Quantum Electronics **34** (4) 320 – 324 (2004)

PACS numbers: 42.55.Ks; 42.60.Jf; 42.60.Lh
DOI: 10.1070/QE2004v034n04ABEH002674

Spectral characteristics of nonchain HF and DF electric-discharge lasers in efficient excitation modes

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***Abstract*. The spectral characteristics of efficient nonchain HF and DF chemical lasers are studied. It is found that the emission spectra of nonchain lasers operating with high efficiency are strongly broadened. Almost 30 emission lines of an HF laser and cascade lasing on the $\boldsymbol{v}(3-2) \rightarrow \boldsymbol{v}(2-1) \rightarrow \boldsymbol{v}(1-0)$ vibrational transitions of HF molecules for a number of rotational lines are obtained. It is shown that the development of discharge inhomogeneities significantly reduces the number of lasing lines in the spectra of nonchain chemical lasers. For an SF_6-D_2 mixture excited by a generator with an inductive storage, about 40 lasing lines are observed on four vibrational transitions of DF molecules and the $\boldsymbol{v}(4-3) \rightarrow \boldsymbol{v}(3-2) \rightarrow \boldsymbol{v}(2-1) \rightarrow \boldsymbol{v}(1-0)$ cascade lasing is obtained at several rotational lines. Nonchain HF and DF electric-discharge lasers with a total and intrinsic efficiency of up to 6 % and 10 %, respectively, pumped from capacitive and inductive generators are developed.**

Keywords: *nonchain HF and DF lasers, emission spectrum, cascade lasing.*

1. Introduction

Studies of nonchain HF and DF electric-discharge lasers and discharges in SF_6-based mixtures currently attract considerable attention [1 – 7]. However, most of the published works are devoted to studying the effect of various experimental factors on the energy characteristics and efficiency of these lasers. The data on the spectra of HF-laser radiation under various excitation conditions are poor, and the data on the DF-laser spectra are virtually absent, while the understanding of the processes occurring in the working mixtures of nonchain chemical lasers requires that the spectral characteristics of laser radiation be studied.

It is known that a population inversion in nonchain lasers is produced in exothermic chemical reactions [8]

$$F + H_2 \rightarrow HF^*(\upsilon \leqslant 3) + H + 139 \text{ kJ mol}^{-1}, \quad (1)$$

$$F + D_2 \rightarrow DF^*(\upsilon \leqslant 4) + D + 133 \text{ kJ mol}^{-1}, \quad (2)$$

where υ is a vibrational level of HF or DF molecules populated in this chemical reaction. Atomic fluorine forms upon the SF_6 dissociation in an electric discharge, and the maximum fraction of the chemical energy is spent on the population of the vibrational levels of HF molecules with $\upsilon = 2$ [9] and DF molecules with $\upsilon = 3$ [8].

Transitions from three excited vibrational levels (the P_3, P_2 and P_1 transitions) are observed in the emission spectra of HF lasers. The number of individual lasing lines may vary within wide limits. As the cavity Q factor increases, up to 20 lasing lines can be observed [10]. About 30 lines were obtained in a laser with an active length of 2 m at a mixture pressure of < 10 Torr [11], but the output energy and efficiency of this laser were very low. The spectrum of a nonchain HF laser consists mainly of 10 – 15 lines, and most of the energy is on the P_2 transition [10, 12 – 16]. A considerable spread in the time of onset of lasing on different vibrational – rotational transitions is observed, and the integral laser pulse has a complex structure consisting of spikes.

Lasing on three lines of the P_4 transition was observed in Ref. [17] using a high-Q cavity with freons – deuterium mixtures pumped by a longitudinal discharge 2 m long. In SF_6-D_2 mixtures excited by a transverse discharge, lasing on the P_4 transition was achieved only at a low cavity loss [18]. As the Q factor decreased, lasing was obtained only on the P_3, P_2, and P_1 transitions. The number of detected vibrational – rotational lasing lines was 15 – 27 [18, 19]. Note that a significant part of energy can be emitted due to cascade transitions [20], and the resulting laser pulse has two or more spikes, as for an HF-laser pulse.

