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SPECTRAL CHARACTERISTICS OF THE BARRIER DISCHARGE ON MERCURY DIIODIDE VAPOR WITH NEON MIXTURE

Abstract

8 **Aims:** To investigate spectral characteristics of a high-frequency barrier discharge plasma of 9 atmospheric pressure on mercury diiodide vapor with neon mixture.

Study design: A small cylindrical radiator with a single dielectric barrier of capacitance $C_d = 40 \text{ pF}$ made of a quartz tube was used for the experiments. Inside along the axis there was a molybdenum electrode with a diameter 1 mm, an external electrode 30 mm in length was made of a mesh with a transmittance 70%.

Place and Duration of Study: Department of Quantum electronics, Faculty of Physics, Uzhhorod National University, between June 2017 and September 2017.

Methodology: The high-voltage generator with an average power of up to 100 W was used to apply sinusoidal voltage (up to 7 kV and 130 kHz) to the electrodes of the radiator. The radiation spectra were recorded using a Jobin Yvon TRIAX 550 spectrometer (1200 lines / mm and 3600 lines / mm, a slit 0.02 mm, a quartz optical fiber and a Spectrum ONE CCD detector cooled with liquid nitrogen.

Results: Intense quasicontinuous emission of spectral bands with a maximum at λ = 443 nm and 306 nm, as well as 342 nm of exciplex molecules mercury monoiodide and iodide molecules have been revealed. The main part of the emission of spectral bands is concentrated in violet-blue spectral region, namely, 439-446 nm, the relative brightness of which is higher than the brightness of other spectral bands and lines.

Conclusion: The gas discharge excilamp at high pumping frequencies on mercury diiodide with neon mixture can be created with operating life higher than the excilamp, where the buffer gas is used helium.

28 Key words: high-frequency barrier discharge, atmospheric pressure, radiation, spectral bands,

29 mixture components, mercury diiodide, neon.

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31 **1.INTRODUCTION.**

32 The creation of new sources of optical radiation, which are important in modern technologies 33 and scientific research, requires studies of their optical characteristics and parameters of working 34 media [1 - 3]. Studies of such characteristics in gas-discharge exciplex sources of radiation on 35 mixtures of mercury diiodide vapor with helium and small additions of nitrogen and xenon at a 36 repetition rate of pump pulses of working mixtures up to 20 kHz were carried out in [4-6]. In radiators 37 on these mixtures the emission of a spectral band in the violet-blue spectral range (λ_{max} = 444 nm) 38 was revealed and regularities in the optical characteristics of the working medium (gas-discharge 39 barrier-discharge plasma) depending on the partial pressures of the mixture components were 40 established. For a number of scientific and technological applications, it is necessary to provide a high 41 radiation power and the use of more "heavy" buffer gases in the working mixture than helium, which 42 have less penetrating power through the walls of the radiator and thereby providing a longer life of the 43 radiation source [7-9]. The creation of radiators (excilamps) of higher power in violet-blue spectral 44 range requires the diagnostics of spectral characteristics of the plasma at higher pumping frequencies 45 of working mixtures, which was the aim of our studies of spectral characteristics of the plasma of a high-frequency (f = 130 kHz) atmospheric pressure barrier discharge on mixtures of mercury diiodide 46 47 vapor with neon .

48 2. EXPERIMENTAL INSTALLATION AND MEASUREMENT TECHNIQUE.

To carry out experiments to study the spectral characteristics of the atmospheric pressure barrier discharge (DBD) plasma on a mixture of mercury diiodide vapor with neon a small cylindrical radiator (Figure 1) with a single dielectric barrier (1) of capacitance $C_d = 40$ pF made of a quartz tube was used for the experiments. Inside along the axis there was a molybdenum electrode (2) with a diameter 1 mm.



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Figure 1. The scheme of the experimental setup: 1 - quartz tube, 2 - internal electrode, 3 - external electrode (mesh), 4- system of pumping and gas inlet, 5 - pulse generator, 6 - high-voltage transformer, 7 - digital oscilloscoper, 8 - voltage divider, 9 - current shunt, 10 - monochromator, 11 optical fiber, 12 - CCD detector, 13 - computer.

