

PACS numbers: 42.55.Ks; 42.60.Lh
DOI: 10.1070/QE2004v034n03ABEH002611

A pulsed hydrogen fluoride amplifier initiated by vibrational excitation of HF molecules by laser radiation

V.I. Igoshin, S.Yu. Pichugin

***Abstract*. An $H_2 - F_2$ amplifier initiated by a resonant vibrational excitation of HF molecules by a pulsed hydrogen fluorine laser is theoretically simulated. Numerical calculations are performed with allowance for an inhomogeneity of the initial HF concentration that appears due to specific features of preparing the experimental laser mixture. The estimated energy output for a mixture of $H_2 : F_2 : O_2 : He = 100 : 600 : 30 : 100$ Torr is $10 - 20$ J L^{-1} for an average initial HF pressure along the amplifier axis of 0.1 – 0.5 Torr. The laser-radiation energy gain on a length of 4 m is 5 – 10.**

Keywords: *$H_2 - F_2$ amplifier, resonant vibrational excitation of molecules, initiation by IR laser radiation.*

An approach to the problem of creating an HF chemical laser initiated by its intrinsic emission and consuming no energy from other external sources is a resonant vibrational excitation of HF molecules followed by transfer of the vibrational energy to H_2 molecules [1, 2]. Subsequently, vibrationally excited H_2 molecules react with F_2 with the formation of free atoms (the energy branching of the chain). The laser characteristics were calculated in Ref. [3] under conditions of the lasing development due to the energy branching of the chain in the $HF - H_2 - O_2 - He$ mixture initiated by IR laser radiation. In this study, a chain reaction in this mixture is initiated experimentally for the first time by HF-laser radiation. However, no lasing was achieved in experiments [3]. In our opinion, this was caused mainly by the impossibility of obtaining a sufficiently homogeneous spatial composition of the working mixture in the experiments. Hydrogen was last to be admitted into the $F_2 - O_2 - He$ mixture through holes arranged with a ~ 10-cm spacing. This resulted in an inhomogeneous production of HF along the reactor length. The aim of this work is a theoretical study of an $H_2 - F_2$ amplifier, which is initiated due to a resonant excitation of HF molecules by a pulsed HF laser under the working-mixture inhomogeneity mentioned above.

Consider an $HF - H_2 - F_2 - O_2 - He$ medium irradiated by a pulse from an HF laser. HF molecules are resonantly excited to the vibrational level $HF(v)$ (v is the level number). The $HF(v)$ molecules transfer their vibrational energy to hydrogen molecules, and the vibrationally excited $H_2(v)$ molecules begin to react with F_2 with the formation of free atoms,

$$H_2(v) + F_2 \rightarrow F + HF + H.$$

Thus, under the action of an HF-laser pulse, a certain concentration N_a of free atoms is produced in the $H_2 - F_2 - O_2 - He$ medium and an $H_2 + F_2$ chain reaction is initiated.

Vibrationally excited $HF(v)$ ($v = 1 - 9$) molecules are formed in this reaction, thus again leading to the production of additional free atoms (the energy branching of the chain). If the production rate of the active centres during the energy branching exceeds their loss rate in the chain-disruption reactions in collisions with oxygen molecules, then a self-acceleration process develops, which is accompanied by an increase in N_a, in the temperature T of the gaseous medium, and in the concentration of vibrationally excited HF molecules.

Some time after the passage of the initiating HF-laser pulse through the medium, favourable conditions for an efficient amplification of the emission determined by vibrational – rotational transitions in HF molecules are created in this medium. This can be either HF-laser radiation, or part of spontaneous emission of vibrationally excited HF molecules that propagates along the amplifier axis in the direction parallel to that of the initiating pulse. As a result, the radiation pulse propagating in the medium considered is amplified at the frequency of HF molecular transitions, as the distance from the entrance to the amplifier increases.

