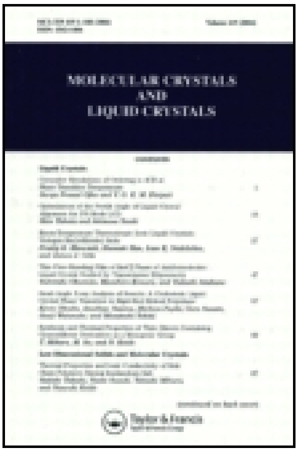

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Fluctuation-Induced Effects in Smectic Films with Nontrivial Boundary Conditions

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Fluctuation-Induced Effects in Smectic Films with Nontrivial Boundary Conditions

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This work generalizes Li-Kardar functional integral approach to a smectic film, wetting the boundary of the isotropic liquid phase. Isotropic-smectic A (IA) interface is considered as an "internal" (bulk) boundary of the film. Previously we generalized this approach to the wetting smectic film with so called Neumann boundary condition, where normal gradients of the smectic layers' displacements at distorted IA-interface were suppressed. In the present paper a nontrivial boundary condition of the vanishing normal forces at the fluctuating IA-interface is considered. Elastic, fluctuation-induced effects for the wetting smectic film with this boundary condition are found.

Keywords Fluctuation-induced effects; isotropic-smectic A-interface; smectic film; wetting

1. Introduction

It is known that thermotropic liquid crystals (LC) exhibit a wealth of pretransitional surface phenomena [1–4]. Just above the bulk isotropic liquid-smectic A (IA) phase transition the smectic layers are observed close to the external surface, bounding an isotropic phase of a smectic LC [1–4]. The smectic layering is a special case of smectic wetting when the thickness growth of the smectic film (WSF) proceeds via a series of discrete layering transitions [5]. In constructing the interface model of the smectic layering [5] a question appears of mutual influence of thermal displacements of the smectic layers and the IA-interface. Here the IA-interface is defined as a boundary separating the isotropic and the smectic A LC phases and acting as an internal (bulk) boundary of the wetting smectic film (WSF) (see Figure). Note that the problem of thermal elastic, fluctuation-induced effects in the WSF is the so called thermal Casimir effect.

The elastic, fluctuation-induced contribution to the free energy density of a smectic film as a function of its equilibrium thickness was first calculated by Mikheev [6] within a "hydrodynamic" approach. However, that approach allowed consideration of only the strong anchoring case of surface smectic layers at smectic interfaces. Functional integral approach [7,8] gives the fluctuation-induced corrections to the interface Hamiltonians of the so-called "correlated" liquids [8] for

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different types of surface anchoring. However, only trivial zero (Dirichlet) boundary conditions for the elastic displacements of the smectic film were considered in [8].

Previously we studied WSF with suppressed normal gradients of the smectic layers' displacements at the interface (so called Neumann boundary condition) [9]. In this paper we generalize functional integral approach [7,8] for the WSF assuming nontrivial (non-Dirichlet and non-Neumann) boundary condition.

2. Formulation of a Problem. Boundary Conditions

Our purpose is to work out the effect of thermal fluctuations of smectic layers on the effective Hamiltonian of the fluctuating IA-interface (see Figure) in the case of smectic wetting in the vicinity of an external flat bounding surface (substrate). It follows from the known interface models [6,10,11] that the bare effective Hamiltonian of the IA-interface, disregarding the influence of thermal fluctuations of smectic layers, can be written as [5]

$$H_{int}[h(\mathbf{y})] = \int_S d^2\mathbf{y} \left\{ V_{int}(h(\mathbf{y})) + \frac{\gamma_{\mathrm{IA}}}{2}(\nabla h(\mathbf{y}))^2 + \frac{W_{\mathrm{IA}}}{2}(\nabla h(\mathbf{y}) - \nabla\tilde{u}(\mathbf{y}, h(\mathbf{y})))^2 \right\}, \quad (1)$$

where $h(\mathbf{y})$ is the local thickness of WSF determined as local removal of the IA-interface from the wetted surface (see Figure); $V_{int}(h(\mathbf{y}))$ is the bare potential of interaction of the IA-interface with a flat bounding surface (substrate) and with smectic layers; γ_{IA} is the bare surface tension of the IA-interface; W_{IA} is the bare amplitude of the potential of the IA-interface local orientation at distorted smectic layers ($W_{\mathrm{IA}} > 0$); $\tilde{u}(\mathbf{y}, h(\mathbf{y}))$ is the elastic thermal displacement in a point ($\mathbf{y}$, $h(\mathbf{y})$) (see below); S is the area of an external flat bounding substrate.

We specify two steps of the functional integral formalism [7,8] as applied to the considered problem. First, the fluctuation displacements of the smectic layers are described by the bulk Grinstein-Pelcovits Hamiltonian [12]. Taken in the quadratic approximation it reads

$$H_0[\tilde{u}(\mathrm{z}, \mathbf{x})] = \int_S d^2x \int dz \, \frac{C_{33}}{2} \left\{ (\partial_z \tilde{u}(\mathbf{x}, z))^2 + \lambda_0^2 \left(\nabla^2 \tilde{u}(\mathbf{x}, z)\right)^2 \right\}, \quad (2)$$

where $\tilde{u}(\mathbf{x}, z)$ is the non-uniform elastic thermal displacement of the smectic layer in a point ($\mathbf{x}$, z); (is the gradient operating in a plane of a wetted surface; $\partial_z \equiv \partial/\partial z$; C_{33} is the compression modulus of smectic layers; λ_0 is the deGennes elastic "cross-length" [13]. Second, the boundary conditions for the elastic displacements $\tilde{u}(\mathbf{x}, z)$ at two surfaces bounding the smectic film are regarded as perturbations acting on an unperturbed bulk system. The boundary conditions are imposed by inserting auxiliary fluctuating fields and by using an integral representation for the δ-function through these auxiliary fields.

In order to develop the approach to WSF [7,8] we assume a strong anchoring of the surface smectic layer at the substrate with coordinate $z = 0$: $\tilde{u}(\mathbf{x}, 0) = 0$. In addition, we assume that normal forces [14,15], acting per unit area of the IA-interface, vanish at the interface

$$\sigma_{ik}\big|_{z=h(\mathbf{y})} \, \boldsymbol{n}_k = f \boldsymbol{n}_i. \quad (3)$$

Here σ_{ik} is the internal smectic stress tensor, f is the sum of additional normal forces acting on the IA-interface, and $\boldsymbol{n}$ is the local normal to the IA-interface. Note that f is obtained from variation of the interface Hamiltonian (1) by infinitesimally small normal displacement $\delta\zeta$ of the interface.

3. Basic Assumptions

Now we introduce the assumptions upon which the proposed generalized functional integral approach is constructed. It is assumed that the IA-interface is essentially more "soft" than the "surface" smectic layer (the smectic layer with the same average position h [6,13]). Accordingly, the following conditions are satisfied

$$\gamma_{\text{IA}} \ll C_{33}\lambda_0, \quad W_{\text{IA}} \ll C_{33}\lambda_0 \quad \text{and} \quad |V''_{int}| \ll C_{33}/h. \tag{4}$$

As shown below (see Sec. 7), the conditions (4) imply that the mean square distortions δh and $\tilde{u}$, reaching the IA-interface, satisfy the following inequalities

$$\langle \tilde{u}^2 \rangle_{z=h(y)} \ll \langle \delta h^2 \rangle \quad \text{and} \quad \langle \delta h^2 \rangle / h^2 \ll 1. \tag{5}$$

We shall consider the static elastic distortions in WSF, which give rise the long-range fluctuations in the smectic film [6], and neglect the density change caused by deformations [14]. Each point at the bounding surfaces is described by three-dimensional radius-vectors

$$\mathbf{r}_1(\mathbf{x}) = (\mathbf{x}, 0), \quad \mathbf{r}_2(\mathbf{y}) = (\mathbf{y}, h + \delta h(\mathbf{y})), \tag{6}$$

where **x, y** are the two-dimensional radius-vectors in the plane of each of the surfaces, respectively; $\delta h(\mathbf{y})$ is the non-uniform fluctuation distortion of the IA-interface relative to its equilibrium position $z = h$ ($\int d^2y \; \delta h(\mathbf{y}) = 0$, see Figure). Then, each point at the surface of equilibrium positions is described by three-dimensional radius-vector $\tilde{\mathbf{r}}_2(\mathbf{y}) = (\mathbf{y}, h)$.