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The diameter of the radiator (tube) was 6 mm, the wall thickness - 1 mm. The discharge gap was 1.5 mm. An external electrode 30 mm in length (3) is made of a mesh with a transmittance T = 70%. At the end of the quartz tube was a 1.5 mm diameter capillary that served to pump the radiator, fill it with working mixtures, as well as to reduce the removal of mercury diiodide vapor from the radiator to the pumping and gas inlet system (4).

The high-voltage generator (5, Figure 1) with an average power of up to 100 W allowed to change the amplitude and frequency of the sinusoidal voltage applied to the electrodes to 7 kV and 130 kHz, respectively. The temporal dependences of the voltage at the source electrodes and the discharge current were recorded with a help of a dual-beam digital oscilloscope HP 54820A Infinium (7). The signals to the oscilloscope were fed from the Tektronix P6015A (8) voltage divider and the current shunt (9), respectively. The error in measuring the discharge current and voltage at the electrodes was 10%.

The radiation of the barrier discharge was recorded in a direction perpendicular to the lateral surface of the quartz tube and was analyzed in the spectral range 200- 900 nm. The radiation spectra were recorded using a Jobin Yvon TRIAX 550 spectrometer (10) (1200 lines / mm and 3600 lines / mm, a slit 0.02 mm, a quartz optical fiber (11), a Spectrum ONE (12) CCD detector cooled with liquid nitrogen). The spectral resolution of the recording system was 0.05 nm when using a 1200 lines / mm grating and 0.01 nm using a 3600 lines / mm grating. The registration system was calibrated on a
 relative intensity in the range 200-900 nm using a standart tungsten lamp.

79 Working mixtures were prepared directly in the radiation source. Hgl₂ mercury diodide in the 80 amount 60 mg was preloaded inside the radiator. The partial pressure of saturated vapor of mercury 81 diiodide was created by heating the working mixture with the energy of the discharge. After loading 82 the salt, dehydration and degassing of the inner surface of the source was carried out by heating at a temperature 70-100 ° C and evacuation for 2 hours. The partial pressure of neon was measured to 83 84 within ± 10 Pa. The value of Hgl₂ vapor partial pressure was determined from the temperature of the 85 coolest place of the radiator (the coolest place of the radiator was determined experimentally and in 86 our emitter it was at the opposite end of the tube relative to the external input 2 of the internal 87 electrode) on the basis of interpolation of the reference data [10] and under our conditions was 0.7 88 kPa.

89 **3.RESULTS AND DISCUSSION**.

Spectral characteristics of the plasma of a high-frequency barrier discharge were studied in a mixture Hgl_2 : Ne.= 0.7:110 kPa. After the initiation of the barrier discharge, a multi-avalanchstreamer type of discharge burning was observed visualy, typical for high frequencies-a set of coneshaped microdischarges (filaments) with a vertex on the electrode and a base on the inner surface of the quartz tube [11]. With increasing mixture temperature, the contrast of brightness in the volume discharge and filaments was smoothed out.

The survey spectrum of the radiation of a barrier discharge plasma on a mixture of mercury diiodide vapor with neon is shown in Figure 2.



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Figure 2. The survey spectrum of the radiation of a barrier discharge plasma on a mixture HgI_2 / Ne = 0.7 / 110 kPa.

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102 The most intense spectral bands and emission lines of the barrier discharge plasma of this mixture, 103 their relative brightness and intensity, taking into account the spectral sensitivity of the recording

104 system (J/k_{λ}) , as well as the excitation energy (E_{ex} , eV) are given in the Table. Data on the excitation

105 energies are taken from [13, 14, 16-18]. The width of the contours of the spectral atomic lines at half-

height ($\Delta\lambda_{0.5}$) was within the range 0.01-0.05 nm. For HgI (B \rightarrow X), the value of $\Delta\lambda_{0.5}$ was equal to 6.5

107 nm.

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109 vapor with neon Hgl_2 / Ne = 0.7 / 110 kPa.

