PROPAGATION OF A DIFFRACTING LIGHT BEAM IN A MOVING NONLINEARLY ABSORBING AND CHEMICALLY ACTIVE MIXTURE OF GASES

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We study the propagation pattern of a diffracting optical beam in a moving nonlinearly absorbing and chemically active gas mixture. The case of absorption of light energy by a reaction product is analyzed. It is demonstrated that the dependence of concentration of the reaction product on the input beam intensity is bistable in character, as in stationary medium. This property of interaction manifests itself through the formation of a high-absorption region. The absorption of light energy by the reaction product intensifies the effect of thermal defocusing with increasing shift of the beam center counter to the direction of motion of the medium. The intensity distribution over the beam cross section, which is initially Gaussian, may be transformed into the distribution with two maxima upon exiting the medium.

1. INTRODUCTION

Stimulation of chemical gas mixtures in a flow reactor is of practical interest in connection with the feasibility of efficient and technically simple conveyance and disposal of mixture components. The other known application is connected with the feasibility of control of chemical reactions in the atmosphere. Therefore, the study of interaction patterns of an optical beam with a chemically active mixture in the presence of transverse motion of the medium is an urgent problem. However, its analysis taking into account the diffraction of optical radiation is absent in the literature. It has been just this paper which is devoted to the study of this problem.

It should be emphasized that, in contrast to the reactor with closed mass exchange, the temperature and concentration profiles (and, as a result, the beam intensity profile) become asymmetric in the case of transverse motion of the medium. The reaction product is carried out from the region occupied by the beam. As a result, in the case of weak thermal blooming of a beam due to its diffraction, the optical radiation is reflected on the amplitude grating induced by the optical radiation. This process is similar to the well-studied process of propagation of light beams and pulses in a cloudy medium (see, for instance, Refs. 1–6). However, there are significant distinctions.

First, for the interaction under study here, the concentration of the absorbing component of the mixture increases, because the optical radiation is absorbed by the reaction product, as a rule. However, it decreases in a cloudy medium. Therefore, the direction of shift of the beam energy center, when the beam propagates in a chemically active medium and its energy is absorbed by the reaction product, is opposite in sign to that of the optical radiation in the case of clearing of clouds and fogs.

Second, it is important to emphasize that for the analyzed case of interaction between the laser beam and the medium, the energy center shift caused by the concentration (amplitude) lens and the shift due to beam defocusing are of the same sign and directed counter to the motion of the medium. Therefore, these two mechanisms of nonlinearity intensify each other. Self-focusing of the beam causes the shift of its center in the opposite direction.

Third, upon exposure to a wide-aperture light beam in a chemically active gas mixture (neglecting thermal blooming), optical bistability (OB)[7,8] can occur under certain conditions. This means that two stable states of the medium and, consequently, two values of temperature and concentration of the reaction product correspond to one input value of the beam intensity. Bistability leads to the formation of regions of high absorption (explosive absorption): at a certain moment the absorption sharply increases, and then it does not vary remaining equal to the new high value. This also fundamentally changes the character of the considered interaction as compared with the propagation of optical beams in a water-droplet medium.

Note that the salient features of OB realization in narrow-temperature (diffracting) beams for a stationary medium were analyzed, in particular, in Ref. 9, where by computer simulation based on the difference schemes constructed in Refs. 10 and 11 it was demonstrated that the diffraction of optical radiation qualitatively changes the process of formation of high-absorption regions and may lead to such phenomena as moving and jumping focuses of the beam. Continuing this study in the present paper, we consider the propagation of laser beams in a moving mixture of gases under conditions of a running reversible chemical reaction.

