Casimir forces in modulated systems.

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Abstract

For the first time we present analytical results for the contribution of electromagnetic fluctuations into thermodynamic properties of modulated systems, like cholesteric or smectic liquid crystalline films. In the case of small dielectric anisotropy we have derived explicit analytical expressions for the chemical potential of such systems. Two limiting cases were specifically considered: (i) the Van der Waals (VdW) limit, i.e., in the case when the retardation of the electromagnetic interactions can be neglected; and (ii) the Casimir limit, i.e., when the effects of retardation becomes considerable. It was shown that in the Casimir limit, the film chemical potential oscillates with the thickness of the film. This non-monotonic dependence of the chemical potential on the film thickness can lead to step-wise wetting phenomena, surface anchoring reorientation and other important effects. Applications of the results may concern the various systems in soft matter or condensed matter physics with multilayer or modulated structures.

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I. INTRODUCTION

Stratified systems are omnipresent in soft matter and solid state physics. Understanding of molecular interactions in these systems is an important step for controlling their properties and aggregation processes. Though these interactions may have many different specific sources, the electromagnetic fluctuations (VdW and Casimir forces) are their common cause. These interactions become appreciable in the submicron range and rapidly increase in the nanometer scale. Although, evidently, the interactions would not scale nor ad infinitum, neither at shorter scales, where steric short range interactions, i.e. interaction forces whose origin is in the Pauli principle, become dominating ones. Still, submicron - nanometer range, where long range forces are dominant ones, is the most important range for applications in nanotechnology processing. Several approaches have been employed to calculate these fluctuational forces. The simplest one is Hamaker approach [1], summing all pairwise interactions, which is evidently not applicable to condensed media, where all molecular units are strongly correlated. The rigorous continuum method was developed in [2] (see also [3]), by Lifshitz, Pitaevskii, and one of the authors of the present paper (I.D.) who derived the general expression for the electromagnetic fluctuational interaction between two macroscopic isotropic bodies separated by an isotropic film. Although the generalization of the analysis [2] for the anisotropic and multilayer systems is conceptually straightforward, it deserves some attention, as it implies prohibitively tedious and bulky calculations, which could be done analytically only under certain rather restrictive approximations (see e.g., [4], [5]). Luckily, however, there are some cases when these approximative calculations can be performed in a well controlled way, and besides they may be useful for systems interesting for applications. For example when dielectric anisotropy is small in the whole region of frequencies relevant for the interactions, one can use the regular perturbation theory [6], which is a typical case for liquid crystalline films [7], [8]. Moreover, cholesteric liquid crystals having a long wavelength helical orientational modulation can be treated as a continuous model of multilayer systems.

In what follows we calculate the electromagnetic fluctuational contributions to the chemical potential of a cholesteric or smectic liquid crystal film (thickness $l$) confined between two semi infinite isotropic media (having in mind experimentally interesting cases, the latter can be, e.g., solid, glass-like substrate and air). Our approach is a macroscopical one,
i.e. we consider quantities averaged over physically infinitesimal volumes. Thus we restrict ourselves to only the long wavelength part of the electromagnetic field fluctuations. In this range everything can be expressed in terms of macroscopic characteristics of the system (dielectric and magnetic permeabilities).

Since for cholesteric liquid crystals typical scales of orientational modulations are very large (of the order of 500\textit{nm} and even larger for nematic - cholesteric mixtures \cite{7}, \cite{8}), they may be the perfect candidates for observing of predicted (by our macroscopic continuum approach) behavior. With a certain modification in order to take into account magnetic properties, one can also perform analogous calculations for large pitch magnetic spiral structures. The same is true for so-called lyotropic smectic liquid crystals where density modulations occur at scales larger than characteristic molecular lengths \((2−5)\textit{nm})\), however, for thermotropic smectic liquid crystals this window of validity of our approach is much more narrow (if it exists at all). In this case the continuum theory loses all pretense of quantitative predictability and a complete description should incorporate microscopic structure and molecular short range forces what can be done only numerically (see e.g., \cite{9}, \cite{10}).

Our motivation for presenting this discussion is two new predictions which have emanated from our investigation. Namely, in the Casimir limit we have found spatially oscillating terms in the chemical potential of the film. In the VdW limit (small film thickness) we have found a monotonic in \(l\) finite anisotropic correction to the Hamaker constant, making possible a reorientation anchoring phase transition with variation of the film thickness.

