Casimir interaction in smectic-A liquid crystals caused by coupled fluctuations of positional and orientational order

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A theoretical study of the Casimir interaction in smectic-A systems, considering fluctuations of both types of smectic ordering—positional and orientational—including the coupling between them, is presented. Two model systems with plan-parallel geometry are studied: homeotropic cell and free-standing film. At large thicknesses of the system the behavior of the Casimir force is found to be primarily determined by positional fluctuations, whereas at small thicknesses also the orientational degrees of freedom greatly contribute to the interaction. The influence of different coupling strengths between orientational and positional order is presented. The dependence of the Casimir force on the director anchoring and surface-tension parameters is studied. The possibilities of experimental detection of the interaction are discussed.

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I. INTRODUCTION

Since the original study of Casimir in 1948 [1], the interest in the ubiquitous Casimir effect has grown enormously. Every year a large number of studies from various fields of physics addressing this phenomenon are published [2–6]. These studies mostly deal with theoretical aspects of the Casimir effect, whereas the experimental work is rather scarce [7–10]. This is to be ascribed to extremely difficult measurements of the Casimir force due to its small magnitude. However, with the modern technologies approaching smaller and smaller length scales, the Casimir effect also gains its importance, for example, in (sub)microelectromechanical devices [11,12].

Liquid crystals are among the systems where the Casimir effect has been intensively studied [13–28]. In these soft systems, which exhibit a large variety of different phases, the impact of thermal fluctuations on the liquid crystalline order has been established long ago [29–31]. The theoretical studies of the Casimir force mostly addressed the simplest liquid crystal—nematic—phase, while some work was also done for smectic and columnar phases. However, despite the theoretical efforts, the experimental confirmation is still lacking. The possibilities of the measurement of the Casimir force in nematic systems have been thoroughly discussed in Ref. [32]. Although the magnitude of the force seems to be achievable (at least in very thin samples) by modern force measurement techniques, such as atomic-force microscopy (AFM) and surface-force apparatus (SFA), some serious difficulties remain. First, it is not easy to avoid elastic deformations of the nematic director, which lead to strong interactions and screen the Casimir force. Second, the magnitude and behavior of the Casimir force strongly depend on the specific anchoring conditions, which makes the identification of the force rather difficult. In view of these drawbacks of nematic systems, it seems promising to extend the research to the smectic systems where the Casimir force is expected to be of even longer range than in nematics [13–15].

The Casimir force in confined smectic systems has been studied by Mikheev [13] and Ajdari *et al.* [14,15] using the continuous description of smectics and by de Oliveira and Lyra [25,28] within the discrete model. All these studies focused on the Casimir force induced by the fluctuations of positional order (i.e., smectic layers) while assuming that the director rigidly follows the layers—is fixed perpendicular to the layers in the smectic-A phase. However, this assumption may not be entirely correct, especially in the vicinity of the smectic-A to smectic-C phase transition, where the fluctuations of the director away from the layer normal become very pronounced. The behavior of the Casimir force at this phase transition was addressed in Ref. [26], but it considered only the director fluctuations that are critical at this transition. It therefore seems necessary to implement a more complex description of smectics that considers both positional and orientational order, taking into account a realistic coupling between them. It is the aim of this paper to calculate and analyze the Casimir force within such an extended model. We will consider two confined smectic-A systems with plan-parallel geometry: a homeotropic cell and a free-standing film. We will study the effect of different coupling strengths between the director and smectic layers and the effect of different boundary conditions (anchoring). We will also discuss the observability of the Casimir force.

II. THEORETICAL MODEL

Smectic liquid crystals are characterized by one-dimensional positional order and by orientational order of molecules [31]. The positional order can be described by a complex order parameter $\Psi=\psi\exp(i\phi)$, where ψ is the degree of smectic order and the phase ϕ is related to the deformation of smectic layers (u). The orientational order can be described by the director $\mathbf{n}$, which gives the average direction of the orientation of the molecules. In smectic-A phase, the molecules are preferentially oriented perpendicular to the

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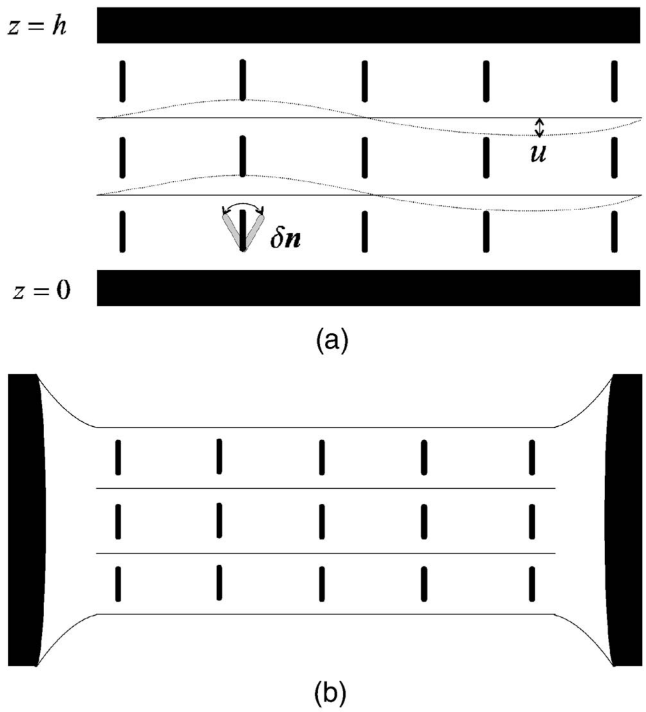

