Effect of orienting surfaces on the director fluctuations in nematic liquid crystals

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We consider the director fluctuations in nematic liquid crystals in a bounded volume, with allowance for the energy of cohesion to the surface. We consider in detail the cases of planarly and homotropically oriented cells. The correlation functions are calculated by a method that leads to a solution in closed form rather than to infinite series. We analyze the dependence of the director fluctuations on the dimensions of the system and of the coupling energy. The angular dependence of the intensity of the scattered light is calculated for normal incidence on an homotropically oriented cell.

1. INTRODUCTION

It is known that nematic liquid crystals (NLC) are, by virtue of their orientational ordering, optically anisotropic media. Since the preferred orientation (of the director) can be easily controlled with the aid of external fields, liquid crystals are used in various optical devices. Owing to the thermal motion, the direction vector fluctuates considerably in NLC. This, on the one hand, influences the characteristics of the apparatus, and on the other serves as a basis for investigating the properties of nematics by light-scattering methods.

Director fluctuations and light scattering from them have by now been thoroughly investigated for an infinite medium. It is known, in particular, that these fluctuations have a Goldston character, which leads, for example, to a divergence of the extinction coefficient in the Born approximation, and to a difference in the conditions of propagation and scattering of ordinary and extraordinary beams.

As a rule, the orientation of the liquid crystals is the result of their interaction with the substrate. It is therefore of considerable interest to investigate NLC in a confined volume. The prescence of boundaries influence substantially not only the director orientation in the entire volume,[1] but also alters the character of its fluctuations. This effect is the basis of one of the method of measuring the energy of the cohesion of the director to the substrate.[2]

We are faced thus with the problem of describing the director fluctuations in a bounded volume. This problem was considered in Refs. 2–4. The standard approach is to expand the fluctuations in natural modes. The solution takes the form of an infinite series, each term of which must be found by solving a complicated transcendental equation.

We propose here a closed solution of this problem for the usual conditions of the behavior of the director n on the boundary. We calculate the correlation function of the director fluctuations for homotropically and planarly oriented cells of the nematic. We analyze the dependence of the behavior of the director fluctuations on the layer thickness and on the constants indicative of the cohesion energy on the boundary. In the case of an homotropically oriented cell, we obtain an expression for the intensity of singly scattered light.

Let a liquid crystal be housed in a cell of thickness L between plane-parallel plates. We introduce a Cartesian coordinate frame with origin at the center of the cell and with a z axis normal to the plates.

To analyze the director fluctuations in an NLC of finite dimensions it is necessary to include in the description of the energy of interaction with the boundary of the medium. We shall assume that this interaction is taken into account by a Papini potential.[5,6] If the surfaces of the substrates between which the nematic is contained are isotropic, there is only one preferred direction—the normal to this surface. The director-substrate interaction energy per unit area is then given by $-\frac{1}{2}W\cos^2\theta$, where θ is the angle between the director and the normal (the z axis), and W is the cohesion energy. If, however, the substrate is not isotropic, and an easy orientation axis exists and is located in the xy plane (we assume henceforth that it is directed along the x axis), the energy takes the form $\frac{1}{2}(W_y n_y^2 + W_z n_z^2)$ (see also Ref. 2), where W_y and W_z are the cohesion energies and are generally speaking unequal.

The total free energy F can thus be represented by a sum of three terms

$$F=F_0+F_{bulk}+F_{surf}, \tag{1}$$

where F_0 is a contribution not connected with the director field, F_{bulk} is the Frank energy

$$F_{bulk}=\frac{1}{2}\int d^3r[K_{11}(\operatorname{div}\mathbf{n})^2+K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n})^2+K_{33}[\mathbf{n}\operatorname{rot}\mathbf{n}]^2], \tag{2}$$

K_{ii} are Frank moduli, and F_{surf} is the surface contribution. If the substrates are isotropic and the equilibrium field of the director $\mathbf{n}^0(\mathbf{r}) = \mathbf{e}_z$, then

$$F_{surf}=-\frac{W}{2}\int d^2\mathbf{r}_\perp[n_z^2(\mathbf{r}_\perp,L/2)+n_z^2(\mathbf{r}_\perp,-L/2)], \tag{3}$$

and if x is the easy-orientation axis, while the equilibrium field of the director $\mathbf{n}^0(\mathbf{r}) = \mathbf{e}_x$ is

$$F_{surf}=\frac{1}{2}\int d^2\mathbf{r}_\perp[W_y n_y^2(\mathbf{r}_\perp,L/2)+W_y n_y^2(\mathbf{r}_\perp,-L/2)+W_z n_z^2(\mathbf{r}_\perp,L/2)+W_z n_z^2(\mathbf{r}_\perp,-L/2)], \tag{4}$$

where we put $\mathbf{r} = (\mathbf{r}_\perp, z)$.

Let us consider the director fluctuations in the first case.

2. DIRECTOR FLUCTUATIONS IN AN HOMOTROPICALLY ORIENTED CELL

Assuming the deviation of the director $\mathbf{n}$ from the equilibrium value $\mathbf{n}^0 = \mathbf{e}_z$ to be small, we can put

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$\delta\mathbf{n}(\mathbf{r}) = \mathbf{n}(\mathbf{r}) - \mathbf{n}^0 = (n_x, n_y, 0)$. The free-energy change is defined in this case as

$$\Delta F = \frac{1}{2}\int d^3\mathbf{r}[K_{11}(\partial_x n_x(\mathbf{r})+\partial_y n_y(\mathbf{r}))^2 + K_{22}(\partial_x n_y(\mathbf{r})-\partial_y n_x(\mathbf{r}))^2 + K_{33}(\partial_z n_x(\mathbf{r}))^2+K_{33}(\partial_z n_y(\mathbf{r}))^2]+\frac{W}{2}\int d^2\mathbf{r}_\perp[n_x^2(\mathbf{r}_\perp, L/2) + n_x^2(\mathbf{r}_\perp, -L/2)+n_y^2(\mathbf{r}_\perp, L/2)+n_y^2(\mathbf{r}_\perp, -L/2)]. \quad (5)$$

