HIGH-POWER SOURCES OF SPONTANEOUS UV RADIATION PUMPED BY PULSED AND CONTINUOUS DISCHARGES

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Design and performance parameters of high-power noble gas and halide excilamps, radiating at λ=193 (ArF*), 222 (KrCl*), 248 (KrF*), 308 (XeCl*), *and* 353 nm (XeF*) *and excilamps on* Cl_2^* *molecules at* λ = 258 nm *are presented. Pulsed longitudinal and barrier discharges and low-pressure continuous glow discharges were used to generate spontaneous UV radiation. Maximum mean power of excilamps up to* 40 W *at* λ = 258 nm, 130 W *at* λ = 222 nm, and 110 W *at* λ = 308 nm *and the efficiency up to* 10–20% *were achieved.*

INTRODUCTION

Sources of spontaneous radiation in the UV and VUV regions can be applied to stimulation of various photoprocesses. These sources are considerably simpler in design and operation as compared to lasers and emit in a wider range of wavelengths. Sources of spontaneous radiation on transitions of rare gas dimers R_2^* and rare gas monohalides RX* are the most efficient ones (Refs. 1–12). Hence, these sources are usually called "excilamps."

Excilamps are pumped by barrier discharge (Refs. 1, 5, 13-16), transverse discharge with a UV-preionization (Refs. 6, 10, 12, 17-19), longitudinal discharge (Refs. 16, 20), microwave discharge (Refs. 2, 3, 21, 22), and a spark discharge (Ref. 11). Recently, efficient emission from RX* molecules has been obtained in a low-pressure glow discharge (Ref. 7, 8). The maximum output of several tens of watts and efficiency about 10% were obtained under pumping by a microwave discharge.

EXPERIMENTAL SETUPS AND MEASUREMENT TECHNIQUES

Construction of excilamps studied is presented in Fig. 1. The excilamps are made of two coaxial quartz tubes of high quality with the transmission coefficient at λ = 222 nm no less than 80%. Diameter of the outer tube was typically 60 mm and discharge length did not exceed 40 cm. The gap between the tubes could be changed from 2.5 to 8 mm. In a lamp with longitudinal discharge two discharge gaps each 15 cm long were used. An additional metal electrode connected with a pulsed power supply was placed into the inner tube to improve the formation of longitudinal discharge. This electrode was not used in the glow discharge excilamp. In that case, electrodes of the excilamp were fed from a dc power supply, that provided the discharge current up to 0.5 A and voltage across the tube up to 10 kV. Cooling water circulates through the inner tube.

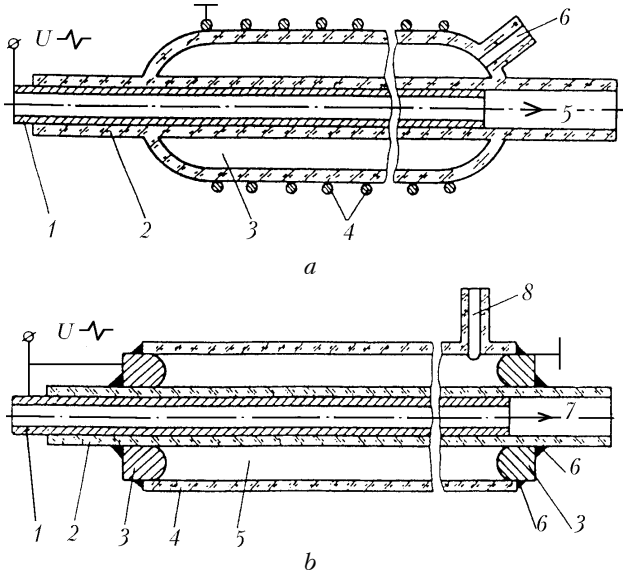

FIG. 1. Design of coaxial excilamps, pumped by barrier discharge (a), longitudinal pulsed and continuous glow discharge (b): a metal tube (reflector) 1, a coaxial lamp 2, a gas volume 3, an external mesh electrode 4, direction of water flow 5, a gas inlet 6 (a); a metal tube 1, an internal quartz tube 2, electrodes 3, an external quartz tube 4, a gas volume 5, seals 6, direction of water flow 7, a gas inlet 8 (b).

Two pulsed generators are used to produce barrier or longitudinal discharge. The first one includes storage and peaking capacitors and "Extra-2" or "Extra-3" thyratrons as a switch. Storage and peaking capacitance are varied in the range from 0.1 to 12 nF

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and from 0 to 6 nF, respectively. Pulse repetition rate is up to 2 kHz. The second generator produced ac pulses with the frequency of 2–15 kHz and voltage amplitude up to 15 kV.

