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The Field of Radiative Forces and the Acoustic Streaming in a Liquid Layer on a Solid Half-Space

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Abstract—The acoustic field and the field of radiative forces that are formed in a liquid layer on a solid substrate are calculated for the case of wave propagation along the interface. The calculations take into account the effects produced by surface tension, viscous stresses at the boundary, and attenuation in the liquid volume on the field characteristics. The dispersion equations and the velocities of wave propagation are determined. The radiative forces acting on a liquid volume element in a standing wave are calculated. The structure of streaming is studied. The effect of streaming on small-size particles is considered, and the possibilities of ordered structure formation from them are discussed.

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INTRODUCTION

Today, an increase in the number of publications devoted to the acoustic effect on microdrops and to the streaming induced in them is being observed. Devices (microboards, microchips) implementing the motion of drops under the effect of surface acoustic waves (SAWs) are under development [1–10]. As a rule, SAWs are excited in a plate made from piezoelectric material. After the first experiments [11], many studies concerned with this subject had been carried out, which resulted in the formation of an important field of applied research: acoustoelectronics [12]. In this field, the studies were stimulated by the needs of biology, cytology, chemistry, and, in the last few years, nanotechnologies. One of the topical problems is the effect of SAWs on the content of a microdrop and, in particular, its mixing by SAW-induced streaming. The effect of SAWs on a drop is also used for breaking the drop into smaller droplets, i.e., for “microatomization” due to the capillary wave generation.

An important application of this is the formation of structures with a preset morphology on a substrate by drying a liquid with suspended particles in a wave field. Today, methods of structure formation due to the self-assembly of nanoparticles in the course of evaporation of a solution are widely used. The acoustic effect not only introduces a new ordering mechanism, but also allows controlling the parameters of the structure to be formed.

Most of the studies concerned with the aforementioned problems are experimental ones. This is related to the technological needs. An appropriate theoretical description may be useful for understanding the physics of the processes and also for optimizing the parameters of both the system and the acoustic action. To develop the theory, it is necessary to calculate the acoustic fields, the radiative forces, the acoustic streaming, and, finally, their effects on the particles.

Below, we consider the following system. A liquid layer with a thickness h overlies a solid substrate. A wave travels along the interface and penetrates into both the solid half-space and the liquid layer. The field formed in the layer acts upon the particles suspended in it so that it causes the formation of a periodic structure.

ACOUSTIC FIELDS IN THE LIQUID LAYER AND IN THE SUBSTRATE

The Basic Problem

First, we consider the simplest statement of the problem [13]. The (x, y) plane of the Cartesian coordinate system coincides with the boundary $z = 0$ between the solid substrate and the liquid layer. The z axis is directed vertically downwards. The upper boundary of the liquid layer is the plane $z = -h$. The acoustic fields in such a system were considered earlier and described by I.A. Viktorov [13]. However, for new applications, a development of the theory is necessary. Below, we briefly review the known results in order to generalize them.

The displacement $\mathbf{U}$ of an element of the solid medium is expressed through the scalar Φ and vector $\mathbf{\Psi}$ potentials. For the two-dimensional problem (in which the displacement along the y axis is absent and the potentials are independent of the y coordinate), the displacements along the x and z axes have the form

$$\mathbf{U} = (U, V = 0, W) = \nabla\Phi + \mathrm{rot}\boldsymbol{\Psi},$$

$$U = \frac{\partial\Phi}{\partial x} - \frac{\partial\Psi}{\partial z}, \quad W = \frac{\partial\Phi}{\partial z} + \frac{\partial\Psi}{\partial x}.$$

The potentials satisfy the wave equations and, for monochromatic waves, the Helmholtz equations:

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial z^2}\right)\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} + \begin{pmatrix}k_l^2\Phi\\ k_t^2\Psi\end{pmatrix} = 0. \qquad (1)$$

Equations (1) involve the wave numbers $k_l = \omega/c_l$ for longitudinal waves, and $k_t = \omega/c_t$ for transverse waves. The velocities of the waves are expressed through the Lame parameters λ and μ:

$$c_l = \sqrt{(\lambda + 2\mu)/\rho}, \quad c_t = \sqrt{\mu/\rho}.$$

Solutions to Eqs. (1) are sought in the form of plane waves traveling along the horizontal axis, i.e., the x axis, and decreasing with the z coordinate:

$$\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} = \begin{pmatrix}A\exp(-qz)\\ B\exp(-sz)\end{pmatrix}\exp(-i\omega t + ikx). \qquad (2)$$

Here, $q = \sqrt{k^2 - k_l^2}$ and $s = \sqrt{k^2 - k_t^2}$ are the scales characterizing the decrease in the longitudinal and transverse field components with depth in the substrate. According to Eq. (2), we seek a wave with a frequency ω and a horizontal wave number k determined from the dispersion equation.

Let us consider the field in the liquid layer $-h < z < 0$. First, we assume that the liquid is an ideal one and apply the linearized Euler and continuity equations:

$$\rho_0\frac{\partial\mathbf{u}}{\partial t} + \boldsymbol{\nabla}p' = 0, \quad \frac{\partial p'}{\partial t} + c_0^2\rho_0\mathrm{div}\mathbf{u} = 0. \qquad (3)$$

Here, $\mathbf{u}$ is the oscillation velocity vector of the liquid, ρ_0 and ρ' are the equilibrium density and its acoustic increment, p' is the acoustic pressure, and c_0 is the velocity of sound.

For the liquid, it is convenient to introduce the scalar potential φ. The corresponding expressions for acoustic variables and the wave equation derived from Eq. (3) have the form

$$\mathbf{u} = \boldsymbol{\nabla}\varphi, \quad p' = -\rho_0\frac{\partial\varphi}{\partial t}, \quad \frac{\partial^2\varphi}{\partial t^2} - c_0^2\Delta\varphi = 0. \qquad (4)$$

We seek the solution to Eqs. (4) in the form of a wave traveling along the boundary with an unknown dependence $D(z)$ of its amplitude on the vertical coordinate:

$$\varphi = D(z)\exp(-i\omega t + ikx),$$

$$\frac{d^2D}{dz^2} + (k_0^2 - k^2)D = 0.$$

Here, $k_0 = \omega/c_0$ is the wave number in the liquid. Determining the function $D(z)$, we write the solution

$$\varphi = (C_1e^{irz} + C_2e^{-irz})\exp(-i\omega t + ikx), \qquad r = \sqrt{k_0^2 - k^2}. \qquad (5)$$

Thus, the acoustic field in the substrate is described by potential (2), while the acoustic field in the liquid layer is described by potential (5). The relations between wave amplitudes, as well as the wave number k, are determined from the characteristic system and the dispersion equation that correspond to the boundary conditions of the problem.

