the particles (in case we have more than one particle suspended in a nematic host). It has been shown in [14] that one can easily derive these interactions by replacing the surface energy (3) of every particle with an effective pointlike source of the director distortions. In particular, for a system of N axially symmetric particles instead of the total energy $\tilde{F}^{\text{har}}_{\text{bulk}} + \sum_{i=1}^N F^i_{\text{surf}}$ we have $\tilde{F}^{\text{har}}_{\text{bulk}} + F^{\text{AS}}_{\text{source}}$, where

$$F^{\text{AS}}_{\text{source}} = -4\pi K \int d\mathbf{r}[\tilde{P}(\mathbf{r})\partial_\mu n_\mu + \tilde{C}(\mathbf{r})\partial_z\partial_\mu n_\mu], \quad (6)$$

with $\tilde{P}(\mathbf{r}) = \sum_{i=1}^N \tilde{p}_i\delta(\mathbf{r}-\mathbf{r}_i)$ and $\tilde{C}(\mathbf{r}) = \sum_{i=1}^N \tilde{Q}_i\delta(\mathbf{r}-\mathbf{r}_i)$ being dipole and quadrupole moment densities. Although (6) was initially derived for an infinite nematic, it is suitable for confined systems as well. For instance, for beads of radius a accompanied by hyperbolic hedgehogs the multipole moments $\tilde{p} = 2.04a^2$ and $\tilde{Q} = -0.72a^3$ provide good agreement between theoretical and experimental results in a planar nematic cell [23]. However, this approximation relies on the electrostatic analogy rooted in (4). Therefore, it seems that the source (6) may be not valid when dealing with the elastically anisotropic bulk energy (1). Fortunately, for this case (in an infinite nematic) the authors of [26] have obtained qualitatively the same representation directly from the Rapini-Papoular energy (3). Assuming weak anchoring ($Wd/K < 1$, where d is the particle size) at the particle surface and expanding $\mathbf{n}(\mathbf{s})$ into the Taylor series about its center they showed that

$$\sum_{i=1}^N F^i_{\text{surf}} \approx \sum_{i=1}^N \{\alpha^i_{z\mu} n_\mu + \beta^i_{z\mu\alpha}\partial_\alpha n_\mu + \gamma^i_{z\mu\alpha\beta}\partial_\alpha\partial_\beta n_\mu\}, \quad (7)$$

where n_μ, similarly to (6), have to be taken at the center $\mathbf{r}^i$ of the ith particle and $\alpha_{z\mu} = 2\oint d\mathbf{s}\, W\nu_z\nu_\mu$, $\beta_{z\mu\alpha} = 2\oint d\mathbf{s}\, W\nu_z\nu_\mu d_\alpha$, and $\gamma_{z\mu\alpha\beta} = \oint d\mathbf{s}\, W\nu_z\nu_\mu d_\alpha d_\beta$ with $\mathbf{d} = \mathbf{s} - \mathbf{r}^i$. These quantities $\alpha_{z\mu}$, $\beta_{z\mu\alpha}$, and $\gamma_{z\mu\alpha\beta}$ are not elastic multipole moments *per se*, but they obey the same symmetry requirements [28,29]. On these grounds, we may conclude that for our case the effective source of the distortions can be written as a generalization of (6) on colloidal particles of arbitrary shape

$$F_{\text{source}} = -4\pi K \int d\mathbf{r}\big[q_\mu(\mathbf{r})n_\mu + p^\alpha_\mu(\mathbf{r})\partial_\alpha n_\mu + Q^{\alpha\beta}_\mu(\mathbf{r})\partial_\alpha\partial_\beta n_\mu\big], \quad (8)$$

where K is an effective elastic constant that allows the elastically isotropic description of the host medium, $q_\mu(\mathbf{r}) = \sum_{i=1}^N q^i_\mu\delta(\mathbf{r}-\mathbf{r}_i)$, $p^\alpha_\mu(\mathbf{r}) = \sum_{i=1}^N p^{\alpha,i}_\mu\delta(\mathbf{r}-\mathbf{r}_i)$, and $Q^{\alpha\beta}_\mu(\mathbf{r}) = \sum_{i=1}^N Q^{\alpha\beta,i}_\mu\delta(\mathbf{r}-\mathbf{r}_i)$. Strictly speaking, the multipole expansion (5), which gives rise to the concept of the elastic multipoles, does not minimize the anisotropic free energy (1). Therefore, beyond the one-constant approximation Eq. (5) should be taken as an ansatz for the director field. In the general case, the parameters of this ansatz q_μ, p^α_μ, and $Q^{\alpha\beta}_\mu$ may differ from those found in the one-constant limit (denoted by tildes). Nevertheless, we can still refer to these parameters as the elastic multipoles since the interparticle interaction, as will be seen below, exhibits an appropriate power-law behavior despite the anisotropy of the elastic constants.

It is worth noting that within the one-constant approximation $K = K_1 = K_2 = K_3$, but beyond it the exact value of K is in fact unknown. This means that the choice of K influences the values of all multipole moments $(q_\mu, p^\alpha_\mu, Q^{\alpha\beta}_\mu)$ corresponding to a given particle. If we fix $(q_\mu, p^\alpha_\mu, Q^{\alpha\beta}_\mu)$, then K can be found as a fitting parameter and vice versa. The only way for unambiguous experimental measurement of the values of $(q_\mu, p^\alpha_\mu, Q^{\alpha\beta}_\mu)$ is by light scattering on the director field $\mathbf{n}(\mathbf{r})$.