It is convenient to represent the fluctuation $\delta\mathbf{n}(\mathbf{r})$ by a two-dimensional Fourier integral

$$\delta\mathbf{n}(\mathbf{r}) = \frac{1}{(2\pi)^2}\int d^2\varkappa \exp[i(\varkappa, \mathbf{r}_\perp)]\delta\mathbf{n}(\varkappa, z), \quad (6)$$

where $\varkappa$ is a wave vector located in the xy plane $\varkappa = (\varkappa\cos(\varphi), \varkappa\sin(\varphi), 0)$. For the change of the free energy (5) we obtain

$$\Delta F = \frac{1}{(2\pi)^2}\int d^2\varkappa\, \Delta F_\varkappa,$$

where

$$\Delta F_\varkappa = \frac{1}{2}\Big\{\int_{-L/2}^{L/2} dz[K_{11}|\varkappa_x n_x+\varkappa_y n_y|^2+K_{22}|\varkappa_x n_y-\varkappa_y n_x|^2 + K_{33}(|\partial_z n_x|^2+|\partial_z n_y|^2)]+W[|n_x(L/2)|^2+|n_x(-L/2)|^2 + |n_y(L/2)|^2+|n_y(-L/2)|^2]\Big\}. \quad (7)$$

We have left out of (7) the argument $\varkappa$ of the function $\delta\mathbf{n}(\varkappa, z)$.

The following procedure is used to find the correlation function in an infinite medium. Differentiating by parts in the equation for ΔF and neglecting the terms outside the integral sign we get an expression of type $\Delta F = (\delta\mathbf{n}, \hat{A}\delta\mathbf{n})/2$, where $\hat{A}$ is a certain differential operator, and the scalar product implies summation over the indices and integration over the continuous variables. The correlation function is then obtained in the form $\langle\delta\mathbf{n}\otimes\delta\mathbf{n}\rangle = k_B T\hat{A}^{-1}$. A feature of organic media is the ΔF contains terms outside the integral sign, which must be taken into account when the inverse of the operator $\hat{A}$ is taken. The problem reduces then to choosing for the functions $\delta\mathbf{n}(\varkappa, z)$ boundary conditions such that, first, ΔF is a quadratic form and, second, the corresponding operator $\hat{A}$ be self-adjoint.

In the present case these boundary conditions are

$$W\delta\mathbf{n}+K_{33}\partial_z\delta\mathbf{n}=0 \quad (8)$$

for $z = L/2$ and

$$W\delta\mathbf{n}-K_{33}\partial_z\delta\mathbf{n}=0 \quad (9)$$

for $z = -L/2$, and the expression for $\Delta F_\varkappa$ has a quadratic form

$$\Delta F_\varkappa = \frac{1}{2}\int_{-L/2}^{L/2} dz(n_x^*, n_y^*)\hat{A}\begin{pmatrix} n_x \\ n_y \end{pmatrix}, \quad (10)$$

where

$$\hat{A} = \begin{pmatrix} \varkappa_x^2 K_{11}+\varkappa_y^2 K_{22}-K_{33}\partial_z^2 & (K_{11}-K_{22})\varkappa_x\varkappa_y \\ (K_{11}-K_{22})\varkappa_x\varkappa_y & \varkappa_y^2 K_{11}+\varkappa_x^2 K_{22}-K_{33}\partial_z^2 \end{pmatrix}. \quad (11)$$

Thus, the boundary conditions (8) and (9) obviate the need for the terms outside the integral sign in the expression for the change of the free energy. Since the operator $\hat{A}$ is symmetric and its adjoint $\hat{A}^*$ is defined on the same class of function, it follows that $\hat{A}$ is a self-adjoint operator.[8] Our aim is to determine the correlation function of the director fluctuations $\hat{G}_h(\varkappa, z, z')$:

$$\hat{G}_h(\varkappa, z, z') = \int d^2\mathbf{r}_\perp \exp[-i(\varkappa, \mathbf{r}_\perp)]\langle\delta\mathbf{n}(\mathbf{r}_\perp, z)\otimes\delta\mathbf{n}(0, z')\rangle, \quad (12)$$

which should satisfy the equation

$$\hat{A}\hat{G}_h = k_B T\hat{I}\delta(z-z'), \quad (13)$$

where $\hat{I}$ is a unit matrix, i.e., it is necessary to invert the operator $\hat{A}$ with account taken of the boundary conditions (8) and (9).

Note that the matrix $\hat{A}$ reduces to diagonal by the transformation $\hat{R}^{-1}\hat{A}\hat{R}$, where $\hat{R}$ is the matrix of rotation through an angle φ in the xy plane

$$\hat{R} = \begin{pmatrix} \cos\varphi & -\sin\varphi \\ \sin\varphi & \cos\varphi \end{pmatrix}. \quad (14)$$

Equation (13) for the correlation matrix $\hat{G}_h$ breaks up in the new coordinate frame $\hat{G}' = \hat{R}^{-1}\hat{G}_h\hat{R}$ into two independent equations for the diagonal elements of the matrix $\hat{G}'$

$$(K_{ii}\varkappa^2-K_{33}\partial_z^2)\hat{G}_{ii}'(\varkappa, z, z')=k_B T\delta(z-z'), \quad i=1, 2. \quad (15)$$

On the boundary of the medium ($z = \pm L/2$) the elements of the matrix $\hat{G}'$ can, according to (8) and (9) satisfy the equations

$$WG_{ii}'(\varkappa, \pm L/2, z')\pm K_{33}\partial_z G_{ii}'(\varkappa, \pm L/2, z')=0. \quad (16)$$

The solution of Eq. (15) with allowance for the boundary conditions (16) can be represented in the form[9]

$$\hat{G}'(\varkappa, z, z') = \frac{k_B T}{K_{33}(u_+^i\partial_z u_-^i - u_-^i\partial_z u_+^i)}\begin{cases} u_+^i(z)u_-^i(z'), & z>z' \\ u_-^i(z)u_+^i(z'), & z<z' \end{cases}, \quad (17)$$

$$G_{12}'=G_{21}'=0,$$

where functions $u_\pm^i(z)$ are solutions of the corresponding homogeneous equations

$$(\varkappa^2 K_{ii}-K_{33}\partial_z^2)u_\pm^i(z)=0 \quad (18)$$

and satisfy the boundary conditions

$$Wu_\pm^i(\pm L/2)\pm K_{33}\partial_z u_\pm^i(\pm L/2)=0, \quad (19)$$

with the Wronskian of these functions independent of z, since Eqs. (18) do not contain first derivatives with respect to z. The solutions of Eqs. (18) with allowance for the conditions (19) are easy to find. For $u_\pm^i$ one can choose the functions

$$u_+^i(z)=(\beta_i+w)\exp(-\beta_i z-L/2)+(\beta_i-w)\exp(\beta_i z-L/2), \quad (20)$$

$$u_-^i(z)=(\beta_i-w)\exp(-\beta_i z+L/2)+(\beta_i+w)\exp(\beta_i z+L/2), \quad (21)$$

where the notation $\beta_i = \varkappa(K_{ii}/K_{33})^{1/2}$ is used, and $w = W/K_{33}$ is the reciprocal length indicative of the cohesion of the nematic with the substrate.[6]

