Optical discharge in aerosols at propagation of laser radiation

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Experimental results on optical discharge in aerodisperse media containing solid aerosol particles at propagation of weakly focused laser beams are discussed. It is shown that appearance of two regimes of optical discharge depends on the wavelength and duration of the laser pulse, the particle size distribution function, the particle material, and the energy characteristics of laser radiation. An analytical equation is proposed that quite satisfactorily describes propagation of laser beams at optical discharge excitation.

In recent years, different research groups have studied nonlinear effects arising in aerodisperse media at propagation of laser beams. The results of several papers devoted to the study of physical processes occurring in clouds and fogs under the action of a high-intensity laser beam are summarized in Ref. 1. The studies of interaction of high-power laser radiation with aerodisperse media including solid aerosol particles have some peculiarities because of a wide variety of physical and optical characteristics of such particles. This directly relates to the phenomenon of optical discharge, which arises once the intensity of laser radiation propagating in an aerodisperse medium exceeds the value of 10^7–10^9 W/cm^2 [see, e.g., Refs. 2–7].

It is known that the optical discharge in gas media containing aerosol particles (e.g., in the atmosphere) is initiated at lower radiation intensity as compared with the discharge in pure gases. Although the theory of optical discharge in aerosols is developed insufficiently, there are no doubts that the optical breakdown in such media is initiated by particles. Unlike a considerable number of papers devoted to the study of optical discharge in highly focused laser beams, whose focal zones are comparable in size with the aerosol particles [for example, Refs. 2–4 and 7], this paper considers appearance of optical discharge in weakly focused beams under conditions close to the atmospheric ones. Based on the obtained experimental data, a phenomenological equation is proposed that well describes propagation of laser radiation under conditions of initiation of optical discharges both in a model medium and in the atmosphere.

Discharge threshold in laser beams of different wavelengths

In spite of a certain difference, the physical mechanisms of optical breakdown near aerosol particles in weakly focused beams at different wavelengths have many common features, at least in the spectral region of 0.3–10.6 μm. Analysis of data obtained at the Institute of Experimental Meteorology[8–12] for laser beams with the focal spot of 3 to 120 m in length (Fig. 1*a*) shows that, in this spectral region, the wavelength dependence of the threshold intensity of optical discharge excitation I_{th} at the standard atmospheric pressure is well approximated by the equation[13]

$$I_{th} = g\lambda^{-2},$$

where g is a parameter, λ is the wavelength in cm. Calculation with the model of thermal explosion of an aerosol particle gives the following dependence of the parameter g on the characteristic size a of the particle and pulse duration t_p (Ref. 16):

$$g = 2.5 \cdot 10^{-4} a^{-1.2}\, t_p^{-0.8},$$

where a is in μm; t_p is in s.

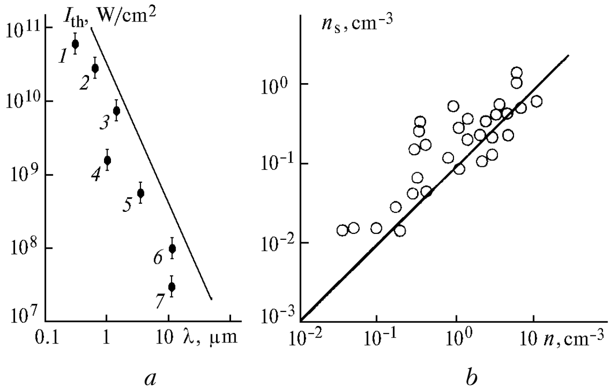

Fig. 1. Wavelength dependence of the threshold of a long laser spark occurrence in aerosol media (*a*): experimental data for air containing aerosol particles (*1–7*) at λ = 0.35 (*1*), 0.53 (*2*), 1.06 μm (*3*) (Ref. 10) λ = 1.06 (*4*), 3.8 (*5*), 10.6 (*6*), 10.6 μm (*7*) (Refs. 8, 9, 11, and 12) and dependence of the concentration of laser sparks n_s on the concentration of aerosol particles at the pulse energy of 100 J (*b*).

