Fluctuation-Induced Forces between Rough Surfaces

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External boundaries change the fluctuations of a correlated fluid, such as a liquid crystal, thereby causing long-range interactions. We compute how the interaction between two parallel plates is modified by the deformations of one plate. Corrections to the leading interaction decay with the average separation of the plates through an exponent related to the roughness of the boundary. These interactions may also modify the surface fluctuations of thin liquid films.

External bodies (point particles, polymers, membranes, etc.) immersed in a fluid modify its fluctuations by imposing boundary conditions. Variations in the fluctuation free energy in turn induce interactions between the external bodies. Examples of these phenomena are the van der Waals interaction between particles, and the Casimir force between two plates [1], both due to *quantum fluctuations* of the electromagnetic field. For bodies immersed in a classical fluid, similar forces arise from reduced *thermal fluctuations*. If the fluid has short-range correlations, the resulting forces are also short ranged, while for a fluid with long-range correlations (e.g., binary mixtures close to a critical point, superfluids, or liquid crystals [2]), they are long ranged with universal characteristics. We shall call all such fluctuation-induced interactions Casimir forces. Thermal Casimir effects in critical systems are closely related to finite-size corrections to the free energy [3]. They have also been discussed in several interesting contexts, such as wetting close to a tricritical point [4], unbinding of fluid membranes in liquid crystals [2,5], and the elongation of surface domains in epitaxial growth [6].

Most computations of Casimir forces are for simple geometries, e.g., between two parallel plates. It is natural to consider how these forces are modified by the roughness that is present in most "random" surfaces. The interactions can in turn alter the thermal fluctuations of a fluid surface, or a liquid-crystal film. There are several approaches to calculating Casimir interactions [1,7]. In principle, a multiple-scattering approach [7] gives the interactions for arbitrary geometry in a perturbation series. Since we are only interested in relatively small deformations around "flat" geometries, we introduce a new method of directly integrating the thermal fluctuations. The method is quite generally applicable to many manifolds with arbitrary intrinsic, D, and embedding, d, dimensions.

Here we present results for interactions between two plates with average separation H, one of which is deformed from a flat geometry. We show that if the roughness of the surface is self-similar [8], and characterized by a roughness exponent ζ_S, the leading $1/H^2$ Casimir interaction has a correction that decays as $1/H^{4-2\zeta_S}$. We

estimate the magnitude of this correction using suitable parameters and find that it may be detectable by current force apparatus [9]. This provides a possibility of determining ζ_S by measuring the force. The Casimir interactions also modify the fluctuations of a fluid/air interface: For "like" boundaries, as in the case of a free-standing liquid-crystal film, the interface deformations are enhanced to the lowest order, while for "unlike" boundaries, e.g., a smectic liquid-crystal film on a solid substrate, they are suppressed.

We first consider a fluid with fluctuations described by a one-component, isotropic field $\phi(\mathbf{r})$, subject to a simple quadratic Hamiltonian,

$$\mathcal{H}_0[\phi] = \int d^d r \, \tfrac{1}{2} K [\nabla \phi(\mathbf{r})]^2 . \tag{1}$$

This is a correct description for the Goldstone modes of a superfluid. Fluctuations of a liquid crystal involve more components (for a nematic), and are anisotropic [2], while those of a fluid at a critical point require a nonquadratic action. These generalizations will be discussed later on. We distinguish between two types of couplings between the external bodies and the fluid: The field fluctuations may be suppressed (type I), e.g., by strong anchoring for liquid crystals, and by substrates that prefer one of the coexisting fluids in a critical mixture (a magnetic field in the spin analogy). Another possibility occurs at open boundaries (type II) of the fluid, and corresponds to the suppression of the normal gradient of ϕ. It is known that Casimir forces have universal amplitudes which depend only on the universality class and the type of the boundary conditions; the strength of the boundary couplings is irrelevant [10–12]. Anticipating such universality, we implement the two types of boundary effects by requiring either the field ϕ (type I) or its normal derivative $\partial_\perp \phi$ (type II) to vanish on the surface of the external bodies.