Rare gas (Ne, He, Xe, Kr) – halogen (F_2, NF_3, HCl, Cl_2) mixtures were prepared in a separate gas-container. Gas purity was better than 99.9%. Three-gas mixtures composed of He(Ne), Xe(Kr), and halogen donor at a pressure up to 1-2 atm were used in pulsed excilamps. The glow discharge was investigated in Xe(Kr)–HCl (Cl_2) mixtures at a pressure up to 30 Torr.

Discharge current and voltage across the excilamps were monitored by means of a resistive shunt and divider that were coupled to a fast oscilloscope S8–14 or using a dc milliammeter and kilovoltmeter. Pulse waveform was fixed with a photodiode FEK–22SPU and an oscilloscope S8-14. Average output power was measured by a calorimeter IMO-2N or a photodiode FEK-22SPU. These measurements have been carried out in the following way. A diaphragm of 1 cm^2 area was placed directly against the excilamp surface. Powermeter was located at a distance of 20 cm from the diaphragm. The emitting area was considered as a point source, emitting uniformly in a solid angle of 4π. In this case, the portion of radiation power that enters powermeter can be calculated according to the following expression.

$$k = \sin^{-2} \alpha/2, \quad (1)$$

where $\alpha = \arctan A/2L$, A is the input aperture of the powermeter. Then the total radiation power was calculated taking into account the area of radiating surface as well as the angular and axial distribution of the output power.

Spontaneous radiation spectra were recorded by a spectrometer ISP-30 on RF-3 film or by a monochromator MDR-23, equipped with a photomultiplier FEU-100. The output signal from the PMT was amplified with a dc amplifier and then recorded with an X-Y recorder.

EXPERIMENTAL RESULTS AND DISCUSSION

Similar to results obtained in Ref. 4, 7, and 8, B-X and D-X bands of RX* molecules dominated in spectra of low-pressure glow discharge. In Kr–HCL(Cl_2) gas mixtures, up to 80% of the output was emitted in those bands. Besides, in Kr–HCL(Cl_2) gas mixtures $^3\Pi_{2g} - {}^3\Pi_{2u}$ band of chlorine molecules at $\lambda = 258$ nm was observed. In high pressure three-gas mixtures, intensity of D-X band decreased, the spectral width of B-X band decreased too. These facts indicate that in these mixtures the rate of collisional relaxation of RX* molecules is high. Spectra of gas mixtures containing F_2 and NF_3 are quite similar.

Figure 2 depicts pulsed power of KrCl* and KrF* excilamps pumped by a barrier discharge versus storage capacitance C_0 and charging voltage U_0. Output power increases with C_0 and U_0 increase, but the efficiency does not improve. That seems to be caused by the fact that the fraction of energy stored in C_0 and deposited into the discharge plasma is lower. Barrier discharge occurs in the form of bright thin filaments chaotically moving against the background of low intensity volume luminescence. In our experiments, output pulse duration varied in the range from 20 to 30 ns, whereas current duration was no longer than 200 ns. It is likely that the main part of the discharge current flows in those filaments with the current density about 1 kA/cm^2 (Ref. 14). At this current density the efficiency of the luminescence of the RX* molecules is low because of a high rate of their quenching by the electron impact. This leads to lower efficiency of RX* excilamp. However, these pumping conditions are preferable for rare-gas dimer formation. So, the efficiency of R_2^* excilamp pumped by a barrier discharge may be as high as 10% (Ref. 14).

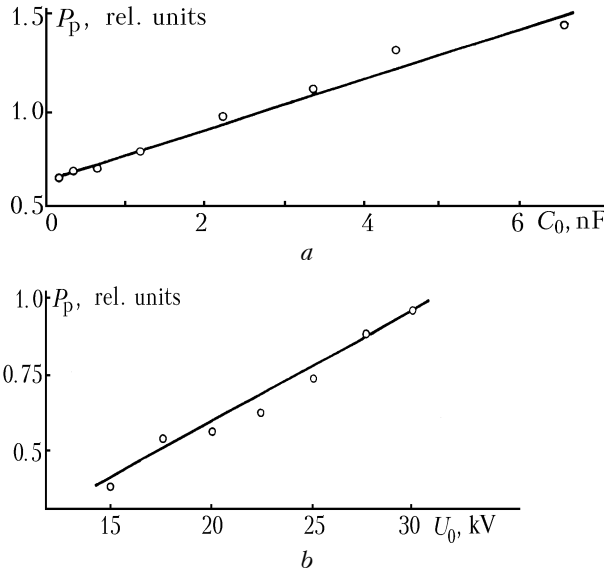

FIG. 2. Pulsed output power of an excilamp, pumped by a barrier discharge, versus storage capacitance C_0 (a) and charging voltage U_0 (b), measured in a gas mixture of He–Kr–HCl *at* $U_0 = 20$ kV *(a) and in a gas mixture of* He–Kr–NF_3 *at* $C_0 = 1.1$ nF *(b).*

