# Chemical hydrogen fluoride laser on a thermal chain explosion initiated by resonance IR radiation

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Abstract. Characteristics of a chemical $H_2 - F_2$ laser initiated by resonant vibrational excitation of HF molecules upon the development of a thermal chain branching reaction are theoretically studied. After the addition of N_2F_2 or F_2SO_3 molecules with partial pressure of 20 Torr to the $HF : H_2 : F_2 : O_2 : He = 0.5 : 76 : 228 : 23 : 100$-Torr mixture, a specific output energy of a hydrogen fluoride laser initiated by a 2-μs pulse from an $H_2 - F_2$ laser with the energy density of $0.01 - 10$ J cm^{-2} amounted to $30 - 60$ J litre^{-1}, exceeding the specific initiation energy by a factor of 100 – 4000.

In Ref. [1], the mechanism of $H_2 - F_2$ laser initiation using resonant excitation of HF molecules by a pulsed hydrogen laser was studied , with subsequent vibrational-energy transfer to H_2 molecules, which enter into the energy chain branching reaction

$$H_2(v \geqslant 1) + F_2 \rightarrow F + HF + H.$$
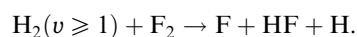
It was theoretically shown that under the action of an $H_2 - F_2$ laser pulse with the energy density of $0.1 - 10$ J litre^{-1} on an $HF - H_2 - F_2 - O_2$ mixture at a pressure of 1.1 bar, lasing can be observed with the specific output energy of about 10 J litre^{-1}. We observed the initiation of the chain $H_2 + F_2$ reaction by radiation from a pulsed hydrogen fluoride laser. However, the experiments gave no evidence of lasing in the $H_2 - F_2$ laser initiated in this way, which was probably explained by an insufficient homogeneity of the laser mixtures prepared.

The specific output energy of a hydrogen fluoride laser at a low level of photolytic initiation can be considerably increased when a chain reaction with a thermal branching mechanism takes place in the laser medium [2]; we will call this reaction the thermal chain explosion. This branching mechanism is caused by thermal dissociation of fluorine-containing RF molecules introduced into an initial laser mixture. The dissociation of such molecules is accompanied by the formation of free F atoms, and it proceeds during heating of the mixture by heat evolved in the chain $H_2 + F_2$ reaction.

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In Ref. [2], the rate constants of thermal dissociation of RF molecules required to realise the thermal mechanism of chain reaction branching and obtain a high specific output power in an $H_2 - F_2$ laser were studied in detail. A number of suitable molecules (F_2SO_3, KrF_2, etc.) making it possible to obtain specific output powers of $100 - 200$ J litre^{-1} at a very low initiation level were found. In this case, the dominant portion of laser radiation is produced in an extremely short time (~ 100 ns) after a rather long heating (during tens of microseconds) of a laser mixture. Note that the hydrogen fluoride model used in Ref. [2] did not take into account the fact that HF molecules are not in a rotational equilibrium. Including this into consideration can substantially decrease the calculated laser energy output the case of a short laser pulse.

In this paper, we use the model taking into account a finite rate of rotational relaxation and, based on it, study a laser operating on an $HF - H_2 - F_2 - O_2 - He - F_2SO_3$ (N_2F_2) mixture and initiated by resonant vibrational excitation of HF molecules with the $H_2 - F_2$ laser radiation in many spectral lines. In this medium, combined energy and thermal branching of the chain $H_2 + F_2$ reaction is possible, which is accompanied by a substantial increase in the specific output power in the laser under study initiated by IR laser radiation.

Consider the medium of an $H_2 - F_2$ laser, which represents the $HF - H_2 - F_2 - O_2 - He$ mixture containing an easily dissociating RF addition. The feasibility of preparing this medium and its stability were analysed in Refs [2, 3]. Under the action of an $H_2 - F_2$ laser pulse with duration t_i, resonant vibrational excitation of HF molecules with vibrational-energy transfer to H_2 molecules takes place in this medium [1]. As a result of VV' exchange reactions, H_2 molecules excited to vibrational levels with $v \geqslant 1$ are formed. These molecules enter into the reaction with F_2, which produces free atoms. Thus, a certain concentration $N_a(t_i)$ of free atoms is produced in the $HF - H_2 - F_2 - O_2 - He - RF$ medium under the action of external $H_2 - F_2$ laser radiation, and the chain reaction is initiated in the $H_2 - F_2$ mixture.

If the rate of production of free atoms in the course of energy chain branching exceeds the rate of their loss in collisions with O_2 molecules, a self-accelerating process is developed, which is accompanied by an increase in N_a and the temperature T of a gas medium. As a result, the rate of the chain reaction at a certain moment becomes sufficient for lasing.