The remainder of our paper has the following structure. Section II contains basic methodical details and equations necessary for our investigation in the framework of a regular method for calculating higher order perturbative corrections. New results are discussed in section III in subsections III A and III B, where we present the electromagnetic fluctuational contributions into the cholesteric film chemical potential. Finally, Section IV deals with miscellaneous subjects related to the physical consequences of our results for wetting, stability and anchoring phenomena in cholesteric films. Some technical material is collected in Appendix to the paper, and those readers who are not very interested in derivations can skip this Appendix finding all essential physical results in the main text of the paper.
II. ELECTROMAGNETIC FLUCTUATIONS IN CHOLESTERIC FILMS

We limit our analysis to the long range forces induced by electromagnetic fluctuations, thus neglecting all kinds of structural thermal fluctuations. We consider a leftmost half space, \(-\infty < x < 0\), as an isotropic body (denoted in what follows by the indices 1 (solid, e.g., glass substrate) and a rightmost half space \(l < x < +\infty\), (vacuum or air), separated by non-homogeneous and anisotropic film, \(0 \leq x \leq l\), (denoted by the index 3) with thickness \(l\).

First closely following the general method [2] we study the case when the substance 3 is a cholesteric liquid crystal film. We chose the system for our benchmark to determine all specific features of electromagnetic fluctuational forces in anisotropic modulated systems. We take the \(x\)-axis perpendicular to the separating surfaces (planes \(x = 0\) and \(x = l\)). This is not the place to explain the general approach to fluctuational electromagnetic forces in detailed, however, in a stripped down version the theory [2] is reduced to the calculation of Green functions \(D_{ik}\) to the Maxwell equations, i.e.,

\[
[\epsilon_{il}(r, i|\omega_n|)\omega_n^2 - \text{curl}_{im}\text{curl}_{ml}]D_{ik}(r, r'; \omega_n) = -4\pi\delta(r - r')\delta_{ik},
\]

(2.1)

where \(\epsilon_{il}(r, i|\omega_n|)\) is dielectric permeability at Matsubara imaginary frequencies \(i\omega_n\). It is worth noting that the dielectric function is always real on the imaginary axis irrespective of whether the system is absorbing or not. In what follows we use atomic units, i.e. put light velocity \(c\) and Planck constant \(\hbar\) as 1 (except where explicitly stated to the contrary and the occurrences of \(\hbar\) or \(c\) are necessary for understanding).

Calculating somehow \(D_{ik}\) one can find the stress tensor \(\sigma_{ik}\) in the film

\[
\sigma'_{ik} = -\frac{T}{2\pi} \sum_{n=0}^{\infty} \left\{ \epsilon^{(3)}_{im} \left[ D^{E}_{mk}(r, r; i\omega_n) - \frac{1}{2} \delta_{mk} D^{E}_{jj}(r, r; i\omega_n) \right] - \frac{1}{2} \delta_{ik} D^{H}_{jj}(r, r; i\omega_n) \right\}.
\]

(2.2)

Here, \(\sigma'_{ik}\) is electromagnetic fluctuation part of \(\sigma_{ik}\), and functions \(D^{E}_{ik}\) and \(D^{H}_{ik}\) are corresponding averages of fluctuating electric and magnetic field components

\[
D^{E}_{ik}(r, r'; i\omega_n) = -\omega_n^2 D_{il}(r, r'; i\omega_n); D^{H}_{il}(r, r'; i\omega_n) = \text{curl}_{il}\text{curl}_{km}D_{lm}(r, r'; i\omega_n).
\]

(2.3)

Because the problem is homogeneous in \(z - y\) plane we make a Fourier transform with respect to \(z - z'\) and \(y - y'\) coordinates, thus getting \(D_{ik}(x, x', \mathbf{q}; i\omega_n)\). Taking into account that
\(\epsilon_{ik}^{(1)} = \epsilon_1 \delta_{ik}, \ \epsilon_{ik}^{(2)} = \delta_{ik}, \) and

\[
\epsilon_{ik}^{(3)} = \begin{bmatrix}
\epsilon_3 & 0 & 0 \\
0 & \epsilon_3 + \epsilon_a (1 + \cos(2q_0x)) & \epsilon_a \sin(2q_0x) \\
0 & \epsilon_a \sin(2q_0x) & \epsilon_3 + \epsilon_a (1 - \cos(2q_0x))
\end{bmatrix},
\]

