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Barrier-discharge-excited coaxial excilamps with the enhanced pulse energy

A.N. Panchenko, V.F. Tarasenko

Abstract. The parameters of sealed off barrier excilamps are studied at high excitation powers. The total output pulse energy up to 25 mJ is achieved (the emitting area of a KrCl excilamp was up to 1500 cm², the output power was above 100 kW, and the efficiency achieved 10 %). It is shown that a volume discharge was formed in the coaxial excilamp when the energy supplied to the working mixture was increased and the pulse repetition rate was increased up to 50 Hz. The peak radiation intensity on the excilamp surface achieved ~ 100 W cm⁻². The optimal excitation energy of a barrier excilamp was found to be 0.1–0.2 mJ cm⁻³. The excilamp efficiency rapidly decreases with further increasing the input energy.

Keywords: coaxial UV excilamps, high-power spontaneous radiation, barrier-discharge excitation.

1. Introduction

Spontaneous radiation sources emitting in the UV and VUV spectral ranges (excilamps) are widely used at present in a variety of fields in science, technology, and medicine [1-4]. As a rule, barrier-discharge-excited coaxial excilamps are used in many applications. Such excilamps have a rather simple design and are reliable, their average output power achieving 100 W [5, 6]. High average output powers of excilamps of this type are obtained due to a high pulse repetition rate (up to 100 kHz), whereas their pulse energy and power are comparatively low and are insufficient for a number of applications. The efficiency of excilamps is several times higher compared to UV and VUV lasers, they emit a greater number of lines, have high average output powers, are sealed off, and have a long service life and simple and various designs. Lasers in turn are sources of narrowband directional radiation and provide high radiation powers and considerable pulse energies.

The radiation intensity of an excilamp can be increased by exciting it by a transverse discharge, which is used in pulsed lasers. The radiation intensity of planar excilamps

A.N. Panchenko, V.F. Tarasenko Institute of High Current Electronics, Siberian Branch, Russian Academy of Sciences, prosp. Akademicheskii 2/3, 634055 Tomsk, Russia; e-mail: alexei@loi.hcei.tsc.ru, VFT@loi.hcei.tsc.ru

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can achieve a few kW cm⁻² [7, 8]; however, their working mixture contacts with metal electrodes. This considerably restricts the operation time of one portion of the mixture and makes impossible the fabrication of sealed off lamps.

The pulsed parameters of coaxial excilamps excited by a barrier discharge are restricted by the energy E stored in the barrier capacitance C_{bar} $(E = C_{\text{bar}}U^2/2)$. The use of ceramics with a high permittivity as a barrier (which increases E) and the second metal electrode with a small radius of curvature provided radiation parameters close to these of planar excilamps (~ 1 kW cm⁻²) [9]. However, the working mixture of this excilamp containing halogen molecules also was in contact with the metal electrode, which restricted the operation time of one portion of the mixture. In addition, due to the high radiation extraction loss from the discharge plasma, the spontaneous radiation pulse energy of the excilamp did not exceed a few millijoules and its efficiency was 1 %. In [10], the parameters of barrier excilamps were studied upon pumping by high-voltage pulses at a pulse repetition rate of up to 12 kHz. However, the efficiency of these excilamps was low because the excitation conditions and gas mixtures were not optimal.

The aim of this paper is to fabricate coaxial sealed off excilamps with high pulsed power, efficiency, and radiation energy. The lamps were excited by a barrier discharge and their electrodes were isolated from the working mixture by two barriers.

2. Excilamp design and measurement method

Figure 1 shows the design of a barrier-discharge-excited excilamp. The barrier discharge was produced in a gap between two coaxial quartz tubes filled with a mixture of krypton or xenon with various halogens. The optimal pressure ratio of inert gas and halogen for all operation regimes of excilamps was $\sim 100:1$ for the total pressure of the mixture 150-200 Torr. The length of the excited region weas 10-90 cm. The diameter of the external tube was 40-60 mm and the distance between quartz barriers was 8-12 mm. For electrodes we used a metal grid and a polished aluminium foil. In this case, radiation could be both extracted from the excilamp and concentrated at its centre through the walls of the inner quartz tube. To increase the excitation power and energy supplied to the mixture, the voltage across excilamp electrodes was increased. This was achieved by using a pulsed generator based on a TGI-1-270/12 thyratron and a pulse transformer. This generator formed voltage pulses of duration 200-500 ns with the amplitude up to 60 kV. The



