Pseudo-Casimir force in chiral smectic liquid crystals

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We present a theoretical study of the pseudo-Casimir force in two chiral smectic systems: a homeotropic cell and a free-standing film. We consider the interaction induced by the fluctuations of orientational order. We demonstrate how the character of the force depends on the type of fluctuation modes and on boundary conditions. We focus on the temperature dependence of the force, which is marked by the vicinity of the smectic-$A^*\rightarrow$smectic-C^* phase transition. We find that at this transition the force diverges if the system is frustrated; otherwise it remains finite. We expose the analogy between the force in these smectic systems and in previously studied nematic systems, thus demonstrating the universality of the pseudo-Casimir interaction.

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I. INTRODUCTION

The interest in fluctuation-induced interactions started with Casimir's pioneering work on the attraction between isolated conducting walls induced by the vacuum fluctuations of the electromagnetic field [1]. This work was followed by studies of the Casimir effect in a large variety of physical systems [2] such as liquid crystals, which are characterized by ''softness'' and richness of phases. Ajdari *et al.* considered the pseudo-Casimir force for the simple geometry of two flat parallel plates immersed in nematic, smectic, and columnar liquid crystal phases [3,4]. The smectic case was actually studied even earlier by Mikheev [5]. These results were generalized to rough substrates [6,7] and finite strength of surface interaction [8]. Prenematic and presmectic wetting systems with inhomogeneous equilibrium ordering (i.e., nontrivial ground state) were studied in Refs. [9,10]. Recently, most of attention was paid to the so-called ''frustrated systems'' such as the nematic hybrid and the Freedericksz cell [11]. Studies were also extended to the chiral nematics [12]. The extensive theoretical efforts have unfortunately not yet been followed by experimental confirmations. The possibilities of the experimental detection of the pseudo-Casimir force in liquid crystals were thoroughly discussed in Ref. [13]. Some recently performed experiments on the spinodal dewetting of thin nematic films [14,15] seem to offer a promising method for the indirect identification of the pseudo-Casimir and van der Waals forces [16,17].

In this paper we present a theoretical study of the pseudo-Casimir effect in chiral smectics. We consider the interaction induced by the thermal fluctuations of the orientational order. The effect of the fluctuations of spontaneous polarization is simplified by the use of the adiabatic approximation [18]. The fluctuations of the positional order, discussed in Refs. [3–5], are not treated here. Our analysis is based on the phenomenological Landau-type description of the Sm-$A^*\rightarrow$Sm-C^* phase transition, while fluctuations are treated within the Gaussian approximation. The surface interaction is described by the Rapini-Papoular model. The details of the theoretical model are explained in Sec. II.

We calculate and analyze the pseudo-Casimir force in two confined smectic systems with planar geometry. In Sec. III we consider the pseudo-Casimir interaction in the homeotropic cell. We distinguish two cases with the temperature of the system T being either above or below the Sm-$A^*\rightarrow$Sm-C^* bulk phase transition temperature T_c. In the first case ($T>T_c$) the emphasis is given to the effect of anchoring strengths on the force, and we extend the conclusions of the previous studies [8,11,13] to the smectic case. In the case of $T<T_c$ we restrict our analysis to cells thin enough such that the equilibrium structure is still homogeneous Sm-A^*, due to the homeotropic anchoring. This is an example of a ''frustrated'' system as the tendency of the smectic director to tilt is suppressed by the surface interaction. In Sec. IV we address the pseudo-Casimir interaction in free-standing smectic films. We consider two cases: the free-standing Sm-A^* film and the free-standing Sm-C^* film. We introduce a simple model of internal anchoring at the free surfaces. We restrict our attention to the films with the homogeneous equilibrium profile, and do not consider the possibility that in free-standing smectic films the ordering of molecules in surface layers can be different from the ordering in interior layers [19]. Throughout this paper we compare our results with the previous studies in nematic systems and demonstrate the universality of the pseudo-Casimir interaction. Its behavior depends on the type of fluctuating modes (i.e., ''massive'' or ''massless'') and on the boundary conditions, while other details of the system do not play an important role. We summarize and comment our results in Sec. V.

II. THEORETICAL MODEL

The theoretical model is based on the Landau free energy expansion in the vicinity of the Sm-$A^*\rightarrow$Sm-C^* phase transition [18,20,21]. The primary order parameter of this transition—a two-dimensional vector $\boldsymbol{\xi}=(\xi_x,\xi_y)$—represents the average tilt of molecules (i.e., director) with respect to the normal to smectic layers. ξ_x,ξ_y are the projections of the tilt onto the smectic plane $(x\text{-}y)$. The secondary order parameter is the spontaneous polarization $\mathbf{P}=(P_x,P_y)$, which is oriented parallel to smectic planes—perpendicular

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to the director and to the layer normal. The polarization fluctuations are much faster than the director fluctuations [18,21]. We shall therefore make the usual assumption that the polarization is always in equilibrium with the director, and perform our calculations retaining only the orientational order parameter $\boldsymbol{\xi}$. This is the so-called ''adiabatic'' approximation [18]. We will also not consider the fluctuations of the positional order, which are not directly related to the Sm-$A^*\rightarrow$Sm-C^* phase transition. In this approximation the Landau free energy density reads

$$f=f_A+\frac{1}{2}a(T)(\xi_x^2+\xi_y^2)+\frac{1}{4}b(\xi_x^2+\xi_y^2)^2 - \Lambda\left(\xi_x\frac{d\xi_y}{dz}-\xi_y\frac{d\xi_x}{dz}\right)+\frac{1}{2}K_1\left(\frac{d\xi_x}{dx}+\frac{d\xi_y}{dy}\right)^2 +\frac{1}{2}K_2\left(\frac{d\xi_x}{dy}-\frac{d\xi_y}{dx}\right)^2+\frac{1}{2}K_3\left[\left(\frac{d\xi_x}{dz}\right)^2+\left(\frac{d\xi_y}{dz}\right)^2\right], \quad (1)$$

