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On the mechanism of selective population of the $3p_1$ level of the neon atom in He–Ne plasma

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In the spectroscopic studies of helium-neon plasma carried out earlier, a pronounced selectivity of the population of the $3p_1$ level (according to Paschen), the upper one from the group of levels of the $2p^54p$ configuration of the Ne atom, was revealed. As the helium pressure increased, up to 60% of the intensity of all transitions $2p^54p \to 2p^53s$ was concentrated in the 352.05 nm line ($3p_1 \to 1s_2$). In the present work, a mechanism is proposed for the growth of the relative population of the $3p_1$ level with increasing He pressure, associated with the features of the collisional kinetics of the $3p_i$ states of the $2p^54p$ configuration. The reason for the significantly faster depletion of the lower $3p_i$ levels ($i \geq 2$) compared to $3p_1$, apparently, is the features of the mutual arrangement of the adiabatic terms of the $\mathrm{Ne}(2p^54p)+\mathrm{He}(1s^2\,{}^1S_0)$ system. The mechanism of formation of selective population of the $3p_1$ level considered in this work can be realized in a mixture of helium with neon and cannot be realized in pure neon, which corresponds to the results of spectroscopic studies. Model calculations of the part of the spectrum related to transitions $3p_1$, $3p_4$, $3p_2 \to 1s_j$ were carried out for a fixed distribution of population flows $3p_1$, $3p_4$, $3p_2$ and different helium pressures. Good agreement between the measurement results and the results of numerical modeling is observed.

Keywords: helium-neon plasma, selective population, inelastic collisions of atoms, adiabatic terms, numerical modeling of excited state population.

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Introduction

Selective population of individual excited states of atoms and molecules is usually a prerequisite for gas medium to be capable of enhancing a light wave transmitted through it. Study [1] addressed possible selective population of the excited neon atom states in the $2p^54s$: $2s_2$, $2s_3$, $2s_4$, $2s_5$configuration (Paschen notation) due to excitation transfer when metastable $\mathrm{He}(1s2s\,{}^3S_1)$ atoms collide with neon atoms in the ground state. Two years later, the first gas laser was demonstrated [2]. The laser used discharge plasma in a helium-neon mixture as an active medium and lasing occurred at the infrared transition 1152.3 nm ($2p^54s$, $2s_2 \to 2p^53p$, $2p_4$). The upper laser level was populated in the course of inelastic collisions of metastable atoms in the triplet state $\mathrm{He}(1s2s\,{}^3S_1)$ with Ne atoms [3]:

$$\mathrm{Ne}(2p^6)+\mathrm{He}(1s2s\,{}^3S_1) \to \mathrm{Ne}(2p^54s,\,2s_2)+\mathrm{He}(1s^2\,{}^1S_0).$$

Helium-neon laser oscillation in the visible region on the red line 632.8 nm ($2p^55s$, $3s_2 \to 2p^53p$, $2p_4$) was achieved in [4]. Upper level $3s_2$ was populated as a result of excitation transfer from the metastable helium atoms in the singlet state $\mathrm{He}(1s2s\,{}^1S_0)$ [3]:

$$\mathrm{Ne}(2p^6)+\mathrm{He}(1s2s\,{}^1S_0) \to \mathrm{Ne}(2p^55s,\,3s_2)+\mathrm{He}(1s^2\,{}^1S_0).$$

Creation of a helium-neon laser served as a driver of explosive growth of the number theoretical and experimental investigations of inelastic processes in the helium-neon mixture. Findings of these investigations are summarized in [5]. Despite the comprehensive information about inelastic processes in the helium-neon plasma available in the literature, far from all in this area have been solved.

Thus, the experiments [6] investigating the processes that form plasma radiation spectra in helium with trace neon impurity detected a strongly pronounced selectivity of population of the upper level from the group of $\mathrm{Ne}(2p^54p)$ levels (Figure 1). The authors of this study don't know whether there are any studies investigating this selectivity and primarily the light flux concentration of the transitions $3p_i \to 1s_j$ in the 352.05 nm line ($3p_1 \to 1s_2$) with helium pressure growth.

In [6], experiments were performed in the helium pressure range $p_{\mathrm{He}} = 0.08-20$ Torr and neon pressure range $p_{\mathrm{Ne}} = 0.0005-0.003$ Torr. Plasma was generated by a low-frequency (40–160 Hz) pulse discharge with two dielectric barriers (discharge tube arrangement is shown in Figure 2), which removed the cataphoretic effect of He-Ne mixture separation.

