# **Coulomb-like interaction in nematic emulsions induced by external torques exerted on the colloids**

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An external mechanical torque on colloids immersed in a nematic liquid crystal can induce a Coulomb-like 1/*r* interaction between them Lev and Tomchuk, Phys. Rev. E **59**, 591 1999-; Lev *et al.*, *ibid.* **65**, 021709 (2002)]. In this paper we show that the director-mediated Coulomb-like interaction of two colloids is determined by the vectors  $\Gamma_{\perp}^{(1)}$  and  $\Gamma_{\perp}^{(2)}$  of the transverse external torques exerted upon these colloids. We derive the 1/*r* potential in which the scalar product  $-(\Gamma_{\perp}^{(1)} \cdot \Gamma_{\perp}^{(2)})$  of the two torques plays the role of the product of two electrostatic charges. The  $1/r$  interaction is attractive for  $(\Gamma_{\perp}^{(1)} \cdot \Gamma_{\perp}^{(2)}) > 0$  and repulsive for  $(\Gamma_{\perp}^{(1)} \cdot \Gamma_{\perp}^{(2)}) < 0$ ("parallel torques" attract whereas "antiparallel torques" repel each other). The vector of transverse torque determines the two-component "elastic charge" (dyad), which is illustrated by the  $1/r^2$  and  $1/r^3$  terms in the elastic energy (the elastic analogs of the monopole-dipole and dipole-dipole interactions). The general status of the pairwise approach to nematic emulsions is considered in terms of the elastic charge density.

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### **I. INTRODUCTION**

Particles of submicrometer and micrometer size immersed in a nematic liquid crystal interact via the director field **n** which mediates the distortions induced by their surfaces  $[1-11]$  $[1-11]$  $[1-11]$ . The new field of nematic colloidal systems, or nematic emulsions  $[11,12]$  $[11,12]$  $[11,12]$  $[11,12]$ , has gained continuously growing interest over the past few years. The physics of these anisotropic colloidal systems has a deep similarity to electrostatics. It has been shown that the director-mediated interaction is of a long range and possesses many other properties characteristic of the interaction between electric dipoles and quadrupoles  $[1-11]$  $[1-11]$  $[1-11]$ , and even charges  $[1,2]$  $[1,2]$  $[1,2]$ . Particle trapping techniques  $[14]$  $[14]$  $[14]$  have been used to test this analogy and demonstrate experimentally the dipole-dipole [[15](#page-7-5)[,16](#page-7-6)] and quadrupole-quadrupole  $[17,18]$  $[17,18]$  $[17,18]$  $[17,18]$  pair interactions. Very recently, reorientation of the elastic dipoles was shown to be responsible for phase transitions between different twodimensional colloidal lattices on a nematic-air interface  $\lceil 13 \rceil$  $\lceil 13 \rceil$  $\lceil 13 \rceil$ . In the context of this analogy it is natural to expect that the Coulomb interaction, which is fundamental to the electrostatics, has an important implication in the physics of nematic emulsions, too. However, the electrostatic analogy has not been fully developed to this level.

Mathematically, the similarity between the directormediated and electrostatic interactions lies in the massless nature of both theories and the Coulomb-like behavior of the Green functions which derives from it. The massive term, which has the form of the square of the order parameter, is absent in the fundamental functionals of both theories. The theory of the electrostatic field  $\varphi$  is massless as the term  $m\varphi^2/2$  is absent in its energy. It is this property that gives rise to the Coulomb 1/*r* potential: a finite massive term would have made it short range, i.e., exp(*-mr*)/*r*. In turn, the elastic theory of a nematic liquid crystal describes the field of the director **n** which is a unit vector. The massless nature of the director field follows from this definition: the energy term quadratic in **n** is trivial as  $n^2 = 1$  [[19](#page-7-10)]. As a result, the elastic theory allows for director components that far from the distortion source behave as 1/*r* and its higher powers, which is formally similar to the potential of an electric charge, electric dipole, and so on.

The two systems are not, however, completely similar. The 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 the electric field. At the same time, **n** is a vector field described by a linear equation (in the one-constant approximation) only in two dimensions. Owing to the linearity, the deformation source can be straightforwardly established: the core of a point defect plays the role of a charge in two dimensions [19](#page-7-10)[–22](#page-7-11). But in three dimensions the field **n** is described by highly nonlinear equations  $[19]$  $[19]$  $[19]$  and point defects cannot be linearly connected with the distortions of **n** they induce  $[9,11]$  $[9,11]$  $[9,11]$  $[9,11]$ . In principle, solutions to these nonlinear equations with appropriate boundary conditions determine the director field produced by any source, but in most cases an analytical solution to this problem is not known.

The deformations, however, are decreasing functions of the distance *r* from the source so that far enough they become sufficiently weak to allow for a linear description and thus for the electrostatic analogy. Then the problem is to make a connection between the asymptotic electrostaticlike far director field and its nonlinear source. Lev *et al.* [[2](#page-7-3)] explicitly introduced a surface separating the nonlinear near zone (called the elastic coat in  $[2]$  $[2]$  $[2]$ ) from the linear far zone. \*Electronic address: victorpergam@yahoo.com The director in the far zone is connected to the source via

<span id="page-0-0"></span>

this phenomenological surface and polar anchoring on it; the surface shape is chosen so as to reflect the distortion symmetry in the near zone and the coordinate-dependent anchoring so as to provide the correct deformation magnitude in the far zone. The result is a pair interaction potential that contains all powers of  $1/r$  $1/r$ : the  $1/r$  term first obtained in [1] is interpreted as a Coulomb interaction,  $1/r^3$  as the dipoledipole interaction, and so on  $[2]$  $[2]$  $[2]$ . This enabled the authors of [[2](#page-7-3)] to express the dipole and quadrupole coefficients, earlier introduced phenomenologically in  $[9,11]$  $[9,11]$  $[9,11]$  $[9,11]$ , as integrals over the chosen surface. Moreover, in  $[2]$  $[2]$  $[2]$ , a Coulomb-like attraction of two similarly oriented colloids was discussed and a scalar quantity for the elastic charge was introduced. Based on the well-known result  $\left[19\right]$  $\left[19\right]$  $\left[19\right]$  that the source of the  $1/r$ director behavior is the mechanical torque exerted on the colloid, Lev *et al.* emphasized that the Coulomb-like attraction can be induced by the vector of external torque  $\Gamma$ . In [[2](#page-7-3)], however,  $\Gamma$  does not explicitly determine the magnitude of the 1/*r* attraction; instead, it is expressed in terms of the specific colloid's shape and anchoring. Apparently for this reason, the Coulomb term is suggested to be nonzero when the elastic coat has no horizontal or vertical mirror symmetries. But this does not require any torque if the particle is asymmetric.