where $a(T)$ is a temperature dependent coefficient that drives the phase transition and b is a constant positive coefficient. The so-called Lifshitz term $\Lambda[\xi_x(d\xi_y/dz)-\xi_y(d\xi_x/dz)]$ is responsible for the helical structure of the Sm-C^* phase, while K_i are the elastic constants. It is more convenient to represent the order parameter $\boldsymbol{\xi}$ in the rotating reference frame, which follows the helical structure of the Sm-C^* phase. The transformation reads $\xi_x=\xi_\parallel\cos(q_cz)-\xi_\perp\sin(q_cz),\xi_y=\xi_\parallel\sin(q_cz)+\xi_\perp\cos(q_cz)$, where q_c is the wave vector of the helix. The free energy density is now expressed with new parameters $\xi_\parallel$ and $\xi_\perp$:

$$f=f_A+\frac{1}{2}[a(T)-K_3q_c^2](\xi_\parallel^2+\xi_\perp^2)+\frac{1}{4}b(\xi_\parallel^2+\xi_\perp^2)^2 +\frac{1}{2}K\left[\left(\frac{d\xi_\parallel}{dx}+\frac{d\xi_\perp}{dy}\right)^2+\left(\frac{d\xi_\parallel}{dy}-\frac{d\xi_\perp}{dx}\right)^2\right] +\frac{1}{2}K_3\left[\left(\frac{d\xi_\parallel}{dz}\right)^2+\left(\frac{d\xi_\perp}{dz}\right)^2\right], \quad (2)$$

where we have set $K_1=K_2=K$ for simplicity. The coefficient $a(T)-K_3q_c^2$ has the form $\alpha(T-T_c)$, where T_c is the temperature of the Sm-$A^*\rightarrow$Sm-C^* phase transition in bulk material and α is a positive material constant.

The surface interaction free energy is in both studied systems described by the phenomenological Rapini-Papoular form $F_{RP}=\frac{1}{2}W_i\int\sin^2(\xi_i-\xi_{is})dS$, where ξ_{is} is the preferred value of the order parameter at the surface, and i stands either for $\parallel$ or $\perp$. The anchoring strength is characterized by the coefficient W_i.

The Hamiltonian of fluctuations is obtained by expanding the free energy of a system around the equilibrium configuration. The order parameter is written as the sum of the mean-field value plus the fluctuating part, $\boldsymbol{\xi}=\boldsymbol{\xi}_0+\delta\boldsymbol{\xi}=(\xi_{\parallel 0}+\delta\xi_\parallel,\xi_{\perp 0}+\delta\xi_\perp)$, and inserted into the free energy expression [Eq. (2)]. We neglect higher order fluctuation terms, keeping only the harmonic part of the Hamiltonian. From here on Sm-A^* and Sm-C^* phases have to be treated separately. In the Sm-A^* phase the mean-field value of the order parameter $\boldsymbol{\xi}$ is equal to $\mathbf{0}$. In the Sm-C^* phase the mean-field value of $\xi_\parallel$ is equal to $\xi_{\parallel 0}=\sqrt{(\alpha/b)(T_c-T)}$, while $\xi_{\perp 0}=0$. The Hamiltonian densities of fluctuations then read

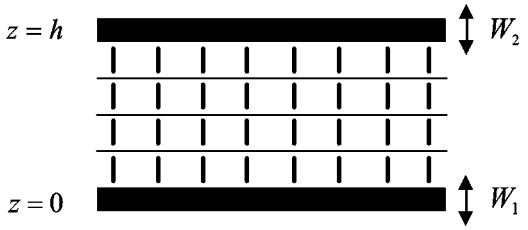

FIG. 1. Homeotropic cell. The arrows indicate the preferential orientation of the director at the plates.

$$h_{fluc}=\frac{1}{2}K_3\begin{Bmatrix}\eta^{-2}(\delta\xi_\parallel^2+\delta\xi_\perp^2)\\ \rho^{-2}\delta\xi_\parallel^2\end{Bmatrix}+\frac{1}{2}K_3\left[\left(\frac{d(\delta\xi_\parallel)}{dz}\right)^2+\left(\frac{d(\delta\xi_\perp)}{dz}\right)^2\right]+\frac{1}{2}K\left[\left(\frac{d(\delta\xi_\parallel)}{dx}+\frac{d(\delta\xi_\perp)}{dy}\right)^2+\left(\frac{d(\delta\xi_\parallel)}{dy}-\frac{d(\delta\xi_\perp)}{dx}\right)^2\right], \quad (3)$$

where in the first term the upper line corresponds to the Sm-A^* phase and the lower to the Sm-C^* phase. We have introduced the correlation lengths of fluctuations: $\eta=(a/K_3-q_c^2)^{-1/2}$ and $\rho=[2(-a/K_3+q_c^2)]^{-1/2}$. From Eq. (3) it is seen that in the Sm-A^* phase both $\delta\xi_\parallel$ and $\delta\xi_\perp$ modes are ''massive'' with the correlation length η, whereas in the Sm-C^* phase the $\delta\xi_\parallel$ mode is ''massive'' with the correlation length ρ and the $\delta\xi_\perp$ mode is ''massless'' with infinite correlation length.