As the He pressure grew, more than 60% of the whole light flux emitted in all transitions from the $2p^54p$ configuration levels concentrated in the 352.05 nm line ($3p_1 \to 1s_2$) (Figure 3). There was an opportunity in [6] to record plasma radiation with high time resolution both in the discharge phase and afterglow phase. Radiation

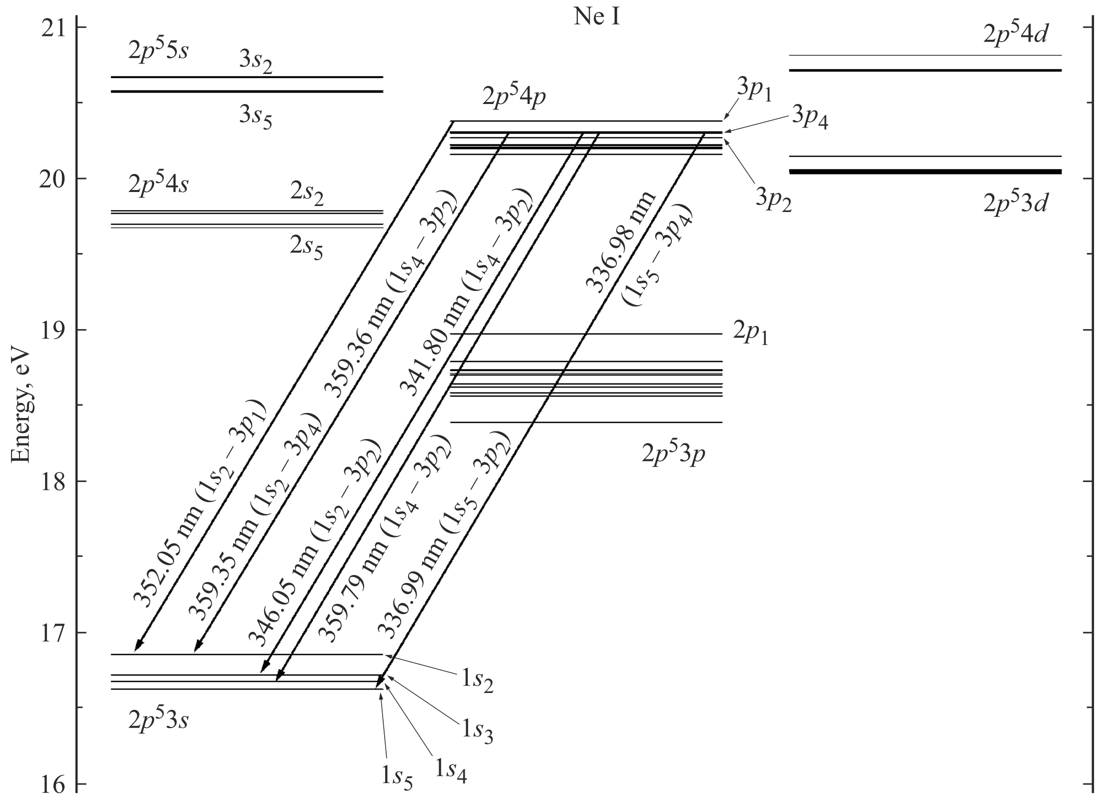

Figure 1. System of neon atom energy levels.

