

Optical Characteristics
of Gas-Discharge
Plasma in Mixtures
of Mercury Dibromide
Vapor with Gases

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INTRODUCTION

The optical characteristics of gas-discharge plasma in mixtures of different compositions are its most important characteristics. They carry information about the physicochemical processes occurring in plasma and its parameters and characteristics, including: the concentrations of electrons, atoms and molecules; and the energy distribution, temperature and drift velocities of electrons. These optical characteristics also characterize the efficiency of elastic and inelastic collisions of electrons with working gases, quenching the energy states of excited molecules, as well as the discharge power losses in electronic processes [1].

Intensive study into the optical characteristics of gas-discharge plasma began with the discovery of the effect of lasers on mixtures of metal vapors, salts and gases. This led to a need to increase their energy parameters and search for new active media [2–4]. Studies of the characteristic effects of gas-discharge plasma in mixtures of mercury dibromide vapor with gases are particularly prominent. Such plasma is used as a working medium for exciplex sources of pulse-periodic coherent (lasers) and spontaneous radiation (excilamps) in the blue-green region of the spectrum and with a wavelength at maximum intensity of ($\lambda_{\max.}$) = 502 nm [5–14]. These sources of radiation have attracted the attention of researchers in connection with the high specific power of radiation, which is recorded in the visible spectral range, by tuning the wavelength of radiation across a wide range of the spectrum, and also with the possibility of operating in a pulse-periodic mode. The average power of laser radiators reaches ~200 W at an efficiency

of 2.1 % with energy in a single pulse of 9.8 Joules (2.4 % efficiency) [8–11]. Such sources of radiation are needed for a number of important applications: the monitoring of air and water basins; range finding of marine objects; underwater communications; the technology of electronic equipment materials; increasing the efficiency of photosynthesis in plants (in greenhouses, space vehicles, submarines, etc.); and pumping solid-state and liquid lasers [15–19].

The data on optical characteristics of the working medium of radiation sources (gas-discharge plasma in mercury dibomide vapor and gas mixtures) make it possible: to diagnose the spectral composition of the discharge plasma radiation; to reveal the optimal component composition of the working mixtures, at which the maximum radiation intensity in the investigated spectral range is reached; and to determine the pulsed radiation characteristics. In addition, such characteristics allow us to establish the physicochemical regularities in the processes that occur in the plasma of a gas discharge in working mixtures, to find ways to increase their energetic and spectral characteristics [3].

During the first studies on optical characteristics, i.e., the emission spectra of gas discharge plasma in a mixture of mercury dibromide with small additions of inert gases (<10 kPa), the emission of spectral bands in the electron-vibrational transitions $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ of mercury monobromide molecules was revealed—later, these were called exciplex ($HgBr^*$) molecules [3]. By nature these spectral radiation bands broaden from the side of short wavelengths [20, 21]. When the total pressure of the mixture increases, they become narrower due to the relaxation process of the population of the vibrational levels of the $B^2\Sigma^+_{1/2}$ state [22–24]. The radiation pulses were delayed relative to the discharge current pulses by several tens of nanoseconds and their duration (generation) was 30–70 ns

[5–7, 25, 26]. In the dependencies of the radiation intensity of the exciplex HgBr^* molecule on the partial pressures of mercury dibromide and inert gases, the following regularities can be observed: an increase in intensity with an increase in the partial pressures of the mixture components, until a maximum is reached; and a decrease in the radiation intensity with a subsequent increase in the partial pressures of the components of the mixture. The radiation intensity with increasing partial pressures after reaching the maximum decreases with a lower rate for inert gases than for mercury dibromide vapor. The partial pressures at which the maximum radiation power is reached are 0.27–1.06 kPa for mercury dibromide vapor and 90–120 kPa for inert gases [26–28]. The most intense emission of mercury monobromide occurs in a mixture of mercury dibromide and helium. The authors of these studies explained their results by changing the reduced electric field strength E/N (where E is the electric field applied to the plasma and N is the total concentration of the particles in the working mixture), and also by quenching the $B^2\Sigma^+_{1/2}$ state of mercury monobromide molecules with the components of the plasma working mixture. A change in the reduced electric field strength leads to a redistribution of the discharge power into various processes that affect the intensity of the radiation, and also to either an increase or decrease in the efficiency of the processes of populating and depopulating the energy states of the components of the working mixtures. The rate constants of elastic and inelastic processes in the collision of electrons with atoms and molecules, which are a quantitative measure of process efficiency, are mainly determined by the theoretical method, which is associated with methodical difficulties and poor knowledge about the efficiency of plasma-chemical processes occurring in plasma in working mixtures [29–32]. In turn, the quenching rate constants of the $B^2\Sigma^+_{1/2}$ states of mercury monobromide molecules by the components

of the mixture can be determined by optical methods for mercury dibromide vapor with helium and neon mixtures. These are in the range $(1.7\text{--}2.7) \cdot 10^{10} \text{ cm}^3/\text{s}$ and $(1.5\text{--}5.5) \cdot 10^{14} \text{ cm}^3/\text{s}$ for quenching by mercury dibromide and helium (neon) molecules, respectively [33–36]. The rate constant of the main population process of the $B^2\Sigma^+_{1/2}$ state of mercury monobromide molecules (dissociative excitation of $B^2\Sigma^+_{1/2}$ states of mercury monobromide molecules in the collision of plasma electrons with mercury dibromide molecules) in mixtures of mercury dibromide vapor with helium and neon varies in the limits of the values $1 \cdot 10^{10}\text{--}9 \cdot 10^9 \text{ cm}^3/\text{s}$ with a change in the reduced electric field strength from 3 to 100 Td [32].

