Colloid-wall interaction in a nematic liquid crystal: The mirror-image method of colloidal nematostatics

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The new area of nematic colloidal systems (or nematic emulsions) has been greatly guided by the fruitful analogy between the colloidal nematostatics and electrostatics. The elastic charge density representation of the colloidal nematostatics [V. M. Pergamenshchik and V. O. Uzunova, Eur. Phys. J. E **23**, 161 (2007); Phys. Rev. E **76**, 011707 (2007)] develops this analogy at the level of charge density and Coulomb interaction. It shows, however, that the colloidal nematostatics in three dimensions substantially differs from the electrostatics both in its mathematical structure and physical implications: the elastic charge and multipoles are dyads; similar charges attract while opposite charges repel each other, and so on. In this paper we consider the interaction between an elastic charge and elastic dipole with a nematic surface (wall) at which the director alignment is fixed. Using the mirror image method of electrostatics as a guiding idea, we develop the mirror image method in the nematostatics for arbitrary director tilt at the wall. A wall is shown to induce a repulsive $1/R^4$ force on the elastic dipole which, in general, is accompanied by its reorientation. External torque on the colloid induces an elastic charge therein and triggers switching to the $1/R^2$ repulsion. The dyadic nature of an elastic dipole is shown to be essential: a particle-wall interaction potential cannot be obtained in phenomenological theories with a single component dipole. In the introductory sections we discuss connection between the director-mediated interaction in two and three dimensions and the electrostatic interaction and consider different symmetries of elastic dipoles. Conservation of the torque components exerted upon colloids is shown to play the role of Gauss' theorem and determines the elastic charge dyad.

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I. INTRODUCTION

Particles immersed in a nematic liquid crystal (NLC) interact via the director field **n** which mediates the distortions induced thereby. The fact that this interaction is of a long range and reminds one the electrostatic interaction was brought to the broad attention by Brochar and de Gennes [1] almost 40 years ago and later by Lopatnikov and Namiot [2]. Over the last 15 years the study of this director-mediated interaction has developed into a rapidly growing branch of the physics of LCs, i.e., the physics of nematic colloidal systems or nematic emulsions [3] (to appreciate the latest scale of this area, see [4]). Depending on the colloids' size and nature, these systems can be very different. The recent development of the field has been inspired mostly by a great diversity of a few- and many-body ordering phenomena in microemulsions with micrometer and submicrometer size colloids [5–11]. However, there is also an important class of nematic nanoemulsions with molecular size colloids (dopants) and supramolecular size colloids. One example is a NLC doped with chiral molecules [12] that induce the well-known macroscopic cholesteric ordering [12]. Moreover, nowadays an intensive investigation of thermodynamics [13–16] and physical chemistry [17] of a NLC doped with a great variety of different solute molecules and micelles [18] is under way. A very special example comprises dye-doped NLC: under the action of light some of them demonstrate spectacular orientational Jánossy effect [19] which is possible only in anisotropic media. It has been recently shown that the supramolecular aggregates [20], interacting via the director field, and other supramolecular complexes, including dye molecules [21], can also play the role of colloids and facilitate the light-induced orientational effect in these "supra" nanoemulsions. One more class comprises nematic ferroemulsions [22]. Colloids of different origin (e.g., inclusions and defects, soft and rigid) and their interaction have also been studied in different smectic LC phases and their free standing films and membranes (see, e.g., [23–27] and review [28]).

This ordering diversity owes its existence to the long-range electrostaticlike interaction via the director field. The rapid development of the field of nematic emulsions has been greatly guided by this important similarity between the nematic emulsions and electrostatics. The director-mediated interaction possesses many properties characteristic of the interaction between electric dipoles and quadrupoles [2,3,5,6,29–37] which has been verified experimentally [38–44]. The deep analogy between the two very different areas of physics not only provides a useful theoretical tool to study the field of anisotropic colloidal systems, but has been perceived as the fact of a fundamental physical significance.

Recently we developed the electrostatic analogy in nematic emulsions at the level of charge and its density [45,46] and showed that the Coulomb interaction, which is fundamental to the electrostatics, has an important implication in the physics of nematic emulsions, too. The director-mediated Coulomblike interaction of two colloids was shown to be fully determined by external torques applied to the colloids. Let the unperturbed director at infinity be along the z axis and $\mathbf{\Gamma}_\perp$ be the vector of a transverse external torque exerted on a particle, Fig. 1: $\mathbf{\Gamma}_\perp$ has nonzero transverse x and y components and zero z component, i.e., $\mathbf{\Gamma}_\perp=(\Gamma_{\perp x},\Gamma_{\perp y},0)$.

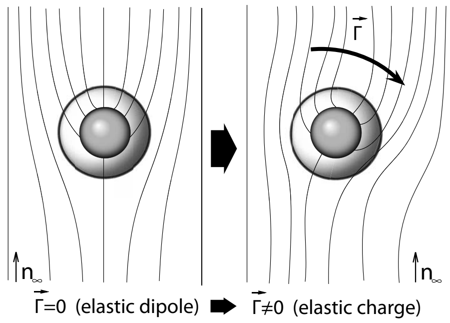

FIG. 1. General deformation source ("particle") in 3D. Inside the gray sphere the deformations can be very large (e.g., induced by point defects or strong anchoring of a real colloid). But outside the larger sphere the deformations are weak and linear which allows for the electrostatic analogy. The particle itself is of the dipolar type, but an external torque upon it can charge it, and it becomes an elastic charge. The arrow conventionally shows the director deformation by the torque Γ normal to the unperturbed director $\mathbf{n}_\infty$ (and to the figure plane).

This torque induces and fully determines a two component elastic charge of a particle which manifests itself both in the two transverse director components, scaling with the distance R as $1/R$, and in the interparticle interaction. Two particles under the action of external torques interact via a Coulomb-like $1/R$ potential where the scalar product $-(\mathbf{\Gamma}_\perp^{(1)}\cdot\mathbf{\Gamma}_\perp^{(2)})=-(\Gamma_{\perp,x}^{(1)}\Gamma_{\perp,x}^{(2)}+\Gamma_{\perp,y}^{(1)}\Gamma_{\perp,y}^{(2)})$ plays the role of the product of two electrostatic charges [45,46]. Because of the difference between the scalar electrostatics and vector nematostatics, the elastic analogs of the surface charge density, charge, and higher multipole moments consist of two tensors (dyads). The multipole moments are naturally expressed via the elastic charge density which is determined by the two transverse director components on the surface imposing the director deformations. The interaction of the axially symmetric sources, considered phenomenologically in [35], obtains as a particular case of the interaction of the two correspondent (diagonal, cf. see Sec. IV B) multipole dyads. The small parameter of the theory is the ratio a/R =(colloid size/distance between colloids). For small a/R the theory provides all the tools available in the electrostatics, e.g., for solving different boundary problems that can occur in the nematostatics of anisotropic emulsions. In this paper we apply the nematostatics developed in [45,46] to the interaction between an elastic charge (dyad) and elastic dipole (dyad) with a wall (surface bounding the NLC) with different director alignments, which is the elastic counterpart of the well-known electrostatic problem solved by the method of images (such experimental situation with a planar wall was recently explored in [44]). In the next section we briefly introduce the colloidal nematostatics of Refs. [45,46], discuss the connection between the director-mediated interaction in two and three dimensions and the electrostatic interaction, and show that the integral form of the torque balance plays the role similar to that of Gauss' theorem in the electrostatics. Then the theory is applied to the colloid-surface interaction.