Consider n manifolds embedded in a fluid, each described by its coordinates $\mathbf{r}_a(x_a)$. Here x_a is a D_a-dimensional internal coordinate for the manifold ($D_a = 1$ for a polymer and $D_a = 2$ for a membrane), and $\mathbf{r}_a$ indicates a position in the d-dimensional fluid. The fluctuation-induced interactions between the manifolds are

obtained by integrating over all configurations of the field ϕ, with the constraints imposed by the external manifolds. Type-I boundary conditions correspond to the constraints $\phi(r_\alpha(x_\alpha)) = 0$, for $\alpha = 1, 2, \ldots n$, which can be imposed by inserting delta functions. Using the integral representation of the delta function, we obtain

$$\exp\left[-\frac{\mathcal{H}_{\text{eff}}[r_\alpha(x_\alpha)]}{kT}\right] = \frac{1}{Z_0}\int \mathcal{D}\phi(\mathbf{r})\prod_{\alpha=1}^{n}\mathcal{D}\psi_\alpha(x_\alpha)\exp\left[-\mathcal{H}_0[\phi] + i\int dx_\alpha\,\psi_\alpha(x_\alpha)\phi(r_\alpha(x_\alpha))\right],\tag{2}$$

where Z_0 is the partition function for the unperturbed fluid, and $\psi_\alpha(x_\alpha)$ are the auxiliary fields defined on the n manifolds, acting as sources coupled to ϕ. After integrating over the field ϕ, we obtain the long-range interactions between the sources as

$$\exp\left[-\frac{\mathcal{H}_{\text{eff}}}{kT}\right] = \int \prod_{\alpha=1}^{n}\mathcal{D}\psi_\alpha(x_\alpha)\exp\{-\mathcal{H}_1[\psi_\alpha(x_\alpha)]\}.\tag{3}$$

The action $\mathcal{H}_1[\psi_\alpha(x_\alpha)]$ for the n-component field $\Psi \equiv (\psi_1, \psi_2, \ldots, \psi_n)$ is given by

$$\mathcal{H}_1[\Psi] \equiv \Psi M \Psi^T = \sum_{\alpha=1}^{n}\sum_{\beta=1}^{n}\int dx_\alpha dx_\beta\,\psi_\alpha(x_\alpha)G^d(r_\alpha(x_\alpha) - r_\beta(x_\beta))\psi_\beta(x_\beta),\tag{4}$$

where $G^d(\mathbf{r}) \equiv \langle\phi(\mathbf{r})\phi(0)\rangle_0$ is the two-point correlation function of ϕ in free space. Finally, the effective interaction between the manifolds is obtained as

$$\mathcal{H}_{\text{eff}}[r_\alpha(x_\alpha)] = \tfrac{1}{2}kT\ln\text{Det}(M[r_\alpha(x_\alpha)]).\tag{5}$$

The matrix M is a functional of $r_\alpha(x_\alpha)$ and its determinant is in general difficult to evaluate. It is possible, however, to perturbatively calculate the corrections due to small deformations around simple geometries. As an explicit example, we computed the interaction energy between two surfaces in $d = 3$, with average separation H, and one plate deformed, i.e., $r_1(x) = (x_1, x_2, 0)$, $r_2(x) = (x_1, x_2, H + h(x))$. The result is $\mathcal{H}_{\text{eff}} = \mathcal{H}_{\text{flat}} + \mathcal{H}_{\text{corr}}$, where

$$\frac{\mathcal{H}_{\text{flat}}}{A} = kT\int\frac{d^2p}{(2\pi)^2}\ln\left[\frac{1}{2ap}\right] - \frac{\zeta(3)}{16\pi}\frac{kT}{H^2}\tag{6}$$

is the Casimir interaction per unit area of two flat plates. The first term in Eq. (6) is a contribution to the surface tension which depends on a lattice cutoff. The second term, decaying as $1/H^2$, has a universal amplitude $-\zeta(3)/16\pi \approx -0.02391$. The energy cost of the deformations is given by

$$\mathcal{H}_{\text{corr}} = -\frac{3\zeta(3)kT}{16\pi H^4}\int d^2x\,h^2(\mathbf{x}) + \frac{kT}{4}\int d^2x\,d^2y[h(\mathbf{x}) - h(\mathbf{y})]^2$$

$$\times\left\{\frac{1}{8\pi^2|\mathbf{x}-\mathbf{y}|^6} - \frac{1}{2\pi|\mathbf{x}-\mathbf{y}|^3 H^3}K_1(t) + \frac{1}{H^6}[K_1^2(t) + K_2^2(t)]\right\},\tag{7}$$

