

## Optical Emission of Atmospheric - Pressure Dielectric Barrier Discharge Plasma on Mercury Diiodide/Rare Gases Mixtures

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**Abstract.** New results of research of spectral and electrical characteristics of the plasma high-frequency atmospheric - pressure dielectric barrier discharge (DBD) at mixtures of mercury diiodide vapor with xenon and neon are presented. Gas-discharge plasma creation and excitation of the working mixture components was carried out by sinusoidal form voltage at a repetition rate 125 kHz. Simultaneous emission of mercury monoiodide exciplex molecules ( $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ ,  $C^2\Pi_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ ), xenon iodide ( $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ ,  $D_{1/2} \rightarrow A^2\Pi_{1/2}$ ) and iodine ( $D' \rightarrow A'$ ) have been revealed. On the basis of comparing the experimental data of the behavior of the spectra B-X and C-X transitions of molecule HgI and depending on the component composition the mechanism of emission brightness enhancement of exciplex molecule HgI in mixtures with addition of xenon have been defined and conclusions about the mechanism of exciplex molecules HgI, XeI and I<sub>2</sub> formation in the discharge have been made. High-frequency atmospheric - pressure barrier discharge on mercury diiodide vapor with xenon and neon mixture is of interest for use in excilamp that emits simultaneously spectral bands in the violet - blue and ultraviolet spectral ranges.

**Keywords:** dielectric barrier discharge, high-frequency, radiation, atmospheric - pressure plasma, mercury diiodide vapor, xenon, neon.

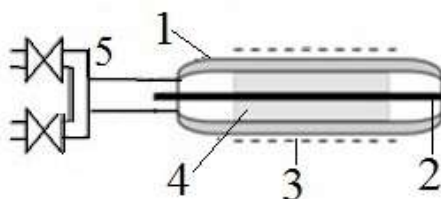
### I. Introduction

Creation of new optical radiation sources, including excimer (or exciplex), that are important in modern science and technology requires the studies of optical emission and parameters of the working media of such sources [1-3]. Plasma in the mixtures of mercury diiodide vapor with atomic and molecular gases is an efficient exciplex source of coherent and spontaneous emission in the violet-blue range of the spectrum with a maximum emission at wavelengths  $\lambda_{\max.} = 441.4, 443, 444, 445$  nm [4-12]. Exciplex radiators (lasers and excilamps) that work on such mixtures provide high spectral radiation power that is important for practical applications in plant photoregulation, biotechnology and medicine. [13-14]. Optical emission of gaseous mixtures, including mercury diiodide vapor, inert gases, molecular nitrogen was studied in [4-11]. Atmospheric pressure plasma in the working mixtures was produced in a glow and barrier discharges under repetitively pulsed or sinusoidal voltage form. High radiation efficiency demonstrated recently for gas mixtures containing mercury diiodide vapor, helium and xenon in the studies in which the experiments were carried out under pulsed pumping (pulse duration  $\leq 150$  nanoseconds) at a frequency 1-20 kHz [10 - 12]. Creation more powerful emitters (excilamps) in this spectral range requires (makes it necessary) carrying out diagnostics of optical emission of atmospheric pressure plasma on mercury diiodide vapor, neon and xenon mixtures at high pumping frequencies (over 20 kHz) of working mixtures, which was the purpose of our research.

### II. Experimental Setup

The experimental setup for the study of atmospheric pressure dielectric barrier discharge (DBD) plasma, initiated by the sinusoidal form of pump pulses in mixtures of mercury diiodide vapor with inert gases neon and xenon was close to that used by us for the investigation of atmospheric-pressure dielectric barrier discharge plasma with mixtures of mercury dibromide vapor with inert gases [15].

For the experiments a small cylindrical radiator (figure 1) with one dielectric barrier (1) with a capacity  $C_d = 40$  pF, made of a quartz tube was used. The molybdenum electrode (2) with a diameter of 1 mm was located

