Multipole expansion for inclusions in a lamellar phase

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Inclusions in a bulk smectic phase distort the ordering of nearby layers. We argue that a multipole expansion for this distortion represents a powerful technique for understanding the linear interactions between inclusions with arbitrary boundary conditions. The fields for the first few higher-order moments are derived and some specific examples are discussed. Our results show the importance of the orientation and/or configuration of the inclusions. [S1063-651X(98)04201-9]

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I. INTRODUCTION

In recent years much progress has been made towards an understanding of the physical, chemical, and biological properties of fluid membranes [1–5]. Such systems are of interest to physicists due to their unique static and dynamic properties and also to biophysicists who see them as models of cell membranes. A cell membrane is primarily a lipid bilayer, albeit one with a wide spectrum of embedded proteins [3–5]. Many fundamental biological processes take place in these membranes and these are often controlled by one or more of the species of embedded proteins. Similar *layered* membrane phases in solution provide one of the most extensively studied examples of the smectic-*A* liquid crystal [6,7] and are exploited in numerous industrial contexts.

While much of the earlier studies of fluid membrane phases focused on essentially homogeneous systems there has been increasing recent attention given to heterogeneous systems. Many of these have sought to understand the membrane-mediated interactions between localized inclusions residing in or between one or two [8,9] layers. Although nonlinear studies have recently appeared [10] most of the theoretical studies have been at the level of a linearized theory. These usually involve minimizing the energy of deformation of the membrane(s) given specific boundary conditions for the membrane near the inclusion. The precise choice of boundary conditions depends on the system under consideration and this in turn strongly effects the results. For example, the interaction potential between inclusions has been shown to be highly sensitive to this choice, even at the level of whether it is attractive or repulsive [9].

Some studies of interactions between pointlike inclusions in bulk nematic [11] and smectic-*A* phases [12,13] also exist. It is the aim of the present work to extend this formalism to deal with more complex inclusions in the smectic phase using a multipole expansion. We argue that this represents a powerful technique as it allows us to extract the far-field interaction between inclusions with arbitrary boundary conditions to any order. This may often be preferable to direct, and rather tedious, numerical calculation with the usual drawback of lack of clear physical insight.

This paper is constructed as follows. In Sec. II we introduce and describe the Hamiltonian for a smectic liquid crystal with general inclusions present. This is then minimized and the results are employed in Sec. III where we introduce a multipole treatment for the inclusions. This gives both the two body interactions and distortion field to arbitrary order. In Sec. IV we discuss two specific examples, relevant for inclusions that induce a local curvature in the layers and linear rodlike inclusions [14]. After the conclusions there follow two Appendices that contain some cumbersome results that are nonetheless central to this paper.

II. GENERALIZED INCLUSIONS IN A BULK SMECTIC-*A* PHASE

In a bulk smectic-*A* phase the layers are stacked atop one another with average layer spacing d and the z direction conveniently defined by the average layer normal. Deformation of the layer away from its average position may conveniently be parametrized by the continuous scalar displacement field u, which represents the normal displacement of the layers in the z direction. Such a description leads to the so-called Landau–de Gennes Hamiltonian [1,2,15]. We model the effect of our inclusions in the smectic phase by including a term $\sim\psi\partial_z u$ in the energy density. The field $\psi(\mathbf{r})$, which can have either sign, can be chosen so as to mimic the effect of an arbitrary inclusion (or distribution of inclusions) as discussed below. This term represents the lowest-order coupling between the inclusions and the local layer compression (or expansion) $\partial_z u$. We reject the other acceptable term $\sim\psi\nabla_\parallel^2 u$, which does not preserve up-down symmetry for a point particle and which would require the introduction of additional Lagrange fields to ensure that all stresses are continuous across the plane $z=$const through the particle. It will turn out that *all* linear perturbations are most easily introduced via the term $\sim\psi\partial_z u$. Finally we omit all terms, including those scaling like $\sim\psi^2$, which correspond to direct interparticle interactions as we wish to study here only the membrane-mediated interparticle interactions. Thus we write the Hamiltonian $\mathcal{H}$ for our system as

$$\mathcal{H}=\frac{B}{2}\int d^3\mathbf{r}[(\partial_z u)^2+\lambda^2(\nabla_\parallel^2 u)^2+\psi\partial_z u]. \qquad (1)$$

