DYNAMICS OF A VAPOR – GAS BUBBLE FORMED BY LASER BREAKDOWN IN A LIQUID

Yu. I. Lysikov

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§1. Laser breakdown in a liquid is accompanied by the formation of a pulsating vapor–gas bubble in the breakdown zone. The behavior of the bubble pulsations has been studied in adequate detail experimentally. Buzukov and others [1] have published the results of a high-speed motion-picture study of a vapor–gas void in the focal region from the instant of formation to a time corresponding to termination of the second pulsation. The reported experimental data indicate a certain complexity in the overall behavior of the void.

On the other hand, certain physical phenomena attending the collapse of a void depend significantly upon the conditions existing during collapse. The primary considerations here are laser sonoluminescence [2], which arises in the final stage of compression of a bubble in the breakdown zone, as well as shock waves propagating in the liquid away from the focal region. The pulsation period and the values of various parameters of the vapor–gas void (pressures and temperatures during collapse) are estimated theoretically in most cases on the basis of an analysis of the spherically symmetrical problem. This approach is fully justified in calculations of powerful explosive processes in a liquid, where the source effectively has point size relative to the dimensions of the zone of outrushing matter. The vapor–gas void formed in the case of laser breakdown has at the instant of conception quite an elongated configuration, which significantly affects its subsequent growth and pulsation. Spherical symmetry can no longer be presumed, because the void has the shape of an ellipsoid of revolution with different ratios between the major and minor axes at different times. In the theoretical calculations it is necessary to invoke methods accounting for the lack of spherical symmetry of the problem.

For the dynamical analysis of a vapor–gas void under the stated conditions during the nascent period corresponding to the first pulsation it should be possible to use the approach developed in [3–5] in order to describe the explosive expansion of a nonspherical gas cloud. The principal hypothesis underlying this approach is that the motion of the gas can be described by the relation $x_i = \sum_{k=1}^{3} F_{ik}(t)\, a_k$ (i, k = 1, 2, 3), in which x_i denotes the coordinates of the i-th gas particle and a_k, its Lagrangian coordinates. From the hydrodynamical equations we obtain in this case nine second-order ordinary differential equations for the functions $F_{ik}(t)$. However, in the fairly simple case of radial expansion of a gaseous spheroid having the shape of an ellipsoid of revolution we assume adiabaticity of the expansion process to obtain two equations in two variables (F_1 and F_3) corresponding to the values of the minor and major axes of the spheroid at different times. The equations for the corresponding case of expansion of a gas cloud in vacuum have already been solved [5]. The counterpressure of the liquid becomes essential in the calculations for pulsations of a vapor–gas void in a liquid, insofar as it induces pulsations and compression rather than pure unbounded expansion. Consequently, in order to be able to apply the equations of the cited works to the case at hand we must augment those equations to account for the counterpressure of the medium.

§2. To derive the analytical relations we use the hydrodynamical equations

$$d\rho/dt = \partial\rho/\partial t + \mathbf{v}\partial\rho/\partial\mathbf{x} = -\rho\partial\mathbf{v}/\partial\mathbf{x},$$
$$d\mathbf{v}/dt = \partial\mathbf{v}/\partial t + (\mathbf{v}\partial/\partial\mathbf{x})\mathbf{v} = -(1/\rho)\partial p/\partial\mathbf{x} \qquad (2.1)$$

and the first law of thermodynamics for a gaseous medium expanding adiabatically

$$dU = (p/\rho^2)d\rho,\ p = (2/i)\rho U, \qquad (2.2)$$

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where U is the internal energy of the gas per unit mass, ρ is the density of the gas, i is the number of molecular degrees of freedom, and p is the pressure. For a comparatively rarefied gas the density and pressure inside the void depend weakly on the coordinate, so that the corresponding dependences can be neglected. Introducing the transformation of variables

$$x=F_1a_1,\ y=F_2a_2,\ z=F_3a_3\quad (F_1=F_2), \tag{2.3}$$

we obtain from (2.1) and (2.2) by analogy with Dyson [4]

$$\rho=\frac{\rho_0}{\varphi},\quad U=\frac{U_0}{\varphi^{2/i}},\quad \varphi=F_1^2F_3. \tag{2.4}$$

The transformation to the new coordinates in the second equation (2.1) is more complicated than in the cases treated in [4, 5]. The derivative of the pressure with respect to the coordinate at the boundary of the spheroid is not as trivial a function of the coordinate normal to the surface of the spheroid as in the main gas interior. In transition from the gaseous to the liquid medium the density and pressure change abruptly (but not so for the velocity), whereupon the derivative of the pressure along the normal to the surface behaves as a delta function. With this fact in mind we discern the boundary value of the derivative of the pressure:

$$dv_i/dt = -(1/\rho)\partial p/\partial x_i - (1/\rho)(\partial p/\partial x_i)_0. \tag{2.5}$$