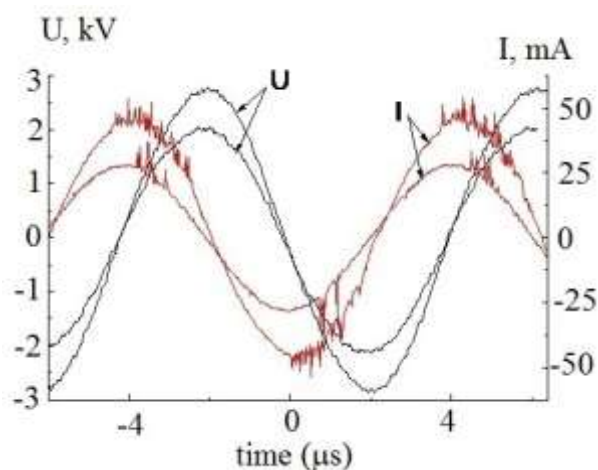


**Figure 1.** Radiator: 1- quartz tube, 2-internal electrode, 3 - external electrode (mesh), 4 –discharge gap, 5- pumping system and a gas inlet.

axially inside. The outer diameter of the radiator (tube) was equal to 6 mm, wall thickness - 1mm. The discharge gap (4) was 1.5 mm. The outer electrode of 30 mm in length (3) is made of mesh with radiation transmittance  $T = 70\%$ . At the end of the quartz tube was located the capillary (5) with a diameter of 1.5 mm, which served to pump the radiator, filling it with working mixtures, as well as to reduce the removal of mercury diiodide vapors from radiator in the pumping system. The dielectric barrier discharge was excited at a total pressure of a mixture of 110.6 - 110.7 kPa. The high-voltage generator allowed changing the amplitude and frequency of the applied voltage to the electrodes of sinusoidal form up to 3 kV and 125 kHz, respectively. Discharge emission was analyzed in the spectral range 200 - 900 nm. The spectral resolution of the registration system was 0.05 nm. Working mixtures were prepared directly in the radiation source. Mercury diiodide ( $HgI_2$ ) in an amount of 60 mg pre-loaded into the radiator. After loading the salt dehydration and outgassing of the internal surface of the source by heating at a temperature of 70 - 100<sup>o</sup>C and pumping during 2 hours was carried out. The partial pressure of gases was measured with an accuracy of 10 Pa. The value of the partial pressure of the vapor  $HgI_2$  was determined by the temperature at the coldest point of the radiator based on the interpolation reference data of [16] and in our conditions was equal to 600 - 700 Pa.

### III. Results And Discussion

Spectral and electrical characteristics of the plasma high-frequency barrier discharge were studied in mixtures of the following compositions:  $HgI_2$ : Ne and  $HgI_2$ :Xe: Ne. In the initial stage (first 20 – 30 s) the discharge color was dependent on the component composition of the mixture. In a binary mixture with neon red color prevailed, while xenon supplements appeared blue. As the self-heating of the mixture discharge color becomes bright blue. After the initiation of the DBD many avalanche-streamer mechanism of discharge burning was observed, typical for frequency ~ 100 kHz - a set of conical micro-discharges with vertex at the electrode and the base on the inner surface of the quartz tube [17]. With increasing temperature, the discharge becomes more diffuse and uniform, the contrast brightness smoothed noticeably in volume discharge and filaments. Figure 2 shows a typical waveform of current and voltage. In each half-cycle of the applied voltage on the waveform of the recorded current on the current bias curve observed series of sharp bursts of different amplitude and approximately the same duration. Bursts on current waveform reflect the value of the conduction current in the discharge. Each current burst is due to the number of threadlike micro discharges-filaments that occur in the discharge gap and are randomly distributed in time. For a mixture  $HgI_2$ : Ne the peaks amplitude has not exceeded 10 mA when the duration was 50-80 ns. The first burst of current with increasing external voltage amplitude (U) all moved to the left of the applied voltage maximum and at large amplitudes U first burst already fitted to the negative phase of the applied voltage. Current bursts of positive polarity always fitted to voltage phase with positive derivative  $dU/dt > 0$ . The number of current bursts increased while the voltage raised. The end of the last



**Figure 2.** Oscillograms of voltage and current pulses of a barrier discharge in the mixture  $HgI_2/Ne = 0.6/110$  kPa at  $f = 125$  kHz for different values of the voltage applied.

pulse coincided with the maximum of applied voltage, i.e., the discharge continued as long as the instantaneous voltage value on the electrode system did not reach U.

At the same value U at the DBD on  $HgI_2$ :Xe: Ne mixtures, breakdown occurred at 150-200 ns earlier as on the  $HgI_2$ : Ne mixture. With the growth of voltage U, the active component gives an increasing contribution to the total current. The maximum value of the conduction current in the DBD at  $HgI_2$ :Xe: Ne mixture at an operating temperature 170<sup>o</sup>C was equal to 15 mA.

