Third continuum in argon and krypton in the case of pumping by an electron beam and a discharge initiated by the electron beam

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The experimental results on pumping of Ar and Kr inert gases and their mixtures with Ne at different pressure by a pulsed electron beam and a discharge initiated by the electron beam are discussed. They allow the conclusion on the role of metastable atoms and dimer molecules in formation of the states yielding the emission in the molecular ion continuum in the afterglow.

1. Introduction

Starting from the 50's, spectral characteristics of plasma in the inert gases were studied extensively using different ways of excitation mostly because of the presence of intense bands of spontaneous emission in the VUV and UV region.[1,2] Lasing at bounded-free transitions of excited molecules, such as Ar_2^* (126 nm), Kr_2^* (146 nm), and Xe_2^* (178 nm) was obtained in the 70's (Ref. 2). However, the broad-band radiation in dimers (or "second continuum of inert gases") remained the subject of theoretical and experimental investigations. Incidentally, the investigators have noticed the presence of radiation, which was called the third continuum, in the VUV and UV spectra of the inert gases. This radiation has much lower intensity, but a wider spectrum. The nature of this radiation is still to be understood. Langhoff[3] supposed this radiation to be caused by transitions of double-ionized excited molecular systems of the Rg_2^{++} type, which experience radiative decay into $Rg^+ + Rg^+$. The theoretical and experimental studies of the spectral and temporal characteristics of the third continuum of inert gases initiated by different pumping mechanisms and the problem on identifying the origin of this radiation have been considered in detail in Ref. 1 and in a more recent papers, Refs. 4–9. "oichenko et al.[1] have considered some molecular and ion transitions of the inert gases, which were proposed by different authors to explain the emission in the third continuum. "ased on detailed analysis, the most probable candidates were revealed. They were the transitions of molecular ions from the states of Rg_2^{+*} (asymptotically corresponding to the ground state of the Rg^+ ion and the excited state ($^3P_{1,2}$) of the Rg^* atom) to the states of Rg_2^+ (asymptotically corresponding to the ground states of the Rg^+ ion and Rg atom). In the authors' opinion, trimers of these states can also contribute to the emission. The constructed model of Rg_2^{+*} formation and decay was then further developed in Refs. 8 and 9.

The still unclear nature of the broad-band emission from the inert gases can be explained, in particular, by a wide variety of experimental conditions under which this radiation was studied. "esides, when interpreting the results, many authors stated that they observed the transitions from the states of either Rg_2^{+*} or Rg_2^{++}, whereas analysis of the available experimental data shows that the number of peaks and their positions in the spectrum heavily depend on the gas pressure, as well as on the pumping mode and pump power.

The detailed study[6] of spectral and temporal characteristics of the broad-band emission from plasma in the inert gases in a wide pressure range (from 0.1 to 30 atm) in the case of excitation by a pulsed X-ray radiation allowed unambiguous determination of local peaks, their dynamics, and change in response to the changing gas pressure. Robert et al.[6] state that the broad-band emission, or the so-called third continuum, is a complex superposition of a great number of lines caused by transitions of both diatomic and more complex molecular-ion complexes. The assumptions by "oichenko et al.[1] and Langhoff[3] on the influence of more complex molecular-ion complexes emitting in the long-wave wing of the broad-band transitions were ignored when constructing the models. These complexes were taken into account only recently in Ref. 7 in simulating the molecular-ion continuum (MIC) excited by X-rays in the inert gases at high pressure.

This work continues the experimental studies[8,9] aimed at revealing the influence of the electric field on the temporal and spectral characteristics of the broad-band emission from the inert gases. We have conducted the experiments on pumping the Ar and Kr inert gases and their mixtures with Ne at different pressure by a pulsed electron beam and electron-beam-initiated discharge. The results obtained allow the conclusion to be drawn that metastable atoms play a significant part in formation of the states emitting the molecular-ion continuum.

