Dynamic fluctuation effects in dilute lyotropic systems

E. I. Kats and V. V. Lebedev

L. D. Landau Institute for Theoretical Physics, Academy of Sciences of Russia, 117940, GSP-1, Moscow V-334,
Kosygina 2, Russia
and Department of Physics, Weizmann Institute of Science 76100, Rehovot, Israel
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Dynamical phenomena in dilute lyotropic solutions are investigated. We consider the case when lyotropic molecules form a system of membranes determining the main peculiarities of these systems. We are interested in the effect of fluctuations of the membranes on dynamical characteristics of the systems. A membrane possesses two soft degrees of freedom associated with its bending deformations and with variations of the surface density n_s of molecules constituting the membrane, which we term elastic deformations. Bending fluctuations are governed by the Helfrich module κ whereas the elastic fluctuations (variations of n_s) are governed by the "internal" compressibility of the membrane which we characterize by an elastic module B. Correspondingly, there are two characteristic surface modes where the motion of the solvent is localized near the membrane. In the linear approximation these modes prove to be overdamped. Due to the softness of these modes, dynamic fluctuation effects related to nonlinear interaction of the modes should be taken into account. To investigate these effects the Wyld-diagram technique is used. It is constructed on the basis of nonlinear equations describing the dynamics of a membrane immersed into a liquid. We have shown that bending fluctuations are most important. Our investigation is performed in the framework of the perturbation theory in $g \simeq T/\kappa$ which is believed to be a small dimensionless parameter, describing the "strength" of the bending fluctuations. These fluctuations produce only small logarithmic corrections to the characteristics of the bending mode whereas they essentially modify the dynamical behavior of the elastic degree of freedom. Namely, these fluctuations drastically change the frequency dependence of the susceptibility describing the relaxation of n_s to the equilibrium. We consider also the influence of dynamical fluctuations of membranes on the macroscopic characteristics of the system such as viscosity coefficients. To find the fluctuation contribution η_{fl} to the viscosity coefficients, we calculate a response of the system of membranes to a macroscopic (long-wavelength) motion of the liquid, with fluctuations of the membranes taken into account. The main part of η_{fl} is associated with the elastic part of the membrane stress tensor but is strongly renormalized by the bending fluctuations. The quantity η_{fl} possesses a complicated frequency dispersion. In the high-frequency region it behaves as $\omega^{-5/3}$, in the intermediate frequency region it behaves as $\omega^{-1/3}$, and for small ω it remains constant. The ratio of this constant to the viscosity of the solvent is of the order of g^{-1}, i.e., η_{fl} exceeds the viscosity of a pure solvent.

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INTRODUCTION

In recent years considerable attention has been paid to the investigation of dilute lyotropic phases. Equilibrium (static) properties of these phases have been extensively examined (see, e.g., books [1–3] and reviews [4–6]), but research of dynamical characteristics (such as viscosity, spectrum of eigenmodes, etc.) is unfortunately in a rather primitive stage. It is largely accounted for by a complexity of dynamical phenomena in these systems. The present work is devoted to the theoretical investigation of dynamical properties of lyotropic systems which prove to be sensitive to nonlinear fluctuation processes.

The most distinctive property of amphiphilic molecules is their ability to spontaneously self-assemble into aggregates of various shapes. This polymorphism is indeed a source of a very rich variety of ordered phases. In our paper we consider particular situations where molecules spontaneously self-assemble into membranes which are bilayers of a thickness of the order of a molecular length.

As a rule, these bilayers are built up from two identical monolayers, oriented opposite each other. Hence a membrane is locally symmetrical with respect to its midsurface.

At present different structures of lyotropic membrane systems are identified in experiments. Lamellar phases exhibit smectic order, cubic phases possess three-dimensional long-range translational order, and sponge or disordered phases show no long-range positional order. Besides, solutions of vesicles or of fragments of membranes are observed. Each type of these structures may be characterized by a typical length scale ξ. For lamellar phases it is an intermembrane distance, for cubic phases it is a period of the structure, for solutions it is a characteristic size of vesicles or of fragments of membranes, and for sponge phases the role of ξ is played by the characteristic size of pores of the "sponge." On scales smaller than ξ all the phases may be regarded as systems of independent membranes possessing a flat shape at zero approximation.

As was shown by Porte *et al.* (see, e.g., [5]) the statis-

tics of dilute membrane solutions in the main approximation is scale invariant (since Helfrich's energy is invariant with respect to any isotropic dilation [7]). It means, for example, that the free energy has to be proportional to ϕ^3, where ϕ is the membrane volume fraction. Fluctuation corrections break up the scale invariance, since each change in scale implies a renormalization of membrane parameters. But in static the renormalization is only logarithmic and hence it is weak. More dramatic are consequences of fluctuation effects in dynamics, since fluctuation contributions to the dynamical coefficients stronger depend on scale.

It proves that a separately taken membrane has two characteristic modes where the motion of the liquid is localized near the membrane. One of these modes is related to the relaxation of the shape of a membrane towards a flat shape. We will term this mode a bending mode. It occurs instead of conventional capillary waves, propagating over the surface with nonzero surface tension. The other characteristic mode is related to the relaxation of the surface density of molecules, constituting the membrane. We will term this mode an elastic mode, since it is related to variations of the density of the membrane. This mode can be interpreted as a surface sound, overdamped due to the bulk shear motion. The said above in this paragraph is valid for isotropic liquid membranes. Anisotropic and crystalline membranes need separate consideration, and these objects are beyond the scope of our paper.

In the present work we are interested in fluctuation effects induced by a nonlinear interaction of modes, associated with a membrane. These effects should be considered in the framework of the mode-coupling theory. We examine also the influence of bending and elastic fluctuations on the dynamics of long-wavelength degrees of freedom. These effects prove to be important due to the softness of the bending mode. In many respects the situation resembles the critical dynamics (see, e.g., review [8]) associated with a critical soft mode. To take into account dynamic fluctuation effects we may first study only self-interaction of this soft mode and then include the interaction of this mode with other degrees of freedom. For orientational phase transitions in liquid crystals such a program was performed in [9].

Our paper is organized as follows.

Section I covers static properties of a separately taken membrane. As is well known, thermal fluctuations of a membrane lead to a logarithmic renormalization of different parameters, also discussed in this section. Dynamic fluctuation effects prove to be stronger than static effects. To study these effects we must utilize nonlinear dynamical equations of a membrane, derived in Sec. II. Using these nonlinear equations we construct an effective action, enabling us to represent dynamical correlation functions in terms of functional integrals. Such a representation generates a diagram technique of the Wyld type [10]. The effective action for systems of membranes is derived in Sec. III.

Further we study fluctuation contributions to observable physical characteristics. To do this we perform an expansion of the effective action in variables, describing

surface degrees of freedom (Sec. IV). The renormalization of the bending mode is studied in Sec. V. It turns out that fluctuation corrections to the dispersion law of the bending mode are logarithmic and therefore small in the region of scales under study. The influence of the fluctuations on the elastic degree of freedom is discussed in Sec. VI. The dynamics of the elastic degree of freedom is essentially modified by fluctuations.

Finally, the fluctuation contribution to the viscosity is calculated (Sec. VII). The main fluctuation contribution is related to fluctuations of membranes with wave vectors $q \geq \xi^{-1}$. Therefore fluctuation effects manifest themselves explicitly in a relatively high-frequency (but reasonable for measurements) region, but the low-frequency viscosity will, of course, also be influenced by these fluctuations. Let us stress that in spite of the fact that the lyotropic systems are very dilute, fluctuations of membranes lead to an important contribution to viscosity coefficients. In this respect dynamical fluctuations drastically differ from static fluctuations which in this region of parameters give only small logarithmic corrections to thermodynamical properties. In the Conclusion we summarize and discuss briefly the results of our work.

There are three appendixes to our paper devoted to bulky computations. The problem is that any dynamical process in the membrane induces a motion of the liquid surrounding the membrane. Therefore at the investigation of eigenmodes one must solve the overall system of equations, including conventional hydrodynamical equations. The solution of the equations expressed through the surface value of the velocity should be utilized to determine the right-hand side of the equation for the surface momentum density, which enables us to close the system of equations for the surface variables. In the three appendixes we collected these technical (but important for the theoretical investigation of dynamical properties of lyotropic systems) details.

Some preliminary results concerning fluctuation contributions to the viscosity have been published in our paper [11].

I. THERMODYNAMICS OF A MEMBRANE

In this section we will discuss static properties of a free membrane. As was explained in the Introduction, we may apply results of this investigation to dilute lyotropic systems at scales smaller than the characteristic scale ξ.

The energy related to variations of a shape of the membrane can be written in the form proposed by Canham [12] and Helfrich [7]

$$E_s = \int dS \left(\frac{\kappa}{2} (R_1^{-1} + R_2^{-1})^2 + \bar{\kappa} R_1^{-1} R_2^{-1} \right). \quad (1)$$

Here R_1 and R_2 are local radii of the membrane curvature and the coefficients $\kappa, \bar{\kappa}$ are elastic modules. The quantity $R_1^{-1} R_2^{-1}$ is the Gaussian curvature of the membrane and the combination $R_1^{-1} + R_2^{-1}$ is referred to as its mean curvature. The term (1) may be called a bending energy. Since we consider bilayer membranes, the bending energy of the membrane must be invariant to the

change of the sign of the curvature, and therefore terms odd in radii R_1, R_2 are missing from the expansion of E_s, which particularly explains the absence of the term with the spontaneous curvature in (1).

One of the conditions of the equilibrium of a free membrane with the solution of monomers, constituting the membrane, is the zero value of its surface tension. For lyotropic systems under study the surface tension of membranes may be finite as a consequence of a long-range interaction between membranes. But on the relatively short scales we are interested in, the role of this surface tension is negligible. Therefore we have omitted the corresponding term in expression (1).

In addition to the energy (1) one must also take into account the "elastic" energy, related to variations of the surface density of molecules n_s, constituting the membrane. In the approximation we need, such a contribution to the energy reads

$$E_n = \frac{1}{2} \int dS B \varsigma^2. \qquad (2)$$

Here

$$\varsigma = (n_s - n_0)/n_0, \qquad (3)$$

where $n_s - n_0$ is a 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.

When the surface tension coefficient of the membrane is negligible, thermal fluctuations of the shape of the membrane are relevant. As was noted by de Gennes and Taupin, these fluctuations give rise to the destruction of a correlation between orientations of sufficiently distant pieces of the membrane [13]. The scale, starting from which this destruction happens, is called a persistent length ξ_p. A membrane of sizes exceeding the persistent length cannot in any approximation be treated as flat.

These fluctuations also lead to the logarithmic renormalization of the modules κ and $\bar{\kappa}$ and of other quantities, characterizing the surface energy of the membrane. First, an attempt to calculate the renormalization of the module κ was taken by Helfrich [14] and later by Förster [15]. The correct renormalization-group (RG) equation for the module κ in the one-loop approximation was derived by Peliti and Leibler [16], Kleinert [17], and Polyakov [18] and the equations for $\bar{\kappa}$ and for the spontaneous curvature in the same approximation were found by Kleinert [19]. Besides, the module B, introduced by (2), is logarithmically renormalized.

The explicit form of the one-loop RG equations for the introduced quantities is

$$\frac{d\kappa}{dL} = -\frac{3T}{4\pi}, \qquad (4)$$

$$\frac{d\bar{\kappa}}{dL} = \frac{5T}{6\pi}, \qquad (5)$$

$$\frac{dB}{dL} = \frac{TB}{4\pi\kappa}. \qquad (6)$$

Here T is the temperature (expressed in energy units) and $L = \ln(r/a_m)$, where r is a characteristic scale and a_m is a cutoff parameter (the length of the order of the molecular size). These equations describe the behavior of the modules $\kappa, \bar{\kappa}, B$ at a variation of the characteristic scale r.

The right-hand sides of the renormalization-group equations, found in the framework of the perturbation theory, always contain the module κ but not $\bar{\kappa}$. The problem is that the term in the energy (1), proportional to the module $\bar{\kappa}$ (i.e., the surface integral of its Gaussian curvature), is a topological invariant, therefore it is not changed at small perturbations of the shape of the membrane.

