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MECHANISM OF GROWTH OF THE INTENSITY OF RADIATION EMITTED IN THE BLUE-VIOLET SPECTRAL INTERVAL BY GAS-DISCHARGE PLASMA GENERATED IN THE MIXTURES OF MERCURY DIIODIDE VAPOR, XENON, AND NEON

A mechanism allowing the intensity of radiation emitted in the blue-violet spectral interval by gas-discharge plasma created in the mixtures of mercury diiodide vapor, xenon, and neon to be increased in comparison with the intensity of radiation from gas-discharge plasma in the mixtures of mercury diiodide vapor and neon is established. The plasma parameters and the reduced electric field, at which the specific discharge power spent for the excitation of mercury monoiodide exciplex molecules is maximum, are determined. The research results can be used for the creation of a more efficient exciplex lamp with bands emitted in the blue-violet spectral interval.

Keywords: gas-discharge plasma, radiation emission by exciplex molecules, plasma parameters, mercury diiodide, xenon, neon.

1. Introduction

Gas-discharge plasma generated in the mixtures of mercury diiodide vapor with various gases is a working medium of exciplex sources of coherent and spontaneous radiation (lasers and excilamps) in the blue-violet spectral interval, with the corresponding intensity maxima being located at the wavelengths $\lambda =$ 441.4, 443, 444, and 445 nm [1–7]. Those sources can be used in scientific researches, photonics, biotechnology, medicine, in the manufacture of gas-discharge display panels, as well as to provide an effective light-assisted control over the processes of photosynthesis, plant growth and development, and phytocenosis [8–13].

Unlike other luminescent lamps and thermal sources, excilamps possess a number of advantages. In particular, this is their radiation emission spectrum. Up to 90% and more of the total emitted power can be concentrated in a rather narrow (the half-width is less than 10 nm) spectral band of a mercury monoiodide exciplex molecule. The corresponding specific emission powers exceed the magnitudes typical of low-pressure lamps based on resonance transitions in atoms [8–10, 14].

In the last decade, light-emitting diode lamps emitting in the blue-violet spectral interval obtained wide applications. They have the highest light efficiency (~100 lm/W) among other light sources. However, the application of powerful (>100 W) light-emitting diode lamps is restricted, because of the necessity to cool them down in order not to lose their productivity [15]. Excilamps emitting in the visible spectral interval have no such a restriction, because their emitting surface can be scaled without losing its specific energy parameters [8, 9].

In our research [14], we revealed the simultaneous emission of mercury monoiodide exciplex molecules in the blue-violet and ultra-violet spectral intervals ($B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$, $C^2\Pi_{1/2} \to X^2\Sigma^+_{1/2}$), as well as the emission of xenon iodide ($B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$, $D_{1/2} \to A^2\Pi_{1/2}$) and iodine ($D' \to A'$) in gas-discharge plasma of the barrier discharge created in the mixture of mercury diiodide vapor, xenon, and neon. We also found that the most intense radiation emission is observed in the spectral band ($B \to X$) with a power maximum at the wavelength $\lambda = 443$ nm, whose intensity is 1.6 times higher than that in plasma in the mixture of only mercury diiodide vapor and neon. The creation of efficient exciplex sources with the simultaneous emission in the blue-violet and ultra-violet spectral intervals can be used for activating the photosynthesis process with the simultaneous destruction of viruses and bacteria. It is also important for the solution of engineering problems dealing with the preservation of energy resources on our planet and the improvement of "artificial" photosynthesis [16–18].

The researches performed by us in work [14] did not adequately elucidate, why the intensity of radiation emission increases in the spectral band of the electron-vibration transition $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$ in mercury monoiodide exciplex molecules, which produces radiation in the blue-violet spectral interval. This issue stimulated further researches aimed at establishing the mechanism that is responsible for the increase of the radiation intensity emitted by plasma generated in the mixture of mercury diiodide vapor, xenon, and neon in the blue-violet spectral interval. This mechanism was determined theoretically; namely, by comparing and classifying the data concerning the plasma parameters and the results of experimental researches.

2. Method of Plasma Parameter Determination

The experimental physics still has no satisfactory diagnostic methods for dense gas-discharge plasma. Therefore, the parameters of barrier discharge plasma were determined numerically on the basis of electron energy distribution functions (EEDFs) in discharge [19]. The EEDFs were found by solving the corresponding kinetic Boltzmann equation in the binomial approximation with the use of the known code "Bolsig+" [20]. The obtained EEDFs were applied to the calculation of the average energy and mobility of electrons, specific losses of discharge power, and the rate constants for elastic and inelastic electron scatterings by mercury diiodide molecules and neon atoms, as well as their dependences on the magnitude of reduced electric field E/N, where E is the electric field strength, and N the total concentration of components in the gas mixture. The variation interval for this parameter, $E/N = 1 \div 100$ Td ($1 \times 10^{-17} \div 1 \times 10^{-15}$ V cm^2), included values that were realized in our experiment.

