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Photochemical Waves in Gases

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Processes involving the propagation of quasimonochromatic radiation in a binary gas medium in which chemical chain reactions may occur (according to the Nernst scheme) are investigated. It is shown that two modes of photochemical wave propagation may occur, depending on the ratio of the initial reagent densities, photodissociation rates, and rates of elementary stages of the chain reaction. The ranges of physical parameters to which these regimes of propagation correspond are found. The profile and velocity of the photochemical wave are calculated for each case.

1. INTRODUCTION

WITH further development of lasers (increase of the directed-radiation power, extension of the frequency band), there is considerable interest in problems connected with the qualitative changes initiated by stimulated laser radiation in the physical or chemical properties of a medium. In particular, a timely and interesting problem is that of the action of electromagnetic radiation on intermolecular processes such as non-quilibrium excitation of internal degrees of freedom of molecular formations, realignment of internal bonds of molecules, etc.[1-7]. Processes of this kind lead to a perturbed state of the medium. If this state is "transparent" to an external radiation flux, then this flux, in principle, can ensure displacement of the perturbed state through the medium. In the case of continuous radiation, the displacement will have a wave-like character, and the propagation velocity of the perturbation depends on the mechanism of the interaction of the radiation with the medium. The process of the propagation of photodissociation waves in gas media was investigated theoretically in[8,9]. It follows from these papers that the character of the propagation of such waves is analogous to the character of propagation of bleaching waves in two-level media[10,11] whose absorption coefficients depend in nonlinear fashion on the intensity of the incident monochromatic radiation. It was established in[8,9] that the velocity of the photodissociation wave is determined by the relation $D = cI_0/(I_0 + cN_0)$, where I_0 is the radiation flux density, N_0 is the initial density of the reagent that dissociates in the photolysis process, and c the velocity of light in the medium. It is of interest to note that in certain real cases the velocity of the photodissociation waves can reach subluminal values[9].

We have previously studied[12,13] photodissociation waves the propagation of which is accompanied by elementary acts of chemical reactions. The initial gas considered was a binary mixture $A_2 - B_2$ in which the transformations occur by a chain mechanism in accord with the Nernst scheme (see[12]). In[12], principal attention was paid to the fundamental aspects of the questions connected with the propagation of photochemical waves. The performed analysis pertains to a particular situation, namely, an excess concentration of the photodissociating reagent, and an excess of the rate constant of the secondary link of the chain over the rate constant of the primary link $(k_1 \ll k_2)$. The purpose of the present study was to investigate the possible stationary wave-like regimes that can be realized for an initial binary gas mixture, and the conditions for realization of these regimes, which must be satisfied by the density of its chemically-active reagents and by the constants characterizing the kinetic processes.

2. PHOTOCHEMICAL WAVES IN THE CASE OF "RAPID" FIRST LINK OF THE CHAIN REACTION $(k_1 \gg k_2)$

If the frequency ν_0 of the quasimonochromatic radiation lies in the photoabsorption band of the reagent A_2 and if a chain reaction is possible in the binary gas medium (it is assumed that the absorption bands of the initial reagents and of the reaction products do not overlap), then the propagation of the photochemical wave can be described by a system of equations consisting of the radiation transfer equation and the equations describing the chemical kinetics of the chain reaction with photoinitiation[12]. An examination of the initial scheme of the reaction shows that after passage of the wave the chemical reactions in the medium can terminate if either all the active centers A[1] or the entire reagent B_2 are consumed. We shall say that the first or second regime of wave propagation takes place if the chemical reactions are terminated because of a shortage of the reagent A or B_2. Taking this circumstance into account and using integral relations that are valid for stationary waves[12], we have for the density of the corresponding reagents behind the wave front $(x' = x - Dt = -\infty)$ in the case of the first regime

$$(A)_- = 0, \quad (B)_- = 2I_0\left(\frac{1}{D} - \frac{1}{c}\right), \quad (A_2)_- = 0,$$

$$(B_2)_- = -I_0\left(\frac{1}{D} - \frac{1}{c}\right) + (B_2^0) - (A_2^0) \geqslant 0; \tag{2.1}$$

and in the case of the second regime

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[1] Since recombination processes are not considered here, it is obvious that the density of the photodissociating reagent A_2 should vanish after the passage of the wave.

