

# Stratified-medium model in studying propagation of high-power femtosecond laser radiation through atmospheric aerosol

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The paper presents the stratified-medium model developed for describing coherent scattering of high-power laser radiation by an ensemble of water aerosol particles applied to solving problems in the femtosecond nonlinear optics. The model presents the aerosol medium in the form of a sequence of layers, with the particles forming thin aerosol screens. The qualitative and quantitative comparison of the results obtained using the model constructed and the Bouguer law for fine aerosol media has been done. The numerical experiment on laser beam propagation through the clouds was performed. The possibility is shown of generating multiple filaments due to perturbations of the intensity of high-power laser beam in aerosol media.

## Introduction

At high-power femtosecond laser pulse propagation through the atmosphere the filamentation is observed, i.e., the spatiotemporal localization of the radiation energy.<sup>1–3</sup> More than 10% of pulse energy remains localized in the near axial region of 100- $\mu\text{m}$  diameter along the whole filament reaching the length about hundred meters.<sup>3</sup> This phenomenon is accompanied by generation of the optical supercontinuum,<sup>2,4,5</sup> which is considered as a promising light source for (the wide-band atmospheric sensing) monitoring of the environment.<sup>6</sup>

The filamentation of a femtosecond laser pulse in the atmosphere, which is essentially heterogeneous multicomponent medium,<sup>7</sup> is characterized by some peculiarities. Perturbations of the refractive index in the turbulent atmosphere result in random shifts of the filament from pulse to pulse<sup>8,9</sup> and in the formation of a stochastic bundle of filaments in a terawatt power pulse.<sup>10</sup>

Particles of atmospheric aerosol cause an increase in the local field of the incident radiation.<sup>11</sup> In this case the light field concentration inside a particle leads to formation of a plasma source in it.<sup>12,13</sup> The formation of plasma channel at propagation of a femtosecond laser pulse through a disperse medium of atmospheric aerosol has been observed experimentally.<sup>14</sup> According to the assessment obtained in Ref. 15 the breakdown threshold for a femtosecond laser pulse in a transparent particle is two orders of magnitude lower than in clear air. The localization of laser radiation in an aerosol particle results in an increase of a fluorescence signal.<sup>16</sup> As shown experimentally<sup>17</sup> and theoretically,<sup>18</sup> the fluorescence at multiphoton molecular excitation in a particle has maximum in the backward direction that makes a prerequisite for sounding atmospheric aerosol using femtosecond laser pulses.<sup>19</sup>

The scattering and absorption of radiation by aerosol particles in the turbid atmosphere can affect the generation of filaments and their extension. In Ref. 14 a series of experiments was described on the interaction of a filament with separate water droplets. Large water particles of 30 to 100- $\mu\text{m}$  diameter were placed along the propagation path of a filament. Droplets of such sizes very often occur in rain clouds.

In the experiment it was detected that such a “large” particle, i.e., comparable with the filament diameter, makes a negligible effect on the further filamentation process. The authors explain this phenomenon by the fact that in the course of formation of a filament interacting with a droplet the peripheral regions of laser beam cross section prevail, whereas the near axial regions are of minor importance.

This interpretation is consistent with the conclusions drawn in Ref. 20, where the energy flows in the pulse cross section were shown to be caused by the Kerr self-focusing in air and defocusing in the laser-induced plasma. Experiments<sup>14</sup> with a separate droplet agree with the results from Ref. 21 in which the stop screens were set on the filament way, namely, the ones that can either transmit or block the radiation of its central part. Theoretically the influence of a separate droplet on the filament was considered in Ref. 22 in the approximation of the simplest model, in which the particle was presented as an absorbing disk.

The filamentation of a laser pulse in a disperse medium of atmospheric aerosol was studied experimentally in Refs. 14 and 23. In Ref. 14 it is shown that at the filament propagation through the aerosol with high particle concentration ( $10^5 \text{ cm}^{-3}$ ) the energy of the entire light beam decreases due to light scattering and its power is insufficient to maintain the filamentation regime. In the experiments<sup>23</sup> it was demonstrated that the nonlinear interaction with aerosol did not affect significantly the energy characteristics of radiation, and the water

aerosol transparency did not change under the action of a femtosecond laser pulse at wavelength of  $0.8 \mu\text{m}$  with the pulse energy up to 17 mJ.

