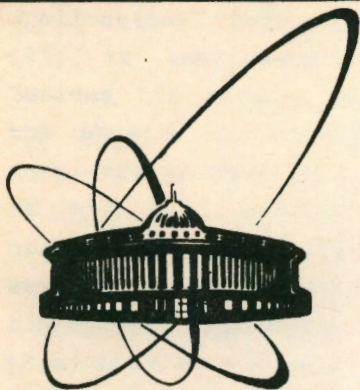


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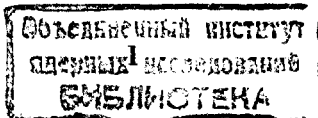
ON THE SURFACE VIBRATION SPECTRA
OF MICELLES AND VESICLES

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1. The study of the dynamics of a liquid drop is a problem of self-dependent interest with a wide variety of applications. Beginning from Kelvin's work related to 1863 [1], it continuously attracts attention of physicists. Besides its natural exploitation and exploration, e.g., in the physics of aerosols, the liquid drop model has been considerably developed in various phenomenological theories. It has been successfully applied in the theory of a gravitating sphere (see, e.g., [2,3]) as well as in the collective model of atomic nuclei [4]. There are also biophysical applications of the model [5-7]. Lately, the physics of microemulsions is of great interest [8,9]. As is known, the microemulsions are formed after the surface active substances (surfactants) are added into the mixture of two liquid components that are insoluble in each other (usually oil and water). At some critical concentration of the surfactants the components after mixing yield a clear solution of high stability. It consists of micelles that are the drops of one liquid (oil) in the other (water). The environments of the liquids are saturated with the surface-active molecules. Other objects of our interest will be vesicles [10,11]. Vesicles appear in solutions of molecules that have a tendency to form bilayer membranes. In the frame of the phenomenological approach (if the surface layer or bilayer is considered to be infinitely thin) the micelles and vesicles can be studied simultaneously.

In this note, we shall be interested in the dynamical properties of micelles and vesicles, namely, in the spectra of the vibration modes connected with the interface between the drops and the surrounding liquid. The relaxation of the micelle and vesicle surface form to its equilibrium (spherical) one has been already investigated in [12,13] where the dispersion law of the relaxation mode has been calculated. However, as has been underlined in [14], these calculations have been carried out using the incorrect assumption of the constant area of the interface. It is pointed out in [14] that in a correct consideration it is



necessary to take into account dynamical equations both for the displacement vector of the surface and the concentration of the surface-active molecules on this surface. It is known that the coefficient of the micelle surface tension is anomalously small in microemulsions and the dependence of the surface energy density on the curvature should be taken into account [15]. In the recent work [16], where the surface modes of fluid droplets are investigated in detail, this dependence was not taken into account (note, however, that in [16] the effects of surface viscosity and compressibility have been studied). In the mentioned work [14], among all, nonlinear equations (boundary conditions) have been obtained to describe the dynamics of the micelles and vesicles with the inclusion of the curvature energy and redistribution of the surfactant molecules on the micelle surface. However, the linear surface modes have been found after a number of approximations in a too simplified form.

In this paper, we base on the linearized boundary conditions for micelles and vesicles found in [14]. Solving exactly the hydrodynamical problem for bulk viscous fluids exterior and interior to the drop (in the linearized case for incompressible fluids), we obtain an exact equation for the frequency of the proper vibrations in the shape of micelles and vesicles. This equation gives wide possibilities for analytical and numerical studies of the frequency spectra. To demonstrate it, we consider the limiting cases of i) the weakly-damped capillary waves for a liquid drop without the surfactant molecules; ii) the pure relaxation mode of the surface micelle vibrations, and iii) the surface mode connected with the redistribution of the surface active molecules on the micelle surface. The obtained analytical expressions make the formulae found in [14] more precise even within the same degree of accuracy. Moreover, we have obtained the frequencies in the higher order of small quantities.