FIG. 1. Schematic presentation of a homeotropic cell (a) and a free-standing film (b).

smectic layers. In this paper we will use the following phenomenological free-energy model for describing smectic-A systems [33,34]:

$$F_{\mathrm{SmA}} = \frac{1}{2}\int \mathrm{d}V[B(\nabla_{\|}u)^2 + K_L(\nabla_{\perp}^2 u)^2 + D(\nabla_{\perp}u + \delta\mathbf{n})^2] + \frac{1}{2}\int \mathrm{d}V\{K_1(\nabla\cdot\mathbf{n})^2 + K_2[\mathbf{n}\cdot(\nabla\times\mathbf{n})]^2 + K_3[\mathbf{n}\times(\nabla\times\mathbf{n})]^2\}. \quad (1)$$

Here and henceforth we assume that the degree of the smectic order ψ is constant, which holds reasonably well when the system is deep in smectic-A phase, i.e., far from the smectic-A–nematic phase transition. Therefore, the parameter ψ does not enter the free energy explicitly. The first and second terms in F_{SmA} are related to the layer compression and to the layer bending, respectively. The third term $D(\nabla_{\perp}u+\delta\mathbf{n})^2/2$ describes the coupling between the orientational order (director) and positional order (layers). It simply states that if the coupling constant D is positive, the director will tend to orient perpendicular to the layers ($\delta\mathbf{n}=-\nabla_{\perp}u$). Here $\delta\mathbf{n}$ is the deviation of the director away from the normal to the unperturbed layers. The last three terms in F_{SmA} describe the Frank director elastic energy. It is worth noting that our model [Eq. (1)] applies to thermotropic as well as to lyotropic smectic systems.

In this paper, we study the Casimir force in two confined smectic systems with plan-parallel geometry: homeotropic cell and free-standing film (Fig. 1). In a homeotropic cell the smectic material is bounded by two parallel plates separated by a distance h. We assume that, at the boundaries, the smectic layers rigidly adjust to the plates, i.e., $u(z=0)=u(z=h)=0$. We also assume that the plates favor homeotropic ordering of the director. The director anchoring will be described by the Rapini-Papoular model

$$F_S[\mathbf{n}] = \frac{1}{2}W\int \sin^2(|\delta\mathbf{n}|)\mathrm{d}S, \quad (2)$$

where W is the anchoring energy per surface unit. The geometry of a free-standing film is similar to the homeotropic cell, except that the smectic material is now bounded by air. Therefore, one has to account for positional fluctuations of the free surfaces of the film. We will ascribe surface-tension energy to deformations of the free surfaces:

$$F_S[u] = \frac{1}{2}\gamma\int (\nabla_{\perp}u)^2\mathrm{d}S. \quad (3)$$

We also assume that there is some effective anchoring at the free surface of the smectic-A film that favors homeotropic orientation of the director and is described by Eq. (2).

III. FREE ENERGY OF FLUCTUATIONS

In equilibrium, the smectic film (both in homeotropic cell and free-standing film) consists of an integer number of smectic layers, which are neither compressed nor dilated $[u_{eq}(\mathbf{r})=0]$. The equilibrium configuration of the director is homeotropic $[\mathbf{n}(\mathbf{r})=\mathbf{n}_z,\ \delta\mathbf{n}(\mathbf{r})=\mathbf{0}]$. What we are interested in are the thermal fluctuations of the smectic structure around this equilibrium, which are the source of the Casimir interaction. More precisely, we need to calculate the free energy of fluctuations in confined system, which can then be related to the interaction between confining boundaries. The first step in this procedure is to calculate the partition function of fluctuations

$$Z_{\mathrm{fluc}} = \exp(-\beta F_{\mathrm{fluc}}) = \int_{\mathrm{bc}} \exp(-\beta H)\mathcal{D}u\mathcal{D}\delta\mathbf{n} \quad (4)$$

where H is the Hamiltonian of fluctuations and $\beta=1/k_BT$. The integral is to be performed over all possible configurations of u and $\delta\mathbf{n}$, taking into account specific boundary conditions. As both studied systems are extensive in horizontal directions (x-y), it is convenient to Fourier transform the fluctuating fields: $u(\mathbf{r})=\Sigma_{\mathbf{q}}u_q(z)\exp(i\mathbf{q}\boldsymbol{\rho})$ and $\delta\mathbf{n}(\mathbf{r})=\Sigma_{\mathbf{q}}\mathbf{n}_q(z)\exp(i\mathbf{q}\cdot\boldsymbol{\rho})$. Here $\mathbf{q}=(q_x,q_y)$ and $\boldsymbol{\rho}=(x,y)$. Performing this transformation on Eq. (1) while keeping only harmonic terms, we obtain the Hamiltonian of fluctuations

$$H = \frac{1}{2}S\sum_{\mathbf{q}}\int_0^h \mathrm{d}z\Bigg[B\left|\frac{\partial u_q}{\partial z}\right|^2 + Dq^2|u_q|^2 + K_Lq^4|u_q|^2 + D(|n_{1q}|^2+|n_{2q}|^2) + iqD(u_qn_{1q}^* - u_q^*n_{1q}) + K_1q^2|n_{1q}|^2 + K_2q^2|n_{2q}|^2 + K_3\left(\left|\frac{\partial n_{1q}}{\partial z}\right|^2 + \left|\frac{\partial n_{2q}}{\partial z}\right|^2\right)\Bigg]. \quad (5)$$