III. PAIRWISE INTERACTIONS

Let us now address the geometry of our problem. The system under consideration is confined by two parallel planes with normal boundary conditions, that is, $n_\mu(z=0) = n_\mu(z=L) = 0$, where L stands for the distance between the planes. To satisfy these constraints, we seek $n_\mu(\mathbf{r})$ in the form

$$n_\mu(\mathbf{r}) = \frac{1}{4\pi^2}\int d^2\mathbf{q}_\perp \frac{2}{L}\sum_{m=1}^\infty n_\mu(\mathbf{q})e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}\sin q_z z, \quad (9)$$

where $q_z = \frac{m\pi}{L}$, $\mathbf{q} = (\mathbf{q}_\perp, q_z)$, and $\mathbf{r} = (\mathbf{r}_\perp, z)$. To second order in small director distortions n_x and n_y the bulk free energy (1) transforms into

$$F_{\text{bulk}} = \frac{1}{4\pi^2 L}\int d^2\mathbf{q}_\perp \sum_{m=1}^\infty \{K_1|n_x(\mathbf{q})q_x + n_y(\mathbf{q})q_y|^2 + K_2|n_x(\mathbf{q})q_y - n_y(\mathbf{q})q_x|^2 + K_3 q_z^2[|n_x(\mathbf{q})|^2 + |n_y(\mathbf{q})|^2]\}. \quad (10)$$

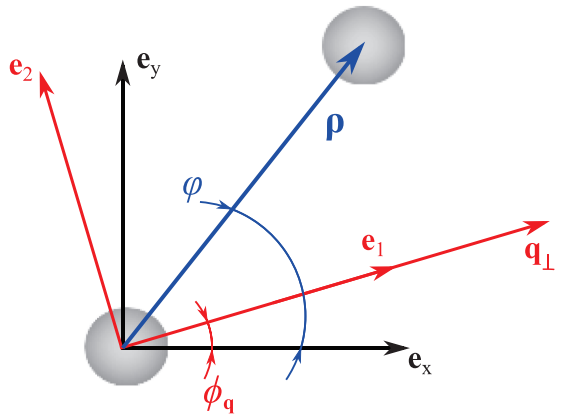

FIG. 1. (Color online) Coordinate system used in this study. For a given wave vector $\mathbf{q}$ the basis $(\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3)$ is rotated with respect to the fixed basis $(\mathbf{e}_x,\mathbf{e}_y,\mathbf{e}_z)$ by the angle $\phi_\mathbf{q}$ around the axis $\mathbf{e}_z\|\mathbf{e}_3$, which is normal to the plane of the drawing, in such a way that $\mathbf{e}_1\|\mathbf{q}_\perp$.

It follows from (10) that the distortion profiles $n_x(\mathbf{r})$ and $n_y(\mathbf{r})$, which minimize the total energy $F_{\rm tot}=F_{\rm bulk}+F_{\rm source}$, obey a system of two coupled equations $\frac{\delta}{\delta n_\mu}F_{\rm tot}=0$. A simple way to decouple them is to introduce an orthogonal basis $(\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3)$ associated with the wave vectors $\mathbf{q}$. For a given $\mathbf{q}$ the basis $(\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3)$ is rotated with respect to the $(\mathbf{e}_x,\mathbf{e}_y,\mathbf{e}_z)$ by the angle $\phi_\mathbf{q}=\arccos(\mathbf{e}_x\cdot\mathbf{q}_\perp/q_\perp)$ around the axis $\mathbf{e}_z\|\mathbf{e}_3$. From the sketch in Fig. 1 we can easily find that

$$\begin{aligned} n_x &= n_1\cos\phi_\mathbf{q}-n_2\sin\phi_\mathbf{q},\\ n_y &= n_1\sin\phi_\mathbf{q}+n_2\cos\phi_\mathbf{q}. \end{aligned} \tag{11}$$

Then $F_{\rm bulk}$ reduces to

$$F_{\rm bulk}=\frac{1}{4\pi^2L}\int d^2\mathbf{q}_\perp\sum_{m=1}^{\infty}\{|n_1(\mathbf{q})|^2\big(K_1q_\perp^2+K_3q_z^2\big)+|n_2(\mathbf{q})|^2\big(K_2q_\perp^2+K_3q_z^2\big)\}. \tag{12}$$

The effective part of the energy given by (8) can be expressed in terms of n_1 and n_2 as well,

$$F_{\rm source}=-\frac{2K}{\pi L}\int d^2\mathbf{q}_\perp\sum_{m=1}^{\infty}\sum_{i=1}^{N}\{\widehat{N}_x^i[n_1\cos\phi_\mathbf{q}-n_2\sin\phi_\mathbf{q}]+\widehat{N}_y^i[n_1\sin\phi_\mathbf{q}+n_2\cos\phi_\mathbf{q}]\}\exp[i\mathbf{q}_\perp\cdot\mathbf{r}_\perp^i]\sin q_zz^i. \tag{13}$$

For the sake of compactness we introduced the operators $\widehat{N}_\mu=q_\mu+p_\mu^\alpha\partial_\alpha+Q_\mu^{\alpha\beta}\partial_\alpha\partial_\beta$. The superscript i denotes that $\widehat{N}_\mu^i$ is constructed from the multipole moments of the ith particle and acts on its coordinates $\mathbf{r}_\perp^i$ and z^i.

Now $F_{\rm tot}$ is the sum of $F_{\rm bulk}$ and $F_{\rm source}$ given by (12) and (13), respectively. The Euler-Lagrange equations for $n_1(\mathbf{q})$ and $n_2(\mathbf{q})$ are independent and their solutions are easy to find. In particular,

$$n_1(\mathbf{q})=\frac{4\pi K}{K_1q_\perp^2+K_3q_z^2}\sum_{i=1}^{N}\{\widehat{N}_x^i\cos\phi_\mathbf{q}+\widehat{N}_y^i\sin\phi_\mathbf{q}\}\times\exp[-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp^i]\sin q_zz^i \tag{14}$$

and

$$n_2(\mathbf{q})=\frac{4\pi K}{K_2q_\perp^2+K_3q_z^2}\sum_{i=1}^{N}\{-\widehat{N}_x^i\sin\phi_\mathbf{q}+\widehat{N}_y^i\cos\phi_\mathbf{q}\}\exp[-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp^i]\sin q_zz^i. \tag{15}$$