2. In the case of an extremely small surface tension coefficient at the interface of two liquids, the dependence

of the surface energy density ϵ_s on the curvature must be taken into account [15]. If R_1 and R_2 are the local curvature radii, then the expansion of ϵ_s (which is symmetrical in R_1 and R_2) including the second order of $1/R$ is

$$\epsilon_s = \alpha - \beta \left(\frac{1}{R_1} + \frac{1}{R_2} \right) + \frac{\kappa}{2} \left(\frac{1}{R_1} + \frac{1}{R_2} \right)^2 + \frac{\bar{\kappa}}{R_1 R_2}. \quad (1)$$

The coefficients α, β, κ and $\bar{\kappa}$ are the functions of temperature and chemical potentials. If the dependence on the curvature can be neglected, α is the usual surface tension coefficient. The conditions $\kappa > 0$ and $2\kappa + \bar{\kappa} > 0$ must be fulfilled to make the function (1) positively defined. The quantity β equals zero for vesicles and $\beta > 0$ for micelles (the curvature radii are measured from the interior to the exterior of the micelle). The phenomenological analysis of the micelle formation shows [14] that after the addition of surface active substance into the oil-water mixture, the coefficient α decreases to the value $\alpha = \beta^2 / (2\kappa + \bar{\kappa})$. Then, the formation of the micelles with the radii $R_m = \beta / \alpha$ begins. Such a picture is true if

$$k_B T / 8\pi(2\kappa + \bar{\kappa}) \ll 1, \quad (2)$$

when the micelle distribution in radii has a sharp maximum near $R = R_m$. The stability of the spherical micelle with respect to small perturbations is ensured by the condition

$$\alpha_1 = \alpha - 2\beta/R_0 + \kappa l(l+1)/R_0^2 > 0. \quad (3)$$

For micelles with the radii $R = R_m$ this also gives $\bar{\kappa} < 4\kappa$. In (3), R_0 is the equilibrium radius of the micelle and the orbital number l appears after the expansion of the deviation $R(\theta, \phi) - R_0$ in spherical harmonics,

$$R(\theta, \phi) - R_0 = u(\theta, \phi) = R_0 \sum_{l,m} a_{lm}(t) Y_{lm}(\theta, \phi). \quad (4)$$

The index m runs from $-l$ to l and l changes from $l=2$ to some $l_{\max} \sim \pi R_0 / d$ where d is the mean interatomic distance [16,17] (the mode $l=0$ is excluded since by assumption the surface encloses an incompressible fluid and there is no change in

the total amount of the micelle material; $l=1$ is also excluded since it corresponds to motion of the center of mass of the droplet). Thermal fluctuations of the surface form which may play a significant role for small β [18] and lead to logarithmic corrections to α, β, κ and $\bar{\kappa}$ are not considered. This is possible if the condition (2) is satisfied.

After this necessary introduction we can formulate the dynamical equations for the micelle surface quantities as they have been found in [14]. In the linearized form (the small quantities are u from (4), the deviation n'_s from the equilibrium value of the surfactant density n_s , and the surface velocity v^s) they are as follows:

$$\frac{\partial u}{\partial t} = v_r^s, \quad (5)$$

$$\rho_s \frac{\partial v_r^s}{\partial t} - \frac{\Delta_{\perp} + 2}{R_0^2} \left[\left(\alpha - 2 \frac{\beta}{R_0} \right) - \frac{\kappa \Delta_{\perp}}{R_0^2} \right] u = p'_1 - p'_2 - 2 \frac{\partial}{\partial r} (\eta_1 v_{r1} - \eta_2 v_{r2}), \quad (6)$$

$$\text{with } \Delta_{\perp} = \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \frac{\partial}{\partial \theta} + \frac{1}{\sin^2 \theta} \frac{\partial}{\partial \phi} \frac{\partial}{\partial \phi}, \quad \Delta_{\perp} Y_{1m} = -1(1+1)Y_{1m},$$

