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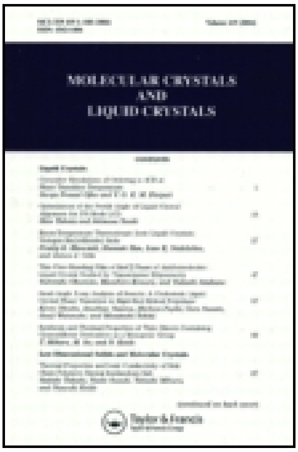

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Crossover From Under to Overdamped Dynamics in Free-Standing Smectic Films Close to the Smectic-A-Nematic Second Order Phase Transition

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Crossover From Under to Overdamped Dynamics in Free-Standing Smectic Films Close to the Smectic-A-Nematic Second Order Phase Transition

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The hydrodynamic properties of free-standing smectic films close to a second order smectic-A (SmA) — nematic (N) phase transition are investigated. By using an extended McMillan model, the profiles of the smectic elastic constants are computed at different temperatures. We show that the amplitude of smectic fluctuations has a strong dependence on the film temperature. By considering the critical behavior of viscoelastic coefficients of thin films, we show that the typical underdamped relaxation of the smectic order is replaced by an overdamped behavior close to the bulk transition temperature.

Keywords dynamic light scattering; smectic films; critical behavior; relaxation dynamics

I. Introduction

Hydrodynamic properties of liquid-cristalline membranes have attracted considerable interest over the past decade, being a subject of several theoretical and experimental investigations [1–5]. Because their common features with some biological systems, the study of smectic films has acquired a multidisciplinar character, with impacting results on different areas. Further, the possibility of drawing free-standing smectic films with few molecular layers of thickness makes them an ideal setup to investigate the interplay of surface and finite-size effects [1,6,7].

The relaxation dynamics of free-standing smectic films has been extensively investigated due to the rich phenomenology associated with the surface enhanced ordering [1,4,5]. Using the x-ray photon correlation spectroscopy technique, several works have demonstrated the existence of distinct relaxation regimes for the smectic order, depending on the wavelength of the in-plane fluctuations and the film thickness [6,8–10]. In thick films, it was observed that the relaxation dynamic occurs through a typical overdamped process which is associated with bulk-elasticity modes [9,11]. In such case, the smectic order presents a slow relaxation dynamic as a consequence of the negligible contribution of the inertial terms [6]. A different scenario has been observed in thin films due to the contribution of the inertial terms. In the limit of long wavelength fluctuations, the density-density correlation function presents an oscillatory-exponential behavior which is governed by the

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surface tension [1,8,9], with the smectic order exhibiting a fast relaxation dynamics. In all cases, the dynamical properties of freestanding smectic films have been investigated in a range of temperatures where the smectic order is well established [3,4].

Recently, several works have been devoted to the study of thermal effects on the hydrodynamic properties of free-standing smectic films close to the smectic-A-nematic phase transition [12–15]. Due to the strong surface anchoring, the elastic constants of smectic phase exhibit a non-uniform profile close to the bulk transition temperature, which reflects the heterogeneous structure presented by the order parameters profiles along the film [12]. In this scenario, smectic layer fluctuations present a strong temperature dependence in the vicinity of smectic-A-nematic transition temperature [12], as it was experimentally demonstrated by specular and diffuse x-ray scattering measurements [16]. Concerning with the dynamic properties, x-ray scattering investigations of capillarity waves on the free-surface of smectic films have demonstrated that the shear layer viscosity exhibits a diverging behavior close to the second order smectic-nematic phase transition [17]. Although some theoretical works have investigated the thermal effects on the relaxation dynamics of smectic order close to first order smectic-isotropic and smectic-nematic phase transitions [18,19], the critical behavior of viscoelastic properties of the smectic phase has not been explored so far.