As follows from Eq. (4), the role of the "invariant charge" (dimensionless coupling constant) is played by the quantity

$$g = \frac{3T}{4\pi\kappa}. \qquad (7)$$

For the perturbation theory to be applicable and, consequently, for Eqs. (4)–(6) to hold, the coupling constant g must be small. Henceforth we will treat g as a small quantity, enabling us to employ the perturbation theory.

A consequence of Eq. (4) is an expression

$$g = \frac{g_0}{1 - g_0 L}. \qquad (8)$$

Here g_0 is the short-wavelength coupling constant. We see that with increasing scale (i.e., with increasing L) the coupling constant grows. Thus we come to a situation which in the quantum field theory is called asymptotic freedom. The scale on which g reaches a value of the order of unity, and the perturbation theory becomes inapplicable, is no other than the persistent length ξ_p. Using (8) we may derive an estimation

$$\xi_p \sim a_m \exp(g_0^{-1}). \qquad (9)$$

Note the exponential character of the dependence of ξ_p on g_0.

The static behavior of the modules $\kappa, \bar{\kappa}, B$ at increasing scale is determined by Eqs. (4)–(6) from which it follows that at increasing scale the module κ becomes smaller, whereas the modules $\bar{\kappa}, B$ grow together with the coupling constant. Using the relation (7) we find the following first corrections:

$$\Delta\kappa = -g_0\kappa L, \quad \Delta B = \frac{1}{3}g_0 B L. \qquad (10)$$

One can say that due to thermal fluctuations the membrane becomes less compressible and softer with respect to fluctuations of the shape.

II. DYNAMIC EQUATIONS

To formulate equations describing dynamics of a membrane it is necessary to introduce a certain parametrization, setting its position in space. In this section we will use a general parametrization demonstrating the explicit

rotational invariance of the equations. Namely, we will assume that the position of the membrane is set by an equation

$$\Phi(\mathbf{r}) = 0, \qquad (11)$$

where Φ is a function of three coordinates.

All expressions presented below have to be invariant under a transformation

$$\Phi \to f(\Phi), \qquad (12)$$

where f is an arbitrary function satisfying the conditions

$$f(0) = 0, \quad f'(0) \neq 0. \qquad (13)$$

The transformation (12) does not influence the position of the membrane in space and, consequently, cannot change any physical quantity. For example, the expression for the unit vector, normal to the membrane

$$l_i = \frac{\nabla_i \Phi}{|\nabla \Phi|}, \qquad (14)$$

is invariant with respect to the transformation (12).

The vector l_i is formally defined in the whole space. It is a general feature of all surface quantities (e.g., the surface density of molecules n_s) which are introduced below. Formally they will be considered as functions of all three coordinates, whereas they are meaningful, of course, only on the surface (11).

Nonlinear equations, describing the dynamics of the membrane in terms of Φ, are presented in the work by Lebedev and Muratov [20]. They can be derived as equations of dynamics of a Langmuir film (see the work by Kats and Lebedev [21]). We give here the ultimate form of these equations without deriving them in detail.

The dynamical equation for the quantity Φ, setting the position of the membrane, has the form

$$\frac{\partial \Phi}{\partial t} = -v_{si} \nabla_i \Phi, \qquad (15)$$

where $\mathbf{v}_s$ is a velocity of the membrane which should coincide with the velocity of a liquid near the membrane. This fact is a consequence of correct boundary conditions of hydrodynamical equations on the membrane [21,20]. Let us stress that derivatives of the velocity (as we will see below) have jumps on the membrane. The meaning of Eq. (15) is that the membrane moves together with the liquid, surrounding it. From Eq. (15) and from the relation (14) the following equation for the unit vector l ensues:

$$\frac{\partial l_i}{\partial t} = -v_{sk} \nabla_k l_i - l_k \nabla_i^\perp v_{sk}. \qquad (16)$$

Here and henceforth we utilize designations

$$\delta_{ik}^\perp = \delta_{ik} - l_i l_k, \qquad \nabla_i^\perp = \delta_{ik}^\perp \nabla_k. \qquad (17)$$

Note that not only the surface quantities such as $l_i, \mathbf{v}_s$ figure in the right-hand side of Eq. (16), but also three-dimensional derivatives. Nevertheless one can check that this relation does not depend on the concrete form of a

continuation of functions l and $\mathbf{v}_s$ from the membrane into the third dimension. Any redefinition of this continuation implies the transformation of the following kind:

$$l_i(\mathbf{r}) \to l_i(\mathbf{r} + \mathbf{R}(\Phi)),$$

where functions $\mathbf{R}(\Phi)$ possess the same properties (13) as the function f in (12). Bearing in mind Eq. (15) for Φ it is not difficult to prove that at such redefinition of l and $\mathbf{v}_s$ Eq. (16) does not change its form. Therefore Eq. (16) is indeed the equation for the surface variable. The same statement is valid for all equations presented below in this section.

The dynamical equation for the surface density of molecules n_s of the membrane can be written as

$$\frac{\partial n_s}{\partial t} = -v_{si} \nabla_i n_s - n_s \nabla_i^\perp v_{si}. \qquad (18)$$

Using Eq. (15) we may rewrite Eq. (18) as

$$\frac{\partial (n_s \mid \nabla \Phi \mid)}{\partial t} = -\nabla_i (\mid \nabla \Phi \mid n_s v_{si}). \qquad (19)$$

Equation (19) has the meaning of the conservation law of the number of molecules, constituting the membrane. Therefore this equation becomes inapplicable in the frequency range, where the exchange of molecules between the membrane and the liquid surrounding it becomes relevant. Apparently this range is beyond our consideration.

The dynamical equation for the surface momentum density of the membrane $\mathbf{j}_s = \rho_s \mathbf{v}_s$ (where ρ_s is the surface density of mass) is

$$\frac{\partial (\mid \nabla \Phi \mid j_{si})}{\partial t} + \nabla_k \left[\mid \nabla \Phi \mid (\rho_s v_{si} v_{sk} + T_{sik}) \right]$$
$$= -\lfloor P \rfloor \nabla_i \Phi - \lfloor \Pi_{ik} \rfloor \nabla_k \Phi. \qquad (20)$$

Here T_{sik} is the surface stress tensor, P is the bulk pressure of the liquid, and Π_{ik} is its viscous stress tensor. Namely,

$$\Pi_{ik} = -\eta \left(\nabla_i v_k + \nabla_k v_i - \frac{2}{3} \delta_{ik} \nabla \mathbf{v} \right) - \zeta \nabla \mathbf{v} \delta_{ik}, \qquad (21)$$

where η and ζ are coefficients of the first and second viscosities. The "floors" in (20) designate the jump of the corresponding variable on the membrane. For example, $\lfloor P \rfloor$ is the difference of values of the pressure "above" and "below" the membrane, with the vector l marking the "top" of the membrane.

An expression for the surface stress tensor T_{sik} entering Eq. (20) was derived in the work by Lebedev and Muratov [20] by means of the Poisson brackets method. We will cite the explicit form of the surface stress tensor, obtained from expressions (1) and (2) for the main contributions to the surface energy of the membrane

$$T_{sik} = \left(B\varsigma - \frac{\kappa}{2} (\nabla_n l_n)^2 \right) \delta_{ik}^\perp$$
$$+ \kappa \nabla_n l_n \nabla_i^\perp l_k - \kappa l_i \nabla_k^\perp \nabla_n l_n. \qquad (22)$$

For the derivation of (22) the quantities B, κ were regarded as constants, and the quantity ς is defined by (3).

The stress tensor T_{sik} given by (22) is not symmetric. Therefore a special concern is to formulate the angular momentum conservation law. Formally the divergence of the tensor figuring in Eq. (20) can be rewritten as a divergence of a symmetric tensor. Unfortunately the expression for this symmetric tensor proposed in the work by Lebedev and Muratov [20] proves to depend not only on the position of the membrane, but also on the function Φ. Nevertheless the angular momentum conservation law can be formulated in a form invariant with respect to the choice of the function Φ. Let us demonstrate this.

It is not difficult to check that for any function A it is

$$\epsilon_{jik} \mid \nabla\Phi \mid l_i\nabla_k^\perp A = \epsilon_{jik}\nabla_k(\mid \nabla\Phi \mid l_iA). \qquad (23)$$

The nonsymmetric part of the stress tensor (22) has just the form $l_i\nabla_k^\perp A$, and therefore the angular momentum conservation law may be written in the form

$$\frac{\partial}{\partial t}\left(\epsilon_{jni}r_n j_{si} \mid \nabla\Phi \mid\right) + \nabla_n\left(\epsilon_{jni} \mid \nabla\Phi \mid \kappa l_i\nabla_k l_k\right)$$
$$+ \nabla_k\left(\epsilon_{jni}r_n(T_{sik} + \rho_s v_i v_k) \mid \nabla\Phi \mid\right)$$
$$= -\epsilon_{jni}r_n(\lfloor P\rfloor\nabla_i\Phi + \lfloor\Pi_{ik}\rfloor\nabla_k\Phi). \qquad (24)$$

On the right-hand side of (24) there is a torque applied to the membrane from the liquid. The explicit expression for this torque was derived by substitution of the right-hand side of Eq. (20).

These equations can be employed to study the eigenmodes of a membrane. The linear dispersion law of the bending mode [see (50) below] was derived first in [22]. The linear dispersion law of the elastic mode [see (54) below] is analogous to the one for Langmuir films [21]. The results for a membrane may be deduced from the results for micelles in the limit of zero spontaneous curvature (see the work by Lebedev [23]).

III. EFFECTIVE ACTION

To investigate dynamic fluctuation effects we will make use of the diagram technique, especially adapted for hydrodynamical systems. Such diagram technique was first developed by Wyld [10], who studied velocity fluctuations in a turbulent fluid. The next step was made in the work by Martin, Siggia, and Rose [24], where the Wyld technique was generalized for a broad class of dynamical systems. The textbook description of the diagram technique can be found in the monograph by Ma [25]. The diagram technique may be formulated in terms of path integrals as was first suggested by de Dominicis [26] and Janssen [27]. We will use this representation in our work.

In the framework of this approach apart from hydrodynamical variables φ_a one should introduce auxiliary fields p_a "conjugated" to the hydrodynamical variables φ_a. Then dynamical correlation functions of the hydrodynamic variables can be presented as functional integrals over both types of fields–conventional and auxiliary. These integrals are taken with the weight $\exp(iI)$, where I is an effective action, which is a functional of both the hydrodynamical and auxiliary variables. Namely, the

pair correlation function is

$$\langle\varphi_a\varphi_b\rangle = \int D\varphi\,Dp\,\exp(iI)\varphi_a\varphi_b. \qquad (25)$$

We will use a special designation for this correlation function

$$D_{ab}(t_1 - t_2, \mathbf{r}_1 - \mathbf{r}_2) = \langle\varphi_a(t_1, \mathbf{r}_1)\varphi_b(t_2, \mathbf{r}_2)\rangle. \qquad (26)$$

It is also useful to introduce a correlation function

$$G_{ab}(t_1 - t_2, \mathbf{r}_1 - \mathbf{r}_2) = \langle\varphi_a(t_1, \mathbf{r}_1)p_b(t_2, \mathbf{r}_2)\rangle, \qquad (27)$$

where the average is defined as in (25). The function G is the response function (or the generalized susceptibility) of the system under study, and therefore G is equal to zero at $t_1 - t_2 < 0$. In the following we will present expressions for the function $G(\omega, \mathbf{q})$ (where ω is a frequency and $\mathbf{q}$ is a wave vector), which is (27) in the Fourier representation. The function $G(\omega)$ is analytical in the upper ω half plane and its singularities in the lower ω half plane determine dispersion laws of eigenmodes of the system. At the thermal equilibrium the functions G and D are related by the fluctuation-dissipation theorem. Note that the third pair correlation function $\langle p_a p_b\rangle$ is identically equal to zero.

The effective action I is constructed on the basis of nonlinear hydrodynamical equations of the system. As was noted by de Dominicis and Peliti [28], apart from the terms directly related to the equations, in the effective action I there appears a logarithm of a functional determinant which may also be represented as an integral over auxiliary Fermi fields (see the papers by Feigelman and Tsvelik [29] and Lebedev, Sukhorukov, and Khalatnikov [30], and our book [31]). It can be demonstrated that for the system under consideration the determinant is equal to unity because of the causality properties of the Green's function G. Therefore we will omit the corresponding term in the effective action.

