CHARACTERISTICS OF $3s_2 \rightarrow 3p_1$ ($\lambda = 4218$ nm) LASER ACTION IN NEON

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The conditions of occurrence of $3s_2 \rightarrow 3p_1$ ($\lambda = 4218$ nm) laser oscillations were studied by the method of induced changes of populations of the levels. The ratio of probabilities of the spontaneous radiative transitions $3s_2 \rightarrow 3p_4$ and $3s_2 \rightarrow 3p_1$ was determined, and a good agreement with the data of other authors was obtained. The ratio of populations of the levels $3s_2$ and $3p_1$, and the ratio of the decay constants $\gamma_2 : \gamma_1$ were estimated.

1. *Introduction*

In the previous paper we reported the observation of $3s_2 \rightarrow 3p_1$ laser action in neon [2]. This transition, which was observed for the first time by Brunet and Laures [1], can be obtained when the competitive laser action 3391 nm (Fig. 2) is attenuated and the appropriate optical materials are used. The measurement of the changes in the intensities of spontaneously emitted selected spectral lines caused by the stopping of the mentioned laser action allows one to estimate the values of the parameters such as the decay constants of the levels, the ratios of populations, *etc.*

Consider a three-level system 2, 1, 0 (Fig. 1) with the laser actions $2 \rightarrow 1$ and $2 \rightarrow 0$ being possible, and assume that the transitions $1 \rightarrow 0$ do not occur. In these conditions one can distinguish three different cases:

0) absence of any laser action,
1) presence of the laser action $2 \rightarrow 1$,
2) presence of the laser action $2 \rightarrow 0$.

The mixed cases, *i.e.* the occurrence of several laser actions simultaneously, are also possible. The covering of one of the mirrors of the optical resonator corresponds to the case 0, *i.e.* to the absence of oscillations. The parameters of the case 0 will be used as the reference ones for the other cases. We denote the populations of the levels in the absence of laser oscillations by N_2^0, N_1^0, N_0^0, and analogously N_2', N_1', N_0' for the case 1, and

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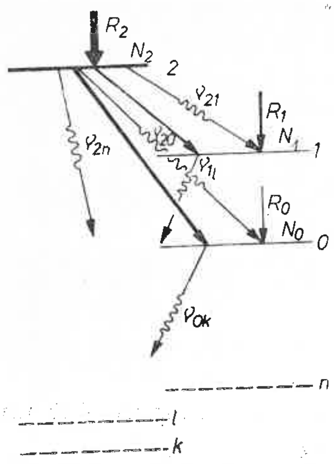

Fig. 1

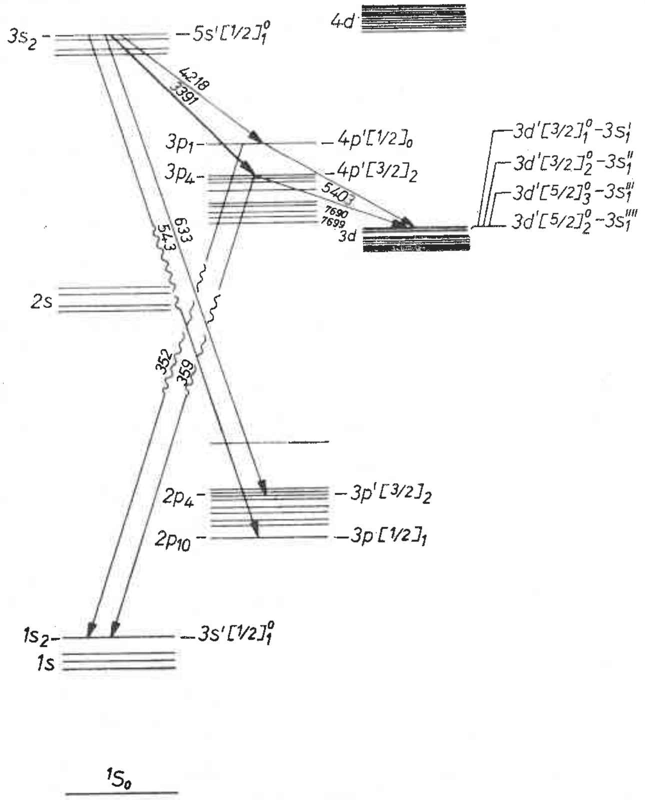

Fig. 2. Scheme of neon levels showing laser transitions concerning this paper [10]

