Experimental investigation into interaction between femtosecond laser pulses and aerosol

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The interaction of femtosecond laser pulses with various-composition liquid droplet aerosol, as well as with individual particles has been investigated experimentally. Acoustic and optical methods were applied to study the energy decay of femtosecond pulses traveling through the aerosol layer and the energy losses of filaments formed at different focusing. It was found experimentally that the nonlinear interaction with atmospheric aerosol does not affect significantly the energy characteristics of radiation.

Nonlinear optical effects arising upon propagation of ultrashort laser pulses in aerosol, as well as propagation of femtosecond laser pulses in the case of realization of filamentation in gas and aerosol atmospheres, are of great importance for studying laser energy transportation through the atmosphere and for atmospheric sensing.

We have carried out the experimental investigations of interaction of laser pulses of femto- and nanosecond duration with model aerosol media and individual particles. We used a Ti:Sa laser emitting at $\lambda = 0.8$ μm with the pulse duration $t_p = 80$ fs and 9 ns and the pulse energy < 17 mJ. The full width at half-maximum (FWHM) of the spectrum of the femto- and nanosecond pulses was ~ 25 nm. The intensity distribution over the beam cross section was close to Gaussian; the beam diameter at the $0.135I_{max}$ level was 8 mm. The aerosol medium with particles of the radius $r = 2.5$ μm had the length of 1.3 cm and the particle number concentration $N < 10^7$ cm^{-3}.

To model the aerosol medium, we dispersed pure water and water including silver nanoparticles. The nanoparticles were used to increase the absorbing properties of aerosol in order to estimate the difference in the energy loss of the femto- and nanosecond pulses traversing through the aerosol medium. The energy loss was measured using two measurement channels: optical and acoustic. The optical channel was used to measure the radiation extinction in the aerosol medium. The measurements of the acoustic response allowed the estimation of the radiation energy lost for dissipation into the thermal energy upon its absorption by the particulate matter.

The acoustic part of the setup included two measurement channels, calibrated against the sound pressure, with the linear frequency range 2–100 kHz. The optical and acoustic signals were measured by Tetronix and Infinium digital oscilloscopes. Filaments were studied using a focusing mirror with the focal length f equal to 120 or 86.5 cm. When studying the interaction between non-focused laser pulses and the aerosol medium, the focusing mirror was replaced by a beam-turning plate. The schematic layout of the experimental setup is shown in Fig. 1.

The aerosol layer transmittance was measured at two wavelengths: $\lambda = 0.8$ μm of the pumping high-power laser and $\lambda = 0.63$ μm of the probing cw He–Ne laser. As is well known, nonlinear effects do not take place in the case of the probing low-power radiation, so it was used to control the particle number concentration N in the aerosol medium at each shot of the Ti:Sa laser.

The medium transmittance for femto- and nanosecond pulses was calculated from the measurements of the pulse transmitted by the aerosol layer and the reference signal, calibrated against the measurements of the pulse energy with IMO-2M for every measurement series.

The measurement results are shown in Fig. 2. It is seen that for both wavelengths ($\lambda = 0.63$ μm and 0.8 μm) the transmittance obeys the Bouguer law, although for the latter the experimental values lie somewhat lower than the calculated curve, which, possibly, corresponds to increase of extinction due to effects of multiphoton absorption in aerosol.

In the case of realization of nonlinear optical effects upon interaction of radiation with the matter, the measurement of the acoustic response allowed us to monitor, though indirectly, the radiation energy, in particular, in the case of filamentation.