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λ, nm	Molecule, atom	J/k_{λ} , a. u.	E _{ex.} , eV
253.6	Hg I	1.51	4.88 [15]
306	HgI(C→X)	4.80	8 [12]
342	l ₂	20.11	5 [16]
365.0	Hg I	0.64	8.86 [15]
404.6	Hg I	0.58	7.73 [13]
435.8	Hg I	0.67	7.73 [13]
443	Hgl(<i>B</i> →X)	102	7 [12, 17]
546.1	Hg I	0.67	7.73 [15]
585.2	Ne I	0.88	18.97 [15, 18]
640.2	Ne I	0.63	18.55 [15, 18]
703.2	Ne I	0.57	18.38 [15, 18]

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The spectrum in the visible range is dominated by a system of bands with a maximum at λ = 112 443 nm, which has a weakly resolved vibrational structure and corresponds to the $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ 113 electron-vibrational transition of HgI molecule [13]. The relative brightness (J/k_{λ}, a. u.) of this system 114 115 of bands was equal to the value 88 (Table). For this system of bands, a sharp increase in intensity 116 from the long-wavelength region and a slow decline in the short-wavelength region are characteristic. 117 In the conditions of atmospheric pressure barrier discharge, the edges of the spectral bands overlap 118 the wavelength range 350-450 nm. In addition to these spectral bands, intense lines of the neon atom 119 were observed in the region 580 - 740 nm, for which the relative brightness was more than 100 times less than the brightness of the system of bands with a maximum at λ = 443 nm (Table). 120

The spectrum also contains the emission of HgI molecules (C \rightarrow X) with a maximum intensity at $\lambda = 306$ nm (J/k_{λ} = 4.80 a. u.), the radiation of decay products of mercury diiodide is the band of molecular iodine I₂(D' \rightarrow A') with a maximum at $\lambda = 342$ nm (J/k_{$\lambda} = 20.11$ a.u.) and atomic mercury lines in the UV and visible range, the most intense of which are - 253.6 ($6^{3}P_{1} \rightarrow 6^{1}S_{0}$), 365.0 ($6^{3}D_{3} \rightarrow$ $6^{3}P_{2}$), 404.6 ($7^{3}S_{1} \rightarrow 6^{3}P_{0}$), 435.8 ($7^{3}S_{1} \rightarrow 6^{3}P_{1}$) and 546.1 nm ($7^{3}S_{1} \rightarrow 6^{3}P_{2}$) [12, 13]. For these lines, the relative brightness was between 1.51 and 0.58 (Table).</sub>

127 In more detail, the emission spectrum of HgI($B \rightarrow X$) with a resolved vibrational structure is 128 shown in Figure 3.

¹⁰⁸ Table. Spectral lines and emission bands of barrier-discharge plasma on mixtures of mercury diiodide



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Figure 3. The emission spectrum of HgI($B \rightarrow X$) with a resolution of 0.01 nm. A mixture HgI₂ / Ne = 0.7 / 110 kPa.

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133 The intensity is concentrated mainly in the long-wave part of the spectrum in the region 439-446 nm with a maximum of radiation at a wavelength λ = 443 nm, which corresponds to a minimum difference 134 in the potential curves of the upper $B^2 \Sigma_{1/2}^+$ and lower $X^2 \Sigma_{1/2}^+$ -states. The difference between the 135 equilibrium internuclear distances for these states is 0.4-0.5 Å [15]. Consequently, under conditions of 136 137 atmospheric pressure BR, transitions occur predominantly from the lower vibrational levels of the B state (v'=0÷5) to the upper vibrational levels of the X state (v"=9÷19). The bands shaded in the red 138 region for small values of v' were observed. At the same time, the shape of the bands (v'=0- v''=17, 139 v'=1- v"=18) is close to symmetrical, since other bands (v'=6- v"=24, v'= 6- v"=25) overlap at a 140 141 given wavelength.

Let us discuss the results of investigations of spectral characteristics of the plasma of the high-frequency atmospheric pressure BR of the mixture. The presence of certain bands and lines in the emission spectrum of a discharge, first of all, depends on the component composition of each mixture and the partial pressures of the components. The radiation intensity of the identified spectral bands and lines at working partial pressures is determined by the rate constants of excitation and quenching of the energy states of atoms and molecules by electrons, concentrations of electrons and mixture components in gas-discharge plasma [19].

