We consider the thermal undulation, or shape fluctuation, of an almost planar fluid membrane surrounded by the same near-critical binary fluid mixtures on both sides. A weak preferential attraction is assumed between the membrane and one component of the mixture. We use the Gaussian free-energy functional to study the equilibrium average of the undulation amplitude within the linear approximation with respect to the amplitude. According to our result given by a simple analytic formula, the ambient near-criticality tends to suppress the undulation of a membrane without the lateral tension. This suppression effect can overwhelm that of the bending rigidity for small wave numbers. Thus, the ambient near-criticality can prevent a large membrane from becoming floppy even if it has no lateral tension.

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I. INTRODUCTION

Amphiphilic molecules can accumulate to form a monolayer at the interface between two phases, working as surfactants, and can also form a bilayer in a one-phase solvent. In either case, a resultant fluid membrane has the restoring force against bending [1], and its shape fluctuates at the equilibrium. Fluid membranes are often stacked regularly to form a lamellar phase because of the balance of their interactions, including the interaction due to steric hindrance of undulating membranes [2–4]. The lamellar structure can work as a photonic device [5].

The thermal undulation, or shape fluctuation, of the lipid-bilayer membrane [6] can explain the flicker phenomenon of red blood cells [7]. When the cell is not swollen, the surface tension, or the lateral tension, of the membrane vanishes because the membrane area is determined so that the free energy is minimized [8–10]. Then, the undulation amplitude is determined by the bending energy and becomes scale invariant. This causes decrease in the effective bending rigidity as the membrane area is larger; a sufficiently large membrane loses its orientation to become floppy [2, 3, 11]. The oil/water interface can have the same property when saturated by surfactants [12, 13].

It is well known that a fluid mixture shows marked concentration fluctuation with longer correlation length as it approaches the demixing critical point. If a colloidal particle is immersed in such a mixture, it has been recently studied in theoretical aspects [17–20]. The concentration gradient due to the adsorption layer generates additional stress including the osmotic pressure, and affects the flow around the particle. Accordingly, for example, the drag coefficient deviates from the Stokes law even if the viscosity is homogeneous in the mixture. Similar deviation for a raftlike region, or a two-dimensional droplet, embedded in a binary fluid membrane is studied in Ref. 21. In fact, the biomembrane has several components; the critical concentration fluctuation of the membrane is measured experimentally, with its possible biological implication suggested [22], and is studied theoretically [23].

In this paper, we consider the thermal undulation of a fluid membrane which is not near critical but is surrounded by near-critical binary fluid mixtures. The ambient near-criticality should influence the undulation when one component of the mixture is preferentially attracted by the membrane. We simplify the problem as follows to study how it influences the average of the undulation amplitude. The temperature is assumed to be homogeneous. We regard the membrane, having a single component, as a thin film fluctuating around a plane, assuming that the typical radius of curvature of the undulation is much larger than the membrane thickness and neglecting the structure of the membrane itself. The semi-infinite regions on both sides of the membrane is assumed to be occupied by incompressible binary fluid mixtures sharing the same properties. Far from the membrane, they are static and in the homogeneous phase near the demixing critical point. Assuming it not to be very close to the critical point, we use the Gaussian free-energy functional. Our calculation is performed within the linear approximation; sufficiently small undulation amplitude and sufficiently weak preferential attraction are assumed.

Our formulation is stated in the next section; some
parts are the same as used in Ref. $17$. The amplitude average considered here is an equal-time correlation at the equilibrium and does not involve the dissipation. Our perturbation scheme is shown in Sect. III, the differential equations are solved in Sect. IV, and the results are shown in Sect. V. The last section is devoted to discussion.

II. FORMULATION

Suppose that the binary fluid mixture consisting of two components $A$ and $B$. We write $\rho_A$ and $\rho_B$ for their mass densities, and $\mu_A$ and $\mu_B$ for the conjugate chemical potentials. In general, introducing the sum $\rho_s = \rho_A + \rho_B$ and the difference $\varphi = \rho_A - \rho_B$, we have

$$\mu_A \delta \rho_A + \mu_B \delta \rho_B = \frac{(\mu_A + \mu_B) \delta \rho_B}{2} + \frac{(\mu_A - \mu_B) \delta \rho_A}{2},$$

(1)

where $\delta$ implies the infinitesimal change. Thus, the intensive variable conjugate to $\varphi$ is $(\mu_A - \mu_B)/2$, which is denoted by $\mu$.

The concentration difference $\varphi$ depends on the position $r$ in the binary mixture. The $\varphi$-dependent part of the free-energy density of the mixture bulk is assumed to be the sum of the term independent of its gradient, denoted by $f$, and the term proportional to its square gradient. This kind of free-energy density is usual in the effective coarse-grained formulation $[24, 25]$. The contribution from the interfaces between the membrane and the mixtures on both sides of the membrane is simply assumed to be given by the surface integral of the potential $f_s$ determined by the value of $\varphi$ immediately near the membrane $[26]$. This potential represents the preferential attraction. These assumptions enable us to write the $\varphi$-dependent part of the free-energy functional of the mixtures as

$$\int_{C^o} dr \left\{ f(\varphi(r)) + \frac{1}{2} M |\nabla \varphi(r)|^2 \right\}$$

$$+ \int_{\partial C} dS f_s(\varphi(r)),$$

(2)

The first term implies the volume integral over the semi-infinite regions ($C^o$) on both sides of the membrane, while the second term implies the surface integral over the interfaces ($\partial C$) on both sides of the membrane. The coefficient $M$ is a positive constant shared by the mixtures on both sides of the membrane. Later we will assume $f$ to be a quadratic function and $f_s$ to be a linear function. The total free-energy functional of our system consists of Eq. (2), the part dependent on $\rho_s$, and the part involving the membrane compressibility and bending.

The undulation deforms the profile of $\varphi$, and changes the value of Eq. (2), which plays a role of a part of the potential energy for the membrane oscillation. This resembles the situation that the membrane is surrounded by elastic medium $[27]$. Here, to calculate the restoring force due to Eq. (2), we need to know how the undulation deforms the profile reversibly. To do so, we consider the reversible, or nondissipative, dynamics of the fluids. The time dependences of $\varphi$ and local intensive variables are thus considered below. The Cartesian coordinate system $(x, y, z)$ is set so that the membrane fluctuates around the $xy$-plane (Fig. 1). The unit vectors along the coordinate axes are denoted by $e_x, e_y$ and $e_z$, respectively. The $z$-coordinate of the membrane is referred to as $\zeta$, which is a function of $(x, y)$ and the time $t$.