In this paper we show that the director-mediated Coulomb-like interaction of two colloids is fully determined by the vectors  $\Gamma_{\perp}^{(1)}$  and  $\Gamma_{\perp}^{(2)}$  of the transverse external torques (perpendicular to the unperturbed director at infinity) applied on the colloids. We derive the 1/*r* interaction potential in which the scalar product  $-(\Gamma_{\perp}^{(1)} \cdot \Gamma_{\perp}^{(2)})$  of the two torques plays the role of the product of two electrostatic charges. Consequently, the interaction can be both attractive and repulsive: it is attractive for  $(\Gamma_{\perp}^{(1)} \cdot \Gamma_{\perp}^{(2)}) > 0$  and repulsive for  $(\Gamma_{\perp}^{(1)}\cdot\Gamma_{\perp}^{(2)})$  < 0 ("parallel torques" attract whereas "antiparallel torques" repel each other). 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 a permanent electric and magnetic dipole or the electric and magnetic polarizability tensors of a given colloid. We also propose a representation in which, 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 (a dyad). 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.

# **II. LINEARIZED DIRECTOR DISTORTIONS AND THEIR SOURCE**

Consider a three-dimensional (3D) director field  $n(r)$ , uniform and parallel to the *z* axis at infinity,  $\mathbf{n}_{\infty} = (0,0,1)$ , and distorted in a finite number of particlelike (compact) domains. We call such deformation domains particles though

<span id="page-1-0"></span>

FIG. 1. Particlelike distortion domain with nonlinear core (darkened). At the enclosing spherical surface *S* the director component normal to the unperturbed uniform director  $\mathbf{n}_{\infty}$  is small, whereas at the nonlinear core it can be large. **i** is the image of the point **r** in *S*.

the distortion therein can be induced by the surface of a real particle, by topological defects with zero total topological charge  $[9,11]$  $[9,11]$  $[9,11]$  $[9,11]$ , or by an external field dying out outside the domain area. Generally, the deformations at the domain center are strong and satisfy nonlinear equations, but sufficiently far away they become weak (Fig. [1](#page-1-0)). Here the transverse director  $\mathbf{n}_{\perp} = (n_x, n_y, 0)$  (perturbation to  $\mathbf{n}_{\infty}$ ) is small,  $|\mathbf{n}_{\perp}|$  $\leq 1$ , and satisfies linear equations.

Enclose the *i*th domain by a spherical surface  $S_i$  of radius  $a_i$  such that outside it  $|\mathbf{n}| \leq 1$ . At the same time, the sphere can be small as compared to the interparticle distances, and the radius-to-distance ratio is a small parameter. Inside  $S_i$  the theory is nonlinear, intractable in standard terms (maybe experimentally inaccessible), and we will consider the director  $\mathbf{n}_{\perp}(\mathbf{s})$  on each  $S_i$  as a given function of  $\mathbf{s} \in S_i$ . A sphere has the following unique advantages: first, it does not introduce any symmetry element so that the outer director is fully determined by  $\mathbf{n}_{\perp}$  on the sphere, and, second, it enables us to calculate Green functions, which seems to be a task impossible for other surfaces. The final results do not depend on the choice of the surface.

It is implied in what follows that the index *t* takes values *x* or *y*, greek indices run over *x*, *y*, and *z*, the index *i* stands for the particle number, summation over repeating indices is performed, and vector **s** refers to a point on the surface of any of the spheres. For simplicity, we omit the divergence free energy (FE) terms and work in the one-constant approximation  $[19]$  $[19]$  $[19]$ . Then the distortion FE functional for the area *Vout* outside all the enclosing spheres is given by

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

<span id="page-1-1"></span>The static transverse director component  $n_t$  satisfies the Laplace equation  $\Delta n_t = 0$ . Integrating ([1](#page-1-1)) by parts in the con<span id="page-2-8"></span>text of this equation, the equilibrium FE reduces to the integral over the total surface  $\cup S_i$  of all spheres:

$$
F = \frac{K}{2} \int_{\cup S_i} n_t(\boldsymbol{\nu} \cdot \nabla) n_t d^2 s,
$$
 (2)

where  $\nu = \nu(s)$  is the unit outer normal to the surface  $\cup S_i$  at **s** [[23](#page-7-13)]. If  $G(\mathbf{r}, \mathbf{r}')$  is the Green function of the Laplace equation subject to the boundary condition  $G(\mathbf{r}, \mathbf{s}) = 0$  for **s**  $\in$  ∪ *S<sub>i</sub>*, *G*(**r**, **s**) → 0 for  $|r$ −**s** $| \rightarrow \infty$ , then the solution to the Laplace equation in the area  $V_{out}$  is fully determined by the distribution of  $n_t$  on its inner boundary  $\bigcup S_i$  via the following Green formula  $|24|$  $|24|$  $|24|$ :

$$
n_t(\mathbf{r}) = -\int_{\bigcup S_i} n_t(\mathbf{s}) [\boldsymbol{\nu}(\mathbf{s}) \cdot \nabla_{\mathbf{s}}] G(\mathbf{r}, \mathbf{s}) d^2 s. \tag{3}
$$

<span id="page-2-1"></span>We see that in the linearized theory, the surface director component  $n_t$ (s) plays the role of the source of the director component  $n_t(\mathbf{r})$  in the outer area. We show below that  $n_x(\mathbf{s})$ and  $n_y(s)$  determine the surface densities of what can be considered as a two-component elastic charge.

# **III. ELASTIC CHARGE DENSITY AND MULTIPOLES OF A SINGLE PARTICLE**

Consider first a single particle enclosed by a sphere *S* of radius *a* and set the coordinate origin at its center. The oneparticle Green function  $G_1$  subject to the boundary condition  $G_1(\mathbf{r}, \mathbf{s}) = 0$  for  $\mathbf{s} \in S$  is known exactly [[24,](#page-7-14)[25](#page-7-15)]:

$$
G_1(\mathbf{r}, \mathbf{r}') = \frac{1}{4\pi} \left( \frac{1}{|\mathbf{r} - \mathbf{r}'|} - \frac{a}{r} \frac{1}{|\mathbf{i}(\mathbf{r}) - \mathbf{r}'|} \right),\tag{4}
$$

<span id="page-2-12"></span>where  $\mathbf{i}(\mathbf{r}) = (a/r)^2 \mathbf{r}$  is the location of the image of the point **r** in the spherical surface which has to be taken with the coefficient *a*/*r*. The normal surface derivative of the Green function  $G_1$  is obtained in the form [[24](#page-7-14)]

$$
[\boldsymbol{\nu}(\mathbf{s}) \cdot \nabla_{\mathbf{s}}] G_1(\mathbf{r}, \mathbf{s}) = -\frac{1}{4\pi} \frac{r^2 - a^2}{a |\mathbf{r} - \mathbf{s}|^3}.
$$
 (5)