Let us introduce the auxiliary fluctuating fields: $\Omega_1(\mathbf{x})$ at the substrate and $\Omega_2(\mathbf{y})$ at the IA-interface. Then the Dirichlet boundary condition at the flat bounding surface takes the form

$$\delta(\tilde{u}(0, \mathbf{x})) = \int D\Omega_1(\mathbf{x}) \exp\left[i \int d^2x \, \Omega_1(\mathbf{x}) \, \tilde{u}(\mathbf{r}_1(\mathbf{x})) \right]. \tag{7}$$

Using (2), (4–5), one can also write

$$\sigma_{zz}(\mathbf{y}, z = h(\mathbf{y})) \approx C_{33}(\partial \tilde{u}/\partial z)_{z=h(\mathbf{y})}, \quad \mathbf{n}(\mathbf{y}) \approx \mathbf{e}_z / \sqrt{1 + (\nabla \delta h)^2}, \tag{8}$$

and for the normal gradient of thermal elastic displacements at the IA-interface

$$\nabla_{\mathbf{m}(\mathbf{y})} \tilde{u}(\mathbf{y}, h(\mathbf{y})) \approx \nabla_{z_y} \tilde{u}(\mathbf{y}, z)\big|_{z=h(\mathbf{y})} \equiv \nabla_{z_y} \tilde{u}(\mathbf{r}_2(\mathbf{y})).$$

Thus, after variation of H_{int} with respect to $\delta\zeta$ the boundary condition (3) becomes reduced to

$$(C_{33}(\partial\tilde{u}/\partial z)_{z=h(\mathbf{y})} - [\gamma_{\mathrm{IA}}\nabla^2\delta h + W_{\mathrm{IA}}\nabla^2(\delta h(\mathbf{y}) - \tilde{u}(\mathbf{r}_2(\mathbf{y}))) + V'_{int}(h(\mathbf{y}))]) / \sqrt{1+(\nabla\delta h)^2} = 0. \quad (9)$$

From a comparison of the Neumann boundary condition at the fluctuating IA-interface in [9] ($(\partial\tilde{u}/\partial z)_{z=h(\mathbf{y})} = 0$) and boundary conditions (3), (9) it is evident that the surface tension, γ_{IA}, of the IA-interface was omitted in [9] and [7,8]. In the present paper, starting from expressions (3) and (9), the surface tension γ_{IA} is taken into account. Clearly, γ_{IA} is treated as a small parameter, which implies that $\gamma_{\mathrm{IA}}/C_{33}\lambda_0 << 1$. However, it is for the first time in the present paper that the elastic, fluctuation-induced effects in WSF are obtained for the case of nontrivial boundary condition (3), (9). In addition, a general expression for the correlation function of thermally induced smectic layer displacements in WSF, close to the IA-interface, is derived (C.8). Finally, the leading contribution in γ_{IA} to the two-dimensional Fourier transform of this correlation function is found.

To separate the variables δh and $\tilde{u}(\mathbf{y}, h(\mathbf{y}))$ we introduce the relative thermal displacements $\psi(\mathbf{y}) = \delta h(\mathbf{y}) - \tilde{u}(\mathbf{y}, h(\mathbf{y}))$ of the IA-interface (so called roughening fluctuations, see [5,6,9]) and change the variables from δh to ψ in (1) and (9). Before making further calculations we simplify the interface Hamiltonian (1). This is achieved, in analogy to [9], by eliminating the 'fast' parts of ψ and $\tilde{u}$ with the renormalization-group (RG) procedure [16]. Note that RG generates the terms proportional to V'_{int}, W_{IA} and $\gamma_{\mathrm{IA}}\partial^2\psi(\mathbf{y})/\partial y^2$, which are absent in H_{int}. However, as can be shown (again in analogy to [16]), under conditions (4–5) all these terms are irrelevant and can be neglected. The relevant part of the bare interface Hamiltonian (1), expressed in terms of ψ and under conditions (4–5), is then given by

$$H_{int}[\psi(\mathbf{y})] = \int_S d^2y \left\{ V_{int}(h+\psi(\mathbf{y})) + \frac{\tilde{\gamma}_{\mathrm{IA}}}{2}(\nabla\psi(\mathbf{y}))^2 \right\}, \quad (10)$$

where $\tilde{\gamma}_{\mathrm{IA}} = \gamma_{\mathrm{IA}} + W_{\mathrm{IA}}$ is the bare stiffness of the IA-interface.

When expressing (9) through integral representation of the δ-function, the proper integration measure: $\sqrt{1+(\nabla\delta h)^2}$ should be inserted under the integrals [17]. But this measure is cancelled by the factor $\sqrt{1+(\nabla\delta h)^2}$ in (9). Thus expanding $\tilde{u}(\mathbf{y}, h(\mathbf{y}))$ in powers of $\psi(\mathbf{y})$ up to the second order and retaining only the relevant terms, the boundary condition (9) can be written as

$$\int D\Omega_2(\mathbf{y}) \exp\left[i \int d^2y\, \Omega_2(\mathbf{y})\, \Pi(\mathbf{r}_2(\mathbf{y})) \right], \quad (11)$$

where

$$\Pi(\mathbf{r}_2(\mathbf{y})) = \left(\frac{\partial\tilde{u}}{\partial z}\right)_{\tilde{\mathbf{r}}_2(y)} + \left(\frac{\partial^2\tilde{u}}{\partial z^2}\right)_{\tilde{\mathbf{r}}_2(\mathbf{y})}\psi(\mathbf{y}) + \left(\frac{\partial^3\tilde{u}}{\partial z^3}\right)_{\tilde{\mathbf{r}}_2(\mathbf{y})}\psi^2(\mathbf{y})/2 - \frac{\gamma_{\mathrm{IA}}}{C_{33}}\left(\frac{\partial^2\tilde{u}}{\partial y^2}\right)_{\tilde{\mathbf{r}}_2(\mathbf{y})} - \frac{\gamma_{\mathrm{IA}}}{C_{33}}\left(\frac{\partial}{\partial z}\frac{\partial^2\tilde{u}}{\partial y^2}\right)_{\tilde{\mathbf{r}}_2(\mathbf{y})}\psi(\mathbf{y}) - \frac{\gamma_{\mathrm{IA}}}{C_{33}}\left(\frac{\partial^2}{\partial z^2}\frac{\partial^2\tilde{u}}{\partial y^2}\right)_{\tilde{\mathbf{r}}_2(\mathbf{y})}\psi^2(\mathbf{y})/2.$$