The representation (25) of the correlation function enables us to develop a diagram technique where bare values of the correlation functions are determined by the linearized hydrodynamical equations, and the interaction vertices are related to the nonlinear terms in the equations. A textbook description of the functional integration methods can be found in the monograph by Popov [32].

The dynamics of a pure liquid is described by the conventional hydrodynamical equations (see, e.g., the volume [33] by the Landau-Lifshitz course). The corresponding part of the effective action is represented as

$$I = -\int dt\,d^3r\left(\nabla_k p_i(P\delta_{ik} + \rho v_i v_k + \tilde{\Pi}_{ik}) + \frac{\partial p_i}{\partial t}\rho v_i\right), \qquad (28)$$

where ρ is the mass density of the liquid, $\mathbf{v}$ is its velocity, $\mathbf{p}$ is the auxiliary field conjugated to the momentum density of the liquid $\rho\mathbf{v}$, P is the pressure, and

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$$\tilde{\Pi}_{ik} = -\eta \left(\nabla_i \tilde{v}_k + \nabla_k \tilde{v}_i - \frac{2}{3} \delta_{ik} \nabla \tilde{\mathbf{v}} \right) - \zeta \nabla \tilde{\mathbf{v}} \delta_{ik}. \quad (29)$$

Here η and ζ are coefficients of the first and second viscosity and

$$\tilde{v}_i = v_i + iTp_i, \quad (30)$$

where T is the temperature of the system (expressed in energy units). This combination will often appear in the following sections.

The form of the effective action of the liquid in the presence of membranes immersed in the liquid was found in the work by Lebedev and Muratov [20]. The method of its construction is close to the one for Langmuir films (see the work by Kats and Lebedev [21]). We will use the results of noted works referring the reader interested in their derivation to the original papers.

The contribution to the effective action supplied by the membrane is

$$I_s = - \int dt \, d^3r \, \delta(\Phi) \mid \boldsymbol{\nabla}\Phi \mid \left(\nabla_k p_i T_{sik} + \frac{\partial p_i}{\partial t} \rho_s v_i \right). \quad (31)$$

The variables figuring here were introduced in Sec. II; we have omitted in (31) the term nonlinear in $\mathbf{v}$. Although formally there is a three-dimensional integral in expression (31), I is indeed determined by the surface integral due to the presence of the factor $\delta(\Phi)$ in the integrand. The latter term in expression (31) may be neglected. The problem is that this term is proportional to the frequency of fluctuations and, consequently, is small in comparison with the first term in (31) for eigenmodes of the membrane.

In the study of the dynamics of the membrane, apart from the effective action (31), we have to take into account the term attributed to the bulk motion of the liquid. This contribution appears as a boundary term after exclusion of the bulk degrees of freedom from the action (28). As was demonstrated in the work by Lebedev and Muratov [20], this term is equal to

$$I_{b-s} = - \int dt \, d^3r \, \delta(\Phi) \mid \boldsymbol{\nabla}\Phi \mid$$
$$\times l_k(\rho p_k \lfloor \partial \tilde{\psi}/\partial t \rfloor + \eta p_i \lfloor \nabla_k \tilde{v}_i + \nabla_i \tilde{v}_k \rfloor), \quad (32)$$

where the floors, as in (20), designate jumps of the bulk values on the membrane and $\tilde{\psi}$ is the potential of the variable $\tilde{\mathbf{v}}$ (30), i.e., the potential part of $\tilde{\mathbf{v}}$ is equal to $\boldsymbol{\nabla}\tilde{\psi}$.

To exploit the contribution (32) for the description of the surface dynamic phenomena we should express all quantities entering (32) through their surface values and derivatives of the values only *along* the surface. To do it we have to use a solution of the bulk dynamical equations which are the extremum conditions for the bulk effective action (28). This problem is considered in Appendixes B and C.

IV. EXPANSION IN U

In what follows we will consider a piece of a membrane, which may be treated as approximately flat. We will believe also that in the equilibrium it is arrayed along the X-Y plane. In this case deviations of the shape of the membrane from the equilibrium shape can be characterized by a displacement $u(x, y)$ of the membrane along the Z direction. The corresponding function Φ can be written in the form

$$\Phi = z - u(x, y), \quad (33)$$

since the condition $\Phi = 0$ sets the surface $z = u(x, y)$.

We will derive results concerning the fluctuation dynamics of the membrane in the framework of the perturbation theory in u. It is a bulky procedure and to make the text more clear we formulate auxiliary results in the Appendixes. In Appendix A we present the expressions derived in the preceding sections in terms of surface variables, assumed to be functions of x, y like the displacement u. In Appendix B we develop a technique enabling us to express three-dimensional derivatives entering the effective action in terms of surface quantities. In Appendix C we present integral relations for $\tilde{\mathbf{v}}$, $\tilde{\psi}$, which we need to get expressions for these derivatives.

To investigate dynamic fluctuation effects we have to calculate the integrals of the type figuring in the definition of the pair correlation function (25). Therefore we should find the principal terms of the expansion of (A10) and (A11) in u.

First, we examine the surface effective action (A10). Taking into account the explicit form (A2) for the components of l and keeping only the principal nonlinear terms (up to the third order in u) one can find

$$I_s = \int dt \, dx \, dy \left[-Bp_l \nabla^2 \varsigma + Bp_\| \varsigma \nabla^2 u \right.$$
$$+ \kappa p_\| \left(\nabla^4 u - \frac{1}{2} (\boldsymbol{\nabla} u)^2 \nabla^4 u \right.$$
$$\left. \left. - \frac{1}{2} \nabla^2 \nabla_\gamma \left[\nabla_\gamma u (\boldsymbol{\nabla} u)^2 \right] \right) \right]. \quad (34)$$

The quantities $p_\|, p_l$ are defined by (A8) and (A9). Here and henceforth Greek subscripts designate components of the vectors along the X and Y axes. Note the symmetry of this expression with respect to the transformation $u, p_\| \to -u, -p_\|$.

The variables u and ς figuring in expression (34) should be regarded as related to $v_\|, v_l$ by the dynamical equation (A6)

$$\frac{\partial u}{\partial t} = v_\|, \quad (35)$$

following from (15) and by the equation following from (18) and rewritten as (A7). With the accuracy which we need, the latter equation acquires the form

$$\frac{\partial \varsigma}{\partial t} = -\nabla^2 v_l + v_\parallel \nabla^2 u. \tag{36}$$

At the derivation of (36) we have used the definition (3) and have omitted the nonlinear term in the left-hand side of Eq. (A7). The quantities $v_\parallel$ and v_l are defined by (A4) and (A5).

The term (A11) supplied by a bulk motion contains the derivative of the quantity $\tilde{v}$ in the direction perpendicular to the membrane. Therefore the term (A11) should be primarily expressed through the surface variables and their derivatives along the membrane.

First we will consider a region of low frequencies

$$\omega \ll \eta q^2/\rho,$$

where q is a characteristic wave vector. The expressions for jumps of the derivatives entering (A11) in this region are determined by (B19)–(B21). Using these relations we can find the expansion of (A11) in u up to the fourth order. The second-order term is

$$I_{b-s}^{(2)} = 4\eta \int dt\, dx\, dy \left(p_\parallel \hat{q} \tilde{v}_\parallel + p_l \hat{q}^3 \tilde{v}_l + p_{\text{tr}} \hat{q}^3 \tilde{v}_{\text{tr}} \right). \tag{37}$$

Here $\hat{q}$ is the nonlocal operator reduced in the Fourier representation to the multiplication by an absolute value q of the wave vector and the components v_{tr}, and p_{tr} are defined by (A5) and (A8).

Before presenting the third- and fourth-order terms it is convenient effectively to exclude the variables $p_{\text{tr}}, v_{\text{tr}}$ from the partition function $\exp(iI)$ by its integration over these variables. Since the variable p_{tr} does not figure in expression (A10) or (34) for the surface effective action I_s and the variable v_{tr} does not enter the right-hand side of Eq. (36) for ς, we should consider only the effective action I_{b-s}. Since I_{b-s} in our approximation is the second-order function of p_{tr}, v_{tr}, the integration of $\exp(iI)$, is reduced to minimization of the effective action I_{b-s} over these variables. After this procedure we get the following terms of the third and fourth order:

$$I_{b-s}^{(3)} = -4\eta \int dt\, dx\, dy\, p_\parallel \left(\hat{q}_\alpha [\hat{q}, u] \hat{q}_\alpha + \nabla_\alpha u i \hat{q}_\alpha \hat{q} \right) \tilde{v}_l$$

$$+ 4\eta \int dt\, dx\, dy\, p_l \left(i \hat{q}_\alpha \hat{q} \nabla_\alpha u + \hat{q}_\alpha [\hat{q}, u] \hat{q}_\alpha \right) \tilde{v}_\parallel, \tag{38}$$

$$I_{b-s}^{(4)} \Rightarrow 2\eta \int dt\, dx\, dy\, (\nabla u)^2 p_\parallel \hat{q} \tilde{v}_\parallel. \tag{39}$$

The square brackets in (38) denote the commutator and $\hat{q}_\alpha = -i\nabla_\alpha$. The arrow in expression (39) means that on the right-hand side of it we have retained only the relevant term. The comments concerning selection rules leading to this expression are given in Sec. V.

Let us discuss now the opposite case, i.e., $\omega \gg \eta q^2/\rho$. We will be interested here only in the second-order term of the expansion of I_{b-s}.

Using expressions (B31) and (B32) from Appendix B and substituting these expressions into (A11) one can find

$$I_{b-s}^{(2)} = 2 \int dt\, dx\, dy\, \{ \sqrt{\eta\rho}[p_l \hat{q}^2 (-i\omega)^{1/2} \tilde{v}_l$$

$$+ p_{\text{tr}} \hat{q}^2 (-i\omega)^{1/2} \tilde{v}_{\text{tr}}]$$

$$- i\rho p_\parallel \hat{\omega} \hat{q}^{-1} \tilde{v}_\parallel \}. \tag{40}$$

Here we introduced the designation $\hat{\omega} = i\partial/\partial t$. The first two terms in (40) originate from the terms with $\nabla_\parallel \tilde{v}_i$ in (A11) and the last term in (40) comes from the contribution to the effective action (A11) related to $\lfloor \partial \psi/\partial t \rfloor$.

The expressions for the effective action given above reduce our problem to the conventional field theory problem. The harmonic part of the effective action determined by (34), (37), or (40) gives bare correlation functions and the higher-order terms presented by (34), (38), and (39) give interaction vertices. We should remember also the relations (35) and (36).

V. BENDING MODE

This section is devoted to the bending mode. Our investigation is based on the calculation of correlation functions with the partition function $\exp(iI)$, where I is the effective action. Due to the softness of the bending mode the corresponding effective action I is defined as a sum of terms (34), (37), (38), and (39), which are correct for frequencies $\omega \ll \eta q^2/\rho$.

The bending mode is described by the displacement u of the membrane in the Z direction and by the auxiliary variable $p_\parallel$. It is important that the bending mode is softer than the elastic one. Therefore at the investigation of this mode one may effectively exclude elastic degrees of freedom from the consideration. Namely, we should exclude the variables p_l, v_l and ς related to v_l by Eq. (36). To do this we have to integrate the partition function $\exp(iI)$ over these variables. Since this action and relations (35) and (36) are linear in v_l and p_l, the integration of $\exp(iI)$ over v_l, p_l, and ς is Gaussian and gives the partition function $\exp(iI_u)$, where I_u is the extremum of I over p_l and v_l at the condition (36).

At studying bending mode we may omit the term with the time derivative in (36). Then we find a relation

$$\hat{q}^2 v_l = -v_l \nabla^2 u. \tag{41}$$

Since the variable ς does not enter the relation (41), we may find the extremum of I over ς without any condition. As a result, we get a relation between p_l and $p_\parallel$. The explicit form of this relation may be found from (34); it is

$$\hat{q}^2 p_l = -\nabla^2 u p_\parallel, \tag{42}$$

analogous to (41).