Substituting expressions (20) and (21) in Eq. (17) for the correlation-matrix elements, we get

$$G_{ii}'(\varkappa,z,z')=\frac{k_BT}{2\beta_iK_{33}\Delta_i}\times\{(\beta_i^2-w^2)\mathrm{ch}(\beta_i(z+z'))+[(\beta_i^2+w^2)\mathrm{ch}(\beta_iL)+2\beta_iw\,\mathrm{sh}(\beta_iL)]\mathrm{ch}(\beta_i(z-z'))-\Delta_i\,\mathrm{sh}(\beta_i|z-z'|)\},\tag{22}$$

where

$$\Delta_i=(\beta_i^2+w^2)\,\mathrm{sh}(\beta_iL)+2w\beta_i\,\mathrm{ch}(\beta_iL).$$

The initial matrix $\hat G_h$ can be obtained by the inverse transformation $\hat G_h=\hat R\hat G'R^{-1}$. We have thus ultimately for the correlation functions

$$\begin{gathered}\langle n_x(\varkappa,z)n_x^*(\varkappa,z')\rangle=\cos^2\varphi G_{11}'(\varkappa,z,z')+\sin^2\varphi G_{22}'(\varkappa,z,z'),\\ \langle n_y(\varkappa,z)n_y^*(\varkappa,z')\rangle=\cos^2\varphi G_{22}'(\varkappa,z,z')+\sin^2\varphi G_{11}'(\varkappa,z,z'),\\ \langle n_x(\varkappa,z)n_y^*(\varkappa,z')\rangle=\langle n_y(\varkappa,z)n_x^*(\varkappa,z')\rangle\\ =\cos\varphi\sin\varphi[G_{11}'(\varkappa,z,z')-G_{22}'(\varkappa,z,z')].\end{gathered}\tag{23}$$

Note that the series obtained in Refs. 2, 3, and 4 for the correlation function of the director fluctuations coincide with the expansions of the meromorphic functions $G'_{ii}(\varkappa,z,z')$ of argument $\varkappa$ into simple fractions. For example, at $\varphi=0$, $z=z'$, $w=\infty$ (stringent boundary conditions) and for identical Frank moduli $K_{ii}=K$ we have for the fluctuations of n_x according to (22) and (23)

$$\langle n_x(\varkappa,z)n_x^*(\varkappa,z)\rangle=\frac{k_BT}{2\varkappa K\,\mathrm{sh}(\varkappa L)}\{\mathrm{ch}(\varkappa L)-\mathrm{ch}(2\varkappa z)\}.\tag{24}$$

This function has poles at $\varkappa=in\pi/L$ (n is the first nonzero number), and its principal parts at these points are equal to

$$\frac{\cos(\pi n)-\cos(2\pi nz/L)}{2i\pi n\cos(\pi n)}\,\frac{1}{\varkappa-i\pi n/L}\,\frac{k_BT}{K}.$$

Summation over n leads to the expression

$$\langle n_x(\varkappa,z)n_x^*(\varkappa,z')\rangle=\frac{k_BT}{LK}\sum_{n>0}\frac{1+(-1)^{n+1}\cos(2\pi nz/L)}{\varkappa^2+\pi^2n^2/L^2},$$

which coincides with the results of Ref. 4.

It is known that the director fluctuations in an infinite nematic are of the Goldston type ($\langle\delta n_{\mathbf q}\otimes\delta n_{-\mathbf q}\rangle\sim q^{-2}$, where $\mathbf q$ is a three-dimensional wave vector). They make the rms director fluctuation at a point infinite on account of the contribution of fluctuations with small q. The nonzero adhesion energy on the boundary of the medium causes the director fluctuations at any point of the volume to become finite. Indeed, eliminating the uncertainty $\varkappa=0$ in Eq. (22), we obtain

$$G_{ii}'(0,z,z')=\frac{1+w(L-|z-z'|)+w^2((L-|z-z'|)^2-(z+z')^2)/4}{w(Lw+2)}\,\frac{k_BT}{K_{33}},\tag{25}$$

i.e., the integral $\int d^2\varkappa\hat G_h(\varkappa,z,z')$ does not diverge in the region $\varkappa=0$. It diverges only at large $\varkappa$, but values of $\varkappa^{-1}$ are smaller than the intermolecular distance have no physical meaning,[3] therefore the integral must be cut off at a certain value k_m. A divergence in the region $\varkappa=0$ appears only when the adhesion energy W tends to zero, with $\langle n_{x,y}(0,z)n_{x,y}(0,z)\rangle\sim k_BT/(2W)$. Thus, in the absence of adhesion to the substrate the director fluctuations are of the Goldston type not only in an infinite medium but also in thin films.

3. DIRECTOR FLUCTUATIONS IN A PLANARLY ORIENTED CELL

We shall assume that the director $\mathbf n^0$ is directed along the x axis, while the z axis is perpendicular, as before, to the nematic boundary. A small deviation $\delta\mathbf n$ of the director from equilibrium has no component along the x axis, i.e., $\delta\mathbf n=(0,n_y,n_z)$. The surface contribution to the free energy change, a contribution connected with the fluctuation δn, is given by Eq. (4). The bulk contribution to this change takes for such a geometry the form

$$\Delta F_{bulk}=\frac12\int d^3\mathbf r\{K_{11}(\partial_yn_y+\partial_zn_z)^2+K_{22}(\partial_yn_z-\partial_zn_y)^2+K_{33}[(\partial_xn_z)^2+(\partial_xn_y)^2]\}.\tag{26}$$

To avoid cumbersome calculations, we confine ourselves in this case to the single-constant approximation ($K_{ii}=K$).

