Pseudo-Casimir interactions and surface anchoring duality in bookshelf geometry of smectic-A liquid crystals

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We analyze the transverse intersubstrate pseudo-Casimir force, arising as a result of thermal fluctuations of the liquid crystalline layers of a smectic-A film confined between two planar substrates in a bookshelf geometry, in which the equidistant smectic layers are placed perpendicular to the bounding surfaces. We discuss the variation of the interaction force as a function of the intersubstrate separation in the presence of surface anchoring to the substrates, showing that the force induced by confined fluctuations is attractive and depends on the penetration length as well as the layer spacing. The strongest effect occurs for tightly confined fluctuations, in which the surface anchoring energy is set to infinity, where the force per area scales linearly with the thermal energy and inversely with the fourth power of the intersubstrate separation. By reducing the strength of the surface anchoring energy, the force first becomes weaker in magnitude but then increases in magnitude as the surface anchoring strength is further reduced down to zero, in which case the force tends to that obtained in the limit of strong anchoring.

I. INTRODUCTION

Liquid crystals [1] are soft materials characterized by long-range, correlated thermal fluctuations of their order parameter. The external inclusions in these media, perturbing their ordered state, are thus expected to experience long-range fluctuation-induced forces akin to the van der Waals-Casimir force, which is induced between dielectric bodies by thermal as well as quantum fluctuations of the electromagnetic field inside and in the environment of the interacting bodies [2-4]. While this standard fluctuation-induced force also exhibits a thermal component, which is the zero-Matsubara-frequency term, the thermal fluctuation-induced interactions in the context of soft-matter systems came to be referred more generally as the pseudo-Casimir force. Such fluctuation-induced effects were studied extensively in various contexts of the soft materials such as liquid crystals or gels [5,24], specifically also in the case of the smectic-A phase [15,16]. In this latter instance, the molecules are arranged in a series of parallel, equidistant, liquidlike smectic layers. While the molecules exhibit no particular positional or-
field fluctuations, describing the orientational order of the molecules within the smectic phase. The role of such a coupling has been studied in the case of a homeotropic and a free-standing smectic film in Ref. [15]. We introduce our model in Section II and discuss the functional-integral methods used to evaluate the partition function and, hence, the free energy of the system in Section III, where the analytical and numerical results are discussed in detail (Sections III C and III D), with the concluding remarks to be followed in Section IV.

II. MODEL

We consider a smectic-A phase of a liquid-crystalline material 1 with parallel layers of separation $a_0$, stacked up in the $z$ direction. The long axes of the molecules, comprising each of the liquidlike smectic layers, are thus aligned on average in the $z$ (normal-to-layer) direction. The smectic-A phase is assumed to be confined between two plane-parallel walls (substrates) placed perpendicular to the smectic layers at positions $y = 0$ and $d$; see Fig. 1. The displacement field due to thermal (in-plane and out-of-plane) undulations of the smectic layers is denoted by $u(r_z,y)$, while the director field of the molecules is denoted by $n(r_z,y)$, where $r_z = (r_z,y)$ is the Cartesian spatial position with the transverse coordinates denoted by $r_z = (x,z)$.

The Hamiltonian (deformation energy) associated with such displacements is given by

$$H_b = \frac{K}{2} \int_V \text{d}r \left[ (\partial_x^2 u + \partial_y^2 u)^2 + \frac{B}{K} (\partial_z u)^2 \right],$$

(1)

where $K$ and $B$ are elastic moduli corresponding to bending and compressing (or, dilating) of the layers, respectively, and $V$ is the volume of the smectic slab.

