

PRESSURE DEPENDENCE OF THE RADIATION EFFICIENCY IN XeCl* MIXTURES PUMPED BY A LONGITUDINAL GLOW DISCHARGE

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Here we present some results of our experimental study of the influence of a low gas pressure on the XeCl radiation efficiency in Xe-Cl₂ mixtures pumped by a longitudinal glow discharge. When the pressure varied from 0.5 to 2 Torr the efficiency of XeCl radiation was shown to remain at the level of 10%. Maximum efficiency was obtained at a pumping power of $7 \cdot 10^{-18}$ W per molecule with the spectral bandwidth increasing with gas pressure decrease reaching 12 nm at the pressure of 1 Torr. Processes involved in XeCl molecule formation are discussed.*

1. In recent years much attention is paid to sources of powerful UV and VUV radiation¹⁻⁵ called the excilamps, which use the radiation of excimer and exciplex molecules that are formed in mixtures of rare gases with halogen pumped by different types of electric discharge. Gas pressure from 0.1 to several bars is conventionally used in excilamps since in this pressure range maximum output power or maximum excilamp efficiency could be obtained. High efficiency of spontaneous radiation (~10%) of XeCl* and KrCl* molecules was demonstrated in Xe(Kr)-Cl₂ gas mixtures under pressures between 10 and 20 Torr pumped by a dc glow discharge.^{2,3} In Refs. 4 and 5, efficiency of radiation of XeCl* and KrCl* molecules was found to be 10% and 14%, respectively, as gas pressure was decreased down to 5 Torr.

In this paper, efficiency of Xe-Cl₂ radiation in low pressure gas mixtures pumped by glow discharge was studied in pressure range from 0.5 to 2 Torr. Besides, XeCl* bandwidth was also investigated.

2. Experimental setup and measurement techniques were similar to those described in Refs. 4 and 5. Radiator of excilamp consists of two coaxial quartz tubes of high quality and two electrodes, fitted at their ends. The length of the discharge gap was 40 cm, the gap between the tubes was 3.5 mm and outer diameter of coaxial discharge gap was 50 mm. Inner quartz tube and cathode were cooled by water. The excilamp was connected to a power supply, providing smoothly adjustable initial voltage and discharge current in the ranges from 1 to 30 kV and from 1 to 50 mA, respectively. Voltage of the discharge initiation depended on pressure and composition of the gas mixture and typically did not exceed 10 kV. We have studied normal form of the glow discharge. In our experiments, voltage across the discharge gap was 2-4 kV, the discharge current varied from 5 to 50 mA corresponding to pumping power from 5 to 300 W. Optimal Xe:Cl₂ = 3:1 gas mixture had been determined

in our previous investigations (see Refs. 4 and 5) and was prepared in a separate gas container. Duration of one cycle of experiments was shorter than time of a substantial mixture degradation.

Light flux from a unit length of the excilamp was taken as a measure of total output power. The radiating area was assumed to be a point source of light since its diameter was less than the distance to photocathode by a factor of 10. FEK-22SPU photocathode was calibrated using radiation of XeCl* laser. We used filters to separate UV radiation. Radiation spectra of excilamp were recorded with a monochromator equipped with a FEU-100 photomultiplier and a grating with 1200 grooves/mm. The photomultiplier was connected to a current amplifier and a recorder.

3. The main experimental results are presented in Figs. 1 and 2. An increase of the efficiency of XeCl* radiation by about 30% as well as increase of its bandwidth by about 15% was observed with the gas pressure decrease from 6 to 1 Torr. Maximum efficiency was obtained in the pressure range from 1 to 2 Torr. Further decrease of the gas pressure down to 0.5 Torr resulted only in a slight decrease of the efficiency by about 10%. Lower gas pressure caused a significant drop of the output power and efficiency (see Fig. 1, curves 1 and 2). Examination of the influence of input power on the efficiency of low pressure excilamp indicates that in the pressure range from 0.5 to 2 Torr the maximum efficiency is achieved at a specific input power of about $0.3 \text{ W}/(\text{cm}^3 \cdot \text{Torr})$ or at a specific input power about $7 \cdot 10^{-18}$ W per one particle of the initial mixture.

High uniformity is a well-known peculiarity of a low-pressure discharge. Indeed, in our experiments at a pressure of 0.5 and 1 Torr we have observed quite homogeneous discharge, that occupied the whole discharge volume. Certain inhomogeneity occurred only in the cathode and anode regions.

