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POPULATION CHANGES OF Ne LEVELS INDUCED IN He—Ne MIXTURE BY LASER ACTIONS IN THE INTERMEDIATE IR. PART II. THE MUTUAL INFLUENCE OF LASER ACTIONS INTERACTING IN 3.3913 μm AND 7.6994 μm, AND 4.218 μm AND 5.403 μm

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The investigation of relative changes in the intensity of selected spontaneously emitted Ne lines in a He—Ne laser, when the laser actions were switched on, interacting in cascade, made it possible to analyse the mutual influence of these actions. This influence is quantitatively discussed for the changes in population ratio for levels involved in transitions, for gain factors and pumping rate constants as a result of stimulated transitions.

It is possible to evaluate some atomic parameters of the medium (see Part I, [1] quoted hereafter as I) when investigating induced changes in level populations and using balance equations. This is not the only information which one can get from that kind of investigations. Following these changes it is possible e.g. to study the mutual influence of the laser actions generated simultaneously, moreover, one may identify the laser transitions.

In the experimental set-up whose description and scheme are shown in I (under the optimum conditions for stimulated transition 0.63 μm), a laser action has been obtained. At first it was only known that its wavelength exceeds that for the 7.6994 μm oscillation $4p'[3/2]_2 \rightarrow 3d'[5/2]_3^0$ — Fig. 1. This conclusion follows from the geometry of the set-up namely from the rotation angle of the resonance cavity mirror at the prism side needed for changing the 7.6994 μm generation into the obtained one. In order to identify the transition we have studied the relative population changes, for levels of the $4p$ and $3d$ groups[1], observed by switching this transition on and/or off. The results are presented in

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[1] All levels with the same principal and orbital quantum numbers of the valence electron belong to this group. The symbols of the level groups are shown in Fig. 1.

Table I, in the third column. Only the levels showing the significant changes are gathered in the table. One can see from the presented results that the biggest population changes were observed for the $4p'[3/2]_1$ and $3d'[5/2]_2^0$ levels. There is no doubt that the laser action was obtained for $4p'[3/2]_1 \rightarrow 3d'[5/2]_2^0$ transition — Fig. 1. It follows from reference [2] that the wavelength of this transition is 8.0066 μm and was obtained for the first time by the authors of papers [3–5].

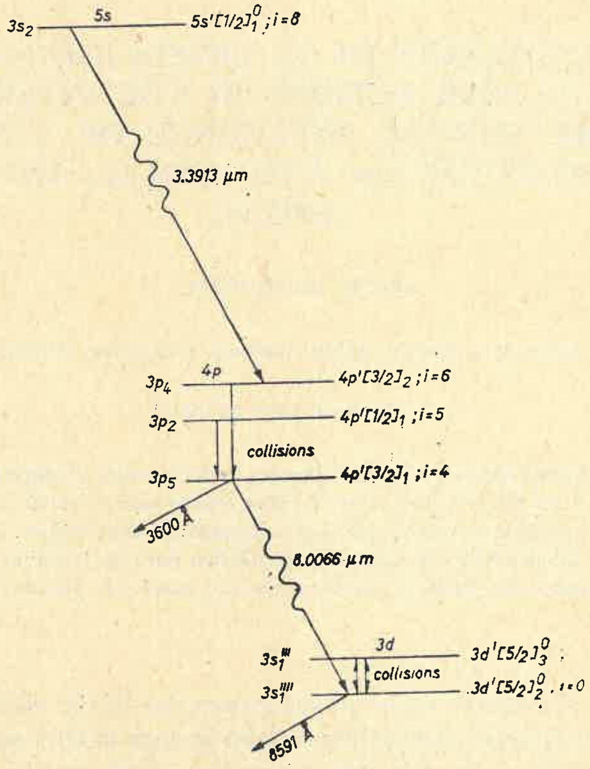

Fig. 1. The scheme of excitation for stimulated transition on 8.0066 μm: levels are labeled in Paschen and Racah notation [11]; wavy lines, transitions in the stimulated emission; solid lines, transitions in the spontaneous emission