Figure 1. Scheme of a barrier excilamp with a pump generator: (*C*) storage capacitor; (*1*) thyratron; (*2*) pulse transformer; (I_d , U_{lamp}) output signals of a current shunt R_{sh} and a voltage divider $R_1 - R_2$, respectively.

capacitance C in the primary winding circuit of the pulse transformer was 8-15 nF and the charging voltage U_0 was varied from 5 to 20 kV.

The pulsed power and radiation energy were measured with a FEK-22 vacuum photodiode. Measurements were performed by the method described in [11]. The lamp was shielded with an opaque screen with a hole of diameter *d*. Behind the hole at a distance of $L \ge d$, a photodiode was located. In this case, the hole in the screen can be treated as a point source emitting uniformly within the solid angle 2π . The fraction of radiation from the hole detected with the photocathode is $k = \sin^2(\alpha/2)$, where $\alpha = \arctan(A/2L)$ and *A* is the photocathode diameter. The total spontaneous radiation power and energy of the active medium of the excilamp were calculated taking into account the photocathode sensitivity and the emitting area of the excilamp.

The energy supplied to the lamp was calculated by measuring the current in the secondary winding of the pulse transformer and voltage across the excilamp with a shunt and a voltage divider, respectively. Electric signals were recorded with a TDS-224 digital oscilloscope.

3. Experimental results and discussion

Figure 2 presents the typical oscillograms of voltage across the excilamp, the discharge current, and spontaneous radiation power at ~ 222 nm. After the voltage pulse is fed into the excilamp, barrier capacitances are charged for 50 ns. The breakdown of the discharge gap occurs when the voltage across the excilamp achieves 20 kV, and the first excitation and radiation pulses of duration 150 ns are formed. During this time, the barrier capacitances of the excilamp are further charged. Then, the current through the gap ceases and the radiation pulse ends. The repeated increase in the current and, hence, the spontaneous radiation pulse appear within 200 ns after the beginning of the discharge of barriers through the secondary winding of the pulse transformer.

A distinct feature of the discharge formed in the twobarrier excilamp at a high voltage across the gap is the discharge uniformity and filling of the entire space between electrodes by the discharge. It is known [5] that in conventional coaxial excilamps operating with pulse repetition rates up to a few hundreds of kilohertz a discharge has a different form. Under optimal excitation conditions, it consists of many conical diffuse microdischarges, each of them in turn consisting of two cones with connected tops at the gap centre, and the bases of microdischarges fill virtually the entire surface of quartz tubes between electrodes. The generation of doubled radiation pulses is caused by the presence of a flat top of the voltage pulse and is realised both at high voltages across the excilamp gap and in conventional regimes with voltages of a few kilovolts. The difference is manifested only in a change in the delay time between the beginning of excitation and the appearance of UV radiation. At high voltages across the gap and, hence, discharge currents of hundreds of amperes, the delay decreases to several nanoseconds.



Figure 2. Typical oscillograms of voltage on the excilamp and of current pulses in the excilamp circuit, and the time dependence of the total spontaneous radiation power at 222 nm in the usual regime (a) and upon shorting the secondary winding of a transformer by a discharger (b); the Kr: $Cl_2 = 150:1$ mixture at a pressure of 210 Torr, C = 14 nF, and $U_0 = 8$ kV.