Just as in the case of a homotropically oriented cell, we can take Fourier transforms with respect to the coordinates x and y. We then obtain for the contribution of $\Delta F_\varkappa$ to ΔF due to the $\delta\mathbf n(\varkappa,z)$ fluctuations, taking (4) into account

$$\Delta F_\varkappa=\frac12K\int_{-L/2}^{L/2}dz[\varkappa^2(|n_y|^2+|n_z|^2)+|\partial_zn_z|^2+|\partial_zn_y|^2+i\varkappa\partial_z(n_z^*n_y-n_y^*n_z)]+\frac12W_z[|n_z(L/2)|^2+|n_z(-L/2)|^2]+\frac12W_y[|n_y(L/2)|^2+|n_y(-L/2)|^2].\tag{27}$$

We omit here, just as in Sec. 2, the argument $\varkappa$ in $\mathbf n(\varkappa,z)$. The boundary conditions that make it possible in this case to reduce the problem to inversion of a self-adjoint operator, are, as follows from (27),

$$\begin{gathered}K[\partial_zn_z(\pm L/2)+i\varkappa_yn_y(\pm L/2)]\pm W_zn_z(\pm L/2)=0,\\ K[\partial_zn_y(\pm L/2)-i\varkappa_yn_z(\pm L/2)]\pm W_yn_y(\pm L/2)=0,\end{gathered}\tag{28}$$

and the corresponding expression for $\Delta F_\varkappa$ takes the form

$$\Delta F_\varkappa=\frac12\int_{-L/2}^{L/2}dz(n_y^*(z),n_z^*(z))\hat B\begin{pmatrix}n_y(z)\\ n_z(z)\end{pmatrix},\tag{29}$$

where

$$\hat B=K(\varkappa^2-\partial_z^2)\hat I.\tag{30}$$

Putting

$$\hat G(\varkappa,z,z')=\begin{pmatrix}\langle n_y(\varkappa,z)n_y^*(\varkappa,z')\rangle & \langle n_y(\varkappa,z)n_z^*(\varkappa,z')\rangle\\ \langle n_z(\varkappa,z)n_y^*(\varkappa,z')\rangle & \langle n_z(\varkappa,z)n_z^*(\varkappa,z')\rangle\end{pmatrix},\tag{31}$$

we obtain the equation

$$\hat B\hat G_p(\varkappa,z,z')=k_BT\hat I\delta(z-z'),\tag{32}$$

which must be solved subject to the boundary conditions

$$\begin{pmatrix}K\partial_z-i\varkappa_y & \sigma W_z\\ \sigma W_y & K\partial_z+i\varkappa_y\end{pmatrix}\hat G_p(\varkappa,\sigma L/2,z')=0,\tag{33}$$

where $\sigma=\pm1$. The values $\sigma=1$ and $\sigma=-1$ correspond respectively to the boundary conditions at $z = L/2$ and $z = -L/2$.

Although the matrix $\hat{B}$ is diagonal, the matrix of the correlators $\hat{G}_p$ is not, for in contrast to the considered homotropic orientation the boundary conditions constitute a system of two equations. To find the solution in this case, let us consider a rectangular 2×4 matrix $\hat{V}$

$$\hat{V} = \begin{pmatrix} \hat{G}_p \\ \partial_z \hat{G}_p \end{pmatrix}. \tag{34}$$

This change makes is possible to reduce the problem to a first-order differential equation

$$\partial_z \hat{V}(z, z') - \hat{M}\hat{V}(z, z') = -\hat{D}\delta(z - z'), \tag{35}$$

where

$$\hat{M} = \begin{pmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ \varkappa^2 & 0 & 0 & 0 \\ 0 & \varkappa^2 & 0 & 0 \end{pmatrix}, \qquad \hat{D} = \frac{k_B T}{K}\begin{pmatrix} 0 & 0 \\ 0 & 0 \\ 1 & 0 \\ 0 & 1 \end{pmatrix}.$$

The boundary conditions have in this notation the form $\hat{\Gamma}_\sigma \hat{V}(\sigma L/2, z') = 0$, where

$$\hat{\Gamma}_\sigma = \begin{pmatrix} \sigma w_y & -i\varkappa_y & 1 & 0 \\ i\varkappa_y & \sigma w_y & 0 & 1 \end{pmatrix}, \quad w_y = \frac{W_y}{K}.$$

The general solution o the homogeneous equation corresponding to (35) is $\exp(z\hat{M})\hat{C}$, where $\hat{C}$ is an arbitrary matrix of dimension 2×4. We denote by $\hat{V}_+$ the solution for $z > z'$, and by $\hat{V}_-$ the solution for $z < z'$. This can be represented in the form

$$\hat{V}_\pm(z, z') = \exp(z\hat{M})\hat{C}_\pm. \tag{36}$$

To obtain a δ-function in the right-hand side of (35) we must choose matrices $\hat{C}_\pm(z')$ such as to satisfy the relation

$$\exp(z'\hat{M})[\hat{C}_+(z') - \hat{C}_-(z')] = 0. \tag{37}$$

This equation, however, is insufficient to determine the 16 elements of the matrices $\hat{C}_\pm$. Equation (37) must be solved jointly with the equations that ensure satisfaction the boundary conditions $\hat{\Gamma}_\sigma \exp[(L/2)\hat{M}]\hat{C}_\sigma(z') = 0$.

Thus, the determination of the matrix of the correlators $\hat{G}_p(\varkappa, z, z')$ reduces to a solution of a system of 16 linear equations. After solving these equations, we can determine $\hat{V}_\pm(z, z')$ with the aid of (36), and consequently also the correlation matrix $\hat{G}_p(\varkappa, z, z')$ in the entire range of variation of the agruments z and z'. As a result we get (see the Appendix)

$$\begin{aligned} G_{p11}(\varkappa, z, z') = {} & \frac{k_B T}{2\varkappa[\varkappa^2(w_y - w_z)^2 - \Delta^2]K}\{\operatorname{sh}(\varkappa|z - z'|) \\ & \times[\Delta^2 - \varkappa^2(w_y - w_z)^2] + \operatorname{ch}(\varkappa(z + z')) \\ & \times[\Delta(w_y w_z - \varkappa^2 - \varkappa_y^2) - \varkappa(w_y - w_z)(w_y w_z + \varkappa_x^2)\operatorname{ch}(\varkappa L) \\ & + \varkappa^2(w_y^2 - w_z^2)\operatorname{sh}(\varkappa L)] + \operatorname{ch}(\varkappa(z - z')) \\ & \times[\varkappa(w_y - w_z)(\varkappa^2 + \varkappa_y^2 - w_y w_z) \\ & - \Delta(w_y w_z + \varkappa_x^2)\operatorname{ch}(\varkappa L) - \varkappa\Delta(w_y w_z + \varkappa_x^2)\operatorname{sh}(\varkappa L)]\}, \end{aligned} \tag{38}$$