In this expression $\nabla_{\parallel}$ is the gradient operator in the x-y plane, $\lambda=\sqrt{K/B}$ is a length characteristic of the smectic (typically of the order of the layer spacing d), and B and K are respectively the compressional and bending moduli of the smectic. The dimensionless field $\psi(\mathbf{r})$ controls the amplitude of the local deformation and depends on the microscopic details of the inclusion-layer interactions. For example, $\psi(\mathbf{r})\propto-\delta(\mathbf{r})$ corresponds to a pointlike inclusion that exerts a local outward push on the neighboring layers. Examples of other more complex inclusions will be discussed in Sec. IV below.

Equation (1) represents a linearized theory and includes the leading-order terms in an expansion in powers of derivatives of u. We require that the inclusions couple sufficiently weakly to the layers so as not to induce large deformation gradients. This expansion can be shown to break down only when $\psi\gtrsim 1$. In what follows we assume that we can safely neglect thermal fluctuations either due to negligible amplitude or because they can be integrated out, renormalizing K and B. Since the smectic-A exhibits quasi-long-range order our results are applicable only on length scales less than the layer orientation correlation length. However, since this varies exponentially with the layer rigidity (in k_BT units) it can be much larger than d [2]. Thus our treatment is appropriate for many thermotropic phases, such as the low temperature diblock copolymer lamellar phase, and may also be appropriate for certain lyotropic systems.

Minimization of Eq. (1) is most easily performed in Fourier space, as discussed in more detail elsewhere [13]. We find that the distortion field is given by

$$u(\mathbf{r})=\int d^3\mathbf{r}'\,G^{(u)}(\mathbf{r}-\mathbf{r}')\,\psi(\mathbf{r}'),\tag{2}$$

where the Green's function $G^{(u)}(\mathbf{r})$ is

$$G^{(u)}(\mathbf{r})=\frac{-1}{16\pi\lambda z}\,e^{-r_{\parallel}^2/4\lambda|z|}.\tag{3}$$

The energy follows by substitution

$$U=\int d^3\mathbf{r}'\int d^3\mathbf{r}\;G(\mathbf{r}-\mathbf{r}')\,\psi(\mathbf{r})\,\psi(\mathbf{r}'),\tag{4}$$

where

$$G(\mathbf{r})=\frac{B}{64\pi\lambda z^2}\left(1-\frac{r_{\parallel}^2}{4\lambda|z|}\right)\exp-\frac{r_{\parallel}^2}{4\lambda|z|}.\tag{5}$$

As mentioned above these results are not new [13] and were previously employed in the context of interactions between pointlike particles. In the present work the field ψ has a different interpretation and can be chosen to model the interactions between particles with arbitrary particle-layer boundary conditions. Equations (2)–(5) will be required in Sec. III below, which contains the main results of this paper.

III. MULTIPOLE EXPANSION

In this section we will present a multipole expansion for the interaction potential between generalized inclusions. We define the smectic potential field ϕ:

$$\phi(\mathbf{r})=\int d^3\mathbf{r}'\,G(\mathbf{r}-\mathbf{r}')\,\psi(\mathbf{r}').\tag{6}$$

This may be thought of as the energy cost of inserting a unit pointlike inclusion at $\mathbf{r}$ (one with unit ''charge,'' as discussed below).