The nature of a surface-colloid interaction is very different when the separation between a colloid surface and a plane-wall surface is microscopic and macroscopic. A microscopic scale is determined by the nematic coherence length which is just a few molecular lengths, i.e., a few nanometers. At a microscopic separation, two surfaces interact via the van der Waals, wetting, and Casimir forces [50,51]. In this case, a spherical colloid-wall attraction force has been measured by atomic force microscopy (AFM) [52–54]. As all the three forces vanish a few nanometers away from the surface, the aim of this study was not the colloid's behavior in itself, but the surface nematic and smectic ordering (wetting) at a nematic-isotropic transition.

The field of colloidal emulsions deals with macroscopic distances, and here we are interested in macroscopic colloid-wall separations of order of micrometers. In this case the interaction is mediated by macroscopic director distortions and the origin of the forces acting in the system is elastic. So far the problem of calculating a mechanical torque on a particle near a NLC surface and its energy has been specified by a particular particle's shape, anchoring, and orientation, and thus could be addressed only numerically [55,56]. The results of Refs. [45,46] considerably simplify the problem. The specific parameters of a particle now enter the problem via its multipole moment, and the problem reduces to the interaction between a wall and an elastic multipole. Here we show that this problem can be solved analytically in a universal form by means of an image method specific for the nematostatics. Using the mirror image method of electrostatics as a guiding idea, we develop the mirror image method in the nematostatics for arbitrary director tilt at the wall. A wall is shown to induce a repulsive tilt-dependent $1/R^4$ force on an elastic dipole and its reorientation to the minimum energy alignment. The external torque, however, induces the elastic charge in this colloid and triggers switching to the $1/R^2$ repulsion. The calculations demonstrate that the dyadic nature of an elastic dipole is essential. In particular, our exact result, that the repulsion from a homeotropic wall is 1.5 times weaker than that from a planar wall, cannot be obtained in phenomenological theories with a single component elastic dipole.

II. ELASTIC CHARGE DENSITY REPRESENTATION OF THE COLLOIDAL NEMATOSTATICS

A. Torque balance, Gauss' theorem, and elastic charge in three dimensions

The fundamental physical quantity of electric charge is purely phenomenological and must be *postulated* in the theory of elementary particles. In contrast, the nematostatics of the director field $\mathbf{n}$ allows for *introduction* of two different charges. Electrostatic potential is a scalar described by the linear Laplace (or Poisson) equation. It is the linearity that underlies the definition of the electric charge and its density as the source of electric field. At the same time, $\mathbf{n}$ is a vector field which reduces to a single variable, described by a linear equation (in the one constant approximation) only in two dimensions (2D). Owing to the linearity, the deformation source can be straightforwardly established: core of a point defect plays the role of a charge in 2D [12,47–49]. The integral, expressing the topological invariant, is independent of the integration contour. This property plays the role analogous to Gauss' theorem in electrostatics while the invariant itself plays the role of a conserved charge. As a result, the two-dimensional nematostatics is similar to the two-dimensional electrostatics with its logarithmic potential: disclinations with topological charges of the same sign repel one another and those with topological charges of opposite sign attract one another.

In three dimensions (3D), however, the analogy between topological defects and charge is completely lost. In 3D, the field $\mathbf{n}$ is described by highly nonlinear equations [12] so that point defects, though remain topological invariants, cannot be linearly connected with the distortions of $\mathbf{n}$ they induce [35,3]. Here the deformation source is the director distribution in a domain of the size $\sim a$ [45,46], Fig. 1. This director distribution can be induced by surface of a real particle, by topological defects with zero total topological charge [3,35], or by an external torque concentrated inside the domain [20]. We call such a compact deformation domain a particle. Consider a three-dimensional director field $\mathbf{n}(\mathbf{r})$ which far from the particles coincides with the uniform unperturbed director $\mathbf{n}_\infty=(0,0,1)$ parallel to the z axis. In the one-constant approximations assumed in this paper, the distortion free energy functional for the area V outside all the particles has the form

$$F\{\mathbf{n}\} = \frac{K}{2}\int (\boldsymbol{\nabla} n_t \cdot \boldsymbol{\nabla} n_t) d^3V. \tag{1}$$

A small perturbation of $\mathbf{n}_\infty$, induced by a particle at a distance $r \gg a$, satisfies the Laplace equation $\Delta n_t=0$ and has a transverse form $(n_x,n_y,0)$: the perturbation of the director's z component is negligible and n_z can be set equal to 1. From the Laplace equation the transverse components are obtained as

$$n_t(\mathbf{r}) = \frac{q_t}{r} + 3\frac{(\mathbf{d}_t \cdot \mathbf{r})}{r^3} + 5\frac{(\mathbf{Q}_t{:}\mathbf{r}{:}\mathbf{r})}{r^5} + \cdots, \tag{2}$$

where $t=x,y$. It is natural to identify the coefficients with the subscript t in this expansion with the tth component of elastic charge, elastic dipole, and elastic quadrupole, respectively. We seek the elastic analog of charge, following de Gennes' idea outlined in [12].

Due to its elasticity, a NLC transfers mechanical torque. A torque, exerted on a particle, induces director deformations in the ambient medium. Nonzero deformations result in a nontrivial torque balance in the director field. It is this torque balance that is described by the standard Euler-Lagrange equations minimizing the free energy functional. The torque balance implies two torques at any spatial point. Namely, the balance at a given point shows that an elastic torque is applied at both sides of any virtual surface passing through this point, and that these two torques have equal magnitudes and opposite signs [12]. Thus, a torque on a particle, located within a closed surface S, is transferred from inside S to outside S, the total torque being conserved. It means that certain torque density, integrated over an arbitrary closed surface, is equal to the total torque applied inside the surface irrespective of its form. In the static equilibrium, only a transverse external torque $\boldsymbol{\Gamma}_\perp=(\Gamma_x,\Gamma_y,0)$ can be exerted on a particle; a longitudinal torque Γ_z cannot be balanced as a rotation about the z axis does not change the elastic energy, $\Gamma_z=0$. In the one-constant approximation, the integral, which expresses the conservation and transfer of a torque $\boldsymbol{\Gamma}_\perp$, is of the form [12]

