POPULATION CHANGES OF Ne LEVELS INDUCED IN He—Ne MIXTURE BY LASER ACTIONS IN THE INTERMEDIATE IR. PART III. POPULATION TRANSFER BY THE NONRADIATIVE TRANSITIONS IN THE 3*d* GROUP OF LEVELS IN NEON

By M. Kaniewska

Institute of Physics, Polish Academy of Sciences, Warsaw*

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Relative changes of intensities appearing when laser actions were switched on and off were studied for chosen Neon lines emitted spontaneously in the He—Ne laser. On the ground of obtained results an excitation transfer due to collisional processes was analized for the 3*d* group of levels in Neon. The ratio of the decay constant for the population of the $3d'[5/2]^0_3$ ($3d'[5/2]^0_2$) level for nonradiative transitions $3d'[5/2]^0_3 \to 3d'[5/2]^0_2$ ($3d'[5/2]^0_2 \to \to 3d'[5/2]^0_3$) and the decay constant for the population of this level for the other transitions was determined (and estimated).

1. *Introduction*

The phenomenon of induced population changes (described in Part I [1]) may be used for the investigation of nonradiative transitions between the atomic levels. We can conclude from the direction of population changes for the studied and the laser levels, that such transitions exist. We may expect these transitions in the case when the sign of population changes is for a group of levels the same. The less energy separation between the levels, the greater probability of these transitions. Moreover, it depends on the fact whether the Wigner rule is fulfilled [2].

The existence of nonradiative transitions between the levels in Neon has been reported in many works. Weaver and Freiberg [3] write about the collisions between the atoms in the $4p'[3/2]_2$, $4p'[1/2]_1$, $4p'[3/2]_1$; $3p'[3/2]_2$ and $3p[1/2]_0$ states and between the atoms in the 5*s* states. In the former two cases it might have been expected that the excitation will be transferred because of a small energy separation between the levels. The decrease in the $5s[3/2]^0_2$ level population, however, in presence of 3.3913 μm oscillation, has been quite unexpected when we take into account the fact that the energy separation between the $5s[3/2]^0_2$ level and the most strongly perturbed by a laser action the $5s'[1/2]^0_1$ level, is as much as 828 cm^{-1}. The thermal motion energy of atoms at the temperature of 400 K

* Present address: Instytut Technologii Elektronowej, Lotników 32/46, 02-668 Warszawa, Poland.

(that is more or less the temperature in the discharge tube) is about 300 cm^{-1}. The similar case has been also observed in the *3d* group (more details will be given later on).

The results of Massey and his colaborators [4] occur very useful for our suggestion on the mechanism of the excitation transfer in the collisional processes. The authors underline the dependence of the cross section for the atomic collisions on the Wigner rule fulfilment. They found that the cross section is higher when the spin quantum numbers of the levels belonging to the colliding atoms fulfil this rule.

The works of Parks and Javan [5], Lilly [6] and Lis [7] are very important for us. They give an example for using the balance equations for determining the physical quantities, characteristic for the nonradiative processes, by means of the induced population changes method. The authors of two former papers obtained the cross sections for collisions in the *4s* and *5s* levels, respectively. The latter work is on the nonradiative processes for the *4p* group of levels.

2. Analysis of population changes for the 3d levels, induced by the 7.6994 μm and 5.403 μm laser actions

In the present work the population changes for the *3d* levels in Neon have been studied. They have been caused by applying the 7.6994 μm laser action in cooperation with the 3.3913 μm laser action and the 5.403 μm one in the presence of the 4.218 μm oscillation. The diagram of the studied levels and laser transitions is shown in Fig. 1.