![Figure 1](xy-plane membrane z zeta(x,y,t))

We write $\mathbf{V}$ for the velocity field in the mixture. The stress of the fluid is separated in general into the reversible part and the irreversible, or dissipative, part involving the viscosity. We can obtain the reversible part of the pressure tensor $\Pi$ and half the local chemical potential difference $\mu$ by studying how Eq. (2) is changed by an infinitesimal virtual deformation of the fluids. This need not follow the perfect fluid dynamics. In the bulk, the results are the same as those in the model $H$, which is a standard model for the dynamics of a near-critical binary fluid $[25, 28]$, because the same free-energy density is used $[17]$. Below, the prime indicates the derivative with respect to the variable. Introducing

$$p_{\text{osm}} \equiv \varphi f'(\varphi) - f(\varphi),$$

(3)

which is called the osmotic pressure, and

$$\Pi_{\text{grad}} \equiv -(1/2) \left( \nabla \varphi^2 + \varphi \nabla \varphi \right) 1 + M \nabla \varphi \nabla \varphi,$$

(4)

where $1$ denotes the isotropic tensor, we find

$$\Pi = p_0 1 + p_{\text{osm}} 1 + \Pi_{\text{grad}}.$$

(5)

The scalar $p_0$ originally comes from the dependence of the free-energy density on $\rho_s$. As in the previous works $[4, 29, 31]$, we regard $p_0$ as constant with respect to the space and time, which leads to

$$\nabla \cdot \mathbf{V} = 0.$$

(6)

We thus neglect the change in the $p_0$-dependent part of the free-energy density and regard $p_0$ as dependent on $r$ and $t$ irrespective of the local state. We also find

$$\mu(r, t) = f'(\varphi(r, t)) - M \Delta \varphi(r, t),$$

(7)
which satisfies
\[ \varphi \nabla \mu = \nabla p_{\text{osm}} + \nabla \cdot \Pi \nabla \mu. \]  
(8)

We need not assume viscosity to calculate the equal-time correlation. The dynamics of the mixture follows
\[ \rho_b \frac{\partial V}{\partial t} = -\nabla p_b - \varphi \cdot \nabla \mu, \]  
(9)

where the convective term is neglected in anticipation of the later linear approximation. The incompressibility condition Eq. (6) can affect on the trajectory of \( \varphi \).

Far from the membrane, the mixture is assumed to be static and in the homogeneous phase, i.e., \( V \) vanishes and \( \varphi \) is constant. There, each of \( \mu, p_b, \) and \( p_{\text{osm}} \) is constant, considering Eqs. (3), (7), and (9). We assume the symmetric surroundings; the constant values of \( \varphi, \mu, \) and \( p_b \) are respectively denoted by \( \varphi_{\infty}, \mu^{(0)}, \) and \( p_b^{(0)} \), which are shared by the mixtures on both sides of the membrane. The stress exerted on the membrane by the surrounding mixture on the positive-\( z \) (negative-\( z \)) side depends on \( (x, y, t) \) and is denoted by \( F^{(+)} \) (\( F^{(-)} \)). We write \( n \) for the unit vector which is normal to the membrane and is directed towards the positive-\( z \) side, and define the mean curvature of the membrane \( H \) so that its sign is positive when the center of curvature lies on the side towards which \( n \) is directed.

In Eq. (2), \( f_s \) simply represents that part of the free-energy density in the bulk which occurs only near the membrane. Thus, as \( f \) generates the osmotic pressure Eq. (3), \( f_s \) generates two-dimensional pressure working at the interfaces. See Appendix A of Ref. [20] for the detail. We have
\[ F^{(\pm)} = \lim_{z \to \pm_\varepsilon} \left\{ \mp \Pi \cdot n + \nabla \cdot f_s + 2Hf_s n \right\}, \]  
(10)

where \( \nabla_{\parallel} \) implies the projection of \( \nabla \) on the tangent plane and \( z \to \varepsilon_+ (\varepsilon_-) \) means that \( z \) approaches \( \varepsilon(x, y, t) \) with \( z - \varepsilon > 0 \) \( (z - \varepsilon < 0) \) maintained. The last two terms above come from the stress due to the two-dimensional pressure \( -f_s \). The boundary condition
\[ \pm Mn \cdot \nabla \varphi = f_s'(\varphi) \quad \text{as} \quad z \to \varepsilon_{\pm}, \]  
(11)

should hold in the local equilibrium as well as in the global equilibrium [17]. The tangential components of \( F^{(\pm)} \) vanishes; the contribution from \( M \nabla \varphi \nabla \varphi \) of Eq. (11) cancels with the tangential stress due to \( f_s \), as described in Appendix D of Ref. [20].

In the range of \( \varphi \) considered, approximating \( f_s(\varphi) \) to be a linear function, we put the right-hand side (rhs) of Eq. (11) equal to \(-h\), where \( h \) is a constant. We can rewrite Eq. (10) as
\[ F^{(\pm)} = \mp \frac{h^2}{M} n + \lim_{z \to \pm_\varepsilon} \left\{ \mp \rho_b \pm f(\varphi) \mp \frac{M}{2} \varphi^2 \right\} \]  
(12)

In the previous works [17,21], the diffusive flux between the two components is considered. Because it is proportional to the gradient of \( \mu \), the mass conservation leads to
\[ \frac{\partial \varphi}{\partial t} = -V \cdot \nabla \varphi + L \Delta \mu, \]  
(13)

where the Onsager coefficient \( L \) is assumed to be a positive constant. Assuming that the diffusion flux cannot pass across the membrane leads to
\[ n \cdot L \nabla \mu = 0 \quad \text{as} \quad z \to \varepsilon_{\pm}. \]  
(14)

The diffusion should not be involved in the equal-time correlation considered here; we will take the limit of \( L \to 0^+ \) later. Still, we use these two equations at this stage because, as shown later, this limit gives rise to the boundary layer problem, which is unfamiliar in comparison with the problem occurring in the limit of zero viscosity.