III. HOMEOTROPIC CELL

The homeotropic cell consists of a Sm-A^* material trapped between two parallel flat plates separated by the distance h (Fig. 1). Smectic layers are parallel to the plates. The preferential orientation of the director at the boundaries is perpendicular to the plates. In this case the surface contribution to the free energy density reads

$$f_S=\frac{1}{2}W_1\boldsymbol{\xi}^2\delta(z)+\frac{1}{2}W_2\boldsymbol{\xi}^2\delta(z-h), \quad (4)$$

where we allow for different anchoring strengths W_1,W_2 at the plates, and retain only the harmonic term of the Rapini-Papoular form. It is more common to represent anchoring strengths in terms of extrapolation lengths defined by $\lambda_j=K_3/W_j$.

As a consequence of these boundary conditions, the Sm-A^* structure in the homeotropic cell is stable even below the bulk Sm-$A^*\rightarrow$Sm-C^* phase transition temperature T_c. In this case the system is in a ''frustrated'' state. Until the temperature is low enough, the anchoring on the plates prevails over the tendency of smectic to tilt. The temperature of the transition from the Sm-A^* structure to the deformed

Sm-C^* structure depends on the thickness of the cell. The critical thickness h_c (at some fixed temperature $T<T_c$) is evaluated by minimizing the free energy of the system while retaining only quadratic terms in the order parameter $\boldsymbol{\xi}$:

$$h_c=\sqrt{2}\rho\ \mathrm{arccot}\left(\frac{\lambda_1\lambda_2-(\sqrt{2}\rho)^2}{\sqrt{2}\rho(\lambda_1+\lambda_2)}\right). \tag{5}$$

For the limiting case of the infinitely strong anchoring ($\lambda_1=0$, $\lambda_2=0$) the critical thickness is equal to $h_c=\sqrt{2}\pi\rho$. The transition from the Sm-A^* to the Sm-C^* structure in the homeotropic cell is analogous to the Fréedericksz transition in the nematic homeotropic cell [22]. While the Fréedericksz transition is driven by the quadratic coupling between an external magnetic field and the director, in our case the transition is induced by an ''internal,'' temperature dependent, smectic field. In this paper we will consider only the case of $h<h_c$, where the equilibrium structure between the plates is homogeneous Sm-A^*.

A. Fluctuations free energy

In order to obtain the free energy of fluctuations, F_{fluc}, we have to evaluate the partition function Z:

$$Z=\exp(-\beta F_{fluc})=\int \exp(-\beta H[\,\delta\xi_\parallel(\mathbf{r}),\delta\xi_\perp(\mathbf{r})\,]) \times\mathcal{D}(\delta\xi_\parallel(\mathbf{r}))\mathcal{D}(\delta\xi_\perp(\mathbf{r})), \tag{6}$$

where $\beta=1/k_BT$. Due to the translational invariance of the studied system in the x-y plane, it is convenient to introduce the Fourier transformations of fluctuating fields $\delta\xi_{\parallel,\perp}(\mathbf{r})=\Sigma_{\mathbf{q}}\exp[i(q_xx+q_yy)]\tilde{\xi}_{\parallel,\perp}(\mathbf{q},z)$, where $\mathbf{q}=(q_x,q_y)$. The Hamiltonian is now reduced to an ensemble of independent harmonic oscillators, $H=\Sigma_{\mathbf{q}}(H_{\mathbf{q}}[\tilde{\xi}_\parallel]+H_{\mathbf{q}}[\tilde{\xi}_\perp])$. In the Sm-$A^*$ phase, fluctuation modes $\tilde{\xi}_\parallel$ and $\tilde{\xi}_\perp$ are degenerate. The Hamiltonian $H_{\mathbf{q}}[\tilde{\xi}_\parallel]=H_{\mathbf{q}}[\tilde{\xi}_\perp]$ reads

$$H_{\mathbf{q}}[\tilde{\xi}_{\parallel,\perp}]=\frac{1}{2}K_3S\left\{\int_0^h\left[\left(\eta^{-2}+\frac{K}{K_3}q^2\right)\tilde{\xi}_{\parallel,\perp}^2+\left(\frac{d\tilde{\xi}_{\parallel,\perp}}{dz}\right)^2\right]dz +\lambda_1^{-1}\tilde{\xi}_{\parallel,\perp}^{-2}+\lambda_2^{-1}\tilde{\xi}_{\parallel,\perp}^{+2}\right\}, \tag{7}$$

where S is the area of the plates, $\tilde{\xi}_{\parallel,\perp}^-=\tilde{\xi}_{\parallel,\perp}(z=0)$, and $\tilde{\xi}_{\parallel,\perp}^+=\tilde{\xi}_{\parallel,\perp}(z=h)$. The partition function can now be factorized as $Z=\Pi_{\mathbf{q}}Z_{\mathbf{q}}[\tilde{\xi}_\parallel]Z_{\mathbf{q}}[\tilde{\xi}_\perp]=\Pi_{\mathbf{q}}Z_{\mathbf{q}}^2$ and the free energy of fluctuations can be written as $F_{fluc}=-2k_BT\Sigma_{\mathbf{q}}\ln Z_{\mathbf{q}}$. The partial partition function $Z_{\mathbf{q}}$ is analogous to the propagator of a quantum-mechanical harmonic oscillator [23], with the exception that in the case of finite anchoring strengths, fluctuations at the boundaries should also be allowed. Such a problem was treated in Ref. [11].