To determine the characteristics of the hydrogen fluoride amplifier initiated by HF-laser radiation, we performed numerical calculations based on the multilevel model developed earlier with regard to the rotational nonequilibrium [4]. In this model, for the gains α_v on the $(v, j-1) \rightarrow (v-1, j)$ transitions of HF molecules ($v = 1, 2, \ldots, R$), we have

$$\alpha_R = hv_R \left(\frac{n_R}{M_{j-1}\tau} - \frac{2j-1}{2j+1} \frac{n_{R-1}}{M_j \tau} \right) \frac{1}{I_R + I_R^s},$$

$$\alpha_{R-1} = \left[\frac{v_{R-1}}{v_R} \alpha_R I_R + hv_{R-1} \left(\frac{n_{R-1}}{M_{j-1}\tau} - \frac{2j-1}{2j+1} \frac{n_{R-2}}{M_j \tau} \right) \right] \times \frac{1}{I_{R-1} + I_{R-1}^s}, \quad (1)$$

V.I. Igoshin, S.Yu. Pichugin P.N. Lebedev Physics Institute, Samara Branch, ul. Novo-Sadovaya 221, 443011 Samara, Russia;
web-site: www.fian.smr.ru; e-mail: theor@fian.smr.ru

Received 8 April 2003; revision received 22 October 2003
Kvantovaya Elektronika **34** (3) 203 – 205 (2004)
Translated by A.S. Seferov

. . . . . .

$$\alpha_1 = \frac{2j+1}{4j}\left[\frac{v_1}{v_2}\,\alpha_2 I_2 + hv_1\left(\frac{n_1}{M_{j-1}\tau} - \frac{2j-1}{2j+1}\,\frac{n_0}{M_j\tau}\right)\right]\frac{1}{I_1 + I_1^{\S}},$$

where I_υ is the radiation intensity at the frequency v_υ corresponding to the $(\upsilon, j-1) \to (\upsilon-1, j)$ transition; $I_\upsilon^{\S}$ is the corresponding saturation intensity [4]; n_υ is the population of the υth HF vibrational level; τ is the characteristic rotational-relaxation time; $M_j = [1/(2j+1)]\exp[j(j+1)\times Q/T]T/Q - 1$; and Q is the characteristic rotational temperature of an HF molecule. The change in the radiation intensity I_υ with its propagation along the x axis in the amplifier medium is described by the radiation-transfer equation

$$\frac{1}{c}\frac{\partial I_\upsilon}{\partial t} + \frac{\partial I_\upsilon}{\partial x} = \alpha_\upsilon I_\upsilon. \qquad (2)$$

The chemical and vibrational kinetic processes in the $H_2 - F_2$-laser medium considered by us and the rate constants of these processes used in our calculations are presented in Ref. [5]. When calculating numerically the characteristics of $H_2 - F_2$ amplifier, we solved the equations for the populations n_υ of the vibrational levels of HF molecules ($\upsilon = 0, 1, \ldots, 7$), equations of chemical kinetics, and equations for the average reserve of the H_2 vibrational quanta and gaseous-medium temperature. It was assumed that the characteristic time of the rotational relaxation is $\tau = 1/(\pi\Delta v_L)$, where Δv_L is the homogeneous half-width of the HF line. In these calculations, the amplification was also assumed to occur simultaneously on the $(\upsilon, j-1) \to (\upsilon-1, j)$ transitions of HF molecular with $\upsilon = 1, 2, \ldots, 6$. The change in the corresponding radiation intensities with the propagation of light along the x axis in the $H_2 - F_2$ amplifier medium is described by Eqns (2). Setting the intensities $I_\upsilon(t)$ of the amplified radiation at the entrance to the amplifier medium ($x = 0$), we can find the radiation intensities at each $(\upsilon, j-1) \to (\upsilon-1, j)$ transition at an arbitrary point x. The specific energy output at a distance x from the entrance of radiation to the amplifier medium is determined by the expression

$$\mathscr{E}(x) = \sum_{\upsilon=1}^{6}\int \alpha_\upsilon(x,t) I_\upsilon(x,t)\mathrm{d}t.$$

