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Pulsed $H_2 - F_2$ laser with simultaneous lasing on rotational and vibrational – rotational transitions

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***Abstract*. The possibility of operation of a pulsed chemical $H_2 - F_2$ laser on purely rotational transitions with simultaneous lasing on vibrational – rotational transitions is theoretically investigated. The lasing of a hydrogen – fluorine laser (mixture pressure 1.1 atm) on both $\upsilon, j-1 \rightarrow \upsilon-1, j$ ($j = 2 - 20$) and $\upsilon, j \rightarrow \upsilon, j-1$ ($j = 12 - 19$) transitions has been modelled taking into account the resonant one- and two-quanta vibrational – rotational processes. The calculated specific energy extraction for purely rotational transitions reaches 0.3 – 0.5 J L^{-1} at $j = 18$ and 19 (wavelength 15.5 – 14 μm) and 0.4 – 0.7 J L^{-1} at $j = 13$ and 14 (wavelength 21 – 19 μm).**

Keywords: *pulsed chemical $H_2 - F_2$ laser, vibrational – rotational processes, purely rotational transitions.*

There is much interest in the study and development of IR lasers with radiation wavelengths above 10 μm because they can be used to separate isotopes, probe atmosphere, analyse semiconductor materials, etc. A new mechanism of pumping far-IR lasers on rotational transitions in halogen hydrides by preliminary excitation of their vibrational states was proposed in [1 – 4]. This pumping scheme takes into account that the vibrational relaxation of some diatomic hydrogen-containing molecules, including halogen hydrides, occurs through the vibrational – rotational – translational (VRT) mechanism [5 – 9]. A specific feature of this mechanism is the excitation of higher lying rotational states of these molecules via resonant energy transfer from the vibrational states of the same molecule. In particular, levels with quantum numbers $j \geqslant 14$ correspond to these rotational states in the HF molecule. Under certain conditions this excitation (with allowance for the slower thermal relaxation of the upper rotational levels in comparison with the lower ones) may lead to inverse population of the rotational states. The thermal excitation of the vibrational levels of HF molecules with their subsequent cooling under rapid expansion in vacuum was considered in [1, 2]. A chemical HF laser with a $F : F_2 : H_2 : He = 0.02 : 0.99 : 1 : 20$ mixture composition ($T = 300$ K, $p = 2700$ Pa) was numerically simulated in [10]. The calculation showed that, when the VRT relaxation dominates over the VT relaxation, lasing with a specific energy of ~ 0.1 J L^{-1} may occur on rotational transitions in a chemical $H_2 - F_2$ laser. This lasing was observed in the experiments [11 – 20] and in many other studies. For example, an atmospheric-pressure chemical laser with the ratio $F_2 : H_2 : He = 1 : 1 : 9$ of the main components in the gas mixture was investigated in [11]. It was found that about 10 % of the output energy covers the long-wavelength range ($\lambda > 15$ μm), which corresponds to purely rotational transitions. Lasing on rotational transitions in different hydrogen-containing molecules (OH, OD, NH, and HF) was obtained in [14 – 17] as a result of studying various mixtures. The VRT relaxation was theoretically found to be dominant for them. In the overwhelming majority of experiments the strong rotational excitation was explained by the direct excitation of rotational states during VRT relaxation.

The purpose of this study was to theoretically investigate the possibility of effective lasing on purely rotational transitions of HF molecules in a pulsed chemical $H_2 - F_2$ laser. A multilevel model of a pulsed hydrogen – fluorine laser was developed in [21]; this model allows one to investigate the lasing (amplification of radiation) on each individual vibrational – rotational transition $\upsilon, j-1 \rightarrow \upsilon-1, j$ with allowance for the nonequilibrium population of the rotational sublevels of HF molecules. An application of this model made it possible to calculate the spectral and temporal characteristics, as well as the energy parameters of the $H_2 - F_2$ laser on vibrational – rotational transitions, which showed good agreement with the experimental data [21]. We modified this model in [22] by taking into account the VRT relaxation of HF molecules. Here, we simulated the lasing of a pulsed chemical $H_2 - F_2$ laser on purely rotational transitions, taking into account the one-quantum resonant VR transitions. It was assumed for simplicity that simultaneous lasing on vibrational – rotational transitions is absent. The calculations for a working mixture of an $H_2 - F_2$ laser with a total pressure of 1 atm showed the possibility of effective lasing on the $\upsilon, j \rightarrow \upsilon, j-1$ transitions with $\upsilon = 1 - 4$ and $j = 14 - 12$. The calculated specific energy extraction for lasing on purely rotational transitions with wavelengths above 19 μm for $\upsilon = 2$ and $j = 14$, 13 and 12 was, respectively, 1.4, 0.9, and 0.5 J L^{-1}. There is a hypothesis about the important role of multiple quanta VR transitions, in which higher rotational levels are excited [6]. In particular, two-quantum VR processes allow for excitation of rotational levels of HF molecules with $j \leqslant 19$; hence, lasing

