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Diagnostics of Optical Characteristics and Parameters of Gas-Discharge Plasma Based on Mercury Diiodide and Helium Mixture

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Abstract

The results of diagnostics of spectral, temporal and energy characteristics of the radiation of gas-discharge plasma in a mixture of mercury diiodide vapor with helium in the spectral range of 350 - 900 nm, and the plasma parameters in the range of reduced electric field E/N = 1 - 100 Td are presented. The plasma is created in the barrier discharge device with a cylindrical aperture. The electrodes are placed 0.2 m in length at a distance of 0.015 m. The amplitude of the pump pulse, the duration and frequency were 20 - 30 kV, 150 ns and 1 - 20 kHz, respectively. Radiation in the visible region of the spectrum of mercury monoiodide exciplex molecule is revealed. Regularities in the optical characteristics of the plasma, depending on the partial pressures of the components of the mixture, the electron energy distribution function, mean electron energy, specific losses of discharge power on the process of dissociative excitation of mercury monoiodide (state $B^2\Sigma^+_{1/2}$) molecules as well as the rate constant of dissociative excitation of mercury monoiodide molecules in working mixture depending on the given reduced electric field are established.

Keywords

Diagnostics, Optical Characteristics, Barrier Discharge, Gas-Discharge Plasma, Mercury Diiodide, Inert gases, The Plasma Parameters

1. Introduction

Gas-discharge plasma in mixtures of mercury diiodide vapor (HgJ_2) is a working medium of exciplex sources of

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coherent and spontaneous radiation in the visible spectral range of the spectral band with a maximum intensity at a wavelength (λ) 444 nm [1]-[5]. Interest in the study and creation of exciplex sources of spontaneous radiation of the visible spectrum is caused by the need to determine the quantitative characteristics of various processes occurring in their working mediums, creation of visible light source more effective than the existing ones, which would emit in the range of photosynthetic active radiation (PAR) and the largest plant sensitivity of biomolecules to solve a number of practical problems. These are tasks such as the intensive light therapy of humans and animals, effective light control of the production process (photosynthesis, growth, development, etc.) of plants and phytocenoses [6]-[8].

To date, several created and studied types of exciplex spontaneous sources of visible light in which to create gas-discharge plasma on working mixtures barrier discharge and other types of discharges have been used. In these works, for the most part, the main attention was focused on gas-discharge plasma on mixture of mercury dibromide vapor with gases: helium, neon, argon, xenon, krypton, nitrogen, and sulfur hexafluoride [9]-[17].

The aim of studies was to make a diagnosis of the spectral, energy and temporal characteristics of the radiation and the parameters of the gas-discharge plasma in mixtures of mercury diiodide vapor with helium, to identify regularities in these characteristics and to determine the partial pressures of the mixture components, as well as plasma parameters at which the maximum power of the radiation in the blue spectral range is reached.

2. Experimental Setup and Measurement Techniques

Plasma was created by barrier discharge in the device with the construction similar to that used in studies [18]. In this device the inter-electrode distance (was 0. 015 m), the length of the electrodes (0.2 m) and working volume (was equal to 111 cm^3) have been increased. Excitation of working mixture was performed by generator of repetitively pulsed form of the output voltage with the possibility of frequency adjustment in the range of 1 - 20 kHz and the amplitude of the voltage pulses in the range of 10 - 30 kV. Diagnostics of spectral, temporal and energy characteristics of the radiation of gas-discharge plasma was carried out on the experimental setup description of which is presented in our articles [15] [17].

3. Results and Discussion

The emission spectra of gas-discharge plasma were investigated in the field of partial vapor pressure of mercury diiodide 2 - 100 Pa and helium 120 - 200 kPa. Voltage, current, and pulse repetition rate of the pump pulses was 20 - 30 kV, 250 A and 10 - 20 kHz, respectively.

The characteristic emission spectrum is shown in **Figure 1(a)**. There is only the radiation of electron-vibrational bands of the transition B → X, υ' = 0 - 5, υ'' = 9 - 19 of exciplex molecules mercury monoiodide (HgI*) with maximum radiation at a wavelength λ = 444 nm, with steep increase in intensity from the longwave region and the slow decrease in the short-wave region [18].

The results of studies of integral characteristics (dependence of the average power of the radiation on the partial pressures of helium buffer gas and mercury diiodide vapor), as well as operation time of the radiation source on one portion of the working mixture are shown in **Figure 1(b)**, **Figure 1(c)** and **Figure 1(d)**. The maximum output power 0.6 watts is achieved at partial pressures of helium and mercury diiodide vapor 170 kPa and 100 Pa, respectively.