From (34) and (37) one can find the second-order part of the effective action I_u

$$I_u^{(2)} = -\int dt\, dx\, dy \left(\kappa \nabla^2 p_\parallel \nabla^2 u + 4\eta p_\parallel \hat{q} \tilde{v}_\parallel \right). \tag{43}$$

The substitution of (41) and (42) into (38) does not pro-

duce essential terms. Therefore the interaction term in I_u originates only from (34) and (39). It is

$$I_u^{(4)} = -\int dt\, dx\, dy \left(2\eta p_\parallel (\nabla u)^2 \hat{q} \tilde{v}_\parallel \right.$$
$$\left. -\frac{1}{2}\kappa p_\parallel \{(\nabla u)^2 \nabla^4 u + \nabla^2 \nabla_\alpha [(\nabla u)^2 \nabla_\alpha u]\} \right).$$
$$(44)$$

The sum $I_u^{(2)} + I_u^{(4)}$ determines the effective action I_u figuring in the partition function $\exp(iI_u)$. It enables us to calculate the dynamical correlation functions defined by (26) and (27).

Namely, we will consider

$$G = \langle u\, p_\parallel \rangle, \qquad D = \langle u\, u \rangle. \qquad (45)$$

The function D, according to the definition (25), is the functional integral

$$D(t_1 - t_2, \mathbf{r}_1 - \mathbf{r}_2)$$

$$= \int Du\, Dp_\parallel \, \exp(iI_u)\, u(t_1, \mathbf{r}_1) u_\parallel (t_2, \mathbf{r}_2), \qquad (46)$$

and there is an analogous expression for the function G. The function D is the pair correlation function of displacements of the membrane and G is the susceptibility determining the value of the average displacement $\langle u \rangle$ arising as a response to an external force applied to the membrane in the l-direction. Namely,

$$\langle u(t', r'_\alpha) \rangle = -i \int dt\, dx\, dy\, G(t' - t, r'_\alpha - r_\alpha) f_\parallel (t, r_\alpha), \qquad (47)$$

where $f_\parallel$ is the surface density of the force.

To determine bare correlation functions we may restrict ourselves only to the harmonic part of the effective action (43). Recall that u is related to $\mathbf{v}_s$ by Eq. (35). It is convenient to present a result of calculations for the Fourier components of the correlation functions (45). They are

$$G(\omega, q) = -(4\eta q\omega + i\kappa q^4)^{-1}, \qquad (48)$$

$$D(\omega, q) = \frac{8\eta T}{q(16\eta^2\omega^2 + \kappa^2 q^6)}. \qquad (49)$$

Here ω is the frequency and $\mathbf{q}$ is the two-dimensional wave vector with the components along the X and Y axes.

Since G is the susceptibility of the system its poles must determine the dispersion law of the bending mode. The function (48) has the pole at the following frequency:

$$\omega = -i\kappa q^3 / 4\eta. \qquad (50)$$

This dispersion law determines an overdamped mode. It

was found in the paper by Broshard and Lennon [22] (see also the papers [23] and [34]). There is also the relation

$$D(\omega) = -\frac{T}{\omega}[G(\omega) - G(-\omega)], \qquad (51)$$

which has the meaning of the fluctuation-dissipation theorem relating the pair correlation function D to the susceptibility G.

To investigate fluctuation corrections to the bare expressions (48) and (49) for the correlation functions (45) we have to construct the perturbation theory, where interaction vertices are determined by the effective action (44) [let us also recall the relation (35)]. The first corrections are determined by the diagrams in Figs. 1 and 2. In these figures dashed lines represent the bare correlation functions D and solid lines represent the bare correlation functions G with the arrow directed from $p_\parallel$ to u. White quadrangles and dark ones designate the fourth-order vertices determined by (44).

Using the explicit forms of the functions G and D (48) and (49) one can find analytical expressions corresponding to these diagrams. The loops in these figures may be treated as self-energy blocks. After summation of all diagrams for G and D with these self-energy corrections we find that the functions D and G in this one-loop approximation are the same as bare functions up to redefinition of parameters $\kappa \to \kappa_1, \eta \to \eta_1$ and satisfy the same fluctuation-dissipation theorem (51) as the bare functions.

The correction $\Delta\kappa = \kappa_1 - \kappa$ is

$$\Delta\kappa = -\frac{3}{2}\kappa \int \frac{d\omega\, d^2 q}{(2\pi)^3} q^2 D(\omega, \mathbf{q}). \qquad (52)$$

The integral in (52) is logarithmic. It can be found explicitly

$$\Delta\kappa = -\frac{3T}{4\pi}L, \qquad (53)$$

where $L = \ln[\max(qa_m; \omega\tau_m)]$ and a_m, τ_m are certain characteristic molecular size and relaxation time, respectively. Note that in the static limit (53) coincides with (10) for a static renormalization of κ.

In the same manner one can calculate the renormalization of the coefficient η. As a result, we obtain

$$\eta_1 = \eta + \Delta\eta,$$

where

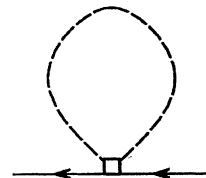$$\Delta\eta = \frac{T\eta}{4\pi\kappa}L = \frac{1}{3}g_0 L$$

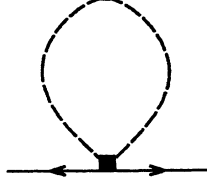FIG. 1. The first fluctuation correction to the pair correlation function G.

FIG. 2. The first fluctuation correction to the pair correlation function D.

and g is the coupling constant (7). We have to stress that such a renormalization of a parameter η does not imply an increase of the viscosity of the solvent.

Now we can explain selection rules of relevant second-order terms in u leading to (39) [and consequently to (44)]. We have taken into account only the terms leading to logarithmic renormalizations of the parameters κ and η, describing the bending mode, assuming that the value of the logarithmic factor L is large. Actually, it is not difficult to perform this selection straightforwardly.

We have investigated one-loop corrections to the bare correlation functions. The small parameter of the expansion in "loops" is the parameter $g_0 L$ mentioned in Sec. I; this parameter has to be supposed to be somehow small. In principle, one can pose a task to renormalize of the bending mode not for small values of the parameter $g_0 L$. If this parameter is of the order of unity, we may not restrict ourselves to the first one-loop correction. To take into account main logarithmic corrections one could hope to use the renormalization-group theory. Unfortunately it is not effective in our case, due to the nonlocal form of the effective action leading to an infinite number of "charges."

VI. ELASTIC DEGREE OF FREEDOM

In the preceeding section we have considered the bending degree of freedom of a membrane. Let us now take the elastic degree. First we cite the results of a linear analysis. The linear dispersion law of the elastic mode is practically identical to the dispersion law for the analogous mode of a Langmuir film [21]

$$\omega = \frac{\pm\sqrt{3} - i}{2} \left(\frac{B^2}{4\eta\rho} \right)^{1/3} q^{4/3}. \qquad (54)$$

The difference in the numerical factor is accounted for by the fact that the membrane is surrounded by a liquid on both sides. It should be borne in mind that this mode exists only if the number of molecules constituting the film is conserved. Apparently, this condition is violated only for very low frequencies, when the exchange of molecules between the solution and the membrane becomes relevant.

It stands to reason to study fluctuation corrections to the dispersion law (54). For the elastic mode the high-frequency region $\omega \gg \eta q^2/\rho$ is relevant, where q is a characteristic wave vector. On the other hand, the fluctuation corrections are related to bending fluctuations,

and therefore (at the same frequency) a characteristic wave vector of the bending mode estimated from (50) is $k \simeq (\eta\omega/\kappa)^{1/3}$. Comparing k and q one can see that actually in the whole region of the applicability of the hydrodynamical approach $k \gg q$. Indeed for this inequality to be true it should be $\omega \ll \eta^5/\rho^3\kappa^2$, which is not a strong restriction.

In Sec. V at the investigation of the bending mode we have exploited the conditions (41) and (42) for variables p_l and v_l. The characteristic frequency of the bending mode $\omega \ll \eta k^2/\rho$, and therefore actually we have implied low-frequency components of these variables. In the high-frequency region the conditions (41) and (42) are not valid, and we should perform a new procedure. Namely, the variable ς has to be expressed in terms of other hydrodynamical variables entering the effective action. To do this we can use the formal solution of Eq. (36)

$$\varsigma = \left(\frac{\partial}{\partial t} \right)^{-1} (\hat{q}^2 v_l + v_\| \nabla^2 u). \qquad (55)$$

Substituting this expression into the effective action (34) we get up to fourth-order terms

$$I_\varsigma = \int dt\, dx\, dy\, B \left(\nabla^2 p_l + \nabla_\alpha p_\| \nabla_\alpha u \right) \partial_t^{-1}$$
$$\times \left(\nabla^2 v_l + \frac{1}{2}\partial_t(\nabla u)^2 \right). \qquad (56)$$

Here we have omitted the terms $\nabla_\alpha(p_\| \nabla_\alpha u)$ and $\nabla_\alpha(v_\| \nabla_\alpha u)$ since they are small over the parameter q/k. Let us consider now the second part of the surface effective action coming from the bulk (A11). The analysis of this term shows that with the same accuracy as at the derivation of (56) we may restrict ourselves to the second-order action (40). For our investigation only the term

$$I_{b-s} = 2\sqrt{\eta\rho} \int dt\, dx\, dy\, \nabla_\alpha p_l \sqrt{\partial_t} \nabla_\alpha \tilde{v}_l \qquad (57)$$

is needed.

The dynamics of the elastic degree of freedom is characterized by the following correlation functions:

$$\langle v_l p_l \rangle_{\omega q} = G_l, \qquad (58)$$

$$\langle v_l v_l \rangle_{\omega q} = D_l. \qquad (59)$$

The bare values of G_l, D_l are determined by the harmonic part of the effective action $I_{el} = I_\varsigma + I_{b-s}$. Fluctuation corrections to the bare values are determined by the interaction terms in I_{el}. It is convenient to represent these correlation functions introducing as usual the self-energy function Σ_l and the polarization operator Π_l, related to the interaction. In the Fourier representation one can write

$$G_l = [B_R q^4/\omega - 2i\sqrt{\eta\rho}(-i\omega)^{1/2}q^2]^{-1}, \qquad (60)$$

where we have included the self-energy function Σ_l into

FIG. 3. The first contribution to the self-energy function Σ_l.

the quantity B_R

$$B_R = B + i\omega q^{-4}\Sigma_l. \tag{61}$$

The pair correlation function is

$$D_l(\omega) = -G_l(\omega)[2T\sqrt{2\eta\rho|\omega|}q^2 + \Pi_l(\omega)]G_l(-\omega), \tag{62}$$

where the first term in the square brackets is induced by the harmonic part of I_{el}.

The first fluctuation contributions to Σ_l, Π_l are presented in Figs. 3 and 4. The dashed and solid lines in these diagrams have the same meaning as in Sec. V (D and G functions); the white and dark circles designate third-order vertices whose structure is determined by the term (56).

The main contributions to Σ_l are represented by the ladder series of diagrams, depicted in Fig. 5, where quadrangles designate the fourth-order vertices determined by (56). Performing the summation of this ladder series one can find

$$B_R = \frac{B}{1 - iBF}. \tag{63}$$

The function F corresponds to a loop in the diagram constructed from G and D (as is depicted in Fig. 5)

$$F = \int \frac{d\nu\, d^2k}{(2\pi)^3} q^4 D(\nu)G(\omega + \nu). \tag{64}$$

Using expressions (48) and (49) we find this function explicitly

$$F(\omega) = \frac{iT\sqrt{3}}{2\kappa^{4/3}(2\eta)^{2/3}}\left(\frac{i}{\omega}\right)^{2/3}. \tag{65}$$

At the derivation of (65) we assumed that the condition $k\xi \gg 1$ is fulfilled, which means $\omega \gg \kappa/(\eta\xi^3)$. Note also that for a strong renormalization of B to be possible we have to suppose

$$\xi^2 \gg \kappa^2/(TB), \tag{66}$$

which ensures that the condition $BF \gg 1$ is satisfied for frequencies $\omega \gtrsim \kappa/(\eta\xi^3)$.

