

Study of an electric-discharge molecular fluorine VUV laser

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Abstract. The effect of the discharge voltage, composition and pressure of the He–F₂ gas mixture on the output radiation parameters of a VUV F₂ laser is studied. It is shown that reaction of dissociative attachment of electrons to fluorine molecules plays a significant role in the process of active medium excitation. The condition for choosing the optimal mixture composition is determined. The use of a pump generator with multistage magnetic compression of high-voltage pulses and preionisation system with a barrier discharge at the surface of a ceramic tube ensures a high homogeneity of the gas discharge (without cathode spots). A small-size 2.5-mJ F₂ laser (with an active volume of ~9 cm³) with a pulse repetition rate of up to 1 kHz operating on a He–F₂ mixture at a pressure of up to 3500 mbar is developed.

Keywords: F₂ laser, dissociative attachment of electrons to fluorine molecules, oscillator with magnetic compression of high-voltage pulses, matched pump mode.

1. Introduction

A 157-nm molecular fluorine laser is one of the most powerful sources of VUV radiation [1–3]. At present, it has no alternatives even among diode-pumped lasers because of the absence of nonlinear crystals converting radiation to wavelength regions below 180 nm [4].

The laser considered in this study has unique properties. Its radiation is absorbed by almost all materials, which makes it possible to use this laser for processing even such materials as silica, Teflon, ultra-hard alloys and ceramics. An extremely short wavelength of the laser allows the fabrication of photonic structures of size smaller down to 100 nm, which is not possible with other high-precision processing equipment. It is proposed to use such structures

in near future for constructing multifunctional single-chip optical devices capable of replacing modern microelectronics devices [5–7]. The F₂ laser is used for writing fibre Bragg gratings (FBGs) in optical fibres used in telecommunications [8–10]. FBG interferometers are also used as sensors of physical quantities and their fields (temperature, pressure, electromagnetic fields, acoustic fields, etc.). Laser radiation at 157 nm is used in photolithographic microtechnology with a spatial resolution of less than 100 nm [1–4, 11]. Finally, the F₂ laser is indispensable in the preparation of components for microelectromechanical systems that are being used extensively in security appliances for intelligence data collection and for diagnostic purposes in medicine. Microelectromechanical systems are quite important for the rapidly growing market of mobile telephones [5].

The traditional approach towards the creation of high-power F₂ lasers is based on high-power pumping, which can be achieved by using an active medium with extremely high pressures (6–10 bar). We have proposed in [2, 3] another approach based on increasing the laser pump power by optimising the conditions of energy input. The advantages of such an approach, which allows a considerable lowering of working pressure of the F₂ laser, have been substantiated. The F₂ laser capable of competing with the latest achievements of leading foreign manufacturers has been fabricated. This laser efficiently generated 28 mJ at a pressure of 3300 mbar. However, a number of questions concerning the effect of mixture composition on the attainment of optimal pump conditions or the possibility of a further decrease in the working pressure of the mixture remained unanswered.

In modern technologies, high-energy F₂ lasers (tens of millijoules) are usually employed together with photomasks. Up to 90% of the light beam energy may be lost in such a mask [5–10]. Moreover, such lasers are very expensive and have a large size and weight.

In a number of technologies, it is more advantageous to use a small F₂ laser with a pulse energy of about 1 mJ. In this case, the optical radiation can be focused almost entirely into a spot of diameter less than 1 μm, and a scanning mirror system can be used to act selectively on the surface of the exposed target. So far, such lasers were efficient only at pressures exceeding 6 atm [12, 13].

The aim of this paper is to study the influence of pressure and mixture composition on the attainment of optimal pump conditions for F₂ laser and to develop a small-size laser with an output energy exceeding 1 mJ for mixture pressures of 3000–3500 mbar.

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2. Basic requirements to laser pumping systems and conditions

The development of a high-power F_2 laser requires a proper choice of the excitation system including the pump generator, preionisation system and discharge circuit, as well as a correct evaluation of the optical resonator parameters and selection of the optimal conditions for excitation of the active medium. We shall consider these requirements one by one.

