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# Interaction between Particles in a Nematic Liquid Crystal: Numerical Study Using the Landau-de Gennes Continuum Theory

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*We study numerically the interaction between particles in a nematic liquid crystal mediated by its elastic distortions with the aid of the Landau-de Gennes continuum theory. We consider the cases where two particles impose rigid normal anchoring on their surfaces and are accompanied by a hyperbolic hedgehog defect. As a function of the distance between the centers of the particles D, we evaluate the force f acting on the particles by integrating the stress tensor. The result is well described by a power law* $f \propto D^{-x}$. *When the "dipoles", composed of a particle and a hyperbolic hedgehog, are in parallel directions, the interaction is attractive and the exponent is* $x \simeq 4$, *consistent with the experimental observations together with the theoretical expectation of the dipole-dipole interaction. For antiparallel dipoles, repulsive interaction is observed and* $x \simeq 3.6$, *slightly stronger than the dipole-dipole interaction.*

**Keywords:** colloid; elastic deformation; interaction; Landau-de Gennes theory; nematic liquid crystal; stress tensor

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## 1. INTRODUCTION

Recently considerable attention has been paid to liquid crystal colloidal dispersions [1–8] as a novel class of composite materials that show structures and mechanical properties different from conventional colloidal systems with isotropic host fluids. Many of the fascinating properties of liquid crystal colloidal dispersions arise from the elastic distortions of the host liquid crystals due to the surface anchoring of immersed particles or droplets. Such elastic distortions can mediate a long-range interaction between particles, which yields various types of superstructures such as linear chains [2,5], anisotropic clusters [1], periodic lattices [7], cellular structures [4], and finger-print textures that mimic those of cholesteric liquid crystals [8]. Detailed investigations of the elastic-distortion-mediated interactions in a liquid crystal are therefore crucially important for the understanding of the mechanism underlying the formation of such superstructures in liquid crystal colloidal dispersions.

There have been several analytic studies that deal with the interaction between particles in a liquid crystal. In the case of a nematic liquid crystal as a host fluid, Ramaswamy *et al.* [9] and Ruhwandl and Terentjev [10] found that the interaction between spherical particles with weak surface anchoring is quadrupolar with the interaction potential proportional to $D^{-5}$, where $D$ is the inter-particle distance. Lubensky *et al*. [11] showed by a phenomenological argument that a particle accompanied by a topological defect called a hyperbolic hedgehog acts as a "dipole" whose interaction potential is proportional to $D^{-3}$. This dipole-dipole interaction is known to be responsible for the chain-like superstructures observed experimentally [2,5]. Lev and co-workers [12] stressed that the symmetry of the particle shape is one of the crucial factors determining the type of the interaction.

The validity of those analytic studies of the elastic-distortion-mediated interaction is, however, somewhat limited because of the intrinsic difficulties encountered in the analytic treatment of the host liquid crystals, including the non-linear nature of the elastic energy or the presence of topological defects. Most of the previous analytic studies implicitly or explicitly assumed that the inter-particle distance is much larger than the characteristic dimension of the particles, or that the elastic distortion of the host liquid crystal is weak enough to allow the description of the elastic energy in a bilinear form. Therefore numerical calculations will be absolutely necessary for a detailed investigation of the particle interaction in a liquid crystal, in particular when the inter-particle distance is not large as compared to the size of the particles or when topological defects are present in the host liquid crystal.

