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# Analysis of Shape Perturbations of a Drop on a Vibrating Substrate for Different Wetting Angles

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**Abstract**—An exact solution is obtained to the problem of axisymmetric normal modes and natural frequencies characterizing surface perturbations of a drop that sits with an arbitrary wetting angle on a substrate and experiences only gravity and surface tension. The resulting mode solutions are used to calculate and analyze different shapes of the perturbed surface for the same drop placed on a vibrating base. The distinctive feature of the present study is the explicit representation of the results in the form of calculated shapes of the surface of a vibrating drop, comparison of the parameters of actual drops with resonance frequencies, and comparison with experimental data.

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Such simple (and even ordinary) objects as a drop of a liquid and its antipode, i.e., a gas bubble (a gas-filled cavity) in a liquid, had been studied for years, and they still attract the attention of researchers working in different fields of modern science and technology. Moreover, recently, it was found that these simple objects can be used as microreactors for conducting important physical technological operations (see below). These findings, in their turn, stimulated further research into the nature of drops and bubbles.

The theory of the dynamic behavior of bubbles and drops that describes their shape variations due to natural oscillations appeared at the boundary of the 19th and 20th centuries. The development of the theory began with the first publications [1–3], which now have become classical. Later, the initial models were improved, primarily, by taking into account explicit physical effects, such as viscosity, compressibility of a fluid medium, thermal and diffusion processes, etc. The basic stages and results of these studies can be found in [4–6] and in references therein.

Apart from detailed investigations of natural surface oscillations in bubbles and drops, considerable interest has been attracted to the behavior of these objects under the action of external driving forces. In the studies of the latter effects, external actions were primarily represented by acoustic waves. The models and results concerned with acoustic actions on gas-vapor cavities in a liquid have formed a special area of research: acoustic cavitation [7, 8]. It is important to note that, approximately 20 years ago, the fully developed theory of acoustic cavitation laid the foundation for another new area of research: studies of the interaction of acoustic waves with bubbles enclosed in artificial shells; these studies are important for the development of modern technologies in medical acoustics (see, e.g., [9]).

Unlike acoustic cavitation, the effect of acoustic waves on drops has been rather poorly investigated. Scarce papers devoted to this subject [10–13] were mainly related to the development of principles of acoustic levitation and its use for remote manipulations with liquid drops and for their diagnostics.

An important step forward in the theoretical and experimental investigations of the dynamic behavior of bubbles and drops was related to finding the possibilities for practical application of the dynamically varying inner volumes of these objects, in which, owing to the unusual conditions and regimes, some specific physicochemical effects and reactions can be run. Acoustic actions can effectively control the dynamics and, primarily, the volume variations of the aforementioned micro-objects and, hence, their internal processes. Consequently, microbubbles and microdrops can operate as microreactors that perform the necessary physicochemical processes under an acoustic control. For microbubbles, the development of the microreactor concept began with the internal luminescence effect, which was first described in [14]; the modern interpretation can be found in [15]. The latest idea concerned with this phenomenon is the "acoustic fusion (sonofusion) effect" inside a collapsing bubble [16].

Implementation of the microreactor concept for a drop requires considering the system formed by a drop on a solid substrate, which makes it necessary to take into account the wetting effect [17]. The "technological process" of such a microreactor consists in that, in

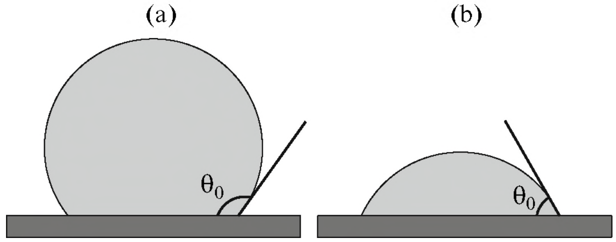


**Fig. 1.** A drop on a substrate: (a) the wetting angle noticeably exceeds 90° (weak wetting, a lyophobic surface of the substrate with respect to the drop liquid); (b) the wetting angle is noticeably smaller than 90° (strong wetting, a lyophilic surface of the substrate).

