Optical pumping of chemical HF lasers on the basis of $NF_3 - H_2$ and $ClF_5 - H_2$ mixtures by an open surface discharge in the bleaching-wave mode

M Yu Artem'ev, A S Bashkin, L D Mikheev, V M Nesterov, M L Sentis, V I Cheremiskin

Abstract. **Lasing on HF upon optical pumping by emission of an open discharge, using NF_3 and ClF_5 as donors of fluorine atoms, was obtained for the first time in a chemical laser and the bleaching-wave mode was realised in a chemical HF laser. An open surface discharge was used as a pump source. The velocity of the bleaching wave, which was formed under its action, reached ~ 8 km s^{-1}. The formation of this wave leads to a rapid (with an ultrasonic velocity) replacement of the working medium in the lasing region, which provides a quantum laser efficiency close to unity. The optimum compo-sition of the working mixture was found to be $NF_3 : H_2 : Kr = 6 : 10 : 125$ Torr. For this composition, the output laser energy in a 3.2-μs pulse reached ~ 0.4 J and the specific output energy was 3.5 J litre^{-1}. Approximately the same output characteristics of laser emission (0.35 J in a 3.5-μs pulse in the $ClF_5 : H_2 : Kr = 3 : 20 : 50$ Torr mixture) were obtained in the system using ClF_5 as donors of fluorine atoms.**

Keywords: chemical laser, photodissociation, surface discharge

1. Introduction

The principle of action of HF lasers being considered here is based on nonchain excitation mechanisms. Although such lasers have a lower efficiency than the chain reaction lasers, they are simpler and safer from the viewpoint of pre-paration of a working mixture and, therefore, are more suitable for some applications in practice. Despite a variety of nonchain reactions used for obtaining lasing and a large number of papers devoted to the study of laser characteristics of media on their basis, potentialities of nonchain reaction HF lasers are far from being completely understood.

The point is that the majority of studies were made upon flashlamp pumping. As for nonchain HF excitation reac-

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tions initiated by emission of an open discharge, their systematic studies are virtually absent. Because of the ab-sence of a shell separating a discharge plasma from an active laser medium, an open discharge differs from a flashlamp by a considerably higher radiation intensity, which reaches se-veral hundred kW cm^{-2} in the spectral band of photodis-sociation of atomic-fluorine donors, the possibility of pro-ducing a considerably shorter pump pulse, and a wider spectral range of emission, which includes the VUV region. The latter circumstance allows one to increase the pump effi-ciency for the known working mixtures which were previo-usly used at flashlamp pumping, and to extend considerably the class of donors of atomic fluorine (or hydrogen) by using molecules having absorption continua in the VUV region.

Moreover, using optical pumping by an open discharge, one can realise a new type of excitation, which consists in the initiation of a free bleaching wave in an active medium and causes a rapid (with ultrasonic velocity) replacement of the active medium in the lasing region for $0.1 - 1$ μs. This eliminates the effect of self-deactivation of active HF mole-cules on the output laser parameters for long (longer than 10 μs) pumping and retains a high specific output energy.

In this paper, we demonstrated for the first time lasing on $NF_3 - H_2$ and $ClF_5 - H_2$ mixtures initiated by emission of an open surface discharge. Previous attempts to obtain lasing by pumping the $NF_3 - H_2$ mixture by a flashlamp failed [1]. An advantage of the NF_3 molecule is that it has a wide absorption spectrum, which lies predominantly in the VUV region. In addition, the secondary photodecomposi-tion of NF_2 radicals, which have diffuse absorption in ranges of $250 - 270$ and $158 - 170$ nm [2, 3], can also make a certain (although small) contribution to the formation of atomic fluorine along with the primary photolysis.

Interest in the ClF_5 molecule as a donor of atomic fluorine, which was earlier studied in detail in experiments on flashlamp-pumping, is caused by the following reasons. On the one hand, gaseous ClF_5 in a mixture with hydrogen is not subject to spontaneous combustion, and on the other hand, this molecule has a high efficiency of initiation of a chemical pump reaction. In Ref. [4], one absorbed photon of initiating emission gave three photons of laser emission.

Both compounds are characterised by a good spectral matching of absorption bands to the Planck's photon distri-bution over wavelengths for the black-body emission with a temperature of $20 - 30$ kK, which is typical of an open dis-charge (Fig. 1).

