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SOME REMARKS ON THE LASER TRANSITIONS AT 3071 nm, 5665 nm and 10353 nm in He-He ELECTRIC DISCHARGE

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The conditions for developing laser oscillations for the transitions $5s'[1/2]_1^0 \rightarrow 4p[1/2]_0$ 3071 nm, $4p[1/2]_0 \rightarrow 3d'[3/2]_1^0$ 10353 nm, $4p[1/2]_0 \rightarrow 3d[3/2]_1^0$ 5665 nm, and $4p[1/2]_0 \rightarrow 3d[1/2]_1^0$ 5326 nm have been discussed on the basis of induced changes in level populations. A great perturbation in population of the $4p[1/2]_0$ level induced by laser oscillation at 3071 nm has been obtained. An excitation flow between the $4p[1/2]_0$ and $4p'[3/2]_2$ levels has been analized.

The laser oscillations at 3391 nm, 3392 nm and 4218 nm are known to induce a very strong perturbation in the populations of neon levels, resonant at the laser frequency [1-4]. In this paper another strong laser perturbation induced by the transition $5s'[1/2]_1^0 \rightarrow 4p[1/2]_0$, 3071 nm, is reported. Though, that laser action is rather weak the increase in the $4p[1\ 2]_0$ level population, induced by the above mentioned laser oscillations, exceeds a factor of 3. It is obvious, that induced changes in level populations depend on population ratios in non-perturbed states: for example, very strong laser radiation at wavelength 633 nm induces in a neon electric discharge (depending on neon pressure) different changes in population of the $5s'[1/2]_1^0$ level. Those changes reach their maximal value at a neon pressure of 4 Tr, that is, 1300%! (factor 14).

The laser action at 3071 nm has been reported for the first time by Brunet and Laures [5]. Because we were not able to obtain that laser action with high intensity by method described in the above mentioned work, so in addition to a prism, a methane cell was inserted inside the laser cavity. Besides methane, as an absorber of the 3391 nm laser line, carbon dioxide was used to suppress laser oscillations at 4218 nm.

The experiments were performed with several laser tubes od different lengths, and Brewster windows made of sodium chloride, fused silica glass, and calcium chloride. These laser tubes (i.d. 8 mm) were filled with He and Ne at a ratio of 13:1 to a total pressure of 1.9 Tr. Two nearly plain mirrors, coated with aluminium on fused silica or calcium chloride substrata, formed a laser resonant cavity. Stimulated emission at 3071 nm

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was observed by means of a prism monochromator with a PbS-photocell; another grating monochromator with a photomultiplier was used to measure the intensities of spectral lines emitted perpendicular to the discharge tube.

Results

(a) Laser transition at 3071 nm

Action of the laser radiation at 3071 nm on excited neon atoms, inside laser tube, produces sizable changes in the populations of the $4p[1/2]_0$ and $5s'[1/2]^0_1$ levels. As seen from the data of Table I, those changes, when conditions for their saturation are fulfilled for a tube of length above 2000 mm, reach +250%, and — 14% for the $4p[1/2]_0$ and $5s'[1/2]^0_1$ levels, respectively. These data allowed one to estimate the population ratio of the levels at 1:12, see Fig. 1. All level populations given in Fig. 1 were estimated by this method, that is, assuming $(N_2/g_2)-(N_1/g_1) = 0$ when conditions of "saturation" were fulfilled.

TABLE I

Wavelengths of transitions in nm	Upper levels	Population changes in %	Lower levels	Population changes in %	Remarks and References
3071	$5s'[1/2]^0_1$	−14	$4p[1/2]_0$	+250	[5]
5665*	$4p[1/2]_0$	+151	$3d[3/2]^0_1$	+34	[13-15]. In [15] the wrong transition is given.
5326*	$4p[1/2]_0$	+141	$3d[1/2]^0_1$	+23	Action at this wavelength has been observed, but on another transition [15]?
10353	$4p[1/2]_0$		$3d'[3/2]^0_1$		
3774	$4p'[1/2]_0$		$3d[3/2]^0_1$		[13]
3620	$4p'[1/2]_0$		$3d[1/2]^0_1$		
5404	$4p'[1/2]_0$	−31	$3d'[3/2]^0_1$	+8.8	These are well known laser actions; their maximal population changes are given for exact calculations of population ratios.
4218	$5s'[1/2]^0_1$	−10.4	$4p'[1/2]_0$	+156	
3391	$5s'[1/2]^0_1$	−56	$4p'[3/2]_2$	+115	

* These laser actions were observed simultaneously with laser action at 3071 mm only — (CO_2 and CH_4 inside the laser cavity).