Research results of temporal characteristics of the discharge plasma are shown in **Figure 1(e)** and **Figure 1(f)**. The current pulses are of different polarity. Their complex shape is caused by the recharge “dielectric-plasma” circuit. Under the influence of the voltage pulse applied to the electrodes of the gas discharge device after the breakdown of the gas electrode gap a current of charging capacity of a dielectric (quartz glass) appears. This current flows in the forward direction. After charging the capacitance of the dielectric its discharge occurs and a current appears, direction of which is opposite to the charging current, therefore positive andnegative polarity is observed on the waveform of the current pulse.

The amplitude and duration of the current pulses is 250 A and 150 ns, respectively. The radiation pulses are of the same polarity **Figure 1(f)**. The radiation pulses duration on FWHM was ~150 ns. Accuracy and reproducibility of the measurement was 10% and 90%, respectively.

Since the experimental physics has no satisfactory method of diagnosis of a dense gas-discharge plasma, barrier discharge plasma parameters under optimal conditions to obtain the maximum emission power of HgJ_2-He (0.04% - 99.96%) mixture were determined and numerically calculated as the total integrals of the electron

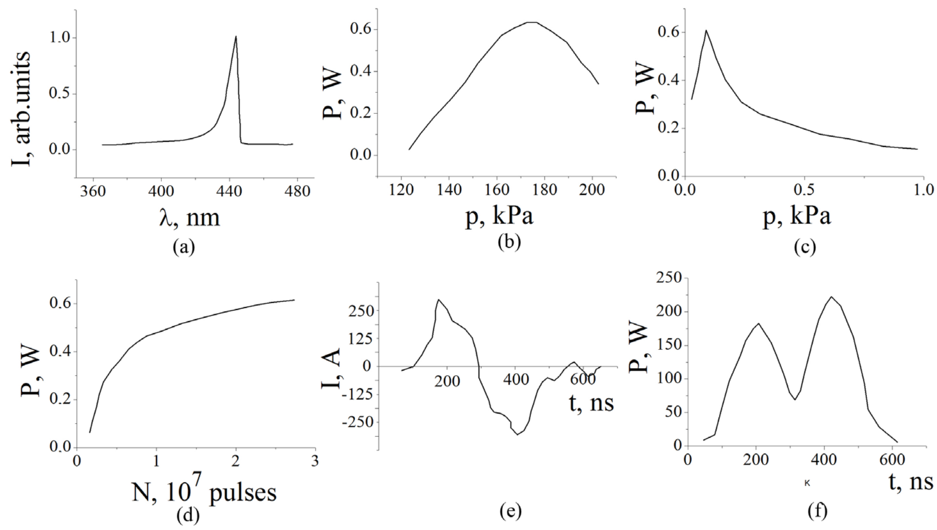

Figure 1. (a). The emission spectrum of a gas discharge plasma, (b) and (c). The dependence of the average radiation power of HgJ* exciplex molecules on partial pressure: (b) helium at a partial pressure of mercury diiodide vapor 100 Pa, (c) mercury diiodide vapor at helium partial pressure of 170 kPa, (d) the dependence of the average radiation power of HgJ* exciplex molecules on the total pulses number, (e) and (f). Oscillograms: the discharge current pulses (e), and the radiation power (f). The mixture HgI2: He = 100 Pa: 170 kPa. The amplitude of the voltage pulses and repetition rate are equal to 23 kV and 18 kHz respectively.

energy distribution function (EEDF) in the discharge. EEDF have been found numerically by solving the kinetic Boltzmann equation in the two-term approximation [19]. Calculations of EEDF were performed using program [20].

On the basis of the obtained EEDF the mean electron energy, specific power losses of electrical discharge on various elementary processes in plasma, as well as the rate constants of elastic and inelastic scattering of electrons on mercury diiodide molecules and helium atoms as a function of the reduced electric field (the ratio of the electric field (E) to the total concentration of mercury diiodide molecules and helium atoms (N)) are determined. The range of variation of the parameter E/N = 1 - 100 Td (1×10^{-17} - 1×10^{-15} V·cm^2) and included the values of parameter E/N = 25 - 75 Td that have been implemented in the experiment.

In the integral of electron collisions with helium atoms and mercury diiodide molecules the following processes are considered: elastic scattering, excitation of atom He energy states (23S, 21S, 23P, 21P, 3SPD, 4SPD, 5SPD), ionization of helium atom, dissociative excitation of electronic state of mercury monoiodide (B2Σ+/1/2) and ionization of mercury dibromide molecules. Data on the absolute values of the effective cross sections of these processes as well as their dependences on the electron energies are taken from works [20]-[22].

Figure 2(a) shows a characteristic EEDF form as the parameter E/N changes in the range 1 - 100 Td. An increase of the parameter E/N leads to an increase in the number of "fast" electrons in the discharge and to decrease the electron density in the work range of the radiator. The average electron energy of the discharge is most strongly dependent on the parameter E/N = 1 - 18 Td, while it increases linearly from 0.6 to 7.5 eV. In the range of the parameter E/N = 18 - 100 Td the mean electron energy is also increased from 7.5 to 13.25 eV, but at a slower rate.

