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Research on sound fields generated by laser-induced liquid breakdown

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The sound field excited by laser-induced liquid breakdown is studied through theory and experiment by using fundamental theories of acoustics. It is shown that there is a logarithmic linear relationship between the photoacoustic signals produced by a pulsed laser in liquid and the laser energy. Using the continuity of photoacoustic signals in liquids, a method is obtained to compute the breakdown threshold in liquids, when the distance between the observation point and the laser-induced breakdown area is much longer than cylinder length. The acoustic pressure amplitude is inversely proportional to the distance but proportional to the length of laser-induced breakdown. The acoustic pulse signal amplitude becomes largest in the direction perpendicular to the spreading direction of light. The acoustic pulse signal amplitude becomes smallest if parallel to the direction of light.

Keywords: photoacoustic effect, optical breakdown, radiated sound pressure, plasma column.

1. Introduction

In the study of liquid materials, the relationship between the micro- and macro-parameters has been the least well understood in condensed matter physics. Since the interaction of liquid molecules strongly depends on the specific variety of the liquid, people cannot fully understand how they move. Therefore, the study of acoustic parameters has great significance. It is important in molecular acoustics to explore liquids at the micro-information level and to explain the fluid nature of acoustics. Due to their non-invasive characteristics and large bandwidth, lasers are suitable for the measurement of the medium's sound parameters, and high measurement accuracy will help the study of the micro-structure of the material.

When a certain laser irradiates on objects, the laser and material will interact and generate sound waves in the material, thus producing the photoacoustic effect.

The main theories of the photoacoustic effect in liquids are the thermal expansion theory, vaporization theory and optical breakdown theory. When the absorption coefficient of the sample is small and the optical and laser energy are not too large, thermal expansion is the main mechanism; when the laser energy is more than the dielectric breakdown threshold, optical breakdown is the main mechanism. Through the mechanism of laser-induced breakdown in liquid, strong acoustic pulse waves can be obtained [1–3]. The conversion efficiency of optical breakdown produced by sound waves is as high as 30%. The photoacoustic effect in liquids can be used to monitor chemical processes and many other areas [4–7].

In this paper, we have established a new model which distributes evenly many spherical pulse sources. Such a series of spheres of plasma superpose to form a plasma cylinder. The source radius of the sphere is equal to the radius of the plasma column. Through the establishment of the model, using the theory of the photoacoustic effect in liquids, we investigate the intensity of photoacoustic signal generated by optical breakdown in liquids with laser energy changing, and discuss a new method for finding the optical breakdown threshold of a liquid. Also, the model is used to obtain the excitation of acoustic pressure by optical breakdown in liquids. The results of experiment fit the theory well.

2. Principle and model

The plasma column is modeled as shown in Fig. 1. Optical breakdown in the liquid will produce a plasma cavity. This plasma chamber is considered as a homogenous cylinder. In the body of the cylinder, ignoring the behavior of individual particles, we only consider the fluid element of the movement. The movement produces sound waves, so classical theory can be used to deal with it. We treat the movement of the plasma column as adiabatic. Because cylinder length $L \gg$ the cylinder radius R, we can consider a one-dimensional situation. Based on this, a number of acoustic parameters of the plasma column can be obtained, and thus the optical breakdown of liquid in the sound field can be discussed.

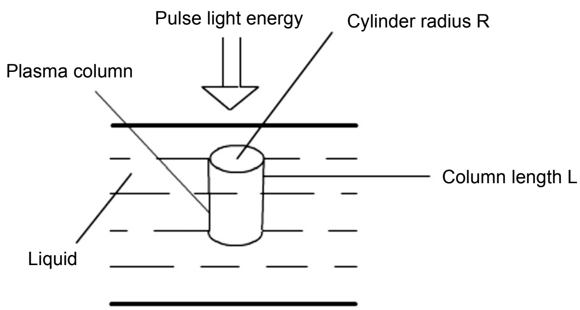

Fig. 1. Plasma column model.