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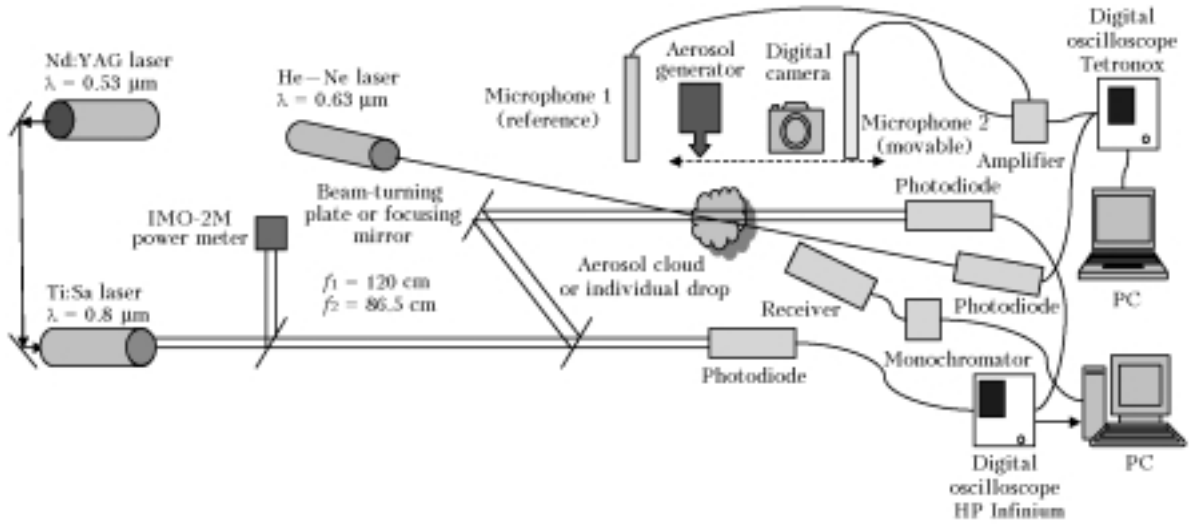


Fig. 1. Experimental setup.

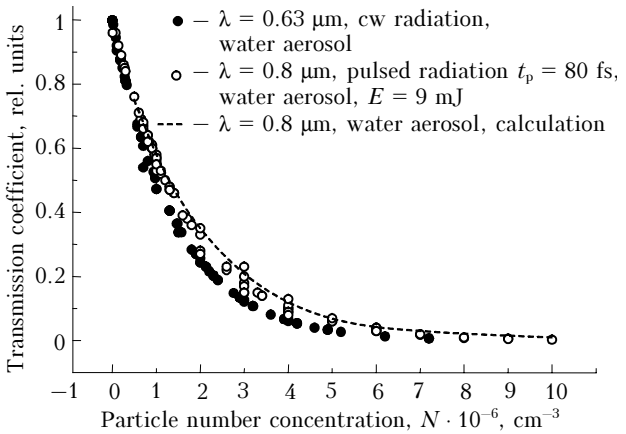


Fig. 2. Transmittance of the aerosol layer vs the particle number concentration.

No special measures were applied for sound-proofing of measuring microphones from the external acoustic noise (operating setup), achieving a sound pressure of ~0.025 Pa. Therefore, for processing of low acoustic responses at a sound pressure up to ~0.001 Pa we used our own Atmospheric Photoacoustic software,[1] including extended tools for optimal processing of photoacoustic digital data, namely, for compensation of frequency and phase distortions in the measurement channels, optimal bandpass filtering of useful signals against the background of external acoustic and instrumental noise, and compensation of nonlinear, diffraction, and dissipative distortions of the measured photoacoustic signals based on the particular experimental conditions.

Measurements of the acoustic response from a filament at different distances R from the mirror focus has shown that a significant extinction of the filament is observed in the case of focusing by the mirrors with the focal lengths f = 86.5 and 120 cm at R = 0.1–4.2 m (Fig. 3).

The receiving microphone of the reference acoustic signal (microphone 1) was spaced by 0.5 m from the mirror focus. Microphone 2 moved along the optical axis of the femtosecond laser radiation from the focus to the distance of 4.2 m. The microphones were set 1.5 cm far away from the optical axis of radiation. The maximum of the filament acoustic spectrum varied within 38–65 kHz, and the peak sound pressure varied roughly within 10^{-3}–5 Pa. To increase the signal-to-noise ratio, the initial data were filtered in the 10–250 kHz range and additionally averaged over series of 64 pulses. The mean energy of the femtosecond pulse for the dependences shown in Fig. 3 was 12 mJ. In Fig. 3, the parameter S is the integral of the absolute value of the acoustic pressure for the period of the generated acoustic response.