Previous investigations into the optical characteristics of gas-discharge plasma in mixtures of mercury dibromide vapor and gases saw the following experimental conditions: the amplitude of the voltage and current pulses reached 50 kV and several kA, respectively; the repetition rate of the pump pulses was in the range 1–2,000 Hz; and total pressures of the mixtures reached 200 kPa. The studies were carried out with mixtures of mercury dibromide vapor and the following gases: helium, neon, argon, nitrogen, xenon, and krypton. Operating devices with a volume of more than 200 cm^3 were used. The highest radiation powers of mercury monobromide exciplexes were obtained for mixtures of mercury dibromide vapor with helium and small nitrogen additions. This is explained by the addition of a channel for the population of $B^2\Sigma^+_{1/2}$ states of mercury monobromide in collisions of mercury dibromide molecules with metastable nitrogen molecules [37].

The study of the parameters and characteristics of plasma discharge in working mixtures is of paramount importance in the study of their optical characteristics, since the optical characteristics of the plasma discharge constitute their functions [1–3]. The parameters and characteristics of the

plasma discharge in the working mixtures were determined using data on the efficiency of electron collisions with the components of the working mixtures. On the basis of these data, the optimum reduced electric field strength (E/N) was established for which the efficiency of the sources on the mixtures increased. To establish this data, a theoretical method was used [29–32]. In the mixture of mercury dibromide vapor with helium, or neon (argon), and also with the addition of nitrogen (a small amount in the mixture), the transport and energy characteristics; the specific power losses of the discharge during the elastic and inelastic processes of collisions of the discharge electrons with the atoms and molecules of a particular mixture; and the rate constants of these processes were calculated [32].

An analysis of the results available in the scientific literature on the investigation into the optical characteristics of gas-discharge plasma in mixtures of mercury dibromide vapor and gases gave us new research problems. These problems included the establishment of the physicochemical regularities that occur in new working mixtures based on mixtures of mercury dibromide vapor with gases, to reveal ways to increase the energy characteristics of radiation from lasers and excilamps in the blue-green spectral range. In addition, there was a need to carry out investigations into the optical characteristics of gas-discharge plasma in the region of large pump repetition rates (above 2,000 Hz), and the duration of the pump pulses close to the lifetime of the $B^2\Sigma^+_{1/2}$ state of mercury monobromide (≤ 30 ns), to reveal new and more effective mixtures of mercury dibromide vapor with gases. Finally, we sought to determine the characteristics and parameters of the plasma discharge under the experimental conditions at which the maximum specific energy characteristics of the radiation of mercury monobromide exciplex molecules were observed.

CHAPTER 1

THE RESULTS OF PRELIMINARY INVESTIGATIONS INTO GAS-DISCHARGE PLASMA IN MIXTURES OF MERCURY DIBROMIDE VAPOR WITH GASES

1.1. Optical characteristics of radiation

Investigations into the optical characteristics of the radiation of gas-discharge plasma in mixtures of mercury dibromide vapor with gases have developed apace since the late 1980s with the creation of a laser that could generate radiation in the spectral range of 502 to 506 nm [5]. To obtain an inversion, gas-discharge plasma was used, which created an atmospheric pressure volume discharge in a mixture of mercury dibromide vapor with helium. The active element of the laser was made of a sealed pyrex tube with an inner diameter of 4 cm and a length of 35 cm. The electrodes of the laser were two tungsten rods 6 mm in diameter and 25 cm in length; the distance between them was 1 cm. Near the cathode (parallel to it) an electrode was located, which was placed inside the glass tube and was used to pre-ionize the working mixture in the inter-electrode gap. The laser was pumped by a generator that provided a pulse voltage to the laser electrodes with an amplitude above 8 kV, a duration of 70 ns, and a pulsed voltage on the pre-ionization electrode. Crystals of mercury dibromide were placed in the active element of the laser and filled with a buffer gas—helium—at a pressure of 1,000 mm Hg. The partial pressure of the mercury dibromide

vapor was created by heating the powder with an external electric heater. The laser-generated energy of several tens of millijoules had a pulse duration of 50 ns. The efficiency of the laser did not exceed 1 %. The authors of [5] have established that the radiation in this spectral region appears in the electron-vibrational transition $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ from $v' = 0-3$ $v'' = 22-23$ diatomic molecules of mercury monobromide. In subsequent work on the optical characteristics, the design of the active element and the parameters of the pump pulses, as well as the component compositions of the working mixtures, were changed, making it possible to increase the energetic characteristics of the laser. When the working mixtures were pumped by a discharge controlled by an electron beam, the radiation energy and efficiency reached 9.8 J and 2.4 %, respectively. In the pulse-periodic mode with electric-discharge pumping and X-ray pre-ionization, the average radiation power was soon brought to 200 W, and the efficiency to 2.1 % [8–11].