The aim of this paper was to study specific features of lasing in mixtures containing the above-mentioned molecu-les pumped by an open discharge.
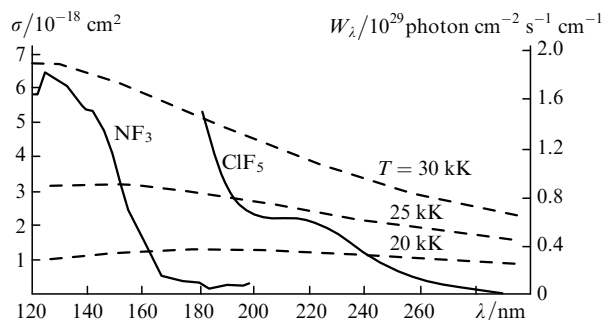
Figure 1. Absorption spectra of NF_3 [5] and ClF_5 [6] and the spectral distribution of emissive power of a black body for different temperatures T. The vertical scale of the curve for ClF_5 is magnified by a factor of 3.5.

2. Experimental setup

Schematic diagram of the experimental setup is presented in Fig. 2. A rectangular laser cell 5 cm × 7 cm × 27 cm in size was made of Teflon, which is chemically stable against the action of F_2 and HF. The cell had CaF_2 windows on its ends. The clear laser aperture was 2.8 cm in diameter. The laser cavity was formed by a flat aluminium mirror and the output window of the laser cell. The surface discharge 19 cm long was formed between two electrodes along the surface of a Teflon tube 7 mm in diameter, which was positioned in the lower part of the laser cell. For the linear stabilisation of the surface discharge, a metal rod, which was electrically connected to a high-voltage electrode, was inserted into the Teflon tube.
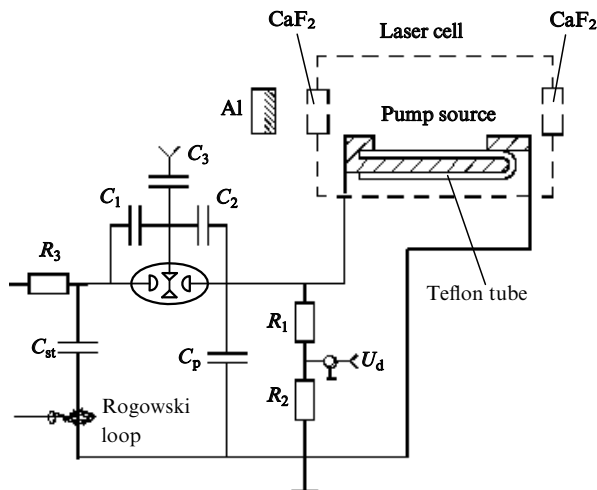

Figure 2. Schematic diagram of the experimental setup.

The discharge was excited by an 11-μF capacitor C_{st}, which was charged to the voltage $U_C = 20$ kV (it stored an energy of 2.2 kJ). The current was switched by a nitrogen-filled gap. When voltage is applied to the electrodes, a sliding barrier discharge propagates along the surface of the Teflon tube. It is caused by the displacement currents in the material of the tube. The barrier discharge closes the inter-electrode gap and causes the development of the main dis-

charge. As a result, a filament of strongly emitting plasma is formed, which expands with a radial velocity of $1-2$ km s^{-1}. We did not measure the brightness temperature of the discharge, but its value for the given electric parameters of the feed circuit is typically $20-30$ kK [7].

The time profiles of the discharge current and the voltage across the discharge gap were measured by a Rogowski loop and a low-inductance resistive divider (resistors R_1 and R_2), respectively. The output laser energy and the laser pulse shape were measured with a calorimeter and a Ge–Au detector. A photodetector with a filter measured the shape of a UV emission pulse of the surface discharge (Fig. 3) outgoing from the window of the laser cell.
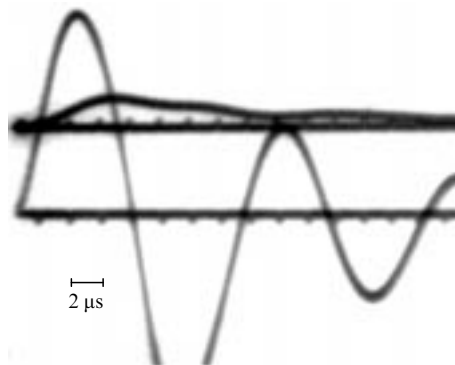

Figure 3. Oscillograms of an UV emission from the surface discharge (upper trace) and discharge current (lower trace)

