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Efficient lasing in mixtures of helium and fluorine in diffuse discharges formed by runaway electrons

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Abstract. **The parameters of stimulated lasing in diffuse discharges formed in mixtures of helium and fluorine in a strongly inhomogeneous electric field are investigated. Lasing is obtained in the visible and VUV spectral regions on the transitions of fluorine atoms and molecules. It is shown that lasing in He–F_2 mixtures at a wavelength of 157 nm continues for several half-periods of the discharge current. Due to the homogeneity of the diffuse discharge, the maximum lasing efficiency of the F_2 laser is 0.15%, which corresponds to the efficiency of this type of lasers pumped by pre-ionised transverse volume discharges.**

Keywords: *effective VUV generation, diffuse discharge, inhomogeneous electric field, runaway electrons.*

1. Introduction

Electric-discharge lasers on fluorine molecules with a lasing wavelength of 157 nm are powerful sources of stimulated radiation in the vacuum ultraviolet region. This radiation is strongly absorbed by most materials, which makes it possible to widely use such lasers for their purification and processing [1], in lithography [2–4], for photochemical deposition [5], etc. For pumping F_2 lasers, transverse volume discharges with preionisation of the gas mixture are commonly used. However, in mixtures with fluorine, a rapid contraction of the volume discharge is observed, which occurs within 20–30 ns [6]. This makes it necessary to use short high-power voltage pulses of large amplitude and high pressure of the working mixture for excitation [7, 8]. Under these conditions, such an important laser parameter as its efficiency is usually less than 0.1% due to the mismatch between the generator impedance and the discharge resistance. The calculated efficiency $\eta_{\rm int}$ of the F_2 laser relative to the deposited energy is 1% [9] and can be implemented using double-discharge generators with a decrease in the pump power and pressure of the active gas mixture [10].

The maximum efficiency of electric-discharge lasers based on molecular fluorine relative to the energy stored in the generator (0.15%) was obtained in works [11, 12] in pumping by short high-power pulses of ~10 ns duration.

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It should be noted that volume diffuse plasma can be formed by initiation with runaway electrons in gas gaps with electrodes of small curvature radius, to which high-voltage pulses with fronts of ~1 ns are applied [13]. In work [14], it was proposed to call such diffuse discharges 'runaway electron preionised diffuse discharges' (REP DD). It has been shown that REP DD can be used to obtain efficient lasing in various spectral ranges [15].

The aim of this work is to study the parameters of stimulated emission in mixtures of helium and fluorine in pumping by a pulsed diffuse discharge.

2. Experimental equipment and measurement techniques

To study the characteristics of radiation generated in nanosecond diffuse discharges that are formed in various gas mixtures by high voltage pulses, a laser chamber connected to a RADAN-220 pulse generator was designed and described in detail in [16]. The maximum energy E stored in the generator's forming line with a capacity of C = 50 pF [17] was 2.1 J. This value is determined by the breakdown voltage U of a P-49 serial spark gap-sharpener, which in our experiments was equal to 280±10 kV.

A high-voltage pulse from the RADAN-220 generator was fed to stainless steel electrodes made in the form of blades with rounded edges and an apex angle of 5°. The curvature radius of the sharp edges was 0.05 mm. This provided an inhomogeneous distribution of the electric field strength in the discharge gap and, accordingly, its increase near the electrodes, which made it possible to form volume diffuse discharges in various active gas mixtures at pressures up to 10 atm. The length of the electrodes was 30 cm, while the gap d between them was 1.8 cm. There was an additional window on the side wall of the chamber for photographing the discharge and recording the parameters of its spontaneous emission pulses. The resonator was formed by a flat aluminium mirror and a plane-parallel MgF_2 plate.

To measure the energy of laser radiation, an OPHIR-type meter (Ophir Optronics LTD, Inc.) with a PE50BB sensor head was used, which was installed at a minimum distance from the resonator's output mirror; the gap between them was blown with helium. Since simultaneous generation of fluorine atoms and molecules in the visible and VUV spectral regions was observed in He–F_2 mixtures, measurements were performed without blowing the gap to determine the energy of visible radiation.