$$(A)_- = I_0\left(\frac{1}{D}-\frac{1}{c}\right)+(A_2{}^0)-(B_2{}^0)\geqslant 0,$$

$$(B_2)_- = 0,\quad (B)_- = I_0\left(\frac{1}{D}-\frac{1}{c}\right)+(B_2{}^0)-(A_2{}^0)\geqslant 0,\quad (A_2)_- = 0, \tag{2.2}$$

where I_0 is the intensity of the incident radiation, which has a photoabsorption cross section σ, (A_2^0) and (B_2^0) are the initial densities of the reagents, c is the velocity of light in the medium, and D is the wave propagation velocity. Inequalities resulting simply from the fact that the densities cannot be negative impose limitations on the wave propagation velocity, namely:

1) the velocity of the wave in which the reaction terminates because of shortage of the active centers A satisfies the condition

$$D > c\Big/\left[1+c\frac{(B_2{}^0)-(A_2{}^0)}{I_0}\right]; \tag{2.3}$$

2) the velocity of the wave in which the reaction terminates because of the shortage of the molecules B_2 should satisfy the condition

$$D < c\Big/\left[1+c\frac{|(B_2{}^0)-(A_2{}^0)|}{I_0}\right]. \tag{2.4}$$

It is clear from physical considerations that the wave propagation velocity increases with increasing rates of the chemical reaction[12,13]. If the second wave propagation regime obtains, then, as seen from (2.4), there exists a limiting velocity, which corresponds to propagation of a photodissociation wave over the products of the instantaneously occurring chemical reactions. In the case of the first regime, the increase of the rates of the chemical reactions can lead, in principle, to subluminal velocities of wave propagation.

The solution of the system describing the stationary wave becomes much simpler if it is assumed that the rate of one of the links of the chemical chain reaction greatly exceeds the rate of the other link. In a number of cases of practical importance (e.g., for the gas mixture $H_2 - Cl_2$ or $H_2 - F_2$) this condition is satisfied. In this part of the paper, we consider the case when the first link of the chain reaction is rapid ($k_1 \gg k_2$). Introducing the dimensionless quantities

$$a_2=\frac{(A_2)}{(A_2{}^0)},\quad a=\frac{(A)}{(A_2{}^0)},\quad \alpha=\frac{\sigma}{k_2}\left(\frac{1}{D}-\frac{1}{c}\right)^{-1},$$

$$u=\frac{I}{(A_2{}^0)}\left(\frac{1}{D}-\frac{1}{c}\right),$$

$$\eta=x'\frac{k_2(A_2{}^0)}{D},\quad \Delta=\frac{(B_2{}^0)}{(A_2{}^0)}-1,\quad \varepsilon=\frac{k_2}{k_1}\ll 1,$$

we obtain the following system of equations:

$$\frac{du}{d\eta}=-\alpha a_2 u,\quad \frac{da_2}{d\eta}=\alpha a_2 u+a_2(2u-a),$$
$$\varepsilon\frac{da}{d\eta}=a(\Delta+a_2-u+a)-\varepsilon a_2(2\alpha u+2u-a), \tag{2.5}$$

The smallness of the parameter ϵ makes it possible to break up the phase space $(u, a_2, a, \dot{u}, \dot{a}_2, \dot{a})$ into regions of "fast" (variation of a) and "slow" (variation of u and a_2) motions[14], so that in the zeroth approximation the variation of the variable a will be determined from the algebraic relation[2)]

$$0=a[a-(u-a_2-\Delta)]. \tag{2.6}$$

Since we are not considering processes accompanied by photon emission and by recovery of the reagent A_2, the function $f = u - a_2 - \Delta$ decreases monotonically with increasing η. Consequently, in order for the function f to vanish at a certain unique value $\eta = \eta_0$, it suffices to satisfy the following inequalities:

$$f(\eta=-\infty)>0,\quad f(\eta=+\infty)<0.$$

If the first regime of propagation of the photochemical wave takes place, then, using relation (2.1), we can show that $f < 0$ for all values of η. In this case we get from (2.6) the solution $a = 0$, and can thus determine the connection between the variables a_2 and u from the first two equations of (2.5):

$$a_2+(1+2/\alpha)u=\text{const.}$$

Using the conditions at $\eta = \pm\infty$, we obtain an expression for the wave propagation velocity:

$$D=c\left\{1+c\frac{\sigma}{4k_2}\left[\sqrt{1+\frac{8k_2(A_2{}^0)}{\sigma I_0}}-1\right]\right\}^{-1}. \tag{2.7}$$