Our calculation is performed within the linear approximation with respect to the undulation amplitude. Introducing a dimensionless parameter \( \epsilon \), we define nonzero \( \varepsilon^{(1)} \) so that we have
\[ \varepsilon(x, t) = \epsilon \varepsilon^{(1)}(x, t). \]  
(15)

Hereafter, \( x \) represents a position on the membrane and has coordinates \((x, y)\), in contrast with \( r \) representing a position in the mixture. Up to the order of \( \epsilon \), the components of the metric tensor of the membrane with respect to \( x \) and \( y \) are the same as those of the \( xy \)-plane, the unit normal vector is
\[ n = e_x - \frac{\partial \varepsilon}{\partial x} e_x - \frac{\partial \varepsilon}{\partial y} e_y, \]  
(16)

and the mean curvature is given by
\[ H = \frac{1}{2} \left( \frac{\partial^2 \varepsilon}{\partial x^2} + \frac{\partial^2 \varepsilon}{\partial y^2} \right). \]  
(17)

We write \( v(x, t) \) for the velocity field of the membrane. Assuming it to be compressible, we write \( p(x, t) \) for the membrane mass per unit area, and \( p(x, t) \) for its in-plane pressure field. This field not only comes from the interaction between lipids [2,32] but can contain the interfacial tension between the membrane and the surrounding fluid. The interfacial tension should be distinguished from the stress due to \( f_s \). The former involves the density profile of the lipids across the interface, while the latter does not. We assume that the components of the mixture do not work as surfactants, and thus \( p \) does not depend explicitly on the value of \( \varphi \) immediately near the membrane.

The equations of motion for a viscous compressible membrane can be found in the previous works [29,32].
Neglecting the membrane viscosity and using the approximate geometrical quantities above, we can write the momentum conservation in the tangential direction as

\[
\rho \frac{\partial v_x}{\partial t} = F_x - \frac{\partial p}{\partial x} \quad \text{and} \quad \rho \frac{\partial v_y}{\partial t} = F_y - \frac{\partial p}{\partial y} \quad (18)
\]

up to the order of \( \epsilon \). Here, \( F \equiv F^{(+)} + F^{(-)} \) denotes the total stress exerted on the mixtures. Assuming the spontaneous curvature to vanish, we write \( cH^2 \) for the bending energy per unit area of the membrane, where \( c \) is the bending rigidity. The restoring force is normal to the membrane, and its component along \( n \) is given by

\[
F_n = -c \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) H \quad (19)
\]

Up to the order of \( \epsilon \), the momentum conservation in the normal direction is represented by

\[
\rho \frac{\partial v_z}{\partial t} = F_z + F_r = 2\eta \frac{\rho}{\partial t} + 2\eta \frac{\rho v_z}{\partial t} - 2\eta \frac{\rho v_z}{\partial t} \quad (20)
\]

while the mass conservation is represented by

\[
\frac{\partial p}{\partial t} = -\frac{\partial \rho v_x}{\partial x} - \frac{\partial \rho v_y}{\partial y} \quad (21)
\]

The limit of zero viscosity in the mixture causes the well-known boundary layer problem of the velocity field, which we deal with by imposing the slip boundary condition between the membrane and the inviscid fluid. We proceed with the calculation after taking this limit, and evaluate \( F^{(\pm)} \) immediately outside these boundary layers on both sides of the membrane. The tangential components of the velocity need not be continuous across the membrane, while the normal component is continuous. In the limit of \( L \to 0^+ \), as is shown later, \( \varphi \) and \( \mu \) have boundary layers. However, at this stage, we do not take this limit and their spacial profiles have no rapid changes near the membrane.

### III. PERTURBATION

In the unperturbed state \( (\epsilon = 0) \), where the membrane is fixed on the \( xy \)-plane, \( \mu \) is homogeneous over a mixture region \( [17] \) and so is \( \rho_b \) because of Eq. (11). They are respectively given by the constants \( \mu^{(0)} \) and \( \rho_b^{(0)} \). Up to the order of \( \epsilon \), we expand the fields as

\[
\varphi(r, t) = \varphi^{(0)}(z) + \epsilon \varphi^{(1)}(r, t) \quad , \\
\mu(r, t) = \mu^{(0)} + \epsilon \mu^{(1)}(r, t) \quad , \\
p_b(r, t) = p_b^{(0)} + \epsilon p_b^{(1)}(r, t) \quad , \\
\text{and} \quad V(r, t) = \epsilon V^{(1)}(r, t) \quad .
\]

On the rhs of each of these equations, the field with the superscript \( (0) \) is defined so that it is independent of \( \epsilon \), while the field with the superscript \( (1) \) is defined so that it becomes proportional to \( \epsilon \) after being multiplied by \( \epsilon \). As shown later, \( \varphi^{(0)} \) depends only on \( z \). For the membranous fields, we use similar expansions,

\[
\rho(x, t) = \rho^{(0)} + \epsilon \rho^{(1)}(x, t) \quad , \\
p(x, t) = p^{(0)} + \epsilon p^{(1)}(x, t) \quad , \\
v(x, t) = v^{(1)}(x, t) \quad , \\
\text{and} \quad F(x, t) = \epsilon F^{(1)}(x, t) \quad ,
\]

where \( \rho^{(0)} \) and \( p^{(0)} \) are constants.

#### A. Unperturbed state

We here consider the equilibrium profile of \( \varphi \) with the membrane fixed on the \( xy \)-plane; this situation is essentially the same as argued in Ref. [26]. From Eq. (7), we have

\[
f'\varphi^{(0)} - M \Delta \varphi^{(0)} = \mu^{(0)} \quad \text{for} \ z \neq 0 \quad .
\]

The correlation length far from the membrane,

\[
\xi_c = \sqrt{\frac{M}{f''(\varphi_{\infty})}} \quad ,
\]

is assumed to be much larger than the microscopic length; the free-energy functional Eq. (2) is a result of coarse-graining. Here, \( f''(\varphi_{\infty}) \) is positive because of the thermodynamic stability. Equation (11) leads to

\[
M \frac{\partial}{\partial z} \varphi^{(0)} = \mp h \quad \text{as} \ z \to 0 \pm \quad (26)
\]

Linearizing Eq. (21) by approximating \( f'(\varphi) \) as \( \mu^{(0)} + f''(\varphi_{\infty})(\varphi - \varphi_{\infty}) \), we obtain the equilibrium profile,

\[
\varphi^{(0)}(z) = \varphi_{\infty} + \frac{h \xi_c}{M} e^{-|z|/\xi_c} \quad (27)
\]

for \( z \neq 0 \). The preferential attraction, represented by \( h \), causes the concentration difference to deviate from its value far from the membrane; the characteristic length is given by the bulk correlation length \( \xi_c \). The approximation is valid when

\[
|h f''(\varphi_{\infty})| \xi_c \ll M f''(\varphi_{\infty}) \quad ,
\]

as discussed in Ref. [17]. We later use the Gaussian model, where Eq. (27) becomes exact.