Laser pulses on FI lines were measured with a FEK-29SPU photocell. To measure the VUV pulses, the photocathode in the standard FEK-22 photocell was replaced with a polished copper plate. The photocell was vacuum-tightly attached to the resonator's output mirror and pumped out to a residual pressure of 5×10^{-4} mm Hg. The work function of the copper cathode was 4.53–5.10 eV, which corresponds to the photoelectric threshold of λ = 274–245 nm [18].

A photocell with a copper cathode was tested by laser pulses at λ = 351–353 nm (XeF^* laser), 337.1 nm (nitrogen laser), and 248 nm (KrF^* laser). The laser pulse shapes at λ = 248 nm, recorded by FEK-22 and the modified photocell, coincided, while the radiation at λ = 337 and 351–353 nm was not detected by the copper-cathode photocell. It should be noted that the techniques for measuring VUV pulses of plasma radiation by devices with metal photocathodes are well developed, provide high accuracy, and are quite widespread [19–21]. The emission spectra of diffuse discharge plasma in the wavelength range of 200–1100 nm were recorded by the HR400 (Ocean Optics) spectrometer.

To measure the discharge current pulses, an ohmic shunt assembled from low-inductive microchip resistors was used. Electrical signals were recorded using a TDS 3054 digital oscilloscope.

3. Results of experiments and their discussion

3.1. Generation on atomic fluorine lines

In the discharge plasma in mixtures of helium with fluorine, the kinetic processes that form inversion at atomic fluorine transitions are part of the kinetics of VUV lasers on F_2 molecules. One of the channels for populating the upper laser level of the VUV transition $D'(^3\Pi_{2g}) \rightarrow A'(^3\Pi_{2u})$ of the fluorine molecule is the energy transfer reaction

$$F^* + F_2 \rightarrow F_2^* + F, \qquad (1)$$

where F^* are excited fluorine atoms in metastable states $3s^4P_j$ (j = 5/2, 3/2, 1/2), which are the lowest laser levels for a laser on fluorine atomic lines [11]. Therefore, the study of lasing on FI lines is important for a more complete understanding of the kinetics of the F_2 laser.

A distinctive feature of the FI laser is the short delay time of the laser pulse relative to the start of the pump pulse and a wide lasing region that reaches 1 cm at a gas mixture pressure of 1–3 atm, whereas in the case of an N_2 laser [16], the output beam is much narrower. This can be attributed to the fact that the lasing threshold on fluorine lines is quite low, while the current flow region in the initial stage of the REP DD is rather wide [22]. Then the flow region of the main discharge current narrows, and since a higher current density is required to reach the lasing threshold on fluorine molecules, the width of the VUV radiation autograph does not exceed 3–4 mm [7].

The duration of laser radiation on FI lines with an excess of the resonator Q-factor is equal to several half-periods of the discharge current, which indicates a high stability of the REP DD in mixtures with fluorine. The laser radiation spectrum contains five lines in the range of 634–755 nm. At the same time, the laser spot periphery at low pressure is dominated by radiation with λ = 634 nm, while lines with λ > 700 nm are observed at the aperture centre. This looks like a drop in the radiation intensity at the lasing spot centre, since the eye is unable to see IR radiation.

Figure 1 shows the intensities of spontaneous and stimulated emission on the atomic fluorine lines versus the helium pressure in the active mixture. The letters Q and D denote quartets (transitions from $3p^4S^0_{3/2}$ and $3p^4P^0_{5/2,3/2,1/2}$ levels) and doublets (transitions from the $3p^2S^0_{1/2}$ and $3p^2P^0_{1/2,3/2}$ levels), respectively. According to work [23], the spontaneous emission intensity P is equal to the product of the population n[FI] of the upper transition level and the probability of spontaneous emission A: $P = n[\mathrm{FI}]A$. It follows from Fig. 1a that the populations of the upper levels of transitions are weakly dependent on the helium pressure. The exception is the quartet transition at λ = 739.8 nm, where the radiation intensity increases significantly. The dependences of the laser line intensities on the helium pressure behave in a very different way (Fig. 1b). As the buffer gas pressure increases, a sharp increase in the lasing power is observed at the $3p^4P^0_{5/2}$ – $3s^4P_{5/2,3/2}$ quartet transitions for λ = 739.8 and 755.2 nm, while the emission intensity on the other lines decreases to almost zero. It can be assumed that these dependences are due to the rapid relaxation of the upper levels of doublet transitions in collisions of fluorine and helium atoms. As the mixture pressure increases, the lasing region becomes the same as that of the F_2 laser. The total radiation energy on the FI lines increases linearly as the mixture pressure increases, and exceeds 0.3 mJ at p = 8 atm. This also confirms the REP DD stability in mixtures with fluorine, since the lasing energy on the FI lines drops sharply [24] when the discharge is contracted.