Survey DBD plasma emission spectra in the HgI<sub>2</sub>: Ne and HgI<sub>2</sub>:Xe: Ne mixtures at f = 125 kHz are shown in figure 3 and figure 4. As can be seen in the spectra in the visible range dominates the system of bands with a peak at λ = 443 nm, which has weakly allowed vibrational structure and corresponds to the electron-vibrational transition B<sup>2</sup>Σ<sup>+</sup><sub>1/2</sub> → X<sup>2</sup>Σ<sup>+</sup><sub>1/2</sub> of mercury monoiodide molecules [18].

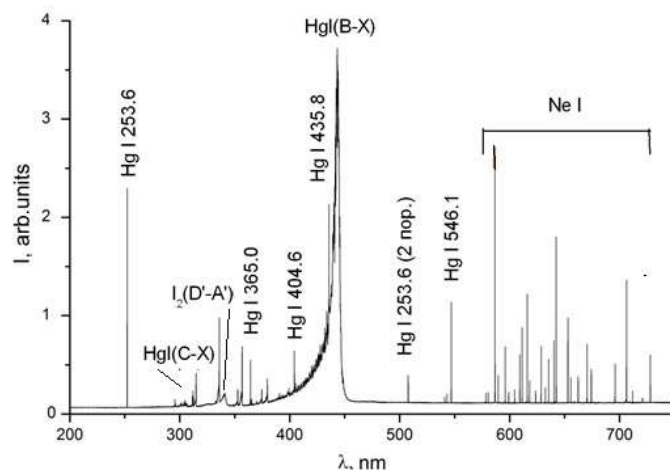


Figure 3. Survey emission spectrum of barrier discharge plasma based on the mixtures HgI<sub>2</sub>/Ne = 0.6/110 kPa.

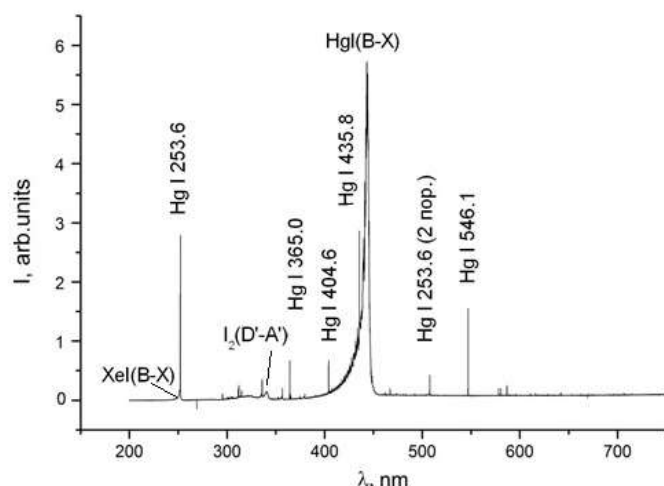


Figure 4. Survey emission spectrum of barrier discharge plasma based on the mixtures HgI<sub>2</sub>/Xe/Ne = 0.7/10/100 kPa.

For this band system characteristic is a sharp increase in intensity from the long-wavelength region and a slow decrease in the short-wavelength region. In terms of atmospheric pressure barrier discharge edges of the spectral bands overlap the range of wavelengths 350-450 nm. In the mixture HgI<sub>2</sub>: Ne = 0.7:100 kPa (figure 3), in addition to these spectral bands in the range 580 - 740 nm observed sufficiently intense lines of neon atoms. In all spectra were registered the radiation of HgI molecules (C → X) with a maximum at λ = 306 nm, the radiation of decay products of mercury diiodide – the band of molecular iodine I<sub>2</sub> (D' → A') with a maximum at λ = 342 nm and lines of atomic mercury in UV and visible range, the most intensive of which are - 253.6 (6<sup>3</sup>P<sub>1</sub> → 6<sup>1</sup>S<sub>0</sub>), 365.0 (6<sup>3</sup>D<sub>3</sub> → 6<sup>3</sup>P<sub>2</sub>), 404.6 (7<sup>3</sup>S<sub>1</sub> → 6<sup>3</sup>P<sub>0</sub>), 435.8 (7<sup>3</sup>S<sub>1</sub> → 6<sup>3</sup>P<sub>1</sub>) and 546.1 nm (7<sup>3</sup>S<sub>1</sub> → 6<sup>3</sup>P<sub>2</sub>) [18, 19]. The addition to the mixture HgI<sub>2</sub>: Ne of xenon led to a significant change in the plasma emission spectrum DBD. For this mixture (HgI<sub>2</sub>: Xe: He = 0.7: 10: 100 kPa) is characteristic that in the emission spectrum, in addition to systems of spectral bands HgI (B → X) and HgI (C → X), are present Xe atoms lines at λ = 462.4 nm (7p[3/2]<sub>2</sub> → 6s[3/2]<sub>2</sub><sup>0</sup>), λ = 467.1 nm (7p[5/2]<sub>3</sub> → 6s[3/2]<sub>2</sub><sup>0</sup>), λ = 823.1 nm (6p[3/2]<sub>2</sub> → 6s[3/2]<sub>2</sub><sup>0</sup>) and λ = 828 nm (6p[1/2]<sub>0</sub> → 6s[3/2]<sub>1</sub><sup>0</sup>) [19, 20]. Also the emission bands of exciplex molecules XeI(B → X) and XeI (D → A) with maxima at 253 and 320 nm were observed, respectively (figure 4). Compared