In our previous study [13], we investigated the interaction between two spherical particles accompanied by a hyperbolic hedgehog defect using the Landau-de Gennes continuum theory that describes the orientational order in terms of a second-rank tensor $Q_{ij}$. The use of $Q_{ij}$ allows one to deal with topological defects in the liquid crystal without introducing any singularities. For the description of the geometry of the system containing two spheres, we made use of bispherical coordinates [14–16] that proved to be useful in the implementation of the boundary conditions at the particle surfaces and at infinity. We showed in another study [17] that the same numerical procedure is applicable to the problem of capillary condensation of a nematic phase just above the nematic-isotropic transition point. We calculated the equilibrium orientation profiles for various inter-particle distances $D$ and evaluated the free energy $F$ of the host liquid crystal. The force acting on the particles $f$ is determined by the differentiation of the free energy $F$ with respect to $D$. In this contribution, we present another method of evaluating the force by calculating the surface integral of the hydrostatic stress tensor. One of the great advantages of evaluating the force using the stress tensor is that for a given inter-particle distance $D$, only the equilibrium orientation profile for that $D$ is required; on the other hand when one wants to calculate the force using the free energy $F$, orientation profiles for distances other than $D$ will be necessary, because one has to know the dependence of $F$ on $D$. In the present article we show results for the interactions of two spherical particles accompanied by a hyperbolic hedgehog defect, which are expressed quite well by a power law $f \propto D^{-x}$ with very small numerical errors as compared to our previous attempts of evaluating $f$ by differentiating the free energy.

## 2. MODEL

In our previous article [13] we have already presented in detail the numerical model for the description of the orientational order of a nematic liquid crystal around two particle and in this article we give only its brief outline.

In the Landau-de Gennes theory, a second-rank traceless tensor $Q_{ij}$ is employed for the description of the orientational order of a nematic liquid crystal. After some appropriate rescaling of the order parameter as presented in our previous article [13], the free energy of the host nematic liquid crystal in terms of $Q_{ij}$ is written as

$$F = \int_{\Omega} d\boldsymbol{r} \left[ \frac{1}{2}\tau \mathrm{Tr}\mathbf{Q}^2 - \frac{\sqrt{6}}{4}\mathrm{Tr}\mathbf{Q}^3 + \frac{1}{4}(\mathrm{Tr}\mathbf{Q}^2)^2 + \frac{1}{2}\xi_R^2 \partial_k Q_{ij} \partial_k Q_{ij} \right], \qquad (1)$$

where $\tau$ serves as the reduced temperature. Hereafter all the lengths are given in units of the particle radius $R_0$ and $\xi_R$ corresponds to the nematic coherence length at the nematic-isotropic phase transition. $\Omega$ is the region outside the two particles occupied by the liquid crystal. Here summations over repeated indices are implied, $\mathrm{Tr}\mathbf{Q}^2 = Q_{ij}Q_{ji}$ and $\mathrm{Tr}\mathbf{Q}^3 = Q_{ij}Q_{jk}Q_{ki}$. We notice that a simplified one-constant form of the elastic energy has been employed and that other terms allowed from symmetry such as $\partial_i Q_{ij}\partial_k Q_{kj}$ are not taken into account. In the numerical calculations presented below, we set $\tau = (3\sqrt{6} - 8)/12 < 0$, where a nematic state $Q_{ij} = Q_{\mathrm{bulk}}(n_i n_j - (1/3)\delta_{ij})$ with $Q_{\mathrm{bulk}} = 1$ minimizes the bulk energy ($\boldsymbol{n}$ is a unit vector corresponding to the director).

Next we describe the boundary conditions. At the surfaces of two particles we impose rigid normal anchoring and fix the order parameter as $Q_{ij} = Q_{\mathrm{bulk}}(\nu_i\nu_j - (1/3)\delta_{ij})$, where $\nu$ is a unit vector normal to the particle surfaces. At infinity, we assume uniform alignment along the $z$-axis and set $Q_{ij} = Q_{\mathrm{bulk}}(e^z_i e^z_j - (1/3)\delta_{ij}) = Q_{\mathrm{bulk}}(\delta_{iz}\delta_{jz} - (1/3)\delta_{ij})$, where $\boldsymbol{e}^z$ is a unit vector along the $z$-direction. This setup corresponds to placing particles with rigid normal surface anchoring in a uniformly aligned nematic liquid crystal. We also notice that since the order parameter at the surfaces is fixed, the particle surfaces do not contribute to the free energy of the system; therefore Eq. (1) constitutes the total free energy of the system.