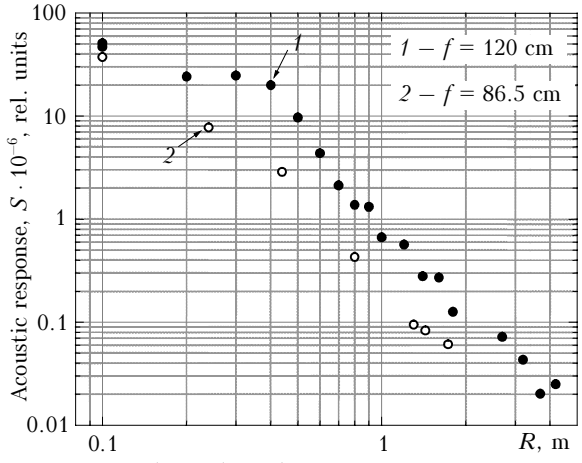


Fig. 3. Longitudinal dependence of the integral acoustic response generated by the channel of propagation of femtosecond laser pulse; the distance from microphone to filament is 1.5 cm.

Figure 3 shows that at a small R the filament energy decreases by orders of magnitude. In this case, the filament energy is understood as a part of the energy, which forms the weakly ionized plasma and then dissipates into thermal energy of the medium due to electron and ion recombination. Thus, the

acoustic signal is a measure of thermal loss of the laser radiation connected with the beam filamentation.

The observed decrease of the acoustic response indicates that behind the focus the filament already does not exist as a self-sustaining formation. The divergent beam does not deliver to the filament the energy, sufficient for the balance of two effects: Kerr nonlinearity for focusing and plasma formation for defocusing. Thus, most probably, we experimentally observe the decay of the filament formed still at the focus. This is also confirmed by results of the comparison of energy losses at different focusing. At a less focusing, the filament still exists as a stable formation at the distance of 0.4 m from the focus, but then its energy sharply decreases. The similar tendency of the sharp decrease of medium excitation behind the focus was mentioned in Ref. 2 when measuring the degree of ionization in the channel of propagation of the focused laser radiation of the femtosecond duration.

It is seen in Fig. 4 that, based on the recordings of the acoustic signal along the laser radiation propagation, it is possible to reconstruct the geometric size of the area with the increased ionization.

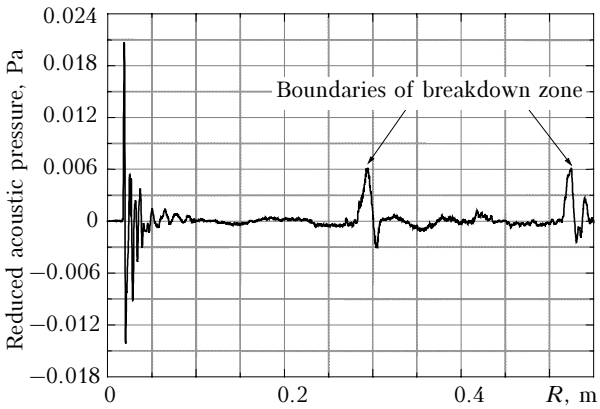


Fig. 4. Acoustic pressure reduced to the distance of 1 m, $P_{red} = P_{exp}R$; microphone is set 1.5 cm far from the laser beam.

The first pulse corresponds to the signal received from the laser beam as it passed just near (1.5 cm) the microphone. The second and third signals correspond to the boundaries of the zone of the increased ionization before and after the focus. Thus, for the case shown in Fig. 4, this zone is 23 cm long.