First consider a distribution $\psi(\mathbf{r}')$ that is localized near the origin (say) and expand the kernel of Eq. (6) for $|\mathbf{r}'|\ll|\mathbf{r}|$ using

$$f(\mathbf{r}-\mathbf{r}')=f(\mathbf{r})-r_i'\nabla_i f(\mathbf{r})+\tfrac{1}{2}r_i' r_j'\nabla_i\nabla_j f(\mathbf{r})+\cdots,\tag{7}$$

where ∇_i denotes the gradient in the $\mathbf{r}_i$th direction. Thus the potential ϕ becomes

$$\phi(\mathbf{r})=G(\mathbf{r})\int\psi(\mathbf{r}')d^3\mathbf{r}'-\nabla_i G(\mathbf{r})\int r_i'\psi(\mathbf{r}')d^3\mathbf{r}'+\frac{1}{2}\nabla_i\nabla_j G(\mathbf{r})\int r_i' r_j'\psi(\mathbf{r}')d^3\mathbf{r}'+\cdots.\tag{8}$$

This expression is essentially the same as the corresponding expansion of the electrostatic potential far from some charge distribution [16]. We define quantities analogous to charge

$$Q=\int\psi\,d^3\mathbf{r}',\tag{9}$$

dipole moment

$$\mathbf{p}=\int\mathbf{r}'\psi\,d^3\mathbf{r}',\tag{10}$$

and quadrupole moment

$$D_{ij}=\int r_i' r_j'\psi\,d^3\mathbf{r}'.\tag{11}$$

However, in the present case $G(\mathbf{r})$ is not the Coulomb potential but is instead given by Eq. (5). The interaction potential $U(\mathbf{r})$ between two generalized inclusions separated by $\mathbf{r}$ follows by inserting a second particle at $\mathbf{r}$. Since the particles need not be identical this second particle, and its moments, are labeled with a tilde:

$$U(\mathbf{r})=\int\phi(\mathbf{r}+\mathbf{r}'')\tilde{\psi}(\mathbf{r}'')d^3\mathbf{r}''=G(\mathbf{r})Q\tilde{Q}+\nabla_i G(\mathbf{r})[Q\tilde{p}_i-\tilde{Q}p_i]-\nabla_i\nabla_j G(\mathbf{r})[p_i\tilde{p}_j-\tfrac{1}{2}(Q\tilde{D}_{ij}+\tilde{Q}D_{ij})]-\tfrac{1}{2}\nabla_i\nabla_j\nabla_k G(\mathbf{r})\times[p_i\tilde{D}_{jk}-\tilde{p}_i D_{jk}+\cdots]+\tfrac{1}{4}\nabla_i\nabla_j\nabla_k\nabla_l G(\mathbf{r})[D_{ij}\tilde{D}_{kl}+\cdots]+\cdots.\tag{12}$$

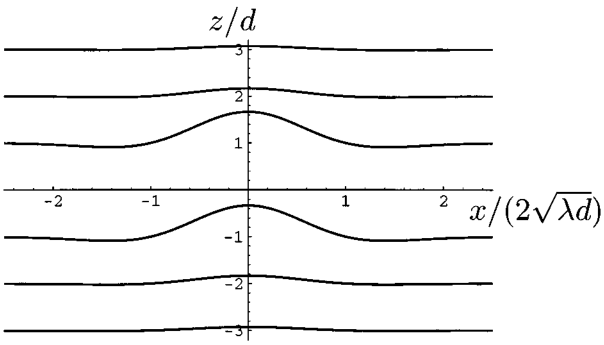

FIG. 1. The smectic deformation field induced by a point z-dipole inclusion at the origin (see text) is shown. A planar slice in the x-z plane is shown. The amplitude of the deformation is in arbitrary units while the z and x axes are in d and $2\sqrt{\lambda d}$ units, respectively.

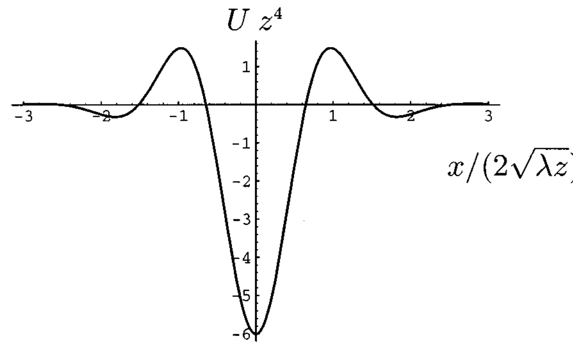

FIG. 2. The interaction potential in arbitrary units between two parallel point z dipoles as a function of their radial separation (along x) in units of $2\sqrt{\lambda z}$. The energy scales with their vertical displacement as $1/z^4$ and changes sign with each flip of either dipole.