We assume in general that a preferred order exists for the director field $n$ at the confining substrates of the smectic film, that is, $n(r_z,0)$ and $n(r_z,d)$ orient preferentially along the $z$-axis. The latter is modeled using the Rapini-Papoular model by assuming the surface-interaction Hamiltonian

$$H_s = -\frac{W}{2} \int_{\partial V} \text{d}r_z (n \cdot \hat{z})^2,$$

(2)

where $W$ is the anchoring energy per unit area and the integration is taken over the surface area of the confining boundaries (substrates) [24]. Considering small thermal fluctuations of the director field around its ground-state configuration, that is, by writing $n = \hat{z} + \delta n = (\delta n_x, \delta n_y, \sqrt{1 - \delta n_x^2 - \delta n_y^2})$, we find

$$H_s = \frac{W}{2} \int_{\partial V} \text{d}r_z (\partial_x u)^2 + (\partial_y u)^2$$

(3)

up to the quadratic order in the fluctuating field $\delta n = (\delta n_x, \delta n_y, \delta n_z)$, where we have also used the relation $(\partial_x, \partial_y)^2 u + \delta n = 0$, asserting that the local director field remains perpendicular to the smectic layers [24, 29].

Because of the translational invariance of the Hamiltonian across the $(x,z)$-plane, we can use the Fourier representation $u(r_z,y) = \sum_{p_z} u_{p_z}(y) e^{i p_z \cdot r_z}$, with $p_z = (p_x, p_z)$ being the two-dimensional wavevector, to express the total Hamiltonian $H = H_b + H_s$ in terms of the terms contributed by individual transverse modes $p_z$. Hence, the total Hamiltonian as well as its bulk and surface parts can be additively decomposed as $H = \sum_{p_z} h[u_{p_z}(y)]$, into a bulk functional $H_b = \sum_{p_z} h_b[u_{p_z}(y)]$ and a surface function $H_s = \sum_{p_z} h_s(u_{p_z}(0), u_{p_z}(d))$, where obviously

$$h[u_{p_z}(y)] = h_b[u_{p_z}(y)] + h_s(u_{p_z}(0), u_{p_z}(d)),$$

(4)

and the bulk and surface terms associated with each transverse mode are obtained, respectively, as

$$h_b[u_{p_z}(y)] = \frac{KA}{2} \int_0^d \text{d}y \left\{ |\dot{u}_{p_z}(y)|^2 + 2p_z^2 |\ddot{u}_{p_z}(y)|^2 \right\},$$

(5)

$$h_s(u_{p_z}(0), u_{p_z}(d)) = \frac{WA}{2} \left\{ |\dot{u}_{p_z}(0)|^2 + |\dot{u}_{p_z}(d)|^2 \right\}$$

$$+ p_z^2 (|u_{p_z}(0)|^2 + |u_{p_z}(d)|^2)$$

$$- \frac{KA}{2} p_z^2 \left\{ (u_{p_z}(d)\dot{u}_{p_z}(d) - u_{p_z}(0)\dot{u}_{p_z}(0)) + c.c. \right\},$$

(6)
where the penetration length is defined as \( \lambda = \sqrt{K/B} \) \[1\], \( A \) is the surface area of each confining plate, \( u_{p_\perp}^* (y) = u_{p_\perp} (y) \), and the dots over the symbols denote the \( y \)-derivative, i.e., \( \dot{u}_{p_\perp} \equiv \partial_y u_{p_\perp} \).

### III. FORMALISM AND RESULTS

#### A. Partition function

The partition function of the system described in the previous section follows by integrating over all field fluctuations as

\[
Z = \prod_p p \int D u_{p_\perp} (y) e^{-\beta \mathcal{H}[u_{p_\perp} (y)]} = \prod_p e^{-\beta \mathcal{F}(p_\perp)}, \tag{7}
\]

where the propagator \( \mathcal{G}(\dot{u}_{p_\perp}(0), u_{p_\perp}(0); \dot{u}_{p_\perp}(d), u_{p_\perp}(d)) \), giving the statistical weight of bounding surfaces field configurations characterized by \( u_{p_\perp}(0), u_{p_\perp}(d) \), and \( \dot{u}_{p_\perp}(0), \dot{u}_{p_\perp}(d) \), is defined as

\[
\mathcal{G}(\dot{u}_{p_\perp}(0), u_{p_\perp}(0); \dot{u}_{p_\perp}(d), u_{p_\perp}(d)) = \int D u_{p_\perp} (y) e^{-\beta h_s[u_{p_\perp}(y)]}. \tag{10}
\]