These experiments were preceded by studies into the emission spectra of a high-frequency discharge in mixtures of mercury dihalides with small additions of inert gases (several kPa) [20, 21]. The emission of electron-vibrational spectral bands for the transition $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ of the mercury monohalide molecule was observed. At the atmospheric pressure of working mixtures necessary for more efficient laser generation of radiation, the emission spectra were almost the same as for mixtures with the addition of buffer gases of several kPa. However, the emission bands were narrower due to the rapid relaxation of the population of vibrational levels of $B^2\Sigma^+_{1/2}$ -state mercury monobromide [22–24].

A weak dependency of the intensity of radiation on the partial pressure of an inert gas for two-component mixtures was noted in [26–28], as well as the presence of an interval of the partial pressure of dihalides and inert

gases (for multicomponent mixtures) at which the radiation intensity was high.

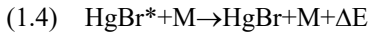
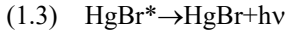
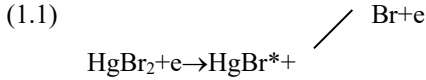
The temporal characteristics of plasma radiation in mixtures of mercury dihalides and inert gases were investigated in [5–7, 25, 26]. The half-widths of the current pulses through the gas-discharge gap and its generation were 30–70 ns. Current pulses outran radiation pulses by several tens of nanoseconds.

The necessity of explaining the optical characteristics of plasma radiation discharge in mercury dibromide vapor mixtures has stimulated interest in the molecular terms of mercury monobromide and mercury dibromide [20, 21, 38, 39].

Figure 1.1 shows a simplified diagram illustrating the formation of HgBr molecules ($B^2\Sigma^+_{1/2}$) under conditions of gas-discharge excitation of a mercury dibromide with gas mixture. According to the data of [38], the $B^2\Sigma^+_{1/2}$ state of mercury monobromide has a strongly bound ionic character, while the ground $X^2\Sigma^+_{1/2}$ state is weakly bound and has a covalent character. The minimum of the potential curve of the B-state of mercury monobromide shifts by 0.6 Å relative to the X-state, which causes a preferential transition from lower vibrational levels of the $B^2\Sigma^+_{1/2}$ state to the upper $X^2\Sigma^+_{1/2}$ states. Such molecules are usually called exciplex-like and are denoted by an asterisk (HgBr*) [2, 3].

It has been established that dissociative excitation in the collision of mercury dibromide molecules with electrons is the main mechanism for the population of the upper energy level ($B^2\Sigma^+_{1/2}$ state) of mercury monobromide in gas-discharge plasma in a mercury dibromide vapor and helium (neon) mixture [3, 13, 40–43]. Immediately after the formation of this excited state of mercury monobromide in the plasma of a gas discharge,

its radiative decay begins. In addition, the $B^2\Sigma^+_{1/2}$ state of $HgBr^*$ is quenched by heavy particles and electrons [3]:



where M represents the particles that quench the $B^2\Sigma^+_{1/2}$ state of mercury monobromide and ΔE is the energy difference in the reaction.

These processes (1.1–1.4) primarily explain the dependency of the intensity of emission of mercury monobromide exciplex molecules on the partial pressures of mercury dibromide. The contribution of processes (1.1–1.4) to radiation intensity depends on the rate constants of excitation and quenching of the $B^2\Sigma^+_{1/2}$ state and particle concentrations [1, 3].

For the same mixtures with the addition of molecular nitrogen or xenon, the excitation of the $HgBr B^2\Sigma^+_{1/2}$ state can occur via an additional channel due to the transfer of energy from nitrogen molecules that are in the metastable state $A^3\Sigma^+_u$ and xenon atoms in the metastable state 3P_2 . The rate constants of this process are rather high: $1 \cdot 10^{-10} \text{ cm}^3/\text{s}$ and $5.3 \cdot 10^{-10} \text{ cm}^3/\text{s}$, respectively [37].

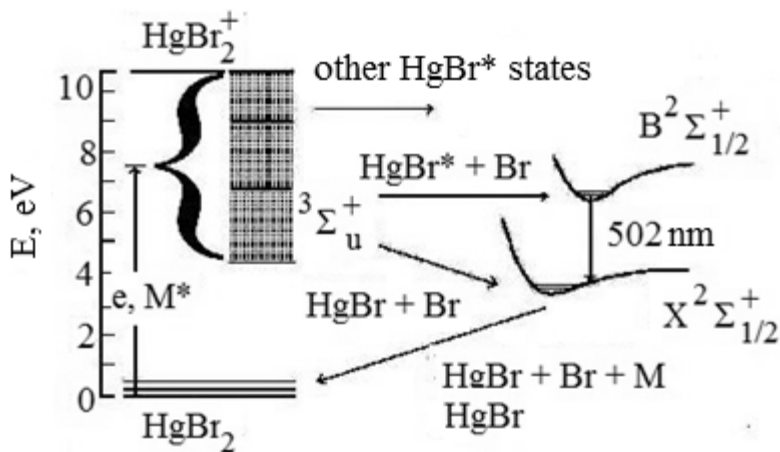


Figure 1.1. A simplified diagram illustrating the formation of HgBr ($B^2\Sigma_{1/2}^+$) molecules under gas discharge excitation of mercury dibromide with gas mixtures; M^* —metastable atoms and molecules [30, 39].