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with a wavelength of 14 μm and longer may occur beginning with these levels. In this study we simulated the lasing of a pulsed chemical H_2-F_2 laser, taking into account one- and two-quanta resonant VR processes. The case of simultaneous lasing on purely rotational and vibrational–rotational transitions of HF molecules was analysed, which corresponds to real experimental conditions.

Let us consider an H_2-F_2 laser generating simultaneously on the vibrational–rotational transitions $\upsilon,j-1 \to \upsilon-1,j$ and purely rotational transitions $\upsilon,j \to \upsilon,j-1$. First, we write the equations for the populations of the rotational sublevels of HF molecules, which are similar to the equations for a hydrogen–fluorine laser operating on only vibrational–rotational transitions [21], additionally taking into account one- and two-quanta resonant VR transitions $[\mathrm{HF}(\upsilon+1,j_1)+\mathrm{M} \to \mathrm{HF}(\upsilon,j)+\mathrm{M}$ or $\mathrm{HF}(\upsilon+2,j_2)+\mathrm{M} \to \mathrm{HF}(\upsilon,j)+\mathrm{M}]$:

$$\frac{\mathrm{d}n_\upsilon^j}{\mathrm{d}t} = -\frac{\alpha_{\upsilon,j}I_{\upsilon,j}}{h\nu_{\upsilon,j}} + \frac{\alpha_{\upsilon,j+1}I_{\upsilon,j+1}}{h\nu_{\upsilon,j+1}} - \frac{g_{\upsilon,j+1}Y_{\upsilon,j+1}}{hw_{\upsilon,j+1}}$$

$$+\frac{g_{\upsilon+1,j}Y_{\upsilon+1,j}}{hw_{\upsilon+1,j}} + \frac{n_\upsilon - n_\upsilon^j}{M_{\upsilon,j}\tau_{\upsilon,j}} - \frac{n_\upsilon^j}{\tau_{\upsilon,j}} + R_{\upsilon,j}. \qquad (1)$$

Here, n_υ^j is the concentration of HF molecules on the υth vibrational and jth rotational levels; n_υ is the total population of the υth vibrational level of HF; $\alpha_{\upsilon,j} = \sigma_{\upsilon,j}[n_\upsilon^j - n_\upsilon^{j-1}(2j+1)/(2j-1)]$; $g_{\upsilon,j} = \sum_{\upsilon,j}[n_\upsilon^{j-1} - n_{\upsilon-1}^j \times(2j-1)/(2j+1)]$; $\sigma_{\upsilon,j}$ and $\sum_{\upsilon,j}$ are the induced-emission cross sections for the $\upsilon,j \to \upsilon,j-1$ and $\upsilon,j-1 \to \upsilon-1,j$ transitions, respectively; $I_{\upsilon,j}$ and $Y_{\upsilon,j}$ are the intracavity lasing intensities for these transitions; $\nu_{\upsilon,j}$ and $w_{\upsilon,j}$ are the corresponding radiation frequencies; $\tau_{\upsilon,j}$ is the characteristic rotational relaxation time in the rotational reservoir model [23]; $M_{\upsilon,j} = (2j+1)^{-1}(T/Q_\upsilon)\exp[j(j+1)Q_\upsilon/T]-1$; Q_υ is the characteristic rotational temperature of HF molecule for the υth vibrational level; and $R_{\upsilon,j}$ is the term taking into account the resonant VR processes (if the corresponding transition to this sublevel occurs). The rotational relaxation time $\tau_{\upsilon,j}$ is related to the rates $k_\upsilon^{\mathrm{M}}(j \to k)$ of the RT processes of the $\mathrm{HF}(\upsilon,j)+\mathrm{M} \to \mathrm{HF}(\upsilon,k)+\mathrm{M}$ type by the expression

$$\frac{1}{\tau_{\upsilon,j}} = \sum_{k,\mathrm{M}} k_\upsilon^{\mathrm{M}}(j \to k)N_{\mathrm{M}},$$

where N_{M} is the concentration of the M component in the laser mixture. The change in the intensities $I_{\upsilon,j}$ and $Y_{\upsilon,j}$ is described by the rate equations

$$\frac{\mathrm{d}I_{\upsilon,j}}{\mathrm{d}t} = c(\alpha_{\upsilon,j}-g)I_{\upsilon,j} + V_{\upsilon,j}, \qquad (2)$$

$$\frac{\mathrm{d}Y_{\upsilon,j}}{\mathrm{d}t} = c(g_{\upsilon,j}-G)Y_{\upsilon,j} + W_{\upsilon,j}. \qquad (3)$$