$$\begin{aligned} & G_{p22}(\varkappa, z, z') \\ & = \frac{k_B T}{2\varkappa[\varkappa^2(w_y - w_z)^2 - \Delta^2]K}\{\operatorname{sh}(\varkappa|z - z'|)[\Delta^2 - \varkappa^2(w_y - w_z)^2] \\ & + \operatorname{ch}(\varkappa(z + z'))[\Delta(w_y w_z - \varkappa^2 - \varkappa_y^2) \\ & - \varkappa(w_y - w_z)(w_y w_z + \varkappa_x^2)\operatorname{ch}(\varkappa L) \\ & - \varkappa^2(w_y^2 - w_z^2)\operatorname{sh}(\varkappa L)] + \operatorname{ch}(\varkappa(z - z')) \\ & \times[\varkappa(w_z - w_y)(\varkappa^2 + \varkappa_y^2 - w_y w_z) \\ & - \Delta(w_y w_z + \varkappa_x^2)\operatorname{ch}(\varkappa L) - \varkappa\Delta(w_y w_z + \varkappa_x^2)\operatorname{sh}(\varkappa L)]\}, \end{aligned} \tag{39}$$

$$\begin{aligned} G_{p12}(\varkappa, z, z') = {} & \frac{-i\varkappa_y k_B T}{[\varkappa^2(w_y - w_z)^2 - \Delta^2]K} \\ & \times[\varkappa(w_y - w_z)\operatorname{sh}(\varkappa(z - z')) + \Delta\operatorname{sh}(\varkappa(z + z'))], \end{aligned} \tag{40}$$

$$G_{p21}(\varkappa, z, z') = G^*_{p12}(\varkappa, z', z),$$

where

$$\Delta = (w_y w_z + \varkappa_x^2)\operatorname{sh}(\varkappa L) + \varkappa(w_y + w_z)\operatorname{ch}(\varkappa L).$$

Equations (38)–(40) go over at very large thicknesses L into the usual correlation function of the director fluctuations in the $(\varkappa, z, z')$ representation for an infinite medium

$$\begin{aligned} \langle n_y(\varkappa, z) n_y^*(\varkappa, z')\rangle &= \langle n_z(\varkappa, z) n_z^*(\varkappa, z')\rangle \\ &= \frac{k_B T}{2\varkappa K}\exp[-\varkappa|z - z'|], \\ \langle n_y(\varkappa, z) n_z^*(\varkappa, z')\rangle &= \langle n_z(\varkappa, z) n_y^*(\varkappa, z')\rangle = 0. \end{aligned} \tag{41}$$

If at least one of the energies W_y or W_z differs from zero, the correlation functions have no singularities at $\varkappa = 0$ (there are no Goldston fluctuations). Expressions (38)–(40) admit also of various limiting transitions $W_{x,y} \to 0$, $W_{x,y} \to \infty$. In particular, if the director can rotate freely in the xy plane on surfaces that border on the substrate, but a deviation from this surface entails very large energy losses ($W_y = 0$, $W_z = \infty$), we obtain

$$\begin{aligned} & \langle n_y(\varkappa, z) n_y^*(\varkappa, z')\rangle \\ & = \frac{k_B T}{2\varkappa K\operatorname{sh}(\varkappa L)}[\operatorname{ch}(\varkappa(z + z')) + \operatorname{ch}(\varkappa L)\operatorname{ch}(\varkappa(z - z')) \\ & \quad - \operatorname{sh}(\varkappa L)\operatorname{sh}(\varkappa|z - z'|)], \end{aligned} \tag{42}$$

$$\begin{aligned} \langle n_z(\varkappa, z) n_z^*(\varkappa, z')\rangle = {} & \frac{k_B T}{2\varkappa K\operatorname{sh}(\varkappa L)}[-\operatorname{ch}(\varkappa(z + z')) \\ & + \operatorname{ch}(\varkappa L)\operatorname{ch}(\varkappa(z - z')) - \operatorname{sh}(\varkappa L)\operatorname{sh}(\varkappa|z - z'|)], \\ & \langle n_y(\varkappa, z) n_z^*(\varkappa, z')\rangle = 0. \end{aligned} \tag{43}$$

For fluctuations that are homogeneous in the cell plane ($\varkappa = 0$), Eqs. (42) and (43) lead to the expressions

$$\begin{aligned} \langle n_y(\varkappa, z) n_y^*(\varkappa, z')\rangle &= \frac{k_B T}{4KL}[L^2 + 2z^2 + 2z'^2 - 2L|z - z'|], \\ \langle n_z(\varkappa, z) n_z^*(\varkappa, z')\rangle &= \frac{k_B T}{4KL}[L^2 - 4zz' - 2L|z - z'|]. \end{aligned} \tag{44}$$

Evidently, fluctuations of rotation about the z axis that are homogeneous in the xy plane are maximal on the layer boundary and minimal at its center, i.e., $\langle n_y(0,z) n_y(0,z)\rangle \sim L^2 + 4z^2$, while fluctuational deflections of the director from the xy plane are, on the contrary, maximal at the center of the layer and equal to zero on its boundaries, i.e., $\langle n_z(0,z) n_z(0,z)\rangle \sim L^2 - 4z^2$.

In the other case, when the director is freely deflected from the plane on the boundary of the medium, and rotation about the z axis entails appreciable energy losses ($W_z = 0$, $W_y = \infty$), we obtain Eqs. (44) in which the subscripts z and y must be interchanged. Thus, $\langle n_z(0,z) n_z(0,z)\rangle \sim L^2 + 4z^2$ and $\langle n_y(0,z) n_y(0,z)\rangle \sim L^2 - 4z^2$. If, however, both energies W_z and W_y tend to zero, the fluctuations, as already noted, become of the Goldston type.

It is of interest to track the manifestation of the singularity of the correlation functions at $\varkappa = 0$. Putting for simplicity $W_y = W_z = W$, we obtain for small W

$$\langle n_y(0,z)n_y(0,z')\rangle=\langle n_z(0,z)n_z(0,z')\rangle\sim\frac{k_BT}{2W}, \quad (45)$$

$$\langle n_y(\varkappa,z)n_y^*(\varkappa,z')\rangle=i\frac{k_BT}{2W}\varkappa_y(z+z')+o(\varkappa^2),$$

i.e., the correlators diverge as $W\to 0$.