Besides, at laser pulse propagation in the turbid atmosphere (cloudiness, fog, solid-state aerosol) perturbations of light field may occur due to aerosol particles. Because of scattering by particles, the regions with high intensity can be formed, which may serve centers of the filamentation.

Until so far, the influence of light scattering by the atmospheric aerosol particles on the filamentation of a femtosecond laser pulse has not been investigated theoretically. The role was not considered of coherent scattering by a polydisperse ensemble of aerosol particles affecting the generation and formation of the filament in the field of a high-power femtosecond laser pulse. However, chaotically located aerosol particles can essentially influence the filamentation.

In this paper we numerically study the coherent scattering of radiation by aerosol particles and the process of generation of filaments at the places of intensity perturbations caused by the interference of scattered radiation. To describe the field scattering on particles, a model was constructed where the phase wave relations are taken into account at their superposition outside a particle.

## The model of stratified aerosol

It should be noted that the laser pulse filamentation takes place due to strong nonlinear-optical interaction between the radiation and a medium, and for its investigation the theory of transfer and the diffusion approximation are inapplicable,<sup>24</sup> as those operate with the beam intensity, which equals the overall density of power flux at incoherent scattering of light field on a large number of disordered particles. Whereas the coherent scattering can significantly affect the pulse filamentation, determining the generation and formation of the multiple filaments, the processes of mutual energy exchange between filaments and the beam.

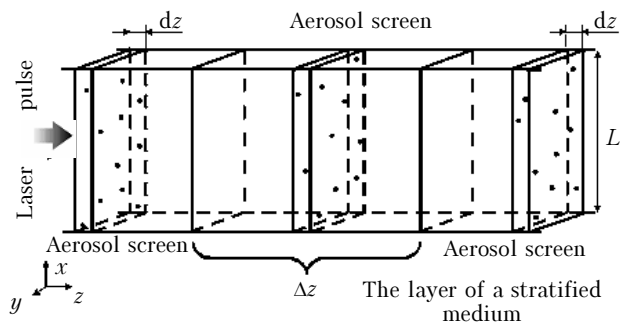
In calculating the laser pulse field in aerosol, it is necessary to solve the problem of coherent scattering of the radiation for each particle of the ensemble. The filament length may be several hundreds of meters, and at propagation in the atmospheric cloudiness a pulse interacts with several tens and hundreds thousands of water drops. Analysis of scattering by the Mie theory for such a large ensemble was impossible. Therefore, to solve the above problem it is essential to construct a simple wave model of scattering, which could take into account the phase difference between scattered and transmitted field components. The grounds and possibilities of constructing such a model are the following.

In the case of a laser radiation there exists a separated coordinate, along which the propagation occurs. The existence of such an evolution coordinate makes it possible to construct the propagation models in which the radiation successively passes the layers

of continuous medium. For the problem of wave propagation through a randomly inhomogeneous medium the stratified-medium model is the basis for the method of small local perturbations<sup>25</sup> in the turbulent atmosphere, as well as for the phase screens method<sup>26</sup> in the case of a nonlinear medium, the screens of nonlinear phase inversion.<sup>27</sup> From the standpoint of the calculus, the stratified-medium model is a physical prototype of the splitting method by physical factors.<sup>28</sup> The stratified-medium model of laser pulse propagation or the splitting method by physical factors, enable one to make use of the advantages of the numerical methods and algorithms for analysis of separate processes. In our case these are the scattering, diffraction, and nonlinear interaction with the atmosphere. As a result, the stratified-medium model gives a considerable decrease of computing time.

The basic idea of the stratified-medium model in application to laser radiation propagation through water aerosol is that the disperse medium is presented in the form of a series of layers of the width  $\Delta z$ , whose particles are located in plane "aerosol" screens (Fig. 1).

Between the screens free diffraction of radiation with the medium take place. Thus, all the particles of a real medium within a layer of width  $\Delta z$  are considered, within the stratified-medium model, to be concentrated in a thin "aerosol" screen.



**Fig. 1.** The model of stratified aerosol medium. Dots are water aerosol particles.  $L$  is the transverse dimension of the considered medium,  $\Delta z$  is the distance between the neighbor aerosol screens.