2.1. Optical breakdown threshold of liquid [8]

From the plasma cylinder model, when the laser energy is higher than the threshold, the medium is completely ionized. The relationship between the laser light energy and the sound energy is

$$\Delta E = \eta q \tag{1}$$

where ΔE stands for the sound field energy, η the photoacoustic liquid conversion efficiency, which for the same material is a constant, and q for the laser energy. The acoustic energy can also be expressed as:

$$\Delta E = \frac{V_0 p^2}{\rho_0 c_0^2} \tag{2}$$

where V_0 stands for the initial volume of the plasma cavity, ρ_0, c_0 are the density and sound velocity of the plasma, respectively, p is the sound pressure. Substituting formula (2) into formula (1) yields:

$$q = \frac{V_0 p^2}{\rho_0 c_0^2 \eta} \tag{3}$$

Taking the base 10 logarithm of formula (3) on both sides,

$$\lg q = 2\lg p + \lg V_0 - \lg \eta - 2\lg c_0 - \lg \rho_0 \tag{4}$$

Let $\lg V_0 - \lg \eta - 2\lg c_0 - \lg \rho_0 = k_1$. For the same substances, k_1 is constant. Then, formula (4) becomes:

$$\lg q = 2\lg p + k_1 \tag{5}$$

The above formula shows that when optical breakdown occurs in liquids, the laser energy and intensity of the sound field have a linear logarithmic relationship. Actually formula (5) is a line segment because laser energy is limited and has a threshold, namely the laser intensity is greater than the threshold intensity. In the case of the thermal expansion mechanism, the relationship between the laser intensity and the intensity of the sound field is [9]

$$p = kI \tag{6}$$

where k is a constant for specific material, and I stands for the laser intensity. Taking the base 10 logarithm of formula (6) on both sides,

$$\lg q = \lg p + k_2 \tag{7}$$

where k_2 is a constant. Comparing formula (5) with formula (7), we can see that the relationship is very similar, both of them show logarithmic linear relations, only the slope of the straight line is different. This shows that the photoacoustic signal intensity increases with the enhancement of the incident laser power. When the laser power is increased further, reaching or exceeding the breakdown threshold of the sample, the signal strength dramatically increases. Laser-generated sound in a liquid can be seen as a continuous process, which is composed of thermal expansion and optical breakdown. Therefore, the two straight lines in formulas (5) and (7) must have a point of intersection; this intersection point is the threshold point, and from this the optical breakdown threshold can be obtained.

2.2. The radiation field of a plasma column [10]

When optical breakdown occurs in liquid, it can produce a plasma cavity which is full of plasma. Pulsed laser beams through a lens perpendicular to the liquid medium can form a series of spherical cavities containing the plasma in focused areas. Each cavity can be seen as a spherical pulse source. These pulse sources form a plasma column by linear superposition of the spheres, and the radius of the spherical pulse source is the same as the cylinder radius. The cylinder length $L \gg$ the cylinder radius R, as shown in Fig. 2. Suppose the length of the line segment is L, with n uniformly distributed identical small pulsating sphere sources, then

$$n = L/2r_0 \tag{8}$$

For a single source sphere, we can use the special wave equation,

$$\frac{\partial^2 p}{\partial r^2} + \frac{\partial p}{\partial r}\frac{\partial \ln S}{\partial r} = \frac{1}{C_0^2}\frac{\partial^2 p}{\partial t^2} \tag{9}$$

The characteristic solution of formula (9) is

$$p = \frac{A}{r}\exp\left[j(\omega t - kx)\right] \tag{10}$$

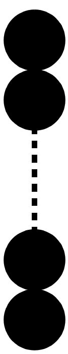

Fig. 2. Simplified plasma column model.

where $|A/r| = p_A$ stands for the acoustic pressure amplitude, r is the distance from the observation point to a small pulse source, $k = \omega/c$ is the wave number. The radial speed of the whole particle,

$$v_r = -\frac{1}{j\omega\rho_0}\frac{\partial\rho}{\partial r} = \frac{A}{r\rho_0 c_0}\left|1 + \frac{1}{jkr}\right| \bullet \exp\Big[j(\omega t - kr)\Big] \tag{11}$$

where ρ_0 and c_0 stand for the density and sound velocity of the liquid, respectively. The speed is continuous on the plasma source surface, so there exists the boundary condition:

$$(v_r)_{r = r_0} = u_0 \tag{12}$$

where u is the vibration velocity of the spherical source surface, $u = u_A \exp[j(\omega t - kr_0)]$, u_A is the amplitude of vibration velocity. Substituting formula (11) into formula (12) yields:

$$A = \frac{\rho_0 c_0 \kappa r_0^2}{1 + (kr_0)^2} \bullet u_A(kr_0 + j) = |A| e^{j\alpha} \tag{13}$$

where $A = \dfrac{\rho_0 c_0 \kappa r_0^2}{1 + (kr_0)^2} \bullet u_A$, $\alpha = \tan^{-1}\left(\dfrac{1}{kr_0}\right)$. Substituting A into formula (10), one can get the radiated sound pressure of a single pulsating spherical source,

$$p = \frac{|A|}{r}\exp\Big[j(\omega t - kr + a)\Big] \tag{14}$$

From formula (14), we can see that the sound pressure amplitude is determined by the value of $|A|$, where the distance from the pulse sphere is r. From formula (13), we can see that the value of $|A|$ is not only related to the vibration velocity u_A of the source sphere, but also to the frequency of sound waves, the radius of the spherical source, and so on. If the radius of the sphere is small or the sound frequency is low, then $kr_0 \ll 1$. A pulse source which meets this condition is called a spherical-point source. For a radiated acoustic field (as shown in Fig. 3), we can get:

$$p = \sum_{i = 1}^{n} \frac{|A|}{r_i}\exp\Big[j(\omega t - kr_i)\Big] \tag{15}$$