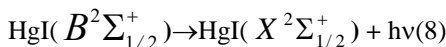
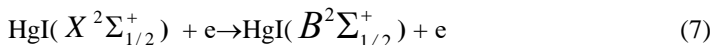
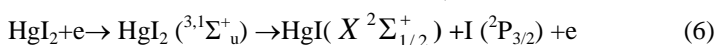
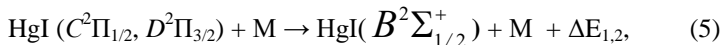
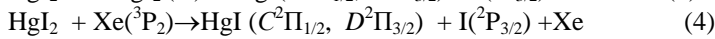
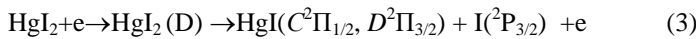
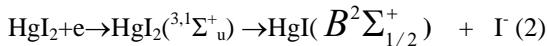
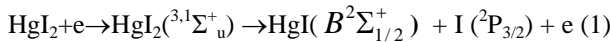
with a mixture HgI<sub>2</sub>: Ne, Ne atomic line intensity at 580 - 750 nm decreased to about 1-2 times, while the emission brightness HgI(B→X) increased by 1.6 times, and the brightness HgI(C→X) decreased by 1.8 times. Under molecular band brightness understood spectrum in the area under the curve.

The most intense spectral bands and plasma emission lines on a mixture of mercury diiodide vapor with neon and xenon for the pump pulse repetition frequency 125 kHz, the relative brightness and intensity based on the spectral sensitivity of the registration system (J/k<sub>λ</sub>), and also the excitation energy is given in table. Data on the excitation energies were taken from works [18-23].

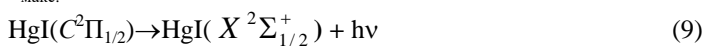
**Table.** Bands and lines of barrier discharge plasma radiation in mixtures of mercury diiodide vapor with neon and xenon

λ, nm	Molecule, atom	J/k <sub>λ</sub> , arb. units		E <sub>e</sub> , eV
		HgI <sub>2</sub> /Ne= 0.6/110 kPa	HgI <sub>2</sub> /Xe/Ne= 0.7/10/100 kPa	
253	XeI(B→X)	-	6.55	4.85 [21]
253.6	HgI	1.3	2.02	4.88 [20]
306	HgI(C→X)	10.0	4.78	8 [18]
320	XeI(B→A)	-	2.05	4.85 [21]
342	I <sub>2</sub>	17.34	22.93	5 [21]
365.0	Hg I	0.55	0.93	8.86 [20]
404.6	Hg I	0.50	0.88	7.73 [19]
435.8	Hg I	0.58	0.97	7.73 [19]
443	HgI(B→X)	88	141	7 [18, 22]
462.4	Xe I	-	0.2	11.00 [20,23]
467.1	Xe I	-	0.21	10.97 [20,23]
546.1	Hg I	0.58	1.42	7.73 [20]
585.2	Ne I	0.88	0.08	18.97 [20,23 ]
640.2	Ne I	0.63	0.02	18.55 [20,23]
703.2	Ne I	0.57	0.02	18.38 [20,23]
823.1	Xe I	-	1.2	9.82 [20,23]
828	Xe I	-	0.5	9.94 [20,23]