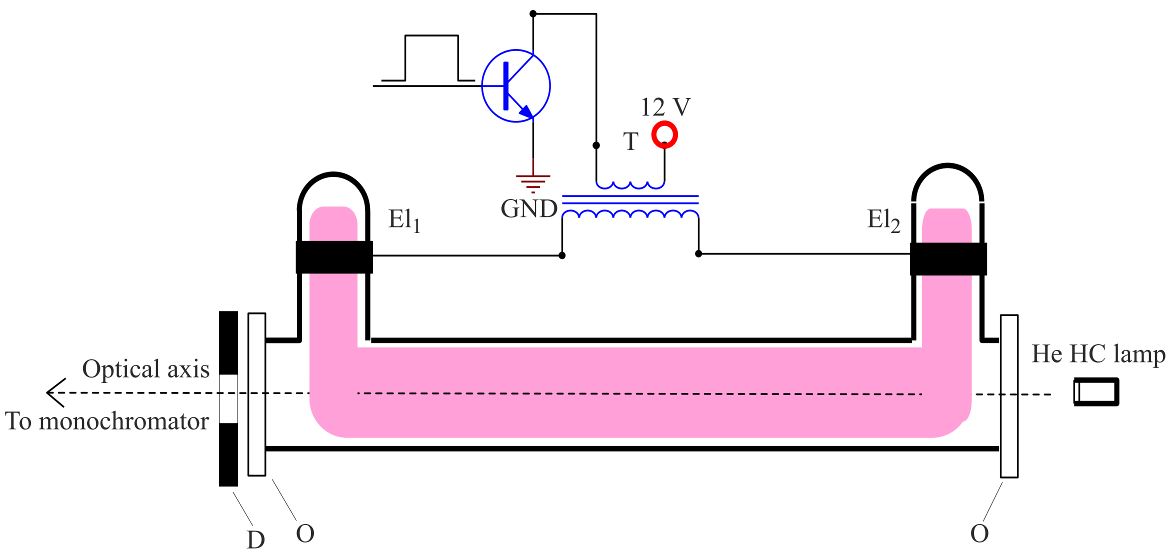

Figure 2. Discharge tube 3.8 cm in diameter, horizontal discharge portion length is 25 cm. O — quartz windows, D — diaphragm 5 mm in diameter, T — pulse transformer, El_1, El_2 — electrodes in the form of copper foil on the outer surface of the glass tube.

concentration of the transitions from the $3p_i$ levels in the 352.05 nm line was observed not only in the afterglow phase (Figure 3), but also in the discharge phase.

This fact is unexpected at first sight because the pressure growth and, consequently, increase in atom concentration and collision frequency shall lead to an accelerated relaxation in the energy level system. The experiment, on the contrary, shows that a transition to a higher helium pressure leads to an increase in population of the upper $3p_1$ $2p^54p$-configuration level with respect to the populations of the $3p_i$ $(i \geq 2)$ levels lying below. It is obvious that selective population of any level is attributable either to heavy population flux in this level or by more intense depletion processes in other levels or by both. The objective