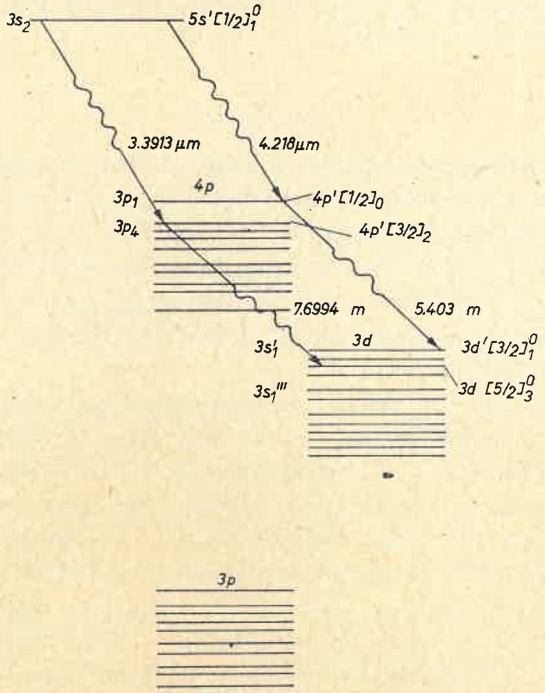


Fig. 1. Diagram of chosen Neon levels and laser transitions. On behalf of clarity the proportions in distances between levels are not in scale

The cooperating actions 3.3913 μm and 4.218 μm were excited in the system in order to obtain a larger population change induced by the 7.6994 μm and 5.403 μm oscillation in the levels mentioned before. The scheme and description of the experimental set-up which was used for the population changes determination are presented in Part I.

The analysis of these changes will be made using the experimental results which are gathered in Table I. The population changes for the $4p$ and $3p$ group of levels are also collected in the same table for comparison. The latter (i.e. for the $4p$ and $3p$ levels) have the same character when caused by the 3.3913 μm laser action (Table I, column III) as the changes described in [9] and this consistence confirms the conclusions made by the author of the quoted work.

The symbols used in this part are essentially the same as the symbols in two previous parts. An additional index "l" which enables one to distinguish the population changes caused by the cascade $5s'[1/2]_1^0 \rightarrow 4p'[1/2]_0 \rightarrow 3d'[3/2]_1^0$ from the changes induced by the oscillations $5s'[1/2]_1^0 \rightarrow 4p'[3/2]_2 \rightarrow 3d'[5/2]_3^0$ is the only change. In the case of the "cascade" $5s'[1/2]_1^0 \rightarrow 4p'[3/2]_2$, $4p'[3/2]_1 \rightarrow 3d'[5/2]_2^0$ the index "m" will be used in the following part of this paper.

The analysis of the values presented in Table I, column III shows that for all the levels belonging to the group $3d$ there is an increase in population induced by the oscillation 3.3913 μm, hence they behave identically with the $4p'[1/2]_1 \ldots 4p[1/2]_1$ levels (except the $4p'[1/2]_0$ level). Whereas, besides a very small increase in the $3d'[3/2]_1^0$ level population no influence of the 4.218 μm oscillation on the populations of all the other $3d$ levels (the last row in Table I) has been observed despite the 106% increase in the $4p'[1/2]_0$ level population (this value is not given in the table). The population change for the $3d[3/2]_1^0$ and $3d[1/2]_1^0$ levels coupled through radiative transitions with the $4p'[1/2]_0$ level should be seen according to [11] as well; beside $3d'[3/2]_1^0$. Two facts noticed in [12] explain the absence of this effect: the oscillation 4.218 μm is due to the small number of the stimulated transitions $5s'[1/2]_1^0 \rightarrow 4p'[1/2]_0$; the population of the $4p'[1/2]_0$ level is low. It is worth-while to point out here that this transition induces very slight (−7.1 %) decrease in population of the upper laser level $5s'[1/2]_1^0$ (this value is not given in the table). Very weak population changes for the $4p$ levels are the consequence of the same facts. In such a situation, we may neglect the population changes of the $3d$ levels which might be transferred from the $4p$ levels.

The laser transitions of the 7.6994 μm and 5.403 μm wave length have a considerable influence on the populations of some $3d$ levels mainly on the $3d'[3/2]_1^0$, $3d'[3/2]_2^0$, $3d'[5/2]_3^0$, $3d'[5/2]_2^0$ levels which indicate the same sign of population changes after switching on the generations mentioned above (Table I, columns IV and V). This situation is due to the existence of the collisions mixing the population of specified levels. The distances between these levels are very small (Table II) and the levels $3d'[5/2]_3^0$ and $3d'[5/2]_2^0$ are closest to each other.

The phenomenon of the excitation transfer in collisions may be described as follows. When the laser generation, e.g. for the transition $4p'[3/2]_2 \rightarrow 3d'[5/2]_3^0$ is excited in the system, the population of the $3d'[5/2]_3^0$ level increases. The population of the neighbouring levels which are situated close to this level increases too, as a result of the atomic collisions.