For the geometry of the system, we deal with the cases where two spherical particles with equal radii ($R_0$) are placed in an infinite nematic medium and where the centers of the particles are located on the $z$-axis at $z = \pm D/2$ ($D$ is the distance between the centers of the particles). Bispherical coordinates [13–16] have proven quite useful and practical in describing the geometry of two non-intersecting spheres. The relation between the usual cylindrical coordinates $(\rho, z, \phi)$ and the bispherical coordinates $(\zeta, \mu, \varphi)$ is written as

$$\rho = \frac{a\sin\mu}{\cosh\zeta - \cos\mu}, \quad z = \frac{a\sin\zeta}{\cosh\zeta - \cos\mu}, \quad \phi = \varphi \tag{2}$$

where $a = \sqrt{(D/2)^2 - 1}$ in our setup. The surfaces of two spheres are simply represented by $\zeta = \pm\zeta_0$ with $\zeta = \cosh^{-1}(D/2)$. The region outside the spheres is mapped onto a rectangle in the $(\zeta, \mu)$ space given by $-\zeta_0 < \zeta < \zeta_0$ and $0 \le \mu \le \pi$, where $\zeta = \mu = 0$ corresponds to infinity.

For simplicity, we assume rotational symmetry about the $z$-axis, which renders our numerical problem an effectively two-dimensional one. Since the $z$ direction is parallel to the orientation of the nematic liquid crystal at infinity, this assumption leads to the situations where two particles are located along the orientation of the nematic at

infinity. The treatment of the order parameter under the assumption of rotational symmetry is described in detail in Refs. [13,18].

To obtain the equilibrium orientation profile for a given interparticle distance $D$, we prepare an initial condition and let it relax via a simple equation of motion: $\partial \boldsymbol{Q}_{ij}/\partial t = -\Gamma(\delta \boldsymbol{F}/\delta \boldsymbol{Q}_{ij} + \lambda \delta_{ij})$. This equation is referred to model A in the notation of Hohenberg and Halperin [19]. $\Gamma$ is a kinetic coefficient inversely proportional to the rotational viscosity and $\lambda$ is a Lagrange multiplier ensuring $\mathrm{Tr}\mathbf{Q} = 0$. How the initial conditions are prepared is presented in Ref. [13].

When the host nematic liquid crystal is in an equilibrium state, the hydrostatic stress tensor is written as $\sigma^e_{ij} = \sigma^d_{ij} - p\delta_{ij}$. Here $\sigma^d_{ij}$ and $p$ are the elastic stress and the hydrostatic pressure, respectively. The expressions of $\sigma^d_{ij}$ and $p$ consistent with the free energy (1) are

$$\sigma^d_{ij} = -\xi_R^2 \partial_i \boldsymbol{Q}_{kl} \partial_j \boldsymbol{Q}_{kl}, \tag{3}$$

$$p = -\left(\frac{1}{2}\tau \mathrm{Tr}\mathbf{Q}^2 - \frac{\sqrt{6}}{4}\mathrm{Tr}\mathbf{Q}^3 + \frac{1}{4}\left(\mathrm{Tr}\mathbf{Q}^2\right)^2 + \frac{1}{2}\xi_R^2 \partial_k \boldsymbol{Q}_{ij} \partial_k \boldsymbol{Q}_{ij} - f_{bulk}\right), \tag{4}$$

where $f_{bulk}$ is the bulk energy density of an undistorted nematic, and $p = 0$ at infinity. Equations (3) and (4) can be derived along the same way as the derivations of $\sigma^d_{ij}$ and $p$ in terms of $\boldsymbol{n}$ presented in Ref. [20].

In principle, the $j$-th component of the force $f_j$ acting on the body $V$ from the liquid crystal can be calculated as

$$f_j = \int_{\partial V} dS \nu_i \sigma^e_{ij}, \tag{5}$$

where $\nu$ is the unit normal pointing outwards from $V$. However, the evaluation of the integral on the particle surface did not give reliable results for the force acting on the particles, possibly due to the numerical errors in the evaluation of the first order derivatives and the ambiguity in the definition of $p$. Therefore we calculate the integral (5) at the plane $z = 0$ as illustrated in Figure 1. Then $f_j$ is the force acting on the liquid crystal in $z < 0$ from that in $z > 0$. In an equilibrium state, the force $f_j$ as calculated above is equal to the force acting on the left-hand-side sphere from the host liquid crystal. Since we consider the cases with rotational symmetry about the $z$-axis, it is obvious that the force has only the component along the $z$-direction, $f_z$. Here we give an explicit expression for $f_z$ in terms of $\mu$ which can be derived using Eq. (2):