Figure 5 shows a dependence of the peak pressure in the acoustic signal on the transmission coefficient of the aerosol medium at the wavelength of the probing radiation.

It is interesting that upon transition from the femtosecond pulse to the nanosecond one with the pulse energy kept unchanged, the amplitude of the acoustic response decreases by two orders of magnitude. Two scenarios are possible. First, for the femtosecond pulse, since the action differs only in the intensity (by five orders of magnitude!), the multiphoton absorption in the particulate matter is realized. Second, for the femtosecond pulse, it is possible to neglect the heat outflow from silver nanoparticles, integrated into aerosol particles. Then the conditions of overheating and explosive water boiling take place in some local areas of the droplet. The acoustic response in this case is higher than in the case of thermal and evaporating mechanisms of its generation. However, the interpretation of the obtained result with the use of some or other mechanism requires a thorough assessment. In addition, these mechanisms are not alternative.

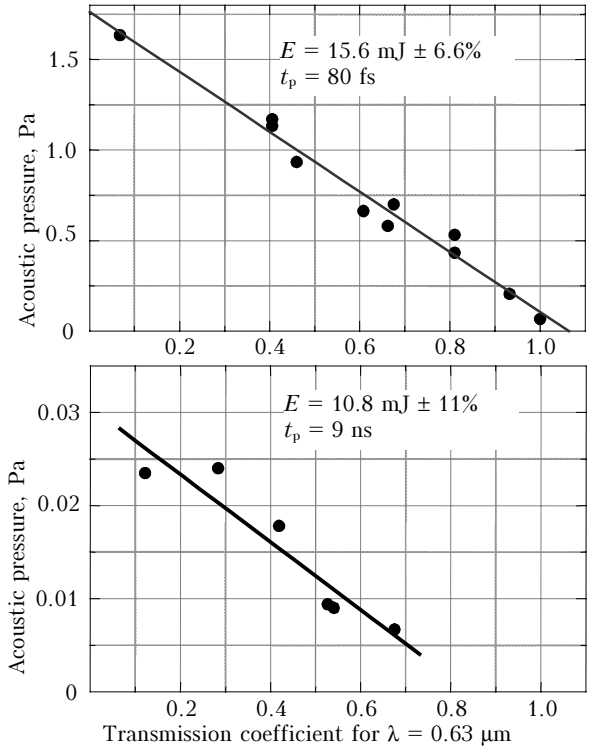


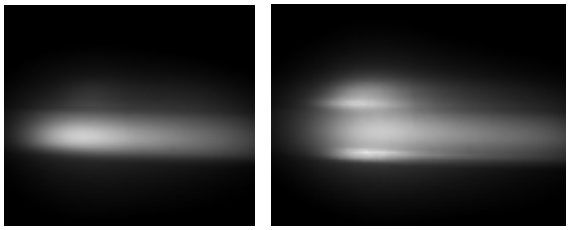
Fig. 5. Peak acoustic pressure generated by the water aerosol with nanoparticles vs. transmission coefficient; microphone is spaced by 5 cm from the aerosol medium; E is the laser pulse energy.

Another cycle of investigations involved the study of luminescence in a dyed (Rhodamine 6G) drop fixed on a capillary in the case of two-photon absorption of femtosecond pulses at λ = 0.8 μm.

The spectrum of the two-photon-induced luminescence of the dye in the drop was recorded with a monochromator and a CCD camera. The dispersion of the monochromator with the grating of 1200 lines/mm was 2.4 nm/mm. The typical spectrograms obtained for different values of the excitation energy are shown in Fig. 6. The vertical dimension of the image in Fig. 6 corresponds to the drop size (2 mm), while the horizontal one corresponds to the spectral distribution (within about 45 nm), the spectral pattern is centered at 580 nm.

Comparison of the spectrograms obtained at different energies of the exciting radiation (7.3 mJ (spectrogram *a* in Fig. 6) and 16.5 mJ (spectrogram

b)), shows a significant spatial-spectral inhomogeneity in the glow of different areas of the drop. Namely, zones corresponding to the drop's top and bottom are spectrally brighter at a high pump energy.



a *b*

Fig. 6. Spectrogram of luminescence of a dyed drop upon the excitation by femtosecond laser pulse.