As described here this laser action has an analogy to another one at 4218 nm. Both transitions have a common upper level $5s'[1/2]^0_1$, but their lower levels differ from one another by a quanta number $j(2p)^5$ neon core. Now we will try to compare those laser transitions.

Let us begin from amplification coefficients. For that we shall use the formula [6],

$$k_0(\lambda) \sim \lambda^3 \gamma_{21} \cdot N_2 \left(1 - \frac{N_1}{N_2} \frac{g_2}{g_1}\right). \tag{1}$$

Here $k_0(\lambda)$ is the coefficient of amplification in the middle of a spectral line broadened by the Doppler effect; λ — wavelength of the spectral lines; γ_{21} — probability of transition $2 \rightarrow 1$; N_2, N_1 — populations of the upper and lower levels; g_2, g_1 — their level statistical weigths. Applying to the transitions at 4218 nm, and 3071 nm: this formula, given in Fig. 1 level populations, and respectively probabilities of transitions [7] we found, $k_0(4218) : k_0(3071) = 5$. So it results that when both laser action can occur, first develop oscillations at 4218 nm, and reducing the population of the $5s'[1/2]_1^0$ level, they suppress the laser action at 3071 nm. This was confirmed in the experiment; in the case of a laser set-up with only the methane cell within the resonator, no oscillations were observed at 3071 nm, but cascade ones at 4218 nm and 5404 nm were observed. However, if those at 4218 nm are removed by means of carbon dioxide, not only the laser action at 3071 nm, but two other at 5665 nm and 5326 nm appeared as well.

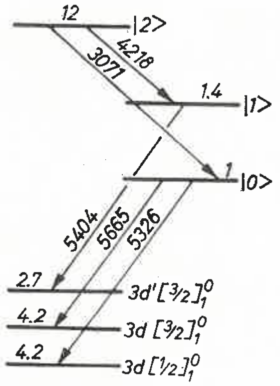

Fig. 1. Diagram of neon levels discussed in this paper. Numbers on the figure are relative populations of levels in the non-perturbed state

The next question is concerned with the ratio of decay constants of the $4p'[1/2]_0$ and $4p[1/2]_0$ levels. Populations of levels, when laser oscillation occur between them, are known to fulfill the relation [8],

$$\alpha_{21} = \frac{N_1 - N_1^0}{N_1^0} : \frac{N_2^0 - N_2}{N_2^0} = \frac{\gamma_2 - \gamma_{21}}{\gamma_1} \cdot \frac{N_2^0}{N_1^0}, \tag{2}$$

where N_l, N_l^0 — the level populations when laser oscillations $|2\rangle \rightarrow |1\rangle$ are present or absent, respectively; γ_l — decay constant of $|l\rangle$ level; γ_{21} — as in formula (1). In the following discussion the $5s'[1/2]_1^0$, $4p'[1/2]_0$ and $4p[1/2]_0$ levels are denoted by $|2\rangle$, $|1\rangle$, and $|0\rangle$ respectively.

Applying Eq. (2) to the laser transitions at 4218 nm and 3071 nm, and neglecting γ_{20}, γ_{21} in respect to γ_2 we obtain

$$\frac{\gamma_0}{\gamma_1} = \frac{\alpha_{21}}{\alpha_{20}} \cdot \frac{N_1^0}{N_0^0}. \tag{3}$$

Because $N_0^0 : N_1^0 = 1.4$; $\alpha_{21} : \alpha_{20} = 15 : 18$ [3], so the decay ratio is found to be $\gamma_0 : \gamma_1 = 1.2$.

It is easy to make sure, that radiative decay constants of the $|1\rangle$ and $|0\rangle$ levels fulfill the relation $\gamma_1^r > \gamma_0^r$ [9, 7] which is inconsistent with the previous result. These conflicting conclusions may be explained by collisional population transfer from the $4p[1/2]_0$ to $4p'[3/2]_2$, $4p'[3/2]_1$ and $4p'[1/2]_1$ levels, or vice versa. It is known that such a collisional population transfer for the $4p'[1/2]_0$ level does not occur [2], [10]. Some remarks on this effect will be presented in the next section.