where $t \equiv |\mathbf{x}-\mathbf{y}|/H$, and the two kernel functions are defined by

$$K_1(t) \equiv \int_0^\infty du\frac{u^2}{2\pi(e^{2u}-1)}J_0(tu),$$

$$K_2(t) \equiv \int_0^\infty du\frac{u^2 e^u}{2\pi(e^{2u}-1)}J_0(tu).\tag{8}$$

There is an implicit short-distance cutoff a for the power laws in Eq. (7). The first term in Eq. (7) represents an instability to deformations related to the attraction between plates. Remarkably, this term is also obtained by replacing $1/H^2$ with $1/[H + h(\mathbf{x})]^2$ in Eq. (6) and averaging over the position $\mathbf{x}$. The second term represents long-range interactions between deformations induced by the fluctuations of the field. The first term in the curly brackets is the conformation energy of the deformed surface in the absence of the second plate, and is independent of H. The remaining terms represent corre-

lations due to the presence of the second plate. Both $K_1(t)$ and $K_2(t)$ approach a constant as $t \to 0$. As $t \to \infty$, $K_1(t) \sim 1/t^3$, and $K_2(t) \sim \exp(-bt)$, with $b \approx 3.3$. The large-t behaviors of $K_1(t)$ and $K_2(t)$ determine the long-range interactions between height fluctuations.

Equation (7) can be used to calculate the Casimir force between a flat and a quenched rough surface. Many solid surfaces produced by rapid growth or deposition processes are characterized by self-similar fluctuations [13]. The fluctuations of a self-affine surface [8] grow as

$$\overline{[h(\mathbf{x}) - h(\mathbf{y})]^2} = A_S|\mathbf{x}-\mathbf{y}|^{2\zeta_S},\tag{9}$$

where the overbar denotes quenched average, and ζ_S is a characteristic roughness exponent. Using Eq. (9) we can take the average of the H-dependent terms in $\mathcal{H}_{\text{eff}}$ to ob-

tain a free energy per unit area,

$$f(H) = -\frac{\zeta(3)}{16\pi}\frac{kT}{H^2} - \frac{3\zeta(3)}{16\pi}\frac{kTA_SL^{2\zeta_s}}{H^4} + \frac{C_1}{4}\frac{kTA_S}{H^{4-2\zeta_s}},$$

$$(10)$$

where L is the extent (upper cutoff) of the self-affine structure, satisfying $\Delta H \equiv A_S^{1/2}L^{\zeta_s} \ll H$ to avoid contact between plates. The coefficient C_1 in Eq. (10), given by

$$C_1 = \int_{a/H}^{L/H}\{-t^{2\zeta_s-2}K_1(t) + 2\pi t^{2\zeta_s+1}[K_1(t)]^2$$
$$+ 2\pi t^{2\zeta_s+1}[K_2(t)]^2\}\,dt,$$

$$(11)$$

does weakly depend on the ratio L/H, but since the functions K_1 and K_2 decay rapidly at a large distance, it is quite insensitive to L as long as $L \gg H$. For $L \gg H \gg \Delta H$, the interactions in Eq. (10) are arranged in order of decreasing strength. The largest effect of randomness is to increase the Casimir attraction by an amount proportional to $(\Delta H/H)^2$. The last term in Eq. (10) decays as $1/H^{4-2\zeta_s}$ and in principle can be used to indirectly measure the roughness exponent ζ_s. In Eq. (10), if all lengths are measured in units of an atomic scale a_0 (e.g., the diameter of a surface atom), A_S becomes dimensionless. Using a reasonable set of parameters, $\zeta_s \approx 0.35$, $a_0 \approx 5$ Å, $A_S \approx 1$, and $L \approx 300$ Å, we estimate that for surfaces of 1 mm size, and 100 Å apart, the forces generated by the three terms in Eq. (10) are 1.9×10^{-4}, 4.9×10^{-5}, and 3.7×10^{-6} N, respectively (with an appropriate lower cutoff of ~ 20 Å). The force generated by the last term is in fact measurable with current force apparatus [9] provided that one can subtract the strong background forces generated by the first two terms.