Efficient regimes of exciting nonchain HF lasers providing an intrinsic efficiency of up to 10 % were attained by us earlier [21, 22]. The objective of this work is a detailed study of the spectral characteristics of efficient HF and DF lasers under various conditions of their initiation with a self-sustained discharge.

2. Experimental

The electric-discharge laser used in our experiments and described in detail in Ref. [22] had an active length of 72 cm and a 3.8-cm interelectrode gap between profiled polished

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Received 11 July 2003; revision received 11 November 2003
Kvantovaya Elektronika **34** (4) 320 – 324 (2004)
Translated by A.S. Seferov

stainless-steel electrodes. The laser cavity was composed of a flat aluminium mirror and the exit KPC-5 or KPC-6 plate. Inductive and capacitive energy-storage units were used to pump the laser. The inductive storage ensured a discharge-current duration of ~ 100 ns, an energy input of 15–100 J L^{-1}, and an intrinsic efficiency of HF and DF lasers of up to 10 % and 7 %, respectively. The capacitance of the capacitive storage C_0 was 70 or 13 nF. The discharge-current duration and energy input in the first and second cases were 200 ns, 10–150 J L^{-1} and 100 ns, 10–50 J L^{-1}, respectively. The pump generators provided total efficiencies of the HF and DF lasers of up to 6 % and 5% and intrinsic efficiencies of up to 10 % and 7 %, respectively. The lasing characteristics were studied in mixtures with a composition of $SF_6 : H_2(D_2) = 8 : 1$ at pressures of < 70 Torr. The laser output energy was measured using an IKT-1N or OPHIR calorimeter with an FL-250A-EX measurement head. The radiation pulse power and shape were measured by an FSG-22 photodetector cooled with liquid nitrogen. The spectral characteristics of radiation were determined using an MDR-12 monochromator and an FSG-22 photodetector. The amplitude–time characteristics of the discharge were recorded using voltage dividers, Rogowsky coils, and shunts. Electric signals were fed to a TDS-220 or TDS-224 oscilloscope.

3. Lasing spectra of HF and DF lasers in high-efficiency operating modes

Figure 1 shows the lasing spectrum of the HF laser pumped from a capacitive storage with $C_0 = 13$ nF at a specific energy input of 25 J L^{-1} and a 6 % total laser efficiency: 21 lasing lines were obtained. The lines of the $\upsilon(1-0)$ vibrational transition had the maximum intensity. Lasing was initiated on the P_2 lines, and the P_1 and P_3 lines appeared 20–40 ns later. The delay time of the appearance of lines of the $\upsilon(3-2)$ and $\upsilon(2-1)$ transitions slightly increases with an increase in the rotational quantum number j. The laser pulse duration at each transition also increases. Subsequently, lasing continues simultaneously at all of the vibrational–rotational transitions. The intensity of a multifrequency laser pulse rises during the pump pulse, after which the radiation intensity exponentially decays for > 1500 ns at a laser-pulse half-height duration of 300 ns. The DF-laser pulse has the same profile, and its total duration is longer by ~ 0.5 μs. The experimental results for the HF laser are shown in Fig. 2.

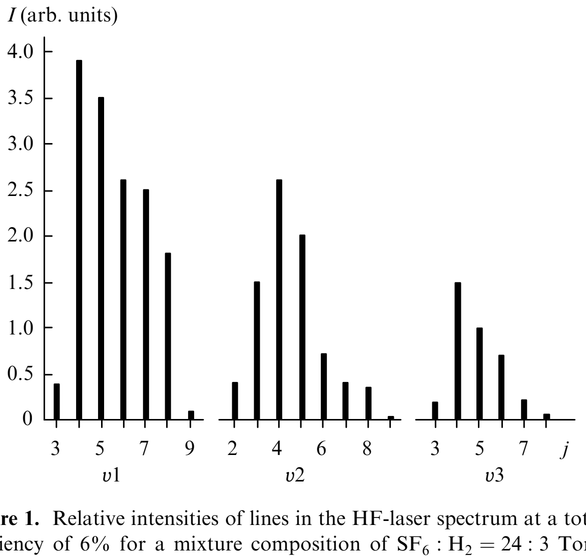

Figure 1. Relative intensities of lines in the HF-laser spectrum at a total efficiency of 6% for a mixture composition of $SF_6 : H_2 = 24 : 3$ Torr. The laser is pumped from the capacitive storage at $C_0 = 13$ nF, a charging voltage $U_0 = 30$ kV, and a specific energy input $E_{in} = 25$ J L^{-1}.