In the absence of RF molecules in an initial laser mixture, the specific output power of the $H_2 - F_2$ laser initiated by IR radiation is rather low (~ 10 J litre^{-1}) [1]. As for the medium considered here, thermal dissociation of RF molecules and

the formation of free atoms in it become of substantial importance upon reaching a certain temperature T. In this case, their concentration sharply increases, which leads to the production of a short laser pulse with a rather large energy [2]. As a result, the specific output power ε of the $H_2 - F_2$ laser initiated by hydrogen fluoride laser radiation can considerably increase (by an order of magnitude). The energy gain of IR laser radiation $K = \varepsilon/\varepsilon_0$, where ε_0 is the specific initiation energy, will also considerably increase [4].

To find characteristics of a hydrogen fluoride laser operating on a thermal chain branching reaction initiated by $H_2 - F_2$ laser radiation, we performed numerical calculations based on the model [5]. To take into account resonant excitation of HF molecules by external hydrogen fluoride laser radiation, we used the equations

$$\frac{dn_v^j}{dt} = \frac{\alpha_{v,j+1}I_{v,j+1}}{hv_{v,j+1}} - \frac{\alpha_{v+1,j}I_{v+1,j}}{hv_{v+1,j}} + \frac{n_v - n_v^j}{M_j\tau_j} - \frac{n_v^j}{\tau_j}. \quad (1)$$

Here, n_v^j is the specific concentration of HF molecules at the level with vibrational and rotational numbers vth and jth, respectively; n_v is the total population of the vth vibrational level of HF; $\alpha_{v,j}$ is the coefficient of absorption of hydrogen fluoride laser radiation at the $(v, j-1) \rightarrow (v-1, j)$ transition with the intensity $I_{v,j}$ by HF molecules; $v_{v,j}$ is the corresponding radiation frequency; τ_j is the characteristic rotational relaxation time [1, 6]; $M_j = (2j+1)\exp[-j(j+1) \times Q/T]Q/T - 1$; and Q is the characteristic rotational temperature of the HF molecule. To calculate rotational relaxation times τ_j, we took the rate constants of RT processes with an exponential dependence on the rotational energy, which were used in Ref. [7].

In the calculations, an initiating radiation pulse for each $(v, j-1) \rightarrow (v-1, j)$ transition, where $v = 1 - 6$ and $j = 4 - 12$, was assumed to have the shape of an isosceles triangle with a base stretching from the beginning of emission in the given spectral line to its termination, which corresponds to the experimental data [1, 7]. The resulting time dependence of the total intensity of an initiating pulse was rather close to the experimental shape of the 2-μs $H_2 - F_2$ laser pulse [1].

We performed calculations for the $H_2 : F_2 : O_2 : He = 76 : 228 : 23 : 100$-Torr mixture ($T = 290$ K). The mixture was assumed to contain an easily dissociating addition of F_2SO_3 or trans-N_2F_2 at a pressure of 20 Torr. The dissociation rate constants of the molecules in the medium under study were taken as follows [2]: $k_d(F_2SO_3) = 3 \times 10^{13} \times \exp(-16500\,\text{K}/T)\ \text{s}^{-1}$ and $k_d(N_2F_2) = 5 \times 10^{11} \times \exp(-15500\,\text{K}/T)\ \text{s}^{-1}$. The initial HF pressure was set equal to 0.5 Torr. The specific output energy ε for laser radiation produced in this medium was calculated for a threshold gain of 0.005 cm^{-1} in the cavity.

The specific initiation energy, which represents the specific energy of the initiating laser absorbed in the working medium of the laser being initiated was calculated by the formula

$$\varepsilon_0 = \int_0^{t_i} \left(\sum_{v,j} \alpha_{v,j} I_{v,j} \right) dt,$$

where $\alpha_{v,j}$ was calculated using equations (1). The results of calculations of characteristics of the $H_2 - F_2$ laser on a thermal chain explosion initiated by hydrogen fluoride laser radiation at different energy densities E_1 are presented in the table. In it, we also give time intervals Δt in which a laser pulse reaches its maximum.

One can see from the table 1 that the calculated specific output energy of the given oscillator is almost independent of E_1: $\varepsilon = 60$ J litre^{-1} in the presence of F_2SO_3 in the initial mixture and $\varepsilon = 33$ J litre^{-1} in the presence of the N_2F_2 addition. The energy gain of the hydrogen fluoride laser decreased from $K = 3900 - 2140$ at $E_1 = 0.01$ J cm^{-2} to $K = 170 - 95$ at $E_1 = 10$ J cm^{-2}, respectively. The interval t_d between the laser pulse produced by a thermal chain explosion and the initiating pulse was $310 - 85$ μs. Thus, the use of an easily dissociating addition provides a considerable increase (by a factor of $5 - 10$) in the energy of the $H_2 - F_2$ laser pulse initiated by hydrogen fluoride laser radiation.

Table 1.

