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Physics of Soap Films

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Soap films constituting from two surfactant monolayers sandwiching a thin layer of water are investigated. The structure of such films is not static because thermal fluctuations induce corrugations on the monolayers which can be decompose into two principle types of modes: undulation (or bending) modes for which the monolayers oscillate in phase, and squeezing modes for which these oscillations have a 180° phase difference. The eigen-modes spectrum for both types of motions is found. It is shown that the squeezing mode (where the water is pumped back and forth) induces fluctuational (anharmonic) contributions to viscosity and diffusion coefficients which are divergent at large scales, leading to anomalous scaling behavior of the coefficients.

Keywords: Fluctuations; fluid-fluid interfaces; soap films; anomalous scaling

1. INTRODUCTION

Seminal contributions by prof. Alfred Saupe to the field of lyotropic liquid crystals and particularly his discovery of biaxial liquid crystals [1] have triggered numerous experimental and theoretical investigations of lyotropic systems. It was crucial for comprehension of physics of these systems and demonstrated the extraordinary richness of lyotropic systems. One perhaps most old example (soap films) will be considered in this paper.

Free standing films attract attention of investigators from the early 70-th when the methods of production such films has been proposed till nowadays (see e.g., the monograph [2] and the survey [3]). There are many reasons for this interest, but, probably the most important fact is that compared to other systems with lowered dimensions free standing films have considerable advantages for experimental investigations. Firstly one can investigate physical phenomena in a broad range of film thicknesses.

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Secondly film thickness can be controlled with high precision and thirdly–influence of background effects and substrates is very small.

There is also one rather general theoretical reason for the interest. In fact free – standing films can be considered as a new class of physical systems: two dimensional systems embedded and living in three dimensional space (i.e. having three dimensional degrees of freedom). But these degrees of freedom are relevant only for dynamical phenomena.

In the most papers devoted to free-standing films the influence of the finite size on phase transitions and structures has been considered (see references in [3]). However the main feature of free standing films is just the absence of a substrate and this feature almost does not manifest itself on thermodynamic properties. The absence of substrates leads to the possibility of bending deformations of free-suspended films. Therefore this film has three dimensional degrees of freedom and since we should not take into account any effects of the interaction with the substrate (that are quite important for conventional systems formed on the surface of solids or liquids) fluctuational effects related to the possibility of bending deformations of films are very noticeable in dynamics on large scales. One can say that specific physics of soap films are manifested mainly in dynamical phenomena and to describe these phenomena is the aim of the paper.

Very thin films consisting of few molecular layers may be pulled from smectic liquid crystals. Just these films have been studied experimentally (see e.g. [3] and references herein) and theoretically in the papers [4], [5] (see also the monograph [6]). Thin free-suspended films may be treated as two dimensional systems. This confines our study to scales much exceeding the thickness of the film.

The same characteristic property (the absence of substrates) is intrinsic for classical soap films known for a long time. However physical quantitative investigations of such systems have been started only recently in connection with revival of general interest to colloidal systems and due to the progress of preparation of soap films with given composition and controlled thickness. Such films are preparing using special dopes to conventional soap films, the stability of which is supplied by surfactant monolayers on the surface of water [7], [8], [9].

Such films are organized more complex manner than free standing films of one component liquid crystals. Soap films are constructed from two different substances and these substances are separated in space. These facts lead to some specific unusual properties of soap films [5].

The main method of investigations of soap films is a light-scattering experiment. Because of relaxation of surface fluctuations, the scattered light

has a broadened spectral distribution compared with the incident light. This broadening is small, however, the modern technique of light beating (intensity fluctuation spectroscopy) is allowed to obtain informations about eigen-modes of the system. Theoretical investigations of dynamic fluctuation phenomena in free standing soap films is the aim of this work.

The content of the paper is the following. In Section 2 results of calculations of the eigen-modes spectrum of a soap film in linear approximation will be presented. Section 3 is devoted to the analysis of fluctuation effects and in the last Section the possibility of experimental verifications of the effects predicted in the work will be discussed shortly.

2. DYNAMIC EQUATIONS OF SOAP FILMS

Let us remind that the film is considered to consist of a volume part, containing almost pure water, bounded by surface layers containing almost pure soap. We believe the surface layers as idealized surfaces having a surface tension γ and a surface elasticity ε_s.