In the present work, we study the hydrodynamic properties of free-standing smectic films close to the second order SmA-N phase transition. By using an extended McMillan model to compute the smectic and nematic order parameters, we properly account the profile of smectic elastic constants for a film at different temperatures. By considering the critical behavior of viscoelastic coeffcients of thin films, we show that the typical underdamped relaxation of the smectic order is replaced by an overdamped behavior close to the bulk transition temperature.

II. Theory

Free-standing smectic films can be described as a stack of smectic layers confined by a surrounding gas. The main surface ordering is represented by a surface tension which strongly reduces the smectic fluctuations close to the surface. As a result, smectic films present a quasi-long range positional order characterized by a logarithmic divergence of fluctuations in the limit of large film thickness [20]. For a thin film with N layers, the elastic deformations on the smectic order are described by

$$\mathrm{H} = \int_{a_0}^{L} d^2r \Bigg[\sum_{n=1}^{N} \frac{dK_n}{2} \left[\nabla_\perp^2 u_n\left(\vec{r}\right) \right]^2 + \sum_{n=1}^{N} \left(\frac{B_n + B_{n+1}}{4d} \right) \left[u_{n+1}\left(\vec{r}\right) - u_n\left(\vec{r}\right) \right]^2 + \frac{\gamma}{2} \left(\left| \nabla_\perp u_1\left(\vec{r}\right) \right|^2 + \left| \nabla_\perp u_N\left(\vec{r}\right) \right|^2 \right) \Bigg] \tag{1}$$

Here, $u_n(\vec{r})$ describes the displacement of the n-th smectic layer from the equilibrium position at point $\vec{r}$. d is the average layer spacing, L is the transversal size of the film and a_0 is a microscopic cutoff length of the order of molecular diameter. B_n and K_n are the compressibility and bending elastic constants of the n-th layer, respectively. The additional energy cost associated with any gradient on the area of the free surfaces is represented by the surface tension γ. In this case, the equilibrium position is determined by the holder used on the free-standing technique with the z axis being normal to the surface film [21]. A characteristic surface tension $\gamma_c = \sqrt{KB}$ delimits the regimes of strong ($\gamma > \gamma_c$) and weak ($\gamma < \gamma_c$) surface anchoring. Here, K and B are the bulk elastic constants which depend on the temperature [11]. At temperatures on which the smectic order is well established, the elastic constants present a flat profile along the whole film with $B_n = B_0$ and $K_n = K_0$ [12,14].

In order to study the relaxation dynamics of the smectic order, we consider a set of linearized motion equations for small deformations in the smectic order of the nth layer [3,10]

$$\rho \frac{\partial^2 u_n(\vec{r}, t)}{\partial t^2} = \eta_3 \nabla_\perp^2 \frac{\partial u_n(\vec{r}, t)}{\partial t} - \frac{1}{d}\left(\frac{\delta H}{\delta u_n}\right), \tag{2}$$

where ρ is the average mass density. The first term on the right side of the Eq. (2) is the viscous force, where η_3 is the layer sliding viscosity. Recent experimental works have demonstrated that η_3 presents a critical behavior close to a second order SmA-N phase transition, which is characterized by $\eta_3 = \eta_0 + \eta_1 \left|(T - T_{AN})/T_{AN}\right|^{-x}$. T is the film temperature and T_{AN} is the the bulk transition temperature. The exponent x is related to the critical exponents $\nu_{\parallel}$ and $\nu_\perp$ of the correlation lengths $\xi_{\parallel}$ and $\nu_\perp$, with $x = 3\nu_{\parallel} - 2\nu_\perp$ [17,22]. The second term in Eq. (2) represents the elastic force which can easily be derived from Eq. (1). It is important to stress that we have neglected the permeation process because it presents a large characteristic time scale [4,23]. Also, we consider that the external forces are negligible.