(2.4)

where we define the cholesteric dielectric permeability as \(\epsilon_{ik}^{(3)} = \epsilon_3 \delta_{ik} + 2 \epsilon_a n_i n_k \) \((n = (0, \cos(q_0x), \sin(q_0x)))\) is cholesteric equilibrium director, \(q_0\) is cholesteric spiral modulation wave vector, related to cholesteric pitch \(p\) as \(q_0 = 2\pi/p\), and in what follows we assume a small dielectric anisotropy \(\epsilon_a < \epsilon_3\), what is the case for all known cholesteric liquid crystals [7], [8].

In the natural coordinate frame \(x, y, z\) (2.2) is a set of nine coupled equations (presented in the appendix to the paper). This set should be supplemented by the usual boundary conditions (corresponding to the continuity of the tangential components of the electric and magnetic fields) which are the continuity conditions for

\[
D_{yk}, \ D_{zk}, \ curl_y D_{lk}, \ curl_z D_{lk}.
\]

(2.5)

In principle the full information concerning electromagnetic fluctuational forces is contained in solution of (A1) - (A9) supplemented by the boundary conditions (2.5). In practice, however, for a given cholesteric dielectric permeability (2.4) the analytic solution is infeasible, thus some approximations are definitely needed not only to make calculations possible, but also to understand correctly underlying physical phenomena. Luckily since \(\epsilon_a < \epsilon_3\), for all known cholesteric liquid crystals, one can solve the equations using a regular perturbation theory with respect to the small parameter \(\epsilon_a/\epsilon_3\).

Omitting a large amount of tedious (although straightforward) algebra, we get in the first order approximation the set of four equations to find \(D_{zz}, D_{yy}, D_{xy}, \) and \(D_{xx}\). Two latter equations are simple relations

\[
D_{xy} = -\frac{iq}{w^2} \frac{\partial}{\partial x} D_{yy}, \ D_{xx} = -\frac{iq}{w^2} \frac{\partial}{\partial x} D_{xy} - \frac{4\pi}{w^2} \delta(x - x')
\]

(2.6)

where we designate \(w^2 \equiv \epsilon \omega_n^2 + q^2\), and as \(\epsilon\) one should take \(1, \epsilon_2\) and \(\epsilon_3 + \epsilon_a\), for air, substrate, and cholesteric film respectively. Two other equations (for \(D_{zz}\) and \(D_{yy}\)) deserve some precaution only in the cholesteric film region \((0 < x < l)\) where they are equivalent to one particle Schrödinger equations in a periodic potential (see appendix, (A10), (A11)).
However, since to derive these equations we already used the perturbation theory, we have to consider only so-called almost free particle approximation (weak coupling limit), which gives for our case (A12), (A13). Determining arbitrary constants $A_i$, $C_i$ entering these expressions from the boundary conditions (2.5) and subtracting homogeneous system contribution, we find finally almost conventional [2] expressions for the Green functions

$$D_{zz} = \frac{4\pi}{w_3\Delta} \cosh[w_3(x - x')] ,$$

(2.7)

where, however, $\Delta$ contains oscillating terms

$$\Delta = 1 - \exp(2w_3l) \frac{(w_3 + w_1)(w_3 + w_2)}{(w_3 - w_1)(w_3 - w_2)} [1 + \alpha_1 \sin(2q_0l) + \alpha_2 \cos(2q_0l)] ,$$

(2.8)

and we designate

$$\alpha_1 = \frac{\epsilon_0 \omega_2^2}{2(q_0^2 + w_3^2)} \left[ -\frac{w_3}{2q_0} + \frac{q_0}{w_3 + w_2} \right] ,$$

(2.9)

and

$$\alpha_2 = \frac{\epsilon_0 \omega_2^2}{4(q_0^2 + w_3^2)} .$$

(2.10)

Analogously

$$D_{yy} = \frac{4\pi w_3}{\omega_2^4 \epsilon_3 \Delta_1} \cosh[w_3(x - x')] ,$$