#### B. Terms at the order of \( \epsilon \)

From Eqs. (11) and (24), we have

\[
\lim_{z \to 0^\pm} \frac{\partial \varphi^{(1)}}{\partial z} = -\xi^{(1)} \lim_{z \to 0} \varphi^{(0)''}(z) \quad .
\]

\[
\frac{\partial \varphi^{(1)}}{\partial z} = -\xi^{(1)} \lim_{z \to 0} \varphi^{(0)''}(z) \quad .
\]
Considering Eq. (14), we have
\[ L \frac{\partial \mu^{(1)}}{\partial z} \to 0 \quad \text{as} \quad z \to 0 \pm . \] (30)

Up to the order of \( \epsilon \), we have
\[ f(\varphi(\zeta +)) = f(\varphi^{(0)}(\zeta +)) + \epsilon \varphi^{(1)}(0+) f'(\varphi^{(0)}(0+)) , \] (31)
where \( \varphi \) and \( \varphi^{(1)} \) depend on \( (r, t) \), \( \zeta \) depends on \( (x, t) \), and \( \varphi(\zeta +) \) means \( \lim_{z \to \zeta^+} \varphi(x, y, z, t) \), while the first term on the rhs above equals
\[ f(\varphi^{(0)}(0+)) + \epsilon \varphi^{(1)}(0+) f'(\varphi^{(0)}(0+) ) . \] (32)
Calculating similarly the other terms in Eq. (12), we use Eqs. (24), (27), and (29) to obtain \( F_x^{(1)} = F_y^{(1)} = 0 \) and
\[ F_z^{(1)} = \left[ -\frac{h^{(1)}_0 - \mu^{(1)}(\varphi^{(0)})}{\xi^2} \right] \pm \left\{ -\frac{2h^2 \xi^2}{M \xi^2} + 4H^{(1)} f_s(\varphi^{(0)}(0+)) \right\} , \] (33)
where \( \cdot \cdot \cdot \) is defined as \( \lim_{x \to 0^+} \cdots - \lim_{x \to 0^-} \cdots \), and \( H^{(1)} \) is defined as Eq. (17) with \( \zeta \) replaced by \( \zeta^{(1)} \). As is mentioned at the end of Sect. II, \( z \to 0 \pm \) above means that the stress is evaluated immediately outside the boundary layers occurring in the limit of zero viscosity.

In the directions of \( x \) and \( y \), we impose the periodic boundary condition, e.g.,
\[ \tilde{p}_b^{(1)}(k, z, t) \equiv \frac{1}{2} \int_{-l/2}^{l/2} dx \int_{-l/2}^{l/2} dy \tilde{p}_b^{(1)}(x, z, t) e^{-ik x} , \] (34)
where \( k \) represents \( (k_x, k_y) \) with \( lk_x/(2\pi) \) and \( lk_y/(2\pi) \) being integers and the period \( l \) is assumed to be sufficiently large. We have
\[ \lim_{z \to 0^\pm} \tilde{V}^{(1)}_z = \tilde{\xi}^{(1)} = \frac{\partial \zeta^{(1)}}{\partial t} . \] (35)

We add an overtile to the further Fourier transform with respect to \( t \), e.g.,
\[ \tilde{p}_b^{(1)}(k, z, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \tilde{p}_b^{(1)}(k, z, t) e^{i\omega t} . \] (36)
Using \( k \equiv \sqrt{k_x^2 + k_y^2} \), we define
\[ \tilde{V}^{\parallel}_x \equiv \left( k_x \tilde{V}_x + k_y \tilde{V}_y / k , \right. \] (37)
and define \( \tilde{v}_y \) similarly. From Eq. (27), we obtain
\[ -i\omega \rho_b \tilde{V}^{(1)}_\parallel = -ik \tilde{p}_b^{(1)} - ik \varphi^{(0)} \tilde{\mu}^{(1)} \] (38)
and
\[ -i\omega \rho_b \tilde{V}^{(1)}_z = -\frac{\partial \tilde{p}_b^{(1)}}{\partial z} - \varphi^{(0)} \frac{\partial \tilde{\mu}^{(1)}}{\partial z} . \] (39)
The solenoidal component \( k_x \tilde{V}_y - k_y \tilde{V}_x \) is time-invariant irrespective of the dynamics above in the inviscid mixture, and is assumed to vanish in the calculation for the equal-time correlation. Equation (10) leads to
\[ i k \tilde{V}^{(1)}_\parallel + \frac{\partial \tilde{V}^{(1)}_\parallel}{\partial z} = 0 . \] (40)

Deleting \( \partial \tilde{p}_b^{(1)}/\partial z \) from Eq. (39) and the \( z \)-derivative of Eq. (38), we use Eq. (40) to derive
\[ \left( \frac{\partial^2}{\partial z^2} - k^2 \right) \tilde{V}^{(1)}_z = -\frac{i k^2}{\rho_b \varphi} \tilde{\mu}^{(1)} . \] (41)
A boundary condition is given by the Fourier transform of Eq. (35) with respect to \( t \). The fields with the superscript \( (1) \) in Eq. (22) vanish far from the membrane. From Eq. (7), we have
\[ \left\{ M \Delta - f''(\varphi^{(0)}) \right\} \varphi^{(1)} = -\mu^{(1)} . \] (42)
Substituting Eq. (22) into Eq. (13), we pick up an equation at the order of \( \epsilon \). The first term on the rhs of Eq. (13) generates \(-\epsilon \tilde{V}_z^{(1)} \varphi^{(0)} \) in the resultant equation, the Fourier transform of which gives
\[ -i\omega \varphi^{(1)} = -\tilde{V}_z^{(1)} \varphi^{(0)} + L \left( \frac{\partial^2}{\partial z^2} - k^2 \right) \tilde{\mu}^{(1)} . \] (43)
To solve these three simultaneous equations, we assume the weak preferential attraction and the Gaussian model, as described below. Let us introduce a dimensionless parameter,
\[ \lambda \equiv \frac{h^2 \xi^2}{\sqrt{c/M}} , \] (44)
which is assumed to be sufficiently small in calculating Eq. (33).

The conditions and equations for \( \mu^{(1)} \) and \( \varphi^{(1)} \), given in the preceding paragraph and by Eqs. (29) and (30), are satisfied by \( \mu^{(1)} = \varphi^{(1)} = 0 \) when \( h \) vanishes, considering Eq. (27). Thus, using \( Z \equiv z/\xi_c \), we can introduce dimensionless fields,

\begin{align*}
Q(Z, \omega) &\equiv \frac{\xi^2 \tilde{\mu}^{(1)}(k, z, \omega)}{h \xi_c^{(1)}(k, \omega)} , \\
G(k, Z, \omega) &\equiv \frac{M \tilde{\varphi}^{(1)}(k, z, \omega)}{h \xi_c^{(1)}(k, \omega)} , \\
and \quad U(k, Z, \omega) &\equiv \frac{i \tilde{V}_z^{(1)}(k, z, \omega)}{\omega \xi_c^{(1)}(k, \omega)} ,
\end{align*}
(45)
where the kernel is defined as
\[ (\partial^2_Z - K^2) U(Z) = \mp \lambda^2 A Q(Z) e^{\mp Z} \] (46)
for \( Z > 0 \), where \( K \equiv k \xi \) and \( A \equiv \epsilon k^2 / (\rho \omega^2 \xi^3) \).