The following conditions should be satisfied.

(i) At the interface $z = 0$, the vertical displacements should be identical, i.e.,

$$W|_{z=0} = \left(\frac{\partial\Phi}{\partial z} + \frac{\partial\Psi}{\partial x}\right)_{z=0}$$

$$= (-qA + ikB)\exp(-i\omega t + ikx)$$

$$= -\frac{u_z}{i\omega}\bigg|_{z=0} = -\frac{1}{i\omega}\frac{\partial\varphi}{\partial z} = -\frac{r}{\omega}(C_1 - C_2)\exp(-i\omega t + ikx).$$

This relation should be satisfied for any x and t. Hence, we obtain the relation between the constants A, B, C_1, and C_2:

$$C_1 - C_2 = -\frac{\omega}{r}(-qA + ikB). \qquad (6)$$

In what follows, the exponential factor corresponding to the wave propagation along the interface will be omitted.

(ii) At the boundary $z = 0$, the normal stresses should be identical: $\sigma_{zz} = -p$. The normal stress in the solid is

$$\sigma_{zz} = \lambda\left(\frac{\partial^2\Phi}{\partial x^2} + \frac{\partial^2\Phi}{\partial z^2}\right) + 2\mu\left(\frac{\partial^2\Phi}{\partial z^2} + \frac{\partial^2\Psi}{\partial x\partial z}\right),$$

$$\sigma_{zz}|_{z=0} = \lambda(-k^2 + q^2)A + 2\mu(q^2A - iksB).$$

The pressure in the liquid at the boundary is

$$p' = -\rho_0\frac{\partial\varphi}{\partial t} = i\omega\rho_0\varphi = i\omega\rho_0(C_1 + C_2).$$

Equating the last two expressions, we arrive at the second relation between the constants:

$$(-\lambda k^2 + \lambda q^2 + 2\mu q^2)A - 2i\mu ksB = -i\omega\rho_0(C_1 + C_2). \qquad (7)$$

(iii) At the boundary $z = 0$, the tangential stresses in the solid are zero, because the liquid is assumed to be ideal. The tangential stress is

$$\sigma_{xz}|_{z=0} = \mu\left(2\frac{\partial^2\Phi}{\partial x\partial z} + \frac{\partial^2\Psi}{\partial x^2} - \frac{\partial^2\Psi}{\partial z^2}\right)_{z=0}$$
$$= -2ikqA - k^2B - s^2B = 0.$$

This yields the third relation,

$$B = -2i\frac{kq}{k^2+s^2}A. \quad (8)$$

(iv) At the free surface of the liquid, i.e., at the upper boundary of the liquid layer $z = -h$, the acoustic pressure p' is zero:

$$p'|_{z=-h} = i\omega\rho_0\varphi|_{z=-h} = C_1e^{-irh} + C_2e^{irh} = 0.$$

This yields the fourth (and last) relation,

$$C_2 = -C_1\exp(-2irh). \quad (9)$$

Thus, we have four homogeneous equations (6)–(9) for determining the unknown amplitudes A, B, C_1, and C_2 and the wave number k. The dispersion equation is derived from the condition that the determinant of the system is zero:

$$4k^2qs - (k^2+s^2)^2 = \frac{\rho_0}{\rho}qk_t^4\begin{cases} r^{-1}\tan(rh) \\ r_*^{-1}\tanh(r_*h). \end{cases} \quad (10)$$

We note that Eq. (10) differs from the dispersion equation obtained in [13] (see Eq. (1.58) in [13]). If, in Eq. (10), we set the liquid density ρ_0 or the layer thickness h to zero, we obtain a simple dispersion equation for Rayleigh waves traveling along the solid–vacuum boundary. The right-hand side of Eq. (10) takes into account the effect of the liquid layer.

The upper row on the right-hand side of Eq. (10) corresponds to such a solution to this equation that the wave velocity in the system is greater than the sound velocity in the liquid but smaller than the longitudinal and transverse wave velocities in the solid: $c > c_0$, $c < c_t < c_l$. The lower row on the right-hand side of Eq. (10) corresponds to a wave velocity smaller than the sound velocity in the liquid: $c < c_0$. In this case, $r = ir_*$, $r_* = \sqrt{k^2 - k_0^2}$.

The Dispersion Curves

Substituting the expressions $k = \omega/c$, $k_l = \omega/c_l$, $k_t = \omega/c_t$, $q = \omega\sqrt{c^{-2} - c_l^{-2}}$, $s = \omega\sqrt{c^{-2} - c_t^{-2}}$, and $r = \omega\sqrt{c_0^{-2} - c^{-2}}$ in Eq. (10), we represent dispersion equation (10) in the form

$$\sqrt{1-\frac{c^2}{c_l^2}}\sqrt{1-\frac{c^2}{c_t^2}} - \left(1-\frac{c^2}{2c_t^2}\right)^2$$
$$= \frac{1}{4}\frac{\rho_0}{\rho}\left(\frac{c}{c_t}\right)^4\sqrt{1-\frac{c^2}{c_l^2}} \quad (11)$$
$$\times\begin{cases} \left(\dfrac{c^2}{c_0^2}-1\right)^{-1/2}\tan\left(H\dfrac{c_0}{c}\sqrt{\dfrac{c^2}{c}-1}\right), & c > c_0, \\ \left(1-\dfrac{c^2}{c_0^2}\right)^{-1/2}\tanh\left(H\dfrac{c_0}{c}\sqrt{1-\dfrac{c^2}{c_0^2}}\right), & c < c_0. \end{cases}$$

Here, $H = \omega h/c_0$ is the wave thickness of the layer. Since the variable H contains the frequency, for this type of wave a dispersion takes place.