In this case the transition was excited in the presence of a cooperating action at 3.3913 μm ($5s'[1/2]_1^0 \rightarrow 4p'[3/2]_2$). There is no typical example of a cascade transition (as e.g. in I) since the upper laser level $4p'[3/2]_1$ is pumped by an excitation transfer from the $4p'[3/2]_2$ level in an atomic collision process and it is not the result of a direct radiative transition. Because of a very small energy separation these levels are very strongly coupled as one may see from the results of paper [6] which are in good agreement with ours, presented in Table I. The collisional excitation transfer $4p'[3/2]_1$ level occurs likewise from the $4p'[1/2]_1$ level. Let us notice, that the change in $4p'[1/2]_1$ equal to 109 % is associated with the excitation of laser action 3.3913 μm causing a strong increase of the $4p'[3/2]_2$ level population (+112 %). The levels $4p'[3/2]_2$ and $4p'[1/2]_1$ are very close to each other —

TABLE I

Investigated level in Racah notation	Relative change (in per cent) of the level population induced by laser action:		Transition assignement and wavelength of spontaneously emitted line used as indicators of the population change [Å]
	3.3913 μm $(N^{b}_{ik}-N^{a}_{i})/N^{a}_{i}$	8.0066 μm in presence of 3.3913 μm $(N^{c}_{im}-N^{b}_{ik})/N^{b}_{ik}$	
$4p'[3/2]_2$	+112.0	− 4.0	3594* $4p'[3/2]_2 \rightarrow 3s'[1/2]^0_1$
			3594* $4p'[1/2]_1 \rightarrow 3s'[1/2]^0_1$
$4p'[1/2]_1$	+109.0	− 5.2	3461 $4p'[1/2]_1 \rightarrow 3s'[1/2]^0_0$
$4p'[3/2]_1$	+ 78.5	−13.0	3600 $4p'[3/2]_1 \rightarrow 3s'[1/2]^0_1$
$3d'[3/2]^0_1$	+ 12.1	weak increase	8118 $3d'[3/2]^0_1 \rightarrow 3p[3/2]_1$
$3d'[3/2]^0_2$	+ 12.5	weak increase	8259 $3d'[3/2]^0_2 \rightarrow 3p[3/2]_2$
$3d'[5/2]^0_3$	+ 11.8	+ 3.9	7943* $3d'[5/2]^0_3 \rightarrow 3p[5/2]_2$
			7944* $3d'[5/2]^0_2 \rightarrow 3p[5/2]_2$
$3d'[5/2]^0_2$	+ 13.7	+13.2	8591 $3d'[5/2]^0_2 \rightarrow 3p'[3/2]_1$

$(N^{b}_{ik}-N^{a}_{i})/N^{a}_{i}$ — the relative change in population of "i" level induced by 3.3913 μm laser action in comparison to level population in absence of laser actions

$(N^{c}_{im}-N^{b}_{ik})/N^{b}_{ik}$ — the relative change in population of "i" level induced by laser actions 3.3913 μm and 8.0066 μm generating simultaneously in comparison to level population in presence of cooperating laser action only

* the asterisk indicates the line used as an indicator of the population change, overlapping with the neighbour line

the distance is 0.9 cm^{-1}. From [7] where the ratio of gain factors for laser transitions $5s'[1/2]^0_1 \rightarrow 4p'[3/2]_2$ and $5s'[1/2]^0_1 \rightarrow 4p'[1/2]_1$ was obtained (level symbols as in Fig. 1)

$$k_{86}/k_{85} = 2.7$$

one can conclude, that at a pressure of about 1 Tr there is the detailed thermal equilibrium between the levels mentioned above. It follows, from the quoted value, that the ratio of the level population for $4p'[3/2]_2$, $4p'[1/2]_1$ unperturbed by a laser action is (the symbols are the same as in I)

$$N^{a}_{6}/N^{a}_{5} = 1.76$$

and is approximately to the ratio of statistical weights

$$N^{a}_{6}/N^{a}_{5} \approx g_6/g_5 = 1.67.$$

Therefore the population of the upper level for the laser action 8.0066 μm increases equally by the excitation transfer from both the $4p'[3/2]_2$ and $4p'[1/2]_1$ levels. From the ratio of the line of 3600 Å ($4p'[3/2]_1 \rightarrow 3s'[1/2]^0_1$) intensities with and without 3.3913 μm laser action we obtain (the symbols are the same as in I)

$$N^{b}_{4k}/N^{a}_{4} \approx 1.8.$$

However, the increase of population inversion for laser levels in the case of the laser action at 8.0066 μm is not yet obvious. Following the increase of the $4p'[3/2]_2$ level population, the one for $3d'[5/2]^0_2$ level also increases as a result of direct spontaneous transitions $4p'[3/2]_2 \rightarrow 3d'[5/2]^0_2$. The same occurs for the $3d'[5/2]^0_3$ level which is coupled with $3d'[5/2]^0_2$ by collisions (Part III). But this increase is rather small as can be deduced from the intensity change of the 8591 Å line ($3d'[5/2]^0_2 \rightarrow 3p'[3/2]_1$)

$$N^{\mathrm{b}}_{0k}/N^{\mathrm{a}}_0 \approx 1.1.$$

The population ratio for $4p'[3/2]_1$ and $3d'[5/2]^0_2$ levels increases as a result of switching on the 3.3913 μm laser action

$$N^{\mathrm{b}}_{4k}/N^{\mathrm{b}}_{0k} \approx 1.6 N^{\mathrm{a}}_4/N^{\mathrm{a}}_0.$$