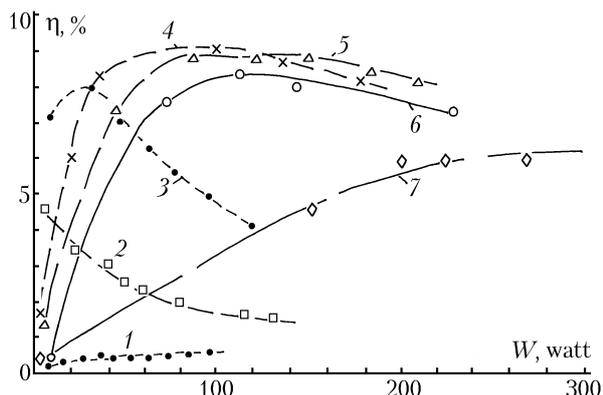


FIG. 1. Efficiency of XeCl* radiation versus pumping power, Xe:Cl₂ = 3:1 gas mixture at the total pressure < 0.25, 0.25, 0.5, 1, 2, 4, and 6 Torr.

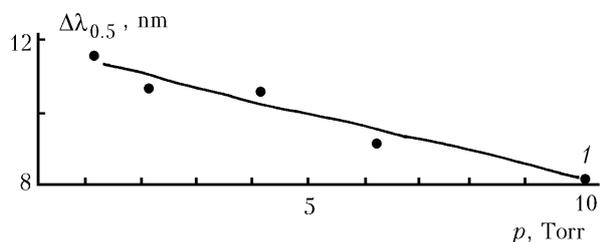


FIG. 2. Bandwidth (FWHM) of spontaneous radiation of XeCl* molecules versus total pressure of Xe:Cl₂ = 3:1. Point 1 was obtained in Xe:HCl = 3:1 gas mixture.

4. Let us analyze the main processes, which are responsible for XeCl* molecule formation. Consider that XeCl* molecules are formed in harpoon reaction or in the process of ion-to-ion recombination, their decay is caused by spontaneous emission and quenching processes in the discharge plasma. To simplify calculations, assume that effect of tube walls can be ignored. The excited xenon atoms Xe*, involved in the harpoon reaction are formed in collisions of xenon atoms with electrons (Penning ionization). Xenon ions are produced in the process of step ionization or by collisions of excited xenon atoms with each other. Negative chlorine ions Cl⁻ are formed in the process of dissociative attachment of electrons to chlorine molecules Cl₂ and are eliminated in the counter process. To simplify the model, photodetachment is ignored, since it plays insignificant role in the discharge kinetics. Since degree of ionization of the discharge plasma is not higher than 10⁻⁵–10⁻⁶, concentrations of the initial components of the gas mixture are considered to be constant. Thus, in the case of stationary discharge the kinetic model includes a set of five nonlinear algebraic equations:

$$\begin{aligned} d/dt [\text{Xe Cl}^*] = & - [\text{Xe Cl}^*] A_1 + [\text{Xe}^*] [\text{Cl}_2] k_1 + \\ & + [\text{Xe}^+] [\text{Cl}^-] k_2 - \sum_i [\text{Xe Cl}^*] N_i k_i = 0; \end{aligned} \quad (1)$$

$$\begin{aligned} d/dt [\text{Xe}^*] = & [\text{Xe}] n_e k_3 - [\text{Xe}^*] [\text{Cl}_2] k_1 - \\ & - [\text{Xe}^*] n_e k_4 - [\text{Xe}^*]^2 k_5 - [\text{Xe}^*] A_2 = 0; \end{aligned} \quad (2)$$

$$d/dt [\text{Xe}^+] = [\text{Xe}^*] n_e k_4 - [\text{Xe}^+] [\text{Cl}^-] k_2 + [\text{Xe}^*]^2 k_5; \quad (3)$$

$$d/dt [\text{Cl}^-] = [\text{Cl}_2] n_e k_6 - [\text{Xe}^+] [\text{Cl}^-] k_2 - [\text{Cl}^-] n_e k_7; \quad (4)$$

$$n_e + [\text{Cl}^-] = [\text{Xe}^+], \quad (5)$$

where $k_1, k_2, k_i, k_3, k_4, k_5, k_6, k_7$ are the rate constants of harpoon process, process of ion-to-ion recombination, process of quenching by various particles, process of excitation, process of Penning ionization, process of attachment, and process of detachment in collisions with electrons, respectively; A_1 and A_2 are the probabilities of spontaneous decay of XeCl* molecules and excited xenon atoms Xe*, respectively; N_i are concentrations of electrons, xenon atoms, and chlorine molecules. The last equation that is the condition of plasma quasineutrality can be omitted. Election concentration is assumed to be 10¹¹ cm⁻³ (Ref. 5).