It is common to use buffer gases, such as N_2 or inert gases in lasers optically pumped by emission of an open discharge in order to increase the pump radiation power and suppress an increase in temperature of a gas mixture caused by the energy released in exothermic reactions. Our preliminary studies of He and Kr as buffer gases showed that Kr gave the best results. It is likely that this is explained by the fact that Kr, which has a lower ionisation potential, provides a higher brightness temperature of the surface discharge. Because of this, we used in the experiments Kr as a buffer gas. All reagents (NF_3 (ClF_5), H_2, and Kr) were mixed directly in the laser cell. The gases had the following purity: 99.63 % (NF_3), 99.9 % (ClF_5), 99.995 % (H_2), and 99.98 % (Kr).

3. Results and discussion

The study of the output energy of the HF laser as a function of partial pressures of NF_3, H_2, and Kr gases shows that the optimum pressures of components in the $NF_3 : H_2 : Kr$ mixture are approximately 6 : 10 : 125 Torr. It follows from Fig. 4 that the output energy of the HF laser with fixed partial pressures of NF_3 and H_2 weakly depends on the buffer gas pressure, i.e., the brightness temperature of the surface discharge is almost independent of the Kr pressure at pressures above 25 Torr. The majority of experiments discussed here were made for a Kr pressure of 50 Torr.

Let us begin the analysis of kinetics of the $NF_3 - H_2$ active medium from the following reactions that are of primary importance in the formation of HF:
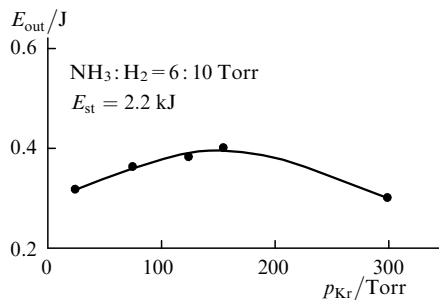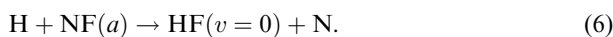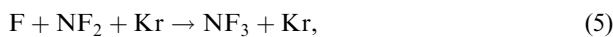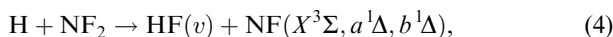
Figure 4. Dependence of the output energy of the HF laser on the partial pressure of Kr for the stored energy $E_{st} = 2.2$ kJ.

$$NF_3 + hv \rightarrow NF_2 + F, \quad (1)$$

$$NF_2 + hv \rightarrow NF(X) + F, \quad (2)$$

$$F + H_2 \rightarrow HF(v) + H, \quad (3)$$

$$H + NF_2 \rightarrow HF(v) + NF(X^3\Sigma, a\,^1\Delta, b\,^1\Delta), \quad (4)$$

$$F + NF_2 + Kr \rightarrow NF_3 + Kr, \quad (5)$$

$$H + NF(a) \rightarrow HF(v = 0) + N. \quad (6)$$

These reactions have the following rate constants: $k_{3v} = f_v \exp(-800\text{K}/T)2.7 \times 10^{-10}$ cm³ s⁻¹, $f_1:f_2:f_3 = 0.17:0.55:0.28$ [8], $k_{4v} = g_v 1.3 \times 10^{-11}$ cm³ s⁻¹, $g_0:g_1:g_2:g_3 = 0.65:0.27:0.07:0.01$, $k_X:k_a:k_b = 0.07:0.91:0.02$ [9], $k_5(\text{Ar}) \approx 10^{-30}$ cm⁶ s⁻¹ [10], $k_6 = 2.5 \times 10^{-13}$ cm³ s⁻¹ [10].

The rate constant for the recombination of atoms F with NF₂ with the participation of Kr as a third particle is unknown. We took for this process the value of k_5 measured for Ar as a third particle [10]. The contribution of the secondary photolysis of NF₂ radicals to the pump rate is insignificant because these radicals have rather narrow (~ 20 and ~ 10 nm) absorption continua in the UV ($\lambda = 260$ nm) and VUV ($\lambda = 164$ nm) regions. The absorption cross sections σ_λ at the maxima are 1.5×10^{-18} cm² [2] and 2.1×10^{-18} cm² [3], respectively.