To analyze Eqs. (11), it is convenient to normalize all the velocities in Eqs. (11) by the transverse wave velocity c_t. In order to avoid the inaccurate statements encountered in [13], we begin with considering the simple specific case where $c_l^2/c_t^2 = 2$. In this case, the root of the dispersion equation that corresponds to the Rayleigh wave (traveling along the boundary between the solid half-space and the vacuum) is determined analytically: $c_R^2/c_t^2 \equiv X_R = 3 - \sqrt{5} \approx 0.764$. For this case, dispersion equation (11) is simplified. The explicit expression for the wave thickness of the layer has the form

$$H = \sqrt{\frac{2X}{1-2X}}$$
$$\times\,\text{arctanh}\left(\left[\frac{4\rho}{\rho_0}\frac{\sqrt{1-2X}}{X^2}\left(\sqrt{1-X} - \left(1-\frac{X}{2}\right)^{3/2}\right)\right],\right)$$
$$X_S = 0.442 < X < X_0 = \frac{1}{2}, \quad (12)$$
$$H = \sqrt{\frac{2X}{2X-1}}\left\{\pi n + \text{arctanh}\left[\frac{4\rho}{\rho_0}\frac{\sqrt{2X-1}}{X^2}\right.\right.$$
$$\left.\left.\times\left(\sqrt{1-X} - \left(1-\frac{X}{2}\right)^{3/2}\right)\right]\right\}, \quad X_0 < X < 1.$$

Here, $X = c^2/c_t^2$; for definiteness, we assumed that $c_0^2/c_t^2 \equiv X_0 = 0.5$. The quantity $X_S \approx 0.442$ is identical to the value of X at which the argument of the hyperbolic arctangent in Eqs. (12) is unity. The corresponding velocity of the wave proves to be identical to the velocity of the Stoneley wave c_S at the boundary between the solid and liquid half-spaces. This velocity

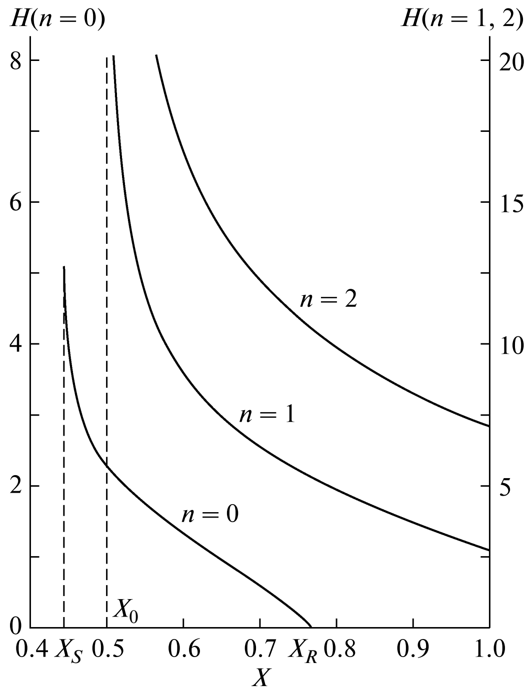

Fig. 1. Illustration of dispersion relation (12). The vertical axis represents the wave thickness of the liquid layer $H = \omega h/c_0$, and the horizontal axis, the normalized square of the wave propagation velocity in the structure $X = c^2/c_t^2$. The following parameters are preset: $4\rho/\rho_0 = 10$, $c_l^2/c_t^2 = 2$, and $X_0 = c_0^2/c_t^2 = 0.5$. In this case, for the Rayleigh wave, $X_R = c_R^2/c_t^2 \approx 0.764$; for the Stoneley wave, $X_S = c_S^2/c_t^2 \approx 0.442$.

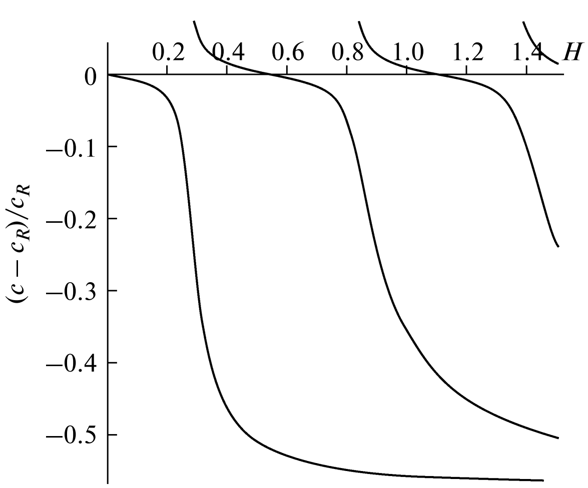

Fig. 2. Dependence of the velocity of wave propagation along the boundary between water and lithium niobate on the thickness of the liquid layer. The abscissa axis represents the layer thickness normalized to the wavelength in the liquid, and the ordinate axis, the relative deviation of the velocities of the surface and Rayleigh waves.

is known [13] to be somewhat smaller than the wave velocity in the unbounded liquid, $c_S < c_0$, for which $X = X_0 = 0.5$ in Eqs. (12). The point $X_0 = 0.5$ separates the regions of applicability of the first and second formulas (12). The value $n = 0$ corresponds to the zero-order mode, and the values $n = 1, 2, \ldots$ correspond to modes of higher orders.

In Fig. 1, dispersion curves (12) are plotted for the modes with the numbers 0, 1, and 2. It should be noted that, as the layer thickness H increases, the propagation velocities of higher-order modes tend to the velocity of sound in the liquid, whereas the velocity of the zero-order mode tends to the velocity of the Stoneley wave.

Figure 2 shows the dispersion dependence for the system that was used in the experiments described in [14]: a film of an aqueous solution on a lithium niobate substrate. The following parameters are preset: the liquid is represented by water with the density $\rho_0 = 1$ g/cm^3 and the velocity of sound $c_0 = 1500$ m/s; the lithium niobate substrate has the density $\rho = 4.7$ g/cm^3, the longitudinal wave velocity $c_l = 7250$ m/s, the transverse wave velocity $c_t = 3750$ m/s, and the Rayleigh wave velocity $c_R = 3480$ m/s; the frequency of waves is 15 MHz. Figure 2 shows the dependence of the relative deviation of the wave velocity c from the Rayleigh wave velocity c_R on the liquid layer thickness H. For the principal mode, when the thickness H is small, these velocities differ only slightly. As H increases, the velocity c decreases; when the thickness is large, this velocity tends to the Stoneley wave velocity c_S, which, at the given parameters, is very close to the velocity of sound in the liquid c_0. At the same time, as H increases, the next (first order) mode arises, its velocity tending to the sound velocity in the liquid when $H \to \infty$. At the point of the first mode generation, the velocity of this mode is identical to the transverse wave velocity in the substrate c_t. As a rule, in the experiments, the layer thickness is much smaller than the wavelength ($H \ll 1$) and the velocity c little differs from the Rayleigh wave velocity c_R. Thus, one can expect that the period of the structure formed from nanoparticles will be approximately identical to half the Rayleigh wavelength.