Optical breakdown in the field of radiation at λ = 1.06 μm

As a high-power laser beam propagates in the atmosphere containing aerosol particles, several, rather

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than one, initiation cells of optical breakdown arise, as a rule, near the beam axis. Such a form of the optical breakdown is called a “long laser spark” (LLS). Formation of laser sparks depends on the chemical composition, concentration, and size of aerosol particles. These dependences were studied experimentally for the case of LLS formation in the Nd laser beam using a setup that included a laser, an aerosol chamber, and recording instrumentation.[14]

The laser emitted radiation with the pulse duration $t_p = 80$ ns (at the intensity half maximum). The radiation was focused at a distance of 10 m by a confocal laser cavity; the mean area of the spot on the 2-m long path was 0.5 cm^2. Beam-splitters directed a part of radiation to the measuring unit (IKT–1N calorimeter, FEK–20 electronic recorder). Aerosol particles were injected into the 2-m long chamber. The concentration, size distribution, chemical composition (natural dust, soot, corundum, etc.) of aerosol particles varied widely. The aerosol microstructure was monitored by a photoelectric or TV counter. The number of breakdown cells was recorded with a camera. The energy and shape of radiation pulses were measured at the chamber entrance and exit.

Dependence of the discharge centers concentration on the concentration of aerosol particles

The concentration of LLS centers was measured at the laser output energy of 100 J and different concentration of corundum (Al_2O_3) aerosol particles. No LLS were observed, if the air in the chamber was carefully cleaned from aerosol particles (with Petryanov filters). As a result, we obtained a nearly linear dependence of the concentration of sparks n_s in LLS on the concentration of aerosol particles with the size $a \geq 1.4$ µm. This dependence is shown in Fig. 1*b*. To determine the physical processes in plasma evolution, the glow of the breakdown plasma was studied using spectral methods. The analysis revealed spectral lines of atomic aluminum, which is a chemical component of corundum, and nitrogen ions, as well as the continuum glow. The presence of aluminum lines indicates that LLS centers are formed near aerosol particles, and the presence of nitrogen ion lines indicates that not only particulate vapor, but air plasma glows too, i.e., optical discharge evolves in the air. The plasma temperature, as estimated from the spectral data, is $T = (2–3)\cdot 10^4$ K.

Relation of the concentration of sparks and the size of particles initiating optical breakdown with the radiation energy density

At the unchanged size distribution of corundum particles, the number of plasma centers n_s was analyzed as a function of the energy density and intensity of laser radiation at the chamber entrance. The experiments showed that n_s increases, as the energy density (intensity) of radiation increases. The comparison of the n_s dependence on I_{th} with the measured aerosol particle size distribution showed that as the radiation intensity increases, smaller aerosol particles take part in the formation of sparks. This comparison also allowed us to find the dependence of I_{th} on the minimum size a_{min} of particles initiating the breakdown. The dependence of I_{th} on a_{min} is shown in Fig. 2.

Figure 2 also shows the dependence (solid line) of the radiation intensity I_{th} on the least size a_{min} that approximates the experimental data by the function

$$I_{th} = b / a_{min}^k, \quad (1)$$

where $k = 1.5$, $b = 2.9\cdot 10^9$ W·cm^{-2}·µm$^{-1.5}$.

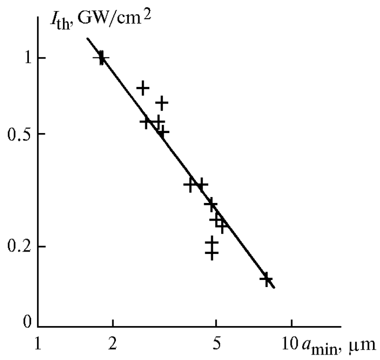

Fig. 2. Dependence of breakdown threshold I_{th} on the minimum size a_{min} of aerosol particles taking part in the formation of sparks; calculation by Eq. (1) (solid line).