2. PRINCIPLE EQUATIONS

The process of thermal blooming of slit diffracting beams in a moving medium, where a reversible chemical reaction runs and the light energy is absorbed by a reaction product, is described by the following system of dimensionless equations[9]:

$$\frac{\partial A}{\partial z} + iD\,\Delta_\perp A + i\varepsilon_{\rm nl} A + \delta_0 NA = 0,$$

$$0 < z \le L_z\,;$$

$$\varepsilon_{\rm nl} = \alpha(T - T_0) + \beta(N - N_0),\ \Delta_\perp = \frac{\partial^2}{\partial x^2}\,,$$

$$0 < x \le L_x\,;$$

$$\frac{\partial T}{\partial t} + V\frac{\partial T}{\partial x} = qNI + \gamma f, \quad I = |A|^2; \qquad (1)$$

$$\frac{\partial N}{\partial t} + V\frac{\partial N}{\partial x} = f\,,$$

$$f = (1 - N)\exp(-1/T) - kN\exp(-T_c/T)$$

with the initial conditions

$$A\,|_{z=0} = \exp(-(x - L_x/2)^2\,(1 + iF)); \qquad (2)$$

$$T\,|_{t=0} = T\,|_{x=0} = T_0;\ f\,|_{x=0} = f\,|_{t=0} = 0;$$

$$A\,|_{x=0,L_x} = 0.$$

Here, z is the longitudinal coordinate scaled to the diffraction length $l_{\rm d} = 2ka^2$; k is the wave number; a is the characteristic radius of the beam upon entering the nonlinear medium; x is the transverse coordinate measured in units of a. Note that the parameter D is remained in Eq. (1) for convenience of numerical experiments illustrating the influence of beam diffraction on the characteristics of beam propagation. We recall that the values $D \ll 1$ correspond to wide-aperture beams for which diffraction can be neglected. The factor δ_0 characterizes the absorption of optical radiation on the diffraction length; q is the beam power; γ is the thermal effect of the reaction proportional to the term $(T_c - 1)$; T_c is the ratio of activation energy of the reverse reaction to that of the direct one; k is the ratio of the reverse reaction rate to that of the direct one. The parameter V equals to the ratio of medium displacement during the characteristic time of the reaction to the beam radius. Thermal blooming of optical beams is described by the term with $\varepsilon_{\rm nl}$, in which α is the ratio of the beam input power to the characteristic power of thermal blooming, and the factor β is proportional to the difference between the refractive indices of the initial substance and the reaction product; T is the dimensionless temperature of the medium; T_0 is its equilibrium value; N is concentration of the reaction product scaled to its maximum value attainable under the given conditions; N_0 is its initial value; the parameter F specifies the light beam focusing. The parameters L_x and L_z are the transverse and longitudinal dimensions of the region considered.

As for the equation of chemical kinetics (third equation in system (1)), it should be noted that it is written for the reaction $A + B \leftrightarrow q$ under assumption that the concentration of one of the components (for instance, the substance B) is much higher than the concentration of another component. It should be emphasized that the change of the equation for concentration of the reaction product has no significant effect on the propagation pattern analyzed here. For instance, in case of replacement of $(1 - N)$ with $(1 - N)^2$, which corresponds to the dissociation reaction $A \leftrightarrow 2q$, as was mentioned in our earlier papers, bistable dependence of the reaction product concentration and the temperature of the medium on the input beam intensity is observed for a wider set of interaction parameters. In this paper, we restrict ourselves to the examination of a chemical process written in Eq. (1).

In addition, we note that for the studied interaction the heat transfer is realized due to removing heat and the reaction product from the region occupied by the beam owing to the motion of the medium. This mechanism of heat redistribution predominates, when the transit time of the medium across the beam is much less than the characteristic transit time of particles of the medium due to diffusion and heat conductivity.

