Emission spectra of capacitive, glow, and barrier discharge $Kr-Cl_2$, $Xe-Cl_2$, $Xe-Br_2$, $Xe-I_2$ excilamps

A.A. Lisenko and M.I. Lomaev

Institute of High-Current Electronics,
Siberian Branch of the Russian Academy of Sciences, Tomsk

Received December 26, 2001

The spectra of $Xe-Cl_2$, $Xe-Br_2$, and $Xe-I_2$ excilamps excited by capacitive, glow, and barrier discharges are presented. The spectra of a barrier discharge $Xe-Cl_2$ excilamp for different gas mixture pressure (60 and 120 Torr) are obtained and analyzed. The dependence of $Kr-Cl_2$ capacitive discharge excilamp spectrum on the operation time is presented.

1. Excilamps present a relatively recent class of sources of spontaneous UV and VUV radiation that are based on the use of nonequilibrium emission of so-called excimer and exciplex molecules. A feature of these molecules is their stability in the excited electronic state and the absence of a strong bond in the ground state. Some such molecules have the intense B–X transition in the UV or VUV regions. This allows high-efficiency (up to ≈ 60%) transformation of the pumped energy into optical radiation. Excilamps are now used in photochemistry, microelectronics, for modification of surface properties, in medicine, in industrial waste, air, and water decontamination technologies, etc., and this explains the great attention paid to their study.[1–11] For exciting working medium, various types of self-sustained discharge, such as glow discharge, barrier discharge, capacitive discharge, and high-pressure discharge with UV preionization are commonly used in practice.

The main features distinguishing excilamps from available luminescent (mercury and hydrogen) lamps, as well as thermal sources of spontaneous UV and VUV radiation are their spectrum and output power. Up to ~ 80% and more of the total output power can be concentrated in a rather narrow (less than 10 nm at halfmaximum) band of the B–X transition of a corresponding molecule. The conditions for emission of two and more exciplex molecules are realized as well. The characteristic output power of excilamps exceeds that for low-pressure hydrogen and mercury lamps.

Potential advantages of excilamps from the viewpoint of their application are the photon energy (4–10 eV), spectral selectivity, high output power, feasibility of scaling and selecting random geometry of the emitting surface. Of special note is the absence of mercury in such lamps, what gives them an advantage over relatively widely used, but ecologically hazardous mercury-containing lamps.

From the point of view of practical application, emission spectrum and its stability, along with such parameters as efficiency, output power, and service life, are among the most important characteristics of an excilamp. This paper is devoted to determination and comparison of emission spectra of capacitive, glow, and barrier discharge excilamps designed in the Laboratory of Optical Radiation of IHCE SB RAS.[9–14]

2. Discharge emission spectra were recorded with a specialized system including an MDR-23 monochromator (inverse linear dispersion of 1.3 nm/mm) with a grating of 1200 lines/mm, a broadband FEU-100 photomultiplier, a TDS 3032 oscilloscope, and a computer. An optical signal at excilamp excitation from a step-up network transformer (glow discharge lamp) or a pulse generator with the pulse repetition rate up to 100 kHz (capacitive or barrier discharge) came to the entrance slit of the monochromator. The electrical signal from the photomultiplier came to the oscilloscope, then it was digitized, and the information was transferred into the computer and stored there.

With the proper processing of the obtained information, we could get the emission spectrum of an excilamp for a given time from the beginning of the excitation pulse, as well as the spectrum integral over the whole duration of the excitation pulse. In the first case, we obtained the power spectrum for a given time, and in the second case it was the pulse energy spectrum, as in the traditional photographic recording of a spectrum. In this paper, we present the emission spectra integral over the pulse duration. The spectral halfwidth of the instrumental function of the system at the minimal revolution rate of the diffraction grating was no wider than 0.02 nm. This mode was used for determination of line profiles or emission bands. When recording survey spectra (in a wide spectral region), the revolution rate of the diffraction grating increased, thus leading to the increasing halfwidth of the instrumental function.

In the experiments we used sealed-off $Kr-Cl_2$, $Xe-Cl_2$, $Xe-Br_2$, $Xe-I_2$ capacitive discharge excilamps, $Kr-Cl_2$ and $Xe-Cl_2$ barrier and glow discharge excilamps. In Refs. 9, 12, and 14 the designs of the used excilamps are described, and the Table presents their general characteristics.