Substituting these solutions into the total energy, one sees that $F_{\rm tot}=\sum_{i>j}U^{ij}+\sum_iU^i$, where U^i is the self-energy of the ith particle and U^{ij} is the energy of the interaction between ith and jth particles,

$$\begin{aligned} U^{ij}=-\frac{8K}{L}\int d^2\mathbf{q}_\perp\sum_{m=1}^{\infty}\Bigg\{&\widehat{N}_x^i\widehat{N}_x^j\left[\frac{\cos^2\phi_\mathbf{q}}{\kappa_1q_\perp^2+\kappa_3q_z^2}+\frac{\sin^2\phi_\mathbf{q}}{\kappa_2q_\perp^2+\kappa_3q_z^2}\right]+\widehat{N}_y^i\widehat{N}_y^j\left[\frac{\sin^2\phi_\mathbf{q}}{\kappa_1q_\perp^2+\kappa_3q_z^2}+\frac{\cos^2\phi_\mathbf{q}}{\kappa_2q_\perp^2+\kappa_3q_z^2}\right]\\ &+\big[\widehat{N}_x^i\widehat{N}_y^j+\widehat{N}_y^i\widehat{N}_x^j\big]\left[\frac{\sin\phi_\mathbf{q}\cos\phi_\mathbf{q}}{\kappa_1q_\perp^2+\kappa_3q_z^2}-\frac{\sin\phi_\mathbf{q}\cos\phi_\mathbf{q}}{\kappa_2q_\perp^2+\kappa_3q_z^2}\right]\Bigg\}\exp[-i\mathbf{q}_\perp\cdot(\mathbf{r}_\perp^i-\mathbf{r}_\perp^j)]\sin q_zz^i\sin q_zz^j. \end{aligned} \tag{16}$$

Hereafter $\kappa_s=K_s/K$, $s=1,2,3$. The integration is quite straightforward and yields

$$\begin{aligned} U^{ij}=-\frac{16\pi K}{L}\sum_{m=1}^{\infty}\Bigg\{&\widehat{N}_x^i\widehat{N}_x^j\left[\frac{1}{\kappa_1}\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right)\cos^2\varphi+\frac{1}{\kappa_1}\mathbb{K}_1\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right)\frac{\sqrt{\kappa_1/\kappa_3}}{\rho_m}\cos2\varphi\right.\\ &\left.+\frac{1}{\kappa_2}\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right)\sin^2\varphi-\frac{1}{\kappa_2}\mathbb{K}_1\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right)\frac{\sqrt{\kappa_2/\kappa_3}}{\rho_m}\cos2\varphi\right]\\ &+\widehat{N}_y^i\widehat{N}_y^j\left[\frac{1}{\kappa_1}\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right)\sin^2\varphi-\frac{1}{\kappa_1}\mathbb{K}_1\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right)\frac{\sqrt{\kappa_1/\kappa_3}}{\rho_m}\cos2\varphi\right.\\ &\left.+\frac{1}{\kappa_2}\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right)\cos^2\varphi+\frac{1}{\kappa_2}\mathbb{K}_1\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right)\frac{\sqrt{\kappa_2/\kappa_3}}{\rho_m}\cos2\varphi\right]\\ &+\big[\widehat{N}_x^i\widehat{N}_y^j+\widehat{N}_x^j\widehat{N}_y^i\big]\left[\frac{1}{\kappa_1}\mathbb{K}_2\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right)-\frac{1}{\kappa_2}\mathbb{K}_2\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right)\right]\sin\varphi\cos\varphi\Bigg\}\sin\frac{m\pi z^i}{L}\sin\frac{m\pi z^j}{L}, \end{aligned} \tag{17}$$

where φ denotes the angle between $\boldsymbol{\rho} = \mathbf{r}_\perp^i - \mathbf{r}_\perp^j$ and $\mathbf{e}_x$, $\mathbb{K}_n(x)$ is the modified Bessel function of the second kind, and $\rho_m = m\pi\rho/L$. This general expression describes the interaction between arbitrary colloidal particles suspended in a homeotropic nematic cell with different elastic constants. If we replace q_μ, p_μ^α, and $Q_\mu^{\alpha\beta}$ in (17) by $-\alpha_{z\mu}/4\pi K$, $-\beta_{z\mu\alpha}/4\pi K$, and $-\gamma_{z\mu\alpha\beta}/4\pi K$, respectively, then at short distances Eq. (17) will transform into that found in [26] for an infinite nematic medium

$$
\begin{aligned}
U^{ij,0} = -4\pi K \Bigg\{ & \widehat{N}_x^i \widehat{N}_x^j \Bigg[\frac{1}{\sqrt{\kappa_1}} \frac{\cos^2\varphi}{\sqrt{\kappa_3\rho^2+\kappa_1 l^2}} - \frac{\cos 2\varphi}{\sqrt{\kappa_1}} \frac{\sqrt{\kappa_3\rho^2+\kappa_1 l^2}-\sqrt{\kappa_1 l^2}}{\kappa_3\rho^2} \\
& + \frac{1}{\sqrt{\kappa_2}} \frac{\sin^2\varphi}{\sqrt{\kappa_3\rho^2+\kappa_2 l^2}} + \frac{\cos 2\varphi}{\sqrt{\kappa_2}} \frac{\sqrt{\kappa_3\rho^2+\kappa_2 l^2}-\sqrt{\kappa_2 l^2}}{\kappa_3\rho^2} \Bigg] \\
& + \widehat{N}_y^i \widehat{N}_y^j \Bigg[\frac{1}{\sqrt{\kappa_1}} \frac{\sin^2\varphi}{\sqrt{\kappa_3\rho^2+\kappa_1 l^2}} + \frac{\cos 2\varphi}{\sqrt{\kappa_1}} \frac{\sqrt{\kappa_3\rho^2+\kappa_1 l^2}-\sqrt{\kappa_1 l^2}}{\kappa_3\rho^2} + \frac{1}{\sqrt{\kappa_2}} \frac{\cos^2\varphi}{\sqrt{\kappa_3\rho^2+\kappa_2 l^2}} - \frac{\cos 2\varphi}{\sqrt{\kappa_2}} \frac{\sqrt{\kappa_3\rho^2+\kappa_2 l^2}-\sqrt{\kappa_2 l^2}}{\kappa_3\rho^2} \Bigg] \\
& + \left[\widehat{N}_x^i \widehat{N}_y^j + \widehat{N}_x^j \widehat{N}_y^i \right] \Bigg[\frac{1}{\sqrt{\kappa_1}} \frac{(\sqrt{\kappa_3\rho^2+\kappa_1 l^2}-\sqrt{\kappa_1 l^2})^2}{\kappa_3\rho^2\sqrt{\kappa_3\rho^2+\kappa_1 l^2}} - \frac{1}{\sqrt{\kappa_2}} \frac{(\sqrt{\kappa_3\rho^2+\kappa_2 l^2}-\sqrt{\kappa_2 l^2})^2}{\kappa_3\rho^2\sqrt{\kappa_3\rho^2+\kappa_2 l^2}} \Bigg] \sin\varphi\cos\varphi \Bigg\},
\end{aligned}
\tag{18}
$$

