



Casimir force in confined biomembranes

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abstract

We reexamine the computation of the Casimir force between two parallel interacting plates delimitating a liquid with an immersed biomembrane. We denote by D their separation, which is assumed to be much smaller than the bulk roughness, in order to ensure the membrane confinement. This repulsive force originates from the thermal undulations of the membrane. To this end, we first introduce a field theory, where the field is nothing else but the height-function. The field model depends on two parameters, namely the membrane bending rigidity constant, κ , and some elastic constant, $\mu \sim D^{-4}$. We first compute the static Casimir force (per unit area), Π , and find that the latter decays with separation D as : $\Pi \sim D^{-3}$, with a known amplitude scaling as κ^{-1} . Therefore, the force has significant values only for those biomembranes of small enough κ . Second, we consider a biomembrane (at temperature T) that is initially in a flat state away from thermal equilibrium, and we are interested in how the dynamic force, $\Pi(t)$, grows in time. To do calculations, use is made of a non-dissipative Langevin equation (with noise) that is solved by the time height-field. We first show that the membrane roughness, $L_{\perp}(t)$, increases with time as : $L_{\perp}(t) \sim t^{1/4}$ ($t < \tau$), with the final time $\tau \sim D^4$ (required time over which the final equilibrium state is reached). Also, we find that the force increases in time according to : $\Pi(t) \sim t^{1/2}$ ($t < \tau$). The discussion is extended to the real situation where the biomembrane is subject to hydrodynamic interactions caused by the surrounding liquid. In this case, we show that : $L_{\perp}(t) \sim t^{1/3}$ ($t < \tau_h$) and $\Pi_h(t) \sim t^{2/3}$ ($t < \tau_h$), with the new final time $\tau_h \sim D^3$. Consequently, the hydrodynamic interactions lead to substantial changes of the dynamic properties of the confined membrane, because both roughness and induced force grow more rapidly. Finally, the study may be extended, in a straightforward way, to bilayer surfactants confined to the same geometry.

Key words: Biomembranes - Confinement - Casimir force - Dynamics.

I. INTRODUCTION

The cell membranes are of great importance to life, because they separate the cell from the surrounding environment and act as a selective barrier for the import and export of materials. More details concerning the structural organization and basic functions of biomembranes can be found in Refs. [1 – 7]. It is well-recognized by the scientific community that the cell membranes essentially present as a phospholipid bilayer combined with a variety of proteins and cholesterol (*mosaic fluid model*). In particular, the function of the cholesterol molecules is to ensure the bilayer fluidity. A phospholipid is an amphiphile

molecule possessing a hydrophilic polar head attached to two hydrophobic (fatty acyl) chains. The phospholipids move freely on the membrane surface. On the other hand, the thickness of a bilayer membrane is of the order of 50 Angstroms. These two properties allow to consider it as a two-dimensional fluid membrane. The fluid membranes, self-assembled from surfactant solutions, may have a variety of shapes and topologies [8], which have been explained in terms of bending energy [9, 10].

In real situations, the biomembranes are not trapped in liquids of infinite extent, but they rather confined to geometrical boundaries, such as white and red globules or liposomes (as drugs transport agents [11 – 14]) in blood vessels. For simplicity, we consider the situation where the biomembrane is confined in a liquid domain that is finite in one spatial direction. We denote by D its size in this direction. For a tube, D being the diameter, and for a liquid domain delimited by two parallel plates, this size is simply the separation between walls. Naturally, the length D must be compared to the bulk roughness, L_{\perp}^0 , which is the typical size of humps caused by the thermal fluctuations of the membrane. The latter depends on the nature of lipid molecules forming the bilayer. The biomembrane is confined only when D is much smaller than the bulk roughness L_{\perp}^0 . This condition is similar to that usually encountered in confined polymers context [15].

The membrane undulations give rise to repulsive effective interactions between the confining geometrical boundaries. The induced force we term *Casimir force* is naturally a function of the size D , and must decays as this scale is increased. In this paper, we are interested in how this force decays with distance. To simplify calculations, we assume that the membrane is confined to two parallel plates that are a finite distance $D < L_{\perp}^0$ apart.

The word "Casimir" is inspired from the traditional Casimir effect. Such an effect, predicted, for the first time, by Hendrick Casimir in 1948 [16], is one of the fundamental discoveries in the last century. According to Casimir, the vacuum quantum fluctuations of a confined electromagnetic field generate an attractive force between two parallel uncharged conducting plates. The Casimir effect has been confirmed in more recent experiments by Lamoreaux [17] and by Mohideen and Roy [18]. Thereafter, Fisher and de Gennes [19], in a short note, remarked that the Casimir effect also appears in the context of critical systems, such as fluids, simple liquid mixtures, polymer blends, liquid ^4He , or liquid-crystals, confined to restricted geometries or in the presence of immersed colloidal particles. For these systems, the critical fluctuations of the order parameter play the role of the vacuum quantum fluctuations, and then, they lead to long-ranged forces between the confining walls or between immersed colloids [20, 21].