We applied the usual decomposition of $\mathbf{n}_q(z)$ into a component n_{1q} parallel to $\mathbf{q}$ and a component n_{2q} perpendicular to

$\mathbf{q}$. As it is seen from Eq. (5) only n_{1q} modes are coupled to the layer fluctuations, while n_{2q} modes represent "pure" director fluctuations. The Fourier transformed surface contributions [Eqs. (2) and (3)] to the Hamiltonian of fluctuations read

$$H_S[\mathbf{n}] = \frac{1}{2}K_3 S L^{-1}\sum_{\mathbf{q}} (|n_{1q}^-|^2 + |n_{1q}^+|^2 + |n_{2q}^-|^2 + |n_{2q}^+|^2), \quad (6)$$

for the director anchoring and

$$H_S[u] = \frac{1}{2}K_3 S \chi^{-1}\sum_{\mathbf{q}} q^2(|u_q^-|^2 + |u_q^+|^2), \quad (7)$$

for the surface tension effect. We introduced the extrapolation lengths $L=K_3/W$, $\chi=K_3/\gamma$ and the following notation $n_{1,2q}^-=n_{1,2q}(z=0)$, $n_{1,2q}^+=n_{1,2q}(z=h)$, $u_q^-=u_q(z=0)$, $u_q^+=u_q(z=h)$ while S is the surface area.

As it can be seen from Eqs. (5)–(7), the modes with different wave vectors $\mathbf{q}$ are decoupled and the Hamiltonian can be written as $H=\Sigma_{\mathbf{q}}(H_{\mathbf{q}}[n_{1q},u_q]+H_{\mathbf{q}}[n_{2q}])$. Therefore, the partition function can be factorized: $Z_{\text{fluc}}=\Pi_{\mathbf{q}}Z_{\mathbf{q}}[n_{1q},u_q]\cdot Z_{\mathbf{q}}[n_{2q}]$. The problem is now reduced to the calculation of partial partition functions $Z_{\mathbf{q}}$

$$Z_{\mathbf{q}}[n_{1q},u_q] = \int \mathrm{d}n_{1q}^- \int \mathrm{d}n_{1q}^+ \int \mathrm{d}u_q^- \int \mathrm{d}u_q^+ \exp(-\beta H_S[n_{1q}^{\pm},u_q^{\pm}]) \int_{n_{1q}(z=0)=n_{1q}^-}^{n_{1q}(z=h)=n_{1q}^+} \int_{u_q(z=0)=u_q^-}^{u_q(z=h)=u_q^+} \exp(-\beta H_q[u_q,n_{1q}]) \times \mathcal{D}u_q(z)\mathcal{D}n_{1q}(z), \quad (8)$$

$$Z_{\mathbf{q}}[n_{2q}] = \int \mathrm{d}n_{2q}^- \int \mathrm{d}n_{2q}^+ \exp(-\beta H_S[n_{2q}^{\pm}]) \int_{n_{2q}(z=0)=n_{2q}^-}^{n_{2q}(z=h)=n_{2q}^+} \exp(-\beta H_q[n_{2q}])\mathcal{D}n_{2q}(z). \quad (9)$$

The partition function $Z_{\mathbf{q}}[n_{1q},u_q]$ is analogous to the quantum propagator of two coupled harmonic oscillators and can be readily evaluated [35,36]

$$Z_{\mathbf{q}}[n_{1q},u_q] \propto [\sinh(\Omega_1 h)\sinh(\Omega_2 h)]^{-1/2} \times[\Omega_1\Omega_2\lambda^{-2}A_1^-A_2^- + \chi^{-1}q^2(\Omega_1 S^2 A_1^- + \Omega_2 C^2 A_2^-) + L^{-1}(\Omega_1 C^2\lambda^{-2}A_1^- + \Omega_2 S^2\lambda^{-2}A_2^-) + \chi^{-1}L^{-1}q^2]^{-1/2}[\Omega_1\Omega_2\lambda^{-2}A_1^+A_2^+ + L^{-1}(\Omega_1 C^2\lambda^{-2}A_1^+ + \Omega_2 S^2\lambda^{-2}A_2^+) + \chi^{-1}q^2(\Omega_1 S^2 A_1^+ + \Omega_2 C^2 A_2^+) + \chi^{-1}L^{-1}q^2]^{-1/2}. \quad (10)$$

The partition function $Z_{\mathbf{q}}[n_{2q}]$ can be evaluated with analogy to the propagator of a single quantum harmonic oscillator [36]

$$Z_{\mathbf{q}}[n_{2q}] \propto \left[\frac{L^{-2}+\Omega_3^2}{2\Omega_3 L^{-1}}\sinh(\Omega_3 h) + \cosh(\Omega_3 h)\right]^{-1/2}. \quad (11)$$