Taking the Fourier transform of the layer displacement written in the form

$$u_n(r, t) = L^2 \int_{2\pi/L}^{2\pi/a_0} \frac{d^2 q}{(2\pi)^2} u_n(q, t)\, e^{i\vec{q}.\vec{r}}, \tag{3}$$

one can write the set of hydrodynamical equations for free-standing smectic-A films in a compact form

$$\rho \frac{\partial^2 \tilde{u}(q, t)}{\partial t^2} + \eta_3 q^2 \frac{\partial \tilde{u}(q, t)}{\partial t} + \frac{B_0}{d^2} M \tilde{u}(q, t) = 0. \tag{4}$$

$\tilde{u}(q, t)$ is a $N \times 1$ matrix with $u_n(q, t)$ components, while M is the interaction matrix defined by

$$M_{1,1} = M_{N,N} = \left(\frac{d\gamma}{B_0}\right) q^2 + \left(\frac{K_1 d^2}{B_0}\right) q^4 + \left(\frac{B_1 + B_2}{2B_0}\right) \tag{5}$$

$$M_{n,n} = \left(\frac{K_n d^2}{B_0}\right) q^4 + \left(\frac{B_{n-1} + 2B_n + B_{n+1}}{2B_0}\right), \quad n = 2, \ldots, N-1 \tag{6}$$

$$M_{n,n+1} = M_{n+1,n} = -\left(\frac{B_n + B_{n+1}}{2B_0}\right), \quad n = 1, \ldots, N-1. \tag{7}$$

The formal solution of the Eq. (4) can be obtained from the eigenvalues λ_p and their associated eigenvectors ϕ_p of the interaction matrix

$$\tilde{u}(q, t) = \sum_{p=1}^{N} C_p(q, t)\, \phi_p(q), \tag{8}$$

where

$$C_p\left(q,t\right)=C_+^p\left(q,0\right)e^{-i\alpha_+^p t}-C_-^p\left(q,0\right)e^{-i\alpha_-^p t}. \tag{9}$$

The coeffcients $C_\pm^p\left(q,0\right)$ are determined from the initial conditions ($t=0$) of the system. By using the equipartition theorem and the condition of statistical independence, we can write the dynamic layer displacement correlation function as [3,20]

$$\langle u_m\left(q,t\right)u_n\left(-q,0\right)\rangle=\sum_{p=1}^{N}\frac{k_BT}{\lambda_p}\phi_m^{(p)}\phi_n^{(p)}\times\frac{\alpha_-^p e^{-i\alpha_+^p t}-\alpha_+^p e^{-i\alpha_-^p t}}{\alpha_-^p-\alpha_+^p}. \tag{10}$$

Here, $\alpha_\pm^p$ are the eigenfrequencies of the p-th mode which are given by

$$i\alpha_\pm^p=\left(\frac{\eta_3q^2}{2\rho}\right)\left[1\mp\sqrt{1-\frac{4\lambda_p\rho B_0}{d^2\eta_3^2q^4}}\,\right]. \tag{11}$$

Depending on the ratio $4\lambda_p\rho B_0/d^2\eta_3^2q^4$, the dynamic correlation function may present two distinct behaviors. For $4\lambda_p\rho B_0/d^2\eta_3^2q^4>1$, the p-th mode relaxes through an oscillatory exponential process, where the damping rate Γ_p is given by the real part of Eq. (11) while the imaginary part corresponds to the oscillation frequency ω_p. On the order hand, a purely exponential relaxation is observed for the pth mode when $4\lambda_p\rho B_0/d^2\eta_3^2q^4<1$. Previous theoretical and experimental works have demonstrated that the observation of these relaxation regimes depends on the film thickness l as well as the in-plane wave vector q [4,8,9]. However, the effects associated with the temperature dependence of the viscoelastic coefficients on the smectic relaxation dynamics has not been investigated so far.