where $l = |z - z'|$. Within the one-constant approximation $K_1 = K_2 = K_3 = K$, Eq. (18) reduces to the well-known multipole interactions $\tilde{U}^{ij,0} = -4\pi K \widehat{N}_\mu^i \widehat{N}_\mu^j |\mathbf{r}_i - \mathbf{r}_j|^{-1}$ considered in [14,15,29].

Before proceeding to a detailed analysis of (17), suppose that we have only two particles in the cell. Then we can omit superscripts i and j and use primed and nonprimed quantities instead, i.e., $\widehat{N}_\mu^i \to \widehat{N}_\mu$, $\widehat{N}_\mu^j \to \widehat{N}'_\mu$, etc.

A. Monopole interaction

Colloidal particles without both the symmetry axis parallel to $\mathbf{n}_0$ and the symmetry plane normal to $\mathbf{n}_0$ possess monopole moments [28,29]. Assume for simplicity $q_y = q'_y = 0$, $q_x = q$, and $q'_x = q'$ (this means that the particles are in addition symmetric with respect to the xz plane). Suppose furthermore that $qq' < 0$. Below we will see that namely the case of the monopoles of opposite sign is of more interest. Then the energy of the corresponding monopole-monopole interaction in the bulk host U_{qq}^0 reads [see (18)]

$$
\begin{aligned}
U_{\mathrm{qq}}^0 = -2\pi K q q' \Bigg[& \frac{1}{\sqrt{\kappa_1}} \frac{1}{\sqrt{\kappa_3\rho^2+\kappa_1 l^2}} - \frac{\cos 2\varphi}{\sqrt{\kappa_1}} \frac{(\sqrt{\kappa_3\rho^2+\kappa_1 l^2}-\sqrt{\kappa_1 l^2})^2}{\kappa_3\rho^2\sqrt{\kappa_3\rho^2+\kappa_1 l^2}} \\
& + \frac{1}{\sqrt{\kappa_2}} \frac{1}{\sqrt{\kappa_3\rho^2+\kappa_2 l^2}} + \frac{\cos 2\varphi}{\sqrt{\kappa_2}} \frac{(\sqrt{\kappa_3\rho^2+\kappa_2 l^2}-\sqrt{\kappa_2 l^2})^2}{\kappa_3\rho^2\sqrt{\kappa_3\rho^2+\kappa_2 l^2}} \Bigg].
\end{aligned}
\tag{19}
$$

Within the one-constant approximation ($\kappa_s = 1$) Eq. (19) yields a Coulomb-like interaction between elastic monopoles $\tilde{U}_{\mathrm{qq}}^0 = -4\pi K \tilde{q}\tilde{q}'/r$, where $r = \sqrt{\rho^2 + l^2} = |\mathbf{r} - \mathbf{r}'|$.

When the host medium is confined to the cell, the energy of the monopole-monopole interaction takes the form

$$
\begin{aligned}
U_{\mathrm{qq}} = -\frac{16\pi K q q'}{L} \sum_{m=1}^{\infty} \Bigg[& \frac{1}{\kappa_1} \mathbb{K}_0\!\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right) \cos^2\varphi + \frac{1}{\kappa_1} \mathbb{K}_1\!\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right) \frac{\sqrt{\kappa_1/\kappa_3}}{\rho_m} \cos 2\varphi \\
& + \frac{1}{\kappa_2} \mathbb{K}_0\!\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right) \sin^2\varphi - \frac{1}{\kappa_2} \mathbb{K}_1\!\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right) \frac{\sqrt{\kappa_2/\kappa_3}}{\rho_m} \cos 2\varphi \Bigg] \sin\frac{m\pi z}{L} \sin\frac{m\pi z'}{L},
\end{aligned}
\tag{20}
$$

which reduces to

$$
\tilde{U}_{\mathrm{qq}} = -\frac{16\pi K \tilde{q}\tilde{q}'}{L} \sum_{m=1}^{\infty} \sin\frac{m\pi z}{L} \sin\frac{m\pi z'}{L} \mathbb{K}_0(\rho_m) \tag{21}
$$

in the one-constant limit ($\kappa_s = 1$). Comparing U_{qq} and $\tilde{U}_{\mathrm{qq}}$ we see a substantial difference. While the latter is isotropic throughout the planes $z = z'$, the former is not. Indeed, at short distances ($\rho \ll \sqrt{\kappa_{1,2}/\kappa_3} L$) between the particles located in the middle of the cell ($z = z' = L/2$) the interaction (20), as well as (21), is repulsive $U_{\mathrm{qq}} = -\frac{4\pi K q q'}{\rho\sqrt{\kappa_3}} \left(\frac{\cos^2\varphi}{\sqrt{\kappa_2}} + \frac{\sin^2\varphi}{\sqrt{\kappa_1}} \right) > 0$ [see (19) with $l = 0$], but its isolines are not circles Moreover, when the distance increases two zones of attraction appear [see Fig. 2(a)]. They are localized either along the x axis (if $\kappa_1 < \kappa_2$) or along the y (if $\kappa_1 > \kappa_2$). In usual nematics, e.g., 4-pentyl-4′-cyanobiphenyl (5CB), $\kappa_1/\kappa_2 \approx 1.5$ and the attraction is strongly suppressed by the cell walls (so-called confinement effect [22]). However, the greater the difference between κ_1 and κ_2, the closer to the particle the attraction zones are and the larger κ_1/κ_3 or κ_2/κ_3, the slower the interaction decays with ρ. Say for definiteness $K_1 = 3.1$ pN, $K_2 = 0.31$ pN, and $K_3 = 0.88$ pN (these values were reported in [25] for 4-chloro-1,3-phenylene bis 4-[4′-(9-decenyloxy)benzoyloxy] benzoate (C1Pbis10BB) in a nematic phase). Under these

