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Planar excilamp on rare gas chlorides pumped by a transverse self-sustained discharge

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***Abstract*. The design and parameters of a UV – VUV spontaneous radiation source – an excilamp operating on chlorides of rare gases $ArCl^*$, $KrCl^*$, and $XeCl^*$ in the wavelength range 175 – 308 nm are presented. The Ne – Xe (Kr, Ar) – HCl mixtures were excited by a high-pressure self-sustained discharge with spark preionisation. It is shown that upon pumping mixtures of rare gases and halogens by a transverse discharge, the intensities of the B – X emission band of molecules $ArCl^*$, $KrCl^*$, and $XeCl^*$ are comparable and up to 90 % of the emission energy of excilamps can be concentrated in the UV region. The peak UV power density at 222 ad 308 nm on the output window of the excilamp was ~ 2 kW cm^{-2} for the pulse energy up to ~ 3 mJ. The output emission energy of the excilamp at 175 nm achieved ~ 0.6 mJ and the peak power density was ~ 0.4 kW cm^{-2}.**

Keywords: *efficient high-power spontaneous UV radiation, high-pressure volume discharge*.

1. Introduction

High-power VUV – UV radiation sources are required for a number of applications in various fields in science, technology, medicine, etc. One of the types of UV radiation sources is electric-discharge exciplex lasers on molecules of rare-gas halides, however, they quite complicated and expensive in operation. In the cases not requiring coherent radiation, lasers can be replaced by simpler sources of spontaneous radiation – excilamps. The methods of excitation of excilamps have much in common with the well developed methods of pumping exciplex lasers. As a rule, as a source of high-power UV radiation a self-sustained discharge in various mixtures of rare gases and halogens is used, which is produced by pulsed high-voltage generators of various designs [1 – 11]. The average power of excilamps excited by a glow discharge exceeds 1 kW [10, 11]. Excilamps efficiently operate in broader ranges of pump parameters, composition, and pressure of gas mixtures than exciplex lasers, and their efficiency is, as a rule, higher. Excilamps are not critical to the excitation power and the active-region geometry, which simplifies the production of a volume discharge and allows the development of both wide-aperture and miniature spontaneous UV radiation sources. However, excilamps with a high average radiation power were earlier mainly developed, whereas a number of applications require a high radiation power density. At present only a few papers are known in which the $XeCl^*$ and $KrCl^*$ excilamps with a high radiation power density were studied [4 – 6]. At the same time, the emission characteristics of $ArCl^*$ molecules in a volume transverse discharge were studied insufficiently and the radiation power at 175 nm was not measured [7, 9].

The aim of this paper is to develop a pulsed planar excilamp on chlorides of rare gases with the UV – VUV radiation power density ~ 2 kW cm^{-2}. The excilamp was excited by a transverse discharge with UV preionisation, which allowed us to increase the pressure of the working mixture up to several atmospheres.

2. Excilamp design and measurement methods

Figure 1 shows the design of a planar excimer lamp pumped by a transverse discharge. The volume discharge was produced by using, as in [4 – 6], a pulsed generator consisting of a storage ($C_0 = 3.3 - 13.2$ nF) and peaking ($C_1 = 0.9$ nF) capacitors and a controlled RU-78 spark gap. The charge voltage of a storage capacitor C_0 was typically 25 kV. The discharge region had a diameter of 5 cm and the distance between a solid and grid electrodes was $d = 2.5$ cm. The transmission of the grid electrode was 65 %. The preionisation of a gas between the electrodes was performed by radiation from spark gaps located around the discharge region. Radiation was coupled out through a 12-mm thick quartz window with transmission 90 %, 80 %, and 37 % at wavelengths 308, 222, and 175 nm, respectively.

Gas mixtures consisting of rare gases Ne, He, Xe, Kr, Ar and halogens HCl and Cl_2 at the buffer gas pressure p_b up to 2.5 atm were prepared directly in a discharge chamber.