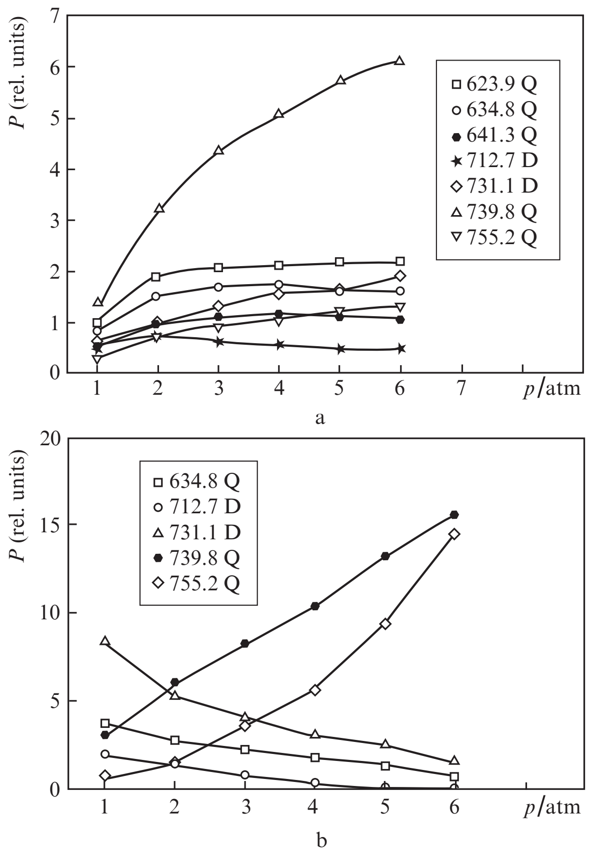

Figure 1. Dependences of the intensities P of (a) spontaneous and (b) stimulated emission on the FI lines on the pressure p of helium mixed with F_2 (5 Torr). Letters Q and D represent quartets and doublets, wavelengths are given in nanometres.

If we assume that a single fluorine atom in $3s^4P_{5/2,\,3/2}$ states produces a single laser photon at λ = 157 157 nm, reaction (1) can provide the lasing energy on F_2 molecules up to 1.6 mJ.

3.2. VUV laser on fluorine molecules

The operation of an F_2 laser pumped by a diffuse discharge is illustrated in Fig. 2. Figure 2a shows typical waveforms of a current pulse I_d in the discharge gap, a pulse of spontaneous emission of a diffuse discharge in the region λ = 200 – 800 nm, the maxima of which correspond to the maxima of I_d, and a laser pulse at λ = 157 nm. The VUV lasing threshold is reached at $p \approx 3$ atm. The laser pulse at λ = 157 nm starts near the first maximum I_d and has three pronounced peaks corresponding to three successive oscillations in the diffuse discharge current. The total VUV laser pulse duration reached 30 ns. The radiation power in the first two peaks increased with increasing pressure of the active mixture, while the intensity of third peak decreased. This is due to the fact that the amplitudes of the first current peaks were weakly dependent on the helium pressure, while the pump current amplitude in the third peak was already noticeably reduced.

