Dynamic fluctuation phenomena in double membrane films

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Dynamics of double membrane films is investigated in the long-wavelength limit $qh \ll 1$ (q is the wave vector, and h is the thickness of the film) including the overdamped squeezing mode. We demonstrate that thermal fluctuations essentially modify the character of the mode due to its nonlinear coupling to the transverse shear hydrodynamic mode. The renormalization can be analyzed if the condition $g \ll 1$ is satisfied (where $g \sim T/\kappa$, T is the temperature, and κ is the bending modulus). The corresponding Green's function acquires as a function of the frequency ω a cut along the imaginary semiaxis. At $qh > \sqrt{g}$ the effective length of the cut is $\sim Tq^3/\eta$ (where η is the shear viscosity of the liquid). At $qh < \sqrt{g}$ the fluctuations lead to an increase in the attenuation of the squeezing mode: it is larger than the 'bare' value by a factor $1/\sqrt{g}$. We also present the analysis of the elastic modes. © *1998 American Institute of Physics.* [S1063-7761(98)01406-1]

1. INTRODUCTION

The most distinctive property of amphiphilic molecules is their ability to spontaneously self-assemble into aggregates of various shapes. Typically, the molecules spontaneously self-assemble into membranes which are bilayers of a thickness of the order of a molecular length. Different lyotropic structures consisting of these membranes have generated considerable current interest (see Refs. 1–3 and review articles[4–6]). Films composed of two bilayer membranes sandwiching a thin layer of a liquid are widely used in the lyotropic systems. They play also an essential role in various biological processes (note the so-called flickering phenomena in erythrocytes or red blood cells). In this paper we will examine the dynamic properties of such double membrane films.

The main peculiarity of a membrane is its negligible surface tension. The membrane is immersed into a liquid and consequently its area can vary. Zero surface tension is the equilibrium condition with respect to the variations. In this case the shape fluctuations of the membrane are determined by the bending elasticity; the corresponding energy is[7,8]

$$\mathscr{H}_{\text{curv}} = \frac{\kappa}{2} \int dA \left(\frac{1}{R_1} + \frac{1}{R_2} \right)^2, \tag{1}$$

where the integral is taken over the membrane which is considered as a two-dimensional object, R_1 and R_2 are its local curvature radii, and κ is the bending rigidity modulus. Corrugations of the membrane induced by the thermal noise lead to the loss of the orientation correlation of the membrane pieces at separations larger than the so-called persistent length[9] ξ_p, which is estimated to be

$$\xi_p \sim a \, \exp(2\pi\kappa/T),$$

where T is the temperature, and a is the thickness of the membrane. The shape fluctuations of the membrane lead to the logarithmic renormalization of the bending modulus κ, which was examined first by Helfrich[10] and later by Förster.[11] The correct renormalization-group (RG) equation was derived by Peliti and Liebler,[12] Kleinert,[13] and Polyakov.[14] The explicit form of the one-loop RG equation is

$$\frac{d\kappa}{d\xi} = -\frac{3T}{4\pi}.$$

Here $\xi = \ln(r/a)$, and r is the characteristic scale. As follows from the equation, the role of the dimensionless coupling constant is played by the quantity

$$g = \frac{3T}{4\pi\kappa}. \tag{2}$$

Note that $\ln(\xi_p/a) \sim g^{-1}$. For real membranes $g \sim 10^{-2}$–10^{-3} and, consequently, we can treat g as a small parameter. The smallness of g means that there exists a wide range of scales $r < \xi_p$ where the thermal fluctuations can be treated in the framework of the perturbation theory.

Below we consider a double membrane film. We assume that at equilibrium the film is parallel to the xy plane. Corrugations of the membranes in a double film can be decomposed into undulation (or bending) deformations and squeez-

ing deformations. The bending deformations are characterized by the displacement u of the film as a whole from its equilibrium position along the z axis and the squeezing deformation is characterized by variations of the film thickness h (which is the separation between the membranes). In the harmonic approximation we obtain from (1) the energy

$$\mathscr{H}=\int dx\,dy\left[\kappa(\nabla^2 u)^2+\frac{\kappa}{4}(\nabla^2 h)^2\right],\tag{3}$$

where u and h are treated as functions of x and y, and ∇ is the two-dimensional gradient.