The discharge current and voltage across the discharge gap were measured with a Rogowski loop and a voltage divider. The emission spectrum of the excilamp in the range 200 – 800 nm was recorded with an EPP-2000 spectrometer (StellarNet Inc.) and in the range 150 – 500 nm with a VM-502 vacuum monochromator (Acton Research Corp.). The time dependence of the UV radiation power was measured with a FEK-22 SPU vacuum photocell. The shape of a radiation pulse at 175 nm was measured by covering the

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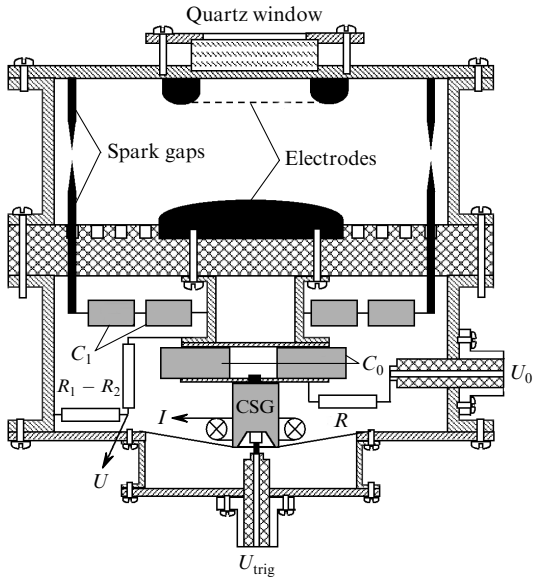

Figure 1. Design of an excilamp pumped by a transverse volume discharge: (C_0, C_1) storage and peaking capacitors; (CSG) controlled spark gap; (U, I) output signals of the voltage divider $R_1 - R_2$ and Rogowski loop, respectively; (U_0, U_{trig}) charge voltage and trigger pulse leads, respectively.

output window of the excilamp with sodium salicylate. The radiation energy at the wavelengths 175 and 222 nm was measured with a C8026 power meter with a CH8025 sensor head (Hamamatsu, the 150 – 250-nm sensitivity band) and at 222 and 308 nm with an OPHIR power meter with a PE-50BB sensor head (with the measuring area diameter of 50 mm) or with a FEK-22 SPU photocell (with the photocathode diameter of 40 mm), which was preliminary calibrated by using a DRL-400 mercury lamp, a MUM-2 monochromator, and an OPHIR power meter with a PD-300UV sensor head. The radiation energy at 175 nm was measured in the argon atmosphere. Electric signals were recorded with TDS-224 or TDS-220 digital oscilloscopes.

3. Emission spectrum of an excilamp

Figure 2 shows the survey emission spectrum of the volume discharge in the Ne : Xe : HCl = 830 : 3 : 1 mixture at the pressure $p_b = 2.5$ atm. The emission spectra of the excilamp operating on chlorides of different rare gases are close to each other. The radiation energy is mainly emitted in the UV – VUV range on the transitions of RCl^* molecules, where R is the Xe, Kr or Ar atom. Almost no radiation is observed outside this range except the separate weak lines of excited atoms and ions R^+, R^*, which contain only a few percent of the total radiation energy.

Figure 3 presents the detailed emission spectra of the volume transverse discharge in the Ne – Xe(Kr, Ar) – HCl mixtures. The relatively narrow D – X transition bands at 236, 199, and 169 nm, the B – X bands at 308, 222, and 175 nm, and a broad C – A band of RCl^* molecules dominate in the spectrum. Similar emission spectra were observed upon excitation of excilamps by different methods [1 – 7]. These spectra are related to the structure of molecules of halides of rare gases and are similar due to a similarity of kinetic processes proceeding in gases upon different methods of discharge excitation. The intensity ratio for the bands