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2. Experimental setup and measurement technique

The working gases were excited by an electron beam and an electron-beam-initiated discharge. An electron accelerator[1] provided the beam current density of 3–4 A/cm^2 behind a foil at the mean electron energy of 150 to 170 eV, pulse duration at halfmaximum of 50 ns, and the output foil window of 42×2 cm in cross section. The discharge was initiated between a metal mesh protecting the foil and an electrode spaced by 4 cm from it. The electrode was set at the opposite side of the chamber. The discharge capacitor C = 100 nF was charged up to the voltage of 15 kV. Oscillograms of the discharge current and the current through a diode of the electron accelerator were recorded with an S8–14 oscilloscope using low-inductance and low-resistance current shunts. The intensity and time behavior of spontaneous emission in the spectral regions isolated with an MDR–12 monochromator were recorded with a FEU–100 photomultiplier tube and an S8–14 oscilloscope. The output window of the discharge chamber was protected with a quartz plate of 3-cm working diameter. Monochromator slits were 150–250 μm (0.36–0.6 nm) wide depending on the intensity of optical signals. Spectral ranges of the broad-band emission free of intense lines and bands of admixtures (N_2, OH, and others) were selected based on analysis of spectrograms recorded with an ISP–30 device in the region of 200–400 nm, as well as the data from Refs. 8–10. The intensity of the emission observed is given in relative units, ignoring the spectral sensitivity of the recording instrumentation.

3. Experimental results and tentative discussion

3.1. Broad-band emission from argon

The oscillograms of optical signals had two maxima in time in the case of pumping by the electron beam and electron-beam-initiated discharge regardless of the experimental conditions in the voltage across the discharge gap and pressure in the working chamber. The first maximum was achieved in 30 ns after the beginning of the current pulse; it corresponded to the time of achieving the maximum energy contribution from the electron beam. The amplitude of the first maximum increased as the gas pressure increased and was practically independent of the discharge voltage. In contrast, the second maximum and the amplitude ratio of the maxima strongly depended on the presence of the electric field (oscillograms in Fig. 1). The highest relative (relative to the main maximum) value of the signal in the afterglow was observed at λ = 190 and 263.6 nm. Note that two maxima in the broad-band emission pulse from argon were also observed at pumping by a pulsed discharge[10] and by an electron beam.[1,11]

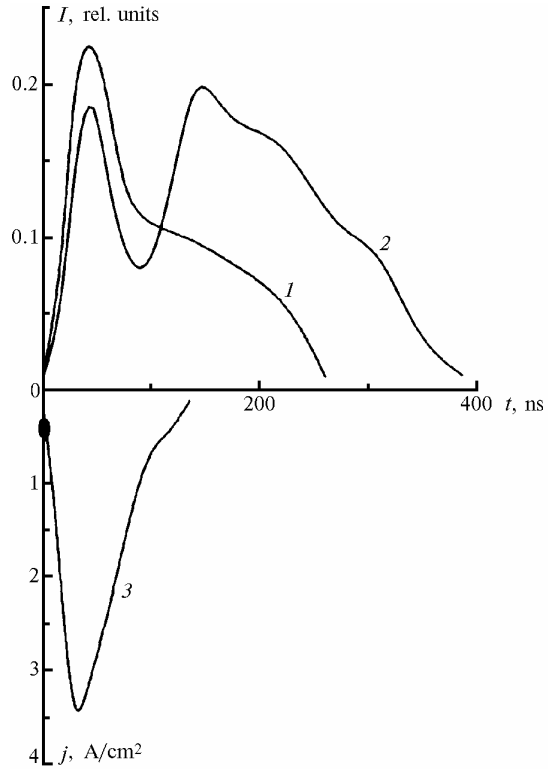

Fig. 1. Oscillograms of spontaneous emission (*1*, *2*) at λ = 190 nm from argon at the pressure of 0.25 atm and the oscillogram of the electron beam current (*3*). Voltage across the discharge gap were: 0 (*1*) and 3 kV (*2*).

”ased on our tentative analysis, the radiation pulse can be divided in time into two periods: one that occurred during the action of the electron beam (first 100 ns) and in the afterglow. Figure 2 shows the energy of spontaneous emission in the afterglow in some bands of the third continuum in argon at different pressure and voltage applied to the discharge gap. At the argon pressure of 1 atm the energy of optical signal in the afterglow increases with the increasing discharge voltage in almost all the ranges of the recorded spectrum, except for λ = 220 nm. The maximum change was noticed in the bands at λ = 200 and 263 nm, where the energy of the afterglow increased by 70–80% at U = 7.5 kV relative to the case of pumping only by the electron beam. At a lower pressure, we can separate two regions in the spectrum of the third continuum of argon, in which the afterglow energy increases significantly. Those are at λ = 180–190 and 263 nm. In the region of λ = 220 nm we observed practically no changes in the afterglow energy. Some increase at the pressure of 0.25 atm in this region is apparently caused by the influence of the band at λ = 263 nm.

