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LASER SONOLUMINESCENCE IN WATER UNDER INCREASED HYDROSTATIC PRESSURE

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UDC 534.29:535.37

The nature of the fairly recently discovered [1-3] pulsed luminescence accompanying the collapse of a single cavity initiated by laser breakdown in a liquid has still not been clarified, despite the importance of solving questions associated with the determination of the state variables of a substance in the vicinity of the special point during spherically symmetric collapse [4-6], which is manifested most distinctly, as was pointed out in [7], in the case of the laser method (as distinct from a high-voltage discharge or exploding wires) of initiation of inhomogeneities.

In [8] attention was concentrated on the purely recombinational mechanism of generation of the luminescence accompanying the collapse of a cavity initiated by laser breakdown in liquid nitrogen. We refer here to the recombination of active particles formed at the instant of spark breakdown. It is quite natural to postulate the role of processes of this type in sonoluminescent effects in water and aqueous solutions. The active particles contributing to laser sonoluminescence (LSL) in water could be, at least for several collapse regimes, H and OH radicals, which have been shown to play an appreciable role in the luminescence accompanying the pulsations of a microscopic cavity in an ultrasonic field of subcavitation intensity [9].

In the present investigation we availed ourselves of the opportunity to "control" the moment of collapse by using the hydrostatic pressure (due to the sharp dependence of the cavity pulsation period on the hydrostatic pressure: $T \sim 1/p^{5/6}$ [10]) and attempted an experimental and theoretical evaluation of the contribution of the luminescence component due to recombination of active particles (particularly H and OH radicals), initiated at different stages in the life of the cavity, including the moment of breakdown.

In addition, by conducting pressure experiments in water at temperatures close to the boiling point we could stimulate to some extent the thermodynamic situation in the induction of cavitation luminescence in liquid nitrogen and other cryogenic liquids (Ar, Xe, Ne).

1. Experimental Results and Their Brief Analysis. The apparatus used for the experiment was similar to that described in [11]. The working chamber, containing the water in which laser breakdown occurred ($E_L = 0.02$ J, $\tau \sim 10^{-8}$ sec), could withstand hydrostatic pressures up to $8 \cdot 10^6$ Pa. The luminous effects accompanying the breakdown of the liquid and the subsequent collapse of the cavity were detected by an FÉU-13 photomultiplier. The intensity of the shock-acoustic effects was determined by means of a hydrophone, based on the piezoceramic TsTS-19, with a resolving time of $\sim 3 \cdot 10^{-7}$ sec. The information obtained through the acoustic and light channels, i.e., from the FÉU and the hydrophone, was fed to an S8-2A two-beam storage oscillograph.

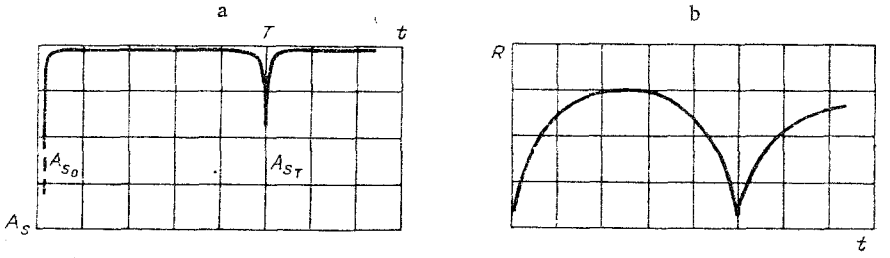

Fig. 1

Voroshilovgrad. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 4, pp. 28-33, July-August, 1981. Original article submitted May 5, 1980.