We introduced the following notation:

$$\Omega_{1,2} = \frac{1}{\sqrt{2}}\frac{1}{\Lambda}\sqrt{1 + (\rho^2+\lambda^2)q^2 + \frac{K_L}{K_3}\lambda^2\Lambda^2 q^4 \mp \sqrt{\left[1 - (\lambda^2-\rho^2)q^2 - \frac{K_L}{K_3}\Lambda^2\lambda^2 q^4\right]^2 + 4\lambda^2 q^2}}, \quad (12)$$

$$\Omega_3 = \sqrt{\Lambda^{-2} + \frac{K_2}{K_3}q^2}, \quad (13)$$

$$C^2 = \frac{1}{2} + \frac{1}{2}\sqrt{\frac{\left[1 + (\rho^2-\lambda^2)q^2 - \frac{K_L}{K_3}\lambda^2\Lambda^2 q^4\right]^2}{\left[1 + (\rho^2-\lambda^2)q^2 - \frac{K_L}{K_3}\Lambda^2\lambda^2 q^4\right]^2 + 4q^2\lambda^2}}, \quad (14)$$

$$S^2 = 1 - C^2,$$

$$A_{1,2}^{\pm} = \frac{\cosh(\Omega_{1,2}h) \pm 1}{\sinh(\Omega_{1,2}h)} \quad (15)$$

and the correlation lengths $\Lambda=(K_3/D)^{1/2}$, $\lambda=(K_3/B)^{1/2}$, $\rho=(K_1/D)^{1/2}$. The free energy of fluctuations can now be written as

$$F_{\text{fluc}} = -k_B T\sum_{\mathbf{q}} (\ln Z_{\mathbf{q}}[n_{1q},u_q] + \ln Z_{\mathbf{q}}[n_{2q}]). \quad (16)$$

IV. CASIMIR FORCE

The force between the confining substrates in plan-parallel geometry is defined as

$$\mathcal{F} = -\frac{\partial F_{\text{int}}}{\partial h}. \tag{17}$$

The interaction free energy F_{int} is defined as a difference between the free energy of a confined system and the free energy of a corresponding bulk reference system, at the same time also subtracting the surface terms that do not depend on the separation between boundaries h: $F_{\text{int}}=F_{\text{fluc}}-F_{\text{fluc}}^{\text{bulk}}-F_{\text{fluc}}^{\text{surf}}$. The procedure for identifying the bulk and surface contribution from the total free energy of fluctuations was described in Ref. [20].

A. Homeotropic cell

The Casimir force in homeotropic cell is obtained from Eqs. (10) and (11) using the definition (17) and taking the limit of hard plates ($\gamma\to\infty$). It consists of four terms

$$\mathcal{F}_{\text{Cas}} = \mathcal{F}[n_2;L] + \mathcal{F}_1[n_1,u] + \mathcal{F}_2[n_1,u] + \mathcal{F}_3[n_1,u;L], \tag{18}$$

where

$$\mathcal{F}[n_2;L] = -\frac{k_BTS}{2\pi}\int_0^\infty \frac{\Omega_3 q \mathrm{d}q}{\dfrac{(\Omega_3+L^{-1})^2}{(\Omega_3-L^{-1})^2}\exp(2\Omega_3 h)-1}, \tag{19}$$

$$\mathcal{F}_1[n_1,u] = -\frac{k_BTS}{2\pi}\int_0^\infty \frac{\Omega_1 q \mathrm{d}q}{\exp(2\Omega_1 h)-1}, \tag{20}$$

$$\mathcal{F}_2[n_1,u] = -\frac{k_BTS}{2\pi}\int_0^\infty \frac{\Omega_2 q \mathrm{d}q}{\exp(2\Omega_2 h)-1}, \tag{21}$$

$$\begin{aligned}\mathcal{F}_3[n_1,u;L] &= -\frac{k_BTS}{4\pi}\int_0^\infty q\mathrm{d}q\left[\frac{\dfrac{\Omega_1^2S^2}{1+\cosh(\Omega_1 h)}+\dfrac{\Omega_2^2C^2}{1+\cosh(\Omega_2 h)}}{\Omega_1S^2A_1^-+\Omega_2C^2A_2^-+L^{-1}}\right]\\ &\quad -\frac{k_BTS}{4\pi}\int_0^\infty q\mathrm{d}q\left[\frac{\dfrac{\Omega_1^2S^2}{1-\cosh(\Omega_1 h)}+\dfrac{\Omega_2^2C^2}{1-\cosh(\Omega_2 h)}}{\Omega_1S^2A_1^++\Omega_2C^2A_2^++L^{-1}}\right].\end{aligned} \tag{22}$$

The first term $\mathcal{F}[n_2;L]$ represents the contribution of "pure" director fluctuation modes and was already considered in Ref. [26]. These director fluctuations are "massive"; therefore, the resulting force is short-range decaying as $\exp(-2h/\Lambda)/h$ at large separations ($h/\Lambda \gg 1$). In analogy with eigenmodes of two coupled harmonic oscillators, the terms $\mathcal{F}_1[n_1,u]$ and $\mathcal{F}_2[n_1,u]$ represent the contributions of "in-phase" [Fig. 2(a)] and "out-of-phase" [Fig. 2(b)] fluctuation modes of the director and layers. The in-phase fluctuations are "massless" $[\Omega_1(q=0)=0]$; therefore, the resulting force $\mathcal{F}_1[n_1,u]$ is long range. The contribution of the out-of-phase fluctuations $\mathcal{F}_2[n_1,u]$ has similar characteristics as the