$$\Gamma_t = K\int_S \varepsilon_{\alpha t\rho}(r_\rho \partial_\beta n_\gamma \partial_\alpha n_\gamma + n_\rho \partial_\beta n_\alpha) dS_\beta, \tag{3}$$

where K is the elastic constant, $\varepsilon_{\alpha t\rho}$ is the absolute antisymmetric tensor, all indices but t run over x,y,z, and summation over the repeated indices is implied. The integral in the right-hand side of Eq. (3) does not depend on the choice of enclosing surface S, and the equality (3) reminds one Gauss' theorem with Γ_t in place of the electric charge. To further justify this connection one notices that integral (3) over a remote surface S vanishes for any term in the expansion (2) except for the first one. Substituting $n_t=q_t/r$ in Eq. (3) and integrating over a sphere gives $\Gamma_y=-4\pi K q_x$, $\Gamma_x=4\pi K q_y$, or

$$q_t = \frac{[\boldsymbol{\Gamma} \times \mathbf{n}_\infty]_t}{4\pi K}. \tag{4}$$

Thus, the tentative conclusion is that, in 3D, the role of Gauss' theorem and a charge is played, respectively, by the conservation of an elastic torque, transferred via the director field, and by two transverse components of an external torque exerted on a particle, Fig. 1. This is fully justified by calculating the Coulomb-like interaction in the elastic charge density representation [45,46].

B. Dyads of elastic mutipoles and their interaction via the director field

The results of Refs. [45,46] can be summarized as follows. The director field outside a single particle is fully determined by the director distribution on its surface. Consider a deformation source specified by the director field n_t on the surface of an enclosing sphere S with radius a. Elastic multipoles must be defined as to reproduce the solution of the Laplace equation outside S exactly in the form (2). This is achieved as follows. We introduce the quantity

$$\sigma_t(\mathbf{s}) = n_t(\mathbf{s})/a^2, \tag{5}$$

which plays the role of a two component surface elastic charge density at point $\mathbf{s}$ on the sphere S. Using natural analogy with the electrostatics, we define the two component elastic multipoles via the surface charge density as the following integrals over the sphere S enclosing the particle:

$$q_t = \frac{a}{4\pi}\int_S \sigma_t d^2s, \tag{6}$$

$$d_{t,\alpha} = \frac{a^2}{4\pi}\int_S \sigma_t \nu_\alpha d^2s, \tag{7}$$

$$Q_{t,\alpha\beta} = \frac{a^3}{8\pi}\int_S \sigma_t(3\nu_\alpha\nu_\beta - \delta_{\alpha\beta})d^2s, \tag{8}$$

where ν is the vector of unit outer normal to S. These definitions result exactly in the director expansion (2). Making use of these definitions, we obtained interaction potentials of two particles with similar multipoles in the leading order in a small parameter (a/R), where R is the modulus of the separation vector $\mathbf{R}$. Interaction of two "charged " particles with nonzero q_t is Coulomb-like,

$$U_{Coulomb} = -4\pi K\frac{q_t^{(1)}q_t^{(2)}}{R} = -\frac{(\mathbf{\Gamma}_\perp^{(1)}\cdot\mathbf{\Gamma}_\perp^{(2)})}{4\pi KR}, \tag{9}$$

where we used relation (4), and $\mathbf{\Gamma}_\perp^{(i)}$ is the transverse component of the torque exerted upon the ith particles. The above connection between the elastic charge and external torque is thus fully justified. Equation (9) shows that, depending on the sign of the scalar product $(\mathbf{\Gamma}_\perp^{(1)}\cdot\mathbf{\Gamma}_\perp^{(2)})=\Gamma_{\perp,t}^{(1)}\Gamma_{\perp,t}^{(2)}$, the elastic Coulomb interaction can be attractive or repulsive. In contrast to the electrostatics and two-dimensional nematostatics, the charges with the same sign attract and with different signs repel one another ("parallel torques" attract whereas "antiparallel torques" repel one another). Although the colloids must be anchored to the director, the Coulomb-like interaction does not directly depend on their specific shape and anchoring. Instead, the elastic charge is determined by the coefficients describing the torque exerted upon the colloid by a given type of external field. For instance, this can be the vector of permanent electric and magnetic dipole or electric and magnetic polarizability tensors of a given colloid.

If external torques are absent, the interaction energy is expressed solely in terms of particles' multipoles. The interaction between two "dipolar" particles is of the form

$$U_{dd} = -12\pi K\frac{(\mathbf{d}_t^{(1)}\cdot\mathbf{d}_t^{(2)}) - 3(\mathbf{d}_t^{(1)}\cdot\mathbf{u})(\mathbf{d}_t^{(2)}\cdot\mathbf{u})}{R^3}, \tag{10}$$

where $\mathbf{u}=\mathbf{R}/R$ is a unit vector along the separation direction [57]. Equations (2) and (5)–(10) (along with the quadrupole-quadrupole potential derived in [45,46]) suggest the following interpretation. q_t is the tth component of the elastic charge and $\sigma_t(\mathbf{s})$ is its surface density at point $\mathbf{s}$ on the sphere. The vector $\mathbf{d}_t$ and tensor $\mathbf{Q}_t$ are the tth dipole and quadrupole moments determined in the standard way by the surface charge density σ_t on the sphere. As σ_x and σ_y are separate sources, they determine not only the x and y director components outside the particle, Eq. (2), but also two independent components (dyad) for each multipole moment, i.e., q_x and q_y, $\mathbf{d}_x$ and $\mathbf{d}_y$, $\mathbf{Q}_x$ and $\mathbf{Q}_y$, and so on.

As the nematostatics is actually nonlinear, the above results of the linearized theory are fully justified under the assumption $|n_t|\ll 1$. In Refs. [45,46] we addressed the natural and practically important question as to how large can be $|n_t|$ that the nonlinear corrections can be safely neglected. In turns out that the restriction on $|n_t|$ on a sphere, enclosing the particle, is not strong: $|n_t|\lesssim\sqrt{a/R}$. Moreover, calculations in [45,46] showed that Eq. (7) gives quite an accurate director field for the topological dipole of Ref. [35] even when $|n_t|$ is as large as 0.8.