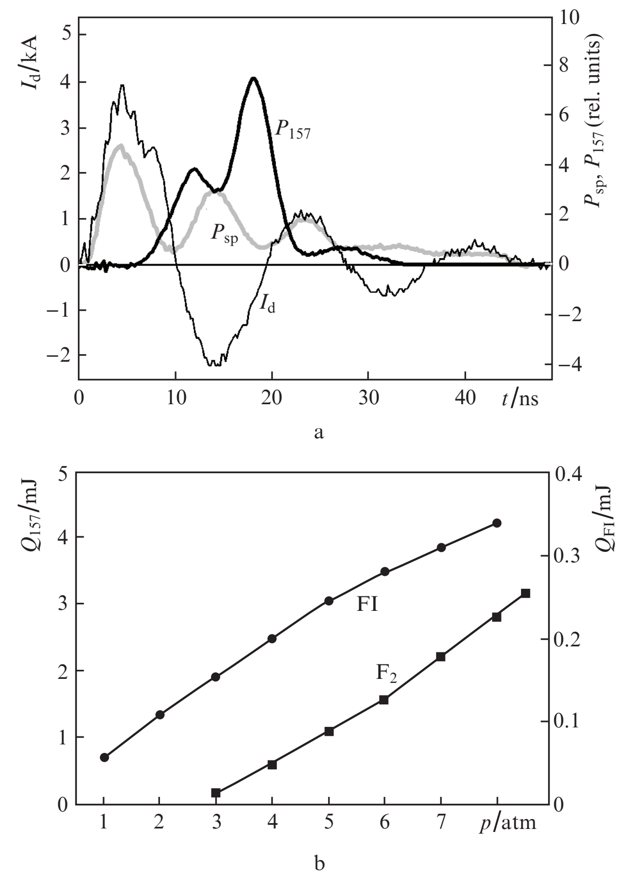

Figure 2. (a) Typical oscillograms of spontaneous emission pulses from diffuse discharge P_{sp} and F_2 laser P_{157}, as well as the discharge current I_d; a mixture of He (7 atm) and F_2 (5 mm Hg) as well as (b) the dependences of the radiation energy of the F laser Q_{FI} and F_2 laser Q_{157} on the pressure of He in a mixture with F_2 (5 mm Hg).

Similar to papers [8, 9, 11, 12], the radiation energy of the molecular fluorine laser increased linearly with increasing pressure of the active mixture. This is due to an increase in the energy supplied to the active medium, since the discharge gap voltage is proportional to the pressure of the mixture [25], while the discharge current in the first two peaks, during which the main energy input occurs, is mainly determined in our experiments by the wave impedance of the RADAN-220 generator and weakly depends on pressure. The maximum lasing energy Q_{157} at a He pressure of 8.5 atm is 3 mJ. In this case, according to the dependence shown in Fig. 2b, its further growth is possible. This energy corresponds to the F_2 laser efficiency of 0.15% obtained by pumping the F_2 laser with a transverse self-sustained discharge with preionisation [11, 12]. This also means that approximately half of the F_2^* molecules in the $D'(^3\Pi_{2g})$ state are formed through the neutral channel (1).

According to estimates, the specific pump power in our experiments, with regard to the data on the voltage U_{qs} in the quasi-stationary stage of the volume discharge given in [25], is 6 – 7 MW cm^{-3}, which corresponds to the optimal value given in [12] and required to obtain the maximum efficiency of the F_2 laser.

The existence of a VUV laser pulse during several current oscillations in the discharge gap also indicates high stability of a diffuse discharge formed in an inhomogeneous electric field due to preionisation by runaway electrons [14]. This effect can be related to conditions similar to those described in [26] for the formation of a diffuse discharge at a voltage pulse amplitude exceeding 250 kV and a rapid increase in the current density in the discharge gap ($dj/dt >$ 50 A cm^{-2} ns^{-1}). A characteristic feature of this pump regime is a rapid increase in the concentration of electrons and ions in direct ionisation processes. Under these conditions, plasma is formed without fluctuations in the concentration of charged particles in the volume, which leads to high stability of the diffuse discharge.

4. Conclusions

Lasing in mixtures of helium with fluorine when pumped by a diffuse discharge formed by runaway electrons in an inhomogeneous electric field has been studied. Lasing in the visible and VUV spectral ranges on transitions of fluorine atoms and molecules is obtained. It is shown that the volume stage of a diffuse discharge is preserved for several successive current oscillations in the laser gap. The lasing energy of up to 3 mJ in the VUV region was attained with a VUV laser pulse duration of up to 30 ns. Due to the short duration and optimal pump power, the maximum lasing efficiency of 0.15% was obtained for the F_2 laser, which is equal to the efficiency of lasers of this type when excited by pre-ionised transverse volume discharges.

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