Our numerical experiments were performed, in particular, for the following values of parameters:

$$q = 50,\quad \delta_0 = 8,\quad \gamma = 0,\quad T_0 = 0.125,\quad T_c = 0.375,$$
$$k = 0.5,\quad L_x = 10,\quad \text{and}\quad L_z = 0.5, \qquad (3)$$

which correspond to OB in the stationary medium.[9]

Simulating the interaction, we are interested not only in spatiotemporal distribution of the beam intensity and concentration of the reaction product, but also in the following integral and local characteristics of the medium and optical beam:

– integral yield of the reaction product

$$J_c(t) = \int_0^{L_z} {\rm d}z \int_0^{L_x} N(z, x, t)\,{\rm d}x\,; \qquad (4)$$

– radius of the optical beam determined through the second moment of intensity distribution

$$\alpha_x^2 = \int_0^{L_x} (x - L_x/2)^2 \, |A(z, x, t)|^2 \, dx/P(z, t), \qquad (5)$$

$$P(z, t) = \int_0^{L_x} |A(z, x, t)|^2 \, dx \; ;$$

– shift of the energy center of the beam with respect to its initial center

$$X_c(z, t) = \int_0^{L_x} (x - L_x/2) \, |A(z, x, t)|^2 \, dx/P(z, t) \, ; \qquad (6)$$

– maximum intensity in a given cross section z, the time t, and the coordinate X_m of the intensity maximum

$$I_m = \max_x |A(z, x, t)|^2 \, . \qquad (7)$$

The study of the three last characteristics is caused by the shift of the beam center and its maximum intensity in the process of interaction of optical radiation with the moving chemically active medium.

3. RESULTS OF NUMERICAL EXPERIMENTS

Let us consider the following parameters as functions of the speed of medium: the integral yield of the reaction, the time t_{st} of reaching the stationary state, the time t_{expl} of beginning of the explosive absorption near the cross section $z = 0$, the value of the transverse coordinate x_{cr} at which high concentration of the reaction product is reached (the system chemical medium – laser radiation is in the upper state), and the longitudinal coordinates at which high concentration is realized. The results of calculations are presented in Table I for $F = \alpha = \beta = 0$, and $D = 1$.

TABLE I. Time t_{expl} of beginning of the explosive absorption, time t_{st} of reaching the stationary state, longitudinal z_{cr} and transverse x_{cr} dimensions of the region of efficient reaction, and integral yield J_c of the reaction product as functions of the speed of particles V of the medium upon exposure to a wide-aperture beam (D = 1).

V	t_{expl}	t_{st}	$\cdot_{cr}$	z_{cr}	J_c
0.5	0	10	3.9	0.5	0.18
1	0	5	4	0.45	0.16
5	0.5	10	4.2	0.3	0.11
10	2	20	4.5	0.25	0.07

As expected, the region of efficient reaction decreases with increasing speed V at the fixed input power. The time of beginning of the explosive absorption increases in this case. The time of reaching the stationary distribution of concentration of the product first decreases and only then increases. The dynamics of the integral yield of the reaction product is shown in Fig. 1.

We emphasize that evolution of $J_c(t)$ is in fact independent of the diffraction of optical radiation: the functions shown in Fig. 1 do not vary when the coefficient D decreases, for instance, down to 10^{-3}. This conclusion also follows from the invariant under the integral absorbed light power,[9] which is independent of the spatial distribution of intensity and is determined only by the initial power of the beam. As known, the diffraction of optical radiation only redistributes the light energy along the transverse coordinate. However, it should be emphasized that for a medium of finite optical thickness the situation can be imagined when, due to absorption saturation, the parameters $J_c(t)$ for focused beams differ from those for initially collimated beams. As an illustration, Fig. 1 shows the values of integral yield of the reaction product for the focused beam with $F = 1$ and 3.

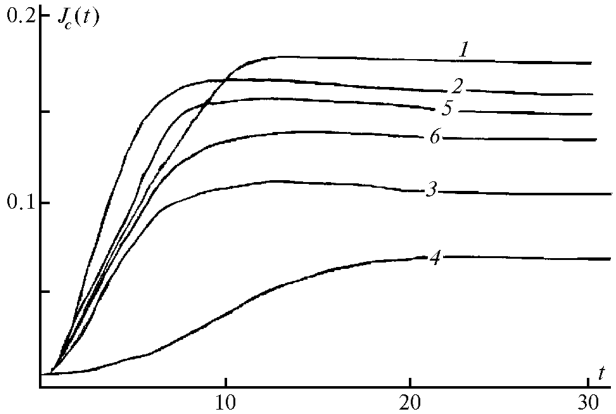

FIG. 1. Evolution of the integral yield of the reaction product for the following values of parameters: $D = 1$; $\alpha = \beta = F = 0$; $V = 0.5$ (1), 1 (2), 5 (3), 10 (4); $V = 1$, $F = 1$ (5), 3 (6).