With the viscosity considered, Eq. [46] would have some terms multiplied by the viscosity coefficient which include a higher derivative of \( U \) with respect to \( Z \).

As mentioned at the end of Sect. [III] we consider the solution in the limit of zero viscosity, \( \epsilon \), we consider only the regions outside the resultant boundary layers. Thus, no more boundary layer of \( U \) remains in Eq. [46].

Because of Eq. [39], we have
\[ U(k, Z, \omega) \to 1 \quad \text{as} \quad Z \to 0 \pm . \] (47)

Thus, applying the method of variation of parameters to Eq. [40], we obtain for \( Z > 0 \)
\[ U(Z) = \left\{ 1 - \frac{\lambda^2 A}{2K} \int_{0}^{\infty} dZ_1 Q(Z_1) e^{-(K+1)Z_1} \right\} e^{-KZ} \]
\[ -\lambda^2 A \int_{-\infty}^{0} dZ_1 \Gamma_K(Z, Z_1) Q(Z_1) e^{-Z_1}, \] (48)
where the kernel is defined as
\[ \Gamma_K(Z, Z_1) = -\frac{1}{2K} e^{-K\mid Z - Z_1 \mid} \] (49)
for \( Z > 0 \) and \( Z_1 > 0 \). For \( Z < 0 \), we likewise find
\[ U(Z) = \left\{ 1 + \frac{\lambda^2 A}{2K} \int_{0}^{\infty} dZ_1 Q(Z_1) e^{(K+1)Z_1} \right\} e^{KZ} \]
\[ +\lambda^2 A \int_{-\infty}^{0} dZ_1 \Gamma_K(Z, Z_1) Q(Z_1) e^{Z_1}, \] (50)
where the kernel is also given by Eq. [49] for \( Z < 0 \) and \( Z_1 < 0 \). Thus, we have
\[ U(Z) = e^{-K\mid Z \mid} + O(\lambda^2), \] (51)
where \( O(\lambda^2) \) represents the term whose quotient divided by \( \lambda^2 \) does not diverge in the limit of \( \lambda \to 0 \). Later we use the term independent of \( \lambda, e^{-K\mid Z \mid} \), to calculate \( Q \) and \( G \).

The free-energy functional Eq. [2] is considered as obtained after renormalization up to the correlation length \( \xi_0 \). Assuming the correlation length to be so short that the higher-order terms are negligible in \( f \), we use the Gaussian model
\[ f(\varphi) = \frac{1}{2} (\varphi - \varphi_\infty)^2 + \mu(0) (\varphi - \varphi_\infty), \] (52)
where \( a \) is a positive constant. This constant, being the reciprocal susceptibility, can be assumed to be proportional to the temperature measured from the critical point. Using Eq. [52] in Eq. [2] amounts to assuming that the mixture is near, but not very close to, the demixing critical point \( [25] \). Equations [29] and [42] respectively become
\[ \lim_{Z \to 0} \partial_Z G(Z) = -1 \] (53)
and
\[ (\partial^2_Z - K^2) G(Z) = -Q(Z), \] (54)
where \( K_1 \equiv \sqrt{K^2 + 1} \). Thus, we find
\[ G(Z) = \left\{ 1 + \frac{1}{2} \int_{0}^{\infty} dZ_1 e^{-K_1 Z_1} Q(Z_1) \right\} e^{-K_1 Z} \]
\[ -\int_{0}^{\infty} dZ_1 \Gamma_{K_1}(Z, Z_1) Q(Z_1) \] (55)
for \( Z > 0 \), and
\[ G(Z) = \left\{ 1 + \frac{1}{2} \int_{-\infty}^{0} dZ_1 e^{-K_1 Z_1} Q(Z_1) \right\} e^{-K_1 Z} \]
\[ -\int_{-\infty}^{0} dZ_1 \Gamma_{K_1}(Z, Z_1) Q(Z_1) \] (56)
for \( Z < 0 \). Using \( \nu \equiv i LM / (\omega \xi^2) \), we rewrite Eq. [30] as
\[ \nu \partial_Z Q(Z) \to 0 \quad \text{as} \quad Z \to 0 \pm . \] (57)
From Eq. [43], we obtain for \( \pm Z > 0 \)
\[ \nu (\partial^2_Z - K^2) Q(Z) = \mp U(Z) e^{-|Z|} + G(Z). \] (58)
Substituting Eq. [54] into Eq. [58], we obtain
\[ -Q(Z) = \nu (\partial^2_Z - K^2) (\partial^2_Z - K^2) Q(Z) \]
\[ \pm 2K e^{-(K^2 + 1)|Z|} + O(\lambda^2) \] (59)
with the aid of Eq. [51]. We are interested in the limit of \( L \to 0+, \text{ i.e.,} -i \nu \to 0+, \) in Eq. [59], which gives the singular perturbation problem \( [31] \). For \( 0 < |Z| \ll 1 \), we introduce \( u \equiv -\nu^{-1/4} Z \) and \( q(u) \equiv Q(\nu^{1/4} u) \) to rewrite Eq. [59] as
\[ -q(u) = \left( \frac{\partial^2}{\partial u^2} - \sqrt{\nu} K_1^2 \right) \left( \frac{\partial^2}{\partial u^2} - \sqrt{\nu} K_2^2 \right) q(u) \]
\[ \pm 2K \exp \left\{ -(K + 1) \nu^{1/4} |u| \right\} + O(\lambda^2), \] (60)
which has regular solutions of \( q \) even in the limit of \( -i \nu \to 0+ \). The highest derivative is free from \( \nu \) in the above, unlike in Eq. [59]. Thus, on each side near \( Z = 0, \) there is a boundary layer, whose thickness tends to zero as \( L \to 0+. \) In this limit, considering Eq. [59], \( Q(Z) \) is asymptotically equal to
\[ \pm 2K e^{-(K^2 + 1)|Z|} + O(\lambda^2) \] (61)
for $Z > 0$ outside the thin layers. This outer solution satisfies the boundary condition for $|Z| \to \infty$, mentioned just below Eq. (53), while does not satisfy Eq. (57). An alternative way to Eq. (58) is as follows. The boundary layer of $Q(Z)$ yields that of $G(Z)$ because of Eq. (54). The outer solution of $G(Z)$ is $\pm U(Z)e^{-|Z|}$ considering Eq. (58). Substituting this into Eq. (54) gives the outer solution of $G(Z)$, i.e., Eq. (51), with the aid of Eq. (54). Thus, once the boundary layers are recognized, Eqs. (59) and (60) are dispensable in deriving Eq. (61).