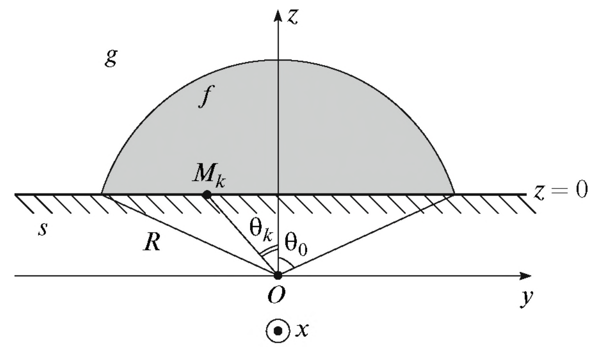


**Fig. 2.** Geometry of the problem.

the course of evaporation (drying) of a drop, which is a disperse system with a filler in the form of some kind of micro- or nanoparticles, the latter undergo self-organization (self-assembly). As a result, a certain micro- or nanostructure is formed on the substrate, such a structure being suitable for various applications [18–21]. The aforementioned process depends on natural factors: the size and composition of the drop, the relative properties determining the wetting of the substrate material with the drop liquid, etc. However, it can also be actively controlled by acoustic methods. In particular, in [23], a microdrop on a substrate and the processes in it were studied under the action of a surface acoustic wave (SAW) propagating in the substrate. It is also possible to consider a vibrational action on the processes in a microdrop: the mechanism of this action should be basically different from the effect of SAW because of the much lower frequency range (from tens of hertz to one or two kilohertz). Indeed, by acting on the entire drop through the temporal variation of the "effective gravity force" in combination with the surface tension, vibration excites certain modes of spatial oscillations of the drop as a whole. This manifests itself in a specific shape of the perturbed surface of the drop (see below).

In the present paper, we calculate the perturbed shape of a drop of an ideal incompressible liquid on a vertically vibrating substrate, which determines the vibration effect on the self-assembly of micro- and nanostructures in the disperse system represented by the drop on the substrate. A necessary step in solving this problem is the initial calculation of the eigenmodes and natural frequencies of a drop on an immobile substrate with allowance for holonomic constraints at the boundary between the liquid drop and the solid surface. The fundamental distinctive feature of the problem solved in this paper is the arbitrary wetting angle (see Fig. 1) corresponding to different liquids and solutions placed on the substrate. In the few publications concerned with the theoretical analysis of such systems [23, 24], calculations were performed for hemispherical drops on the substrate, i.e., for a wetting angle of 90°. In this particular case, calculation and analysis of modes are considerably simplified, some of the modes being degenerate. However, the results of such calculations give no idea of the behavior of a vibrating drop in the cases of strong and weak wetting (Fig. 1). In our paper, we also demonstrate possible deformations of the surface of a vibrating drop on a substrate (such data are absent in the cited publications).

The geometry of the problem is shown in Fig. 2. The drop in the form of a sphere segment with a characteristic opening angle (wetting angle) $\theta_0$ and an initial sphere radius $R$ sits on a horizontal substrate so that the circular contour of its contact with the substrate remains invariable in the course of vibrations. Taking into account the surface tension $\sigma$ at the gas–liquid interface and the gravity force, we consider perturbations of the drop surface in the spherical coordinate system, these perturbations being axially symmetric about the vertical axis (i.e., in the azimuth angle $\psi$). First, we determine the normal modes of natural oscillations of the drop in the form of a sphere segment attached to the substrate. Then, on the basis of the calculated normal mode system, we analyze the vibration-caused forced variations of the drop surface. At each of these two steps, the boundary conditions in the region of contact between the drop and the substrate are considered as imposed holonomic constraints (boundary conditions) and, with the use of the D'Alembert–Lagrange principle, the generalized constraint forces are eliminated. After this, a change to normal generalized coordinates is performed. In addition, the following effective approach to the problem is applied: we consider oscillations of the whole spherical drop with holonomic constraints in the chosen cut plane, which represents the actual substrate. As a result, we simultaneously obtain the solution for two cases (two parts of the drop), one of which is for the sphere segment with an angle $\theta_0$ and the other is for the sphere segment with the angle $\pi - \theta_0$.

We assume that the motion of the liquid drop that affects the shape of its surface is potential, which allows us to use the velocity potential $\varphi$ in our subsequent equations. The varying surface of the spherical (in the absence of perturbations) drop is described as

$$r(\theta, t) = R + \xi(\theta, t),$$

where $\xi(\theta, t)$ is a small deviation of the drop surface from the initial spherical surface.