We performed numerical calculations taking into account the inhomogeneity of the initial HF concentration in the volume that resulted from the specific procedure used to prepare the laser mixture. The time dependence of the total intensity I_{in} of the input laser pulse was assumed to be equal to that measured experimentally for a 2-μs pulse of an $H_2 - F_2$ laser [3], and for $t > 2$ μs we assumed that $I_{in} = 1$ W cm^{-2}. In accordance with the experimental spectral and energy characteristics of the $H_2 - F_2$-laser pulse, it was assumed in the calculations that this pulse includes radiation of six HF vibrational bands on the $(\upsilon, j-1) \to (\upsilon-1, j)$ transitions, where $\upsilon = 1-6$, $j = 7$ [6]. The calculations were performed for an energy-consuming mixture of $H_2 : F_2 : O_2 : He = 100 : 600 : 30 : 100$ Torr. The change in the initial partial pressure of HF in the mixture as a function of the distance along the x axis was described by the expression $p_{HF} = p_0 + C\sin(2\pi x/d)$, where C is the amplitude of the p_{HF} deviation from the average value and d is the period of the HF-pressure inhomogeneity along the amplifier axis. As a result of numerical calculations, we find the local (not averaged over the inhomogeneity period d) parameters of the amplifier under study, in particular, the specific laser energy output $\mathscr{E}$ as a function of x for various p_0, C, and the energy density $E_{in} = E(x=0)$ of the input initiating pulse.

Table 1.

p_0/Torr	E_{in}/J cm^{-2}	E_{out}/J cm^{-2}	K
0.1	0.25	0.75	3
	0.5	2.5	5
	1	4.7	4.7
	2	6.8	3.4
0.25	0.5	0.65	1.3
	1	2.5	2.5
	2	5.4	2.7
	3	7.3	2.4
0.5	1	1.1	1.1
	2	3.4	1.7
	3	5.7	1.9
	4	7.4	1.85

Table 1 presents the calculated output-pulse energy density $E_{out} = E(x=L)$ and the energy gain $K = E_{out}/E_{in}$ at various p_0 and E_{in} values for the amplifier length $L =$ 4 m. The calculations were performed at a comparatively small deviation of the HF partial pressure from the average value ($C/p_0 = 5\,\%$) with a period $d = 20$ cm. Note that calculations for $d = 10$ cm lead to almost the same results. The maximum attainable lasing-energy gain increases with a decrease in the initial HF pressure in the mixture and reaches a value of 5 for $p_0 = 0.1$ Torr (see Table 1). This is determined by a decrease in the laser-radiation losses in the $H_2 - F_2$-amplifier medium observed with decreasing p_{HF}, which primarily leads to a decrease in the energy density E_{in} required to amplify efficiently the emission on the HF transitions. Decreasing the oxygen pressure in the mixture also results in increased calculated K values. For example, at an O_2 pressure of 10 Torr, the energy gain for a 4-m amplifier length is 10. The time profile of the output laser pulse calculated for $p_0 = 0.25$ Torr and $E_{in} = 2$ J cm^{-2} is shown in Fig. 1. The first pulse observed at the output of the $H_2 - F_2$-amplifier is the slightly attenuated initiating pulse, and the amplified HF-laser pulse is delayed by ~ 20 μs.

In order to determine the acceptable HF inhomogeneity in the medium of the amplifier under study, the deviation of the initial HF-pressure amplitude from its average value was

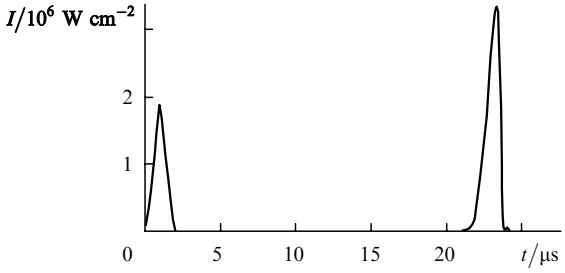

Figure 1. Calculated time dependence of the total laser-radiation intensity I at the amplifier output for $p_0 = 0.25$ Torr and $E_{in} = 2$ J cm^{-2} ($C/p_0 = 5\,\%$).