In deriving (3) we disregarded the interaction between the membranes. First, one should remember the steric interaction, which is associated with a certain restriction of accessible configurations for one membrane in the presence of the second membrane.[8] The explicit expression for the energy is[15]

$$\mathscr{H}_{\text{ster}}=\int dx\,dy\,\frac{3\pi^2 T^2}{128\kappa h^2}.\tag{4}$$

Due to the interaction (4) two membranes can be treated independently only on scales smaller than $g^{-1/2}h$. Therefore (3) is the main contribution to the energy if

$$qh>\sqrt{g},\tag{5}$$

where q is the characteristic wave vector. Second, we should take into account the van der Waals interaction. We assume that the same liquid is inside and outside the film. We can then write the van der Waals energy as[16]

$$\mathscr{H}_{\text{vdw}}=\int dx\,dy\,\frac{Ha^2}{2\pi h^4},\tag{6}$$

where H is the Hamaker constant. We can disregard this energy, in contrast with (3), if

$$(qh)^4>\frac{H}{\kappa}\left(\frac{a}{h}\right)^2.$$

Let us assume that the thickness of the film is large enough to satisfy the inequality

$$g^2>\frac{H}{\kappa}\left(\frac{a}{h}\right)^2.$$

Then (5) is the only restriction that enables us to treat the energy (3) as the main contribution to the film energy.

2. DYNAMICS

We will examine the dynamics of the double membrane film in the long-wavelength limit $qh\ll 1$, where q is the wave vector of the eigenmodes of the film. Note that the inequality $qh\ll 1$ is compatible with (5) since $g\ll 1$. In the limit $qh\ll 1$ we should take into account the following variables which describe the dynamics: the velocity of the film $\mathbf{v}$, the displacement of the film u, the film thickness h, and the densities of the two membranes since they are conserved quantities. We are interested mainly in the squeezing mode, which is associated with the relaxation of the thickness h.

To find dynamical characteristics of the film one should solve the conventional hydrodynamic equations in bulk supplemented by boundary conditions on both membranes. In the linear approximation the problem was solved by Brochard and Lennon,[17] who found the dispersion relation of the squeezing mode

$$\omega=-i\,\frac{\kappa h_0^3 q^6}{24\eta},\tag{7}$$

where ω is the frequency of the mode, h_0 is the equilibrium separation between the membranes, and η is the viscosity of the liquid surrounding the membranes. In deriving (7) it was assumed that at equilibrium the film is flat. The dispersion relation

$$\omega=-i\,\frac{\kappa q^3}{2\eta}\tag{8}$$

of the bending mode was also found in the linear approximation. Note that the dispersion relation (7) is correct only if one neglects the direct interaction of the membranes upon satisfaction of the condition (5), whereas the region of applicability of the dispersion relation (8) does not depend on the interaction of the membranes, since they move in-phase in the bending mode. The elastic modes associated with variations of the membrane densities are harder than (7) and (8).[18] Therefore the only effect of the elastic degrees of freedom upon examination of the squeezing mode is the incompressibility condition

$$\nabla_\alpha v_\alpha=0.\tag{9}$$

Here and below we believe that all variables characterizing the film are functions of x and y and we assume that the Greek subscripts run over x and y.

We will consider the renormalization of the dispersion law (7) of the squeezing mode due to fluctuational effects. Nonlinear dynamical equations of the film should be utilized for this purpose. In the long-wavelength limit $qh_0\ll 1$ the equations can be derived phenomenologically. The reactive (nondissipative) part of the equations can be found by using the Poisson brackets method (see Ref. 19 and also Ref. 18), whereas the dissipative part of the equations is expressed in terms of the kinetic coefficients. One should know the expression for the energy $\mathscr{H}$ of the system to write both contributions. Actually, we will need the expression for one Poisson bracket:

$$\{j_\alpha(x_1,y_1),h(x_2,y_2)\}=h(x_1,y_1)\nabla_\alpha[\delta(x_1-x_2)\delta(y_1-y_2)],\tag{10}$$

where j_α is the two-dimensional momentum density of the film. The expression (10) (which is characteristic of two-dimensional density of any conserved scalar quantity on a film[18]) is motivated by the fact that the two-dimensional mass density of the film is ρh, where ρ is the three-dimensional density of the liquid. Note that $j_\alpha\approx\rho h v_\alpha$ since we believe that the membrane thickness a can be ignored in comparison with the film thickness h.

The dynamic equation for the thickness h has the standard form following from (10)

$$\partial_t h + \nabla_\alpha (v_\alpha h) = \Gamma \nabla^2 \frac{\delta \mathscr{H}}{\delta h}, \tag{11}$$

where $\partial_t \equiv \partial/\partial t$, and Γ is the kinetic coefficient. The second power of the gradient appears in (11) since the equation should support the conservation law of the liquid inside the film and therefore the right-hand side of the equation should be a full derivative at any $\mathscr{H}$. Due to (9) the second term on the left-hand side of (11) describes the sweeping of h by the velocity v_α. In the linear approximation we can ignore the sweeping term. Substituting the harmonic expression (3) for the energy $\mathscr{H}$ into (11) and comparing the result with (7), we obtain

$$\Gamma = h_0^3 / 12\eta. \tag{12}$$

Note that Γ is inversely proportional to the shear viscosity coefficient. The point is that the dissipation described by Γ comes from viscous motion of the liquid surrounding the double membrane film which is excited only slightly at large η.