C. Rotation about the homogeneous director n_∞ and second rank tensor of the elastic dipole

The interaction potentials (9) and (10) suggest that there are two elastic charge scalars and two elastic dipole vectors (and two second rank quadrupole tensors). When is this interpretation correct? Imagine that the director distribution at the source, n_x and n_y, changes but this change has nothing to do with a transformation of the reference frame. Then the new components after the change, n_x' and n_y', have no connection with their old values, n_x and n_y, via a tensorial transform in the the real x, y space. Similarly, the new values of q_t and $d_{t,\alpha}$ have no connection with their old values via a tensorial transform in the real x, y space. Such a change can be considered as a transform in some intrinsic space with the coordinates n_x and n_y or $\mathbf{d}_x$ and $\mathbf{d}_y$, reminiscent of the isotopic space of nuclear physics. Thus, under such an "intrinsic" transformation, n_x and n_y do not transform as components of a spatial vector. Then q_t is not a spatial vector but can be viewed as two independent charges, a dyad; similarly, $d_{t,\alpha}$ is not a spatial tensor but a dyad of two independent vectors $d_{x,\alpha}$ and $d_{y,\alpha}$. Another obstacle for viewing the object $d_{t,\alpha}$ as a tensor is that t runs over 1 and 2 whereas α runs over 1, 2, and 3. There is an important case, however, when q_t and $d_{t,\alpha}$ are a two-dimensional vector and second rank tensor, respectively.

As a rotation about the homogeneous director $\mathbf{n}_\infty$, which is along the z axis, does not alter the free energy of a particle, all the elastic multipoles should be determined up to an arbitrary azimuthal angular variable ϕ. Let us consider the subscript $\alpha=t'$ with the values 1 and 2 and with the value $\alpha=3$ separately. The indices taking values 1 and 2 will be denoted t or t'. Then q_t and $d_{t,3}$ are transformed as a two-dimensional vector, and $d_{tt'}$ is transformed as a second rank tensor. The vector q_t is fully determined by the external torque vector; $d_{t,3}$ will be addressed later on. Consider $d_{t,t'}$ and find its general ϕ-dependent form induced by a rotation of the particle about the z axis. A general second rank tensor $d_{tt'}$ is a sum of its symmetric and antisymmetric part. Here we consider only symmetric tensors. In the proper reference frame O_0, a symmetric tensor $\mathbf{d}=\|d_{tt'}\|$ reduces to its diagonal form $\mathbf{d}_0$, i.e.,

$$\mathbf{d}_0 = \begin{pmatrix} D_{11} & 0 \\ 0 & D_{22}\end{pmatrix}. \tag{11}$$

A symmetric second rank tensor can be decomposed into an isotropic part $\propto\delta_{ij}$ and anisotropic traceless part. In particular, $\mathbf{d}_0$ (11) (i.e., $\mathbf{d}$ in the particular reference frame O_0) can be written as

$$\mathbf{d}_0 = d\begin{pmatrix} 1 & 0 \\ 0 & 1\end{pmatrix} + \Delta\begin{pmatrix} 1 & 0 \\ 0 & -1\end{pmatrix}, \tag{12}$$

where $d=(D_{11}+D_{22})/2$, $\Delta=(D_{11}-D_{22})/2$. Now we want to find the form of the tensor $\mathbf{d}$ in an arbitrary reference frame

O_ϕ that can be obtained from O_0 by rotating it by an angle ϕ *anticlockwise* (the angle ϕ is counted from the x axis). Such a rotation induces the following transformation of an arbitrary vector $\mathbf{r}=(r_1,r_2)$: $\mathbf{r}\rightarrow\mathbf{r}'=(r_1',r_2')$, where $r_t'=R_{tt'}r_{t'}$ and the matrix $\|R_{tt'}\|$ is of the form

$$\|R_{tt'}\| = \begin{pmatrix} \cos\phi & \sin\phi \\ -\sin\phi & \cos\phi \end{pmatrix}. \tag{13}$$

The second rank tensor $\mathbf{d}_0$ is transformed to the reference frame O_ϕ as $d_{tt'}=R_{ts}R_{t's'}d_{0ss'}$. In the context of Eqs. (12) and (13) this gives

$$\mathbf{d} = d\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \Delta\begin{pmatrix} \cos 2\phi & -\sin 2\phi \\ -\sin 2\phi & -\cos 2\phi \end{pmatrix}. \tag{14}$$

As discussed below, if the tensor $d_{tt'}$ is symmetric then the vector $d_{t,3}$ vanishes. Thus, the dyad of dipole "vectors" in an arbitrary reference frame O_ϕ has the form

$$\mathbf{d}_x = (d + \Delta\cos 2\phi, -\Delta\sin 2\phi, 0),$$

$$\mathbf{d}_y = (-\Delta\sin 2\phi, d - \Delta\cos 2\phi, 0). \tag{15}$$

The angle ϕ can also be viewed as a rotation angle of the particle itself in the fixed reference frame O_0 where the dipole has the diagonal form $\mathbf{d}_0$ (14). Then ϕ is the angle of an *clockwise* particle rotation from its special diagonal state $\mathbf{d}(\phi=0)=\mathbf{d}_0$ to an arbitrary state $\mathbf{d}$: after this rotation the form of the dipole tensor $\mathbf{d}(\phi)$ will be given by formula (15). Note that formula (7) gives the following estimates for the magnitude of the coefficients d and Δ: $d\sim a^2\langle|n_t|\rangle$ where $\langle|n_t|\rangle$ is the average of the absolute value of the transverse director component over the enclosing sphere of radius a; while $\Delta\sim a^2(\langle|n_x|\rangle-\langle|n_y|\rangle)$.

In the case of a general asymmetric particle (which implies an asymmetric director distribution in its vicinity), the tensor $d_{tt'}$ has a finite antisymmetric part and the components $d_{t,3}$ are nonzero. The case of a symmetric tensor considered above corresponds to certain symmetry of the source particle. The rotational symmetry C_n of any order n about the unperturbed director is not sufficient to eliminate an antisymmetric part of $d_{tt'}$ and vector $d_{t,3}$. In general, these two quantities are finite if the director distribution has a helical component related to twist distortions. Such a deformation source has chiral properties and will be considered elsewhere [58]. Important symmetry elements are reflection planes passing through the z axis. A particle with just a single such reflection plane has no chirality, its tensor $d_{tt'}$ is symmetric, but the vector $d_{t,3}$ is not generally zero. We call such particles *general biaxial*. Two mutually perpendicular reflection planes passing through the z axis eliminate the vector $d_{t,3}$, i.e., $d_{x,3}=d_{y,3}=0$, and particles with this symmetry constitute an important particular case of colloids which we call *(just) biaxial*. It is this case of a biaxial colloid that was considered above.

A general biaxial particle is biaxial in the x, y plane, its director distribution in the x, y cross sections has mirror symmetry with respect to one of the two principal axes. A just biaxial particle is more symmetric: its director distribution in the x, y cross sections has mirror symmetry with respect to both principal axes. An important particular case of a biaxial particle is an azimuthally symmetric particle with the symmetry $C_{\infty v}$ (infinite order rotational symmetry about the z axis plus symmetry with respect to any plane passing through the z axis). Such a particle is both uniaxial and nonchiral; here we call such a particle uniaxial. Note, however, that an azimuthally symmetric (uniaxial) particle with the symmetry C_∞ without reflection planes (e.g., uniaxial helicoid) is a chiral source and has nonzero both antisymmetric part of $d_{tt'}$ and vector $d_{t,3}$ [58].

