POPULATION CHANGES OF Ne LEVELS INDUCED IN He—Ne MIXTURE BY LASER ACTIONS IN THE INTERMEDIATE IR. PART I. THE EVALUATION OF Ne ATOM PARAMETERS IN He—Ne LASERING MEDIUM

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The measurements of the relative changes in the intensity of selected spontaneously emitted Ne lines in a He—Ne mixture, when the laser actions were switched on or off, made possible the evaluation of the ratio of the total decay constants for the populations of $4p'[3/2]_2$ and $3d'[5/2]^0_3$ levels and the probability of the spontaneous $4p'[3/2]_2 \rightarrow 3d'[5/2]^0_3$ transition.

1. Introduction

The methods for evaluating the atomic transition probabilities in the lasering medium and the relative population numbers for the corresponding levels based on the measurement of the intensity changes in spontaneously emitted radiation, when specified laser actions are switched on and/or off, were developed successively by different authors (for example [1–3]). Lis ([4, 5]) has considered the case when several laser oscillations in Ne (stimulated alternatively) have a common upper level. In the present paper a similar treatment was generalized for the case when two laser oscillations have a common intermediate level (in other words: when they can cooperate in a cascade).

The main idea of this method consists in the study of relative population changes of levels induced by stimulating laser oscillations. For a theoretical description of those changes, the balance equations are applied connecting the populations of the corresponding levels, transition probabilities and pumping rates. In the simplest case we take into account two levels only (also only a single laser oscillation). It is obvious that when it is switched on, simultaneously the population of the upper level decreases while that of the lower level increases, depending on the intensity of the laser action. The effect of population changes is reflected by the corresponding intensity changes of spontaneously emitted lines beginning at the discussed levels.

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The effect of this change however is not limited to the levels involved directly in the corresponding transition but it can be transferred to the other levels by radiative or non-radiative transitions.

2. *Three level set*

Let us consider a He—Ne laser with the broad-band resonator. Even, when adjusted to the optimal conditions for the laser oscillation at 0.63 μm one can ascertain that in the spectrum emitted by it several other stimulated transitions (particulary the 3.39 μm transition) are present independently of the 0.63 μm transition. Therefore the cascade transitions can be observed. Now we proceed to a discussion of the case of a two-step cas-

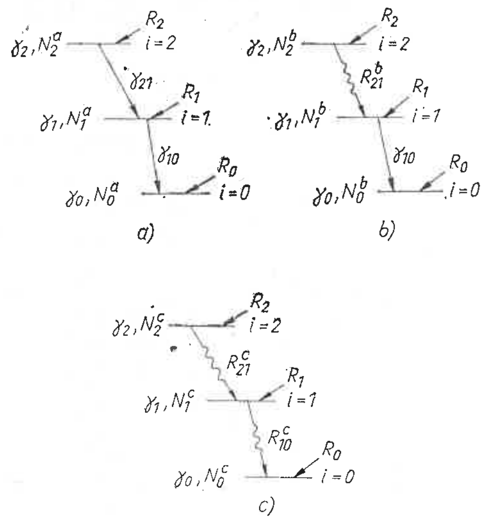

Fig. 1. Simplified level schema for a two step cascade: solid lines — transitions in the spontaneous emission, wavy lines — transitions in the stimulated emission; (a), (b), (c) correspond to the three cases discussed. Meaning of the symbols is explained in the text

cade between $2 \to 1$ and $1 \to 0$ levels. Fig. 1a, 1b, and 1c visualize three possibilities how different transitions can combine in such system, so we will have:

(a) absence of laser actions (spontaneous transitions only)
(b) presence of laser action $2 \to 1$
(c) presence of laser actions $2 \to 1$ and $1 \to 0$

The corresponding balanced equations for the level populations in the stationary state are: in case (a):

$$R_2 = \gamma_2 N_2^a, \tag{I.1}$$ [1]

$$R_1 + \gamma_{21} N_2^a = \gamma_1 N_1^a, \tag{I.2}$$

$$R_0 + \gamma_{10} N_1^a = \gamma_0 N_0^a. \tag{I.3}$$

[1] This work consists of three parts. To distinguish the particular parts, display formulae are numbered the supplementary Roman indices. Following this convention for notation (I.1) and so on will have the meaning of formulae 1 in part I.