4. General Expression for the Elastic, Fluctuation-Induced Contribution to H_{int}

By generalizing [7–9] and using (2,6,7,11) we obtain the equation for calculating the fluctuation-induced contribution, H_{eff}, to the effective interface Hamiltonian (10). It reads

$$\begin{aligned}\exp\left[-H_{eff}/k_BT\right] &= Z_0^{-1}\int D\tilde{u}(\mathbf{r})\,\exp[-H_0[\tilde{u}]/k_BT]\iint D\Omega_1(\mathbf{x})\,D\Omega_2(\mathbf{y})\\ &\times\, exp\left[i\int d^2x\,\Omega_1(\mathbf{x})\,\tilde{u}(\mathbf{r}_1(\mathbf{x}))\,+\,i\int d^2y\,\Omega_2(\mathbf{y})\,\Pi(\mathbf{r}_2(\mathbf{y}))\right],\end{aligned}\tag{12}$$

where $Z_0=\int D\tilde{u}(\mathbf{r})\,\exp[-H_0[\tilde{u}]/k_BT]$. Note that the expression (12) is a functional integral over thermal displacements of smectic layers and over auxiliary fields. Also note that $G_b(\mathbf{r})=\langle\tilde{u}(0)\,\tilde{u}(\mathbf{r})\rangle_0$ is the two-point correlation function of the bulk smectic, where $\langle\cdots\rangle_0= Z_0^{-1}\int D\tilde{u}(\mathbf{r})\,(\cdots)\exp[-H_0[\tilde{u}]/k_BT]$. In our case

$$G_b(\mathbf{y}-\mathbf{x},\,z_{\mathbf{y}}-z_{\mathbf{x}})\,=\,\frac{k_BT}{C_{33}}\int\frac{d^2q}{(2\,\pi)^2}\,\frac{\exp\left(i\,\mathbf{q}\,(\mathbf{y}-\mathbf{x})\,\exp\left(-\,\lambda_0q^2z\right)\right.}{2\lambda_0q^2},\tag{13}$$

where, following the choice (6), we assume that $z_{\mathbf{y}}\geq z_{\mathbf{x}}$ and introduce $z=z_{\mathbf{y}}-z_{\mathbf{x}}$. Accordingly, $\nabla_{z_y}G_b(\mathbf{y}-\mathbf{x},\,z_{\mathbf{y}}-z_{\mathbf{x}})$ and $\nabla_{z_{\mathbf{x}}}\nabla_{z_y}G_b(\mathbf{y}-\mathbf{x},\,z_{\mathbf{y}}-z_{\mathbf{x}})$ can be found in analogy to [9].

To proceed further, we expand the expression in braces, on the right-hand side of Eq. (12), in powers of i. Then by performing the Gaussian integration over the elastic variables we find

$$\exp\left[-H_{eff}/k_BT\right]\;=\;\iint D\Omega_1(\mathbf{x})\,D\Omega_2(\mathbf{y})\;\exp[-H_1[\Omega_1(\mathbf{x}),\,\Omega_2(\mathbf{y})]],\tag{14}$$

where the effective Hamiltonian of the two-component field $\Omega\equiv(\Omega_1(\mathbf{x}),\,\Omega_2(\mathbf{y}))$ is given by

$$\begin{aligned}H_1[\Omega]=\frac{1}{2}\iint d^2x\,d^2y\,\{&\Omega_1(\mathbf{x})\,G_b(\mathbf{y}-\mathbf{x},\,0)\,\Omega_1(\mathbf{y})\\ &+\Omega_1(\mathbf{x})\,[\widehat{D}_{12}(\mathbf{y})\,G_b(\mathbf{y}-\mathbf{x},\,h)]\,\Omega_2(\mathbf{y})+\\ &+\Omega_1(\mathbf{y})[\widehat{D}_{12}(\mathbf{x})\,G_b(\mathbf{x}-\mathbf{y},\,h)]\Omega_2(\mathbf{x})\\ &+\Omega_2(\mathbf{x})[\widehat{D}_{22}(\mathbf{x},\mathbf{y})\,G_b(\mathbf{y}-\mathbf{x},\,z)]_{z=0}\Omega_2(\mathbf{y})\}\equiv\Omega M\Omega^T,\end{aligned}\tag{15}$$

and where

$$\begin{aligned}\widehat{D}_{12}(\mathbf{y})&=[1+\psi(\mathbf{y})\,\partial/\partial h+(\psi^2(\mathbf{y})/2)\partial^2/\partial h^2]\;[\partial/\partial h-(\gamma_{\mathrm{IA}}/C_{33})\,\partial^2/\partial y^2],\\ \widehat{D}_{22}(\mathbf{x},\mathbf{y})&=\lfloor 1+((\psi(\mathbf{y})-\psi(\mathbf{x}))^2/2)\,\partial^2/\partial z^2\rfloor\\ &\quad\times[-\partial^2/\partial z^2+(\gamma_{\mathrm{IA}}/C_{33})^{\,2}(\partial^2/\partial y^2)(\partial^2/\partial x^2)].\end{aligned}$$

Note that the matrix M in Eq. (15) is a functional of the radius-vectors $r_1(\mathbf{x})$ and $r_2(\mathbf{y})$.

When deriving (15) we make use of the equality $\langle \tilde{u}(r_1)\cdots \tilde{u}(r_{2m})\tilde{u}(r_{2m+1})\rangle_0 = 0$. In obtaining the part $(\ldots)$ between $\Omega_2(\mathbf{x})$ and $\Omega_2(\mathbf{y})$ we observed that the term linear in $(\psi(\mathbf{y})$ -$\psi(\mathbf{x}))$ vanishes, which can be easily verified using (13). The quadratic form of $H_1[\Omega]$ allows us to perform the integration over fields Ω_i in (14). Thus we obtain the general expression for the effective Hamiltonian, describing the additional elastic, fluctuation-induced interaction between the perturbed IA-interface and the flat surface bounding the WSF. It reads

$$H_{eff}[\mathbf{r}_1(\mathbf{x}), \mathbf{r}_2(\mathbf{y})] \ = (k_B T/2) \ \ln \mathrm{Det}\{M[\mathbf{r}_1(\mathbf{x}), \mathbf{r}_2(\mathbf{y})]/\pi\}, \tag{16}$$

where

$$M(\mathbf{x},\ \mathbf{y}) \ = \frac{1}{2}\begin{pmatrix} G_b(\mathbf{y}-\mathbf{x},\,0) & \widehat{D}_{12}(\mathbf{y})\,G_b(\mathbf{y}-\mathbf{x},\,h) \\ \widehat{D}_{12}(\mathbf{x})\,G_b(\mathbf{x}-\mathbf{y},\,h) & \widehat{D}_{22}(\mathbf{x},\mathbf{y})\,G_b(\mathbf{y}-\mathbf{x},\,z)\Big|_{z=0} \end{pmatrix}. \tag{17}$$

5. H_{eff} for Small Distortions of the IA-Interface

For small $\delta h(\mathbf{y})$ and $\psi(\mathbf{y})$ (see Sec. 7), the matrix $M(\mathbf{x},\ \mathbf{y})$ can be approximated by the second order expansion in $\psi(\mathbf{x})$. In this expansion $M(\mathbf{x},\ \mathbf{y}) = M_0(\mathbf{x},\ \mathbf{y}) + \delta M(\mathbf{x},\ \mathbf{y})$, where $M_0(\mathbf{x},\ \mathbf{y})$ is the functional matrix for a flat bounding surface and $\delta M(\mathbf{x},\ \mathbf{y})$ is the correction caused by relative fluctuations of the IA-interface. Note that a two-dimensional Fourier transform of M (denoted $\widetilde{M}$, see Appendix A) can be written down as $\widetilde{M} = \widetilde{M}_0 \widetilde{W}$ [7,8], where $\widetilde{W} = 1 + \widetilde{M}_0^{-1}\delta\widetilde{M}$. Then the effective Hamiltonian (16) can be decomposed as $H_{eff} = H_{flat} + H_{corr}$, where (compare with [7–9])

$$H_{flat} \ = (k_B T/2) \ \ln \mathrm{Det}\left\{ \widetilde{M}_0/\pi \right\} \tag{18}$$

is the effective Hamiltonian, describing the elastic fluctuation-induced interaction between unperturbed (flat) IA-interface and the flat external surface, bounding the WSF. The second of the two terms is the additional elastic fluctuation-induced contribution to the H_{eff}, caused by relative thermal distortions of the IA-interface. It reads

$$H_{corr} = (k_B T/2) \ \ln \mathrm{Det}\left\{ \widetilde{W} \right\}. \tag{19}$$

