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Spectral Characteristics of the Barrier Discharge on Mercury Diiodide Vapor with Neon Mixture

Antonina A. Malinina[1*]

[1]Department of Quantum Electronics, Uzhhorod National University, Narodna Square 3, Uzhgorod 88000, Ukraine.

Author's contribution

The sole author designed, analyzed and interpreted and prepared the manuscript.

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Short Research Article

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ABSTRACT

Aims: To investigate spectral characteristics of a high-frequency barrier discharge plasma of 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 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

**Corresponding author: E-mail: antonina.malinina@uzhnu.edu.ua;*

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.

Keywords: High-frequency barrier discharge; atmospheric pressure; radiation; spectral bands; mixture components; mercury diiodide; neon.

1. INTRODUCTION

The creation of new sources of optical radiation, which are important in modern technologies and scientific research, requires studies of their optical characteristics and parameters of working media [1-3]. Studies of such characteristics in gas-discharge exciplex sources of radiation on mixtures of mercury diiodide vapor with helium and small additions of nitrogen and xenon at a repetition rate of pump pulses of working mixtures up to 20 kHz were carried out in [4-6]. In radiators on these mixtures the emission of a spectral band in the violet-blue spectral range (λ_{max} = 444 nm) was revealed and regularities in the optical characteristics of the working medium (gas-discharge barrier-discharge plasma) depending on the partial pressures of the mixture components were established. For a number of scientific and technological applications, it is necessary to provide a high radiation power and the use of more "heavy" buffer gases in the working mixture than helium, which have less penetrating power through the walls of the radiator and thereby providing a longer life of the radiation source. In addition, for the solution of a number of problems of artificial photosynthesis, it is also important to create a radiation source that simultaneously radiates spectral bands and lines in the visible and ultraviolet spectral ranges [7-9].

The creation of such emitters of higher power in the visible and ultraviolet spectral ranges requires the diagnostics of spectral characteristics of the plasma at higher pumping frequencies 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 vapor with neon.

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 (Fig. 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. The experimental setup consists of: 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.

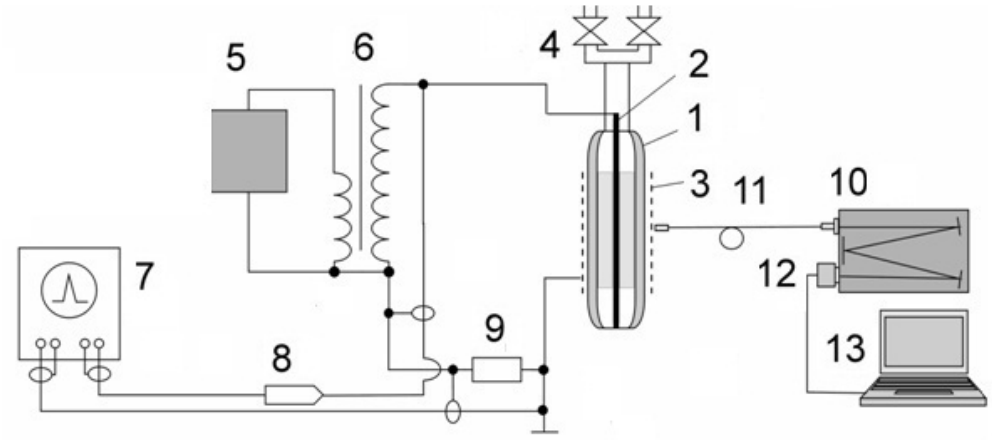

Fig. 1. The scheme of the experimental setup

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, Fig. 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.

Working mixtures were prepared directly in the radiation source. HgI_2 mercury diodide in the amount 60 mg was preloaded inside the radiator. The partial pressure of saturated vapor of mercury diiodide was created by heating the working mixture with the energy of the discharge. After loading 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 within ±10 Pa. The value of HgI_2 vapor partial pressure was determined from the temperature of the coolest place of the radiator (the coolest place of the radiator was determined experimentally and in our emitter it was at the opposite end of the tube relative to the external input 2 of the internal electrode) on the basis of interpolation of the reference data [10] and under our conditions was 0.7 kPa.

