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Kinetics of the nitrogen first negative system excitation by ionising radiation

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***Abstract*. The rate constants of N_2^+(B) quenching by nitrogen and helium and of two- and three-body charge exchange of He_2^+ on H_2, D_2, and Kr are measured from luminescence at the 0 – 0 transition of the first negative system of nitrogen in mixtures of helium and nitrogen with hydrogen, krypton or deuterium excited by alpha particles emitted by ^{210}Po .**

Keywords: *nitrogen, first negative system, kinetics, ionising pump.*

The efficient quasi-continuous laser on the first negative system of nitrogen has been built due to the selective depopulation of the $B\,^2\Sigma_u^+$ and $X\,^2\Sigma_g^+$ states of the N_2^+ ion by hydrogen molecules [1, 2]. The kinetics of processes in the $He - N_2 - H_2$ mixture was studied in [3] from luminescence at the 0 – 0 transition of the first negative system of nitrogen excited an electron beam with a pulse duration of 1.5 μs. However, the rates constants of processes measured in [3] noticeably differ from those reported in [4, 5].

In this paper, the rate constants of processes in $He - N_2 - H_2$ (Kr, D_2) mixtures were determined from luminescence at the 0 – 0 transition of the first negative system of nitrogen at 391 nm excited by alpha particles emitted by ^{210}Po. Krypton and deuterium can efficiently depopulate the $X\,^2\Sigma_g^+$ state of N_2^+ [6], as hydrogen does, and are also of interest for obtaining quasi-continuous lasing in the first negative system of nitrogen. The use of alpha sources for ionisation provides a highly stable pump power compared to that of an electron beam or other excitation sources.

The experimental setup is described in [7]. Eighteen ^{210}Po sources with a total activity of $\sim 7 \times 10^9$ Bq were accommodated in a stainless steel chamber. The excitation region volume was 40 cm^3 and the specific pump power was $\sim 5 \times 10^{-5}$ W cm^{-3} (at a helium pressure of 4 atm). Emission spectra were analysed with an SPM-2 monochromator equipped with a FEU-106 photomultiplier operating in the photon counting regime. He (of purity 99.99 %), N_2 (99.998 %), and Kr (99.999 %) were used. Deuterium (no more than 0.2 %) and technical hydrogen were passed through traps with silica gel and active copper, vessels with these gases being cooled with liquid nitrogen during gas admission.

The basic kinetic processes proceeding in high-pressure $He - N_2 - H_2$ (Kr, D_2) mixtures upon ionising pumping are summarised in Table 1. The measured rate constants are mainly compared with the results obtained in [3 – 5, 10], while references to other works and their discussion are contained in [3]. The data on electron – ion recombination are absent in Table 1 because this recombination is completely suppressed by competing processes (1 – 5 and others). According to estimates, the electron concentration n_e in mixtures with He (at a pressure of up to 6 atm) does not exceed $\sim 10^{12}$ cm^{-3} even in the alpha-particle track. Because the luminescence intensity was measured in arbitrary units, to determine the absolute value of rate constants, it is necessary to use standard rates at the corresponding stages of the kinetic chain of processes [3]. In analysis of quenching of the upper laser level, such a standard was the spontaneous decay probability v_{sp} (process 9 in Table 1), while for the charge exchange of He_2^+ on the mixture components, these standards were the two- and three-body rate constants of the charge exchange of He_2^+ on nitrogen (processes 4 and 5 in Table 1). The pump power in the pressure range of the mixture components studied was proportional to the total pressure of the gas mixture.

Figure 1 shows the dependence of $1/I$ on the nitrogen pressure and Fig. 2 presents the dependence of the para-

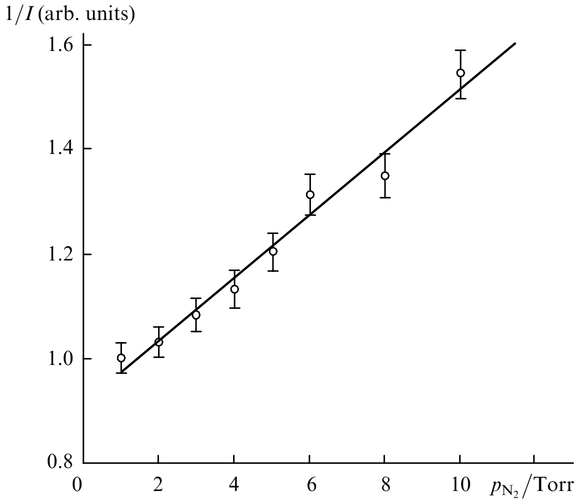

Figure 1. Dependence of the inverse luminescence intensity on the nitrogen pressure p_{N_2} in the He (4 atm) – N_2 mixture.

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Табл.1.