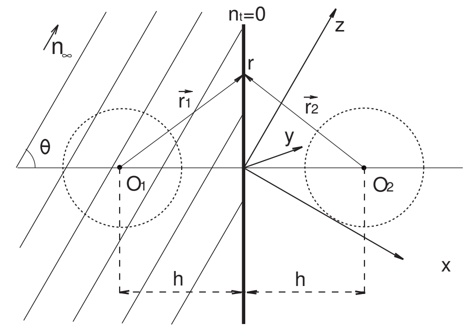

FIG. 2. Geometry of the problem considered. A wall (bold line), a particle at O_1, and its image at O_2. The z axis, which is along the unperturbed director n_∞, makes the angle θ with the normal to the wall. The x axis lies in the figure plane whereas the y axis is normal to it and directed from the reader. The perturbation n_t of n_∞ vanishes on the wall: $n_t=0$, $t=x,y$. The director field outside the dashed spheres, representing a finite size of the particle and its image, obeys linear equations.

Thus, under a rotation of a biaxial particle about the z axis its elastic dipole transforms as a second rank 2×2 tensor (14) with the isotropic fraction of magnitude d and anisotropic fraction of magnitude Δ, in which ϕ is the angle of the anticlockwise rotation of the particle from the diagonal dipolar state. All of the x, y cross sections of a uniaxial $C_{\infty v}$ particle are circular and have no anisotropy, $\Delta=0$; this can be easily verified by choosing an arbitrary azimuthally symmetric director distribution [surface charge density (5)] and integrating over the sphere in Eq. (7). Particles with a finite Δ are biaxial in the x, y plane, their director distribution in the x, y cross sections is not azimuthally symmetric, e.g., elliptical.

The above formulas can be used to solve boundary problems similar to those of electrostatics. The simplest boundary problem is the interaction of an elastic multipole with a plane surface bounding the nematic sample and imposing a fixed homogeneous director alignment. Here we consider this problem for the elastic charge and dipole.

III. THE MIRROR IMAGE METHOD

Consider a single particle at distance h from a plane surface of a NLC sample (wall). We assume that the wall's anchoring is strong, the director alignment in the sample far from the particle is homogeneous and parallel to the z axis, $\mathbf{n}_\infty=(0,0,1)$, but the angle θ it makes to the surface normal is arbitrary, Fig. 2; $\theta=\pi/2$ and $\theta=0$ correspond to the planar and homeotropic surface director alignment, respectively. To justify the linearized theory, h is assumed to be large compared to the particle size $\sim a$. As the director on the wall is fixed, the boundary condition consists of two equations

$$\mathbf{n}_t(\mathbf{r}_{wall}) = 0, \quad t = x,y, \tag{16}$$

which have to be satisfied for any point $\mathbf{r}_{wall}$ of the wall. The problem of a particle-wall interaction can be solved using the mirror-image method. Let us show that the boundary conditions (16) can be satisfied by placing an image particle with the multipole moment of the same order on the other side of wall.

Consider an elastic multipole at a distance h from a wall and its image multipole located at the mirror point behind the wall at the distance $R=2h$ from the real particle, Fig. 2. Variables, attributed to the particle and its image, will be indicated by index 1 and index 2, respectively. The director field, which determines the multipoles, is fixed at the spheres S_1 and S_2 of radius a enclosing, respectively, the particle and its image. To the order a/R, the transverse director components n_t on the wall are given by the sum [46]

$$n_t(\mathbf{r}) = n_t^{(1)}(\mathbf{r})\left(1 - \frac{a}{r_2}\right) + n_t^{(2)}(\mathbf{r})\left(1 - \frac{a}{r_1}\right), \tag{17}$$

where $n_t^{(1)}$ is the far field (2) of the single particle, $\mathbf{r}_1=\mathbf{r}-\mathbf{o}_1$, $\mathbf{r}_2=\mathbf{r}-\mathbf{o}_2$, where $\mathbf{o}_1$ and $\mathbf{o}_2$ are the radius vectors of the two centers. Equation (17) explicitly shows nonadditivity of the fields produced by the two particles: $n_t=n_t^{(1)}$ on S_1 ($r_1=a$) and $n_t=n_t^{(2)}$ on S_2 ($r_2=a$); the additivity $n_t(\mathbf{r})\sim n_t^{(1)}+n_t^{(2)}$ takes place only when both $a/r_1 \ll 1$ and $a/r_2 \ll 1$, i.e., far from the particles.

Let us set the reference frame with the onset on the wall between the particles so that the xz plane (with z axis along $\mathbf{n}_\infty$) is perpendicular to the wall, and the y axis is normal to the figure plane, Fig. 2. Then $\mathbf{o}_1=(-h \sin\theta, 0, -h\cos\theta)$, $\mathbf{o}_2=(h\sin\theta, 0, h\cos\theta)$, an arbitrary point of the wall $\mathbf{r}_{wall}=(-z\cos\theta, y, z\sin\theta)$, and

$$\mathbf{r}_1 = (-z\cos\theta + h\sin\theta,\ y,\ z\sin\theta + h\cos\theta),$$

$$\mathbf{r}_2 = (-z\cos\theta - h\sin\theta,\ y,\ z\sin\theta - h\cos\theta). \tag{18}$$

The particle-image separation vector is $\mathbf{R}=\mathbf{o}_2-\mathbf{o}_1=R\mathbf{u}$, where $\mathbf{u}=(\sin\theta, 0, \cos\theta)$. In the context of Eq. (17) and the equality $r_1=r_2$, the condition $\mathbf{n}_t=0$ on the wall gives

$$n_t^{(1)}(\mathbf{r}_{wall}) + n_t^{(2)}(\mathbf{r}_{wall}) = 0, \quad t = x,y. \tag{19}$$

These equations determine the components of the image multipole. We will find the image charge and image dipole for arbitrary tilt θ and then, by substituting its components in the interaction energy (9) or (10), calculate the interaction between the colloid and the wall. Below we consider a charged, nonchiral $C_{\infty v}$ uniaxial, biaxial, and general dipolar particle individually.