We now consider how interactions in Eqs. (6) and (7) are modified for more complex fluids. Fluctuations at a critical point cannot be described by a quadratic action, and hence it is not easy to integrate out the field ϕ in Eq. (2). Exact results in two dimensions [12] indicate that the Casimir interaction between two flat plates decays as $-c\pi kT/24H$ for like boundaries, where c is the central charge of the critical system. Repeating the computations leading to Eq. (6) in two dimensions yields $-\pi kT/24H$, corresponding to $c = 1$, as expected for a free field theory. We make the likely conjecture that for deformed surfaces, results from a quadratic action are valid, except for a similar change of amplitude. We can somewhat improve upon the magnitude of the amplitude by using the correct two-point correlation functions of the field theory for $G(\mathbf{r})$ in Eq. (4). For example, using $G(\mathbf{r}) \sim 1/r^{1/4}$ for the Ising model in $d = 2$ reduces the amplitude by a factor of 1.5, giving rise to a value much closer to the exact result ($c_{\text{Ising}} = 0.5$).

The above results can be easily generalized to anisotropic fluids. With two coupling constants $K_\perp$ and $K_\parallel$ for fluctuations with wave vectors perpendicular and parallel to the plates in Eq. (1), the amplitude of the Casimir interaction is multiplied by $(K_\perp/K_\parallel)^{d-1}$. Such anisotropy is inherent in nematic liquid crystals: The energy cost of fluctuations of the nematic director $\mathbf{n}$ are given by [14]

$$\mathcal{H}_N = \int d^3r\, \tfrac{1}{2}\,[\kappa_1(\nabla\cdot\mathbf{n})^2 + \kappa_2(\mathbf{n}\cdot\nabla\times\mathbf{n})^2 + \kappa_3(\mathbf{n}\times\nabla\times\mathbf{n})^2].$$

$$(12)$$

If the nematic director is on average perpendicular to the plates, its fluctuations parallel to the plate can be decomposed into transverse and longitudinal components. The transverse (longitudinal) component is an anisotropic field, with coupling constants κ_3 and κ_2 (κ_1) perpendicular and parallel to the plates. Adding up the two contributions, we find that the Casimir energies in Eqs. (6) are multiplied by $\kappa_3/\kappa_1 + \kappa_3/\kappa_2$. This is precisely the result obtained by Ajdari, Peliti, and Prost [2].

An extreme limit of anisotropy is exhibited by smectic liquid crystals, with a deformation energy

$$\mathcal{H}_S = \int d^dr\, \frac{1}{2}\left[B\left(\frac{\partial u}{\partial z}\right)^2 + \kappa(\nabla^2u)^2\right].$$

$$(13)$$

The anisotropy introduces a length scale $\lambda \equiv \sqrt{\kappa/B}$ into the problem. For smectic layers parallel to the external plates, the interaction now decays as $1/H^{(d-1)/2}$. The deformations of one plate modify the interaction, and the analog of Eq. (10) is

$$f(H) = -\frac{\zeta(2)}{16\pi}\frac{kT}{H\lambda} - \frac{\zeta(2)}{16\pi}\frac{kTA_SL^{2\zeta_s}}{\lambda H^3}$$
$$+ \frac{C_2}{4}\frac{kTA_S}{\lambda^{1-\zeta_s}H^{3-\zeta_s}},$$

$$(14)$$

with C_2 expressible in terms of a different set of kernel functions. The first term in Eq. (14) is identical to that obtained by Ajdari, Peliti, and Prost [2]. The decay of the last term is again related to the roughness exponents of the surface. Clearly these forces have the same magnitude as in the isotropic case of Eq. (10) for $H \approx \lambda$, but decay more slowly, and hence become comparatively stronger for $H \gg \lambda$.

So far, we have calculated Casimir effects for manifolds with type-I boundary conditions. Manifolds with type-II or mixed boundary conditions can be handled similarly. The type-II constraint, $\partial_\perp\phi = 0$, is inserted into the functional integral via $\int \mathcal{D}\psi\exp(i\psi\partial_\perp\phi)$, thus representing a dipole source. After integrating over ϕ, we obtain a quadratic action for the auxiliary fields Ψ, as in Eq. (4). However, whereas the coupling between two type-I manifolds is $G^d(\mathbf{r}-\mathbf{r}')$, it is $\partial_\perp G^d(\mathbf{r}-\mathbf{r}')$ between type I and type II, and $\partial_\perp\partial'_\perp G^d(\mathbf{r}-\mathbf{r}')$ between two type-II manifolds. The remaining computations can proceed as before. We find that two type-II boundaries still attract with an amplitude $1/(4\pi)^{d/2}\Gamma(d/2)\zeta(d)$, while mixed boundary conditions result in a repulsion of amplitude $1 - 2^{1-d}$ times that of like boundaries. Similar behavior is observed for smectic liquid-crystal layers [2], where the ratio of the interactions between I-I, II-II, and I-II

boundary conditions is given by $1:1:2^{(1-d)/2}-1$.