2.1 Pump generator

For a laser to operate under relatively low pressures of the gas mixture, a pump generator with a steep voltage build-up front (having a duration of less than 100 ns) and providing a sufficiently high voltage across the discharge gap is required. The most suitable oscillator for this purpose is one with magnetic compression of high-voltage pulses, which is capable of producing pulses with a front duration of several tens of nanoseconds [14].

Figure 1 shows the electrical circuit of the pump generator used by us. It contains two magnetic elements for compressing high-voltage pulses and a Fitch LC oscillator [15]. A cold-cathode thyatron of the type TPI1-1k/20 was used as a switch. The storage capacitors had a total capacitance $C_1 + C_2 = 12$ nF. The peaking capacitors had a capacitance $C_4 = 3.5$ nF. The inductances L_1 , L_2 , and L_3 of the magnetic compression elements shown in the circuit diagram correspond to the saturated state of their cores. The induction coils L_1 and L_3 are wound on ferrite rings 1000NN K100 \times 60 \times 15, while the induction coil L_2 is wound on a magnetic amorphous iron core. Magnetisation reversal of the core of L_1 is carried out by the pulsed charge current of storage capacitors flowing from a high-voltage source, while in the core of inductances L_2 and L_3 it is carried out by bias coils fed by a dc power supply ($J = 2$ A).

2.2 Preionisation system

The spark preionisation system, which is employed frequently in excimer lasers, has two significant drawbacks: a nonuniform UV illumination along the discharge gap due to a nonuniform distribution of the pump energy, and the necessity of using a large number of inductive or capacitive decouplings for a uniform current distribution in individual

sparks [1–3, 16]. Instead of the spark system, we used for preionisation an extended barrier discharge through a dielectric. The preioniser was installed on one side of the discharge gap and consisted of two parallel electrodes, one of which was a knife-edge electrode and the other was a cylindrical electrode. The electrodes were separated by a ceramic tube. The knife-edge electrode was connected to the high-voltage electrode of the laser. A uniform barrier discharge spread over the surface of the tube and ensured a fairly uniform illumination of the working volume, as well as the required initial concentration of electrons. Owing to a limited barrier discharge current density, the erosion of the knife-edge electrode is hampered and this considerably slows down the undesirable process of metallisation of the optical elements of the laser.

2.3 Discharge circuit

A small CL5000 excimer laser (emitting at 308, 248 and 193 nm) produced by the Physical Instrumentation Centre, Prokhorov General Physics Institute, Russian Academy of Sciences, was used for developing the F_2 laser [17]. The specific features of the excimer laser are its metal ceramic construction and computer control, which ensure a long service life, possibility of smooth variation and a high stability of laser pulses. The active volume of the laser was $V = d \times w \times l \approx 9$ cm³ (where $d = 1.2$ cm is the interelectrode spacing, $w \approx 0.3$ cm is the discharge width and $l \approx 25$ cm is the length of the discharge region). Experiments were performed at a gas pressure $p \leq 3500$ mbar in the discharge chamber.

The radiation lifetime τ_r of the upper laser level $D^3\Pi_{2g}$ is only 3.7 ns (the effective lifetime is even shorter) [18]. Pumping by such short pulses is a difficult task. The F_2 laser is usually pumped in the quasi-stationary regime, when the pump pulse duration is much longer than the characteristic time of the processes occurring in the laser. As a result, about 30 % of radiation energy is lost due to spontaneous emission [18, 19]. In other words, for shorter pump pulses, the radiative losses must be lower and the lasing efficiency must be higher.

On the other hand, the pump pulse duration affects the properties of self-sustained volume discharge. During the discharge, local overheated regions with profuse emission of electrons may be formed at the cathode surface (cathode spots), which cause an enhanced erosion of electrodes and