The first two fields in the expansion ($\boldsymbol{\nabla} G$ and $-\boldsymbol{\nabla\nabla} G$) are given in Appendix A. We see from the results of this Appendix that the higher moments of the inclusions interact radially with essentially the same range $\simeq\sqrt{\lambda z}$. In addition the contribution to the potential from each higher moment has (i) an increasingly rich form with more turning points, corresponding to more metastable states, and (ii) an increasingly strong power law decay in the z direction.

The distortion field u

The distortion field u has already been shown to be related to the inclusion field ψ according to Eq. (2). Not only can we calculate $u(\psi)$ but, by inversion, also $\psi(u)$. This is a powerful feature, particularly if we wish to impose boundary conditions, e.g., on the shape of the distortion near the inclusion. An example of how such boundary conditions can be imposed is given in Sec. IV below [17].

Furthermore we can obtain the smectic distortion field around a generalized inclusion by a similar multipole expansion of Eq. (3). Using the expansion (7) we find that an inclusion at the origin induces a distortion field at $\mathbf{r}$ given by

$$u(\mathbf{r})=G^{(u)}(\mathbf{r})Q-\boldsymbol{\nabla} G^{(u)}(\mathbf{r})\cdot\mathbf{p}+\tfrac{1}{2}\nabla_i\nabla_j G^{(u)}(\mathbf{r})D_{ij}+\cdots. \tag{13}$$

Thus we are able to visualize the distortion field induced by each moment of ψ; see Fig. 1.

The multipole expansions discussed in this section converge for $z>d$. A discussion of the case when the particles reside in the same layer ($z\lesssim d$) can be found in Appendix B.

IV. SOME SPECIFIC EXAMPLES

We have already emphasized that the multipole expansion can greatly simplify the calculation of the interaction potential. In this section we will demonstrate this with two specific examples, a unit dipole in the z direction and a linear rodlike inclusion.

The smectic deformation field around a unit dipole in the z direction is given by $-\nabla_z G^{(u)}$ [see Eq. (13) and the results given in Appendix A]. It induces a local curvature in the layers without any net change in the spacing of the nearest layers, as shown in Fig. 1. Thus we have been able to deal with curvature-inducing inclusions without the direct coupling term $\sim\psi\nabla_\parallel^2 u$ discarded from Eq. (1). As mentioned in Sec. II this offers considerable simplification, with no additional Lagrange fields required. The ''trick'' of constructing a dipolar inclusion to induce curvature is, in a somewhat contrived way, similar to the use of the method of image charges in electrostatics.

Such dipolar inclusions are very similar to those considered elsewhere [9], where the magnitude of the angle that the layers make with the inclusion was fixed as a boundary condition. This comparison is easily understood when we realize that we can control the *maximum* slope of the neighboring layers induced by the dipole. The maximum slope is found a microscopic length $\simeq\sqrt{\lambda d}$ away from the dipole and is simply linear in the dipole moment p_z. The maximum slope of the layers at $z=\pm d$ (say) is

$$|\partial_x u|_{\substack{\max\\ z=\pm d}}=\frac{0.69 p_z}{16\pi\lambda^{3/2}d^{5/2}}. \tag{14}$$

Thus the fixed contact angle boundary condition crudely corresponds to a z dipole where the angle is fixed by the amplitude of the moment p_z. This identification delivers a considerable algebraic simplification.

We may also go on to ask about the interaction between two such dipoles. Their interaction is given by $-\nabla_z\nabla_z G$ [see Eq. (12) and the results given in Appendix A], which is plotted in Fig. 2. This potential is nonmonotonic and changes sign each time either one of the dipoles changes sign. Thus there are regions in space that are attractive for parallel dipoles but repulsive for antiparallel and vice versa. This may have interesting implications for dipoles that are not permanently anchored either up or down in the membrane.