3. Figure 1 shows the emission spectra of the positive discharge column of capacitive excilamps for the

0235-6880/02/03 263-04 $02.00

conditions given in the Table. The spectral halfwidth of the most intense B–X transition of KrCl* (λ = 222 nm), XeCl* (λ = 308 nm), XeBr* (λ = 282 nm) molecules is, respectively, ~ 4.5, 8, and 7 nm. The spectra in this region are characterized, first, by the presence of pronounced D–X and C–A transitions of the above molecules. Second, in the case of XeBr* and XeCl* molecules, a wide and intense blue wing of the B–X transition was observed. The atomic iodine line at λ = 206 nm (Fig. 1*d*) turned out to be the most intense line in the spectrum of the $Xe–I_2$ excilamp at low pressure (~ 1.5 Torr), and the band corresponding to the B–X transition of the XeI* molecule (λ = 253 nm) had much lower intensity. As the pressure increased, the intensity ratio of this line and the band changed, and at the pressure ~ 10 Torr they had comparable amplitudes.

Figure 2 shows the emission spectra of $Kr–Cl_2$ and $Xe–Cl_2$ excilamps at glow discharge excitation. Comparison of Figs. 1 and 2 allows the conclusion that the spectral distribution of radiation is almost the same in the both cases. This is likely the consequence of the fact that such characteristics of the working mixture as the pressure and composition, as well as the reduced electric field strength E/P (E is the electric field strength, P is pressure) and efficiency are similar.

Table. General characteristics of the studied capacitive, barrier, and glow discharge excilamps

Characteristic	Type of discharge							
	capacitive				barrier		glow	
Working mixture	$Xe–Cl_2$	$Kr–Cl_2$	$Xe–Br_2$	$Xe–I_2$	$Xe–Cl_2$	$Kr–Cl_2$	$Xe–Cl_2$	$Kr–Cl_2$
Working mixture pressure, Torr	6	6	4.5	1.5	120	120	6–9	6–9
Interelectrode gap, cm	20	20	20	50	0.8	0.8	20–80	20–80
Outer tube diameter, cm	3.8	4.2	4.2	3.5	6.5	6.5	1-6	1-6
Wavelength of the maximum of spectral distribution, λ, nm	308	222	282	206	308	222	308	222