The opposite case, when W is large, Eqs. (38)–(40) go over into

$$\langle n_y(0,z)n_y(0,z')\rangle$$
$$=\langle n_z(0,z)n_z(0,z')\rangle\sim\frac{k_BT}{4KL}[L^2-4zz'-2L|z-z'|], \quad (46)$$
$$\langle n_y(\varkappa,z)n_y^*(\varkappa,z')\rangle\sim i\frac{k_BTK}{LW^2}\varkappa_y(z+z')+o(\varkappa^2).$$

4. LIGHT SCATTERING BY DIRECTOR FLUCTUATIONS IN A CELL WITH AN HOMOTROPICALLY ORIENTED NEMATIC

A nematic liquid crystal is an optically anisotropic medium, and its dielectric-constant tensor is uniquely connected with the director field[7]

$$\varepsilon_{\alpha\beta}(\mathbf{r})=\varepsilon_{\parallel}n_\alpha(\mathbf{r})n_\alpha(\mathbf{r})+\varepsilon_\perp(\delta_{\alpha\beta}-n_\alpha(\mathbf{r})n_\beta(\mathbf{r})).$$

The director fluctuations $\delta\mathbf{n}(\mathbf{r})$ lead to changes $\delta\varepsilon(\mathbf{r})$ of the dielectric-constant tensor

$$\delta\varepsilon_{\alpha\beta}(\mathbf{r})=\varepsilon_a(n_\alpha^0\delta n_\beta(\mathbf{r})+n_\beta^0\delta n_\alpha(\mathbf{r})),$$

where $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$ is the anisotropy of the medium. This, in turn, causes light scattering in the medium. The intensity I of the scattered light is proportional to $\langle E'_\alpha(\mathbf{r})E'^*_\alpha(\mathbf{r})\rangle$, where $\mathbf{E}'$ is the scattered-wave field. Light scattering in unbounded NLC have been investigated in sufficient detail (see, e.g., Ref. 10). Let us analyze the influence of the director-substrate interaction on the light-scattering process.

If a plane wave with amplitude $\mathbf{E}^0$ and wave vector $\mathbf{k}_i$ propagates in a medium, the value $\langle E'_\alpha(\mathbf{r})E'^*_\beta(\mathbf{r})\rangle$ of singly scattered waves $\mathbf{E}'$ can be determined by an integral over the scattering volume

$$\langle E_\alpha'(\mathbf{r})E_\beta'^*(\mathbf{r})\rangle=\frac{\omega^4}{c^4}\int d^3\mathbf{r}'\,d^3\mathbf{r}''T_{\alpha\gamma}(\mathbf{r},\mathbf{r}')T^*_{\beta\lambda}(\mathbf{r},\mathbf{r}'')$$
$$\times\langle\delta\varepsilon_{\gamma\mu}(\mathbf{r}')\delta\varepsilon_{\lambda\nu}(\mathbf{r}'')\rangle E^0_\mu E^{0*}_\nu\exp(i\mathbf{k}_i(\mathbf{r}'-\mathbf{r}'')), \quad (47)$$

where $T_{\alpha\gamma}(\mathbf{r},\mathbf{r}')$ is the Green's functions of the Maxwell equations, and the anisotropy must be taken into account in it. Since we are interested in the basic influence of the director cohesion energy on the scattering, we confine ourselves to the asymptote $T_{\alpha\gamma}(\mathbf{r},\mathbf{r}')$ at large $\mathbf{R}=\mathbf{r}-\mathbf{r}'$ for an isotropic medium

$$T_{\alpha\beta}(\mathbf{r},\mathbf{r}')=\frac{1}{4\pi R}e^{ikR}(\delta_{\alpha\beta}-s_\alpha s_\beta), \quad (48)$$

where $k=\omega\varepsilon^{1/2}/c$, $\varepsilon=(\varepsilon_\parallel+\varepsilon_\perp)/2$, $\mathbf{s}=\mathbf{R}/R$. Substitution of (48) in (47) leads to

$$\langle E_\alpha'(\mathbf{r})E_\beta'^*(\mathbf{r})\rangle=\frac{\omega^4V}{c^4(4\pi)^2R^2}(\delta_{\alpha\gamma}-s_\alpha s_\gamma)(\delta_{\beta\lambda}-s_\beta s_\lambda)$$
$$\times\frac{1}{L}\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''\exp(-iq_z(z'-z''))$$
$$\times\langle\delta\varepsilon_{\gamma\mu}(\varkappa,z')\delta\varepsilon^*_{\lambda\nu}(\varkappa,z'')\rangle E_\mu^0E_\nu^{0*}, \quad (49)$$

where V is the scattering volume ($V=LS$ and S is the illuminated area of the cell),

$$\langle\delta\varepsilon_{\gamma\mu}(\varkappa,z')\delta\varepsilon^*_{\lambda\nu}(\varkappa,z'')\rangle$$
$$=\varepsilon_a^2[n_\gamma^0n_\lambda^0\langle\delta n_\mu(\varkappa,z')\delta n_\nu^*(\varkappa,z'')\rangle$$
$$+n_\mu^0n_\nu^0\langle\delta n_\gamma(\varkappa,z')\delta n_\lambda^*(\varkappa,z'')\rangle$$
$$+n_\gamma^0n_\nu^0\langle\delta n_\mu(\varkappa,z')\delta n_\lambda^*(\varkappa,z'')\rangle$$
$$+n_\mu^0n_\lambda^0\langle\delta n_\gamma(\varkappa,z')\delta n_\nu^*(\varkappa,z'')\rangle], \quad (50)$$