We define $Q_{\text{in}}(Z)$ so that $Q(Z)$ equals the sum of $Q_{\text{in}}(Z)$ and Eq. (61); $Q_{\text{in}}(Z)$ rapidly becomes zero as $|Z|$ increases beyond the thickness of the boundary layer. In the limit of $L \to 0+$, substituting the sum into Eq. (59) gives

$$G(Z) = e^{-(K+1)Z} - e^{-K Z} \left\{ K - \int_0^\infty dZ_1 Q_{\text{in}}(Z_1) \right\} + O(\lambda^2) \quad (62)$$

for $Z > 0$ outside the boundary layer. There, we should have $G(Z) = U(Z)e^{-Z}$ from Eq. (58). Thus, we use Eqs. (51) to find

$$\lim_{L \to 0^+} \int_0^\infty dZ_1 Q_{\text{in}}(Z_1) = K + O(\lambda^2) ,$$

and thus Eq. (63) in the limit of $Z \to 0+$ gives

$$\lim_{L \to 0^+} G(0+) = 1 + O(\lambda^2) . \quad (64)$$

This happens to be equal to the same limit of the outer solution of $G(Z)$. If $L$ were assumed to vanish from the beginning, Eq. (57) would be trivial and Eq. (58) would give $G(Z) = \pm U(Z)e^{-|Z|}$ for $\pm Z > 0$. This overall solution contradicts with Eqs. (53) and (54); substituting this solution into Eq. (54) and substituting the resultant $Q$ into Eq. (55) gives Eq. (62) without the integral of $Q_{\text{in}}$ for $Z > 0$, i.e., a different solution of $G$. This means that we cannot assume $L$ to vanish from the beginning. We use Eq. (29) to derive Eq. (33) because of the statements in the last paragraph of Sect. III. Thus, we cannot take the limit of $L \to 0+$ before taking the limit of $Z \to 0\pm$ in evaluating $F(\pm)$ in the reversible dynamics.

Using Eqs. (18), (61) and (63), we obtain

$$\lim_{L \to 0^+} \lim_{Z \to 0^+} \partial_Z U(Z) = -K - \frac{\lambda^2 A K^2}{K + 1} + \lambda^2 A \int_0^\infty dZ_1 Q_{\text{in}}(Z_1) + O(\lambda^4)$$

$$= -K + \frac{\lambda^2 A K^2}{K + 1} + O(\lambda^4) . \quad (65)$$

For $Z < 0$, using the procedure leading to Eq. (63), we obtain

$$\lim_{L \to 0^+} \int_{-\infty}^0 dZ_1 Q_{\text{in}}(Z_1) = -K + O(\lambda^2) . \quad (66)$$

Thus, considering Eqs. (18), (50), (61) and (66), we find $U(Z)$ to be even with respect to $Z$ up to the order of $\lambda^2$. With the aid of Eq. (41), $V_j(z)$ is found to be odd with respect to $z$ up to this order, in spite of which $v_j$ does not vanish because of the slip boundary condition. From Eqs. (55), (60), (61), (63) and (66), $G(Z)$ turns out to be odd with respect to $Z$ up to the order of $\lambda$. There are three terms on the rhs of Eq. (53). The Fourier transform of its first term is thus found, with the aid of Eqs. (59) and (60), to be given by

$$2 \lim_{z \to 0^+} \left\{ \left( -\frac{i \omega p_0}{k^2} \right) \frac{\partial V_j^{(1)}(1)}{\partial z} + \frac{\hbar \phi^{(1)}(1)}{\xi_c} \right\} = \frac{2 \xi_c^{(1)}(1)}{\xi_c^4} \lim_{z \to 0^+} \left\{ - \frac{\partial_z U(Z)}{A} + \lambda^2 G(Z) \right\} \quad (67)$$

up to the order of $\lambda^3$. Substituting Eqs. (59) and (65) into Eq. (67), we find the Fourier transform of the sum of the first and second terms on the rhs of Eq. (53) in the limit of $L \to 0^+$ to be

$$2 \rho_0 \omega \xi_c^{(1)}(1) - \frac{2 \xi_c^{(1)}(1)}{\xi_c^4} \lambda^2 d(K) \quad (68)$$

up to the order of $\lambda^3$, where

$$d(K) \equiv \frac{K^2}{K + 1} . \quad (69)$$

Equation (68) originates from Eq. (4); the term $M \nabla \phi \nabla \phi$ in Eq. (4) does not contribute to this result, as mentioned below Eq. (11). Which component is preferred by the membrane is not involved in deriving Eq. (68), which does not contain a term with odd powers of $h$. Using Eqs. (68) and (69), we can evaluate $F$ in the limit of $L \to 0+$.

Introducing the isothermal compressibility of the membrane $\kappa$, we assume $\kappa^{(1)} = \rho^{(1)}(1)/\rho^{(1)}(0)$. Noting the statement above Eq. (33), we use Eqs. (18) and (21) to obtain

$$\frac{\partial}{\partial \phi} \tilde{\rho}^{(1)} = -i k \rho^{(1)}(1) \parallel \frac{\partial}{\partial \phi} \tilde{\phi}^{(1)} = -i k \rho^{(1)} \kappa \tilde{\rho}^{(1)} , \quad (70)$$

which describe the nondissipative oscillation in the tangential direction. That in the normal direction, independent of Eq. (70), leads to the equilibrium average of the undulation amplitude, as shown in the next section.

V. RESULTS

The stress $-\rho^{(0)}$ gives the lateral tension or surface tension referred to in Refs. [2, 3, 4, 9, and 32] where the preferential attraction is not considered. It vanishes when the membrane is not forced to be stretched or compressed, as is mentioned in the second paragraph of Sect. I. Then,
if the preferential attraction occurs in the ambient criticality, the total stress
\[ \sigma \equiv -p(0) + 2f_x(\varphi(0)(0+)) \] (71)
vanishes. The factor 2 above comes from the two interfaces on both sides of the membrane. Thus, in more general, \( \sigma \) is regarded as the lateral tension.