It is seen that the experimentally determined value of the parameter k ($k = 1.5$) differs from the theoretical value ($k = 1.2$, see Ref. 13). In our experiments, the change of a_{min} from 0.5 to 5 µm leads to the change of I_{th}, at which LLS arose at unchanged spark concentration n_s, from 10^9 to $1.5\cdot 10^8$ W·cm^{-2}, i.e., roughly by seven times. Replacement of corundum particles with particles having far different optical constants (natural dust, soot, graphite, etc.) at roughly the same characteristic size of particles leads to the change of the breakdown threshold by no more than 2–3 times.

Radiation attenuation by plasma cells

Attenuation of radiation by plasma cells T_λ was estimated by measuring the radiation energy density at the chamber entrance E_0 and at the exit from it E. The dependences of $T_\lambda = E/E_0$ were obtained in two series of experiments. In the first series it was the dependence of T_λ on the concentration of aerosol particles n characterizing the initial aerosol optical depth in the chamber at the constant value $E_0 = (24 \pm 1)$ J/cm^2, and in the second series it was the dependence on E_0 at the constant concentration $n = (1.4 \pm 0.2)$ cm^{-3}. The experimental results are shown in Fig. 3.

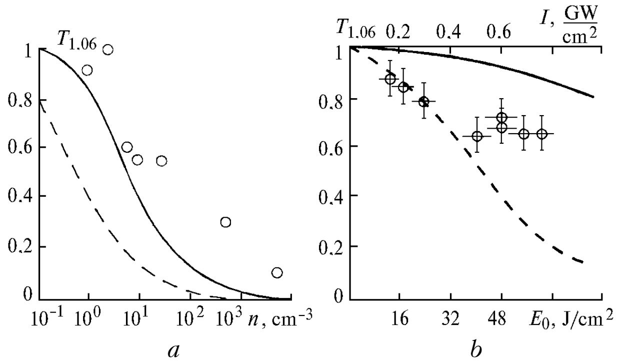

Fig. 3. Dependence of $T_{1.6} = E/E_0$ on the concentration of aerosol particles n (a) and on E_0 at n = const (b). Calculation by Eq. (13) (solid line) and Eq. (12) (dashed line) with the parameters $b = 2.9 \cdot 10^9$ W·μm$^{1.5}$cm^{-2}, $k = 1.5$, $S = 4$, $z = 2$ m.

As expected, the radiation was more strongly attenuated, as the concentration n increased at E_0 = const, because of the growth in the number of discharge cells (Fig. 3a). On the other hand, the increase in the number of large aerosol particles, because of the increase in the energy at n = const, also led to a stronger attenuation of the radiation, since discharge cells arise at smaller particles with the increase of E_0. Note that the concentration of small particles under natural conditions far exceeds the concentration of large particles ($a \gg 1$ μm) and, as a rule, increases strongly with the decrease in the particle size.[15] Below we present a comparison between the experimental results and theoretical calculations.

Optical discharge in CO_2 laser beam and radiation extinction

Extinction of CO_2 laser radiation ($\lambda = 10.6$ μm) by the breakdown cells formed on particles of natural aerosol was studied under field conditions along the path several tens meters long at the wind speed up to 0.5 m·s^{-1}. The radiation energy density averaged over the beam cross section reached E_0 = 12 J·cm^{-2}. The breakdown occurred at the energy density E_0 = 4.5–6 J·cm^{-2}. The mean concentration of sparks at the energy density E_0 = 10 J·cm^{-2} was $n_s = 8 \cdot 10^{-5}$ cm^{-3}. The dependence of $T_\lambda = E/E_0$ on E_0 is shown in Fig. 4; it is analogous to the dependence shown in Fig. 3b for λ = 1.06 μm. The threshold intensities of the CO_2 laser radiation, at which we observed the breakdown and formation of plasma centers, were lower than those for the Nd laser radiation (see Fig. 1).