$$f_z = 2\pi a^2 \int_0^\pi d\mu \frac{\sin\mu}{(1-\cos\mu)^2}\ \sigma^e_{zz}\big|_{z=0}, \tag{6}$$

where the definition of $a$ can be found after Eq. (2).

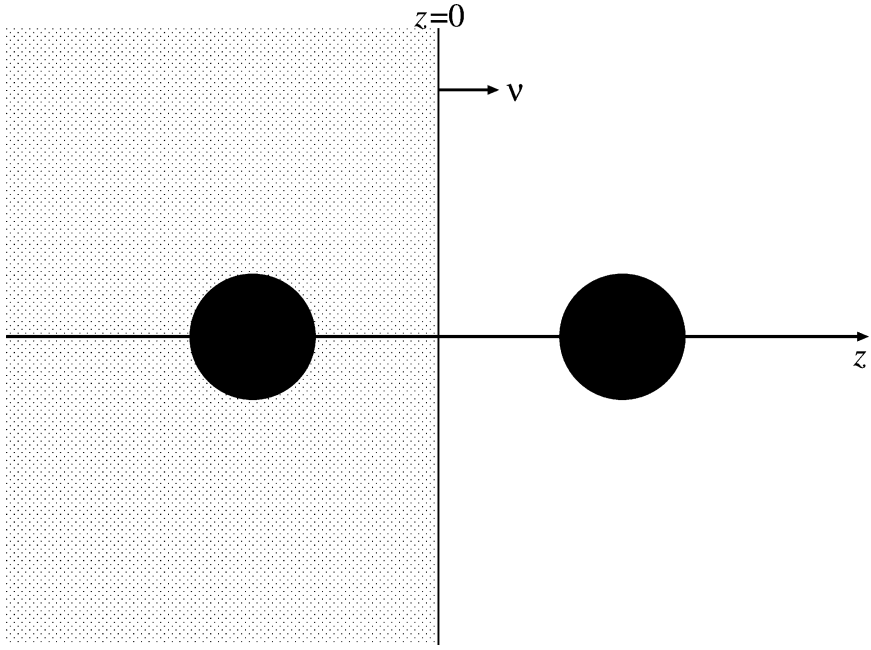


**FIGURE 1** The evaluation of the integral (5) at the plane $z = 0$ yields the force acting on the liquid crystal in $z < 0$ (shaded region) from that in $z > 0$ (white region).

We notice that the force acting on a particle can be calculated by any closed surface integral of the stress tensor enclosing the particle. We choose the $z = 0$ plane just for simplicity and clarity of the numerical calculation.

## 3. RESULTS

As noted in the Introduction, we shall consider the cases where two particles are accompanied by a hyperbolic hedgehog defect. Moreover, since we assume rotational symmetry about the $z$-axis where the centers of the two particles lie, we restrict ourselves to the situations where the hedgehog defects also lie on the $z$-axis. We show in Figure 2 some typical orientation profiles of a nematic liquid crystal around two particles by gray-scale plots of $Q_{zz}^2$. In Figure 2(a) the "dipoles" composed of a particle and a hedgehog defect take parallel orientations, while antiparallel dipoles can be seen in Figure 2(b). We note that for the nematic coherence length, we have chosen the same parameter $\xi_R = 5 \times 10^{-3}$ as in our previous study [13].