The partial partition function $Z_{\mathbf{q}}$ depends on the value of parameter $p^2=\eta^{-2}+(K/K_3)q^2$. If $p^2>0$, then $Z_{\mathbf{q}}$ is proportional to

$$Z_{\mathbf{q}}\propto\left(\frac{\lambda_1^{-1}\lambda_2^{-1}+p^2}{p(\lambda_1^{-1}+\lambda_2^{-1})}\sinh(ph)+\cosh(ph)\right)^{-1/2}, \tag{8}$$

and in the case of $p^2<0$, $Z_{\mathbf{q}}$ reads

$$Z_{\mathbf{q}}\propto\left(\frac{\lambda_1^{-1}\lambda_2^{-1}-p^2}{p(\lambda_1^{-1}+\lambda_2^{-1})}\sin(ph)+\cos(ph)\right)^{-1/2}, \tag{9}$$

where with p we now denote the absolute value. The first case corresponds to a harmonic oscillator in repulsive potential and the second case to a harmonic oscillator in attractive potential, as can be seen from Eq. (7). The total free energy of fluctuations can now be written as

$$F_{fluc}(T>T_c)=\frac{k_BTS}{2\pi}\frac{K_3}{K}\int_{1/\eta}^{\infty}\ln\left(\frac{\lambda_1^{-1}\lambda_2^{-1}+p^2}{p(\lambda_1^{-1}+\lambda_2^{-1})}\sinh(ph)+\cosh(ph)\right)pdp \tag{10}$$

for $T>T_c$ and

$$F_{fluc}(T<T_c)=\frac{k_BTS}{2\pi}\frac{K_3}{K}\int_0^{\infty}\ln\left(\frac{\lambda_1^{-1}\lambda_2^{-1}+p^2}{p(\lambda_1^{-1}+\lambda_2^{-1})}\sinh(ph)+\cosh(ph)\right)pdp+\frac{k_BTS}{2\pi}\frac{K_3}{K}\int_0^{(\sqrt{2}\rho)^{-1}}\times\ln\left(\frac{\lambda_1^{-1}\lambda_2^{-1}-p^2}{p(\lambda_1^{-1}+\lambda_2^{-1})}\sin(ph)+\cos(ph)\right)pdp, \tag{11}$$

for $T<T_c$, meanwhile replacing the sum over $\mathbf{q}$ by an integral.

B. Pseudo-Casimir force

The structural force is defined as

$$\mathcal{F}=-\frac{\partial F_{int}}{\partial h}. \tag{12}$$

Here the interaction free energy F_{int} is measured from a reference bulk free energy [11]. We follow the procedure where the interaction part of the free energies is identified by factorizing F_{fluc} [Eqs. (10) and (11)] into a bulk contribution proportional to Sh, a surface contribution independent of h, and an interaction contribution, as described in Ref. [11].

For $T>T_c$, the force reads

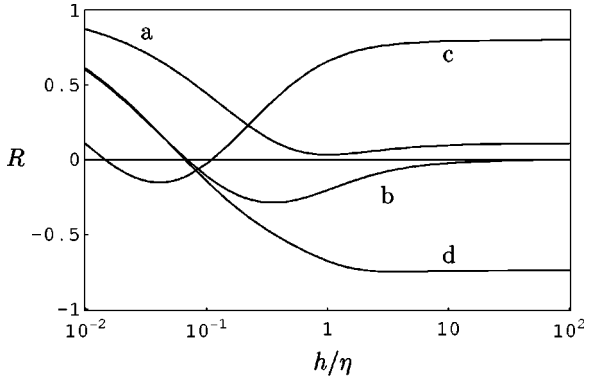

FIG. 2. Pseudo-Casimir force in the homeotropic cell for $T > T_c$. The dependence of the reduced amplitude R on the parameter h/η is presented for different sets of anchoring strengths: (a) $\lambda_1/\eta = 0.5$, $\lambda_2/\eta = 0.5$; (b) $\lambda_1/\eta = 1$, $\lambda_2/\eta = 0.05$; (c) $\lambda_1/\eta = 0.1$, $\lambda_2/\eta = 0.01$; and (d) $\lambda_1/\eta = 10$, $\lambda_2/\eta = 0.05$.

$$\mathcal{F}_{Cas} = -\frac{k_B T S}{\pi}\frac{K_3}{K}\int_{1/\eta}^{\infty}\frac{p^2 dp}{\frac{(p+\lambda_1^{-1})(p+\lambda_2^{-1})}{(p-\lambda_1^{-1})(p-\lambda_2^{-1})}\exp(2ph)-1}. \tag{13}$$

This integral can be evaluated numerically. An analytical expression can be obtained for the limiting case of infinitely strong anchoring at the plates ($\lambda_1 = 0, \lambda_2 = 0$),

$$\mathcal{F}_{Cas}(\lambda_1 = 0, \lambda_2 = 0) = -\frac{k_B T S}{2\pi}\frac{K_3}{K}\frac{1}{h^3}\sum_{k=1}^{\infty}\frac{\exp\left(-2\frac{h}{\eta}k\right)}{k^3} \times\left(\frac{1}{2}+\frac{h}{\eta}k+\frac{h^2}{\eta^2}k^2\right). \tag{14}$$

This result is analogous to the pseudo-Casimir force induced by massive fluctuation modes in the homeotropic nematic cell [8]. The character of the pseudo-Casimir force is determined by the character of fluctuation modes, and corresponding boundary conditions. In this case the fluctuation modes are massive, and therefore the force is short range. Some profiles of the pseudo-Casimir force for different sets of anchoring strengths are shown in Fig. 2. Presented is the reduced amplitude of the pseudo-Casimir force as compared to the force in the case of symmetric infinitely strong anchoring conditions:

$$R = \frac{\mathcal{F}_{Cas}(\lambda_1, \lambda_2, h, \eta)}{\mathcal{F}_{Cas}(\lambda_1 = 0, \lambda_2 = 0, h, \eta)}. \tag{15}$$