To compute the Casimir force between the confining walls, we first elaborate a more general field theory that takes into account the primitive interactions experienced by the confined membrane. As we shall see below, in confinement regime, the field model depends only on two parameters that are the membrane bending rigidity constant and a coupling constant containing all information concerning the interaction potential exerting by the walls. In addition, the last parameter is a known function of the separation D . With the help of the constructed free energy, we first computed the static Casimir force (per unit area), Π . The exact calculations show that the latter decays with separation D according to a power law, that is $\Pi \sim \kappa^{-1} (k_B T)^2 D^{-3}$, with a known amplitude. Here, $k_B T$ denotes the thermal energy, and κ the membrane bending rigidity constant. Of course, this force increases with temperature, and has significant values only for those biomembranes of small enough κ . The second problem we examined is the computation of the dynamic Casimir force, $\Pi(t)$. More precisely, we considered a biomembrane at temperature T that is initially in a flat state away from the thermal equilibrium, and we were interested in how the expected force grows in time, before the final state is reached. Using a scaling argument, we first showed that the membrane roughness, $L_{\perp}(t)$, grows with time as : $L_{\perp}(t) \sim t^{1/4}$ ($t < \tau$), with the final time $\tau \sim D^4$. The latter can be interpreted as the required time over which the final equilibrium state is reached. Second, using a non-dissipative Langevin equation, we found that the force increases in time according to the power law : $\Pi(t) \sim t^{1/2}$ ($t < \tau$). Third, the discussion is extended to the real situation where the biomembrane is subject to hydrodynamic interactions caused by the flow of the surrounding liquid. In this case, we show that : $L_{\perp}(t) \sim t^{1/3}$ ($t < \tau_h$) and $\Pi(t) \sim t^{2/3}$ ($t < \tau_h$), with the new final time $\tau_h \sim D^3$. Consequently, the hydrodynamic interactions give rise to drastic changes of the dynamic properties of the confined membrane, since both roughness and induced force grow more rapidly.

This paper is organized as follows. In Sec. II, we present the field model allowing the determination of the Casimir force from a static and dynamic point of view. The Sec. III and Sec. IV are devoted to the computation of the static and dynamic induced forces, respectively. We draw our conclusions in the last section. Some technical details are presented in Appendix.

II. THEORETICAL FORMULATION

Consider a fluctuating fluid membrane that is confined to two interacting parallel walls 1 and 2. We denote by D their finite separation. Naturally, the separation D must be compared to the bulk membrane roughness, L_{\perp}^0 , when the system is unconfined (free membrane). The membrane is confined only when the condition $L \ll L_{\perp}^0$ is fulfilled. For the opposite condition, that is $L \gg L_{\perp}^0$, we expect finite size corrections.

We assume that these walls are located at $z = -D/2$ and $z = D/2$, respectively. Here, z means the perpendicular distance. For simplicity, we suppose that the two surfaces are physically equivalent. We design by $V(z)$ the interaction potential exerted by one wall on the fluid membrane, in the absence of the other. Usually, $V(z)$ is the sum of a repulsive and an attractive potentials. A typical example is provided by the following potential [22]

$$V(z) = V_h(z) + V_{vdW}(z) , \tag{2.0}$$

where

$$V_h(z) = A_h e^{-z/\lambda_h} \tag{2.0}$$

represents the repulsive hydration potential due to the water molecules inserted between hydrophilic lipid heads [22]. The amplitude A_h and the potential-range λ_h are of the order of : $A_h \simeq 0.2 \text{ J/m}^2$ and $\lambda_h \simeq 0.2 - 0.3 \text{ nm}$. In fact, the amplitude A_h is $A_h = P_h \times \lambda_h$, with the hydration pressure $P_h \simeq 10^8 - 10^9 \text{ Pa}$. There, $V_{vdW}(z)$ accounts for the van der Waals potential between one wall and biomembrane, which are a distance z apart. Its form is as follows

$$V_{vdW}(z) = -\frac{H}{12\pi} \left[\frac{1}{z^2} - \frac{2}{(z + \delta)^2} + \frac{1}{(z + 2\delta)^2} \right] , \tag{2.0}$$

with the Hamaker constant $H \simeq 10^{-22} - 10^{-21} \text{ J}$, and $\delta \simeq 4 \text{ nm}$ denotes the membrane thickness. For large distance z , this implies

$$V_{vdW}(z) \sim \frac{W\delta^2}{z^4} . \tag{2.0}$$

Generally, in addition to the distance z , the interaction potential $V(z)$ depends on certain length-scales, (ξ_1, \dots, ξ_n) , which are the interactions ranges. The fluid membrane then experiences the following total potential

$$U(z) = V\left(\frac{D}{2} - z\right) + V\left(\frac{D}{2} + z\right) , \quad -\frac{D}{2} \leq z \leq \frac{D}{2} . \tag{2.0}$$

In the Monge representation, a point on the membrane can be described by the three-dimensional position vector $\mathbf{r} = (x, y, z = h(x, y))$, where $h(x, y) \in [-D/2, D/2]$ is the height-field. The latter then fluctuates around the mid-plane located at $z = 0$.

The Statistical Mechanics for the description of such a (tensionless) fluid membrane is based on the standard Canham-Helfrich Hamiltonian [9, 23]

$$\mathcal{H}[h] = \int dx dy \left[\frac{\kappa}{2} (\Delta h)^2 + W(h) \right] , \tag{2.0}$$

with the membrane bending rigidity constant κ . The latter is comparable to the thermal energy $k_B T$, where T is the absolute temperature and k_B is the Boltzmann's constant. There, $W(h)$ is the interaction potential per unit area, that is

$$W(h) = \frac{U(h)}{L^2} , \tag{2.0}$$

where the potential $U(h)$ is defined in Eq. (2), and L is the lateral linear size of the biomembrane. Let us discuss the pair-potential $W(h)$.

Firstly, Eq. (2) suggests that this total potential is an *even* function of the perpendicular distance h , that is

$$W(-h) = W(h) . \tag{2.0}$$

In particular, we have $W(-D/2) = W(D/2)$.

Secondly, when they exist, the zeros h_0 's of the potential function $U(h)$ are such that

$$V\left(\frac{D}{2} - h_0\right) = -V\left(\frac{D}{2} + h_0\right) . \tag{2.0}$$

This equality indicates that, if h_0 is a zero of the potential function, then, $-h_0$ is a zero too. The number of zeros is then an *even* number. In addition, the zero h_0 's are different from 0, in all cases. Indeed, the quantity $V(D/2)$ does not vanish, since it represents the potential created by one wall at the middle of the film. We emphasize that, when the potential processes no zero, it is either repulsive or attractive. When this same potential vanishes at some points, then, it is either repulsive or attractive between two consecutive zeros.