For a brightness temperature of the pump source of 30 kK, the photon fluxes into these absorption bands are approximately identical and equal to $\sim 1.6 \times 10^{23}$ photon cm⁻² s⁻¹, which corresponds to the rate of formation of F atoms in the photolysis of NF₂ by emission of an infinite flat source $\sigma_\lambda I_0 = 5.8 \times 10^5$ s⁻¹, where I_0 is the summary photon flux into the absorption bands of NF₂. This rate is considerably lower than the corresponding value for the dissociation of NF₃ molecules, which is 2.4×10^6 s⁻¹. Moreover, as will be shown below, the radical concentration is several times lower than the concentration of initial molecules.

Under conditions of the excess H₂, when the rate of pump reaction (3) exceeds the rate of formation of F atoms in the photolysis, the spatial distribution of the pump rate $Q_p(r,t)$ in reaction (3) should virtually coincide with the spatial distribution of the rate of formation of F atoms. In the simplified model that describes excitation of the active medium by emission of a cylindrical pump source, the pump rate can be represented in the form

$$Q_p(r,t) \equiv k_{3v} N_F N_{H_2} \sim N_{NF_3} f(r,t),$$

where N_F, N_{H_2}, and N_{NF_3} are the concentrations of F, H₂,

and NF₃; and the distribution $f(r,t)$ describes the excitation rate as a function of the distance r from the axis of the surface discharge.

The spatial distribution of the function $f(r,t)$ is determined by the attenuation of radiation by NF₃ molecules according to Bouguer's law and the cylindrical geometry of the pump source, which gives a correction in the form of the factor R/r, where R is the source radius:

$$f(r,t) = \frac{R}{r} \int \sigma_\lambda I_0^\lambda \exp\left[-\sigma_\lambda \int_R^r N_{NH_3}(r,t)dr\right]d\lambda.$$

Here I_0^λ is the spectral density of Planck's emissive power.

Fig. 5 presents the pump distribution functions for $R = 0.3$ and 0.5 cm, which were numerically obtained for $T = 30$ kK assuming that $N_{NF_3}(r,t) = N_0 = \text{const}$. One can see that the excitation of the active medium is efficient only inside a thin (thinner than 1 cm) cylindrical layer adjacent to the pump source. However, the experimental measurements (using apertures placed outside the cavity) of specific energies of laser emission from the region adjacent to the pump source ($r < 1.4$ cm) and the periphery region located at the distance $r > 1.4$ cm from the pump source showed that they were approximately identical and equal to 3.0 and 3.4 J litre⁻¹, respectively.
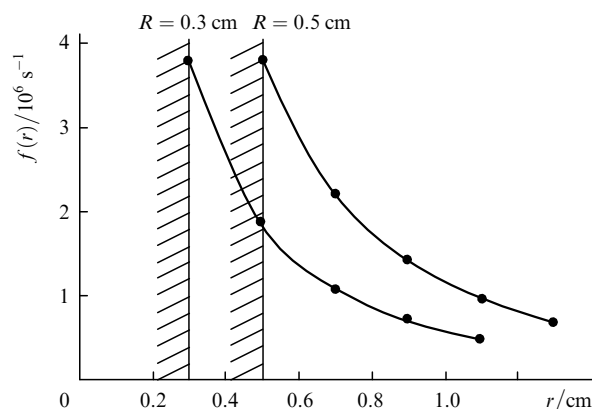

Figure 5. Pump distribution function $f(r)$.

This result suggests that the bleaching wave is formed in the active medium [11]. The essence of the phenomenon is that NF₃ molecules are completely dissociated within a layer of unit optical thickness adjacent to the pump source and, after that, radiation penetrates into deeper layers of substance and produces there the same effect. As a result, the absorbing layer and the excitation layer corresponding to it run away from the pump source. At a sufficiently high intensity of initiating radiation, which is typical of the pump source used in our study, the velocity of the bleaching wave should considerably exceed the velocity of discharge plasma expansion. Indeed, the brightness temperature of microsecond discharges for an electric energy of about several kilojoules is typically $20-30$ kK [11]. For the 40-nm effective width of the absorption spectrum of NF₃ (see Fig. 1), the photon flux I_0 into the absorption band reaches 8×10^{23} photon cm⁻² s⁻¹ ($T = 30$ kK) and the bleaching wave velocity at the boundary with the source is $V = I_0/N_0 = 40$ km s⁻¹, where N_0 is the concentration of NF₃ molecules corresponding to an optimum pressure of 6 Torr [11]. This

velocity considerably exceeds the typical velocity of discharge expansion, which is $1-2$ km s^{-1}.