The Acoustic Field in the Liquid Layer

In view of Eq. (5), after determining the constants C_1 and C_2, we write

$$\varphi = -i\omega A\frac{q}{r}\frac{k_t^2}{k^2+s^2}\frac{\sin r(z+h)}{\cos rh}e^{-i(\omega t - kx)}. \qquad (13)$$

From Eq. (13), we determine the components of the oscillation velocity

$$\mathbf{u} = \nabla(D(z)e^{ikx})e^{-i\omega t} = \left\{ikD; \frac{\partial D}{\partial z}\right\}e^{-i\omega t + ikx}.$$

Thus, the amplitudes of the horizontal and vertical velocity components are

$$u_x = \frac{\omega k}{k^2+s^2}\frac{q}{r}k_t^2\frac{\sin r(z+h)}{\cos rh}A,$$

$$u_z = -\frac{i\omega}{k^2+s^2}qk_t^2\frac{\cos r(z+h)}{\cos rh}A.$$

The Inclusion of Surface Tension

In calculating the acoustic field and the radiation forces, it is necessary to estimate the effect of additional factors. One of them is the surface tension. With this factor taken into account, the basic problem acquires another condition instead of condition (iv) (Eq. (9)) at the free surface of the liquid. All the other boundary conditions (Eqs. (6)–(8)) remain the same. The new condition involves the pressure at the free boundary $z = -h$ in the presence of surface tension, which is determined by the Laplace formula

$$p - p_0 = \sigma\left(\frac{\partial^2\xi}{\partial x^2} + \frac{\partial^2\xi}{\partial y^2}\right). \qquad (14)$$

Here, σ is the surface tension coefficient and ξ is the surface displacement. Let us differentiate Eq. (14) with respect to time. Then, we express the pressure p given by Eq. (14) through the velocity potential φ and use the evident relation $\partial\xi/\partial t = u_z = \partial\varphi/\partial z$ for the vertical velocity component. As a result, we obtain

$$\left.\left(\rho_0\frac{\partial^2\varphi}{\partial t^2} + \sigma\frac{\partial^2}{\partial x^2}\frac{\partial\varphi}{\partial z}\right)\right|_{z=-h} = 0. \qquad (15)$$

Substituting potential (5) in boundary condition (15), we arrive at the equation $\rho_0\omega^2 D + \sigma k^2 D' = 0$. This leads to the following generalization of relation (9) between the constants:

$$C_2 = -C_1\frac{1+i\Sigma}{1-i\Sigma}\exp(-2irh) = -C_1\exp(-2irh_{\text{eff}}),$$
$$\Sigma = \sigma k^2 r/\rho_0\omega^2. \qquad (16)$$

In Eq. (16), we introduced the effective thickness of the layer:

$$h_{\text{eff}} = h - \frac{1}{r}\arctan\Sigma. \qquad (17)$$

The dispersion equation is similar to that obtained in the problem without surface tension, namely, in the previous equation (10), it is necessary to make the substitution $h \to h_{\text{eff}}$.

Since the combination of parameters Σ is small in all the cases, effective layer thickness (17) differs from the true thickness h by approximately $\Sigma/r = \sigma/(\rho_0 c^2)$. The latter quantity is independent of h and ω. For a thin layer of pure water, this quantity is very small ($\sim 10^{-11}$ m). However, in some specific cases, for example, when particles suspended in a solution form a polymer film as a result of solvent evaporation, the effect of surface tension may be of interest.

The Inclusion of Sound Attenuation in the Liquid

One more factor that may affect the field structure is the attenuation of sound in the liquid. In the presence of attenuation, equation of motion (3) acquires an additional term that contains the effective viscosity coefficient b. Then, wave equation (4) takes the form

$$\frac{\partial^2\varphi}{\partial t^2} - c_0^2\Delta\varphi - \frac{b}{\rho_0}\Delta\frac{\partial\varphi}{\partial t} = 0. \qquad (18)$$

The solution to Eq. (18) has the form of expression (5) with the parameter r being replaced by $r_1 = \sqrt{k_1^2 - k^2}$, where $k_1^2 = k_0^2(1 - i\delta)^{-1}$ is the complex wave number in the liquid. The parameter $\delta = \omega b/\rho_0 c_0^2$ is small if the absorption in the liquid at distances on the order of wavelength can be considered to be weak. The same replacement $r \to r_1$ should be made in dispersion equation (10).

Since the object of most interest is a thin layer, namely, a drying liquid film, we restrict our consideration to the presence of zero-order mode alone in dispersion equations (10) and (11). Expanding $\tan(r_1 h)$ in a series for a small layer thickness and assuming that the values of parameters are the same as those in Eq. (12), we obtain a simplified dispersion relation:

$$\sqrt{1-X} - \left(1 - \frac{X}{2}\right)^{3/2}$$
$$= \frac{H}{4\sqrt{2}}\frac{\rho_0}{\rho}X^{3/2}\left(1 + \frac{H^2}{3}\frac{2X-1}{2X} - i\delta\frac{H^2}{3}\right). \qquad (19)$$

One can see that the only imaginary term on the right-hand side of Eq. (19) is proportional not only to the small parameter δ, but also to the cube of the small wave thickness of the layer H. A similar dependence on the parameters δ and H will occur for the imaginary additions to X, c, and the wave number. This means that, as the liquid film dries, attenuation rapidly decreases and becomes negligibly small. Evidently, in all the cases, attenuation in the system is smaller than in the unbounded liquid, because the major part of wave energy is concentrated in the ideal medium, i.e., in the solid half-space.

