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An $H_2 - F_2$ amplifier initiated by IR laser radiation in an inhomogeneous working mixture

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***Abstract*.** **An $H_2 - F_2$ amplifier initiated by radiation from a pulsed hydrogen fluoride laser is studied theoretically. Numerical calculations are made by taking into account the inhomogeneity of the initial HF concentration appearing upon preparation of the laser mixture in experiments. The mean theoretical output energy for the $H_2 : F_2 : O_2 : He = 100 : 600 : 30 : 100$-Torr mixture is about $15\,J\,L^{-1}$ for an average initial pressure of hydrogen fluoride equal to 0.5 Torr along the amplifier axis. The results of calculations show that, if a sufficiently homogeneous medium is obtained in experiments, lasing becomes possible according to the kinetic scheme of initiation proposed for the $H_2 - F_2$ laser.**

Keywords: *$H_2 - F_2$ amplifier, resonance vibrational excitation of molecules, initiation of chain reaction.*

One of the possible approaches towards the creation of a chemical laser initiated by its intrinsic radiation and not requiring any external energy sources was tested by us and our coworkers in Ref. [1], where the laser parameters were calculated during the development of lasing due to energy branching of the chain reaction initiated by IR laser radiation in the $HF - H_2 - F_2 - O_2 - He$ mixture. The chain reaction in the $H_2 - F_2 - O_2 - He$ mixture was initiated for the first time by hydrogen fluoride laser radiation. A comparison of the experimental and theoretical lasing buildup times revealed a satisfactory agreement between the experimental and theoretical times of the development of the chain reaction. However, lasing was not observed in the experiments [1]. In our opinion, this is due to the fact that the working mixture used in the experiments was not sufficiently homogeneous. The last component to be added in the $H_2 - F_2 - O_2 - He$ mixture was hydrogen, which was introduced through a number of holes separated by a distance of ~ 5 cm. This resulted in an inhomogeneous distribution of HF along the reactor length. The aim of this paper is to calculate the development of lasing and the laser parameters attainable in an inhomogeneous working mixture, and also to determine quantitative criterion specifying the admissible degree of inhomogeneity of HF.

We will study an $H_2 - F_2$ amplifier initiated by resonance excitation of HF molecules by pulsed hydrogen fluoride laser radiation, followed by a transfer of the vibrational energy of these molecules to H_2 molecules. The reaction of vibrationally excited H_2 molecules with F_2 molecules leads to the formation of free atoms. We will calculate the laser parameters under conditions of amplification due to the energy branching of the chain reaction in the $HF - H_2 - F_2 - O_2 - He$ mixture for an initiating pulse whose spectrum and energy parameters correspond to those for the $H_2 - F_2$ laser pulse [1].

Consider the $HF - H_2 - F_2 - O_2 - He$ medium exposed to a multiple line emission pulse from an $H_2 - F_2$ laser having a duration t_{in}. The hydrogen fluoride molecules resonantly absorb the radiation corresponding to each line of the input pulse, resulting in the formation of vibrationally excited HF(v) molecules (v is the vibrational level number):

$$HF(0,j) + h\nu_{1j} \to HF(1,j-1),$$

$$HF(1,j) + h\nu_{2j} \to HF(2,j-1),$$

$$\dots\dots\dots\dots\dots\dots\dots\dots$$

Here, j is the rotational quantum number, and ν_{vj} is the frequency of the P-line of the input pulse at the (v, $j-1) \to (v-1, j)$ transition. The HF(v) molecules transfer their vibrational energy to the hydrogen molecules:

$$\begin{aligned} &HF(v) + H_2(0) \to HF(v-1) + H_2(1), \\ &HF(v) + H_2(1) \to HF(v-1) + H_2(2). \end{aligned} \qquad (1)$$

The resulting molecules of vibrationally excited hydrogen H_2 react with F_2 molecules to form free atoms:

$$H_2(v \geqslant 1) + F_2 \to F + HF + H\,. \qquad (2)$$

Thus, the action of an external $H_2 - F_2$ laser pulse in the $H_2 - F_2 - O_2 - He$ medium leads to the formation of a certain number of free atoms with a concentration $N_a(t_{\text{in}})$, and to the initiation of a chain reaction between H_2 and F_2. Vibrationally excited molecules HF(v) ($v = 1 - 9$) are formed in the course of this reaction, and the processes (1) and (2) lead to the appearance of additional free atoms (energy branching of the chain). If the rate of formation of the active centres during energy branching of the chain

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exceeds the rate of their loss in the reactions of chain rupture caused by collisions with oxygen molecules, a self-accelerating process will be initiated, which is accompanied by an increase in the concentration N_a and the temperature T of the gas.

As a result, the rate of the chain reaction becomes sufficiently high at a certain instant t_1 to ensure an efficient amplification of laser radiation, and emission at the vibrational–rotational transitions of HF molecules will be amplified in the medium after the passage of an initiating HF laser pulse. This may be the weak emission of a cw HF laser acting on the medium for a time $t > t_1$, or spontaneous emission of vibrationally excited HF molecules in the direction of propagation of the initiating pulse. Thus, the HF laser radiation pulse propagating in the medium will be amplified more and more with increasing distance from the amplifier input.