The formation of the bleaching wave is also evident from the following reasoning. Assuming that the dominant contribution to lasing is made by the $2 \to 1$ transition and neglecting the lasing threshold , one can easily estimate the specific output laser energy ε in the case of complete photodecomposition of NF$_3$ molecules as

$$\varepsilon \approx N_0[\gamma_2 - (\gamma_1 + \gamma_2)/2]hv_{\rm L} = 2.8 \text{ J litre}^{-1},$$

where γ_1 and γ_2 are the fractions of HF molecules formed in the vibrational states $v = 1$ and 2, respectively; and $hv_{\rm L}$ is the laser photon energy. The difference between the specific laser energy ε obtained in this way and the experimental value can be explained by the contribution of other transitions and the secondary photolysis. In any case, this estimate of the specific energy is close to the experimental value. Thus, to explain the experimentally observed specific output energy, one should assume that there occurs complete photodecomposition of NF$_3$ molecules in the active region. Moreover, the comparison of the estimate with the experimental result suggests that the quantum yield of laser emission per one fluorine atom formed through photodissociation is close to unity.

Another evidence in favour of complete decomposition of NF$_3$ under the action of initiating radiation is the behaviour of the laser emission. Fig. 6 presents oscillograms of a laser pulse and discharge current for the gas mixture with the optimum composition NF$_3$:H$_2$:Kr = 6:10:125 Torr. One can see that the laser pulse is considerably shorter than the first half-period of current and, therefore, than the pump pulse (Fig. 3). This may be caused by complete photodecomposition of absorbing molecules long before the end of the pump pulse. Thus, an increase in laser aperture should lead to a more complete use of the pump energy.
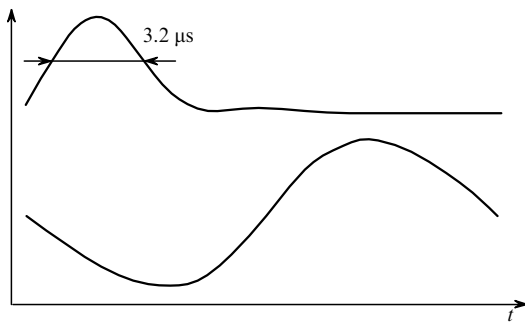

Figure 6. Oscillograms of a laser pulse (top) and current (bottom) for the mixture NF$_3$:H$_2$:Kr = 6:10:125 Torr.

The space-averaged velocity V of the bleaching wave can be estimated as the ratio of the laser aperture ($d = 2.8$ cm) to the laser pulse duration τ. For the duration $\tau = 3.5$ µs, which is observed for the gas mixture NF$_3$:H$_2$:Kr = 6:10:50 Torr, we have $V = 8$ km s^{-1}. Taking into account that $V \simeq RI_0/\rho N_0$ [11] (ρ is the radius of the bleaching wave), one can also estimate the photon flux I_0 in the absorption band of NF$_3$ at the source surface and the brightness temperature of the open discharge. Assuming that $R = 0.5$ cm and $\rho = 2.5$ cm at the end of a laser pulse, we obtain

$I_0 = 8 \times 10^{23}$ photon cm^{-2} s^{-1}, which corresponds to a brightness temperature of the pump source of about 30 kK for the 40-nm effective spectral width of the absorption band of NF$_3$ (Fig. 1). In the approximation of monochromatic excitation (σ_λ = const), the lifetime $\tau_{\rm p}$ of NF$_3$ molecules at the front of the bleaching wave can be estimated from the relation $\tau_{\rm p} = (\sigma_\lambda N_0)^{-1}/V$, where $(\sigma_\lambda N_0)^{-1}$ is the thickness of the absorbing layer; σ_λ is the absorption cross section; and N_0 is the initial concentration of NF$_3$ molecules. For the effective absorption cross section of about 3×10^{-18} cm^2 (Fig. 1), the estimate gives $\tau_{\rm p} = 2 \times 10^{-6}$ s. Thus, owing to the formation of the bleaching wave, the working substance in the active laser region is replaced every 2 µs.