The Fourier transform of Eq. (19) is given by \(-ck^4\tilde{\zeta}/2\) because of Eq. (17). Equation (20) yields
\[ -i\omega p(0)\tilde{v}_z = \tilde{F}^{(1)} - \left( \frac{ck^4}{2} - p(0)k^2 \right) \tilde{\zeta}^{(1)} , \] (72)
which is combined with Eqs. (35) and (73) to yield
\[ \rho_k^{(\text{eff})} \frac{\partial \tilde{v}_z^{(1)}}{\partial t} = - \left\{ \frac{c}{\xi_c^2} \left[ \frac{K^4}{2} + 2\lambda^2 d(K) \right] + \sigma k^2 \right\} \tilde{\zeta}^{(1)} \] (73)
up to the order of \( \lambda^3 \). Here, we use \( \rho_k^{(\text{eff})} \equiv \rho(0) + 2\rho^\varphi/k, \) the second term of which represents the induced mass \( \rho^\varphi \). Equations (35) and (73) describe the nondissipative oscillation in the normal direction. We thus find
\[ \frac{1}{2} \left\{ \rho_k^{(\text{eff})} \left| \tilde{v}_z^{(1)}(k,t) \right| ^2 + \left( \frac{ck^4}{2} + \sigma k^2 + \frac{2h^2}{M\xi_c} d(k\xi_c) \right) \left| \tilde{\zeta}^{(1)}(k,t) \right| ^2 \right\} \] (74)
to be time-independent. As shown in the next paragraph, the above represents the total energy of the oscillation in the normal direction. Let \( \langle \cdots \rangle \) indicate the equilibrium average at the temperature \( T \), and \( k_B \) denote the Boltzmann constant. Using the equipartition theorem, we find
\[ \langle \tilde{\zeta}(k,t)\tilde{\zeta}(k',t) \rangle = \delta_{k,-k'} k_B T \left( \frac{ck^4}{2} + \sigma k^2 + \frac{2h^2}{M\xi_c} d(k\xi_c) \right)^{-1} \] (75)
up to the order of \( h^3 \), where \( d \) is defined by Eq. (69). This is our main result. The average of the squared undulation amplitude can be calculated from Eq. (75) by means of
\[ \langle \tilde{\zeta}(x,t)\tilde{\zeta}(x,t) \rangle = \sum_k \langle \tilde{\zeta}(k,t)\tilde{\zeta}(-k,t) \rangle . \] (76)

Only in this paragraph, we suppose an external stress field exerted on the membrane. We write \( \eta_z(x,t) \) for its \( z \)-component, which should be added to the rhs of Eq. (20). Its Fourier transform \( \tilde{\eta}_z(k,t) \) should appear on the rhs of Eq. (73) multiplied by \( \epsilon \). Let us multiply this modified equation with \( l^2\tilde{v}_z(-k,t) = l^2\tilde{v}_z^{(1)}(-k,t) \) and sum the resultant product over \( k \). Then, with the aid of Eq. (55), we find the time derivative of Eq. (74) to be given by
\[ l^2 \sum_k \eta_z(k,t)\tilde{v}_z(-k,t) \]
\[ = \int_{-l/2}^{l/2} dx \int_{-l/2}^{l/2} dy \eta_z(x,t)v_z(x,t) , \] (77)
which is the work done to the membrane per unit time by the external stress field. This means that Eq. (74) is the total energy, or the effective Hamiltonian, of the oscillation in the normal direction.

Judging from Eq. (69), \( d(K) \) is positive and increases with \( K = k\xi_c. \) Thus, the term involving \( d \) in Eq. (75) tends to suppress each wave-number component of the undulation amplitude. The suppression is more remarkable as \( k \) is larger for a given \( \xi_c \). Equation (69) leads to \( d(K) \approx K^2 \) as \( K \) is smaller and \( d(K) \approx K \) as \( K \) is larger, as shown in Fig. 2. We use the hydrodynamics based on Eqs. (2) and (52). Considering the statement above Eq. (52), our result ceases to be valid for the length scale much smaller than the correlation length, \( \xi_c \), for \( K \approx 1 \). Suppose that \( K \) is small enough to give our reliable result \( d(K) \approx K^2 \), and Eq. (75) informs that the suppression is more remarkable as \( \xi_c \) is larger and as \( h^2 \) is larger. Then, the adsorption layer is also remarkable in its thickness and amplitude, considering Eq. (27).

VI. DISCUSSION

The average of the undulation amplitude is not determined only by the lateral tension and bending energy of the membrane in particular when the undulation causes a significant change in \( \varphi \). This surely occurs when near-critical binary fluid mixtures surround the membrane with the preferential attraction to make the adsorption layer remarkable. To find the change, we study the reversible dynamics by considering Eq. (2) and the bending energy of the membrane. We assume the fluids to be inviscid from the beginning and then take the limit of \( L \to 0^+. \) Discontinuous motion is inevitable immediately near the membrane in these limits for the reversibility. The incompressibility is assumed for the surrounding fluids. Using all these conditions, we can...
determine the reversible oscillation of the profile of ϕ associated with that of the membrane shape ζ.

The equal-time correlation can be calculated in terms of the statics. However, the sum of Eq. (2) and the bending energy of the membrane does not have all the information. Formally, as shown in Appendix, we can calculate how the sum is changed by changes in ϕ and ζ up to the second order with respect to them, and integrate out the former to obtain the effective Hamiltonian only for the latter. This integration amounts to specifying the profile of ϕ for a given ζ, considering that the involved probability distribution is Gaussian. In the oscillation of ζ, the trajectory of ϕ thus determined is not necessarily the same as considered in our formulation, where the discontinuous motion and the incompressibility give constraints on the trajectory. When the membrane is surrounded by incompressible one-component fluids without ϕ, we need not care their motion in calculating the undulation amplitude at the equilibrium because the motion contributes only to the kinetic energy. However, it is not the case in our problem, where ϕ influences the stress exerted on the membrane and the potential-energy part, dependent on the shape ζ, in the effective Hamiltonian. In this paper, we calculate the dependence of ϕ on ζ by using the reversible dynamics, instead of studying the free-energy functional including all the required information in terms of the statics.

As mentioned at the last paragraph in the preceding section, the ambient near-criticality tends to suppress the undulation amplitude more remarkably as the adsorption layer is more remarkable. As far as our result remains valid, the suppression is also more remarkable as the wave number of the undulation is larger. Then, like the membrane, the adsorption layer would wrinkle more severely. If there is no preferential attraction, Eq. (75) is reduced to the previous result [2, 3, 7]. Then, Eq. (76) can be calculated as

\[
\frac{k_B T}{(2\pi)^2} \frac{1}{2l} \int dk \frac{1}{2\pi k} \left( \frac{ck^4}{2} - p^{(0)} k^2 \right)^{-1},
\]

(78)

where s is the lower cut-off length. For the membrane without the lateral tension, we neglect \( p^{(0)} \) to obtain

\[
\langle \zeta(x, t) \zeta(x, t) \rangle = \frac{k_B T}{2\pi c} \left( \frac{l}{2\pi} \right)^2,
\]

(79)

which implies that the averaged undulation amplitude is scale invariant [2]. This is derived by the \( k^{-3} \) dependence of the integrand of Eq. (78), which dependence comes from the bending energy. If the preferential attraction occurs to give \( h \neq 0 \) and if the lateral tension vanishes to give \( \sigma = 0 \), the last term \( \propto d(K) \propto k^2 \) can overwhelm the term \( c k^4/2 \) in the brackets of Eq. (75) for small values of \( k \). Thus, even if there is no lateral tension, the ambient near-criticality combined with the preferential attraction can suppress the small-wavenumber components of the undulation more than the bending rigidity, and can prevent a large membrane from becoming floppy.