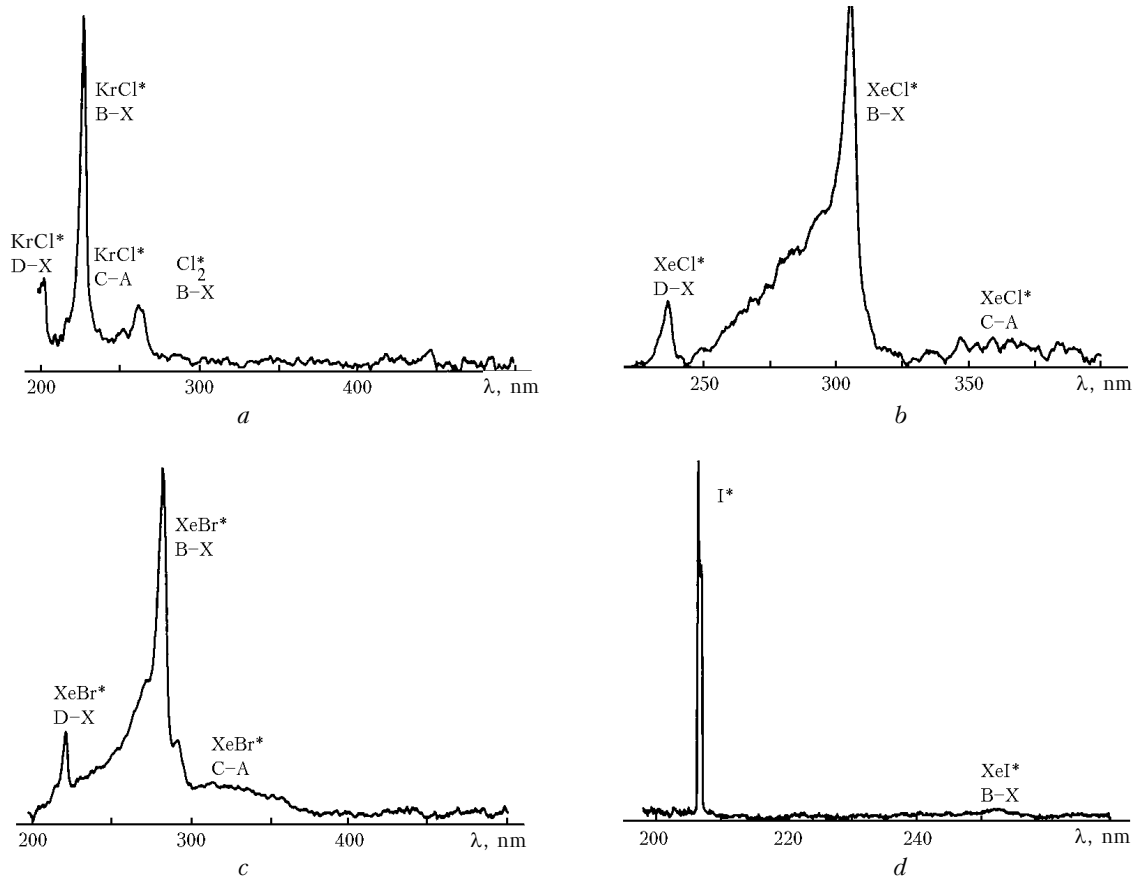

Fig. 1. Emission spectra of capacitive discharge excilamps for mixtures: $Kr–Cl_2$ at pressure of 6 Torr (*a*), $Xe–Cl_2$ at pressure of 6 Torr (*b*), $Xe–Br_2$ at pressure of 4.4 Torr (*c*), and $Xe–I_2$ at pressure of 1.5 Torr (*d*).

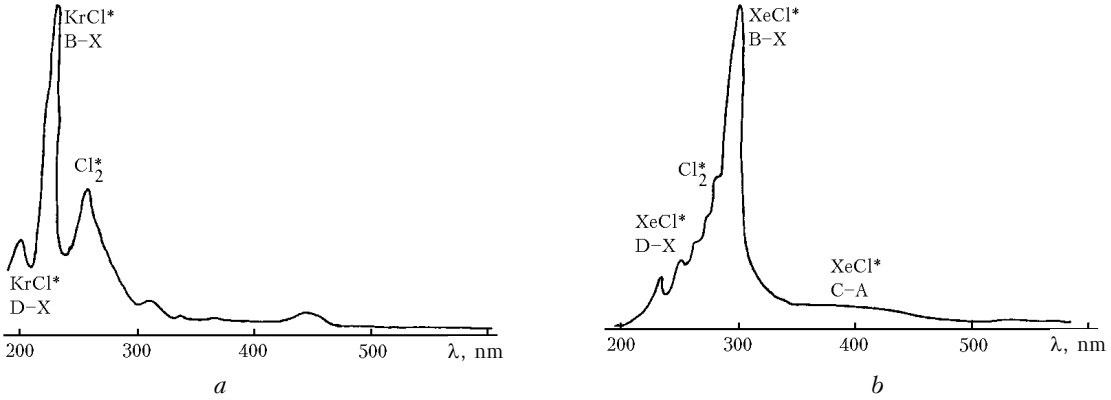

Fig. 2. Emission spectra of glow discharge excilamps for mixtures: $Kr–Cl_2$ at pressure of 6 Torr (*a*) and $Xe–Cl_2$ at pressure of 6 Torr (*b*).

