Optical cavitation of transparent liquids exposed to broad-band laser light

N. F. Bunkin and V. B. Karpov
Institute of General Physics, Academy of Sciences of the USSR

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A new cavitation regime has been observed as laser light with a wide frequency spectrum $\Delta\omega$ interacts with a transparent liquid. The effect is shown to arise under the condition $\Delta\omega \gg (v/c)\omega_0 \equiv \Omega$ (ω_0 is the central frequency of the spectrum, c is the velocity of light, and v is the hypersound velocity in the liquid). A theoretical model is proposed on the basis of a new representation of a liquid: as a colloid of metastable clusters of ultramicroscopic gas bubbles.

The present experiments used ÉLI-91M XeCl lasers ($\lambda = 308$ nm, pulse length $\tau_p = 20$ ns, energy of 50 mJ) with an output linewidth $\Delta\omega/2\pi c \approx 15$ cm^{-1} $\gg \omega/2\pi c \approx 0.1$ cm^{-1}. The pump spectrum was also thinned out in the experiments (to obtain two lines of width <0.1 cm^{-1} separated by a frequency interval ≈ 0.7 cm^{-1}). The energy in the pulse in this case was 10 mJ.

Lenses directed the light pulse to a cell holding the test liquid (the size of the cell was either 5 cm or 20 μm). When an intensity J_{thr} was reached in the 5-cm cell, cavitation was observed over the length of the beam waist: Tracks of rising bubbles appeared. In the process we observed an abrupt increase (by a factor of about five) in the cross-sectional area of the beam transmitted through the cell, as a result of the light-scattering bubbles which formed at the waist over a time τ_p. The onset of cavitation in the 5-cm cell was accompanied by the simulations appearance of a Stokes signal of stimulated Brillouin scattering (with an intensity $\sim 10\%$ of J_{thr}). It was established that cavitation did not occur in the case of the thinned spectrum at the same pump intensity (or even at an intensity higher by one or two orders of magnitude). In this case, we observed a Stokes signal, and its intensity was on the order of the pump intensity.

In order to determine J_{thr} correctly, we used a cell 20 μm in size (the uncertainty in the value of J_{thr} in the 5-cm cell stemmed from the appearance of cavitation along the waist, i.e., over a distance of 0.5 cm). In addition, it was possible to observe and photograph the resulting bubbles in the case of the 20-μm cell. For these observations we used a fluorescent screen, which we placed 1–2 m from the cell along the beam path. The time evolution of the processes occurring within the cell was imaged on the fluorescent screen. The time at which J_{thr} was reached was determined from the appearance of characteristic circles on the screen (Fig. 1). Since the exposure time for these photographs was actually τ_p, it can be regarded as an established fact that the bubbles manage to form over ultrashort times. The 20-μm cell has the further convenience that the size of the bubbles is on the order to the size of the cell itself, so the

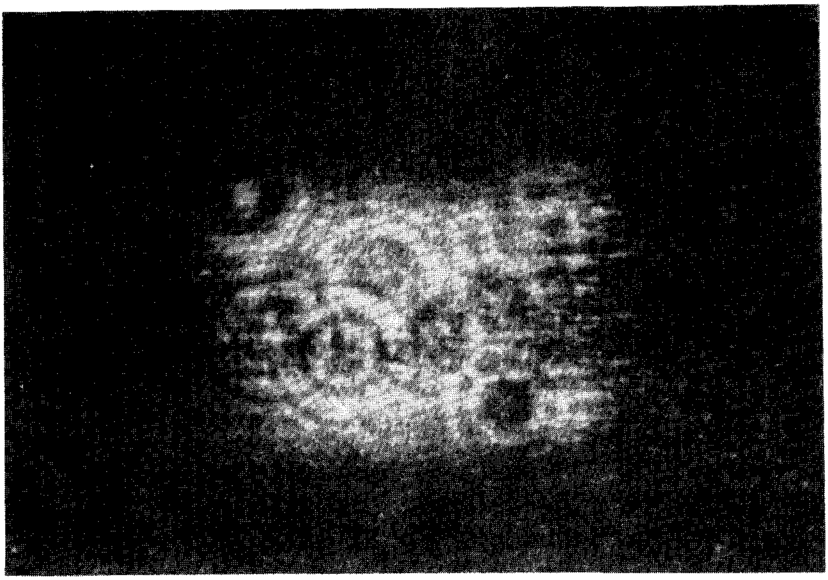

FIG. 1. Formation of cavitation bubbles in the 20-μm cell.