Process number	Process	Rate constant, probability	Value	References
1	$He^+ + 2He \rightarrow He_2^+ + He$	$k_1/10^{-31}$ cm^6 s^{-1}	1.1	[6]
2	$He^+ + N_2 \rightarrow$ products	$k_2/10^{-9}$ cm^3 s^{-1}	1.2	[6]
3	$He^+ + N_2 + He \rightarrow$ products	$k_3/10^{-29}$ cm^6 s^{-1}	2.2	[8]
4	$He_2^+ + N_2 \rightarrow N_2^+ + 2He$	$k_4/10^{-10}$ cm^3 s^{-1}	11	[9]
4a	$He_2^+ + N_2 \rightarrow N_2^+(B_{v=0}) + 2He$		3 ± 1	[4, 5]
5	$He_2^+ + N_2 + He \rightarrow N_2^+ + 3He$	$k_5/10^{-30}$ cm^6 s^{-1}	16 ± 3	[3, 9]
5a	$He_2^+ + N_2 + He \rightarrow N_2^+(B_{v=0}) + 3He$		3 ± 1	[4, 5]
6	$N_2^+(B) + N_2 \rightarrow$ products	$k_6/10^{-11}$ cm^3 s^{-1}	9 ± 2 5.1 ± 0.9	This paper, [3] [4]
7	$N_2^+(B) + He \rightarrow$ products	$k_7/10^{-13}$ cm^3 s^{-1}	11 ± 3 8 ± 2 5.4 ± 0.8	This paper [3] [4]
8	$N_2^+(B) + N_2 + He \rightarrow$ products	$k_8/10^{-30}$ cm^6 s^{-1}	2 ± 0.5 $\leqslant 1$	This paper [4, 5]
9	$N_2^+(B) \rightarrow N_2(X) + h\nu$	$\nu_{sp}/10^7$ s^{-1}	1.6	[11]
10	$He_2^+ + H_2 \rightarrow$ products	$k_{10}/10^{-10}$ cm^3 s^{-1}	10 ± 3 4.1 24 ± 4 4.5 ± 1	This paper [10] [3] [4]
11	$He_2^+ + H_2 + He \rightarrow$ products	$k_{11}/10^{-30}$ cm^6 s^{-1}	15 ± 5 $\leqslant 1$ 9 ± 5	This paper [4, 5] [10]
12	$He_2^+ + Kr \rightarrow$ products	$k_{12}/10^{-11}$ cm^3 s^{-1}	8 ± 3 $\leqslant 8$	This paper [10]
13	$He_2^+ + Kr + He \rightarrow$ products	$k_{13}/10^{-30}$ cm^6 s^{-1}	3 ± 1 17 ± 3	This paper [10]
14	$He_2^+ + D_2 \rightarrow$ products	$k_{14}/10^{-10}$ cm^3 s^{-1}	8 ± 3	This paper
15	$He_2^+ + D_2 + He \rightarrow$ products	$k_{15}/10^{-30}$ cm^6 s^{-1}	< 2	This paper

meter p/I on the helium pressure in the $He - N_2$ mixture (where p is the mixture pressure and I is the luminescence intensity at 391 nm). The data processing by the method [3] gave negative deactivation rates for the $B_{v=0}$ state in some mixtures (ν_B is denoted as in [3]). Therefore, unlike [3], where the data processing was performed assuming that the only population channel of the upper laser level is the charge exchange on N_2 of only the He_2^+ ions that have been produced due to the He^+ conversion (processes 1, 4 and 5 in Table 1), it was assumed here that the other $N_2^+(B)$ population channels also exist (processes 4 and 5 being dominant), so that the population rate of the $B_{v=0}$ state is proportional to the mixture ionisation rate. One of such channels is the associative ionisation of the excited states of helium: $He^* + He \rightarrow He_2^+ + e$ followed by the charge exchange of the molecular ion on the nitrogen atom.

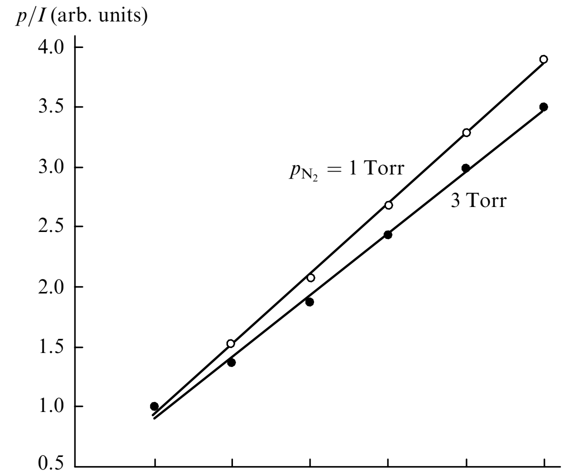

Figure 2. Dependences of the parameter p/I on the helium pressure p_{He} in $He - N_2$ mixtures at different nitrogen pressures p_{N_2}.

In this case, the luminescence intensity of the mixture at 391 nm is described by the expression

$$I = CW/q, \tag{1}$$

where C is a coefficient proportional to the sensitivity of a detection system; W is the pump power of the mixture; and q is the total deactivation rate of the $B_{v=0}$ state, i.e., $q \sim W/I \sim p/I$.

