Influence of volume dynamics of aerosol particles exposed to laser radiation on temporal characteristics of the acoustic response

N.N. Bochkarev, A.M. Kabanov, and V.A. Pogodaev

Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences, Tomsk

Received April 20, 2000

We present results of experimental investigations of time behavior of an acoustic pulse formed at various regimes of thermal interaction of high-power pulses of CO2-laser radiation and small volumes of an absorbing liquid. A connection between the shape of the generated pressure pulse and regimes of laser radiation interaction with a substance is established. The model of pulsing sphere is applied to interpretation of the obtained dependence of characteristics under study on the laser energy density. The comparison of computed and experimental data is carried out.

Interaction of high-power IR laser radiation with atmospheric aerosol is accompanied by a large number of nonlinear optical effects causing transformations of the propagation channel properties. In particular, such processes as evaporation, explosion, and optical breakdown are known as the main nonlinear optical effects associated with the liquid-droplet aerosol. They can be observed in a wide range of radiation energy and significantly differ by their influence on the characteristics of medium and laser beams. The on-line contactless monitoring of interaction of high-power laser radiation with an absorbing substance is needed in predicting the efficiency of laser radiation transfer through the atmosphere¹ as well as in some other applications.²

From the standpoint of creating a physical basis for such diagnostic methods, the process of acoustic signal generation at phase transitions in small volumes of some substance is rather attractive. 1 In contrast to thermooptical regime of the acoustic response formation, in this case the acoustic signal is reliably recorded in the open air both under laboratory and field conditions without any acoustic insulation of the object irradiated and a microphone.

Investigation into the generation of acoustic signals emitted by aerosol particles and changing their properties under the action of high-power optical fields, allowed us to study the processes of explosive boiling up and destruction of the particles. $^{3-5}$ It was shown that in transition from regular surface evaporation of particles to their volume explosive boiling up and destruction, a change of the mode of the excess pressure pulse formation takes place resulting in a change of the dependence of acoustic signal amplitude on the energy density of incident radiation. Earlier⁶ it was established that aerosol microstructure can influence amplitude characteristics of the acoustic response and the energy thresholds for destruction of aerosol particles by laser

radiation as well as the dependence of limiting values of the radiant energy density on the droplet sizes and rates of their laser-induced heating were determined. ⁷

It was noticed, in the experiments, 8 that the shape of the acoustic pulse generated depends on the type of laser radiation interaction with the particulate matter, taking place in each particular case. It was established, in particular, that the delay time t_d of the acoustic pulse recoding shortens as the energy density Eof incident radiation increases, starting from the energy threshold of the explosive boiling up $(E \sim 2 \text{ J/cm}^2).$

One of the main parameters of the explosive boiling up affecting the magnitude $t_{\rm d}$ is the so-called explosion time $t_{\rm e}$, i.e., the time interval between the onset of the radiation action on a particle and the moment the vapor emission starts from its surface. The explosion time includes the time needed for the particle substance to warm up to the temperature of explosive boiling up, the time during which vapor bubbles must appear in the overheated liquid, and the time of the bubbles growth and floating up to the surface, what results in destruction of the whole particle, in the case of small homogeneously absorbing droplets $\alpha_n r_0 < 1$, or only in its fragmentation in the case of large inhomogeneously absorbing droplets $\alpha_n r_0 > 1$. Here α_n is the volume absorption coefficient of the particle substance at the wavelength of incident radiation; r_0 is the particle radius. In both of these cases, the start of vapor emission is the start of formation of the pressure pulse to be recorded. In Ref. 9 the time of explosion was studied optically by following up variations in the aerosol layer transparency and the light scattering signal. A decrease of $t_{\rm e}$ from 3 to 1 μs was noticed at the increase of E from 2 to 10 J/cm². These results agree with the experimental data, obtained by other authors using different independent methods, 10 and with the theoretical estimates, 9 as well.