Extinction of the CO_2 laser radiation was also studied under model conditions. A model medium was generated by spraying the corundum (Al_2O_3) powder in the chamber with the dimensions 5 × 2.5 × 1.2 m. The chamber was set 30 m far from the laser exit window. The size distribution of corundum particles $f(a)$ measured by the photoelectric counter was close to the exponential Junge distribution. A 30-μs long pulse of the CO_2 laser was focused at the center of the chamber. The pulse shape was the ordinary shape of a pulse of an electroionization CO_2 laser. The intensity in the leading peak lasting several hundreds nanoseconds was about ~10^8 W/cm^2.

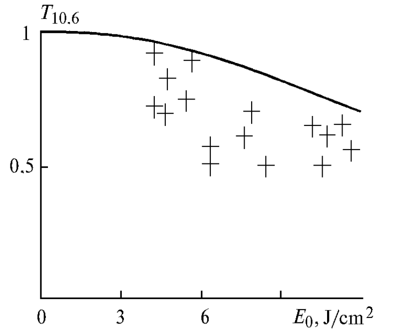

Fig. 4. Dependence of $T_{10.6} = E/E_0$ on the initial energy density E_0; calculation by Eq. (13) (solid line); $b = 1.6 \cdot 10^9$ W·μm^{2}cm^{-2}, $k = 2$, $F = 0.5$, $S = 4$, $m = 1/3$, $z = 100$ m.

The measurement results on the pulse energy after passage through the chamber with sprayed aerosol particles are shown in Fig. 5. In spite of twofold variation of the pulse energy, the energy of radiation passed through the chamber is rather stable. This can be explained by the formation of discharge cells. Since the discharge cells overlap almost completely with the waist cross section the energy measured at the exit was the energy of radiation passed through the chamber before the formation of discharge cells. The plasma in the developed cells almost completely absorbed radiation. The plasma temperature was estimated from the data of spectroscopic measurements to be $(2.5–3.5) \cdot 10^4$ K; this value depended on the pulse energy.

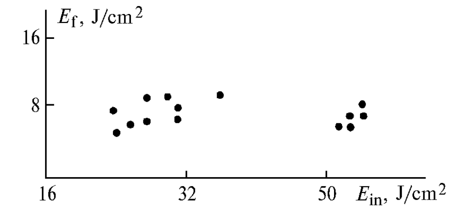

Fig. 5. Transmission of the pulse energy through Al_2O_3 aerosol: E_{in} and E_f are the initial and final energy of the pulse.

The possibility of initiating optical breakdown in the aerosol medium was also studied by sending double pulses of the CO_2 laser through the chamber. The initial energy density in the pulses ranged from 15 to 35 J/cm^2. The mean intensity varied from 1.5 to $3.5 \cdot 10^7$ W/cm^2. As before, the radiation was focused at the center of the chamber. The duration of the first pulse was 1.5–2 μs, and the duration of the second pulse was 2.5–3 μs. The gap between the pulses varied from 20 to 100 μs. In the corundum powder sprayed in the chamber, the mass fraction of particles with the radius of 1–3 μm was 70%, and that of the particles with radii larger than 5 μm was less than 0.5%.

The breakdown cells arose in the medium at passage of the first pulse. Comparing the number of the centers with the particle size distribution, we found that at the pulse energy density ≥ 20 J/cm^2 the breakdown was initiated on particles with radii ≥ 5 μm. As the second pulse propagated, the discharge centers arose at the same places as those in the first pulse. This indicates that solid particles were not burned out at the formation of the discharge cells by the first pulse and were able of initiating the breakdown.

Evolution of plasma cells in space and time

As follows from the above results, after appearance of the plasma cells on aerosol particles in the field of high-power laser radiation, attenuation of laser radiation by the plasma cells in time depends on the rate of the plasma cell growth or, what is the same, on the speed of propagation of the laser radiation absorption wave (LRAW).