The rate equations corresponding to the kinetic model (1) – (6) have the form

$$\frac{\partial[\text{NF}_3]}{\partial t} = -Q(t) + k_5[\text{F}][\text{NF}_2][\text{Kr}], \tag{7}$$

$$\frac{\partial[\text{F}]}{\partial t} = Q(t) - k_3[\text{F}][\text{H}_2] - k_5[\text{F}][\text{HF}_2][\text{Kr}], \tag{8}$$

$$\frac{\partial[\text{H}]}{\partial t} = k_3[\text{F}][\text{H}_2] - k_4[\text{H}][\text{NF}_2], \tag{9}$$

$$\frac{\partial[\text{NF}_2]}{\partial t} = Q(t) - k_4[\text{H}][\text{NF}_2] - k_5[\text{F}][\text{NF}_2][\text{Kr}], \tag{10}$$

where $Q(t)$ is the rate of NF$_3$ photolysis and square brackets denote concentration. Here, we ignored the secondary photolysis of NF$_2$ radicals and reaction (6), which is slow compared to other processes.

We can also show that for the Kr pressure of about 50 Torr, the regeneration of NF$_3$ molecules due to recombination of NF$_2$ radicals with atomic fluorine almost does not affect the formation of the beaching wave. Indeed, assuming [NF$_2$] = N_0 in Eqn (8), one can easily find that the recombination rate is considerably lower than the rate of pump reaction (3) in Eqn (8) and, therefore, the photodissociation rate Q as well because $\partial[\text{F}]/\partial t \simeq 0$ for the excess of H$_2$. Moreover, one can show that the recombination of NF$_2$ has no noticeable effect on the kinetics of the active medium.

For this purpose, we will first estimate the maximum concentration [F]$_{\rm max}$ of atomic fluorine achievable under the experimental conditions. It follows from (8) that

$$[\text{F}]_{\rm max} \simeq Q_{\rm max}(k_3[\text{H}_2])^{-1} = 0.25\sigma_\lambda I_0 N_0(k_3[\text{H}_2])^{-1}$$

$$\simeq 4 \times 10^{15} \text{ cm}^{-3},$$

where $Q_{\rm max} = 0.25\sigma_\lambda I_0 N_0 = 0.25(\tau_{\rm p})^{-1}N_0$ is the maximum photodissociation rate at the front of the bleaching wave (the factor 0.25 is caused by the fact that in the case of free propagation of the bleaching wave the region of the maximum photodissociation rate spatially coincides with the wave front section in which $I = 0.5I_0$ and $N = 0.5N_0$ [11]).

Assuming further that [H] $\simeq$ [NF$_2$] in (9), which will be evident later, and $\partial H/\partial t = 0$ and using the estimate obtained for [F]$_{\rm max}$, one can easily see that the rate of reaction of hydrogen atoms with NF$_2$ becomes comparable to the rate of their formation in reaction (3) even for the concentration as low as [H]$_{\rm max} \simeq$ [NF$_2$]$_{\rm max} \simeq 5 \times 10^{16}$ cm^{-3}. Therefore, the recombination rate for radicals in Eqn (10) is lower

than the rate of their reaction with atomic hydrogen by more than an order of magnitude. Taking this fact into account and comparing Eqns (9) and (10), one can easily verify the validity of the relation $[H] \simeq [NF_2]$. Thus, the radical recombination indeed has no effect on laser kinetics in our experimental conditions.

The relaxation of excited $HF(v)$ molecules occurs in their interaction with the components of the initial mixture (NF_3, H_2, and Kr) and the products of photochemical reactions (H, F, NF_2, NF, and HF). The rate constant for quenching of $HF(v)$ molecules by krypton is unknown, but, taking into account an extremely weak quenching of $HF(v)$ molecules by argon [6] and a low concentration of krypton, which was used in the experiments, it is reasonable to expect that the quenching by krypton is negligibly weak. The same may be said about the quenching by initial NF_3 molecules (the rate constant is 3×10^2 s^{-1} Torr^{-1} [6]), NF_2 radicals and atomic fluorine (the rate constants for $HF(v = 1, 2, 3)$ quenching are, respectively, 1.4×10^{-14}, 9.7×10^{-14}, and 2.5×10^{-13} cm^3 s^{-1} for NF_2 [10] and $(2-4) \times 10^{-13}$ cm^3 s^{-1} for H [12]), and atomic fluorine, whose concentration does not exceed 4×10^{15} cm^{-3} and rate constant for quenching is $2.7 \times 10^{-11} \exp(-1350\,\text{K}/T)$ cm^3 s^{-1} [6].