Adding neon to argon in the case of pumping by the electron beam resulted in suppression of the broad-band emission from argon.[8] In this case, the band in the region of 180–190 nm exhibited the lowest sensitivity to the influence from neon. Pumping of argon mixtures with neon by a semi-selfmaintained discharge gives similar results.

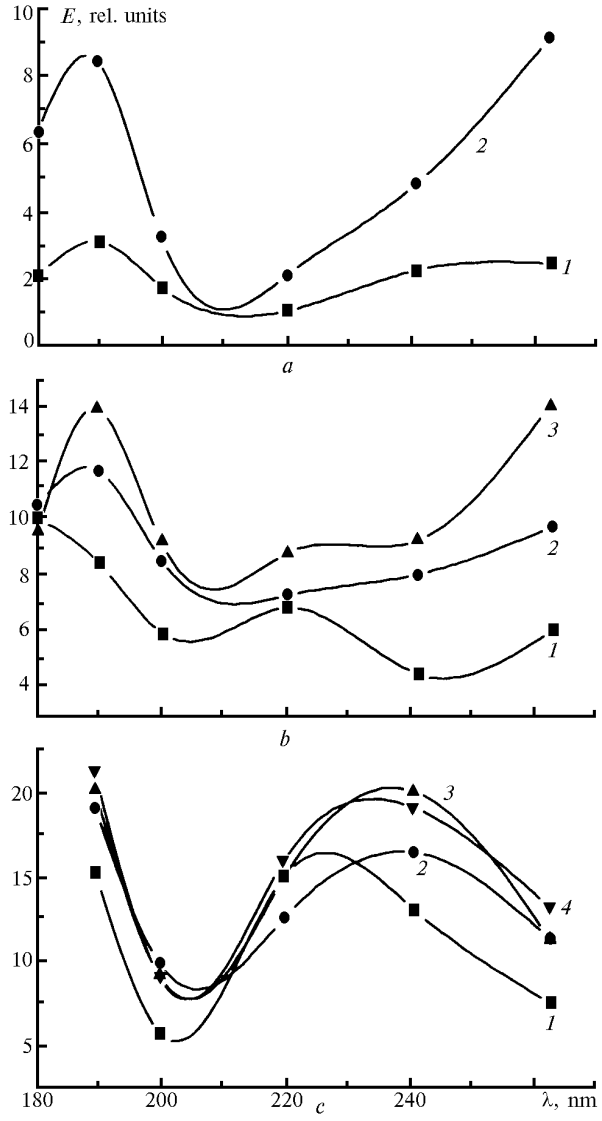

Fig. 2. Energy of spontaneous emission from the afterglow in some bands of the molecular-ion continuum in argon at the pressure of 0.25 (*a*), 0.5 (*b*), and 1 atm (*c*) and the voltage across the discharge gap: 0 (*1*), 3 (*2*), 5 (*3*), and 7.5 kV (*4*).

Figure 3 shows the dependences similar to those in Fig. 2 but for the mixtures of argon (0.5 atm) with neon (30, 150 Torr, and 0.5 atm, respectively). With small additions of neon, the energy of the afterglow increases only slightly in the spectral region of 220–260 nm, in the absence of the discharge voltage. The increase in the neon content results in suppression of the afterglow within the entire spectral region of the third continuum of argon, while the emission in the regions of 190 and 260 nm remains significant even at the argon–neon ratio of 1:1. Application of the discharge voltage in the case of neon content in the mixture up to 150 Torr results in relatively more sharp increase of the afterglow energy in the region of λ = 190 and 260 nm than in the case of pure argon. However, at equal contents of argon and neon, some decrease of the energy in the region of λ = 190 nm is observed, and the increase of energy in the afterglow slows down in the region of 260 nm (Fig. 3*c*). The medium part of the argon broad-band emission spectrum (200–240 nm) is almost completely suppressed at high concentrations of neon in the mixture, and application of the voltage to the discharge gap does not change the energy of the afterglow in this case.