The Inclusion of Viscous Stresses at the Boundary between the Liquid Layer and the Substrate

Shear viscosity creates an additional mechanism of wave interaction at the boundary between the liquid layer and the substrate. If shear viscous stresses are taken into account, only the third boundary condition given by Eq. (8) changes. The new condition is the equality of tangential stresses at $z = 0$:

$$\sigma_{xz} = \mu\left(2\frac{\partial^2\Phi}{\partial x\partial z} + \frac{\partial^2\Psi}{\partial x^2} - \frac{\partial^2\Psi}{\partial z^2}\right) = 2\eta\frac{\partial^2\varphi}{\partial x\partial z}.$$

Substituting expressions (2) and (5) for the potentials in this formula and applying the first boundary condition given by Eq. (6), we obtain the relation

$$B = -2ikq\frac{1+i\beta}{k^2+s^2+2i\beta k^2}A, \quad \beta = \omega\frac{\eta}{\mu}.$$

Using the remaining boundary conditions, we determine the constants

$$C_{1,2} = \pm\frac{\omega q}{2r}A\frac{\exp(\pm irh)}{\cos rh}\frac{s^2-k^2}{k^2+s^2+2i\beta k^2}$$

and, finally, the dispersion equation

$$4k^2qs(1+i\beta) - (k^2+s^2)^2\left(1+i\beta\frac{2k^2}{k^2+s^2}\right) = \frac{\rho_0}{\rho}qk_t^4\frac{\tan rh}{r}. \quad (20)$$

The contribution of tangential stresses appears on the left-hand side of Eq. (20) and is determined by the parameter β, which is usually small. With allowance for the smallness of β and the wave thickness H of the liquid layer, we obtain a dispersion relation similar to Eq. (19):

$$\sqrt{1-X} - \left(1-\frac{X}{2}\right)^{3/2} + i\beta\left(\sqrt{1-X} - \sqrt{1-\frac{X}{2}}\right) = \frac{H}{4\sqrt{2}}\frac{\rho_0}{\rho}X^{3/2}. \quad (21)$$

In Eq. (21), we replace X by $X(1 + i\Delta)$, where Δ is a small imaginary addition. After calculating Δ, we calculate the attenuation coefficient, i.e., the imaginary part of the wave number:

$$k'' = -\frac{k}{2}\Delta = \frac{\eta\omega^2}{2cc_t^2}\left(1-\frac{X}{2}\right)^2\left[1-\frac{3}{2}\left(1-\frac{X}{2}\right)^2\right]^{-1}.$$

One can see that, when the thickness of the liquid layer is small, the wave attenuation occurs approximately in the same way as it would occur for a shear wave in a solid with a viscosity identical to the viscosity of the liquid.

A Standing Wave in the Liquid Layer

Now, let two waves propagate in the substrate in opposite directions. The potential of the wave traveling in the positive direction of the x axis is determined by Eq. (13). The wave propagating in the negative direction $(-x)$ can be obtained by the formal replacement $k \to -k$ in Eq. (13). The total potential is as follows:

$$\varphi = \varphi_1 + \varphi_2 = 2D(z)\cos kx e^{-i\omega t} = -i\omega A\frac{2q}{r}\frac{k_t^2}{k^2+s^2}\frac{\sin r(z+h)}{\cos rh}\cos kx e^{-i\omega t}. \quad (22)$$

Potential (22) corresponds to a standing wave with an amplitude periodically varying with the horizontal coordinate x.

From Eq. (22), we determine the real components of oscillation velocity and the acoustic pressure:

$$u_x = \frac{k}{r}U_0\sin r(z+h)\sin kx\sin\omega t,$$

$$u_z = -U_0\cos r(z+h)\cos kx\sin\omega t,$$

$$p' = -\frac{\rho_0\omega}{r}U_0\sin r(z+h)\cos kx\cos\omega t.$$

Here, we introduce the notation for the amplitude factor

$$U_0 \equiv \frac{2\omega q}{\cos rh}\frac{k_t^2}{(k^2+s^2)}A.$$

Evidently, attenuation should cause variations in the wave numbers k and r, each of which will acquire an imaginary addition. In an ideal medium, potential (22) has only the cosine component in the dependence on the x coordinate, whereas, in the presence of attenuation, a sine component appears.

THE RADIATION PRESSURE OF AN ACOUSTIC WAVE AND THE STREAMING IN THE LIQUID LAYER

Above, we calculated the characteristics of the acoustic field in the liquid layer. The characteristics vary according to the harmonic law, and their average values are zero. Therefore, the period average force acting on a liquid volume element and on the particles suspended in the liquid should also be zero. In this approximation, the expected formation of structures should not occur. Nonzero average values appear when the quadratically nonlinear terms are taken into account in the initial hydrodynamic equations. Since the average values of quadratic combinations of oscillating variables are not identically zero, we obtain a nonzero average force that leads to structuring of the ensemble of particles.

The Behavior of Small-Size Particles in the Liquid

The radiation pressure of sound was described in many reviews (see, e.g., [15]). The pressure acting from the side of an acoustic field of arbitrary configuration on a suspended particle was calculated in [16]. The origin of the radiative force is the response of a particle to the scattering of the incident wave. However, for small-size particles, such a mechanism is presumably not the governing one. As is known, the fraction of the wave energy scattered by a particle is proportional to $(kR)^4$, where R is the particle radius (see, e.g., [17]). For example, for polystyrene particles with a radius of 100 nm under an incident wave with a frequency of 15 MHz [14], this parameter is on the order of 10^{-9}. Therefore, the predominant mechanism is as follows: because of their small size, the particles can be carried away by the acoustic streaming in the liquid and can move together with the latter.

Let us consider a small particle in an oscillating liquid. The equation of motion has the form (see, e.g., [18])

$$m\frac{d^2}{dt^2}(X-\xi) = -m'\frac{d^2\xi}{dt^2} + F_{\rm diss} + F. \quad (23)$$

Here, X is the displacement of a particle, ξ is the acoustic displacement of the liquid at the particle site, $F_{\rm diss}$ is the dissipative force due to the flow around the particle, and F is the radiative force. If the particle has a spherical shape, we have

$$m = m_S + \frac{1}{2}m_0, \quad m' = m_S - m_0 = \frac{4}{3}\pi R^3(\rho_S - \rho_0),$$

where m_S and R are the mass of the particle and its radius, m_0 is the mass of the displaced liquid, and ρ_S and ρ_0 are the respective densities. If the densities are different, the particle placed in the acoustic field experiences the action of forces both depending on the viscosity of the medium and independent of it.