<span id="page-2-0"></span>Substituting  $(5)$  $(5)$  $(5)$  in integral  $(3)$  $(3)$  $(3)$  gives the outer Poisson integral, which is the exact formula for the director induced outside the single sphere by the given director on its surface, i.e.,

$$
n_t(\mathbf{r}) = \frac{1}{4\pi} \int_S n_t(\mathbf{s}) \frac{r^2 - a^2}{a|\mathbf{r} - \mathbf{s}|^3} d^2 s.
$$
 (6)

<span id="page-2-3"></span><span id="page-2-2"></span>Expanding ([6](#page-2-2)) in a power series of the inverse distance *r* from the origin yields

$$
n_t(\mathbf{r}) = \frac{q_t}{r} + 3\frac{(\mathbf{d}_t \cdot \mathbf{r})}{r^3} + 5\frac{(\mathbf{Q}_t \cdot \mathbf{r})}{r^5} + \cdots, \tag{7}
$$

<span id="page-2-10"></span><span id="page-2-7"></span>where

$$
\sigma_t(\mathbf{s}) = n_t(\mathbf{s})/a^2,\tag{8}
$$

$$
q_t = \frac{a}{4\pi} \int_S \sigma_t d^2 s,\tag{9}
$$

$$
d_{t,\alpha} = \frac{a^2}{4\pi} \int_S \sigma_t \nu_\alpha d^2 s,\tag{10}
$$

<span id="page-2-11"></span>
$$
Q_{t,\alpha\beta} = \frac{a^3}{8\pi} \int_S \sigma_t (3\nu_\alpha \nu_\beta - \delta_{\alpha\beta}) d^2 s. \tag{11}
$$

<span id="page-2-4"></span>Equations  $(7)$  $(7)$  $(7)$ – $(11)$  $(11)$  $(11)$  suggest the following interpretation.  $q_t$ is the *t*th component of the elastic charge and  $\sigma_t(s)$  is its surface density at point **s** on the sphere. The vector  $\mathbf{d}_t$  and tensor  $Q_t$  are the *t*th dipole and quadrupole moments determined in the standard way by the surface charge density  $\sigma_t$ on the sphere. As  $\sigma_r$  and  $\sigma_v$  are separate sources, they determine not only the *x* and *y* director components outside the particle, Eq. ([7](#page-2-3)), but also two independent tensors (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.

It is known that the Coulomb term in the director expan-sion ([7](#page-2-3)) is fully determined by an external torque  $\Gamma$  applied on the particle  $[19]$  $[19]$  $[19]$ , i.e.,

$$
n_t(\mathbf{r}) = \frac{\left[\Gamma \times \mathbf{n}_{\infty}\right]_t}{4\pi Kr} + O\left(\frac{1}{r^2}\right). \tag{12}
$$

<span id="page-2-5"></span>Comparing  $(7)$  $(7)$  $(7)$  with  $(12)$  $(12)$  $(12)$ , we find the formula that expresses the two-component elastic charge via the external torque:

$$
q_t = \frac{\left[\Gamma \times \mathbf{n}_{\infty}\right]_t}{4\pi K}.\tag{13}
$$

<span id="page-2-6"></span>The charge components  $q_x$  and  $q_y$  can be induced only by the transverse torque components as  $(13)$  $(13)$  $(13)$  reads  $q_x$  $=\Gamma_y/(4\pi K)$ ,  $q_y = -\Gamma_x/(4\pi K)$ . Notice that multipoles of a higher order than  $q_t$  also depend on  $\Gamma$ , but, in contrast to the charge, they can be nonzero even for  $\Gamma = 0$ .

From the physical point of view, all we know about the source is either the director distribution in its close proximity, or the asymptotic far distortions induced by it in the homogeneous director field. The general problem of finding elastic multipole moments of a particle is therefore solved by Eqs.  $(8)$  $(8)$  $(8)$ – $(11)$  $(11)$  $(11)$ , in the first case, and by Eqs.  $(7)$  $(7)$  $(7)$  and  $(13)$  $(13)$  $(13)$ , in the second case. Moreover, in Sec. V we show that the multipoles are unambiguously determined by the single-particle director distribution  $n_t(s)$  [elastic charge density  $\sigma_t(s)$ ] unperturbed by other particles.

### **IV. COULOMB-LIKE INTERACTION OF TWO PARTICLES**

Now consider two particles enclosed by the spheres  $S_1$ and  $S_2$  with the centers at  $\mathbf{o}_1$  and  $\mathbf{o}_2$  and radii  $a_1$  and  $a_2$ , respectively (Fig. [2](#page-3-0)). The two-particle FE  $F_{1, \& 2}$  is obtained by substituting ([3](#page-2-1)) into ([2](#page-2-8)), which gives  $F_{1\&2} = F_1 + F_2 + F_{12}$ , where  $F_1$  and  $F_2$  are the self-energies of the particles 1 and 2, and  $F_{12}$  is the interaction energy, which is of the form

$$
F_{12} = \frac{K}{2} \int_{S_1} \int_{S_2} \sigma_t(\mathbf{s}_2) \sigma_t'(\mathbf{s}_1) U(\mathbf{s}_2, \mathbf{s}_1) d^2 s_2 d^2 s_1, \qquad (14)
$$

<span id="page-2-9"></span>where

<span id="page-3-0"></span>

FIG. 2. Two spheres enclosing particles 1 and 2.

<span id="page-3-4"></span>
$$
U(\mathbf{s}_2, \mathbf{s}_1) = -a_1 a_2 [(\boldsymbol{\nu}_2 \cdot \nabla_2)(\boldsymbol{\nu}_1 \cdot \nabla_1) G_2(\mathbf{s}_2, \mathbf{s}_1) + (\boldsymbol{\nu}_1 \cdot \nabla_1)
$$
  
×( $\boldsymbol{\nu}_2 \cdot \nabla_2$ ) G<sub>2</sub>( $\mathbf{s}_2$ ,  $\mathbf{s}_1$ )], (15)

and  $v_i$  is the outer normal to the surface  $S_i$  at  $s_i \in S_i$ . The potential *U* is expressed via the two-particle Green function  $G_2(\mathbf{r}, \mathbf{r}')$ , which satisfies the boundary condition  $G_2(\mathbf{r}, \mathbf{s})$  $=0$  for  $s \in S_1 \cup S_2$ . As it cannot be found exactly  $\left|25\right|$  $\left|25\right|$  $\left|25\right|$ , we developed a successive perturbation procedure to find  $G_2$  in the form of an expansion in a power series of the small parameter  $a/R$ , where  $\mathbf{R} = \mathbf{o}_1 - \mathbf{o}_2$  is the center-to-center separation vector (see the Appendix). In terms of the unit vector  $\mathbf{u} = \mathbf{u}_{12} = \mathbf{R}/R$ , this expansion of *U* takes the form