This is a generalization of the former work in nematics [8], where the contribution of massive fluctuation modes was considered only for the case of symmetric anchoring conditions ($\lambda_1 = \lambda_2$). The profiles in Fig. 2 can be explained by the interplay of four characteristic lengths: distance between plates (h), correlation length of fluctuations (η), and two extrapolation lengths (λ_1, λ_2). It is known from the previous studies of the pseudo-Casimir effect [11] that in the case of symmetric boundary conditions (strong-strong or weak-weak anchoring at the plates) the force is attractive, whereas in the case of antisymmetric boundary conditions (strong-weak anchoring at the plates) the force is repulsive. In our system it is not very obvious which parameters determine the effective anchoring strengths. It seems (Fig. 2) that there are actually two different regimes. When $h/\eta < 1$, the effective anchoring strengths are determined by λ_1/h and λ_2/h. In the case of $\lambda_i/h < 1$ the anchoring is effectively strong; and correspondingly, if $\lambda_i/h > 1$, the anchoring is effectively weak. In the second regime, where $h/\eta > 1$, the effective anchoring strengths are determined by parameters λ_1/η and λ_2/η, using the same criteria as in the first regime. This can be explained if we recall that the anchoring is effectively strong when the interaction between the substrate and liquid crystal is stronger than the internal interaction in the liquid crystal [24]. The strength of the surface interaction is measured by the extrapolation lengths λ_i. The internal interaction includes two contributions, as can be seen from Eq. (3): the "massive" contribution whose strength is characterized by η^{-1}, and the elastic contribution which scales as h^{-1}. At small h/η the elastic contribution dominates, and the effective strength of the anchoring is obtained by comparing parameters λ_i and h. At large h/η the "massive" contribution is dominant, and consequently, the effective strength of the anchoring depends on parameters λ_i and η.

All the lengths in Fig. 2 are scaled by the correlation length η. The values of parameters λ_i/h change by varying the parameter h/η. Therefore the pseudo-Casimir force in the first regime ($h/\eta < 1$) exhibits crossovers from attractive to repulsive and vice versa [Figs. 2(b)–(d)]. The parameters λ_i/η are fixed, therefore the character of the force in the second regime ($h/\eta > 1$) does not change. It should be kept in mind that η is temperature dependent, and that the character of the force is, consequently, also temperature dependent. At large separations ($h/\eta \gg 1$), the reduced amplitude R saturates at a constant value. This shows that in this regime the force has the same functional form as the leading term in the case of infinitely strong anchoring [Eq. (14)], which decays as $\exp(-2h/\eta)/h$. The saturation value is the largest when the anchoring at the plates is either very strong or very weak. It can be shown that in the case of very strong anchoring at both plates ($\lambda_1/\eta, \lambda_2/\eta \ll 1$) the reduced amplitude saturates at $R = 1 - 2(\lambda_1/\eta + \lambda_2/\eta)$ [Fig. 2(c)], whereas in the case of very weak anchoring at both plates ($\lambda_1/\eta, \lambda_2/\eta \gg 1$) the saturation value is $R = 1 - 2(\eta/\lambda_1 + \eta/\lambda_2)$. In the antisymmetric case where the anchoring at one plate is very weak ($\lambda_1/\eta \gg 1$) and at the other plate very strong ($\lambda_2/\eta \ll 1$), the reduced amplitude saturates at $R = -1 + 2(\eta/\lambda_1 + \lambda_2/\eta)$ [Fig. 2(d)]. The behavior of the force at large separations is substantially modified in the case of $\lambda_i/\eta = 1$, where the anchoring at one or both plates is neither strong nor weak. It can be shown that in the first case the force decays as $\exp(-2h/\eta)/h^2$, and consequently, the reduced amplitude goes to zero at $h/\eta \gg 1$ [Fig. 2(b)]. In the case of $\lambda_1/\eta = \lambda_2/\eta = 1$, the force decays even faster—as $\exp(-2h/\eta)/h^3$.

The evaluation of the pseudo-Casimir force in the case of ''frustrated'' homeotropic system ($T<T_c$) is performed following the same procedure as described above. However, it should be noted that now the bulk reference structure is Sm-C^*, while the structure between the plates is still Sm-A^*. The pseudo-Casimir force in the case of $T<T_c$ reads

$$\mathcal{F}_{Cas}=-\frac{k_BTS}{\pi}\frac{K_3}{K}\left[\int_0^\infty \frac{p^2dp}{\frac{(p+\lambda_1^{-1})(p+\lambda_2^{-1})}{(p-\lambda_1^{-1})(p-\lambda_2^{-1})}\exp(2ph)-1}\right.$$
$$+\frac{1}{2}\int_0^{(\sqrt{2}\rho)^{-1}}\frac{(\lambda_1^{-1}\lambda_2^{-1}-p^2)\cot(ph)-p(\lambda_1^{-1}+\lambda_2^{-1})}{(\lambda_1^{-1}\lambda_2^{-1}-p^2)+p(\lambda_1^{-1}+\lambda_2^{-1})\cot(ph)}$$
$$\left.\times p^2dp+\frac{1}{12\rho^3}\right]. \qquad (16)$$

It is again instructive to consider the pseudo-Casimir force in the limiting case of infinitely strong anchoring ($\lambda_1=0$, $\lambda_2=0$),

$$\mathcal{F}_{Cas}(\lambda_1=0,\lambda_2=0)=-\frac{k_BTS}{4\pi}\frac{K_3}{K}\left[\frac{\zeta(3)}{h^3}+2\int_0^{(\sqrt{2}\rho)^{-1}}\right.$$
$$\left.\times\cot(ph)p^2dp+\frac{1}{3\rho^3}\right]. \qquad (17)$$