$$\rho_s \frac{\partial v_{\theta}^s}{\partial t} - \frac{1}{R_0} \frac{\partial \alpha}{\partial n_s} \frac{\partial n'_s}{\partial \theta} = \frac{1}{R_0} \frac{\partial}{\partial \theta} (-\eta_1 v_{r1} + \eta_2 v_{r2}) + \left(\frac{\partial}{\partial r} - \frac{1}{R_0} \right) (-\eta_1 v_{\theta 1} + \eta_2 v_{\theta 2}), \quad (7)$$

$$\rho_s \frac{\partial v_{\phi}^s}{\partial t} - \frac{1}{R_0 \sin \theta} \frac{\partial \alpha}{\partial n_s} \frac{\partial n'_s}{\partial \phi} = \frac{1}{R_0 \sin \theta} \frac{\partial}{\partial \phi} (-\eta_1 v_{r1} + \eta_2 v_{r2}) + \left(\frac{\partial}{\partial r} - \frac{1}{R_0} \right) (-\eta_1 v_{\phi 1} + \eta_2 v_{\phi 2}), \quad (8)$$

$$\frac{\partial n'_s}{\partial t} + \frac{n_s}{R_0 \sin \theta} \left[\frac{\partial}{\partial \theta} \sin \theta v_{\theta}^s + \frac{\partial v_{\phi}^s}{\partial \phi} \right] = 0. \quad (9)$$

In these equations, ρ_s is the mass density of the surface layer, v_r^s , v_{θ}^s and v_{ϕ}^s are the components of the surface velocity v^s , p' is the variable part of the pressure, and η is the constant shear viscosity. The indices '1' and '2' relate the quantities to the bulk fluids interior and exterior to the droplet. All variable quantities (after differentiation) are related to $r=R_0$. In addition to (5)-(9), the following condition must be satisfied at the interface:

$$v_1 = v_2 = v^s \quad \text{at } r=R_0. \quad (10)$$

The detailed obtaining of (5)-(9) is given in [14]. We only note that in these equations the flow of the surface-active molecules to the micelle surface is neglected (this flow is connected with very slow diffusion processes). Small surface kinetic terms are also omitted, which is true if the condition (2) is satisfied. Due to the same reason, the terms with $\partial \beta / \partial n_s$ and $\partial \kappa / \partial n_s$ are neglected. Finally, the incompressibility of the bulk fluids is assumed ($\text{div} v = 0$). Note that there is no terms with $\bar{\kappa}$ in the equations, which is due to the invariance with respect to small variations of the term proportional to $\bar{\kappa}$ in the surface energy.

3. To obtain the solution of the above formulated problem, one must know the distribution of the bulk fluids outside and inside the drop. This means, in our case we have to solve the linearized Navier-Stokes equation

$$\frac{\partial v}{\partial t} = \frac{\eta}{\rho} \Delta v - \frac{1}{\rho} \text{grad } p' \quad (11)$$

in both regions. As distinct from [14], we follow Chandrasekhar's approach [3], that allows us to obtain the exact solution of (11) in a quite simple way. Namely, we express the velocity as a sum of a poloidal and a toroidal part as follows: $v = v_{\text{pol}} + v_{\text{tor}}$,

$$v_{\text{tor}} = \sum_{1m} \left[e_{\theta} \frac{1}{r \sin \theta} \frac{\partial}{\partial \phi} - e_{\phi} \frac{1}{r} \frac{\partial}{\partial \theta} \right] T_{1m}(r, t) Y_{1m}(\theta, \phi), \quad (12)$$

$$v_{\text{pol}} = \sum_{1m} \left[e_r \frac{1(1+1)}{r^2} + e_{\theta} \frac{1}{r} \frac{\partial^2}{\partial r \partial \theta} + \frac{1}{r \sin \theta} \frac{\partial^2}{\partial r \partial \phi} \right] U_{1m}(r, t) Y_{1m}(\theta, \phi). \quad (13)$$