After some calculations (see Appendix A) we obtain the following h-depended contribution to H_{flat}

$$H_{flat}(h) = S\ \frac{k_B T}{2} \int \frac{d^2k}{(2\,\pi)^2} \ln\left[1 - \chi_M\ \exp\left(-2\,\lambda_0 q^2 h\right)\right] = S\ V_{Mikh}(h), \tag{20}$$

where

$$V_{Mikh}(h) = -\ k_B T\ \mathrm{Li}_2(\chi_M)/(16\,\pi\lambda_0 h), \qquad \left(\mathrm{Li}_n(\nu) = \sum_{k=1}^{\infty} \nu^k/k^n\right), \tag{21}$$

is the long-range "repulsive" contribution to $V_{int}(h)$, or more accurately, to the free energy density of the WSF with equilibrium thickness h ($\chi_M < 0 \rightarrow \mathrm{Li}_2(\chi_M) < 0$). Note that the long-range "repulsive" potential $V_{Mikh}(h)$ coincides in the limit of $\gamma_0 \gg C_{33}\lambda_0$ with the "hydrodynamic" Mikheev interaction [6], where γ_0 is the stiffness of external boundary of WSF. This result confirms the correctness of the boundary conditions imposed in Sec. 2.

6. Local and Nonlocal Corrections to H_{corr}

The evaluation of H_{corr} from Eq. (19) is given in the Appendix B along with the resulting general expression for H_{corr} (B.14). It is possible to decompose H_{corr} into local and nonlocal contributions (see Appendix B). The local contribution is given by

$$\begin{aligned} H_{corr}^{(loc)} = \int d^2y\,\psi^2(\mathbf{y}) \Bigg[\frac{k_B T}{2} \int \frac{d^2k}{(2\pi)^2} \Bigg\{ - \frac{[\widehat{D}_2(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},\,h)]}{4\,D_0(\mathbf{k})} \frac{\partial}{\partial h} \\ \times [\widehat{D}_{hh}(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},\,h)] - ([\widehat{D}_2(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},\,h)]^2 \\ + \widetilde{G}_b(\mathbf{k},\,0)\,[\widehat{D}_{zz}(\mathbf{k})\,\widetilde{G}_b(\mathbf{k},\,z)]_{z=0}) \frac{[\widehat{D}_{hh}(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},\,h)]^2}{16\,D_0^2(\mathbf{k})} \Bigg\} \Bigg]. \end{aligned} \tag{22}$$

Note that the expression in square brackets is the fluctuation-induced correction to the gap of mode ψ. Inserting (A.1), (A.3), (B.12), (B.4) in (22) and extending the integration over k from 0 up to ∞ (due to the presence of rapidly decreasing exponential), we find

$$H_{corr}^{(loc)}[\,\psi(\mathbf{y})\,] = -k_B T\,\frac{\mathrm{Li}_2(\chi_M)}{8\,\pi\lambda_0 h^3} \int d^2y\,\frac{\psi^2(\mathbf{y})}{2} = V''_{Mikh}(h) \int d^2y \frac{\psi^2(\mathbf{y})}{2}. \tag{23}$$

If we keep the term linear in $\psi(\mathbf{y})$ in $A(\mathbf{k},\mathbf{k})$, Eq. (B.13), an additional contribution to $H_{corr}^{(loc)}$ of the form $V'_{Mikh}(h)\int d^2y\,\psi(\mathbf{y})$ will appear. We can formally combine this term with the leading corrections $V_{Mikh}(h)$ and $(1/2)\,V''_{Mikh}(h)\,\psi^2(\mathbf{y})$ into the total long-range elastic fluctuation-induced potential. It gives

$$V_{Mikh}^{(loc)}(h + \psi(\mathbf{y})) = -\,k_B T\,\mathrm{Li}_2(\chi_M)/(16\,\pi\lambda_0(h + \psi(\mathbf{y}))), \tag{24}$$

which accounts for the repulsion of the distorted IA-interface from the flat bounding surface.

In turn we obtain the corresponding total local contribution to the interface Hamiltonian (10):

$$H_{Mikh}^{(loc)}(h + \psi(\mathbf{y})) = \int d^2y V_{Mikh}^{(loc)}(h + \psi(\mathbf{y})). \tag{25}$$

The nonlocal contribution to H_{corr} is given in Appendix B (Eq. B.15). In case of weak nonlocality ($\psi(\mathbf{x}) \approx \psi(\mathbf{y}) + ((_{\mathbf{y}}\psi(\mathbf{y}))\;(\mathbf{x} - \mathbf{y})$), the nonlocal contribution (B.15) describes the occurrence of an elastic fluctuation-induced correction $\delta\gamma_{el}$ to the stiffness of the relative capillary mode and is given by

$$H_{corr}^{(nonloc)}[\,\psi(\mathbf{y})\,] \approx \int d^2y\ \delta\gamma_{el}(h)(\nabla\psi(\mathbf{y}))^2/2\,, \tag{26}$$

where

$$\begin{aligned}\delta\gamma_{el}(h) = k_BT\,/(8\pi^2)\ &\int d^2x\,(\mathbf{y}-\mathbf{x})^2\Bigg[\int kdk\ J_0(k|(\mathbf{y}-\mathbf{x})|)\ (\lambda_0k^2)^3\\ &\times\int\frac{qdq}{\lambda_0q^2}\ \frac{J_0(q|(\mathbf{y}-\mathbf{x})|)}{1-\chi_M\exp(-2\lambda_0q^2h)}+\Bigg(\int qdq\ J_0(q|(\mathbf{y}-\mathbf{x})|)\\ &\times\frac{\lambda_0q^2\chi_M\exp(-2\lambda_0q^2h)}{1-\chi_M\exp(-2\lambda_0q^2h)}\Bigg)^2+\int qdq\ J_0(q|(\mathbf{y}-\mathbf{x})|)\\ &\times\frac{(\lambda_0q^2)^2\chi_M\exp(-\lambda_0q^2h)}{1-\chi_M\exp(-2\lambda_0q^2h)}\int kdk\ \frac{J_0(k|(\mathbf{y}-\mathbf{x})|)\ \exp(-\lambda_0k^2h)}{1-\chi_M\exp(-2\lambda_0k^2h)}\Bigg].\end{aligned} \tag{27}$$

It is obvious that the integrals over a wave vector in the first term of square brackets in (27) are defined by the cutoff parameters. Thus the corresponding contribution to $\delta\gamma_{el}$ is traditionally included into a redefined stiffness of the mode ψ (see [16]). The other two terms in (27) give h-dependent (dimensional) correction $\delta\gamma_{el}(h)$ to the stiffness of the mode ψ, caused by thermal displacements of smectic layers in WSF. The integrals in these terms can be calculated approximately in view of their fast convergence due to rapidly decreasing exponential factor exp(- λ_0q^2h). Indeed, the wave vectors $q\lesssim q_C = 1\big/\sqrt{\lambda_0h}$ provide the leading contribution to these integrals and it is possible to omit the decreasing exponentials in denominators, which reduces the integrals to tabulated ones. Making the substitutions $Q = q/q_C$ and $\rho = q_C\mid\mathbf{y}-\mathbf{x}|$, we find

$$\begin{aligned}\delta\gamma_{el}(h) \approx \frac{k_BT}{4\pi}\frac{1}{h^2}\ &\int\rho^3d\rho\left[\left(\chi_M\int Q^3dQ\ J_0(Q\rho)\ \exp(-2Q^2)\right)^2\right.\\ &\left.+\,\chi_M\int Q_1^5dQ_1\ J_0(Q_1\rho)\exp(-Q_1^2)\int Q_2\,dQ_2\ J_0(Q_2\rho)\exp(-Q_2^2)\right].\end{aligned} \tag{28}$$

and finally get

$$\delta\gamma_{el}(h) \approx \frac{k_BT}{16\pi}\frac{\chi_M^2}{h^2}\left(\frac{1}{4}-\frac{1}{\chi_M}\right) > 0. \tag{29}$$

In the limit of $\gamma_{\mathrm{IA}} \ll C_{33}\lambda_0(\chi_M \to -1)$ the corrections (23), (28–29) along with (23), (28) coincide with the results in [9].