The only quenching processes that can noticeably affect the kinetic of the active medium are the quenching by $NF(a)$ molecules and the VV exchange in HF (the VT relaxation is slower [12]):

$$HF(v) + NF(a) \rightarrow NF(b) + HF(v-2), \quad (11)$$

$$HF(v) + HF(v+2) \rightarrow 2HF(v+1). \quad (12)$$
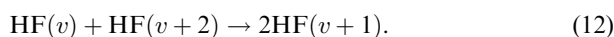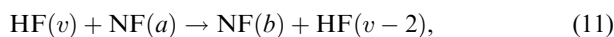
In this case, $k_{11v} = 8.3 \times 10^{-12}$ and 7.5×10^{-11} cm^3 s^{-1} for $v = 2$ and 3, respectively [10], and $k_{12v} = T^{-1}\{3\exp[-0.5 \times (300\,\text{K}/T)^{1/2}] - 1\} 6 \times 10^{-9}$ cm^3 s^{-1} for $v = 0$ [13].

Indeed, as noted above, the rate of formation of $NF(a)$ and $HF(v = 0)$ in reaction (4) is found to be comparable to the photodissociation rate even for concentrations as low as $[H] \simeq [NF_2] \simeq 5 \times 10^{16}$ cm^{-3}. Because of this, it is reasonable to expect that the concentrations of $NF(a)$ and HF $(v = 0)$ in the active medium of the optimum composition lie in the range of $10^{16} - 10^{17}$ cm^{-3}. Using the rate con-stants presented above, one can easily see that the quenching of $HF(v = 2, 3)$ in processes (11) and (12) may be of noticeable importance. Although the VV exchange does not change the energy accumulated in the vibrational reservoir, the effect of this process on laser characteristics of the active medium under consideration is not surprising because there, in contrast to the HF chain reaction laser, the population of the zero vibrational level takes place in reaction (4) in addition to excitation reaction (3), and these reactions have comparable rates.

The estimates presented above suggest that the optimum NF_3 concentration (2×10^{17} cm^{-3}) is indeed determined by the quenching of $HF(v)$ in processes (11) and (12). Moreover, the exact numerical calculations [14] for short (1 μs) pumping, which does not cause the formation of the bleaching wave, give the optimum concentration $[NF_3] = 2.3 \times 10^{17}$ cm^{-3}, which is close to the experimental value obtained in the present study.

As for the study of ClF_5 as a donor of fluorine atoms, the results obtained here should be treated as preliminary because they proved to be critical to the method of pre-

paration of the $ClF_5 - H_2$ mixture directly in the laser cell due to homogeneous and/or heterogeneous dark reactions proceeding in the mixture.

The optimum proportion $ClF_5 : H_2 = 3 : 20$ Torr was obtained for a krypton pressure of 50 Torr. In this mixture, we obtained an output laser energy of about 0.35 J using an 11-μF capacitor charged to 20 kV. The laser pulse had almost the same shape as in the experiments with NF_3 used as a donor of fluorine atoms.

The formation of the bleaching wave in the $ClF_5 - H_2$ mixture does not obviously follow from the absorption spectrum of ClF_5 in Fig. 1 because, according to our estimates, the probability of photodecomposition near the surface of the pump source emitting as a black body with a temperature of 30 kK is as low as $\sim 5 \times 10^5$ s^{-1}, which is almost an order of magnitude lower than the value for NF_3. Moreover, the excitation of the active medium in the UV region is homogeneous because we have in this spectral region $(\sigma_\lambda N_0)^{-1} > d$, where $(\sigma_\lambda N_0)^{-1}$ is the absorption depth for pump photons and d is the laser aperture. However, the bleaching-wave mode is possible if ClF_5 molecules have a sufficiently strong absorption continuum in the VUV region.

4. Conclusions

Thus, we observed for the first time lasing in a chemical HF laser, with NF_3 molecules used as a donor of fluorine atoms, using optical initiation of the active medium by emission of an open surface discharge and realised the bleaching-wave mode in a chemical HF laser. For the $NF_3 : H_2 : Kr = 6 : 10 : 125$ Torr mixture, the output laser energy reached 0.4 J in a 3.2-μs pulse, and the specific output power was 3.5 J litre^{-1}. The experimental results are qualitatively explained within the framework of a simple analytical model, whose analysis gives evidence of the formation of the bleaching wave in the active medium containing NF_3 molecules. Under these conditions, donors of fluorine atoms are decomposed in the region limited by the laser aperture long before the end of the pump. Therefore, an increase in the laser aperture should lead to a considerable increase in the output energy for a fixed pump energy. The bleaching-wave mode in the chemical HF laser offers new possibilities in the development of chemical lasers.