0021-8944/81/2204-0461$07.50

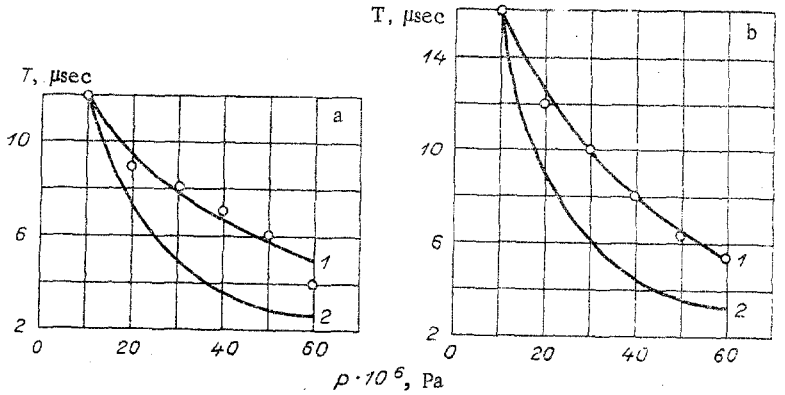

Fig. 2

Figure 1a schematically illustrates a typical result, from the analysis of which we obtained the data presented below. The first pulses on the time sweep correspond to laser breakdown, i.e., to the moment of initiation; the second correspond to collapse of the cavity. The time interval between breakdown and collapse will be called the pulsation period T of the cavity. Figure 1b shows the change in cavity radius during this period of time.

Using distilled water at two temperatures (t = 20 and 80°C) we obtained data for the relation between the cavity pulsation period and the hydrostatic pressure (Fig. 2a, b, curve 1), for the relation between the total amplitude A_S of laser sonoluminescence and the hydrostatic pressure (Fig. 3a, b, curve 1), and also for the parameter $K = A_{SV}/A_{SR}$ (Fig. 4, curves 1, 2), which characterizes the ratio of the violet and red LSL components and which, in our view, must be sensitive to any alteration of the photon generation mechanism by external factors — temperature and hydrostatic pressure.

An analysis of the curves in Figs. 2-4 reveals the following. First, the expected mechanism — recombination of H and OH radical pairs — will most probably operate at t = 80°C and low hydrostatic pressures in view of the relatively low degree of compression of the cavity and the greatest value of the pulsation period T in such conditions, i.e., we are actually saying that in such conditions there is hardly any temperature boost at the moment of collapse and there is a relatively long time ($\sim$T) for cooling of the postbreakdown plasma. Hence, the best agreement between the experiment and a theory that takes into account the considered process is to be expected when the thermodynamic conditions are not severe [t = 80°C, $p_{st} \sim (0.1\text{-}3)\cdot 10^6$ Pa]. This is confirmed by the sharp change and subsequent leveling out of the parameter K (see Fig. 4), which can be interpreted as a contest of several competing mechanisms [in the range $p_{st} > (0.1\text{-}3)\cdot 10^6$ Pa, where K varies strongly] with the subsequent predominance of one of them (in the range $p_{st} \sim 4\cdot 10^6$ Pa, where K is practically constant).

Second, the regions which differ sharply from one another in magnitude of K (in this case the range of static pressures is the same and the temperatures are different) are of interest. It is apparent also that in this region K remains constant, which probably indicates a predominant contribution of some specific mechanism of generating electromagnetic radiation quanta. The scope of this work did not include an analysis of these mechanisms, which are

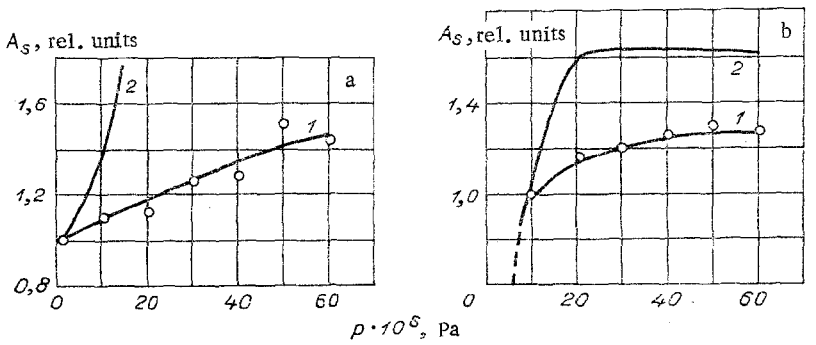

Fig. 3

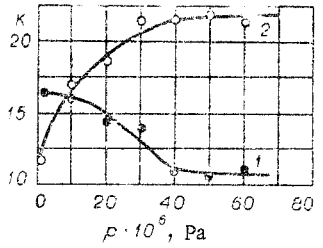

Fig. 4

obviously connected with the contribution of such processes as radiative recombination of electrons and ions, and also the bremsstrahlung of electrons on ions (at high degrees of ionization) and neutral atoms (at degrees of ionization $<10^{-2}$).