Choosing the origin such that $u(\eta = 0) = u_2/2$, we can easily obtain for the profile of the photochemical wave in the case under consideration the following formulas:

$$u=\left[\frac{2+\alpha}{\alpha}+\left(\frac{2}{u_0}-\frac{2+\alpha}{\alpha}\right)e^{\alpha\eta}\right]^{-1},$$

$$a=0,\quad a_2=1-\left(1+\frac{2}{\alpha}\right)u,$$

$$b=2u,\quad b_2=\Delta+1-2\left(1+\frac{1}{\alpha}\right)u.$$

The variation of the reagent densities and of the radiation flux density is illustrated by Fig. 1, regime I.

We assume next that the second regime takes place. It is seen from (2.2) that in this case $f(\eta = +\infty) < 0$ and $f(\eta = -\infty) > 0$. To solve this system, it is convenient to choose as the origin the point at function f vanishes $f(0) = 0$. We then get from (2.6)

$$a=0 \text{ if } \eta>0,$$

$$a=u-a_2-\Delta \text{ if } \eta<0. \tag{2.8}$$

When (2.8) is taken into account, we can obtain the integrals of the initial system for the corresponding regions:

$$a_2=1-(1+2/\alpha)u \quad \text{if} \quad \eta>0,$$

$$a_2=(u_0+\Delta)u_0{}^{1/\alpha}u^{-1/\alpha}-\Lambda-u \quad \text{if} \quad \eta<0.$$

Joining the solutions at the zero point, we obtain an equation for the wave velocity:

$$\frac{1}{1+t}=\left(\frac{2}{\xi}t+1-r\right)\left[\frac{4}{\xi}t(1+t)\right]^t,$$

$$\xi=\frac{2k_2(B_2{}^0)}{\sigma I_0},\quad r=\frac{(A_2{}^0)}{(B_2{}^0)}\quad t=\frac{k_2}{\sigma}\left(\frac{1}{D}-\frac{1}{c}\right). \tag{2.9}$$

[2)] It is easy to prove the stability of the fast motions in accordance with the general theory[14].

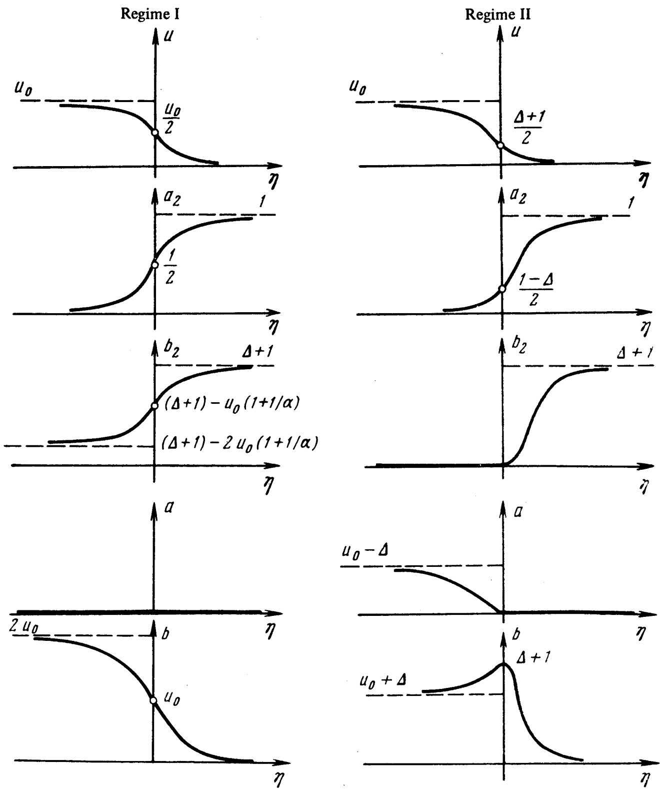


FIG. 1. Wave profiles for the case $k_1 \gg k_2$.