All calculations were carried out for the partial pressures of mixture components, at which the maximum in the radiation emission intensity was obtained experimentally [14]. In particular, these were partial pressures of 0.6 kPa for mercury diiodide vapor and 110 kPa for neon in the case of the mixture "mercury diiodide + neon" and partial pressures of 0.7 kPa for mercury diiodide vapor, 10 kPa for xenon, and 100 kPa for neon in the case of the mixture "mercury diiodide + xenon + neon".

The following elementary processes made allowance for in the collision integral of electrons with neon atoms and mercury diiodide molecules: elastic scattering by xenon atoms; excitation of the energy levels of xenon atoms with threshold energies of 3.4, 8.31, 8.44, 9.69, 10.0, 11.0, and 11.7 eV; ionization of xenon atoms; elastic scattering by neon atoms; excitation of the energy levels of neon atoms with threshold energies of 16.62, 16.67 ($1s^1$), 16.84 ($1s^2$), 18.72 ($2p$), 20.0 ($2s + 3d$), 20.65 ($3p$), and 4.9 eV; ionization of neon atoms; ionization of mercury diiodide molecules; and dissociative excitation of the $B^2\Sigma^+_{1/2}$ electron state of monoiodide mercury. The absolute values of the effective cross-sections of those processes, as well as their dependences on the electron energy were taken from works [21, 22].

The magnitude of the reduced electric field E/N, at which the experimental researches of work [14] were carried out, was calculated using the formulas presented in work [23]. The corresponding values are: 46 Td for the mixture HgI_2–Ne and 49 Td for the mixture HgI_2–Xe–Ne.

3. Results of Plasma Parameter Research

In Fig. 1, typical forms of the EEDF are shown for various values of the parameter E/N from the interval 1–100 Td. The increase of this parameter gives

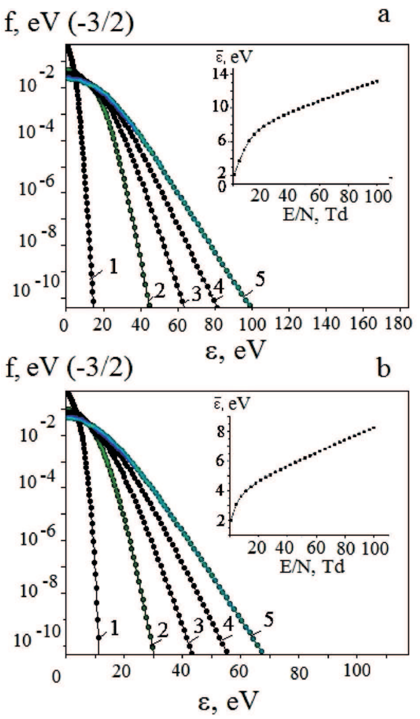

Fig. 1. Energy distribution functions of electrons in a discharge in the mixtures HgI_2(0.54%) + Ne(99.46%) (*a*) and HgI_2(0.64%) + Xe(9.03%) + Ne (90.33%) (*b*) for various parameter values $E/N = 1$ (*1*), 25.8 (*2*), 50.5 (*3*), 75.3 (*4*), and 100 (*5*). The corresponding dependence of the average electron energy on E/N is shown in the inset

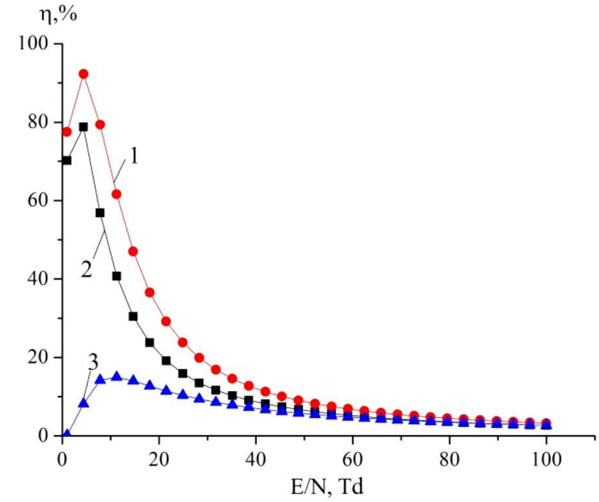

Fig. 2. Dependences of the specific losses of discharge power spent on the electron-impact dissociative excitation of the $B^2\Sigma^+_{1/2}$ state of a mercury monoiodide molecule in discharge plasma generated in the HgI_2(0.64%) + Xe(9.03%) (*1*) and HgI_2(0.64%) + Xe(9.03%) + Ne (90.33%) (*2*) mixtures, and on the excitation of the metastable $^3P_2$ state of xenon atoms on the parameter E/N (*3*)

rise to a growth in the number of "fast" electrons in the discharge and a reduction of the electron concentration in the operational interval of an emitter. The electron energy maxima in plasma in the HgI_2–Ne (Fig. 1, *a*) and HgI_2–Xe–Ne (Fig. 1, *b*) mixtures at $E/N = 100$ Td were equal to 114 and 76.2 eV, respectively.