Thirdly, we first note that, from relation (2), we deduce that the first derivative of the potential function, with respect to distance h , is an *odd* function, that is $W'(-h) = -W'(h)$. Applying this relation to the midpoint $h = 0$ yields : $W'(0) = 0$. Therefore, the potential W exhibits an extremum at $h = 0$, whatever the form of the function $V(h)$. We find that this extremum is a maximum, if $V''(D/2) < 0$, and a minimum, if $V''(D/2) > 0$. The potential U presents an horizontal tangent at $h = 0$, if only if $V''(D/2) = 0$. On the other hand, the general condition giving the extrema $\{h_m\}$ is

$$\left. \frac{dV}{dh} \right|_{h=\frac{D}{2}-h_m} = \left. \frac{dV}{dh} \right|_{h=\frac{D}{2}+h_m} . \tag{2.0}$$

Since the first derivative $W'(h)$ is an odd function of distance h , it must have an *odd* number of extremum points. The point $h = h_m$ is a maximum, if

$$\left. \frac{d^2V}{dh^2} \right|_{h=\frac{D}{2}-h_m} < - \left. \frac{d^2V}{dh^2} \right|_{h=\frac{D}{2}+h_m} , \tag{2.0}$$

and a minimum, if

$$\left. \frac{d^2V}{dh^2} \right|_{h=\frac{D}{2}-h_m} > - \left. \frac{d^2V}{dh^2} \right|_{h=\frac{D}{2}+h_m} . \tag{2.0}$$

At point $h = h_m$, we have an *horizontal* tangent, if

$$\left. \frac{d^2V}{dh^2} \right|_{h=\frac{D}{2}-h_m} = - \left. \frac{d^2V}{dh^2} \right|_{h=\frac{D}{2}+h_m} . \tag{2.0}$$

The above deductions depends, of course, on the form of the interaction potential $V(h)$.

Fourthly, a simple dimensional analysis shows that the total interaction potential can be rewritten on the following scaling form

$$\frac{W(h)}{k_B T} = \frac{1}{D^2} \Phi\left(\frac{h}{D}, \frac{\xi_1}{D}, \dots, \frac{\xi_n}{D}\right) , \tag{2.0}$$

where (ξ_1, \dots, ξ_n) are the ranges of various interactions experienced by the membrane, and $\Phi(x_1, \dots, x_{n+1})$ is a $(n + 1)$ -factor scaling-function.

Finally, we note that the pair-potential $W(h)$ cannot be singular at $h = 0$. It is rather an analytic function in the h variable. Therefore, at fixed ratios ξ_i/D , an expansion of the scaling-function Φ , around the value $h = 0$, yields

$$\frac{W(h)}{k_B T} = \frac{\gamma}{2} \frac{h^2}{D^4} + \mathcal{O}(h^4) . \tag{2.0}$$

We restrict ourselves to the class of potentials that exhibit a minimum at the mid-plane $h = 0$. This assumption implies that the coefficient γ is positive definite, i.e. $\gamma > 0$. Of course, such a coefficient depends on the ratios of the scale-lengths ξ_i to the separation D .

In confinement regime where the distance h is small enough, we can approximate the total interaction potential by its quadratic part. In these conditions, the Canham-Helfrich Hamiltonian becomes

$$\mathcal{H}_0 [h] = \frac{1}{2} \int dx dy \left[\kappa (\Delta h)^2 + \mu h^2 \right] , \tag{2.0}$$

with the elastic constant

$$\mu = \gamma \frac{k_B T}{D^4} . \tag{2.0}$$

The prefactor γ will be computed below. The above expression for the elastic constant μ gives an idea on its dependance on the film thickness D . In addition, we state that this coefficient may be regarded as a *Lagrange multiplier* that fixes the value of the membrane roughness.

Thanks to the above Hamiltonian, we calculate the mean-expectation value of the physical quantities, like the height-correlation function (propagator or Green function), defined by

$$G(x - x', y - y') = \langle h(x, y) h(x', y') \rangle - \langle h(x, y) \rangle \langle h(x', y') \rangle . \tag{2.0}$$

The latter solves the linear differential equation

$$(\widehat{\kappa} \Delta^2 + \widehat{\mu}) G(x - x', y - y') = \delta(x - x') \delta(y - y') , \tag{2.0}$$

where $\delta(x)$ denotes the one-dimensional Dirac function, and $\Delta = \partial^2/\partial x^2 + \partial^2/\partial y^2$ represents the two-dimensional Laplacian operator. We have used the notations : $\widehat{\kappa} = \kappa/k_B T$ and $\widehat{\mu} = \mu/k_B T$, to mean the reduced membrane elastic constants.

From the propagator, we deduce the expression of the membrane roughness

$$L_{\perp}^2 = \langle h^2 \rangle - \langle h \rangle^2 = G(0, 0) . \tag{2.0}$$

Such a quantity measures the fluctuations of the height-function (fluctuations amplitude) around the equilibrium plane located at $z = 0$. We show in Appendix that the membrane roughness is exactly given by

$$L_{\perp}^2 = \frac{D^2}{12} , \tag{2.0}$$

provided that one is in the confinement-regime, i.e. $D \ll L_{\perp}^0$. Notice that the above equality indicates that the roughness is independent on the geometrical properties of the membrane (through κ). We emphasize that this relation can be recovered using the argument that each point of the membrane has equal probability to be found anywhere between the walls [24].