3. RESULTS AND DISCUSSION

Spectral characteristics of the plasma of a high-frequency barrier discharge were studied in a mixture HgI_2 : Ne.= 0.7:110 kPa. After the initiation of the barrier discharge, a multi-avalanch- streamer type of discharge burning was observed visualy, typical for high frequencies-a set of cone-shaped microdischarges (filaments) with a vertex on the electrode and a base on the inner surface of the quartz tube [11,12]. 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 Fig. 2.

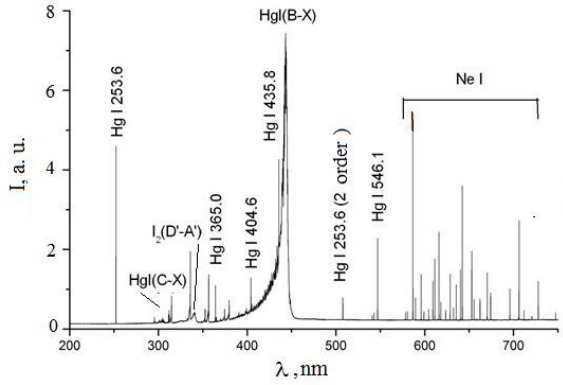

Fig. 2. The survey spectrum of the radiation of a barrier discharge plasma on a mixture HgI_2 / Ne = 0.7 / 110 kPa

The most intense spectral bands and emission lines of the barrier discharge plasma of this mixture, their relative brightness and intensity, taking into account the spectral sensitivity of the recording system (J/k_λ), as well as the excitation energy (E_{ex}, eV) are given in the Table 1. Data on the excitation energies are taken from [13,14, 15-17]. 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 → X), the value of $\Delta\lambda_{0.5}$ was equal to 6.5 nm.

The spectrum in the visible range is dominated by a system of bands with a maximum at λ = 443 nm, which has a weakly resolved vibrational structure and corresponds to the $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ electron-vibrational transition of HgI molecule [13]. The relative brightness (J/k_λ, a. u.) of this system of bands was equal to the value 102 (Table 1). For this system of bands, a sharp increase in intensity from the long-wavelength region and a slow decline in the short-wavelength region are characteristic. In the conditions of atmospheric pressure barrier discharge, the edges of the spectral bands overlap the wavelength range 350-450 nm. In addition to these spectral bands, intense lines of the neon atom 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 1).

The spectrum also contains the emission of HgI molecules (C → X) with a maximum intensity at λ = 306 nm (J/k_λ = 4.80 a. u.), the radiation of decay products of mercury diiodide is the band of molecular iodine $I_2(D' \rightarrow A')$ with a maximum at λ = 342 nm (J/k_λ = 20.11 a.u.) and atomic mercury lines in the UV and visible range, the most intense of which are - 253.6 ($6^3P_1 \rightarrow 6^1S_0$), 365.0 ($6^3D_3 \rightarrow 6^3P_2$), 404.6 ($7^3S_1 \rightarrow 6^3P_0$), 435.8 ($7^3S_1 \rightarrow 6^3P_1$) and 546.1 nm ($7^3S_1 \rightarrow 6^3P_2$) [12,13]. For these lines, the relative brightness was between 1.51 and 0.58 (Table).

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

Table 1. Spectral lines and emission bands of barrier-discharge plasma on mixtures of mercury diiodide vapor with neon HgI_2 / Ne = 0.7 / 110 kPa

λ, nm	Molecule, atom	J/k_λ, a. u.	$E_{ex.}$, eV
253.6	Hg I	1.51	4.88 [18]
306	HgI($C \rightarrow X$)	4.80	8 [12]
342	I_2	20.11	5 [15]
365.0	Hg I	0.64	8.86 [18]
404.6	Hg I	0.58	7.73 [13]
435.8	Hg I	0.67	7.73 [13]
443	HgI($B \rightarrow X$)	102	7 [12, 16]
546.1	Hg I	0.67	7.73 [18]
585.2	Ne I	0.88	18.97 [17, 18]
640.2	Ne I	0.63	18.55 [17, 18]
703.2	Ne I	0.57	18.38 [15, 18]