An obtained laser action was weak (generation threshold 0.6) and it was observed merely in the presence of the 3.3913 μm oscillation. This may be caused by a decrease in the transmission of the fluorite window and prism in this spectral region.

The mutual influence of laser actions interacting in the cascade 3.3913 μm ($5s'[1/2]^0_1 \rightarrow 4p'[3/2]_2$) and 7.6994 μm ($4p'[3/2]_2 \rightarrow 3d'[5/2]^0_3$) as well as 4.218 μm ($5s'[1/2]^0_1 \rightarrow 4p'[1/2]_0$) and 5.403 μm ($4p'[1/2]_0 \rightarrow 3d'[3/2]^0_1$) has been studied in the experimental set-up mentioned before. A scheme of the levels and transitions between them for the first and second cascade is shown in Fig. 2 and 3, respectively. The additional mechanism

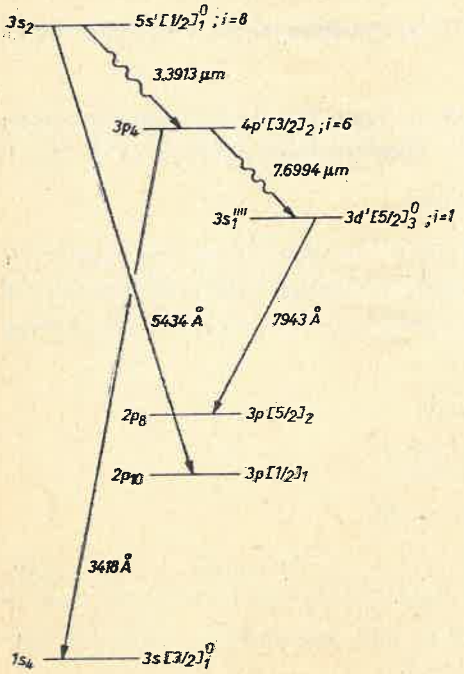

Fig. 2

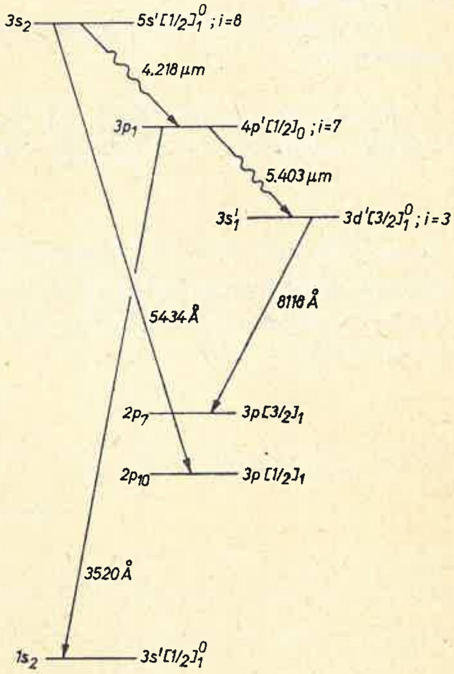

Fig. 3

Fig. 2. The diagram of levels and transitions between them which are connected with cascade $5s'[1/2]^0_1 \rightarrow 4p'[3/2]_2 \rightarrow 3d'[5/2]^0_3$

Fig. 3. The diagram of levels and transitions between them which are connected with cascade $5s'[1/2]^0_1 \rightarrow 4p'[1/2]_0 \rightarrow 3d'[3/2]^0_1$

of pumping for the upper levels $4p'[3/2]_2$, $4p'[1/2]_0$ of the laser transitions 7.6994 μm and 5.403 μm is connected with 3.3913 μm and 4.218 μm laser transitions. They cause the increase in population inversion. The influence of pumping transition on lower laser action in cascade can be presented quantitatively (as in the case of the 8.0066 μm action) by means of changing the population ratio for levels involved in a given laser transition caused by switching the cooperating action on and off.