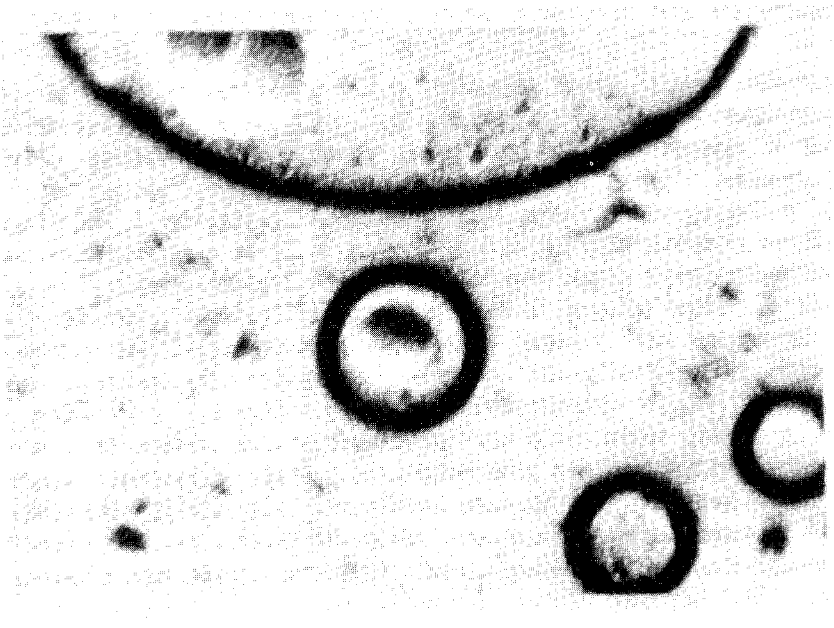

FIG. 2. Appearance of the cavitation bubbles in the 20-μm cell after the passage of a laser pulse. The bubble radius is $R^* \sim 10^{-4}$–10^{-3} cm.

bubbles adhere to the walls and can be studied with a microscope (Fig. 2).

Table I shows values of J_{thr} measured for several liquids, along with the values of $(\rho\partial\epsilon/\partial\rho)^2$.

It follows from this table that J_{thr} is inversely proportional to $(\rho\partial\epsilon/\partial\rho)^2$, indicating a relationship between the cavitation and the generation of hypersound (further evidence for this possibility comes from the fact that the appearance of cavitation is accompanied by the simultaneous appearance of Brillouin scattering in the 5-cm cell).

We checked to see whether the observed effect was related to the standard mechanisms for laser cavitation (see the review by Lyamshev and Naugol'nykh,[1] for example): the thermal effect and breakdown. The hydrodynamic mechanism and the acoustic mechanism for cavitation obviously have no bearing on the effect observed here.

The observed effect could not be related to surges of a fluctuation nature in the energy of the wide-band laser light, since in this case the effect would also be of a probabilistic nature (rather than of a regular nature, as it was in our experiments), and hydrodynamic processes would be totally impossible at a surge duration $t \sim 1/\Delta\omega \sim 10^{-11}$ s.

Let us take a theoretical look at this effect. Armandilo and Proch[2] have also observed cavitation accompanied by stimulated Brillouin scattering in experiments with a XeCl laser. They also noted, as a purely experimental fact, that the appearance of the cavitation depended on the spectral width $\Delta\omega$.

We believe that this effect stems from the final stage of coalescence (or collapse) of gas bubbles in the liquid. This coalescence would be stimulated by two oppositely directed hypersonic beams, which would result from the wide-band nature of the light. Their appearance in the case of the 5-cm cell is a consequence of the stimulated Brillouin scattering. Specifically, under the condition $\Delta\omega \gg \Omega$, it is possible to find a component of the pump with a frequency $(\omega - 2\Omega)$ for every spectral component of the pump with frequency ω. When the threshold for stimulated Brillouin scattering is reached, this $(\omega - 2\Omega)$ component becomes the Stokes component with respect to the $(\omega - \Omega)$ component propagating in the direction opposite the pump. In the case of the 20-μm cell, in which stimulated Brillouin scattering is not possible, but the incident laser light overlaps the laser light rereflected from the cell walls, oppositely directed hypersonic waves are excited by virtue of an electrostrictive effect [the hypersound intensity in either case is $J_s\,[\rho(\partial\epsilon/\partial\rho)]^2$; Table I]. As these hypersonic waves propagates through the medium and are absorbed in it, they give rise to a body force $f = 2\xi(\delta_0/v)$, where δ_0 is the amplitude attenuation coefficient of the hypersound, and ξ is an efficiency parameter. Since there are two oppositely directed waves in the medium, the resultant force is of a compressional nature.

TABLE I.