For the far-field, when the sound waves of every small spherical source spread to the observation point P, the amplitudes of the sound waves are similar. So, in formula (15), for the amplitude part, r_i can be replaced by r, which is the distance

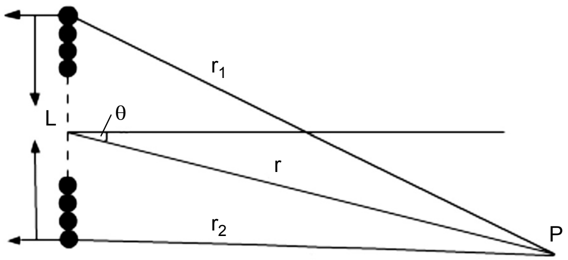

Fig. 3. Radial acoustic field.

from approximate center line to the observation point. Considering phase difference, the following can be obtained from Fig. 3,

$$r_2 = r_1 + 2r_0 \sin\theta$$

$$r_3 = r_1 + 4r_0 \sin\theta$$

$$\vdots$$

$$r_n = r_1 + 2(n-1)r_0 \sin\theta$$

Let $h = r_0 \sin\theta$. Formula (15) can then be written,

$$(p_A)_{\theta=0} = \frac{A}{r}\exp\left[j(\omega t - kr_1)\right]\left[1 + \exp(-2jkh) + \ldots + \exp(-j4k(n-1)h)\right] =$$

$$= \frac{A}{r}\exp\left[j(\omega t - kr)\right]\frac{\sin(knh)}{\sin(kh)}$$

It can be seen that since the acoustic radiation from each sphere reaches the observation point in different directions, there will be interference which makes the directions of the sound field varied, so directivity emerges. When $\theta = 0$,

$$(p_A)_{\theta=0} = n\frac{A}{r}\exp\left[j(\omega t - kr)\right] \tag{16}$$

The directional function is then:

$$D(\theta) = \frac{(p_A)_\theta}{(p_A)_{\theta=0}} = \left|\frac{\sin(knh)}{n\sin(kh)}\right| \tag{17}$$

3. Experimental results and analysis

3.1. Experimental set-up

The experimental set-up is shown in Fig. 4. The light source was a YAG adjuster (YG581, Quantel Corporation), the output of the laser wavelength was 1.06 μm, pulse width 8–10 nm, repetition rate was 10 Hz and the output laser spot diameter was 6 mm.

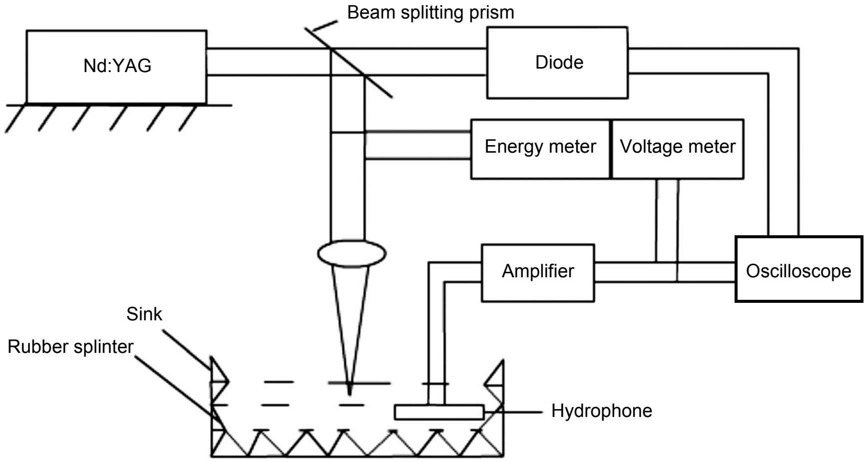

Fig. 4. Schematic of experimental set-up.