The liquid satisfies the hydrodynamic equations of motion supplemented with boundary conditions at the surfaces. The liquid is assumed to be incompressible and the effects of gravity are neglected. In this Section we use the linearized hydrodynamic equations, the role of nonlinear terms will be discussed in Section 3.

We will neglect also a motion of a gas surrounding film. Therefore the boundary conditions are reduced to the equality of the momentum flux density to the surface stress tensor of the film (see [10]). As a result one can get the system of linear equations and the condition of existence of nontrivial solutions determines the dispersion laws of eigen-modes. Omitting details which can be found in the papers, [11], [12] and [13], we will present here only results. It appears that zero equating the determinant of the mentioned above system of linear equations gives two relations between the frequency ω and the wave-vector k of any perturbation of the film:

$$\left(\omega^2 + \frac{2i\eta k^2\omega}{\rho}\right)^2 - \omega^2\left(\omega_1^2 + \frac{\varepsilon_s m k^2}{\rho} cth\frac{mh}{2} - \frac{4\eta^2 k^3 m}{\rho^2} th\frac{kh}{2} cth\frac{mh}{2}\right)$$

$$+ \frac{\varepsilon_s k^2 \omega_1^2}{\rho}\left(m\, cth\frac{mh}{2} - k\, cth\frac{kh}{2}\right) = 0, \qquad (1)$$

$$\left(\omega^2+\frac{2i\eta k^2\omega}{\rho}\right)^2-\omega^2\left(\omega_2^2+\frac{\varepsilon_s mk^2}{\rho}th\frac{mh}{2}-\frac{4\eta^2k^3m}{\rho^2}cth\frac{kh}{2}th\frac{mh}{2}\right)$$

$$+\frac{\varepsilon_s k^2\omega_2^2}{\rho}\left(mth\frac{mh}{2}-k\,th\frac{kh}{2}\right)=0, \tag{2}$$

In these equations η and ρ-viscosity and mass density of the liquid, h is the thickness of the film, the surface elasticity $\varepsilon_s=-n_s\partial\gamma/\partial n_s$, where n_s is the surface concentration of soap molecules. In addition

$$\omega_1^2=\frac{\gamma\mathbf{k}^3+2kV''}{\rho}th(kh/2),\quad \omega_2^2=\frac{\gamma k^3}{\rho}cth(kh/2),$$

where $V''=d^2V/dh^2$ and $V(h)$ - the interaction potential per unit area, retaining the distance between the surfaces of the film.

Let us discuss shortly the dependence of the interaction potential on h. The most well-known forces responsible for this interaction are Van der Waals forces [14]. For non-retarded case when $h\ll\lambda_0$, where λ_0 is some characteristic wavelength, depending on the dispersion of dielectric permeabilities have

$$V(h)=\frac{\Theta}{h^2}, \tag{3}$$

where the parameter Θ has the dimension of the energy.

Expression (3) is correct only for absolutely flat films. One can study the more general case when boundaries of the film are not flat. The corrections to (3) due to roughness of surfaces can be expressed as [15]

$$V(h)=\frac{\Theta\Delta^2}{h^4}, \tag{4}$$

where Δ is the mean square value of the roughness.

For $h\ll\lambda_0$ the retardation effects should be taken into account. The potential $V(h)$ in this case will be

$$V(h)=\frac{\Theta\lambda_0}{h^3}. \tag{5}$$

Of course there are contributions to $V(h)$ from other forces (electrostatic, steric, hydratation and so on). We are not going to discuss here the concrete form of $V(h)$ referring readers to the papers [16], [17] , where this problem was investigated (note also the paper [18] where the Van der Waals interaction in the case of thin soap films has been studied). It can be concluded from these papers that a fair description for the Van der Waals energy and related quantities for soap films is given by the empirical formulas including five (!) adjustable parameters. Nevertheless deviations from approximate expressions given above are not very large (for low film thickness). We will use further for estimations two particular forms of Van der Waals interaction (3) and and (5) and the potential proportional to h^{-4} which will be written in the following form

$$V(h) = \frac{\Theta_1^2}{\gamma h^4}, \tag{6}$$

where the parameter Θ_1 has the dimension of the energy. Note that for the stability of the film the condition $V'' > 0$ should be satisfied.