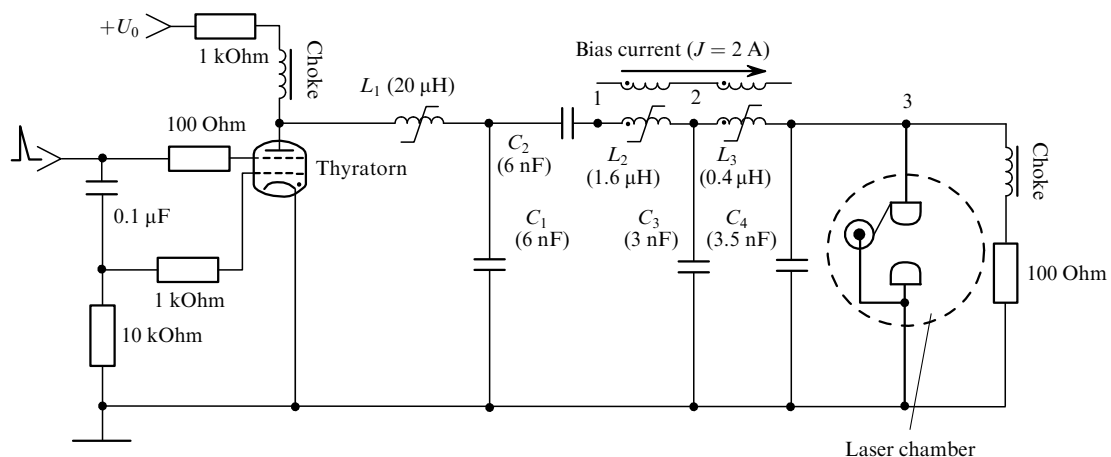


Figure 1. Electrical circuit of the pump oscillator.

can provoke the formation of sparks, thus shortening the service life of the laser, especially for high pulse repetition rates. The time of formation of cathode spots is inversely proportional to the square of the discharge current density and is longer than 20 ns under conditions typical of the F₂ laser (current density $j > 1 \text{ kA cm}^{-2}$) [12]. Hence a short duration of the pump pulse is also important for obtaining a homogeneous self-sustained volume discharge without cathode spots. The duration of the pump current pulse is proportional to $(L_d C_4)^{1/2}$, where L_d is the inductance of the discharge circuit. While constructing the laser, steps were taken to minimise the value of L_d by reducing the discharge circuit cross section and the length of the current leads.

2.4 Optical resonator

In the case of lower working pressures and a smaller resonator length, it is necessary to optimise properly the reflectivity of the resonator mirrors. The condition for the maximum radiation coupling out of the resonator has the form [20]

$$\frac{\ln(R_1 R_2)}{2l} = \beta \left[1 - \left(\frac{\alpha}{\beta} \right)^{1/2} \right], \quad (1)$$

where α is the small-signal gain; β is the absorption coefficient (sum of the coefficients of ‘transient’ absorption during the excitation pulse and steady-state absorption caused by the presence of alien inclusions in the gas mixture); R_1 and R_2 are reflectivities of the highly reflecting mirror and the output resonator mirror, respectively. Experimental dependences of α on p for the F₂ laser are presented in [21]. It follows from these dependences that under the conditions considered by us (for $p \approx 3500 \text{ mbar}$), $\alpha \approx 0.14 \text{ cm}^{-2}$ and $\alpha/\beta \approx 10$ [18]. Assuming further that $R_1 = 0.98$, we obtain from Eqn (1) the value $R_2 = 0.23$ required for ensuring the maximum energy extraction. The reflectivity of one plane–parallel CaF₂ plate (from both surfaces) is about 10%. Obviously, at least two such plates must be used to improve the Q -factor of the resonator.

2.5 Optimal conditions for excitation of the active medium

To pump the F₂ laser efficiently, the energy of the storage capacitors ($C_1 + C_2$) must be supplied to the discharge plasma with minimal losses in the highest pump power regime [2, 3]. If we neglect the unavoidable losses in the pump generator circuit, such a loss-free supply is possible only if the limiting voltage (corresponding to the idling voltage for the given pump generator system) across the discharge gap (DG) and the matched pump regime are realised simultaneously.

The condition for the matched pump regime has the form

$$\frac{E_m}{p} = \frac{2E_{qs}}{p}, \quad (2)$$

where E_m is the maximum field strength and E_{qs}/p is the ratio of the field strength to the gas pressure in the quasi-stationary regime, corresponding to the maximum self-sustained discharge current. Condition (2) indicates the equality of the discharge gap resistance $R_{qs} = U_{qs}/J_m$ at the highest current and the wave impedance $Z = (L_d/C_4)^{1/2}$ of the C_4 discharge circuit through the DG. Practically all the energy stored in the peaking capacitor C_4 can be supplied

only in a matched pump regime to the discharge plasma during the pump current pulse.