The YAG output infrared beam was divided into two beams through a mirror: one went through the mirror and then a synchronization signal generator connected with the oscilloscope, as a synchronization signal. Another was reflected at 45°, with a Model-DG measurable reflection to measure energy value of the reflected beam. Then the reflected beam was focused by a focusing lens with a focal length of 292 mm, and the focal point below the liquid level. The laser spot diameter was about 0.8 mm at the focus spot (the spot is formed after laser go through the focusing lens). The tank (600 mm×250 mm×350 mm) was plexiglass internally muffled by a rubber wedge. A hydrophone was used as a photoacoustic signal detector. The distance between the focus areas was measured by specialized equipment. An HFM-1 charge amplifier was used to measure the maximum voltage of the photoacoustic signal after sending the enlarged photoacoustic signal received by the hydrophone to a DHF-4 charge amplifier. The maximum frequency of the charge amplifier was set to 100 kHz and its minimum frequency was set to 0.3 Hz. In order to monitor and observe the size and waveform of the photoacoustic signal, an SBM-10 multi-purpose oscilloscope was joined to both ends of the pulse millivoltmeter. The beam splitter and focusing lens were fixed to the magnetic platform by the light bench base.

3.2. Results and analysis

3.2.1. Optical breakdown threshold of liquid

The laser energy was changed, while the distance between the hydrophone and the focal point of the laser was kept constant and the magnification of the charge amplifier was regulated. At each magnification, the photoacoustic signal maximum voltage was found by the pulsing millivoltmeter, with five figures recorded at each point and an average taken. Then, according to the hydrophone sensitivity and electric charge magnification, the acoustic pressure figures were obtained. Experimental

Table 1. Intensity of acoustic signals, when the liquid is tap water, r = 100 mm.

Laser energy [mJ]	35.6	30.8	27.5	22.5	16.8	14.5	11.5	10.0
Voltage signal [mV]	250.8	204.2	153.0	121.0	60.2	51.8	14.6	13.9
Sound pressure signals [Pa]	25832.4	21032.6	15759.0	12463.0	6200.6	5335.4	1503.8	1431.7

Table 2. Intensity of acoustic signals, when the liquid is saline water, r = 100 mm.

Laser energy [mJ]	36.2	28.4	22.4	17.6	6.2	5.1
Voltage signal [mV]	285.2	271.0	250.8	204.2	81.4	60.2
Sound pressure signals [Pa]	29072.6	27963.0	25872.4	20985.6	8427.2	6543.6

samples came from tap water and normal saline water. The results are listed in Tabs. 1 and 2.

Putting the data from Tabs. 1 and 2 into formulas (5) and (7) gives:

– Water: $k_1 \approx -9.3596$, $k_2 \approx -5.6285$;

– Saline: $k_1 \approx -10.4078$, $k_2 \approx -6.2931$.

Putting k_1 and k_2 into formulas (5) and (7), we find that the relationships between the laser energy and intensity of acoustic signals in tap water and saline water, are as follows:

– Water: $\lg q = 2\lg p - 9.3596$ for $q \geq$ threshold and $\lg q = \lg p - 5.6285$ for $q <$ threshold;

– Saline: $\lg q = 2\lg p - 10.4078$ for $q \geq$ threshold and $\lg q = \lg p - 6.2931$ for $q <$ threshold

By solving the equations or plotting a graph we find that optical breakdown threshold of a single pulse of laser energy is 11.7 mJ in tap water and 6.6 mJ in normal saline.

3.2.2. Optical breakdown of liquid sound field excitation

In experimental measurements, the distance between the hydrophone and the laser focal point was changed while keeping the laser energy constant and placing the hydrophone perpendicular to the plasma cylinder. The maximum voltage of the photoacoustic signal was then measured by the pulsing millivoltmeter, with five values measured at each point and an average taken. The acoustic pressure values were then calculated from hydrophone sensitivity and charge magnification. Experimental samples were tap water and normal saline water. The results are listed in Tabs. 3 and 4 and Fig. 5.

Table 3. Intensity of photoacoustic signals in water (40 mJ).

Distance [mm]	100	120	150	170	190	210	230	250
Voltage signal [mV]	318.6	285.3	260.3	231.3	202	161.2	140.2	131.1
Sound pressure signals [Pa]	32 815.8	29 437.4	26 810.9	23 823.9	20 888.4	16 603.6	14 440.6	13 503.

T a b l e 4. Intensity of photoacoustic signals in physiological saline (36 mJ).