We represent Eq. (23) in terms of the velocities of the particle $v = dX/dt$ and the liquid $u = d\xi/dt$, and, specifying the expression for the dissipative force [18], we obtain the equation

$$\frac{4}{3}\pi R^3\left(\rho_S + \frac{\rho_0}{2}\right)\frac{d}{dt}(v-u) = -\frac{4}{3}\pi R^3(\rho_S - \rho_0)\frac{du}{dt}$$
$$- 6\pi\eta R\left(1 + R\sqrt{\frac{\rho_0\omega}{2\eta}}\right)(v-u) \quad (24)$$
$$- 3\pi R^2\sqrt{\frac{2\eta\rho_0}{\omega}}\left(1 + \frac{2R}{9}\sqrt{\frac{\rho_0\omega}{2\eta}}\right)\frac{d}{dt}(v-u) + F.$$

In a steady-state flow, where the velocities v and u are time independent, their difference is determined by the effects of the radiative force F and the Stokes force. The radiative force tends to increase the velocity of the particles with respect to the flow, whereas the Stokes force tends to decrease it. Using the Gor'kov formula for the radiation pressure on a particle in a standing wave field, we reduce Eq. (24) to the form

$$R(kR)\left[\frac{\rho_S + (2/3)(\rho_S - \rho_0)}{2\rho_S + \rho_0} - \frac{1}{3}\frac{c_0^2\rho_0}{c_S^2\rho_S}\right]\rho u_0^2$$
$$= 6\eta\left(1 + R\sqrt{\frac{\rho_0\omega}{2\eta}}\right)(v-u).$$

Here, u_0 is the amplitude of the oscillation velocity of the liquid. Estimates by this formula show that, for the particles used in the experiments [14], the difference between the velocities did not exceed 10^{-7}–10^{-8} m/s. Thus, the hypothesis that small-size particles are almost completely carried away by the acoustic streaming while the radiative force acting on the particles can be neglected seems plausible.

The Radiation Pressure on the Liquid

The radiative force F_i is expressed through the radiation stress tensor Π_{ik} [19]:

$$F_i = -\frac{\partial}{\partial x_k}\Pi_{ik}, \quad \Pi_{ik} = \frac{\varepsilon}{\rho_0 c_0^2}\langle p'^2\rangle\delta_{ik} + \rho_0\langle u_i u_k\rangle. \quad (25)$$

Here, the angular brackets denote averaging over the period of the acoustic wave and ε is the nonlinearity parameter of the liquid. For the nonzero components of the tensor, from Eq. (25) we obtain the expressions

$$\Pi_{xx} = \frac{\varepsilon}{\rho_0 c_0^2}\langle p'^2\rangle + \rho_0\langle u_x^2\rangle,$$
$$\Pi_{xz} = \Pi_{zx} = \rho_0\langle u_x u_z\rangle,$$
$$\Pi_{zz} = \frac{\varepsilon}{\rho_0 c_0^2}\langle p'^2\rangle + \rho_0\langle u_z^2\rangle.$$

Thus, the contribution to the force is made by both diagonal and nondiagonal elements of the radiation stress tensor. Calculating the average values of acoustic quantities, we obtain

$$\langle u_x^2\rangle = \frac{k^2}{2r^2}U_0^2\sin^2 r(z+h)\sin^2 kx,$$
$$\langle u_z^2\rangle = \frac{U_0^2}{2}\cos^2 r(z+h)\cos^2 kx,$$
$$\langle u_x u_z\rangle = -\frac{k}{8r}U_0^2\sin 2r(z+h)\sin 2kx,$$
$$\langle p'^2\rangle = \frac{\rho_0^2\omega^2}{2r^2}U_0^2\sin^2 r(z+h)\cos^2 kx.$$

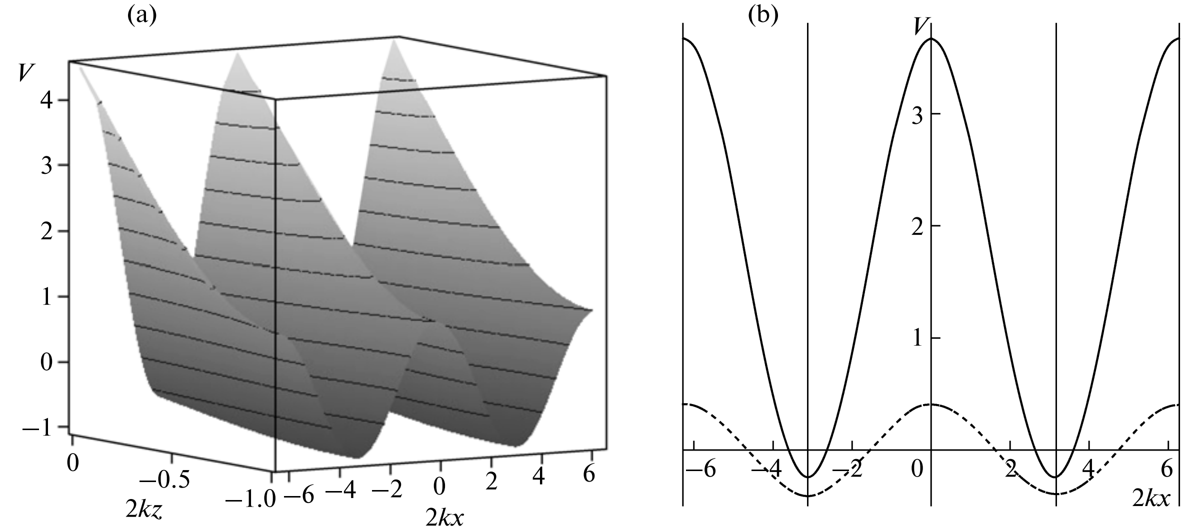

Fig. 3. Radiation pressure potential: (a) the three-dimensional representation and (b) the sections of the potential profile at the interface $z = 0$ (the solid line) and at the free surface $z = -h$ (the dotted line).

Correspondingly, the components of the radiation stress tensor are

$$\Pi_{xx} = \frac{\rho_0 U_0^2}{2r^2}\sin^2 r(z+h)(\varepsilon k_0^2 + (k^2 - \varepsilon k_0^2)\sin^2 kx),$$

$$\Pi_{xz} = \Pi_{zx} = -\frac{k\rho_0}{8r}U_0^2 \sin 2r(z+h)\sin 2kx,$$

$$\Pi_{zz} = \frac{\rho_0 U_0^2}{2}\cos^2 kx\left(\left(\frac{\varepsilon k_0^2}{r^2} - 1\right)\sin^2 r(z+h) + 1\right).$$