Average output power of the KrCl* excilamp pumped by a barrier discharge versus pulse repetition rate is presented in Fig. 3. The power increases with pulse repetition rate up to 10 kHz. Excilamp can operate at higher pulse repetition rate since at 10 kHz no overheat is evident. Maximum output of the excilamp with the barrier discharge measured was 0.6 W when operating with KrCl* molecule and 1 W with KrF* molecule. It is significant that the lifetime of a gas mixture is long. For example, output power at $\lambda = 222$ nm did not change during 400 hours when the lamp was switched on for short time periods (total number of pulses was over 10^6).

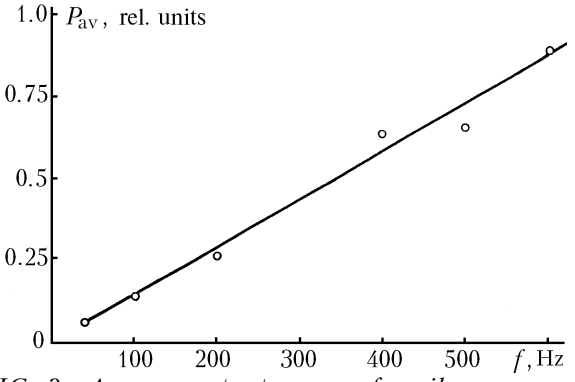

FIG. 3. Average output power of excilamp pumped by a barrier discharge versus pulse repetition rate, obtained in a gas mixture of He–Kr–HCl *at* U_0 = 20 kV *and* C_0 = 1.1 nF.

Figure 4 depicts output power at λ = 222 nm versus charging voltage of the storage capacitor U_0. Excilamps with one or two gaps were pumped by a longitudinal pulsed discharge. When charging voltage increased, a dramatic increase in output power was observed. Moreover, the picture of a discharge glow changed. An absolutely homogeneous discharge was obtained at low U_0 value. When U_0 was close but lower than 20 kV, a diffuse channel, chaotically moving in the discharge gap, occurred. Volume discharge with bright filaments bridging the gap was formed as U_0 value exceeded 20 kV. In the last case, the output energy was below 5 mJ in 250 ns pulse. Similarly to the barrier discharge pumping, efficiency of excilamp was low. This is probably caused by inhomogeneities in the longitudinal discharge.

Longitudinal discharge lamp showed linear increase in output power with the pulse repetition rate growing up to 800 Hz. Fast overheat of the excilamp and mixture degradation prevented from further increase of pulse repetition rate. Nevertheless, output of 4 W was obtained at a pulse repetition rate up to 2 kHz when the lamp was switched on for a short period of time.

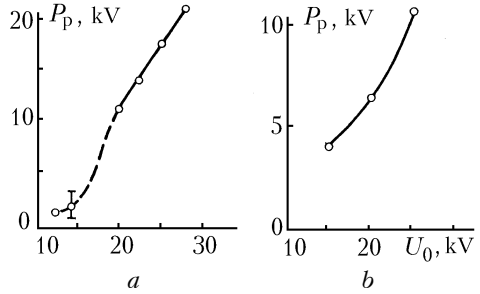

FIG. 4. Pulsed output power of excilamp, pumped by a longitudinal discharge as a function of charging voltage of a storage capacitor, observed under the following conditions: Ne–Kr–HCl *gas mixture,* C_0 = 4.4 nF, C_1 = 4.7 nF, *excilamp with one discharge gap (a);* $C_0 = C_1$ = 1.1 nF, *excilamp with two discharge gaps (b).*

Excilamp pumped by a continuous glow discharge in low-pressure Xe(Kr)–HCl(Cl_2) mixtures demonstrated the maximum output power and efficiency. In our experiments, glow discharge was easily formed at gas pressure up to 30 Torr. Besides, two forms of the glow discharge were found in agreement with the data reported in Refs. 7 and 8. Voltage across the excilamp $U_0 > 6$ kV and current not higher than $I_p \sim 2$–3 mA are typical for the first stage. Further increase of the current resulted in a step-wise change of the discharge form. The second form of the glow discharge was characterized by the voltage across the excilamp U_g lower approximately by a factor of two as compared to the previous case. Moreover, under these conditions discharge current was determined only by a dc power supply and could reach 0.5 A.