A. Structure Factor

From the dynamic correlation function of the smectic layer displacement, we can write the structure factor $S(t)$ of free-standing films. Assuming that the layer fluctuations are Gaussian, it is straightforward to show that the time dependent part of the structure factor is given by [3,6,23]

$$S(t)=g(q,q_z)\sum_{n,m=1}^{N}C_{n,m}(q_z)\left\langle u_n\left(q,t\right)u_m\left(-q,0\right)\right\rangle, \tag{12}$$

where

$$g\left(q,q_z\right)=\frac{q_z^2q^2L}{2\pi}\int_{\frac{2\pi}{L}}^{\frac{2\pi}{a_0}}\frac{dq}{q'\left(q^2-q'^2\right)}\left[q\,J_0\left(q'L\right)J_1\left(qL\right)-q'J_0\left(qL\right)J_1\left(q'L\right)\right] \tag{13}$$

and

$$C_{n.m}(q_z)=2\pi\rho_s\left|\rho_M\left(q_z\right)\right|^2e^{-iq_z(m-n)d}\times\exp\left(-\frac{q_z^2}{2}\left[\left\langle u_n^2\left(0,0\right)\right\rangle+\left\langle u_m^2\left(0,0\right)\right\rangle\right]\right). \tag{14}$$

Here, $q_z=2\pi/d$, ρ_s is the surface molecular density and ρ_M is the linear electron density. $J_n\left(x\right)$ is the n-th order Bessel function. As we can see from Eq. (12), the time dependence of the structure factor is determined by the temporal correlations in the smectic order. The discrete description of the structure factor of smectic films is expected to be equivalent to the continuum theoretical models used to describe the dynamic x-ray scattering measurements in free-standing films [4,8,9]. However, the discrete model allows to easily explore the spatial variability of the viscoelastic coefficients due to the boundary conditions. In spite of the fact that a great variety of phenomena associated with surface effects have motivated several investigations about the static properties of free-standing films, there are just a few works about the surface effects on dynamical properties of these systems.

In order to analyze how the critical behavior of the viscoelastic coefficients affects the relaxation dynamics of the smectic order, we use the discrete extension of the McMillan mean-field model to properly account the orientational s_i and translational σ_i order parameters of each smectic layer [24]. From this model, the one-particle effective potential within a smectic layer is given by

$$\begin{aligned} V_1\left(z_1,\theta_1\right) &= -\frac{V_0}{3}\Big[s_1+s_2+3W_0/V_0 \\ &\quad +\alpha\cos\left(2\pi z_1/d\right)\left(\sigma_1+\sigma_2\right)\Big]P_2\left(\cos\theta_1\right) \end{aligned} \tag{15}$$

$$V_i\left(z_i,\theta_i\right) = -\frac{V_0}{3}\left[\sum_{i-1}^{i+1} s_i+\alpha\cos\left(2\pi z_i/d\right)\left(\sum_{i-1}^{i+1}\sigma_i\right)\right]P_2\left(\cos\theta_i\right) \tag{16}$$

$$\begin{aligned} V_N\left(z_N,\theta_N\right) &= -\frac{V_0}{3}\Big[s_N+s_{N-1}+3W_0/V_0 \\ &\quad +\alpha\cos\left(2\pi z_N/d\right)\left(\sigma_N+\sigma_{N-1}\right)\Big]P_2\left(\cos\theta_N\right) \end{aligned} \tag{17}$$

Here, $P_2\left(\cos\theta_i\right)$ is the second order Legendre polynomial with θ_i being the angle in the i-th layer between the long axis of the molecule and the z direction. V_0 is a parameter of the microscopic model [25] that determines the scale of the nematic-isotropic transition temperature, and k_B is the Boltzmann's constant. Another parameter of the model is α, that according to the McMillan theory is related to the length of the alkyl tails of calamitic molecules of smectogenic compounds. In the extended mean-field model for finite films [24], the strength of surface anchoring is determined by the parameter W_0. This parameter couples with the orientational order parameter s and represents the surface-induced homeotropic alignment in the film. In free-standing smectic-A films, the absence of a substrate gives rise to an almost perfect homeotropic alignment at the film surface [1].