IV. PARTICLE-WALL INTERACTION IN A NLC

A. Elastic charge-wall interaction

The charge $q_t^{(1)}$ can be induced by an external field exerting the torque Γ with the components $\Gamma_y=4\pi K q_x^{(1)}$ and $\Gamma_x=-4\pi K q_y^{(1)}$; the image charge $q_t^{(2)}$ is on the other side of the wall at the distance h from it, Fig. 3. The director field of a single elastic charge is $n_t^{(i)}=q_t^{(i)}/r_i$, Eq. (2). Then from Eq. (19) we obtain $q_t^{(2)}=-q_t^{(1)}$, $t=x,y$. Thus, the particle and its image have opposite charges which corresponds to $\mathbf{\Gamma}_\perp^{(2)}=-\mathbf{\Gamma}_\perp^{(1)}$, Fig. 3. As two opposite elastic charges repel one another, the elastic charge-wall interaction is repulsive. The repulsion force obtains from the interaction energy (9) of the torques $\mathbf{\Gamma}_\perp$ and $-\mathbf{\Gamma}_\perp$ by differentiating with respect to R at $R=2h$, i.e.,

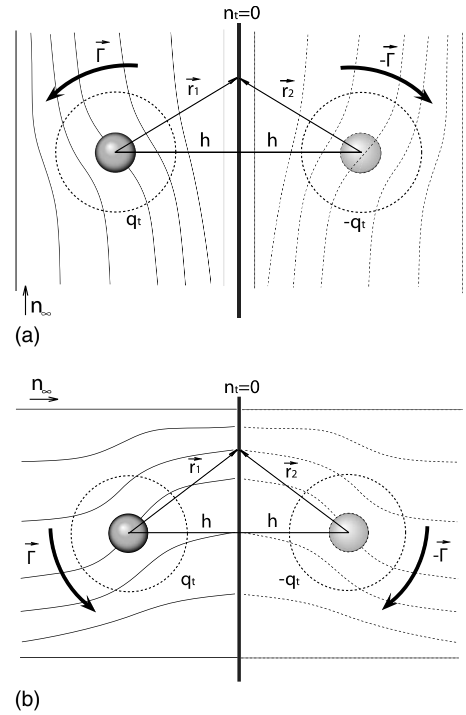

FIG. 3. Elastic charge at a wall with fixed planar (upper sketch) and homeotropic (lower sketch) director alignment. Elastic charge q_t induced by an external torque $\mathbf{\Gamma}$ and its image $-q_t$ induced by the image torque $-\mathbf{\Gamma}$. The director at the wall remains unperturbed and equal to $\mathbf{n}_\infty$.

$$F_{q\text{-}wall} = \frac{\mathbf{\Gamma}_\perp^2}{16\pi K h^2}. \tag{20}$$

The result depends on the direction of $\mathbf{n}_\infty$ and thus on the surface tilt only via the trivial relation $\mathbf{\Gamma}_\perp=\mathbf{\Gamma}-(\mathbf{\Gamma}\cdot\mathbf{n}_\infty)$.

B. Interaction of a uniaxial $C_{\infty v}$ dipole with a wall

Now consider the interaction between a wall and an elastic dipole dyad $(\mathbf{d}_x, \mathbf{d}_y)$, Eqs. (7) and (15). In general, each of $\mathbf{d}_x$ and $\mathbf{d}_y$ has three nonzero components. Symmetry makes some of them vanish. For a uniaxial $C_{\infty v}$ particle the dipole dyad is isotropic: $d_{tt'}=d\delta_{tt'}$. Consider this simple and practically important case of colloids. For instance, such are the so-called topological dipoles, i.e., spherical particles with homeotropic boundary conditions with a companion hyperbolic hedgehog or disclination ring [3,35].

The dipole dyad of a uniaxial particle obtains from Eq. (15) by setting $\Delta=0$,

$$\mathbf{d}_x^{(1)} = (d,0,0), \quad \mathbf{d}_y^{(1)} = (0,d,0). \tag{21}$$

The interaction energy (10) of two uniaxial $C_{\infty v}$ dipolar particles with isotropic coefficients $d^{(1)}$ and $d^{(2)}$ is obtained in the form

$$U_{1d-1d} = \frac{12\pi K d^{(1)} d^{(2)}}{R^3}(1-3\cos^2\theta), \tag{22}$$

where θ is the angle the separation vector $\mathbf{R}$ (which is along the surface normal) makes with the far homogeneous director $\mathbf{n}_\infty$. To solve the boundary condition (19), the image-dipole $\mathbf{d}_t^{(2)}$ is chosen in the form

$$\mathbf{d}_x^{(2)} = (d_x^{(2)}\cos\psi, 0, -d_x^{(2)}\sin\psi),$$

$$\mathbf{d}_y^{(2)} = (0, d_y^{(2)}, 0), \tag{23}$$

where ψ is the angle $\mathbf{d}_x^{(2)}$ makes with the x axis. The director field of a single ith dipole at point $\mathbf{r}_{wall}$ of the surface is $n_t^{(i)} = 3(\mathbf{d}_t^{(i)}\cdot\mathbf{r}_i)/r_i^3$, see Eq. (2), and the boundary conditions read

$$(\mathbf{d}_t^{(1)}\cdot\mathbf{r}_1) + (\mathbf{d}_t^{(2)}\cdot\mathbf{r}_2) = 0, \quad t = x,y. \tag{24}$$

Substituting Eqs. (21), (23), and (18), into Eq. (24) gives the equations

$$h(d\sin\theta - d_x^{(2)}\sin\theta\cos\psi + d_x^{(2)}\cos\theta\sin\psi) - z(d\cos\theta + d_x^{(2)}\cos\theta\cos\psi + d_x^{(2)}\sin\theta\sin\psi) = 0,$$

$$d + d_y^{(2)} = 0, \tag{25}$$

which are equivalent to the system

$$d\sin\theta - d_x^{(2)}\sin(\theta-\psi) = 0,$$

$$d\cos\theta + d_x^{(2)}\cos(\theta-\psi) = 0,$$

$$d + d_y^{(2)} = 0. \tag{26}$$

This system is solved by $d_x^{(2)} = d_y^{(2)} = -d$, $\psi = 2\theta$, i.e.,

$$\mathbf{d}_x^{(2)} = (-d\cos 2\theta, 0, d\sin 2\theta),$$

$$\mathbf{d}_y^{(2)} = (0, -d, 0). \tag{27}$$

Substituting Eq. (27) into (10) with $\mathbf{u} = (\sin\theta, 0, \cos\theta)$ gives

$$U_{1d} = 12\pi K \frac{d^2(\sin^2\theta + 2)}{R^3}. \tag{28}$$

Differentiating (28) with respect to R at $R = 2h$, we obtain the force with which the wall repels a uniaxial dipole,

$$F_{1d\text{-}wall} = \pi K \frac{9d^2(2+\sin^2\theta)}{4h^4}. \tag{29}$$

Now consider two important particular cases of the planar and homeotropic wall.