Optics

The decrease of the delay time $t_{\rm d}$ recorded in Ref. 8 at increasing the E value, within the same limits as in Ref. 9, is about an order of magnitude higher than t_e that cannot be explained by variations in the explosion time. There are two factors more that can cause such a decrease. The one is a significant growth in the excess pressure in the region of laser radiation interaction with aerosol and, as a consequence, enhancement of nonlinear acoustic effects such as distortions of the acoustic wave profile and the increase in shock wave velocity at the early stage of its appearance. The second one is an increase in the size of a region where the acoustic pulse generation occurs because of flying apart fragments of the exploded aerosol particles after their exposure to laser radiation. The first cause may be ignored, in accordance with the measurement conditions, because nonlinear effects in the generated acoustic signals are negligibly small. The corresponding estimate shows that the parameter N(the Khokhlov number¹¹) becomes

$$N = (\gamma + 1)(\rho_a/\rho)(\pi a/\lambda)^2 \ll 1, \tag{1}$$

where $\gamma = 1.4$ is the ratio of the medium heat capacities at constant pressure and volume; λ is the measured wavelength of the sound wave generated; a is the efficient size of the region the acoustic pulse is generated in; ρ is the air density, $\rho_a \sim \gamma \, P_a/c^2$ is the density perturbation and P_a is the corresponding acoustic pressure within the volume of the acoustic pulse generation; and c is the speed of sound in the air. It is obvious that the acoustic wave generated may be treated as a linear one even at early stages of its propagation from the interaction volume.

Thus, we may consider the change in the size of the region the acoustic pulse is formed in as a possible cause of t_d decrease with the growth of the radiant energy density.

The goal of this work was to obtain quantitative data on variations of the principal characteristics of the recorded acoustic response generated due to evaporative and explosive interactions between laser radiation and an absorbing substance of a small volume and to identify the causes of such variations.

The radiation of pulsed TEA CO₂ laser at 10.6 µm wavelength was used as the acting one. It was directed through a focusing lens with 0.12 m focal length to the interaction region, where the beam's cross section area was 4 mm². The radiation pulse shape was typical for CO₂ laser emission at the atmospheric pressure. The affecting pulse energy was 3 J at its total length of 3.10^{-6} s. The duration of the leading edge of the pulse was 3.10^{-8} s. The radiant energy was measured with an IMO-2 calorimeter. A water stream of 250-µm diameter was used as the object to be irradiated. Its volume absorption coefficient α_n was equal to 800 cm⁻¹ at the wavelength of incident radiation used. The water cylinder of 250-µm diameter and 2 mm long served as a source of acoustic perturbation in the medium in this geometry of the experiment.

To record the acoustic signal, we have used a 7-mm-diameter wideband capacitor microphone and a precision pulsed noise meter with the bandwidth of 10⁵ Hz. The maximum admissible amplitude of the peak pressures recorded was 172 dB at the absolute measurement error of 0.5 dB. The acoustic perturbation was measured from the distance of 7.2 mm in the direction perpendicular to the plane formed by the axes of the acting optical beam and the water cylinder to be irradiated. The geometry of the experiment is shown in Fig. 1. There were two reasons for choosing this geometry. First, we aimed at providing a reliable recording of the photo-acoustic pressure pulse against the background of laboratory noises (no less than 68 dB). Second, we had to meet the requirements to the error in determination of both the time behavior of the pressure in the acoustic wave and the time delay t_d (the time between the onset of the laser pulse action and appearance of the acoustic signal). To provide for coincidence of the acting laser beam with the object to be irradiated, a collimated low-power beam of He-Ne laser was introduced into the interaction region to facilitate the proper alignment and as a reference one. The optical signal at the wavelength of sensing radiation (0.63 µm) served as an additional check of temporal characteristics of the processes under study. The recording of acoustic and optical channels was performed with a storage oscilloscopes.

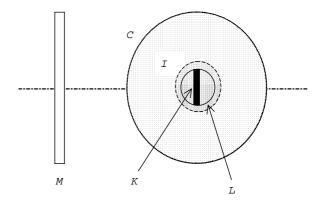


Fig. 1. The geometry of experiments: M is the microphone, Kis the water cylinder, L is the laser beam, I is the effective region of sound emission at evaporation, C is the effective region of the sound emission due to explosion.

In studying parameters of the acoustic response at different regimes of thermal interaction between the laser radiation and the absorbing substance, the following characteristics were recorded: the amplitudes and lengths of positive and negative phases of the acoustic pulse P_+ , P_- , t_+ , and t_- ; the duration of the positive phase leading edge $t_{\rm ph}$; the delay time of the acoustic signal t_d determined by the distance from the volume of the pressure pulse formation to microphone membrane and sound speed in the medium.

Relative variability of the temporal parameters of the acoustic response versus laser energy density E is presented in Fig. 2. The values of temporal parameters of pressure pulse at $E=0.14~\mathrm{J/cm^2}$ were used as the reference, t_0 , value. At $E<0.14~\mathrm{J/cm^2}$ the recording instruments failed to reliably record the acoustic pulse parameters under study.