The addition of neon, along with nitrogen and sulfur hexafluoride, to the mixture leads to an increase in the population of metastable state $A^3\Sigma_u^+$ nitrogen molecules due to the relaxation of the population from higher-lying nitrogen levels in collisions with sulfur hexafluoride molecules and the fragments thereof. This leads to a 50 % increase in the energy of HgBr laser radiation [4]. In addition to the above processes, the HgBr $B^2\Sigma_{1/2}^+$ state can also be excited through collision reactions with the ions of inert gases and nitrogen, as the rate constants of these processes reach high values $(3-7) \times 10^{-10} \text{ cm}^3/\text{s}$ [44]. However, the energy deposition of such processes to the excitation of mercury monobromide $B^2\Sigma_{1/2}^+$ state in gas-discharge plasma in mixtures of mercury dibromide vapor with gases has not yet been determined.

In most studies of the characteristics of gas-discharge plasma in mixtures of mercury dibromide vapor with gases, excitation of the working mixtures was done by discharge through barrier-dielectric quartz glass, which provided a uniform burning of the discharge without the use of preliminary ionization at typical atmospheric pressures of the working mixtures and simplification of the design of the gas-discharge device [12, 14, 26–28, 45–47]. The first report on the studies of the spectral, integral, and temporal characteristics of gas-discharge plasma in mixtures of mercury dibromide vapor with gases for excitation, using a single-barrier discharge, dates from April 1980 [12]. The excitation of HgBr_2 vapor in a mixture with helium, as an inert gas, was carried out in a gas discharge cuvette and used a thin-walled quartz tube. Two tungsten electrodes were located inside the gas discharge cuvette, one of which was fitted with a quartz tube. To fill the gases in the side of the cuvette, a quartz glass tube was welded in place. Inside the tube there was a capillary, which reduced the removal of mercury dibromide vapor to the pumping and gas inlet system. To create the desired partial pressure of mercury dibromide vapor, the gas discharge cuvette was heated using an electric heater, which was located on an additional external quartz tube. A high-voltage pulse-periodic voltage of 30–40 kV, with a pulse duration of 50 ns and a repetition rate of 10–100 Hz, was applied to the electrodes. In these studies, only one spectral band with a maximum intensity of $\lambda = 502$ nm was detected in the spectral range 300–700 nm. A weak dependency of the radiation intensity on the partial pressure of helium was observed in the temperature range 100–2,000 °C in the heating of the gas-discharge cuvette. Experiments on the temporal characteristics of gas-discharge plasma in a mixture of mercury dibromide vapor with gases used a mixture of mercury dibromide vapor with helium [12]. Current pulses were twofold. Each individual current pulse had a duration of 50 ns with a

leading rise-front of ~ 10 ns. The time between pulses was 150 ns. The radiation pulses were also twofold. The amplitude of the second pulse in the radiation was higher than in the first one. As the pressure of helium increased, the amplitude of both pulses decreased. However, for the first pulse, this decrease was faster. The authors suggested that the first pump pulse generated excited and non-excited molecules of mercury monobromide. The radiation of the excited molecules was determined by the amplitude of the first optical pulse. Under the action of the second pump pulse, molecules of mercury monobromide from the $X^2\Sigma^+_{1/2}$ state were excited. As a result of this, there was an additional increase in the population of the $B^2\Sigma^+_{1/2}$ state, leading to an increase in the amplitude of the second radiation pulse. The decrease in the amplitude of the radiation pulses with an increase in the partial pressure of helium was caused by a decrease in mean electron energy, leading to a decrease in the efficiency of the process of excitation of the $B^2\Sigma^+_{1/2}$ state and, accordingly, its population decreased.

Figure 1.2 shows the results of the author's investigations into the optical characteristics of gas-discharge plasma in mixtures of mercury dibromide vapor with inert gases, [26]. It shows the characteristic emission spectrum of a mixture of HgBr_2 vapor with helium (neon, xenon) and the dependency of the emission intensity of the spectral band of $\lambda = 502$ nm HgBr^* on the partial pressures of mercury dibromide vapor with helium, neon and xenon [26]. The spectra differed in the rate of the decrease in the intensity of the spectral band of $\lambda = 502$ nm HgBr^* molecules in the ultraviolet part, as well as in width, depending on the partial pressure of the inert gases. The emission spectra of a mixture of HgBr_2 vapor with helium (neon, xenon) for low pressures (less than atmospheric) continued further into the region of shorter wavelengths. The dependency of the emission intensity of the spectral band in the short-wave region can be explained by

the relaxation processes of the population of the upper ($v' > 1$) vibrational levels of the $B^2\Sigma^+_{1/2}$ states, which occur more rapidly than the electron-vibrational transition to the ground $X^2\Sigma^+_{1/2}$ state [22–24]. The dependency of the radiation intensity on the partial pressure of mercury dibromide shows an increasing trend, and the region of maximum radiation is found at $HgBr^*$ 0.27–1.06 kPa. The results of the studies presented in Figure 1.2 show that the most intense emission of mercury monobromide molecules takes place in a mixture of mercury dibromide vapor with a light helium gas. The behavior of the radiation intensity of gas-discharge plasma in the partial pressures of mercury dibromide vapor and inert gases can be explained by the rate of the processes of population and depopulation of the mercury monobromide $B^2\Sigma^+_{1/2}$ state, which, in turn, depends on the parameter E/p (where E is the electric field strength, which is applied to the plasma gap, and p is the total pressure of the mixture). For a mixture with helium, the excitation efficiency of the mercury monobromide $B^2\Sigma^+_{1/2}$ state is higher, and the quenching process by helium is lower [30–36]. With increasing voltage of up to 40 kV and a repetition rate of the generator pulses from 10 Hz to 100 Hz, a proportional increase in the radiation intensity can be observed. Over a period of time (>50 hours), the decrease in the radiation intensity in the spectral band is negligible [26].