Now, we calculate the radiative forces:

$$-F_x = \frac{\partial \Pi_{xx}}{\partial x} + \frac{\partial \Pi_{xz}}{\partial z} = \frac{\rho_0 k U_0^2}{4} \times \sin 2kx\left((\varepsilon - 1)\frac{k_0^2}{r^2}\cos 2r(z+h) - \left((\varepsilon - 1)\frac{k_0^2}{r^2} + 1\right)\right), \quad (26)$$

$$-F_z = \frac{\partial \Pi_{zz}}{\partial z} + \frac{\partial \Pi_{zx}}{\partial x} = \frac{\rho_0 r U_0^2}{4} \times \sin 2r(z+h)\left((\varepsilon - 1)\frac{k_0^2}{r^2}\cos 2kx + \frac{(\varepsilon - 1)k_0^2 + k^2}{r^2}\right). \quad (27)$$

Expressions (26) and (27) are convenient for analyzing the radiative forces in a liquid layer with a small thickness, when the surface wave velocity is greater than the velocity of sound in the liquid, because, in this case, all the coefficients are positive and the signs of the terms appearing in the formulas can be easily determined. For the horizontal force F_x, the factor in parentheses is always negative for any values of z, so that the direction of F_x is only determined by the horizontal coordinate x. Since the dependence on x is periodic, the force exhibits maxima and minima, the latter corresponding to zero force value. Presumably, the particles suspended in the liquid are mainly grouped in the regions of these minima. For the vertical force, the factor in parentheses is positive for any value of x, so that the direction of the vertical force component does not depend on the horizontal coordinate x and is determined by the z coordinate alone. An analysis of the dispersion curves shows that the parameter rh can vary from 0 to $\pi/2$ with a subsequent periodic shift by πn, where n is an integer. This means that the factor $\sin 2r(z + h)$ is positive and the vertical component of the radiative force tends to gather the suspended particles at the free surface of the liquid. In other words, as the thickness of the layer decreases in the course of evaporation, the grouped particles are deposited on the substrate surface.

Since the radiative forces are quadratic in the acoustic field, the period of the spatial structure formed under their action in the horizontal direction is identical not to the acoustic wavelength, but to half the acoustic wavelength, because $2k = 2 \times 2\pi/\lambda = 2\pi/(\lambda/2)$.

The Radiation Pressure Potential

In the general case, the radiation pressure is a tensor quantity, but, in the problem under study, $\mathrm{rot}\,\mathbf{F} = 0$; i.e., the radiation pressure force can be represented as a gradient of a certain potential:

$\mathbf{F} = -\mathrm{grad}\, V$. Let us determine this potential by using Eqs. (26) and (27):

$$V = \frac{\rho_0 U_0^2}{8}\left[(\varepsilon - 1)\frac{k_0^2}{r^2}\cos 2kx \cos 2r(z+h) + \frac{(\varepsilon - 1)k_0^2 + k^2}{r^2}(\cos 2r(z+h) - 1) - \left((\varepsilon - 1)\frac{k_0^2}{r^2} + 1\right)\cos 2kx\right].$$

Here, the integration constant is introduced in such a way that, at the layer surface $z = -h$, the hydrodynamic pressure related to acoustic streaming (see the next section) is zero. Figure 3a shows the characteristic form of the potential for the parameters of the media considered above. Figure 3b shows the profiles of the potential for two sections: near the boundary between the solid substrate and the liquid layer and near the free surface of the liquid layer. The solid line corresponds to the potential near the interface, and the dotted line, near the surface. The vertical lines indicate the positions of the extrema of the potential. One can see that the horizontal structure of the potential does not depend on the vertical coordinate and contains minima, which points to the possibility of particle concentration in these regions. The absolute minima of the potential are close to the free surface of the liquid layer; at the same time, near the interface, the potential well is narrower. Hence, as the layer thickness decreases, the particles are additionally concentrated in the regions corresponding to the minima of the potential.

The Acoustic Streaming Caused by the Radiation Pressure

The radiation pressure sets the liquid in motion and causes it to flow. The structure of the steady-state streaming at small hydrodynamic Reynolds numbers is calculated using the system of equations [20]

$$-\eta \Delta \mathbf{U} = -\nabla P + \mathbf{F}, \quad \mathrm{div}\mathbf{U} = 0. \qquad (28)$$

Here, $\mathbf{U}$ is the velocity of acoustic streaming and P is the flow pressure. Applying the rot operation to the first of Eqs. (28) and taking into account the potential nature of the radiation force $\mathbf{F}$, we obtain a biharmonic equation for the stream function:

$$\Delta\Delta\Psi = 0, \quad U_x = \partial\Psi/\partial z, \quad U_z = -\partial\Psi/\partial x. \qquad (29)$$

These equations should be complemented with boundary conditions. At the boundary $z = 0$ of the liquid with the solid half-space, the flow velocity is zero ($U_x = U_z = 0$); at the free surface of the liquid $z = -h$, the vertical flow is absent ($U_z = 0$) and the pressure on the surface is also absent ($P = 0$).

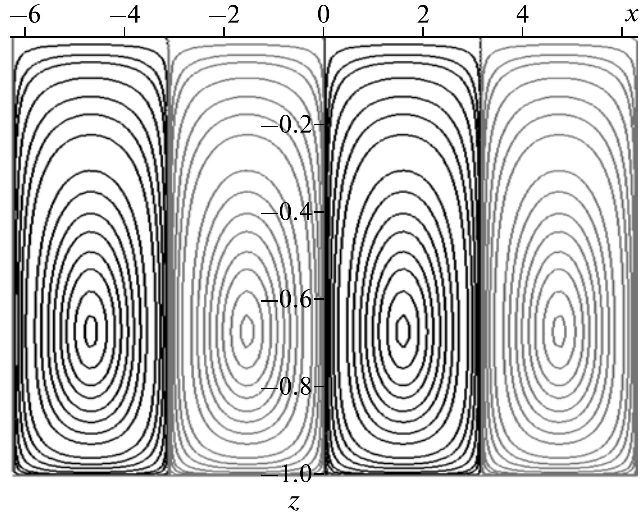

Fig. 4. Streamlines. The abscissa axis represents the quantity $x_1 = 2kx$, and the ordinate axis, $z_1 = 2kz$.

Taking into account the structure of the calculated field of radiation forces, we seek the solution with the following dependence on the horizontal coordinate:

$$\Psi = \Psi_0(z)\sin(2kx), \quad U_x = A(z)\sin(2kx), \quad U_z = B(z)\cos(2kx).$$

Formulas (29), which determine the stream function, suggest the relations $A = \Psi_0'$, $B = -2k\Psi_0$, and $B' = -2kA$. Integrating the equation for the streamlines $dx/U_x = dz/U_z$ and taking into account the aforementioned relations, we obtain the equation $\Psi_0 \sin(2kx) = \mathrm{const}$. One can see that the flow velocity is zero on the lines $x = \pi n/2k$ lying in the (x, z) plane. These lines separate the regions with oppositely directed velocities (see Fig. 4).