The dynamic equation for j_α has the form[21]

$$\partial_t j_\alpha - \{\mathscr{H}, j_\alpha\} = J_\alpha, \tag{13}$$

where **J** is the momentum flow from the bulk to the film. Since this term supplies the main dissipation of the film momentum, we ignored the internal viscosity. The Poisson bracket $\{\mathscr{H}, j_\alpha\}$ can be reduced to the divergence of the symmetric stress tensor for any energy $\mathscr{H}$.[18] Actually, only the contribution associated with the Poisson bracket (10) and created by the harmonic energy (3) is relevant for us. We can then write Eq. (13) in the form

$$\partial_t j_\alpha + \frac{\kappa}{2} h \nabla_\alpha \nabla^4 h + \nabla_\alpha P_s = J_\alpha, \tag{14}$$

where P_s is the two-dimensional pressure, which is related to the elastic degrees of freedom (see the Appendix). In the linear approximation relevant for us we can write[21]

$$J_\alpha = -2\eta \hat{q} v_\alpha, \tag{15}$$

where $\hat{q}$ is the nonlocal operator, which is reduced to multiplying by the absolute value of the wave vector q in the Fourier representation. The expression (15) implies the inequality $\omega \ll \eta q^2 / \rho$, which is satisfied for the squeezing mode.

We will not present here dynamical equations for the variables j_z and u and for the densities of the membranes. The reason is that the equations for j_z and u, which describe the bending mode, decouple in the approximation used from Eqs. (11) and (14). Actually, the equations describing the bending motion of the double film are the same as for a single membrane and the corresponding nonlinear equations can be found in Ref. 18 and also in Refs. 20 and 21. One should remember only that the bending modulus of the double film is 2κ, as follows from (3). As to the equations for the densities of the membranes, they need a separate analysis, which is presented in the Appendix. The only role of the degrees of freedom in analyzing the squeezing mode reduces to the incompressibility condition (9).

3. RENORMALIZATION OF THE SQUEEZING MODE

As can be seen from (7), in the long-wavelength limit the squeezing mode is very soft. This is the reason why one anticipates that fluctuational effects which are related to the mode are relevant. The effects are associated with the non-linear terms in the dynamic equations and can be examined in terms of the diagrammatic technique of the first type which was developed by Wyld,[22] who studied the velocity fluctuations in a turbulent fluid. In Ref. 23 the Wyld technique was generalized for a broad class of dynamical systems. A textbook description of the diagram technique can be found in the book by Ma.[24] The diagram technique can be formulated in terms of path integrals, as was first suggested by de Dominicis[25] and Janssen.[26] In the framework of this approach apart from conventional dynamic variables one should also introduce auxiliary fields conjugated to the variables. The dynamic correlation functions of the variables can then be presented as functional integrals over both types of fields: conventional and auxiliary. The integrals are taken with the weight $\exp(i\mathscr{T})$, where $\mathscr{T}$ is the effective action which is constructed on the basis of nonlinear dynamic equations of the system.

Since we are interested in the renormalization of the squeezing mode of the double membrane film, we will take into account only the variables h and v_α and the corresponding auxiliary conjugated fields p and μ_α. We should also remember the incompressibility condition (9) and impose an analogous constraint $\nabla_\alpha \mu_\alpha = 0$ on the field μ_α. We can then write the correlation function of the film thickness h in the form

$$\langle h_1 h_2 \rangle = \int \mathscr{D}h \mathscr{D}\mathbf{v}_{tr} \mathscr{D}p \mathscr{D}\mu_{tr} \exp(i\mathscr{T}) h_1 h_2, \tag{16}$$

where the subscript "*tr*" implies that in the Fourier representation we should use only the components of the fields **v** and $\boldsymbol{\mu}$, which are transverse to the wave vector **q**. The explicit expression for the effective action in (16) can be found by using the dynamic equations (11) and (14). It can be written as the sum of the reactive part and the dissipative part $\mathscr{T} = \mathscr{T}_{\text{reac}} + \mathscr{T}_{\text{diss}}$, where

$$\mathscr{T}_{\text{reac}} = \int dt\, d^2r \left\{ p\partial_t h + pv_\alpha \nabla_\alpha h \right.$$
$$\left. + \mu_\alpha \partial_t j_\alpha - \frac{\kappa}{2} \mu_\alpha \nabla^4 h \nabla_\alpha h \right\}, \tag{17}$$

$$\mathscr{T}_{\text{diss}} = \int dt\, d^2r \left\{ -\frac{1}{2} \Gamma \kappa p \nabla^6 h \right.$$
$$\left. + iT\Gamma (\nabla p)^2 + 2\eta \mu \hat{q} (\mathbf{v} + iT\mu) \right\}. \tag{18}$$

The detailed derivation of the effective action for the problem can be found in Refs. 20 and 21.