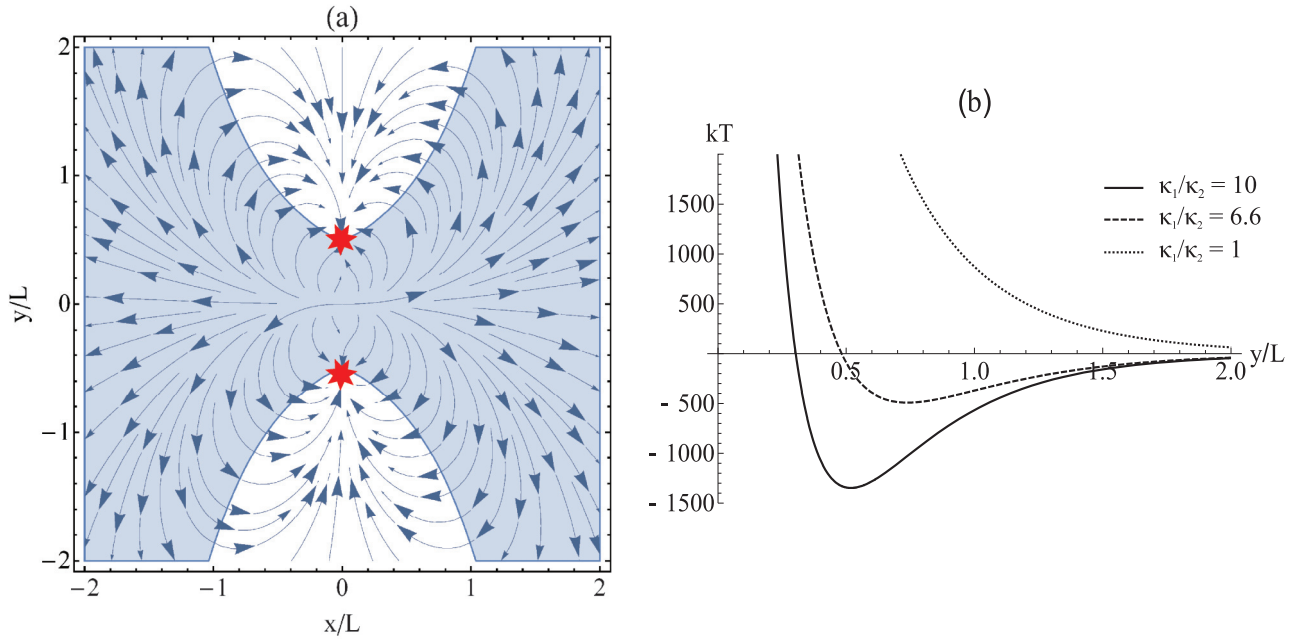

FIG. 2. (Color online) Interaction between two monopoles q_x and q'_x of opposite sign. The particles are placed in the middle of the cell ($z = z' = L/2$). (a) Map of the interaction. Arrow lines indicate the local direction of the force $\mathbf{F} = -\nabla U_{\rm qq}$. The shaded region is a repulsion zone $\frac{\partial U_{\rm qq}}{\partial \rho} < 0$. The stars indicate minima of the energy. Here $\kappa_1/\kappa_3 = 3.5$ and $\kappa_2/\kappa_3 = 0.35$ [25]. (b) Energy of the interaction along the y axis ($\varphi = \pi/2$). Here $q_x = q'_x = d$, $d = 3$ μm, $L = 12$ μm, $K_3 = 0.88$ pN, and $K = 1.4$ pN, the average elastic constant of C1Pbis10BB [25].

conditions, the interaction between the monopoles of opposite sign is attractive along the y axis when $\rho \gtrsim 0.5L$. In order to estimate the energy in that region, let us assume by analogy with [14] that $q = q' \approx d$, where $d = 3$ μm is the particles size, and take $L = 12$ μm and $K = (K_1 + K_2 + K_3)/3$. This rough estimate shows that the attraction is quite strong; its energy is of the order of 1000 kT [see solid line in Fig. 2(b)]. Thus, it might result in the formation of linear chains with large separation between the particles.

B. Dipole interaction

Consider now the interaction between colloidal inclusions with axially symmetric director configurations in their vicinity. Widely used representatives of such colloids are spherical particles with homeotropic anchoring. When the radius of the particle a is large enough, a topological defect called a hyperbolic hedgehog appears in its vicinity [1]. Such a configuration has the symmetry of a dipole aligned along $\mathbf{n}_0$. It is characterized by equal elastic moments $p_x^x = p_y^y = p$. Consequently, $\widehat{N}_\mu = p\partial_\mu$, $\widehat{N}'_\mu = p'\partial'_\mu$, and the energy of the interaction in the bulk nematic can be written as [see (18)]

$$U^0_{\rm pp} = \frac{4\pi K p p'}{r^3}\,\frac{\kappa_3}{\sqrt{\kappa_1}}\,\frac{\kappa_3\sin^2\theta - 2\kappa_1\cos^2\theta}{(\kappa_3\sin^2\theta + \kappa_1\cos^2\theta)^{5/2}}, \qquad (22)$$