The elastic constant μ may be calculated using the known relation

$$L_{\perp}^2 = \frac{1}{8} \frac{k_B T}{\sqrt{\mu \kappa}} . \tag{2.0}$$

This gives

$$\mu = \frac{9}{4} \frac{(k_B T)^2}{\kappa D^4} . \tag{2.0}$$

This formula clearly shows that this elastic constant decays with separation D as D^{-4} . The term $\mu h^2/2$ then describes a confinement potential that ensures the localization of the membrane around the mid-plane. Integral over the hole plane \mathbf{R}^2 of this term represents the loss entropy due to the confinement of the membrane. The value (19) of the elastic constant is compatible with the constraint (17).

Therefore, the elaborated model is based on the Hamiltonian (13), with a quadratic confinement potential. We can say that the presence of the walls simply leads to a confinement of the membrane in a region

of the infinite space of perpendicular size L_{\perp} .

We define now another length-scale that is the in-plane correlation length, L_{\parallel} . The latter measures the correlations extent along the parallel directions to the walls. More precisely, the propagator $G(x-x', y-y')$ falls exponentially beyond L_{\parallel} , that is for distances d such that $d =$

$$\sqrt{(x-x')^2 + (y-y')^2} > L_{\parallel}.$$

From the standard relation

$$L_{\perp}^2 = \frac{k_B T}{16\kappa} L_{\parallel}^2, \quad (2.0)$$

we deduce

$$L_{\parallel} = \frac{2}{\sqrt{3}} \left(\frac{\kappa}{k_B T} \right)^{1/2} D. \quad (2.0)$$

In contrary to L_{\perp} , the length-scale L_{\parallel} depends on the geometrical characteristics of the membrane (through κ).

The next steps consist in the computation of the Casimir force at and out equilibrium.

III. STATIC CASIMIR FORCE

When viewed under the microscope, the membranes of vesicles present thermally excited shape fluctuations. Generally, objects such as interfaces, membranes or polymers undergo such fluctuations, in order to increase their configurational entropy. For bilayer biomembranes and surfactants, the consequence of these undulations is that, they give rise to an induced force called *Casimir force*.

To compute the desired force, we start from the partition function constructed with the Hamiltonian defined in Eq. (13). This partition function is the following functional integral

$$\mathcal{Z} = \int \mathcal{D}h \exp \left\{ -\frac{\mathcal{H}_0[h]}{k_B T} \right\}, \quad (3.0)$$

where integration is performed over all height-field configurations. The associated free energy is such that : $\mathcal{F} = -k_B T \ln \mathcal{Z}$, which is, of course, a function of the separation D . If we denote by $\Sigma = L^2$ the common area of plates, the Casimir force (per unit area) is *minus* the first derivative of the free energy (per unit area) with respect to the film-thickness D , that is

$$\Pi = -\frac{1}{\Sigma} \frac{\partial \mathcal{F}}{\partial D}. \quad (3.0)$$

This force per unit area is called disjoining pressure. In fact, Π is the required pressure to maintain the two plates at some distance D apart. In term of the partition function, the disjoining pressure rewrites

$$\frac{\Pi}{k_B T} = \frac{1}{\Sigma} \frac{\partial \ln \mathcal{Z}}{\partial D} = \frac{1}{\Sigma} \frac{\partial \mu}{\partial D} \frac{\partial \ln \mathcal{Z}}{\partial \mu}. \quad (3.0)$$

Using definition (19) together with Eqs. (23) and (24) yields

$$\Pi = -\frac{1}{2} \frac{\partial \mu}{\partial D} L_{\perp}^2. \quad (3.0)$$

Explicitly, we obtain the desired formula

$$\Pi = \frac{3}{8} \frac{(k_B T)^2}{\kappa D^3}. \quad (3.0)$$

From this relation, we extract the expression of the disjoining potential (per unit area) [25]

$$V_d(D) = -\int_{\infty}^D \Pi(D') dD' = \frac{3}{16} \frac{(k_B T)^2}{\kappa D^2}. \quad (3.0)$$

The above expression of the Casimir force (per unit area) calls the following remarks.

Firstly, this force decays with distance more slowly in comparison to the Coulombian one that decreases rather as D^{-2} .

Secondly, this same force depends on the nature of lipids forming the bilayer (through κ). In this sense, contrarily to the Casimir effect in Quantum Field Theory [16] and in Critical Phenomena [20], the present force is *not* universal. Incidentally, if this force is multiplied by κ , then, it will become a universal quantity. Thirdly, at fixed temperature and distance, the force amplitude has significant values only for those bilayer membrane of small bending rigidity constant.

Fourthly, as it should be, such a force increases with increasing temperature. Indeed, at high temperature, the membrane undulations are strong enough.

Finally, the numerical prefactor $3/8$ (Helfrich's c_H -amplitude [9]) is close to the value obtained using Monte Carlo simulation [26].

In Fig. 1, we superpose the variations of the reduced static Casimir force $\Pi/k_B T$ upon separation D , for two lipid systems, namely SOPC and DAPC [27], at temperature $T = 18^\circ\text{C}$. The respective membrane bending rigidity constants are : $\kappa = 0.96 \times 10^{-19}$ J and $\kappa = 0.49 \times 10^{-19}$ J. These values correspond to the renormalized bending rigidity constants : $\hat{\kappa} = 23.9$ and $\hat{\kappa} = 12.2$. The used methods for the measurement of these rigidity constants were *entropic tension* and *micropipet* [27]. These curves reflect the discussion made above.