E_1/J cm^{-2}	ε_0/J litre^{-1}	N_a/cm^{-1}	ε/J litre^{-1}	Δt/μs	K
0.01	0.15	1.6×10^{10}	60 (33)	311 (311)	3900 (2140)
0.1	0.11	1.2×10^{11}	60 (33)	154 (154)	545 (300)
1	0.28	3.7×10^{11}	61 (34)	97 (97)	220 (120)
10	0.36	5×10^{11}	61 (34)	85 (85)	170 (95)

Note: Data in the parentheses correspond to the results of calculations for the initial mixture containing the trans-N_2F_2 addition.

Fig. 1 presents the time dependence of the power density of the thermal chain explosion $H_2 - F_2$ laser in a medium with the initial composition HF : H_2 : F_2 : O_2 : He : F_2SO_3 = $0.5 : 76 : 228 : 23 : 100 : 20$ Torr for $E_1 = 1$ J cm^{-2}. Our calculations show that the laser pulse caused by a thermal chain explosion is rather short (~ 20 ns) and it is important to take into account the deviation from the rotational equilibrium in the calculation of specific output laser energy under these conditions. Figs 2 and 3 present the corresponding time dependences of the concentration of free atoms and the gas temperature. In this case, a substantial increase in the concentration of active centres and in the laser radiation power begins upon achieving a temperature of $1200 - 1300$ K.
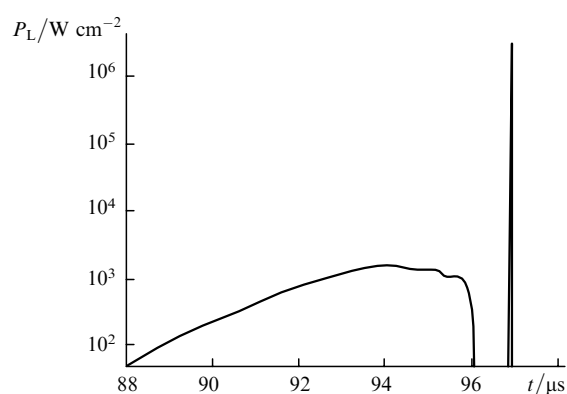

Figure 1. Time dependence of the output power density of the $H_2 - F_2$ laser on a thermal chain explosion in an HF $-$ H_2 $-$ F_2 $-$ O_2 $-$ He $-$ F_2SO_3 mixture for $E_1 = 1$ J cm^{-2}.

Thus, we theoretically studied an $H_2 - F_2$ laser on a thermal chain explosion initiated by multiline radiation of a hydrogen fluoride laser using resonant vibrational excitation of HF molecules. In the presence of easily dissociating additions of trans-N_2F_2 or F_2SO_3 with partial pressure of 20 Torr in the HF : H_2 : F_2 : O_2 : He = $0.5 : 76 : 228 : 23 : 100$-Torr
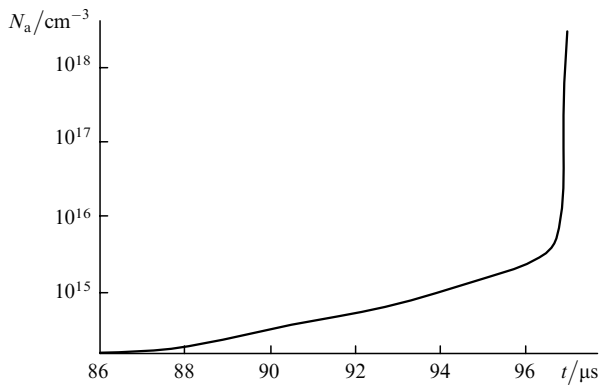
Figure 2. Time dependence of the concentration of free atoms in the medium of the $H_2 - F_2$ laser where a thermal chain reaction is developed.
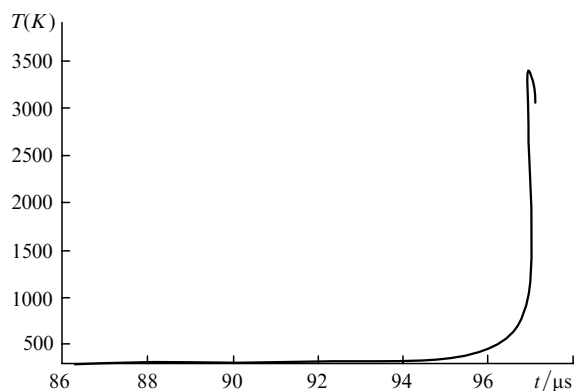

Figure 3. Time dependence of the temperature of the laser medium.

mixture, the specific output energy of the $H_2 - F_2$ laser initiated by a hydrogen fluoride laser pulse with energy density of $0.01 - 10$ J cm^{-2} can reach 60 J litre^{-1}, which exceeds the specific initiation energy by a factor of $100 - 4000$.

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