Any poloidal field of the form (13) is orthogonal to any toroidal field (12). Substituting (12) and (13) into (11) in spherical coordinates, we obtain

$$L_1 T_{1m}^{(1)} = 0, \quad L_1 U_{1m}^{(1)} + \frac{B_1^{(1)}}{\rho_1} r^{1+1} = 0, \quad L_2 U_{1m}^{(2)} - \frac{B_2^{(2)}}{\rho_2} r^{-1} = 0, \quad (14)$$

where the differential operator L_1 reads

$$L_1 = \frac{\partial}{\partial t} + \frac{\eta_1}{\rho_1} \left[\frac{1(1+1)}{r^2} - \frac{\partial^2}{\partial r^2} \right], \quad i=1,2, \quad (15)$$

and the solutions of the Laplace equation (it follows from (11)) have been used

$$p_1' = \sum_{1m} B_1^{(1)} (1+1) r^1 Y_{1m}, \quad p_2' = \sum_{1m} B_1^{(2)} 1r^{-1-1} Y_{1m}. \quad (16)$$

After some transformations, (5)-(10) with the use of (4) can be now rewritten in terms of the functions T and U in the following form:

$$\dot{a}_{1m}(t) = 1(1+1)R_0^{-3} U_{1m}^{(1)}(R_0, t), \quad (17)$$

$$\rho_s \ddot{a}_{1m} + \alpha_1 a_{1m} (1-1)(1+2)R_0^{-2} = B_1^{(1)} (1+1)R_0^{1-1} - B_1^{(2)} 1R_0^{-1-2} - 21(1+1)R_0^{-3} (\eta_1 - \eta_2) (\partial/\partial r - 2/R_0) U_{1m}^{(1)}, \quad (18)$$

$$\dot{\nu}_{1m}(t) = 1(1+1)R_0^{-2} \partial U_{1m}^{(1)} / \partial r, \quad (19)$$

$$\rho_s \frac{\partial T_{1m}^{(1)}}{\partial t} = \frac{2}{R_0} T_{1m}^{(1)} (\eta_1 - \eta_2) + \frac{\partial}{\partial r} (-\eta_1 T_{1m}^{(1)} + \eta_2 T_{1m}^{(2)}), \quad (20)$$

$$\rho_s \frac{\partial^2 U_{1m}^{(1)}}{\partial t^2} - n_s \frac{\partial \alpha}{\partial n_s} \nu_{1m} = \left(\frac{\partial^2}{\partial r^2} - \frac{2}{R_0} \frac{\partial}{\partial r} + \frac{1(1+1)}{R_0^2} \right) (-\eta_1 U_{1m}^{(1)} + \eta_2 U_{1m}^{(2)}). \quad (21)$$

After differentiation, $r=R_0$ must be set in these equations. In (19) and (21), the expansion

$$n_s' = n_s \sum_{1m} \nu_{1m} Y_{1m} \quad (22)$$

has been used. Moreover, the conditions (10) together with (12) and (13) give the equations

$$U_{1m}^{(1)}(R_0, t) = U_{1m}^{(2)}(R_0, t), \quad T_{1m}^{(1)}(R_0, t) = T_{1m}^{(2)}(R_0, t),$$

$$\frac{\partial U_{1m}^{(1)}}{\partial r} = \frac{\partial U_{1m}^{(2)}}{\partial r}, \quad r=R_0. \quad (23)$$

Eqs. (17)-(23) are the complete set of the equations necessary to determine the frequency spectrum of the small micelle surface vibrations. All poloidal quantities connected with the velocity field will be considered to depend on time as $\exp(-i\omega t)$. The toroidal functions that are independent of the poloidal ones will be assumed to be proportional to $\exp(-i\Omega t)$. That is, we suppose $a_{1m}(t) = a_{1m}(0) e^{-i\omega t}$, and so on, and $T_{1m}^{(1)}(r, t) = T_{1m}^{(1)}(r, 0) e^{-i\Omega t}$. After substitution of U and T into (14), we come to equations determining the spherical Bessel functions [19] with the following general solutions finite as $r \rightarrow \infty$:

$$U_{1m}^{(1)}(r, 0) = C_{1m}^{(1)} r j_1(z_1) - i\beta_1^{(1)} r^{1+1} / \rho_1 \omega, \quad 0 \leq r \leq R_0, \quad (24)$$

$$U_{1m}^{(2)}(r, 0) = C_{1m}^{(2)} r h_1^{(s)}(z_2) + i\beta_1^{(2)} r^{-1} / \rho_2 \omega, \quad r \geq R_0; \quad z_1 = r(i\omega \rho_1 / \eta_1)^{1/2}$$

$$T_{1m}^{(1)}(r, 0) = D_{1m}^{(1)} r j_1(z_1), \quad (25)$$

$$T_{1m}^{(2)}(r, 0) = D_{1m}^{(2)} r h_1^{(1)}(z_2) + E_{1m}^{(2)} r h_1^{(2)}(z_2), \quad z_1 = r(i\Omega \rho_1 / \eta_1)^{1/2}.$$

To exclude the exponential divergency as $r \rightarrow \infty$, we must use in (24) $s=1$ for the function $h_1^{(1)}$ if from two roots $iz_2 \sim i\sqrt{1-i\omega}$ we choose the one with $\text{Re} i\sqrt{1-i\omega} < 0$. If the root with $\text{Re} i\sqrt{1-i\omega} > 0$ is chosen, one must set $s=2$.

As to the toroidal mode, note that in our problem it is absent. Really, the equations for $T_{1m}^{(1)}$ contain no physical quantities which may lead to surface vibrations. So $\text{Re} \Omega = 0 = \text{Re} i\sqrt{1-i\omega}$. To exclude the divergency of $T_{1m}^{(2)}$ for large r , we must set $D_{1m}^{(2)} = E_{1m}^{(2)} = 0$. Together with (20), (23) and (25) this gives $D_{1m}^{(1)} j_1(z_1) = 0$ and $D_{1m}^{(1)} [j_1(z_1) + z_1 j_1'(z_1)] = 0$. These equations are compatible with each other if $D_{1m}^{(1)} = 0$ or, if simultaneously $j_1(z_1) = 0$ and $z_1 j_1'(z_1) = 0$ with the only possible common root $z_1 = 0$ (that is, $\Omega = 0$). This proves our statement because in both cases there is no toroidal mode.

Combining (17)-(24), we come after some simple but cumbersome algebra to the following equations for determination of the coefficients $C^{(1)}$ from (24):

$$X C_{1m}^{(1)} + Y C_{1m}^{(2)} = 0, \quad Z C_{1m}^{(1)} + W C_{1m}^{(2)} = 0, \quad (26)$$

where (in what follows, $r=R_0$ in z_1)

$$\begin{aligned} X &= \frac{\rho_1}{1} - \frac{z_1}{2l+1} \frac{j_{1+1}(z_1)}{j_1(z_1)} \left[2(1+2)P + \rho_1 - A_1/\omega^2 \right], \\ Y &= \frac{\rho_2}{1+1} + \frac{z_2}{2l+1} \frac{h_{1-1}^{(1)}(z_2)}{h_1^{(1)}(z_2)} \left[2(1-1)P - \rho_1 + A_1/\omega^2 \right], \\ Z &= \rho_1 - \frac{1z_1}{2l+1} \frac{j_{1+1}(z_1)}{j_1(z_1)} \left[2(1+2)P + \rho_s/R_0 + n_1/\omega^2 \right] \\ W &= -\rho_2 - \frac{1+1}{2l+1} \frac{z_2 h_{1-1}^{(1)}(z_2)}{h_1^{(1)}(z_2)} \left[2(1-1)P - \rho_s/R_0 - n_1/\omega^2 \right]. \end{aligned} \quad (27)$$