Very uniform emission from the whole discharge volume was observed at the first stage. Besides, no reduction of the UV output was recorded in the cathode region. A detailed study of this form of the glow discharge was performed with the optimal gas mixture Kr:HCL(Cl_2) = 10:1 at a total pressure of 6 Torr. Output power about 2.5 W at λ = 222 nm and lamp efficiency of 10–15% were obtained. The output was found to be nearly the same when HCl and Cl_2 containing gas mixtures were used. This result contradicts the data reported in Refs. 7 and 8. Addition of helium into the mixture increased the output by a factor of two. So, radiation power of 5 W at λ = 222 nm and lamp efficiency up to 20% were obtained with the gas mixture He:Kr:Cl_2 = 20:10:1 at a total pressure of 6 Torr. In this pumping regime, no overheat of excilamp was evident and, hence, no changes in its output were observed over a long period of operation. With an active volume of excilamp being 200 cm^3, the specific output power was calculated to be P_{sp} = 25 mW/cm^3. Further increase of output can be achieved due to an increase in the excilamp dimensions. For instance, to obtain output of P = 100 W and P = 1 kW, active volume V = 4 liters and V = 40 liters is required, respectively.

High value of E/p parameter is typical of the first stage of discharge. It is caused, probably, by high rate of the electron attachment to halogen molecules. This assumption is supported by the fact that there is emission of Cl_2^* molecules in the gas mixture containing Cl_2 and HCl with nearly equal intensities. Hence, fast dissociation of Cl_2 and HCl molecules occurs providing chlorine ions and further formation of Cl_2^* molecules. The E/p value is as large as 30 V/(cm·Torr) in this case. Therefore, direct ionization of inert gas atoms by electron impact dominates in the discharge plasma (see Ref. 28). Then, it can be concluded that radiating molecules are formed in the process of ion-to-ion recombination:

$$Kr^+(Kr_2^+) + Cl^- \rightarrow KrCl^* + (Kr). \quad (2)$$

The increase in the excilamp output power observed when helium is added to the mixture is connected, probably, with the additional formation of RX* molecules due to the following process.

$$Kr^+(Kr_2^+) + Cl^- + He \rightarrow KrCl^* + He + (Kr). \quad (3)$$

As was mentioned above, discharge current increased by a factor of about 100 when transition from the first form of the discharge to the second one was observed. Similar increase in the output power was also observed. Output power and efficiency of XeCl* and KrCl* excilamp versus discharge current are presented in Fig. 5.

Maximum output of 110 W at $\lambda = 308$ nm and 130 W at $\lambda = 222$ nm was obtained at the efficiency up to 14%. Note that discharge in this form occupies only a part of the active volume, but remains diffuse and uniform. This can cause overheat of the excilamp in the discharge region and reduction of the output. So, gas mixture lifetime was not longer than one hour. In order to reach longer lifetime, water cooling should be improved. Our experiments indicate that the discharge in xenon-containing mixtures is wider than it is in krypton-containing mixtures. Besides, it can be expanded by increase of discharge current. Discharge expansion in xenon-containing mixture resulted from higher electron drift velocity in xenon as compared to that in krypton (see Ref. 28).

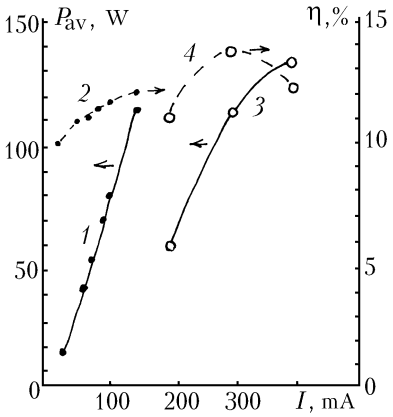

FIG. 5. Average output power (curves 1, 3) and efficiency (curve 2, 4) of excilamp with a continuous glow discharge versus discharge current in the following gas mixtures: Xe:Cl_2 = 3:1 *at a total pressure* P = 3 Torr *(curves 1, 2),* Kr:Cl_2 = 10:1 *at a total pressure* P = 6 Torr *(curves 3, 4).*