Equations (1), (2) are too complex to show simply what the general solution for ω is. The equation (1) corresponds to motion when the both surfaces of the film move synchro and the equation (2) describes asynchronous motion of the film boundaries. One can see that the both equations (1) and (2) have three solutions in the long wavelength limit $kh \ll 1$. The explicit form for dispersion laws of eigen-modes depends on relations among dimensionless parameters kh, $|mh|$, $\eta^2/\varepsilon_s \rho h$, $\eta^2/\gamma \rho h$. In the limit $kh \ll 1$ and $|mh| \ll 1$ the dispersion laws have been found in [12] for the case $\varepsilon_s \gg \gamma$. Here we will give the results in the wide region of parameters kh and $|mh|$ for a more natural case $\varepsilon_s \sim \gamma$. In this case we have one small dimensionless parameter $\eta^2/\varepsilon_s \rho h$.

For $kh \ll \eta (\varepsilon_s \rho h)^{-1/2}$ one can get from (1) the dispersion law of so called bending mode

$$\omega = \pm c_t k - i \frac{\varepsilon_s^2 h^3}{16 \eta \gamma} k^4, \tag{7}$$

where $c_t = (2\gamma/\rho h)^{1/2}$. Note that in the conformity with the general consideration of [5], [6] the attenuation of this mode is proportional to k^4 but not k^2 as for the case of ordinary viscous damping.

The second propagating mode can be found from (2). It may be called the longitudinal sound and it has the following dispersion law:

$$\omega = \pm c_l k - i\frac{2}{\rho}\left(6\eta + \frac{\varepsilon_s \rho h}{48\eta}\right)k^2. \tag{8}$$

Here $c_l = (2\varepsilon_s/\rho h)^{1/2}$. One can see that for $kh \ll \eta(\varepsilon_s \rho h)^{-1/2}$ we have pure viscous damping, whereas for $\eta(\varepsilon_s \rho h)^{-1/2} \ll kh \ll 1$ the attenuation of longitudinal sound is inversely proportional to η. Physically this fact is related to the unusual mechanism of attenuation in this limit. The longitudinal sound is attenuated due to excitation of motion in the bulk of the film which is inversely proportional to η.

In the limit $kh \ll 1$ for each of the equation (1) and (2) there is also one pure relaxational mode. We will see below that for our consideration the most relevant will be the squeezing mode which dispersion law can be easy found from (2):

$$\omega = -i\frac{h^3 k^2}{24\eta}(\gamma k^2 + 2V''). \tag{9}$$

Physically this mode corresponds to such motion when film liquid is pumped back and forth through a slab with thickness h, formed by soap monolayers, according to the well-known Reynolds law.

For the completeness we will present also the dispersion laws for the other regions of parameters. In the limit $\eta(\varepsilon_s \rho h)^{-1/2} \ll kh \ll 1$ instead of the longitudinal sound we have the elastic mode:

$$\omega = \frac{\pm(3)^{1/2} - i}{2}\left(\frac{\varepsilon_s^2}{\eta\rho}\right)^{1/3} k^{4/3}. \tag{10}$$

In the same region of scales the squeezing mode is transformed into the following:

$$\omega = \pm c_t h k^2 - i\left(\frac{2\eta c_t}{\rho h}\right)^{1/2} k. \tag{11}$$

At last for $kh \gg 1$ instead of bending and squeezing modes we have ordinary capillary waves:

$$\omega = \left(\frac{\gamma k^3}{\rho}\right)^{1/2}\left(1 - \frac{i}{2}\left(\frac{\eta^2}{4\rho\gamma}\right)^{1/4} k^{1/4}\right). \tag{12}$$

All our consideration has assumed that $\gamma \sim \varepsilon_s$. In the case $\varepsilon_s \gg \gamma$ there is the region of the usual viscous attenuation of the bending mode. Namely in this case for scales

$$\eta(\varepsilon_s \rho h)^{-1/2} \ll kh \ll \eta(\gamma\rho h)^{-1/2} \tag{13}$$

one can easy find from (1):

$$\omega = \pm c_t k - \frac{i\eta}{2\rho} k^2. \tag{14}$$

3. NONLINEAR DYNAMIC EQUATIONS AND FLUCTUATIONS

From the analysis presented in the preceding Section one can easy understand that at the scales $kh \gg (V''h^2/\gamma)^{1/2}$ the squeezing mode is the most soft one ($\omega \sim k^4$) and therefore at these scales thermal fluctuation effects can be relevant. Our aim in this Section is the investigation of these effects.