Diffraction of the optical beam leads to considerable transformation of its intensity profile as compared with the case of propagation of wide-aperture beams, when the beam either remains Gaussian or has a dip in the region of high absorption. A diffracting beam shifts counter to the direction of transverse motion of the medium (Fig. 2) even in the absence of phase grating (purely absorbing medium), similarly to its propagation in a cloudy medium.[4]

However, the direction of shift changes. If perturbations of permittivity due to changes in mixture composition or its heating are significant, the beam acquires asymmetric shape and may even break into two subbeams (Fig. 2b). In addition, it is important to emphasize that two independent subbeams may occur upon exiting the medium under conditions of thermal blooming. One of them (with lower intensity) is practically in the center of the initial beam. In the case of defocusing due to changes in mixture composition, such a structure of the intensity profile is formed in a cross section of the medium nearer to its input. However, the intensity between two maxima does not vanish.

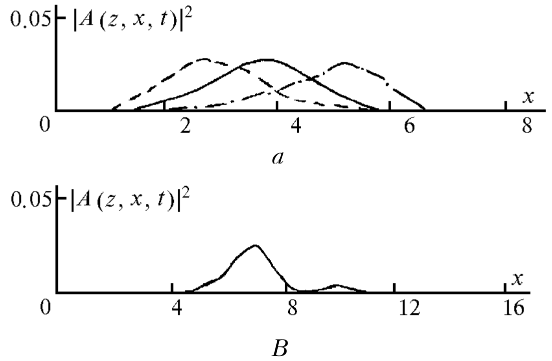

FIG. 2. Distribution of the light beam intensity at $t = 30$ *over the beam cross section* $z = 0.5$ *for* $V = 1$, $F = 0$, $D = 1$ *with* $\alpha = \beta = 0$ *(solid curve);* $\alpha = 0$, $\beta = -10$ *(dashed curve);* $\alpha = 0$, $\beta = 10$ *(dot-and-dash curve) (a);* $\alpha = -2$, $\beta = 0$ *(B).*

As was stated above, the interaction under consideration has the property of optical bistability. In a distributed medium, OB manifests itself through the formation of high-absorption regions. For instance, a region with high concentration of the reaction product and its low concentration can be formed along the z coordinate under certain conditions (for $z \le z_{\rm cr}$). Because it determines the absorption coefficient, its values significantly differ before and after $z_{\rm cr}$. As a result, intensity of optical radiation as a function of the longitudinal coordinate has two parts of evolution, rapid ($z \le z_{\rm cr}$) and slow decrease. To illustrate this, stationary variation of maximum intensity of a wide-aperture beam along z is shown in Fig. 3 by the solid curve.

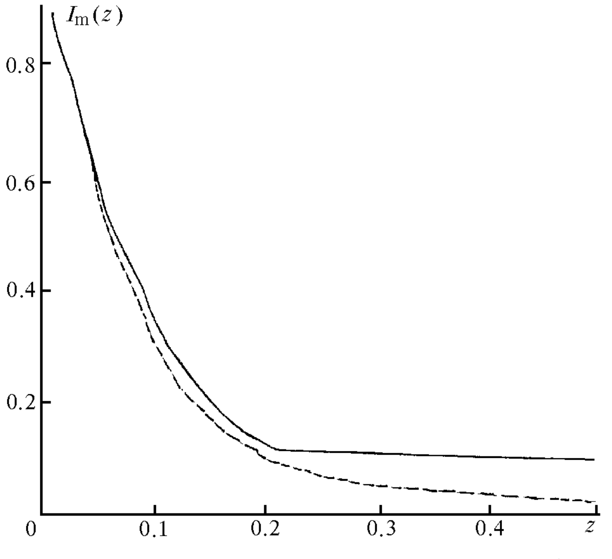

FIG. 3. Longitudinal stationary distribution of the maximum intensity upon exposure to wide-aperture beams for $D = 0.001$ *(solid curve) and* 1 *(dashed curve),* $\alpha = \beta = 0$, *and* $V = 1$.