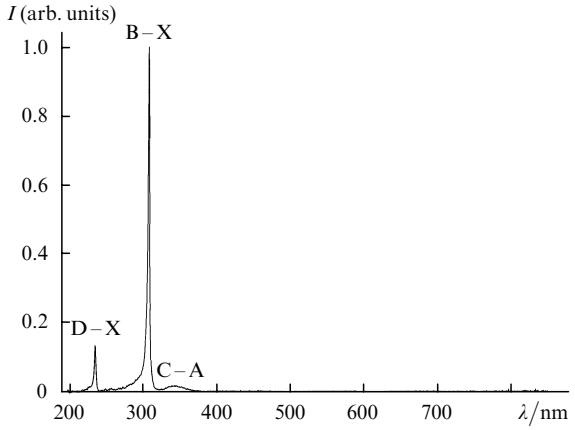

Figure 2. Survey emission spectrum of the XeCl excilamp on the Ne : Xe : HCl = 840 : 3 : 1 mixture in the wavelength range 190 – 850 nm for $p_b = 2.5$ atm.

depends on the method of discharge excitation. Thus, the intensity ratio for the D – X and B – X transitions in the glow and capacity discharges is higher than that in a high-pressure transverse discharge, and the half-width of the B – X band increases, achieving 10 nm [11].

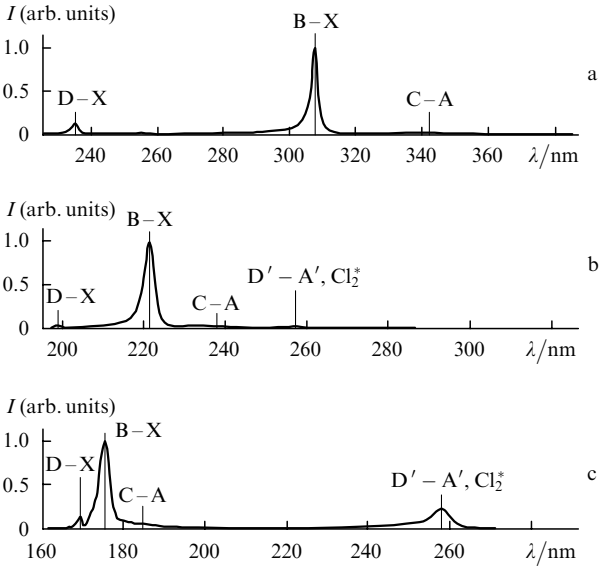

Figure 3. Detailed emission spectra of the excilamp on mixtures Ne : Xe : HCl = 840 : 3 : 1 for $p_b = 2.5$ atm (a), Ne : Kr : HCl = 1300 : 15 : 1 for $p_b = 2$ atm (b), and Ne : Ar : HCl = 2200 : 20 : 1 for $p_b = 2.5$ atm (c). The intensities of the bands of $ArCl^*$ molecules are corrected taking into account the transmission of the output quartz plate.

In addition, in the emission spectra of a discharge in mixtures with Kr and especially Ar, the 258-nm D′ – A′ band of excited Cl_2^* molecules is observed. The intensity of the chlorine band in the emission spectrum of a KrCl lamp is 3 % of the intensity of the B – X band of $KrCl^*$ molecules, which is close to the intensity of the spontaneous emission spectrum of the active medium of an electric-discharge KrCl laser [12]. The relative intensity of the Cl_2^* band in the Ne – Ar – HCl mixture increases by a factor of 5 – 7. The chlorine band in the emission spectra of ArCl and KrCl excilamps

can be related to reactions involving excited chlorine atoms in different states. These states can be populated due to the predissociation of exciplex molecules produced in the following processes:

$$R^+ + Cl^- + Ne \rightarrow RCl^* + Ne(H), \quad (1)$$

$$R^* + HCl(v) \rightarrow R + Cl^* + Ne(H). \quad (2)$$

Here, R^+ and R^* are the ion or metastable state of krypton or argon and $HCl(v)$ is a hydrogen chlorides molecule in an excited vibrational state. The high probability of reactions in channel (2) for R = Kr was pointed out in [13], and the concentration of Cl^* measured in the medium of an electric-discharge KrCl laser was comparable with that of the metastable Kr^* ions. The fraction of reactions proceeding via channel (2) in low-pressure mixtures with argon (R = Ar) can achieve 80 % [14, 15]. The excited chlorine molecules can be produced then both in the harpoon reaction [6]