Finally we mention briefly the interaction between rodlike inclusions that couple uniformly to the layer spacing along their length. Such rods have a net charge-charge interaction but this is insensitive to their relative orientations. Furthermore rods with indistinguishable ends have no dipole moment. Thus the leading-order orientationally dependant term enters at the quadrupolar order. The quadrupole moment of a simple rod is easily calculated: for a rod confined to the x-y plane that makes an angle θ with the x axis it is

$$D_{ij}=\beta\begin{pmatrix}\cos^2\theta & \sin\theta\cos\theta\\ \sin\theta\cos\theta & \sin^2\theta\end{pmatrix}, \tag{15}$$

where β is a coupling constant. For two such quadrupoles, making angles θ_1 and θ_2 with their line of separation (the x axis) the interaction potential U is obtained from Eq. (15) and $\nabla_i\nabla_j\nabla_k\nabla_l G$ (this last tensor is easily calculated but is too cumbersome to give in full). The energy of the two quadrupoles is found to be

$$\begin{aligned}U(\overline{x},\theta_1,\theta_2)=\frac{\beta^2}{2^5\lambda^2 d^4 z^4}\,[&12-36\overline{x^2}+18\overline{x^4}-2\overline{x^6}\\&-(24\overline{x^2}-19\overline{x^4}+2\overline{x^6})(\cos 2\theta_1+\cos 2\theta_2)\\&+(6-18\overline{x^2}+9\overline{x^4}-\overline{x^6})\cos 2(\theta_1-\theta_2)\\&+(5\overline{x^4}-\overline{x^6})\cos 2(\theta_1+\theta_2)],\end{aligned} \tag{16}$$

where $\overline{x^2}=x^2/(4\lambda|z|)$. Plots of the relative orientations $\{\theta_1,\theta_2\}$ that minimize this energy are given in Fig. 3.

V. CONCLUSIONS

The multipole analysis presented here represents a powerful technique for describing the distortion and interaction fields induced by general inclusions. Our analysis offers a considerable saving over direct calculations. We have demonstrated the 1-to-1 relationship between the inclusion field ψ and the smectic distortion field $u(\mathbf{r})$. Hence it is possible to choose ψ to mimic the effect of any inclusion. Thus our results are relevant for quite general inclusions. We have shown that certain features of simple inclusions can be described adequately by considering only a single higher moment of ψ.

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APPENDIX A: MULTIPOLAR FIELDS

Consider two inclusions separated by $\mathbf{r}$ and choose a coordinate system in which the x axis passes through the projection of the particles onto the x-y plane. We employ the notational simplification $\overline{x^2}=x^2/(4\lambda|z|)$ and $\overline{z}=|z|/d$ in which units we have the potential field between unit ''charges'' (5)