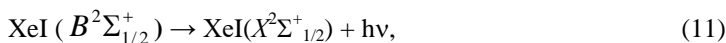
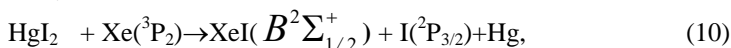
The emission of observed spectral transitions may be initiated by the following reactions [24-31]:



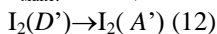
$$\lambda_{\text{max}} = 443 \text{ nm}$$



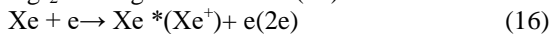
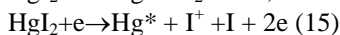
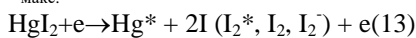
$$\lambda_{\text{max}} = 306 \text{ nm}$$

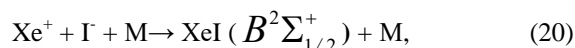
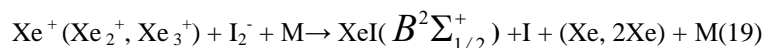
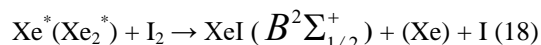


$$\lambda_{\text{max}} = 253, 320 \text{ nm}$$



$$\lambda_{\text{max}} = 342 \text{ nm}$$





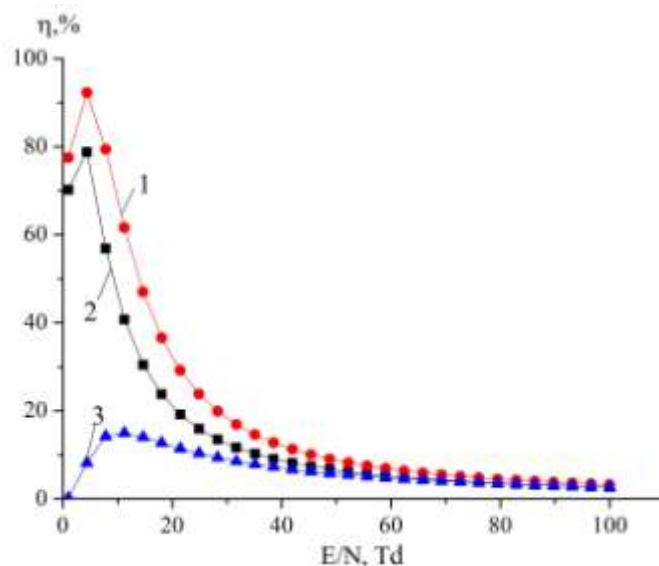
where M – concentrations of quenching molecules or atoms (HgI<sub>2</sub>, Xe, Ne), ΔE<sub>1,2</sub> - the energy difference between the states C<sup>2</sup>Π<sub>1/2</sub>, D<sup>2</sup>Π<sub>3/2</sub> and B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup> molecules HgI.

Reactions (1) and (2) are known as major sources of HgI\* molecules, effective cross section for which is equal the value (1.2 - 3.0) × 10<sup>-17</sup> cm<sup>2</sup> [24-25]. In addition, mercury monoiodide can be formed in the reaction (3) as a result of mercury diiodide molecules excitation in the state D under collisions with electrons (D - the state is the sum of a number of states, which are located between 5.7 eV and 9.5 eV (ionization threshold HgI<sub>2</sub>) [26 - 28]. Emission from the D - state HgI<sub>2</sub> molecules is not observed, as this state quickly dissociates with formation of electronically excited molecules HgI\* to C<sup>2</sup>Π<sub>1/2</sub>, D<sup>2</sup>Π<sub>3/2</sub> - states [27]. These states are excited also by mercury diiodide molecules collisions with xenon atoms in the metastable state (<sup>3</sup>P<sub>2</sub>) in the reaction (4). In the reaction (5) they relax with high efficiency in mercury monoiodide state B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup> [27].