We introduce the notation for the pair correlation functions. Taking into account only the transverse components of the fields **v** and $\boldsymbol{\mu}$, we can write

$$\langle h(t,\mathbf{r})p(0,0)\rangle = \int \frac{d\omega\, d^2q}{(2\pi)^3}\exp(-i\omega t+i\mathbf{q}\mathbf{r})G(\omega,\mathbf{q}),$$

$$\langle v_\alpha(t,\mathbf{r})\mu_\beta(0,0)\rangle = \int \frac{d\omega\, d^2q}{(2\pi)^3}\exp(-i\omega t+i\mathbf{q}\mathbf{r})$$

$$\times\left[\delta_{\alpha\beta}-\frac{q_\alpha q_\beta}{q^2}\right]G_{tr}(\omega,\mathbf{q}), \qquad (19)$$

$$\langle h(t,\mathbf{r})h(0,0)\rangle = \int \frac{d\omega\, d^2q}{(2\pi)^3}\exp(-i\omega t+i\mathbf{q}\mathbf{r})D(\omega,\mathbf{q}),$$

$$\langle v_\alpha(t,\mathbf{r})v_\beta(0,0)\rangle = \int \frac{d\omega\, d^2q}{(2\pi)^3}\exp(-i\omega t+i\mathbf{q}\mathbf{r})$$

$$\times\left[\delta_{\alpha\beta}-\frac{q_\alpha q_\beta}{q^2}\right]D_{tr}(\omega,\mathbf{q}). \qquad (20)$$

The correlation functions $\langle pp\rangle$ and $\langle\boldsymbol{\mu}\boldsymbol{\mu}\rangle$ are equal to zero (for the general property of the technique, see, e.g., Ref. 18). The functions D and D_{tr} determine the pair correlation functions of the observable quantities and the functions G and G_{tr} are the response functions. Therefore, the function $G(\omega)$ is analytic in the upper ω half-plane.

It is possible to formulate the diagram technique for calculating the correlation functions (19) and (20). The harmonic part of the effective action $\mathscr{T}=\mathscr{T}_{\text{reac}}+\mathscr{T}_{\text{diss}}$ determines the bare values of the response functions

$$G_0(\omega,\mathbf{q})=-\frac{1}{\omega+i\Gamma\kappa q^6/2}, \quad G_{tr,0}(\omega,\mathbf{q})=-\frac{1}{\rho h\omega+2i\eta q}. \qquad (21)$$

The values of the 'bare' pair correlation functions satisfy the relations

$$\text{Im}\,G = \frac{\kappa q^4}{4T}D, \quad \text{Im}\,G_{tr}=\frac{1}{2T}D_{tr}, \qquad (22)$$

which are the consequences of the fluctuation-dissipation theorem. In addition to the harmonic part, the effective action $\mathscr{T}$ contains terms of the third order, which determine the third-order vertices which figure on the diagrams representing the perturbation series for the correlation functions (19) and (20). One can check the relations (22) order by order. Consequently, these relations are valid for the ''dressed'' correlation functions (19) and (20). Note that the relation

$$\int \frac{d\omega}{2\pi}D(\omega,q)=\frac{2T}{\kappa q^4}, \qquad (23)$$

which can be proved by using (22), the analyticity of $G(\omega)$ in the upper half-plane, and the asymptotic law $G(\omega)\approx-\omega^{-1}$, which is correct for large ω. Actually, (23) is a direct consequence of (3), since the integral over frequencies is just the simultaneous correlation function.

Analysis of the diagrams shows that they contain infrared logarithms, which are related to the lines representing the correlation function D in Eq. (20). The lines produce the factors

$$(\nabla_\alpha h(t,\mathbf{r})\nabla_\beta h(t,0)) = \frac{TL}{2\pi\kappa}\delta_{\alpha\beta}, \qquad (24)$$

where $L=\ln[hg^{-1/2}/r]$, and r^{-1} is determined by the characteristic external wave vector of the diagram. The expression (24) can be found from (23) if one recalls condition (5). The presence of the logarithmic contributions implies that the main renormalization of a correlation function like $G(\omega,\mathbf{q})$ is produced by the degrees of freedom with the wave vectors much smaller than q. Therefore, we should extract from the diagrammatic expressions for $G(\omega,\mathbf{q})$ only the contributions corresponding to the interaction with the degrees of freedom.