The results of calculations performed within this model using rate constants presented in Refs. 6–11 allow us to come to the following conclusions.

a) Under conditions of our experiment XeCl* molecules are mainly formed in harpoon reactions. Contribution of ion-to-ion recombination does not exceed several percent. This fact allows further simplification of the model and leads to consideration of a set of only two equations. It should be noted that a significant contribution of harpoon reaction to formation of exciplex molecules in Cl₂-containing gas mixtures was reported in Refs. 2–5.

b) Electron energy that determines the rate of excitation of xenon atoms strongly affects the efficiency of XeCl* formation. Nevertheless, under conditions of our experiments, the ratio between the rates of two formation processes of exciplex molecules only slightly depends on the rate constant of excitation.

c) Only one process of quenching of XeCl* molecules in the discharge volume is to be considered significant. That is quenching by halogen molecules whose contribution is as low as 5–10%.

5. Thus, investigations of the efficiency of XeCl* radiation in low-pressure gas mixture pumped by a glow discharge were performed. Radiation efficiency was shown to remain as high as 10% within the pressure range from 0.5 to 2 Torr, whereas radiation bandwidth (FWHM) increased monotonously with the gas pressure decrease reaching 12 nm at a pressure of 1 Torr. In the pressure range from 0.5 to 2 Torr, the maximum efficiency was obtained at a specific input power about 7·10⁻¹⁸ W per one particle of the mixture. One can assume that efficiency of radiation of other rare-gas-monohalides is high in a low-pressure discharge. Low-pressure glow discharge can be used in excilamps due to its high uniformity in discharge tubes of various design and dimensions.

REFERENCES

1. A.M. Boichenko, V.F. Tarasenko, E.A. Fomin, and S.I. Yakovlenko, *Kvant. Elektron.* **20**, No. 1, 7–30 (1993).
2. A.P. Golovinskii, *Pis'ma Zh. Tekh. Fiz.* **18**, No. 8, 73–76 (1992).
3. A.P. Golovinskii and V.S. Kan, *Opt. Spektrosk.* **75**, No. 3, 604–609 (1993).
4. A.N. Panchenko, É.A. Sosnin, V.S. Skakun, V.F. Tarasenko, and M.I. Lomaev, *Pis'ma Zh. Tekh. Fiz.* **21**, No. 21, 47–51 (1995).
5. M.I. Lomaev, A.N. Panchenko, É.A. Sosnin, V.S. Skakun, and V.F. Tarasenko, *Atmos. Oceanic Opt.* **9**, No. 2, 125–129 (1996).
6. Yu.P. Raizer, *Physics of Gas Discharge* (Nauka, Moscow, 1992), 592 pp.
7. A.E. Kuklin and Yu.I. Khapov, “Rate constants of reactions involved in the formation and quenching of exciplex molecules,” Preprint No. 301, Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR, Novosibirsk (1986), 23 pp.
8. V.M. Baginskii, V.V. Vladimirov, P.M. Golovinskii, and A.I. Shchedrin, “Optimization and stability of discharge in excimer lasers with He/Xe/HCl gas mixture,” Preprint No. 2, Institute of Physics of the Academy of Sciences of the Ukraine, Kiev (1988), 37 pp.
9. B.D. Kulagin, “Kinetic processes in plasma of excimer lasers,” Author's Abstract of Cand. Phys. Math. Sci. Dissert., I.V. Kurchatov Institute of Atomic Energy, Moscow (1980), 11 pp.
10. A.M. Boichenko, V.I. Derzhiev, A.G. Zhidkov, A.V. Karelin, A.V. Koval', and O.V. Sereda, *Trudy IOFAN* **21**, 44–115 (1989).
11. B.M. Smirnov, *Negative Ions* (Atomizdat, Moscow, 1978), 174 pp.