N_2'', N_1'', N_0'' for the case 2. Using the balance equations for the populations we obtain in the stationary conditions the relations [3]:

$$N_1'-N_1^0 = \frac{\gamma_2-\gamma_{21}}{\gamma_1}(N_2^0-N_2') \tag{1a}$$

$$N_0'-N_0^0 = \frac{\gamma_{20}}{\gamma_0}(N_2'-N_2^0) \tag{1b}$$

for the laser action 2 → 1, and:

$$N_1''-N_1^0 = \frac{\gamma_{21}}{\gamma_1}(N_2''-N_2^0) \tag{2a}$$

$$N_0''-N_0^0 = \frac{\gamma_2-\gamma_{20}}{\gamma_0}(N_2^0-N_2'') \tag{2b}$$

for the laser action 2 → 0. The quanties γ_2, γ_1, γ_0 are the decay constants of the corresponding levels (taking into account the radiationless depopulation), and γ_{21}, γ_{20} are the probabilities of the corresponding radiative transitions. From the equations (1a) and (2a), and (1b) and (2b) we obtain:

$$\frac{\gamma_2}{\gamma_{21}}-1 = \frac{\alpha_1}{\alpha_2} \tag{3}$$

and

$$\frac{\gamma_2}{\gamma_{20}}-1 = \frac{\beta_2}{\beta_1}, \tag{4}$$

where

$$\alpha_1 = \frac{N_1'-N_1^0}{N_1^0}:\frac{N_2^0-N_2'}{N_2^0}, \qquad \alpha_2 = \frac{N_1''-N_1^0}{N_1^0}:\frac{N_2''-N_2^0}{N_2^0}$$

$$\beta_2 = \frac{N_0''-N_0^0}{N_0^0}:\frac{N_2^0-N_2''}{N_2^0}, \qquad \beta_1 = \frac{N_0'-N_0^0}{N_0^0}:\frac{N_2'-N_2^0}{N_2^0}.$$

The quantities α_1, α_2, β_1, β_2 can be determined experimentally by measuring the relative intensities of spectral lines from the levels 2, 1, 0 in the cases 0, 1, and 2.

2. *Experimental set-up*

In the experiment two lasers were used:

1) the laser described in [2] in order to obtain the laser transition $3s_2 \rightarrow 3p_1$,

2) the laser with a quartz prism stuck at one end of the tube in order to obtain the pure laser action 633 nm [4]. The discharge tube of the first laser was closed at one end by a fluorite prism, and at the other one by a fluorite window. In these conditions the possibility

of appearance of the laser action 633 nm was excluded because of colouring of fluorite under vacuum ultraviolet irradiation and, in consequence, of small transmission of fluorite in the visible region. Both laser tubes of the same dimensions (1500 nm long and 8 nm in internal diameter) were connected to the common vacuum system in order to fill them with the some He–Ne mixture. The pressures of the gases corresponded to optimum conditions for the laser action 633 nm. In the direction perpendicular to the laser tubes a SPM-2 grating monochromator followed by a detection system for measurement of intensities of spectral lines (1P28 photomultiplier) was placed. D.C. signals due to the particular lines in the conditions of covering of one of the mirrors or of maximum intensity of given laser oscillations were measured. Spectral lines 543 nm, 352 nm, and 359 nm were used to determine the changes of populations of the levels $3s_2$, $3p_1$, and $3p_4$. The spatial distribution of intensity of these lines in the plane perpendicular to the laser tube in the presence of laser oscillations 3391 nm, 633 nm, and 4218 nm was also studied. The intensity of spectral lines from the studied levels in the presence of laser oscillations was independent of the direction of observation.

3. *Experimental results*

In Table I the ratios of relative changes of populations of the levels $3s_2$, $3p_4$, and $3p_1$ for various laser actions are given. Neon levels $3s_2$, $3p_1$, and $3p_4$ are denoted by subscripts 2, 1, and 0, respectively.