in case (b):

$$R_2 = \gamma_2 N_2^b + R_{21}^b, \tag{I.4}$$

$$R_1 + R_{21}^b + \gamma_{21} N_2^b = \gamma_1 N_1^b, \tag{I.5}$$

$$R_0 + \gamma_{10} N_1^b = \gamma_0 N_0^b. \tag{I.6}$$

in case (c):

$$R_2 = \gamma_2 N_2^c + R_{21}^c, \tag{I.7}$$

$$R_1 + R_{21}^c + \gamma_{21} N_2^c = \gamma_1 N_1^c + R_{10}^c, \tag{I.8}$$

$$R_0 + \gamma_{10} N_1^c + R_{10}^c = \gamma_0 N_0^c. \tag{I.9}$$

Here N_i^a, N_i^b, N_i^c (i = 0, 1, 2) — are numbers of atoms per unit volume in the level i for each of the cases (a), (b), (c). R_i — the pumping rate of the level i ($i = 0, 1, 2$), i.e. the number of atoms per unit time and volume arriving at the given state as a result of collisions with electrons, He atoms and so on. γ_i — the decay constant of the i level population resulting from both: radiative and nonradiative transitions. γ_{21}, γ_{10} — the probabilities of the radiative spontaneous transitions $2 \to 1$, $1 \to 0$, respectively. R_{21}^b, R_{21}^c, R_{10}^c — pumping rates of the levels 1 or 0 caused by the laser actions: R_{21}^b — from the level 2 or 1 induced by the laser action $2 \to 1$, R_{21}^c — from the level 2 to 1 induced by the laser action $2 \to 1$, in the presence of the simultaneous action $1 \to 0$ and: R_{10}^c — from the level 1 to 0 as a result of the laser action $1 \to 0$ in the presence of the stimulated transition $2 \to 1$; respectively.

In (I.1)—(1.9) one assumes that the R_i's are constant and independent of the laser action intensity. This assumption is consistent with the results reported in Schiffner's [6], Garscadden's [7], and Yano's [8] papers, concerning the change in the discharge current intensity induced by the laser action (therefore indirectly involving conclusions about R_i). The mentioned effect depending on the gas pressure in the discharge tube was very weak and could be neglected. Other parameters, which could influence the values R_i like the gas pressure, were kept constant during the whole experiment.

From the equations (I.1)—(I.9) we deduce the ratio of the decay constant of the level population — γ_1 to the probability of the spontaneous $1 \to 0$ transition — γ_{10}

$$\frac{\gamma_1}{\gamma_{10}} = \frac{\dfrac{N_0^a - N_0^c}{N_0^a - N_0^b} - \dfrac{N_1^a - N_1^c}{N_1^a - N_1^b}}{\dfrac{N_2^a - N_2^c}{N_2^a - N_2^b} - \dfrac{N_1^a - N_1^c}{N_1^a - N_1^b}}. \tag{I.10}$$

Values of the relative population changes $-\dfrac{N_0^a - N_0^c}{N_0^a - N_0^b}$ and so on, were evaluated directly from the intensity changes of the corresponding lines emitted spontaneously.

3. *Experimental set-up*

The intensity changes of lines in spontaneous emission has been measured in the standard system shown in Fig. 2. The lines emitted spontaneously were observed perpendicularly to the laser axis. The radiation was focussed on the entrance slit of the SPM-2 grating monochromator. The intensity of selected lines was measured by means of the EMI 9659QA photomultiplier and sensitive galvanometer (10^{-9} A/division). The photo-

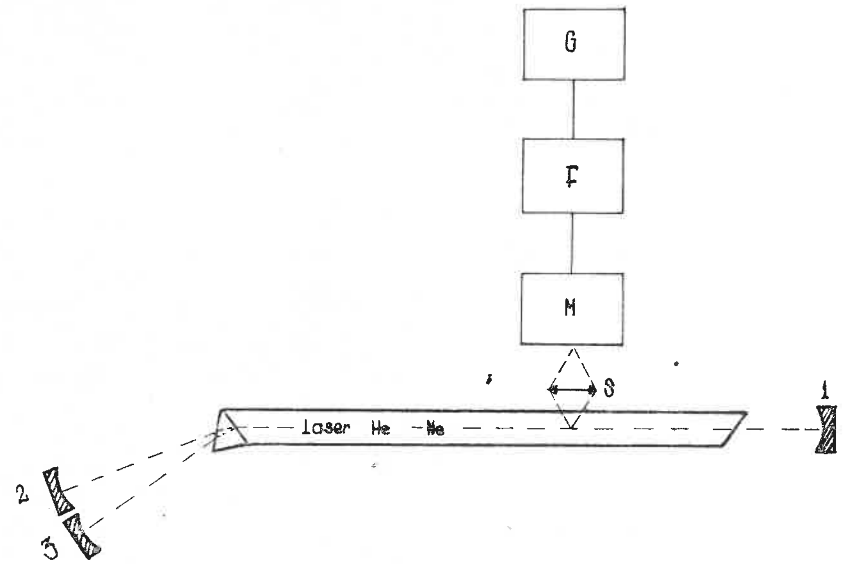

Fig. 2. Schema of the experimental set-up. 1, 2, 3 — mirrors, S — lens, M — grating monochromator, F — photomultiplier, G — galvanometer