Note that the surface term \( \mathcal{F}(p_\perp) \) is additive in \( u_{p_\perp}(0) \) and \( u_{p_\perp}(d) \), and, as such, is routinely grouped into two separate terms, denoted by \( h_s(u_{p_\perp}(0)) \) and \( h_s(u_{p_\perp}(d)) \). The propagator \( \mathcal{G} \) involves functional integration over a Boltzmann-weighted Hamiltonian involving the second-order derivative of the field \( u_{p_\perp}(y) \); see Eq. \[1\], which can be calculated using the methods introduced in Ref. \[30\], or the final expression provided in Ref. \[31\]. As a matter of fact, this methodology has already been used in the context of Casimir interactions, e.g., in Ref. \[32\], dealing with the pseudo-Casimir force in a confined nematic polymer, and in Ref. \[33\], which analyzes the Casimir effect in fluids above the isotropic-lamellar transition within the Brazovskii mesoscopic theory, with formal developments discussed further in Ref. \[34\]. Hence, using the same methodology and basically following Ref. \[30\], we derive a closed form expression for the integrand entering the partition function \( \mathcal{F}(p_\perp) \) as

\[
e^{-\beta h_s(u_{p_\perp}(0))} \mathcal{G}(\dot{u}_{p_\perp}(0), u_{p_\perp}(0); \dot{u}_{p_\perp}(d), u_{p_\perp}(d)) e^{-\beta h_s(u_{p_\perp}(d))} = \mathcal{A}(\omega_1, \omega_2, d) e^{-\beta h_{\text{eff}}(u_{p_\perp}(0), u_{p_\perp}(d); \dot{u}_{p_\perp}(d), u_{p_\perp}(d))} \tag{11}
\]

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\]

with an effective field surface-Hamiltonian that can be obtained as

\[
h_{\text{eff}}(u_{p_\perp}(0), u_{p_\perp}(d); \dot{u}_{p_\perp}(d), u_{p_\perp}(d))
\]

\[
= m_{11} (|u_{p_\perp}(0)|^2 + u_{p_\perp}(d)^2)^2 \\
+ m_{12} (u_{p_\perp}(0) u_{p_\perp}^*(d) + c.c.) \\
+ m_{13} (\dot{u}_{p_\perp}(d) u^*(d) - \dot{u}_{p_\perp}(0) u_p^*(0) + c.c.) \\
+ m_{14} (\dot{u}(d) u^*(d) - \dot{u}(0) u^*(d) + c.c.) \\
+ m_{22} (\dot{u}(0)^2 + u^*(d)^2) \\
+ m_{33} (\dot{u}(0)^2 + (\dot{u}(d))^2) \\
+ m_{44} (\dot{u}(0) \dot{u}^*(d) + c.c.). \tag{12}
\]

We make use of the eigenfrequencies \( \omega_1 \) and \( \omega_2 \) [see Eq. \[5\]] defined through

\[
\omega_1^2 + \omega_2^2 = 2p_z^2,
\]

\[
\omega_1^2 \omega_2^2 = p_z^4 + \lambda^{-2} p_z^2, \tag{13}
\]

where again \( \lambda = \sqrt{K/B} \) is the penetration length, \( \ell = K/W \) is the extrapolation (or pinning) length, and the shorthand notations \( c_i \equiv \cosh(\omega_i d) \) and \( s_i \equiv \sinh(\omega_i d) \) for \( i = 1, 2 \) \[30\] to express the elements \( m_{ik} \) of the matrix...
and the prefactor $\mathcal{A}$ in Eq. \eqref{eq:prefactor} as
\begin{equation}
\mathcal{A}(\omega_1, \omega_2, d) = \frac{1}{2\pi} \frac{\sqrt{\omega_1 \omega_2}}{\sqrt{M}} \left| \frac{\omega_1^2 - \omega_2^2}{M} \right|^{\frac{1}{2}},
\end{equation}
where
\begin{equation}
M = (\omega_1^2 + \omega_2^2) s_1 s_2 - 2\omega_1 \omega_2 (c_1 c_2 - 1).
\end{equation}
The final step of calculating $Z$ involves Gaussian functional integrals over the fluctuating surface fields \{\hat{u}_p(0), \hat{u}_p(d), \hat{u}_{p_2}(d)\} of the factor $e^{-\beta \hat{H}_{\text{ext}}}$, see Eq. \eqref{eq:prefactor}.