TABLE I

$\frac{\Delta N_2}{N_2^0}:\frac{\Delta N_0}{N_0^0}$ α_2^{-1} 633 nm	$\frac{\Delta N_0}{N_0^0}:\frac{\Delta N_2}{N_2^0}$ α_1 3391 nm	$\frac{\gamma_2}{\gamma_{20}}=\frac{\alpha_1}{\alpha_2}-1$	$\frac{\Delta N_2''}{N_2^0}:\frac{\Delta N_1''}{N_1^0}$ α_2^{-1} 633 and 3391 nm	$\frac{\Delta N_1'}{N_1^0}:\frac{\Delta N_2'}{N_2^0}$ α_1 4218 nm	$\frac{\gamma_2}{\gamma_{21}}=\frac{\alpha_1}{\alpha_2}-1$	γ_{20}/γ_{21}		
						Murphy [5]	Weaver, Freiberg [6]	Author
1.48 ± 0.04	2.1 ± 0.1	4.1 ± 0.2	2.63 ± 0.04	14.8 ± 0.3	39.9 ± 1.6	10.8	9.6	9.7 ± 0.9

The obtained values of the ratios $\gamma_2:\gamma_{20}$ and $\gamma_2:\gamma_{21}$ agree with the literature data [6]. One can then conclude that with the used diameter of the tube and the pressures of the gases, the collisional depopulation of the level $3s_2$ is negligible compared with the radiative one. When the laser tube of smaller internal diameter (*e.g.* 4 mm) was used, considerably greater value of the ratio $\gamma_2:\gamma_{20}$ was obtained.

For the ratio of probabilities of the radiative transitions $3s_2 \rightarrow 3p_4$ and $3s_2 \rightarrow 3p_1$, $\gamma_{20}:\gamma_{21}$, very good agreement with the theoretical data [6] was also found. The latter ratio does not depend on the diameter of the tube nor on the pressures of the gases.

The examples of results obtained for the relative changes of populations of the considered levels are given in Table II for several cases. Column 1 refers to the pure 4218 nm laser action obtained with the selection of laser transitions by means of two fluorite prisms. The quoted changes of populations are the maximum ones for our experimental conditions.

TABLE II

	1	2	3*	4
	4218 nm	4218 and 3391 nm	3391,7690 or 7699 nm	3391 nm
$3s_2$	−8%	−33%	−61%	−54%
$3p_1$	+121%	+63%	−23%	−19%
$3p_4$			+77%	+116%

* When in the considered conditions one quenches the laser action 3391 nm by means of a polyethylene foil, the laser action 4218 nm appears.

It seems that with increasing length of the active medium above the one used in the experiment, further increase in changes of populations of atomic levels is not observed. This may mean that the difference $\frac{N_2}{g_2} - \frac{N_1}{g_1}$ is close to zero in the conditions of maximum $3s_2 \rightarrow 3p_1$ oscillations.

The construction of the laser tube with the prism stuck at one end does not ensure a good separation of laser actions 4218 nm and 3391 nm, and consequently the complete quenching of the latter. In this case we observe both laser actions simultaneously with considerable attenuation of the laser transition $3s_2 \rightarrow 3p_1$ (column 2). The position of the mirror corresponded to the maximum population of $3p_1$ level, and then to the maximum intensity of laser transition $3s_2 \rightarrow 3p_1$.

The conditions obtained when the additional fluorite prism is used to compensate the splitting caused by the stuck prism correspond to those obtained when the resonator without selection of laser transitions is used (column 3).

The results listed in column 4 were obtained when, in addition to the compensation of splitting caused by the prism, a LiF plate was introduced into the resonator.

4. *Conclusions*

From the data contained in Table II the ratio of populations of the levels $3s_2$ and $3p_1$, $N_2^0 : N_1^0$, can be estimated. In the presence of laser oscillations the condition $\frac{N_2}{g_2} - \frac{N_1}{g_1} \geqslant 0$ must be fulfilled, and then $\frac{0.92\, N_2^0}{3} - \frac{2.21\, N_1^0}{1} \geqslant 0$, and $N_2^0 : N_1^0 \approx 7.2$.

The ratio $\gamma_1 : \gamma_2$ can be calculated from the equation obtained from (2a)

$$\gamma_1 = \left(\frac{N_2 - N_2^0}{N_2^0} : \frac{N_1 - N_1^0}{N_1^0} \right) \frac{N_2^0}{N_1^0} \gamma_{21}$$

and the data of Table I. We find $\gamma_1 : \gamma_2 \approx 0.5$.