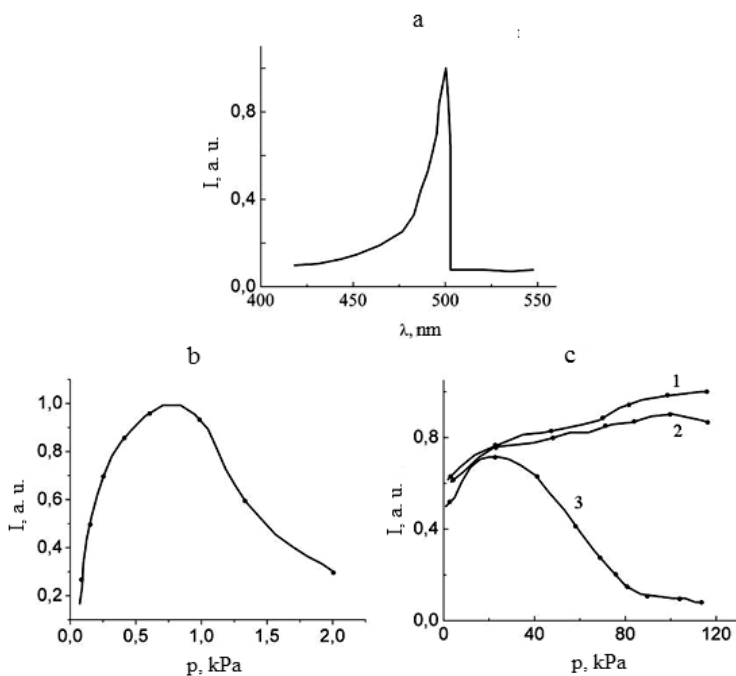


Figure 1.2. a) The radiation spectrum of a pulsed discharge in a He:HgBr₂ = 119.7:0.27 kPa mixture; b) the dependency of the emission intensity of the spectral band $\lambda = 502 \text{ nm HgBr}^*$ on the saturated vapor pressure HgBr₂ for the He:HgBr₂ mixture where the partial pressure of helium is 106.4 kPa; c) the dependency of the emission intensity of the $\lambda = 502 \text{ nm HgBr}^*$ band on the buffer gas pressure for the mixture: (1) He:HgBr₂, (2) Ne:HgBr₂; (3) Xe:HgBr₂—the saturated vapor pressure of HgBr₂ was 0.27 kPa [26].

A number of articles [14, 27, 28, 45, 46] present data on the optical characteristics of gas-discharge plasma at high pumping frequencies of working mixtures (up to 2,000 Hz). When pumping a mixture of mercury dibromide and neon with a microwave discharge for a discharge volume of 14.13 cm³, an average radiation power of 42.8 W and an emission efficiency

of 9.4 % [47] can be achieved. [14, 27, 28, 45, 46] have all reported on the optimization of the energy and resource characteristics of working mixtures of HgBr laser and an excilamp in a pulse-periodic discharge plasma at pump repetition rates of 400—1,900 Hz. The gas-discharge plasma was created by combined surface and barrier discharge. Investigations were carried out on two, three and four-component mixtures of the following compositions: HgBr₂:He; HgBr₂:N₂:He; HgBr₂:Xe:He; HgBr₂:Xe:N₂:He. The partial vapor pressure of HgBr₂ in the working mixtures was created by dissipation of the discharge energy. The dependency of the radiation intensity on the composition of the mixture was measured at a pump pulse repetition rate of 1,000 Hz and pulse voltage of 18 kV. For a double HgBr₂:He mixture, the increase in helium pressure from 141 kPa to 200 kPa resulted in an increase in the average radiation power of 1.7 times. For the three-component mixtures (curves 1 and 2, Figure 1.3a), maximum radiation powers were achieved with xenon partial pressures of 2.03–4.05 kPa; for nitrogen the range was 12.13–47.96 kPa. The average radiation power for four-component mixtures was greater than for a ternary mixture with xenon, but smaller than with nitrogen. The optimal ratio of the average power for the component composition mixtures was: 1:1.9:4.2:21.5 = mixture 1:mixture 2:mixture 3:mixture 4, where: 1 - HgBr₂:He (helium pressure 121.6 kPa); 2 - HgBr₂:Xe: He (Xe: He = 1:39); 3 - HgBr₂:Xe:N₂:He (the ratio of Xe:N₂:He = 1:10:29); and 4 - HgBr₂:N₂:He (the ratio of N₂:He = 1:3). Fig. 1.3b shows the dependency of the average radiation power on the number of pump pulses on one portion of the working mixture. The growth rate of the average radiation power was larger for the HgBr₂:N₂:He mixture than for the HgBr₂:Xe:He mixture. At the same time, when the maximum value of the average radiation power was reached, mixtures with added nitrogen showed more steady behaviour than mixtures with added xenon. Thus, for