The profile of the photochemical wave corresponds in this case to the following expressions (see Fig. 1, regime II):

if $\eta \geq 0$

$$u = \left[\frac{2+\alpha}{\alpha} + \left(\frac{2}{u^0} - \frac{2+\alpha}{\alpha}\right)e^{\alpha\eta}\right]^{-1},$$

$$a = 0, \quad a_2 = 1 - (1 + 2/\alpha)u, \quad b = 2u, \quad b_2 = \Delta + 1 - 2(1 + 1/\alpha)u,$$

if $\eta \leq 0$

$$\eta = -\frac{1}{\alpha}\int_{u^0}^{u} dx\, \{x[(u_0 + \Delta)u_0^{1/\alpha}x^{-1/\alpha} - x - \Delta]\}^{-1};$$

$$a = 2u - (u_0 + \Delta)u_0^{1/\alpha}u^{-1/\alpha}, \qquad a_2 = (u_0 + \Delta)u_0^{1/\alpha}u^{-1/\alpha} - \Delta - u,$$

$$b = (u_0 + \Delta)u_0^{1/\alpha}u^{-1/\alpha}, \qquad b_2 = 0,$$

where

$$u^0 \equiv u(\eta = 0) = (\Delta + 1)/2.$$

The inequalities in (2.1) and (2.2) yield the necessary conditions for the realization of the first or second regime. These inequalities, in the new notation, take the form

$$t < {}^1\!/_2(1 - r)\xi \qquad \text{(first regime)},$$

$$t > {}^1\!/_2(1 - r)\xi, \quad t > {}^1\!/_2(r - 1)\xi \qquad \text{(second regime)}. \quad (2.10)$$

Relations (2.7) and (2.9), with allowance for (2.10), separate the physical-parameter regions for which the first and second regimes of wave propagation are realized. It is convenient to plot these regimes graphically on a plane in which the abscissas represent the ratio of double the rate of the chemical reaction to the photodissociation rate ($\xi = 2k_2(B_2^0)/\sigma I_2$), and the ordinates represent the ratio of the initial densities of the components of the binary gas ($r = (A_2^0)/(B_2^0)$). The plane breaks up into two regions (Fig. 2). If the problem parameters I_0, (A_2^0), (B_2^0), k_2, and σ are such that the point with the coordinates (ξ, r) belongs to region I, then the first wave-propagation regime is realized. The region II corresponds to the second regime. It can be shown that the boundary between the regions is the curve

$$r = 1 - \xi^{-1}[\sqrt{1+\xi} - 1].$$

3. PHOTOCHEMICAL WAVES IN THE CASE OF "FAST" SECOND LINK OF THE CHEMICAL REACTION ($k_2 \gg k_1$)

Let us consider the opposite limiting case, when the second link of the chemical reaction is the "fast" one. Since it can be shown that in this case the "fast" motions are stable, we shall solve the system in the zeroth approximation with respect to the small parameter $\mu = k_1/k_2 \ll 1$. The zeroth-approximation equations, which are analogous (2.5), take the form

$$\frac{du}{d\eta} = -\alpha u(b_2 + b - u + \Delta), \qquad \frac{db_2}{d\eta} = b_2(2u - b),$$

$$0 = b[b - (u - b_2 - \Delta)];$$

$$b_2 = \frac{(B_2)}{(B_2^0)}, \qquad b = \frac{(B)}{(B_2^0)}, \qquad \eta = x'\frac{k_1(B_2^0)}{D},$$

$$u = \frac{I}{(B_2^0)}\left(\frac{1}{D} - \frac{1}{c}\right), \quad \alpha = \frac{\sigma}{k_1}\left(\frac{1}{D} - \frac{1}{c}\right)^{-1}, \quad \Delta = \frac{(A_2^0)}{(B_2^0)} - 1. \tag{3.1}$$

Using relations (2.1) and (2.2), we can show that for both possible propagation regimes of the photochemical wave, the function $\varphi = u - b_2 - \Delta$ vanishes at a certain point $\eta = \eta_0$. Taking this circumstance into account, we get from the third equation of (3.1) for the density of the active centers B:

$$b = 0, \quad \eta \geqslant 0, \qquad b = u - b_2 - \Delta, \quad \eta \leqslant 0 \tag{3.2}$$

(the origin is chosen to be the point $\eta_0 = 0$).

