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Dissociative Recombination in the Afterglow of Low-pressure Barrier Discharge. Population of Ne($2p^5 3d$) Atoms

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The decaying neon plasma produced by the dielectric barrier discharge (DBD) in a cylindrical tube at a pressure of 0.1−40 Torr has been spectroscopically investigated to analyze the dissociative recombination (DR) of molecular ions with electrons as a mechanism for the formation of excited atoms. It is shown that, at the electron density in the afterglow less than $5 \cdot 10^{10}\,\mathrm{cm}^{-3}$ the DR is the dominant source of population of $3d$ levels at pressures $P_{\mathrm{Ne}} \geq 0.6$ Torr. At lower pressures, the optical properties of the decaying plasma are formed to a greater extent by the collisional-radiative recombination of Ne^+ ions. A significant variation of the relative intensities of the $3d \to 3p$ transition lines in the afterglow with a change in gas pressure was found, reflecting the effect of inelastic collisions on the formation of the spectrum of decaying plasma in the near infrared region. From measurements carried out at a pressure of 0.6 Torr, the relative values of the partial DR coefficients for the $3d_j$ levels of the neon atom were found. Comparison of these data with measurements in the near ultraviolet region, containing the lines of $4p \to 3s$ transitions, indicates the need to take into account the cascade $4p \to 3d$ transitions to correctly solve the problem of the final products of dissociative recombination.

Keywords: dielectric-barrier discharge, low-pressure plasma, optical emission spectroscopy, dissociative recombination, molecular ions, collisional-radiative recombination, cascade transitions.

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Introduction

The constant interest of researchers in the process of dissociative recombination (DR) of molecular ions, first proposed by the authors [1] to explain the phenomena involving charged particles in the ionosphere, is explained by two circumstances. On the one hand, this deionization mechanism determines the properties of a wide range of laboratory plasma objects and is a necessary link in models of stellar and planetary atmospheres [2]. On the other hand, despite the long history of studying the process, previously unknown manifestations of DR are found even in the case of the simplest molecular ions, such as H_2^+ [3] or HeNe^+ [4], which stimulates the development of new approaches to the calculation of its main characteristics i.e. cross-sections and rate constants [5]. The process discussed in most detail has been studied in the plasma of inert gases, and the largest number of papers has been related to the recombination of Ne_2^+ ions. The DR reaction in the case of neon is described by the equation

$$\mathrm{Ne}_2^+(\upsilon) + e \to (\mathrm{Ne}_2^*)_{\mathrm{unstable}} \xrightarrow{\alpha_j} \mathrm{Ne}^*(j) + \mathrm{Ne} + E_j^{\mathrm{kin}}, \quad (1)$$

where α_j are partial coefficients of recombination proportional to the probabilities of excited atoms formation in the final state j. Process (1) is multi-channel in nature both in input and output channels. Ions in various vibrational-rotational states υ can participate in recombination, and as a result of the reaction the atoms appear in a limited set of excited states j, which carry away the dissociation energy together with the atom in the ground state. Obviously, the partial coefficients, like the dissociation energy, must be characterized by at least two indices υ and j. However, we are forced out to simplify the analysis by considering the problem under the assumption that the ions distribution over internal states does not depend on the experiment conditions. The reasons are, firstly, the lack of reliable experimental data on the kinetics of excited molecular ions in plasma and, secondly, the prevailing in the literature after calculations [6] opinion about the extremely high rate of vibrational relaxation of molecular ions in collisions with atoms, according to which the rate constant k_υ of the process exceeds $10^{-10}\,\mathrm{cm}^3/\mathrm{s}$. At gas pressure ≈ 1 Torr the relaxation time is $\tau_r < 1\,\mu\mathrm{s}$, which is at least two orders of magnitude smaller than the recombination time $1/\alpha_s\,[e]$ under the conditions of our experiment (electron density $[e] < 5 \cdot 10^{10}\,\mathrm{cm}^{-3}$), if we use the DR rate constant [7]

$$\alpha_s = 1.7 \cdot 10^{-7}(300/T_e)^{0.43}\,(\mathrm{cm}^3/\mathrm{s}).$$

Given the above, α_s can be represented as a sum of partial coefficients: $\alpha_s = \sum \alpha_j$. In this approximation, the coefficients α_s and α_j completely characterize the recombination process and make it possible to express the population fluxes F_j of excited levels of atoms as follows:

$$F_j = \alpha_j[\mathrm{Ne}_2^+][e]. \quad (2)$$

Spectroscopic observations [8–14] of the afterglow of a direct current (DC) or high-frequency (HF) current glow discharge in inert gases (except for helium [15]) have shown that DR populates the excited levels $np^5(n+1)p$, $np^5(n+1)d$ and $np^5(n+2)p$ (n is the principal quantum number of an unexcited electron). The radiation at transitions from higher levels In the papers, where it was noticed [8–10,14], the authors associated with collision-radiative recombination (CRR) of Ne^+ ions:

$$Ne^+ + e + e \xrightarrow{\alpha_{cr}} Ne^+ + e \tag{3}$$

(α_{cr} — rate coefficient of CRR).

According to the process mechanism [16,17], the primary products of electron capture by ion in a triple collision (3) are highly excited states of the atom, and the distribution of populations over atomic levels is formed as a result of competition between collisional and radiative transitions

$$Ne_k^+ + e \Leftrightarrow Ne_j^+ + e, \tag{4}$$

$$Ne_k^+ \to Ne_j^+ + hv, \tag{5}$$

so in plasma with atomic ions the excited atoms can appear in any state, and the radiation caused by CRR is carried by all afterglow lines.

In experiments on measuring the α_j coefficients, this „background“ was eliminated by choosing a residually high gas pressure (for example, $P_{Ne} = 20$ Torr [11], 10 Torr [13]) when the ionic composition of the plasma is such that $[Ne_2^+] \gg [Ne^+]$ due to the rapid conversion of atomic ions into molecular ions

$$Ne^+ + Ne + Ne \xrightarrow{\beta} Ne_2^+ + Ne \tag{6}$$

($\beta \approx 5 \cdot 10^{-32}\,cm^6/s$ [18] is conversion rate constant).

However, the data of the cited papers hardly reflect the actual distribution of the flux (2) over the levels j due to the uncertainty that can be introduced by inelastic atomic-atomic collisions

$$Ne_k^* + Ne \Leftrightarrow Ne_j^* + Ne. \tag{7}$$

This is confirmed by the results of experiments in a wide range of pressure changes [12,14,19], which clearly indicate the participation of collisions (7) in the formation of the afterglow spectrum at pressures above 1 Torr. It is obvious that correct data on α_j values can be obtained at $P_{Ne} < 1$ Torr only. The study of DR of molecular ions in a spectroscopic experiment at such low pressures is still limited by data [14,20]. In the paper [14] relative populations of $4p$-levels of the neon atom were measured in the pressure range 150−0.2 Torr, and it was shown that up to $P_{Ne} \approx 0.6$ Torr at the electrons density $[e] \leq 5 \cdot 10^{10}\,cm^{-3}$ it is possible, based on the difference between the time characteristics of the spectral line intensities $J(t)$, and their reactions to pulsed „heating“ of electrons in the afterglow, to reliably separate the DR and CRR fluxes and to find the relative values of the coefficients α_j. Under these conditions, the conversion of atomic ions (6) is suppressed, and other mechanisms for the molecular ions formation are inefficient [20], so their density is low ($[Ne_2^+] \ll [Ne^+]$), and the decaying plasma radiation turns out to be close to the threshold of possibilities for registration by the method of multichannel photon counting. However, a well-known property of DR i.e. high selectivity of the process compared to CRR and at least two orders of magnitude higher recombination coefficient (at $[e] \approx 5 \cdot 10^{10}\,cm^{-3}$) [16,17] ensure the problem solution. The DR selectivity is especially pronounced when the levels of the $2p^5 4p$ configuration are populated. According to the results [14], at electron temperatures close to room temperature DR is available for only those levels of the neon atom that are located on the energy scale below $4p_4$[1] ($3p_3$ in Paschen notations), i. e. $4p_5$−$4p_{10}$. To complete the picture of the neon afterglow, note that, as we know, currently there are no data on DR role in the population of the levels of $2p^5 4s$ configuration.