Pure Cl_2 at a pressure of 3 Torr and He:Cl_2 = 1:1 gas mixture at a pressure of 6 Torr were used to obtain emission at $\lambda = 258$ nm. Cl_2^*–excilamp with the output of 40 W and efficiency of 5% was developed. Output power in He–Cl_2 mixtures was higher by a factor of two than in pure Cl_2. Similarly to the previous cases, this improvement of Cl_2^*-excilamp operation is caused by higher rate of formation of Cl_2^* molecules in the three-body recombination process (see Ref. 25):

$$Cl^+ + Cl^- + He \rightarrow Cl_2^* + He. \quad (4)$$

It is worth noting that emission of Cl_2^*–excilamp produces ozone in the ambient air. Hence, VUV radiation with the wavelength shorter than 200 nm is present in the output. This radiation is related to transitions from the upper excited levels of chlorine molecules (see Ref. 26).

Experiments with the discharge in the second form demonstrated that in agreement with data from Refs. 7 and 8 the output decreased appreciably when chlorine in the mixture was replaced by hydrogen chlorine. Lower intensity of the emission of chlorine molecules was observed as well. This demonstrates lower rate of chlorine ion formation in the discharge plasma (see reaction (4)) in the mixtures containing hydrogen chlorine. Besides, production rate of rare gas metastable atoms is increased at low E/p value. Based on these facts, one can assume that when the discharge is in the second form, RX* molecules are mainly formed by the impacts of metastable rare gas atoms on chlorine molecules:

$$Xe^*(Kr^*) + Cl_2 \rightarrow XeCl^*(KrCl^*) + Cl. \quad (5)$$

Let us estimate the rate of formation of RX* molecules dN_{RX^*}/dt in the processes (3) and (5) in a dc glow discharge plasma using XeCl* molecules as an example. Relevant rate constants were taken from Refs. 25 and 27. Electron number density $N_e \sim 10^{11}$ cm^{-3} can be easily estimated based on the discharge current density and data on electron drift velocity in rare gases, presented in Ref. 28. Since the discharge plasma is quasineutral, the relationship between concentrations of xenon ions N_{Xe^+} and chlorine N_{Cl^-} is as follows:

$$N_{Xe^+} = (N_{Cl^-} + N_e).$$

Positive and negative ion concentrations may be close in glow discharge electronegative gas plasma (see Ref 28). So, if we assume that $N_{Cl^-} \sim N_e$, the rate of formation of exciplex molecules in process (3) can be estimated to be not higher than $dN_{XeCl^*}/dt \sim 10^{16}$–$10^{17}$cm^{-3}s^{-1}.

Concentration of metastable rare gas atoms can be estimated using the following expression:

$$N_{Xe^+} = K_b \times N_{Xe} \times N_e / K_g \times N_{Cl_2} \sim 5 \times 10^{10} \text{ cm}^{-3}. \quad (6)$$

Here K_b is the rate constant of xenon excitation by electron impact, K_g is that of process (5). Hence, the rate of process (5) $dN_{XeCl^*}/dt \sim 10^{18}$ cm^{-3} s^{-1} is much higher than that of process (3). Equilibrium concentration of XeCl* molecules at a given value of dN_{XeCl^*}/dt may reach $N_{XeCl^*} \sim 10^{10}$ cm^{-3}. This

calculated value is in a good agreement with the experimental results presented in Refs. 7 and 8.

Specific output power of XeCl*–excilamp pumped by a glow discharge in the second form reached 1 W/cm^3. This value corresponds to the emission from about 10^{18} XeCl* molecules in 1 cm^3 during 1 s. Close values of the rate of formation of exciplex molecules and the rate of their radiative decay indicate that radiationless decay of these molecules contributes to the glow discharge plasma kinetics. This result provides an explanation of high efficiency of excilapms pumped by a glow discharge. It should be noted that active volume of 1 kW excilamp comes to only 1 liter.

CONCLUSION

Thus, in the present paper, development of coaxial excilamps pumped by discharges of various types is reported. The maximum output power and efficiency were obtained in excilapms with a dc low–pressure glow discharge. Output over 100 W and efficiency over 10% were demonstrated. High output parameters of excilamps, pumped by glow discharge, are related to low rate of radiationless decay of the exciplex molecules and higher homogeneity of the glow discharge as compared to pulsed high-pressure discharges. Output scaling up to 1 kW is possible if the active volume of coaxial excilamp is increased. Besides, the use of gas mixtures with different composition can lead to appreciably wider range of wavelengths of the output radiation.