For shortening of expressions let us rewrite the dispersion law (9) in the new notations

$$\omega = -i\zeta k^2(\alpha k^2 + \alpha_1). \tag{15}$$

Here $\alpha_1 \equiv 2h^2 d^2V/dh^2$, $\alpha \equiv \gamma h^2$, $\zeta \equiv h/24\eta$. The convenience of such notations in particular is in the fact that the coefficient α has the meaning of the elastic modulus of the soap film, ζ is the kinetic coefficient which may be called a diffusion one.

To neglect α_1 in the dispersion law (15) we have to restrict ourselves only to the scales

$$kh \gg (\alpha_1 h^2/\alpha)^{1/2}. \tag{16}$$

This condition for non-retarded Van der Waals interaction (3) leads to

$$kh \gg (\Theta/\gamma h^2)^{1/2} \tag{17}$$

and for the potential (6) it is

$$kh \gg \Theta_1/\gamma h^2. \tag{18}$$

Let us note that the natural upper bound for parameters Θ and Θ_1 is the temperature and it is convenient to introduce the parameter

$$x \equiv T/\gamma h^2 \tag{19}$$

which is small. Actually $x \sim 1$ for $h \sim a_m$ where a_m – molecular size and therefore for $h \gg a_m$ we have $x \ll 1$. Therefore the inequalities (17) and (18) can be satisfied in the long wavelength limit $kh \ll 1$.

Let us consider now the region of scales (16) where we may neglect α_1 in the dispersion law (9) of the squeezing mode. In this region the squeezing mode is a very soft one which explains the essential role of the squeezing fluctuations in the dynamics of soap films.

We are interested in fluctuational effects related to the squeezing mode here. Therefore we may restrict ourselves only to this soft mode and neglect the other ones. To describe the motion in the squeezing mode one should add to the set of hydrodynamical variables of isotropic liquids, one additional variable ψ determining for example the two dimensional density of the bulk liquid in the film. After elimination of all hard degrees of freedom we come to the closed system of nonlinear equations for the variable ψ describing variations of the film thickness in the squeezing mode (playing the same role as the concentration in binary solutions [19]) and the transversal with respect to a wave vector component of the velocity $\mathbf{v}_\perp$.

Let us describe shortly the derivation of these equations. Suppose in the equilibrium state a soap film is stretched along the plane $x - y$. The energy of a soap film can be represented in the following form

$$H = \int dx\, dy\, (\mathbf{j}^2/2\rho + g^{1/2}\, \epsilon\, (\rho g^{-1/2}, \sigma, \psi, \nabla\psi)). \tag{20}$$

Here ϵ is energy density referred to the unit area of x-y plane, ρ is the mass density, σ is specific entropy and

$$g = 1 + (\nabla_\alpha u)^2. \tag{21}$$

The quantity g entering (20) is associated with the fact that the combination $g^{1/2}\, dx\, dy$ determines an element of the surface of the film and therefore the

quantity $\rho g^{-1/2}$ equals the mass of a unit area of the film irrespective to the choice of the axis x and y. Introduction of the quantity g into (20) ensures the invariance of the energy H with respect to the choice of the coordinate system. In (21) and below the Greek indices designate components of the vectors along the x, y axes. The momentum density $\mathbf{j} = (j_\alpha, j_z)$ and therefore in (20) $\mathbf{j}^2 = j_\alpha^2 + j_z^2$.

The natural dynamic equation for ψ can be written in a standard form:

$$\partial\psi/\partial t + v_\alpha \nabla_\alpha \psi = -\zeta \delta H/\delta\psi. \tag{22}$$

To find the explicit form of the dissipative term we have to know the dependence of ϵ on ψ and $\nabla\psi$. It can be found directly from the dispersion law (9) of the squeezing mode. Indeed two terms in the brackets in the expression (9) describe two contributions to the energy ϵ from the interaction between two surfaces of the film and from the elastic deformation of the film. Finally one can find the following expression for the energy ϵ:

$$\epsilon = V'' h^2 \psi^2/2 + \gamma h^2 (\nabla\psi)^2/4 \equiv (\alpha_1 \psi^2 + \alpha(\nabla\psi)^2)/2, \tag{23}$$

and the explicit form for the dynamical equation for ψ

$$\partial\psi/\partial t + v_\alpha \nabla_\alpha \psi = -\zeta(\alpha_1 \psi + \alpha \nabla^2 \psi), \tag{24}$$