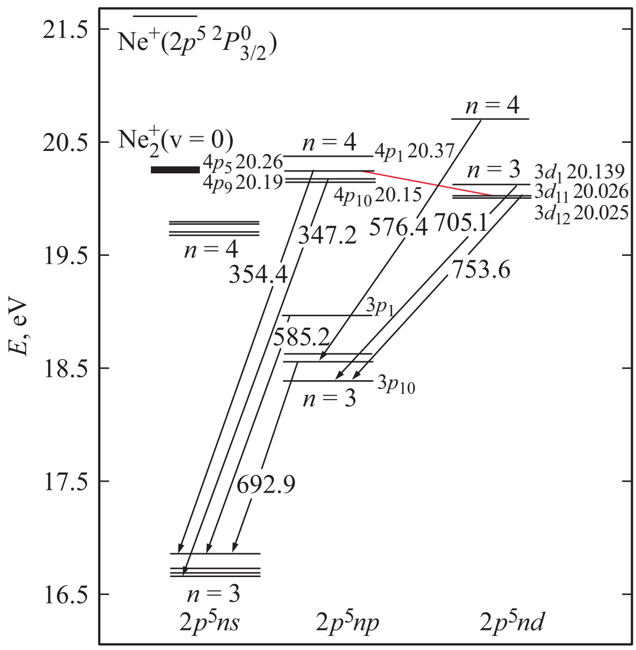

Figure 1. Scheme of excited levels and lines of neon atom (in nm), and energy of atomic and molecular ions. The position of the vibrational level $v = 0$ is indicated in accordance with the results [3].

The aim of this work was to solve the problem of the DR flux distribution over $3d$-levels of neon atom at the lowest possible neon pressure.

Experimental setup

A dielectric barrier discharge (DBD) in a glass cylindrical tube 20 cm long and inner diameter 3.9 cm was used as a plasma source. The advantages of DR study in the afterglow of such discharge and the details of the experiment are

[1] Hereinafter, we use the simplest way to designate levels according to their position in the energy scale: $4p_1$ is the top level of the configuration $2p^5 4p$, $4p_{10}$ is bottom. Similarly $3p_1$−$3p_{10}$, $3d_1$−$3d_{12}$.

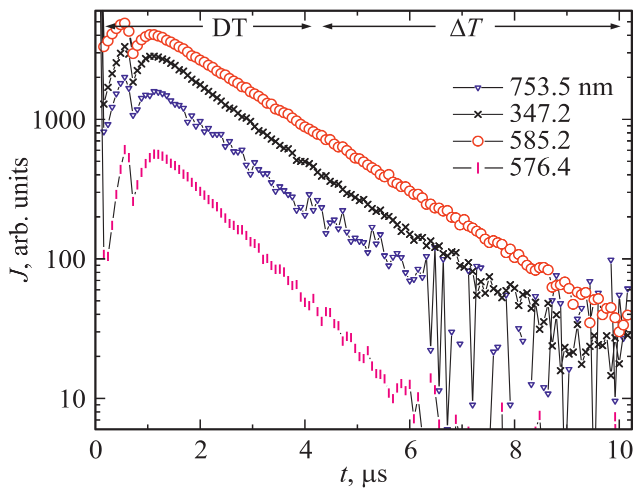

Figure 2. DBD afterglow on the tube axis. $P_{\text{Ne}} = 0.65$ Torr. $[e](t = 1.5\,\text{ms}) \approx 4 \cdot 10^{10}\,\text{cm}^{-3}$. For the convenience of results presenting, the numbers of photoelectrons are multiplied by the following coefficients: 585.2 nm — 1, 576.4 nm — 1, 347.2 nm — 3.5, 753.5 nm — 10.

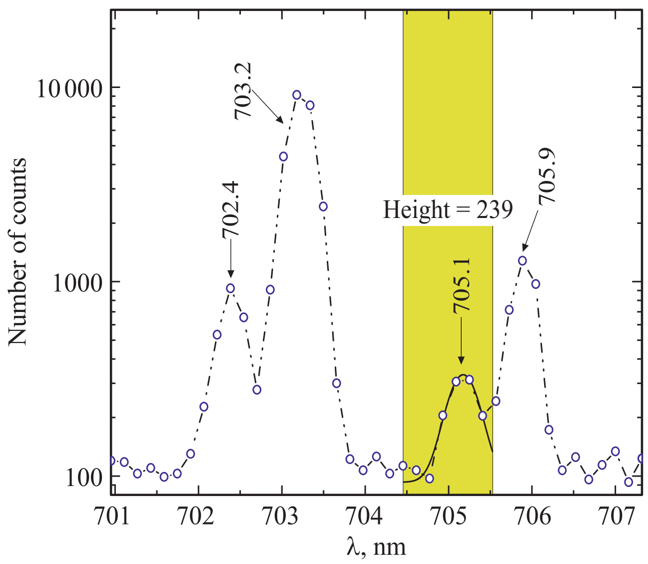

Figure 3. A fragment of the afterglow spectrum at $P_{\text{Ne}} = 10$ Torr.