(b) Collisional excitation transfer $4p[1/2]_0 \rightleftarrows 4p'[3/2]_2$

Information on this excitation flow can be found in works [1, 10]. The data presented there show that the population of the $4p[1/2]_0$ level increases by 10% when, due to oscillations at 3391 nm, the population of the $4p'[3/2]_2$ level increases by 100%. Because the discussed levels are not radiatively coupled to each other, this 10% increase may be caused by atomic collisions only. In the following discussion another neon level $4p'[3/2]_2$ will be considered. It is denoted by $|k\rangle$, coupled with level $|2\rangle$ by radiative transition, and with $|0\rangle$ — only by atomic collisions. We can write the number of atoms transfering between states $|k\rangle$ and $|0\rangle$, in unit volume and per unit time, by formula

$$Q_{k0} = \gamma_{k0} N_k - \gamma_{0k} N_0, \tag{4}$$

where γ_{k0}, γ_{0k} are the rates of the collisional processes $|k\rangle \to |0\rangle$ and $|0\rangle \to |k\rangle$. Using formula (4), and the balanced equations for the populations, we obtain for stationary conditions a relation analogous to Eq. (2) but with altered decay constants

$$\alpha_{20} = \frac{\gamma_2 - \gamma_{20}^*}{\gamma_0^*} \cdot \frac{N_2^0}{N_0^0}, \tag{5}$$

where

$$\gamma_{20}^* = \gamma_{20} + \frac{\gamma_{k0} \cdot \gamma_{2k}}{\gamma_k + \gamma_{k0}} \tag{6}$$

and

$$\gamma_0^* = \gamma_0 + \frac{\gamma_{0k} \cdot \gamma_k}{\gamma_k + \gamma_{k0}}. \tag{7}$$

Next, neglecting $\gamma_{21} : \gamma_2$ and $\gamma_{20} : \gamma_2$ we obtain

$$\frac{\gamma_0}{\gamma_1} = \frac{\alpha_{21}}{\alpha_{20}} \cdot \frac{N_1^0}{N_0^0} \cdot \frac{1 - \dfrac{\gamma_{k0}}{\gamma_k + \gamma_{k0}} \cdot \dfrac{\gamma_{2k}}{\gamma_2}}{1 + \dfrac{\gamma_{0k}}{\gamma_k + \gamma_{k0}} \cdot \dfrac{\gamma_k}{\gamma_0}}. \tag{8}$$

The new formula (8) differs from (3) by a factor below unity, so real ratio decay constants are less than the obtained value 1.2.

Besides formula (5) another one, for the case when level $|2\rangle$ changes its population may be derived, but there are no laser oscillations which end on levels $|k\rangle$ and $|0\rangle$, or which might produce a cascade into them. This formula is

$$\beta_{20} = \frac{N_0^0 - N_0}{N_0^0} : \frac{N_2^0 - N_2}{N_2^0} = \frac{\gamma_{20}^*}{\gamma_0^*} \cdot \frac{N_2^0}{N_0^0}. \tag{9}$$

Dividing (5) by (9) we can write

$$\frac{\gamma_2}{\gamma_{20}^*} = \frac{\alpha_{20}}{\beta_{20}} + 1. \tag{10}$$

A formula similar to (10) is known to have been derived in work [10], but the above takes into account collision effects of neon atoms in states $|k\rangle$, $|0\rangle$ exactly. Because the $4p'[3/2]_2$ level is strongly coupled, due to atomic collisions, with other levels $4p$, the discussion of results from the above work should be repeted.

After this digression, let us return to the main subject. Let us try to find the rates γ_{k0} and γ_{0k}. With this object in view, consider a case when the population of level $|2\rangle$ changes, but the population of level $|0\rangle$ does not. That case may be realized when very strong laser oscillations on transitions $|2\rangle \rightarrow |k\rangle$ and $|k\rangle \rightarrow |l\rangle$ can be obtained simultaneously; $|l\rangle$ is one of the $3d$ or $4s$ levels. The balanced equations for populations in stationary conditions are (see Fig. 2):

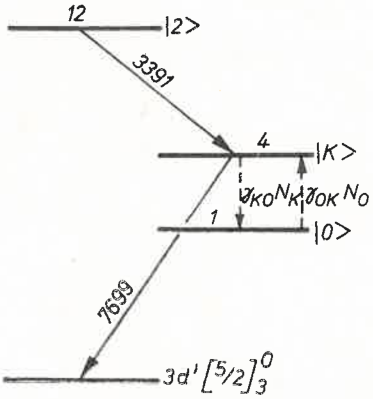

Fig. 2. The neon level diagram "used to derive" formula (11). Numbers on the figure represent the relative populations of levels in the non-perturbed state

$$\gamma_0 N_0^0 = \gamma_{20} N_2^0 + \gamma_{k0} N_k^0 - \gamma_{0k} N_0^0 + R_0$$

when there is no perturbation, and

$$\gamma_0 N_0 = \gamma_{20} N_2 + \gamma_{k0} N_k - \gamma_{0k} N_0 + R_0$$

when there is a perturbation. Solving these equations, and substituting $N_0 - N_0^0 = 0$, we get

$$\gamma_{k0} = \gamma_{20} \cdot \frac{N_2^0}{N_k^0} \cdot \frac{\dfrac{N_2^0 - N_2}{N_2^0}}{\dfrac{N_k - N_k^0}{N_k^0}} . \tag{11}$$