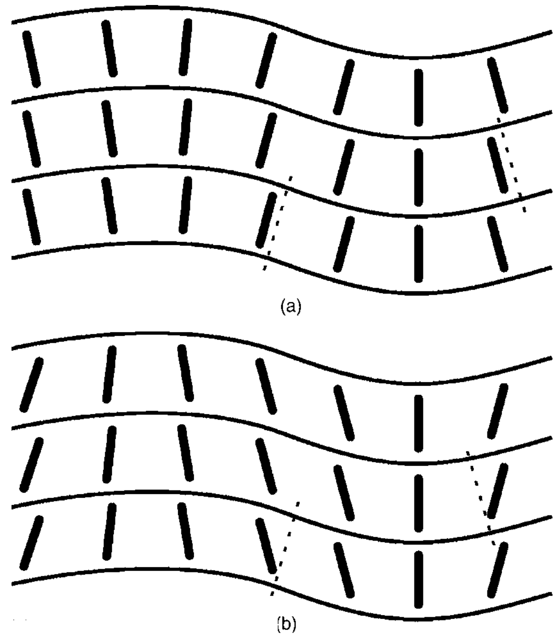

FIG. 2. Schematic presentation of fluctuation modes: (a) in-phase fluctuations of director and layers, and (b) out-of-phase fluctuations of director and layers. The dotted lines show local normal to the layers.

$\mathcal{F}[n_2;L]$ term. The last term $\mathcal{F}_3[n_1,u;L]$ is a correction to the $\mathcal{F}_1[n_1,u]$ and $\mathcal{F}_2[n_1,u]$ terms due to the finite director anchoring strengths W at the plates. This correction is short range and equal to 0 in the limit of very strong anchoring ($W\to\infty$, $L=0$).

We will compare our result [Eqs. (18)–(22)] to the Casimir force obtained by Mikheev [13] and Ajdari *et al.* [15] within a simplified model of smectic-A phase. In this simplified model, only fluctuations of smectic layers are considered while the director is assumed to be fixed perpendicular to the layers. The corresponding Hamiltonian reads $H_{\text{lay}}=1/2\int \mathrm{d}V[B(\partial u/\partial z)^2+K_L'(\nabla_\perp^2 u)^2]$. With the boundary conditions $u(z=0)=u(z=h)=0$, this gives the Casimir force $\mathcal{F}_{\text{Cas}}^{\text{lay}}=-k_BTS\zeta(2)/16\pi h^2\sqrt{K_L'/B}$ (where $K_L'=K_L+K_1$). The

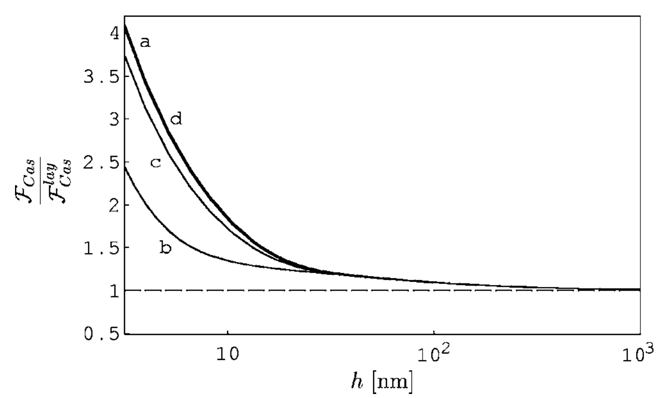

FIG. 3. Casimir force $\mathcal{F}_{\text{Cas}}$ in homeotropic smectic-A cell compared to approximate force $\mathcal{F}_{\text{Cas}}^{\text{lay}}$ for different director anchoring strengths: (a) $W\to\infty$, (b) $W=10^{-3}$ J/m^2, (c) $W=10^{-4}$ J/m^2, and (d) $W=10^{-5}$ J/m^2.

comparison between this approximate force $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}$ and our model, where the director degrees of freedom are included, is presented in Fig. 3. We used the following material constants: $B=2\times10^6\ \mathrm{N/m^2}$, $D=10^5\ \mathrm{N/m^2}$, $K_1=K_2=K_3=K_L=10^{-11}$ N. The exact Casimir force $\mathcal{F}_{\mathrm{Cas}}$ is significantly larger than the approximate force $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}$ only up to the thickness of a few correlation lengths Λ ($\Lambda=10$ nm) where the short-range contributions $\mathcal{F}[n_2;L]$ and $\mathcal{F}_2[n_1,u]$ are important. At larger thicknesses, only the long-range contribution of the in-phase director-layer fluctuations $\mathcal{F}_1[n_1,u]$ needs to be considered. In the limit of $h/\Lambda \gg 1$, this contribution ($\mathcal{F}_1[n_1,u]$) exactly matches $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}$, as can be seen from Fig. 3 and can also be shown analytically. A finite strength of the director anchoring generally reduces the magnitude of the Casimir force, as was explained in previous studies [21,26]. The force is the strongest when the anchoring is either very weak or very strong. When the anchoring is somewhere in between these limits, in the sense that the extrapolation length L is comparable to typical lengths of the system (correlation length or separation), then the magnitude is strongly reduced. This is seen in the case of $W=10^{-3}\ \mathrm{J/m^2}$ ($L=10$ nm) in Fig. 3, while in other cases the anchoring does not have an important effect.