Here, we have denoted

$$\begin{aligned} A_1 &= \alpha_1(1-1)(1+2)R_0^{-3}, & n_1 &= n_s \frac{\partial \alpha}{\partial n_s} 1(1+1)R_0^{-3}, \\ \rho_1 &= \frac{\rho_1}{1} + \frac{\rho_2}{1+1} + \frac{\rho_s}{R_0}, & P &= \frac{\rho_1}{z_1^2} + \frac{\rho_2}{z_2^2}. \end{aligned} \quad (28)$$

It is assumed that from two above mentioned roots $i\sqrt{1\omega}$ the root is chosen for which $\text{Re} i\sqrt{1\omega} < 0$. The second root does not change (27) due to the property $h_1^{(2)}(-z) = (-1)^1 h_1^{(1)}(z)$ [19]. The condition of nontrivial solvability of (26) leads to the equation

$$XW - YZ = 0. \quad (29)$$

This is the main result of the paper. Eq.(29) is the equation for determination of the proper frequencies of the surface vibrations of the considered liquid drops. Within the linear approximation this equation is exact giving wide possibilities for both numerical and analytical analysis.

4. Let us consider a few simple examples of the application of eq.(29). For the beginning, we give asymptotic expressions for the Bessel functions $j_1(z)$ and $h_1^{(1)}(z)$ [19] we shall use:

$$\frac{j_{1+1}}{j_1} \approx \frac{z}{2l+3} \left[1 + \frac{z^2}{(2l+3)(2l+5)} + \dots \right],$$

$$\frac{h_{1+1}^{(1)}}{h_1^{(1)}} \approx \frac{z}{2l-1} \left[1 + \frac{z^2}{(2l-3)(2l-1)} + \dots \right], \quad \text{for } |z| \ll 1, \quad (30)$$

$$\frac{j_{1+1}}{j_1} \approx i + \frac{1+1}{z} + \dots, \quad \frac{h_{1-1}^{(1)}}{h_1^{(1)}} \approx i + \frac{1}{z} + \dots, \quad \text{if } |z| \gg 1. \quad (31)$$

i) Consider the spectrum of the capillary waves of the liquid drop. We will neglect the quantities ρ_s and n_1 but, contrary to [14], no additional conditions will be used as to the velocity distribution of the bulk fluids. Substituting the asymptotics (31) for $|z_{1,2}| \gg 1$ into (27) and keeping in (29) only the terms of the order z , we obtain in the first approximation the well-known result [1] (if α_1 is replaced by α in (28))

$$\omega_0^2 = A_1/\rho_1. \quad (32)$$

In the next approximation we retain in (29) the terms of the orders z and 1 (then in X and Y the terms -1 and $-z$ and, correspondingly, in Z and W the terms -1 and $-1/z$ should be retained; this means that, contrary to [14], the viscosity terms in (6)-(8) must also be taken into account in such an approximation). The equation for ω then reads

$$[\omega^2 - \omega_0^2][(-i\omega)^{1/2}a + b] = \omega_0^2(2l+1)/1(1+1), \quad (33)$$

with

$$\begin{aligned} a &= R_0 \rho_1 [1/\sqrt{\rho_1 \eta_1} + 1/\sqrt{\rho_2 \eta_2}], \\ b &= (\rho_1 - \rho_2)^2 / \rho_1 \rho_2 + 2\rho_1 (\eta_1 - \eta_2) / \sqrt{\rho_1 \rho_2 \eta_1 \eta_2}. \end{aligned}$$

For vesicles, the quantity b equals zero identically. For our liquid drop we have $|b| \ll |a\sqrt{1\omega}|$. Representing

$$\omega \approx \omega_0(1+\Delta), \quad |\Delta| \ll 1, \quad (34)$$

and substituting it into (33), we obtain in the first

$$\Delta = \frac{1}{2} \frac{(2l+1)^2}{1(1+1)} [b - a\sqrt{\omega_0/2}(1-i)]^{-1}. \quad (34a)$$

