

# On the possibility of obtaining high energy parameters from an IR-initiated thermal-chain explosion H<sub>2</sub>–F<sub>2</sub> laser

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**Abstract.** A chemical H<sub>2</sub>–F<sub>2</sub> laser initiated by IR radiation through vibrational excitation of HF molecules is studied theoretically under the conditions of a branched thermal-chain reaction caused by the thermal dissociation of gaseous additions to the laser mixture. It is demonstrated that, using the F<sub>2</sub>SO<sub>3</sub> addition at a partial pressure of 40 Torr, the H<sub>2</sub>–F<sub>2</sub> laser with a mixture pressure of 1 bar, initiated by a pulse of a hydrogen–fluorine laser can provide the output exceeding 200 J litre<sup>-1</sup> in 20-ns laser pulses.

**Keywords:** pulsed chemical H<sub>2</sub>–F<sub>2</sub> laser, thermal-chain explosion, energy parameters.

The problem of initiating chemical lasers by IR radiation has long been studied [1, 2]. In Ref. [2], an IR-initiated, chemically active molecular system was proposed and substantiated theoretically, whose energy output exceeds the input energy of initiation photons. This was the CH<sub>3</sub>F–D<sub>2</sub>–F<sub>2</sub>–CO<sub>2</sub>–He system emitting at the 9.6- $\mu\text{m}$  00<sup>0</sup>1→02<sup>0</sup> transition of CO<sub>2</sub> molecules. This paper also introduced the term ‘photon-branched (PB) chain reaction’, which characterises the kinetics of physicochemical processes in the active medium.

Note that a general suggestion about the possibility of chain branching with the help of the energy of reemitted photons in such molecular systems was first expressed in Ref. [3], which studied photochemical action of IR radiation. The authors of [3, 4] discovered the possibility of initiating chemical reactions by IR radiation in mixtures containing molecules N<sub>2</sub>F<sub>4</sub>, BCl<sub>3</sub>, and SF<sub>6</sub> that resonantly absorb the 10.6- $\mu\text{m}$  emission of a CO<sub>2</sub> laser and whose dissociation produces free atoms.

The authors of Refs [5, 6] carried out experiments devoted to creating an IR-initiated chemical HF laser using the SF<sub>6</sub>–H<sub>2</sub> mixture. In Refs [7, 8], lasing on rovibronic transitions of HF (DF) molecules was obtained using reactions initiated in SF<sub>6</sub>–H<sub>2</sub> and N<sub>2</sub>F<sub>4</sub>–H<sub>2</sub> (D<sub>2</sub>) mixtures. In all these experiments, the energy of the output emission was extremely small, being  $\sim 10^{-3}$ – $10^{-4}$  of the energy of the initiating pulse of a CO<sub>2</sub> laser. This indicates a small

efficiency of forming active centres during the dissociation of SF<sub>6</sub> and N<sub>2</sub>F<sub>4</sub> molecules, which resonantly absorb the IR radiation. The fast relaxation of vibrational energy of these molecules in the laser medium can be responsible for this small efficiency.

Free atoms can also be produced by other mechanisms caused by nonresonance IR irradiation of the medium. For example, the experiment on developing a pulsed HF laser initiated by the emission of a CO<sub>2</sub> laser that was focused on an aluminium plate placed in a CF<sub>4</sub>–H<sub>2</sub> gas mixture was described in [9]. The IR radiation produces an electron plasma in the region near the plate. CF<sub>4</sub> molecules dissociate in the plasma layer, producing atomic F. The energy output of this laser was very small: much less than the energy of the initiating pulse. The paper [10] was devoted to the experimental study of thermal decomposition of solid NaN<sub>3</sub> particles heated by a CO<sub>2</sub> laser. It was found that Na atoms produced upon the decomposition of NaN<sub>3</sub> could react with F<sub>2</sub>, producing active F centres.

These works stimulated theoretical studies on the possibility of creating pulsed PB reaction chemical lasers that would be based on new mechanisms of IR-induced production of free atoms. A novel method of realising PB chain reaction was proposed which is based on the non-resonance interaction of IR radiation with dispersed chemically active media [11–13].

The idea was to introduce finely dispersed particles that strongly absorb the IR radiation (e.g., passivated metal particles) into the active medium of the chemical HF (DF) laser. These particles can be heated by sufficiently intense IR radiation to high temperatures in about 1  $\mu\text{s}$  while leaving the temperature of the ambient medium virtually unchanged. This results in the evaporation or thermal decomposition of the particle matter and the initiation of a chain reaction.