The first term in $\mathcal{F}_{Cas}$ [Eqs. (16) and (17)] has the typical form of the pseudo-Casimir interaction induced by massless fluctuation modes with infinite correlation lengths and is dominant at small h/ρ. This term is actually the same as the interaction induced by director fluctuations in the homeotropic nematic cell and was analyzed in detail in Ref. [13]. The second term becomes prominent in the vicinity of the structural transition from Sm-A^* to Sm-C^*, where it diverges. This repulsive divergence of the fluctuation-induced force is characteristic for the second-order transitions and is logarithmic, as noted in Ref. [11]. The last term results from the difference of the bulk free energy of fluctuations in Sm-A^* and reference Sm-C^* configuration. The behavior of this system is analogous to the nematic Fréedericksz cell [11], as we have already mentioned.

The temperature profile of the pseudo-Casimir force in the homeotropic cell, at some fixed thickness h, is shown in Fig. 3. We measure the temperature in dimensionless units: $t=(\alpha h^2/K_3)T$. The behavior of the force in the regime $t>t_c$ was commented along with Fig. 2. On supercooling the system ($t<t_c$) the force approaches some local minimum, and eventually it diverges at the structural transition to the Sm-C^* structure. The stronger the anchoring at the plates, the deeper the supercooling limit, and the more pronounced the minimum. Except at small t_c-t, the behavior of the force in this regime is dominated by the frustration driven part of the interaction—the last two terms in Eqs. (16) and (17). The last term is attractive and increases as $(t_c-t)^{3/2}$. Near the structural transition, the repulsive divergence of the second term prevails.

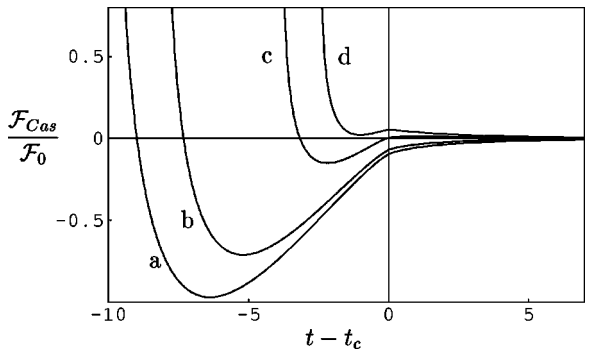

FIG. 3. Temperature profile of the pseudo-Casimir force in the homeotropic cell. We introduced a unitless temperature $t=\alpha h^2T/K_3$. The amplitude of the force is given in the natural unit $\mathcal{F}_0=k_BTSK_3/Kh^3$ (we neglect a weak temperature dependence of $\mathcal{F}_0$). The force is plotted for different sets of anchoring strengths: (a) $\lambda_1/h=0$, $\lambda_2/h=0$; (b) $\lambda_1/h=0.1$, $\lambda_2/h=0.01$; (c) $\lambda_1/h=1$, $\lambda_2/h=0.05$; and (d) $\lambda_1/h=10$, $\lambda_2/h=0.05$.

It is also important to note that because in the case of $T<T_c$ and $h<h_c$ the system is in a ''frustrated'' state, the mean-field force is present as well. The mean-field force is a consequence of a difference between mean-field free energy densities of the bulk reference (i.e., Sm-C^*) and the structure between plates (i.e., Sm-A^*). The mean-field force is equal to

$$\mathcal{F}_{MF}=(f_C-f_A)S=-\frac{1}{4}\frac{\alpha^2}{b}(T_c-T)^2S, \qquad (18)$$

where f_C and f_A are the free energy densities of the Sm-C^* and Sm-A^* phases.

To get an impression of the magnitudes of the pseudo-Casimir and mean-field force, we use a reasonable set of material constants: $\alpha=4\times10^4$ N/m^2 K, $b=8\times10^5$ N/m^2, $K_3=3\times10^{-12}$ N, $T_c=368$ K, and $K_3/K=0.1$ [18,25,26]. We take the thickness of the cell to be $h=20$ nm and consider the limiting case of infinitely strong anchoring. In this case the system can be supercooled down to $T\approx T_c-1.8$ K. The pseudo-Casimir force obviously dominates over the mean-field force at very small T_c-T, however, its amplitude in this region is also rather small [$\mathcal{F}_{Cas}/S(T=T_c)\approx-5$ pN/μm^2]. It is more instructive to estimate the magnitudes near the minimum of the pseudo-Casimir force (Fig. 3). Its amplitude is here about 50 pN/μm^2, which amounts to approximately 10% of the mean-field force. The pseudo-Casimir force could become more important near the structural transition where it diverges. But this divergence is weak. We estimate that when approaching the transition to approximately 0.01 K, the amplitude of the pseudo-Casimir interaction can reach about 200 pN/μm^2, which is still less than 20% of the mean-field interaction. In a more realistic case of finite anchoring strengths, the amplitude of the pseudo-Casimir interaction is reduced, as can be seen in Fig.

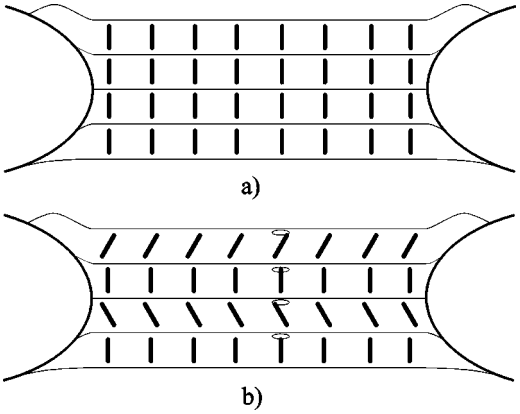

FIG. 4. Schematic presentation of a free-standing smectic film: (a) Sm-A^*, (b) Sm-C^*. (The period of the indicated helical modulation in the Sm-C^* film is not plotted to scale.)