For the planar wall $\theta = \pi/2$, the image has the form $\mathbf{d}_x^{(2)} = (d,0,0)$, $\mathbf{d}_y^{(2)} = (0,-d,0)$, Fig. 4, and the repulsive force has the magnitude

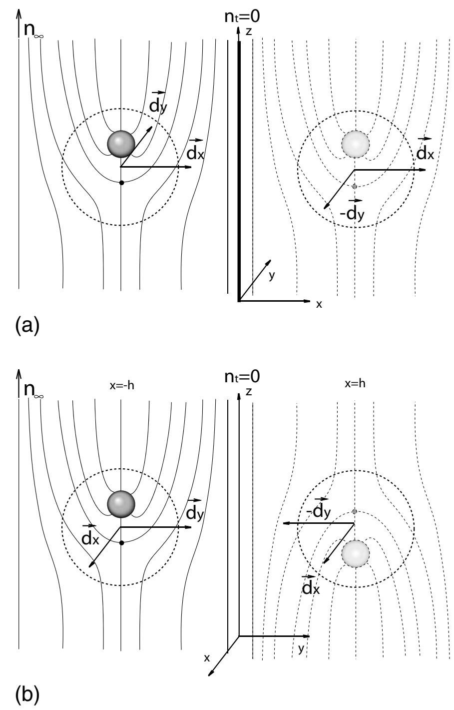

FIG. 4. Dyad of elastic dipole at a wall with fixed planar director alignment. The dipole (left) with $\mathbf{d}_x = (d,0,0)$ and $\mathbf{d}_y = (0,d,0)$ and the image (right) with $\mathbf{d}_x^{(2)} = (d,0,0)$ and $\mathbf{d}_y^{(2)} = (0,-d,0)$ shown in two mutually perpendicular planes: xz plane normal to the wall (upper sketch) and planes $x = -h$ (left) and $x = h$ (right) parallel to the wall (lower sketch).

$$F_{1d\text{-}wall,planar} = \pi K \frac{27d^2}{4h^4}. \tag{30}$$

For the homeotropic wall $\theta = 0$, $\mathbf{d}_x^{(2)} = (-d,0,0)$, $\mathbf{d}_y^{(2)} = (0,-d,0)$, Fig. 5, and the repulsive force has the magnitude

$$F_{1d\text{-}wall,hom} = \pi K \frac{9d^2}{2h^4}. \tag{31}$$

Thus, the force $F_{1d\text{-}wall,hom}$ is 1.5 times weaker than $F_{1d\text{-}wall,planar}$. In this concern we would like to emphasize that the results, obtained above by means of the nematostatics of

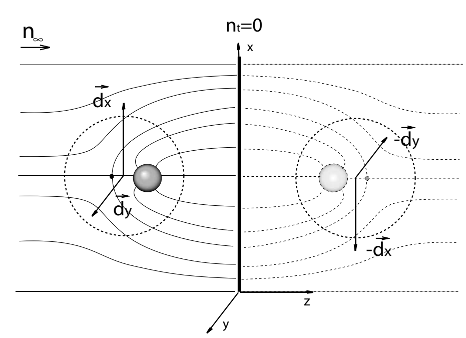

FIG. 5. Dyad of elastic dipole at a wall with fixed homeotropic director alignment. The dipole (left) with $\mathbf{d}_x = (d,0,0)$ and $\mathbf{d}_y = (0,d,0)$ and the image (right) with $\mathbf{d}_x^{(2)} = (-d,0,0)$ and $\mathbf{d}_y^{(2)} = (0,-d,0)$ shown in the xz plane normal to the wall.

Refs. [45,46], show that even in the simplest case of a homeotropic and planar wall the dyadic nature of the elastic dipole is essential. Indeed, a single dipole component is insufficient to satisfy the two boundary conditions (16) on the wall. Therefore, phenomenological theories, such as [35], which deal solely with a uniaxial dipolar particle and describe it by a single coefficient d, cannot be used in the problem of a particle-wall interaction. Nevertheless, let us compare our result with what could be "naively" obtained from the phenomenological approach in the simplest cases. When the poles of a single dipole interchange, the dipole changes its orientation to the inverse one and is described by the coefficient $(-d)$. In this situation, the image dipole can be chosen only in the form of the single-component dipole with the same or inverted sign. In the homeotropic case, the image dipole $d^{(2)}=-d$ allows to eliminate the particle-induced perturbation n_t on the wall and thus to satisfy the boundary condition (19). In the planar case, however, the y equation cannot be satisfied: the choice $d^{(2)}=-d$ is unacceptable as it would result in attraction which is obviously incorrect, whereas the choice $d^{(2)}=d$ satisfies only the x equation (19). Nevertheless, this last choice is the only possible one in the sense that at least it results in a repulsion from the wall. Then, using Eq. (22) with $|d^{(1)}d^{(2)}|=d^2$, one obtains for the planar ($\theta=\pi/2$) and homeotropic ($\theta=0$) cases, respectively, the following formulas:

$$\tilde{F}_{1d\text{-}wall,planar}=\pi K\frac{9d^2}{4h^4},$$

$$\tilde{F}_{1d\text{-}wall,hom}=\pi K\frac{18d^2}{4h^4}. \tag{32}$$

Apart from the difference in the coefficients, the "naive" prediction is that the repulsion from a wall with homeotropic anchoring is twice as strong than that from a wall with planar anchoring, $\tilde{F}_{1d\text{-}wall,hom}/\tilde{F}_{1d\text{-}wall,planar}=2$. This is in contrast to the above exact result

$$F_{1d\text{-}wall,hom}/F_{1d\text{-}wall,planar}=2/3. \tag{33}$$

Prediction (33) can be verified in an experiment similar to that recently reported by Pishnyak *et al.* [44] who studied the interaction between colloids of the topological dipole type [35] and a NLC surface with a planar anchoring. In a thick cell with horizontal surfaces, the distance h from the lower surface is set in the balance between the attraction due to colloid's weight and the elastic dipole-surface repulsion. The ratio h_h/h_p of the equilibrium distance h_h of a topological dipole from a lower surface with a homeotropic anchoring to the distance h_p from that with a planar anchoring can be estimated experimentally and compared both with $h_h/h_p=\sqrt[4]{2/3}\simeq 0.9$, following from the prediction (33) of this paper, and with $h_h/h_p=\sqrt[4]{2}\simeq 1.2$, following from the "naive" prediction (32). Note that the value of the coefficient d in our theory, which does not enter the ratio h_h/h_p, is $1/3$ of that calculated numerically in [35] (see Refs. [45,46]).

Another interesting effect to observe is "charging" of a colloid, i.e., inducing an elastic charge by exerting external torque [e.g., by applying an electric (magnetic) field if the particle is ferroelectric (ferromagnetic)]. A torque on a colloid creates an elastic charge which is repelled from the wall with a force (20). In the case of a dipole-type colloid, an external torque would switch the h^{-4} dipole-wall repulsion to a h^{-2} charge-wall repulsion which can manifest itself, in particular, in a sharp increase of the equilibrium distance from the wall.