On the basis of the above we will try to construct, for conditions t = 80°C, $p_{st} \sim (0.1-3)\cdot 10^6$ Pa, a simple theoretical model that takes into account mainly the recombination of radical pairs that survive from the moment of breakdown till the moment of collapse.

2. Discussion of Experimental Results. Theoretical Model. Laser breakdown of distilled water produces a low-temperature plasmoid, which cools down during the subsequent pulsation of the cavity. Thus, at the time of formation of the cavity it contains a mixture of ions, electrons, and radicals [10]. During the subsequent expansion of the cavity rapid recombination of ions with electrons occurs and by the moment of maximum expansion of the cavity H and OH radicals and neutral molecules can predominate in it.

We consider the recombination of H and OH radicals within the pulsating vapor–gas cavity on the basis of very simple ideas of classical collision theory [6]. Let us assume that the breakdown of water gives rise to N_0 radical H and OH pairs in the cavity, which in the course of time recombine to form H_2O molecules. We consider the recombination of radicals together with the reverse process – the dissociation of vapor molecules to radicals, i.e., during collapse the thermodynamic conditions promote considerable dissociation within the cavity. Hence, the rate of radical recombination is given by the equation

$$dN/dt = -\sigma_r v_r N^2/V + \sigma_d v_d N_M^2/V, \tag{2.1}$$

where σ_r and σ_d are the recombination and dissociation cross sections, respectively; v_r and v_d are the mean relative velocities of the radicals and molecules, respectively; N is the number of radical pairs; N_M is the number of neutral H_2O molecules; V is the volume of the cavity.

Recombination of H and OH radicals to form H_2O molecules releases a large amount of energy, which must be removed for stabilization of the molecule. If all the energy is converted to vibrational energy, then after one oscillation the molecule will dissociate again. An obvious channel for energy removal is the radiative mechanism, but with increase in vapor–gas concentration in the collapsing cavity the probability of radiative recombination is reduced and the probability of exchange recombination is increased, i.e., at sufficiently high concentrations excess energy is removed by a third particle as a result of a triple collision [12]. Proceeding from the above considerations we express the rate of radiative recombination in the form

$$\frac{dN'}{dt} = \frac{k'}{(k_d + k_M) n_M} \frac{dN}{dt}, \tag{2.2}$$

where N' is the number of radical pairs recombining with radiation; k' is the rate constant of radiation; k_d is the rate constant of dissociation of excited molecules; k_M is the rate constant for exchange of energy of an excited molecule with a stable molecule; n_M is the concentration of stable H_2O molecules.

The rate of recombination of H and OH radicals within the cavity depends significantly on its dynamics. We assume that the vapor–gas cavity pulsates in an incompressible viscous liquid, and the processes within the cavity follow an adiabatic law. With those assumptions the dynamics of the cavity can be represented by the known equation [13]

$$R\frac{d^2R}{dt^2}+\frac{3}{2}\left(\frac{dR}{dt}\right)^2=\frac{1}{\rho}\left[p_0\left(\frac{R_0}{R}\right)^{3\gamma}-p_{st}-\frac{2\sigma}{R}-\frac{4\mu}{R}\frac{dR}{dt}\right]. \quad (2.3)$$

Equations (2.1)-(2.3) were solved numerically on a computer by the Runge-Kutta method for different hydrostatic pressures and temperatures of distilled water.

1. The numerical solution of the above equations shows pulsating variations of the rate of radiative recombination during pulsation of the cavity, which is qualitatively consistent with the experimental data.

2. The pulsation periods of cavities in water under increased hydrostatic pressure are an order lower than the pulsation periods at normal static pressure, given by the known relative $T \sim (p_{st} - p_p)^{-5/6}$. A comparison of the experimental relation with the theoretical one (see Fig. 2a, b, curve 2) shows that for both temperatures the experimental values of the periods are somewhat greater (by up to 30%) than the theoretical values, and the divergence increases with increase in pressure and temperature. This fact probably indicates that the cavity contents (mainly laser breakdown products) do not cool to the ambient temperature on attainment of its maximum volume, as is implied by an adiabatic process. This circumstance and also the probable contribution of other ignored physical effects and processes will, of course, affect the degree of agreement between the theoretical (based on the developed recombination model) and experimental relations: $A_s = f(p_{st})$.