$$Cl^* + HCl(v = 1) \rightarrow Cl_2^* + H, \quad (3)$$

and during the ion–ion recombination process

$$Cl^+ + Cl^- + Ne \rightarrow Cl_2^* + Ne, \quad (4)$$

where the positive chlorine ions are produced upon step ionisation of Cl^*.

The main energy emitted by a transverse-discharge pumped excilamp is concentrated in the B–X band of RCl^* molecules; the width of this band caused by rapid collision relaxation in a high-pressure gas does not exceed ~ 2 nm. Thus, under experimental conditions presented in Fig. 3, about 80 % of energy is emitted in the B–X band of $KrCl^*$ molecules in the wavelength range 200–230 nm and $XeCl^*$ molecules in the wavelength range 275–320 nm. In the case of an ArCl excilamp, up to 60 % of energy is emitted in the VUV region, the rest of the energy being mainly emitted on the 258-nm $D' - A'$ band of molecular chlorine. This means that the volume transverse discharge in the Ne–Ar–HCl mixture can be used as an efficient high-power VUV radiation source.

4. Voltage, current and emission characteristics of an excilamp

Figure 4 shows the oscillograms of voltage pulses across the discharge gap of an excilamp, of the discharge current of a storage capacitor, and radiation power for molecules $KrCl^*$ and $XeCl^*$. Because the active resistance of the transverse discharge under our experimental conditions did not exceed 30 % of the impedance of a pump generator, the discharge current of a storage capacitor oscillated and its amplitude weakly depended on the pressure of the working mixture. The UV radiation pulses appeared after the breakdown of the discharge gap of the excilamp. Before the breakdown, the current is caused by the charging of peaking capacitors through spark preionisation gaps, the maximum emission intensity of RCl^* molecules being achieved at instants close to those for the maxima of the discharge current. The duration of UV radiation pulses coincided with the pump pulse duration, which demonstrates a high homogeneity of the active medium of the excilamp. As the pressure of a gas

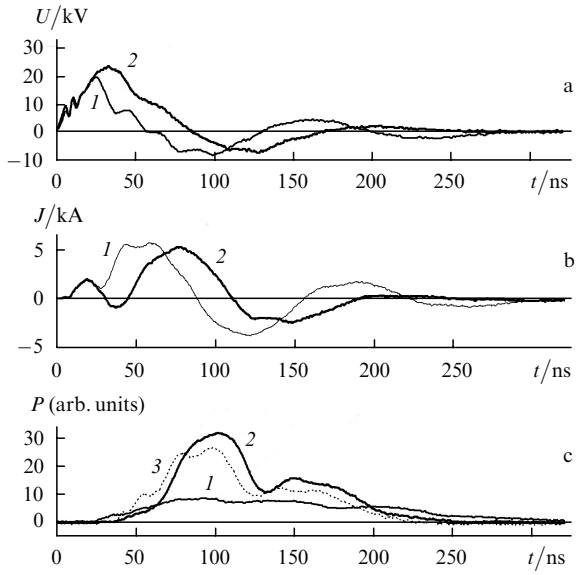

Figure 4. Oscillograms of voltage pulses on the discharge gap of the excilamp (a), of the discharge current of the storage capacitor (b), and radiation power at 222 nm (*1*, *2*) and 308 nm (*3*) obtained for mixtures Kr : HCl = 15 : 1 (p = 18 Torr) at the neon pressure 0.5 atm (*1*) and 2.5 atm (*2*) and Xe : HCl = 3 : 1 (p = 9 Torr) at the neon pressure 2.5 atm (*3*).

mixture was increased, the active resistance of the discharge gap also increased, resulting in an increase in the pump power and a drastic increase in the emission power and energy of chlorides of rare gases.