REFERENCES

1. V.I. Buinov, G.A. Voikova, and I.V. Podmoshenskii, Zh. Prikl. Spektrosk. **54**, No. 1, 164–166 (1991).
2. T. Hatakeyama, F. Kannari, and M. Obara, Appl. Phys. Lett. **59**, No. 4, 387–389 (1991).
3. H. Kumagai and K. Toyoda, Appl. Phys. Lett. **59**, No. 22, 2811–2813 (1991).
4. V.T. Mikxelso, A.B. Treshchalov, V.A. Peet, et al., Kvant. Elektron. **14**, No. 7, 1404–1406 (1987).
5. B. Gellert and V. Kogelschatz, Appl. Phys. B. **52**, No. 1, 14–21 (1991).
6. B.A. Koval', V.S. Skakun, V.F. Tarasenko, E.A. Fomin, and E.B. Yankelevich, Prib. Tekh. Eksp., No. 4, 244–245 (1992).
7. A.P. Golovitskii, Pis'ma Zh. Tekhn. Fiz. **18**, No. 8, 604–609 (1993).
8. A.P. Golovitskii and S.N. Kan, Opt. Spektrosk. **75**, No. 3, 604–609 (1993).
9. A.M. Boichenko, V.F. Tarasenko, E.A. Fomin, and S.I. Yakovlenko, Kvant. Elektron. **23**, No. 1, 3–25 (1993).
10. A.A. Kuznetsov, V.S. Skakun, V.F. Tarasenko, and E.A. Fomin, Pis'ma Zh. Tekhn. Fiz. **19**, No. 5, 1–5 (1993).
11. G.V. Runev and V.B. Saenko, Pis'ma Zh. Tekhn. Fiz. **19**, No. 21, 53–56 (1993).
12. A.M. Boichenko, V.S. Skakun, V.F. Tarasenko, E.A. Fomin, and S.I. Yakovlenko, Laser Physics **3**, No. 4, 838–843 (1993).
13. G.A. Voikova, N.N. Kirillova, E.N. Pavlovskaya, and A.V. Yakovleva, Zh. Prikl. Spektrosk. **41**, 681–695 (1984).
14. B. Eliasson and U. Kogelschatz, Pure and Appl. Chem. **62**, No. 9, 1667–1674 (1990).
15. U. Kogelschatz and H. Esrom, Laser and Optoelectronics **22**, 55–59 (1990).
16. V.A. Vizir', V.S. Skakun, G.V. Smorudov, E.A. Fomin, V.F. Tarasenko, and V.V. Chervyakov, Kvant. Elektron. **22**, No. 5, 541–545 (1995).
17. V.S. Skakun, V.F. Tarasenko, and E.A. Fomin, Zh. Prikl. Spektrosk. **56**, No. 2, 331–333 (1992).
18. A.M. Boichenko, V.S. Skakun, V.F. Tarasenko, E.A. Fomin, and S.I. Yakovlenko, Kvant. Elektron. **23**, No. 6, 532–534 (1993).
19. A.A. Kuznetsov, V.S. Skakun, V.F. Tarasenko, and E.A. Fomin, Atmos. Oceanic. Opt. **6**, No. 6, 408–409 (1993).
20. T. Gerber, W. Luthy, and P. Burkhard, Opt. Commun. **35**, No. 2, 242–244 (1980).
21. H. Kumagai and M. Obara, Appl. Phys. Lett. **54**, No. 26, 2619–2621 (1989).
22. H. Kumagai and M. Obara, Appl. Phys. Lett. **55**, No. 15, 1583–1584 (1989).
23. Ch.K. Rhodes, ed., *Excimer Lasers* (Springer Verlag, Berlin, Heidelberg, New York, 1979).
24. G.N. Gerasimov, B.E. Krylov, A.V. Loginov, and S.A. Shchukin, Usp. Fiz. Nauk **162**, 123–159 (1992).
25 A.M. Boichenko, V.I. Derzhiev, A.G. Zhidkov, et al., Trudy FIAN **21**, 44–115 (1989).
26. V.S. Zuev, A.V. Kanaev, and L.D. Mikheev, Kvant. Elektron. **11**, No. 2, 354–360 (1984).
27. H. Hokozono, K. Midorikava, M. Obara, et al., J. Appl. Phys. **52**, No. 3, 680–690 (1984).
28. Yu.P. Raizer, *Gas Discharge Physics* (Nauka, Moscow, 1987), 592 pp.