Reaction (6) is another dissociation channel of mercury diiodide molecules compared to reaction (1), wherein a rate constant is equal to 2.5 × 10<sup>-11</sup> cm<sup>3</sup>s<sup>-1</sup> [29]. When electrons collide with mercury monoiodide molecules in ground state there B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup> state excites (reaction 7), rate constant of which is equal to 2.7 × 10<sup>-7</sup> cm<sup>3</sup>s<sup>-1</sup> [30]. Electron-vibrational transitions B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup> → (X<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup>) and (C<sup>2</sup>Π<sub>1/2</sub>) → (X<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup>) of mercury monoiodide molecules lead to the emission of spectral bands (τ<sub>r</sub> = 27.3 × 10<sup>-9</sup> s) with a maximum intensity at a wavelength λ<sub>max.</sub> = 443 nm (reaction (8)) and λ<sub>max.</sub> = 306 nm (reaction (9)), respectively [18,31]. The reaction of the collision of xenon atoms in the metastable <sup>3</sup>P<sub>2</sub> with the molecules of mercury diiodide (10) is possible, quite effective channel of the formation of exciplex molecules xenon iodide. Electron-vibrational transition B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup> → (X<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup>, A<sup>2</sup>Π<sub>1/2</sub>) of xenon iodide molecules leads to the emission of spectral bands with a maximum intensity at a wavelength λ<sub>max.</sub> = 253 nm and λ<sub>max.</sub> = 320 nm (reaction (11)) [14,23]. The emission of spectral bands with a maximum intensity at a wavelength λ<sub>max.</sub> = 342 nm is caused by electron-vibrational transitions D' → A' of iodine molecules (reaction 12), which are formed in the reaction (13) [14]. The excited mercury atoms are formed due to the reaction (13,14,15) due to the large effective cross section of dissociative excitation of mercury diiodide molecules by electrons. The emission of spectral lines of atoms and ions of xenon and neon caused by reactions (16, 17).

In addition to the reactions (10), the reactions (18-20) can be an effective source of exciplexes XeI (B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup>) formation. Reaction (18) is a harpoon reaction between Xe\* (Xe<sub>2</sub>) and (I<sub>2</sub>) molecules, while (19-20) are ion - ion recombination [14, 23].

A possible reason for increase of emission brightness in HgI (B → X) can be the difference in the plasma parameters in a mixture of mercury diiodide vapor, xenon and neon, in comparison with the mixture without xenon. For their study, we used a technique which is described in detail in [15, 32, 33]. As a result, an electronic concentration, rate constants of dissociative excitation of electrons of the molecules of mercury monoiodide (in these mixtures) and the distribution of the specific discharge power losses on these processes. The value of the electron density 2.4 · 10<sup>16</sup> m<sup>-3</sup> and 2.8 · 10<sup>16</sup> m<sup>-3</sup>, the mean electron energy of 9.8 eV and 6.0 eV, the rate constant of the B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup> state excitation is equal to 2.2 · 10<sup>-14</sup> m<sup>3</sup>/s and 1.2 · 10<sup>-14</sup> m<sup>3</sup>/s for the experimental value of reduced electric field in plasma (E / N) = 46 Td and 49 Td for HgI<sub>2</sub>- Ne and HgI<sub>2</sub>- Xe - Ne gas mixtures, respectively. The distribution of the specific discharge power losses for the processes of dissociative excitation of mercury monoiodide molecules increase with increasing of E/N. For the HgI(B<sup>2</sup>Σ<sub>1/2</sub><sup>+</sup>) electronic states the losses reach their maximum values of 92 % and 79 % at E/N = 4.4 Td for HgI<sub>2</sub>- Ne and HgI<sub>2</sub>- Xe - Ne gas mixtures, respectively, and then as E/N increases further, the power losses decrease (figure 5). Specific losses of discharge power for process of excitation of the metastable state <sup>3</sup>P<sub>2</sub> of xenon atoms (figure 5, curve 3) have a similar dependence on the reduced electric field strength, reach a maximum 15%, for the E/N = 11.2 Td. For the experimental value of the parameter E/N = 46 Td and 49 Td, they are equal to the 10% and 7% respectively for HgI<sub>2</sub>- Ne and HgI<sub>2</sub>- Xe - Ne gas mixtures, respectively.



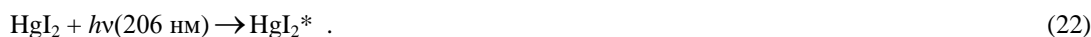
**Figure 5.** Specific discharge power losses for collisional interactions of electrons with mercury diiodide molecules and xenon atoms in the  $\text{HgI}_2\text{-Ne} = 0.54 - 99.46\%$  mixture as function of  $E/N$  (1) and in the  $\text{HgI}_2\text{-Xe-Ne} 0.64 - 9.03 - 90.33\%$  mixture as function of  $E/N$  (2) - dissociative excitation of the electronic states of mercury diiodide with the production of mercury monoiodide in the  $B^2\Sigma^+_{1/2}$  state, (3) - excitation of the metastable state  $^3P_2$  of xenon atoms.