described in [20]. The discharge frequency was 80–200 Hz, the observations were carried out along the axis of the discharge tube. Neon pressure 0.1–40 Torr, electrons density in the afterglow on the tube axis $[e] \leq 5 \cdot 10^{10}\,\text{cm}^{-3}$. The value $[e]$ was estimated from the rate of destruction of atoms in the resonance state $2p^5 3s(^3P_1)$ by electrons in the afterglow [20]. The temperature of the tube wall under all conditions was equal to room temperature. The light fluxes were registered by a multichannel photon counter, the spectral sensitivity $S(\lambda)$ of the setup was determined in the range 345–850 nm by radiation of the continuous spectrum source LS-1-CAL. As in the paper [14], in order to identify the source of population of the excited levels, we analyzed the reaction of decaying plasma radiation to pulsed heating of electrons. The only difference from [14] is that in this paper we used a high-frequency electric field as a heating source, as described in [20]. Some of the studied lines and their corresponding excited levels are shown in Fig. 1. We used two methods for measuring the line intensities in the afterglow. At pressures above several Torr, i.e. with relatively bright lines of the neon atom, afterglow spectra were registered in the region 700–850 nm. In this mode the photon counting circuit summed the pulses of the photomultiplier during the strobe gate pulse ΔT, the parameters of which, — width and delay DT — were chosen based on the observations $J_{jk}(t)$ with the desired time resolution. Figs. 2, 3 show an example of such measurements. The data in Fig. 2 confirm the identity of the change in time of photon fluxes at the transitions $3p \to 3s$, $3d \to 3p$, and $4p \to 3s$ in the sufficiently distant afterglow stage, and their clear difference from the intensity $J_{576.4}(t)$ of line 576.4 nm, the top level $4d$ of which lies much higher than the ground $v = 0$ vibrational level of Ne_2^+ ion (Fig. 1), and for this reason is populated exclusively by the CRR of Ne^+ (3) ions. This difference is also confirmed by the intensities response to pulsed heating of electrons. The line 585.2 nm was chosen as the reference line for $3p \to 3s$ transitions, the line radiation of which carries over a significant fraction of the DR flux [12]. It is also seen that by placing the gate pulse in the sufficiently late afterglow, it is possible to minimize the CRR contribution (Fig. 2), even if it is noticeable at the initial stage. The fragment of the afterglow spectrum shown in Fig. 3 explains the method for calculating the line intensities. Using the Origin program, approximating $J(\lambda)$ by the Gaussian function (solid curve in Fig. 3), we found the amplitude at the maximum and took it equal to the line intensity. The reason for the relatively low spectral resolution is the need to work with wide input and output slits of the monochromator to increase the luminous flux. At lower pressures a long-term (up to several hours) measurement of two signals turned out to be more efficient: the first one is intensity $J_{jk}(t)$, and the second one is signal $J_D(t)$ with tuning monochromator to the nearest spectral interval free of lines. For example, in the case of line 705.1 nm line, the monochromator was re-tuned to 704.5 nm. (Fig. 3). The subtraction $J_{jk}(t) - J_D(t)$ made it possible to take into account not only „dark“ photoelectrons, but also weak illumination from light scattered in the monochromator of strong lines of transitions $3p{-}3s$. To compare the luminous fluxes, the sums of the numbers of photoelectrons in the photon counter channels corresponding to the range ΔT were calculated (Fig. 2).