The authors of [12] studied numerically the operation of a pulsed chemical HF laser initiated by the IR radiation, which evaporated finely dispersed aluminium particles injected into the laser medium. The specific energy output of the hydrogen–fluorine laser reached 100–200 J litre<sup>-1</sup> in the mixture at a pressure of 1 bar containing aluminium particles with diameters of 0.1–0.2  $\mu\text{m}$  and a concentration of 10<sup>9</sup> cm<sup>-3</sup>. This exceeds the specific input energy of the initiating radiation by a factor of 20. Under these conditions, the overlap time of the diffuse clouds of evaporated particles ( $\sim 0.1$   $\mu\text{s}$ ) is much shorter than the duration of the chemical lasing ( $> 1$   $\mu\text{s}$ ), ensuring a high degree of homogeneity of the medium.

In previous papers [14, 15], we have proposed a new approach to initiating an H<sub>2</sub>–F<sub>2</sub> laser by external IR radi-

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ation, where the energy output exceeds the input. At low rates of IR initiation, the energy output of a hydrogen–fluorine laser can be increased greatly by realising a thermal chain explosion in the medium, made possible by the dissociation of specially selected fluorine-containing molecules, RF.

The authors of [16] studied in detail the question of the thermal dissociation rates of RF molecules at which the thermal branching mechanism of the UV-initiated  $\text{H}_2\text{--F}_2$  chain reaction can be realised. In this case, most of the output energy is emitted in a very short period of time ( $\sim 100$  ns) after a long (tens of microseconds) heating of the mixture. The calculations of Ref. [16] used a model of a hydrogen–fluorine laser that neglects the deviation of HF molecules from the rotational equilibrium, which can significantly lower the calculated laser energy output in the case of short laser pulses.

Among the molecules proposed for thermal branching, we consider the  $\text{F}_2\text{SO}_3$  molecule to be the most suitable. With the specific dissociation energy of  $31 \text{ kcal mol}^{-1}$ , it is sufficiently stable; however, at temperatures exceeding  $1000 \text{ K}$ , its rate of thermal dissociation is high enough. In this work, we use the model that takes into account the finite rotational relaxation rate to optimise the energy characteristics of a laser on the  $\text{HF--H}_2\text{--F}_2\text{--O}_2\text{--He--F}_2\text{SO}_3$  mixture initiated by a pulse of a hydrogen–fluorine laser. In doing so, we vary the mixture composition and the rotational quantum number of the generated radiation.

Consider the medium of a hydrogen–fluorine laser on the  $\text{HF--H}_2\text{--F}_2\text{--O}_2\text{--He--F}_2\text{SO}_3$  mixture. An incident pulse of duration  $t_i$  from an  $\text{H}_2\text{--F}_2$  laser resonantly excites vibrational levels of HF molecules, whose vibrational energy is then transferred to  $\text{H}_2$  molecules as a result of VV' exchange processes, producing vibrationally excited  $\text{H}_2$  molecules ( $v \geq 1$ ) [14]. These molecules react with  $\text{F}_2$ , producing free fluorine atoms. Thus, under the action of an external pulse of an  $\text{H}_2\text{--F}_2$  laser, the  $\text{HF--H}_2\text{--F}_2\text{--O}_2\text{--He--F}_2\text{SO}_3$  mixture produces a certain concentration  $N_a$  of free fluorine atoms, which initiate the  $\text{H}_2\text{--F}_2$  chain reaction. If in the process of the energy chain branching, the production of free atoms dominates their loss due to collisions with  $\text{O}_2$  molecules, a self-accelerating process increasing the concentration of active centres and the temperature of the gas medium  $T$  will start.

Therefore, at a certain moment the rate of the chain reaction becomes sufficiently large to start lasing. If there are no  $\text{F}_2\text{SO}_3$  molecules in the laser medium, the specific energy output of this laser will be relatively small ( $\sim 10 \text{ J litre}^{-1}$ ) [14]. In the mixture under study, however, the process of thermal dissociation of  $\text{F}_2\text{SO}_3$  molecules, producing free F atoms in the reaction  $\text{F}_2\text{SO}_3 \rightarrow \text{F} + \text{FSO}_3$ , becomes significant at some temperature  $T$ . This will drastically increase the concentration of atomic fluorine, leading to the generation of a relatively powerful and short laser pulse at time instant  $t_1$  after the initiation [15, 16]. As a result, the specific energy output  $\varepsilon_L$  of the laser under study can be greatly enhanced.