Thus, these plasma parameters differ slightly depending on the composition of the mixture ( $\text{HgI}_2\text{-Ne}$  or  $\text{HgI}_2\text{-Xe-Ne}$ ) and therefore it is necessary to involve other processes that explain the significant increase in brightness of  $\text{HgI}$  radiation ( $B \rightarrow X$ ) in a mixture of mercury diiodide vapor, xenon and neon (figure 4, table) in comparison with the mixture without xenon (figure 3, table). Such processes can be energy transfer to mercury diiodide molecules in collisions with xenon atoms in a metastable state ( $^3P_2$ ) and the process of quenching of  $C^2\Pi_{1/2}$ ,  $D^2\Pi_{3/2}$ -states of mercury monoiodide molecules by xenon atoms with a nonradiative transition to the  $B^2\Sigma^+_{1/2}$ -state (4,5). Process (5) has been found in experiments on photodissociation of mercury diiodide [27, 34], as well as in our experiments on  $\text{HgI}^*$  molecules excitation in terms of DBD on the mixture  $\text{HgI}_2\text{-Xe-He}$  at the frequency of the pump pulse of the mixture up to 20 kHz [12]. The obvious argument in favor of this is the decrease in brightness of  $\text{HgI}(C \rightarrow X)$  bands system of radiation in mercury diiodide vapor, xenon and neon mixture in comparison with mercury diiodide vapor and neon mixture (table).

The sharp decrease in the intensity of the spectral lines of neon atoms in 580-750 nm range in this mixture can be explained by the process of Penning ionization of xenon [35]:



In these experiments in the emission spectra was not manifested resonance line of the iodine atom at  $\lambda = 206$  nm, due to absorption of radiation by molecules of mercury diiodide in the process [36]:



Thus, the numerical simulation results give the reason to conclude about the possibility to increase the radiation power in the violet-blue spectral range of the multi-wavelength DBD-driven exciplex lamp based on a mixture of the mercury diiodide vapor, xenon and neon by means of the reduction of the parameter  $E/N$  to the value of 4.4 Td. The simplest way to achieve such values is to change the design of the lamp, namely the combination of pulse of high electric field on the front of the discharge and the low field behind the front.

#### IV. Conclusion

The spectral and electrical characteristics of high-frequency (125kHz) atmospheric-pressure dielectric barrier discharge in mixtures of mercury diiodide vapor with neon and additives of xenon have been studied. Intense quasi-continuous emission with a maximum at  $\lambda = 443$  nm of mercury monoiodide exciplex molecules, the main part of which is concentrated in the region 439-446 nm have been revealed. The optimum to obtain maximum emission of  $\text{HgI}$  molecules ( $B \rightarrow X$ ) is a mixture  $\text{HgI}_2\text{:Xe: Ne}$ . On  $\text{HgI}(B \rightarrow X)$  bands system accounts for about 77 % of the radiation of gas-discharge plasma of dielectric barrier discharge in the region of

400 - 900 nm. Emission brightness of HgI (B → X) in dielectric barrier gas-discharge plasma in a mixture with the addition of xenon was increased in 1.6 times in comparison with a mixture HgI<sub>2</sub>: Ne due to the presence of quenching processes of C<sup>2</sup>Π<sub>1/2</sub> and D<sup>2</sup>Π<sub>3/2</sub> states of mercury moniodide molecules by Xe atoms and atomic collisions of xenon in <sup>3</sup>P<sub>2</sub> metastable state with the molecules of mercury diiodide, that increases its population in B<sup>2</sup>Σ<sup>+</sup><sub>1/2</sub> state. The analysis of the distribution of energy in the internal degrees of freedom of the gas and the reaction rates of collisions with electrons for different electric field concluded that the combination of a high electric field pulse at the front of the discharge and the low field behind the front can be used to increase the emission intensity in mercury diiodide vapor with neon and xenon additives mixtures. Atmospheric - pressure dielectric barrier discharge on multicomponent mixtures of mercury diiodide vapor with gases can be used to create self-heating gas discharge excilamp which emits spectral bands in the violet - blue and ultraviolet spectral ranges. The emission of this spectral band can be effectively used to activate the process of photosynthesis with the simultaneous destruction of viruses and bacteria.

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