The problem we consider here is characterized by a wide range of spatial scales of variation of light field and aerosol particle size, which can be described by the following system of inequalities:

$$L > a \gg d_f > D \geq h, \quad (1)$$

where  $a$  is the beam radius,  $d_f$  is the cross size of a filament, and  $D$  is the diameter of an aerosol particle. The inequality (1) is supplemented with the limitations imposed on the size of the calculation region  $L$  and the transverse step of the grid  $h$  to provide adequate results of the numerical investigations. In the air the filament diameter  $d_f$  is  $\sim 100 \mu\text{m}$ , and for a laser beam of radius 1 cm the size of the region  $L$  must exceed the step  $h$  by four

orders of magnitude  $L/h \sim 10^4$ . Fulfillment of this condition in the rectangular coordinate system on the plane of the pulse cross section requires excessive computer resources. Therefore, the optimization of the physical model and the construction of an effective calculation algorithm are very important in solving the problem considered.

The aerosol screen in the disperse medium, in contrast to the phase screen in the turbulent atmosphere, has the finite thickness and consists of two parallel planes located at the distance  $dz$  (Fig. 1). At the first plane the particle ensemble is given, which scatters the field; at the second plane – the interference of radiation scattered by droplets with an unperturbed field is calculated. In this model, it is generally taken that the aerosol particles are located at the nodes of the design grid. The scattering by different particles of one aerosol screen occurs independently. The water aerosol particles are “soft” and to determine the field scattered by particles the method of anomalous diffraction is used.<sup>24</sup> The field  $E(\mathbf{r}, z)$ , scattered by a particle, is calculated using the Kirchoff integral<sup>29</sup>:

$$E(\mathbf{r}, z) = -\frac{i}{\lambda} \iint_{S_D} (e^{-ip(\xi, \eta)} - 1) e^{-ikl(\xi, \eta, \mathbf{r}, z)} \frac{E_0}{l(\xi, \eta, \mathbf{r}, z)} d\xi d\eta, \quad (2)$$

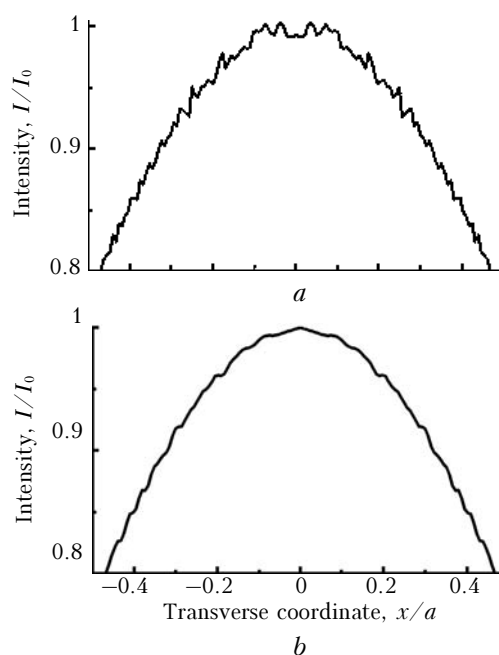
where  $E_0$  is the electric field strength of the incident light;  $\lambda$  and  $k$  are the wavelength and the wave number, respectively;  $l(\xi, \eta, \mathbf{r}, z)$  is the distance from the point  $(\xi, \eta)$ , located in the diameter plane of the particle  $z = 0$ , to the observation point  $(\mathbf{r}, z)$ . The parameter  $p(\xi, \eta)$ , being equal to the phase shift of the beam transmitted through a particle at the point with coordinates  $(\xi, \eta)$ , is determined depending on the method the anomalous diffraction is treated with. The integral is taken over the cross section  $S_D$  of a spherical particle. The constructed model of aerosol screen enables us to describe the coherent scattering of the wave by particles and the interference of scattered waves.

The scattering by particles results in the angular deviation of the wave vector  $k$ , and, hence, in the appearance of the field components divergent from the direction of propagation. In the numerical experiment, the calculation area of the finite dimension  $L$  is used, and these components can be reflected from its boundaries. To eliminate the possibility of reflection of radiation from the boundaries, the absorption coefficient is introduced in the boundary area of the grid, which increases smoothly along the direction toward the boundary. Such absorption removes the scattered field from the field of consideration and thus is responsible for the decrease of the beam energy at scattering.