It has been already taken into account here that $\text{Re}\sqrt{-i\omega} < 0$. Note that these weakly damping capillary waves differ from the result [14] even in the most rough approximation corresponding to $b=0$.

ii) Now, consider the opposite case $|z_{1,2}| \ll 1$ that seems to take place for micelles. Neglected ρ_s and n_1 and keeping in (29) the terms of the orders 1 and z^2 , we obtain the following quadratic equation with respect to ω :

$$(2l+1)(\eta_1 + \eta_2) [\rho_1(1+1) + \rho_2] + 2(\eta_1 - \eta_2) \left[\frac{q_1 \rho_1}{\eta_1} \frac{1(1+2)}{(2l+3)(2l+5)} - \frac{p_1 \rho_2}{\eta_2} \frac{1^2 - 1}{(2l-3)(2l-1)} \right] + i \frac{p_1 q_1}{\omega R_0^2} = A_1 (\eta_1 + \eta_2) l(1+1)(2l+1) \omega^{-2}, \quad (35)$$

with $p_1 = \eta_1(2l^2 + 4l + 3) + \eta_2 2l(1+2)$ and $q_1 = \eta_1 2(1^2 - 1) + \eta_2(2l^2 + 1)$.

It is not difficult to solve and discuss this equation. Depending on the determinant of the quadratic equation one can obtain the solutions of various characters. We omit here such an analysis that is connected with concrete values of the quantities entering into (35) and write down only the case when the first term in (35) can be neglected. Then, analogously as in [14], we obtain the pure relaxation mode, however, its analytical expression differs from that in [14]:

$$\omega \approx -i A_1 R_0^2 l(1+1)(2l+1)(\eta_1 + \eta_2) / p_1 q_1. \quad (36)$$

iii) The last example will be the situation when the main role in eqs.(27)-(29) is played by the quantities connected with the surface -active substance. We consider the case for which the surface vibration frequency ω is much larger than ω_0 from i). Then we can neglect A_1/ω^2 in comparison with ρ_1 . We also will not take into account ρ_s/R_0 and obtain ω for $|z_{1,2}| \gg 1$. Retaining in (29) the terms of the order z^2 and z , the equation for determination of ω is

$$\omega^2 \rho_1 (\sqrt{\rho_1 \eta_1} + \sqrt{\rho_2 \eta_2}) + (-i\omega)^{1/2} R_0 \rho_1 n_1 - n_1 \left(\frac{\rho_1}{\rho_2} \sqrt{\rho_2 \eta_2} + \sqrt{\rho_1 \eta_1} \right) = 0. \quad (37)$$

The solutions of this fourth degree equation can be easily obtained. If the last term in (37) is neglected, we find the following solution that coincides with the result from [14] (this is the only case when our result fully coincides with [14]):

$$\omega_0 = \frac{1}{2} (\pm \sqrt{3} - i) \{-R_0 n_1 / [\sqrt{\rho_1 \eta_1} + \sqrt{\rho_2 \eta_2}] \}^{2/3}. \quad (38)$$

However, (38) is true only if the quantity

$$\Delta = \frac{2}{3} \frac{n_1}{\rho_1 \omega_0^2} \left[\frac{\rho_1}{\rho_2} \sqrt{\rho_2 \eta_2} + \sqrt{\rho_1 \eta_1} \right] [\sqrt{\rho_1 \eta_1} + \sqrt{\rho_2 \eta_2}]^{-1} \quad (39)$$

obeys the condition $|\Delta| \ll 1$. The first correction to the solution (38) has the form $\omega = \omega_0(1 + \Delta)$. This surface mode is connected with the redistribution of surface-active molecules on the micelle surface. The frequency of this mode possesses real and imaginary parts of the same order of magnitude. Note that distinctions of the results i) and ii) from [14] are due to approximate solutions of the dynamical equations given in Section 2 and approximate calculations of the velocity distribution that have been used in [14].