To find the characteristics of a hydrogen–fluorine laser on the branched thermal chain reaction initiated by an  $\text{H}_2\text{--F}_2$  laser, we performed numerical calculations using the model earlier developed for a fixed rotational quantum number of the emitted radiation [17]. At the first stage (initiation), we solved the equations describing resonant excitation of HF molecules by the external radiation of an

$\text{H}_2\text{--F}_2$  laser:

$$\frac{dn_v^j}{dt} = \frac{\alpha_{v,j+1} I_{v,j+1}}{h\nu_{v,j+1}} - \frac{\alpha_{v+1,j} I_{v+1,j}}{h\nu_{v+1,j}} + \frac{n_v - n_v^j}{M_j \tau_j} - \frac{n_v^j}{\tau_j}.$$

Here,  $n_v^j$  is the specific population of the  $v$ th vibrational,  $j$ th rotational level in HF molecules;  $n_v$  is the total population of the  $v$ th vibrational level of HF molecules;  $\alpha_{v,j}$  is the coefficient of absorption of the external radiation of intensity  $I_{v,j}$  that is due to  $(v, j-1) \rightarrow (v-1, j)$  transitions of HF molecules;  $\nu_{v,j}$  is the lasing frequency;  $\tau_j$  is the characteristic time of rotational relaxation [18, 19];  $M_j = [1/(2j+1)] \exp[j(j+1)Q/T] T/Q - 1$ ;  $Q$  is the characteristic rotational temperature of HF molecules.

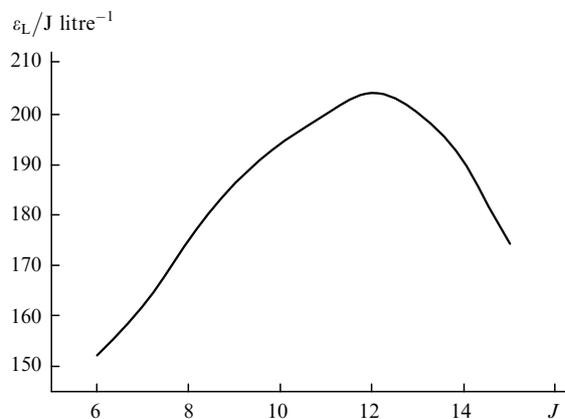
In accordance with experimental data [14, 19], we assumed in the calculations that the initiating pulse had the form of an isosceles triangle for all  $(v, j-1) \rightarrow (v-1, j)$  transitions of the HF molecule with  $v = 1-6$ ,  $j = 4-12$ . The resulting time dependence of the total intensity of the initiating pulse was sufficiently close to the experimentally observed shape of  $2\text{-}\mu\text{s}$  pulses of a hydrogen–fluorine laser [14]. The total energy density of the initiating pulse amounted to  $1 \text{ J cm}^{-2}$ . The actual calculations were carried out for the mixture  $\text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} = X : 400 : 40 : Y \text{ Torr}$  (at  $T = 300 \text{ K}$ ). The mixture was assumed to contain 40 Torr of the easily dissociating  $\text{F}_2\text{SO}_3$  addition. The total pressure of the mixture was 781 Torr. The pressures of hydrogen and helium varied, but their total partial pressure was 300 Torr. The dissociation rate of  $\text{F}_2\text{SO}_3$  molecules in the considered medium was assumed to be given by  $k_d = \exp(-16500 \text{ K}/T) \times 5 \times 10^{13} \text{ s}^{-1}$  [16]. The initial HF pressure was 1 Torr. The specific energy output  $\varepsilon_L$  of the laser was calculated for the threshold cavity gain of  $0.005 \text{ cm}^{-1}$ .

Table 1 lists the calculated values of  $\varepsilon_L$  for the rotational numbers  $J = 7$  and 12 and various pressures of hydrogen and helium. One can see that when the initial mixture contains  $\text{F}_2\text{SO}_3$  molecules, the calculated energy output is the greater, the lower is the concentration of helium. However, since some amount of helium must be present in experiments with hydrogen–fluorine lasers, in the following we will consider the mixture  $\text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} = 200 : 400 : 40 : 100 \text{ Torr}$ , which produces relatively high energy outputs while remaining experimentally realistic. Then, the radiation energy yield of the hydrogen–fluorine thermal chain explosion laser can exceed  $200 \text{ J litre}^{-1}$  in the  $\text{HF--H}_2\text{--F}_2\text{--O}_2\text{--He--F}_2\text{SO}_3$  medium at a pressure of 1 bar.