### B. Interaction free energy

The free energy straightforwardly follows from the final results in the preceding section. By dropping irrelevant additive factors as necessary, the interaction free energy of the system can be obtained from
\begin{equation}
\mathcal{F}(d) = -k_B T \ln Z,
\end{equation}
where
\begin{equation}
Z \propto \prod_{\mathbf{p}_L} \frac{1}{\sqrt{|G(\mathbf{p}_L)|}},
\end{equation}
and $G(\mathbf{p}_L)$ can be written in an explicit form as
\begin{equation}
G(\mathbf{p}_L) = A + B \cosh(2\alpha d) + C \sinh(2\alpha d) + D \cos(2\gamma d) + E \sin(2\gamma d).
\end{equation}
Here $\alpha$ and $\gamma$ are defined through the real and imaginary parts of the solutions of Eqs. \eqref{eq:omega}, as $\omega_1 = \alpha - i\gamma$ and $\omega_2 = \alpha + i\gamma$, where
\begin{align}
\alpha &= \frac{1}{\sqrt{2}} \left( p_x^2 + \sqrt{p_x^2 + \lambda^2 p_z^2} \right)^{1/2}, \\
\gamma &= \frac{1}{\sqrt{2}} \left( -p_x^2 + \sqrt{p_x^2 + \lambda^2 p_z^2} \right)^{1/2},
\end{align}
and we find
\begin{align}
A &= \sqrt{p_x^2 + \lambda^2 p_z^2} \left( \lambda^2 - p_x^2 - \epsilon^2 p_z^2 \right)^2, \\
B &= -\gamma^2 \left[ (\lambda^2 - 2\epsilon^2 p_z^2) (4p_x^2 + \sqrt{p_x^2 + \lambda^2 p_z^2}) \ell^{-1} - \epsilon^2 p_x^2 (8p_x^2 + 8\sqrt{p_x^2 + \lambda^2 p_z^2} + \lambda^2 p_z^2 + \epsilon^2) \right], \\
C &= 2\alpha \lambda \ell p_z^2 \epsilon^{-1} \left( \lambda^2 p_z^2 - \ell^2 p_z^2 \right), \\
D &= -\alpha^2 \lambda^2 \left( \lambda^2 p_z^2 - 2\epsilon^2 p_z^2 (4p_x^2 + \sqrt{p_x^2 + \lambda^2 p_z^2}) \ell^{-2} + \epsilon^2 p_x^2 (8p_x^2 + 8\sqrt{p_x^2 + \lambda^2 p_z^2} + \lambda^2 p_z^2 + \epsilon^2) \right), \\
E &= -2\gamma \lambda \ell p_z^2 \epsilon^{-1} \left( \lambda^2 p_z^2 - \ell^2 p_z^2 \right).
\end{align}

### C. Limiting cases

In the limits of weak and strong anchoring, i.e., $W \to 0$ and $W \to \infty$, or, equivalently, expressed in terms of the extrapolation length, $\ell \to \infty$ and $\ell \to 0$, respectively, $G$ (Eqs. \eqref{eq:G_limit1} - \eqref{eq:G_limit4}), irrespective of irrelevant prefactors, reduces to the exact same expression
\begin{align}
G\big|_{W=0,\infty} &= \alpha^2 + \gamma^2 - \gamma^2 \cosh(2\alpha d) - \alpha^2 \cos(2\gamma d) \\
&= \gamma^2 \sin^2(\alpha d) - \alpha^2 \sin^2(\gamma d),
\end{align}
which is equal to $M/2$ (Eq. \eqref{eq:prefactor}). In other words, the interaction free energy in the strong and the weak anchoring limits is found to coincide. This is a special case of a duality that exists in this system and tallies also with the properties of the pseudo-Casimir interactions in nematic liquid crystals \cite{5,6}.