Discussion of results

The list of lines from which the partial coefficients of recombination were determined, and their characteristics are presented in Table 1. From these data, we see that the configuration $2p^5 3d$ consists of two groups of closely

Table 1. Parameters of spectral lines of transitions $3d \to 3p$. In parentheses — data for resonance lines, in the bottom line — for the line 345.4 nm of the transition $4p_5 - 3d_{11}$transition

Upper level	Energy, eV	λ_{jk}, nm	A_{jk}, 10^6 s^{-1}	$A_j = \Sigma_v(A_{jk})$	$S(\lambda_{jk})$
$3d_1$	20.1394	705.1 (61.56)	2.38 (40.3)	44.4 (84.7)	0.89
$3d_2$	20.1375	705.9	7.46	48.2	0.89
$3d_3$	20.1363	794.3	3.82	47.7	0.25
$3d_4$	20.1361	813.6	11.8	48.6	0.17
$3d_5$	20.0484	830.0	14	46.6	0.11
$3d_6$	20.0482	841.8	15.6	46.5	0.088
$3d_7$	20.0404	747.2 (61.87)	2.96 (80)	41.8 (121.8)	0.58
$3d_8$	20.0367	748.9	23.1	50.5	0.57
$3d_9$	20.0349	849.5	39	48.2	0.068
$3d_{10}$	20.0346	837.7	50	50	0.096
$3d_{11}$	20.0264	753.6 (61.91)	30.6 (26.6)	50.5 (77.1)	0.61
$3d_{12}$	20.0246	754.4	38.7	53.8	0.61
$4p_5$	20.2592	345.4	36.8	112	0.28

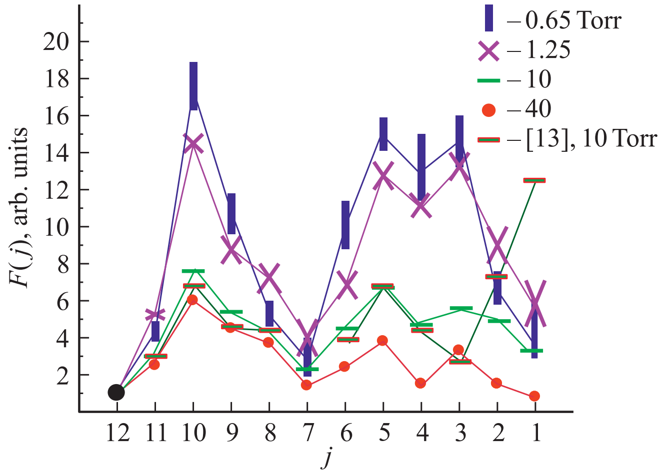

Figure 4. Fluxes of quanta from $3d_j$-levels of neon atom in afterglow, rel. un.

spaced levels corresponding to different moments of the atomic core, the distance between which is less than the thermal energy $kT_a \approx 0.03$ eV. This again emphasizes the necessity of choosing extremely low pressures in such experiment. The minimum pressure, at which we managed to separate the DR and CRR fluxes, was $P_{\mathrm{Ne}} \approx 0.6$ Torr. When calculating the partial coefficients of recombination, the following rather obvious relation was used:

$$\alpha_j = J_{jk} A_j / \left(A_{jk} S(\lambda_{jk})\right), \quad (8)$$

where J_{jk} is the intensities of the spectral lines found by one of the methods described above. In calculations we used the probabilities of transitions A_{jk} from the NIST database presented in Table 1, where A_j is the sum of probabilities over all allowed transitions from level j. For three levels, i.e. $3d_1$, $3d_7$ and $3d_{11}$, we give two values A_j considering (in parentheses) and not considering transitions to the ground state of the neon atom allowed for these levels. The presence of resonant transitions introduces uncertainty into the calculations of α_j (8), the elimination of which requires analysis of the radiation „trapping“ effect under experimental conditions. The solution of this problem [21,22] for various geometries and mechanisms of spectral line broadening shows that for our conditions (the density of atoms is more than 10^{16} cm^{-3}) neon turns out to be optically dense ($\kappa_0 R \gg 1$, κ_0 is absorption coefficient at the center of the line, R is radius of the discharge tube), so that the resonant radiation in the calculation of probabilities A_j can be ignored. However, in our situation the problem becomes more complicated due to the specificity of the spectral line profiles in the afterglow associated with dissociative recombination. In accordance with (1) as a result of the reaction the excited atoms arise with a kinetic energy $\delta E_j^{\mathrm{kin}}/2$, which significantly exceeds the thermal energy kT_a of atoms at $T_A = 300$ K. Comparing the energies of the ground vibrational level of the Ne_2^+ ion and the levels $3d_j$ (Fig. 1), we find that the minimum kinetic energy (corresponding to the population of the level $3d_1$) $\Delta E_j^{\mathrm{kin}}/2 \approx 0.06$ eV, maximum (for $3d_{11}$) $\Delta E_j^{\mathrm{kin}}/2 \approx 0.12$ eV. The spectral width of the lines of such atoms, due to the Doppler effect, turns out to be much larger than the width of the absorption line of atoms in the ground state [10,24]. In these studies the authors observed in the afterglow of neon at pressures less than 3 Torr lines of $3p \to 3s$ transitions by 3 times wider than in the discharge. For the lines $3d \to 3p$, such observations were not made, but from a comparison of the value $\Delta E_j^{\mathrm{kin}}/2 \approx 0.12$ eV and the energy $\Delta E_{3p}^{\mathrm{kin}}/2 \approx 0.63$ eV for the level $3p_1$ (Fig. 1) it is clear that the broadening will be much less significant. From the point of view of this paper problems, the measurements performed at pressures below 1 Torr, when the shape of the absorption line can be considered close to Doppler one, are of greatest interest. In this approximation the violation of the condition $\kappa_0 R \gg 1$, according to our estimates, can only occur on the wings of the resonance line 61.91 nm (upper level $3d_{11}$), which carry over an insignificant fraction of the line's integral radiation. Following these considerations, we did not take into account the resonant transitions when calculating the branching coefficients. Note that it is easy to take them into account by recalculating the branching coefficients in the presence of the exact solution of the problem.

The main results of the paper are presented in Fig. 4 as fluxes of quanta $F(j) = F_j/F_{12}$, emitted by $3d$-levels, related to the flux F_{12} from the lower level at various neon pressures. One can see a significant change in the relative values of the fluxes with pressure decreasing, clearly indicating the increased contribution of the upper levels to the afterglow spectrum. We tried to approximately show the uncertainty of values $F(j)$ due to the small luminous fluxes at pressures 1.25 and 0.65 Torr by the vertical size of the corresponding symbols. It is important to note that the transition from 1.25 to 0.65 Torr leads only

Table 2. Partial coefficients of recombination (in %) to $3d$ and $4p$ levels

j	12	11	10	9	8	7	6	5	4	3	2	1
α_{3dj}	0.01	0.04	0.17	0.1	0.05	0.03	0.1	0.14	0.13	0.14	0.06	0.04
α_{4pj}			0.1	0.22	0.18	0.17	0.22	0.11				

to a slight distribution deformation of the quantum flux, which obviously indicates $F(j)$ stabilization as a function of pressure and, in line with the logic of paper, can be associated with decreased effect of inelastic collisions (7) at low pressure. This means that values $F(j)$ related to $P_{\mathrm{Ne}} = 0.65$ Torr represent the distribution of the DR flux over the configuration levels $2p^5 3d$, i. e. they are proportional to the relative values of the partial coefficients α_j. The data for $3d_1$, $3d_7$, and $3d_{11}$ were obtained under the assumption that the resonant radiation is trapped, i. e. in probabilities A_j only $3d \rightarrow 3p$ transitions are taken into account.

We compared the data of our paper with the only results of a similar experiment available in the literature [13], in which the relative populations N_j of the $3d_J$ levels of the neon atom in the afterglow of a high-frequency discharge in neon at pressure 10 Torr were identified. In calculations the authors [13] used the transition probabilities proposed in [24]. From the values N_j and the transition probabilities [24] we reconstructed the relative fluxes $F(j)$. There are no data in [13] for levels $3d_{12}$ and $3d_7$, therefore, for clarity of comparison in Fig. 4 we present the calculated fluxes, combining them with our data for $3d_{11}$ at pressure 10 Torr. It can be seen that, in general, the results [13] are close to our results, except for the upper levels $3d_2$ and $3d_1$. It is difficult to judge the degree of correctness of these data. Note only that they were obtained from the intensities of the lines 705.9 and 867.9 nm, while when we review the lines 705.9 and 705.1 nm, for which the quantum yield of the photomultiplier is the same, and therefore errors associated with determining the sensitivity of the circuit of radiation registration in a wide range of wavelengths are excluded. From the fragment of the spectrum in Fig. 3 and the transition probabilities of Table 1, which differ little from those used by the authors [13], it follows that the flux ratio $F(j = 2)/F(j = 1)$ proposed in [13] is opposite.