For the process of dissociative excitation of mercury monoiodide molecules ($B^2\Sigma^+_{1/2}$ -state) **Figure 2(b)** specific losses of discharge power increase with the parameter E/N, reach a maximum 12% for E/N = 9 Td, and at further increase of the parameter E/N they decrease. The rate of increase and the decrease of the power losses of the discharge power on this process and its value is associated with the nature of the dependence of the effective excitation cross section of $B^2\Sigma^+_{1/2}$ state, on the electron energies and their absolute values, the dependence of the electron energy distribution function for different values of the parameter E/N and the value of threshold energy of dissociative excitation of $B^2\Sigma^+_{1/2}$ state of the mercury monoiodide molecule [23].

Figure 2 shows the results of a numerical calculation of the rate constant of dissociative excitation of mercury monoiodide molecules for the ratio of the partial pressure in the mixture, in which there is maximum radiated

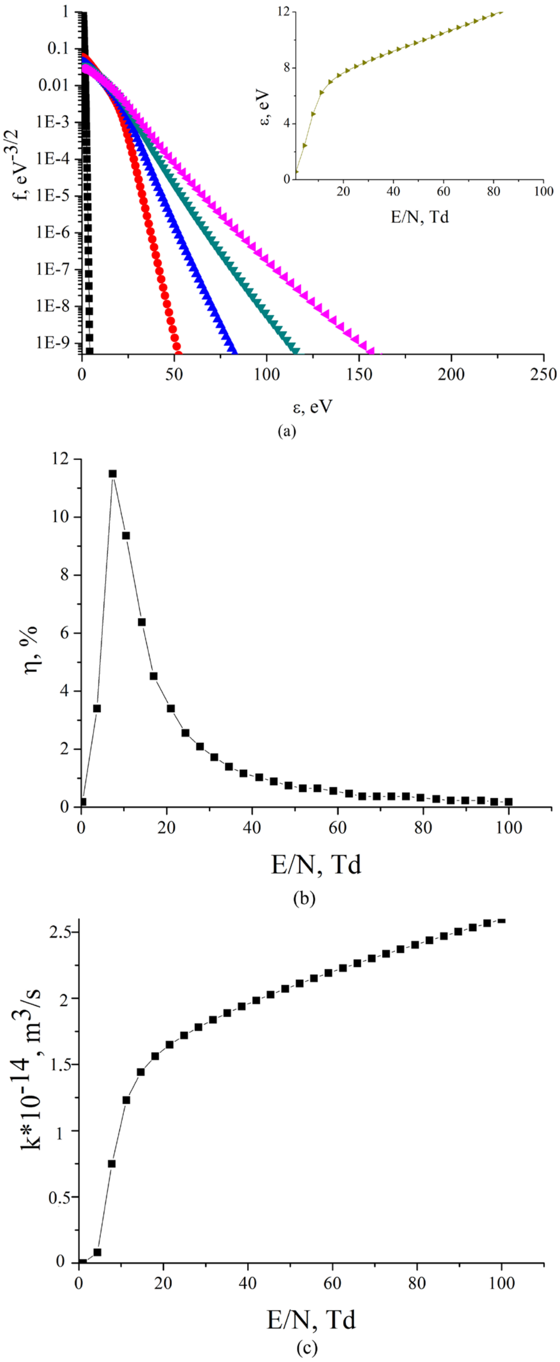

Figure 2. (a). Electron energy distribution functions in the discharge for the values of the parameter E/N: 1 (1), 25.8 (2), 50.5 (3), 75.3 (4), 100 (5) Td; at the insert-the dependence of mean electron energy on parameter E/N, (b) and (c). Dependences of the power losses of the discharge on dissociative excitation of mercury monoiodide molecules $B^2\Sigma^+_{1/2}$ -state and rate constant of dissociative excitation of mercury monoiodide molecules $B^2\Sigma^+_{1/2}$ -state by electrons on the parameter E/N. The total mixture pressure P = 170.1 kPa.

power of spectral band λ_{max} = 444 nm of exciplex molecule HgJ*. The rate constant is characterized by a high value k ≈ 10^{-14} - 10^{-15} m^3/s, that is associated with high values of absolute effective cross sections of the corresponding process. In the range of values of the parameter E/N = 25 - 75 Td, in which a radiation source works, the rate constant of $B^2\Sigma^+_{1/2}$ -state excitation has a value (1.6 - 2.4) 10^{-14} m^3/s.