the mixture $\text{HgBr}_2:\text{N}_2:\text{He}$ at a ratio of the gas components $\text{N}_2:\text{He} = 1:4$, after reaching the maximum value, the radiation power during $8 \cdot 10^5$ pulses decreased by no more than 10 %. In addition to studying the radiative characteristics of the component composition of the working mixture, measurements were made of the dependency of the average radiation power on the pumping voltage and the repetition rate of the pulses. With an increase in the energy stored on the capacitance of 0.06 nF dielectric (quartz glass) from 0.13 mJ/cm^3 to 0.34 mJ/cm^3 , the average radiation power increased proportionally. This also occurs when the repetition rate of pulses varies from 400 to 1,900 Hz. Saturation of power in the frequency of the investigated range was not observed. For the mixture $\text{HgBr}_2:\text{N}_2:\text{He}$ at a ratio $\text{N}_2:\text{He} = 1:3$, a total pressure of

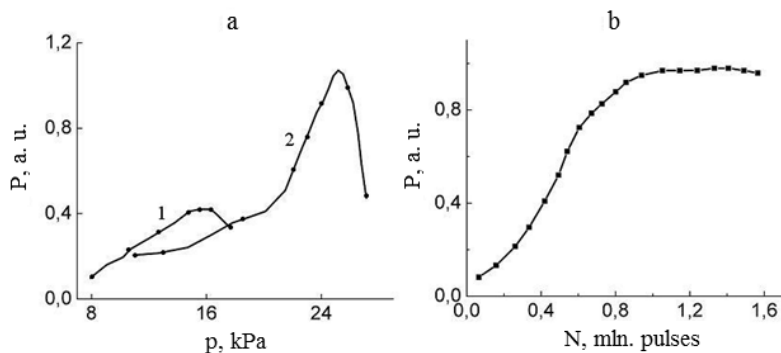


Figure 1.3. a) The dependency of the average radiation power on the partial pressure: 1 - xenon and 2 - nitrogen in the mixtures $\text{HgBr}_2:\text{Xe}:\text{He}$ and $\text{HgBr}_2:\text{N}_2:\text{He}$, respectively; b) the dependency of the average radiation power on the total number of pulses in the $\text{HgBr}_2:\text{N}_2:\text{He}$ (ratio $\text{N}_2:\text{He} = 1:4$). The total pressure of the gas components was 121.6 kPa, the repetition rate of the pump pulses was 1,000 Hz [27].

121.6 kPa and a repetition rate of 1,900 Hz, an average radiation power of 6.8 mW was achieved. When pumping a gas-discharge tube with only a barrier discharge, the average radiation power was less than 1.5 times the value achieved when pumping with a combined (barrier and surface) discharge.

1.2. Parameters and characteristics of plasma

Data on the characteristics and parameters of gas-discharge plasma in some mixtures of mercury dibromide vapor and gases were established in [29–32]. As experimental physics does not yet have satisfactory diagnostic methods for dense gas-discharge plasma, studies in this direction rely heavily on theoretical methods. The kinetic Boltzmann equation for electrons was used to find the electron energy distribution function (EEDF) [1, 48]. In determining the EEDF, it was assumed that under the experimental conditions, the duration of the pump pulses was longer than the time required to set the electron energy distribution function in the plasma. The plasma medium was spatially homogeneous and characterized by the constancy of the composition of all its components. Taking into account these features, which in most experiments are justified, allowed us to use the quasi-stationary Boltzmann equation for electrons in numerical calculation of the EEDF.

Figure 1.4a shows the results of numerical calculation of the EEDF for a mixture of mercury dibromide vapor with helium [32]. In this paper, it is noted that the maximum value of Boltzmann's EEDF shifted to a region of high electron energies in comparison to Maxwell's maximum of EEDF. The energy distribution function of electrons occupies a narrow energy range for small values of the parameter E/p , and when it increases, it expands.

According to the calculated EEDF, the following were determined for gas-discharge plasma in helium (neon) and mercury dibromide vapor mixtures: transport and energy characteristics; electron drift velocity; mean electron energy; specific powers in the elastic and inelastic electron scattering channels for helium (neon) atoms; and dissociative excitation and ionization of mercury dibromide molecules. The drift velocity of the electrons in the mixtures studied increased linearly from 10^6 to 10^8 cm/s with an increase in the parameter E/p in the range $1\text{--}30$ V·cm⁻¹·mm Hg⁻¹. In the parameter region $E/p = 1\text{--}15$ V·cm⁻¹·mm Hg⁻¹, the electron drift velocity for the plasma in a HgBr₂:Ne mixture was greater than for the HgBr₂:He mixture; in the parameter range $E/p = 15\text{--}30$ V·cm⁻¹·mm Hg⁻¹, their values practically coincided [32]. The average electron energies in the same range of changes in the parameter E/p also increased from 4 eV to 12.5 eV. In the range of the parameter $E/p = 5\text{--}30$ V·cm⁻¹·mm Hg⁻¹, mean electron energies for a mixture of mercury dibromide vapor with helium were higher than those in mixtures of mercury dibromide vapor with neon [32]. Fig. 4b presents the dependencies of the specific power losses in the process of electron collision in gas-discharge plasma in mixtures of mercury dibromide vapor with helium at a different ratio of the partial pressures of the components in the value of the parameter E/p [30, 32].