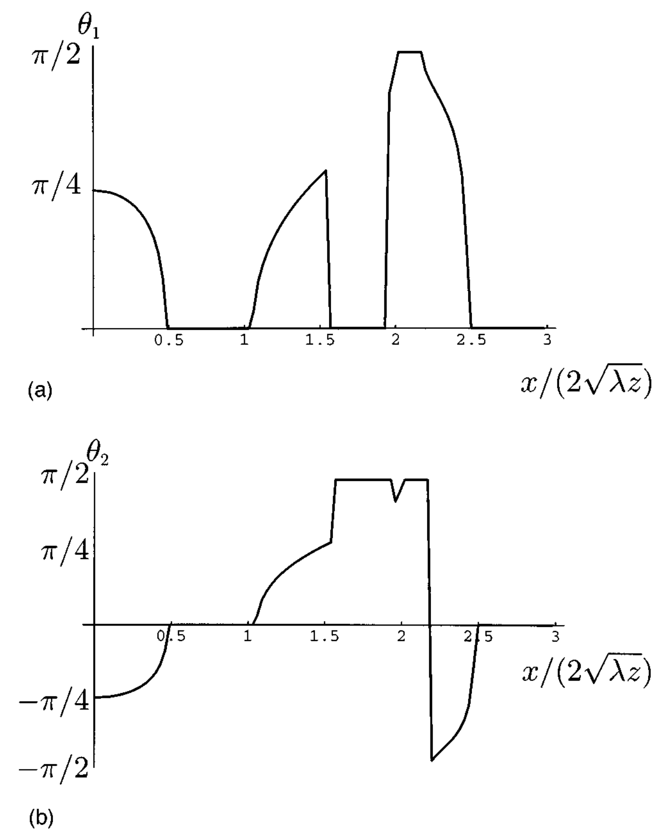

FIG. 3. These plots show the equilibrium angles (a) θ_1 and (b) θ_2 of two point quadrupole inclusions in a smectic phase. The angles correspond to those made by the projections of rodlike inclusions onto the x-y plane relative to a line of separation, chosen to be the x axis. We restrict θ_1 and θ_2 to lie on $[0,\pi/2]$ and $[-\pi/2,\pi/2]$ without loss of generality. These angles correspond to the equilibrium orientation of two rods for large enough separations.

$$G=\frac{B}{64\pi\lambda d^2}\,e^{-\overline{x^2}}(1-\overline{x^2})\overline{z}^{-2} \tag{A1}$$

between a unit charge (at the origin) and a unit dipole

$$\boldsymbol{\nabla}G=\frac{-Be^{-\overline{x^2}}}{64\pi\lambda d^3}\begin{pmatrix}\sqrt{\dfrac{d}{\lambda}}\,\overline{x}(2-\overline{x^2})\overline{z}^{-5/2}\\ 0\\ (2-4\overline{x^2}+\overline{x^4})\overline{z}^{-3}\end{pmatrix} \tag{A2}$$

and between unit dipoles

$$-\boldsymbol{\nabla}\boldsymbol{\nabla}G=\frac{-Be^{-\overline{x^2}}}{64\pi\lambda d^4}\begin{pmatrix}(d/\lambda)\left(-1+\dfrac{7}{2}\overline{x^2}-\overline{x^4}\right)\overline{z}^{-3} & 0 & \sqrt{d/\lambda}\,\overline{x}(6-6\overline{x^2}+\overline{x^4})\overline{z}^{-7/2}\\ 0 & (d/\lambda)\left(-1+\dfrac{1}{2}\overline{x^2}\right)\overline{z}^{-3} & 0\\ \sqrt{d/\lambda}\,\overline{x}(6-6\overline{x^2}+\overline{x^4})\overline{z}^{-7/2} & 0 & (6-18\overline{x^2}+9\overline{x^4}-\overline{x^6})\overline{z}^{-4}\end{pmatrix}. \tag{A3}$$

Any higher-order terms can also readily be calculated.

As discussed in Sec. III A above the smectic displacement field at **r** induced by an inclusion at the origin can also be written as a multipole expansion; see Eq. (13). The deformation field due to a unit charge is given by Eq. (3). This is circularly symmetric in the x-y plane and has odd symmetry in z:

$$G^{(u)}=\frac{-\operatorname{sgn}(z)}{16\pi\lambda d}\, e^{-\overline{r_\parallel^2}}\overline{z}^{-1}, \tag{A4}$$

where $\overline{r_\parallel^2}=r_\parallel^2/(4\lambda|z|)$ and the factor sgn(z)≡z/|z| preserves symmetry. The deformation field u around a unit dipole and a unit quadrupole is found to be

$$-\boldsymbol{\nabla} G^{(u)}=\frac{e^{-\overline{r_\parallel^2}}}{16\pi\lambda d^2}\begin{pmatrix}\operatorname{sgn}(z)\sqrt{d/\lambda}\,\overline{xz}^{-3/2}\\ \operatorname{sgn}(z)\sqrt{d/\lambda}\,\overline{yz}^{-3/2}\\ (1-\overline{r_\parallel^2})\overline{z}^{-2}\end{pmatrix} \tag{A5}$$