<span id="page-3-1"></span>
$$
\frac{2\pi}{a_1 a_2} U(\mathbf{s}_1, \mathbf{s}_2) = -\frac{1}{R} + \frac{a_1(\mathbf{v}_1 \cdot \mathbf{u}) - a_2(\mathbf{v}_2 \cdot \mathbf{u})}{R^2}
$$

$$
-\frac{a_1 a_2 [(\mathbf{v}_1 \cdot \mathbf{v}_2) - 3(\mathbf{v}_1 \cdot \mathbf{u})(\mathbf{v}_2 \cdot \mathbf{u})]}{R^3} + \cdots
$$
 (16)

Substituting  $(16)$  $(16)$  $(16)$  into  $(14)$  $(14)$  $(14)$  gives the interaction energy  $F_{12}$ . Using the definitions ([9](#page-2-10)) and ([10](#page-2-11)) of the elastic charge and dipole, the main-order terms can be reduced to the following form:

<span id="page-3-2"></span>
$$
F_{12} = -\frac{\Gamma_t^{(1)} \cdot \Gamma_t^{(2)}}{4\pi KR} + \frac{\left[\Gamma^{(2)} \times \mathbf{n}_{\infty}\right]_t(\mathbf{d}_t^{(1)} \cdot \mathbf{u}) - \left[\Gamma^{(1)} \times \mathbf{n}_{\infty}\right]_t(\mathbf{d}_t^{(2)} \cdot \mathbf{u})}{R^2} - 4\pi K \frac{(\mathbf{d}_t^{(1)} \cdot \mathbf{d}_t^{(2)}) - 3(\mathbf{d}_t^{(1)} \cdot \mathbf{u})(\mathbf{d}_t^{(2)} \cdot \mathbf{u})}{R^3} + \cdots, (17)
$$

where we replaced expression  $(9)$  $(9)$  $(9)$  for the elastic charge  $q_t$  in terms of the surface director by its expression  $(13)$  $(13)$  $(13)$  in terms of the transverse external torque. The first term in  $(17)$  $(17)$  $(17)$  is the Coulomb-like interaction. The importance of the formula ([9](#page-2-10)) for the two-component elastic charge  $q_t$  is that, owing to it, the Coulomb term  $-4\pi K q_t^{(1)} q_t^{(2)}/R$  is expressed solely via the transverse components  $\Gamma_{\perp} = \Gamma - (\Gamma \cdot \mathbf{n}_{\infty})$  of the torques exerted upon the particles:

$$
F_{\text{Coulomb}} = -\frac{(\Gamma_{\perp}^{(1)} \cdot \Gamma_{\perp}^{(2)})}{4\pi KR}.
$$
 (18)

<span id="page-3-3"></span>This formula shows that the Coulomb attraction, considered in  $[2]$  $[2]$  $[2]$ , can be induced only by more or less parallel torques, while opposite torques induce Coulomb repulsion,

the inversion geometry being  $\Gamma_{\perp}^{(1)} \perp \Gamma_{\perp}^{(2)}$ . The Coulomb term does not directly depend on details of the colloids' shape and surface anchoring (see the discussion below, in Sec. V). We see that breaking the horizontal and vertical mirror symmetry of the director field induced by asymmetrical particles is only a necessary but not a sufficient condition for the Coulomb interaction: irrespective of the symmetry, the 1/*R* term can be induced only by the external torques on colloidal particles. In particular, this interaction does not depend on the radii of the two spheres chosen at the intermediate stage of the derivation.

If one admits the interpretation of Eqs.  $(9)$  $(9)$  $(9)$  and  $(10)$  $(10)$  $(10)$  as the definition of a two-component elastic charge (monopole) and dipole, then the three terms in the elastic energy expansion  $F_{12}$  ([17](#page-3-2)) can be naturally associated with the elastic energies of Coulomb, monopole-dipole, and dipole-dipole interactions, respectively.

# **V. PAIRWISE APPROACH AND THE ELASTIC CHARGE DENSITY REPRESENTATION**

The FE expansion in a power series of the finite parameter  $(a/R)$ , obtained in this paper, allows one to explore the general status of the pairwise approach to nematic emulsions (the details can be found in  $[26]$  $[26]$  $[26]$ ). A system of *N* particles interacting via a pairwise potential has a finite number of degrees of freedom. In contrast, the nematic liquid crystal is a field system which, generally speaking, cannot be reduced to a finite number of degrees of freedom, and thus the pairwise approach to this system is approximate and has a certain range of applicability. Indeed, it is not difficult to see that, if  $N \geq 3$ , the two particles can interact via other particles which gives rise to an irreducible many-body interaction. For instance, the interaction of charged particles 1 and 2 via particle 3 gives rise to an irreducible three-particle correction  $F_{12,3}$  to the pair interaction ([14](#page-2-9)). This term is obtained from the three-particle Green function, which can be derived by the perturbation method developed in the Appendix, and has the form  $\lceil 26 \rceil$  $\lceil 26 \rceil$  $\lceil 26 \rceil$ 

$$
F_{12,3} = -\frac{a_3}{4\pi K} \frac{\Gamma_t^{(1)} \Gamma_t^{(2)}}{R_{13} R_{23}},\tag{19}
$$

where  $R_{13}$  and  $R_{23}$  are the corresponding distances and  $a_3$  is the radius of particle 3. We see that  $F_{12,3} \sim (a/R)F_{12}$ . This shows that the pairwise approximation to elastic charges is violated in the order next to the leading one in the parameter *a*/*R*. Thus, this parameter must be small and the interaction potential ([14](#page-2-9)) is meaningful only to leading order.

This drives us to the following important conclusion: the elastic charge ([9](#page-2-10)) of a particle can be calculated with the elastic charge density  $\sigma_t(s)$  ([8](#page-2-7)) in which the perturbations induced by other particles are neglected. Indeed, the perturbation induced by particle 2 at particle 1 is  $\delta \sigma_{1,2} \sim Cq_2 / R$ , where  $C < 1$  describes the elastic resistance of the director field at particle 1 to the external perturbation. At the same time, the unperturbed elastic charge density  $\sigma_1 \sim q_1/a_1$ . Therefore, the charge induced on particle 1 is  $\delta q_{1,2}$  $\leq (a_1/R)q_1$ , and the correction to the unperturbed Coulomb

term  $F_{12} \sim q_1 q_2 / R$  is  $\Delta F_{1,2} {\delta \sigma} \leq (a_1 / R) F_{12} \sim F_{12,3}$  (for brevity, here and in what follows the factor  $4\pi K$  in the energy estimates is omitted). This correction cannot be consistently incorporated in the pairwise approach and is small within its applicability range. Similarly, it can be shown that, in a system of elastic dipoles or quadrupoles, these multipole moments of each particle can be calculated from formulas  $(10)$  $(10)$  $(10)$  and  $(11)$  $(11)$  $(11)$  with the surface director distribution (charge density)  $\sigma_t(s)$ , unperturbed by other particles.