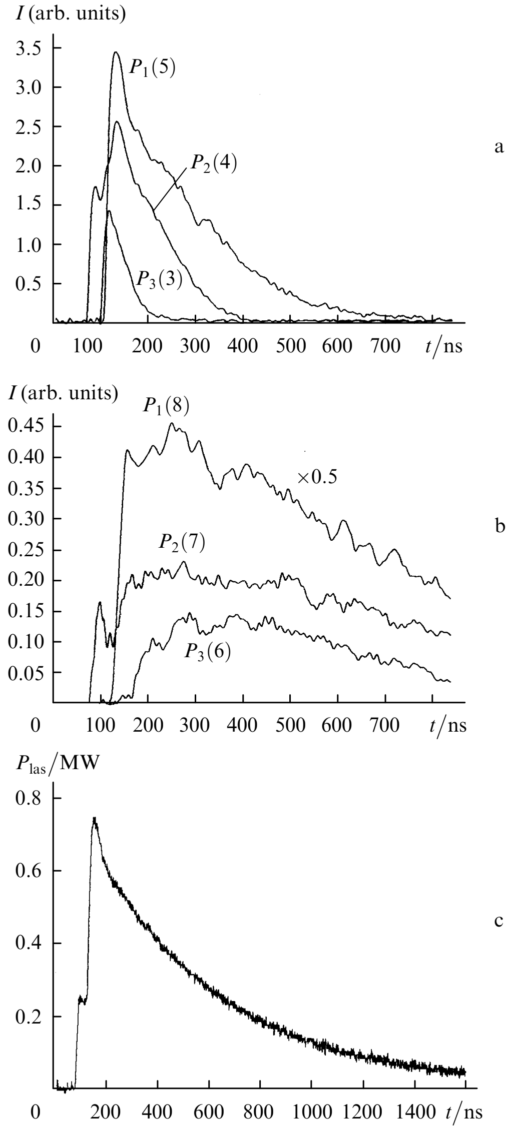

Figure 2. (a) $P_3(3) \rightarrow P_2(4) \rightarrow P_1(5)$ and (b) $P_3(6) \rightarrow P_2(7) \rightarrow P_1(8)$ cascade transitions in the HF-laser spectrum; (c) a multifrequency laser pulse for a mixture of $SF_6 : H_2 = 24 : 3$ Torr efficiently pumped from the capacitive storage at $C_0 = 13$ nF and $U_0 = 30$ kV. The laser gap is broken down at a moment $t = 0$.

The lasing energy distribution over the vibrational transitions has the form $Q(P_1):Q(P_2):Q(P_3) = 7:3:1$. Approximately 60 % of the emitted energy was contained in two lines, $P_1(7)$ and $P_1(8)$. This energy distribution differs significantly from the data obtained in Refs [10, 12–16] and can be associated with intense cascade transitions, which enhance the efficiency of energy extraction from the active medium of a nonchain chemical laser ([20] and Fig. 2). We see that the lines for the transition from the $P_3(j-1)$ level are initiated close to the line peak for the transition from the

$P_2(j)$ level. When lasing appears on the P_1 vibrational transition, the radiation intensity of the P_2 lines begins to grow. More than 85 % of the total energy is emitted in the cascades that terminate in the most intense lines of the $\upsilon(1-0)$ transition.