Here, g and G are the threshold gains for lasing on, respectively, purely rotational and vibrational–rotational transitions in HF and $V_{\upsilon,j}$ and $W_{\upsilon,j}$ are the terms taking into account the spontaneous emission. Equations (2) and (3) must be solved along with Eqns (1), the equations for the populations n_υ of the vibrational levels of HF molecules ($\upsilon = 0,\ 1,...,7$), the chemical kinetics equations, and the equations for the mean reserve of vibrational quanta of H_2 and the gas-medium temperature. The chemical processes in the $H_2-F_2-O_2-\mathrm{He}$ mixture that were considered in the calculations, the corresponding rate constants, and the other calculation parameters were reported in [24].

Specific calculations were performed for the $H_2:F_2:O_2:\mathrm{He} = 1:3:0.3:7$ mixture ($p = 1.1$ atm) at the initiation level set by the initial concentration of free atoms $N_{\mathrm{a}} = 3\times10^{16}$ cm^{-3} [21]. The rate constants of the RT processes, $k_\upsilon^{\mathrm{M}}(j \to k)$, were described by the expressions with an exponential dependence of $k_\upsilon^{\mathrm{M}}(j \to k)$ on the change in the rotational energy, which were used in [21], with the same numerical values of the coefficients. For simplicity the calculations were performed for only the following one- and two-quanta resonant VR processes:

$$\mathrm{HF}(\upsilon,3)+\mathrm{M} \to \mathrm{HF}(\upsilon-1,14)+\mathrm{M}, \qquad (4)$$

$$\mathrm{HF}(\upsilon,1)+\mathrm{M} \to \mathrm{HF}(\upsilon-2,19)+\mathrm{M}, \qquad (5)$$

where $\mathrm{M} = \mathrm{HF}$ or H_2. It was assumed that the rate constant of process (4) doubles that of process (5), and their summary values are assumed to be equal to the corresponding rate constants of the vibrational–translational relaxation of HF molecules upon collisions with HF and H_2 molecules [24]. In this case, lasing on purely rotational transitions in HF may occur on the cascade of $\upsilon,19 \to \upsilon,18$; $\upsilon,18 \to \upsilon,17\ldots$ transitions with wavelengths of ~ 14, ~ 15.5 μm, respectively. There are no experimental data on the induced-emission cross sections for the $\upsilon,j \to \upsilon,j-1$ transitions ($j = 12-19$) in HF molecules; therefore, their values were varied from 10^{-15} to 10^{-14} cm^2 in the calculations. The calculated values of specific extracted laser energy for the aforementioned purely rotational transitions at $\sigma_{\upsilon,j} = 10^{-15}$ and 10^{-14} cm^2 were nearly the same.

Table 1 contains the calculated values of specific energy extraction for a chemical H_2-F_2 laser working on purely rotational transitions ($\upsilon,j \to \upsilon,j-1$ ($\upsilon = 0-3, j = 19-12$) at $\sigma_{\upsilon,j} = 10^{-15}$ cm^2. The results of similar calculations in the absence of simultaneous lasing on the vibrational–rotational transitions (with large G value) are also presented. It can be seen that the calculated specific energy extraction ε_j in the case of lasing on the $\upsilon,j \to \upsilon,j-1$ transitions is 0.3–0.5 J L^{-1} for $\upsilon = 0-1$ and $j = 18-19$ ($\lambda \approx 14-15.5$ μm) and 0.4–0.7 J L^{-1} for $\upsilon = 2-3$ and $j = 13-14$ ($\lambda \approx 19-21$ μm). Note that the absence of lasing on the vibrational–rotational transitions increases the calculated values of energy extraction for purely rotational transitions by 20%–40%. As can be seen in Table 1, the ε_j values gradually decrease in the cascades of $\upsilon,19 \to \upsilon,18$; $\upsilon,18 \to \upsilon,17\ldots$ and $\upsilon,14 \to \upsilon,13$; $\upsilon,13 \to \upsilon,12\ldots$ transitions, beginning with the levels with $j = 19$ and 14 [to which energy is pumped as a result of resonant VR processes (4) and (5)]. This is caused by the energy loss due to the RT relaxation, as a result of which the energy emitted on the $\upsilon,j \to \upsilon,j-1$ transition is only partially re-emitted on the $\upsilon,j-1 \to \upsilon,j-2$ transition in the aforementioned cascades. As a result, for example, the ε_{16} values are fairly small. The total specific energy extraction was 6.8 J L^{-1} for the $\upsilon,j \to \upsilon,j-1$ ($\upsilon = 0-3, j = 12-19$) transitions and 65 J L^{-1} on the vibrational–rotational $\upsilon,j-1 \to \upsilon-1,j$ ($\upsilon = 1-3$, $j = 2-20$) transitions. Figure 1 shows the calculated time dependences of the specific lasing power P_j for the $1,j \to 1,j-1$ transitions with $j = 14$ and 19. It can be