Distance [mm]	50	70	100	120	140	160	180	200
Voltage signal [mV]	260.8	250.0	215.0	115.2	90.2	79.8	69.6	57.8
Sound pressure signals [Pa]	27604.0	25750.0	22145.0	11865.6	8219.4	7168.8	5953.4	

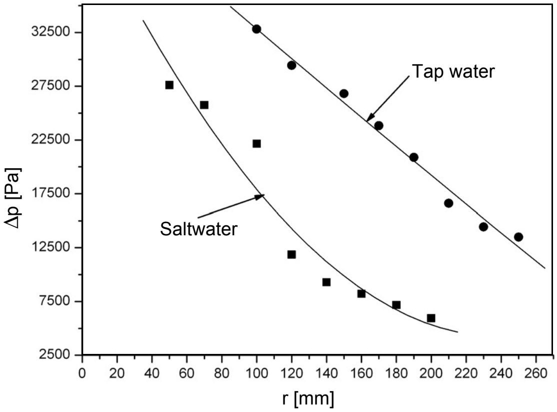

Fig. 5. Acoustic pressure amplitude versus distance.

Figure 5 shows the relationship between the sound pressure amplitude and the distance. It can be seen that acoustic pressure amplitude and distance have an inverse relationship.

When the hydrophone is far from the plasma column, the distance between the photoacoustic signal and the sound pressure is inversely proportional, that is,

$$P_{\max} = \frac{K_1}{r} \tag{18}$$

Substituting the experimental data into formula (18), and taking the average, we have: $K_1 \approx 3.4\times10^3$ Pa·m (tap water), $K_2 \approx 1.3\times10^3$ Pa·m (saline).

Substituting corresponding data into formula (16), we get

$$A = \frac{K_1}{n} \tag{19}$$

where n is the number of spherical pulse sources. From experimental observation $r_0 = 1$ mm, so if the number of source spheres is certain, for a specific material where the optical breakdown length is L, A can be obtained. The sound pressure P can then be obtained.

Table 5 lists the experimental data far the relationship between optical breakdown and acoustic signal intensity.

T a b l e 5. Relationship between optical breakdown length and intensity of photoacoustic signals.

Optical breakdown length [cm]	Water signal Sound pressure [Pa]	Normal saline signal Sound pressure [Pa]
1	10 467.5	9 786.7
2	20 163.9	21 069.4
3	32 815.8	29 435.2

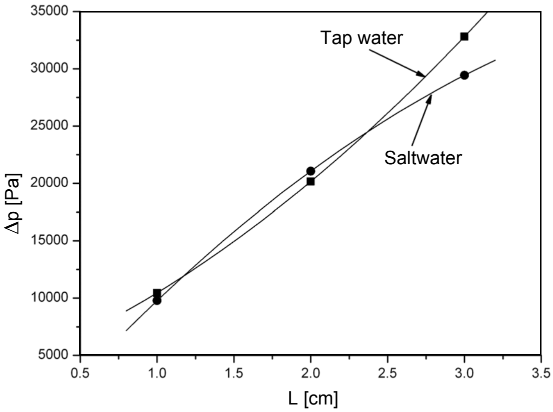

Fig. 6. Acoustic pressure amplitude versus the length of laser-induced breakdown when $r = 100$ mm.

Figure 6 shows the relationship between optical breakdown and acoustic signal intensity. From the chart, we can see that there is a proportional relationship between optical breakdown and acoustic signal intensity. The results fit formula (19) well.

4. Conclusions

The overall conclusions are: 1) There is a threshold value in the optical breakdown region, which can be obtained from the relationships between laser energy and signal intensity. The size of the optical breakdown threshold is related to the nature of the liquid. When there are some charged particles, air bubbles and impurities in liquid, the threshold will be much lower; 2) When the optical breakdown occurs in the liquid, the breakdown region can be treated as a cylinder containing a plasma cylinder of length L and diameter d, with the cylinder seen as a number of spherical pulse sources by linear superposition, the diameter of the spherical pulse sources is equal to the diameter of the cylinder; 3) When the distance from the observation point to the optical breakdown zone is r and the length of cylinder is L, the sound pressure and the directional characteristics of acoustic field by theoretical calculation are basically consistent with experiment. In the direction perpendicular to the light propagation, the acoustic pressure amplitude is inversely proportional to the distance r but proportional to the length L of laser-induced breakdown; 4) In the perpendicular direction to the spread of light, the sound pressure amplitude of acoustic pulse signal reaches maximum. The sound pressure amplitude of acoustic pulse signals reaches minimum when following the direction of the light.

Optical breakdown occurs when a pulsed laser excites a liquid to its threshold intensity, generating plasma which produces acoustic waves by absorbing light energy. It is reasonable to deal with the macroscopic movement of plasma using the classical theory after ignoring the macroscopic effects of a single particle.

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