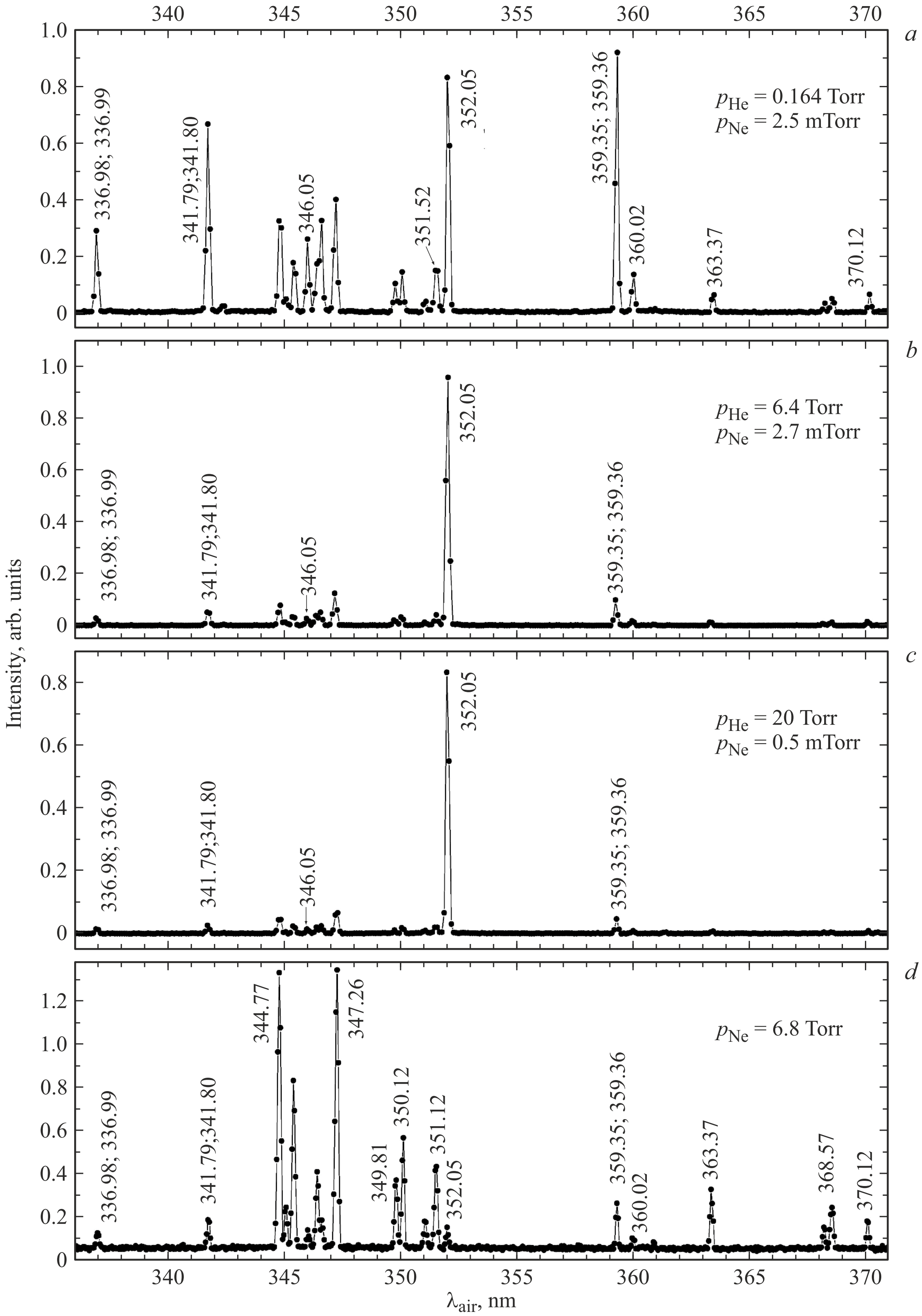

Figure 3. Discharge afterglow spectra in the He-Ne mixture (a, b, c) and pure neon (d).

of this study was to investigate the impact of the second selective population option. In the model, the distribution of population flux was considered to be stationary and population variation was observed due to the pressure

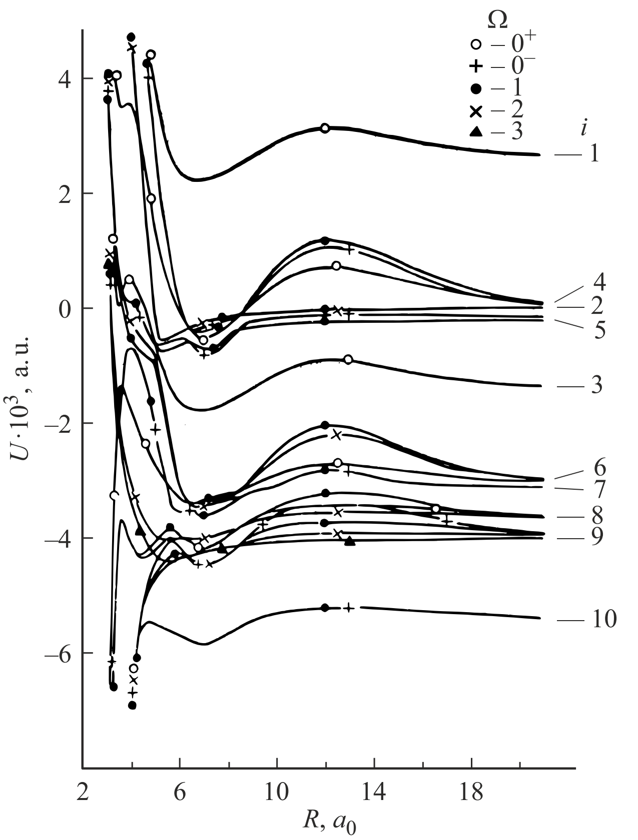

Figure 4. Quasi-molecular terms of the $\mathrm{Ne}(2p^54p,\ 3p_i) + \mathrm{He}$ system [7]. The energy is expressed in atomic units. Ω is the quantum number of the total momentum projection of the electron shell on the molecule axis, i is the $3p_i$ atomic level number according to Paschen notation.

growth and, consequently, growth of transition rate between the excited states of a neon atom in the $2p^54p$ configuration as a result of collisions.