3. An analysis of the variation of the radiative recombination rate in relation to hydrostatic pressure of the water shows that at a water temperature of 80°C the radiative recombination rate increases with increase in water pressure up to $2\cdot10^6$ Pa and with further increase in pressure remains constant (see Fig. 3b, curve 2). This can be attributed to reduction of the radiative recombination rate constant, since an increase in hydrostatic pressure of the water increases the concentration of the cavity contents, and volume recombination begins to predominate. From the obtained results we can conclude that for each water temperature there will be a particular hydrostatic pressure at which the light output will be a maximum, i.e., the conditions for induction of LSL can be optimized.

A numerical analysis of Eq. (2.3) for the cavity dynamics at 20°C and various hydrostatic pressures showed that the temperatures within the cavity correspond to the first ionization region of the molecules and, hence, radical recombination can be appreciable only at hydrostatic water pressures up to $\sim1\cdot10^6$ Pa (see Fig. 3a, curve 2), and the main mechanism, particularly when $p_{st} > 1\cdot10^6$ Pa, is ion-electron recombination, very probably involving triple collisions. The experimentally observed further increase in absolute light output (see Fig. 3a, curve 1) and its subsequent stabilization at a relatively high level indicate that in such conditions an ever-increasing role is played by the bremsstrahlung radiation mechanism, which becomes predominant, according to the variation of K (see Fig. 4, curve 1), at pressures $p_{st} > 4\cdot10^6$ Pa.

We point out in conclusion that the developed approximate theoretical model predicts a number of effects that have not yet been experimentally confirmed. The first one is the relatively long duration of the LSL pulse, which depends appreciably on the hydrostatic pressure and temperature and, second, the increasing (with increase in pressure) time shift of the LSL pulse relative to the time of the "severe" phase of collapse, which is identified experimentally with the time of generation of the shock-acoustic pulse. The observed disagreement is probably due to the fact that either the mentioned effects (owing to their smallness) lies outside the limits of sensitivity of the apparatus used [the error of length measurement is $\sim(6-7)\cdot10^{-8}$ sec, and that of time delays is $\sim3\cdot10^{-7}$ sec], or it is possible that the observed anomalies are due to clustering of the vapor-gas material in the cavity or, in other words, to the "cluster plasma" with its specific instabilities (see [14]), which we think can be formed during laser breakdown and subsequent pulsation.

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HYDRODYNAMICS OF DISPERSED-ANNULAR GAS–LIQUID STREAMS IN BUNDLES OF RODS

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UDC 621.039.5:532.5

On the basis of concepts involving dispersed-annular gas–liquid streams in circular pipes [1, 2], within the framework of the cell model of the stream, we have constructed a one-dimensional stationary hydrodynamic model of the flow of a gas–liquid mixture in a dispersed-annular regime in channels with bundles of rods. We have analyzed the mass and force interactions between the components of the dispersed-film stream within the cells and between the cells. We obtained satisfactory agreement with the numerical and experimental data published in the literature with respect to hydraulic resistance in channels with bundles of heated rods having various geometries. We have shown the variation of the main hydrodynamic characteristics of the steam–water dispersed-annular stream over the cross section and along the length of the channel to be functions of the operational parameters of the mixture.

The development of calculation methods for the hydrodynamics and the heat-transfer crisis in channels with bundles of rods can follow two different lines: in the first place, it can proceed on the basis of methods using averaged parameters of the coolant in the channel; in the second place, it can be based on cell models taking account of convective and turbulent mixing of the phases between the cells. The first line has been developed to a considerable extent in the processing of direct experimental data on hydraulic resistance and resistance crises [3].

The second line is more general and flexible than the first, but it is, naturally, more cumbersome. The investigation of the hydrodynamics and the heat-transfer crisis in channels with bundles of rods, using cell representations, is usually carried out within the framework of a homogeneous model of a gas–liquid stream. This approach has been developed most coherently in [4].

At a volumetric concentration of the gaseous phase higher than 0.6-0.8 [5] the steam–water stream in a channel with bundles of rods moves in a dispersed-film regime of flow. The homogeneous model does not take account of the most characteristic features of such a flow,

Moscow. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 4, pp. 33-44, July-August, 1981. Original article submitted May 29, 1980.

0021-8944/81/2204-0465$07.50