Figure 5 illustrates the influence of the type of a halogen carrier on the excilamp operation. Upon the replacement of hydrogen chloride by chlorine, the shape of the emission pulse of molecules of rare-gas chlorides changed, and the emission energy decreased approximately by a factor of 1.5 both at 222 and 308 nm. The emission power of $KrCl^*$ molecules in a mixture with Cl_2 increased during 10 ns after the discharge-gap breakdown and then gradually decreased. The time dependence of the emission power at 222 nm virtually coincides with the time dependence of the concentration of metastable krypton ions in the volume transverse discharge [16]. This means that RCl^* molecules in mixtures with chlorine are predominantly produced in the harpoon reaction

$$R^* + Cl_2 \rightarrow RCl^* + Cl. \quad (5)$$

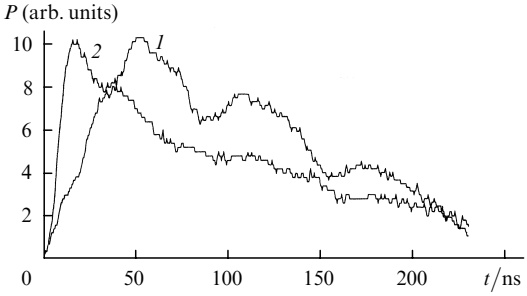

Figure 5. Oscillograms of the radiation power at 222 nm obtained for mixtures Kr : HCl = 15 : 1 (*1*) and Kr : Cl_2 = 15 : 1 (*2*) (p = 36 Torr) at the neon pressure 0.5 atm.

When HCl is used, molecules of rare-gas chlorides are mainly produced due to three-body ion–ion recombination [6]

$$R^+ + Cl^- + Ne \rightarrow RCl^* + Ne, \quad (6)$$

and that is why the emission power of RCl^* molecules has several maxima correlating with the maxima of the concentration of ions in a plasma, which are close in time to those of the discharge current.

The time dependences of the intensity of the $D' - A'$ emission band of the Cl_2^* molecule and RCl^* molecules observed in our experiments were similar. Therefore, we can conclude that the D' state of the Cl_2 molecule in the volume transverse discharge is predominantly populated due to ion–ion recombination [see reaction (4)].

The influence of the buffer-gas type on the emission energy of the excilamp is shown in Fig. 6. At a low pressure, the most efficient buffer gas is argon. However, as the gas pressure is increased, the intensity and duration of emission of RCl^* molecules in mixtures with Ar and He begin to decrease due to the development of spark channels in the discharge gap of the excilamp. The maximum energy of spontaneous radiation was obtained in gas mixtures with neon used as a buffer gas.

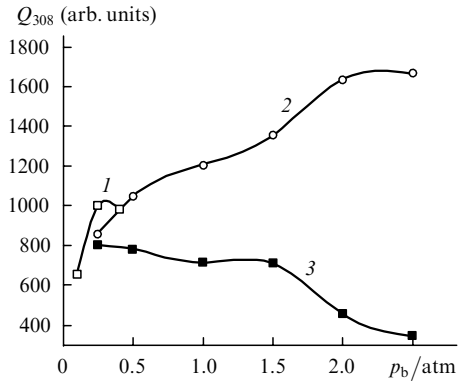

Figure 6. Dependences of the radiation energy of the excilamp at 308 nm on the pressure of buffer argon (*1*), neon (*2*), and helium (*3*) gases for the Xe : HCl = 3 : 1 mixture and $p = 9$ Torr.