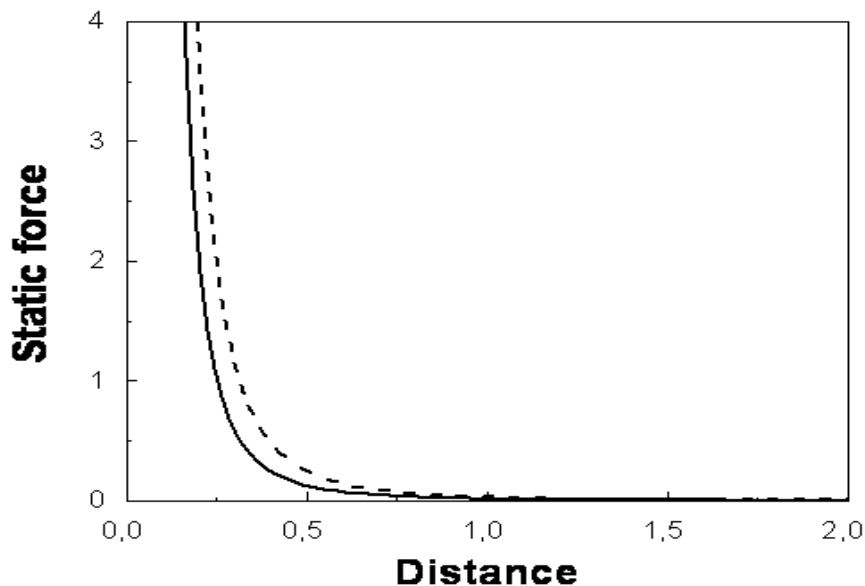


FIG. 1. Reduced static Casimir force, $\Pi/k_B T$, versus separation D , for two lipid systems that are SOPC (solid line) and DAPC (dashed line), of respective membrane bending rigidity constants : $\kappa = 0.96 \times 10^{-19}$ J and $\kappa = 0.49 \times 10^{-19}$ J, at temperature $T = 18^\circ\text{C}$. The reduced force and separation are expressed in arbitrary units.

IV. DYNAMIC CASIMIR FORCE

To study the dynamic phenomena, the main physical quantity to consider is the time height-field, $h(r, t)$, where $r = (x, y) \in \mathbf{R}^2$ denotes the position vector and t the time. The latter represents the time observation of the system before it reaches its final equilibrium state. We recall that the time height function $h(r, t)$ solves a non-dissipative Langevin equation (with noise) [28]

$$\frac{\partial h(r, t)}{\partial t} = -\Gamma \frac{\delta \mathcal{H}_0[h]}{\delta h(r, t)} + \nu(r, t) , \tag{4.0}$$

where $\Gamma > 0$ is a kinetic coefficient. The latter has the dimension : $[\Gamma] = L_0^4 T_0^{-1}$, where L_0 is some length and T_0 the time unit. Here, $\nu(r, t)$ is a Gaussian random force with mean zero and variance

$$\langle \nu(r, t) \nu(r', t') \rangle = 2\Gamma \delta_2(r - r') \delta(t - t') , \tag{4.0}$$

and \mathcal{H}_0 is the static Hamiltonian (divided by $k_B T$), defined in Eq. (13).

The bare time correlation function, whose Fourier transform is the dynamic structure factor, is defined by the expectation mean-value over noise ν

$$G(r - r', t - t') = \langle h(r, t) h(r', t') \rangle_\nu - \langle h(r, t) \rangle_\nu \langle h(r', t') \rangle_\nu , \quad t > t' . \tag{4.0}$$

The dynamic equation (28) shows that the time height function h is a functional of noise ν , and we write : $h = h[\nu]$. Instead of solving the Langevin equation for $h[\nu]$ and then averaging over the noise distribution $\mathcal{P}[\nu]$, the correlation and response functions can be directly computed by means of a suitable field-theory, of action [28 – 31]

$$\mathcal{A}[h, \tilde{h}] = \int dt \int d^2r \left\{ \tilde{h} \partial_t h + \Gamma \tilde{h} \frac{\delta \mathcal{H}_0}{\delta h} - \tilde{h} \Gamma \tilde{h} \right\} , \tag{4.0}$$

so that, for an arbitrary observable, $\mathcal{O}[\zeta]$, one has

$$\langle \mathcal{O} \rangle_\nu = \int [d\nu] \mathcal{O}[\varphi[\nu]] \mathcal{P}[\nu] = \frac{\int \mathcal{D}h \mathcal{D}\tilde{h} \mathcal{O} e^{-\mathcal{A}[h, \tilde{h}]}}{\int \mathcal{D}h \mathcal{D}\tilde{h} e^{-\mathcal{A}[h, \tilde{h}]}} , \tag{4.0}$$

where $\tilde{h}(r, t)$ is an auxiliary field, coupled to an external field $h(r, t)$. The correlation and response functions can be computed replacing the static Hamiltonian \mathcal{H}_0 appearing in Eq. (13), by a new one : $\mathcal{H}_0[h, J] = \mathcal{H}_0[h] - \int d^2r J h$. Consequently, for a given observable \mathcal{O} , we have

$$\left. \frac{\delta \langle \mathcal{O} \rangle_J}{\delta J(r, t)} \right|_{J=0} = \Gamma \langle \tilde{h}(r, t) \mathcal{O} \rangle . \tag{4.0}$$

The notation $\langle \cdot \rangle_J$ means the average taken with respect to the action $\mathcal{A}[h, \tilde{h}, J]$ associated with the Hamiltonian $\mathcal{H}_0[h, J]$. In view of the structure of equality (33), \tilde{h} is called *response field*. Now, if $\mathcal{O} = h$, we get the response of the order parameter field to the external perturbation J

$$R(r - r', t - t') = \left. \frac{\delta \langle h(r', t') \rangle_J}{\delta J(r, t)} \right|_{J=0} = \Gamma \langle \tilde{h}(r, t) h(r', t') \rangle_{J=0} . \tag{4.0}$$

The causality implies that the response function vanishes for $t < t'$. In fact, this function can be related to the time-dependent (connected) correlation function using the fluctuation-dissipation theorem, according to which

$$\Gamma \langle \tilde{h}(r, t) h(r', t') \rangle = -\theta(t - t') \partial_t \langle h(r, t) h(r', t') \rangle_c . \tag{4.0}$$

The above important formula shows that the time correlation function $C(r - r', t - t') = \langle \varphi(r, t) \varphi(r', t') \rangle_c$ may be determined by the knowledge of the response function. In particular, we show that

$$L_\perp^2(t) = \langle h^2(r, t) \rangle_c = -2\Gamma \int_{-\infty}^t dt' \langle \tilde{h}(r, t') h(r, t') \rangle . \tag{4.0}$$

The limit $t \rightarrow -\infty$ gives the natural value $L_\perp^2(-\infty) = 0$, since, as assumed, the initial state corresponds to a completely flat interface.