Figure 3 shows oscillograms of laser pulses from the $P_1(6)$ level at various excitation energies for $C_0 = 13$ nF. As the energy deposited in the active medium of the HF laser increases, the lasing duration for this transition shortens. The relative lasing energies for this transition at specific pump energies of 36, 25, and 18 J L^{-1} were 0.18, 0.65, and 1, respectively. This behaviour of the laser pulse was also observed at other vibrational–rotational transitions for $j < 6$. The total laser-pulse duration also decreased. This was accompanied by an increase in the output energy and extension of the lasing spectrum due to the appearance of lines with larger j. Increasing the energy dissipated in the gas led to an increase in the gain on various transitions and, consequently, to shorter pulse durations due to a fast inversion removal. Decreasing the pump energy (< 25 J L^{-1}) caused a significant spread in the time of the lasing onset for various transitions, the lasing efficiency decreased, and a peaky structure began to appear in the integral laser pulse.

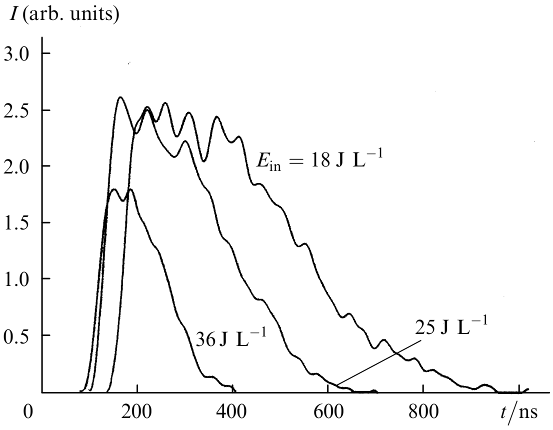

Figure 3. Shapes of laser pulses for the transition from the $P_1(6)$ level at various energies E_{in} deposited in the active medium. A mixture with a composition of $SF_6 : H_2 = 24 : 3$ Torr is pumped from the capacitive storage at $C_0 = 13$ nF.

Figure 4 shows the emission spectrum of the HF laser efficiently pumped from the capacitive storage with $C_0 = 70$ nF and a specific energy input of 90 J L^{-1}. In this case, the output energy was 1.3 J and the total laser efficiency was 4 %. Compared to the experiment with $C_0 = 13$ nF, the number of lasing lines increased to 25 primarily due to the appearance of intense lines with rotational quantum numbers $j \geqslant 9$. At the same time, the lasing lines on the $\upsilon(1-0)$ transition with $j \leqslant 5$ disappear. This can be linked to an increase in the gas temperature accompanying an increase in the energy input and to populating the rotational levels of the HF molecular ground state [23]. An HF-laser emission spectrum close to that in Fig. 4 was observed in Ref. [24] for an $SF_6 - H_2$ mixture initiated by an electron beam, which provided highly homogeneous pumping and a high lasing efficiency, and an energy deposition of 120 J L^{-1}. An HF laser efficiently pumped from an inductive storage yielded up to 30 lasing lines [22].

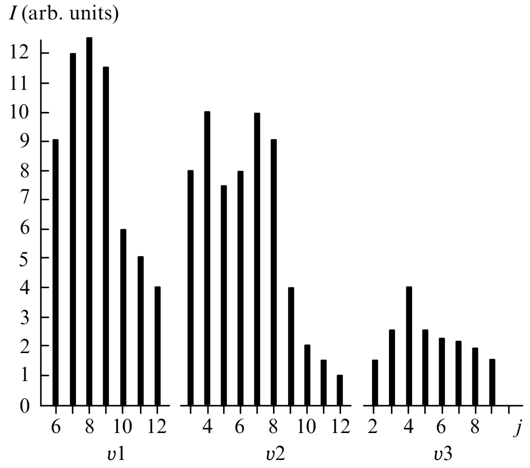

Figure 4. Emission spectrum of the HF laser with a mixture of $SF_6 : H_2 = 36 : 4.5$ Torr efficiently pumped from the capacitive storage: $C_0 = 70$ nF, $U_0 = 30$ kV, and $E_{in} = 90$ J L^{-1}.