For the i-th layer, we will assume that the bending elastic constant K_i is proportional to s_i^2 and the compressibility elastic constant B_i is proportional to σ_i^2 [11,26]. For thick films ($N \to \infty$), the local order parameters should have values predicted by the McMillan theory [25] for the bulk SmA phase. At the temperature T_0 well below the bulk SmA—N transition temperature, we can assume that the bulk order parameters are s_0 and σ_0, corresponding to elastic constants $K\left(T_0\right)=K_0$ and $B\left(T_0\right)=B_0$. From the values of B_0 and K_0 at the reference temperature T_0, we can obtain the elastic constant profiles at a temperature T by using the following expressions:

$$K_i\left(T\right)=K_0\left[\frac{s_i\left(T\right)}{s_0}\right]^2 \tag{18}$$

$$B_i\,(T) = B_0 \left[\frac{\sigma_i\,(T)}{\sigma_0} \right]^2 \tag{19}$$

The elastic constant profiles may have distinct behaviors near the first and second order phase transitions depending on the profiles of the order parameters. The order of the bulk phase transition is determined by the α parameter. McMillan theory predicts that, for $\alpha < 0.70$, the bulk SmA—N transition is continuous while for $0.70 < \alpha < 0.98$ this transition is a first order one. For values of α above 0.98, a first order SmA—I phase transition takes place. We will consider values for α that correspond to SmA—N transitions. In particular, we will use $\alpha = 0.60$ that corresponds to a bulk second order transition temperature, with $T_{AN} = 0.177261\ V_0/k_B$. We performed our calculations using $W_0 = 3V_0$ that corresponds to a strong homeotropic anchoring at the boundary free surfaces of the film.

III. Results

In Fig. 1 we present the phase diagram of the central layer of a free-standing smectic film, with $N = 41$. We consider the regime of strong surface anchoring, with $W_0/V_0 = 3$. Here, one can notice that the sequence of phases is similar to that predicted by McMillan for bulk smectogenic systems. For $\alpha < 0.80$, the system exhibits a second order SmA-N phase transition (solid line) which corresponds to a continuous reduction of the smectic order while the nematic phase stays finite. For $0.80 < \alpha < 0.95$, the smectic order presents a discontinuous reduction to non-null values at the transition temperature, corresponding to a first order phase transition from the smectic-A phase to a surface-induced smectic A phase (si-SmA) [27], as shown in the inset. In particular, the tricritical point of the McMillan phase diagram is split into a critical point and a critical end point. In this case, the end of the coexistence line between the smectic and surface-induced smectic defines the critical point. In addition, the critical end point represents the position at which the line of a continuous transition between the surface-induced smectic and surface-induced nematic

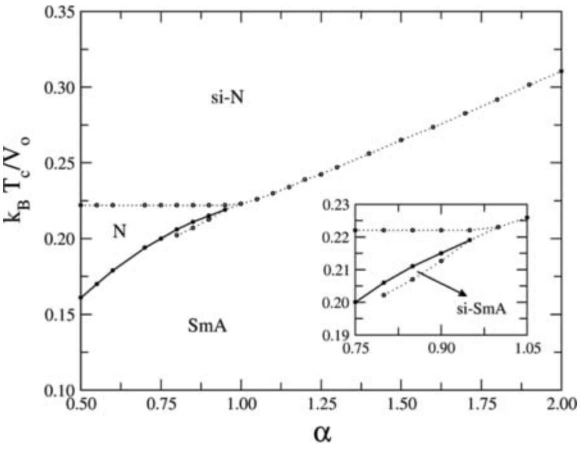

Figure 1. The phase diagram in the T_c *vs* α parameter space for the film thickness $N = 41$. Here, one can notice that inner layers of the film present a phase diagram similar to that of bulk systems. However, the tricritical point splits into a critical point and a critical end point, as shown in the inset.

phases encounters the coexistence line of the smectic and the nematic and surface-induced smectic phases. For $0.95 < \alpha < 1.00$, a first order SmA-N phase transition is observed, while a first order smectic-A-isotropic phase transition takes place for $\alpha > 1.00$.