The program can be realized directly in using the language of the functional integral. Let us separate the variables h, p, $\mathbf{v}$, and $\boldsymbol{\mu}$ into fast parts (with wave vectors larger than q), basic parts (with wave vectors of the order of q), and slow parts (with wave vectors smaller than q). In calculating $G(\omega,\mathbf{q})$ we can forget about the fast parts and keep the interaction of the basic part with the slow part. We then obtain the following expression from (17) and (18):

$$\mathscr{T}=\int dt\, d^2r\left\{p\partial_t h+pv_\alpha m_\alpha+\mu_\alpha\partial_t j_\alpha\right.$$

$$\left.-\frac{\kappa}{2}\mu_\alpha\nabla^4 h m_\alpha-\Gamma\frac{\kappa}{2}p\nabla^6 h+2\eta\mu\hat{q}\mathbf{v}\right\}+\dots, \qquad (25)$$

where h, p, $\mathbf{v}$, and $\boldsymbol{\mu}$ denote the basic parts of the fields, m_α is the gradient of the slow part of h, and the dots designate the irrelevant terms. The action (25) is of the second order over h, p, $\mathbf{v}$, and $\boldsymbol{\mu}$ and, consequently, the integrals over the fields can be taken explicitly. Since $\mathbf{m}$ varies only weakly along the length q^{-1}, we obtain

$$G(\omega,\mathbf{q})=-\langle(\rho h\omega+2i\eta q)\Delta^{-1}\rangle_m, \qquad (26)$$

$$G_{tr}(\omega,\mathbf{q})=-\langle(\omega+i\kappa\Gamma q^6/2)\Delta^{-1}\rangle_m, \qquad (27)$$

$$\Delta=(\rho h\omega+2i\eta q)(\omega+i\kappa\Gamma q^6/2)-\kappa q^4 m_{tr}^2/2, \qquad (28)$$

where

$$m_{tr}^2=\left(\delta_{\alpha\beta}-\frac{q_\alpha q_\beta}{q^2}\right)m_\alpha m_\beta,$$

and the notation $\langle\dots\rangle_m$ means averaging over the statistics of $\mathbf{m}$. In calculating (26) and (27) we substituted $\mathbf{j}=\rho h\mathbf{v}$. Actually, the terms with ρh can be and we omit them below.

In averaging (26) and (27) we can assume that the statistics of $\mathbf{m}$ are Gaussian. The point is that only the simultaneous correlation functions of $\mathbf{m}$ enter the expressions. These functions are described by the harmonic energy (3). The pair correlation function of $\mathbf{m}$ is equal to (24). Therefore,

$$\langle m_{tr}^2\rangle=\frac{TL}{2\pi\kappa},$$

and we find from (26)

$$G(\omega,\mathbf{q})=-\int_{-\infty}^{\infty}\frac{d\mathsf{s}}{\sqrt{2\pi}}\exp(-\mathsf{s}^2/2)$$

$$\times\left(\omega+i\frac{\kappa\Gamma}{2}q^6+i\frac{TL}{8\pi\eta}q^3\mathsf{s}^2\right)^{-1}. \qquad (29)$$

We see that G as a function of the frequency ω has the cut along the imaginary semiaxis, which starts from $\omega = -i\Gamma\kappa q^6/2$ and goes to $-i\infty$. The effective length of the cut can be estimated as Tq^3/η, which is the new characteristic frequency associated with the fluctuations. Let us compare the frequency with the position of the pole in the bare expression:

$$\frac{Tq^3/\eta}{\Gamma\kappa q^6} \sim \frac{g}{(qh_0)^3}. \tag{30}$$

We conclude that the fluctuation effects dominate in the region $g^{1/2} < qh_0 < g^{1/3}$. We can now justify the disregard of $\rho h\omega$ in comparison with ηq in the above expressions. When $qh \sim 1$,

$$\rho h_0\omega/\eta q \sim \rho\kappa/\eta^2 h_0 \sim a/h_0 \ll 1,$$

and at $qh_0 \sim \sqrt{g}$

$$\rho h_0\omega/\eta q \sim \rho\kappa g^2/\eta^2 h_0 \ll 1.$$

Performing Fourier transform of (29) over frequencies, we obtain

$$G(t,\mathbf{q}) = i\left(1 + \frac{TL}{4\pi\eta}q^3 t\right)^{-1/2}\exp\left\{-\frac{\kappa}{2}\Gamma q^6 t\right. \tag{31}$$

The expression (31) is correct for a positive time t. For negative times $G(t) = 0$ due to the causality principle since G is the response function. We see from (31) that in the fluctuation region $g^{1/2} < qh < g^{1/3}$ there appears an intermediate power asymptotics $t^{-1/2}$, which at large times t is changed by the exponential decay. This means that the squeezing mode is described by a dynamic equation, which is nonlocal in time.