Model of population of the upper levels of the Ne atom in the $2p^54p$ configuration

The cause of less effective de-excitation of the $3p_1$ state of the Ne atom in collisions with He atoms than the de-excitation of the $3p_i$ $(i \geq 2)$ states in the same configuration, but with a lower energy may be associated with the features of mutual arrangement of the quasi-molecular terms of the $\mathrm{Ne}(2p^54p,\ 3p_i) + \mathrm{He}(1s_2\ ^1S_0)$ system [7][1] (Figure 4).

It can be seen that the upper quasi-molecular term correlated with the $\mathrm{Ne}(3p_1)$ state is far from other terms of the $\mathrm{Ne}(2p^54p,\ 3p_i) + \mathrm{He}$ $(i \geq 2)$ system. As reported in [5], it follows from the mutual arrangement patterns of the quasi-molecular terms (Figure 4) that the probability of the transitions $3p_1 \rightarrow 3p_i$ $(i \geq 2)$ shall be low in collisions between $\mathrm{Ne}(2p^54p,\ 3p_1)$ and He atoms. The same paper reports that collisions

$$\mathrm{Ne}(2p^54p, 3p_i) + \mathrm{He}(1s^2\ ^1S_0) \xrightarrow{\sigma_{if}} \mathrm{Ne}(2p^54p, 3p_f) + \mathrm{He}(1s^2\ ^1S_0) \quad (1)$$

are much more effective for transitions between the group of $3p_4$, $3p_2$, $3p_5$ levels and the group of $3p_6$, $3p_7$, $3p_8$, $3p_9$ levels. This is supported by the experiment results (Table 1). Cross-sections σ_{if} achieve $10^{-16} - 10^{-15}\,\mathrm{cm}^2$ for the transitions $i = 2,\ 4 \rightarrow f = 2,\ \ldots,\ 10$.

It is worth mentioning here that, for the $2p^53p$ configuration just as for the $2p^54p$ configuration, transitions from the upper level to the lower $2p_1 \rightarrow 2p_2,\ 2p_3, \ldots, 2p_{10}$ levels are doubtful in thermal collisions between $\mathrm{Ne}(2p^53p)$ and He atoms: $\langle \sigma_{1f}(T \sim 300\,\mathrm{K}) \rangle \ll 10^{-16}\,\mathrm{cm}^2$, because the term of the $\mathrm{Ne}(2p_1) + \mathrm{He}$ system does not approach the terms of $\mathrm{Ne}(2p_i,\ i \geq 2) + \mathrm{He}$ [5].

The authors of this study suggest that the increase in the population of the $3p_1$ level with respect to the populations lying below the levels with the helium pressure growth in the helium-neon plasma may be caused by extremely low de-excitation rate of $\mathrm{Ne}(3p_1)$ in collisions with the He atoms due to the features of interaction between the $\mathrm{Ne}(2p^54p)$ and He atoms. It is estimated that, when the helium pressures exceed 3 Torr, the collisional mixing rate (1) starts exceeding the radiative decay rate of the $2p^54p$ states or

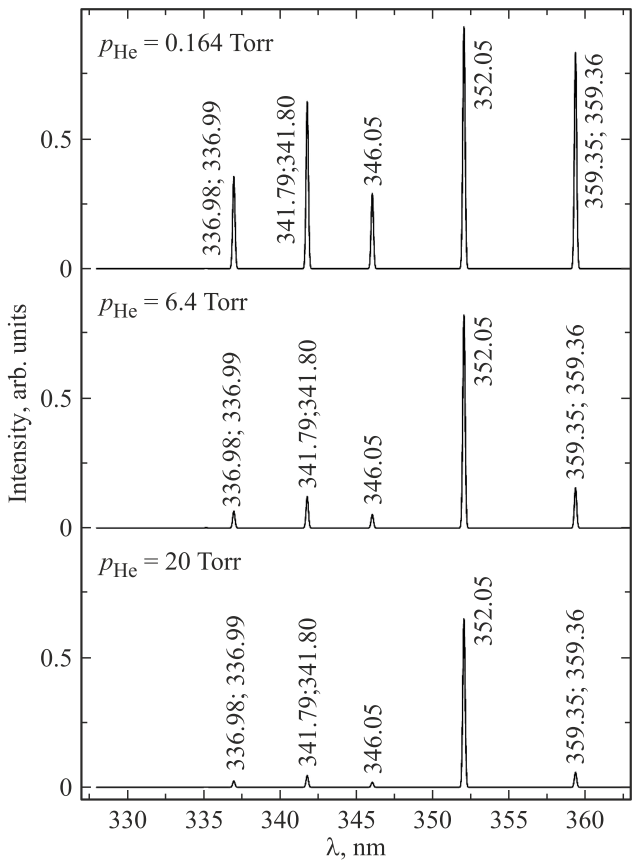

Figure 5. Results of numerical simulation of radiation spectra in the He–Ne mixture.