3. Experiment

As in Refs. [2, 3], we used for studying the optical radiation parameters a special measuring chamber connected hermetically with the output laser window. The chamber was first evacuated and then filled with nitrogen. A pyroelectric Gentech detector was mounted inside the chamber for measuring the output laser energy. The pump generator voltage pulses and the voltage across the discharge gap were recorded with a high-voltage Tektronix P6015A probe and a LeCroy WaveSurfer 432 oscilloscope.

The optical resonator having a total length of 54 cm was formed by a highly reflecting plane mirror, an output window in the form of a plane–parallel CaF₂ plate, as well as an auxiliary plane–parallel CaF₂ plate that was installed inside the measuring chamber at a distance of $\sim 20 \text{ cm}$ from the first plate.

The laser under investigation could work at a pulse repetition rate of up to 1000 Hz, but the measurement of its parameters was carried out at a frequency of 10 Hz. In the course of the experiments, we measured the high-voltage pulses and the laser beam energy for various charge voltages U_0 , compositions and pressures of the gas mixture.

Figure 2 shows the oscillograms of voltage pulses at characteristic points in the pump generator circuit (points 1, 2, 3 in Fig. 1). One can see that the magnetic generator provided a fourteen-fold compression of pulses and formed voltage pulses of duration up to $\sim 70 \text{ ns}$ and amplitude up to 21 kV in the DG.

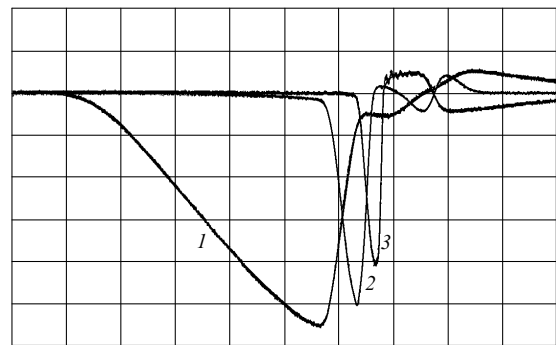


Figure 2. (a) Oscillograms of voltage pulses in the pump oscillator system at points 1 [curve (1)], 2 [curve (2)] and 3 [curve (3)] indicated in Fig. 1. Scale: 1 division – 5 kV along the vertical and – 200 ns along the horizontal direction.

Figure 3 shows the dependence of the F₂ laser energy W_g on the total pressure p of the gas mixture for various values of U_0 for the mixture He–F₂ with different fluorine concentrations [F₂]. Since the lasing energy depends on the energy $W_4 = C_4 U_m^2 / 2$ stored in the peaking capacitor, where U_m is the maximum voltage across the DG, Fig. 4 also shows for comparison the experimental dependences of U_m on p . One can see that the voltages U_m first increase and then saturate with identical limiting values U_{lim} determined by the voltage U_0 [2, 3]. The higher the percentage of fluorine in the mixture, the lower the pressure p at which saturation occurs. However, the dependences $W_g = f(p)$