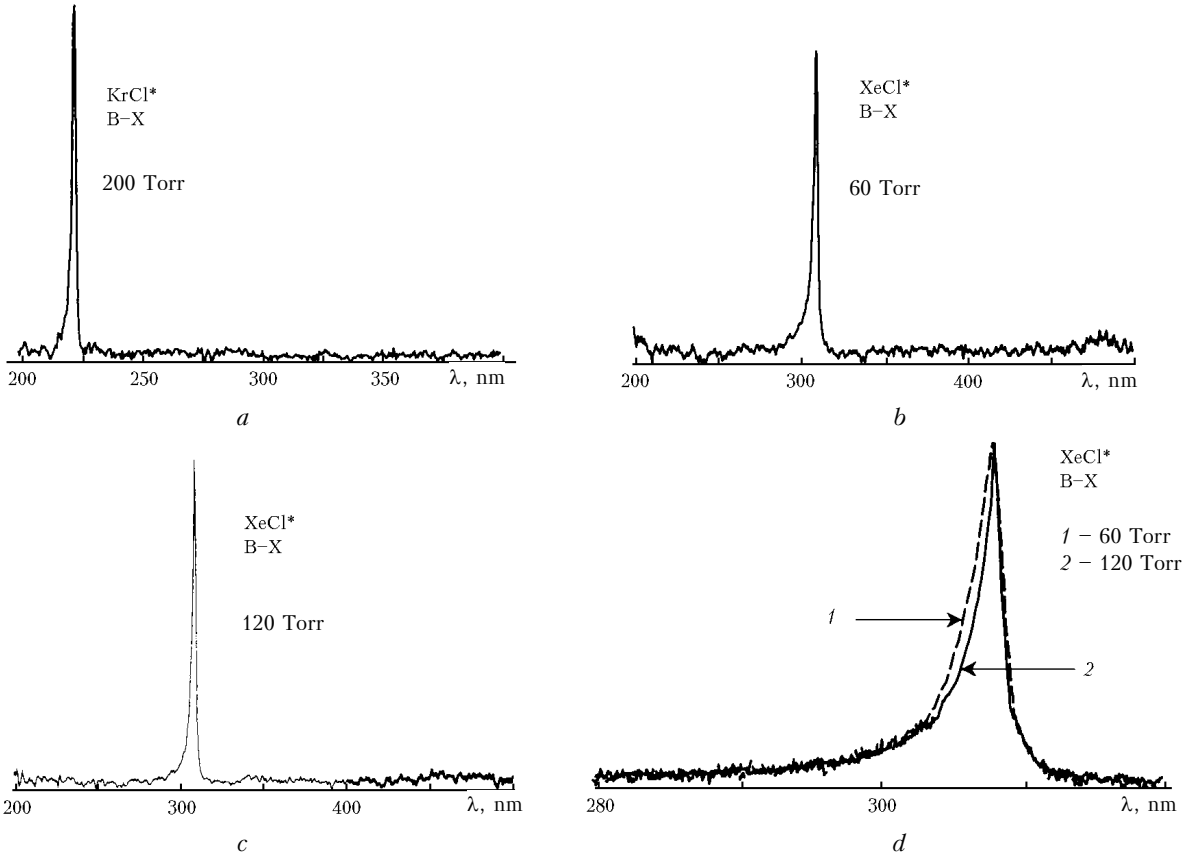

Fig. 3. Emission spectra of barrier discharge KrCl excilamp at mixture pressure of 200 Torr (*a*) and barrier discharge XeCl excilamp at mixture pressure of 60 and 120 Torr (*b–d*).

A characteristic feature of the barrier discharge is a small interelectrode gap, which allows using a high pressure of the working mixture. Figure 3 depicts the emission spectra of $Kr–Cl_2$ and $Xe–Cl_2$ excilamps at this method of excitation. As can be seen from Fig. 3, the main feature is the presence only of bands of B–X transitions in the spectrum. The spectral halfwidth of the B–X transition of the KrCl* molecule is ~ 2 nm. The bands of the D–X and C–A transitions of the KrCl* molecule, as well as the band of the Cl_2^* molecule that were present in the emission spectrum at excitation by the capacitive and glow discharges, are almost absent in the case of the barrier discharge. Figures 3*b*, *c*, and *d* illustrate the effect of the pressure on the halfwidth of the BX transition of the XeCl* molecule. Thus, at the pressure of 60 Torr it is ~ 3.2 nm, and at the pressure of 120 Torr it is ~ 1.8 nm. The decrease in the halfwidth of the emission band is explained by increasing rate of vibrational relaxation of molecules at increasing pressure.[15]

The stability of the emission spectrum was checked for sealed-off capacitive discharge excilamps, whose service life is no less than 1000 h (Refs. 12 and 16). Figure 4 shows the spectral distribution for the $Kr–Cl_2$ as a function of the excilamp operation time. The decrease in the output power was no more than 15%. It can be seen that the emission spectrum remained almost unchanged. In extra experiments on recording of a spectrum after different periods of excilamp operation, the decrease in the fraction of radiation of D–X and C–A transitions was noticed as well.

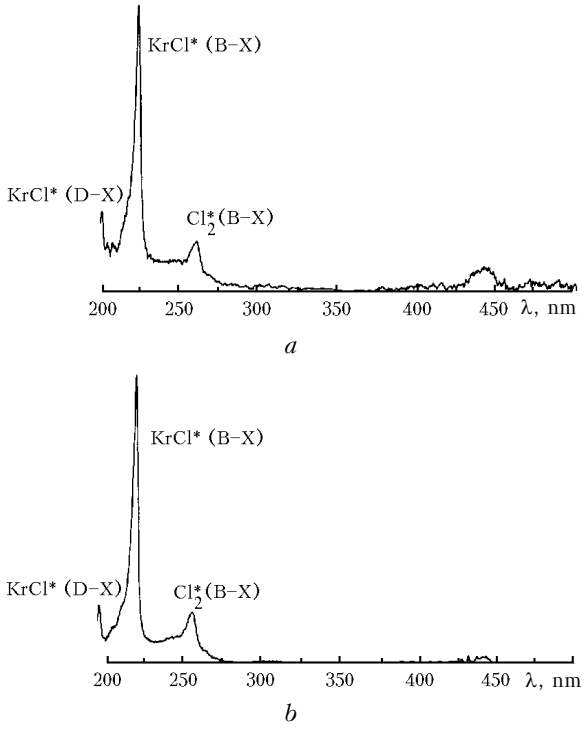

Fig. 4. Spectrum of capacitive discharge $Kr–Cl_2$ excilamp at the operation time of 100 (*a*) and 625 h (*b*).