In order to study the hydrodynamic properties of freestanding smectic films close to the second order SmA-N phase transition, we restrict our attention to the case of $\alpha = 0.60$, with $W_0/V_0 = 3$. For this value of the parameter α, the bulk transition temperature is $T_{AN} = 0.177261\ V_0/k_B$. In Fig. 2, it is exhibited the profile of the elastic constants of a 41-layer free-standing film, at distinct temperatures. With exception of the outermost layers, the bending elastic constant presents a flat profile even close to the transition temperature, as shown in Fig. 2(a). Such a behavior reflects the temperature dependence of the orientational order parameter, which stays finite close to the second order SmA-N phase transition. A different scenario is observed for the compression elastic constant, as presented in Fig. 2(b). At temperatures well below to the bulk transition temperature, we notice that the compression elastic constant presents a flat profile due to the fact that the translational order parameter is well established far from the transition. As the temperature becomes closer to the bulk transition temperature, a pronounced reduction in the compression elastic constant is observed, which develops a non-uniform profile.

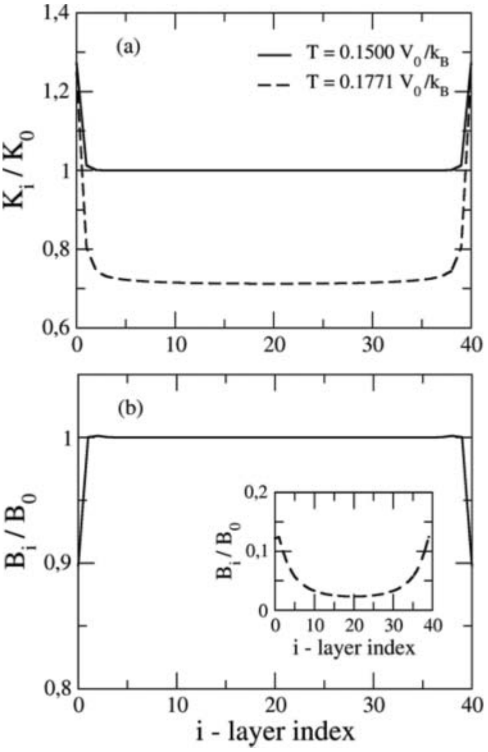

Figure 2. Normalized elastic constants versus layer index for films at distinct temperatures: (a) bending elastic constant and (b) compressibility elastic constant. The parameters of the microscopic model are $W_0/V_0 = 3$ and $\alpha = 0.60$, with a bulk transition temperature $T_{AN} = 0.177261\ V_0/k_B$. Notice that the compressibility elastic constant presents a strong reduction as the film temperature becomes close to T_{AN} (inset).

The temperature dependence of the elastic constants are expected to play an important role in the hydrodynamic properties of free-standing smectic films. In what follows, we analyze the amplitude of the smectic fluctuations and the relaxation dynamic of the smectic order for films with elastic constants presenting the profiles obtained in Fig. 2. In particular, the amplitude of the smectic fluctuations of the *n*th layer is defined by [20]

$$\Delta_i^2 = \left\langle u_i^2\left(0,0\right)\right\rangle = \frac{k_B T d}{B_0}\int_{\frac{2\pi}{L}}^{\frac{2\pi}{a_0}} \frac{d^2 q}{\left(2\pi\right)^2}\left(M^{-1}\right)_{i,i} \quad , \tag{20}$$