(2.11)

where

$$\Delta_1 = 1 - \exp(2w_3l) \frac{(w_3 + w_2)(\epsilon_1 w_3 + w_1)}{(w_3 - w_2)(\epsilon_1 w_3 - w_1)} [1 + \beta_1 \sin(2q_0l) + \beta_2 \cos(2q_0l)] ,$$

(2.12)

and as above

$$\beta_1 = \frac{\epsilon_0 w_3^3}{4(q_0^2 + w_3^2)\epsilon_3(w_3 + w_2)} + \frac{\epsilon_0 w_2 w_3 q_0}{4\epsilon_3(q_0^2 + w_3^2)} ,$$

(2.13)

and

$$\beta_2 = -\frac{\epsilon_0 w_3^3}{4\epsilon_3(q_0^2 + w_3^2)(w_3 + w_2)} - \frac{\epsilon_0 q_0^2}{2\epsilon_3(q_0^2 + w_3^2)} .$$

(2.14)

These expressions (2.7) - (2.14) are our main results and they are ready for further inspection.
III. FLUCTUATIONAL CONTRIBUTION TO THE CHEMICAL POTENTIAL

Our aim is to calculate fluctuational contributions into thermodynamic properties of a thin cholesteric film on an isotropic solid substrate. Since the film should be in equilibrium

\[ P = P_{ch} - \sigma'_{xx}, \]  

(3.1)

where \( \sigma'_{xx} \) is given in (2.2), \( P \) is the air or vapor pressure, and \( P_{ch} \) is the pressure of the bulk cholesteric. Note that using a standard definition for the pressure

\[ P_{ch} = \rho \frac{\partial E}{\partial \rho} - E, \]  

(3.2)

were \( \rho \) is cholesteric mass density, and into the energy density \( E \), orientational deformation energy (Frank energy [7], [8]) has to be added to isotropic liquid pressure \( P_0 \). Therefore,

\[ P_{ch} = P_0 + K_{22} q_0 \gamma (\epsilon_{ikn} n_k n_n - q_0), \]  

(3.3)

where \( \epsilon_{ikn} \) is antisymmetric tensor, \( K_{22} \) - twist orientational elastic modulus, and \( \gamma = - (\partial \ln q_0 / \partial \rho) \) is cholesteric pitch “compressibility”. Now we are in the position to reap fruits of our calculations. From (3.1) and given above expressions (2.2), (2.7) and (2.11) the electromagnetic fluctuational part of the cholesteric film chemical potential \( \mu(l) \) reads

\[ \mu(l) = - \frac{T}{2\pi} \sum_{n=0}^{\infty} \int_0^{\infty} q dq w_3 \left( \frac{1}{\Delta} + \frac{1}{\Delta_1} \right). \]  

(3.4)

Putting all expressions together one can write down (3.4) explicitely. This general expression is a very bulky one, however, it can be considerably simplified in two important limiting cases.

A. VdW interactions

We term VdW interaction the case of small thicknesses, i.e. \( l \ll \lambda^* \), where \( \lambda^* \) characterizes the main absorption band of the cholesteric film. Typically in known cholesteric liquid crystals [8], \( \lambda^* \) is of the order of \( 10^{-4} \text{cm} \) and \( q_0 \lambda^* < 1 \). Thus we can perform an expansion over \( q_0 l \) in (2.7) - (3.4) and get a relatively compact answer for this limit

\[ \mu(l) = \mu_0(l) + a \frac{q_0^2}{l}, \]  

(3.5)
where $\mu_0(l) \propto l^{-3}$ is known [2] for isotropic films VdW contribution, and

$$a = \frac{1}{64\pi^2} \int_0^\infty dp \int_0^\infty d\xi \frac{\epsilon_a(i\xi)}{\epsilon(b(i\xi))} \left[ \frac{(\epsilon_1(i\xi) + \epsilon_3(i\xi))(\epsilon_3(i\xi) + 1)}{\epsilon_3(i\xi) - \epsilon_2(i\xi))(\epsilon_3(i\xi) - 1)} \exp(x) - 1 \right]^{-1}. \quad (3.6)$$