TABLE I

Symbol of analysed level in representation [10]:		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]
Racah	Paschen	3.3913 µm	7.6994 µm in presence of 3.3913 µm	5.403 µm in presence of 4.218 µm	3.3913 µm and 7.6994 µm′ in the case when the population change of the $4p'[3/2]_2$ level is zero	
		$(N^b_{ik}—N^a_i)/N^a_i$	$(N^c_{ik}—N^b_{ik})/N^b_{ik}$	$(N^c_{il}—N^b_{il})/N^b_{il}$	$(N^c_{ik}—N^a_i)/N^a_i$	
I	II	III	IV	V	VI	VII
$5s'[1/2]^0_1$	$3s_2$	−54.5	−21.8	−11.5	−9.7	5434 $5s'[1/2]^0_1 \rightarrow 3p\ [1/2]_1$
$4p'[1/2]_0$	$3p_1$	−19.4	−5.2	−47.0	−3.5	3520 $4p'[1/2]_0 \rightarrow 3s'[1/2]_1$
$4p'[1/2]_1$	$3p_2$	+109.0	−36.6	−7.3	0	3461 $4p'[1/2]_1 \rightarrow 3s'[1/2]^0_0$
$4p'[3/2]_2$	$3p_4$	+117.0	−36.5	−6.9	0	3418* $4p'[3/2]_2 \rightarrow 3s\ [3/2]^0_1$ 3418* $4p'[1/2]_1 \rightarrow 3s\ [3/2]^0_1$
$4p'[3/2]_1$	$3p_5$	+80.0	−32.0	−5.4	0	3600 $4p'[3/2]_1 \rightarrow 3s'[1/2]^0_1$
$4p\ [1/2]_0$	$3p_3$	+17.0	−17.8	−4.5	0	3454 $4p\ [1/2]_0 \rightarrow 3s\ [3/2]^0_1$
$4p\ [3/2]_2$	$3p_6$	+14.3	−13.9	slight decrease	0	3498 $4p\ [3/2]_2 \rightarrow 3s\ [3/2]^0_1$
$4p\ [3/2]_1$	$3p_7$	+11.0	−8.0	slight decrease	0	3685 $4p\ [3/2]_1 \rightarrow 3s'\ [1/2]^0_1$
$4p\ [5/2]_2$	$3p_8$	+11.0	−10.0	slight decrease	0	3701 $4p\ [5/2]_2 \rightarrow 3s'[1/2]^0_1$
$4p\ [5/2]_3$	$3p_9$	+9.8	−7.6	slight decrease	0	3473 $4p\ [5/2]_3 \rightarrow 3s\ [3/2]^0_2$
$4p\ [1/2]_1$	$3p_{10}$	+11.0	0	0	0	3511 $4p\ [1/2]_1 \rightarrow 3s\ [3/2]^0_2$
$3d'[3/2]^0_1$	$3s'$	+11.4	+15.4	+54.0	+5.4	8118 $3d'[3/2]^0_1 \rightarrow 3p\ [3/2]_1$
$3d'[3/2]^0_2$	$3s''_1$	+10.5	+19.0	+8.9	+7.7	8259 $3d'[3/2]^0_2 \rightarrow 3p\ [3/2]_2$
$3d'[5/2]^0_3$	$3s'''_1$	+10.6	+52.0	+9.1	+12.7	7943* $3d'[5/2]^0_3 \rightarrow 3p\ [5/2]_2$ 7944* $3d'[5/2]^0_2 \rightarrow 3p'[5/2]_2$
$3d'[5/2]^0_2$	$3s''''_1$	+12.8	+19.0	+8.9	+6.1	8591 $3d'[5/2]^0_2 \rightarrow 3p'[3/2]_1$
$3d\ [5/2]^0_3$	$3d'_1$	+9.6	slight increase	slight increase	slight increase	8781* $3d\ [5/2]^0_3 \rightarrow 3p\ [3/2]_2$ 8782* $3d\ [5/2]^0_2 \rightarrow 3p\ [3/2]_2$

TABLE I (continued)