To study the effect of the discharge homogeneity on the laser spectral characteristics, one of the profiled electrodes was replaced by a semicircular electrode with a 20-mm radius. This substitution resulted in a discharge-homogeneity loss already 80 ns after a breakdown of the laser gap. The moment of contraction was identified by an abrupt discharge-current rise and a voltage drop across the laser gap. Note that the laser output energy decreased to 0.25 J, while the specific pump energy remained constant. The lasing spectrum obtained under such conditions is shown in Fig. 5. A disturbance of the discharge homogeneity unambiguously leads to a reduced number of lasing lines primarily due to the disappearance of laser transitions with $j > 9$. In this case, only four lines remain in the $\upsilon 1$ band, the intensities of the lines in the $\upsilon 3$ band fall significantly, and the maximum fraction of energy is emitted in the $\upsilon 2$ band. Lasing in $\upsilon 1$ and $\upsilon 3$ bands is delayed relative to lasing in $\upsilon 2$ by 80–100 ns, thus leading to the appearance of two peaks in the integral pulse (Fig. 6). This is accom-

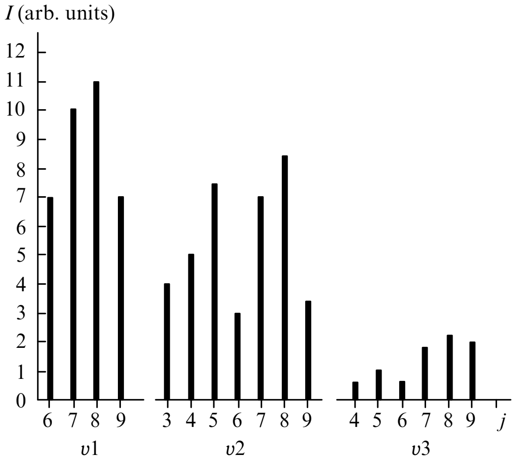

Figure 5. Emission spectrum of the HF laser for an inhomogeneous discharge in a mixture of $SF_6 : H_2 = 36 : 4.5$ Torr pumped from the capacitive storage: $C_0 = 70$ nF, $U_0 = 30$ kV, and $E_{in} = 90$ J L^{-1}.

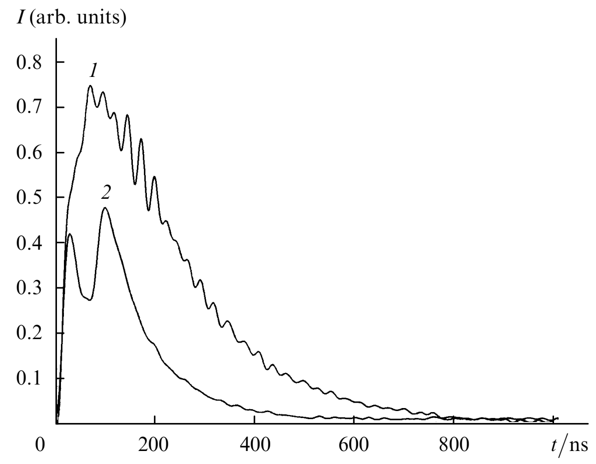

Figure 6. Integral radiation pulses of the HF laser with a mixture of $SF_6 : H_2 = 36 : 4.5$ Torr (*1*) in an efficient excitation mode and (*2*) at a disturbed discharge homogeneity: $C_0 = 70$ nF, $U_0 = 30$ kV, and $E_{in} = 90$ J L^{-1}.

panied by an almost twofold decrease in the laser-pulse duration, since the lines with large rotational quantum numbers j that have the maximum duration disappear from the lasing spectrum.

Figure 7 shows the spectrum of the DF laser efficiently pumped from the inductive storage, and Fig. 8 shows the time-dependent behaviour of individual lasing lines of DF molecules. In this excitation mode, the output energy was 0.7 J and the intrinsic efficiency reached 6.5 %. Laser radiation was obtained in four vibrational bands of DF molecules. The maximum intensities were observed in the $\upsilon1$ and $\upsilon2$ bands, and the total number of lasing lines was 37. For close parameters of the pump pulse and active lengths of the laser and cavity and in the presence of discharge inhomogeneities, the maximum number of individual DF-laser lines observed in Ref. [18] was 27. The lasing threshold was reached first for the lines of the $\upsilon3$ band. Lasing in other bands arose 20 – 80 ns later, and simultaneous lasing was observed at all of 37 lines. The delay times of appearance of individual lines slightly increased with an increase in the rotational quantum number j. The maximum duration of lasing at individual lines was 1500 ns for $j = 9 - 10$ and then slightly decreased. The integral laser pulse had a single peak and had a profile similar to that of the HF laser (Fig. 2c).