To close the system of equations we should add the equation for the velocity v_α (or j_α). The non-dissipative dynamic equation for j_α has the following form:

$$\partial j_\alpha/\partial t = -\nabla_\beta T_{\alpha\beta}, \tag{25}$$

where the stress tensor $T_{\alpha\beta}$ is

$$T_{\alpha\beta} = g^{1/2} P\left(\delta_{\alpha\beta} - \frac{\nabla_\alpha u \nabla_\beta u}{g}\right) + g^{1/2} \frac{\partial\epsilon}{\partial \nabla_\beta \psi} \nabla_\alpha \psi + \mathrm{v}_\beta \mathrm{j}_\alpha. \tag{26}$$

Note that the derivative $\partial j_\alpha/\partial$t is equal to the divergence of the symmetric tensor, hence, the conservation law for the projection of the angular momentum onto the z-axis is valid. Bearing in mind that coordinate axes are chosen arbitrarily we arrive to the conclusion that all components of the angular momentum are conserved.

Figuring in Expression (26) the pressure P is defined by the standard formulae

$$P = \rho \partial \epsilon / \partial \rho - \epsilon. \tag{27}$$

From (26) and (27) it is easy to see that the quantity $-P$ is equal to the variation of the energy of the film at an adiabatic increasing of its area by unity. Therefore $P = -2\gamma$, where the factor 2 appeared since our film has two surfaces.

In the region of scales (16) the squeezing mode is the most soft one of the system. It means that main fluctuation contributions to characteristics of the system are related just to the squeezing mode. The hydrodynamic equations we have just considered have nonlinear couplings among dynamical variables and, therefore, interactions among harmonic modes that lead to corrections to the harmonic modes given in Section 2. These anharmonic fluctuational corrections can be calculated
using rather elegant diagrammatic technique developed by Wyld [20]. A textbook description of the diagram technique can be found in the book by Ma [21] (see also the monograph [6]). Details of calculations are beyond the scope of the paper (all the more that prof. A.Saupe does not like cumbersome calculations). Here only results will be presented. The scheme of the derivation is given shortly in the paper [22].

It turns out that fluctuational contributions to ζ and η_1 diverge at large scales and exceed the bare values at wave vectors

$$k < \left(\frac{T}{\alpha \zeta \eta_1} \right)^{1/2}. \tag{28}$$

In this region we cannot restrict ourselves only to the first corrections and should take into account higher-order contributions. This is the same situation as near a second-order phase transition and therefore we can expect a scaling behavior of the coefficients characterizing the "dressed" correlation functions. Let us introduce scaling exponents Δ_η and Δ_ζ which determine the long wavelength behavior of the coefficients ζ and η_1

$$\zeta \propto k^{-\Delta_\zeta}, \quad \eta_1 \propto k^{-\Delta_\eta}. \tag{29}$$

To estimate the values of these exponents one can use renormalization-group (RG) methods. The marginal dimension is $1 + 4$ (time $+$ 4d space). It is not very difficult to check that there are no corrections to the coefficients

T and α which is accounted for by the fluctuation-dissipation theorem and by the fact that α is the static module (in statics fluctuations are not relevant). In the dimension $d = 4 - \varepsilon$ the one-loop RG equations for the coefficients ζ and η_1 and are

$$\frac{d\zeta}{dL} = (d-1)g\zeta, \quad \frac{d\eta_1}{dL} = \frac{1}{d+2} g\eta_1. \tag{30}$$

Here

$$g = \frac{TS_d}{(2\pi)^d \eta_1 \zeta \alpha d} \Lambda^{-\varepsilon}$$

is an invariant charge, S_d is the area of the d-dimensional sphere, $L = ln(\Lambda/k)$, Λ is a cutoff. For $\varepsilon \ll 1$ the fixed point of (30) is

$$g^* = \frac{6}{19}\varepsilon.$$

Thus the exponents determined by (30) are

$$\Delta_\zeta = \frac{18\varepsilon}{19}, \quad \Delta_\eta = \frac{\varepsilon}{19} \tag{31}$$

For the real soap films $\varepsilon = 2$ which is not a small parameter. Nevertheless we may hope that (31) give a reasonable estimation for the exponents Δ_η and Δ_ζ even at $\varepsilon = 2$. Therefore we may expect that the viscosity coefficient η_1 only weakly depends on the scale whereas ζ diverges with the exponent close to 2 and therefore the dispersion law for the squeezing mode (with the V''-term neglected!) only slightly differs from a diffusion one. Let us note that the behavior of the second viscosity coefficient η_2 is determined by the same exponent Δ_η and therefore the attenuation of the longitudinal sound will be proportional to $\omega^{2-\Delta_\eta}$.