The boundary of transition from the region of high concentration of the reaction product to the region of low efficiency of reaction is blurred upon exposure to narrow-aperture beams (see Fig. 3, dashed curve). In this case, the absorption coefficient varies more smoothly near the boundary of the two regions. This leads to smoother variation of the intensity along the z axis as compared with the case of wide-aperture beams. It is evident that the decrease of maximum intensity of the beam in the region of weak the reaction ($z \ge z_{\rm cr}$) is caused by beam diffraction.

Thermal blooming of optical radiation can significantly change the longitudinal distribution of the region of efficient reaction. For instance, Fig. 4 shows the concentration of the reaction product along z in the cross section . $= L_x/2$ which is the initial axis of the beam.

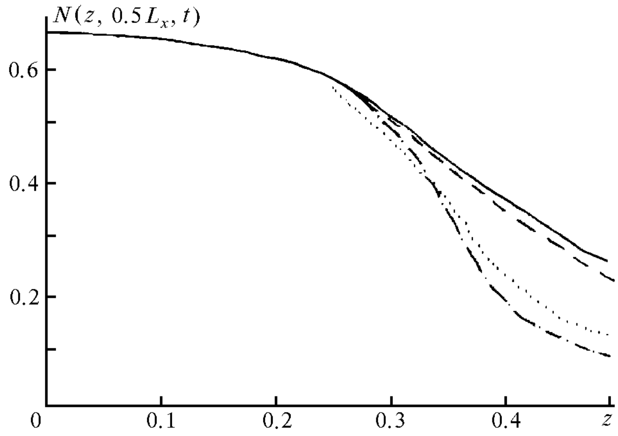

FIG. 4. Longitudinal distribution of concentration of the reaction product at $t = 30$ *for* $V = 1$, $F = 0$, $D = 1$ *with* $\alpha = \beta = 0$ *(solid curve);* $\alpha = 0$, $\beta = -10$ *(dashed curve);* $\alpha = 0$, $\beta = 10$ *(dot-and-dash curve);* $\alpha = -2$, $\beta = 0$ *(dotted curve).*

Comparison of the functions shown in Fig. 4 demonstrates that thermal blooming always decreases the region of high concentration of the reaction product at the beam axis and leads, in contrast to diffraction, to the formation of a more contrast variation structure of the reaction product concentration along the z coordinate. At the same time, for thermal defocusing, concentration value is even somewhat higher upon exiting the medium as compared with the case of self-focusing due to changes in the mixture composition.

During stimulation of a chemical reaction by optical radiation, the beam intensity profile transforms into a structure with an intensity dip at the axis. As a result, the integral broadening of the beam is determined by contribution of profile transformation, its diffraction, and thermal blooming. To highlight the contribution of each of them into the integral radius, Table II presents relative broadening of the beam upon exiting the medium as functions of the diffraction coefficient, focusing, and thermal blooming of optical radiation.

It is important to emphasize that the transverse radius of the beam is more than twice larger even for a weakly diffracting beam ($D = 0.001$). Such an increase is caused, first, by the decrease of light energy in the central part of the beam; second, by tubular structure of the beam profile formed near the input cross section of the cell at the initial stage of formation of the region with high concentration of the reaction product due to higher absorption near the initial maximum of the intensity. As a consequence, we can observe the decrease of diffraction length for subbeams, and then they acquire the Gaussian profile. The path along which this occurs depends on subbeams radii and contrast of the intensity profile with a dip at its axis. A comparison of the results demonstrates that thermal defocusing brings the greatest contribution to the increase of the beam radius. As a result of its effect, the beam radius increases almost five times, which is also connected with its breaking into separate subbeams.