5. Thus, with the use of linear dynamical equations of the phenomenological theory developed by Lebedev and Muratov [14] for the surface vibrations of micelles and vesicles and applying the exact solution of the linear hydrodynamic problem for incompressible bulk fluids inside and outside the drop, the exact equation for the frequency spectrum has been obtained. The redistribution of the surfactant molecules on the micelle surface has been taken into account as well as the influence of the surface layer mass density. The obtained equation gives many possibilities for the analysis of the spectra and further comparison with experiments, e.g., light or neutron scattering, neutron spin-echo studies [20] and others. To demonstrate it, we have considered a few simple examples such as the weakly-damped capillary waves of a liquid drop, the relaxation mode of the micelle and the mode

connected with the redistribution of the surface-active molecules. The obtained analytical expressions make the recent results [14] more precise.

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References

- [1] H.Lamb, Hydrodynamics (Dover, New York, 1945).
- [2] S.Chandrasekhar, Hydrodynamics and Hydrodynamic Stability (Clarendon, Oxford, 1968).
- [3] S.Chandrasekhar, Proc.London Math.Soc. 9 (1959) 141.
- [4] A.Bohr and B.R.Mottelson, Nuclear Structure, vol.2 (Mir, Moscow, 1977).
- [5] M.V.Volkenstein, in: Molecular Biology.Problems and Perspectives (Nauka, Moscow, 1964) p.172 (in Russian).
- [6] A.V.Zatovsky and V.Yu.Klishko, Mol.Biol.(Moscow) 23 (1989) 1091.
- [7] K.A.Landman, J.Theor.Biol. 115 (1985) 9.
- [8] L.M.Prince, Microemulsions. Theory and Practice (Academic, New York, 1977).
- [9] Physics of Complex and Supermolecular Fluids. eds. S.A.Safran and N.A.Clark (John Wiley, New York, 1987).
- [10] F.Szoka, Jr., and D.Papahadjopoulos, Ann.Rev.Biophys. Bioeng. 9 (1980) 467.
- [11] P.Meleard et al., Europhys.Lett. 11 (1990) 355.
- [12] M.B.Schneider, J.T.Jenkins and W.W.Webb, J.Phys.(Paris) 45 (1984) 1457.
- [13] S.T.Milner and S.A.Safran, Phys.Rev 36A (1987) 4371.
- [14] V.V.Lebedev and A.R.Muratov, JETP 95 (1989) 1751.
- [15] W.Helfrich, Z.Naturforsch. 103B (1975) 67.
- [16] L.C.Sparling and J.E.Sedlak, Phys.Rev. 39A (1989) 1352.
- [17] A.Tamura and T.Ichinokawa, Surf.Sc. 136 (1984) 437.
- [18] P.G.De Gennes and S.Taupin, J.Chem.Phys. 86 (1982) 2294.
- [19] Handbook of Mathematical Functions, eds.M.Abramowitz and I.Stegun (Dover, New York, 1972).
- [20] J.S.Huang and S.T.Milner, Phys.Rev.Lett. 59 (1987) 2600.

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Лисы В.

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О спектрах поверхностных колебаний
мицелл и везикул

В рамках линейной феноменологической теории Лебедева и Муратова найдено точное уравнение для собственных поверхностных мод мицелл и везикул. Теория учитывает перераспределение поверхностно-активного вещества на поверхности мицеллы и зависимость плотности поверхностной энергии от кривизны. Полученное уравнение использовано для более точных, по сравнению с ранее известными, вычислений спектров поверхностных колебаний в различных приближениях.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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Lisy V.

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On the Surface Vibration Spectra
of Micelles and Vesicles

Within the linear approximation of the recent Lebedev and Muratov's phenomenological theory an exact equation for the proper surface modes of spherical micelles and vesicles is obtained. The theory takes into account the redistribution of the surfactant molecules on the micelle surface and the dependence on the curvature. The equation found allowed us to calculate the surface vibration spectra in various approximations more precisely if compared with the previously known results.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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