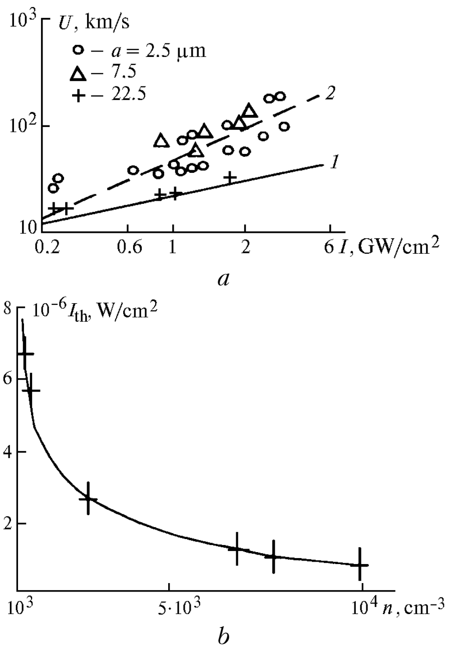

Fig. 6. Dependence of the LRAW speed on the radiation intensity (*a*): LDW (*1*) and FIW (*2*); dependence of the threshold intensity of low-threshold optical discharge on the concentration of aerosol particles (*b*): experimental data (crosses) and calculation (solid line).

The experiments conducted with application of high-resolution equipment showed that the light-detonation mode of LRAW (LDW) takes place for Nd lasers at the intensity $\approx 10^8$ W/cm^2 (Refs. 5, 6, and 16), and at the radiation intensity higher than $(2-3)\cdot 10^8$ W/cm^2 the speed of LRAW can significantly exceed the speed of LDW.[17,18] To study the speed of LRAW in a more detail, we have conducted experiments on the exposure of tungsten threads of 5, 15, and 45 μm in diameter to Nd laser radiation. The data on the maximum speed of LRAW in air shown in Fig. 6*a* indicate that the LRAW speed could exceed the LDW speed by tens times, and the LRAW speed at the initiation of the breakdown on thin threads (5 and 15 μm) are far higher than on the thick threads (45 μm).

Propagation of LRAW in a dense gas ($P = 1$ atm) with the speed far higher than the LDW speeds at the radiation intensity of 10^8-10^9 W·cm^{-2} can be explained by complex mechanisms of gas ionization ahead of the wave front. Such LRAW are usually called fast ionization waves (FIW).[19] Sufficiently rigorous theory of FIW formation in air is not yet developed.

Low-threshold optical discharge

At rather high concentrations of aerosol particles ($n > 10^3$ cm^{-3}), the optical discharge threshold decreases down to $I_{th} = 1-10$ MW/cm^2. This phenomenon of the low-threshold optical discharge was first described in Ref. 20; collective effects in aerosol cause it. At high concentration of large particles ($a \geq a_{min}$), the discharge cells at individual active particles overlap and thus form an optical discharge zone, the absorption of radiation inside which far exceeds the surface losses. Three stages can be isolated in the formation and evolution of the low-threshold optical discharge. At the first stage, nucleation and center growth occur near individual aerosol particles; then the centers merge into one macroscopic center. This stage lasts from 40 to 100 μs what depends on the aerosol microstructure and radiation intensity. The influence of solid particles manifests itself only at the first stage (nucleation and growth of discharge centers). The second stage 0.1–1 ms long is characterized by the growth of the macrocenters. The duration of this stage is determined by the duration of laser pulse. The cell of the low-threshold discharge achieved 2.5–8 cm in size depending on the experimental conditions. The third (dissipation) stage lasts ~0.4 ms. The rate of the plasma cell growth is about tens meters per second. The physical mechanism of its evolution is slow combustion.[5,16,22] The criterion of appearance of the low-threshold optical discharge was established as a condition for the amount of radiation energy absorbed by an ensemble of particles

$$\bar{\sigma}_{ab}(a)nI = S,$$

where $\bar{\sigma}_{ab}$ is the spectrum-mean absorption cross section of a particle; S is a constant depending on particle characteristics. The values of S for some aerosol media are tabulated below.