It is suitable for obtaining high specific output characteristics using initiation of a medium by long (longer than 10^{-5} s) radiation pulses. In the bleaching wave, the active medium in the lasing region is renewed with ultrasonic velocity (10 km s^{-1} and higher), which allows one to avoid self-deactivation of $HF(v)$ in the active medium at a high (several joules per litre) specific output energy. Lasing in a free photodissociation wave was first observed in an optically pumped XeF laser [11], whose output energy under these conditions reached 1 kJ when a strong shock wave in Kr was used as a pump source [15].

As for the use of ClF_5 molecules as donors of fluorine atoms, we observed for the first time lasing upon ClF_5 dissociation by emission of an open discharge. For the $ClF_5 : H_2 : Kr = 3 : 20 : 50$ Torr mixture, the maximum output energy in the 3.5-μs laser pulse reached 0.35 J. The results of our study are preliminary because they were found to be critical to the method of mixing ClF_5 with H_2. A more comprehensive study of ClF_5 as a donor of fluorine atoms, using an improved mixing technique, is of considerable interest be-

cause the use of this donor in an optically pumped HF laser offers a relatively high laser efficiency.

The development of an HF laser with a high total efficiency was not the aim of our study. However, the results obtained in the study of the active medium on the basis of NH_3 lead to some conclusions about a possible laser efficiency. In the laser design used in our experiments, only 1/6 of the total emission of the pump source propagates in the direction of the active medium limited by the laser aperture. Moreover, comparing the durations of the laser pulse (3.2 µs) and the initiating pulse (~ 10 µs), one can easily see that only 1/3 of this energy is used because of the bleaching of the medium. This means that by using all the emission energy in the absorption band of NF_3 (in the case of an axially symmetric arrangement of the pump source and a sufficiently large laser aperture), we can obtain a total laser efficiency (measured with respect to the energy stored in a capacitor) of about 0.4 %. Note that there are additional means of increasing the pump source efficiency by improving its electric matching.

References

1. Gregg D W et al. *Chem. Phys. Lett.* **8** 609 (1971)
2. Heidner R F III, Helvajian H, Koffend J B *J. Chem. Phys.* **87** 1520 (1987)
3. Collins R J, Husain D *J. Photochem.* **2** 459 (1972 – 1973)
4. N F Chebotarev *PhD (Phys) Thesis* (Moscow: L Ya Karpov Research Institute of Physical Chemistry, 1976)
5. Shirafuji T, Tachibana K *Appl. Surf. Sci.* **79/80** 117 (1994)
6. Bashkin A S, Igoshin V I, Oraevskii A N, Shcheglov V A *Khimicheskie Lazery* (Chemical Lasers) (Moscow: Nauka, 1982)
7. Borovich B L, Zuev V S, Katulin V A, Mikheev L D, Nikolaev F A, Nosach O Yu, Rozanov V B *Sil'notochnye Izluchayushchie Razryady i Gazovye Lazery s Opticheskoi Nakachkoi* (High-Current Emitting Discharges and Optically Pumped Gas Lasers) (Moscow: VINITI, 1978) v. 15
8. Gross R W F, Bott J F (Eds) *Handbook of Chemical Lasers* (New York: Wiley, 1976)
9. Malins R J, Setser D W *J. Phys. Chem.* **85** 1342 (1981)
10. Koffend J B, Gardner C E, Heidner R F III *J. Chem. Phys.* **83** 2904 (1985)
11. Zuev V S, Miheev L D *Photochemical Lasers* (Laser Science and Technology Series, Vol. 11) (Harwood Academic, 1991) v. 11
12. Mirels H, Hofland R, Whittier J S *J. Appl. Phys.* **50** 6660 (1974)
13. Igoshin V I, Pichugin S Y *Kvantovaya Elektron.* **21** 417 (1994) [*Quantum Electron.* **24** 384 (1994)]
14. Sabonnadiere M P, Tcheremiskine V I, Sentis M L, Mikheev L D, Delaporte Ph C *Proc. SPIE* **3574** 622 (1998)
15. Anisimov S V, Zemskov E M, Zuev V S, Kazanskii V M, Kashnikov G N, Mikheev L D, Nesterov R O, Sokolov V V, Stavrovskii D B, Cheremiskin V I *Laser Phys.* **4** 416 (1994)