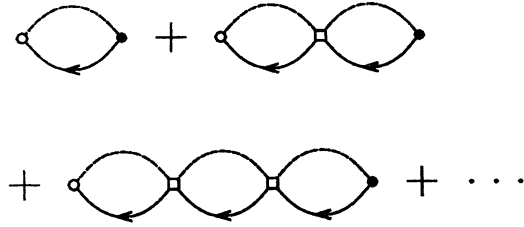FIG. 4. The first contribution to the "polarization operator" Π_l.

FIG. 5. The "ladder" sequence of diagrames determining the principal contribution to Σ_l.

In the same manner one can find the main contribution to Π_l. It is determined by the ladder sequence of the diagrams of the type depicted in Fig. 4. The sum of this series is

$$\Pi_l(\omega) = \frac{q^4}{2}F_D(\omega)B_R(\omega)B_R(-\omega), \tag{67}$$

where the function F_D corresponds to a loop constructed from two D lines

$$F_D(\omega) = \int \frac{d\nu\, d^2k}{(2\pi)^3} q^4 D(\nu)D(\omega + \nu). \tag{68}$$

There is a relation for the functions F and F_D following from (51)

$$F_D(\omega) = -\frac{2T}{\omega}[F(\omega) - F(-\omega)]. \tag{69}$$

One can check that (62), (67), and (69) supply the fluctuation-dissipation theorem for G_l and D_l,

$$D_l(\omega) = -iT[G_l(\omega) + G_l(-\omega)]. \tag{70}$$

The dispersion law for the elastic mode should be determined by the poles of the renormalized Green's function G_l. If $\omega \gg (TB)^{3/2}/(\kappa^2\eta)$, then the renormalization of B is not relevant, i.e., $B_R \simeq B$, and we obtain from (60) the bare dispersion law (54) for the elastic mode. In the region $\omega \ll (TB)^{3/2}/(\kappa^2\eta)$ the renormalization of B is strong and the renormalized value of B does not depend on the bare value

$$B_R \simeq i/F. \tag{71}$$

In this case the condition for the Green's function G_l to have a pole follows from (60) and (71),

$$2i\sqrt{\eta\rho}(-i\omega)^{3/2}F = q^2. \tag{72}$$

However, Eq. (72) has no solutions on the physical sheet of the complex plane ω. This rather unusual phenomenon does not lead to any catastrophic consequence since the response function has the physical meaning only at real frequencies, and there are no restrictions on types of singularities at complex ω. Therefore the disappearance of poles of the response function G_l does not essentially change the physical behavior.

The correlation function D_l is associated with a high-frequency motion of a liquid, concentrated near the mem-
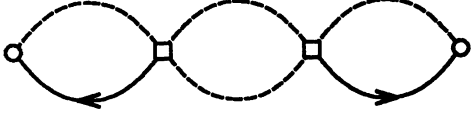
FIG. 6. The typical diagram determining a relevant contribution to Π_l.

brane. Perhaps physically it is more reasonable to examine the correlation function $\langle \varsigma\varsigma\rangle_{\omega q} = D_\varsigma$. The expression for this function can be found directly by means of the representation (55), but technically it is easier to express the effective action I_{el} in terms of the variables ς and p_ς conjugated to it. Then the interaction with bending fluctuations will be determined by I_{b-s} (57). To find the main fluctuation contribution to D_ς we should sum up the ladder series of diagrams of the type depicted in Fig. 6. We omit the calculations and give only the result for the region of frequencies $\omega^5 \gg g^6\eta\kappa^2 q^{12}/\rho^3$, where the fluctuation contribution dominates

$$D_\varsigma(\omega) = \frac{F_D(\omega)}{2\left(1 - iBF(\omega)\right)[1 - iBF(-\omega)]}. \quad (73)$$

Here the function $F(\omega)$ is defined by (65) and the function $F_D(\omega)$ is defined by (68) being related to F by (69).

VII. FLUCTUATION CONTRIBUTION TO VISCOSITY

Above we have examined a separately taken membrane. Now we are going to investigate long-range dynamic effects in dilute lyotropic systems related to membranes. We believe that there is a long-wavelength motion in the liquid and will investigate a response of a system of membranes to the motion.

For a separate membrane it means that we have to regard the variables p_i and v_i in the effective action (34) as the following sum:

$$p_i \to p_i + \bar{p}_i, \quad v_i \to v_i + \bar{v}_i, \quad (74)$$

where the quantities $\bar{p}_i$ and $\bar{v}_i$ represent the long-wavelength motion, and p_i, v_i describe the motion concentrated near the membrane. Let us stress that the variables $\bar{p}_i$ and $\bar{v}_i$ do not enter the contribution I_{b-s} into the effective action related to the bulk motion near the membrane.

Note that at a description of the long-wavelength motion we could not use the variables v_l, p_l which are components in the coordinate system referred to the bending membrane. The fact is that it should be described in the external (laboratory) Cartesian reference system.

After the substitution (74) we find in the main approximation from (34) the following contribution to the effective action containing $\bar{v}, \bar{p}$:

$$I_v = \int dt\, dx\, dy\, B(\nabla_\alpha \bar{p}_\alpha + \nabla_\alpha u \nabla_\alpha p_\parallel)\partial_t^{-1}$$
$$\times \left(\nabla_\alpha \bar{v}_\alpha + \frac{1}{2}\partial_t(\nabla_\beta u)^2\right). \quad (75)$$

There are no "cross" terms proportional to $\bar{p}p$ and so on in this effective action (75). The fact is that $\bar{p}$ and $\bar{v}$ are related to the long-wavelength motion, whereas p and v are related to the motion of the membrane with the wave vectors $k \gtrsim \xi^{-1}$. Therefore these cross terms vanish at the integration over space coordinates. The term $\nabla_\alpha u \nabla_\alpha p_\parallel$ entering (75) should be treated as having a small wave vector, which is possible for large wave vectors of $u, p_\parallel$.

Above we omitted a term with components $\bar{p}_z$ and $\bar{v}_z$. This term originates, for example, from the κ-dependent part of the effective action (31). However, contributions to viscosity coefficients, induced by this term, are higher order in g than those related to p_α, v_α components and therefore are not relevant.

To characterize the long-wavelength motion we should find the response function

$$G_{ik}(t_1 - t_2, \mathbf{r}_1, \mathbf{r}_2) = \langle \bar{v}_i(t_1, \mathbf{r}_1)\bar{p}_k(t_2, \mathbf{r}_2)\rangle. \quad (76)$$

This function can be directly related to viscosity coefficients which we are looking for. Indeed using the definition (27) and the second-order part of (28) one can derive the following equation for G_{ik}:

$$i\left((\partial_t - c^2\nabla^2\partial_t^{-1})\delta_{ik} - [\eta\nabla^2\delta_{ik} + (\tfrac{1}{3}\eta + \zeta)\nabla_i\nabla_k]\right)$$

$$\times G_{kn}(t_1 - t_2, \mathbf{r}_1, \mathbf{r}_2)$$

$$+ \int dt_3\, d^3r_3\, \Sigma_{ik}(t_1 - t_3, \mathbf{r}_1, \mathbf{r}_3)G_{kn}(t_3 - t_2, \mathbf{r}_3, \mathbf{r}_2)$$

$$= -\delta_{in}\delta(t_1 - t_2)\delta(\mathbf{r}_1 - \mathbf{r}_2). \quad (77)$$

Here the self-energy function Σ_{ik} represents fluctuation contributions caused by the surface term (31). In our case this term is reduced to (75) related to bending fluctuations.

To find the contribution to Σ_{ik} determined by (75) we have to sum up the same ladder series of diagrams as is depicted in Fig. 5. As a result, we find

$$\Sigma_{\alpha\beta}(\omega, \mathbf{r}_1, \mathbf{r}_2) = \frac{B_R}{\omega}\delta(z_1)\delta(z_2)$$
$$\times \nabla_\alpha\nabla_\beta\left[\delta(x_1 - x_2)\delta(y_1 - y_2)\right], \quad (78)$$

where the membrane is believed to be arrayed along the plane $z = 0$. More accurately this function $\Sigma_{\alpha\beta}$ should be associated with a piece of the membrane (with a characteristic size ξ) which can be considered as roughly flat.

To find a contribution to the long-wavelength characteristics due to membranes we should average expression (78) over these pieces of membranes. As a result, we obtain the average self-energy function $\bar{\Sigma}_{\alpha\beta}(\omega, \mathbf{r}_1 - \mathbf{r}_2)$, carrying the information needed for us. The real part of this function determines corrections to the sound velocity (or velocities if there are more than one sound mode).

The imaginary part of $\bar{\Sigma}$ determines fluctuation contributions to the viscosity coefficients. For dilute systems corrections to the sound velocity are small (and we will not consider them), whereas contributions to the viscosity prove to be relevant.

This averaging procedure is most simple for isotropic phases (sponge or solutions of vesicles). Then

$$\eta_{fl} = -\frac{7}{15}\frac{S}{V}\frac{\mathrm{Im}B_R}{\omega}, \tag{79}$$

where the factor S/V represents the density of membranes and the numerical factor arises at averaging over angles. Analogously one can find the fluctuation contribution to the second viscosity coefficient ζ. We present here the expression for the combination of the first and second viscosity coefficients determining the sound attenuation

$$\frac{4}{3}\eta_{fl} + \zeta_{fl} = \frac{8}{7}\eta_{fl}. \tag{80}$$

Combining the given above expressions for the renormalization of the module B one can find fluctuation viscosities in different frequency regions (remember that in the expressions given below the numerical factors are associated with the isotropic case). In the region $\omega \gg (BT)^{3/2}/(\kappa^2\eta)$, where $B_R \simeq B + iB^2F$ we get

$$\eta_{fl} = \frac{7}{20}\frac{TB^2S}{\kappa^{4/3}(2\eta)^{2/3}V}\mid\omega\mid^{-5/3}. \tag{81}$$

Note that, strictly speaking, there is one more inequality which should be satisfied for expression (81) to be valid. Namely, we have to suppose $\omega \ll B^{3/2}/(\kappa\eta)^{1/2}$ to neglect nonlinear terms in the effective action provided by the surface stress tensor (22) related to Helfrich's energy (1). The second region $\omega \ll (BT)^{3/2}/(\kappa^2\eta)$ is probably more realistic from the experimental point of view. If (66) is fulfilled, it is consistent with the inequality $\omega \gg \kappa\xi^{-3}/\eta$. In this region $\bar{B} \simeq i/F$ and

$$\eta_{fl} = \frac{7}{15}\frac{\kappa^{4/3}(2\eta)^{2/3}S}{TV}\mid\omega\mid^{-1/3}. \tag{82}$$

For frequencies $\omega \ll \kappa\xi^{-3}/\eta$ expression (65) for the function F is not valid, since in this case the region of integration over k is determined by the characteristic wave vector of the order of ξ^{-1}. If for the characteritic scale ξ the condition (66) is fulfilled, then

$$\mathrm{Im}B_R \sim \frac{\mathrm{Re}F}{(\mathrm{Im}F)^2}. \tag{83}$$

Otherwise (when ξ is smaller than $\kappa/\sqrt{TB}$)

$$\mathrm{Im}B_R \sim \frac{\mathrm{Re}F}{B^2}. \tag{84}$$

The imaginary part of the function F in this low-frequency region does not depend on ω and the real part can be estimated from (69) and (64)

$$\mathrm{Re}F = -\frac{\omega}{T}F_D \sim \frac{\omega}{\kappa}\eta\xi^3\mathrm{Im}F \tag{85}$$

and

$$\mathrm{Im}F \sim T\xi^2\kappa^{-2}. \tag{86}$$

Using these estimates and expressions (83) and (84) we can find the fluctuation viscosity in the low-frequency region. If the renormalization of B due to bending fluctuations is small we have from (84)

$$\frac{\eta_{fl}}{\eta} \sim \frac{S}{V}T\eta\xi^5/\kappa^3. \tag{87}$$

If $\xi^2 \gg \kappa^2/(TB)$, i.e., fluctuations strongly renormalize B, then the ratio of η_{fl} to the viscosity of the solvent η will be of the order of

$$\frac{\eta_{fl}}{\eta} \sim \frac{\kappa S\xi}{TV}. \tag{88}$$

For sponge phases the quantity $S\xi/V$ is of the order of unity. Therefore the ratio $\eta_{fl}/\eta \sim g^{-1}$ and η_{fl} exceeds the bare value η (let us recall that g is a small parameter of the theory). Note also that (87) reveals a rather unexpected phenomenon of an increase of the fluctuation viscosity at the dilution of lyotropic systems.