7. Conclusion

We start by reviewing all the assumptions used here. Let us first illustrate the physical meaning of the inequality (4). Note that the functional integral approach allows calculating the correlation functions of fluctuating fields in bounded systems with long-range correlations [18] (see Appendix C). The leading contribution to the two-dimensional Fourier transform of the correlation $G_{\text{IA}}^{(u)} = \langle \tilde{u}(\mathbf{x}, h(\mathbf{x}))\, \tilde{u}(\mathbf{y}, h(\mathbf{y}))\rangle$ becomes reduced to

$$\widetilde{G}_{\text{IA}}^{(u,0)}(\mathbf{q}) = \frac{k_B T}{S\, C_{33}} \frac{1}{2\lambda_0 q^2} \frac{(1-\chi_M)(1-\exp(-2\lambda_0 q^2 h))}{1-\chi_M \exp(-2\lambda_0 q^2 h)}. \tag{30}$$

The Fourier transformations of the non-uniform fluctuations $\delta h(\mathbf{y})$ and $\psi(\mathbf{y})$ are $\delta h(\mathbf{y}) = \sum'_{\mathbf{q}} h_{\mathbf{q}} \exp(i\mathbf{q}\mathbf{y})$ and $\psi(\mathbf{y}) = \sum'_{\mathbf{q}} \psi_{\mathbf{q}} \exp(i\mathbf{q}\mathbf{y})$, respectively. Using (10) and (23) one can also define the Fourier transform of the correlator $\langle \psi_q \psi_{-q}\rangle$ as

$$\langle \psi_{\mathbf{q}} \psi_{-\mathbf{q}}\rangle \cong k_B T \big/ \left(V''_{int}(h) + V''_{Mikh}(h) + (\tilde{\gamma}_{\text{IA}} + \delta\gamma_{el}(h))\, q^2\right). \tag{31}$$

Now, by a direct comparison of (30) and (31) it follows that the inequality $\widetilde{G}_{\text{IA}}^{(u,0)}(\mathbf{q}) \ll \langle \psi_q \psi_{-q}\rangle$ is valid under conditions (4). It means that the inequality $\widetilde{G}_{\text{IA}}^{(u,0)}(\mathbf{q}) \ll \langle h_q h_{-q}\rangle$ is also satisfied. Hence, the left inequality in (5) is satisfied and

$$\langle h_{\mathbf{q}} h_{-\mathbf{q}}\rangle \approx \langle \psi_{\mathbf{q}} \psi_{-\mathbf{q}}\rangle. \tag{32}$$

We conclude that the condition (4) of "softness" of the IA-interface means, in particular, that the thermal capillary fluctuations δh dominate at the IA-interface and in the roughening fluctuations of IA-interface. Using [4] we can estimate $\tilde{\gamma}_{\text{IA}}$through the classical Gibbs-Kelvin equation [5] as $\tilde{\gamma}_{\text{IA}} \sim 10^0\, \text{erg} \cdot \text{cm}^{-2}$, also confirmed in [19]. In turn, for typical bilayer smectic LC [13] we have $\Delta H_{\text{IA}} \sim C_{33} \sim 10^8\, \text{erg} \cdot \text{cm}^{-3}$, $\lambda_0 \sim d_0 \sim 10^{-7}\, \text{cm}$. In this case a simple analysis shows that the first two inequalities in (4) are satisfied. As it was found for $T_{\text{IA}} \simeq 300\, K$, the right inequality in (4) for $V''_{Mikh}(h)$ is also satisfied for arbitrary value of h and the correction (29) to $\tilde{\gamma}_{\text{IA}}$ is negligibly small even for $h \sim d_0$: $\delta\gamma_{el}(h) \sim 10^{-2} \div 10^{-1}\, \text{erg} \cdot \text{cm}^{-2}$. It can be shown that for the remaining terms in the interface potential, as introduced in [5], the inequalities on the right hand side of (4) and (5) are also satisfied for both partial and complete smectic wetting.

We conclude that taking the thermal displacements of the smectic layers in WSF into account results in adding to the interface potential the additional long-range potential $V_{Mikh}^{(loc)}(h + \psi(y))$; it also generates the correction $\delta\gamma_{el}(h)$ to the stiffness of the IA-interface. One can show (see [9]) that these conclusions are also valid in the case of wetting the LC free surface by the smectic A phase. It is important to note that though $\delta\gamma_{el}(h)$ is infinitesimally small, it can be important for understanding the properties of membranes with an infinitesimally small bare surface tension.

Separating out the smooth part $V_0(h)$ of the interfacial potential V_{int} [5] it is not difficult to derive an equation for the temperature dependence of the average WSF thickness $h_0(t)$ ($V'_{0h} = 0$), where $t = (T - T_{\text{IA}})/T_{\text{IA}}$, ($t \geq 0$) is given by

$$t = A\, \exp(-h_0/\xi_C) - \left(1/\Delta H_{\text{IA}}\right) k_B T\, \text{Li}_2(\chi_M) \big/ \left(16\, \pi \lambda_0 h_0^2\right), \tag{33}$$

and where ξ_C is the correlation length in the bulk smectic; A ($A > 0$) is the reduced amplitude of the short-range repulsive interaction [5]. Fitting Figure 1 in [3] with (33) we obtain $A \approx 0.0455$ and $\xi_C \approx 4.5 \cdot 10^{-7}\, \text{cm}$, which is in agreement with [5]. In turn,

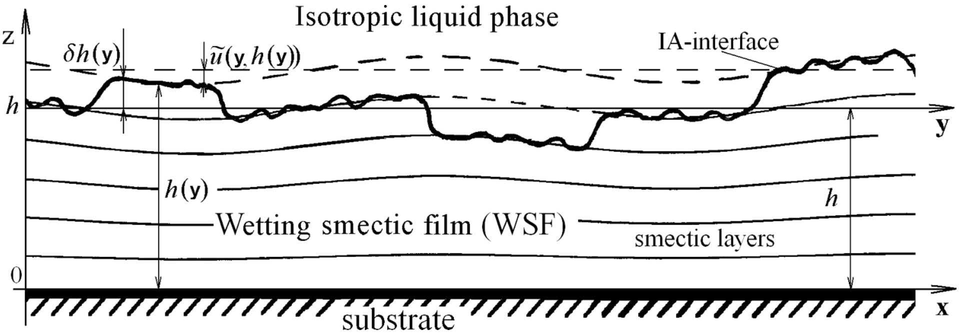

Figure 1. A schematic picture of WSF covering flat bounding surface (substrate). The equilibrium position, h, of the IA-interface is shown by the thin straight line.

the correlators $\langle h_q h_{-q} \rangle$ and (31), respectively, define the IA-interface thickness and the IA-interface-smoothing Gaussian width in structure factors of the x-ray reflectivity from WSF [2,5]. Thus, from a simultaneous fitting of all mentioned experimental dependencies to the same LC compound it would be possible to experimentally determine the value of $\tilde{\gamma}_{\text{IA}}$ and to derive $V''_{Mikh}(h)$ and $\delta\gamma_{el}(h)$ for a comparison with the obtained results.