In the case of HgI_2–Ne mixture, the strongest dependence of the average energy of electrons in discharge plasma on the parameter E/N takes place in the interval 1–14.7 Td, where it linearly increases from 2.1 to 6.8 eV. In the interval $E/N = 14.7\div100$ Td, this parameter also increases from 6.8 to 13.1 eV, but at a lower rate. A slower growth of the average electron energy in this interval of the parameter E/N is a result of the energy losses spent by fast electrons for the excitation of the energy states of mercury diiodide molecules and neon atoms.

For the discharge in the HgI_2–Xe–Ne mixture, the average energy of electrons in plasma depends most strongly on the parameter E/N in the interval 1–11 Td. In this case, it linearly increases from 2.0 to 4.0 eV. Within the interval $E/N = 11\div100$ Td, this parameter increases from 4.0 to 8.2 eV, but also at a lower rate. A slower growth of the average electron energy in this interval of the parameter E/N is associated with the energy losses of fast electrons spent for the excitation of the energy states of mercury diiodide molecules, as well as xenon and neon atoms.

For plasma created in the HgI_2–Ne mixture, the average electron energy amounted to 9.8 eV at an experimental reduced electric field of 46 Td. For plasma in the HgI_2–Xe–Ne mixture, the corresponding values were 6.0 eV and 49 Td.

A decomposition of the specific losses of discharge power into the components spent on main elementary processes in the interval of reduced electric field strength $E/N = 1\div100$ Td is depicted in Fig. 2. The specific losses of discharge power spent on the process of dissociative excitation of the $B^2\Sigma^+_{1/2}$ state of mercury monoiodide molecules increase with the parameter E/N. At $E/N = 44$ Td , they reach maximum values of 92% and 79% for plasma created in the HgI_2–Ne and HgI_2–Xe–Ne mixtures, respectively. The further growth of the parameter E/N results in their reduction. The specific losses of discharge power on the excitation of the metastable $^3P_2$ state of xenon atoms (Fig. 2, curve *3*) have a similar dependence on the reduced electric field strength and reach a maximum of 15% at $E/N = 11.2$ Td. For the processes concerned, the magnitude of the specific losses of discharge power and the rate of their increase or reduction are connected with the magnitudes of the effective cross-sections of energy states, the character of their dependences on the electron energy, the behavior of EEDF at various E/N-values, and the threshold energies of dissociative excitation of mercury monoiodide molecules and xenon atoms. For the process of dissociative excitation of the $B^2\Sigma^+_{1/2}$ state of mercury monoiodide, the specific losses of discharge power amount to 10% in plasma generated in the HgI_2–Ne mixture at the reduced electric field $E/N = 46$ Td and to 7% in plasma in the HgI_2–Xe–Ne mixture at $E/N = 49$ Td.

In Fig. 3, the results of numerical calculations obtained for the rate constants of the processes of electron-impact dissociative excitation of the $B^2\Sigma^+_{1/2}$ state of mercury monoiodide molecules (curves *1* and *2*) and the excitation of the metastable $^3P_2$ state of xenon atoms (curve *3*) are exhibited. The rate constants for the former process change within an interval from 1×10^{-16} to 3.5×10^{-14} m^3/s for the parameter E/N varying from 1 to 100 Td. For the reduced electric fields $E/N = 46$ (HgI_2–Ne) and 49 Td (HgI_2–Xe–Ne), the rate constants for the electron-impact dissociative excitation of the $B^2\Sigma^+_{1/2}$ state of mercury monoiodide molecules equal 2.2×10^{-14} and 1.2×10^{-14} m^3/s, respectively. Hence, these plasma parameters are slightly different, depending on the gas mixture composition (HgI_2–Ne or HgI_2–Xe–Ne).