Since the drop is incompressible, the potential satisfies the two-dimensional Laplace equation, whose solution is determined by the series of $n$th-order Legendre functions $P_n$ multiplied by the desired amplitude functions and integral positive powers of the radius, which excludes singularity at the center of the drop:

$$\varphi(r, \theta, t) = \sum_{n=2}^{N} \tilde{A}_n(t) r^n P_n(\cos\theta). \quad (1)$$

It should be noted that summation in Eq. (1) begins from the third term, since the zero-order partial mode corresponds to isotropic extension (compression) of the drop and is not allowed because of incompressibility. The first partial mode corresponds to translational displacement of the drop and is not considered in our problem. The boundary condition at the drop surface normalized by the unperturbed spherical surface and corresponding to the pressure jump due to surface tension and gravity is determined by equation [25]

$$\rho_f \frac{\partial^2 \varphi}{\partial t^2} - \frac{\sigma}{R^2}(2 + \Delta_\Omega)\frac{\partial \varphi}{\partial r} + \rho_f g \frac{\partial \varphi}{\partial r}\cos\theta = 0, \quad (2)$$

where $\Delta_\Omega = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)$ is the angular part of the Laplace operator in the spherical coordinate system with allowance for symmetry in the axial angle $\psi$.

On a solid substrate at $z = 0$, the no-leakage condition should be satisfied:

$$\mathbf{vn}|_{z=0} = 0, \quad (3)$$

where $\mathbf{n}$ is the normal unit's vector to the substrate and directed toward the upper half-plane; the substrate plane is determined in spherical coordinates by the equation $r\cos\theta = R\cos\theta_0$. For calculation, the no-leakage conditions are assigned on the discrete circles $M_k$ (see Fig. 2). Now, it is necessary to specify the condition at the contact circumference, which should be immobile. This condition has the form

$$\mathrm{grad}\varphi|_{r=R,\,\theta=\theta_0} = 0. \quad (4)$$

In the above expressions, it is convenient to change to the dimensionless variables

$$\tau = t/T_0, \quad T_0 = \sqrt{R^3\rho_f/\sigma}, \quad X = r/R$$

and to the dimensionless physical parameters

$$\begin{aligned}
\Phi(X, \theta, \tau) &= \frac{T_0}{R^2}\varphi(r, \theta, t) = \sum_{n=2}^{N} A_n(\tau) X^n P_n(\cos\theta), \\
\tilde{A}_n(t) &= \frac{1}{T_0 R^{n-2}} A_n(\tau = t/T_0), \\
\frac{\xi(X, \theta, \tau)}{R} &= \sum_{n=2}^{N} f_n(\tau) X^{n-1} P_n(\cos\theta), \\
\frac{df_n}{d\tau} &= nA_n(\tau),
\end{aligned} \quad (5)$$

where $A_n(\tau)$ and $f_n(\tau)$ are the desired dimensionless functions of time.

Substituting Eq. (5) into boundary condition (2), we arrive at a system of dimensionless equations describing the amplitudes of the partial mode of a free spherical drop in the constant gravity field:

$$\ddot{A}_n + \omega_n^2 A_n + K\left[\frac{(n+1)^2}{2n+3}A_{n+1} + \frac{n(n-1)}{2n-1}A_{n-1}\right] = 0,$$

$$\omega_n^2 = n(n+2)(n-1), \quad n = 2, 3, \ldots, N.$$

Substitution of Eq. (5) into boundary condition (3) leads to a system of dimensionless equations describing the holonomic constraints at the liquid–solid boundary inside the circle whose perimeter coincides with the contact line:

$$\begin{aligned}
f_k &= \sum_{n=2}^{N} A_n(\tau)\beta_{nk} = 0, \quad k = 1, 2, \ldots, P-1, \\
\beta_{nk} &= \left(\frac{\cos\theta_0}{\cos\theta_k}\right)^{n-1} \\
&\times [n\cos\theta_k P_n(\cos\theta_k) + \sin\theta_k P_n^1(\cos\theta_k)].
\end{aligned} \quad (6)$$

For the contact line, using Eq. (4), we obtain

$$\begin{aligned}
&\sum_{n=2}^{N-1} A_n(\tau)[nP_N^1(\cos\theta_0)P_n(\cos\theta_0) \\
&- NP_N(\cos\theta_0)P_n^1(\cos\theta_0)] = 0.
\end{aligned} \quad (7)$$