So far we have been considering the elastic energy of the outer zone outside the spheres. The energy  $F_{in}$  of the unperturbed inner zone of a particle has not been considered as it does not contribute to the interaction. But when the inner zone is perturbed by other particles,  $F_{in}$  acquires a dependence on their coordinates which can result in some interaction term  $\Delta F_{in}$ { $\delta \sigma$ }. It is not difficult to see that this  $\Delta F_{in}$  $\sim F_{12,3}$ . By definition of the equilibrium surface director distribution,  $\sigma_t(s)$  satisfies the equation  $\delta F_{in}/\delta \sigma_t = 0$ , so that  $\Delta F_{in}$ { $\delta \sigma$ } is quadratic in  $\delta \sigma$ . As  $F_{in} \sim q^2/a$ , one has  $\Delta F_{in}$  $\sim (\delta q)^2 / a \sim q^2 (a/R)^2 / a \sim (a/R) q^2 / R \sim (a/R) F_{12}$ . Thus, the interaction terms originating from the inner zones of particles can be neglected in the pairwise approach.

Now consider the requirement that the components  $n_t$ should be small at the auxiliary sphere *S* and derive the restriction it implies in the case of elastic charges. When  $n_t$  is large, the inaccuracy is related to neglecting the nonlinear (anharmonic) terms. The estimates for the nonlinear (anharmonic) corrections to the director field  $n_{an}$  and energy  $F_{an}$ were derived in Ref. [[9](#page-7-12)]. In the case of elastic charges,  $F_{an}$  is found to be  $\sim F_{12}(n)^2$  [[26](#page-7-16)] where  $\langle n \rangle$  is the average magnitude of the transverse director on the sphere with the radius  $a, \langle n \rangle \sim q/a$ ; see Eq. ([9](#page-2-10)). At the same time, the many-body correction  $F_{12,3} \sim (a/R)F_{12}$ . The anharmonic correction  $F_{an}$ can be neglected in the pairwise approach if  $F_{an} \le F_{12,3}$ which gives

$$
\langle n \rangle \lesssim \sqrt{a/R}.\tag{20}
$$

Of course, large  $n_t \sim 1$  on the sphere *S* that separates the inner and outer zones can result in an inaccuracy in the values of the multipole moments  $(9)$  $(9)$  $(9)$ – $(11)$  $(11)$  $(11)$ , but below we will see that this might be a rather weak restriction.

The elastic charge is determined solely by the external mechanical torque applied on a deformation source. However, in order to transfer this torque to the director, the source and director must be connected. If the source consists of, e.g., the defect pair radial hedgehog–hyperbolic hedgehog, this connection is provided by the elastic resistance to deviations of the vector connecting the two defects from the equilibrium direction along  $\mathbf{n}_{\infty}$  (see Ref. [[9](#page-7-12)]). If the deformation source is a particle, then the connection is provided by the surface anchoring. Nevertheless, the formula  $(18)$  $(18)$  $(18)$  implies that to establish the Coulomb interaction one needs considerably less information about the particle than is contained in its shape and surface anchoring (whose easy axis and strength depend on a point of the surface). This situation is similar to that in electrostatics: to describe the interaction of charged bodies at large separations, one needs to know just a few electric multipole moments rather than the exact charge distribution in the bodies. For instance, if the anchoring is very strong, the vector of a permanent dipole (electric or magnetic) or the polarizability tensor of the particle determines the torque exerted by a given (electric or magnetic) field, and no further information about the anchoring is needed. If the anchoring is not infinitely strong, then the field can change the orientation of the particle, and to find the torque one needs to know the angular restoring force constant  $k_{\theta}$  of the source [[9](#page-7-12)]. This discussion clearly shows that a deformation source in nematic emulsions has to be characterized by a few parameters such as  $k_{\theta}$ , the vectors of electric and magnetic dipole moments and polarizability tensors, elastic dipoles, and quadrupole tensors. As we showed above, all these quantities are characteristics of an individual unperturbed source which makes the pairwise description uniquely determined. In particular, the elastic dipole and quadrupole moments can be, in principle, obtained from the far field distortions the particle induces in a homogeneous director field.

Let us give some illustration of the elastic charge density representation developed in this paper. In general, the dipoledipole potential, given by the third term in Eq.  $(17)$  $(17)$  $(17)$ , depends on the angles made by  $\mathbf{d}_{x}^{(1)}$  and  $\mathbf{d}_{x}^{(2)}$ ,  $\mathbf{d}_{y}^{(1)}$  $y^{(1)}$  and  $\mathbf{d}^{(1)}_y$  $\chi^{(2)}$ , as well as by these vectors and the separation vector **u**. But if each particle is axially symmetric about the *z* axis (parallel to  $\mathbf{n}_{\infty}$ ), the potential depends solely on the angle  $\theta$ =arccos  $u_z$ ; in this case Eq. ([10](#page-2-11)) gives  $\mathbf{d}_x^{(i)} = (d_i, 0, 0), \mathbf{d}_y^{(i)}$  $\overline{u}^{(i)} = (0, d_i, 0)$ , and the potential simplifies to

$$
F_{dd} = \frac{4\pi K d_1 d_2}{R^3} (1 - 3\cos^2\theta). \tag{21}
$$

This particular case corresponds to the phenomenological dipole-dipole interaction considered in Ref.  $[9]$  $[9]$  $[9]$ .