varied in the calculations. The laser-energy gain as a function of the parameter C/p_0 for $p_0 = 0.1$ Torr and $E_{in} = 1$ J cm^{-2} is plotted in Fig. 2. This dependence and calculations for other versions show that the input laser radiation can be efficiently amplified at $C/p_0 < 20\,\%$. Figure 3 shows the calculated dependences of the energy output on x for $C/p_0 = 5\,\%$ and $50\,\%$ ($d = 20$ cm). Oscillations in the energy output are caused by an inhomogeneity in the initial HF concentration along the x axis. Negative values of the energy output correspond to a predominance of a negative gain, i.e., to an absorption of laser radiation. The average calculated energy output for $C/p_0 = 5\,\%$ and $50\,\%$ is ~ 10 J L^{-1} at $x > 100$ cm and 3 J L^{-1} only at $x > 200$ cm, respectively.

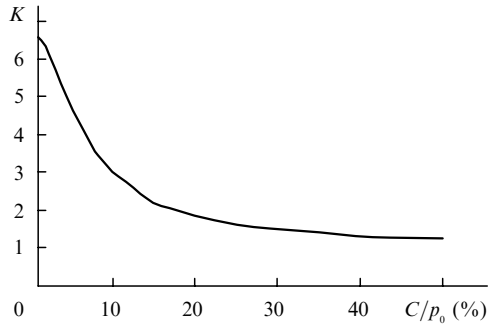

Figure 2. Output-energy gain versus parameter C/p_0 for $p_0 = 0.1$ Torr and $E_{in} = 1$ J cm^{-2}.

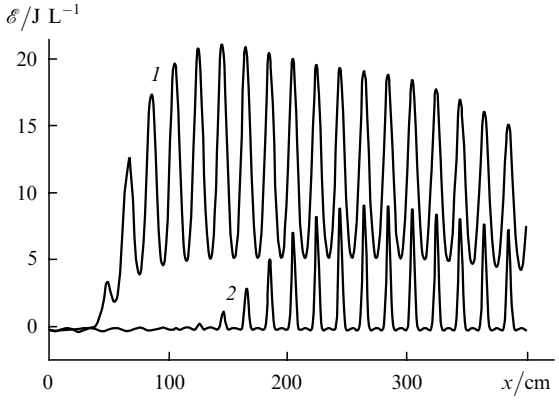

Figure 3. Calculated dependences of the specific energy output on x for $C/p_0 = 5\,\%$ (*1*) and $50\,\%$ (*2*) at $p_0 = 0.1$ Torr and $E_{in} = 1$ J cm^{-2}.

Thus, we have performed a numerical simulation of a pulsed hydrogen fluoride chemical amplifier initiated by input radiation. We have shown that the kinetic scheme for initiating an $H_2 - F_2$ laser through a resonant vibrational excitation of HF molecules can ensure a significant laser-energy gain without external energy sources.

References

1. Bashkin F.S., Igoshin V.I., Oraevskii A.N., Shcheglov V.A. *Khimicheskie lazery* (Chemical Lasers) Ed. by N.G. Basov (Moscow: Nauka, 1982) p. 182.
2. Bashkin A.S., Kurdoglyan M.S., Oraevskii A.N. *Trudy FIAN*, **194**, 45 (1989).
3. Azarov M.A., Drozdov V.A., Igoshin V.I., Katulin V.A., Kurov A.Yu., Petrov A.L., Pichugin S.Yu., Troshchinenko G.A. *Kvantovaya Elektron.*, **25**, 401 (1998) [*Quantum Electron.*, **28**, 388 (1998)].
4. Igoshin V.I., Pichugin S.Yu. *Kvantovaya Elektron.*, **23**, 326 (1996) [*Quantum Electron.*, **26**, 318 (1996)].
5. Igoshin V.I., Pichugin S.Yu. *Kvantovaya Elektron.*, **21**, 417 (1994) [*Quantum Electron.*, **24**, 384 (1994)].
6. Azarov M.A., Igoshin V.I., Pichugin S.Yu., Troshchinenko G.A. *Kvantovaya Elektron.*, **29**, 21 (1999) [*Quantum Electron.*, **29**, 859 (1999)].