In order to reveal the net effect of the director-layer coupling, it is also instructive to compare the "coupled" Casimir force to its "uncoupled" counterpart, which is obtained when director and layer fluctuations are treated independently. This uncoupled force is equal to $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{unc}}=\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}+\mathcal{F}_{\mathrm{Cas}}^{\mathrm{dir}}$, where $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}$ is the contribution of positional fluctuations with the director fixed perpendicular to the layers, as it was introduced in the previous paragraph. The contribution of the director degrees of freedom $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{dir}}$ is just twice the contribution of "pure" director fluctuation modes $\mathcal{F}[n_2;L]$ ($\mathcal{F}_{\mathrm{Cas}}^{\mathrm{dir}}=2\mathcal{F}[n_2;L]$). The comparison between $\mathcal{F}_{\mathrm{Cas}}$ and $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{unc}}$ is shown in Fig. 4 for different coupling constants D. It turns out that the coupling increases the magnitude of the force but for no more than a few ten percents. The increase is larger for weaker coupling constants D (larger correlation length Λ) and is 0 in the limit of $D\to\infty$. The profiles in Fig. 4 can be explained as follows. In the limit of very large thicknesses ($h\gg\Lambda$), the terms $\mathcal{F}_1[n_1,u]$ and $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}$, which are the only long-range contributions, are equal and the ratio between the coupled and uncoupled force is 1. With decreasing distance, the $\mathcal{F}_1[n_1,u]$ term gets larger than $\mathcal{F}_{\mathrm{Cas}}^{\mathrm{lay}}$ and the ratio increases. At thicknesses comparable to correlation length Λ, the short-range contributions from the director and out-of-phase fluctuations set in, which results in reducing the difference between the two forces.

The effect of different coupling strengths between the director and smectic layers on the Casimir force is presented explicitly in Fig. 5. Reduction of the coupling constant D results in an increase of the magnitude of the force. This is first due to the increased correlation length Λ (and, hence, increased range of the director-type contributions) and second also due to the larger coupling effect as it was demonstrated in Fig. 4. This kind of behavior could be observed when cooling the system from the smectic-A to smectic-C phase. In the vicinity of the phase transition, the coupling constant changes as $D\propto(T-T_c)$ within the Landau model. Therefore, the magnitude of the Casimir force is expected to increase while approaching the phase transition. This specific temperature dependence could make the Casimir force distinguishable from other interactions present in the system, thereby facilitating its detection. However, in order to give a correct description of the system at the phase transition, anharmonic terms should also probably be added to the Hamiltonian.

B. Free-standing film

The Casimir force in a free-standing film has a similar form as in the homeotropic cell [Eq. (18)]

$$\mathcal{F}_{\mathrm{Cas}}=\mathcal{F}[n_2;L]+\mathcal{F}_1[n_1,u]+\mathcal{F}_2[n_1,u]+\mathcal{F}_3[n_1,u;L,\chi]. \tag{23}$$

The first three terms are identical in both systems. The last term $\mathcal{F}_3[n_1,u;L,\chi]$, which describes the effect of finite director and layer anchoring strengths, becomes more complicated

$$\begin{aligned}\mathcal{F}_3[n_1,u;L,\chi]=-\frac{k_BTS}{4\pi}\sum_{i=1,2}\int_0^\infty q\,\mathrm{d}q\Bigg[&\Omega_1\Omega_2\lambda^{-2}\left(\frac{\Omega_1A_2^{\mp}}{1\pm\cosh(\Omega_1h)}+\frac{\Omega_2A_1^{\mp}}{1\pm\cosh(\Omega_2h)}\right)\\&+\chi^{-1}q^2\left(\frac{\Omega_1^2S^2}{1\pm\cosh(\Omega_1h)}+\frac{\Omega_2^2C^2}{1\pm\cosh(\Omega_2h)}\right)+L^{-1}\lambda^{-2}\left(\frac{\Omega_1^2C^2}{1\pm\cosh(\Omega_1h)}+\frac{\Omega_2^2S^2}{1\pm\cosh(\Omega_2h)}\right)\Bigg]\\&\times[\Omega_1\Omega_2\lambda^{-2}A_1^{\mp}A_2^{\mp}+\chi^{-1}q^2(\Omega_1S^2A_1^{\mp}+\Omega_2C^2A_2^{\mp}+L^{-1})+L^{-1}\lambda^{-2}(\Omega_1C^2A_1^{\mp}+\Omega_2S^2A_2^{\mp})]^{-1}.\end{aligned} \tag{24}$$

It is a sum of two contributions ($i=1,2$) that differ only by sign alternation ($\pm$) in some terms.