Above we have discussed only isotropic systems. For other cases our results are also qualitatively correct. For example, in cubic phases the fluctuation contributions to the viscosity coefficients will be of the same order as the ones for the sponge phase (and at $g < 1$ they exceed the bare value η), but the numerical factors in the expressions for the contributions differ from the ones for the sponge phase. Lamellar phases need a separate consideration which we present below.

In the lamellar phases orientations of different pieces of membranes are not random. They have a preferred direction characterized by a unit vector $\mathbf{n}$ analogous to the director in thermotropic smectics A. Since the vector l orthogonal to the pieces is on the average directed along $\mathbf{n}$ after averaging of (78) over the pieces, we obtain a contribution to the viscosity coefficient η_{4fl} determining the following contribution to the viscosity tensor:

$$\eta_{4fl}(\delta_{ij} - n_in_j)(\delta_{kl} - n_kn_l). \tag{89}$$

We use the conventional notation for the viscosity coefficient of a system with uniaxial symmetry [35]. It is not very difficult to connect the value of η_{4fl} with the value η_{fl} found for the sponge phase. Namely,

$$\eta_{4fl} = -\frac{S}{TV}\frac{\mathrm{Im}B_R}{\omega} = \frac{15}{7}\eta_{fl}. \tag{90}$$

The fluctuation contribution to η_4 is the leading one; it exceeds the bare value η. But there are also fluctuation contributions to other viscosity coefficients of the lamellar phase arising at averaging over the pieces since they are oriented not precisely along the Z axis. The simplest way to find the contributions is to use expression (89) and to take into account fluctuations of the vector $\mathbf{n}$. Apart

from these fluctuation contributions, there are bare contributions determined by the viscosity of the solvent [coefficients η and ζ in (21)]. Using the explicit expressions for the viscosity tensor of the isotropic liquid and definitions of the viscosity coefficients for the lamellar phase (see [35]), one can find

$$\eta_1 = \frac{4}{3}\eta + \zeta + 4N\eta_{4\text{fl}}, \quad \eta_2 = \eta, \quad \eta_3 = \eta + N\eta_{4\text{fl}},$$

$$\eta_5 = \zeta - \frac{2}{3}\eta + 2N\eta_{4\text{fl}}, \tag{91}$$

and, of course, $\eta_4 \simeq \eta_{4\text{fl}}$.

The factor N entering these expressions is related to averaging over fluctuations of **n**. It is easy to estimate this factor using a conventional expression for the elastic energy of smectics and inserting the values of elastic constants of the lamellar phase (see, e.g., review [6]). Performing this procedure one can find

$$N\delta_{\alpha\beta} = \langle n_\alpha n_\beta \rangle = \int \frac{dq_z d^2 q_\perp}{(2\pi)^3} \frac{T q_\alpha q_\beta}{B_{\text{Sm}} q_z^2 + K q_\perp^4}. \tag{92}$$

The smectic elastic modules B_{Sm} and K entering (92) can be expressed in terms of κ and ξ [6]

$$B_{\text{Sm}} \sim T^2/\kappa\xi^3, \quad K \sim \kappa/\xi. \tag{93}$$

The integral in (92) is determined by the characteristic wave vectors $q \lesssim \xi^{-1}$ and can be estimated by means of (93) as $N \sim g$. Therefore this factor is small due to $g \ll 1$, which is the condition of the applicability of our consideration. But the fluctuation contributions into viscosity coefficients presented in (91) are of the order of the bare coefficients η, ζ, since they originated from the large (proportional to g^{-1}) fluctuation contribution to the coefficient η_4.

The fluctuation viscosity coefficients we have investigated in this section directly determine relaxation of shear motions (since it is the first viscosity that enters the corresponding dispersion law), sound attenuation, and other reological and acoustical properties in lyotropic systems.

CONCLUSION

Let us sum up the results of our paper. We theoretically investigated dynamical properties of dilute lyotropic systems for the case when molecules constitute a system of membranes. The presence of membranes determine the main peculiarities of these systems including dynamical ones. A membrane possesses two soft degrees of freedom associated with its bending deformations and with variations of the surface density of molecules n_s constituting the membrane. Correspondingly, there are two characteristic surfacial modes where the motion of the solvent is localized near the membrane. In the linear approximation these modes prove to be overdamped (50) and (54).

We investigated the role of nonlinear fluctuation effects

in the framework of the mode-coupling theory; bending fluctuations proving to be most important. We developed the perturbation theory over a dimensionless parameter $g \simeq T/\kappa$ (T being the temperature and κ being the Helfrich module) describing the "strength" of the bending fluctuations, g assumed to be small. These fluctuations produce only small logarithmic corrections to the characteristics of the bending mode whereas they essentially modify the dynamical behavior of the elastic degree of freedom. Namely, the fluctuations drastically change the frequency dependence of the susceptibility describing the relaxation of n_s to the equilibrium. This renormalization of the elastic mode manifests itself particularly in a rather complicated frequency dispersion of the corresponding response function. For example, due to fluctuations poles of this response function in the lower half plane of ω disappear. We would like to stress that physically it does not create any problem since the response function is directly observable only at real frequencies.

From the experimental point of view eigenmodes of the system manifest themselves in the behavior of the dynamical structure factor, which can be measured by x-ray or neutron scattering. Of course, it is not an easy problem to observe a motion corresponding to bending and elastic modes of the membranes due to the presence of the bulk motion. However, both these contributions (from the bulk and surfacial motions) can be distinguished, since they have different dependences on frequencies and wave vectors.

We have also studied the influence of dynamical fluctuations of membranes on macroscopic characteristics of a system such as viscosity coefficients. The main part of the fluctuation contribution η_{fl} to viscosity coefficients is associated with the elastic part of the membrane stress tensor but strongly renormalized by bending fluctuations. The value of η_{fl} possesses a complicated frequency dispersion. In the high-frequency region it behaves as $\omega^{-5/3}$, in the intermediate frequency region it behaves as $\omega^{-1/3}$, and for small ω it remains constant. The ratio of this constant to the viscosity of the solvent is of the order of g^{-1}, i.e., η_{fl} exceeds the viscosity of a pure solvent. For this assertion to be correct the inequality (66) should be fulfilled. Our results are in agreement with the experiment [36] where the ratio η_{fl}/η exceeding unity has been observed in the limit of a very dilute sponge phase.

An attempt to estimate a membrane contribution to the low-frequency viscosity of lyotropic systems was taken in the paper by Onuki and Kawasaki [37]. But their estimates (giving a fluctuation contribution of the same order as the viscosity of the pure solvent η) are in fact related to the linear coupling between the membrane modes and long-wavelength degrees of freedom, which is actually nonexistent. There are only fluctuation contributions to viscocity, the principal one (which we have investigated in this paper) being of the order of $g^{-1}\eta$ and, besides, there are contributions to the viscosity related to the κ-dependent part of (22), these contributions are of the order of $g\eta$ and therefore small. The authors of paper [38] using some qualitative arguments discussed fluctuation contributions to the viscosity for lyotropic phases and found the frequency dependence analogous to our ex-

pression (82). But the coefficient figuring in their expression contains a small dimensionless factor dg/ξ (where d is the thickness of a membrane) in comparison with our result (82), and therefore their term is negligible.

Note also that the contribution to the viscosity coefficient η_3 of lamellar phases (which is the same order as the viscosity of the solvent) found in Sec. VII is the most plausible reason for the discrepancy between the values of κ, found in the paper [39] from the relaxation times of the long- and short-wavelength modes characteristic of lamellar phases.

We believe that our predictions can be checked more clearly in acoustic experiments. The problem is that the fluctuation contribution to the viscosity we have found directly determines the attenuation of the sound. Another favorable circumstance is the hardness of a sound, at a given frequency ω the sound wave possesses a small wave vector $q = \omega/c$ (c being the velocity of sound). Therefore in acoustic experiments it is possible to measure a wide region of frequencies without breaking the condition $q\xi \ll 1$ employed at the derivation of the expressions for η_{fl}.

ACKNOWLEDGMENTS

APPENDIX A: PARAMETRIZATION OF SURFACE QUANTITIES

In this appendix we give relations for the surface quantities derived in Secs. I–III in the reference system where the position of a membrane is characterized by the displacement u of the membrane in the z direction. Let us stress that the use of u does not mean that we have to confine ourselves to small displacements u. All relations given below in this appendix are formally exact ones. The only restriction is that the function $z = u(x,y)$ must be single valued.

At the taken parametrization all the functions characterizing the membrane should be treated as functions of x,y. For example, we will assume that the quantity n_s is a function of x,y. The velocity $\mathbf{v}_s$ of the membrane in this case is related to the three-dimensional velocity $\mathbf{v}$ of a liquid by the following way:

$$v_{si}(x,y) = v_i(x,y,u). \tag{A1}$$

Note that now only the derivatives along the X,Y directions appear in terms determined by the surface quantities.

Since the function Φ is now determined by expression (33), the components of the unit vector (14) perpendicular to the membrane are

$$l_\alpha = -\frac{\nabla_\alpha u}{\sqrt{1+(\nabla u)^2}}, \qquad l_z = \frac{1}{\sqrt{1+(\nabla u)^2}}. \tag{A2}$$

With the use of the introduced quantities the energy (1) can be rewritten as

$$\Omega_s = \int dx\, dy\, \sqrt{1+(\nabla u)^2}$$
$$\times \left(\frac{\kappa}{2}(\nabla_\alpha l_\alpha)^2 + \frac{\bar{\kappa}}{2}[(\nabla_\alpha l_\alpha)^2 - \nabla_\alpha l_\beta \nabla_\beta l_\alpha] \right). \tag{A3}$$

Instead of Cartesian components of $\mathbf{v}_s$ it is convenient to introduce new variables according to the following definitions:

$$v_\| = \sqrt{1+(\nabla u)^2}\, l_i v_{si} = v_{sz} - \nabla_\alpha u\, v_{s\alpha}, \tag{A4}$$

$$v_{s\alpha} = \frac{1}{\sqrt{1+(\nabla u)^2}}(v_\| l_\alpha + \nabla_\alpha v_l + \epsilon_{\alpha\beta}\nabla_\beta v_{tr}). \tag{A5}$$

Here we have used the subscript $\|$ to label the components of the corresponding quantities along the unit vector l, the subscript l labels the components longitudinal and the subscript tr labels the components transverse to the wave vector and $\epsilon_{\alpha\beta}$ is the two-rank antisymmetric tensor

$$\epsilon_{xy} = -\epsilon_{yx} = 1.$$

Using these definitions and Eqs. (15) and (19) one can easily find that

$$\frac{\partial u}{\partial t} = v_\| \tag{A6}$$

and

$$\frac{\partial n_s}{\partial t} + v_{s\alpha}\nabla_\alpha n_s = -\frac{n_s}{\sqrt{1+(\nabla u)^2}}(\nabla_\alpha^2 v_l + v_\|\nabla_\alpha l_\alpha). \tag{A7}$$

Note that the component v_{tr} does not enter the right-hand sides of exprssions (A6) and (A7). Actually it was a motivation for such definitions of v_l and v_{tr} in (A5).