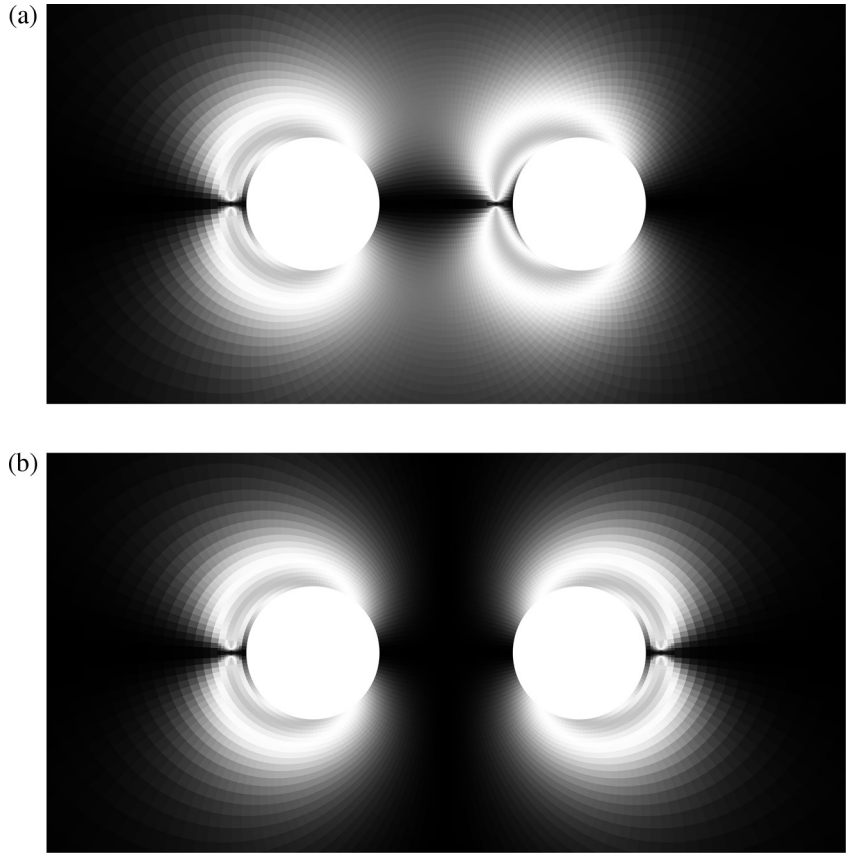


**FIGURE 2** The orientation profiles of a nematic liquid crystal shown by gray-scale plots of $Q_{zz}^2$. (a) "parallel-dipole" and (b) "anti-parallel-dipole" configurations. The inter-particle distance is set to $D = 4.0$. The $z$-axis is along the horizontal direction. In black regions, the liquid crystal is aligned along the $z$-direction, while $Q_{zz} = 0$ in white regions.

In Figure 3 we plot the force $f_z$ acting on the left particle as a function of the inter-particle distance $D$ for parallel "dipoles" (Fig. 2(a)). From Figure 3(a) we find that the particles feel an attractive force ($f > 0$) for large $D$, while the force is repulsive for small $D$. The short-range repulsion arises from the hyperbolic hedgehog defect situated between the two particles. Note that such a short-range repulsion cannot be dealt with in analytic arguments. The distance between the centers of the particles at rest ($f = 0$) is $D \simeq 2.42$, in good agreement with the value $D \simeq 2.46$ deduced from the minimum of the free energy