The effect of the finite surface tension γ on the Casimir force in free-standing smectic films is presented in Fig. 6. We compare the force in a free-standing film to the corresponding force in homeotropic cell for a specific director anchoring strength $W=10^{-5}\ \mathrm{J/m^2}$ (the director anchoring is not essential in this case, and choosing some other value of W would lead to very similar results). As it is seen from Fig. 6 the finite surface tension γ reduces the magnitude of the

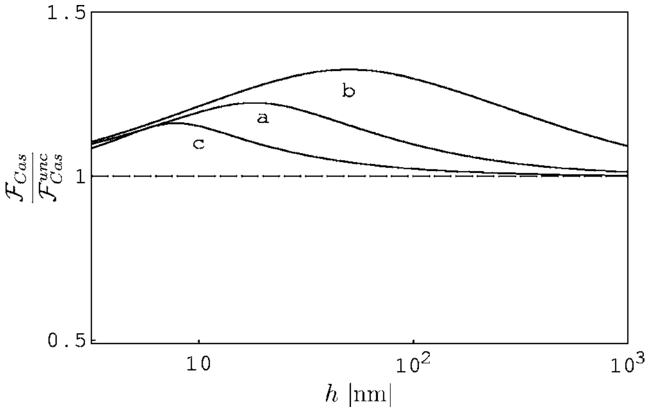

FIG. 4. Comparison between the coupled ($\mathcal{F}_{\rm Cas}$) and uncoupled ($\mathcal{F}_{\rm Cas}^{\rm unc}$) Casimir force in homeotropic cell for infinitely strong director anchoring ($W\to\infty$) and various coupling constants D: (a) $D=10^5$ N/m^2, (b) $D=10^4$ N/m^2, and (c) $D=10^6$ N/m^2.

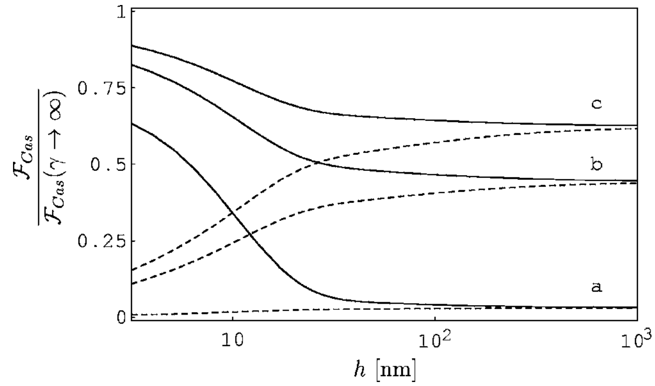

FIG. 6. Casimir force in free-standing film compared to the force in homeotropic cell [$\mathcal{F}_{\rm Cas}(\gamma\to\infty)$] for the director anchoring strength $W=10^{-5}$ J/m^2, coupling constant $D=10^5$ N/m^2, and different surface tensions: (a) $\gamma=10^{-2}$ J/m^2, (b) $\gamma=5\times10^{-2}$ J/m^2, and (c) $\gamma=10^{-1}$ J/m^2. The dashed lines represent $\mathcal{F}_{\rm Cas}^{\rm lay}(\gamma)$.

Casimir force. This effect was already predicted by Mikheev [13] within the simplified model, where only positional fluctuations of smectic layers were taken into account. He obtained the following result for the force: $\mathcal{F}_{\rm Cas}^{\rm lay}(\gamma)=-k_BTS/16\pi h^2\sqrt{K_L'/B}\,Li_2[(\gamma-\sqrt{K_L'B})/(\gamma+\sqrt{K_L'B})]^2$, where Li_2 is the dilogarithm function. Our result is in agreement with this simplified model in the limit of large thicknesses h as it is indicated by the dashed lines in Fig. 6. At smaller thicknesses, the effect of director degrees of freedom becomes important and it eventually reduces the difference between the compared forces.

It is again instructive to compare the coupled Casimir force [Eq. (23)] to the uncoupled force, where the director and layer fluctuations are treated independently: $\mathcal{F}_{\rm Cas}^{\rm unc}=\mathcal{F}_{\rm Cas}^{\rm lay}(\gamma)+2\mathcal{F}[n_2;L]$. As it is shown in Fig. 7, the net effect of coupling is to increase the magnitude of the force, similar as in the homeotropic cell. The increase is substantial in the case of a small surface tension γ [Fig. 7(a)], while it does not exceed a few ten percents otherwise.

V. OBSERVABILITY

The forces in liquid-crystal systems are usually measured by atomic force microscopy (AFM) and a surface-force apparatus (SFA) [37]. The geometry of these experimental setups is not plan-parallel, as in our study, but consists of a sphere and a plane (AFM) or two crossed cylinders (SFA). Fortunately, the force in these curved geometries can be related to the interaction in a plan-parallel system through the Derjaugin approximation [38]

$$\mathcal{F}=2\pi R\frac{F_{\rm int}}{S}, \tag{25}$$

where $F_{\rm int}/S$ is the free energy per surface unit in plan-parallel geometry and R is the radius of the sphere (AFM) or of the cylinders (SFA). The validity of the Derjaugin approximation is limited to the systems where the curved surfaces do not induce significant distortions of the liquid-crystal structure. This is difficult to achieve in nematic liquid crystals, where the director can be easily deformed, which results in an additional mean-field interaction. This mean-field interaction is usually much stronger than the Casimir force, which makes the experimental detection of the latter