The experiments showed that the excitation generator based on a pulse transformer allowed us to change rather simply the delay time between radiation peaks and the power of the second peak. The maximum amplitude U and duration t of pulses generated by a pulse transformer are related to the transformer core area S and the number W of coils by a simple expression [12]

$$Ut = \Delta BSW,\tag{1}$$

where ΔB is the magnetic induction drop in the transformer core. By increasing U or t, we can produce the saturation of the core at a certain instant. This results in a drastic decrease in the inductance in the excilamp circuit, while the power of the second excitation pulse drastically increases due to the increase in the discharge current. A similar result can be achieved by decreasing ΔB with the help of a preliminary magnetisation of the core by a direct current or by shorting excilamp electrodes by means of an additional discharger. Typical oscillograms for this operation regime of the excilamp are shown in Fig. 2b. The radiation peaks in this regime are overlapped. The power of the second peak increases approximately by thee times (up to 150 kW). It is interesting that the ratio Q_1/Q_2 of energies emitted during the first (Q_1) and second (Q_2) pulses is virtually independent of the excitation regime. For conditions of Fig. 2, the total spontaneous radiation energy was 12.5 mJ and $Q_1/Q_2 = 1/1.6$. The efficiency with respect to the input energy was 7 % for the specific input energy of 0.25 mJ cm^{-3} . Because the energy supplied to the medium in each peak is approximately the same and is determined by the voltage to which the barrier capacitance can be charged, the efficiency of the electric energy conversion to spontaneous radiation in the second peak increases. This is related to the energy input to produce conductivity in the discharge gap of the excilamp by ionising inert gas atoms. Exciplex molecules are produced in mixtures with molecular chlorine pumped by a barrier discharge mainly in the harpoon reaction of metastables of inert gas (Kr* or Xe*) with Cl_2 molecules [7]:

$$\operatorname{Kr}^{*}(\operatorname{Xe}^{*}) + \operatorname{Cl}_{2} \to \operatorname{Kr}\operatorname{Cl}^{*}(\operatorname{XeCl}^{*}) + \operatorname{Cl}.$$
 (2)

The contribution of the recombination of the inert gas ions $Kr^{+}\ (Xe^{+})$ and Cl^{-}

$$\operatorname{Kr}^{+}(\operatorname{Xe}^{+}) + \operatorname{Cl}^{-} \to \operatorname{Kr}\operatorname{Cl}^{*}(\operatorname{Xe}\operatorname{Cl}^{*})$$
 (3)

to the population of the B state of exciplex molecules is small. Therefore, the energy spent for ionisation during the discharge formation is lost. Before the second current pulse, the gap strength has no time to recover completely, and the ionisation energy loss decreases.

The replacement of chlorine by hydrogen chloride in the excilamp mixture resulted in the decease in the spontaneous radiation energy and power by approximately five times under the same conditions of discharge formation. This is explained by the low efficiency of harpoon reaction (2) in mixtures with HCl [7]. As the pump power is increased, the degree of ionisation of the working mixture and the fraction of exciplex molecules produced due to recombination (3) increase. However, studies performed in [10] showed that the efficiency of mixtures with HCl pumped by the barrier discharge remains low up to the input energy 8 mJ cm⁻³.

The optimisation of the design of the pulse transformer improved the working parameters of the excilamp (Fig. 3). The radiation peaks of the excilamp in this regime have the same power 80 kW and the total output energy achieves 15 J for efficiency ~ 10 %. As the excilamp length was increased up to 90 cm, the radiation energy increased up to 25 mJ, the efficiency remaining the same. The maximum pulse repetition rate of coaxial excilamps in these experiments was 50 Hz, while the maximum average radiation power of a sealed off excilamp with the active length of 90 cm exceeded 1 W.



Figure 3. Time dependence of the total radiation power of the excilamp at 222 nm in the optimal excitation regime; the Kr: $Cl_2 = 100:1$ mixture at a pressure of 180 Torr, C = 10 nF, $U_0 = 8$ kV.