where M^{-1} is the inverse of the interaction matrix. In Fig. 3, it is shown the profile of the smectic fluctuations of a 41-layer film at distinct temperatures (only half of the symmetric profile is depicted). We considered the typical values for bulk elastic constants $K_0 = 10^{-6}$ dyn and $B_0 = 2.5 \times 10^7$dyn/cm^2, with $\gamma = 20$ dyn/cm, $L = 4$ mm, $a_0 = 4\,\mathring{A}$, $d = 30\,\mathring{A}$, and $V_0 = 2.2 \times 10^{-13}$ erg. For a temperature ($T_{AN} = 0.1500 V_0/k_B$) well below the bulk transition temperature ($T_{AN} = 0.177261 V_0/k_B$), we observe that the smectic fluctuations present a profile with a negative concavity, corresponding to the regime of strong surface tension ($\gamma > \gamma_c$). In fact, the surface tension reduces the smectic fluctuation at the film surfaces, suppressing the effects associated with free boundary conditions. As the film temperature becomes closer to the bulk transition temperature ($T_{AN} = 0.1771 V_0/k_B$), we notice that the negative concavity of the fluctuation profiles persists, with a pronounced enhancement of the amplitude of fluctuations for the inner layers. Such a behavior is mainly associated with the reduction of the compression elastic constant close to second order SmA-N phase transition. Indeed, the surface ordering induced by the surface tension is just partially transmitted to the inner layers as the compression elastic constant decreases. As a consequence, the film behaves in a similar way as in the regime of weak surface tension ($\gamma < \gamma_c$). Such a behavior is in good agreement with the fluctuation profile measured in free standing smectic film of the 7*AB* (4,4′-diheptylazoxybenzene) compound [16].

Now, we analyze the relaxation dynamics of the smectic order close to the second order SmA-N phase transition. In Fig. 4, we show the time evolution of function $G_2(t) = [S(t)/S(0)]^2$ for a 41-layer film at distinct temperatures. We used the same parameters

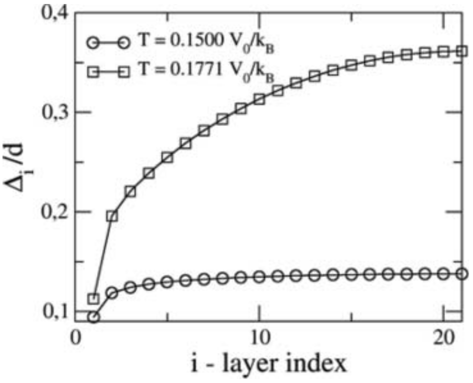

Figure 3. Profile of the smectic fluctuations for a 41-layer film at distinct temperature. The bulk and surface constants were kept equal to experimental typical values. Notice that the pronounced enhancement in the amplitude of smectic fluctuation on the inner layers as the film temperature becomes closer to the bulk transition temperature.

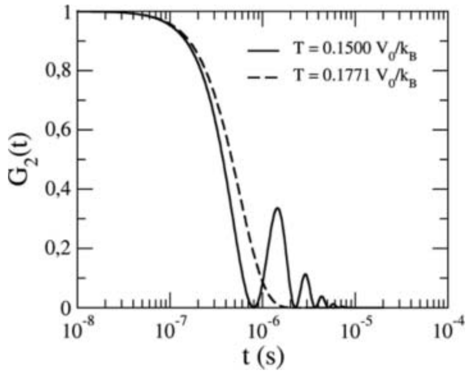

Figure 4. Time evolution of the structure factor for a 41-layers free-standing film at different temperatures. The in plane wavevector is $q = 1000\ cm^{-1}$. Notice that an overdamped relaxation takes place as the film temperature becomes closer to the bulk transition temperature.

used in Fig. 3, with the in-plane wavevector $q = 10^3\ cm^{-1}$. We considered the temperature dependence of the layer sliding viscosity as given by $\eta_3 = \eta_0 + \eta_1 \left|(T - T_{AN})/T_{AN}\right|^{-x}$, with $\eta_0 = 0.5\ g/(cm\,.\,s)$, $\eta_1 = 0.1\ g/(cm\,.\,s)$, and $x = 1\big/2$. For a temperature well below the bulk transition temperature ($T_{AN} = 0.1500V_0/k_B$), we notice that the function $G_2(t)$