In the experiment the intensities of laser actions at 3391 nm and 7699 nm were chosen in such a manner that changes in population of the $4p[1/2]_0$ level vanishes (see Table II). Because, $N_2^0 : N_k^0 = 4$ and $\gamma_{20} = 1.07 \cdot 10^5\ \mathrm{s}^{-1}$ [7] so, γ_{k0} is found to be: $\gamma_{k0} = 5.1 \cdot 10^5\ \mathrm{s}^{-1}$. The second rate γ_{0k} can be easily obtained by means of equation,

$$\frac{\gamma_{0k}}{\gamma_{k0}} = \frac{g_k}{g_0} \exp\left(- \frac{E_k - E_0}{kT} \right),$$

which results from the principle of detailed balancing [11].

TABLE II

Wavelengths of transitions in nm	Level symbols	Population changes in %	Levels symbols	Population changes in %	Level symbols	Population changes in %
4218 and 5404	$5s'[1/2]_1^0$	-20	$4p'[1/2]_0$	$+4$	$3d'[3/2]_1^0$	$+57$
3391 and 7699	$5s'[1/4]_1^0$	-52	$4p'[3/2]_0$	$+44$	$4p[1/2]_0^*$	0

*This level is not the lower one of cascade transition at 3391 nm, 7699 nm.

For the $4p'[3/2]_2$ and $4p[1/2]_0$ levels — $g_0 : g_k = 1 : 5$, $(E_k - E_0) : kT \sim 1$ [4] are fulfilled, so we obtain, $\gamma_{k0} : \gamma_{0k} = 0.54$. Detailed calculations, based on the last result, show, that the resultant excitation flow (4) changes its direction when the population of the $4p[1/2]_0$ level increases by 110%, due to laser oscillations at 3071 nm. This fact has confirmation in experimental data. However, the sign of population changes of the $4p[1/2]_0$ level remains constant; it does not depend on the intensity of laser action at 3071 nm, because it is determined by rates γ_{0k}, $\overset{*}{\gamma}_0$, $\overset{*}{\gamma}_{20}$ and γ_{2k}.

(c) Cascade laser transitions

In the experiment we have obtained two other laser oscillations for the transitions at 5665 nm, $4p[1/2]_0 \rightarrow 3d[3/2]^0$ [13–15], and 5326 nm, $4p[1/2]_0 \rightarrow 3d[1/2]_1^0$. Those laser oscillations developed only when the population of the $4p[1/2]_0$ level was strongly increased by laser action at 3071 nm.

It seems, that laser action at 10353 nm, $4p[1/2]_0 \rightarrow 3d'[3/2]_1^0$ can be obtained as well, particularly when laser oscillations at 3071 nm occur. To confirm that we shall compare the amplification coefficients of laser transitions at 10353 nm and 4218 nm. Using for that purpose: formula (1), data from Fig. 1 and from [7] we get:

$$k_0(4218) : k_0(10353) = \left(\frac{4218}{10353}\right)^{3} \cdot \frac{2.63}{0.281} \cdot 12 \cdot \frac{1 - \frac{1.4}{4}}{1 - \frac{2.7}{3}} = 49,$$

when level $4p[1/2]_0$ is non-perturbed, and 2.4 when its population increases three times. It means that under non-perturbed conditions, in order to achieve laser oscillations at 10353 nm the long tube — above 9 m — has to be used [16]. The conditions of lasering on this transition radically improve when laser oscillations at 3071 nm develop. Then, a tube of length 500 mm is suficient for obtaining the laser action discussed above. However, we did not manage to obtain that lasering, because the windows made of sodium chloride suppressed the laser action at 3071 nm, and the tube was only 1.5 m long.

Among cascade laser transitions originating from level $4p'[1/2]_0$ we observed only one, however well known, at 5404 nm. The remaining laser actions, that is on transitions $4p'[1/2]_0 \rightarrow 3d[3/2]_1^0$ and $4p'[1/2]_0 \rightarrow 3d[1/2]_1^0$ were not observed. Efforts at obtaining those laser actions failed even when the laser oscillations at 4218 nm occur. It was because in this case laser oscillation at 5404 mn developed, and they reduced the population of the $4p'[1/2]_0$ level nearly to its non-perturbed value (see Table II).

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