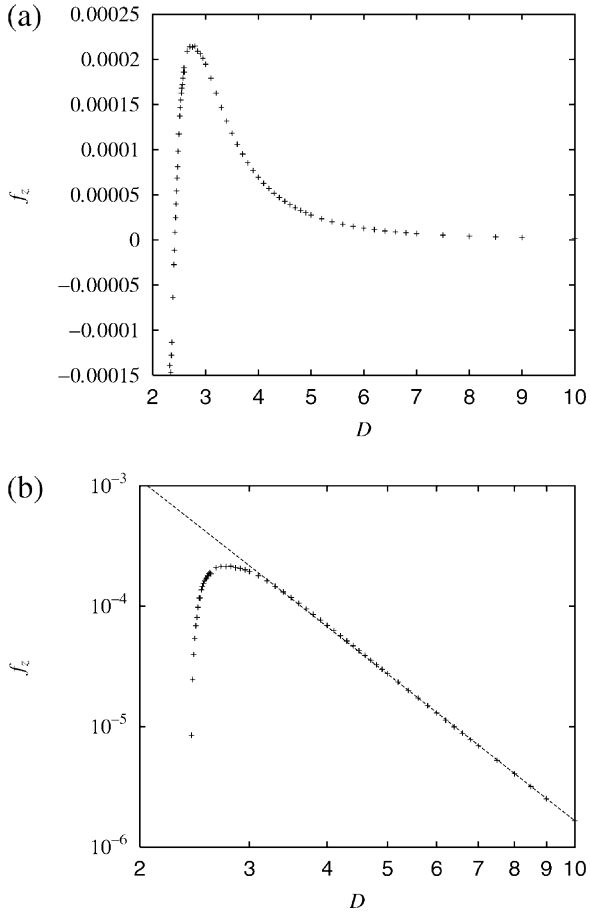


**FIGURE 3** The force $f_z$ acting on the left-hand-side particle as a function of the inter-particle distance $D$ in the case of parallel "dipoles". (a) normal plot, and (b) log-log plot for attractive forces ($f_z > 0$). The dashed line corresponds to $0.01856 \times D^{-4.05}$.

in our previous study [13]. To observe the long-range nature of the interaction force more precisely, we give a log-log plot of $f$ versus $D$ for $f > 0$ (attractive force) in Figure 3(b). The dashed line corresponds to $0.01856 \times D^{-4.05}$. This result is in agreement with the dipolar interaction expected theoretically [11] whose interaction force is proportional to $D^{-4}$. We note that this $D^{-4}$ interaction force has also been observed experimentally [21,22]. It may be worthwhile to notice that our previous calculation of the interaction force by differentiation

of the free energy with respect to $D$ contains some numerical errors (see Figure 5 of Ref. [13]), while the present data in Figure 3(b) fall quite well on a single line. This might indicate that the direct evaluation of the force by integrating the stress tensor is a more reliable method of investigating the interaction in a liquid crystal.

In Figure 4, we give a plot of the force as a function of $D$ for anti-parallel "dipoles" (Fig. 2(b)). The interaction is repulsive ($f_z < 0$) for all $D$ and in Figure 4 only a log-log plot of $-f_z$ versus $D$ is presented. From the theoretical arguments in Refs. [11,12], it is expected that the interaction force is again of dipolar form and proportional to $D^{-4}$. However, the dashed line in Figure 4 is $0.00543 \times D^{-3.63}$ and the exponent deviates from $-4$. Although the origin of this deviation is not clear, as was emphasized in the Introduction, the previous analytic studies of the interaction in a liquid crystal implicitly assume that the inter-particle distance is large enough. Therefore the present result may indicate the limitation of the validity of the previous analytic studies for intermediate inter-particle distances. We note that