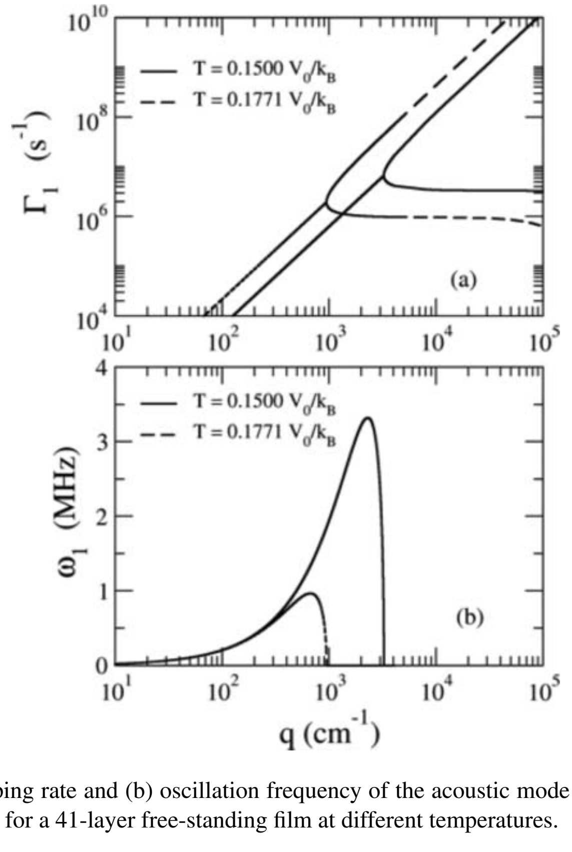

Figure 5. (a) Damping rate and (b) oscillation frequency of the acoustic mode as a function of the in-plane wavevector for a 41-layer free-standing film at different temperatures.

exhibits an underdamped dynamics, with an oscillatory-exponential decay. In this case, the smectic order presents a relaxation dynamics governed by the surface tension [1]. As the temperature becomes closer to the bulk transition temperature ($T_{AN} = 0.1771 V_0/k_B$), we noticed that the smectic order presents an overdamped dynamics, with a simple exponential decay. This result can be understood by considering the acoustic relaxation mode of the smectic order, corresponding to the first term of the sum in Eq. (10). In fact, earlier works have demonstrated that the time evolution of the structure factor is determined by the lowest eigenvalue ($p = 1$) of the interaction matrix which governs the dynamic correlation functions $\langle u_m(q, t)\, u_n(-q, 0)\rangle$ [3]. As the temperature becomes closer to the bulk transition temperature, the range of non-null oscillation frequency ω_1 of the acoustic mode is shifted towards lower wavevectors, as shown in Fig. 5. This shift can be directly associated with the critical divergence of the layer sliding viscosity, which modifies the critical wavevector $q_c = \left(4\lambda_1 \rho B_0/d^2\eta_3^2\right)^{1/4}$ at which a non-null oscillation frequency for the acoustic mode takes place.

IV. Summary and Conclusions

In summary, we studied the hydrodynamic properties of free-standing smectic films close to a second order SmA-N phase transition. By using an extended McMillan model to compute the orientational and translational order parameters, we determined the profile of the elastic constants for films at different temperatures. Close to the transition temperature, we observed a pronounced reduction in the compressibility elastic constant, which presents a non-uniform profile. As a consequence, a significant enhancement in the layer fluctuations was observed due to the partial transmission of the surface ordering induced by the strong surface tension. Concerning the relaxation dynamics, our results showed that an overdamped relaxation takes place due to the shift in the range of wavevectors at which a non-null oscillation frequency for the acoustic mode takes place. Such a crossover in the relaxation dynamics of the smectic order shows that the critical behavior of the viscoelastic constants plays an important role in the hydrodynamic properties of free-standing films.

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