It is also convenient to determine components of $\mathbf{p}_s$ analogously to definitions (A4) and (A5),

$$p_\| = \sqrt{1+(\nabla u)^2}\, l_i p_{si} = p_{sz} - \nabla_\alpha u\, p_{s\alpha}, \tag{A8}$$

$$p_{s\alpha} = \frac{1}{\sqrt{1+(\nabla u)^2}}(p_\| l_\alpha + \nabla_\alpha p_l + \epsilon_{\alpha\beta}\nabla_\beta p_{tr}). \tag{A9}$$

Then the surface contribution to the effective action (31) can be rewritten as

$$I_s = \int dt\, dx\, dy\, \Bigg[-Bp_l\nabla_\alpha^2 \varsigma - p_\| B\varsigma\nabla_\alpha l_\alpha$$
$$+\kappa p_\| \left(-\nabla_\alpha^2\nabla_\gamma l_\gamma \right.$$
$$+l_\alpha l_\beta \nabla_\alpha\nabla_\beta\nabla_\gamma l_\gamma + \nabla_\alpha l_\alpha l_\beta\nabla_\beta\nabla_\gamma l_\gamma$$
$$\left. -\nabla_\alpha l_\alpha\nabla_\beta l_\gamma\nabla_\gamma l_\beta + \frac{1}{2}(\nabla_\alpha l_\alpha)^3 \right) \Bigg]. \tag{A10}$$

Note that the component p_{tr} does not enter expression (A10). This fact is related to the isotropy of the membrane. The contribution (32) in the new terms is written as

$$I_{\mathrm{b-s}} = -\int dt\, dx\, dy \left[\rho p_\|\lfloor\partial\tilde{\psi}/\partial t\rfloor + 2\eta p_\| l_i\lfloor\nabla_\|\tilde{v}_i\rfloor + \eta(p_\| l_\alpha + \nabla_\alpha p_l + \epsilon_{\alpha\beta}\nabla_\beta p_{\mathrm{tr}})(\lfloor\nabla_\|\tilde{v}_\alpha\rfloor + \nabla_\alpha u\lfloor\nabla_\|\tilde{v}_z\rfloor)\right], \qquad \text{(A11)}$$

where $\nabla_\| = l\nabla$ and the floors as before, designate the jumps of the corresponding variables on the membrane.

Formally exact expressions (A10) and (A11) derived above will be a starting point of our investigation of fluctuation dynamics of membranes.

APPENDIX B: EXPRESSIONS FOR DERIVATIVES

In this appendix we derive expressions for derivatives of the quantity $\tilde{v}$ defined by (30) near the membrane. These relations have been used in Sec. IV in the analysis based on expression (A11). To derive these expressions we must use an equation for $\tilde{v}$ which is the extremum condition of the bulk effective action (28). This equation should be solved for a given shape of a membrane and at a given surface value of $\tilde{v}$. We will derive formally exact relations enabling us to find the relations by expansion in the displacement of the membrane u along the Z axis.

It is convenient to represent $\tilde{v}$ as

$$\tilde{v}_i = \nabla_i\tilde{\psi} + \tilde{a}_i. \qquad \text{(B1)}$$

Here $\tilde{\psi}$ is the potential of $\tilde{v}$ and $\tilde{a}$ is the rotational part of $\tilde{v}$. The bulk motion for the problem may be investigated in the linear approximation. In this case the extremum conditions which we need may be written in the following form (see [20]):

$$\nabla^2\tilde{\psi} = 0, \qquad \text{(B2)}$$

$$\nabla^2\tilde{a} = d_t\tilde{a}, \qquad \text{(B3)}$$

where

$$d_t = \frac{\rho}{\eta}\frac{\partial}{\partial t}.$$

Note that these equations are formally identical to the bulk hydrodynamical equations for the velocity $\mathbf{v}$.

First we will consider the case of low frequencies ω

$$\omega \ll \eta q^2/\rho, \qquad \text{(B4)}$$

where q is a characteristic wave vector. Although, as a consequence of (B4), the term on the right-hand side of Eq. (B3) is small, it may not be omited. The problem is that at $d_t \to 0$ Eqs. (B2) and (B3) for $\tilde{\psi}$ and $\tilde{a}$ lead to an equation $\nabla^2\tilde{v} = 0$, the solution of which for an arbitrary surface value of $\tilde{v}$ does not possess the property of "incompressibility" $\nabla\cdot\tilde{v} = 0$ following from (B1) and (B2). Therefore we should take into account the terms proportional to d_t, but we may keep only the terms linear in d_t.

We are going to exploit relations for the derivatives of $\tilde{\psi}$ and $\tilde{a}$ near the membrane in terms of surface operators $\hat{L}$, $\hat{M}$,

$$\lfloor\nabla_z\tilde{\psi}\rfloor_\pm = \hat{L}_\pm\lfloor\tilde{\psi}\rfloor_\pm, \qquad \text{(B5)}$$

$$\lfloor\nabla_z\tilde{a}\rfloor_\pm = \hat{L}_\pm\lfloor\tilde{a}\rfloor_\pm + \hat{M}_\pm\lfloor d_t\tilde{a}\rfloor_\pm. \qquad \text{(B6)}$$

Here $\lfloor\;\rfloor_+$ designates the value of a corresponding variable "above" the membrane and $\lfloor\;\rfloor_-$ – "below" the membrane with the vector l [components of which are defined by (A2)] marking the "top" of the membrane. These quantities (e.g., $\lfloor\tilde{\psi}\rfloor_\pm$) are believed to be functions of x, y and, consequently, the operators $\hat{L}, \hat{M}$ in (B5) and (B6) should be regarded as acting on such functions as well.

The method of the deriving explicit expressions for $L_\pm, M_\pm$ is described in Appendix C. It should be stressed that for the derivatives $d_t\tilde{\psi}, \nabla_i\tilde{\psi}, d_t\tilde{a}, \nabla_i\tilde{a}$ the same relations as (B5) and (B6) for $\tilde{\psi}, \tilde{a}$ are valid. The problem is that the equations controlling the dynamics of the derivatives of $\tilde{\psi}, \tilde{a}$ are the same as (B2) and (B3) for these quantities themselves. For example,

$$\lfloor\nabla_z^2\tilde{\psi}\rfloor_\pm = \hat{L}_\pm\lfloor\nabla_z\tilde{\psi}\rfloor_\pm. \qquad \text{(B7)}$$

From (B1), (B5), and (B6) it follows that

$$\lfloor\nabla_z\tilde{v}_i\rfloor_\pm = \hat{L}_\pm\lfloor\tilde{v}_i\rfloor_\pm + \hat{M}_\pm\lfloor d_t\tilde{a}_i\rfloor_\pm. \qquad \text{(B8)}$$

Relations for the derivatives ∇_α along the X and Y axes will be also needed for us. Using the definition $\lfloor A\rfloor_\pm = A(x, y, z \to u \pm 0)$ we find the relation

$$\lfloor\nabla_\alpha A\rfloor_\pm = \nabla_\alpha\lfloor A\rfloor_\pm - \nabla_\alpha u\lfloor\nabla_z A\rfloor_\pm, \qquad \text{(B9)}$$

valid for any function A. Applying the relation to the variables $\tilde{\psi}, \tilde{a}$, we find

$$\lfloor\nabla_\alpha\tilde{\psi}\rfloor_\pm = \hat{N}_\alpha^\pm\lfloor\tilde{\psi}\rfloor_\pm, \qquad \text{(B10)}$$

$$\lfloor\nabla_\alpha\tilde{a}\rfloor_\pm = \hat{N}_\alpha^\pm\lfloor\tilde{a}\rfloor_\pm - \nabla_\alpha u\hat{M}_\pm\lfloor d_t\tilde{a}\rfloor_\pm, \qquad \text{(B11)}$$

where

$$\hat{N}_\alpha^\pm = \nabla_\alpha - \nabla_\alpha u\hat{L}_\pm. \qquad \text{(B12)}$$

The relations (B10) and (B11) enable us to deduce from (B1),

$$\lfloor\nabla_\alpha\tilde{v}_i\rfloor_\pm = \hat{N}_\alpha^\pm\lfloor\tilde{v}_i\rfloor_\pm - \nabla_\alpha u\hat{M}_\pm\lfloor d_t\tilde{a}_i\rfloor_\pm. \qquad \text{(B13)}$$

It is not difficult to check that at the condition (B4) there is an estimate $\tilde{v}/\tilde{a} \sim \eta\omega/(\rho q^2)$, which is the consequence of an almost degeneracy of Eqs. (B2) and (B3). Therefore $\tilde{v}_i \ll \nabla_i\tilde{\psi}, \tilde{a}_i$, i.e., $\tilde{a}_i \simeq \nabla_i\tilde{\psi}$ and with the accuracy needed for us from (B8) and (B13) we get

$$\lfloor\nabla_z\tilde{v}_i\rfloor_\pm = \hat{L}_\pm\lfloor\tilde{v}_i\rfloor_\pm - \hat{M}_\pm\lfloor\nabla_i d_t\tilde{\psi}\rfloor_\pm, \qquad \text{(B14)}$$

$$\lfloor \nabla_\alpha \tilde{v}_i \rfloor_\pm = \hat{N}_\alpha^\pm \lfloor \tilde{v}_i \rfloor_\pm + \nabla_\alpha u \hat{M}_\pm \lfloor \nabla_i d_t \tilde{\psi} \rfloor_\pm. \qquad (B15)$$

Exploiting the relations (B5) and (B10) for $d_t \tilde{\psi}$ we derive from (B14)

$$\lfloor \nabla_z \tilde{v}_z \rfloor_\pm = \hat{L}_\pm \lfloor \tilde{v}_z \rfloor_\pm - \hat{M}_\pm \hat{L}_\pm \lfloor d_t \tilde{\psi} \rfloor_\pm, \qquad (B16)$$

$$\lfloor \nabla_z \tilde{v}_\alpha \rfloor_\pm = \hat{L}_\pm \lfloor \tilde{v}_\alpha \rfloor_\pm - \hat{M}_\pm \hat{N}_\alpha^\pm \lfloor d_t \tilde{\psi} \rfloor_\pm. \qquad (B17)$$

Taking into account the incompressibility condition $\nabla_z \tilde{v}_z + \nabla_\alpha \tilde{v}_\alpha = 0$, we find from (B15) and (B16)

$$(\hat{M}_\pm \hat{L}_\pm - \nabla_\alpha u \hat{M}_\pm \hat{N}_\alpha^\pm) \lfloor d_t \tilde{\psi} \rfloor_\pm = \hat{L}_\pm \lfloor \tilde{v}_z \rfloor_\pm + \hat{N}_\alpha^\pm \lfloor \tilde{v}_\alpha \rfloor_\pm. \qquad (B18)$$

This relation enables us to express $\lfloor d_t \tilde{\psi} \rfloor_\pm$ and then $\lfloor \nabla_z \tilde{v}_i \rfloor_\pm$ through the value $\lfloor \tilde{v}_i \rfloor_\pm$ figuring on the right-hand sides of (B16)–(B18).

Substituting expressions (C5) and (C6) into (B18) [remember the definition (B12)] and solving the resulting relation, we may find the expression for $\lfloor d_t \tilde{\psi} \rfloor_\pm$ up to the second order in u. This expression is bulky enough. We present here an expression for the difference $\lfloor d_t \tilde{\psi} \rfloor_+ - \lfloor d_t \tilde{\psi} \rfloor_-$ figuring in the effective action (A11). Keeping only relevant (namely producing logarithmic renormalization; see Sec. V) terms of the second order in u we find

$$(\rho/\eta) \left\lfloor \frac{\partial \tilde{\psi}}{\partial t} \right\rfloor \Rightarrow -4\hat{q}\tilde{v}_z + 4\hat{q}\nabla_\alpha u \tilde{v}_\alpha$$
$$-4i\hat{q}_\alpha [\hat{q}, u]\tilde{v}_\alpha + 4\nabla_\beta u \hat{q}_\alpha \hat{q}_\beta \hat{q}^{-1} \tilde{v}_\alpha$$
$$-2(\nabla u)^2 \hat{q} \tilde{v}_z. \qquad (B19)$$

The arrow in this expression means that we restrict ourselves to the relevant second order terms. Remember that $\lfloor \ \rfloor$ designates the jump $\lfloor \ \rfloor_+ - \lfloor \ \rfloor_-$. Substituting this expression into (B16) and (B17), we find in the same approximation

$$\lfloor \nabla_\| \tilde{v}_z \rfloor \Rightarrow -2\nabla_\beta u \hat{q} \tilde{v}_\beta - 2\nabla_\beta u \hat{q}_\alpha \hat{q}_\beta \hat{q}^{-1} \tilde{v}_\alpha, \qquad (B20)$$

$$\lfloor \nabla_\| \tilde{v}_\beta \rfloor \Rightarrow -2\hat{q}\tilde{v}_\beta - 2\hat{q}_\alpha \hat{q}_\beta \hat{q}^{-1} \tilde{v}_\alpha + 2i\hat{q}_\beta \hat{q}^{-1} \nabla_\alpha u i \hat{q}_\alpha \tilde{v}_z$$
$$+2\hat{q}[\hat{q}^{-1}, u]i\hat{q}_\beta \hat{q} \tilde{v}_z + 2\hat{q}^{-1}[\hat{q}, u]i\hat{q}_\beta \tilde{v}_z$$
$$-2\hat{q}^{-1}\nabla_\beta u \hat{q}^2 \tilde{v}_z$$
$$+2(\nabla u)^2 \hat{q}\tilde{v}_\beta + 3(\nabla u)^2 \hat{q}_\alpha \hat{q}_\beta \hat{q}^{-1} \tilde{v}_\alpha. \qquad (B21)$$