It is known (I) that the population ratio for levels connected with the 7.6994 μm transition equals

$$N^{b}_{6k}/N^{b}_{1k} = 1.72 \pm 0.08 \tag{II.1}$$

in presence of the 3.3913 μm laser action.

On the other hand the same population ratio calculated, in the absence of the mentioned laser actions, in the same way as in relation (II.1) is

$$N^{a}_{6}/N^{a}_{1} = 0.87 \pm 0.04. \tag{II.2}$$

One can see that 3.3913 μm laser action almost doubles the population ratio for levels 6 and 1. When we divide (II.1) by (II.2) we get

$$N^{b}_{6k}/N^{b}_{1k} \approx 1.98 N^{a}_{6}/N^{a}_{1}.$$

Let us compare these results with the one obtained from the intensity ratio for 7943 Å ($3d'[5/2]^0_3 \rightarrow 3p[5/2]_2$) and 3418 Å ($4p'[3/2]_2 \rightarrow 3s[3/2]^0_1$ lines under the same condition i.e. with and without 3.3913 μm laser action

$$N^{b}_{6k}/N^{b}_{1k} \approx 1.96 N^{a}_{6}/N^{a}_{1}.$$

The results (II.1) and (II.2), as we remember, were obtained under an assumption that the saturation condition of level population for $4p'[3/2]_2$ and $3d'[5/2]^0_3$ were fulfilled during 7.6994 μm oscillation in the presence of the 3.3913 μm one. This good agreement shows once more that the saturation condition is valid within a good approximation.

The inequality follows from the threshold condition (when we neglect all possible losses: reflection, absorption, diffraction)

$$N^{a}_{6}/N^{a}_{1} \geqslant g_6/g_1 = 0.72. \tag{II.3}$$

One can expect, comparing (II.3) with (II.2), that the oscillation 7.6994 μm is possible even without cooperation with 3.3913 μm. This fact has been confirmed experimentally — see Table II.

The positive influence of the pumping transition 3.3913 μm on 7.6994 μm is clear when we compare gain factors for this transition obtained with the cooperating action switched on and off. For that reason the formula describing gain factors at the center of the line $4p'[3/2]_2 \rightarrow 3d'[5/2]^0_3$ ($6 \rightarrow 1$) under the assumption of doppler distribution [12] was used

$$k_{61} = \frac{\lambda^2_{61}\gamma_{61}\sqrt{\ln 2}}{4\pi^{3/2}\Delta\nu_{61}}\left(N^{a}_{6} - \frac{g_6}{g_1}N^{a}_{1}\right),$$

where $\Delta\nu_{61}$ is the half-width of a line, k_{61} — gain factor for the 7.6994 μm transition. The gain factor for this transition in the presence of cooperative action is denoted by k'_{61}. Then

$$\frac{k'_{61}}{k_{61}} = \frac{N^{b}_{1k}}{N^{a}_{1}}\left(\frac{N^{b}_{6k}/N^{b}_{1k}-g_6/g_1}{N^{a}_{6}/N^{a}_{1}-g_6/g_1}\right).$$

Substituting (II.1), (II.2), $g_6/g_1 = 5/7$ and N^{b}_{1k}/N^{a}_{1}, the latter obtained from intensities of lines originating from level -1, into the above relation we get

$$k'_{61}/k_{61} = 7.5 \pm 0.5.$$

So, switching on the generation 3.3913 μm causes that the gain factor increases 7.5 times for the 7.6994 μm oscillation.

The influence of pumping action is visualized by the changes in populations of laser levels for the supported transition — Table II. As one can see from the data presented there, the laser transitions 7.6994 μm and 5.403 μm are very weak without cooperative action. The changes in level population one order of magnitude lower than those observed in the presence of cooperative action, as well as the small values of gain factors [8] good evidence for the proposed mechanism.