Figure 7 presents the dependences of the energy density Q_{sp} and peak power density I_{sp} of emission at 222 and 308 nm on the distance to the external surface of the quartz window of the excilamp measured with a photodiode. One can see that the radiation parameters of the XeCl and KrCl excilamps virtually coincide. The minimum distance R at which the photodiode could be placed was 1.6 cm. The extrapolation of the experimental dependence to the output window surface ($R \sim 0.1$ cm) gives $Q_{sp} = 0.12$ mJ cm^{-2} at the excilamp output, which corresponds to the total radiation energy $Q_t = 2.4$ mJ for the quartz plate area $S = 20$ cm^2. In this case, I_{sp} achieves ~ 1.8 kW cm^{-2}. The values of Q_{sp} and I_{sp} calculated by the methods presented in [6] differed from the extrapolation data no more than by 20 %. The value of Q_t measured with an OPHIR power meter with a PE-50BB sensor head was 2.5 ± 0.2 mJ at a distance of ~ 5 mm to the output window surface. As the storage capacitance C_0 was increased to 13.2 nF, the total emission energy achieved 3 mJ and the peak power density increased up to ~ 2 kW cm^{-2}.

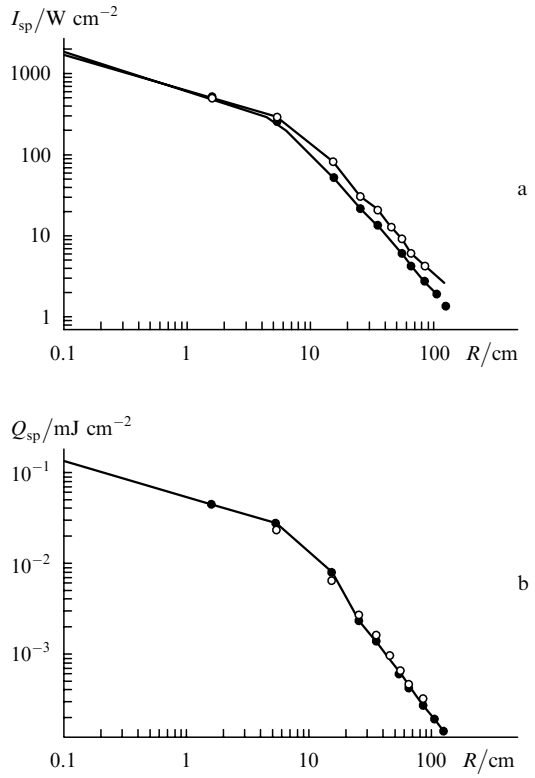

Figure 7. Dependences of the peak power density (a) and energy density (b) for UV radiation at 222 nm (empty circles) and 308 nm (dark circles) on the distance to the external surface of the output quartz mirror of the excilamp for mixtures Ne : Xe : HCl = 830 : 3 : 1 ($p_b = 2.5$ atm) and Ne : Kr : HCl = 680 : 15 : 1 ($p_b = 2$ atm). The experimental data (circles) are extrapolated by curves.

The characteristics of VUV emission of $ArCl^*$ molecules were approximately four times worse than those of $XeCl^*$ and $KrCl^*$ molecules. The output radiation energy Q_t of the excilamp at 175 nm achieved ~ 0.6 mJ for the peak power density $I_{sp} \sim 0.4$ kW cm^{-2}.

One can see from Fig. 7 that the radiation intensity decreases as $1/R^2$ at a great distance from the excilamp ($R \geqslant 10$ cm). This can be used to estimate the total energy emitted to the solid angle 4π assuming that the excilamp is a point light source. The maximum radiation energy, taking into account the transparency of a grid electrode, was 65 mJ for the energy from a storage capacitor supplied to the active medium of the excilamp equal to 1.4–1.6 J. Therefore, the efficiency of conversion of the pump energy to the UV radiation energy was 4 %–4.5 %, which is approximately twice the maximum efficiency of an electric-discharge KrCl laser [17] and coincides with the efficiency of a transverse-discharge-pumped KrCl excilamp calculated in [6].