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Appendix

A *Derivation of the Effective Hamiltonian,* $\mathbf{H_{flat}}$

With (13) the two-dimensional Fourier transforms of the correlation functions in M_0 are

$$\widetilde{G}_b(\mathbf{q},\, 0) \;=\; S^{-1}k_BT/\big(C_{33}2\lambda_0q^2\big) \quad \text{and} \quad \widetilde{G}_b(\mathbf{q},\, h) \;=\; S^{-1}k_BT\exp\left(-\lambda_0q^2h\right)/\big({}_{33}2\lambda_0q^2\big). \tag{A.1}$$

For simplicity we take $S=1$ in all calculations that follow. Then the two-dimensional Fourier-transform of the matrix $M_0(\mathbf{x},\,\mathbf{y})$ reads

$$\widetilde{M}_0(\mathbf{k}) \;=\frac{1}{2}\begin{pmatrix} \widetilde{G}_b(\mathbf{k},\,0) & \widehat{D}_2(\mathbf{k})\,\widetilde{G}_b(\mathbf{k},\,h) \\ \widehat{D}_2(\mathbf{k},\,h)\,\widetilde{G}_b(\mathbf{k},\,h) & \widehat{D}_{zz}(\mathbf{k})\,\widetilde{G}_b(\mathbf{k},\,z)\Big|_{z=0} \end{pmatrix}, \tag{A.2}$$

where

$$\widehat{D}_2(\mathbf{k},h) = \partial/\partial h \;+\; (\gamma_{\text{IA}}/C_{33})\,k^2 \quad \text{and} \quad \widehat{D}_{zz}(\mathbf{k}) \;=\; -\partial^2/\partial z^2 \;+\; (\gamma_{\text{IA}}/C_{33})^2\,k^4. \tag{A.3}$$

Taking discrete wave vector notation, $\mathbf{k}_i$, each block in (A.2) can be viewed as an infinite-dimensional matrix (see [9] for example). After an even number of rearrangements and with the help of (A.1), the determinant of the matrix $\widetilde{M}_0(k)$ is easily calculated giving

$$\mathrm{Det}\,\widetilde{M}_0 = \prod_i \left(\frac{1}{4}\frac{k_B T}{C_{33}}\right)^2 \left(1 \; - \left(\frac{\gamma_{IA}}{C_{33}\lambda_0}\right)^2\right) \left[1 - \chi_M \exp\left(-2\lambda_0 k_i^2 h\right)\right], \qquad (A.4)$$

where

$$\chi_M = (\gamma_{IA} - C_{33}\lambda_0)/(\gamma_{IA} + C_{33}\lambda_0) < 0 \qquad (A.5)$$

is the Mikheev parameter [6]. Substituting (A.4) to (18) and passing from summation over $\mathbf{k}_i$ back to integration, it is easy to find the h-depended contribution to H_{flat}, given by Eq. (20).

B *Derivation of the Effective Hamiltonian* $\mathrm{H}_{\mathrm{corr}}$*, Eq. (19)*

In analogy to [7–9], after carrying out the two-dimensional Fourier transform of matrix δM, we obtain

$$\delta\widetilde{M}(\mathbf{k},\ \mathbf{q}) = \frac{1}{2}\begin{pmatrix} 0 & A(\mathbf{k},\ \mathbf{q}) \\ A(\mathbf{q},\mathbf{k}) & B(\mathbf{k},\ \mathbf{q}) \end{pmatrix}, \qquad (B.1)$$

$$\begin{aligned} A(\mathbf{k},\ \mathbf{q}) \;&= \iint d^2x\, d^2y\ \exp(-i\,\mathbf{k}\mathbf{y})\exp(i\,\mathbf{q}\mathbf{x})\left[\widehat{D}_{hh}(\mathbf{y},\,h)\,G_b(\mathbf{y}-\mathbf{x},\,h)\right]\ \psi(\mathbf{y}) \\ &+ \iint d^2x\, d^2y\ \exp(-i\,\mathbf{k}\mathbf{y})\exp(i\,\mathbf{q}\mathbf{x})\Big(\partial\left[\widehat{D}_{hh}(\mathbf{y},\,h)\,G_b(\mathbf{y}-\mathbf{x},\,h)\right]/\partial h\Big)\ \psi^2(\mathbf{y})\,/2, \end{aligned} \qquad (B.2)$$

$$\begin{aligned} B(\mathbf{k},\ \mathbf{q}) = &\iint d^2x\, d^2y\ \exp(-i\,\mathbf{k}\mathbf{y})\exp(i\,\mathbf{q}\mathbf{x}) \\ &\times\left[\widehat{D}_{4z}(\mathbf{x},\,\mathbf{y})\,G_b(\mathbf{y}-\mathbf{x},\,z)\right]_{z=0}(\psi(\mathbf{y})-\psi(\mathbf{x}))^2/2, \end{aligned} \qquad (B.3)$$

and

$$\widehat{D}_{4z}(\mathbf{x},\mathbf{y}) \;= -\frac{\partial^4}{\partial z^4} + \left(\frac{\gamma_{\mathrm{IA}}}{C_{33}}\right)^2 \frac{\partial^2}{\partial y^2}\frac{\partial^2}{\partial x^2}\frac{\partial^2}{\partial z^2}, \quad \widehat{D}_{hh}(\mathbf{y},h) \;= \frac{\partial^2}{\partial h^2} - \frac{\gamma_{\mathrm{IA}}}{C_{33}}\frac{\partial^2}{\partial y^2}\frac{\partial}{\partial h}. \quad (B.4)$$

Using (A.2) one can now invert the matrix $\widetilde{M}_0$ by switching again to the discrete wave vector notation $\mathbf{k}_i$ (note that $\mathbf{k}_i = \mathbf{q}_i$). Then, each block in $\delta\widetilde{M}(\mathbf{k},\mathbf{q})$, Eq. (B.1), and in $\widetilde{M}_0^{-1}(\mathbf{k})$ should be understood as an infinite-dimensional matrix (see [9] for example). Using (B.1) one can now obtain Eq. (19), where the blocks $\widetilde{W}_{\alpha\beta}(\mathbf{k}_i,\ \mathbf{q}_j)$ are given by

$$\widetilde{W}_{11}(\mathbf{k}_i,\ \mathbf{q}_j) \;=\; \begin{bmatrix} 1 + c(\mathbf{k}_i,\ \mathbf{q}_i) & c(\mathbf{k}_i,\ \mathbf{q}_j) \\ c(\mathbf{k}_j,\ \mathbf{q}_i) & 1 + c(\mathbf{k}_j,\ \mathbf{q}_j) \end{bmatrix}, \qquad (B.5)$$

$$c(\mathbf{k}_i,\ \mathbf{q}_j) \;=\; -A(\mathbf{k}_i,\ \mathbf{q}_j)\ \left[\widehat{D}_2(\mathbf{k}_i,\ h)\,\widetilde{G}_b(\mathbf{k}_i,\ h)\right]/(4\,D_0(\mathbf{k}_i)), \qquad (B.6)$$

$$\widetilde{W}_{12}\,(\mathbf{k}_i,\ \mathbf{q}_j) \;= r\,(\mathbf{k}_i,\ \mathbf{q}_j) - B(\mathbf{k}_i,\ \mathbf{q}_j)\ \left[\widehat{D}_2(\mathbf{k}_i,\ h)\,\widetilde{G}_b(\mathbf{k}_i,\ h)\right]/(4\,D_0(\mathbf{k}_i)), \qquad (B.7)$$

$$r\,(\mathbf{k}_i,\ \mathbf{q}_j) \;=\; A(\mathbf{k}_i,\ \mathbf{q}_j)\ \left[\widehat{D}_{zz}(\mathbf{k}_i)\,\widetilde{G}_b(\mathbf{k}_i,\ z)\right]_{z=0}/(4\,D_0(\mathbf{k}_i)), \qquad (B.8)$$

$$\widetilde{W}_{21}\,(\mathbf{k}_i,\ \mathbf{q}_j) \;=\; A(\mathbf{k}_i,\ \mathbf{q}_j)\,\widetilde{G}_b(\mathbf{k}_i,\ 0)/(4\,D_0(\mathbf{k}_i)), \qquad (B.9)$$