One can see from Fig. 1 that the dependence $q(p_{N_2})$ is approximately linear and the influence of three-body processes involving the N_2^+ ion and two nitrogen molecules on the B state population is insignificant in the pressure range of helium and nitrogen studied, in accordance with the data obtained in [3]. The dependence $q(p_{N_2})$ obtained for different nitrogen pressures (Fig. 2) shows the necessity of consideration of three-body processes (8 in Table 1). Then, the deactivation rate of the $B_{\upsilon=0}$ state is described by the expression

$$q = \nu_{sp} + k_6[N_2] + k_7[He] + k_8[N_2][He]. \qquad (2)$$

The rate constants k_6, k_7 and k_8 can be found from the data presented in Figs 1 and 2 and the known value of ν_{sp} (Table 1). The values of k_6 and k_7 obtained here well agree with those measured in [3]. Recall that the processing of our data according to [3] gave inadmissible values of parameters and the rate constants.

Figure 3 shows the dependences of the inverse luminescence intensity on the krypton pressure (similar dependences were measured for hydrogen and deuterium). They are also linear, indicating to a key role of either the charge exchange of He_2 on Kr or quenching of $N_2^+(B)$ by krypton. For hydrogen, the dominant role of charge exchange over quenching by hydrogen molecules was demonstrated in [1, 3]. We will assume here that the quenching rate constants of the $B_{\upsilon=0}$ state by krypton and deuterium are also small. It is known that the charge exchange of He^+ on H_2 and D_2 is almost absent, and the rate constant of the corresponding process involving krypton should be also low, similarly to Ar and Xe [6]. In the He (3 – 6 atm) – N_2 (4 Torr) mixture, more that 70 % of the He^+ ions are converted to the He_2^+ ions even by the estimate using the $k_1 - k_5$ rate constants. The associative ionisation of the excited helium states also results in the formation of the He_2^+ ions. Therefore, we will assume that the luminescence intensity at 391 nm decreases after the addition of krypton (hydrogen, deuterium) mainly due to the charge exchange of He_2^+ on Kr (H_2, D_2).

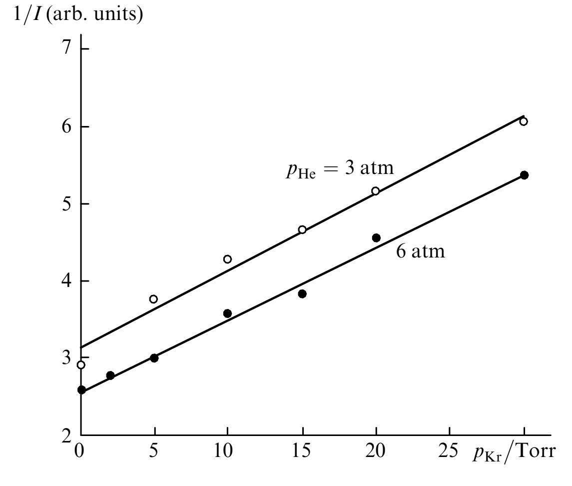

Figure 3. Dependence of the inverse luminescence intensity on the krypton pressure p_{Kr} in the He – N_2 (4 Torr) – Kr mixture at different helium pressures.

Then, the inverse luminescence intensity is described by the expression

$$\frac{I_0}{I} = 1 + \frac{(k_{12} + k_{13}[He])[Kr]}{(k_4 + k_5[He])[N_2]}, \qquad (3)$$

where I_0 is the luminescence intensity for the helium-nitrogen mixture without krypton. By using the known rate constants k_4 and k_5, we find from Fig. 3 the rate constants k_{12} and k_{13}. The rate constants of charge exchange of He_2^+ on Kr (H_2, D_2) measured in this way are presented in Table 1. The rate constants k_{11} and k_{12} well agree with those obtained in [10], whereas k_{13} noticeably differs from this rate constant obtained in [10]. In our opinion, the 'effective' value of the rate constant k_{13} for the three-body charge exchange of He_2^+ on Kr depends on the krypton pressure. A similar conclusion can be made from Fig. 10 of paper [9] for the rate constant of two-body charge exchange of He_2^+ on Ne, where the measured charge exchange rate was saturated at the neon pressure above 0.3 Torr. The partial pressure of krypton in our study was 2 – 30 Torr, which was higher by two orders of magnitude than that in [10]. It probably explains a strong difference between the values of k_{13} measured in our paper and in [10].

Thus, the rate constants of a number of processes proceeding in the active medium of a laser in the first negative system of nitrogen have been measured in this paper. The obtained rate constants well agree as a whole with those reported in [3, 10]. The rate constants of three-particle charge exchange of He_2^+ on H_2 (process 11 in Table 1) and on Kr (process 13) noticeably differ from those obtained in [4, 5] and measured in [10], respectively.

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