The thickness of the aerosol screen  $dz$  was chosen under condition that the fields scattered by particles in one plane do not overlap in the second plane. Besides, the thickness of the aerosol screen  $dz$  should be much less than the layer width  $\Delta z$ .

In the numerical simulation the thickness of aerosol screen was chosen to be equal to  $dz = 2$  mm and the layer thickness  $\Delta z = 15$  cm. In simulating the coherent scattering of a radiation beam with the wavelength  $\lambda = 0.8 \mu\text{m}$  radius  $a = 2.5$  mm in the disperse medium with the particle density up to  $N = 100 \text{ cm}^{-3}$  and their radii  $R = 2-10 \mu\text{m}$  we used the calculation grid of the size  $L = 7.5a$  and the step  $\sim 20 \mu\text{m}$ .

To illustrate the influence of aerosol screen on the radiation propagation, we have considered the screen with one water particle of radius  $R = 2 \mu\text{m}$ , located on the axis of a collimated beam of the Gaussian profile. Figure 2 shows near axial ( $|x| \leq 0.5 a$ ) intensity distribution  $I(x)$  in two planes outside the aerosol screen.



**Fig. 2.** The intensity profile in the near axial region of a collimated Gaussian beam, scattered by a single water droplet of radius  $R = 2 \mu\text{m}$ , located on its axis. The distance of propagation  $z$  behind the aerosol screen:  $z = 15$  cm (a);  $z = 30$  cm (b).  $I_0$  is the intensity on the axis of an incident beam of radius  $a = 2.5$  mm.

The diffraction at a single layer of the width  $\Delta z = 15$  cm smoothes the interference of perturbations occurring due to superposition of unperturbed wave and the wave scattered by a single spherical droplet (Fig. 2a). The contrast of the interference fringes formed at scattering by a single particle is low, and the relative variation of intensity in maxima and minima does not exceed 1%. Later on (Fig. 2b) the contrast of the interference pattern decreases. The effect of a single particle of size  $2 \mu\text{m}$  on the beam intensity distribution is weak. Nevertheless, the laser beam scattering by an ensemble of particles results in a significant distortion of the radiation intensity profile.

This is shown in Fig. 3, where the intensity profile is given in a beam emitted at the distance  $z = 1.5$  m through the chain of 10 aerosol screens, each of those contains  $\sim 5000$  water droplets of the radius  $R = 6 \mu\text{m}$ .

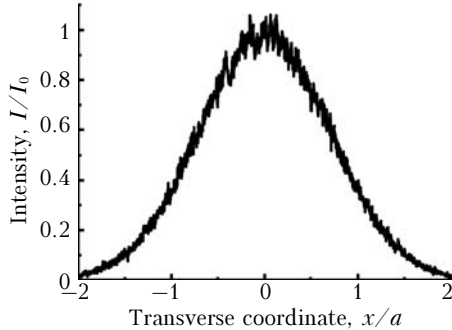


Fig. 3. The beam intensity profile.

In each screen the particles were positioned randomly at the grid nodes according to a uniform distribution. This example corresponds to the stratified-medium model of a monodisperse medium with the particle concentration  $N = 100 \text{ cm}^{-3}$ . In this case, the intensity fluctuations produced by the wave interference scattered by the many randomly located particles, reach 10%.

### Laser beam scattering by a monodisperse aerosol

To demonstrate the stratified-medium model, we consider, as an example, the laser beam scattering by monodisperse media with the sizes of water particles  $R = 2, 6$  and  $10 \mu\text{m}$ . The particle concentration is chosen to be the same and equal to  $N = 100 \text{ cm}^{-3}$ , that corresponds to the atmospheric cloudiness. The absorption in water particles is low, and its effect on the beam propagation is not taken into account. The propagation length in all cases was  $z \approx 24.5$  m that is one half of the diffraction length for a laser beam of the selected radius. The model of stratified-medium contained a series of  $\sim 160$  statistically independent aerosol screens. Profiles of the mean beam intensity in aerosol medium were determined by the Monte Carlo method over the array of 40 statistically independent realizations of the series of screens.