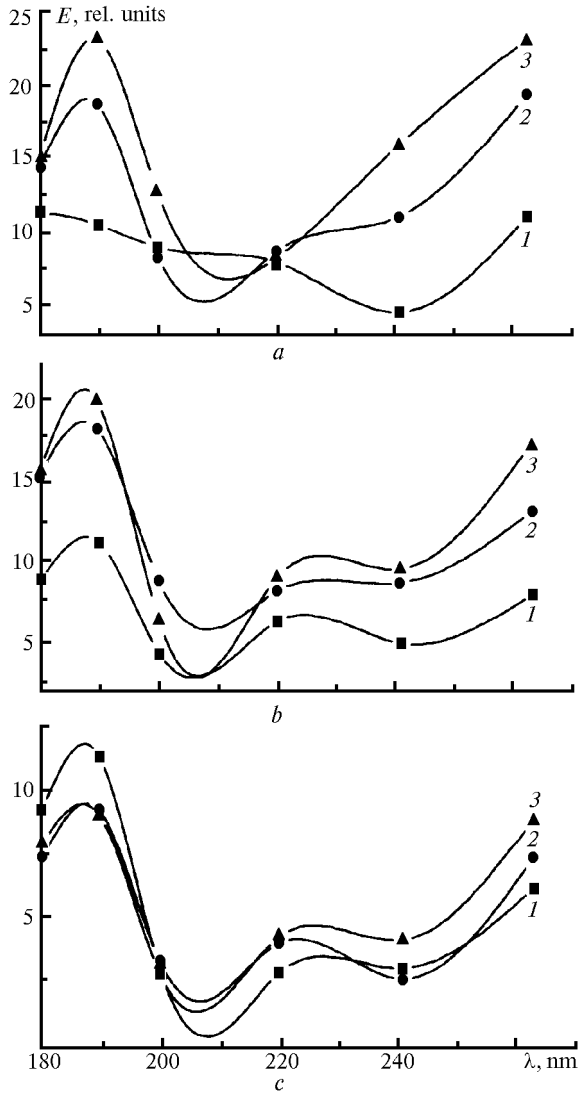

Fig. 3. Energy of spontaneous emission from the afterglow in some bands of the molecular-ion continuum of argon at the pressure of 0.5 atm and addition of neon: 30 Torr (*a*), 150 Torr (*b*), and 0.5 atm (*c*); voltage across the discharge gap: 0 (*1*), 3 (*2*), 5 (*3*), and 7.5 kV (*4*).

3.2. Broad-band emission from krypton

Similar properties have also been observed in the broad-band continuum of krypton. The oscillogram of spontaneous emission from krypton in the spectral region of 200–320 nm had the main peak corresponding to the pulse of excitation by the electron beam and afterglow under the experimental conditions.

Application of the electric field changed the intensity and duration of the afterglow, while practically not affecting the main pulse. The results of analysis of afterglow of the krypton broad-band emission at different pressure and voltage across the discharge gap are shown in Fig. 4.