Fig. 1 shows the calculated energy output as a function of the rotational quantum number  $J$  for the chosen mixture,  $\text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} = 200 : 400 : 40 : 100 \text{ Torr}$ . One can see that the maximum energy output of  $204 \text{ J litre}^{-1}$  is reached

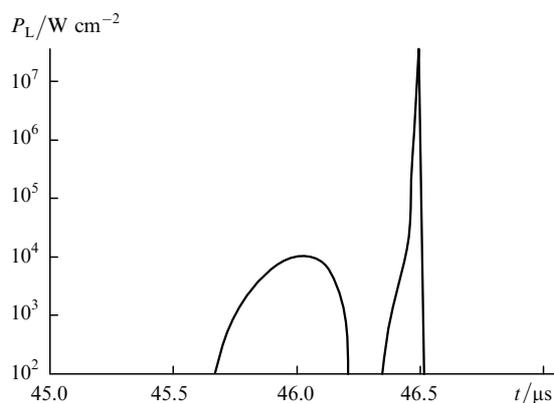
| $[\text{H}_2]/\text{Torr}$ | $[\text{He}]/\text{Torr}$ | $N_a/10^{12} \text{ cm}^{-3}$ | $\varepsilon_L/\text{J litre}^{-1} (J=7)$ | $\varepsilon_L/\text{J litre}^{-1} (J=12)$ | $t_1/\mu\text{s}$ |
|----------------------------|---------------------------|-------------------------------|---|--|-------------------|
| 50                         | 250                       | 3.9                           | –   | 11   | 40                |
| 100                        | 200                       | 4.1                           | 65  | 101  | 39                |
| 150                        | 150                       | 3.8                           | 123                                       | 165  | 42                |
| 200                        | 100                       | 3.4                           | 162                                       | 204  | 46                |
| 250                        | 50                        | 3.1                           | 184                                       | 225  | 51                |
| 300                        | 0                         | 2.8                           | 194                                       | 235  | 58                |

at  $J = 12$ . The rotational quantum number  $J = 12$  was also optimal for all other mixtures. This is because under the conditions of a thermal chain explosion, the lasing occurs at high temperatures (greater than 1000 K) and, therefore, at higher values of  $J$  (greater than 10).



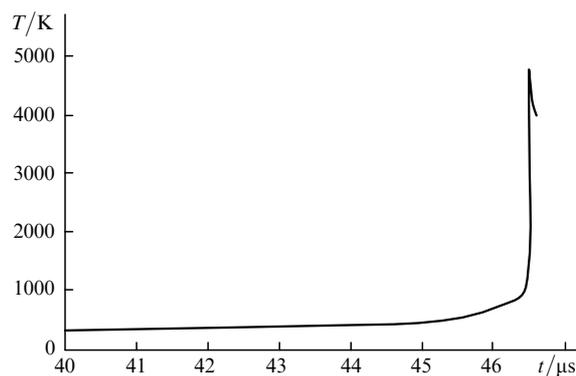
**Figure 1.** Calculated specific energy output of the hydrogen–fluorine thermal-chain explosion laser in the mixture  $\text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} = 200 : 400 : 40 : 100$  Torr as a function of the rotational quantum number  $J$  of the generated radiation.

Fig. 2 shows the calculated temporal dependence of the output intensity of the hydrogen–fluorine laser in the case of a thermal chain reaction in the active mixture  $\text{HF} : \text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} : \text{F}_2\text{SO}_3 = 1 : 200 : 400 : 40 : 100 : 40$  Torr. One can see from Fig. 2 that the duration of the laser pulse is rather small ( $\sim 20$  ns), which makes it important to take the deviation from the rotational equilibrium into account in calculating the specific energy output. Fig. 3 shows the temporal dependence of the temperature of the gas medium. The output power of the considered laser increases strongly when the temperature of the laser mixture reaches 1200–1300 K.



**Figure 2.** Temporal dependence of the output intensity of the hydrogen–fluorine laser in the case of a thermal-chain reaction in the mixture  $\text{HF} : \text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} : \text{F}_2\text{SO}_3 = 1 : 200 : 400 : 40 : 100 : 40$  Torr.

In this work, we have optimised the hydrogen–fluorine thermal-chain explosion laser over the laser mixture composition and the rotational quantum number of the generated radiation. The explosion is initiated by resonant



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

vibrational excitation of HF molecules by multiline emission from a hydrogen–fluorine laser. We have shown that, in the original mixture  $\text{HF} : \text{H}_2 : \text{F}_2 : \text{O}_2 : \text{He} : \text{F}_2\text{SO}_3 = 1 : 200 : 400 : 40 : 100 : 40$  Torr initiated by a pulse of an  $\text{H}_2\text{-F}_2$  laser, the energy output reaches more than  $200 \text{ J litre}^{-1}$  for the optimal rotational number of 12. Importantly, most of the energy is emitted in a short pulse, about 10 ns long. This regime is of interest for plasma research and the technology of laser-shock strengthening of alloys. In this case, the generation of short pulses does not require costly and bulky accelerating equipment.

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