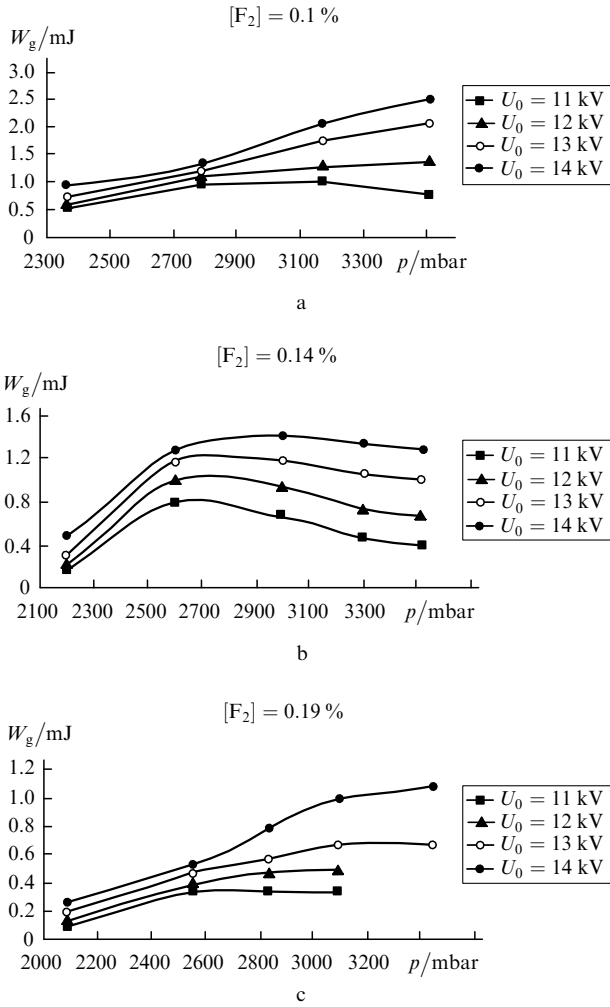


Figure 3. Dependence of the output energy W_g on the total pressure p of the He-F₂ mixture for different values of the charge voltage U_0 and fluorine concentration $[F_2]$.

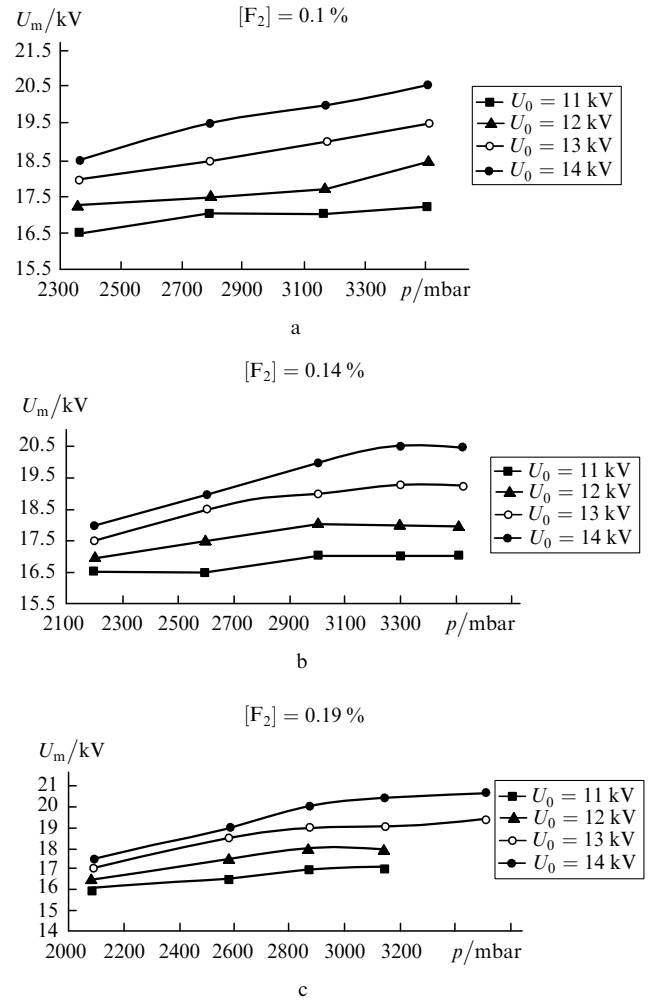


Figure 4. Dependence of the maximum voltage U_m across the DG on the total pressure p of the He-F₂ mixture for different values of the charge voltage U_0 and fluorine concentration $[F_2]$.

behave in an entirely different manner. They differ not only in magnitude, but also in the position of the peaks. Visual observations of the discharge region reveal that the discharge in the DG is quite homogeneous (without cathode spots, sparks and enhanced brightness zones of the glow) owing to a uniform preionisation and a short pump pulse duration. Hence discharge inhomogeneities cannot be responsible for such a strong variation of the output laser energy.

The evolution of the dependences $W_g = f(p)$ can be followed by taking into account the process of matching plasma resistance with the wave impedance of the discharge circuit. In order to attain impedance matching, we must know the value of E_{qs}/p [see Eqn (2)], which is estimated from the following assumptions.