With the aid of (3.2) we establish from the first two equations of the system (3.1) the connection between b_2 and u for the region $\eta \geq 0$:

$$b_2^{-\alpha/2}(u - \Delta) + \frac{\alpha}{2-\alpha}b_2^{-\alpha/2+1} = \text{const.} \tag{3.3}$$

From the known conditions at $\eta = 0$ and $\eta = +\infty$

$$u(0) = u_0, \quad b_2(0) = u_0 - \Delta,$$
$$u(+\infty) = 0, \quad b_2(+\infty) = 1,$$

with allowance for (3.3), we obtain an equation for the wave propagation velocity

$$\left(\frac{1}{\xi Y} + 1 - r\right)^{1-Y} = Yr + 1 - r, \tag{3.4}$$

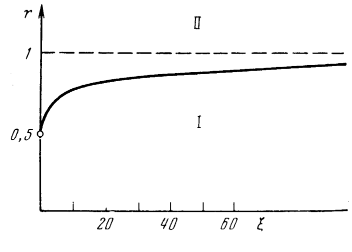


FIG. 2. Illustrating the determination of the propagation regimes of the photochemical wave ($k_1 \gg k_2$).

where

$$r=\frac{(A_2{}^0)}{(B_2{}^0)},\ Y=\frac{\sigma}{2k_1}\left(\frac{1}{D}-\frac{1}{c}\right)^{-1},\ \xi=\frac{2k_1(B_2{}^0)}{\sigma I_0}.$$

In analogy with the analysis given in Sec. 2, we obtain for the wave profile the following equations:

if $\eta \geq 0$

$$2\eta=\int\limits_{u_0-\Delta}^{b_2}\frac{dx}{x}\left[\left(\frac{\alpha}{2-\alpha}-\Delta\right)x^{\alpha/2}-\frac{\alpha}{2-\alpha}x+\Delta\right]^{-1},$$

$$u=\Delta+\left(\frac{\alpha}{2-\alpha}-\Delta\right)b_2{}^{\alpha/2}-\frac{\alpha}{2-\alpha}b_2,$$

$$b=0,\quad a=2u,\ a_2=\Delta+b_2-u; \tag{3.5a}$$

if $\eta \leq 0$

$$u=u_0,\ b_2=(u_0+\Delta)\Big/\left[\frac{2u_0}{u_0-\Delta}e^{-\eta(u_0+\Delta)}-1\right],$$

$$a=u_0+b_2+\Delta,\quad a_2=0,\quad b=u_0-b_2-\Delta. \tag{3.5b}$$

Figure 3 illustrates the changes of the corresponding quantities for both regimes on the wave front. Unlike in the case of Sec. 2, when inequality $k_2 \ll k_1$ is satisfied, the form of Eq. (3.4) for the wave velocity and the relations describing the profile (3.5) remain unchanged for both regimes. An investigation of the transcendental equation (3.4) is conveniently carried out graphically (see Fig. 4). The two points of intersection of the function in the right-hand and left-hand sides of (3.4) correspond to two roots of this equation. It can be noted that one of the roots has a value $Y_0 = 1$. In the general case, however, it does not correspond to any physical situation whatever. Indeed, this root would correspond to a wave propagating with velocity $D = 2k_1c/(2k_1 + \sigma c)$ at an arbitrary value of the rate constant of the slow link of the chain reaction. In the case $k_1 = 0$ this velocity should coincide with the velocity corresponding to ordinary photodissociation wave, i.e., with $D \approx I_0/(A_2^0)$[9], since there are no chemical reactions. At $k_1 = 0$, however, the root $Y_0 = 1$ corresponds to the value $D = 0$.