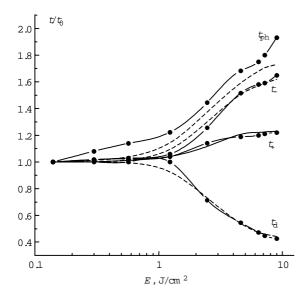


Fig. 2. Relative variability of the acoustic response temporal parameters versus laser radiation energy density: the experiment is presented by solid curves, the theory by dashed curves.

As follows from Fig. 2 the thermal and evaporation mechanisms of acoustic signal generation, do not change the magnitude of t_d , which is equal to the time of pressure pulse passage at the speed of sound from the source to the receiver. Under the regime of explosive boiling up of the object, the magnitude of t_d sharply decreases, what points to a change of the mechanism of the acoustic signal generation. A change in the interaction mode causes a significant change in the pulse shape. This results in a change of the ratio between full duration of the compression phase and its leading edge. Since at excitation of the interaction volume by a short laser pulse the shape of the leading edge must follow the distribution of thermooptical actions, 12 a significant decrease in t_d and the increase in $t_{\rm ph}$ evidence of the fact that the size of the sound generation region, filled with the fragments of the droplet explosion, essentially increases at $E > 2 \text{ J/cm}^2$.

The experimentally obtained dependence of the peak acoustic pressure P_+ in the received signal on the radiation energy density E is shown in Fig. 3 and it is close to those obtained earlier.⁴⁻⁶

A change of the behavior in the region $E \sim 2 \text{ J/cm}^2$ is indicative of reaching the threshold of the explosive boiling up. Figure 3 depicts the corresponding data for both monodisperse and

polydisperse aerosols, as well as for individual water drop⁹ scaled in accordance with the geometry of the experiment (see Fig. 1). It is seen that at $E < 1 \, \mathrm{J/cm^2}$ the measured dependence $P_+(E)$ is close to that measured for an individual water drop $(P \sim E^4)$ and at $E > 1 \, \mathrm{J/cm^2}$ the quantity $(P \sim E^4)$ well agrees with the dependence obtained for a monodisperse aerosol. Based on data presented in Fig. 3, we may conclude that in the experiments with the liquid volume under study at $0.14 \, \mathrm{J/cm^2} < E < 2 \, \mathrm{J/cm^2}$, it is the evaporative mechanism of sound generation that is steadily observed.

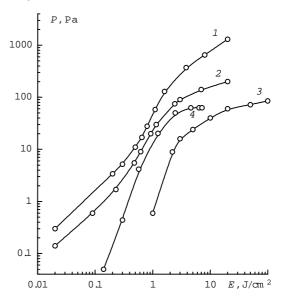


Fig. 3. Dependence of the acoustic pressure on the laser radiation energy density at optical-acoustical effect in liquid aerosols: (1) polydisperse aerosol; (2) monodisperse aerosol of 2.7 μ m radius; (3) water drop of 15 μ m radius; (4) water cylinder.

The measured dependence of peak acoustic pressure at the rarefaction phase P_- turned out to be similar to that presented in Fig. 3 for all values of the acting energy density. The ratio between P_+ and P_- remains constant in all interaction modes, therefore it cannot serve as the information source for determining these parameters.

Based on supposition on variability of the acoustic pulse generation region, we have numerically simulated the process of acoustic response formation. In case of a finite-length pulse of a laser radiation, the recorded acoustic signal is a convolution of the acoustic signal itself with the computed transient function of the spatial conditions for recording the signal. The transient function of the recording instrument itself was ignored because of its negligible effect within the frequency range under study. Based on data presented in Fig. 1 and assuming that the acoustic signal generation region is a sphere, the diffraction distortions of the recorded acoustic response were taken into account based on solution of the problem on the

spherical pulsed emission (the region of acoustic signal onto the piston-type diaphragm¹³ generation) (microphone membrane). Surface oscillations of the sphere were approximated by the following function:

$$f(t) = (nt/t_a)^n \exp(-nt/t_a) \sin(\pi t/t_a),$$
 (2)

where t_a is the acoustic pulse length, n=3-3.8 is the empiric constant.

Since the region of acoustic pulse generation significantly differs bv its thermophysical characteristics from the ambient air, we have supposed a possibility of identifying this region as a sound emitter with the well known model of pulsing sphere (emitter of zero order) of radius a oscillating with the frequency ω and producing a sound pressure ¹³

$$P = \xi \rho c(a/x)^2 (ikx/(1+iX)) \exp i(X + \omega t - kx),$$
 (3)

where X = ka, $k = 2\pi/\lambda$; $\omega = kc$; ξ is the velocity of the sphere's surface; x is the coordinate. The expression (3) shows, in particular, that the efficiency of energy transfer by a sphere at ka < 1 tends to pure imaginary quantity and significantly decreases. Consequently, for a pulse of the type (2), the efficiently emitted wavelength is connected with the size of the sphere through the relation $\lambda < \pi a$.