and

$$\frac{1}{2}\boldsymbol{\nabla}\boldsymbol{\nabla} G^{(u)}=\frac{e^{-\overline{r_\parallel^2}}}{32\pi\lambda d^3}\begin{pmatrix}\operatorname{sgn}(z)\,\frac{d}{\lambda}\left(-\frac{1}{2}+\overline{x^2}\right)\overline{z}^{-2} & \operatorname{sgn}(z)\,\frac{d}{\lambda}\,\overline{xyz}^{-2} & \sqrt{\frac{d}{\lambda}}\,\overline{x}(2-\overline{r_\parallel^2})\overline{z}^{-5/2}\\ \operatorname{sgn}(z)\,\frac{d}{\lambda}\,\overline{xyz}^{-2} & \operatorname{sgn}(z)\,\frac{d}{\lambda}\left(-\frac{1}{2}+\overline{y^2}\right)\overline{z}^{-2} & \sqrt{\frac{d}{\lambda}}\,\overline{y}(2-\overline{r_\parallel^2})\overline{z}^{-5/2}\\ \sqrt{d/\lambda}\,\overline{x}(2-\overline{r_\parallel^2})\overline{z}^{-5/2} & \sqrt{\frac{d}{\lambda}}\,\overline{y}(2-\overline{r_\parallel^2})\overline{z}^{-5/2} & \operatorname{sgn}(z)(2-4\overline{r_\parallel^2}+\overline{r_\parallel^4})\overline{z}^{-3}\end{pmatrix}, \tag{A6}$$

respectively. The symmetry of these results can be checked by carefully transforming $z\to -z$ throughout.

APPENDIX B: DISCUSSION AND REFINEMENTS IN THE ‘‘SHORT RANGE’’ LIMIT $z \lesssim d$

The singular nature of the Green's functions (17), (20) near $z=0$ may, at first sight, appear to be a cause for concern. In fact this behavior is not indicative of a fundamental flaw in the theory provided we adopt a physically motivated ‘‘smoothness criterion’’ for $\psi(\mathbf{r})$, which is discussed in more detail below. Alternatively we can simply argue that a lower cutoff at $z\simeq d$ is a natural consequence of the breakdown of the continuum theory at such length scales. Both approaches produce essentially the same qualitative result.

We can make the physical requirement of smoothness in ψ more explicit by replacing pointlike ‘‘sources’’ of ψ with sources which are Gaussian in $\mathbf{r}_\parallel$ according to

$$\psi_{\text{smooth}}(\mathbf{r})=\frac{1}{\pi b^2}\int \psi(\mathbf{r}')e^{-(r_\parallel-r_\parallel')^2/b^2}d^2\mathbf{r}_\parallel', \tag{B1}$$

where b is some microscopic length of the order of d and the factor $1/(\pi b^2)$ preserves the normalization. The smoothed function ψ_{smooth} can then be substituted into Eq. (6) [or Eq. (2)]. The resulting multipole expansions are identical to Eqs. (12) or (13) except that the variable z is everywhere replaced by z_{smooth} given by

$$z_{\text{smooth}}=z+\begin{cases} b^2/(4\lambda) & \text{for the distortion field } u\\ b^2/(2\lambda) & \text{for the interaction potential } U.\end{cases} \tag{B2}$$

This is an exact result. It is important in that it tells us that a microscopic cutoff in z arises naturally from the requirement that ψ be sufficiently smoothly varying.

While the above treatment reassures us that there is no physical flaw in our theory it is nonetheless still dangerous to attempt a quantitative extrapolation of our results to $z\simeq d$ since there are now no explicit guarantees of convergence for the series (6) or (13). However, we can say that the interaction between two particles in the same layer is short ranged and has a more complex form for higher order moments of ψ.

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[17] Note, however, that any boundary conditions we may wish to impose for u near the particle are strictly only satisfied for an isolated particle. As other particles approach the boundary values may deviate slightly from those at infinity. This is a result of the system minimizing the total distortion energy. In order to understand why this is happening it may help to think of ψ as a chemical potential field for the layer compression (expansion) $\partial_z u$. This field is chosen so as to fix the distortion correctly for infinite particle separation but the boundary values deviate in an elastic fashion as the particles approach from infinity. However, we believe that our description is adequate for large enough separations, in the spirit of the perturbation expansion Eq. (1).