Fluctuation-Induced Interfacial Forces in Ordered Films

In a recent Letter, Ajdari, Peliti, and Prost [1] broached the question of long-range boundary interactions mediated by massless excitations in ordered phases, specifically, liquid crystals. A similar program has, in fact, been undertaken a few years previously [2-4]. Three comments are in order.

(a) The authors of [1] have evidently overlooked an earlier paper [4], where one of the physical effects they address, namely, the interaction between the planar boundaries of a homotropic smectic film of thickness h, was explicitly calculated for *arbitrary* values of the interfacial tensions $c^{\pm}$. The result [see Eq. (15) of [4]] in the notation of [1] is

$$\delta \mathcal{F}_s = \frac{1}{16\pi} \text{Li}_2 \left[\frac{c^+ - (\kappa_1 B)^{1/2}}{c^+ + (\kappa_1 B)^{1/2}} \frac{c^- - (\kappa_1 B)^{1/2}}{c^- + (\kappa_1 B)^{1/2}} \right]$$
$$\times \left(\frac{B}{\kappa_1} \right)^{1/2} \frac{kT}{h}, \qquad (1)$$

where the Franck constant is denoted by κ_1, and the bulk modulus by B; the tension of an interface with a solid substrate is to be taken as infinite here. The dilogarithm is defined by $\text{Li}_2(x) = \sum_{n=1}^{\infty} x^n n^{-2}$, so that $\text{Li}_2(1) = \zeta(2)$. In the limit $c^{\pm} \gg (\kappa_1 B)^{1/2}$ this expression approaches Eq. (3) of [1] [see also Eq. (16) of [4]].

(b) It is important to note, however, that this result for $\delta \mathcal{F}_s$ is valid only at temperatures below the *roughening transition* temperatures T_R^+ and T_R^- of both the smectic interfaces. The specifics of such a transition at a smectic interface were also discussed in [4]. Here the consequences are reported. Above the roughening temperature T_R^+ the smectic fluctuations of the longest wavelengths, which determine the asymptotic form (1), become decoupled from the interface location; the surface tension c^+ in (1) and in the boundary Hamiltonian (15) of [1] should then be replaced by the effective *surface stiffness* $\tilde{c}^+$ of the smectic layers. The qualitative behavior of $\tilde{c}^+$ is quite analogous to that of the capillary stiffness which was discussed in [4]. At the roughening transition $\tilde{c}^+$ jumps from the value c^+ in the low-temperature phase to a smaller value related to the universal value of stiffness at the Kosterlitz-Thouless transition: cf. Eq. (12) of [4]. In the high-temperature phase $\tilde{c}^+(T)$ exhibits a $(T - T_R^+)^{1/2}$ singularity near T_R^+, while for $T \gg T_R^+$ it approaches a value equal to the anchoring energy of the homotropic orientation. An interface with a solid substrate is still reproduced in the $c^+ = \infty$ limit.

(c) The corresponding contribution of phonon modes to the interaction between boundaries of a simple crystal film is not "obviously negligible," as stated in [1]. While the coupling between the longitudinal modes and one set

of the transverse modes in a film makes an application of the continuum methods of [1,2,4] rather difficult, an explicit result, namely,

$$\delta \mathcal{F}_s \simeq -0.347 kT/h^2, \qquad (2)$$

can be obtained for a microscopic model of a simple atomic crystalline film with two free boundaries [2]. The harmonic forces between atoms in the model are central, so that the ratio of longitudinal and transverse sound velocities is fixed at $\sqrt{3}$ [5]. In general, the numerical constant in (2) should depend on dimensionless ratios of the elastic constants of the film and the substrate(s); in particular, we expect that the attractive interaction (2) is changed to a repulsive form when one of the free boundaries is replaced by an interface between a crystal and a rigid solid substrate. In any case, the amplitude of this effect should exceed that induced by typical van der Waals interactions already at room temperatures. Furthermore, these two sources of h^{-2} terms in $\delta \mathcal{F}_s(h)$ can be separated since their amplitudes depend quite differently on temperature: while the phonon effect (2) is determined by thermal excitations and, thus, is directly proportional to the thermal energy kT, the van der Waals (photon) contribution is dominated in most cases by the zero-point fluctuations and does not vary significantly with temperature.

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[2] A. A. Chernov and L. V. Mikheev, Poverkhnost, No. 9, 37 (1987) [in Russian]; here we present an improved numerical value of the prefactor.

[3] A. A. Chernov and L. V. Mikheev, Dokl. Acad. Nauk SSSR **297**, 349 (1987) [Sov. Phys. Dokl. **32**, 906 (1987)]. This paper contains the notion that fluctuations of the director in nematic liquid crystals are similar to phonons in producing interfacial interactions $\propto kT/h^2$, but explicit results were not published.

[4] L. V. Mikheev, Zh. Eksp. Teor. Fiz. **96**, 632 (1989) [Sov. Phys. JETP **69**, 358 (1989)].

[5] A. A. Maradudin, E. W. Montroll, and G. H. Weiss, *Theory of Lattice Dynamics in the Harmonic Approximation* (Academic, New York, 1971).