Let us emphasize in passing that this anisotropic VdW contribution scales as $1/l$, and it is more long ranged than known fluctuational forces in isotropic homogeneous materials. It comes from the interplay of the characteristic lengths (film thickness and cholesteric pitch), and the extra factor $q_0^2$ in (3.5) provides correct dimensionality. Note also that the sign of this contribution (3.6) does not depend on the cholesteric spiral chirality (left or right) as one can certainly expect but it does depend on dielectric properties and could be positive or negative. In the latter case the planar director orientation (cholesteric axis along $x$-axis) is always stable, and in the former case this orientation is energetically unfavorable and could become unstable at a certain thickness $l_{cr}$. The critical thickness is determined by a competition of VdW contribution (3.6) and stabilizing planar orientation short range anchoring energy $W$

$$l_{cr} \propto \frac{W}{aq_0^2}. \quad (3.7)$$

This transition is analogous to the Freedericksz transition known in liquid crystals [7], [8], however, while the Freedericksz transition in liquid crystals is driven by the quadratic coupling between an external magnetic (or electric) field and the director, in our case the transition is induced by internal electromagnetic field fluctuations (VdW forces).

Some comments about the validity of (3.7) seem in order here. The critical thickness (3.7) is determined by a competition of the VdW (long range) contribution into the bulk chemical potential, and phenomenological (basically short range) anchoring energy $W$. Clearly the approach misses short range contributions into the bulk properties. If the only forces between particles had a range of the order of molecular dimensions (short range forces), the corresponding contributions would decrease exponentially with increasing distance, therefore assuming that at the scale (3.7) the VdW contribution is dominating, we believe that $l_{cr}$ is much larger than all molecular scales. This assumption is not always justifiable, and in this case, (3.7) does not maintain numerical accuracy, but nevertheless is useful and instructive for gaining some qualitative insight on interplay short and long range forces.
B. Casimir force

We now turn to the opposite limiting case \( l > \lambda^* \) when electromagnetic interaction retardation effects become relevant, and it is referred as Casimir limit. Closely following to [2] we can replace in this case all dielectric permeabilities entering the general formula (3.4) for \( \mu(l) \) by their static values, and after some algebra end up with

\[
\mu(l) = \mu_0(l) + b \frac{1}{l^4} \cos(2q_0 l),
\]

(3.8)

where \( \mu_0 \propto l^{-4} \) is conventional isotropic Casimir contribution, and the coefficient \( b \) reads as

\[
b = -\frac{1}{32\pi^2} \int_0^\infty dx \int_1^\infty dp \epsilon_a(0) \sqrt{\epsilon_3(0)} \frac{x^3}{p^2} \frac{\phi \exp(x)}{(\phi \exp(x) - 1)^2},
\]

(3.9)

where we designate

\[
\phi = \frac{(s_{10} + p \epsilon_1(0))(s_{20} + p)}{(s_{10} - p \epsilon_1(0))(s_{20} - p)},
\]

(3.10)

and

\[
s_{10} = \sqrt{\frac{\epsilon_1(0)}{\epsilon_3(0)}} - 1 + p^2; \quad s_{20} = \sqrt{\frac{1}{\epsilon_3(0)}} - 1 + p^2.
\]

(3.11)

The Casimir limit (3.8) is particularly instructive, because it shows oscillating with a thickness dependence of the chemical potential. Since the both contributions (monotonic and oscillating ones) decay as the same power of \( l \), it is especially interesting to hypothesize the case (not forbidden in principle although formally beyond our first order perturbation theory approximation) when it becomes negative. Suffice it to say that such an oscillating function \( \mu(l) \) leads to a film formation which is stable only in a certain range of its thickness. This kind of successive thinning or wetting - drying transitions are known for layered smectic phases (see e.g. [11], [12], [13]). We have shown that Casimir forces in cholesteric liquid crystals can be responsible for similar phenomena. Lacking sufficient data on dielectric permeability frequency dispersion we can at present discuss only the general qualitative features of Casimir forces in cholesteric liquid crystals.

Actually our picture is not entirely correct since we have neglected all short range forces, and this relatively weak oscillating Casimir contribution can be swamped by stronger short range forces. However by their nature, the short range forces have no connection with the effects under consideration which are due to cholesteric long wavelength orientational modulations. Short range contributions to the chemical potential are the same in cholesterics and...
in analogous homogeneous systems (e.g., nematics). Thus, even in the case of dominating short range forces, the Casimir contribution (3.8) could be disentangled in differential measurements. We anticipate that the discussed above effects of Casimir film instabilities will be observable and that understanding of underlying mechanisms will be essential to predict and to use these phenomena.