The value of the decay constant of the level $3s_2$ which is twice as big as that of the level $3p_1$ explains the difficulties in obtaining greater number of stimulated transitions

$3s_2 \rightarrow 3p_1$, and consequently in attaining large depopulation of the level $3s_2$ by the considered laser action. In fact, the small number of stimulated transitions $3s_2 \rightarrow 3p_1$ (small decrease in the population of the level $3s_2$) causes the large increase in the population of weakly populated and weakly depopulated level $3p_1$.

When the splitting by the prism is compensated then 1° the stimulated transition $3s_2 \rightarrow 3p_1$ is not observed, and 2° we obtain the ratio of changes of populations of the levels $3s_2$ and $3p_4$ different from that obtained in the case of pure laser action 3391 nm.

In order to explain the former fact we estimate the ratio of amplification factors for the two laser transitions. We make use of the formula for integral amplification factor [7]:

$$C_{ij} = \int\limits_0^\infty K_{ij}(\nu)d\nu = \frac{\lambda_{ij}^2}{8\pi} N_i^0 \gamma_{ij} \left(1 - \frac{g_i}{g_j} \frac{N_j^0}{N_i^0}\right).$$

For the laser transitions $3s_3 \rightarrow 3p_4$ and $3s_2 \rightarrow 3p_1$ we have

$$C_{20} = \frac{3391^2}{8\pi} \gamma_{20} N_2^0 \left(1 - \frac{3}{5} \frac{N_0^0}{N_2^0}\right)$$

and

$$C_{21} = \frac{4219^2}{8\pi} \gamma_{21} N_2^0 \left(1 - 3 \frac{N_1^0}{N_2^0}\right).$$

Substituting $\gamma_{20} : \gamma_{21} = 10$, $N_2^0 : N_1^0 = 7$, and $N_2^0 : N_0^0 = 3$, we obtain $C_{20} : C_{21} = 14$. In consequence when the both laser actions can occur, the oscillations at the transition $3s_2 \rightarrow 3p_4$ develop first. The stimulated transitions $3s_2 \rightarrow 3p_4$ cause the decrease in the population of the level $3s_2$ and make the development of the laser action 4218 nm impossible.

The different ratios of changes of populations of the levels $3s_2$ and $3p_4$ in the case when the splitting by the prism is compensated (61 : 77 — Table II, column 3), and in the case of pure laser action 3391 nm (1 : 2.1 — Table I, column 2) can be explained by assuming the appearance of stimulated transitions from the level $3p_4$. In fact, the presence of these oscillations causes the decrease in the population of the level $3p_4$ in the first case. It seems that these are the laser actions $3p_4 \rightarrow 3s_1''''$ 7690 nm [8], and $3p_4 \rightarrow 3s_1'''$ 7699 nm [9]. When the LiF plate is introduced into the resonator these laser transitions are quenched and we obtain the results identical with those obtained for pure laser action 3391 nm (the transmission of LiF falls rapidly at 7000 nm).

We have also observed stimulated transition $3p_1 \rightarrow 3s_1'$ ($\lambda = 5403$ nm) [8].

REFERENCES

[1] H. Brunet, P. Laures, *Phys. Letters*, **12**, 106 (1964).
[2] L. Lis, *Phys. Letters*, **39A**, 119 (1972).
[3] T. F. Johnston, *Appl. Phys. Letters*, **4**, 161 (1970).
[4] L. Lis, *Acta Phys. Polon.*, **A42**, 307 (1972).
[5] P. W. Murphy, *J. Opt. Soc. Amer.*, **58**, 1200 (1968).
[6] L. A. Weaver, R. J. Freiberg, *J. Appl. Phys.*, **37**, 1528 (1966).
[7] G. Birnbaum, *Optical Masers*, Academic Press, New York and London 1964.
[8] C. K. N. Patel, R. A. McFarlane, W. L. Faust, *Quantum Electronics III*, P. Grivet, N. Bloembergen, eds., New York 1964, p. 561.
[9] W. L. Faust, R. A. McFarlane, C. K. N. Patel, C. G. B. Garrett, *Phys. Rev.*, **133A**, 1476 (1963).
[10] C. E. Moore, *Atomic Energy Levels*, 1949.