Therefore, other elementary processes have to be considered, which could explain a substantial increase of the radiation intensity emitted by a HgI molecule ($B \to X$) in plasma created in the mixture of mercury diiodide vapor with xenon and neon in comparison with that for the mixture without xenon. In particular, it can be the energy transfer to mercury diiodide molecules at their collisions with xenon atoms in the metastable $^3P_2$ state,

$$\mathrm{HgI_2} + \mathrm{Xe}(^3P_2) \to \mathrm{HgI}(C^2\Pi_{1/2}, D^2\Pi_{3/2}) + $$
$$+ \mathrm{I}(^2P_{3/2}) + \mathrm{Xe}, \qquad (1)$$

as well as the quenching of the $C^2\Pi_{1/2}$ and $D^2\Pi_{3/2}$ states of a mercury monoiodide molecule by xenon

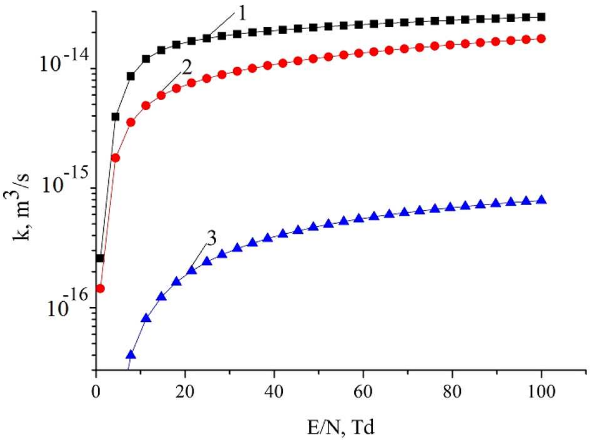

Fig. 3. Dependences of the rate constants of dissociative excitation of the $B^2\Sigma^+_{1/2}$ state of mercury monoiodide molecule by electrons in discharge plasma generated in the $HgI_2(0.64\%)$ + + Xe(9.03%) (*1*) and $HgI_2(0.64\%)$ + Xe(9.03%) + Ne (90.33%) (*2*) mixtures, and the rate constant of excitation of the metastable $^3P_2$ state of xenon atoms on the parameter E/N (*3*)

atoms with the radiationless transition onto the $B^2\Sigma^+_{1/2}$ state,

$$\mathrm{HgI}(C^2\Pi_{1/2}, D^2\Pi_{3/2}) + M \to \mathrm{HgI}(B^2\Sigma^+_{1/2}) +$$
$$+ \mathrm{Xe} + \Delta E_{1,2}, \qquad (2)$$

where M is the concentration of molecules or atoms (HgI_2, Xe, Ne) quenching the $C^2\Pi_{1/2}$ and $D^2\Pi_{3/2}$ states of a mercury monoiodide molecule, and $\Delta E_{1,2}$ the energy difference between the $C^2\Pi_{1/2}$, $D^2\Pi_{3/2}$, and $B^2\Sigma^+_{1/2}$ states. Process (2) was revealed in the experiments dealing with photodissociation of mercury diiodide [24, 25], as well as in our experiments under the conditions of barrier discharge in the mixture of mercury diiodide vapor, xenon, and helium [23].

4. Conclusions

Hence, the comparison of the results of our researches obtained for the parameters of gas-discharge plasma created in the HgI_2–Ne and HgI_2–Xe–Ne mixtures with experimental data made it possible to establish the mechanism giving rise to the increase in the HgI ($B \to X$) emission intensity in the barrier-discharge plasma generated in the gas mixture with xenon admixtures. The mechanism consists in the growth of the population of the $B^2\Sigma^+_{1/2}$ state of a mercury monoiodide molecule due to radiationless transitions from higher energy levels $C^2\Pi_{1/2}$ and $D^2\Pi_{3/2}$ at their quenching by xenon atoms.

The researches allowed us to determine the magnitude of the reduced electric field strength, at which the specific contribution of the electric discharge power spent on the excitation of the $B^2\Sigma^+_{1/2}$ state of mercury monoiodide molecules is maximum; namely, $E/N = 4.4$ Td. The result obtained makes it possible to elevate the power parameters of exciplex lamps emitting in the blue-violet spectral interval.

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МЕХАНІЗМ ЗБІЛЬШЕННЯ ІНТЕНСИВНОСТІ ВИПРОМІНЮВАННЯ ГАЗОРОЗРЯДНОЇ ПЛАЗМИ НА СУМІШАХ ПАРІВ ДИЙОДИДУ РТУТІ, КСЕНОНУ ТА НЕОНУ В ФІОЛЕТОВО-СИНЬОМУ СПЕКТРАЛЬНОМУ ДІАПАЗОНІ

Р е з ю м е

Встановлено механізм збільшення інтенсивності випромінювання газорозрядної плазми на сумішах парів дийодиду ртуті, ксенону та неону в порівнянні з сумішшю парів дийодиду ртуті і неону в фіолетово-синьому спектральному діапазоні. Встановлені параметри плазми, величина приведеного електричного поля, при якому питома потужність розряду, що вноситься в збудження ексиплексних молекул монойодиду ртуті, максимальна. Результати досліджень можуть бути використані для створення більш ефективної ексиплексної лампи, що випромінює спектральні смуги в фіолетово-синьому діапазоні.