The interaction free energy per unit area (after subtracting a trivial additive contribution from the bulk material) is obtained as
\begin{equation}
\frac{\mathcal{F}(d)}{A} = \frac{k_B T \lambda^2}{2\pi^2 d^2} \int_0^\infty dp_x \int_0^\infty dp_z \ln \left\{ \frac{1 - e^{2\alpha}}{1 - \left( \frac{2\alpha \sin \gamma}{(\gamma^2 - 1)} \right) e^{2\gamma}} \right\},
\end{equation}
where we have defined the rescaled variables $p_x d \to p_x$, $p_z (d^2/\lambda) \to p_z$ and have taken the upper limit of $p_z$ to infinity, i.e., $\pi d^2/(\lambda a) \to \infty$. We have also non-dimensionalized $\alpha$ and $\gamma$ (Eqs. \eqref{eq:alpha_limit} - \eqref{eq:gamma_limit}) as
\begin{align}
\alpha d &\to \alpha = \sqrt{q_x^2 + q_y^2 + q_z^2}/\sqrt{2}, \\
\gamma d &\to \gamma = \left( -q_x^2 + \sqrt{q_x^2 + q_z^2} \right)/\sqrt{2}.
\end{align}

### D. Numerical results

The free energy \eqref{eq:free_energy} gives rise to a fluctuation-induced interaction force,
\begin{equation}
f(d) = -\frac{\partial \mathcal{F}(d)}{\partial d},
\end{equation}
which turns out to be attractive for the whole range of the intersubstrate separations. We numerically evaluate
the corresponding dimensionless interaction pressure,
\[ \Pi(\tilde{d}) = \frac{\beta \lambda^3}{A} f(\lambda \tilde{d}), \]  
(37)
and show its behavior as a function of the rescaled inter-substrate separation \( \tilde{d} = d/\lambda \) and other rescaled system parameters, i.e., \( \lambda/\ell \) and \( \lambda/a_0 \), in Figs. 2, 3 and 4 respectively. To produce the numerical results, the model parameters are varied as follows. First, we note that the Casimir force between the bounding substrates, which we investigated in detail in this work.

Figure 2 also indicates that, at a given value of \( \lambda/\ell \), the pressure profiles become increasingly less negative (strong anchoring). The same trend occurs starting with a fixed \( \lambda/\ell < 1 \) and decreasing it to very small values (weak anchoring), corroborating the aforementioned observation that the limiting results for \( \lambda/\ell \to \infty \) and 0 coincide, where the largest magnitude of the attractive (negative) pseudo-Casimir force at a given rescaled separation \( \tilde{d} \) is established.

The interaction pressure in the strict limit of strong (or, weak) anchoring can be quite accurately approximated by a universal expression as
\[ \Pi_0(\tilde{d}) = -C_0/\tilde{d}^4, \]
where \( C_0 \approx 1.76985 \) is obtained from numerical integration of Eq. (33). Equation (38) is shown as a gray solid curve in Fig. 2 and, as seen, it closely approximates the data obtained for large (or, small) \( \lambda/\ell \), when \( \tilde{d} \) is only modestly large (i.e., \( \tilde{d} \gtrsim 1.5 \); the above analytical estimate is in fact expected to remain valid for \( d^2 \gg \lambda a_0 \).

Figure 3 also indicates that, at a given value of \( \lambda/\ell \) and for fixed \( \lambda/a_0 \), the interaction pressure varies non-monotonically with \( \lambda/\ell \) (or, equivalently, with the anchoring energy). This behavior is shown in Fig. 3, where the dimensionless pressure is plotted as a function of \( \ln(\lambda/\ell) \), at fixed \( \tilde{d} = 1.3 \). It turns out that the magnitude of the (attractive) interaction pressure increases as \( \lambda/a_0 \) is increased (or, equivalently, \( a_0 \) is decreased). This can be seen from both Figs. 3 and 4. In the latter figure, we fix \( \lambda/\ell = 100 \) and plot the interaction pressure profiles for three different values of \( \lambda/a_0 \), as indicated on the graph.