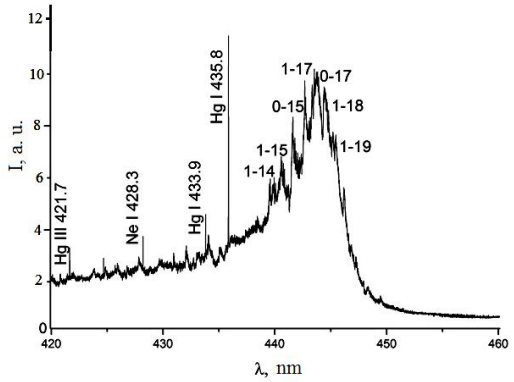

Fig. 3. The emission spectrum of HgI($B \rightarrow X$) with a resolution of 0.01 nm. A mixture HgI_2 / Ne = 0.7 / 110 kPa

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 in the potential curves of the upper *B* $^2\Sigma^+_{1/2}$ and lower *X* $^2\Sigma^+_{1/2}$ -states. The difference between the equilibrium internuclear distances for these states is 0.4-0.5 Å [18]. Consequently, under conditions of 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 region for small values of v' were observed. At the same time, the shape of the bands (v'=0- v''=17, v'=1- v''=18) is close to symmetrical, since other bands (v'=6- v''=24, v'= 6- v''=25) overlap at a 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]:

$HgI_2 + e \rightarrow HgI_2(^{3,1}\Sigma^+_u) \rightarrow HgI(B^2\Sigma^+_{1/2}) + I(^2P_{3/2}) + e$ (1)

$HgI_2 + e \rightarrow HgI_2(^{3,1}\Sigma^+_u) \rightarrow HgI(B^2\Sigma^+_{1/2}) + I^-$ (2)

$HgI_2 + e \rightarrow HgI_2(D) \rightarrow HgI(C\,^2\Pi_{1/2}, D^2\Pi_{3/2}) + I(^2P_{3/2}) + e$ (3)

$HgI(C\,^2\Pi_{1/2}, D^2\Pi_{3/2}) + M \rightarrow HgI(B^2\Sigma^+_{1/2}) + M + \Delta E_{1,2}$ (4)

$HgI_2 + e \rightarrow HgI_2(^{3,1}\Sigma^+_u) \rightarrow HgI(X^2\Sigma^+_{1/2}) + I(^2P_{3/2}) + e$ (5)

$HgI(X^2\Sigma^+_{1/2}) + e \rightarrow HgI(B^2\Sigma^+_{1/2}) + e$ (6)

$HgI(B^2\Sigma^+_{1/2}) \rightarrow HgI(X^2\Sigma^+_{1/2}) + h\nu$ (7)

$\lambda_{max.}$ = 443 nm

$HgI(C\,^2\Pi_{1/2}) \rightarrow HgI(X^2\Sigma^+_{1/2}) + h\nu$ (8)

$\lambda_{max.}$ = 306 nm

$HgI_2 + e \rightarrow Hg^* + 2I\ (I_2^*, I_2, I_2^-) + e$ (9)

$I_2(D') \rightarrow I_2(A')$ (10)

$\lambda_{max.}$ = 342 nm

$HgI_2 + e \rightarrow Hg^* + I_2^+ + 2e$ (11)

$HgI_2 + e \rightarrow Hg^* + I^+ + I + 2e$ (12)

$Ne + e \rightarrow Ne^* + e$ (13)

$Ne^* \rightarrow Ne + h\nu,$ (14)

$Ne^* + M \rightarrow Ne$ (15)

where M – concentration of molecules and atoms (HgI_2, Ne),

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

Reactions (1) and (2) are known as the main sources of exciplex mercury monoiodide (HgI*) molecules, namely these reactions are the channel of the dissociative excitation of the $B^2\Sigma^+_{1/2}$ - state of HgI* molecules as a result of inelastic collision of electrons with mercury diiodide molecules. The rate constant, which according to the data of the studies [26] is equal to $2 \cdot 10^{-15}$ m^3 / s and $2.5 \cdot 10^{-17}$ m^3 / s, respectively.