where θ is the angle between $\mathbf{r} = \mathbf{r} - \mathbf{r}'$ and $\mathbf{n}_0$. Assuming $\kappa_s = 1$, one can readily transform $U^0_{\rm pp}$ to $\tilde{U}^0_{\rm pp} = 4\pi K\tilde{p}\tilde{p}'(1 - 3\cos^2\theta)/r^3$, which is well known from [14]. It follows from (22) that the energies of the interaction along the director ($\theta = 0$) and in the perpendicular direction ($\theta = \pi/2$) depend differently on the bend constant. Indeed, $U^0_{\rm pp}(\theta = 0) = -8\pi pp'(K/K_1)^2|z - z'|^{-3}K_3$, while $U^0_{\rm pp}(\theta = \pi/2) = 4\pi pp'K^2K_1^{-1/2}\rho^{-3}K_3^{-1/2}$. Suppose that K_3 is much larger than K_1. This is the case near a nematic–smectic-A phase transition, for instance. Under these circumstances, bend (and twist) deformations are energetically unfavorable and the elasticity of the liquid crystal is defined primarily by the splay constant. That is, $K \approx K_1$ near the transition point. This means in particular that we may expect a strengthening of the interaction along $\mathbf{n}_0$ and weakening in the perpendicular directions upon approaching a nematic–smectic-A phase transition. It is interesting to note that such a reduction of the interaction potential to one dimension above the transition point is qualitatively consistent with its behavior below $T_{\text{N-Sm}}$, in a smectic-A phase. References [30,31] showed that in a lamellar medium the interaction between two elastic dipoles $\bar{p}$ and $\bar{p}'$,

$$U^{\rm sm}_{\rm pp} = -\pi K_1\bar{p}\bar{p}'\left(\frac{32}{l^3} - \frac{16\rho^2}{\lambda l^4} + \frac{\rho^4}{\lambda^2 l^5}\right)e^{-\rho^2/4\lambda l}, \qquad (23)$$

where $\lambda = \sqrt{K_1/B}$ and B is the compression modulus, vanishes exponentially along the layers ($z \to z'$) and scales as $1/|z - z'|^3$ along the normal to the layers [32].

If the particles with hedgehogs are placed in a homeotropic cell, the energy (17) takes a surprisingly simple form

$$U_{\rm pp} = \frac{16\pi K p p'}{L^3}\,\frac{\kappa_3}{\kappa_1^2}\sum_{m=1}^{\infty} m^2\pi^2\sin\frac{m\pi z}{L} \times \sin\frac{m\pi z'}{L}\,\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right), \qquad (24)$$

which does not contain the twist constant K_2. This sounds reasonable since the director field around such an elastic dipole has no azimuthal component. Equation (24), as well as its

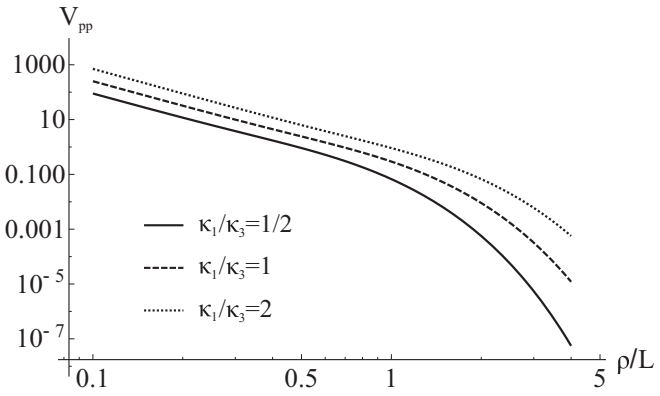

FIG. 3. Dimensionless energy of the dipole-dipole interaction $V_{\rm pp} = U_{\rm pp}L^3\kappa_1^2/16\pi K pp'\kappa_3$ as a function of the particle separation ρ. At short distances $\rho \lesssim L\sqrt{\kappa_1/\kappa_3}$ the energy $U_{\rm pp} \propto 1/\rho^3$. When $\rho \gtrsim L\sqrt{\kappa_1/\kappa_3}$ the interaction decays exponentially. Note that, although the curve $\kappa_1 > \kappa_3$ lies above the others, the corresponding energy of the interaction is not necessarily higher as it depends on the absolute values of the elastic constants.

one-constant counterpart [27]

$$\tilde{U}_{\rm pp} = \frac{16\pi K\tilde{p}\tilde{p}'}{L^3}\sum_{m=1}^{\infty} m^2\pi^2 \sin\frac{m\pi z}{L}\sin\frac{m\pi z'}{L}\mathbb{K}_0(\rho_m),$$

predicts a completely isotropic interaction between the particles located in the middle of the cell ($z = z' = L/2$). Namely, parallel dipoles repel and antiparallel dipoles attract. A common feature of the elastic interaction in nematic cells is its confinement discovered experimentally in [22]. Due to the rigid director orientation on the cell plates, the interaction decays exponentially when the interparticle distance ρ exceeds a threshold value $\rho_{\rm max} \sim L$. Formally, this is because of the asymptotic behavior of the modified Bessel functions $\mathbb{K}_n(x \gg 1) \propto e^{-x}/\sqrt{x}$. One sees from (24) that $\rho_{\rm max}$ depends on the ratio $\sqrt{K_1/K_3}$. In typical calamitic nematics $K_1 < K_3$ and the range of the interaction is about $\sqrt{K_1/K_3}$ times smaller than that predicted by the one-constant theory [27]. If, to the contrary, $K_1 > K_3$, the region of the interaction will be about $\sqrt{K_1/K_3}$ times larger (see Fig. 3).

It should also be remarked that in nematics with high K_1/K_3 and low K_2/K_3 a twist transition of the elastic dipole may occur [33,34]. Such a structural rearrangement probably alters the elastic moments and destroys the axial symmetry of the dipole-dipole interaction transforming it into the aforementioned monopole-monopole. However, this issue requires further investigation.