The above assertion is correct for the wave vectors $q \gtrsim \sqrt{g}/h_0$. In the limit $qh_0 \ll \sqrt{g}$ we return to the local equation (11) but with the renormalized kinetic coefficient $\widetilde{\Gamma}$. The quantity can be found by integrating the weight $\exp(i\mathscr{I})$ over the degrees of freedom with the wave vectors $q \gtrsim \sqrt{g}/h_0$. The main effect is attributed to the sweeping term in the effective action (17). Because of the integration over the degrees of freedom with the wave vectors $q \gtrsim \sqrt{g}/h$, the term $iT\Gamma(\nabla p)^2$ in (18) for the long-wavelength degrees of freedom is renormalized. We find for the renormalized value

$$\bar{\Gamma} - \Gamma = \frac{1}{4T}\int dt\, d^2r\langle\mathbf{v}(t,\mathbf{r})h(t,\mathbf{r})\mathbf{v}(0,\mathbf{0})h(0,\mathbf{0})\rangle, \tag{32}$$

where averaging is performed over the degrees of freedom with the wave vectors $q \gtrsim \sqrt{g}/h_0$. Using the renormalized expressions for the correlation functions, we obtain the estimate $\widetilde{\Gamma} \sim g^{-1/2}\Gamma \gg \Gamma$.

4. CONCLUSIONS

We demonstrated that fluctuations essentially modify the character of the squeezing mode due to its nonlinear coupling with transverse shear hydrodynamic mode. The fluctuation effects lead to nonlocality of the equation for the mode; the corresponding Green's function is (31). The new characteristic frequency of the mode related to the fluctuations is $\omega \sim Tq^3/\eta$ (q is the wave vector); remarkably, it does

not depend on the bending elasticity. It is important to distinguish the characteristic frequency from the attenuation of the membrane bending mode (8), which has the same q^3 dependence on the wave vector. We stress that the strong fluctuation effects are observed only for dynamics. The static characteristics are not influenced by fluctuations because of the smallness of the coupling constant (2). This is the reason why we need only the harmonic part of the energy (3).

Strong dynamic fluctuations of h occur for the wave vectors $q \gtrsim \sqrt{g}/h$. For smaller wave vectors the fluctuations of h are weak. Nevertheless, even for the wave vectors there is a memory of the region of strong fluctuations, which is the renormalized value of the kinetic coefficient Γ in Eq. (11): The bare value (12) is substituted by $\widetilde{\Gamma} \sim g^{-1/2}\Gamma \gg \Gamma$. Note also that to analyze the dispersion relation of the squeezing mode in the limit $q \ll \sqrt{g}/h$ starting from (11) we should take into account in addition to the energy (5), the steric contribution (4) and the van der Waals (6) contribution to the energy. As a result, we find

$$\omega = -i\widetilde{\Gamma}q^2\left(\frac{9\pi^2 T^2}{64\kappa h^4} + \frac{10Ha^2}{\pi h^6}\right).$$

Let us discuss the possibility of checking our predictions experimentally. The membranes can be studied by a variety of experimental techniques. Lately, laser «tweezers» have become a useful tool for probing dynamical properties of membranes. This technique enables us to obtain direct information about amplitudes and characteristic times of dynamical fluctuations of different objects consisting of membranes. For details see the monography[27] and recent experiments.[29–31] We can also mention force apparatus measurements,[28] which make it possible to investigate dynamical response for two very thin lamellar systems confined between the walls, and the classical light-scattering experiments. Because of relaxation of the membrane fluctuations, the scattered light has a broadened spectral distribution compared to the incident light. Despite the small broadening, the modern technique of light beating (intensity fluctuation spectroscopy) allows one to obtain information about eigenmodes of the system.

The conclusions concerning the renormalization of the squeezing mode, in our opinion, are interesting, both in their own right and as a new test of the membrane fluctuations.