This qualitative result is confirmed by the result of streamline calculation. From biharmonic equation (29), we obtain an ordinary differential equation for the function $\Psi_0(z)$, which can be solved:

$$\left(\frac{d^2}{dz^2} - 4k^2\right)^2 \Psi_0(z) = 0,$$

$$\Psi_0 = \alpha[\sinh(2kz) + \beta\cosh(2kz) + \gamma(2kz)\sinh(2kz) + \delta(2kz)\cosh(2kz)].$$

Here, the constants α, β, γ, and δ are determined by the boundary conditions. From the conditions at the interface $z = 0$, we find $\beta = 0$ and $\delta = -1$. Then, for the stream function and the velocity components of streaming, we obtain

$$\Psi = \alpha[\sinh(2kz) + \gamma(2kz)\sinh(2kz) - (2kz)\cosh(2kz)]\sin(2kx),$$

$$U_x = 2k\alpha[\gamma\sinh(2kz) + \gamma(2kz)\cosh(2kz) - (2kz)\sinh(2kz)]\sin(2kx),$$

$$U_z = -2k\alpha[\sinh(2kz) + \gamma(2kz)\sinh(2kz) - (2kz)\cosh(2kz)]\cos(2kx).$$

The condition $U_z(z = -h) = 0$ determines the form of γ, and the constant α is expressed through the radiation pressure:

$$\gamma = \frac{\sinh(2kh) - (2kh)\cosh(2kh)}{(2kh)\sinh(2kh)},$$

$$\alpha = -\frac{\rho_0 U_0^2 h}{32k\eta\sinh(2kh)}.$$

The final solution for the stream function has the form

$$\Psi = -\frac{\rho_0 U_0^2 h}{32k\eta\sinh(2kh)}\Big[\sinh(2kz) + \frac{\sinh(2kh) - (2kh)\cosh(2kh)}{(2kh)\sinh(2kh)}(2kz)\sinh(2kz) - (2kz)\cosh(2kz)\Big]\sin(2kx).$$

This expression was used to plot the streamlines of the acoustic streaming. As one can see from Fig. 4, the streamlines are denser near the lines $2kx = \pi n$. This means that, in the presence of particle interaction forces, which may be either of hydrodynamic or some other origin (e.g., of electric or chemical nature), suspended particles should mainly concentrate in these regions. Thus, a standing surface wave can serve as the controlling factor for the formation of ordered structures from nanoparticles suspended in the liquid.

CONCLUSIONS

We developed a theory that qualitatively explains the experiments on the formation of ordered structures of particles in the course of drying of a colloidal solution on a solid substrate. We demonstrated the possibility to control the process by exciting a wave that propagates along the liquid–solid interface. We calculated the wave field, the radiation forces, and the acoustic streaming in the liquid layer. We considered the main factors that affect the formation of the acoustic field, the vortex streaming, and the nanoparticle structures. The radiative forces arising in the liquid layer serve as the main factor of ordering. The period of the structure formed on the substrate is identical to half the wave length. The radiation pressure that acts on the particles carried by the liquid and is due to the acoustic wave scattering by the particles can presumably be neglected. An important role can be played by forces of nonacoustic origin, which lead to aggregation of particles drawn together under the effect of streaming.

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REFERENCES

1. S. Shiokawa, Y. Matsui, and T. Moriizumi, Jpn. J. Appl. Phys. **28** (Suppl. 1), 126 (1989).
2. S. Shiokawa, Y. Matsui, and T. Ueda, Jpn. J. Appl. Phys. **29** (Suppl. 2-1), 137 (1990).
3. T. Uchida, T. Suzuki, and S. Shiokawa, Proc. IEEE Ultrason. Symp. **2**, 1081 (1995).
4. C. J. Strobl et al., Proc. IEEE Ultrason. Symp. **1**, 255 (2002).
5. A. Wixforth, J. Scriba, and C. Gauer, MST News. **5**, 42 (2002).
6. A. Wixforth, Superlatt. Microstruct. **33**, 389 (2003).
7. C. J. Strobl, Z. V. Guttenberg, and A. Wixforth, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **51**, 1432 (2004).
8. A. Renaudin, P. Tabourier, V. Zhang, J. C. Camart, and C. Druon, Sens. Actuat. B **113**, 389 (2006).
9. M. K. Tan, J. R. Friend, and L. Y. Yeo, in *Proc. of the 16th Austral. Fluid Mechanics Conf.* (Australia, 2007), pp. 790–793.
10. M. K. Tan, J. R. Friend, and L. Y. Yeo, in *Proc. of the 5th Austral. Congress on Appl. Mechanics ACAM 2007*, pp. 348–353.
11. R. M. White and F. W. Voltmer, Appl. Phys. Lett. **7**, 314 (1965).
12. Yu. V. Gulyaev and F. S. Hickernell, Acoust. Phys. **51**, 81 (2005).
13. I. A. Viktorov, *Sonic Surface Waves in Solids* (Nauka, Moscow, 1981) [in Russian].
14. O. V. Rudenko, P. V. Lebedev-Stepanov, V. A. Gusev, A. I. Korobov, B. A. Korshak, N. I. Odina, M. Yu. Izosimova, S. P. Molchanov, and M. V. Alfimov, Akust. Zh. **56** (6) (2010, in press).
15. Z. A. Gol'dberg, in *High-Intensity Ultrasonic Fields*, Ed. by L. D. Rozenberg (Nauka, Moscow, 1968) [in Russian].
16. L. P. Gor'kov, Dokl. Akad. Nauk SSSR **140**, 88 (1961) [Sov. Phys. Dokl. **6**, 773 (1961)].
17. *Acoustics in Problems*, Ed. by S. N. Gurbatov and O. V. Rudenko (Fizmatlit, Moscow, 2009) [in Russian].
18. P. V. Lebedev-Stepanov and O. V. Rudenko, Acoust. Phys **55**, 729 (2009).
19. A. P. Sarvazyan, O. V. Rudenko, and W. L. Nyborg, Ultrasound Med. Biol. (2010, in press).
20. O. V. Rudenko and S. I. Soluyan, *Theoretical Foundations of Nonlinear Acoustics* (Nauka, Moscow, 1975; Consultants Bureau, New York, 1977).

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