The presence of the boundary will necessarily have a significant effect on the behavior of the functions F_i, because they determine the velocity and total range of expansion of the gas, and it is quite obvious that the intrusion of the boundary with the liquid will cause the unbounded expansion process to be replaced by a finite pulsating motion. To obtain the equations for F_i we write (2.5) in the form

$$\ddot{F}_i a_i = -n_i(\partial p/\partial \mathbf{n})_0, \tag{2.6}$$

where $\mathbf{n}$ is the unit vector normal to the spheroid surface. The geometry of the investigated vapor–gas void is such that the level surfaces for the given functions are described by the relation

$$\frac{x^2+y^2}{F_1^2a^2}+\frac{z^2}{F_3^2a^2}=1.$$

The transformation (2.3) takes the spheroid into a sphere, for which it is convenient to introduce polar coordinates:

$$a_1=a\sin\vartheta\cdot\cos\varphi,\quad a_2=a\sin\vartheta\cdot\sin\varphi,\quad a_3=a\cos\vartheta.$$

Recognizing the fact that all the variables depend only on the radial coordinate a and using the definition of the normal to a surface, as well as the normal derivative from (2.6), we obtain

$$\ddot{F}_1 a\sin\vartheta\cdot\cos\varphi=-\frac{\sin\vartheta\cdot\cos\varphi}{\rho F_1}\left(\frac{\partial p}{\partial a}\right)_0,$$
$$\ddot{F}_2 a\sin\vartheta\cdot\sin\varphi=-\frac{\sin\vartheta\cdot\sin\varphi}{\rho F_2}\left(\frac{\partial p}{\partial a}\right)_0,$$
$$\ddot{F}_3 a\cos\vartheta=-\frac{\cos\vartheta}{\rho F_3}\left(\frac{\partial p}{\partial a}\right)_0.$$

In these equations the angular dependence is completely separated, and it is possible to integrate with respect to the radial coordinate from zero to the boundary value a_0. Inasmuch as $F_1=F_2$, only two equations are left:

$$\ddot{F}_1\frac{a_0^2}{2}=-\frac{1}{F_1}\int\limits_{p(a_0-0)}^{p(a_0)}\frac{dp}{\rho},\quad \ddot{F}_3\frac{a_0^2}{2}=-\frac{1}{F_3}\int\limits_{p(a_0-0)}^{p(a_0)}\frac{dp}{\rho}.$$

The integral (which we denote by I) on the right-hand sides of the equations cannot be computed without knowing the detailed behavior of the pressure and density in the boundary layer. However, since ρ is independent of a and $I=p/\rho$ at the boundary on the gas side, a_0-0, while at the boundary on the liquid side, a_0,

$$p=A/\rho_1^n-B,\quad n\sim 7,\quad I=\frac{n}{n+1}\frac{p}{\rho_1}\approx\frac{p}{\rho_1},$$

it may be assumed with good accuracy that

$$I = -\frac{p'}{\rho_1} + \frac{p(a_0 - 0)}{\rho},$$

where ρ_1 is the density in the liquid at the boundary with the gas and p' is the pressure, which must be corrected for the increment due to surface tension. Using (2.2) and (2.4), we arrive at the system of equations

$$\ddot{F}_1 = \frac{4U_0}{ia_0^2 F_1 \varphi^{2/i}} - \frac{2}{a_0^2 F_1}\frac{p'}{\rho_1}, \quad \ddot{F}_3 = \frac{4U_0}{ia_0^2 F_3 \varphi^{2/i}} - \frac{2}{a_0^2 F_3}\frac{p'}{\rho_1}.$$

Rigorous correction for the surface tension in the pressure equation would yield a nonseparable angular dependence. Inasmuch as we are discussing pulsations of a void of fairly large radius, for which surface tension incurs only a minor correction, we can introduce the latter in an approximate fashion by putting $p' = p_1 + (\sigma/a_0)(1/F_1 + 1/F_3)$, where p_1 is the pressure in the liquid and σ is the coefficient of surface tension. The determination of p_1 from the hydrodynamical equations for the liquid poses formidable computational difficulties. The result is not qualitatively affected if we consider p_1 to be equal to the average value of the pressure in the liquid for one pulsation period. We thus obtain finally

$$\ddot{F}_1 = \frac{4U_0}{ia_0^2 F_1 (F_1^2 F_3)^{2/i}} - \frac{2}{a_0^2 F_1}\frac{p_1}{\rho_1} - \frac{2\sigma}{a_0^3 F_1 \rho_1}\left(\frac{1}{F_1} + \frac{1}{F_3}\right),$$
$$\ddot{F}_3 = \frac{4U_0}{ia_0^2 F_3 (F_1^2 F_3)^{2/i}} - \frac{2}{a_0^2 F_3}\frac{p_1}{\rho_1} - \frac{2\sigma}{a_0^3 F_3 \rho_1}\left(\frac{1}{F_1} + \frac{1}{F_3}\right). \quad (2.7)$$

It is clearly impossible to solve these equations analytically, and instead we proceed numerically.