In Table 2 we placed the partial coefficients of recombination $\alpha_j^{\mathrm{rel}} = \alpha_j / \sum \alpha_j$ found from data presented in Fig. 4. These values reflect the contribution of the recombination flux to the $3d_j$ level to the total recombination flux to the $2p^5 3d$ levels. Together with similar data on the results [14] for the levels $4p_5$–$4p_{10}$ of configuration $2p^5 4p$ (also presented in Table 2) they give a complete picture of the DR of Ne_2^+ molecular ions as source of population of the excited levels of the said configurations.

The determination of the partial coefficients of DR must be accompanied by the role analysis of cascaded transitions. For example, according to data of [13], the population of $3p$-levels of the neon atom is almost by a one-forth is due to $3d \rightarrow 3p$ cascades. A similar problem is absent only for the $4p$-levels, the lower six of which [14] are associated in afterglow directly with DR (1). To determine the contribution of cascades to the population of the $3d$-levels, data on the lines of $4p \rightarrow 3d$ transitions located in the far infrared region of the spectrum are required. In the NIST Database we found a single line 5327 nm (Fig. 1 shows the corresponding $4p_5 \rightarrow 3d_{11}$ transition) with transition probability $A_{pd} = 1.74 \cdot 10^5\,\mathrm{s}^{-1}$). Using the full set of data on line intensities and probabilities of $4p \rightarrow 3s$ transitions under similar conditions [14], and the data in Fig. 2, it is easy to estimate the contribution of the flux $4p_5 \rightarrow 3d_{11}$ into the 753.6 nm line emission. At $t \geq 5$ ms from the beginning of the afterglow, when the effect of collision-radiative recombination is excluded, it is $\approx 10\%$. We emphasize that this is the contribution of only one, and the least populated in the afterglow [14] level of the $2p^5 4p$ configuration. If we take into account that due to the selection rules the transitions to $3d_{11}$ are also allowed from levels $4p_6$, $4p_7$, $4p_8$ and $4p_{10}$, whose total population is by an order of magnitude greater than the population of $4p_5$, then the issue of the possibility of direct population of the levels $3d$ at DR of Ne_2^+ ions is legitimate. To answer it, we need a complete set of data on the probabilities of more than fifty allowed $4p \rightarrow 3d$ transitions, which we do not have. Note that if they are the source of population of levels $3d$, the issue of the contours of spectral lines in the problem of radiation trapping ceases to be significant.

Due to the uncertainty with cascade transitions, the coefficients α_{3dj} (Table 2), in contrast to α_{4pj}, can be called partial coefficients of recombination only with a certain stipulation. Note that the problem of determining partial coefficients and the role of cascade transitions arises not only in relation to inert gases, but is relevant for any type of plasma containing molecular ions. These data are of particular interest for predicting the properties of active medium in plasmas with molecular ions [25]. They can also be used as reference data for testing theoretical models (for example, [26,27]) claiming an adequate distribution of the recombination flux over the output channels of the process.

Conclusions

The distribution of the dissociative recombination flux of Ne_2^+ molecular ions with electrons over the levels of the configuration $2p^5 3d$ of the neon atom in the afterglow of low-pressure dielectric barrier discharge is found by optical emission spectroscopy. For the first time, the measurements were carried out under conditions of extremely low (less than 1 Torr) gas pressures, which exclude the effect of inelastic atom-atom collisions on the populations of excited levels. The paper results show the need to analyze cascade transitions $4p \rightarrow 3d$, the uncertainty of their contribution to the population of $3d$ levels does not allow us to identify the obtained data with partial coefficients of dissociative recombination characterizing the distribution of the recombination flux over the output channels of the process. Comparison of the photon fluxes at the transitions from the levels $4p_5$ and $3d_{11}$, which are coupled by an infrared transition, suggests that the dissociative recombination mechanism of Ne_2^+ is such that its output channels do not contain levels of $2p^5 3d$ configuration.

Conflict of interest

The author declares that he has no conflict of interest.

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