electric current I_f was proportional to the intensity of the investigated line $I_{ij}(\nu_{ij})$: $I_f = CI_{ij}(\nu_{ij})$, C is a proportionality factor which includes the spectral response of the photomultiplier. In the adopted method it is meaningless, because the measurements were limited merely to the evaluation of the relative changes in the intensity of the same line.

All investigated lines were confined to the spectral range from 3418 Å to 8782 Å; in some cases it was not possible to avoid the overlapping of the neighbouring lines (in tables these overlapping lines are marked by an asterisk).

The width of the instrumental function was 1.6 Å and was evaluated for the 6328 Å laser line with the monochromator slits equal to 0.02 mm.

All stimulated transitions discussed in the present paper are situated in the infrared spectral range; therefore the fluorite end windows and the prism in the laser resonator were used.

The 90 cm and 174 cm long laser tubes were employed with an inner diameter of 8 mm. They were filled with an He–Ne mixture at a partial pressures and total pressure empirically adjusted for the optimum efficiency for the visible 0.63 μm laser action.

As the laser transitions co-operated in a cascade, therefore a resonator consisted of three mirrors (two on the prism side) covered by an aluminium layer. The losses at the mirrors amounted about 10%. Mirrors were mounted close to the tube ends.

The discharge current was kept constant over all measurements and amounted 30 mA.

Numerical results will be given in Sect. 4 of this paper. The accuracy of the obtained results was estimated using the method of maximum deviation for the mean value. The instability of the laser functioning was a source of random errors. On the contrary when to analyse the population change of a given level, the line was used having not resolved satellite it introduced the systematic error to the evaluated quantities. Nevertheless, in these cases the intensity ratio of the proper line and its satellite, according to [9] was 10:1; the 3418 Å ($4p'[3/2]_2 \rightarrow 3s[3/2]_1^0$) line and the 3418 Å ($4p'[1/2]_1 \rightarrow 3s[3/2]_1^0$) line as well as the 7943 Å ($3d'[5/2]_3^0 \rightarrow 3p[5/2]_2$) and the 7944 Å ($3d'[5/2]_2^0 \rightarrow 3p[5/2]_2$) line — Fig. 3, therefore the systematic error introduced by it was of no practical significance.

The dark current of the photomultiplier was compensated to zero.

4. *Experimental results and discussion*

In this paper we discuss the results of our measurements on relative population changes of levels induced by the $5s'[1/2]_1^0 \rightarrow 4p'[3/2]_2 \rightarrow 3d'[5/2]_3^0$ stimulated transitions 3.39 μm and 7.6994 μm. In the forthcoming parts we will consider the changes of level

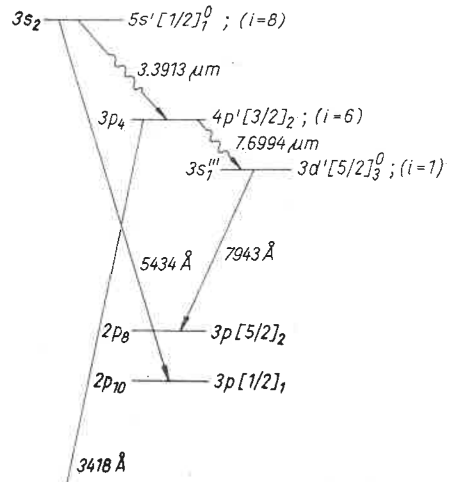

Fig. 3. Selected energy levels of Ne. Levels are labeled in Paschen and Racah notation

populations induced by other cascades. To distinguish the particular cases by suitable notation we will use in the equations (I.1)—(I.9) for the population numbers N the supplementary indices. For the sake of simplicity in notation, levels involved in the analysed processes will be labeled by means of one number only (i — see Fig. 3 and 4). Moreover, to visualize the kind of cascade the second bottom index will be added (for the cascade quoted above index k). Finally the upper indices a, b, c will still be used to distinguish the three possible cases discussed in Section 2 (equations (I.1)—(I.9)). Of course the population of a certain level i for the case (a) will be labeled with two indices N_i^{a} only (according to physical meaning of the case (a)). The indices i will be used both for the labelling of transition probabilities and pumping rates. Following this convention for notation, N_{8k}, N_{6k}, N_{1k}, and so on, will have the meaning of populations of the upper, intermediate and lower level for the $5s'[1/2]_1^0 \to 4p'[3/2]_2 \to 3d'[5/2]_3^0$ cascade. In this notation the equation (I.10) for the considered cascade has an explicit form:

$$\frac{\gamma_6}{\gamma_{61}} = \frac{\dfrac{N_1^{a}-N_{1k}^{c}}{N_1^{a}-N_{1k}^{b}} - \dfrac{N_6^{a}-N_{6k}^{c}}{N_6^{a}-N_{6k}^{b}}}{\dfrac{N_8^{a}-N_{8k}^{c}}{N_8^{a}-N_{8k}^{b}} - \dfrac{N_6^{a}-N_{6k}^{c}}{N_6^{a}-N_{6k}^{b}}}. \tag{I.11}$$

The resulting changes in the level populations as obtained from measurements are collected in the table. The minus (plus) sign at the particular value means the decreasing (increasing) of the level population with switching on of the indicated laser action.

Putting the corresponding data from the Table to the equation (I.11), we obtain the value for the ratio of the decay constant of the $4p'[3/2]_2$ level population and the probability of the spontaneous $4p'[3/2]_2 \to 3d'[5/2]_3^0$ transition corresponding to 7.6994 μm:

$$\frac{\gamma_6}{\gamma_{61}} = 6.4 \pm 0.4. \tag{I.12}$$

In evaluating this ratio from the balanced equations only the $6 \to 1$ radiative transition was taken into consideration in view of its large transition probability [11], exceeding considerably that for all other possible $4p \to 3d'[5/2]_3^0$ transitions. Thereby the transfer of the excitation was neglected as a result of the radiative transitions from the remaining levels of the $4p$ group (i.e. except $4p'[3/2]_2$) to the $3d'[5/2]_3^0$ level. For large probabilities of the transitions mentioned this effect ought to be taken into account, with regard to the strong collisional transfer of the excitation from the $4p'[3/2]_2$ by laser populated level to some others $4p$ levels.

Using the data from Lis's paper [4]:

$$\frac{\gamma_8}{\gamma_{86}} = 4.1 \pm 0.2; \quad \frac{\gamma_8}{\gamma_6} = 1$$

one can compare the probability of one of the strongest transitions in neon — $5s'[1/2]_1^0 \to$ $\to 4p'[3/3]_2$ (3.3913 μm) with the probability of the $4p'[3/2]_2 \to 3d'[5/2]_0^3$ transition:

$$\frac{\gamma_{86}}{\gamma_{61}} = 1.56 \pm 0.20.$$

In theoretical calculations, assuming intermediate coupling, Murphy [11] obtained 1.3 for this ratio.

TABLE I

Investigated level:		Relative change (in parts per cent) of the level population induced by laser action:				Transition assignment and wavelength of spontaneously emitted line used as indicators of the population change
in Racah notation	*i* according to scheme in Fig. 3	3.3913 μm		7.6994 μm in the presence of 3.3913 μm		
		$(N^b_{ik}-N^a_i)/N^b_{ik}$	$(N^b_{ik}-N^a_i)/N^a_i$	$(N^c_{ik}-N^b_{ik})/N^b_{ik}$	$(N^c_{ik}-N^a_i)/N^a_i$	
$5s'[1/2]^0_1$	8		−54.5		−64.5	5434 $5s'[1/2]^0_1 \rightarrow 3p[1/2]_1$
$4p'[3/2]_2$	6	+54.0	+117.0	−36.5	+38.1	3418*$4p'[3/2]_2 \rightarrow 3s[3/2]^0_1$ 3418*$4p'[3/2]_1 \rightarrow 3s[3/2]^0_1$
$3d'[5/2]^0_3$	1	+10.0	+10.6	+52.0	+68.1	7943*$3d'[5/2]^0_3 \rightarrow 3p[5/2]_2$ 7944*$3d'[5/2]^0_2 \rightarrow 3p[5/2]_2$

Let us notice that knowing $N^{b}_{6k}:N^{b}_{1k}$, the population ratio of the $4p'[3/2]_2$, and the $3d'[5/2]^0_3$ levels in the presence of the 3.3913 µm laser action one can, from (I.3), (I.6), calculate the ratio of the decay constant γ_1 of the $3d'[5/2]^0_3$. level population to the probability γ_{61} of the spontaneous transition at 7.6994 µm:

$$\frac{\gamma_1}{\gamma_{61}} = \frac{(N^{a}_{6}-N^{b}_{6k})/N^{b}_{6k}}{(N^{a}_{1}-N^{b}_{1k})/N^{b}_{1k}} \times \frac{N^{b}_{6k}}{N^{b}_{1k}}.$$