$$\widetilde{W}_{22}(\mathbf{k}_i,\ \mathbf{q}_j) = \begin{bmatrix} 1 + c(\mathbf{k}_i,\ \mathbf{q}_i) + s(\mathbf{k}_i,\ \mathbf{q}_i) & c(\mathbf{k}_i,\ \mathbf{q}_j) + s(\mathbf{k}_i,\ \mathbf{q}_j) \\ c(\mathbf{k}_j,\ \mathbf{q}_i) + s(\mathbf{k}_j,\ \mathbf{q}_i) & 1 + c(\mathbf{k}_j,\ \mathbf{q}_j) + s(\mathbf{k}_j,\ \mathbf{q}_j) \end{bmatrix}, \qquad (B.10)$$

$$s\,(\mathbf{k}_i,\ \mathbf{q}_j) \;=\; B(\mathbf{k}_i,\ \mathbf{q}_j)\,\widetilde{G}_b(\mathbf{k}_i,\ 0)/(4\,D_0(\mathbf{k}_i)), \qquad (B.11)$$

$$D_0(\mathbf{k}) = (1/16)\,(k_BT/C_{33})^2\,((\gamma_{IA}/(C_{33}\lambda_0))^2 - 1)\ \left[1 - \chi_M \exp(-2\lambda_0 k^2 h)\right]. \qquad (B.12)$$

We now insert the contributions above, containing terms linear in ψ, into Eq. (10). Summing up all these terms and setting the sum to zero: $(\ldots)\ \psi = 0 \rightarrow F'(h) = 0$, gives an equation for the equilibrium WSF thickness subject to thermal fluctuations of the IA-interface and of the smectic layers. This condition is equivalent to the condition $\int d^2y\ \psi(\mathbf{y}) = 0$, which means $\langle\psi\rangle = 0$ and causes the term linear in $\psi(\mathbf{y})$ disappear in $A(\mathbf{k},\,\mathbf{k})$. Using (B.5)–(B.10), we can obtain Det $\widetilde{W}$ up to the second order in $\psi(\mathbf{y})$. It reads

$$\begin{aligned} \text{Det}\,\widetilde{W} \;&=\; 1 + \sum_{\mathbf{k}} \{s(\mathbf{k},\mathbf{k}) + 2c(\mathbf{k},\mathbf{k})\} \\ &- \sum_{\mathbf{k},\mathbf{q}(\mathbf{q}\neq\mathbf{k})} \{c(\mathbf{k},\mathbf{q})\ c(\mathbf{q},\mathbf{k}) + W_{21}(\mathbf{k},\mathbf{q})\ r(\mathbf{q},\mathbf{k})\}\end{aligned}. \qquad (B.13)$$

Substituting (B.13) into (19), expanding the corresponding logarithm of $\psi(\mathbf{x})$ up to the second order, passing from summation over $\mathbf{k}_i$ to integration, and, finally, using (B.2) and (B.3) we obtain:

$$\begin{aligned} H_{corr} = (k_BT/2)\,[&\int d^2k/(2\pi)^2 \iint d^2x\,d^2y\,\{\widetilde{G}_b(\mathbf{k},0)\,\exp(-i\mathbf{k}(\mathbf{y}-\mathbf{x}))\ (4D_0(\mathbf{k}))^{-1} \\ &\times [\widehat{D}_{4z}(\mathbf{x},\mathbf{y})\,G_b(\mathbf{y}-\mathbf{x},z)]_{z=0}(\psi(\mathbf{y})-\psi(\mathbf{x}))^2/2 - 2\,[\widehat{D}_2(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},h)]\ (4D_0(\mathbf{k}))^{-1} \\ &\times \exp(-i\mathbf{k}(\mathbf{y}-\mathbf{x}))\,(\partial[\widehat{D}_{hh}(\mathbf{y},h)\,G_b(\mathbf{y}-\mathbf{x},h)]/\partial h)\,\psi^2(\mathbf{y})/2\} \\ &- \iint \frac{d^2k\,d^2q}{(2\pi)^4}\,\{[\widehat{D}_2(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},h)]\,[\widehat{D}_2(\mathbf{q},h)\,\widetilde{G}_b(\mathbf{q},h)] + \widetilde{G}_b(\mathbf{k},0)\,[\widehat{D}_{zz}(\mathbf{q})\,\widetilde{G}_b(\mathbf{q},z)]_{z=0}\} \\ &\times \iint d^2y\,d^2v_1\ \exp(i\mathbf{q}\mathbf{y} - i\mathbf{k}\mathbf{v}_1))\,\psi(\mathbf{y})\,[\widehat{D}_{hh}(\mathbf{y},h)\,G_b(\mathbf{v}_1-\mathbf{y},h)]\,(16D_0(\mathbf{k})D_0(\mathbf{q}))^{-1} \\ &\times \iint d^2x\,d^2v_2\ \exp(i\mathbf{k}\mathbf{x} - i\mathbf{q}\mathbf{v}_2))\,\psi(\mathbf{x})\ [\widehat{D}_{hh}(\mathbf{x},h)G_b(\mathbf{v}_2-\mathbf{x},h)]]. \end{aligned} \qquad (B.14)$$

After performing the integration over relative variables $\mathbf{v}_1 - \mathbf{y},\ \mathbf{v}_2 - \mathbf{x}$ in (B.14), it is possible now to decompose H_{corr} into local and nonlocal contributions as discussed in the main text of the present paper. In particular, the nonlocal contribution to H_{corr}

is given by:

$$H_{corr}^{(nonloc)} = (k_B T/2) \iint d^2x\, d^2y\, (\psi(\mathbf{y}) - \psi(\mathbf{x}))^2/2 \times \{[\widehat{D}_{4z}(\mathbf{x},\,\mathbf{y})\, G_b(\mathbf{y}-\mathbf{x},\, z)]_{z=0} \Phi_1(\mathbf{y}-\mathbf{x}) + \Phi_2^2(\mathbf{y}-\mathbf{x}) + \Phi_3(\mathbf{y}-\mathbf{x})\, \Phi_4(\mathbf{y}-\mathbf{x})\}, \qquad (B.15)$$

$$\Phi_1(\mathbf{y}-\mathbf{x}) = \int d^2q/(2\pi)^2\, \widetilde{G}_b(\mathbf{q},0)\exp(-i\mathbf{q}(\mathbf{y}-\mathbf{x}))\,(4D_0(\mathbf{q}))^{-1},$$

$$\Phi_2(\mathbf{y}-\mathbf{x}) = \int \frac{d^2q}{(2\pi)^2}[\widehat{D}_2(\mathbf{q},h)\,\widetilde{G}_b(\mathbf{q},h)]\,[\widehat{D}_{hh}(\mathbf{q},h)\,\widetilde{G}_b(\mathbf{q},h)]\frac{\exp(-i\mathbf{q}(\mathbf{y}-\mathbf{x}))}{4D_0(\mathbf{q})},$$

$$\Phi_3(\mathbf{y}-\mathbf{x}) = \int \frac{d^2q}{(2\pi)^2}[\widehat{D}_{hh}(\mathbf{q},h)\,\widetilde{G}_b(\mathbf{q},h)]\,[\widehat{D}_{zz}(\mathbf{q})\,\widetilde{G}_b(\mathbf{q},z)]_{z=0}\,\frac{\exp(-i\mathbf{q}(\mathbf{y}-\mathbf{x}))}{4D_0(\mathbf{q})},$$

$$\Phi_4(\mathbf{y}-\mathbf{x}) = \iint d^2k/(2\pi)^2[\widehat{D}_{hh}(\mathbf{k},h)\,\widetilde{G}_b(\mathbf{k},h)]\,\widetilde{G}_b(\mathbf{k},0)\exp(-i\mathbf{k}(\mathbf{y}-\mathbf{x}))\,(4D_0(\mathbf{k}))^{-1},$$

$$\widehat{D}_{hh}(\mathbf{k},h) = \partial^2/\partial h^2 + (\gamma_{\mathrm{IA}}/C_{33})\,k^2\,\partial/\partial h. \qquad (B.16)$$