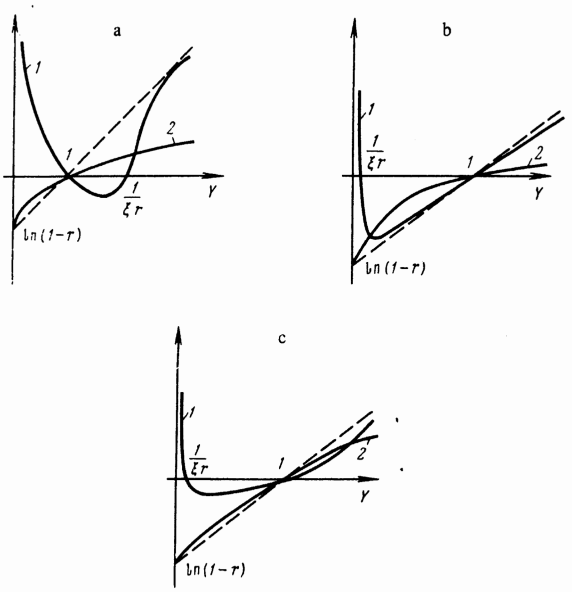


FIG. 4. Graphical investigation of Eq. (3.4) for the wave-propagation velocity ($k_1 \ll k_2$): 1) $f_1 = (1-Y)\ln(1/\xi Y + 1-r)$, 2) $f_2 = \ln(Yr + 1-r)$.

The velocity of the photochemical wave is determined by the second root Y_1 of Eq. (3.4). This root is obtained by numerical methods, and for the velocity we get

$$D=2k_1cY_1/(2k_1Y_1+c\sigma). \tag{3.6}$$

In the limiting case $k_1 \to 0$ (or $B_2^0) \to 0$) relation (3.6) goes over into the formula for the velocity of the ordinary photodissociation wave[9]

$$D=cI_0/(I_0+c(A_2{}^0)). \tag{3.7}$$

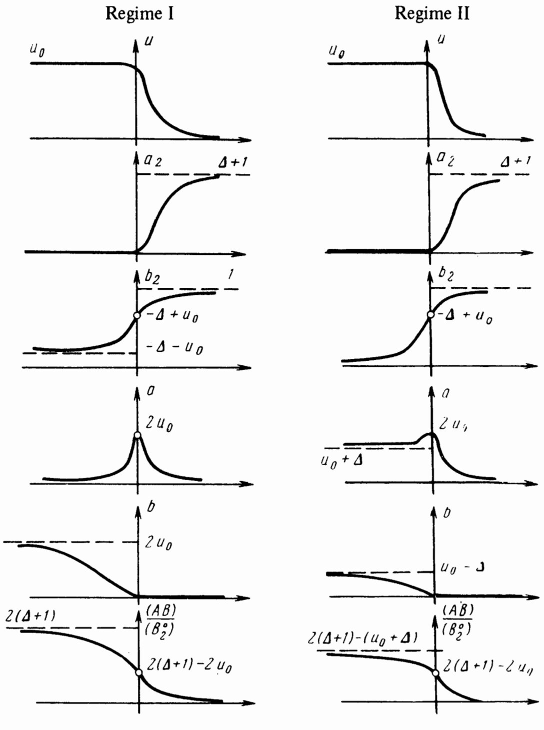
It is obvious that the root Y_0 has a physical meaning only in the unique case when $Y_0 = Y_1 = 1$ (this situation corresponds to tangency of curves 1 and 2 in Fig. 4). The roots coincide for the following relation between the system of parameters:

$$\xi=(e^{-r}+r-1)^{-1}.$$

In analogy with the case considered in Sec. 2, the plane of the physical parameters (ξ, r) breaks up into regions corresponding to the first and second regimes of the photochemical-wave propagation in the initial binary medium (see Fig. 5). The boundary between the regions is determined from the condition under which the boundary regime is realized, namely, at $\eta = -\infty$ both the density of the active centers A and the density of the reagent B_2 are equal to zero. Using this condition, we can obtain the relation for the curve separating the regions of the two regimes:

$$[2(1-r)]^{1-1/\xi(1-r)}=1-r+r/\xi(1-r). \tag{3.8}$$

The transcendental equation (3.8) is solved numerically, and the results of the calculation are shown graphically


Regime I

Regime II

FIG. 3. Wave profiles for the case $k_1 \ll k_2$.

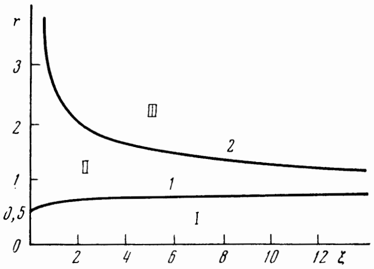


FIG. 5. Wave-propagation regimes for the case $k_1 \ll k_2$: 1) numerical solution of Eq. (3.8), 2) $\xi = r/(r-1)^2$.