The emission of the observed spectral transitions can be initiated by the following reactions [20 - 29]: 151 152 153 $I ({}^{2}P_{3/2}) + e (1)$

155	Hgl_2 +e $ ightarrow$ Hgl $_2({}^{3,1}\Sigma^*_{\mathrm{u}})$ $ ightarrow$ Hgl $(B^2\Sigma^+_{1/2})$ +	Г	(2)
156	$HgI_2+e \rightarrow HgI_2(D) \rightarrow HgI(C \ ^2\Pi_{1/2}, D^2\Pi_{3/2}) + I(^2P_{3/2}) + e$		(3)
157	$\mathrm{Hgl}\;(\mathcal{C}\;^2\Pi_{1/2},\mathcal{D}^2\Pi_{3/2}) + \mathrm{M} \to \mathrm{Hgl}(\mathcal{B}^2\Sigma^+_{1/2}) + \mathrm{M}\; + \Delta \mathrm{E}_{\mathrm{1,2}}$		(4)
158	$\operatorname{Hgl}_{2}\operatorname{+}\!\!\!e \to \operatorname{Hgl}_{2}({}^{3,1}\!\Sigma_{{}^{\!\!\!\!\!\!\!\!\!\!}}^{*})\to \operatorname{Hgl}(X^{2}\Sigma_{1/2}^{+})\operatorname{+}\!$		(5)
159	$\operatorname{Hgl}(X^2\Sigma_{1/2}^+) + \operatorname{e} \to \operatorname{Hgl}(B^2\Sigma_{1/2}^+) + \operatorname{e}$		(6)
160	Hgl($B^2\Sigma^+_{1/2}$) $ ightarrow$ Hgl($X^2\Sigma^+_{1/2}$) + hv		(7)
161	$\lambda_{\text{max.}}$ = 443 nm		
162	HgI(C ² Π _{1/2}) →HgI($X^2\Sigma_{1/2}^+$) + hν		(8)
163	λ _{max.} = 306 nm		
164	$HgI_2 + e \rightarrow Hg^* + 2I(I_2^*, I_2, I_2^-) + e$		(9)
165	$I_2(D') \to I_2(A')$		(10)
166	λ _{max.} = 342 nm		
167	$HgI_2 + e \rightarrow Hg^* + I_2^+ + 2e$		(11)
168	$HgI_2 + e \rightarrow Hg^* + I^+ + I + 2e$		(12)
169	Ne + e \rightarrow Ne* + e		(13)
170	$Ne^* \rightarrow Ne + hv$,		(14)
171	$Ne^{+}M \rightarrow Ne$		(15)
172			

173 where M – concentration of molecules and atoms (Hgl₂, Ne),

174 $\Delta E_{1,2}$ -the difference in energies of C ${}^{2}\Pi_{1/2}$, $D^{2}\Pi_{3/2}$ and $B^{2}\Sigma_{1/2}^{+}$ - states.

175 Reactions (1) and (2) are known as the main sources of exciplex mercury monoiodide (HgI*) 176 molecules. The rate constant, which according to the data of the studies [26] is equal to $2 \cdot 10^{-15}$ m³ / s 177 and $2.5 \cdot 10^{-17}$ m³ / s, respectively.

178 Kinetic equation for population $B^2 \Sigma_{1/2}^+$ - state of Hgl * molecules:

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$$\frac{d[HgI^*]}{dt} = k_{d} [HgI_{2}][N_{e}] - \tau_{r}^{-1} [HgI^*] - k_{q} [HgI^*][M], \qquad (16)$$

where k_d is the rate constant of dissociative excitation of HgI * molecules by electron impact, τ_r is the radiative lifetime of the state of HgI * molecules (27.3 ns [27]), k_q is the rate constant of quenching $B^2 \Sigma_{1/2}^+$ state of HgI * molecules; [HgI*], [HgI₂], [N_e], [M] are the concentrations of the molecules HgI*, HgI₂, electrons and quenching molecules and atoms (HgI₂, Ne), respectively (the quenching rate constants of mercury diiodide and neon molecules are $(3.6\pm0.3)\cdot10^{-16}$ m³/s and 2.9 $\cdot10^{-20}$ m³/s, respectively [30, 23]).

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For the quasi-stationary case, from expression (16), the emission intensity:

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$$I_{HgI^*}^{-1} = \alpha(1 + k_q \tau_r [M]), \qquad (17)$$

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190 where $\alpha = (k_d hv [Hgl_2][N_e])^{-1}$.