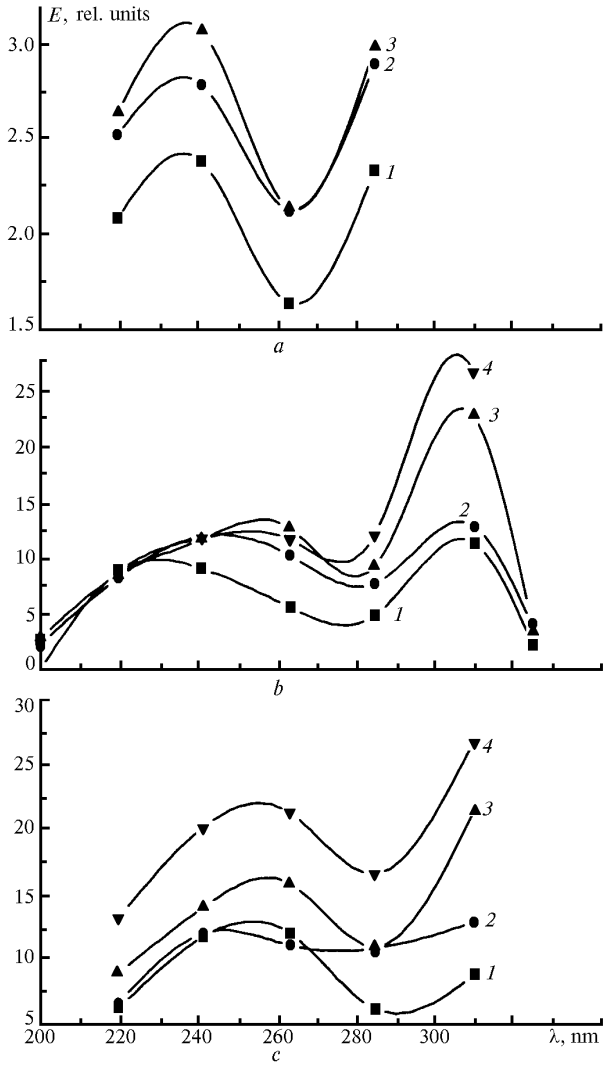

Fig. 4. Energy of the spontaneous emission from the afterglow in some bands of the molecular-ion continuum in krypton at the pressure of 0.25 (*a*), 0.5 (*b*), and 1 atm (*c*) and voltage across the discharge gap: 0 (*1*), 3 (*2*), 5 (*3*), and 7.5 kV (*4*).

It should be noted that the most intense peak in the region of 310 nm (Fig. 4*b*) corresponds to the afterglow of OH and $n\,m^+$ molecules[11] and strongly distorts the pattern of behavior of the krypton continuum in the neighboring spectral regions. In contrast to argon, there are no pronounced regions of the broad-band spectrum of krypton, the intensity of afterglow in which changes steadily with the gas pressure increase in the case of applying the electric field. This is likely connected with a high retarding ability of krypton and, correspondingly, with varying (in response to the pressure) homogeneity of preionization by the electron beam, as well as with a stronger influence of triatomic molecular ions on the emission spectrum. In krypton the ratio between the diatomic and triatomic molecular ions is about 10:90 already at the pressure of 1 atm, whereas in the case of argon this ratio is just the opposite at the pressure of 2 atm: 90:10 (see Ref. 12). At the krypton pressure of 0.25, 0.5, and 1 atm, the sharpest change in the intensity of MIC afterglow under application of the electric field was observed in the regions of 220–260 (maximum at 235 nm), 240–280 (maximum at 260 nm), and 220–300 nm (maximum at 260 nm). Thus, the region of sharpest changes shifts toward sorter waves as the pressure decreases.

4. Discussion and conclusions

The obtained experimental results, together with the results of previous studies,[8,9] support the conclusion drawn in Ref. 7 that the so-called third continuum is a complex superposition of many bands belonging to both diatomic ions and more complex molecular-ion complexes. So, the more correct name for it is the molecular-ion continuum. Appearance, disappearance, width, and intensity of these bands can change depending on the conditions of excitation, pressure, and presence of admixtures.

Since the main maximum keeps almost unchanged at application of the electric field, the principal attention is paid to the emission observed after termination of the beam current. In Ref. 11, at excitation of the inert gases by a pulsed electron beam of 5-ns duration, the presence of a narrow intense peak in the optical signal was also observed. The peak manifested itself all over the spectral region under study, in contrast to the long afterglow, which was observed only in the region of the third continuum. "ased on these findings, Amirov et al.[11] assumed that the short intense peak corresponds to the emission due to transitions of atomic ions. The energy of this emission is insufficient for reliable recording on a photographic film. The third continuum, in its turn, is emitted mainly in the afterglow. In our case, this explanation does not explain the time behavior observed, because in the regions with only broad-band spectrum the energy in the first peak is often higher than or equal to the energy of the afterglow. Moreover, in the region of 180–190 nm at the argon pressure below 0.5 atm the first peak (first 100 ns corresponding to the full length of the electron beam) was not observed under the experimental conditions.

Consider the possible causes of the increase in the intensity of MIC of inert gases in afterglow in the case of pumping by electron-beam initiated discharge. Application of a weak electric field under the conditions of our experiment resulted in a relatively small increase in the temperature of plasma electrons and in significant changes in the intensity and duration of MIC afterglow at

some spectral regions. It is reasonable to conclude that afterglow cannot be caused by double-charged molecular ions, but belongs to single-charged molecular ions of the Rg_2^{+*} type. Therefore, we use the model from Ref. 1. According to this model (for designations see Ref. 1), the $Rg_2^{+*}(1)$ states emitting in the MIC region can be populated through two channels [$Rg_2^{+*}(1)$ is the state which asymptotically corresponds to the ground state of the ion and the excited state of the atom, and $Rg_2^{+*}(2)$ to the state which asymptotically corresponds to the ground state of the atom and the excited state of the ion of Rg^{+*}]:

1. The first channel is the ionization with excitation:

$$Rg + e_p \rightarrow Rg^{+*} + e_p + e, \quad (1)$$

where e_p is for electrons of the pump beam, and the following transition to $Rg_2^{+*}(2)$:

$$Rg^{+*} + 2R \rightarrow Rg_2^{+*}(2) + Rg, \quad (2)$$

and mixing with electrons or gas to the Rg_2^+ (1) state:

$$Rg_2^{+*}(2) + e(Rg) \rightarrow Rg_2^{+*}(1) + e(Rg). \quad (3)$$

2. The second channel starts from conversion of the ion and excited atom:

$$Rg^+ + Rg^* + Rg \rightarrow Rg_2^{+*}(1) + Rg. \quad (4)$$

In Ref. 1 the efficiency of these channels during the action of the electron beam was estimated, and it was shown that at high pump power (ionization frequency of 10^4 s^{-1}) the primary contribution to population of $Rg^{+*}(1)$ comes through the first channel. In the afterglow the population through the second channel can prevail, because at high pump power the high concentration of metastable atoms and electrons keep in plasma for a sufficiently long time after the excitation pulse. At interaction with the plasma electrons, both the reaction of ionization of metastable particles

$$Rg^* + e \rightarrow Rg^+ + 2e \quad (5)$$

and generation of new metastable particles

$$Rg + e \rightarrow Rg^* + e \quad (6)$$

take place.

These reactions are balanced by the opposite ones, and in the case of plasma recombination the opposite processes prevail. Application of the external electric field to plasma results in an increase in the electron temperature, makes the plasma non-equilibrium due to ionization. The direct reactions (5) and (6) start to dominate in this case too. The rate constant of reaction (5) has the exponential dependence on the electron temperature: $k_5 = 4 \cdot 10^{-5} \cdot T_e^{-3} \times$ e. p $(-4.2/T_e)$ (for argon).[1] The increase in the electron temperature leads to the increase in the rate of ionization of metastable particles, and, consequently, the rate of generation of emitting molecular ions through the second channel [reaction (4)] increases, as well as the intensity of the broad-band emission in the afterglow. As the gas pressure grows, the rate of conversion of metastable particles increases. So, the increase of the afterglow intensity must be observed at higher strength of the electric field, what is just the case in the experiment.

The possible influence of the reaction of ionization of excimer molecules

$$Rg_2^* + e \rightarrow Rg_2^+ + 2e, \quad (7)$$

should also be noted. The rate constant of this reaction $k_7 = 4 \cdot 10^{-5} \cdot T_e^{-3}$ e. p $(-3.7/T_e)$ (for argon)[1] also depends on the electron temperature. This reaction must contribute at high gas pressure. In krypton, as the electric field is applied, at the pressure of 0.25 atm the energy of MIC afterglow increases in the region of 200–260 nm with the maximum near 235 nm. As the pressure grows, the band of sharpest changes shifts toward longer waves, what indicates that dimer ions, as emitting molecules, are replaced by more complex molecular-ion complexes.

Ne atoms added to argon only slightly affect the electron temperature of plasma and, correspondingly, the rate of reaction (4). However, they destroy the emitting state Ar_2^{+*} (see reactions in Ref. 8). On the other hand, at high concentrations of neon, argon ions are efficiently generated in the Penning reaction. Then they take part in formation of states emitting in the MIC. Finally, these processes together lead to quenching of the entire broad-band emission of argon, but the intensity of emission in the regions near 190 and 260 nm in the second, in time sequence, maximum drops more slowly. Adding 3% of air to argon (0.5 atm) resulted in a decrease of the intensity and energy of a signal from the afterglow by more than two times. This decrease is connected, in our opinion, with the efficient transfer of the excitation energy from argon metastable particles to nitrogen molecules.

Thus, the second maximum in the time behavior of MIC emission from the inert gases can be explained by accumulation of the pump energy in metastable particles and, at high pressure, in dimers with their following ionization by plasma ions and population of the emitting states of molecular ions in the reactions of conversion of ions and excited atoms [reaction (4)] or excitation of a molecular ion by plasma electrons, correspondingly. Application of the electric field to the plasma gap results in an increase of the electron temperature, growth of the rate of ionization of metastable particles and dimers, and finally to the increase in the rate of generation of the emitting states in reaction (4). That is, the emission bands of argon with maxima at 235 and 260 nm, whose intensity increases significantly with application of the electric field, unambiguously belong to single-ionized molecular ions.

Acknowledgments

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