The glow spectra of different drop areas are shown in Fig. 7: rows 1 and 2 correspond to the top and central parts of the drop in Fig. 6*b,* respectively. If the glow spectrum of the central part is a typical spectrum of spontaneous two-photon-induced luminescence of Rhodamine 6G, then that of the top part is much narrower and shifted to the shortwave region.

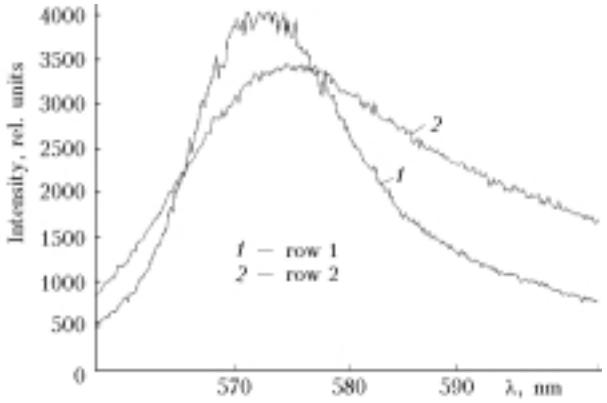


Fig. 7. Luminescence spectrum of a dyed drop excited by femtosecond laser pulse of 16.5 mJ energy.

Since the drop boundaries are whispering gallery boundaries, a possible explanation of the observed spatial-spectral differences is associated with the processes, proceeding in this zone, where, as known, the power density of the pumping radiation is somewhat increased. The upper part of the spectrogram corresponds to the point of contact of the drop and the capillary, that is, the quasispherical drop surface is there broken, and, correspondingly, the total internal reflection for the appropriate rays is distorted. The drop's bottom is characterized by another degree of sphericity as compared to its center.

Thus, in upper and lower parts of the drop, the conditions of total internal reflection are violated for the rays, which have been propagated earlier in the central part at total reflection angles to the drop surface. So, the radiation from the upper and lower parts of the drop includes a larger fraction of rays from the whispering gallery zone (that is, zone with the increased pump power density), as compared to the glow of the whole drop volume. Earlier we observed similar spectral differences in the case of one-photon excitation of dyes by nanosecond pulses under conditions of saturated absorption.[3]

Based on the conducted cycle of experiments, we can draw the following conclusions.

Propagation of femtosecond laser pulses does not change the transmittance of water aerosol measured at the wavelength of the cw probing radiation. Correspondingly, the aerosol microstructure does not change, and, consequently, thermal effects of evaporation and explosive boiling of the particulate matter are absent. In the case of optical breakdown of aerosol, detected by the acoustic method in some cases, the optical thickness of the mist also does not change. This indicates that the breakdown sites are formed inside a small number of aerosol particles and, without a short-pulse energy support they do not develop.

The acoustic response from the interaction between the femtosecond pulse and the aerosol medium, whose water particles are added with silver nanoparticles, two orders of magnitude exceeds the response from interaction between a nanosecond pulse of the same energy and the same medium. This is indicative of the additional energy loss of the laser beam upon its propagation through aerosol medium in the form of a femtosecond pulse. This loss does not exceed several percent, which is in agreement with optical measurements.

The measurements of the acoustic response from propagation of the filament, formed upon focusing of the laser radiation, evidence that the filament exists as a self-regulating form only near the focus.

The measurement of the acoustic signal in an open air is an effective instrument for studying the interaction of femtosecond laser pulses with aerosol media and the filament propagation through aerosol and gas media. This method allows us to reconstruct the energy properties of the filamented laser radiation, the geometric dimensions of the filament, and the transmittance of the aerosol layer for the femtosecond laser pulse both in the case of beam filamentation and without it.

Acknowledgments

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