TABLE II

Investigated level in Racah notation	Relative change (in per cent) of the level population induced by laser action:				Transition assignement and wavelength of spontaneously emitted line used as indicators of the population change [Å]
	7.6994 μm		5.403 μm		
	in presence of 3.3913 μm	in absence of 3.3913 μm	in presence of 4.218 μm	in absence of 4.218 μm	
$4p'[3/2]_2$	-32.6	-6.4			$3594^*(4p'[3/2]_2 \rightarrow 3s'[1/2]^{0}_{1})$ $3594^*(4p'[1/2]_1 \rightarrow 3s'[1/2]^{0}_{1})$
$3d'[5/2]^{0}_{3}$	$+47.0$	$+4.2$			$7943^*(3d'[5/2]^{0}_{3} \rightarrow 3p[5/2]_2)$ $7944^*(3d'[5/2]^{0}_{2} \rightarrow 3p[5/2]_2)$
$4p'[1/2]_0$			-47.0	-12.6	$3520(4p'[1/2]_0 \rightarrow 3s'[1/2]^{0}_{1})$
$3d'[3/2]^{0}_{1}$			$+54.0$	$+\ 4.4$	$8118\ (3d'[3/2]^{0}_{1} \rightarrow 3p[3/2]_1)$

It seems, however, that the generation 5.403 μm is stronger than the 7.6994 μm one. This also follows from the fact that this transition in the absence of the 4.218 μm laser action may be more easily excited than the 7.6994 μm one without cooperative action. It is very difficult in practice to adjust the mirrors in the resonance cavity to obtain 7.6994 μm action without the use of cooperative action, hence one should do it in the presence of the 3.3913 μm generation. The 5.403 μm transition also has a lower generation threshold, $N^{a}_{7}/N^{a}_{1} = g_7/g_1 = 0.33$, it is even lower than for the 3.3913 μm transition (0.6).

The minimum length of an active medium for which the given laser transitions may oscillate are defined in paper [8]. One can see from the table presented there how strongly this length may be reduced in the presence of cooperating action. The length required for the 7.6994 μm action without cooperating action ought to be approximately 0.78 m, while with that action a length of about 0.2 m is sufficient. For comparison, the same length for the strongest transition in the He—Ne laser — 3.3913 μm is 0.03 m only.

Now let us proceed to discussion whether the presence of the 7.6994 μm oscillation has positive influence on 3.3913 μm too. The atoms are transferred from the $4p'[3/2]_2$ level to the $3d'[5/2]^0_3$ level as a result of this transition. It causes the increase in population ratio for levels $5s'[1/2]^0_1$ and $4p'[3/2]_2$. In order to estimate this influence, let us consider how the pumping rate constant changes due to the stimulated emission for the 3.3913 μm transition in the presence of 7.6994 μm action. Dividing side by side in the equations written in I [1]: (I.1)—(I.4) and (I.1)—(I.7) we obtain the relation between the constants R^{b}_{86} and R^{c}_{86}

$$R^{c}_{86} = \frac{N^{a}_{8}-N^{c}_{8}}{N^{a}_{8}-N^{b}_{8}} R^{b}_{86}.$$

After taking into account the experimental data we have

$$R^{c}_{86}/R^{b}_{86} = 1.19 \pm 0.06. \tag{II.4}$$

Hence, the presence of the oscillation at 7.6994 μm only slightly changes the pumping rate constant for a transition at the wavelength 3.3913 μm.

It is quite different for a cascade transition $5s'[1/2]^0_1 \rightarrow 4p'[1/2]_0 \rightarrow 3d'[3/2]^0_1$. Switching on the 5.403 μm oscillation causes the pumping constant for the transition at the wavelength 4.218 μm to increase 2.5 times. One can explain the fact that the number of transitions between the $5s'[1/2]^0_1$ and $4p'[1/2]_0$ levels exceeds twice the usual as follows. When the 5.403 μm oscillation is additionaly excited then

(1) The population inversion between these two levels increases. The number of transitions, connected with a given mode excited in line (4.218 μm), is proportional to the population inversion [9, 10]

$$R_s = \delta u(\nu)\,[B_{87}\delta N_8 - B_{78}\delta N_7] = \delta u(\nu) B_{87}\left[\delta N_8 - \frac{g_8}{g_7}\delta N_7\right]$$

where $\delta u(\nu)$ is the energy density in a given mode, δN_8, δN_7 — the number of atoms interacting with radiation of the resonance frequency (excited mode frequency).

(2) The gain increases. Therefore the number of oscillating modes may also increase. The gain without the 5.403 μm action was too low to compensate for the losses in these modes. It is important to notice that the generation threshold for the transition 4.218 μm (when we neglect all losses) is very high $N^{a}_{8}/N^{a}_{7} = 3$. It may be the reason why in the absence of 5.403 μm only a few modes are excited. Such an interpretation could be applied to the cascade at 3.3913 μm and 7.6994 μm also. In that case, however, the population inversion and gain factor are so high and the generation threshold for 3.3913 μm is so low (0.6) that one can easily neglect the influence of the additional emptying of the $4p'[3/2]_2$ level by the 7.6994 μm transition.

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