3. However, this does not necessarily mean that its ratio to the mean-field force is also smaller. In any case, one could extract the fluctuation-induced interaction from the experimental data, provided that the interaction be measured at several temperatures, and taking into account that the mean-field interaction is thickness independent.

IV. FREE-STANDING SMECTIC FILMS

A free-standing film is formed by drawing a small amount of smectic material over a hole in a metal or glass plate (Fig. 4). The geometry of this system is identical as in the case of the homeotropic cell. The smectic layers are oriented parallel to the free surface. The system again possesses the translational invariance in x-y plane, which enables us to Fourier transform fluctuating fields as shown in the case of the homeotropic cell, thus diagonalizing the Hamiltonian. However, there are now no plates imposing boundary conditions. In our model we will assume that there exists some internal anchoring, which favors the surface ordering of the smectic to be the same as in the bulk system. This means that the preferred value of the order parameter at the free surfaces of a smectic film is equivalent to the equilibrium value inside the film. The mean-field structure of the film is therefore homogeneous. The anchoring is the same at both free surfaces. We will consider two cases: a free-standing Sm-A^* film ($T>T_c$) and a free-standing Sm-C^* film ($T<T_c$).

A. Free-standing Sm-A^* film

The evaluation of the pseudo-Casimir force in the Sm-A^* film is identical as in the above discussed case of the homeotropic cell. The preferential orientation of molecules at the surface is perpendicular to the smectic layers. The surface free energy density is equal to

$$f_S=\frac{1}{2}W\boldsymbol{\xi}^2[\,\delta(z)+\delta(z-h)\,]. \tag{19}$$

The anchoring at both surfaces is characterized by the extrapolation length $\lambda=K_3/W$. The pseudo-Casimir force is then equal to Eq. (13) inserting $\lambda_1=\lambda_2=\lambda$. As mentioned, this result is analogous to the pseudo-Casimir interaction induced by massive fluctuation modes in nematics. Its dependence on the anchoring strength λ was analyzed in Ref. [8].

B. Free-standing Sm-C^* film

In this case the free energy of fluctuations is calculated using the Hamiltonian of the Sm-C^* phase [Eq. (3)]. The boundary conditions are assumed to coincide with the bulk structure. The surface free energy density reads

$$f_S=\left[\frac{1}{2}W_{\parallel}(\xi_{\parallel}-\xi_{\parallel 0})^2+\frac{1}{2}W_{\perp}\xi_{\perp}^2\right][\,\delta(z)+\delta(z-h)\,]. \tag{20}$$

Here we allow for different anchoring strengths for each type of fluctuations. Performing the Fourier transformation $\delta\xi_i(\mathbf{r})=\Sigma_{\mathbf{q}}\exp[i(q_x x+q_y y)]\tilde{\xi}_i(\mathbf{q},z)$, we obtain $H=\Sigma_{\mathbf{q}}(H_{\mathbf{q}}[\tilde{\xi}_{\parallel}]+H_{\mathbf{q}}[\tilde{\xi}_{\perp}])$, where

$$H_{\mathbf{q}}[\tilde{\xi}_i]=\frac{1}{2}K_3 S\left(\int_0^h\left[\begin{Bmatrix}\rho^{-2}+\dfrac{K}{K_3}q^2\\ \dfrac{K}{K_3}q^2\end{Bmatrix}\tilde{\xi}_i^2+\left(\frac{d\tilde{\xi}_i}{dz}\right)^2\right]dz\right.$$
$$\left.+\begin{Bmatrix}\lambda_{\parallel}^{-1}\\ \lambda_{\perp}^{-1}\end{Bmatrix}(\tilde{\xi}_i^{-2}+\tilde{\xi}_i^{+2})\right). \tag{21}$$

The upper line corresponds to $\tilde{\xi}_{\parallel}$ and the lower to $\tilde{\xi}_{\perp}$. We again introduced extrapolation lengths $\lambda_{\parallel}=K_3/W_{\parallel}$ and $\lambda_{\perp}=K_3/W_{\perp}$. The pseudo-Casimir force is calculated following the procedure described in the case of the homeotropic cell. It consists of two terms:

$$\mathcal{F}_{Cas}=-\frac{k_B T S}{2\pi}\frac{K_3}{K}\left[\int_{1/\rho}^{\infty}\frac{r^2dr}{\dfrac{(r+\lambda_{\parallel}^{-1})^2}{(r-\lambda_{\parallel}^{-1})^2}\exp(2rh)-1}\right.$$
$$\left.+\int_0^{\infty}\frac{r^2dr}{\dfrac{(r+\lambda_{\perp}^{-1})^2}{(r-\lambda_{\perp}^{-1})^2}\exp(2rh)-1}\right]. \tag{22}$$

The first one is typical for the pseudo-Casimir interaction induced by massive fluctuations characterized by finite correlation length (ρ), and the second one for the pseudo-Casimir interaction induced by massless fluctuations with infinite correlation length. Their dependence on the thickness of the film h has been already commented in Sec. III.