In the conclusion of this appendix we will consider a situation occuring at "high" frequencies defined by the condition

$$\omega \gg \eta q^2/\rho, \qquad (B22)$$

opposite to (B4). Then, in contrast to the previous consideration, near the membrane

$$\tilde{\mathbf{a}} \sim \nabla \tilde{\psi} \sim \tilde{\mathbf{v}}, \qquad (B23)$$

but

$$\nabla_\| \tilde{\mathbf{a}} \gg \nabla^\perp \tilde{\mathbf{a}}, \qquad (B24)$$

where $\nabla_i^\perp$ is defined in (17) and $\nabla_\| = \mathbf{l} \cdot \nabla$. The reason is that $\nabla^\perp \tilde{\mathbf{a}} \sim q\tilde{\mathbf{a}}$, and therefore a solution of Eq. (B3) at the condition (B22) gives

$$\nabla_\| \tilde{\mathbf{a}} = \mp \sqrt{d_t}\tilde{\mathbf{a}}, \qquad (B25)$$

where the upper sign corresponds to points above the membrane and the lower sign to points below the membrane. The operator $\sqrt{d_t}$ in the Fourier representation is reduced to a multiplier

$$\sqrt{-i\omega\rho/\eta},$$

where the function $\sqrt{-i\omega}$ is positive on the positive imaginary semiaxis and has a cut along the negative imaginary semiaxis. We see now that the condition (B24) is a consequence of (B22).

The inequality (B24) together with the conventional relation $\nabla \cdot \tilde{\mathbf{a}} = 0$ leads to a condition

$$\mathbf{l} \cdot \tilde{\mathbf{a}} = 0. \qquad (B26)$$

Therefore $\mathbf{l} \cdot \tilde{\mathbf{v}} = \nabla_\| \tilde{\psi}$. The derivative $\lfloor \nabla_\| \tilde{\psi} \rfloor_\pm$ may be expressed through $\lfloor \tilde{\psi} \rfloor_\pm$ by means of the relation

$$\lfloor \nabla_\| A \rfloor_\pm = \sqrt{1 + (\nabla u)^2} \lfloor \nabla_z A \rfloor_\pm + l_\alpha \nabla_\alpha \lfloor A \rfloor_\pm, \qquad (B27)$$

valid for any function A. This relation is a consequence of the definition $\nabla_\| = \mathbf{l} \cdot \nabla$ and of (B9). Note that the expressions for $\hat{L}_\pm$ in the frequency region (B22) are the same as above, since Eq. (B2) does not contain d_t. Therefore

$$\lfloor \tilde{\psi} \rfloor_\pm = \left(\sqrt{1 + (\nabla u)^2} \hat{L}_\pm + l_\alpha \nabla_\alpha \right)^{-1} \lfloor \mathbf{l} \cdot \tilde{\mathbf{v}} \rfloor_\pm, \qquad (B28)$$

which is a consequence of (B5) and (B27).

At the condition (B22) there is $\nabla_\| \tilde{\mathbf{v}} = \nabla_\| \tilde{\mathbf{a}}$ and, consequently,

$$\lfloor \nabla_\| \tilde{\mathbf{v}} \rfloor_\pm = \mp \sqrt{d_t} \lfloor \tilde{\mathbf{a}} \rfloor_\pm. \qquad (B29)$$

Here we have taken into account the softness of the bending mode, which enables us to neglect terms with $d_t u$ arising at deriving (B29) from (B25). Formulas (B28) and (B29) solve the problem, since, as a consequence of (B1) and (B26) we find

$$\lfloor \tilde{a}_z \rfloor_\pm = \tilde{v}_{sz} - \hat{L}_\pm \lfloor \tilde{\psi} \rfloor_\pm,$$
$$\lfloor \tilde{a}_\alpha \rfloor_\pm = \tilde{v}_{s\alpha} - \nabla_\alpha \lfloor \tilde{\psi} \rfloor_\pm. \qquad (B30)$$

Here $\tilde{\mathbf{v}}_s$ is the surface value of the variable $\tilde{\mathbf{v}}$, which is continuous on the membrane.

Let us consider the term $\lfloor \nabla_\| \tilde{\mathbf{v}} \rfloor_\pm$ zero order in u. In this case from (B26) one can deduce that $\tilde{a}_z = 0$ and from (C5) and (B28) it follows that

$$\lfloor \tilde{\psi} \rfloor_\pm = \mp \hat{q}^{-1} \tilde{v}_\|. \qquad (B31)$$

Then from (B30) we find

$$\lfloor \tilde{a}_\alpha \rfloor_\pm = \tilde{v}_{s\alpha} \pm \nabla_\alpha \hat{q}^{-1} \tilde{v}_\|.$$

Using these relations we find from (B29)

$$\lfloor \nabla_\parallel \tilde{v}_\alpha \rfloor = \lfloor \nabla_\parallel \tilde{v}_\alpha \rfloor_+ - \lfloor \nabla_\parallel \tilde{v}_\alpha \rfloor_-$$

$$= -\sqrt{d_t}(\lfloor \tilde{a}_\alpha \rfloor_+ + \lfloor \tilde{a}_\alpha \rfloor_-) = -2\sqrt{d_t}\tilde{v}_{s\alpha}. \quad (B32)$$

The terms of $\lfloor \nabla_\parallel \tilde{v} \rfloor_\pm$ of higher order in u will not be needed for us at frequencies (B22).

APPENDIX C: INTEGRAL RELATIONS

In this appendix we formulate integral relations for solutions of (B2) and (B3) making it possible to formulate a procedure for deriving expressions for the operators $\hat{L}, \hat{M}$ figuring on the right-hand sides of (B5) and (B6).

For any function $\tilde{\psi}$ satisfying the Laplace equation (B2) one can get the following representation:

$$4\pi\tilde{\psi}(\mathbf{r}') = \int dS \left(\frac{1}{R}\lfloor \nabla_\parallel \tilde{\psi} \rfloor_- + \frac{\mathbf{R} \cdot \mathbf{l}}{R^3}\lfloor \tilde{\psi} \rfloor_- \right). \quad (C1)$$

Here the integration is performed over an infinite surface, $\mathbf{r}'$ is a point "below" the surface, $\nabla_\parallel = \mathbf{l} \cdot \nabla$ and $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, where $\mathbf{r}$ is a point of the surface and $\mathbf{l}$ is the unit

vector normal to the surface. Below we will believe this surface to be determined by the position of the membrane and therefore $\mathbf{l}$ is the unit vector determined by (A2). Recall that $\lfloor \ \rfloor_\pm$ designates the values of a corresponding variable "above" and "below" the membrane.

Expression (C1) gives the value of the function $\tilde{\psi}$ at any point $\mathbf{r}'$ below the membrane as an integral over surface values of the same function $\lfloor \tilde{\psi} \rfloor_-$ and its derivative $\lfloor \nabla_\parallel \tilde{\psi} \rfloor_-$. In the same manner from (B3) we may derive a relation

$$4\pi\tilde{\mathbf{a}}(\mathbf{r}') = \int dS \left(\frac{1}{R}\lfloor \nabla_\parallel \tilde{\mathbf{a}} \rfloor_- + \frac{\mathbf{R} \cdot \mathbf{l}}{R^3}\lfloor \tilde{\mathbf{a}} \rfloor_- \right)$$

$$+ \frac{1}{2}\int dS \left(R\lfloor \nabla_\parallel d_t\tilde{\mathbf{a}} \rfloor_- + \frac{\mathbf{R} \cdot \mathbf{l}}{R}\lfloor d_t\tilde{\mathbf{a}} \rfloor_- \right), \quad (C2)$$

valid up to the first order in d_t.

To derive a relation for the quantity $\lfloor \tilde{\psi} \rfloor_-$, which will be needed for us, we should take in (C1) the point $\mathbf{r}'$ approaching to the membrane from below. Then after the Fourier transformation over x, y we find

$$2\lfloor \tilde{\psi} \rfloor_-(x', y') = \int dx\, dy \int \frac{d^2q}{(2\pi)^2} \exp(-iq_\alpha R_\alpha - qR_z)\sqrt{1 + (\nabla u)^2} \left[\frac{1}{q}\lfloor \nabla_\parallel \tilde{\psi} \rfloor_- + \left(l_z + \frac{iq_\alpha}{q}l_\alpha \right) \lfloor \tilde{\psi} \rfloor_- \right], \quad (C3)$$

where $R_\alpha = r_\alpha - r'_\alpha$ and $R_z = u(r_\alpha) - u(r'_\alpha)$. Performing the same procedure for (C2) we get

$$2\lfloor \tilde{\mathbf{a}} \rfloor_-(x', y') = \int dx\, dy \int \frac{d^2q}{(2\pi)^2} \exp(-iq_\alpha R_\alpha - qR_z)$$

$$\times \sqrt{1 + (\nabla u)^2} \left[\frac{1}{q}\lfloor \nabla_\parallel \tilde{\mathbf{a}} \rfloor_- + \left(l_z + \frac{iq_\alpha}{q}l_\alpha \right) \lfloor \tilde{\mathbf{a}} \rfloor_- \right.$$

$$- \frac{1}{2}[(R_z q^{-2} + q^{-3}) \lfloor \nabla_\parallel d_t\tilde{\mathbf{a}} \rfloor_- + R_z q^{-2}l_z\lfloor d_t\tilde{\mathbf{a}} \rfloor_-$$

$$\left. + (R_z q^{-2} + q^{-3}) l_\alpha iq_\alpha\lfloor d_t\tilde{\mathbf{a}} \rfloor_-] \right]. \quad (C4)$$

To deduce from the relations (C3) and (C4) the expressions for $\hat{L}$ and $\hat{M}$ figuring in (B5) and (B6) we should first perform the substitution (B27). Then the combination $\lfloor \nabla_z \partial \tilde{\mathbf{a}}/\partial t \rfloor_-$ ensuing from (C4) should be substituted by $\hat{L}\lfloor \partial \tilde{\mathbf{a}}/\partial t \rfloor_-$, since the term with $\hat{M}$ may be omitted as a term of the second order in d_t. And finally we should express the integrands in (C3) and (C4) through the left-hand sides of these relations, i.e., to convert the corresponding integral operators. Unfortunately, in the general case the result of this procedure cannot be represented in the compact form.

We will be interested in the first terms of the expansion of (B5) and (B6) in u. These terms may be derived explicitly after the expansion of the relations (C3) and (C4) in u and by converting these relations step by step. Keeping only terms of the zero, first, and second order in u, we find

$$\hat{L}_\pm = \mp\hat{q} - [\hat{q}, u]\hat{q} \pm (\nabla u)^2\hat{q} \pm \frac{1}{2}\hat{q}[u, [u, \hat{q}]]\hat{q}, \quad (C5)$$

$$\hat{M}_\pm = \mp(2\hat{q})^{-1} + \frac{1}{2}\hat{q}[\hat{q}^{-1}, u] + \frac{1}{2}\hat{q}^{-1}[\hat{q}, u]$$

$$\pm \frac{1}{4}\left\{ \hat{q}^{-2}(\nabla u)^2\hat{q} + 2(\nabla u)^2\hat{q}^{-1} - \hat{q}^{-1}[u, [u, \hat{q}]]\hat{q} - \hat{q}[u, [u, \hat{q}]]\hat{q}^{-1} - \hat{q}[u, [u, \hat{q}^{-1}]]\hat{q} \right\}. \quad (C6)$$

We have introduced here the operator $\hat{q}_\alpha = -i\nabla_\alpha$ and the nonlocal operator $\hat{q}$, which corresponds to the multiplier q in the Fourier representation. The square brackets here designate the commutator (e.g., $[\hat{q}, u] = \hat{q}u - u\hat{q}$). Formulas (C5) and (C6) give the expressions for points on the both sides of the membrane.

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