APPENDIX

Elastic modes

Here we consider the elastic modes associated with the relaxation of the surface density of molecules that comprise the two membranes of a double film. To find the dispersion relation for the modes we should start from the elastic energy associated with the variations of the surface density of molecules n_s. In the harmonic approximation the elastic energy of a single membrane is[21]

$$\mathscr{H}_{el}=\frac{1}{2}\int dAB\mathsf{s}^2. \tag{A1}$$

Here

$$\mathsf{s}=(n_s-n_0)/n_0,$$

where n_s-n_0 is the deviation of the surface density of molecules n_s from its equilibrium value n_0, and the coefficient B has the meaning of the inverse compressibility of the membrane. The elastic energy is the sum of terms (A1) for both membranes that constitute the double film.

Let us consider the elastic modes in the linear approximation. We assume that at equilibrium the membranes lie in the planes $z=\pm h_0/2$. The deviations of the membranes from the positions can then be characterized by their displacements $u_{1,2}$ along the z axis. To find the dispersion relations for the modes one should solve conventional hydrodynamic bulk equations supplemented by boundary conditions at the membranes. As we will see, the frequencies of the elastic modes are small compared to sound frequency. Consequently, we can use the convential linearized equations for an incompressible liquid,[32]

$$\nabla_k v_k=0, \quad \left(\frac{\eta}{\rho}\nabla^2-\partial t\right)v_k=\frac{\nabla_k P}{\rho}, \tag{A2}$$

where $k=x,y,z$. Since the membranes are immersed into the liquid, they move with the velocity of the liquid which is continuous near the membranes. The boundary conditions for Eqs. (A2) for a membrane can be found in Refs. 20 and 21. In the linear approximation they are

$$\rho^s\partial_t v_{z\,1,2}+\kappa\nabla_\alpha^4 u_{1,2}=-\lfloor P\rfloor_{1,2}, \tag{A3}$$

$$\partial_t u_{1,2}=v_{z\,1,2}, \tag{A4}$$

$$\rho^s\partial_t v_{\alpha\,1,2}+B\nabla_\alpha\mathsf{s}_{1,2}=\eta\lfloor\nabla_z v_\alpha\rfloor_{1,2}, \tag{A5}$$

$$\partial_t\mathsf{s}_{1,2}+\nabla_\alpha v_{\alpha\,1,2}=0, \tag{A6}$$

where the "floors" designate a jump at the membranes, ρ^s is the two-dimensional mass density of amphiphilic molecules, and subscripts 1 and 2 numerate the membranes. The terms with ρ^s in Eqs. (A3) and (A5) are negligible.

Now, we will solve Eqs. (A2) with the boundary conditions (A3)–(A6) under the assumption that all variables are proportional to $\exp(-i\omega t+iqx)$, where ω is the frequency, and q is the wave vector.

The velocity of the liquid is divided into two parts: potential and solenoidal. The potential component is related to the pressure which obeys the equation

$$(\nabla_z^2-q^2)P=0.$$

The solenoidal component is described by the equation

$$-i\rho w v=\eta(\nabla_z^2-q^2)v.$$

Thus we can explicitly write the solutions of the equations inside and outside the film in terms of the velocity of the membranes. The expressions are slightly different for the symmetric ($\mathsf{s}_1=\mathsf{s}_2$) and the antisymmetric ($\mathsf{s}_1=-\mathsf{s}_2$) cases. Using the solutions in bulk, we can express the jumps in Eqs. (A3) and (A5) in terms of v_x and v_z on the membranes. As a

result, we find a linear system for v_x and v_z. We can then write the condition for the existence of nontrivial solutions of the system, which for the symmetric case in the simplified form is

$$V^2\left(\coth\frac{Vqh_0}{2}-\coth\frac{qh_0}{2}\right)^2-\left[V^2+(V+\beta)\right.$$
$$\times\left(V\coth\frac{Vqh_0}{2}-\coth\frac{qh_0}{2}\right)\right]$$
$$\times\left[V\coth\frac{qh_0}{2}\coth\frac{Vqh_0}{2}\right.$$
$$\left.+\left(V\coth\frac{Vqh_0}{2}-\coth\frac{qh_0}{2}\right)\right]=0. \tag{A7}$$

Here we introduce the notation

$$V=\sqrt{-i\frac{\omega\rho}{\eta q^2}}, \quad \beta=i\frac{Bq}{\eta\omega},$$

and suggest that in the case of elastic modes one deals with the frequency $\omega\gg\eta q^2/\rho$. For the antisymmetric case the condition can be obtained from (A7) by substituting $\coth\to\tanh$.