The dominating reaction in the He-F₂ mixture is the dissociative attachment of electrons to fluorine molecules [19, 22]:



This reaction controls the electron losses in the laser plasma. The attachment coefficient increases with decreasing electron energy $W_e = (3/2)kT_e$, where k is the Boltzmann constant and T_e is the electron temperature.

The higher the fluorine concentration, the larger the fraction of electrons leaving the plasma as a result of attachment. This leads to a rapid growth of W_e and T_e . The theoretical dependence of T_e on $[F_2]$ in the He-F₂ mixture ionised by a quasi-stationary electron beam was obtained in [19] and is shown in Fig. 5 (lower curve). In the gas discharge, we have $T_e \sim E/p$ [23]. The method used for gas mixture ionisation does not affect the mechanism of variation of T_e and E/p with $[F_2]$. Hence it can be assumed that the dependences $T_e = f([F_2])$ are identical in the plasmas created by an electron beam and a self-sustained discharge. In this case, knowing that $E_{qs}/p = 1.6 \text{ V cm}^{-1} \text{ mbar}^{-1}$ for $[F_2] = 0$ [18], we can easily obtain the dependence $E_{qs}/p = f([F_2])$, which is also shown in Fig. 5 (upper curve). Hence we obtain $E_{qs}/p = 2.6, 3$ and $3.5 \text{ V cm}^{-1} \text{ mbar}^{-1}$ for the values of $[F_2] = 0.1\%, 0.14\%$ and 0.19% respectively.

In accordance with Eqn (2), the pressure of the mixture corresponding to the matched pump regime is determined by the points of intersection of the curves $E_m/p = U_m/(pd) = f(p)$ in Fig. 6, plotted on the basis of the experimental data presented in Fig. 4, with the theoretical values $2E_{qs}/p = 5.2, 6$ and $7 \text{ V cm}^{-1} \text{ mbar}^{-1}$.

A comparison of Figs 3, 4 and 6 shows that for an enhanced fluorine concentration ($[F_2] \geq 0.19\%$, see Figs 3c,

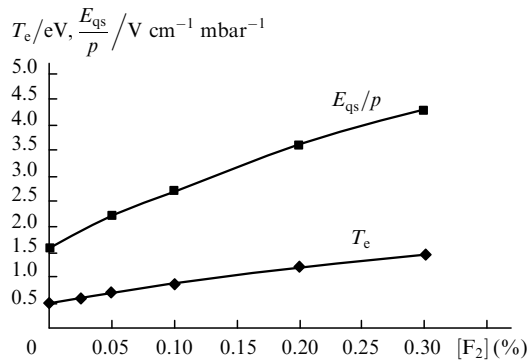


Figure 5. Theoretical dependences of the electron temperature T_e and the ratio E_{qs}/p of the field strength to the gas pressure on the molecular fluorine concentration $[F_2]$ in the F_2 laser plasma.

4c, and 6c), matched regime is possible only for $p \leq 2100$ mbar, under which the voltage across the DG is not so high. In the rest of the working pressure range, the behaviour of the dependences $W_g(p)$ is determined by the dependence $U_m(p)$ and by the increase in the value of α