Let us give the general picture of fluctuation effects in the soap films. At shortest scales the dispersion laws of all modes are determined by the linear dynamic equations. At scales determined by (28), (16 – 19) squeezing fluctuations lead to the scaling behavior discussed above.

At larger scales a new phenomenon should be taken into account. Namely as we have shown above (Section 2) the bending mode (7) has anomalously

weak attenuation. Let us rewrite (7) in more compact designations

$$\omega = \pm c_s k - i\mu k^4, \tag{32}$$

where ω is the frequency, k is the wave vector and as we have seen in Section 2 the viscous damping of this sound proportional to k^2 is absent (in fact it is due to the rotational invariance of the film). For soap films comparing (7) and (32) we have the following estimations

$$c_s \sim (\gamma/\rho h)^{1/2}, \quad \mu \sim \gamma h^3/\eta,$$

where ρ is the $3d$ density of the water and η is its viscosity. Besides the linear attenuation μk^4 figuring in (32) the nonlinear fluctuation contribution to the attenuation of the bending mode βk^3 exists [5,6]. Here $\beta \sim T/(\rho_2 c_s)$ and ρ_2 is the surface mass density of the film (for the soap film $\rho_2 \sim \rho h$). Comparing both contributions ($\propto k^4$ and $\propto k^3$) we conclude that the fluctuation attenuation becomes essential at wave vectors

$$kh \sim (T/\gamma h^2)(\eta^2/\gamma\rho h)^{1/2}, \tag{33}$$

Bending fluctuations (as well as squeezing ones) give anomalous contributions to the viscosity coefficients. If kh exceeds the value (33) the contributions to η_1 and η_2 are of the order of $T/(\mu c_s k)^{1/2}$. For kh smaller than the scale (33) they are of the order of $T/\beta^{2/3} c_s k^{1/3}$. Note that the above contributions to $\eta_1$and η_2 can be neglected in the region (28) since really Θ does not exceed T. Let us remind that actually it is the main condition of the applicability of our theory, because the large role of squeezing fluctuations is based on the possibility to neglect the interaction term in the dispersion law (9) of the squeezing mode.

4. CONCLUSION

Let us sum up results of the work. We have seen that in soap films due to the weakness of the potential $V(h)$, retaining the thickness h of the film, the squeezing mode is the most soft one $\omega \sim k^4$. Therefore in this region of scales fluctuations of this mode may play a relevant role. It turns out indeed that fluctuational effects lead to the strong renormalization of the parameters describing the squeezing mode. Namely, the kinetic coefficient ζ diverges like and $(kh)^{-18\varepsilon/19}$ and viscosity coefficients as $(kh)^{-\varepsilon/19}$

Thus fluctuations make the squeezing mode harder and harder with increasing of scales and such a manner to cut of the mentioned above divergences.

Let us remind that to be a correct such scenario has to neglect the potential $V(h)$. It is the most delicate assumption. The fortunate for us circumstance in this respect, it is a small numerical factor figuring in the Van der Waals contribution. In principal there are other possibilities to diminish further the bulk Van der Waals contribution by using appropriate dopes and liquids for constructing soap films.

Now few words concerning the experimental situation. The studies of soap films [3], [8], [9], [11], [12] after careful deconvolution of the spectra allow to interpret the light scattering results in terms of the bulk values for the surface tension γ and shear viscosity η_1. There is a large discrepancy between data obtained from the measurements of the squeezing and of the bending modes. Although it is not obvious that a film tension and viscosities measured in megahertz frequency range (bending mode) must be equal the values of these quantities obtained in the kilohertz frequency region (squeezing mode) it can not explain the differences.

There are a number of possible reasons for the observed differences. We will not discuss all these explanations here. Note only that the fluctuational effects considered in our work give us a natural (at least partial) explanation for the difference found in the experiments. Further measurements including direct acoustic experiments should be performed to clarify what mechanism for the increase of the viscosity is the most important one.

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