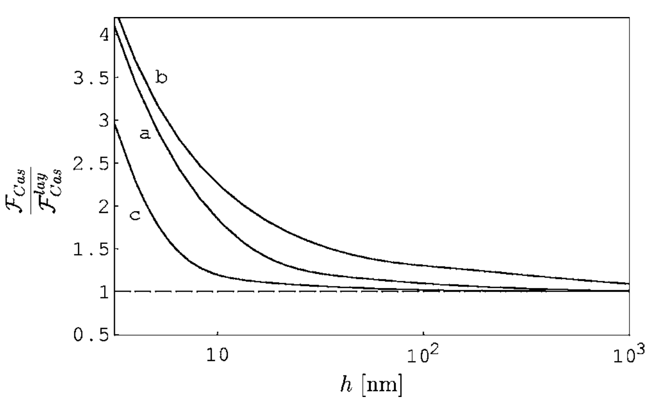

FIG. 5. Effect of director-layer coupling constant D on the Casimir force: (a) $D=10^5$ N/m^2, (b) $D=10^4$ N/m^2, and (c) $D=10^6$ N/m^2. Strong anchoring of the director ($W\to\infty$) is assumed.

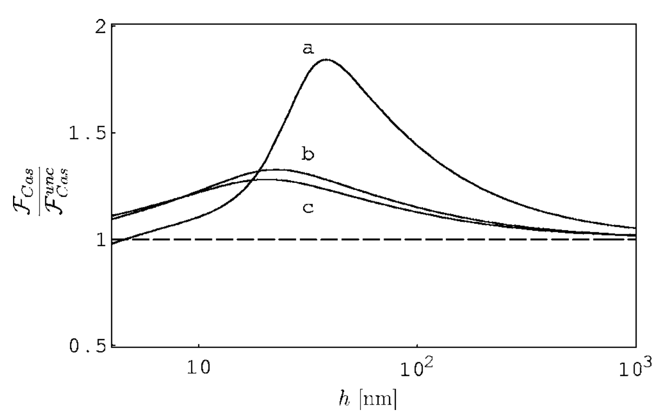

FIG. 7. Comparison between the coupled ($\mathcal{F}_{\rm Cas}$) and uncoupled ($\mathcal{F}_{\rm Cas}^{\rm unc}$) Casimir force in a free-standing film for the director anchoring strength $W=10^{-5}$ J/m^2, coupling constant $D=10^5$ N/m^2, and different surface tensions: (a) $\gamma=10^{-2}$ J/m^2, (b) $\gamma=5\times10^{-2}$ J/m^2, and (c) $\gamma=10^{-1}$ J/m^2.

difficult [32]. Smectic layers are much "stiffer" than the nematic director. Therefore, a smectic system adjusts to the curved surfaces by formation of an array of edge dislocation loops, whereas the smectic layers do not bend considerably [39]. The force profile due to the compression (or dilation) of smectic layers in this arrangement is comprised of quasiperiodic parabolas [39,40]. These parabolas are often found to reside on an attractive background [39,41,42] whose origin is not yet fully understood. It seems that the Casimir force is about an order of magnitude too small to be responsible for this. In addition to the Casimir force, the enhanced positional order at the surfaces has also been suggested as a possible source of the observed attraction [37,39]; but to give a definite answer, more studies should be performed.

Let us evaluate to what extent could the Casimir force in smectics be detected by AFM and SFA. The force sensitivity of AFM is about 10 pN, and the sphere is usually not larger than 20 μm [32,37]. Using the Derjaugin approximation, we can estimate that this suffices for the detection of the Casimir force up to the thickness of about 1 μm, which is quite a lot. For the SFA with typical radius $R=2$ cm and the force sensitivity of about 10 nN [37], we obtain a similar estimate. Both devices are also capable of measuring forces at very small thicknesses of the sample down to just a single molecular layer, which might enable one to detect the influence of the director degrees of freedom. A convenient characteristic of the Casimir force in smectics is also that it does not significantly depend on the specific director anchoring conditions (see Fig. 3), which are not always well known. This is quite the opposite, as in the nematics (where the behavior of the Casimir force strongly depends on the boundary conditions [32]), and facilitates the interpretation of experimental results.

In addition to the direct measurements, the impact of the Casimir force could also be observed indirectly—for example, in wetting behavior [13,15], light scattering from the surface of a free-standing smectic film [43], and in structure of some lamellar lyotropic systems [44].

VI. CONCLUSION

In conclusion, we studied the Casimir force in two confined smectic-A systems: homeotropic cell and free-standing film. We calculated the force considering the fluctuations of smectic layers and director while the degree of smectic order was assumed to be constant. The main focus was on the effect of the director-layer coupling on the Casimir force. We found that the coupling increases the magnitude of the force, compared to the uncoupled system, and analyzed the effect of different coupling strengths. The influence of the director degrees of freedom and their coupling to the smectic layers is most effective at small thicknesses of the system. At very large thicknesses the results of our model are identical to the results of a simplified model [13,15] where only the layer-displacement variable u was considered. Finally, we discussed the possibilities of experimental detection of the Casimir force in smectics.

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