C. Quadrupole interaction

Far from a spherical particle accompanied by a Saturn ring or boojum topological defect the director field can be described by means of the quadrupole moments $Q_x^{xz} = Q_y^{yz} = Q$. Implying $\widehat{N}_\mu = Q\partial_z\partial_\mu$ and $\widehat{N}'_\mu = Q'\partial'_z\partial'_\mu$, one can find that

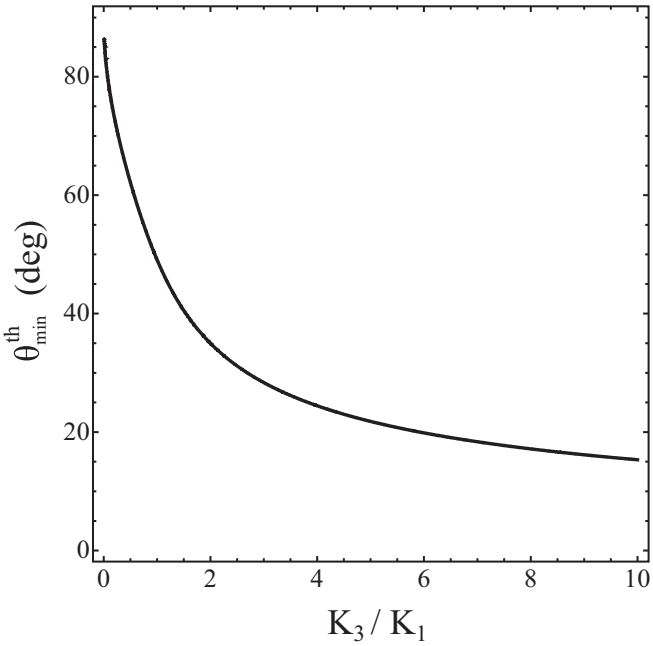

FIG. 4. Dependence of the angle $\theta_{\rm min}^{\rm th}$ that minimizes the energy of the quadrupole-quadrupole interaction (25) on the ratio K_3/K_1. In the limit of vanishing K_3/K_1 the angle $\theta_{\rm min}^{\rm th}$ approaches (but does not reach) 90°. Similarly, if $K_3/K_1 \to \infty$, then $\theta_{\rm min}^{\rm th}$ approaches (but does not reach) 0°.

in the bulk medium

$$U_{\rm QQ}^0 = \frac{4\pi K Q Q'}{r^5}\kappa_3\sqrt{\kappa_1} \times \frac{24\kappa_1^2\cos^4\theta + 9\kappa_3^2\sin^4\theta - 72\kappa_1\kappa_3\sin^2\theta\cos^2\theta}{(\kappa_3\sin^2\theta + \kappa_1\cos^2\theta)^{9/2}}. \quad (25)$$

In the one-constant limit ($\kappa_s = 1$), Eq. (25) reduces to the well-known $\tilde{U}_{\rm QQ}^0 = 4\pi K\tilde{Q}\tilde{Q}'(9 - 90\cos^2\theta + 105\cos^4\theta)/r^5$. The quadrupole-quadrupole interaction (25) is highly anisotropic. If $QQ' > 0$ it is repulsive along the directions $\theta = 0$ and $\theta = \pi/2$ and attractive within an intermediate range of the angles. Such a pattern of the interaction can lead to aggregation of quadrupole particles in chains directed at some angle $0 < \theta_{\rm min} < \pi/2$ to the bulk director $\mathbf{n}_0$. Figure 4 shows that the angle $\theta_{\rm min}^{\rm th}$ that minimizes $U_{\rm QQ}^0$ is a monotonically decreasing function of the ratio K_3/K_1. At the same time, the experimental value of $\theta_{\rm min}$ is about 30° practically regardless of K_3/K_1 [4]. This discrepancy is probably caused by the influence of higher-order elastic multipoles and by nonlinear deformations in the vicinity of the defects [35]. Restricting ourselves to the quadrupole-quadrupole interaction and consequently to a qualitative level only, we see that in the limit of large K_3/K_1 the equilibrium angle $\theta_{\rm min}^{\rm th}$ approaches (but does not equal) 0°. That is, near a nematic–smectic-A phase transition the chains become almost parallel to $\mathbf{n}_0$. Recently, such a decrease of the angle between the rubbing direction and the line joining the centers of two boojum colloids from 33° to 22° across the phase transition was reported in [36].

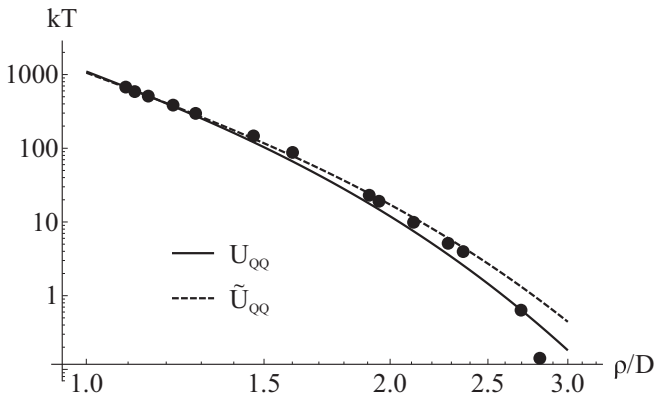

FIG. 5. Quadrupole interaction between two beads with tangential anchoring in the cell filled with 5CB. Here $Q = Q' = -0.4a^3$ and $\tilde{Q} = \tilde{Q}' = -0.31a^3$, with $a = D/2 = 2.2\ \mu$m, $L = 8\ \mu$m, $K_1 = 6.2$ pN, $K_2 = 3.7$ pN, $K_3 = 8.3$ pN [38], and $K = 6.1$ pN. The points depict experimental data from [22].

In the nematic cell the interaction between two quadrupoles is given by

$$U_{\mathrm{QQ}} = \frac{16\pi K Q Q'}{L^5}\frac{\kappa_3}{\kappa_1^2}\sum_{m=1}^{\infty} m^4\pi^4 \cos\frac{m\pi z}{L} \times \cos\frac{m\pi z'}{L}\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_1/\kappa_3}}\right). \quad (26)$$

Naturally, since (26) is quite similar to (24), properties of the quadrupole interaction are similar to those of U_{pp} as well. Indeed, it is isotropic for the particles located in the middle of the cell and scales at short distances as $1/\rho^5$. At $\rho \gtrsim L\sqrt{K_1/K_3}$ the interaction is exponentially decaying.