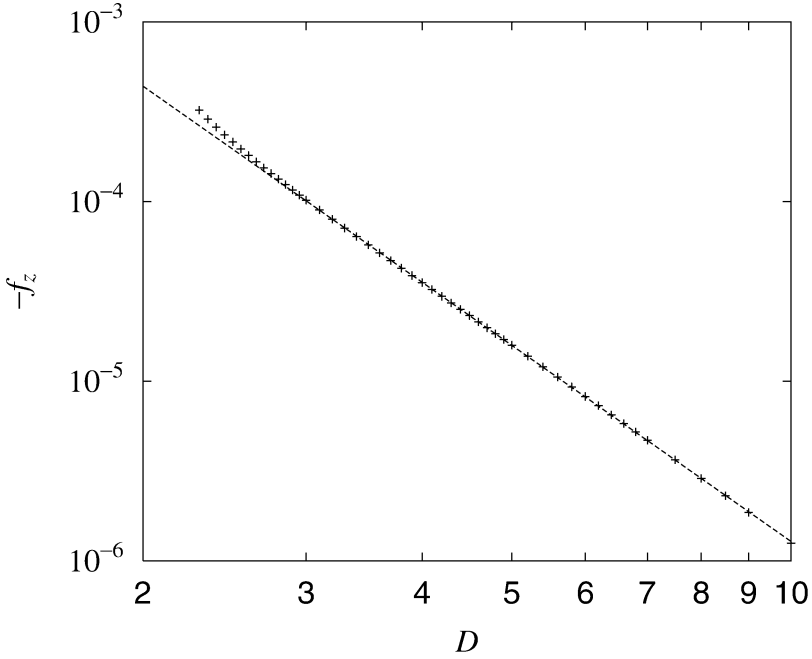


**FIGURE 4** Log-log plot of the force $-f_z$ versus the inter-particle distance $D$ in the case of anti-parallel dipoles. The dashed line corresponds to $0.00543 \times D^{-3.63}$.

the particles behave as radial hedgehogs because of the rigid normal anchoring at the particle surfaces. The bare repulsion between those two radial hedgehogs may render the interaction in the present case stronger than the dipole-dipole one. We also notice that the present result $f_z \propto D^{-3.6}$ is not consistent with our previous result [13] of the interaction force $f_z \propto D^{-3}$ calculated by differentiating the free energy of the host liquid crystal with respect to $D$. So far as we know, there have been no experimental data concerning the interaction between "anti-parallel dipoles" that can be compared with our numerical results, therefore we cannot determine at present which exponent, $-3.6$ or $-3$, is more reliable.

## 4. CONCLUSION

We investigated numerically the elastic-distortion-mediated interaction in a uniformly aligned nematic liquid crystal between two spherical particles. We considered the cases where rigid normal anchoring is imposed at the particle surfaces and each particle is accompanied by a hyperbolic hedgehog defect. We employed the Landau-de Gennes continuum theory in terms of a second-rank tensor order parameter $Q_{ij}$, which enables one to deal with topological defects without introducing any singularities. We also made use of bispherical coordinates, with which the boundary conditions at the particle surfaces and at infinity are naturally implemented.

In our previous study, the free energy $F$ of the host liquid crystal was evaluated for different inter-particle distances $D$ and we calculated the force $f$ acting on the particles by differentiating $F$ with respect to $D$. In this paper, the force was evaluated in a different way by integrating the hydrostatic stress tensor on a plane lying between the two particles. One of the advantages of evaluating $f$ using the stress tensor for a given $D$ is that only the equilibrium orientation profile for that $D$ is necessary, while profiles for other $D$'s will be necessary if one evaluates $f$ by differentiating $F$.

In the case of "dipoles", composed of a particle and a hedgehog defect, in parallel directions, the interaction is attractive for large $D$ and the attractive force behaves as $f \sim D^{-4}$, until the particles feel short-range repulsion due to a hedgehog defect situated between the two particles. The distance between particles at rest ($f = 0$) is $D \simeq 2.42$ in good agreement with $D \simeq 2.46$ deduced from the minimum in $F$ in our previous study. The long-range attraction is of dipolar type and consistent with previous experimental findings and analytic arguments. In the case of anti-parallel "dipoles", the interaction is repulsive for all $D$ and the interaction force obeys the power law

$f \sim D^{-3.6}$. This is not in agreement with the theoretical expectation of the dipolar interaction $f \sim D^{-4}$. This disagreement, whose origin is not clear so far, may indicate the limitation in the validity of analytical studies of the interaction in liquid crystals implicitly assuming that the inter-particle distance is large enough as compared to the particle size.

Finally we notice that the present numerical results of the interaction force evaluated by the stress tensor are described quite well by a power law with very small numerical errors. So far as we know, almost all of the previous similar numerical studies concerning the interaction between particles in a liquid crystal calculated only the free energy of the host liquid crystal and the present work is the first attempt to evaluate the interaction force by directly dealing with the stress tensor. Considering the small numerical errors in our present results, we believe that the evaluation of the stress tensor provides one of the reliable methods of investigating the interaction between particles in a liquid crystal. One of the necessary extensions will be to calculate the force by directly integrating the stress tensor at the particle surface, which was not successful in the present study. Such calculations will require a refined treatment of the stress tensor at a solid surface and will be helpful in investigating many-body problems in liquid crystal colloidal dispersions.



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