[1] Symbols $1s^2\ ^1S_0$ for the ground state of the He atom will be omitted below for clarity.

Table 1. Maxwell-distribution-averaged cross-sections $\langle\sigma_{if}(T\sim 350\,\mathrm{K})\rangle$ of the transitions $3p_i\to 3p_f$ in collisions (1) [5,8], ΔE_{4f} is the $3p_f$ energy level with respect to the $3p_4$ level

f	Paschen state	state, jl-coupling	ΔE_{4f}, eV	$\langle\sigma_{if}\rangle$, 10^{-16} cm^2	
				$i=4$	$i=2$
1	$3p_1$	(1/2)[1/2]0	0.0716	10^{-2}	10^{-2}
4	$3p_4$	(1/2)[3/2]2	0	–	10.6 ± 1.0
2	$3p_2$	(1/2)[1/2]1	−0.0001	7.25 ± 0.45	–
5	$3p_5$	(1/2)[3/2]1	−0.0064	6.72 ± 0.45	9.45 ± 0.90
3	$3p_3$	(3/2)[1/2]0	−0.0381	0.60 ± 0.03	$0.77-0.06$
6	$3p_6$	(3/2)[3/2]2	−0.0831	1.42 ± 0.02	2.22 ± 0.20
7	$3p_7$	(3/2)[3/2]1	−0.0863	0.86 ± 0.03	1.29 ± 0.10
8	$3p_8$	(3/2)[5/2]2	−0.1004	1.80 ± 0.03	2.18 ± 0.20
9	$3p_9$	(3/2)[5/2]3	−0.1089	1.98 ± 0.04	2.72 ± 0.25
10	$3p_{10}$	(3/2)[1/2]0	−0.1477	1.02 ± 0.05	0.72 ± 0.07

all levels, except the upper $3p_1$ level. According to [5] (Table. 1), the Maxwell-distribution-averaged cross-sections $\langle\sigma_{i1}\rangle$ of the excitation transfer to the $3p_1$ level are equal to 10^{-18} cm^2. The relevant rate constants may be calculated as follows

$$k_{q1}=\langle\sigma_{q1}\rangle\sqrt{\frac{8k_{\mathrm{B}}T}{\pi\mu}},$$

where $q=2$, 4; k_{B} is the Boltzmann constant; T is the gas temperature; μ is the reduced mass of colliding particles, in this case He and Ne atoms.

Rate constants k_{14}, k_{12} of the reverse transitions $3p_1\to 3p_4$, $3p_1\to 3p_2$ recalculated using the principle of detailed balance:

$$k_{1q}=k_{q1}\frac{g(3p_q)}{g(3p_1)}\exp\left[\frac{E(3p_1)-E(3p_q)}{k_{\mathrm{B}}T}\right], \qquad (2)$$

are still lower by 1 or 2 orders of magnitude than the rate constants of the transitions between the lower $3p_i$, $i=2,\ldots,10$ levels. In equation (2), $q=2$ or 4, $g(3p_q)$ and $g(3p_1)$ are the degeneracy multiplicities of the $3p_q$ and $3p_1$ levels, respectively, $E(3p_1)-E(3p_q)$ is the difference of the $3p_1$ and $3p_q$ energy levels.

To check whether it is possible to explain the observed selectivity of population of the $3p_1$ level, populations N_{3p_1}, N_{3p_4}, N_{3p_2} of three upper $3p_1$, $3p_4$, $3p_2$ levels of the $2p^54p$ configurations were calculated using the balance equations:

$$\Gamma_{3p_1}=-N_{3p_1}\left\{\Sigma_L A_{3p_1\to L}+\Sigma_f[\mathrm{He}]k_{1f}\right\}+N_{3p_4}[\mathrm{He}]k_{41}+N_{3p_2}[\mathrm{He}]k_{21}, \qquad (3a)$$

$$\Gamma_{3p_4}=-N_{3p_4}\left\{\Sigma_L A_{3p_4\to L}+\Sigma_f[\mathrm{He}]k_{4f}\right\}+N_{3p_1}[\mathrm{He}]k_{14}+N_{3p_2}[\mathrm{He}]k_{24}, \qquad (3b)$$

$$\Gamma_{3p_2}=-N_{3p_2}\left\{\Sigma_L A_{3p_2\to L}+\Sigma_f[\mathrm{He}]k_{2f}\right\}+N_{3p_1}[\mathrm{He}]k_{12}+N_{3p_4}[He]k_{42}, \qquad (3c)$$