Table

Aerosol medium	Al_2O_3	Al	Mg	C
S, W/cm^3	$3.8\cdot 10^4$	$3\cdot 10^5$	$3\cdot 10^5$	$5.5\cdot 10^5$
Aerosol medium	SiO_2	NaCl	S	$Hg(NO_3)_2$
S, W/cm^3	$6\cdot 10^5$	$1.6\cdot 10^6$	10^7	$2\cdot 10^7$

From the equation for the criterion of discharge appearance, it follows that the dependence of the threshold intensity on the particle concentration is hyperbolic. This character of the dependence is supported by the experimental results shown in Fig. 6*b* for the case of aerosol of corundum particles with the mean size $a = 2.4$ μm $(S/\bar{\sigma}(a) = 8.8\cdot 10^9$ W/cm$^5)$ (Refs. 6, 21, and 22).

Calculation of attenuation of laser radiation by the breakdown cells

Propagation of laser radiation in an aerodisperse medium with growing plasma cells is described based on the transfer equation

$$\frac{\partial I}{\partial t} + c\,\frac{\partial I}{\partial z} = -c\alpha(z, t)I, \quad z \geq 0, \quad t \geq 0 \qquad (2)$$

assuming the condition that $I(0, t) = I_0$.

In Eq. (2) the following designations are used: $I(z, t)$ is the radiation intensity; z is the coordinate along the direction of radiation propagation; t is time; c is the speed of light; α is the extinction coefficient.

Assume that a breakdown cell arises at a point z at the time $t = z/c$ and grows at the rate

$$v = v_0 I^m, \quad v_0 = \text{const}. \qquad (3)$$

This dependence on the rate is usually peculiar to the LDW $\{m = 1/3,\ v_0 = 0.375[2(\gamma - 1)\rho]^{1/3},\ \rho$ (g/cm^3) is the air density; γ is the adiabatic exponent$\}$ and FIW ($m = 1$, $v_0 = 6.4\cdot 10^{-3}$ cm^3 s^{-1} W^{-1}) modes.

Assume then, that the breakdown cells are opaque for laser radiation, are spheres with the cross size $R \gg \lambda$. In this approximation (it is violated only in the initial period of nucleation and growth of the breakdown cells), the cross section of radiation attenuation by the breakdown cell is determined by the equation

$$\sigma = 0, \quad t < z/c;$$
$$\sigma = 2\pi v_0^2 \left(\int_{z/c}^{t} I^m(z, t')\,\mathrm{d}t' \right)^2, \quad t \geq z/c. \qquad (4)$$

On the other hand, it follows from the approximation (3) that once the discharge cell arises, the radiation is attenuated by the aerodisperse medium only in the path section preceding the place of the discharge cell. Ignoring the cell overlapping, we have

$$\alpha(z, t) = \alpha_0 + \int_{a_{\min}}^{\infty} \sigma(z, t)\, f(a)\, \mathrm{d}a. \qquad (5)$$

It was mentioned above that the discharge cells arise on large particles: $a \geq a_{\min}(I)$. The size distribution of large particles is described by the exponential function

$$f(a) = Fa^{-s}, \quad a \geq a_{\min}(I). \qquad (6)$$

Taking into account Eq. (6), from Eq. (5) we have

$$\alpha(z, t) = (s - 1)^{-1} F a_{\min}^{(1-s)} \sigma + \alpha_0. \qquad (7)$$

Introduce the auxiliary function

$$G(z, t) = \int_{z/c}^{t} I^m(z, t')\mathrm{e}^{m\alpha_0 z}\, \mathrm{d}t', \ t \geq z/c \qquad (8)$$

and substitute the variables $x = z$, $\tau = t - z/c$.