$\mathbf{q}=\mathbf{s}k-\mathbf{k}_i$ is the scattering vector with components $(\varkappa\cos\varphi,\varkappa\sin\varphi,q_z)$ (we neglect the difference between the wave numbers of the ordinary and extraordinary wave). We shall consider only normal incidence, i.e., $\mathbf{k}_i=k\mathbf{e}_z$. If the scattering is in the direction $\mathbf{s}=(\sin\theta\cos\varphi,\sin\theta\sin\varphi,\cos\theta)$, we have $\mathbf{q}=k(\sin\theta\cos\varphi,\sin\theta\sin\varphi,\cos\theta-1)$, $q=2k\sin(\theta/2)$. Equations (50) and (49) jointly with expressions (38)–(40) make it possible to determine the scattering intensity in an homotropically as well as a planarly oriented cell, and the double integral in (49) can be obtained analytically. For example, for an homotropically oriented cell with identical Frank moduli we obtain for the intensity I

$$I=I_0C\frac{1}{2\varkappa\Delta}\{(\varkappa^2-w^2)J_2+[(\varkappa^2+w^2)\mathrm{ch}(\varkappa L)$$
$$+2w\varkappa\,\mathrm{sh}(\varkappa L)]J_3-J_1\Delta\}, \quad (51)$$

where

$$C=\frac{\omega^4V\varepsilon_a^2k_BT}{c^4(4\pi)^2R^2K},$$

I_0 is the incident-light intensity, and J_i denote the integrals

$$J_1=\frac{1}{L}\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''\exp(-q_z(z'-z''))\mathrm{sh}(\varkappa|z'-z''|)$$
$$=\frac{2}{Lq^2}\Big\{-\varkappa L+\frac{1}{q^2}[(\varkappa^2-q_z^2)\mathrm{sh}(\varkappa L)\cos(q_zL)$$
$$+2\varkappa q_z\mathrm{ch}(\varkappa L)\sin(q_zL)]\Big\}. \quad (52)$$

$$J_2=\frac{1}{L}\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''\exp(-q_z(z'-z''))\mathrm{ch}(\varkappa(z'+z''))$$
$$=\frac{2}{Lq^2}[\mathrm{ch}(\varkappa L)-\cos(q_zL)]. \quad (53)$$

$$J_3=\frac{1}{L}\int_{-L/2}^{L/2}dz'\int_{-L/2}^{L/2}dz''\exp(-q_z(z'-z''))\mathrm{ch}(\varkappa(z'-z''))$$
$$=\frac{2}{Lq^4}\{(\varkappa^2-q_z^2)[\mathrm{ch}(\varkappa L)-\cos(q_zL)-1]$$
$$+2\varkappa q_z\mathrm{sh}(\varkappa L)\sin(q_zL)\}. \quad (54)$$

Recognizing the explicit dependence of the components of the scattering vector $\mathbf{q}$ on the scattering angle θ, we can represent (51) in the form

$$I=I_0C\frac{\cos(\theta/2)}{2\xi\sin(\theta/2)[(\sin\theta+\alpha)^2-\exp(-2\xi\sin\theta)(\sin\theta-\alpha)^2]}$$
$$\times\{\sin^2\theta-\alpha^2+(\sin\theta+\alpha)^2(\xi\sin\theta-\cos\theta)+\exp(-\xi\sin\theta)$$
$$\times[(\sin\theta+\alpha)^2\cos(\theta+2\xi\sin^2(\theta/2))$$
$$+(\sin\theta-\alpha)^2\cos(\theta-2\xi\sin^2(\theta/2))$$
$$-2(\sin^2\theta-\alpha^2)\cos(2\xi\sin^2(\theta/2))]-\exp(-2\xi\sin\theta)$$
$$\times[\alpha^2-\sin^2\theta+(\sin\theta-\alpha)^2(\xi\sin\theta+\cos(\theta))]\}, \quad (55)$$

where $\xi=kL$ is a large quantity and $\alpha=W/(Kk)$. If $\sin\theta$

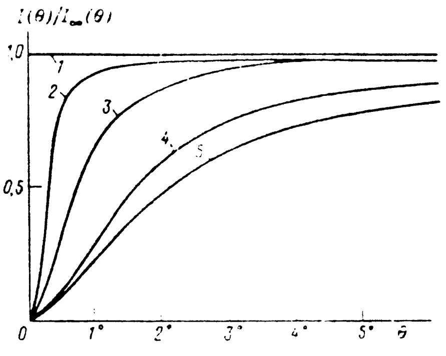

FIG. 1. Angular dependence of the intensity of light scattered by an homotropically oriented liquid-crystal cell, referred to the intensity $I_\infty(\theta) = I_0 C\cos^2(\theta/2)$ in an unbounded medium at various values of $\alpha = W/(Kk) = 0.2$ (*1*), 0.001 (*2*), 0.01 (*3*), 0.1 (*4*), 1 (*5*); $\xi = kL = 100$.

is not small, all the exponentials of type $\exp(-\xi\sin\theta)$ can be neglected, and we obtain the rather simple equation

$$I=I_0C\frac{\cos(\theta/2)}{2\xi\sin(\theta/2)}\left[\xi\sin\theta-\cos\theta+\frac{\sin\theta-\alpha}{\sin\theta+\alpha}\right]. \quad (56)$$

Evidently, at sufficiently large cell thicknesses L Eq. (56) goes over into the usual equation $I_\infty = C\cos^2(\theta/2)$ for scattering in an unbounded medium.

Note that if $\theta = 0$ Eq. (56) becomes infinite. The reason is that we have neglected terms containing $\exp(-\xi\sin\theta)$. The exact expression (55) for the scattering intensity remains finite in this case.

Figure 1 shows the angular dependence, calculated from Eq. (55), of the scattered light for an homotropically oriented liquid crystal at normal incidence. It is seen that the suppression of the long-wave fluctuations by the surface coupling lowers the intensity of the small-angle scattered light. The characteristic width of the dip decreases with decrease of the coupling energy or with increase of the cell thickness. For typical NLC samples and for thickness of the order of 10 μm it amounts to 1–2 deg.

APPENDIX

The exponentials of the matrix M, which enter in expressions (36) and (37), can be easily calculated by diagonalizing the matrix by the transformation $\hat{S}^{-1}\hat{M}\hat{S}$, where

$$\hat{S}=\begin{pmatrix}1&0&1&0\\0&1&0&1\\\varkappa&0&-\varkappa&0\\0&\varkappa&0&-\varkappa\end{pmatrix},$$

$$\hat{S}^{-1}=\frac{1}{2}\begin{pmatrix}1&0&\varkappa^{-1}&0\\0&1&0&\varkappa^{-1}\\1&0&-\varkappa^{-1}&0\\0&1&0&-\varkappa^{-1}\end{pmatrix}, \quad (A1)$$

multiplying $\exp(z\hat{M})$ from the left by $\hat{S}\hat{S}^{-1}$ and from the right by $\hat{S}^{-1}\hat{S}$, where

$$\exp(z\hat{S}^{-1}\hat{M}\hat{S})=\begin{pmatrix}e^{\varkappa z}&0&0&0\\0&e^{\varkappa z}&0&0\\0&0&e^{-\varkappa z}&0\\0&0&0&e^{-\varkappa z}\end{pmatrix}. \quad (A2)$$