TABLE II. The square of relative broadening of the beam $\eta = a_x^2\,(z = 0.5,\ t = \infty)/a_x^2\,(z = 0.5,\ t = 0)$ *as a function of beam thermal blooming and focusing parameters for* $V = 1$.

D	α	β	F	η
0.001	0	0	0	5
1.0	0	0	0	5
1.0	0	0	1	7
1.0	0	0	3	8
1.0	−2	0	0	23
1.0	0	−1	0	14
1.0	0	10	0	3

Let us turn to an analysis of the position of the beam energy center upon exiting the medium as a function of diffraction, thermal blooming, and focusing (Table III, Fig. 5).

TABLE III. Shift of the energy center of the beam X_c *and coordinate* X_m *of the intensity maximum from their positions upon entering the medium and the value of* I_m *in the cross section* $z = 0.5$ *at time* $t = 30$ *(stationary distribution) as functions of the diffraction, thermal blooming, and focusing coefficients of the beam for* $V = 1$. *In the case* $\alpha = -2$, *the size of the region in the transverse direction is* $L_x = -20$.

D	α	β	F	X_c	X_m	I_m
0.001	0	0	0	−0.8	−0.9	0.1
1	0	0	0	−1.2	−1.3	0.03
1	0	0	1	−0.7	−0.8	0.04
1	0	0	3	−1.1	−1.4	0.02
1	−2	0	0	−2.7	−2.9	0.025
1	0	−10	0	−1.7	−1.6	0.028
1	0	10	0	−0.12	0.2	0.025

Note that the shift of the center of gravity of optical beam and the coordinates of the intensity maximum can significantly differ due to the competition between different thermal blooming mechanisms (amplitude and phase gratings). This can be easily seen in Fig. 2. First of all, let us note that the center of the beam shifts nonuniformly in z counter to the direction of motion of the medium even for purely amplitude gratings with $D = 0.001$ (see Fig. 5, solid curve). This emphasizes the influence of diffraction even for its small values because of formation of subbeam structure. With increase of the diffraction coefficient up to $D = 1$, the evolution of position of the beam center remains similar to the previous case: shift of X_c increases approximately as z^2 with increase of z (see Fig. 5, dashed curve).

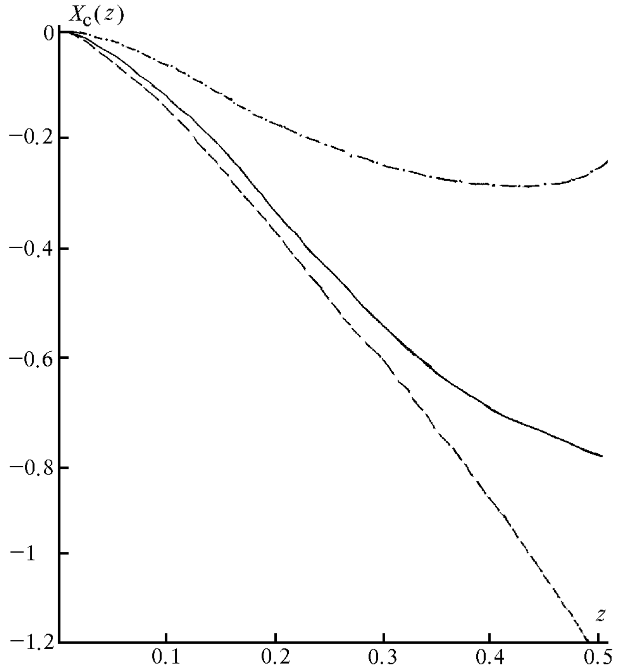

FIG. 5. Evolution of the stationary position of the integral center of gravity of the beam along the longitudinal coordinate for $V = 1$, $F = 0$ *with* $\alpha = \beta = 0$, $D = 0.001$ *(solid curve),* 1 *(dashed curve),* $D = 1$, $\alpha = 0$, $\beta = -10$ *(dot-and-dash curve).*