To determine the parameters of a hydrogen fluoride amplifier initiated by the $H_2 - F_2$ laser radiation, we performed numerical calculations using a previously developed model [2]. In this model, the gains α_v ($v = 1, 2, \ldots, 6$) at the $(v, j-1) \to (v-1, j)$ transitions of HF molecules have the form

$$\alpha_6 = h\nu_6\left(\frac{n_6}{M_{j-1}\tau} - \frac{2j-1}{2j+1}\frac{n_5}{M_j\tau}\right)\frac{1}{I_6 + I_6^{\mathrm{s}}},$$

$$\alpha_5 = \left[\frac{\nu_5}{\nu_6}\alpha_6 I_6 + h\nu_5\left(\frac{n_5}{M_{j-1}\tau} - \frac{2j-1}{2j+1}\frac{n_4}{M_j\tau}\right)\right]\frac{1}{I_5 + I_5^{\mathrm{s}}}, \tag{3}$$

$$\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots$$

$$\alpha_1 = \frac{2j+1}{4j}\left[\frac{\nu_1}{\nu_2}\alpha_2 I_2 + h\nu_1\left(\frac{n_1}{M_{j-1}\tau} - \frac{2j-1}{2j+1}\frac{n_0}{M_j\tau}\right)\right]\frac{1}{I_1 + I_1^{\mathrm{s}}}.$$

Here, I_v is the intensity of radiation with frequency ν_v at the $(v, j-1) \to (v-1, j)$ transition, I_v^{s} is the corresponding saturation intensity, n_v is the total population of the vth vibrational level of HF, τ is the rotational relaxation time in the rotational reservoir model [3], $M_j = [1/(2j+1)](T/Q)\times \exp[j(j+1)Q/T] - 1$, and Q is the characteristic rotational temperature of the HF molecule. The variation in the laser radiation intensity I_v during its propagation along the x axis in the amplifier medium is described by the radiation transport equation

$$\frac{1}{c}\frac{\partial I_v}{\partial t} + \frac{\partial I_v}{\partial x} = \alpha_v I_v. \tag{4}$$

The processes of chemical and vibrational kinetics in the $H_2 - F_2$ laser medium and the rate constants of the processes used in calculations are presented in Ref. [2]. Numerical calculations of the parameters of a hydrogen fluoride amplifier involved the solution of the equations for the populations n_v of the vibrational levels of HF molecules (v =1, 2, ..., 7), equations of chemical kinetics, as well as the equations for the average number of the vibrational quanta of H_2 and the gas temperature. It was assumed that the rotational relaxation time is $\tau = 1/(\pi\Delta\nu_{\mathrm{L}})$, where $\Delta\nu_{\mathrm{L}}$ is the homogeneous half-width of the HF line, and that amplification occurs simultaneously at several transitions of the HF molecule: $(v, j-1) \to (v-1, j)$, where $v = 1, 2, \ldots, 6$. A change in the intensities of laser radiation during its propagation in the direction of the x axis in the $H_2 - F_2$-amplifier medium is described by Eqns (4). By specifying the intensities $I_v(t)$ of the radiation being amplified at the input to the amplifier medium ($x = 0$), we can determine the laser radiation intensity for each $(v, j-1) \to (v-1, j)$ transition at any point x. The specific energy output in the vth band at a distance x from the radiation input to the amplifier medium is determined by the expression $\varepsilon_v(x) = \int \alpha_v(x, t) \times I_v(x, t)\,\mathrm{d}t$.

We carried out numerical calculations by taking into account the spatial inhomogeneity of the initial HF concentration, associated with the peculiarities of laser mixture preparation in the experiments. The time dependence of the total intensity of the initiating pulse was assumed to be identical to the experimental dependence observed for an $H_2 - F_2$ laser pulse of duration 2 μs [1, 4]. In accordance with the spectral and energy characteristics of the $H_2 - F_2$ laser pulse, we assumed that the spectrum of this pulse consists of six vibrational bands of HF corresponding to the $(v, j-1) \to (v-1, j)$ transitions for $j = 7$. The total energy density of the initiating radiation was assumed to be 10 J cm^{-2}. We calculated the dependence of the specific energy output ε on x for the $H_2 : F_2 : O_2 : \mathrm{He} = 76 : 228 : 23 : 578$-Torr mixture used in the experiments [1] and for the $H_2 : F_2 : O_2 : \mathrm{He} = 700 : 600 : 30 : 100$-Torr mixture.