5. Conclusions

We have developed a high-power VUV–UV spontaneous radiation source on chlorides of rare gases pumped by a transverse volume discharge in gas Ne–Xe (Kr, Ar)–HCl mixtures. The emission spectrum of the excilamp mainly consists of the relatively narrow D–X and B–X bands of chlorides of rare gases. The spectra of the $KrCl^*$ and $ArCl^*$ excilamps also exhibit, along with these bands, the 258-nm band of the excited molecular chlorine Cl_2^*.

We have found that upon pumping by a transverse discharge, the intensities of the B–X emission band of molecules $ArCl^*$, $KrCl^*$, and $XeCl^*$ in the volume transverse discharge are comparable and up to 90 % of the radiation energy of the excilamp can be concentrated in the UV region. The peak UV power density on the output window of the excilamp achieved ~ 2 kW cm^{-2} for the pulse energy up to ~ 3 mJ.

It has been shown that the volume discharge in the Ne–Ar–HCl mixture is an efficient high-power VUV radiation source. The radiation energy $Q_t \sim 0.6$ mJ and power density $I_{sp} \sim 0.4$ kW cm^{-2} of this mixture at 175 nm can amount to ~ 25 % of the corresponding values for the $XeCl^*$ and $KrCl^*$ excilamps.

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References

1. Eliasson B., Kogelschatz U. *Appl. Phys. B*, **46**, 299 (1988).
2. Nakamura I., Kannari F., Obara M. *Appl. Phys. Lett.*, **57**, 2057 (1990).
3. Taylor R.S., Leopold K.E., Tan K.O. *Appl. Phys. Lett.*, **59**, 525 (1991).
4. Koval' B.A., Skakum V.S., Tarasenko V.F., Fomin E.A., Yankelevich E.B. *Prib. Tekh. Eksp.*, (4), 244 (1992).
5. Boichenko A.M., Skakun V.S., Tarasenko V.F., Fomin E.A., Yakovlenko S.I. *Laser Phys.*, **3**, 838 (1993).
6. Boichenko A.M., Skakun V.S., Sosnin E.A., Tarasenko V.F., Yakovlenko S.I. *Kvantovaya Elektron.*, **23**, 344 (1996) [*Quantum Electron.*, **26**, 336 (1996)].
7. Shuaibov A.K., Shimon L.L., Dashchenko A.I., Neimet Yu.Yu., Shevera I.V. *Pis'ma Zh. Tekh. Fiz.*, **25**, 29 (1999).
8. Lomaev M.I., Skakun V.S., Sosnin E.A., Tarasenko V.F., Shitts D.V. *Pis'ma Zh. Tekh. Fiz.*, **25**, 27 (1999).
9. Shuaibov A.K., Dashchenko A.I. *Kvantovaya Elektron.*, **30**, 279 (2000) [*Quantum. Elecrtn.*, **30**, 279 (2000)].
10. Skakun V.S., Lomaev M.I., Tarasenko V.F. *Pis'ma Zh. Tekh. Fiz.*, **28**, 42 (2002).
11. Lomaev M.I., Skakun V.S., Sosnin E.A., Tarasenko V.F., Shitts D.V., Erofeev M.V. *Usp. Fiz. Nauk*, **173**, 201 (2004).
12. Peet V.E., Slivinskii E.V., Treshchalov A.B. *Kvantovaya Elektron.*, **17**, 438 (1990) [*Sov. J. Quantum. Electron.*, **20**, 372 (1990)].
13. Andrew E., Dyer P.E., Roebuck J. *Opt. Commun.*, **49**, 149 (1984).
14. Sadeghi N., Cheaib M., Setser D.W. *J. Chem. Phys.*, **90**, 219 (1989).
15. Tsuji M., Ide M., Muraoka T., Nishimura Y. *J. Chem. Phys.*, **101**, 328 (1994).
16. Nakamura K. *J. Appl. Phys.*, **83**, 1840 (1988).
17. Panchenko A.N., Tarasenko V.F. *IEEE J. Quantum Electron.*, **31**, 1231 (1995).