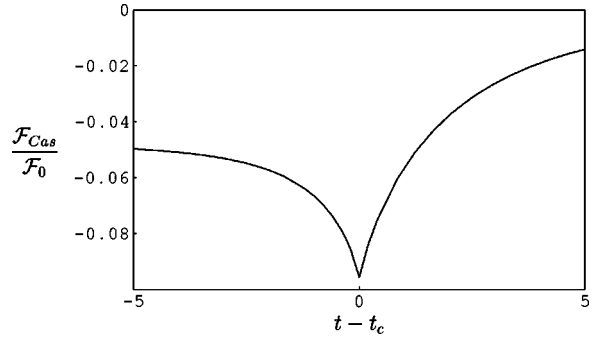

FIG. 5. Temperature dependence of the pseudo-Casimir force in a free-standing smectic film. Again the unitless temperature $t = \alpha h^2 T/K_3$ is introduced. The amplitude of the force is given in the natural unit $\mathcal{F}_0 = k_B T S K_3 / K h^3$ (a weak temperature dependence of $\mathcal{F}_0$ is neglected). Presented is the limiting case of infinitely strong anchoring; $\lambda = 0$ in the Sm-A^* film and $\lambda_\parallel = 0$, $\lambda_\perp = 0$ in the Sm-C^* film.

The temperature dependence of the pseudo-Casimir force in the free-standing smectic film is shown in Fig. 5. We present the force only for the limiting case of infinitely strong anchoring; that is, $\lambda = 0$ in the Sm-A^* film and $\lambda_\parallel = 0$, $\lambda_\perp = 0$ in the Sm-C^* film. Due to the symmetric boundary conditions, the pseudo-Casimir force in the free-standing smectic film is always attractive. It reaches the maximum at the structural transition from the Sm-A^* to the Sm-C^* film ($T = T_c$). Lowering or rising the temperature reduces the amplitude of the force. In the Sm-A^* film there are two degenerate massive fluctuation modes whose contributions to the pseudo-Casimir force decay rapidly while rising the temperature. In the Sm-C^* film the contribution of the massless mode is almost temperature independent, while the contribution of the massive mode again decays rapidly away from $T = T_c$. The profile of the force is therefore asymmetric. There is no divergence of the force at the structural transition from the Sm-A^* to the Sm-C^* film like in the homeotropic cell. In our model of the smectic film, no frustration is induced by the boundary conditions; and consequently, the divergence does not occur. The increase of the amplitude is a consequence of the fact that when approaching $T \to T_c$, all fluctuation modes become massless. We have already estimated in Sec. III that the amplitude of the force at $T = T_c$, for the film thickness $h = 20$ nm, is about 5 pN/μm^2. The implementation of finite anchoring strengths does not significantly alter the temperature profile of the force but merely reduces its amplitude.

V. CONCLUSION

In this study we analyzed the fluctuation-induced force in two smectic systems with simple planar geometry: homeotropic cell and free-standing smectic film. We considered the fluctuations of the orientational order. We demonstrated the universal behavior of the pseudo-Casimir force—short-range interaction induced by massive fluctuation modes and long-range interaction induced by massless fluctuation modes—already discovered in previously studied nematic systems. We discussed the effect of different finite anchoring strengths at the boundaries of the homeotropic cell on the character of the pseudo-Casimir force. We showed that in this case the pseudo-Casimir force can exhibit crossovers from the attractive to the repulsive regime (and vice versa) at small distances (compared to the correlation length of fluctuations) between plates. At large distances the character does not change, and the amplitude of the force is the largest when the anchoring at the plates is either very strong or very weak. In both studied systems we focused on the temperature dependence of the force, which is especially interesting due to the vicinity of the phase transition. In the case of the ''supercooled'' homeotropic cell, which is yet another representative of ''frustrated'' liquid crystal systems, we rediscover the logarithmic divergence of the force at the structural transition, already found in the nematic counterparts. At the transition from the Sm-A^* to the Sm-C^* free-standing film, the amplitude of the force is increased, but it does not diverge as there is no frustration in the system. Here we should again stress that our simple model does not include possible inhomogeneous equilibrium structure of a free-standing smectic film, with different surface and bulk ordering. In this case the critical behavior of the force could be substantially altered.

The pseudo-Casimir force induced by the orientational fluctuations which we studied in our paper, and the long-range interaction induced by the fluctuations of positional order, studied by Mikheev [5] and Ajdari *et al.* [3,4], do not close the discussion on the fluctuation-induced forces in smectics. A complete description should incorporate both effects, considering also the coupling between the orientational and positional order. Due to this coupling the deformation of the positional order (i.e., smectic layers) results in the modification of the equilibrium director profile [27] which complicates the situation considerably. However, in thin smectic films the fluctuations of the positional order are suppressed by the confinement [28]. Therefore we can expect that our model gives physically reasonable results. We also did not take into account that in chiral smectics, as a consequence of orientational fluctuations, the polarization in the system is not homogeneous. This results in the appearance of space charge and leads to the Coulomb interaction in the system [29–34]. We are not able to establish the importance of this effect in our systems at present. However, as this interaction is especially prominent in systems with a large value of spontaneous polarization, it is reasonable to assume that our model works well at least for materials with a small value of spontaneous polarization. Furthermore, as the interest in the pseudo-Casimir interaction is mostly related to very thin liquid crystalline films, the use of a discrete description of smectics instead of our continuous model should be considered [19].

The experimental detection of the pseudo-Casimir interaction induced by the orientational fluctuations could be facilitated by its specific temperature dependence, related to the presence of the phase transition. This could enable to distinguish it from other interactions present in smectic systems. Especially interesting is the ''frustrated'' system where the divergence of the force could be approached by varying the temperature while keeping constant thickness of the sample. In free-standing smectic films, the force between the

surfaces cannot be measured by the standard force-measurement devices such as the atomic force microscope or the surface force apparatus. Some information about the interaction can however be obtained by experiments that measure the intensity of light scattered by capillary waves on the surface of a free-standing film [35].

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