§3. We solved Eq. (2.7) numerically on a BÉSM-4 computer by the Runge–Kutta method. We chose the values for the parameters according to considerations of correspondence with the experimental conditions of [1]. Certain difficulties arise in this selection process, because the experimental data are not such as to permit a clear-cut unambiguous selection and, in addition, the system (2.7) clearly becomes inapplicable for describing the process after a certain lapsed "relaxation" time of the order of a few microseconds following breakdown. At the instant of breakdown the generated void grows rapidly along the beam to large dimensions, since the liquid is strongly heated in that direction and violent vaporization takes place in the resulting void. It is not at all clear whether we are concerned with formation of the void at the initial instant from a large number of minute bubbles produced on the beam axis or with an outrush of gas along the axis away from the focal region, but it is obvious that either way the process is extremely nonadiabatic and is accompanied by vigorous vaporization of liquid from the wall into the void.

Therefore, our model is not suitable for describing the process until the void attains appreciable dimensions in the axial direction. Accordingly, we adopt the following values for F_i after an analysis of motion-picture films and data on the dimensions of the focal region [1]:

$$F_1(0) = 1.25,\ F_3(0) = 50,\ \dot{F}_1(0) = 0,\ \dot{F}_3(0) = 1.5 \cdot 10^5\,\text{sec}^{-1};\ a_0 = 0.5 \cdot 10^{-4}\ \text{m}. \quad (3.1)$$

Allowance is made here for the fact that the void diameter is given by twice the value of $F_i a_0$. The value of p_1 can be taken equal to the pulsation-period-average pressure, which in turn is equal to the pressure at infinity, i.e., in the given situation to atmospheric pressure. The density of water is $\rho_1 = 1$ g/cm^3, and its coefficient of surface tension σ is equal to 74 dyn/cm. To estimate U_0 we note that if the gas expanded at the initial instant adiabatically from a focal region of volume $\sim \frac{4\pi}{3} a_0^3$ to the volume given by (3.1), then with regard for the estimated initial pressure in [1] its pressure would have to be the order of several atmospheres. On the basis of the latter consideration and the fact that the intense vaporization at the initial instant makes the density of the gas at that time close to the density of the liquid, we obtain from the pressure equation $v_0^2 \sim 10^4$ m^2/sec^2. Next we introduce the dimensionless time $t' = t/\tau$, where $\tau = ka_0/c$ and $c = 1.5 \cdot 10^3$ m/sec is the velocity of sound; we adopt as the average characteristic void dimension ka_0 a value of $\sim 15a_0$.

Equations (2.7), subject to the initial conditions (3.1), assume the dimensionless form

$$\ddot{F}_1 = \frac{1}{F_1 (F_1^2 F_3)^{0.67}} - \frac{0.01}{F_1} - \frac{0.001}{F_1}\left(\frac{1}{F_1} + \frac{1}{F_3}\right),$$
$$\ddot{F}_3 = \frac{1}{F_3 (F_1^2 F_3)^{0.67}} - \frac{0.01}{F_3} - \frac{0.001}{F_3}\left(\frac{1}{F_1} + \frac{1}{F_3}\right),$$
$$F_1(0) = 1.25,\ F_3(0) = 50,\ \dot{F}_1(0) = 0,\ \dot{F}_3(0) = 0.07.$$

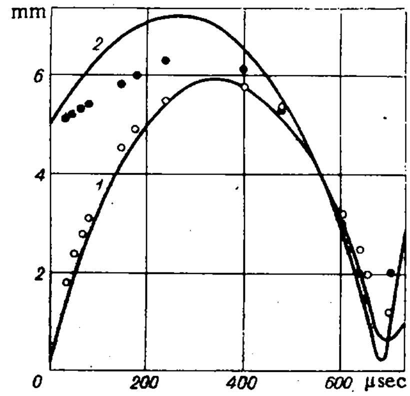

Fig. 1

The solution of this system is given in Fig. 1, along with experimental data obtained on the basis of motion-picture frames [1] for the void dimensions. Curve 1 corresponds to twice the value of $F_1 a_0$, curve 2 to $2F_3 a_0$, and the dark and light circles to the experimental values of the respective dimensions.

The calculations disclose an interesting property of the void, which is also discernible in the motion pictures of [1]. Starting from a shape greatly elongated along the beam axis and evolving through a spherical shape midway in the pulsation cycle, the void ultimately compresses into an oblate spheroid. As we mentioned in the introductury section, the final compression phase is immensely valuable in explaining the intensity of the ensuing laser sonoluminescence. Thus, the final compression phase is characterized by a discus-like void. It is interesting to note that the subsequent expansion takes place in both experiment and calculations in such a way as to interchange the former positions of the major and minor axes of the ellipsoid. The spheroid is drawn out along the beam, and the growth of the transverse dimensions lags behind the growth of the longitudinal. Clearly, at the instant of compression under the experimental conditions the process becomes strongly nonadiabatic with condensation (or vaporization) of the gas on the walls, and the application of our previous calculations to the subsequent stages of motion becomes questionable. However, during the period of the first pulsation the correspondence between the experimental and analytical curves is reasonably good, and the analytically predicted interchange of the major and minor axes of the spheroid in the compression phase is very intriguing from the standpoint of being able to explain the concomitant luminescence intensity.

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