Consider now a membrane at temperature T that is initially in a flat state away from thermal equilibrium. At a later time t , the membrane possesses a certain roughness, $L_\perp(t)$. Of course, the latter is time-dependent, and we are interested in how it increases in time.

We point out that the thermal fluctuations give rise to some roughness that is characterized by the appearance of anisotropic humps. Therefore, a segment of linear size L effectuates excursions of size [32]

$$L_{\perp} = BL^{\zeta} . \tag{4.0}$$

Such a relation defines the *roughness exponent* ζ . Notice that L is of the order of the in-plane correlation length, L_{\parallel} . From relation (20), we deduce the exponent ζ and the amplitude B . Their respective values are : $\zeta = 1$ and $B \sim (k_B T/\kappa)^{1/2}$.

In order to determine the growth of roughness L_{\perp} in time, the key is to consider the excess free energy (per unit area) due to the confinement, ΔF . Such an excess is related to the fact that the confining membrane suffers a loss of entropy. Formula (27) tells us how ΔF must decay with separation. The result reads [32]

$$\Delta F \sim k_B T/L_{\max}^2 \sim k_B T (B/L_{\perp})^{2/\zeta} , \tag{4.0}$$

where L_{\max} represents the wavelength above which all shape fluctuations are not accessible by the confined membrane. The repulsive fluctuation-induced interaction leads to the disjoining pressure

$$\Pi = -\frac{\partial \Delta F}{\partial L_{\perp}} \sim L_{\perp}^{-(1+2/\zeta)} . \tag{4.0}$$

In addition, a care analysis of the Langevin equation (28) shows that

$$\frac{\partial L_{\perp}}{\partial t} \sim -\Gamma \frac{\partial \Delta F}{\partial L_{\perp}} = \Gamma \times \Pi \sim \Gamma L_{\perp}^{-(1+2/\zeta)} . \tag{4.0}$$

We emphasize that this scaling form agrees with Monte Carlo predictions [32, 33]. Solving this first-order differential equation yields [34]

$$L_{\perp}(t) \sim \Gamma^{\theta_{\perp}} t^{\theta_{\perp}} , \quad \theta_{\perp} = \frac{\zeta}{2 + 2\zeta} = \frac{1}{4} . \tag{4.0}$$

This implies the following scaling form for the linear size

$$L(t) \sim \Gamma^{\theta_{\parallel}} t^{\theta_{\parallel}} , \quad \theta_{\parallel} = \frac{1}{2 + 2\zeta} = \frac{1}{4} . \tag{4.0}$$

Let us comment about the obtained result (39).

Firstly, as it should be, the roughness increases with time (the exponent θ_{\perp} is positive definite). In addition, the exponent θ_{\perp} is *universal*, independently on the membrane bending rigidity constant κ . Secondly, we note that, in Eq. (39), we have ignored some non-universal amplitude that scales as $\kappa^{-1/4}$. This means that the time roughness is significant only for those biomembranes of small bending rigidity constant.

Fourthly, this time roughness can be interpreted as the perpendicular size of holes and valleys at time t . Fifthly, the roughness increases until a fine time, τ . The latter can be interpreted as the time over which the system reaches its final equilibrium state. This characteristic time then scales as

$$\tau \sim \Gamma^{-1} L_{\perp}^{1/\theta_{\perp}} , \tag{4.0}$$

where we have ignored some non-universal amplitude that scales as κ . Here, $L_{\perp} \sim D$ is the final roughness. Explicitly, we have

$$\tau \sim \Gamma^{-1} D^4 . \tag{4.0}$$

As it should be, the final time increases with increasing film thickness D . On the other hand, we can rewrite the behavior (39) as

$$\frac{L_{\perp}(t)}{L_{\perp}(\tau)} = \left(\frac{t}{\tau}\right)^{\theta_{\perp}} . \tag{4.0}$$

This equality means that the roughness ratio, as a function of the reduced time, is *universal*. Now, to compute the dynamic Casimir force, we start from a formula analog to that defined in Eq. (24), that is

$$\frac{\Pi(t)}{k_B T} = \frac{1}{\Sigma} \frac{\partial \ln \widehat{Z}}{\partial D} = \frac{1}{\Sigma} \frac{\partial \mu}{\partial D} \frac{\partial \ln \widehat{Z}}{\partial \mu}, \tag{4.0}$$

with the new partition function

$$\widehat{Z} = \int \mathcal{D}h \mathcal{D}\tilde{h} e^{-\mathcal{A}[h, \tilde{h}]}. \tag{4.0}$$

A simple algebra taking into account the basic relation (35a) gives

$$\frac{\Pi(t)}{k_B T} = -\frac{1}{2} \frac{\partial \mu}{\partial D} L_{\perp}^2(t), \tag{4.0}$$

which is very similar to the static relation defined in Eq. (25), but with a time-dependent membrane roughness, $L_{\perp}(t)$.