The beam shift can be reduced by initial focusing of the beam. At the same time, for a certain part of the propagation path, its center position can be constant, for instance, when $F = 3$, X_c does not vary until $z = 0.13$. Defocusing of optical radiation increases the shift of the beam center counter to the medium, because the effect of a distributed lens caused by defocusing of the amplitude grating induced due to concentration variation of the reaction product coincides in the sign of light beam deflection. At self-focusing of optical beam with time, its center begins to shift in the direction of motion of the medium at the end of the medium. As a result, its value in the stable mode is halved as compared with the initial moment of light pulse propagation. The maximum value of X_c for $\beta = 10$ is reached within the thickness of the medium: the beam first shifts counter to the direction of motion of the gas mixture and then in the direction of motion (see Fig. 5, dot-and-dash curve).

The coordinate of the maximum intensity also varies in a similar way (see Table III, Fig. 6). In contrast to the function X_c, it reaches higher values of transverse shift of the intensity maximum and changes the direction of shift of peak intensity in the case of beam self-focusing due to change in gas mixture composition.

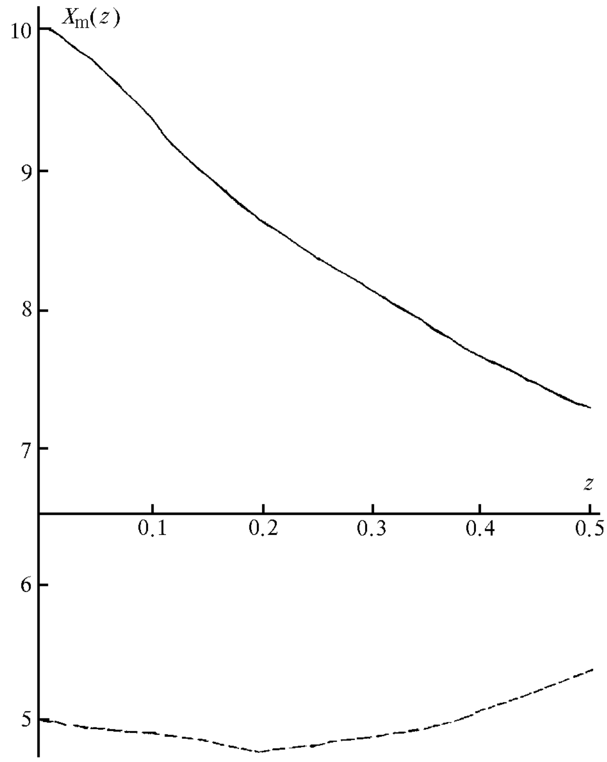

FIG. 6. Evolution of the longitudinal coordinate of stationary maximum intensity of the beam for $V = 1$, $F = 0$ *and* $D = 1$ *with* $\alpha = -2$, $\beta = 0$ *(solid curve);* $\alpha = 0$, $\beta = 10$ *(dashed curve).*

Summarizing this paper, we can conclude that optical bistability may occur in the flow chemical reactor (e.g., in the atmosphere) when light energy is absorbed by a product of a chemical reaction. Bistability manifests itself through the regions of high and low absorption. Diffraction of optical radiation fundamentally changes the evolution of the spatial beam profile as compared with the case of wide-aperture beams. This is manifested through breaking of an initially Gaussian beam into subbeams. In addition, diffraction reduces the contrast of concentration in the regions of high and low absorption.

In this paper we have analyzed thermal blooming of a light beam in a chemically active gas mixture with absorption of light energy by the reaction product. In contrast to the propagation of optical radiation in a cloudy medium, the transverse shift of the beam due to thermal defocusing and shift of the beam center caused by refraction at the amplitude grating intensify each other due to absorption.

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