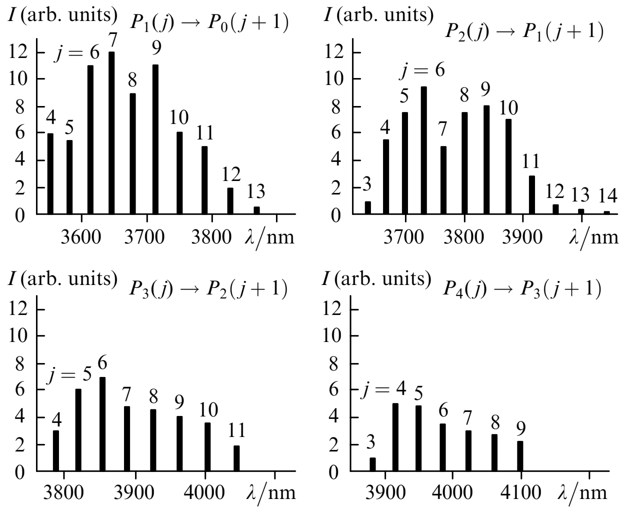

Figure 7. Oscillation spectrum of the DF laser with a mixture of $SF_6 : D_2 = 36 : 4.5$ Torr efficiently pumped from the inductive storage: $C_0 = 70$ nF, $U_0 = 30$ kV, and $E_{in} = 45$ J L^{-1}.

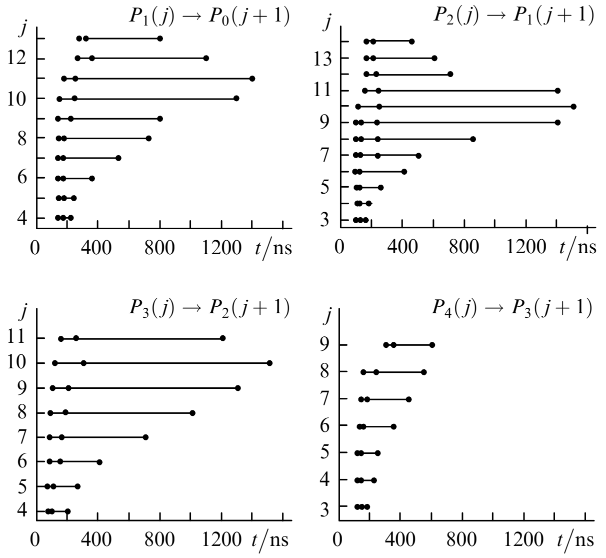

Figure 8. Time-dependent behaviour of laser transitions of the DF laser with a mixture of $SF_6 : D_2 = 36 : 4.5$ Torr efficiently pumped from the inductive storage: $C_0 = 70$ nF, $U_0 = 30$ kV, and $E_{in} = 45$ J L^{-1}. Points denote the onset, intensity peaks, and the end of lasing at individual lines. Moment $t = 0$ corresponds to a breakdown of the laser gap.

Similar to the case of the HF laser [20], the interaction of a number of vibrational – rotational transitions is observed [the $P_4(j) \to P_3(j+1) \to P_2(j+2) \to P_1(j+3)$ cascade lasing]. Figure 9 shows the profiles of laser pulses for cascade lasing. As is seen, lasing on the lines of the $\upsilon4$ band is initiated close to the intensity peaks of the corresponding $\upsilon3$ lines. This may be related to a small fraction of the chemical energy spent on the formation of excited DF* molecules ($\upsilon = 4$), and an inversion for lines of the $\upsilon4$ band can appear only upon lasing on the coupled lines in the $\upsilon3$ band [8, 10]. In addition, lines in the $\upsilon3$ and $\upsilon2$ bands could have second peaks near the lasing maxima of the coupled lines in the $\upsilon2$ and $\upsilon1$ bands, respectively.