Table 1.

v	ε_{19}/J L^{-1}	ε_{18}/J L^{-1}	ε_{17}/J L^{-1}	ε_{16}/J L^{-1}	ε_{14}/J L^{-1}	ε_{13}/J L^{-1}	ε_{12}/J L^{-1}
0	0.47 (0.47)	0.28 (0.3)	0.16 (0.17)	0.07 (0.07)	0.21 (0.17)	0.08 (0.06)	– –
1	0.48 (0.61)	0.32 (0.43)	0.17 (0.28)	0.08 (0.13)	0.5 (0.56)	0.23 (0.29)	0.06 (0.13)
2	0.34 (0.5)	0.23 (0.37)	0.13 (0.25)	0.04 (0.12)	0.7 (0.84)	0.37 (0.51)	0.17 (0.29)
3	0.25 (0.36)	0.17 (0.26)	0.11 (0.18)	0.05 (0.09)	0.6 (0.82)	0.34 (0.51)	0.17 (0.3)

Note: ε_j is the specific laser energy extraction, calculated for the $v,j \rightarrow v, j-1$ transition; the values in parentheses are the results of the calculation in the absence of lasing on the vibrational–rotational transitions.

seen that lasing on these transitions in the absence of lasing on the vibrational–rotational transitions begins somewhat earlier (by 50 ns). This is explained by the more rapid increase in the gas mixture temperature, because the energy is not spent on lasing in the wavelength range of 2.5–3 μm.

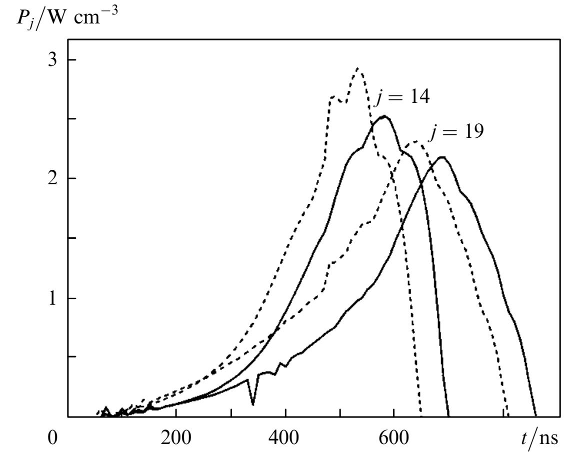

Figure 1. Time dependences of the specific lasing power for an $H_2 - F_2$ laser on purely rotational transitions $1, 14 \rightarrow 1, 13$ and $1, 19 \rightarrow 1, 18$, with simultaneous lasing on the vibrational–rotational transitions (solid lines) and in the absence of lasing on these transitions (dashed lines).

Thus, we simulated the work of a pulsed chemical $H_2 - F_2$ laser with simultaneous lasing on the $v,j-1 \rightarrow v-1,j$ ($j = 2-20$) and $v,j \rightarrow v,j-1$ ($j = 12-19$) transitions, with allowance for the resonant one- and two-quanta VR processes. Specific calculations were performed for the $H_2{:}F_2{:}O_2{:}He = 1{:}3{:}0.3{:}7$ mixture with a total pressure of 1.1 atm at the initiation level set by the initial concentration of free atoms $N_a = 3 \times 10^{16}$ cm^{-3}. The calculated specific energy extraction for lasing on purely rotational transitions was 0.3–0.5 J L^{-1} for the transitions with wavelengths $\lambda \approx 17 - 14$ μm ($v = 0-1$, $j = 18-19$) and 0.4–0.7 J L^{-1} for the transitions with wavelengths of 21–19 μm ($v = 2-3$, $j = 13-14$). Suppression of the lasing on vibrational–rotational transitions increased the energy extraction for purely rotational transitions by 20%–40%.

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