Let us now illustrate our theory by a specific calculation. The authors of Ref.  $[9]$  $[9]$  $[9]$  described and considered the following complicated dipole system: a sphere imposing normal surface director alignment (imaginary radial hedgehog) accompanied by a hyperbolic hedgehog at distance  $r_d$  from the sphere's center **o**. If the sphere's radius is equal to 1, this  $r_d \approx 1.22$ . Using a unique ansatz method, the dipole moment *P* of the pair was found to be *P*=2.04. In our approach the dipole moment can be obtained from Eq.  $(10)$  $(10)$  $(10)$  by integrating the director distribution over the auxiliary sphere *S* enclosing the sphere-hedgehog pair. We thus need the director distribution in a close proximity of this pair. Such a director distribution was obtained by rotating the two-dimensional ansatz about the symmetry axis, Eq.  $(29)$  $(29)$  $(29)$  of Ref.  $[9]$ . Thus obtained function has an incorrect far asymptotics and cannot be used in the far zone. Moreover, it does not satisfies the nonlinear Euler-Lagrange equations for the 3D director in the near zone, but just gives a distribution qualitatively similar to the true one. Nevertheless, we expect that the dipole moment calculated from this ansatz should be close to its true value; the correct asymptotics is guaranteed by our Eq. ([7](#page-2-3)). We choose our auxiliary sphere *S* with the center at **o** and radius  $a > r_d$ . Notice that, while in Ref. [[9](#page-7-12)] the dipole moment was identified with the total prefactor of the  $1/r^2$  term in the far field, our dipole moment is just 1/3 of that in the expansion

([7](#page-2-3)), and thus our dipole moment should be compared with  $P/3 = 0.68$ . Equation ([10](#page-2-11)) with ansatz (2[9](#page-7-12)) of Ref. [9] gives the components of the dipole dyad in the form  $\mathbf{d}_x = (d,0,0)$ ,  $\mathbf{d}_y = (0, d, 0)$ . The numerical integration yields  $d = 0.66$  for *a*  $=1.7$  and  $d=0.54$  for  $a=1.5$  [of course, with ansatz (31) of Ref.  $[9]$  $[9]$  $[9]$ , which has the correct far asymptotics, and for sufficiently large  $a \ge 5$ , Eq. ([10](#page-2-11)) automatically gives  $d=0.68$ with a high accuracy. This shows that the goal of calculating the multipole moments from the director field at the source can be achieved reasonably well even if the director at the source is known only qualitatively. Moreover, although on the above sphere with  $a=1.7$  the maximum transverse director component is as large as  $n_t \approx 0.8$ , the calculated dipole moment 0.66 is quite close to its exact value 0.68. Therefore, the inaccuracy in the elastic dipole moment imposes practically a very weak restriction on the magnitude of  $n_t$  on the integration sphere *S*. Notice that Eq. ([10](#page-2-11)) has already proved very practical in Ref.  $[13]$  $[13]$  $[13]$  where it was used to estimate the dipole moment of liquid droplets trapped at a nematic-air interface.

#### **VI. CONCLUSION**

In this paper we developed an elastic charge density representation of the pairwise approach to the colloidal interaction via the nematic director field. The elastic multipole moments are expressed via the director distribution in the colloid, which plays the role of the elastic charge density. These formulas are practically convenient as they determine the elastic multipoles via the director distribution in the colloid's proximity. It is essential that the multipoles of a particle are its unambiguous individual characteristics, determined from the single-particle problem. In particular, we derived the general formula  $(18)$  $(18)$  $(18)$  which shows that the Coulomb-like interaction is determined by external torques applied on the colloids. This formula can greatly simplify and facilitate the theoretical description of nematic emulsions in external fields. Indeed, external fields exert mechanical torques on colloidal particles thus distorting the nematic director and resulting in the interparticle interaction. The results of this paper show that the very complicated problem of calculating the director distortions induced by the surfaces of specific colloids can be avoided: the field-induced interaction term can be immediately written down provided the fieldinduced torques are known. Thus, the problem of the fieldinduced interaction in nematic emulsions reduces to describing the distribution of torques, which in many cases is equivalent to the distribution of, e.g., the electric or magnetic dipoles of the colloids. The physics of nematic emulsions under the influence of external fields can be very interesting. An example of such a system, where the role of colloids is played by columnar nanosize aggregates, assembled from dye molecules in a nematic host, is considered in Refs.  $\lfloor 27, 28 \rfloor$  $\lfloor 27, 28 \rfloor$  $\lfloor 27, 28 \rfloor$ .

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# **APPENDIX: THE TWO-PARTICLE GREEN FUNCTION**

In contrast to the single-sphere case, the Green function for two spheres cannot be found in the form of a sum of the Coulomb source and a finite number of its pointlike images. Here we obtain the lowest-order terms of expansion of the Green function in a power series of the small radius-todistance ratio by considering a few successive images of the Coulomb source in two spheres.

Figure [2](#page-3-0) shows the two spheres  $S_1$  and  $S_2$  of radii  $a_1$  and  $a<sub>2</sub>$  representing two particles. The radius vectors of the points  $o_1, o_2, r, s_1, \ldots$  will be denoted by the corresponding bold symbols. The Green function  $G_2(\mathbf{r}, \mathbf{r}')$  for the Laplace equation, subject to the boundary condition  $G_2(\mathbf{r}, \mathbf{r}') = 0$  for any  $\mathbf{r}' \in S_1 \cup S_2$ , is of the form

$$
G_2(\mathbf{r}, \mathbf{r}') = (4\pi)^{-1} [J(\mathbf{r} - \mathbf{r}') + I(\mathbf{r}, \mathbf{r}')] , \tag{A1}
$$

where  $I(\mathbf{r}, \mathbf{r}')$  is a field of unknown "image" source that must compensate the field of the Coulomb source  $J=|\mathbf{r}-\mathbf{r}'|^{-1}$  on  $S_1 \cup S_2$  [[24](#page-7-14)]. The vectors **i**<sub>1</sub> and **i**<sub>2</sub> represent the first-order images of **r**, respectively, in  $S_1$  and  $S_2$ . As described after Eq. ([4](#page-2-12)), their dependence on the location **r** of the source and the *i*th sphere center  $\mathbf{o}_i$  is determined by the formula [[24](#page-7-14)[,25](#page-7-15)]

$$
\mathbf{i}_i(\mathbf{r}) - \mathbf{o}_i = \tilde{q}_i^2(\mathbf{r} - \mathbf{o}_i),
$$
 (A2)

<span id="page-5-1"></span><span id="page-5-0"></span>where

$$
\widetilde{q}_i = \frac{-a_i}{|\mathbf{r} - \mathbf{o}_i|}.
$$
 (A3)

The first-order image source  $I_i(\mathbf{r}, \mathbf{r}')$  is in the *i*th sphere and has the strength given by the ratio  $\tilde{q}_i$  ([A3](#page-5-0)), i.e.,

$$
I_i(\mathbf{r}, \mathbf{r}') = \frac{\tilde{q}_i}{|\mathbf{i}_i(\mathbf{r}) - \mathbf{r}'|}.
$$
 (A4)

The unique advantage of the specific geometry of a sphere is that the distance from the image at  $\mathbf{i}_i$  to any point  $\mathbf{r}'$  on the sphere  $S_i$  is proportional to the distance from this point to the source at **r**:

$$
|\mathbf{i}_{i} - \mathbf{r}'| = |\tilde{q}_{i}| |\mathbf{r} - \mathbf{r}'|, \quad \mathbf{r}' \in S_{i}.
$$
 (A5)

Owing to this relation the sum  $J(\mathbf{r}, \mathbf{r}') + I_i(\mathbf{r}, \mathbf{r}')$  vanishes on the sphere  $S_i$  and thus gives the exact single-particle Green function  $(4)$  $(4)$  $(4)$ .