In the discharge in the first mixture at $E/p = 1$ V·cm⁻¹·mm Hg⁻¹, 90 % of the power was lost to elastic collision (heating of the gas). In a mixture of mercury dibromide vapor with neon, this value was 15 % [32]. The dissociative excitation and ionization of mercury dibromide molecules consumes 20–35 % of the discharge power in the parameter range $E/p \sim 1\text{--}4$ V·cm⁻¹·mm Hg⁻¹. With an increase in the parameter E/p , losses entail in the processes of excitation and ionization of inert gas atoms from the ground

state, while part of the power that is transferred to the mercury dibromide molecules and to the helium and neon atoms in elastic and stepwise processes is reduced. The inelastic processes in the plasma mixtures with neon lose several times more power in the range of the changes in the values of the parameter E/p in comparison to mixtures of mercury dibromide vapor with helium [32]. For the mixture $\text{HgBr}_2:\text{Ne} = 0.35\%:99.65\%$, which was used as a working mixture in the active element of an HgBr laser [30], discharge power losses in the dissociative excitation of HgBr (B) at the maximum of its value were $\sim 8\%$. With an increase in the parameter E/N (≥ 4 Td), this power gradually decreased. The power that is deposited in other processes also decreased with the increase of the parameter E/N , with the exception of the power for ionization processes and general electronic excitation. This regularity is related to the dependency of the effective cross section of the processes acting on the electron energy. The difference in the dependency of the specific power losses of discharge power on these processes can be explained by the introduction into the numerical calculation of more effective processes of inelastic cross-sections, namely, the vibrational excitation of mercury dibromide molecules and their attachment. Fig. 1.4c presents data on the dependencies of the rate constants of the elastic and inelastic processes of electron collisions with molecules and atoms in mixtures of mercury dibromide vapor with helium and neon in the parameter E/p , which were determined in [31, 32]. They are a quantitative measure of the efficiency of these processes [1, 3]. The rate constants of processes for both mercury dibromide molecules and for helium and neon atoms increase with an increase in the parameter E/p . In the dissociative excitation and ionization of HgBr_2 molecules, they increase by one to two orders of magnitude when the parameter E/p changes from 1 to $30 \text{ V}\cdot\text{cm}^{-1}\cdot\text{mm Hg}^{-1}$. In the parameter region $E/p \sim 2\text{--}3 \text{ V}\cdot\text{cm}^{-1}\cdot\text{mm Hg}^{-1}$,

where a greater contribution of the discharge power to these processes can be observed, the constants depend weakly on the kind of inert gas. The highest value of the constant ($10^{-7} \text{ cm}^3/\text{s}$) was obtained for excitation of the D-state of HgBr_2 molecules (sums of states above the excitation threshold of 7.9 eV) [31]. For $E/p < 2 \text{ V}\cdot\text{cm}^{-1}\cdot\text{mm Hg}^{-1}$ in a mixture with neon, the rate

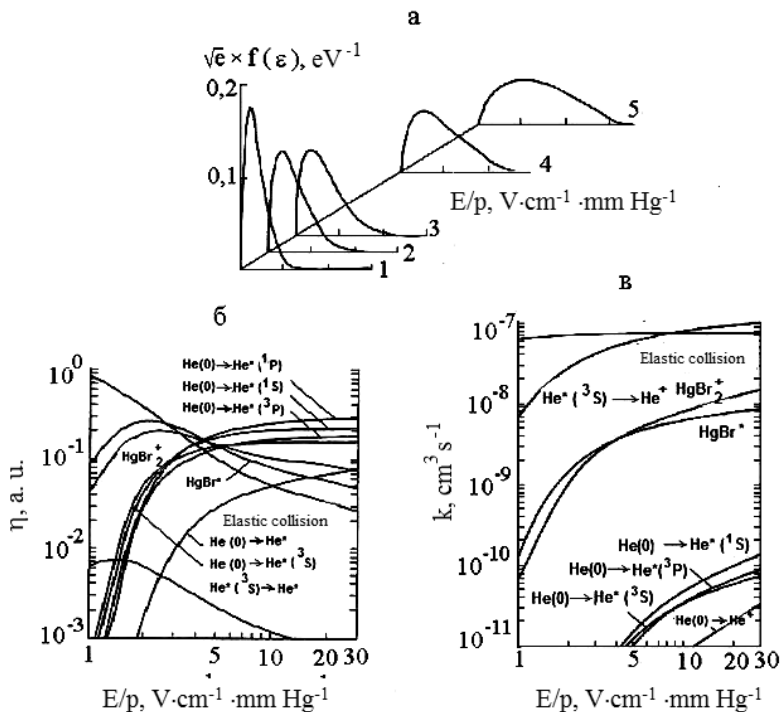


Figure 1.4. a). The electron energy distribution functions for a mixture of $\text{HgBr}_2:\text{He} = 0.5:99.5$; b). Specific power losses from the processes of electron collision with mercury dibromide molecules and helium in the mixture: a) $\text{HgBr}_2:\text{He} = 0.5:99.5$; c). The rate constants of the excitation and ionization processes in the mixture: a) $\text{HgBr}_2:\text{He} = 0.5:99.5$ [32].

constants are of great importance. The rate constants of the processes of elastic scattering of electrons with inert gas atoms and ionization are characterized by high values of $\sim 10^{-8}$ – 10^{-7} cm³/s, which are associated with higher effective cross-sections of these processes [32].