$\Gamma_{3p_i}(i=1,\ 4,\ 2)$ are the population fluxes of the $3p_1$, $3p_4$, $3p_2$ states; $A_{3p_i\to L}$ are the probabilities of radiative transitions: $2p^54p$, $3p_i\to 2p^53s$; $2p^54p$, $3p_i\to 2p^54s$; $2p^54p$, $3p_i\to 2p^53d$ [9]; k_{if} are the process rate constants (1) $3p_i\to 3p_f$, [He] is the He atom concentration. Numerical simulation of radiation spectra was performed in three stages.

1. The experimentally recorded Ne atom radiation spectrum in the region 335–375 nm ($2p^54p\to 2p^53s$) at the He pressure $p_{\mathrm{He}}=0.164$ Torr and the probability of radiative transitions [9] were used to determine the populations N_{3p_1}, N_{3p_4}, N_{3p_2}.

2. The populations N_{3p_1}, N_{3p_4}, N_{3p_2} at $p_{\mathrm{He}}=0.164$ Torr, radiative transition probabilities [5] and excitation transfer cross-sections (1) [5,9] (Table 1) on the basis of the population balance equations ($3a-3c$) were used to determine the fluxes Γ_{3p_1}, Γ_{3p_4}, Γ_{3p_2} of population of the $3p_1$, $3p_4$, $3p_2$ levels. Distribution of population fluxes Γ_{3p_i} was recorded and used to calculate the population fluxes of the $3p_1$, $3p_4$, $3p_2$ levels at other He pressures.

3. The distribution of fluxes Γ_{3p_i} calculated for $p_{\mathrm{He}}=0.164$ Torr was used to determine the population of the $3p_1$, $3p_4$, $3p_2$ levels and the intensity of spectral lines emitted in transitions from these levels for other He pressures.

Table 2. Calculation results

Levels	Jl-coupling	Degeneracy multiplicity $2J+1$	Energy with respect to the $3p_4$, meV	N_{3p_i} $p_{\text{He}} = 0.164$ Torr	Γ_{3p_i}	N_{3p_i} $p_{\text{He}} = 6.4$ Torr	N_{3p_i} $p_{\text{He}} = 20$ Torr
$3p_1$	(1/2)[1/2]0	1	71.6	2314	$3.678 \cdot 10^{10}$	2037	1611
$3p_4$	(1/2)[3/2]2	5	0	11601	$10.21 \cdot 10^{10}$	2195	820
$3p_2$	(1/2)[1/2]1	3	−0.1	7312	$7.217 \cdot 10^{10}$	820	492

Table 2 lists the calculated population fluxes Γ_{3p_i} and populations N_{3p_i} in relative terms. Figure 5 shows the intensities of spectral lines emitted in transitions from the $3p_1$, $3p_4$, $3p_2$ levels that were calculated by the populations N_{3p_1}, N_{3p_4}, N_{3p_2} and probabilities of the radiative transitions $2p^5 4p \rightarrow 2p^5 3s$ [9].

Findings and conclusions

Comparison of spectra in Figure 3, *a, b, c* and Figure 5 indicates that the experimental and simulated spectra modification patterns with an increase in p_{He} are similar. In the authors' opinion, this is an argument in favor of the proposed mechanism of selective population of the $3p_1$ level.

The addressed depletion mechanism of the lower $2p^5 4p$ configuration levels, that, at the same time, does not cover the upper $3p_1$ level, shall be implemented exactly for the He-Ne mixture, but not for the pure Ne because it is considerably associated with the specifics of mutual arrangement of the $\text{Ne}(2p^5 4p) + \text{He}$ quasi-molecule terms. The pure Ne does not also exhibit the selective population of the $3p_1$ level. The 352.05 nm line doesn't differ fundamentally from other near-UV spectrum lines of Ne (Figure 3, *d*).

Conflict of interest

The authors declare that they have no conflict of interest.

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