The vectors  $\mathbf{i}_{12}$  and  $\mathbf{i}_{21}$  are the second-order images of the point **r**;  $\mathbf{i}_{12}$  is the image of the point  $\mathbf{i}_1$  in sphere  $S_2$ , and  $\mathbf{i}_{21}$  is the image of the point  $\mathbf{i}_2$  in sphere  $S_1$ . In the context of Eq. ([A2](#page-5-1)), it follows that the sources  $I_{12}$  and  $I_{21}$  of the secondary images  $\mathbf{i}_{12}$  and  $\mathbf{i}_{21}$  are of the following form:

$$
I_{12}(\mathbf{r}, \mathbf{r}') = \frac{\tilde{q}_1 \tilde{q}_{12}}{|\mathbf{i}_{12} - \mathbf{r}'|},
$$

$$
\tilde{q}_{12} = \frac{-a_2}{|\mathbf{i}_1 - \mathbf{o}_2|},
$$

$$
\mathbf{i}_{12} - \mathbf{o}_2 = \tilde{q}_{12}^2(\mathbf{i}_1 - \mathbf{o}_2),
$$
(A6)

and

$$
I_{21}(\mathbf{r}, \mathbf{r}') = \frac{\tilde{q}_2 \tilde{q}_{21}}{|\mathbf{i}_{21} - \mathbf{r}'|},
$$
  

$$
\tilde{q}_{21} = \frac{-a_1}{|\mathbf{i}_2 - \mathbf{o}_1|},
$$
  

$$
\mathbf{i}_{21} - \mathbf{o}_1 = \tilde{q}_{21}^2 (\mathbf{i}_2 - \mathbf{o}_1).
$$
 (A7)

The point  $\mathbf{i}_{121}$ , which is the image of  $\mathbf{i}_{12}$  in sphere 1, and the corresponding source  $I_{121}$  are determined by the following formulas:

$$
I_{121}(\mathbf{r}, \mathbf{r}') = \frac{\tilde{q}_1 \tilde{q}_{12} \tilde{q}_{121}}{|\mathbf{i}_{121} - \mathbf{r}'|},
$$
  

$$
\tilde{q}_{121} = \frac{-a_1}{|\mathbf{i}_{12} - \mathbf{o}_1|},
$$
  

$$
\mathbf{i}_{121} - \mathbf{o}_1 = \tilde{q}_{121}^2 (\mathbf{i}_{12} - \mathbf{o}_1).
$$
 (A8)

Consider the case when the arguments of the Green function belong to different spheres. Let  $s_1 \in S_1$  and  $s_2 \in S_2$ ; the point **r** is very close to a point  $s_2$  on  $S_2$ ,  $r \approx s_2$ , whereas **r**' lies on  $S_1$ ,  $\mathbf{r}' = \mathbf{s}_1$  (Fig. [2](#page-3-0)); the reason why we do not set **r**  $=$ **s**<sub>2</sub> will be clarified below. The sum  $I(\mathbf{r}, \mathbf{s}_1) = I_1 + I_2 + I_2$  $+I_{12}+I_{121}$  takes now the following explicit form:

<span id="page-6-0"></span>
$$
I(\mathbf{r}, \mathbf{s}_1) = -\frac{a_1}{|\mathbf{r} - \mathbf{o}_1|} \frac{1}{|\mathbf{i}_1 - \mathbf{s}_1|} - \frac{a_2}{|\mathbf{r} - \mathbf{o}_2|} \frac{1}{|\mathbf{i}_2 - \mathbf{s}_1|} + \frac{a_2 a_1}{|\mathbf{i}_2 - \mathbf{o}_1||\mathbf{r} - \mathbf{o}_2|} \frac{1}{|\mathbf{i}_{21} - \mathbf{s}_1|} + \frac{a_1 a_2}{|\mathbf{r} - \mathbf{o}_1||\mathbf{i}_1 - \mathbf{o}_2|} \frac{1}{|\mathbf{i}_{12} - \mathbf{s}_1|} - \frac{a_1 a_2}{|\mathbf{r} - \mathbf{o}_1||\mathbf{i}_1 - \mathbf{o}_2|} \frac{a_1}{|\mathbf{i}_{12} - \mathbf{o}_1|} \frac{1}{|\mathbf{i}_{12} - \mathbf{s}_1|}.
$$
 (A9)

Let us estimate the order of smallness of each term in this sum. To this end we notice (see Fig.  $2$ ) that **s**<sub>2</sub>-**s**<sub>1</sub>  $\sim$   $|\mathbf{o}_2 - \mathbf{o}_1|$   $\sim$   $|\mathbf{r} - \mathbf{o}_1|$   $\sim$   $|\mathbf{i}_2 - \mathbf{s}_1|$   $\sim$   $|\mathbf{i}_2 - \mathbf{o}_1|$   $\sim$   $|\mathbf{i}_1 - \mathbf{o}_2|$   $\sim$   $|\mathbf{i}_{12}$  $-$ **s**<sub>1</sub> $\vert \sim \vert \mathbf{i}_{12} - \mathbf{o}_1 \vert \sim R$ , whereas  $\vert \mathbf{i}_1 - \mathbf{s}_1 \vert \sim \vert \mathbf{r} - \mathbf{o}_2 \vert \sim \vert \mathbf{i}_{21} - \mathbf{s}_1 \vert$  $\sim$   $|\mathbf{i}_{121} - \mathbf{s}_1|$   $\sim a_1 \sim a_2 \sim a$ . It is now seen that  $I_1 \sim I_2 \sim I_{21}$  $\sim R^{-1}$  and  $I_{12} \sim I_{121} \sim (a/R)^2 R^{-1}$ . The reflection process can be continued, but the last terms  $I_{12}$  and  $I_{121}$  in  $(A9)$  $(A9)$  $(A9)$  and higher-order terms do not contribute to Eq.  $(14)$  $(14)$  $(14)$ , and we will restrict  $I(A9)$  $I(A9)$  $I(A9)$  to the first three terms. Then, up to terms  $\sim O[(a/R)^2]$ , the Green function  $G_2 = (4\pi)^{-1}(J+I_1+I_2+I_2)$ and can be written as