C *Correlation Functions for Bounded Systems of "Correlated" Liquids*

Generalized functional integration method allows to obtain the correlation functions $G_{\mathrm{IA}}^{(u)} = \langle \tilde{u}(\mathbf{x},h(x))\,\tilde{u}(\mathbf{y},h(\mathbf{y}))\rangle$ of the thermal displacements $\tilde{u}$. General expression for correlation function $G^{(u)}(\mathbf{r},\mathbf{r}') = \langle \tilde{u}(\mathbf{r})\,\tilde{u}(\mathbf{r}')\rangle$ can be obtained from the generating functional [17,18]

$$Z\{J\} = \left\langle \exp\left[\int d^3r\, J(\mathbf{r})\,\tilde{u}(\mathbf{r})\right]\right\rangle = \exp\left[(1/2)\iint d^3r\, d^3r'\, J(\mathbf{r})\,\langle\,\tilde{u}(\mathbf{r})\,\tilde{u}(\mathbf{r}')\,\rangle\, J(r')\right] \qquad (C.1)$$

using auxiliary field $J(\mathbf{r})$. With the help of Eq. (12), $Z\{J\}$ can be written as:

$$Z\{J\} = Z_{01}^{-1} Z_0^{-1}\int D\tilde{u}(\mathbf{r})\exp[-H_0[\tilde{u}]/k_BT]\iint D\Omega_1(\mathbf{x})\, D\Omega_2(\mathbf{y}) \times \exp\left[i\int d^2x\,\Omega_1(\mathbf{x})\,\tilde{u}(\mathbf{r}_1(\mathbf{x})) + i\int d^2y\,\Omega_2(\mathbf{y})\,\Pi(\mathbf{r}_2(\mathbf{y})) + \int d^3r\, J(\mathbf{r})\,\tilde{u}(\mathbf{r})\right], \qquad (C.2)$$

where $Z_{01} = \exp[-H_{eff}/k_BT]$. By analogy with obtaining (14), the Gaussian integration in (C.2) over $\tilde{u}(\mathbf{r})$ results in

$$Z\{J\} = Z_{01}^{-1} exp[(1/2)\iint d^3r\, d^3r' J(\mathbf{r})\, G_b(\mathbf{r},\mathbf{r}')\, J(\mathbf{r}')]\iint D\Omega_1 D\Omega_2 \exp[-S_{eff}\{\Omega,J\}], \qquad (C.3)$$

where

$$S_{eff}\{\Omega,J\} = H_1[\Omega] - i\int d^2x\,\Omega_1(\mathbf{x})\,P_1(\mathbf{x}) - i\int d^2y\,\Omega_2(\mathbf{y})\,P_2(\mathbf{y})\,, \qquad (C.4)$$

$$P_1(\mathbf{x}) = \int d^3r\,G_b(\mathbf{r}_1(\mathbf{x}),\mathbf{r})\,J(\mathbf{r})\,,\ P_2(\mathbf{y}) = \int d^3r\left[\widehat{D}_2(\mathbf{r}_2(\mathbf{y}))\,G_b(\mathbf{r}_2(\mathbf{y}),\mathbf{r})\right]J(\mathbf{r})\,, \quad (C.5)$$

and where (following (8))

$$\widehat{D}_2(\mathbf{r}_2(\mathbf{y})) = \partial/\partial z_y - (\gamma_{IA}/C_{33})\,\partial^2/\partial y^2. \qquad (C.6)$$

After an even number of matrix transforms for $H_1[\Omega_\alpha, \Omega_\beta]$ and M using (15) and (17), and after change of variables: $\widetilde{\Omega}_\alpha = \Omega_\alpha - (i/2)\sum_\beta M^{-1}_{\alpha\beta}P_\beta$, one obtains S_{eff} in the form

$$S_{eff}\{\,\Omega, J\,\} = H_1\left[\widetilde{\Omega}\right] - (1/4)\sum_{\alpha,\beta} P_\alpha\,M^{-1}_{\alpha\beta}\,P_\beta. \qquad (C.7)$$

Finally, comparing the expressions (C.1) and (C.3) leads to $G^{(u)}(\mathbf{r}, \mathbf{r}')$

$$\begin{aligned} G^{(u)}(\mathbf{r}, \mathbf{r}') = G_b(\mathbf{r}, \mathbf{r}') - \{ &\iint d^2x\,d^2y\ G_b(\mathbf{r}, \mathbf{r}_1(\mathbf{x}))\ M^{-1}_{11}(\mathbf{x}, \mathbf{y})\ G_b(\mathbf{r}', \mathbf{r}_1(\mathbf{y})) \\ &+ \iint d^2x\,d^2y\ G_b(\mathbf{r},\mathbf{r}_1(\mathbf{x}))\ M^{-1}_{12}(\mathbf{x}, \mathbf{y})\ [\widehat{D}_2(\mathbf{r}_2(\mathbf{y}))\ G_b(\mathbf{r}_2(\mathbf{y}),\mathbf{r}')] \\ &+ \iint d^2x\,d^2y\ [\widehat{D}_2(\mathbf{r}_2(\mathbf{x}))\ G_b(\mathbf{r}_2(\mathbf{x}),\mathbf{r})]\ M^{-1}_{21}(\mathbf{x}, \mathbf{y})\ G_b(\mathbf{r}',\mathbf{r}_1(\mathbf{y})) \\ &+ \iint d^2x\,d^2y\ [\widehat{D}_2(\mathbf{r}_2(\mathbf{x}))\ G_b(\mathbf{r}_2(\mathbf{x}),\mathbf{r})]\ M^{-1}_{22}(\mathbf{x}, \mathbf{y})\ [\widehat{D}_2(\mathbf{r}_2(\mathbf{y}))\ G_b(\mathbf{r}_2(\mathbf{y}),\mathbf{r}')]\ \}. \end{aligned} \qquad (C.8)$$

Upon (4–5), $G^{(u)}_{\mathrm{IA}}$ can be calculated approximately by analogy with calculation of $M(\mathbf{x}, \mathbf{y})$. The leading contribution to $G^{(u)}_{\mathrm{IA}}(\mathbf{k},\mathbf{q})$ corresponds to vanishing $\psi(\mathbf{x})$ in (C.8). In this case, M^{-1} coincides with M_0^{-1}, $G^{(u)}_{\mathrm{IA}}$ coincides with $G^{(u,0)}_{\mathrm{IA}} = \langle\tilde{u}(\mathbf{x},h)\,\tilde{u}(\mathbf{y},h)\rangle$ and we find Eq. (30). In the limit when $\gamma_{\mathrm{IA}} \ll C_{33}\lambda_0$, Eq. (30) coincides with $\widetilde{G}^{(u,0)}_{\mathrm{IA}}(q)$ from [9] and corrections to $\widetilde{G}^{(u,0)}_{\mathrm{IA}}(\mathbf{q})$ up to the second order in ψ, for $\mathbf{q} \to 0$, are proportional to

$$k_BT/(2C_{33}\lambda_0q^2)\left\lfloor \int d^2y\,(\,\psi^2(\mathbf{y})/S)(\lambda_0q^2)^2\exp\left(-2\lambda_0q^2h\right)/(1-_M\exp\left(-2\lambda_0q^2h\right))^n \right\rfloor.$$

These corrections are either connected with correction to the stiffness of the mode u or proportional to the powers of $\mathbf{q}$ higher than second and, given the conditions (4-5), are irrelevant for all $\mathbf{q}$.

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