Combining formulae (43) and (46) leads to the desired expression for the time Casimir force (per unit area)

$$\frac{\Pi(t)}{\Pi(\tau)} = \left(\frac{t}{\tau}\right)^{\theta_f}, \tag{4.0}$$

where $\Pi(\tau)$ is the final static Casimir force, relation (25). The *force exponent*, θ_f , is such that

$$\theta_f = 2\theta_{\perp} = \frac{\zeta}{1+\zeta} = \frac{1}{2}. \tag{4.0}$$

The induced force then grows with time as $t^{1/2}$ until it reaches its final value $\Pi(\tau)$. At fixed time and separation D , the force amplitude depends, of course, on κ , and decreases in this parameter according to $\kappa^{-3/2}$. Also, we note that the above equality means that the force ratio as a function of the reduced time is *universal*.

In Fig. 2, we draw the reduced dynamic Casimir force, $\Pi(t)/\Pi(\tau)$, upon the renormalized time t/τ .

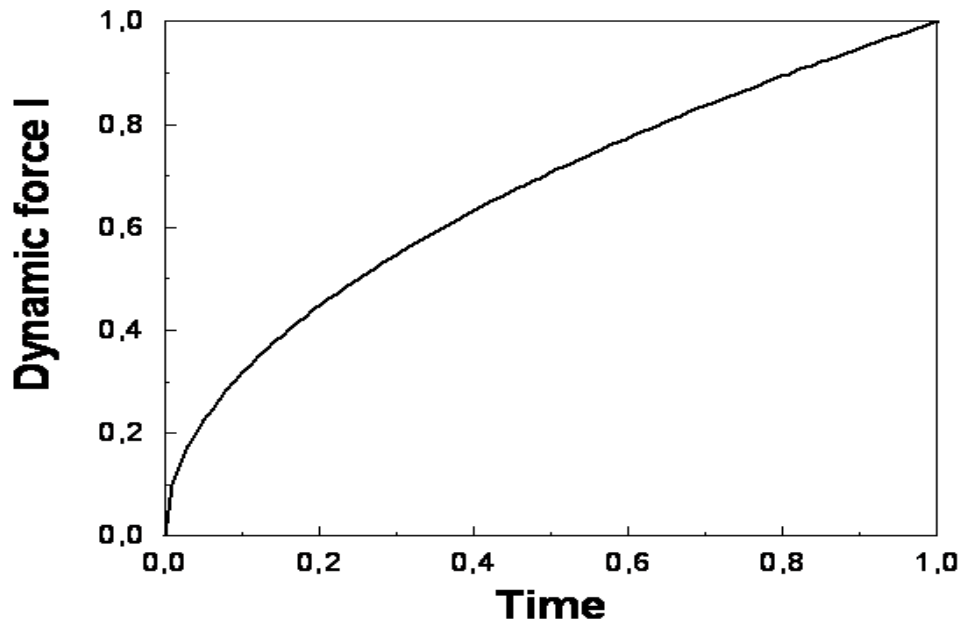


FIG. 2. Reduced dynamic Casimir force, $\Pi(t)/\Pi(\tau)$, upon the renormalized time t/τ .

Finally, consider again a membrane which is initially flat but is now coupled to overdamped surface waves. This real situation corresponds to a confined membrane subject to hydrodynamic interactions. The roughness now grows as [35]

$$\widehat{L}_\perp(t) \sim t^{\widehat{\theta}_\perp}, \quad \widehat{\theta}_\perp = \frac{\zeta}{1+2\zeta} = \frac{1}{3}. \tag{4.0}$$

Therefore, the roughness increases with time *more rapidly* than that relative to biomembranes free from hydrodynamic interactions.

In this case, the dynamic Casimir force is such that

$$\frac{\Pi_h(t)}{\Pi(\tau_h)} = \left(\frac{t}{\tau_h}\right)^{\widehat{\theta}_f}, \tag{4.0}$$

where $\Pi(\tau_h)$ is the final static Casimir force, relation (25). The new force exponent is

$$\widehat{\theta}_f = 2\widehat{\theta}_\perp = \frac{2\zeta}{1+2\zeta} = \frac{2}{3}. \tag{4.0}$$

There, $\tau_h \sim D^3$ accounts for the new time-scale over which the confined membrane reaches its final equilibrium state. Therefore, the dynamic Casimir force decays with time as $t^{2/3}$, that is more rapidly than that where the hydrodynamic interactions are ignored, which scales rather as $t^{1/2}$. As we said before, this drastic change can be attributed to the overdamped surface waves that develop larger and larger humps.

We depict, in Fig. 3, the variation of the reduced dynamic force (with hydrodynamic interactions), $\Pi_h(t)/\Pi(\tau_h)$, upon the renormalized time t/τ_h .

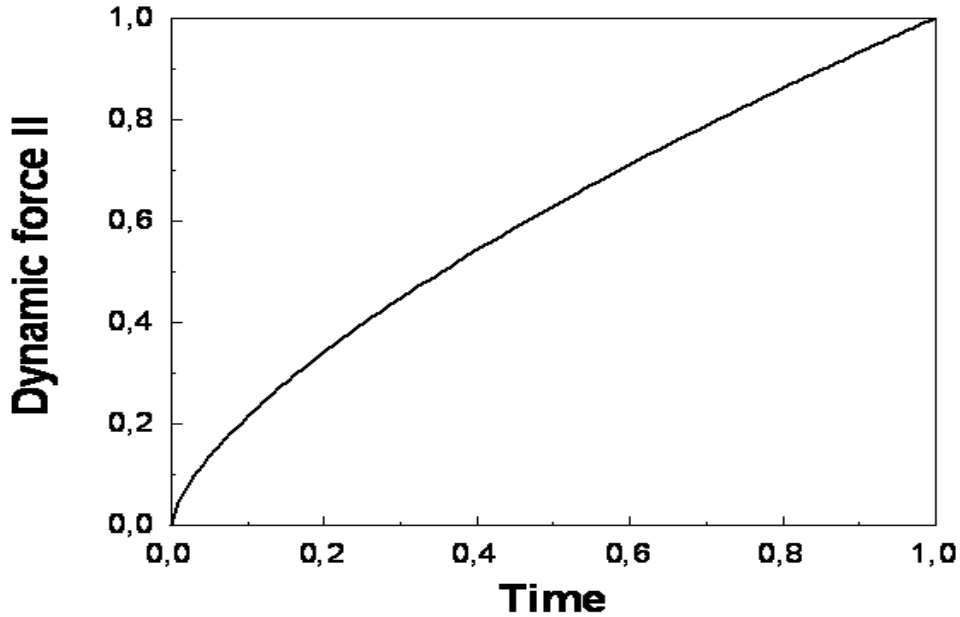


FIG. 3. Reduced dynamic Casimir force (with hydrodynamic interactions), $\Pi_h(t) / \Pi(\tau)$, upon the renormalized time t/τ_h .