IV. CONCLUSION

In summary, in this paper we have calculated electromagnetic fluctuational contributions into cholesteric liquid crystal film chemical potential, and found the oscillating with the film thickness contribution (3.8). Quite remarkably this result illustrates that the collective nature of the Casimir forces suggests it to have a non-trivial dependence on film thickness, and even the sign of the Casimir force is dependent of material parameters and structure.

Our theoretical approach can be extended in several directions. First, our main result (3.8) with evident modifications can be applied to a smectic liquid crystal film, where its dielectric permeability acquires an oscillating contribution owing to one dimensional density modulation. Indeed, for a smectic film we should replace (2.4) by

\[ \epsilon_{yy} = \epsilon_{xz} = \epsilon_3; \epsilon_{xx} = (\epsilon_3 + \epsilon_a) + \left( \frac{\partial(\epsilon_3 + \epsilon_a)}{\partial \rho} \right) \cos(q_{sm},x), \] (4.1)

where we chose \( x \)-axis as a normal to smectic layers, and \( q_{sm} \) is smectic density modulation wave vector. Of course the macroscopic approach, we used in this paper, is valid only when the corresponding modulation periods are large with respect to atomic or molecular scales. It is always the case for cholesteric liquid crystals (a typical pitch is about \((500 - 700)nm\)), but usually not true for standard thermotropic smectics, where their density modulated with a period in the range of \((2 - 5)nm\) [8], [7]. Thus as an application of our results to smectics, we have in mind long period lyotropic smectic liquid crystals and some other lamellar (layered) membrane structures [14] where period can be much larger.

One more comment of caution. The relation between the fluctuational force and chemical potential (in fact the mechanical equilibrium condition of the type of (2.6) we have used for cholesteric films), is valid for liquids (or uniform in density cholesteric liquid crystals having no solid-like elasticity). In smectics, like in solids, an elastic layer compressibility modifies the equilibrium, and there is no simple relation between electromagnetic fluctuation force...
and film chemical potential. For these cases our theoretical predictions should be confronted with direct force or (in anisotropic cases) angular torque measurements.

On equal footing one can consider an apparently unrelated problem of electromagnetic fluctuations in solid long periodic magnetic structures. The issue of Casimir forces in materials with nontrivial magnetic susceptibility has been discussed recently [15], [16], [17]. To treat the magnetic case we have to include magnetic permeabilities $\mu$ of all three media into the equation for the Green function (2.1) and to modify the boundary conditions (2.5). The latter ones are read as the continuity of

$$D_{zz}, \frac{1}{\mu} \frac{dD_{zz}}{dx}, D_{yy}, \frac{\epsilon}{w^2} \frac{dD_{yy}}{dx},$$

(4.2)

and besides $w$ functions are now $w^2 = \epsilon \mu \omega_n^2 + q^2$. Calculations become more involved (see e.g. [18]) but in the framework of perturbation theory are still feasible analytically. However results and even signs of fluctuational contributions depend on many unknown functions describing dielectric and magnetic permeability frequency dispersions. A more specific study might become appropriate should suitable experimental results become available.

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APPENDIX A

We take the $y$-axis along the vector $q$ and in the coordinate frame attached to the system (2.2) in self-evident notations reads

$$\left[ (\epsilon_{zz} \omega_n^2 + q^2) - \frac{\partial^2}{\partial x^2} \right] D_{zz} + \epsilon_{zy} \omega_n^2 D_{yz} = -4\pi \delta(x - x') , \quad \text{(A1)}$$

$$\left[ \epsilon_{yy} \omega_n^2 - \frac{\partial^2}{\partial x^2} \right] D_{yy} + \epsilon_{yz} \omega_n^2 D_{zy} + i \frac{\partial}{\partial x} q D_{xy} = -4\pi \delta(x - x') , \quad \text{(A2)}$$

$$(\epsilon_{xx} \omega_n^2 + q^2) D_{xx} + iq \frac{\partial}{\partial x} D_{yx} = -4\pi \delta(x - x') , \quad \text{(A3)}$$