It is difficult to find the dispersion relations from (A7) and the analogous equation for the antisymmetric case in a general situation. Below we consider two different limiting cases and assume that

$$\frac{\eta^2}{Bph_0}\ll 1, \tag{A8}$$

which is natural since h_0 is much larger than the molecular length for real films. First, we consider the short-wavelength limit

$$qh_0\gg\left(\frac{\eta^2}{B\rho h_0}\right)^{1/2}. \tag{A9}$$

We then obtain the same dispersion relation as for the elastic mode of a single membrane[33,21]

$$\omega=\frac{\pm\sqrt{3}-i}{2}\left(\frac{B^2}{4\eta\rho}\right)^{1/3}q^{4/3}. \tag{A10}$$

This means that the thickness of layers near the membranes, where the hydrodynamic motion occurs, is much less than h_0 and, consequently, the membranes can be assumed to be nearly independent in this case. Note that due to (A8), the condition (A9) is compatible with $qh_0\ll 1$, where the membranes cannot be regarded as independent in considering, say, the squeezing mode. Therefore, one should be careful: under the condition (A9) the membranes can be treated as nearly independent only in examining the elastic modes. In the opposite long-wavelength limit,

$$qh_0\ll\left(\frac{\eta^2}{B\rho h_0}\right)^{1/2}, \tag{A11}$$

we deal with two different dispersion relations. In the symmetric case the dispersion relation is

$$\omega = \frac{\pm\sqrt{3}-1}{2}\left(\frac{B^2}{\eta\rho}\right)^{1/3} q^{4/3}. \qquad (A12)$$

This is the same dispersion relation as (A10) but with the doubled membrane elasticity, which is natural for the double film. In the antisymmetric case the dispersion relation is

$$\omega = -i\,\frac{Bq^2 h_0}{2\eta}. \qquad (A13)$$

Thus we encounter a simple diffusion.

The dispersion laws (A10), (A12), and (A13) show that the frequencies of the elastic modes are small in comparison with the sound frequency cq (where c is the sound velocity), which justifies our using the incompressible hydrodynamic equations (A2). Note also that for the mode (A13) the condition (A8) ensures the inequality $\omega \gg \eta q^2/\rho$, which was suggested in the derivation of the relation (A7) (the inequality enables us to disregard the potential part of the velocity). Thus, the condition (A8) makes our scheme self-consistent.

In the long-wavelength limit the double membrane film can be treated as an effective single membrane. This effective membrane should be framed by hydrodynamic variables, which give the information about the ''microscopic'' construction of the double membrane film. In other words, one should incorporate into the set of ''macroscopic'' variables the surface densities (described by s_1 and s_2) of molecules, which comprise the two membranes, and the two-dimensional mass density ρh of the liquid between the membranes. The dynamic equation for the variable has been derived in the main text of this paper [see Eq. (11)]. The phenomenological dynamic equations for s_1 and s_2 can be derived in the same manner as the Poisson brackets for the density of any conserved scalar quantity, the same structure of the Poisson bracket as (10).[18] In terms of the variables $s_+ = (s_1 + s_2)/2$ and $s_- = s_1 - s_2$ the equations are

$$\partial_t s_+ = -\nabla_\alpha[(1+s_+)v_\alpha] + D_+ \nabla^2 s_+, \qquad (A14)$$

$$\partial_t s_- = -\nabla_\alpha(s - v_\alpha) + D_- \nabla^2 s_-. \qquad (A15)$$

Here we discarded the bending motion. The system of equations (A14) and (A15) should be supplemented by Eq. (14) in the main text of the paper, where

$$P_s = 2B s_+. \qquad (A16)$$

In analyzing the elastic degrees of freedom we should use the following expression for the momentum flow from the bulk:

$$J_\alpha = -2\sqrt{i\,\eta\rho\hat\omega}\,v_\alpha, \qquad (A17)$$

where the velocity v_α is implied to be longitudinal, since just the longitudinal component of the velocity is involved in the elastic motion. We stress that the expression (A17) is correct if $\omega \gg \eta q^2/\rho$, in contrast with the applicability condition of (15).

Linearizing the system of equations (14), (A14), and (A15) (and ignoring the squeezing degrees of freedom), we find the dispersion relations (A12) and $\omega = -iD_- q^2$. The term with D_+ in (A14) appears to be irrelevant. Comparing the dispersion relation $\omega = -iD_- q^2$ with (A13), we obtain

$$D_- = Bh_0/2\eta. \qquad (A18)$$

It can then be verified that the linear coupling between the elastic and the squeezing degrees of freedom described by the term with κ in (14) is negligible. The nonlinear terms in Eqs. (14), (A14), and (A15) lead to the interaction of different modes. Explicit analysis shows that the fluctuation effects do not affect appreciably the linear dispersion relations (A10), (A12), and (A13) due to $g \ll 1$. The same holds for a nonlinear interaction with the bending degree of freedom, which (because of the same inequality $g \ll 1$) does not change the results obtained in the linear approximation.

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