In the frameworks of the assumptions presented, we have numerically simulated the time behavior of the acoustic pressure in the wave generated under different modes of laser radiation interaction with an absorbing substance. The calculated results of simulation are shown in Fig. 2 by dotted curves.

The comparison of calculated and experimental data has led us to the following conclusions. In the evaporative regime, the time delay corresponds to the acoustic wave front movement from the excitation region to the receiver with the speed of sound; and the shape of the compression pulse front corresponds to the region of the acoustic pulse generation, the efficient size of which is a = 1.3 mm. The corresponding condition, $\lambda < \pi a$, for optimal emission 13 by a sphere gives a close value a = 1.5 mm.

Once the irradiated object has blown up, its condensed fragments scatter at various distances and fill up the region of the radius $a \sim 3.4$ mm. The duration of the leading edge increases up to $7 \mu s$, what corresponds to a = 3 mm under conditions of optimal emission by pulsing spheres. Noticeable difference between the calculated and experimental values for the leading edge duration may be due to nonuniform filling up of the sound generation region with the products of explosion.

Thus, the transition from regular evaporation to explosive blowing up is characterized by a sharp increase in the size of the region the acoustic response is generated in. This can be explained by the scatter of the explosion products at a supersonic speed and filling with them the effective volume, as shown in Fig. 1.

The dependence of the pressure pulse amplitude on the energy density of laser emission in the regime of regular evaporation is close to the quadratic one, as has been noted by the authors in their earlier experiments with water aerosols of different microstructure.

Based on the model experiments conducted, it may be deduced that the shape of acoustic signal generated due to thermal interaction of laser radiation with an absorbing substance of small volume is determined by the interaction mode and significantly depends on it. The obtained quantitative data on amplitude and time behavior of the acoustic pressure allowed the identification of the mode of interaction between the laser radiation and the absorbing substance.

References

1. N.N. Bochkarev, Yu.E. Geints, A.A. Zemlyanov, A.M. Kabanov, and V.A. Pogodaev, Atmos. Oceanic Opt. 11, No. 7, 602-608 (1998).

2. S.V. Mel'chenko, E.D. Mel'chenko, M.M. Makogon, et al., Kvant. Elektron. 24, No. 10, 952-954 (1997).

3. A.A. Zemlyanov, Yu.E. Geints, A.M. Kabanov, R.L. Armstrong, Appl. Opt. 35, No. 30, 6062–6068 (1996).

4. N.N. Bochkarev, A.A. Zemlyanov, and A.M. Kabanov, "Acoustic response from explosive boiling up of aerosol particles exposed to pulsed laser radiation", VINITI, No. 28-89-V93, November 29, 1993, Tomsk (1993).

5. R.L. Armstrong, A.A. Zemlyanov, and A.M. Kabanov, Atmos. Oceanic Opt. 7, No. 9, 668-670 (1994).

6. N.N. Bochkarev, Yu.E. Geints, A.A. Zemlyanov, A.M. Kabanov, Opt. Atm. 1, No. 10, 111-112 (1988).

7. A.A. Zemlyanov and A.M. Kabanov, in: Proceedings of II International Symposium on Atmospheric and Oceanic Optics (Tomsk, 1995) pp. 171-172.

8. N.N. Bochkarev, A.M. Kabanov, and V.A. Pogodaev, Atmos. Oceanic Opt. 7, No. 9, 666-667 (1994).

9. Yu.E. Geints, A.A. Zemlyanov, V.E. Zuev, A.M. Kabanov, and V.A. Pogodaev, Nonlinear Optics of Atmospheric Aerosol, (Publishing House of SB RAS, Novosibirsk, 1999), 260 pp.

10. B.S. Park, A. Biswas, P. Armstrong, et al., Opt. Lett. 15, No. 4, 206-208 (1990).

11. N.S. Bakhvalov, Ya.M. Zhileikin, and E.L. Zabolotskaya, Nonlinear Theory of Sound Beams (Nauka, Moscow, 1982),

12. B.E. Gusev, and A.A. Karabutov, Laser Optoacoustics (Nauka, Moscow, 1991), 304 pp.

13. T. Hayasaka, *Electroacoustics* [Russian translation] (Mir, Moscow, 1982), 246 pp.