Combining boundary conditions (6) and (7), we arrive at a single system of equations for the imposed constraints:

$$\begin{aligned}
f_k &= \sum_{n=2}^{N} A_n(\tau)\beta_{nk} = 0, \quad k = 1, 2, \ldots, P, \\
\beta_{nk} &= \left(\frac{\cos\theta_0}{\cos\theta_k}\right)^{n-1} \\
&\times [n\cos\theta_k P_n(\cos\theta_k) + \sin\theta_k P_n^1(\cos\theta_k)],
\end{aligned}$$

$$k = 1, 2, \ldots, P-1, \quad n = 2, 3, \ldots, N,$$
$$\beta_{nk} = nP_N^1(\cos\theta_0)P_n(\cos\theta_0) - NP_N(\cos\theta_0)P_n^1(\cos\theta_0),$$
$$k = P, \quad n = 2, 3, \ldots, N-1,$$
$$\beta_{nk} = 0, \quad k = P, \quad n = N. \tag{8}$$

We note that, in Eqs. (6)–(8) and in the subsequent calculations, the upper infinite limit of summation is replaced by a finite value while continuous boundary condition (3) imposed on the entire contact area is represented in the discrete form. This is necessary for subsequent numerical calculations.

We represent the solution $\Phi(X, \theta, \tau)$ describing arbitrary oscillations (including the forced ones) of the drop on the substrate as a superposition of normal modes $\Phi^m(X, \theta, \tau)$:

$$\Phi(X, \theta, \tau) = \sum_m \Phi^m(X, \theta, \tau),$$
$$\Phi^m(X, \theta, \tau) = \sum_{n=2}^{N} A_n^m(\tau)X^n P_n(\cos\theta). \tag{9}$$

If we represent $A_n^m(\tau)$ in the form

$$A_n^m(\tau) = a_n^m e^{-iW_m\tau}, \tag{10}$$

the dimensionless constants $a_n^m$ and $W_m$ involved in Eq. (10) will describe the relative amplitude of the $n$th partial mode included in the $m$th normal mode and the $m$th natural frequency, respectively.

To obtain a system of differential equations describing the desired functions $A_n^m(\tau)$ appearing in Eq. (9), we apply the D'Alembert–Lagrange principle and eliminate the substrate's reaction forces with the help of constraint equations (8):

$$\begin{cases} \ddot{A}_n + \omega_n^2 A_n \\ + K\left[\dfrac{(n+1)^2}{2n+3}(1-\delta_{nN})A_{n+1} + \dfrac{n(n-1)}{2n-1}(1-\delta_{n2})A_{n-1}\right] \\ = \displaystyle\sum_{k=1}^{P} \mu_k \beta_{nk}, \\ \displaystyle\sum_{n=2}^{N} A_n(\tau)\beta_{nk} = 0, \end{cases}$$

where $\mu_k$ ($k = 1, 2, \ldots, P$) form the set of the desired independent Lagrange factors. After elimination of these factors, the system of equations can be represented in matrix form:

Natural frequencies for two complementary drops of water on a solid substrate with opening angles $\theta_0 = 60°$ and 120° and with three characteristic radii

| Number of normal mode | $R$=1 mm | $R$=5 mm | $R$=10 mm | Dimensionless natural frequencies |
|---|---|---|---|---|
| | Natural frequencies, Hz | | | |
| 3 | 260 | 23 | 8 | 6.0954 |
| 4 | 413 | 37 | 13 | 9.6621 |
| 5 | 476 | 43 | 15 | 11.1511 |
| 6 | 571 | 51 | 18 | 13.3598 |
| 7 | 719 | 64 | 23 | 16.8276 |
| 8 | 950 | 85 | 30 | 22.2372 |
| 9 | 1008 | 90 | 32 | 23.6130 |
| 10 | 1290 | 115 | 41 | 30.1978 |

$$\ddot{X}^m = CX^m. \tag{11}$$

Here, $X^m = [a_2^m; a_3^m; \ldots; a_N^m]$ is the column vector of the unknown amplitudes of partial harmonics for the $m$th normal mode and $C$ is the known constant matrix. The homogeneous system of equations (11) will be solved when, for the matrix $C$, we determine the eigenvalues $\lambda_1, \lambda_2, \ldots, \lambda_{N-1}$ and the eigenvectors $[a_2^m; a_3^m; \ldots; a_N^m]$ for each of the $\lambda_m$. The eigenvalues are found from the characteristic equation $\det(C - \lambda E) = 0$, where $E$ is a $(N-1) \times (N-1)$ unit matrix. The set of the eigenvectors $X^m$ of matrix $C$ satisfies the equation $CX^m = \lambda_m X^m$. The eigenvalues are related to the natural frequency by the formula $W_m = \sqrt{-\lambda_m}$.