Note that the efficiency of conventional barrier excilamps for the maximum average radiation power was also ~ 10 %. However, as mentioned above, the discharge shape in this case was substantially different. At high pulse repetition rates, the discharge gap is filled with many conical microdischarges moving chaotically against the background of a homogeneous low-current discharge [2, 5, 6]. In this case, the excitation power is distributed in the gap nonuniformly and the maximum power is achieved near tops of the cones. The question arises of how the excilamp efficiency will change with increasing excitation power in a volume discharge without microdischarges and filaments. To increase the excitation power, the active length of the excilamp was reduced to 10-12 cm and the diameter of quartz tubes was reduced to 43 and 23 mm. Figure 4 presents the dependences of the efficiency and radiation energy of the XeCl excilamp with the active volume 100 cm³ on the specific energy input. One can see that the radiation efficiency of exciplex molecules drastically decreases with increasing input energy. Similar dependences were also observed for XeI, KrBr, and Cl₂ excilamps. Thus, the optimal powers of excitation by high-voltage pulses for obtaining the maximum efficiency and maximum energy are substantially different. Note that the maximum radiation intensity on the excilamp surface in these experiments achieved 100 W cm⁻².

To explain the decrease in the radiation efficiency with increasing excitation power, we consider basic process proceeding in the active medium of the excilamp. As mentioned above, exciplex molecules in mixtures with chlorine are formed in harpoon reaction (2). The rate of this reaction is



Figure 4. Dependences of the efficiency and the total radiation energy of the XeCl excilamp with the active volume of 100 cm^3 on the specific energy input.

$$\frac{\mathrm{d}[\mathrm{XeCl}^*]}{\mathrm{d}t} = k_{\mathrm{gar}}[\mathrm{Xe}^*][\mathrm{Cl}_2],\tag{4}$$

where k_{har} is the rate constant of the harpoon reaction and [Xe^{*}] and [Cl₂] are the concentrations of particles involved in the reaction. However, as the exaction power is increased, the discharge current and electron concentration increase, resulting in the increase in the step ionisation rate of Xe^{*}

$$\frac{\mathrm{d}[\mathrm{Xe}^+]}{\mathrm{d}t} = k_{\mathrm{sti}}[\mathrm{Xe}^*]n_\mathrm{e} \tag{5}$$

(k_{sti} is the rate constant of step ionisation and n_e is the electron concentration) and in the quenching rate of XeCl^{*} molecules by electrons

$$\frac{\mathrm{d}[\mathrm{XeCl}^*]}{\mathrm{d}t} = k_{\mathrm{d}}[\mathrm{XeCl}^*]n_{\mathrm{e}}$$
(6)

 $(k_{\rm d}$ is the quenching rate constant). Therefore, the loss of Xe* due to step ionisation and the loss of working XeCl* molecules will increase with increasing the discharge current due to their quenching by electrons, and the excilamp efficiency will decrease. A similar mechanism of the decrease in the formation efficiency of exciplex molecules in a filament of a barrier discharge with increasing the degree of gas ionisation was found in [13] upon simulation of the operation of barrier excilamps. For the typical conditions in a barrier discharge [14–16], the rates of reactions (4)–(6) will be equal for $n_e \sim 10^{13} - 10^{14} \text{ cm}^{-3}$. The estimate of the electron concentration in a volume barrier discharge for the drift velocity $V_{\rm dr} \sim 10^6 {\rm ~cm~s^{-1}}$ of electrons in pure xenon [17] gives $n_{\rm e} > 10^{13}$ cm⁻³ already for the specific pump energy $\sim 0.5 \text{ mJ cm}^{-3}$, when the excilamp efficiency rapidly decreases (Fig. 4). We can also make conclusion from this about the operation of barrier excilamps in regimes with high pulse repetition rates. Working molecules in such excilamps are produced most efficiently near quartz walls at the base of conical microdischarges where the excitation power is optimal for obtaining high efficiencies.

4. Conclusions

We have studied the emission of exciplex KrCl and XeCl molecules at high excitation powers. Pulsed excilamps with the radiation energy up to 25 mJ, the peak radiation power up to 150 kW, and the efficiency up to 10% have been fabricated. It has been shown that radiation pulse consists of two peaks. The delay time between the peaks and the amplitude of each of them can be varied in a broad range by retaining the total radiation energy. The peak UV radiation intensity on the excilamp surface achieved $\sim 100 \text{ W cm}^{-2}$.

The optimal specific excitation energy for the barrier excilamp has been found to be $0.1-0.2 \text{ mJ cm}^{-3}$. Further increase in the input energy results in a rapid decrease in the excilamp efficiency.

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