In order to calculate N^{b}_{6k}/N^{b}_{1k} one assumes, that the populations of the laser level involved in the transitions at the 7.6994 µm (6 and 1) satisfy the saturation condition [4] (which means that the increase in length of the active medium at low losses in the optical cavity keep the difference in populations of the laser levels close to zero)

$$N^{c}_{6k} - \frac{g_6}{g_1} N^{c}_{1k} = 0 \tag{I.13}$$

where the g_i's are the statistical weights of the levels. But for this case the switching on of the oscillations ought to change only slightly the relative population change of the laser level with increasing length of the active medium. Such was the case indeed. The relative changes of the $4p'[3/2)_2$ level population obtained in the experiment induced by the 7.6994µm laser action in co-operation with that at the 3.3913 µm were: for the tube length 90 cm and 174 cm — 33% and — 36% respectively. These results justify the assumption, that the condition (I.13) is fulfilled to a good approximation.

From the data which are listed in the table one can deduce the relationship: $N^{c}_{6k} = 0.63\, N^{b}_{6k}$ and $N^{c}_{1k} = 1.51\, N^{b}_{1k}$. Substituting the above values into (I.13) and taking into consideration, that $g_6 = 5$ and $g_1 = 7$, we obtain

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

Hence, we have

$$\frac{\gamma_1}{\gamma_{61}} = 9.6 \pm 0.6. \tag{I.14}$$

From (I.14) and (I.12) we get the relationship between γ_1 and γ_6:

$$\frac{\gamma_1}{\gamma_6} = 1.50 \pm 0.20.$$

From which it can be seen that the lifetime of the lower level of the 7.6994 µm laser transition ($i = 1$) is shorter than the lifetime of the upper level. This conclusion would be qualitatively expected from the general conditions for the excitation of the laser oscillation.

From the numerical data obtained one can draw conclusions about the contribution of particular transitions in the population balance in the stationary state for the $4p'[3/2]_2$ and $3d'[5/2]^0_3$ levels. It is shown schematically in Fig. 4. A similar schema for the $5s'[1/2]^0_1$ and $4p'[3/2]_2$ levels for the 3.3913 µm transition was presented in [4]. The supplementary contribution of the 7.6994 μm transition in the depopulation of the $4p'[3/2]_2$ level and the population of the $3d'[5/2]^0_3$ level is visualized in Fig. 4. It was assumed that the relative population of the $5s'[1/2]^0_1$ level pumped with the rate R_8 is equal to 100 units. We see that the population of the $3d'[5/2]^0_3$ level is due mainly to pumping (with the constant R_1) whereas the contribution of the $4p'[3/2]_2 \rightarrow 3d'[5/2]^0_3$ transition is rather low.

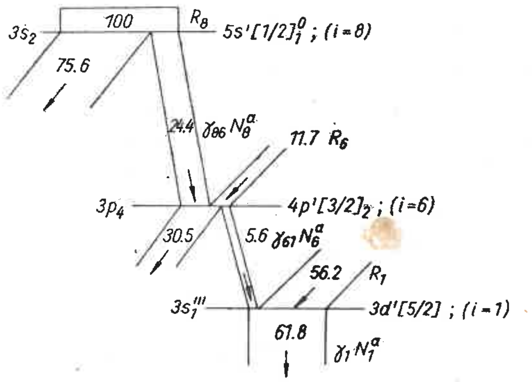

Fig. 4. Contribution of the individual transitions to the population or to the depopulation of the levels. Quoted values refer to the population of the $5s'[1/2]^0_1$ level, taken arbitrarily as equal to 100 units. Values were calculated from the equations: $\frac{\gamma_6}{\gamma_{61}} = 6.4 \rightarrow \gamma_{61}N^a_6 = \frac{\gamma_6 N^a_6}{6.4} = \frac{36.1}{6.4} = 5.6$; $\frac{\gamma_1 N^a_1}{\gamma_{61}N^a_6} = \frac{(N^a_6 - N^b_{6k})/N^a_6}{(N^a_1 - N^b_{1k})/N^a_1} = 11.04 \rightarrow \gamma_1 N^a_1 = 61.8$; $R_1 = \gamma_1 N^a_1 - \gamma_{61}N^a_6 = 56.2$

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