Figure 3 shows the calculated shapes of normal modes and the corresponding natural frequencies (table) for two drops with different wetting angles: $\theta_0 = 60°$ and $\theta_0 = 120°$. The normal modes are numbered according to the same principle as that used in [26]: the number $m$ of the normal mode is identical to the number of nodes within half of the deformed drop's profile. It should be noted that, in the case of drop oscillations at a resonance frequency, the shape of the drop's surface gradually acquires the shape of the corresponding normal mode irrespective of the initial conditions.

Now, we proceed to considering the dynamics of a drop on a substrate performing harmonic oscillations. Let the following law govern the dimensionless displacement of the substrate:

$$\zeta(\tau) = \frac{\zeta_0}{R} e^{-i(W\tau + \pi/2)},$$

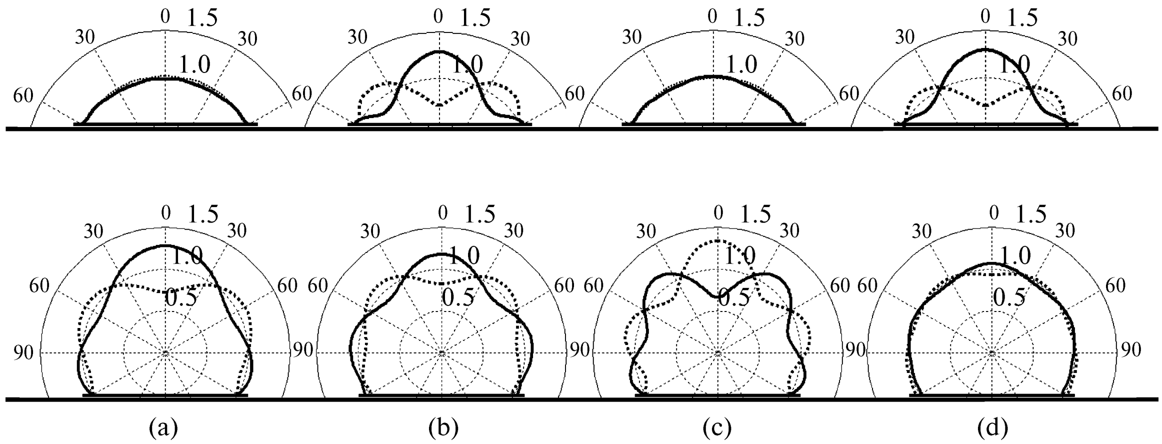


**Fig. 3.** Shapes of normal modes for a water drop with a radius of 1 mm on a substrate (the upper row refers to a drop with an opening angle of 60°, and the lower row to a drop with an opening angle of 120°). The solid and dotted lines show antiphased oscillations for the (a) 3rd, (b) 4th, (c) 5th, and (d) 6th modes. The natural frequencies are given in the table.

where $W$ is the dimensionless cyclic frequency of substrate oscillations. To describe the deformation of the drop surface, we change to the noninertial frame of reference rigidly connected with the substrate. In this case, the external force density field involved in the equation of motion of the liquid in the drop can be represented as $\mathbf{f} = \mathbf{g} - \mathbf{a}$, where $\mathbf{g}$ is the free fall's acceleration vector and $\mathbf{a}$ is the substrate's acceleration vector varying with time. Thus, the change to the noninertial frame of reference is equivalent to the introduction of a time-dependent gravitational field, whose acceleration is directed along the $OZ$ axis (see Fig. 2), in the equation of motion:

$$\frac{\partial}{\partial t}\mathbf{v} + (\mathbf{v}\nabla)\mathbf{v} = -\frac{\nabla p}{\rho_f} + \mathbf{g} - \mathbf{e}_z\frac{\partial^2\zeta}{\partial t^2}, \qquad (12)$$

where $\mathbf{e}_z$ is the unit vector along the $OZ$ axis.