The emergence of the spectral band emission with a maximum at a wavelength λ = 444 nm of electron-vibrational transition $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ of HgJ* exciplex molecules in a gas-discharge plasma in a mixture of mercury diiodide helium occurs in consequence of the processes leading to the formation and destruction of mercury monoiodide $B^2\Sigma^+_{1/2}$ -state, the main of which are [21] [22] [24]:

$$HgJ_2 + e \rightarrow HgJ^* + J + e \tag{1}$$

$$HgJ_2 + e \rightarrow HgJ^* + J^- \tag{2}$$

$$HgJ^* \rightarrow HgJ + h\nu , \tag{3}$$

$$HgJ^* + M \rightarrow HgJ + M + \Delta E \tag{4}$$

M-concentrations of molecules and atoms (HgJ_2, He) respectively.

The steep increase in the intensity from the area in the spectrum with longer wavelengths and a slow decrease in the short wavelengths (**Figure 1(a)**) is due to the progress of the potential curves (excited $B^2\Sigma^+_{1/2}$ -state shifted toward large internuclear distances relative to $X^2\Sigma^+_{1/2}$ -state, the difference between the equilibrium internuclear distances for these states is 0.4 - 0.5 Å [25]) and the relaxation processes population of the upper vibrational levels of the excited electronic state that occur faster than the electronic-vibrational transitions to the ground $X^2\Sigma^+_{1/2}$ -state [4] [23].

Therefore, under atmospheric pressure barrier discharge BD transitions occur predominantly from low vibrational levels of B - state (v'= 0 - 5) to the upper vibrational levels of X-state (v" = 9 - 19) [4].

The dependence of the radiation power of HgJ* exciplex molecules on the partial pressure of helium (**Figure 1(b)**) is caused primarily by the following processes: an increase in the electron concentration with increasing partial pressure of helium in the mixture, changes in portion of discharge energy, which is consumed for the working mixture heating; a change in the mean electron energy and the rate constant of exciplex molecules HgJ* excitation depending on the values of the parameter E/N as well as the process of quenching of the HgJ* molecules $B^2\Sigma^+_{1/2}$ -state in collisions with helium atoms [23] [26]. By increasing the partial pressure of helium in the mixture decreases the value E/N. This leads to an increase in specific losses of discharge power on elastic scattering of electrons by atoms and molecules and, consequently, to an increase in the partial pressure of mercury diiodide vapor and radiation power of mercury monoiodide exciplex molecules. Furthermore, an increase in radiation power with increasing partial pressures as helium and mercury diiodide vapor facilitates an increase of electrons concentration, which increases with the increase of components concentration in the working mixture [23]. The presence of a maximum and a further drop in the radiation power of mercury monoiodide exciplex molecules at increasing the partial pressure of helium is due to a decrease in the proportion of the discharge energy that is spent on the excitation of molecule HgJ* $B^2\Sigma^+_{1/2}$ -state (2b), and the process of quenching this state of mercury monoiodide molecules in collisions with atoms of helium, the rate constant of which equals to 2.9 × 10^{-16} m^3/s [26].

$$HgJ^* + He \rightarrow HgJ + He + \Delta E , \tag{5}$$

where ΔE is the energy difference in the reaction.

The dependence of the radiation power of exciplexes HgJ * on the partial pressure of mercury diiodide vapor (**Figure 1(c)**) and the optimal value of the partial pressure of mercury diiodide vapor is determined by the kinetics of processes leading to excitation and quenching of molecules HgJ* $B^2\Sigma^+_{1/2}$ -state, the rate constant of the process (1) and the effectiveness of the quenching process:

$$HgJ^* + HgJ_2 \rightarrow HgJ + HgJ_2 + \Delta E , \tag{6}$$

where ΔE is the energy difference in the reaction.

Above a certain value of the partial pressure of mercury diiodide vapor quenching process (8) will play an important role in connection with what occurs a decrease in the radiation power. The rate constant of this process is

$(3.6 \pm 0.3) \times 10^{-16}$ m^3/s [27].

4. Conclusions

Atmospheric pressure barrier discharge plasma in a mixture of mercury diiodide vapors with helium gives the spectral emission band in the blue-green region (λ_{max} = 444 nm). An average output power of 0.6 W and pulsed power 220 W are reached at a pulse repetition rate of the pump pulses 18 kHz, the amplitude of voltage and current pulse 23 kV and 250 A, respectively, and duration ~150 ns (the amount of radiation 111 cm^3). The results of numerical modeling of processes in plasma allowed to establish the electron energy distribution function, mean electron energy, specific losses of discharge power on the process of dissociative excitation of mercury monoiodide molecules (state $B^2\Sigma^+_{1/2}$) as well as the rate constant of dissociative excitation of mercury monoiodide molecules in the working mixture depending on the value of the reduced electric field.

Atmospheric pressure barrier discharge plasma in a mixture of mercury diiodide vapors with helium can be used as the working medium of the radiator in the blue spectral range that can be used in scientific researches in the area of biotechnology, photonics, medicine, as well as for creation discharge indicator panels.

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