Mean profiles of laser beam intensity for media with different particle sizes are denoted by the solid curve in Fig. 4. The intensity profiles are shown by dashed line for the radiation propagated through the medium without particles. Note that because of scattering the average intensity of the beam propagated through the disperse medium is less than in the absence of particles. With the increase of the particle diameter their scattering cross section  $\sigma_p$  grows and at a constant concentration  $N$  the power attenuation, caused by the scattering, is more essential.

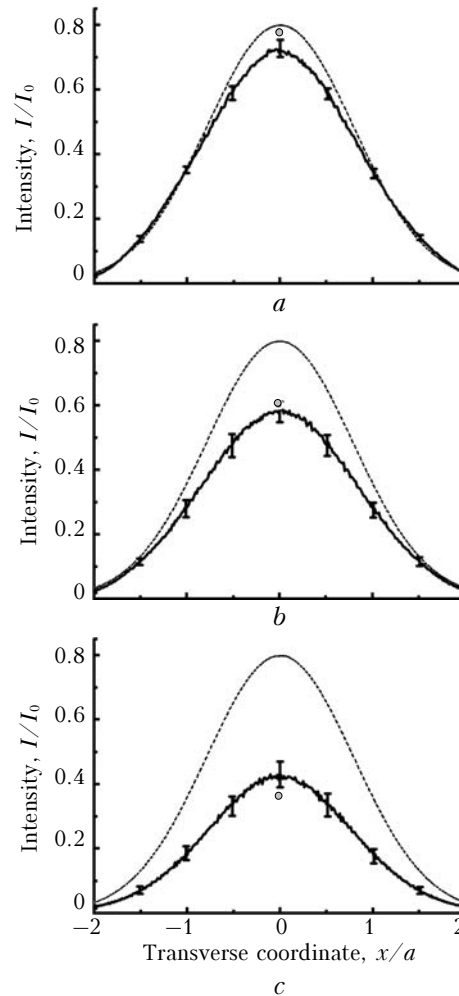


Fig. 4. Mean beam intensity profiles obtained by the method of statistical tests using the ensemble of 40 realizations of a monodisperse aerosol with the particle concentration  $N = 100 \text{ cm}^{-3}$  and size of droplets  $R = 2$  (a);  $6$  (b), and  $10 \mu\text{m}$  (c). The result of estimating by the Bouguer law is shown by a point at  $x = 0$ . The distance of propagation  $z \approx 24.5$  m.

For estimating the obtained results we use the Bouguer law for the intensity  $I(z)$  in the disperse medium<sup>24</sup>:

$$I(z) = I_d(z)e^{-\tau}, \quad (3)$$

where  $\tau(z)$  is the optical depth;  $I_d(z)$  is the radiation intensity in the absence of aerosol at propagation to the distance  $z$  in the scattering medium. In the case of monodisperse aerosol the optical depth  $\tau$  equals:

$$\tau = \sigma_p N z. \quad (4)$$

With the increasing particle size,  $R$ , the optical depth  $\tau$  of the medium rises at equal particle concentration, and the beam intensity is attenuated significantly. Using the tabulated values<sup>30</sup> for the scattering cross section of water droplets at the wavelength  $\lambda = 0.8 \mu\text{m}$ , the intensity on the beam axis in the considered media was calculated by

formulas (3) and (4). The obtained values are shown in Fig. 4 by separate points at  $x = 0$ .

Deviations of the results obtained by simulation from the given estimations can be explained by the fact that the Bouguer law was derived in the approximation of single scattering. The contribution of single scattered radiation to the intensity  $I(z)$  calculated according to the known methods<sup>24</sup> is no more than 0.1% under conditions accepted in simulation. Besides, the influence of multiple scattered radiation on the value  $I(z)$  can be found to be significant. With the increase in the particle size, the width of the beam pattern of scattered radiation is narrowed, and the influence of multiple scattered component on the beam axial intensity increases.