The inclusion of the variable gravitational field leads to modification of boundary condition (2), which, in terms of the dimensionless variables, takes the form

$$\frac{\partial^2\Phi}{\partial\tau^2} + K\left[(1+a)\frac{\partial\Phi}{\partial X} + \frac{\partial a}{\partial\tau}\frac{\xi}{R}\right]\cos\theta - (2+\Delta_\Omega)\frac{\partial\Phi}{\partial X}$$
$$= -K\frac{\partial a}{\partial\tau}(\cos\theta - \cos\theta_0)\bigg|_{X=1}, \qquad (13)$$

where $a$ is the substrate's acceleration normalized by $g$. Note that, in the general case, boundary condition (13) on the free drop's surface is of the integro-differential type because the liquid particle's displacements and the velocity potential are related by an integral dependence. In the particular case of harmonic variation of these quantities with time, the aforementioned dependence degenerates into an algebraic one. Substitution of series (5) in boundary condition (13) according to the scheme described above leads to a system of dimensionless equations describing the partial mode's amplitudes of a free spherical drop in a variable gravitational field:

$$\ddot{A}_n + \omega_n^2 A_n + K(1+a)\left[\frac{(n+1)^2}{2n+3}A_{n+1} + \frac{n(n-1)}{2n-1}A_{n-1}\right]$$
$$+ K\frac{\partial a}{\partial\tau}\left[\frac{n+1}{2n+3}f_{n+1} + \frac{n}{2n-1}f_{n-1}\right] = 0,$$
$$\omega_n^2 = n(n+2)(n-1), \quad n = 2, 3, \ldots, N.$$

Applying the D'Alembert–Lagrange principle to the coupled system in a variable gravitational field, we obtain:

$$\begin{cases} \ddot{A}_n + \omega_n^2 A_n + K(1+a) \\ \times\left[\frac{(n+1)^2}{2n+3}(1-\delta_{nN})A_{n+1} + \frac{n(n-1)}{2n-1}(1-\delta_{n2})A_{n-1}\right] \\ + K\frac{\partial a}{\partial\tau}\left[\frac{n+1}{2n+1}(1-\delta_{nN})f_{n+1} + \frac{n}{2n-1}(1-\delta_{n2})f_{n-1}\right] \\ = \sum_{k=1}^{P}\mu_k\beta_{nk}, \\ n = 2, 3, \ldots, N, \\ \sum_{n=2}^{N}\beta_{nk}A_n(\tau) = 0, \quad k = 1, 2, \ldots, P, \\ \frac{df_n}{d\tau} = nA_n(\tau). \end{cases} \qquad (14)$$

To solve system (14), we applied the Runge–Kutta method accurate to fourth-order terms with the use of the following initial conditions: at zero time $t = 0$, the $z$th velocity component of liquid particles at the substrate surface coincides with the substrate velocity