4. In conclusion let us summarize the main results of this work.

First, only relatively narrow bands of exciplex molecules were observed in the emission spectrum of excilamps in the region from 200 to 500 nm. By selecting a working mixture (and, correspondingly, an emitting molecule) we can provide the emission in a needed spectral region.

Second, it should be noted that the spectra of capacitive discharge and glow discharge excilamps are similar. These spectra included the bands of B–X, D–X, and C–A transitions of the corresponding exciplex molecule, as well as the halogen molecule. At excilamp excitation by a barrier discharge, the emission spectrum included only the narrow B–X band with characteristic FWHM ~ 2–3 nm. The emission spectrum of the $Xe–I_2$ excilamp depended on the mixture pressure and included mainly the atomic iodine line at low pressure and the atomic iodine line and the band of the XeI* molecule at the increased pressure.

Third, the emission spectrum of sealed-off capacitive discharge excilamps remained almost unchanged for a long lamp operation period comparable with the lamp service life.

Acknowledgments

The authors are thankful to V.F. Tarasenko for his support of this work, E.A. Sosnin, M.V. Erofeev, and D.V. Shitz for placing at our disposal the capacitive discharge excilamps and excitation generators.

This work was partly supported by the ISTC (Project No. 1270).

References

1. *Proc. of the 9th Int. Symp. on Science & Technology of Light Sources, LS-9* (Cornell University, Ithaca, NY, USA, 2001).
2. *Proc. of the 8th Int. Symp. on Science & Technology of Light Sources, LS-8* (Greifswald, Germany, 1998).
3. *Proc. of the 7th Int. Symp. on Science & Technology of Light Sources, LS-7* (Kyoto, Japan, 1995).
4. A. Oda, H. Sugawara, Y. Sakai, and H. Akashi, J. Phys. D **33**, 1507–1513 (2000).
5. R.P. Mildren and R.J. Carman, J. Phys. D **34**, L1–L6 (2001).
6. Xueji Xu, Thin Solid Films **390**, 237–242 (2001).
7. J.-Y. Zhang and I.W. Boyd, Appl. Surface Sci. **168**, 296–299 (2000).
8. U. Kogelschatz, B. Eliasson, and W. Egli, Pure Appl. Chem. **71**, No. 10, 1819–1828 (1999).
9. A.N. Panchenko, E.A. Sosnin, and V.F. Tarasenko, Opt. Commun. **161**, 249–252 (1999).
10. M.I. Lomaev, A.N. Panchenko, E.A. Sosnin, and V.F. Tarasenko, Zh. Tekh. Fiz. **68**, No. 2, 64–68 (1998).
11. M.I. Lomaev, A.N. Panchenko, V.S. Skakun, E.A. Sosnin, V.F. Tarasenko, M.G. Adomson, B.R. Myers, and F.T. Wang, Laser and Particle Beams **15**, No. 2, 339–345 (1997).
12. M.I. Lomaev, V.S. Skakun, E.A. Sosnin, V.F. Tarasenko, and D.V. Shitz, Pis'ma Zh. Tekh. Fiz. **25**, No. 21, 27–32 (1999).
13. A.N. Panchenko and V.F. Tarasenko, Optics and Spectroscopy **84**, No. 3, 337–339 (1998).
14. M.I. Lomaev, A.N. Panchenko, V.S. Skakun, E.A. Sosnin, and V.F. Tarasenko, Izv. Vyssh. Uchebn. Zaved., Fiz., No. 5, 69–72 (2000).
15. V.Yu. Baranov, V.M. Borisov, and Yu.Yu. Stepanov, *Electric Discharge Excimer Lasers* (Energoatomizdat, Moscow, 1988), 216 pp.
16. M.V. Erofeev, V.S. Skakun, E.A. Sosnin, V.F. Tarasenko, and E.B. Chernov, Atmos. Oceanic Opt. **13**, No. 3, 286–288 (2000).