Then Eq. (2) with the allowance for Eq. (5) can be transformed into the equation for the function $G(x, \tau + x/c)$

$$\frac{1}{m}\frac{\partial G}{\partial x} = \frac{2\pi v_0^2 F}{3(s - 1)a_{\min}^{(s-1)}}\,\mathrm{e}^{-2m\alpha_0 x}\, G^3, \quad \tau \geq 0 \qquad (9)$$

with the condition $G(0, \tau) = I_0^m\,\tau$.

As was shown above, the value of $a_{\min}$ depends on the intensity of radiation incident on the particle. Taking the experimental dependence for the dependence of $a_{\min}$ on the intensity, Eq. (1), in the form $I_0\mathrm{e}^{-\alpha_0 z} = b/a_{\min}^k$ we can present Eq. (8) as

$$\frac{\partial G}{\partial x} = -\xi \mathrm{e}^{-\mu x} G^3, \qquad (10)$$

where it is denoted

$$\xi = \frac{2\pi v_0^2 m F}{3(s - 1)}\left(\frac{I_0}{b}\right)^{(s-1)/k}; \quad \mu = \left(\frac{s - 1}{k} + 2m\right)\alpha_0.$$

Integrating Eq. (10), we obtain

$$G = \left[\frac{2\xi}{\mu}\,(1 - \mathrm{e}^{-\mu x}) + \frac{1}{I_0^{2m}\tau^2}\right]^{-1/2}. \qquad (11)$$

Now let us pass from the function G to the function I using the equation

$$I(x, \tau + x/c) = \mathrm{e}^{-\alpha_0 x}\left(\frac{\partial G(x, \tau + x/c)}{\partial \tau}\right)^{1/m}$$

and simultaneously come back to the variables z and t. As a result, for the radiation intensity we have the equation

$$I(z, t) = I_0\mathrm{e}^{-\alpha_0 z}\left[1 + \frac{2\xi}{\mu}\, I_0^{2m}\,(t - z/c)^2(1 - \mathrm{e}^{-\mu z})\right]^{-3/2m},$$
$$t \geq z/c.$$

Integrating the latter equation over time, we find the energy density in the cross section z for the time t from the beginning of laser pulse. For the case of fast ionization wave

$$E(z, t) = I_0\mathrm{e}^{-\alpha_0 z}(t - z/c) \times$$
$$\times\left[1 + \frac{2\xi}{\mu}\, I_0^2(t - z/c)^2(1 - \mathrm{e}^{-\mu z})\right]^{1/2},$$
$$t \geq z/c; \qquad (12)$$

and for the case of light-detonation breakdown wave

$$E(z, t) = I_0\mathrm{e}^{-\alpha_0 z}\,\frac{(t - z/c)}{D^{3/2}} \times$$
$$\times\left(1 + \frac{3}{5}\,\frac{h^2(t - z/c)^4}{D} - \frac{1}{7}\,\frac{h^3(t - z/c)^6}{D^2}\right), \qquad (13)$$
$$D = 1 + h(t - z/c)^2, \quad h = \frac{2\xi}{\mu}\, I_0^2(1 - \mathrm{e}^{-\mu z}).$$

Figures 3 and 4 show the calculated results on $T_\lambda = E/E_0$ as a function of the initial radiation energy density E_0 for our experimental conditions ($\alpha_0 z \ll 1$). Some discrepancy between the calculation and the experiment is connected with several causes: imperfection of the calculation procedure, insufficiently accurate experimental data (particle size and concentration, energy distribution over the cross section, errors in determination of the radiation intensity, etc.) [see also Refs. 5, 6, 23, and 24].

From the results presented above it follows that now we have certain ideas on the regularities of optical discharge excitation in aerodisperse media. These regularities were studied mostly in laser beams with the wavelengths of 1.06 and 10.6 μm. Undoubtedly, it would be useful to extend the list of wavelengths and aerodisperse media in the experiments on optical discharge and thus to make more clear the nature of optical discharge initiation in aerosols.

Acknowledgments

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