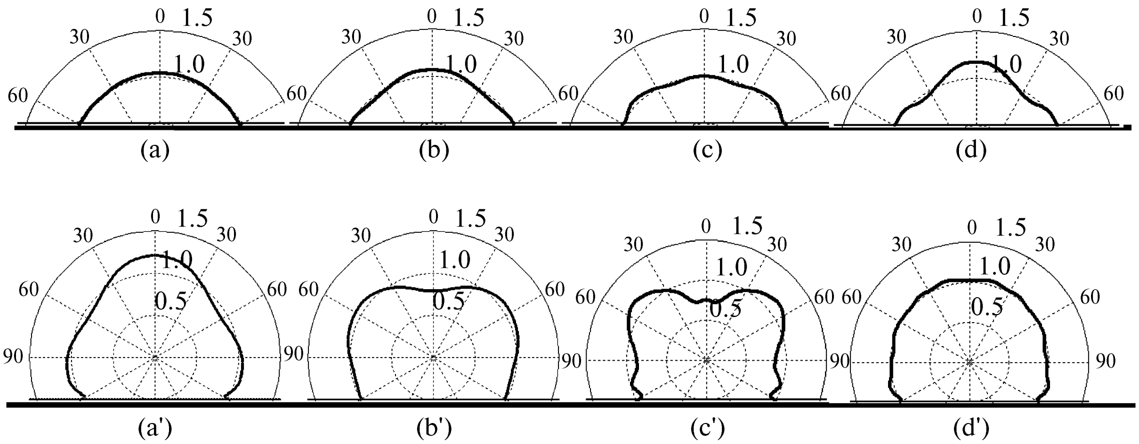


**Fig. 4.** Shapes of drops (the parameters are identical to those in Fig. 3) on a substrate vibrating with a frequency of 1900 Hz and a displacement amplitude of 10 μm for different instants of time: $t$ = (a, a') 0.0149, (b, b') 0.0168, (c, c') 0.0302, and (d, d') 0.0343 s. The period of the vibrating substrate is 0.526 ms.

while the liquid particles outside this thin layer are assumed to be immobile. The law governing the motion of the substrate is chosen so that, at $t = 0$, its displacements and, hence, the displacements of the liquid particles are zero. Figure 4 shows the profiles of the deformed drop's surface for different instants of time.

From the results of numerical simulation, it follows that deformation of the drop surface may be considerable. Presumably, this is related to the potential flows arising inside the drop under the effect of the external force's acceleration $\mathbf{f} = \mathbf{g} - \mathbf{a}$. One can expect that their evolution is determined by not only the magnitude of the external force $\mathbf{F}_{ext} = m\mathbf{f}$ ($m$ is the mass of the drop), but also the inertia of the liquid layers. The latter property can be characterized by the time $\Delta t_i$ that is required for a liquid volume's element to "respond" to a variation in the velocity $v_{surf}$ of the moving substrate. Since $\mathbf{F}_{ext}$ acts in the vertical direction, we conditionally divide the drop into layers in the direction of the $OZ$ axis, i.e., according to their distance from the supporting plane (see Fig. 2). In view of condition (3), the layer adjacent to the solid–liquid interface does not move along the $OZ$ axis with respect to the substrate; therefore, the characteristic time $\Delta t_i$ for this layer is much smaller than that for the layers separated from the substrate by certain distances. If the characteristic time $1/F$ of the variation of the external force is comparable with $\Delta t_i$ and, at the same time, $a$ reaches considerable values (according to the results of numerical simulation, considerable values of $a$ are those greater than 10), it is possible to initiate a regime with a considerable deformation of the drop surface and an effective excitation of low-order modes. At such parameters of external action, for drops with an angular size $\theta_0 < \pi/2$, the system may pass to a nonlinear regime accompanied by the detachment of microdrops from the surface [26]. Based on the results of numerical simulation, we assume that the drop's atomization regime can be initiated if the frequency of substrate oscillations is sufficiently low, so that the drops have enough time to "respond" to changes in the substrate velocity. On the other hand, the frequency should be sufficiently high for the plane to reach sufficiently high acceleration $\mathbf{a}$ in the course of its motion. It is important to note that, in the case of choosing the optimal frequency, the deformation of the drop's surface considerably depends on the substrate's displacement amplitude $\zeta_0$.

At high frequencies of substrate oscillations, when $1/F \ll \Delta t_i$ (according to numerical simulation, for a system with the chosen parameters, this condition is satisfied by frequencies of several kilohertz and higher), one should presumably expect a softer regime, at which the perturbation is localized near the drop surface. Possibly, persistence of such a regime will favor an increase in the rate of drop evaporation without spraying (atomization). Indeed, it is under these conditions that the energy of substrate motion can theoretically be localized in the kinetic energy of the motion of a liquid particle in a thin layer of the drop, which should facilitate separation of molecules from the liquid surface. Generation of such a regime may be useful for practical implementation of technological processes that require fast evaporation of drops from a solid surface in the absence of a pronounced convection of the liquid inside the drops.

Returning to the idea of controlling the self-assembly and self-organization of nanostructures in microdrops by vibration action, we note that such an action on the nanostructure's self-organization process is of a multifactor character. First, vibration directly affects the dynamics of nanoparticles in the drop. Second, it affects (controls) the microdrop's evaporation process. Third, the perturbed structure of the drops surface will "manifest" itself in the pattern that remains on the substrate after evaporation. All of these factors are initially determined by the spatial modes excited in the drop by vibration, i.e., by the modes studied in the present paper.


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