C. Interaction of a general dipole with a wall

The mirror-image method can be applied to dipolar particles of arbitrary shape. In the general case, dipole dyads of a real particle and its image have all the three components, $\mathbf{d}_t^{(1)}=(\alpha_t^{(1)},\beta_t^{(1)},\gamma_t^{(1)})$ and $\mathbf{d}_t^{(2)}=(\alpha_t^{(2)},\beta_t^{(2)},\gamma_t^{(2)})$. Then the two boundary conditions (19) split into the following three equations:

$$\alpha_t^{(1)}+\alpha_t^{(2)}-(\gamma_t^{(1)}+\gamma_t^{(2)})\tan\theta=0,$$

$$(\alpha_t^{(1)}-\alpha_t^{(2)})\tan\theta+\gamma_t^{(1)}-\gamma_t^{(2)}=0,$$

$$\beta_t^{(1)}+\beta_t^{(2)}=0. \tag{34}$$

This system is solved with

$$\alpha_t^{(2)}=-\alpha_t^{(1)}\cos 2\theta+\gamma_t^{(1)}\sin 2\theta,$$

$$\beta_t^{(2)}=-\beta_t^{(1)},$$

$$\gamma_t^{(2)}=\gamma_t^{(1)}\cos 2\theta+\alpha_t^{(1)}\sin 2\theta. \tag{35}$$

Thus, the dyad of the general image dipole has the form

$$\mathbf{d}_t^{(2)}=(-\alpha_t^{(1)}\cos 2\theta+\gamma_t^{(1)}\sin 2\theta,-\beta_t^{(1)},\gamma_t^{(1)}\cos 2\theta+\alpha_t^{(1)}\sin 2\theta). \tag{36}$$

Substituting Eq. (36) into Eq. (10), we obtain

$$U_d=\frac{12\pi K}{R^3}[\alpha_t^{(1)}\alpha_t^{(1)}(1+\sin^2\theta)+\beta_t^{(1)}\beta_t^{(1)}+\gamma_t^{(1)}\gamma_t^{(1)}(1+\cos^2\theta)-\alpha_t^{(1)}\gamma_t^{(1)}\sin^2\theta]. \tag{37}$$

Differentiating (37) with respect to R at $R=2h$ gives the repulsion force of an arbitrary elastic dipole from the wall, i.e.,

$$F_{d\text{-}wall}=\frac{9\pi K}{4h^4}[\alpha_t^{(1)}\alpha_t^{(1)}(1+\sin^2\theta)+\beta_t^{(1)}\beta_t^{(1)}+\gamma_t^{(1)}\gamma_t^{(1)}(1+\cos^2\theta)-\alpha_t^{(1)}\gamma_t^{(1)}\sin^2\theta]. \tag{38}$$

It is important to remember that the components of the dipole dyad entering this formula depend on an arbitrary angle ϕ, Eq. (15). This fact is trivial in the homogeneous space as the particle energy is degenerate in ϕ, but the presence of a wall breaks the azimuthal symmetry around the z axis and removes the degeneracy. This means that the angle ϕ changes as to minimize the interaction energy with the wall. In other words, a particle, approaching wall from a large distance, will turn to assume some particular orientation with $\phi=\phi_m$. Below we consider an important particular case described by formula (38) with an explicit ϕ dependence.

D. Interaction of a biaxial dipole with a wall

Consider a biaxial particle whose elastic dipole, as described in Sec. II C, has no z components: from Eq. (15) such a dipole dyad is obtained as $\mathbf{d}_x^{(1)}=(d+\Delta\cos 2\phi,-\Delta\sin 2\phi,0)$, $\mathbf{d}_y^{(1)}=(-\Delta\sin 2\phi,d-\Delta\cos 2\phi,0)$. Identifying from this expression the components of $\mathbf{d}_t^{(1)}=(\alpha_t^{(1)},\beta_t^{(1)},\gamma_t^{(1)})$ and substituting them in Eq. (38) gives the repulsion force

$$F_{2d\text{-}wall}=\frac{9\pi K}{4h^4}[(d^2+\Delta^2)(2+\sin^2\theta)+2\Delta d\cos 2\phi\sin^2\theta]. \tag{39}$$

Minimization with respect to ϕ [the free energy and the force have the same ϕ dependence (39)] gives (i) ϕ is arbitrary for a uniaxial $C_{\infty v}$ particle with $\Delta=0$, which is obvious as such a particle is invariant with respect to rotation about the z axis; (ii) ϕ is arbitrary at the homeotropic wall, $\theta=0$, which is obvious as such a wall does not break the symmetry about the z axis, Fig. 5; (iii) for $0<\theta\leq\pi/2$, $\phi=0$ if $\Delta<0$ and $\phi=\pi/2$ if $\Delta>0$; this means that the longer axis of the particle's x, y cross section turns parallel to the wall which obviously saves the elastic energy.

Particles producing director deformations with two symmetry planes passing through the z axis, referred to here as just biaxial, can be exemplified by objects of the following geometries: the topological dipole of Ref. [35] but created by an ellipsoid with the normal surface anchoring rather than by a sphere; by an elliptical cone with its long axis along $\mathbf{n}_\infty$ and planar anchoring at its lateral surface. In all of these cases the equilibrium orientation of the long axis in the transverse x, y cross section is parallel to the wall, and thus the repulsion from a wall is accompanied by a reorientation about the homogeneous director. The only exclusion is the case of homeotropic wall when the only effect is the repulsion.

V. CONCLUSION

The nematostatics in 2D and 3D is very different. The former is very similar to the two-dimensional electrostatics where disclination cores are in place of electric charges. The latter is similar to the electrostatics only in that its Green's functions are Coulomb-like [45,46]. In 3D the counterpart of the electric charge density is a dyad, the elastic charge can be induced only by an external torque whose components play the role of the elastic charge dyad. In this three-dimensional colloidal nematostatics, the Coulomb-like interaction has the reverse sign. We described some implications of the colloidal nematostatics in 3D and showed that, in contrast to the electrostatics, the charges and dipoles are repelled from the wall and turn about the homogeneous director. Using the simplest geometries of a homeotropic and planar wall, we demonstrated that the problem of particle-wall interaction in a NLC can be solved only in the framework of the colloidal nematostatics with its dyadic multipole structure. One interesting effect, which can be derived from our calculations is that, applying the field-induced torque on a dipolar colloid, one can charge it, Fig. 1, thus switching the repulsion from the nematic surface from the $1/h^4$ to the $1/h^2$ regime. Our results prompt experimental tests of the interaction in nematic emulsions that, rather than dealing with a pair of particles, can deal with a single colloid at a wall. In particular, the prediction of the nematostatics can be tested by measuring a relative repulsive force from a homeotropic and planar walls with strong anchoring and comparing the results with the prediction expressed by Eq. (33).

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