V. CONCLUSIONS

In this work, we have reexamined the computation of the Casimir force between two parallel walls delimitating a fluctuating fluid membrane that is immersed in some liquid. This force is caused by the thermal fluctuations of the membrane. We have studied the problem from both static and dynamic point of view.

We were first interested in the time variation of the roughening, $L_{\perp}(t)$, starting with a membrane that is initially in a flat state, at a certain temperature. Of course, this length grows with time, and we found that : $L_{\perp}(t) \sim t^{\theta_{\perp}}$ ($\theta_{\perp} = 1/4$), provided that the hydrodynamic interactions are ignored. For real systems, however, these interactions are important, and we have shown that the roughness increases more rapidly as : $\widehat{L}_{\perp}(t) \sim t^{\widehat{\theta}_{\perp}}$ ($\widehat{\theta}_{\perp} = 1/3$). The dynamic process is then stopped at a final τ (or τ_h) that represents the required time over which the biomembrane reaches its final equilibrium state. The final time behaves as : $\tau \sim D^4$ (or $\tau_h \sim D^3$), with D the film thickness.

Now, assume that the system is explored at scales of the order of the wavelength q^{-1} , where $q = (4\pi/\lambda) \sin(\theta/2)$ is the wave vector modulus, with λ the wavelength of the incident radiation and θ the scattering-angle. In these conditions, the relaxation rate, $\tau(q)$, scales with q as : $\tau^{-1}(q) \sim q^{1/\theta_{\perp}} = q^4$ or $(\tau_h^{-1}(q) \sim q^{1/\widehat{\theta}_{\perp}} = q^3)$. Physically speaking, the relaxation rate characterizes the local growth of the height fluctuations.

Afterwards, the question was addressed to the computation of the Casimir force, Π . At equilibrium, using an appropriate field theory, we found that this force decays with separation D as : $\Pi \sim D^{-3}$, with a known amplitude scaling as κ^{-1} , where κ is the membrane bending rigidity constant. Such a force is then very small in comparison with the Coulombian one. In addition, this force disappears when the

temperature of the medium is sufficiently lowered.

The dynamic Casimir force, $\Pi(t)$, was computed using a non-dissipative Langevin equation (with noise), solved by the time height-field. We have shown that : $\Pi(t) \sim t^{\theta_f}$ ($\theta_f = 2\theta_{\perp} = 1/2$). When the hydrodynamic interactions effects are important, we found that the dynamic force increases more rapidly as : $\Pi_h(t) \sim t^{\hat{\theta}_f}$ ($\hat{\theta}_f = 2\hat{\theta}_{\perp} = 2/3$).

Notice that we have ignored some details such as the role of inclusions (proteins, cholesterol, glycolipids, other macromolecules) and chemical mismatch on the force expression. It is well-established that these details simply lead to an additive renormalization of the bending rigidity constant. Indeed, we write $\kappa_{\text{effective}} = \kappa + \delta\kappa$, where κ is the bending rigidity constant of the membrane free from inclusions, and $\delta\kappa$ is the contribution of the incorporated entities. Generally, the shift $\delta\kappa$ is a function of the inclusion concentration and compositions of species of different chemical nature (various phospholipids forming the bilayer). Hence, to take into account the presence of inclusions and chemical mismatch, it would be sufficient to replace κ by $\kappa_{\text{effective}}$, in the above established relations.

As last word, we emphasize that the results derived in this paper may be extended to bilayer surfactants, although the two systems are not of the same structure and composition. One of the differences is the magnitude order of the bending rigidity constant.

APPENDIX

To show formula (17), we start from the partition function that we rewrite on the following form

$$\mathcal{Z} = \int \mathcal{D}h \exp \left\{ -\frac{\mathcal{H}[h]}{k_B T} \right\} = \int_{-D/2}^{D/2} dz \Phi(z) . \quad (5.0)$$

Also, it is easy to see that the membrane mean-roughness is given by

$$L_{\perp}^2 = \frac{\int_{-D/2}^{D/2} dz z^2 \Phi(z)}{\int_{-D/2}^{D/2} dz \Phi(z)} . \quad (5.0)$$

The restricted partition function is

$$\Phi(z) = \int \mathcal{D}h \delta[z - h(x_0, y_0)] \exp \left\{ -\frac{\mathcal{H}[h]}{k_B T} \right\} . \quad (5.0)$$

Here, $\mathcal{H}[h]$ is the original Hamiltonian defined in Eq. (3). Of course, this definition is independent on the chosen point (x_0, y_0) , because of the translation symmetry along the parallel directions to plates. Notice that the above function is not singular, whatever the value of the perpendicular distance.

Since we are interested in the confinement-regime, that is when the separation D is much smaller than the membrane mean-roughness L_{\perp}^0 ($z \sim h \ll L_{\perp}^0$), we can replace the function Φ par its value at $z = 0$, denoted Φ_0 . In this limit, Eq. (A.2) gives the desired result.

This ends the proof of the expected formula.

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