Kinetic equation for population $B^2\Sigma^+_{1/2}$ - state of HgI * molecules:

$$\frac{d[HgI^*]}{dt} = k_d [HgI_2][N_e] - \tau_r^{-1} [HgI^*] - k_q HgI^*][M], \quad (16)$$

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*], [HgI_2], [N_e], [M] are the concentrations of the molecules HgI*, HgI_2, electrons and quenching molecules and atoms (HgI_2, Ne), respectively (the quenching rate constants of mercury diiodide and neon molecules are $(3.6\pm0.3)\cdot10^{-16}$ m^3/s and $2.9\cdot10^{-20}$ m^3/s, respectively [30,23]).

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

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

where $\alpha = (k_d h\nu [HgI_2][N_e])^{-1}$.

In addition, molecules of mercury monoiodide can be formed in reactions (3) due to the excitation of mercury diiodide molecules to state D in collisions with electrons (D-state is the sum of several states that lie between 5.7 and 9.5 eV (ionization threshold HgI_2). Emission from the D-state of HgI_2 is not observed, since this state rapidly predissociates with the formation of electronically excited HgI* 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 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 $8\cdot10^{-15}$ m^3/s [25]. In the collision of electrons with mercury monoidide molecules in $X^2\Sigma^+_{1/2}$ -state, $B^2\Sigma^+_{1/2}$ -state (reaction 6) is excited, the rate constant of which is $2.7\cdot10^{-13}$ m^3/s [26]. Electron-vibrational transitions $B^2\Sigma^+_{1/2} \rightarrow (X^2\Sigma^+_{1/2})$ and $C^2\Pi_{1/2}) \rightarrow (X^2\Sigma^+_{1/2})$ of mercury monoiodide molecules lead to the emission of spectral bands with maximum intensity at a wavelength $\lambda_{max.}$ = 443 nm (reaction (7)) and $\lambda_{max.}$ = 306 nm (reaction (8)), respectively [12, 27]. Excited iodine molecules are formed in reaction 9. Emission of spectral bands with a maximum of intensity at a wavelength $\lambda_{max.}$ = 342 nm is caused by the electronic-vibrational transition *D*' $\rightarrow$ *A*' of iodine molecules (reaction 10) [28]. The excited mercury atoms are formed due to the passage of the reactions (9,11,12) due to the large effective cross section for the dissociative 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) $\cdot10^{-17}m^3/s$ (our calculation data). Excited neon atoms emit quanta ($h\nu$) as a result of radiation decay (reaction 14) as a result of which neon lines were observed in the spectrum (Fig. 2). The expression for the intensity of these lines can be determined from the kinetic equation for the population of the energy states of neon atoms:

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

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], [Ne], [M] are the concentrations of the atoms Ne*, Ne, electrons (N_e) and quenching molecules and atoms (HgI_2, Ne), respectively (data on the rate constants of this quenching process (15) in the literature known to us are not available).

For the quasi-stationary case, from expression (18), the emission intensity:

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

where $\alpha = (k_{ex.} h\nu [Ne][N_e])^{-1}$.

In the emission spectra (Fig. 2), the resonance line of the iodine atom did not appear at λ = 206 nm, which is due to the absorption of radiation by mercury diiodide molecules in the process [29]:

$$HgI_2 + h\nu\ (206\ nm) \rightarrow HgI \quad (20)$$

4. CONCLUSION

Thus, the diagnostics of spectral characteristics of the atmospheric pressure barrier discharge plasma in mixtures of mercury diiodide vapor with neon during high-frequency pumping of the working mixture revealed 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, respectively.

The main part of the emission of spectral bands is concentrated in the region 439-446 nm, the relative brightness of which is higher than the brightness of other spectral bands and lines. Such a discharge of atmospheric pressure on a mixture of mercury diiodide vapor with neon at higher pumping frequencies of the working mixture can be used to create a powerful gas discharge excilamp that emits in the violet-blue spectral range, which operating life will be higher than the excilamp, where the buffer gas is used helium. In addition, emission at these wavelengths, one in the visible range and the other in the UV range, can be efficiently used to activate photosynthesis with simultaneous destruction of viruses and bacteria, as well as solving a number of problems of artificial photosynthesis.

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COMPETING INTERESTS

Author has declared that no competing interests exist.

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