### Scattering by an aerosol polydispersion

The particle size-distribution function in the atmospheric cloudiness was approximated by the modified gamma-distribution<sup>24</sup>:

$$g(R) = AR^\alpha e^{-\eta R^\gamma}, \quad (5)$$

where  $A$  is the normalization constant;  $\alpha$ ,  $\eta$ , and  $\gamma$  are the distribution parameters, which for stratocumulus of C1 class are equal to<sup>24</sup>:  $\alpha = 6$ ,  $\eta = 1.5 \mu\text{m}^{-1}$ , and  $\gamma = 1$ . The characteristic concentration of water drops in such a cloud is  $N = 100 \text{ cm}^{-3}$ . Particles with the radii  $R = 2\text{--}10 \mu\text{m}$  dominate in such a cloud. The stratified-medium model contains a series of statistically independent aerosol screens located every 15 cm along the beam. At generation of random ensemble of particles at the screen the distribution function  $g(R)$  was replaced by the piecewise constant function of the particle radius  $R$  with the step of  $1 \mu\text{m}$ .

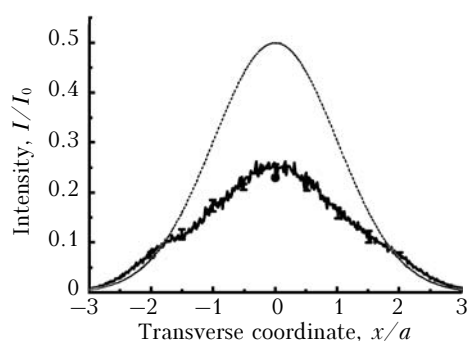


Fig. 5. The average intensity profile of laser beam with the radius  $a = 2.5 \text{ mm}$  at the scattering in stratocumulus.

We have considered the laser beam propagation in a polydisperse cloud medium along the path of the length  $z = 50 \text{ m}$ . The profile of the mean intensity of the beam, obtained by averaging over 10 statistically independent realizations of the series of aerosol screens, is shown in Fig. 5. The cross section of a light beam in a medium without particles is shown

by a dotted line. The intensity value on the beam axis, obtained from the experimental data,<sup>24</sup> is shown by a separate point at  $x = 0$ . We can see a good agreement between the numerically calculated and the experimental data. This example shows that the constructed model describes adequately the process of radiation scattering in a real disperse medium.

### Physical pattern of multifilamentation of pulses in aerosol

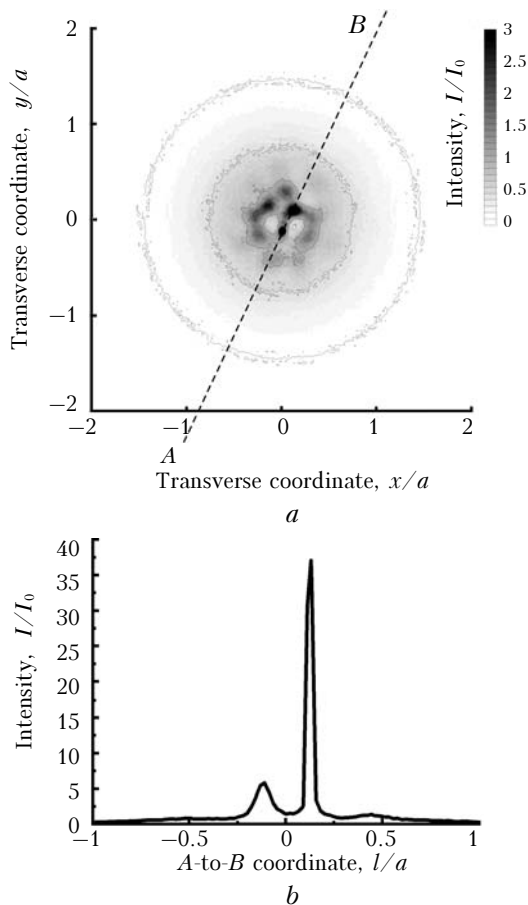
The origin of the filaments depends upon the Kerr self-focusing of pulsed layers, at which the spatial energy concentration occurs. The central pulse layer, containing peak power, is focused at the minimum distance, thus determining the filament initiation. To investigate the formation of filaments in the high-power femtosecond laser pulse, the stationary approximation is used.<sup>26</sup> In this approximation the dispersion of air is not taken into account, because this factor determines the pulse variation in time and its frequency spectrum, and does not affect the origin of filaments. Laser plasma is not considered as well, because its generation begins at the intensity exceeding  $10^{13}\text{--}10^{14} \text{ W/cm}^2$ , i.e., when a nonlinear focus is formed and its intensity increases by  $10\text{--}10^2$  times as compared with the initial one. The mathematical model of filament origin in a disperse medium describes the stationary self-focusing of a laser beam, which undergoes coherent scattering by the ensemble of aerosol particles. In this case, the intensity of a beam coincides with the intensity of central pulsed layer determining the filament origin.