191 In addition, molecules of mercury monoiodide can be formed in reactions (3) due to the 192 excitation of mercury diiodide molecules to state D in collisions with electrons (D-state is the sum of 193 several states that lie between 5.7 and 9.5 eV (ionization threshold Hgl₂). Emission from the D-state 194 of Hgl₂ is not observed, since this state rapidly predissociates with the formation of electronically 195 excited Hgl* molecules in C and D-states [22 - 24]. They are quenched in reaction (4), leading to a high population $B^2 \Sigma^+_{1/2}$ - state of mercury monoiodide [23]. The collision reaction of mercury diiodide 196 197 molecules with electrons (5) is a channel for the formation of molecules of mercury monoiodide in the ground state, which rate constant has the value 8.10⁻¹⁵ m³/s [25]. In the collision of electrons with 198 mercury monoidide molecules in $X^2 \Sigma_{1/2}^+$ -state, $B^2 \Sigma_{1/2}^+$ -state (reaction 6) is excited, the rate 199 constant of which is 2.7·10⁻¹³ m³/s [26]. Electron-vibrational transitions $B^2 \Sigma_{1/2}^+ \rightarrow (X^2 \Sigma_{1/2}^+)$ and 200 $C^2\Pi_{1/2}) \rightarrow (X^2\Sigma_{1/2}^+)$ of mercury monoiodide molecules lead to the emission of spectral bands with 201 202 maximum intensity at a wavelength λ_{max} = 443 nm (reaction (7)) and λ_{max} = 306 nm (reaction (8)), 203 respectively [12, 27]. Excited iodine molecules are formed in reaction 9. Emission of spectral bands with a maximum of intensity at a wavelength λ_{max} = 342 nm is caused by the electronic-vibrational 204 205 transition $D' \rightarrow A'$ of iodine molecules (reaction 10) [28]. The excited mercury atoms are formed due 206 to the passage of the reactions (9,11,12) due to the large effective cross section for the dissociative 207 excitation of mercury diiodide molecules by electrons. The reaction (13) is responsible for the excitation of atoms of neon buffer gas, the rate constant of which is within the range (2-7) $\cdot 10^{-17}$ m³/s 208 209 (our calculation data). Excited neon atoms emit quanta (hv) as a result of radiation decay (reaction 210 14) as a result of which neon lines were observed in the spectrum (Figure 2). The expression for the 211 intensity of these lines can be determined from the kinetic equation for the population of the energy 212 states of neon atoms:

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214
$$\frac{d[Ne^*]}{dt} = k_{ex.} [Ne][N_e] - \tau_P^{-1} [Ne^*] - k_{q.} [Ne^*][M], \qquad (18)$$

215

where $k_{ex.}$ is the rate constant of the excitation of Ne^{*} atoms by electron impact, τ_r is the radiative lifetime of the energy states of the Ne^{*} atoms, k_q is the rate constant of the quenching of the energy states of the Ne^{*} atoms; [Ne^{*}], [Ne], [M] are the concentrations of the atoms Ne^{*}, Ne, electrons (N_e) and quenching molecules and atoms (Hgl₂, Ne), respectively (data on the rate constants of this quenching process (15) in the literature known to us are not available).

223
$$I_{Ne^*}^{-1} = \alpha(1 + k_q \tau_r [M]),$$
 (19)

224		
225	where $\alpha = (k_{ex.} hv [Ne][N_e])^{-1}$.	
226	In the emission spectra (Figure 2), the resonance line of	of the iodine atom did not appear at λ =
227	206 nm, which is due to the absorption of radiation by mercury	diiodide molecules in the process [29]:
228		
229	Hgl_2 + hv (206 nm) \rightarrow Hgl	(20)

230

4. CONCLUSION. 231

232 Thus, the diagnostics of spectral characteristics of the atmospheric pressure barrier discharge 233 plasma in mixtures of mercury diiodide vapor with neon during high-frequency pumping of the working 234 mixture revealed intense quasicontinuous emission of spectral bands with a maximum at λ = 443 nm 235 and 306 nm, as well as 342 nm of exciplex molecules mercury monoiodide and iodide molecules, 236 respectively. The main part of the emission of spectral bands is concentrated in the region 439-446 237 nm, the relative brightness of which is higher than the brightness of other spectral bands and lines. 238 Such a discharge of atmospheric pressure on a mixture of mercury diiodide vapor with neon at higher 239 pumping frequencies of the working mixture can be used to create a powerful gas discharge excilamp 240 that emits in the violet-blue spectral range, which operating life will be higher than the excilamp, 241 where the buffer gas is used helium.

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