(see Fig. 5). Region II, which lies above the curve (3.8) corresponds to the second regime, and region I corresponds to the first regime.

In concluding this section, we note the following singularity connected with the approximation within the framework of which the analysis was carried out. It follows from relations (3.4) and (2.6) that the only roots with physical meaning are those satisfying the inequalities

$$1 - 1/r < Y < 1/\xi(r-1), \qquad (3.9)$$

and this imposes the following condition on the system parameters:

$$1 - 1/r < 1/\xi(r-1). \qquad (3.10)$$

Curve 2 of Fig. 5 is a plot of $\xi = r/(r-1)^2$, and is the upper boundary of the region of the second regime. Above this curve lies region III, for the parameters of which there is no stationary solution. For the points belonging to curve 2, the velocity of propagation of the process is determined by the formula for the wave of photodissociation over the products of the chemical chain reaction:

$$D = c\left[1 + c\frac{(A_2^0)-(B_2^0)}{I_0}\right]^{-1}. \qquad (3.11)$$

It is understandable from physical considerations that a wave-like process with such a velocity can propagate only in the case of infinitely high rates of the chemical reactions ($k_1 \to \infty$). But it follows from the foregoing analysis that the velocity determined by formula (3.11) is realized at the finite values of k_1. The cause of the resultant disparity lies in the fact that the zeroth-approximation formulas cease to be valid in the subregion of the parameters adjacent to the curve $\xi = r/(r-1)^2$, Therefore, for the parameters belonging to region III it is necessary to carry out an investigation in at least the next higher order in μ.

4. CONCLUSION

It follows from the foregoing analysis that the process of propagation of the photochemical wave depends significantly on the rate of the chemical reaction. If the rate of the chemical reaction is slow compared with the photolysis rate, then a usual photodissociation wave is realized. In the opposite case, it is necessary to take into consideration the chemical transformations accompanying the photolysis. The usual bimolecular reactions between two molecules

$$A_2 + B_2 \rightleftarrows 2AB$$

are quite slow, since the activation energy for them is appreciable, ~20–50 kcal/mole. On the other hand, low activation energies (1–5 kcal/mole) of the reactions of atoms or radicals with molecules give rise to exceedingly large reaction rates (e.g., for the mixture $H_2 + F_2$ at $T \approx 300°K$ in the case of equal concentrations $N_0 = [H_2^0] = [F_2^0] \approx 10^{17}$ particles/cm^2 and for a density of the "priming" active centers $\sim 10^{-2} N_0$, the rate of formation of the reaction products is $\approx 3 \times 10^{20}$ particles/cm^3 sec). The mechanism of the development of the chain reaction is based on the principle of non-destructibility of the free valence of the monoradicals[3)]. This general principle makes chain reactions a very widespread type of chemical transformations.

The foregoing analysis, although concentrating on binary mixtures having a chain mechanism of transformations, is more general in character and gives a qualitative idea of the influence of the chemical reactions on the process of propagation of photodissociation waves in gas media. In the case when the chemical reaction contributes to the destruction of the photodissociation reagents, the rate of propagation and the width of the profile of the photochemical wave exceed the corresponding parameters of the photodissociation wave that could propagate in the gas in the absence of the chemical reaction. On the other hand, if the direction of the reaction is such that it results in recovery of the photodissociating reagent, then it is natural to expect a qualitatively opposite picture.

It should be noted that in many cases of practical importance chemical chain reactions are accompanied by the release of heat, and this leads, as a result of the Arrhenius factor $\sim e^{-E/kT}$ (E is the activation energy), to a change in the constant of the elementary links of the reaction. It is therefore of interest to study the process of propagation of photochemical waves with allowance for thermal effects. Preliminary investigations have shown that in this situation one of the possible regimes is a combined radiation-detonation regime, the rate of which is determined from the generalized Jouguet condition. It is obvious that thermal effects can be neglected if the mixture is diluted by some neutral gas or if the ratio of the densities of the initial reagents is sufficiently far from stoichiometric.

In conclusion, we are grateful to V. S. Zuev for useful discussions.

[3)] Each chemical bond in a valence-saturated molecule is ensured by two paired electrons. In a free radical there is one unpaired electron. In a reaction of a free radical (or atom) with a molecule there will always be produced a new free radical, so that the number of electrons of the system is odd[15].

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