$$\left[ (\epsilon_{zz} \omega_n^2 + q^2) - \frac{\partial^2}{\partial x^2} \right] D_{zy} + \epsilon_{zy} \omega_n^2 D_{yy} = 0 , \quad \text{(A4)}$$
From these equations in the first order over the small parameter $\epsilon_a/\epsilon_3$ we get two decoupled equations for $D_{zz}$ and $D_{xx}$, which are in the only non-trivial region $0 < x < l$

$$\left[w_3^2 - \frac{\partial^2}{\partial x^2} + \frac{\epsilon_3}{4w_3} \sin(2q_0x)\right] D_{zz} = -4\pi \delta(x-x'), \tag{A10}$$

and

$$\left[w_3^2 - \frac{\partial^2}{\partial x^2} - \frac{\epsilon_a w_3^2}{\epsilon_3} - \frac{\epsilon_a q^2}{\epsilon_3 w_3^2}\right] D_{yy} = 0, \tag{A11}$$

and from the solution to these equations using two relations (2.6) one can find two others Green functions $D_{xy}$ and $D_{xx}$. General solutions to the equations (A10), (A11) read as

$$D_{zz} = C_1 \exp(w_3x) \left[1 - \frac{\epsilon_a w_3^2}{4w_3q_0} \sin(2q_0x) + \frac{\epsilon_a w_3^2}{4w_3(q_0^2 + w_3^2)}(w_3 \cos(2q_0x) + q_0 \sin(2q_0x))\right]$$

$$+ C_2 \exp(-w_3x) \left[1 + \frac{\epsilon_a w_3^2}{4w_3q_0} \sin(2q_0x) - \frac{\epsilon_a w_3^2}{4w_3(q_0^2 + w_3^2)}(-w_3 \cos(2q_0x) + q_0 \sin(2q_0x))\right]$$

$$- \frac{2\pi}{w_3} \exp(-w_3|x-x'|) \left[1 + \frac{\epsilon_a w_3^2}{4w_3(q_0^2 + w_3^2)}(q_0 \cos(2q_0x) + w_3 \sin(2q_0x))\right] \tag{A12}$$

and

$$D_{yy} = A_1 \exp(w_3x) \left[1 + \frac{\epsilon_a w_3^2}{4w_3q_0} \sin(2q_0x) - \frac{\epsilon_a w_3^2}{4w_3(q_0^2 + w_3^2)}(w_3 \cos(2q_0x) + q_0 \sin(2q_0x)) + \frac{\epsilon_a q^2}{4w_3q_0}\right]$$

$$+ A_2 \exp(-w_3x) \left[1 - \frac{\epsilon_a w_3^2}{4w_3q_0} \sin(2q_0x) + \frac{\epsilon_a w_3^2}{4w_3(q_0^2 + w_3^2)}(-w_3 \cos(2q_0x) + q_0 \sin(2q_0x)) + \frac{\epsilon_a q^2}{4w_3q_0}\right]$$

$$- \frac{2\pi w_3}{\epsilon_3 \omega_n^2} \exp(-w_3|x-x'|) - \frac{\pi w_3^2 \epsilon_a}{\epsilon_3 \omega_n^2} \exp(w_3x)t_+$$

$$+ \frac{\pi q^2 \epsilon_a}{\epsilon_3 \omega_n^2} \exp(w_3x)s_+ + \frac{\pi w_3^2 \epsilon_a}{\epsilon_3 \omega_n^2} \exp(-w_3x)p_+ - \frac{\pi q^2 \epsilon_a}{\epsilon_3 \omega_n^2} \exp(-w_3x)m_+, \tag{A13}$$
where \( C_i, A_i \) are constants which should be found from the \( x = 0 \) and \( x = l \) boundary conditions, and in (A13) we introduce shorthand notations

\[
\begin{align*}
t_+ &= \exp(w_3x' - 2w_3x)\frac{-w_3\cos(2q_0x) + q_0\sin(2q_0x)}{2(q_0^2 + w_3^2)}; \\
s_+ &= -\frac{1}{2w_3}\exp(w_3x' - 2w_3); \\
m_+ &= \frac{1}{2w_3}\exp(-w_3x' + 2w_3x); \\
p_+ &= \exp(w_3x')\frac{1}{2q_0}\sin(2q_0x).
\end{align*}
\]

(A14)