The $h(\mathbf{x})^2$ dependence of the Casimir energy in Eq. (7) may have important consequences for the fluctuations of a fluid surface, or a film. The fluctuations of a free surface are governed by a surface tension energy $\gamma \int d^3x (\nabla h)^2/2$. For sufficiently long wavelengths, the first term in Eq. (7) dominates the surface tension and modifies the fluctuations. [The second term in Eq. (7) is equivalent to an increase in surface tension of roughly 10^{-3} dyn/cm.] For like boundaries (such as a free-standing film) there is an instability to deformations, while for unlike boundaries (e.g., a film on a solid substrate) there is an additional stabilizing force. In the absence of any other interactions, the crossover length is $\lambda_0^f \sim 24 \, (\gamma_{\text{eff}}/kT)^{1/2}H^2$. For a film which is 100 Å thick, typical fluid-air interfacial tension yields $\lambda_0^f \sim 4$ μm. Of course, additional stabilizing forces may be present. For example, gravity produces an energy cost of $\rho g \int d^2x \, h^2/2$ for deformations. This is larger than the Casimir deformation energy for thicknesses $H > 0.6(kT/\rho g)^{1/4} \approx 0.5$ μm. Similarly, for a smectic liquid-crystal film, the crossover length is $\lambda_0^s \sim 35(\gamma_{\text{eff}}/kT)^{1/2}H^{3/2}\lambda^{1/2}$. We estimate that $\lambda_0^s \sim 6$ μm for $H = 100$ Å, and gravity becomes important for thicknesses $H > 2$ μm. There are few experiments measuring the roughness of liquid surfaces [15]. It would be interesting if future experiments can probe the effects of Casimir forces on the surface roughness of thin liquid films.

In summary, we have computed fluctuation-induced long-range interactions between slightly deformed plates for a variety of correlated fluids. These interactions can modify the fluctuations of the surface of thin fluids, and lead to interesting power-law corrections to the Casimir force between self-affinely rough substrates. The methods introduced can be easily generalized to calculate the interaction between other types of manifolds: For example, we find a $1/H$ energy between a surface and a long directed polymer parallel to it. We also computed the in-teraction of two-dimensional domains with fixed boundary shapes. Details of these results will be published elsewhere.

[1] H. B. G. Casimir, Proc. K. Ned. Akad. Wet. **51**, 793 (1948).

[2] A. Ajdari, L. Peliti, and J. Prost, Phys. Rev. Lett. **66**, 1481 (1991).

[3] J. L. Cardy, Phys. Rev. Lett. **65**, 1443 (1990), and references therein.

[4] M. P. Nightingale and J. O. Indekeu, Phys. Rev. Lett. **54**, 1824 (1985).

[5] L. Peliti and J. Prost, J. Phys. **50**, 1557 (1989).

[6] P. Kleban (to be published).

[7] R. Balian and B. Duplantier, Ann. Phys. (N.Y.) **112**, 165 (1978).

[8] B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1981).

[9] For a review, see J. N. Israelachvili and P. M. McGuiggan, Science **241**, 795 (1988).

[10] M. E. Fisher and P.-G. de Gennes, C. R. Acad. Ser. B **287**, 207 (1978).

[11] V. Privman and M. E. Fisher, Phys. Rev. B **30**, 322 (1984).

[12] H. W. J. Blöte, J. L. Cardy, and M. P. Nightingale, Phys. Rev. Lett. **56**, 742 (1986).

[13] P. Pfeifer, Y. J. Wu, M. W. Cole, and J. Krim, Phys. Rev. Lett. **62**, 1997 (1989); M. Kardar and J. O. Indekeu, Phys. Rev. Lett. **65**, 662 (1990).

[14] P.-G. de Gennes, *The Physics of Liquid Crystals* (Oxford Univ. Press, Oxford, 1974).

[15] A. Braslu, M. Deutsch, P. S. Pershan, A. H. Weiss, J. Als-Nielsen, and J. Bohr, Phys. Rev. Lett. **54**, 114 (1985).