According to the accepted approximations the stratified-medium model of the filamentation in a disperse medium comprises a series of layers, each layer containing two screens: aerosol and nonlinear. Such a model enables us to show the nonlinear transformation of the pulse field with the intensity perturbations, occurring at scattering by particles and diffraction in the medium.

To analyze the process of filament generation, we considered the Gaussian beam of radius  $a = 2.5 \text{ mm}$  at the wavelength  $\lambda = 0.8 \mu\text{m}$ . The initial beam power exceeded by a factor of  $10^2$  the critical power of self-focusing  $P_{cr}$  in air, which was  $P_{cr} \sim 2 \cdot 10^2 \text{ W}$  (Ref. 7). The parameters of the disperse medium correspond to the stratocumulus with the particle concentration  $N = 100 \text{ cm}^{-3}$  and the particle size distribution (5). The length of propagation was  $2 \text{ m}$ , that corresponds to  $0.04$  of the beam diffraction length. At this distance, the stratified-medium model assumes 50 layers with statistically independent aerosol screens.

Figure 6 shows a separate realization of the intensity in the central pulse layer at the distance  $z = 2 \text{ m}$ . In the gray-scale picture of the intensity distribution pattern we can see, in the plane of cross section (Fig. 6a), several randomly located "hot" spots, in which the intensity exceeds the initial maximum value  $I_0$ . It should be noted that in the absence of

perturbations in the pulse one filament on the axis is formed. The above experiment has shown that in the presence of drops the formation of several strong maxima takes place.



**Fig. 6.** The formation of filaments in the central pulsed layer with the peak power  $P_0 = 100P_{cr}$  in propagating at the distance  $z \approx 2$  m in the stratocumulus with the particle concentration  $N = 100 \text{ cm}^{-3}$ . The intensity distribution  $I(x, y)$  in the plane of the pulse cross section (a); the intensity profile in the cross section  $AB$  passing through two “hottest” spots in a beam (b).

Figure 6b shows the intensity profile along a straight line  $AB$  passing through two most “hot” spots. The intensity maximum in the left “hot” spot exceeds the initial value  $I_0$  by more than 5 times, and in the right “hot” one by more than 30 times. Thus, in spite of random distribution of drops-scatterers, over the disperse medium the Kerr self-focusing in air forms the perturbations at a relatively short distance, in which the intensity exceeds the initial one by several tens of times.

The obtained intensity distribution  $I(x, y)$  enables one to assess the radiation power  $P_{hs}$  at “hot” spots of the beam. According to the assessments made the power in the left “hot” spot is  $P_{hs} \sim 9P_{cr}$ , in the right spot the power  $P_{hs} \sim 11P_{cr}$ . Because the power within the beam perturbations significantly exceeds the critical power of self-focusing in air  $P_{cr}$ , one can

expect that in laser pulses of terawatt power the intensity maxima become centers of filament formation randomly located in space.

## Conclusions

The stratified-medium model of coherent scattering of high-power laser radiation has been developed using the water aerosol particle ensemble as an example. The model is based on the presentation of aerosol medium in the form of a series of layers, in which particles are grouped in thin aerosol screens. Among the aerosol screens of the model the diffraction and nonlinear interaction of radiation with the medium take place. The model enables one to calculate the laser radiation field at its propagation in mono- and polydisperse clouds.

Analysis of test problems of laser beam propagation through mono- and polydisperse aerosol shows that the proposed model adequately describes the scattering by an ensemble of particles.

It is shown that the occurrence of maxima in the laser beam intensity distribution at coherent radiation scattering by aerosol particles can result in the generation of multiple random filaments at high-power laser pulse propagation through the disperse medium.

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