Around the room temperature, for example, aqueous solutions of 2-methyl propanoic acid and 1-propanol respectively have the upper and lower consolute points \[37\]. It is probable, however, the structure of the lipid-bilayer membrane is disordered when it is immersed in either of these solutions, considering that it has the affinity to alcohol \[38\]. Thus, these solutions would not be available for experimental check of our result. In the coacervation of aqueous solutions of elastin-related polypeptides, the lower consolute points are around the room temperature \[39\]. The vesicle made of the lipid-bilayer membrane can contain polyethylene glycol and dextran aqueous solution \[40\], which has the demixing critical point around the room temperature \[41\]. Our result may be observed in either of these polymer solutions if not blurred by the polymer dynamics. Sodium dodecyl-sulphate, water and pentanol form a lamellar phase with dodecane being the solvent \[42\]. Adding some fluorinated hydrocarbon to the solvent, we may check our result experimentally, considering that perfluorohexane and iso-octane has the upper consolute point around the room temperature \[43\].

Our theory presupposes a sheet of the membrane, on both sides of which the semi-infinite regions are occupied by fluids sharing the same properties. This presupposition of symmetric surroundings should be given up in considering the surfactant monolayer at the oil/water interface. The near-criticality on one side can be also expected to suppress the undulation amplitude, which remains to be studied. It is interesting to calculate how the interval between stacked membranes is changed by the near criticality of the intercalated fluids. To do this, we should study the finite-size effect of the surrounding fluids by also considering the interaction between membranes due to the critical adsorption \[10\]. This study may suggest realization of a photonic device responding to small temperature change.

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Appendix: Fallacious Effective Hamiltonian

Equation (2) is a functional dependent on ϕ and ζ; we add the subscript ζ to \( C^\infty \) and \( \partial C \) to specify the regions for a given membrane shape ζ. The corresponding grand-
potential functional is given by
\[ \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) , \] (A.1)
where \( \bar{f} \) is defined as the difference of \( \mu^{(0)}(\phi) \) subtracted from the integrand of the first integral in Eq. (2). We use Eq. (52) and regard \( f_s \) as the linear function mentioned above Eq. (12). The deviation of \( \zeta \) from zero and that of \( \phi \) from \( \phi(0) \), denoted by \( \varphi_1 \), cause the deviation of the grand-potential functional. We obtain this deviation, denoted by \( \delta \Omega \), by subtracting
\[ \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \] (A.2)
from Eq. (A.1). If necessary to clarify the description, the superscript \( \dagger \) \( (\dagger) \) is added to a quantity and the region of the surrounding fluid on the positive-\( z \) (negative-\( z \) side).

We rewrite the first term of Eq. (A.2) as the sum of
\[ \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \]
and the integral of
\[ \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \]
(A.3)
with respect to \( x \) and \( y \) over the region considered in Eq. (34). Thus, subtracting Eq. (A.2) from Eq. (A.1), we encounter a term
\[ \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \]
(A.4)
the integrand of which is rewritten as
\[ \frac{1}{2} \left[ \frac{\partial}{\partial x} \right]^2 + \frac{\partial}{\partial y} \left( \phi_1 \right) + \frac{c}{\zeta} \left( \phi_1 \right) \]
with the aid of Eq. (24). Up to the order of \( \zeta^2 \), Eq. (A.4) equals \( -2h^2 \zeta^2 / (M \zeta) \) because of Eq. (27). The second term of Eq. (A.1) is rewritten as the integral of
\[ \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \]
with respect to \( x \) and \( y \) over the region considered in Eq. (34). In the above, we have
\[ f_s(\phi(0))(0) = f_s(\phi(0))(0) + h \phi_1 \]
(A.8)
with the higher-order terms neglected. Hence, we use Eqs. (24) and (20) to obtain
\[ \delta \Omega[\varphi_1, \zeta] = \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \]
\[ \delta \Omega[\varphi_1, \zeta] = \int_{C_\zeta} d\zeta \int d\phi \int d\zeta \int_{\partial C_\zeta} dS f_s(\phi) \]
(A.9)
up to the second order with respect to \( \varphi_1 \) and \( \zeta \).

Let us minimize Eq. (A.9) with \( \zeta \) fixed. The stationary condition gives
\[ \left( \frac{\partial}{\partial x} \right)^2 + \frac{\partial}{\partial y} \left( \phi_1 \right) = 0 \] for \( x \neq \zeta \) and
\[ Mn \cdot \nabla \phi_1 = \frac{\zeta}{\zeta} \]
at \( z = \zeta \).

Let \( \phi \) denote \( \varphi_1 \) satisfying the above and \( \varphi_1 \to 0 \) as \( |z| \to \infty \). We can obtain \( \phi \) by using Eqs. (34) to (50) with \( Q \) put equal to 0. The Fourier transform of \( \phi^* \) with respect to \( x \) and \( y \) is given by
\[ \tilde{\phi}^*(k, z) = \frac{h \zeta}{MK} e^{-Kz / \zeta} \]
(A.11)
up to the order of \( \zeta \). The corresponding result for \( \tilde{\phi}^*(k, z) \) coincides with \( -\tilde{\phi}^*(k, -z) \).

The probability distribution functional of \( \varphi_1 \) and \( \zeta \) is proportional to \( e^{-\delta \Omega[\varphi_1, \zeta]/(kT)} \), and its integration with respect to \( \varphi_1 \) gives that of \( \zeta \). The integration amounts to substituting \( \phi \) into Eq. (A.9) because the distribution functional is Gaussian. Thus, the probability distribution functional of \( \zeta \) is proportional to \( e^{-\delta \Omega[\phi, \zeta]/(kT)} \), where \( \delta \Omega[\phi, \zeta] \) is found to be
\[ \int \frac{h^2}{MK} \frac{d(K)}{d(K)} + f_s(\phi_1) \]
(A.12)
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