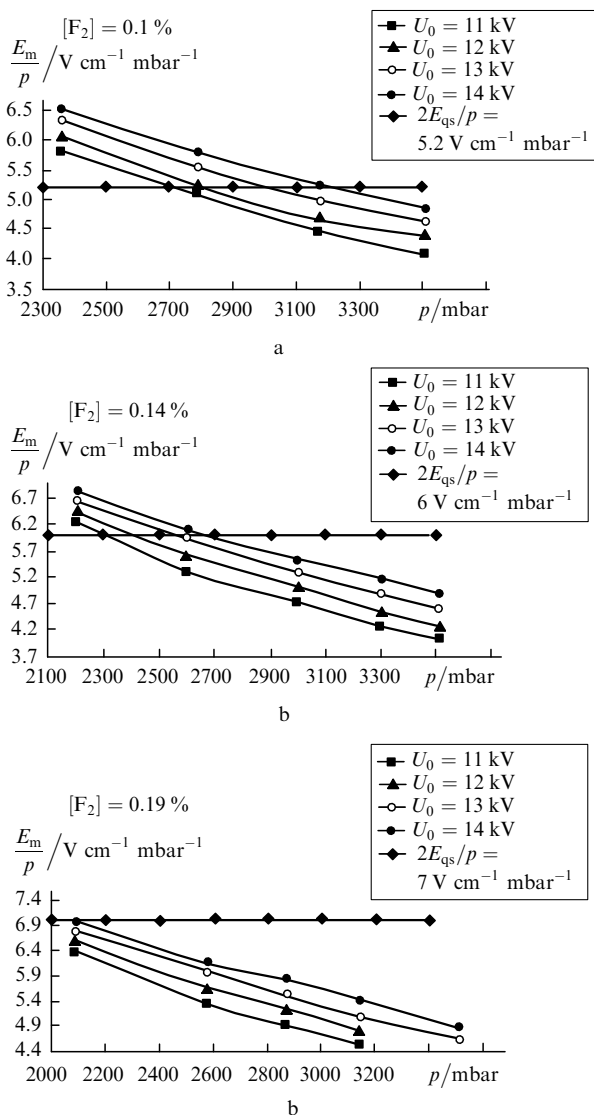


Figure 6. Dependences of the ratio E_m/p of the maximum field strength to the total gas pressure on p for different values of U_0 and $[F_2]$.

with p [18]. However, the higher the pressure p , the smaller the fraction of energy supplied to the active medium from C_4 [2, 3]. As a result, the output laser energy is found to be low.

For $[F_2] = 0.1\%$ and $U_0 = 14$ kV, the matched regime and limiting voltage across the DG are realised under a working pressure close to the limiting value $p_{lim} = 3500$ mbar. Under these conditions, W_g attains its highest values owing to the maximum energy input and the highest gain α (for $p \leq p_{lim}$).

For $[F_2] = 0.14\%$, the highest values of W_g (see Fig. 3b) correspond to the values of p for the matched pump regime (Fig. 6b). In this intermediate region, however, the matched pump regime is attained for values of p and U_m lower than p_{lim} and U_{lim} , and hence intermediate values of W_g are attained in this case. If a resonator with only one plane-parallel output CaF_2 plate is used under the same conditions, the lasing energy drops to about half due to a decrease in the Q -factor of the resonator.

The above results indicate that the attainment of the highest lasing energy in an electric-discharge F_2 laser requires a He- F_2 mixture composition for which the matched active medium pump regime is realised under conditions of limiting voltage across the DG and limiting working pressure (for the given excitation system).

For a transition to even lower working pressures, we must use a mixture with a higher fluorine concentration. However, in order to avoid a further decrease in the value of U_m , we must increase the steepness of the pump voltage front, which is a quite difficult task, or increase the electrode gap, which results in a large difference between the divergence of radiation along the vertical and horizontal coordinates of the beam.

Note that in contrast to the analogous foreign lasers with an energy of 2 mJ at a working pressure of 6 bar [13] and 7 bar [12] of the mixture, the output energy of the laser described in this communication was up to 2.5 mJ under a mixture pressure of 3.5 bar, i.e., much lower than in the above-mentioned works.

4. Conclusions

In view of the peculiarities and the requirements imposed on the operation of electric-discharge F_2 laser, it is proposed to use a modified system of active medium excitation based on a generator for magnetic compression of high-voltage pulses with a preionisation of the electrode gap by a barrier discharge over the surface of a ceramic tube. Such a system ensures highly homogeneous self-sustained volume discharges without cathode spots in He- F_2 mixtures.

An analysis of the dependences of the output laser parameters on the mixture composition, pressure and the discharge voltage showed that the pumping processes are considerably influenced by the reaction of dissociative attachment of electrons to fluorine molecules which controls the electron plasma temperature and the process of energy input to the active medium. It is found that the optimal composition of the He- F_2 mixture is one for which the matched pump regime of the active medium is realised under conditions of limiting voltage across the DG and limiting working pressure (for the given excitation system).

Our investigations have led to the development of a small-size F_2 laser which works at a much lower pressure of

the gas mixture (up to 3500 mbar) and whose radiation parameters (output radiation energy of 2.5 mJ, active volume of $\sim 9 \text{ cm}^3$) are not inferior to those of the foreign analogues.

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