<span id="page-6-1"></span>
$$
4\pi G_2(\mathbf{r}, \mathbf{s}_1) = \left(\frac{1}{|\mathbf{r} - \mathbf{s}_1|} - \frac{a_1}{|\mathbf{r} - \mathbf{o}_1|} \frac{1}{|\mathbf{i}_1 - \mathbf{s}_1|}\right) - \frac{a_2}{|\mathbf{r} - \mathbf{o}_2|} \left(\frac{1}{|\mathbf{i}_2 - \mathbf{s}_1|} - \frac{a_1}{|\mathbf{i}_2 - \mathbf{o}_1|} \frac{1}{|\mathbf{i}_{21} - \mathbf{s}_1|}\right).
$$
(A10)

Equation  $(15)$  $(15)$  $(15)$  contains the two successive normal derivatives of Eq.  $(A9)$  $(A9)$  $(A9)$  on  $S_1$  and  $S_2$ . The normal derivative of  $G_2(\mathbf{r}, \mathbf{r}')$  at  $\mathbf{r}' = \mathbf{s}_1 \in S_1$  can be calculated similarly to the single-particle case as the expressions in the parentheses are merely  $G_1(\mathbf{r}, \mathbf{s}_1)$  and  $G_1(\mathbf{i}_2, \mathbf{s}_1)$  for particle  $S_1$ . One has

$$
4\pi[\nu(\mathbf{s}_1)\cdot\nabla_{\mathbf{s}_1}]G_2(\mathbf{r},\mathbf{s}_1) = \frac{a_1^2 - |\mathbf{r} - \mathbf{o}_1|^2}{a_1|\mathbf{r} - \mathbf{s}_1|^3} - \frac{a_2}{|\mathbf{r} - \mathbf{o}_2|} \frac{a_1^2 - |\mathbf{i}_2(\mathbf{r}) - \mathbf{o}_1|^2}{a_1|\mathbf{i}_2(\mathbf{r}) - \mathbf{s}_1|^3}.
$$
\n(A11)

To get the normal derivative of  $(A10)$  $(A10)$  $(A10)$  at  $\mathbf{r} = \mathbf{s}_2 \in S_2$  we first calculate it at  $\mathbf{r} \approx \mathbf{s}_2$ , taking into account the **r** dependence of all the image sources, and then pass to the limit  $\mathbf{r} \rightarrow \mathbf{s}_2$ ; the reason is that this passage does not commute with the **r** derivative. This gives

<span id="page-6-2"></span>
$$
4 \pi [\nu(\mathbf{s}_2) \cdot \nabla_{\mathbf{r}}] [\nu(\mathbf{s}_1) \cdot \nabla_{\mathbf{s}_1}] G_2(\mathbf{r}, \mathbf{s}_1)
$$
  
\n=
$$
\left[ [\nu(\mathbf{s}_2) \cdot \nabla_{\mathbf{r}}] \left( \frac{a_1^2 - |\mathbf{r} - \mathbf{o}_1|^2}{a_1 |\mathbf{r} - \mathbf{s}_1|^3} \right) - \frac{a_2}{|\mathbf{r} - \mathbf{o}_2|} [\nu(\mathbf{s}) \cdot \nabla_{\mathbf{r}}] \right]
$$
  
\n
$$
\times \left( \frac{a_1^2 - |\mathbf{i}_2(\mathbf{r}) - \mathbf{o}_1|^2}{a_1 |\mathbf{i}_2(\mathbf{r}) - \mathbf{s}_1|^3} \right) - \left( \frac{a_1^2 - |\mathbf{i}_2(\mathbf{r}) - \mathbf{o}_1|^2}{a_1 |\mathbf{i}_2(\mathbf{r}) - \mathbf{s}_1|^3} \right)
$$
  
\n
$$
\times [\nu(\mathbf{s}_2) \cdot \nabla_{\mathbf{r}}] \left( \frac{a_2}{|\mathbf{r} - \mathbf{o}_2|} \right). \tag{A12}
$$

At the limit **r** $\rightarrow$ **s**<sub>2</sub>, the vectors **i**<sub>2</sub>(**r**), **r**, and **s**<sub>2</sub> coincide, **r**−**o**<sub>2</sub> → *a*<sub>2</sub>, and the expression in square brackets tends to zero. Using that  $|s_2 - \mathbf{o}_2| = a_2$  and  $s_2 - \mathbf{o}_1 = \mathbf{R} - \nu(\mathbf{s})a_2$ , we obtain

$$
\lim_{\mathbf{r}\to\mathbf{s}_2} \left[ \nu(\mathbf{s}_2) \cdot \nabla_{\mathbf{r}} \right] \left[ \nu(\mathbf{s}_1) \cdot \nabla_{\mathbf{s}_1} \right] G_2(\mathbf{r}, \mathbf{s}_1)
$$
\n
$$
= -\frac{1}{4\pi a_1 a_2} \frac{a_1^2 - |\mathbf{R} - a_2 \nu(\mathbf{s}_2)|^2}{|\mathbf{s}_2 - \mathbf{s}_1|^3}.
$$
\n(A13)

The second term in Eq.  $(15)$  $(15)$  $(15)$  is obtained from Eq.  $(A12)$  $(A12)$  $(A12)$ by interchanging the indices 1 and 2 implying **R**→−**R**-, i.e.,

$$
\lim_{\mathbf{r}\to\mathbf{s}_1} [\nu(\mathbf{s}_1)\cdot\nabla_{\mathbf{r}}][\nu(\mathbf{s}_2)\cdot\nabla_{\mathbf{s}_2}]G_2(\mathbf{r},\mathbf{s}_2) =
$$

$$
-\frac{1}{4\pi a_1 a_2} \frac{a_2^2 - |\mathbf{R} + a_1 \nu(\mathbf{s}_1)|^2}{|\mathbf{s}_2 - \mathbf{s}_1|^3}.
$$
 (A14)

<span id="page-6-3"></span>Substituting both terms in Eq.  $(15)$  $(15)$  $(15)$  and taking into account that  $\mathbf{s}_2 - \mathbf{s}_1 = \mathbf{R} + a_1 \nu(\mathbf{s}_1) - a_2 \nu(\mathbf{s}_2)$ , yields

$$
U(\mathbf{s}_2, \mathbf{s}_1) = -\frac{a_1 a_2}{2\pi} \frac{\left[\mathbf{R} \cdot (\mathbf{s}_2 - \mathbf{s}_1)\right]}{|\mathbf{s}_2 - \mathbf{s}_1|^3}.
$$
 (A15)

Equation  $(16)$  $(16)$  $(16)$  is obtained by expanding  $(A15)$  $(A15)$  $(A15)$  in a power series of the inverse separation 1/*R*.

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