The dependence of the initial partial pressure p_{HF} (in Torr) of hydrogen fluoride in the mixture on the distance x (in cm) is described by the expression $p_{\mathrm{HF}} = 0.5 + C\sin(\pi x/10)$, where C is the amplitude of deviation of pressure p_{HF} from its mean value. The quantity C was varied in calculations in order to determine the admissible inhomogeneity of HF in the medium of the amplifier under study. Table 1 shows the dependence of the minimum ($\varepsilon_{\min}$) and maximum ($\varepsilon_{\max}$) theoretical values of the specific output energy on C for the $H_2 : F_2 : O_2 : \mathrm{He} = 76 : 228 : 23 : 578$-Torr mixture for $x \simeq 400$ cm. The negative values of the output energy correspond to absorption of laser radiation. One can see from Table 1 that the efficient amplification of the input laser radiation is possible only for $C \leqslant 0.1$ Torr. The same condition was obtained for the $H_2 : F_2 : O_2 : \mathrm{He} = 100 : 600 : 30 : 100$-Torr mixture. The obtained results probably explain the absence of lasing in experiments on initiation of the $H_2 : F_2 : O_2 : \mathrm{He} = 76 : 228 : 23 : 578$-Torr mixture by $H_2 - F_2$ laser radiation.

Table 1.

C/Torr	$\varepsilon_{\min}$/J L^{-1}	$\varepsilon_{\max}$/J L^{-1}
0.04	−0.1	1.5
0.05	−0.3	1.2
0.06	−0.5	0.85
0.1	−0.7	0.01
0.15	−0.8	−0.6
0.2	−0.9	−0.7

Fig. 1 shows the dependence of the theoretical specific output energy and the laser radiation energy density (including initiating radiation) on x for the $H_2 : F_2 : O_2 : \mathrm{He} = 700 : 600 : 30 : 100$-Torr mixture for $C = 0.05$ Torr. One can see that the mean output energy for such an enriched mixture is about 15 J L^{-1} for $x > 100$ cm, which is certainly of practical interest. The oscillations of output energy are due to the inhomogeneous concentration of HF along the x axis. In the theoretical version of the amplifier operating in the single-pass mode, the energy gain (ratio of the energies of the output and initiating laser radiation) is equal to 1.3–1.5 for an amplification length 4–5 m. The gain for the initiating radiation energy absorbed in the medium is much larger. It can be obtained in the regenerative amplifier mode in which almost all the introduced energy remains in the active medium. According to the calculations, the gain in such an amplifier may be as large as 40–50 [5].

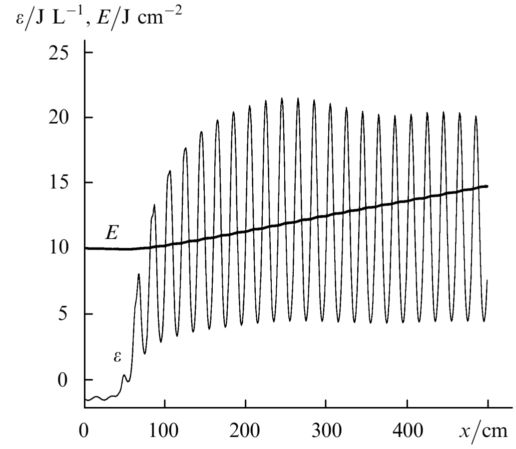

Figure 1. Dependences of the theoretical specific output energy ε and the laser radiation energy density E on x for the $H_2 : F_2 : O_2 : He = 100 : 600 : 30 : 100$-Torr mixture for $C = 0.05$ Torr.

The main purpose of our research was to interpret the experimental results [1] and to find the reasons behind the absence of lasing in these experiments. Fig. 2 shows the dependence of the specific output energy on x for the experimental $H_2 : F_2 : O_2 : He = 76 : 228 : 23 : 578$-Torr mixture for $C = 0.05$ Torr. One can see that the mean theoretical output energy for this mixture is about 0.5 J L^{-1} for $x > 400$ cm.

The theoretical results lead to the conclusion that in the case of a sufficiently homogeneous medium, the kinetic initiation scheme proposed by us can be used to obtain lasing.

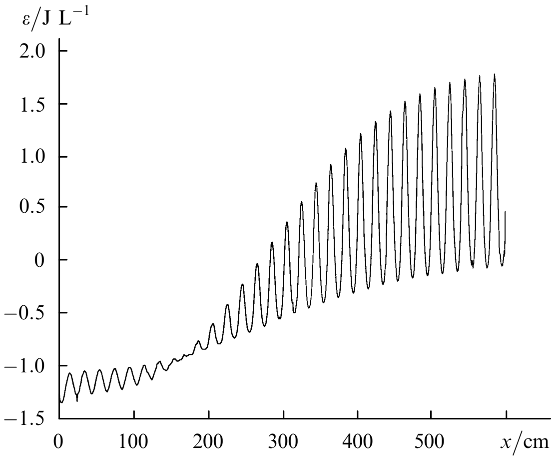

Figure 2. Dependence of the theoretical specific output energy ε on x for the $H_2 : F_2 : O_2 : He = 76 : 228 : 23 : 578$-Torr mixture for $C = 0.05$ Torr.

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