The numerical calculation of the electron distribution functions in gas-discharge plasma in three component mixtures of mercury dibromide vapor and neon with 10 % additions of either nitrogen or xenon are presented in [3]. The value of the E/N parameter was chosen to provide approximately equal mean electron energies for these mixtures. In a mixture with nitrogen, the electron energy distribution was depleted in the energy range 2–5 eV. This is due to the fact that in this range the vibrational excitation cross-sections have high values (above 10^{-16} cm²). In plasma mixed with Xe in this electron energy range, there is an excess of electrons, and for energies exceeding the 8.3 eV excitation threshold of the Xe (³P₂) state a depletion in the distribution is observed. These features have a strong influence on the electron energy distribution function by changing the E/N parameter and, accordingly, on the excitation efficiency of mercury dibromide molecules in the plasma in these mixtures [3].

1.3. Conclusions to Chapter 1

An analysis of the literature on the processes in gas-discharge plasma of mercury dibromide vapor shows the following. The optical characteristics and parameters of plasma in mixtures of mercury dibromide vapor with gases were investigated for a limited set of compositions of working mixtures. There is no data on these characteristics for the pulse-periodic and sinusoidal modes of pumping working mixtures for pulse repetition rates above 2,000 Hz and for short pump pulse durations (at the level of exciplex molecule lifetimes). The possibility of the radiation of

several exciplex molecules simultaneously, which can be formed in gas-discharge plasma in the mixtures studied, has not yet been elucidated. The parameters and characteristics of the plasma for most working mixtures has not yet been clarified. There is no data on the optical characteristics in devices of small dimension (with a radiation area of $<10 \text{ cm}^2$), which are necessary for exciplex sources in a number of practical applications.

These circumstances determined the direction of our work.

CHAPTER 2

TECHNIQUE AND EXPERIMENTAL METHODS

The optical characteristics of plasma in mixtures of mercury dibromide vapor and gases were investigated through barrier discharge (dielectric-quartz glass) [16]. The most important characteristic of this discharge is that the conditions for obtaining a non-equilibrium plasma can be provided at increased (atmospheric) pressures. In a barrier discharge, this can be achieved more simply than in other alternative methods, such as low-pressure discharges, high-pressure pulse discharges, or through the injection of an electron beam into a gas. It provides flexibility in relation to geometry, working media and plasma parameters. Ozone was first produced with the help of a barrier discharge, as well as powerful coherent infrared radiation from a CO₂ laser; and incoherent UV, VUV, and visible radiation from excimer and exciplex molecules [16, 49].

2.1. Experimental installations

Investigation into barrier discharge characteristics was carried out using three experimental installations, the block diagrams of which are shown in figures 2.1–2.3.

The structural elements of the installations are designed to take into account the peculiarities of the experimental conditions with a discharge that is formed in mixtures of mercury dibromide vapor and gases when a high-voltage pulse-periodic voltage or a sinusoidal voltage is applied to the

working electrodes with frequencies of 5 Hz, 3–9 kHz and 120 kHz, respectively; interference in the range (5 Hz–150 MHz); and salt vapor and gases of working mixtures at pressures of 70–200 kPa.

The main units of the first experimental installation (Figure 2.1) are: a gas discharge cuvette (GDC); a vacuum pumping and gas filling system (VPGFS); a high voltage (HVG) generator (pulse-periodic voltage); and a system for recording the optical and electrical discharge characteristics. The generator allows the reception of pulses with an adjustable voltage amplitude in the range 3–10 kV, a duration of 400–600 ns, and a pulse repetition rate of $f = 3,000\text{--}9,000$ Hz. The registration system consists of: a diffraction monochromator SD 7 (with a grating of 600 lines/mm and spectral resolution of the recording system at 2.4 nm); photodetectors (FD); photoelectronic multipliers FEU-106 and 14 ELU-FS; an electrical signal amplifier (A)—U5-9; spectra recorder (SP)—KSP-4; a digital voltmeter (V)—S-4300; an oscilloscope (O)—C1-72 (or C7-10A); a Rogovsky coil (RC); a radiation power meter (PM)—Quartz-01; and a light filter (F)—SZS-16.

The second experimental setup (Figure 2.2) was designed to study sinusoidal optical and electrical signals at high frequency. Its main units are: a high voltage generator with a sinusoidal form of the output voltage; a monochromator—Jobin Yvon FHR 1000 (grating 2,400 lines/mm and spectral resolution of the recording system at 0.08 nm); a photodetector—a high-speed CCD solid-state matrix; a personal computer; a digital oscilloscope—LeCroy WaveRunner 6100A; and a power meter—Newport 1918-C. The generator made it possible to change the amplitude and frequency of the sinusoidal voltage up to 7 kV and 130 kHz, respectively.

The third experimental setup (Figure 2.3) consisted of: a high-voltage generator (FID FPG 10-1MKS20); a coaxial cable (RG213, $R = 50 \Omega$);