4. Conclusions

The spectral characteristics of radiation of HF and DF chemical lasers have been studied upon efficient excitation at specific pump energies of 20 – 100 J L^{-1}. It is shown that the oscillation spectra of nonchain lasers operating in high-efficiency modes widen significantly. Simultaneous lasing on 30 vibrational – rotational transitions of three vibrational bands is obtained for the HF laser. Cascade lasing on the $\upsilon(3-2) \to \upsilon(2-1) \to \upsilon(1-0)$ vibrational transitions of HF molecules is obtained at several rotational lines, and > 85 % of the total energy can be emitted in the cascades.

It is found that, if the discharge homogeneity is disturbed, the number of lasing lines decreases, the spread in the time of the lasing onset at individual lines increases, and the output energy becomes several times lower. A discharge contraction is a factor that shortens multifrequency laser pulses and results in the formation of their peaky profile.

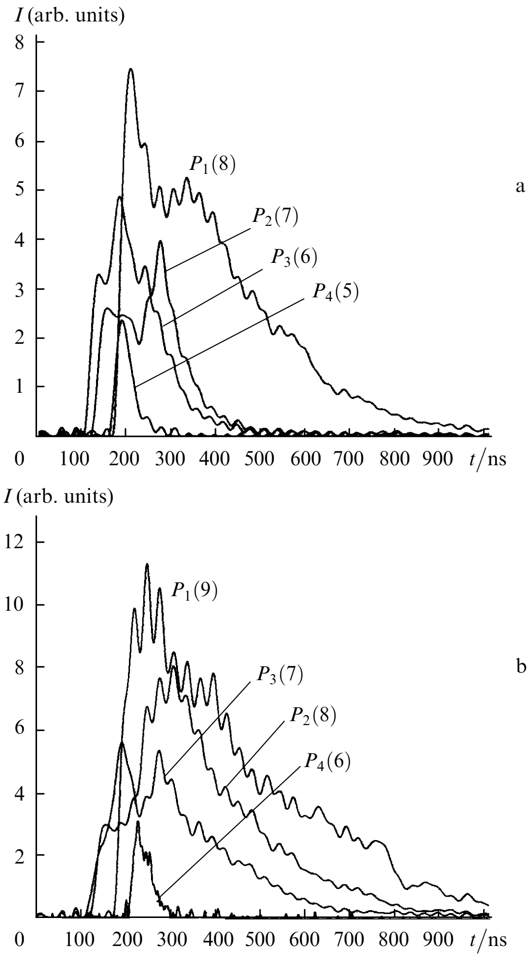

Figure 9. (a) $P_4(5) \rightarrow P_3(6) \rightarrow P_2(7) \rightarrow P_1(8)$ and (b) $P_4(6) \rightarrow P_3(7) \rightarrow P_2(8) \rightarrow P_1(9)$ cascade transitions in the spectrum of the DF laser with a mixture of $SF_6 : D_2 = 36 : 4.5$ Torr efficiently pumped from the inductive storage: $C_0 = 70$ nF, $U_0 = 30$ kV, and $E_{in} = 45$ J L^{-1}. Moment $t = 0$ corresponds to a breakdown of the laser gap.

About 40 lasing lines on four vibrational transitions of DF molecules at $j < 11-14$ and the $v(4-3) \rightarrow v(3-2) \rightarrow v(2-1) \rightarrow v(1-0)$ cascade lasing on a number of rotational lines are obtained in an SF_6-D_2 mixture pumped from the inductive generator.

The following conclusion can be drawn from the experiments performed. A stable volume discharge formed in $SF_6-H_2(D_2)$ mixtures due to an intense preionisation and a uniform electrical field in the laser gap ensures a highly homogeneous active medium of a nonchain HF(DF) laser, thus leading to simultaneous lasing on 30–40 vibrational–rotational transitions at a rotational quantum number of $j \geqslant 10$. Intense cascade transitions that accompany these processes enhance the efficiency of extracting the energy from the active medium of a nonchain chemical laser and increase its efficiency.

Nonchain HF and DF electric-discharge lasers with total efficiencies of 6 % and 5 % and intrinsic efficiencies of up to 10 % and 7 %, respectively, pumped from capacitive and inductive generators are developed.

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