Substance	$(\rho\partial\epsilon/\partial\rho)^2$	J_{thr} (MW/cm^2)
Hexane	1.14	25.1
Methanol	0.83	26.7
Water	0.76	52.13

We make the further assumption that there are clusters of ultramicroscopic gas bubbles with a radius $R_0 \sim 10^{-7}$ cm in the liquid and that these body forces act on the boundaries of these clusters, which collapse, forming a large bubble. This assumption can be made because (first) the cavitation is observed in very small volumes (in the case of the 5-cm cell, the volume in which the bubbles form is on the order of 10^{-3} ml, and that in the 20-μm cell is even smaller); i.e., only ultramicroscopic gas bubbles $\sim 10^{-7}$ cm in size could exist in such volumes (the probability for the presence of a larger ultramicroscopic gas bubble in such a volume is low because the concentration by volume of the gas bubbles decreases with increasing bubble size; Ref. 3). Such ultramicroscopic gas bubbles could not by themselves lead to cavitation, as we know,[3] but (more on this below) they could exist in the form of clusters as a result of a coagulation process. Second, evidence in favor of the hypothesis of a cluster mechanism for cavitation in this case comes from the fact that the cavitation bubbles manage to form over a time τ_p. This formation could not be the result of a diffusion of gas inside an individual bubble, but it could be explained in terms of an avalanche coalescence of clusters [indirect evidence for this possibility comes from the presence of liquid droplets within the cavitation bubbles (Fig. 2); they could be formed by shells of individual ultramicroscopic gas bubbles in a cluster].

The following estimates support the hypothesis of a cluster formation of ultramicroscopic gas bubbles in a liquid (these estimates are made for water). It can be shown that adsorption of surfactants in the shells of ultramicroscopic gas bubbles would cause the shell density to satisfy $\rho_1 > \rho$, where ρ is the density of the liquid. We then find that the condition $\rho_1/\rho = 8/7$ (which is plausible from the physical standpoint) corresponds to neutral buoyancy of the ultramicroscopic gas bubbles. The system consisting of the liquid and the ultramicroscopic gas bubbles tends toward a state with a lower surface energy; i.e., the ultramicroscopic gas bubbles should coalesce. The coalescence process has fast and slow steps. In the fast step, diffusive collisions of the ultramicroscopic gas bubbles result in their coalescence (coagulation) and in the formation of clusters of N ultramicroscopic gas bubbles. The cluster then either coalesces or floats up. The lifetime of a cluster, τ_c, must be such that the gas content in the cluster remains constant by virtue of a dissolution of air from the surface. It is not difficult to show that the concentration of the ultramicroscopic gas bubbles is $n_0 \sim 10^{14}$ cm^{-3}. Using the results of Ref. 4 for the time required for the formation of m clusters ($m = n_0/N$) in a unit volume, we find $\theta \sim 3 \times 10^{-3} [(n_0/m)]^{2/3}$. The value of m is found from the condition that the time over which it subsequently decreases by a factor of two due to a coagulation of the clusters themselves is on the order of τ_c; i.e., we have[4] $[\rho\nu/(mkT)] \sim \tau_c$, where ν is the kinematic viscosity of the liquid, T is its temperature, τ_c is determined by the time required for a cluster to float up to the surface, $\tau_c \sim l/\langle u^2 \rangle^{1/2}$, l is the height of the cell, and $\langle u^2 \rangle^{1/2}$ is the mean square velocity of the floating up (or of the sedimentation; this velocity would be determined by the deviation of ρ_1/ρ from the mean value corresponding to the condition for neutral buoyancy). It can be shown that under our experimental conditions we would have $\langle u^2 \rangle^{1/2} \sim 10^{-10} N^{1/6}$ cm/s. For $l = 1$ cm we have $m \sim 3 \times 10^3$ cm^{-3} and $N \sim 3 \times 10^{10}$. We thus find the estimates $\theta \sim 10^3$ s and $\tau_c \sim 10^8$ s $\simeq 3.5$ yr. In other words, the clusters exist for a long time. This prolonged existence is confirmed by our experiments: The clusters were reproduced over a period of several months without a change of liquid. We can test the estimate of m by counting the number of cavitation bubbles excited by a laser pulse in the 20-μm cell (see Fig. 1; we are allowing for the fact that the size of the laser spot is ~ 3 mm). We find $m \sim 10^3$ cm^{-3}. The estimate of m can also be tested independently if we know the cluster radius R and make use of the simple relation $m \sim n_0(R_0/R)^3$. Assuming that the radius of the bubble formed during the cavitation is $R^* \sim R$, and taking $R^* \sim 2 \times 10^{-4}$ cm (Fig. 2), we find $m \sim 10^3$ cm^{-3}.

This estimate of the cluster formation time θ was tested in an experiment based on the following idea: Since bubbles which float up are formed from clusters, the value of m decreases in the exposure region. In other words, it is possible to arrange a local outgassing. In the 5-cm cell, at a fixed intensity (above the threshold), cavitation is thus excited at the waist. This cavitation comes to a complete halt after a certain time which depends on the pulse repetition frequency. It reappears if the beam is moved to another part of the cell or after a time of 25–30 min, which corresponds to the estimate of θ. Finally, the fact that the coalescence manages to occur over a time on the order of τ_p can be attributed to the onset of an avalanche process with a time scale $R/v_a \sim R^*/v_a \sim 10^{-8}$ s (v_a is the sound velocity in air), i.e., $\sim \tau_p$. This "cluster mechanism" might also play a role in acoustic or hydrodynamic cavitation.

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