IV. CONCLUDING REMARKS

We considered a smectic-A liquid crystalline film confined within a planar gap that quenches the thermal fluctuations at the boundaries of the sample. A bookshelf geometry—in which the molecules vicinal to the two planar bounding substrates are forced to orient in co-planar directions—provides a particularly interesting physical system to study the fluctuating modes of the layering field and the layer displacement of the smectic-A phase. The imposition of the hard boundaries modifies the free-space fluctuation spectrum, allowing only for the modes which are compatible with the presence of the boundaries as well as with the nature of the interaction between the vicinal smectic layers and the boundary itself; hence, leading to a fluctuation-induced pseudo-Casimir force between the bounding substrates, which we investigated in detail in this work.

We use the standard statistical mechanical continuum
field theory paradigm to calculate the fluctuation force, resulting from thermally excited modes within a $k_B T$ energy range of their ground state. While, in general, the context of our calculation is quite close to the pseudo-Casimir interactions, we do explicitly take into account the smectic-A boundary interaction energy, assuming it has a general Rapini-Papoular form [28], and thus avoid the question of the proper boundary condition of the fluctuating displacement fields. In addition, our calculation is interesting also from the purely formal perspective, since it develops further the Casimir methodology of field Hamiltonians, involving higher order field derivatives, which were first used in the context of Casimir interactions for confined nematic polymers [32] and confined Brazovskii-type soft media [33]. The main difference between the Brazovskii-type soft media and the smectic-A liquid crystalline film is the absence of the bound states in the Fourier decomposition in the direction perpendicular to the bounding surfaces. This difference can be tracked to the sign of the first order derivative term in the field Hamiltonian Eq. (5).

The main features of our results may be outlined as follows. First, the fluctuations with vanishingly small values of both the azimuthal and normal derivatives of layer displacement field on the bounding substrates are found to give a long-ranged intersubstrate attraction, which, at sufficiently large separations, is well-described by the universal expression given in Eq. (38). The calculated prefactor $C_0 \approx 1.76985$ differs from what is proposed in Ref. [5], where the prefactor is suggested to be of the form $\zeta(4) \approx 1.08232$. The latter conjecture is indeed not supported by our numerical findings. Second, the interaction force (or pressure) exhibits a non-monotonic dependence on the ratio $\lambda/\ell$ and shows a duality with respect to the magnitude of the surface anchoring energy. Third, the attractive pressure mediated between the bounding substrates is strengthened as $a_0$, the periodicity along the normal of the layers, is decreased, implying that denser smectic systems should show an increased pseudo-Casimir force.

The non-monotonic behavior of the pseudo-Casimir interaction pressure on the surface anchoring coupling strength $\lambda/\ell$ leads to small interaction magnitude for intermediate values, that increases either for very large or very small values of the surface coupling strength. The interaction forces obtained in the two limits of weak ($\lambda/\ell \to 0$) and strong anchoring ($\lambda/\ell \to \infty$) coincide, indicating a special case of a duality in this system, which resembles the behavior previously seen in the case of nematic liquid crystals [5, 7, 36]. This property can be understood by examining the boundary condition equations as follows. Formally, for a bulk free-energy density of the form $f_b = \frac{1}{2}a\phi^2 + \frac{1}{2}b\psi^2 + \frac{1}{2}c\zeta^2$ and the surface energy density of the form $f_s = W(\frac{1}{2}\eta\phi^2 + \frac{1}{2}\zeta\phi^2)$, as we deal with in this paper, the boundary equations at $y = d$ read $b\phi - c\psi + W\eta\phi = 0$ and $c\phi + W\zeta\phi = 0$. We note that the limit $W \to \infty$ corresponds to $\phi = \phi = 0$ on the substrate. Then the partition function directly reads $\mathcal{Z} = \mathcal{A} \propto 1/\sqrt{M}$ [see Eqs. (9), (11), and (20)], where $M$ turns out to be equal to $2\gamma_{W=0,\infty}$ [Eq. (32)]. On the other hand, for $W = 0$, the boundary condition equations lead to undetermined values for $\phi$ and $\psi$. This is why to evaluate $\mathcal{Z}$ we integrate (average) over all possible values of both fields. This averaging reasonably leads to the same result as for the strong anchoring condition.

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