It is expedient to consider in place of the matrices $\hat{C}_\sigma(z')$ the matrices $\hat{H}^{(\sigma)}(z') = \hat{S}^{-1}\hat{C}_\sigma(z')$. The elements of the correlation matrix $\hat{G}_p$ can then be expressed in terms of the elements of the matrices $\hat{H}^{(\sigma)}$

$$\begin{aligned}G_{p11}&=H_{11}^{(\sigma)}e^{\varkappa z}+H_{32}^{(\sigma)}e^{-\varkappa z},\\ G_{p22}&=H_{22}^{(\sigma)}e^{\varkappa z}+H_{42}^{(\sigma)}e^{-\varkappa z},\\ G_{p12}&=H_{12}^{(\sigma)}e^{\varkappa z}+H_{32}^{(\sigma)}e^{-\varkappa z},\\ G_{p21}&=H_{21}^{(\sigma)}e^{\varkappa z}+H_{41}^{(\sigma)}e^{-\varkappa z},\end{aligned} \quad (A3)$$

with $\sigma = 1$ for $z > z'$ and $\sigma = -1$ for $z < z'$ in these equations.

Multiplying both sides of (38) by $\hat{S}\exp(-\hat{S}^{-1}\hat{M}\hat{S}z')$ and taking (A1) into account we get

$$\hat{H}^{(1)}(z')-\hat{H}^{(-1)}(z')=\frac{k_BT}{2K\varkappa}\begin{pmatrix}-\exp(-\varkappa z')&0\\0&-\exp(-\varkappa z')\\\exp(\varkappa z')&0\\0&\exp(\varkappa z')\end{pmatrix}. \quad (A4)$$

The boundary conditions (39) can be written in the form

$$\begin{pmatrix}(\sigma w_y+\varkappa)\exp(\sigma\varkappa L/2)&(\sigma w_y-\varkappa)\exp(-\sigma\varkappa L/2)\\ i\varkappa_y\exp(\sigma\varkappa L/2)&i\varkappa_y\exp(-\sigma\varkappa L/2)\end{pmatrix}\begin{pmatrix}H_{1j}^{(\sigma)}\\H_{3j}^{(\sigma)}\end{pmatrix}$$

$$=\begin{pmatrix}i\varkappa_y\exp(\sigma\varkappa L/2)&i\varkappa_y\exp(-\sigma\varkappa L/2)\\-(\sigma w_z+\varkappa)\exp(\sigma\varkappa L/2)&-(\sigma w_z+\varkappa)\exp(-\sigma\varkappa L/2)\end{pmatrix}$$

$$\times\begin{pmatrix}H_{2j}^{(\sigma)}\\H_{4j}^{(\sigma)}\end{pmatrix}, \quad (A5)$$

where $j = 1$ or 2. For $j = 1$ we obtain from (A5) and (A4) the set of equations

$$\begin{pmatrix}H_{21}^{(1)}\\H_{41}^{(1)}\end{pmatrix}=\begin{pmatrix}H_{21}^{(-1)}\\H_{41}^{(-1)}\end{pmatrix}, \quad (A6)$$

$$\begin{pmatrix}H_{11}^{(1)}\\H_{31}^{(1)}\end{pmatrix}-\begin{pmatrix}H_{11}^{(-1)}\\H_{31}^{(-1)}\end{pmatrix}=\frac{1}{2\varkappa}\begin{pmatrix}-\exp(-\varkappa z')\\\exp(\varkappa z')\end{pmatrix}, \quad (A7)$$

$$\begin{pmatrix}H_{11}^{(\sigma)}\\H_{13}^{(\sigma)}\end{pmatrix}=\frac{1}{2i\varkappa\varkappa_y}$$

$$\times\begin{pmatrix}-\varkappa_y^2+(\sigma w_y-\varkappa)(\sigma w_z+\varkappa)&[-\varkappa_y^2+(\sigma w_y-\varkappa)(\sigma w_z-\varkappa)]e^{-\sigma\varkappa L}\\ [\varkappa_y^2-(\sigma w_y+\varkappa)(\sigma w_z+\varkappa)]e^{\sigma\varkappa L}&\varkappa_y^2-(\sigma w_y+\varkappa)(\sigma w_z-\varkappa)\end{pmatrix}$$

$$\times\begin{pmatrix}H_{21}^{(\sigma)}\\H_{41}^{(\sigma)}\end{pmatrix}, \quad (A8)$$

and for $j = 2$

$$\begin{pmatrix}H_{12}^{(1)}\\H_{32}^{(1)}\end{pmatrix}=\begin{pmatrix}H_{12}^{(-1)}\\H_{32}^{(-1)}\end{pmatrix}, \quad (A9)$$

$$\begin{pmatrix}H_{22}^{(1)}\\H_{42}^{(1)}\end{pmatrix}-\begin{pmatrix}H_{22}^{(-1)}\\H_{42}^{(-1)}\end{pmatrix}=\frac{1}{2\varkappa}\begin{pmatrix}-\exp(-\varkappa z')\\\exp(\varkappa z')\end{pmatrix}, \quad (A10)$$

$$\begin{pmatrix}H_{22}^{(\sigma)}\\H_{42}^{(\sigma)}\end{pmatrix}=\frac{1}{2i\varkappa\varkappa_y}$$

$$\times\begin{pmatrix} \varkappa_y^2-(\sigma w_z-\varkappa)(\sigma w_y+\varkappa) & [\varkappa_y^2-(\sigma w_z-\varkappa)(\sigma w_y-\varkappa)]e^{-\sigma\varkappa L} \\ [(\sigma w_z+\varkappa)(\sigma w_y+\varkappa)-\varkappa_y^2]e^{\sigma\varkappa L} & \varkappa_y^2-(\sigma w_y+\varkappa)(\sigma w_z-\varkappa) \end{pmatrix}$$

$$\times\begin{pmatrix} H_{12}^{(\sigma)} \\ H_{32}^{(\sigma)} \end{pmatrix}. \tag{A11}$$

Substitution of (A8) in (A7) and of (A11) in (A10) now reduces the problem to inversion of two 2×2 matrices. After determining thus all the elements of the matrix $H^{(\sigma)}$ with the aid of Eqs. (A3) we can calculate the elements of the sought correlation matrix $\hat{G}_p$. They are contained in Eqs. (38)–(40).

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