Now we try to examine how the anisotropy of the elastic constants affects the values of the multipole moments. The energy of the interaction between two beads with tangential anchoring in a homeotropic cell filled with 5CB liquid crystal was measured in [22]. The theory we present involves two parameters: the effective elastic constant K and the value of the quadrupole moment $Q = Q'$. Apparently, both these parameters cannot be unambiguously found from an experiment on the energetics of the interaction because $U_{\mathrm{QQ}} \propto K^2 Q Q'$. In order to be clear, we simply define $K = (K_1 + K_2 + K_3)/3$ and $Q = Q' = -\beta a^3$, where a is the particle radius and β is unknown and positive. Then fitting the one-constant curve

$$U_{\mathrm{QQ}} = \frac{16\pi K \tilde{Q}\tilde{Q}'}{L^5}\sum_{m=1}^{\infty} m^4\pi^4 \cos\frac{m\pi z}{L}\cos\frac{m\pi z'}{L}\mathbb{K}_0(\rho_m), \quad (27)$$

with these parameters to the experimental data from [22], one can find that $\tilde{\beta} = 0.31$ (see Fig. 5 for details). The same procedure with the energy (26) results in $\beta = 0.4$. Although β and $\tilde{\beta}$ differ considerably, they both are close to the value 0.36 found in [37] from another experiment.

We have to admit that the fitting of U_{QQ} is somewhat sensitive to the ratio K_1/K_3, which varies with temperature from 0.7 to 0.8 [38]. We employed $K_1/K_3 = 0.75$ as it provides good agreement between all the results. Under that condition, both curves in Fig. 5 describe equally well the interaction at the distances smaller than the cell thickness. At the same time, U_{QQ} gives a slightly more accurate representation of the tail of the potential.

As we have already mentioned, in the bulk nematic host the interaction between two quadrupoles (25) is repulsive along the director. However, recently it was observed that in nematics with $K_1 > 2K_2$, spherical particles with degenerate planar anchoring attract along $\mathbf{n}_0$ [39]. A numerical investigation performed by the same authors revealed that in such a medium the director at the surface of the particle deviates from the meridional directions. The azimuthal component of $\mathbf{n}(\mathbf{s})$ gives rise to a so-called chiral dipole [40]. In terms of the elastic multipoles, this helical configuration can be described by $p_x^y = -p_y^x = -p_c$, where p_c is positive for right-handed helicity and negative otherwise [28]. Then $\widehat{N}_x = -p_c\partial_y$, $\widehat{N}_y = p_c\partial_x$, and Eq. (18) yields

$$U^0_{\mathrm{p_c p_c}} = \frac{4\pi K p_c p_c'}{r^5}\frac{\kappa_3}{\sqrt{\kappa_2}}\frac{\kappa_3\sin^2\theta - 2\kappa_2\cos^2\theta}{(\kappa_3\sin^2\theta + \kappa_2\cos^2\theta)^{5/2}}. \quad (28)$$

Hence, the chiral dipoles of the same handedness attract along the director ($\theta = 0$), while those of opposite handedness repel, in complete agreement with the observations [39].

Interestingly, the interaction between chiral inclusions in the bulk host as well as in the cell

$$U_{\mathrm{p_c p_c}} = \frac{16\pi K p_c p_c'}{L^3}\frac{\kappa_3}{\kappa_2^2}\sum_{m=1}^{\infty} m^2\pi^2 \sin\frac{m\pi z}{L} \times \sin\frac{m\pi z'}{L}\mathbb{K}_0\left(\frac{\rho_m}{\sqrt{\kappa_2/\kappa_3}}\right) \quad (29)$$

depends on the twist constant K_2 and does not depend explicitly on the splay K_1. However, the opposite is true for the interaction of achiral dipoles considered above. The latter does not depend on K_2 but do depend on K_1. These features originate from the types of deformations that dominate in every configuration. Those are primarily twist-bend and splay-bend deformations in the cases of chiral and achiral dipoles, respectively.

IV. CONCLUSION

We obtained a general expression for the energy of the interaction between colloidal particles suspended in a homeotropic nematic cell with different Frank elastic constants. It is suitable for the particles of arbitrary size and shape. In general, our results confirm the validity of the one-constant approximation for calamitic nematic liquid crystals, in which the elastic constants are usually comparable. At the same time, the present study reveals some interesting features of the elastic colloidal interactions in liquid crystal hosts with more specific relations between the elastic constants.

For instance, we predict a decrease of the angle between a chain of quadrupole particles and the bulk director with increasing K_3/K_1. Recently, such a decrease of the angle between the rubbing direction and the line joining the centers of two boojum colloids from 33° to 22° across a nematic–smectic-A phase transition was reported in [36]. This effect is a manifestation of the suppression of the interaction along the

directions normal to $\mathbf{n}_0$ upon approaching the transition point. Such a reduction of the interaction to one dimension above the transition point is qualitatively consistent with its behavior in a smectic-A phase.

Despite the constant's values, the interactions in the cell are confined. That is, if the projection of the interparticle distance onto the plane of the cell exceeds a threshold value $\rho_{\max}$ (close to the cell thickness for calamitic nematics), the interaction will decay exponentially. Hence, there exists a finite region of the interaction. Its size is proportional to $\sqrt{K_1/K_3}$. This in particular means that in nematics with $K_1 \gg K_3$ the threshold $\rho_{\max}$ can increase up to a few cell thicknesses.

The one-constant approximation predicts a completely isotropic interaction between two asymmetric particles (elastic monopoles) located in the middle of the cell. They attract if their elastic monopole moments are of the same sign and repel otherwise. The anisotropy of the elastic constants breaks the symmetry of the interaction by inducing two zones of repulsion (attraction). In weakly elastically anisotropic nematics these zones are of no interest, as they lie beyond the region of the interaction. However, in the hosts, where either $K_1 \gg K_2, K_3$ or $K_2 \gg K_1, K_3$, they are within the region and under certain conditions might lead to the formation of thermodynamically stable linear chains with large separations between the particles.

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[32] Strictly speaking, there is a lack of clear definition of the multipole moments in a smectic-A liquid crystal. We just have to choose them so as to mimic the director field around a given colloidal particle. We use p_z and p_z' (in the notation of [30]) for axially symmetric particles with a broken horizontal (normal to $\mathbf{n}_0$) symmetry plane. Even if this choice is not correct, the main statement remains valid. The particles with any set of the moments will interact mainly along the director.
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