I	II	III	IV	V	VI	VII
$3d\,[5/2]^0_2$	$3d''_1$	+7.1	slight increase	slight increase	+1.8	8418* $3d\,[5/2]^0_2 \to 3p\,[5/2]^0_2$ 8417* $3d\,[5/2]^0_3 \to 3p\,[5/2]_2$
$3d\,[3/2]^0_1$	$3d_2$	+2.8	slight decrease	0	0	7472 $3d\,[3/2]^0_1 \to 3p\,[1/2]_1$
$3d\,[3/2]^0_2$	$3d_3$	+3.3	−1.1	0	0	7488 $3d\,[3/2]^0_2 \to 3p\,[1/2]_1$
$3d\,[7/2]^0_3$	$3d_4$	+3.8	−1.9	0	0	8495 $3d\,[7/2]^0_3 \to 3p\,[5/2]_2$
$3d\,[7/2]^0_4$	$3d'_4$	+2.7	−1.3	0	0	8377* $3d\,[7/2]^0_4 \to 3p\,[5/2]_3$ 8376* $3d\,[7/2]^0_3 \to 3p\,[5/2]_3$
$3d\,[1/2]^0_1$	$3d_5$	+1.2	−1.2	0	0	7536 $3d\,[1/2]^0_1 \to 3p\,[1/2]_1$
$3d\,[1/2]^0_0$	$3d_6$	increase	decrease	0	0	7544 $3d\,[1/2]^0_0 \to 3p\,[1/2]_1$
$3p'[1/2]_0$	$2p_1$	0	−1.0	+3.3	0	5852 $3p'[1/2]_0 \to 3s'[1/2]_0$
$3p'[1/2]_1$	$2p_2$	+1.0	+1.0	+1.2	+0.7	6599 $3p'[1/2]_1 \to 3s'[1/2]^0_1$
$3p\,[1/2]_0$	$2p_3$	−1.4	+2.8	+4.7	+0.7	6074 $3p\,[1/2]_0 \to 3s\,[3/2]^0_1$
$3p'[3/2]_2$	$2p_4$	−1.4	+4.1	increase	+2.1	6678 $3p'[3/2]_2 \to 3s'[1/2]^0_1$
$3p'[3/2]_1$	$2p_5$	+3.9	0	0	0	6266 $3p'[3/2]_2 \to 3s'[1/2]^0_0$
$3p\,[3/2]_2$	$2p_6$	+7.7	−5.1	0	0	6929 $3p\,[3/2]_2 \to 3s'[1/2]^0_1$
$3p\,[3/2]_1$	$2p_7$	+3.2	0	+2.4	+0.7	6383 $3p\,[3/2]_1 \to 3s\,[3/2]^0_1$
$3p\,[5/2]_2$	$2p_8$	+1.7	0	0	0	6506 $3p\,[5/2]_2 \to 3s\,[3/2]^0_1$
$3p\,[5/2]_3$	$2p_9$	0	−1.0	0	0	6402* $3p\,[5/2]_3 \to 3s\,[3/2]^0_2$ 6401* $5s'[1/2]^0_1 \to 3p'[1/2]_1$
$3p\,[1/2]_1$	$2p_{10}$	+2.0	−0.7	0	0	7245 $3p\,[1/2]_1 \to 3s\,[3/2]^0_1$
$3d'[3/2]^0_2$	$3s''_1$	Relative population changes of the mentioned levels, induced by the 4.218 μm laser action (except the $3d'[3/2]^0_1$ level, which the population increased slightly) were zero.				
.	.					
.	.					
.	.					
$3d\,[1/2]^0_0$	$3d_6$					

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

TABLE II

Symbol of analysed level in representation: Racah	Paschen	The energy separation of some 3d levels from the $3d'[5/2]^0_3$ level [cm^{-1}]	Spin quantum number of electronic shell
$3d'[3/2]^0_1$	$3s'_1$	25.5	1
$3d'[3/2]^0_2$	$3s''_1$	9.8	0
$3d'[5/2]^0_3$	$3s'''_1$	0	0
$3d'[5/2]^0_2$	$3s''''_1$	1.5	1
$3d\,[5/2]^0_3$	$3d'_1$	709	1
$3d\,[5/2]^0_2$	$3d''_1$	711	0
Ne(0) — Neon atom in the ground state			0
He(0) — Helium atom in the ground state			0

This process may be expressed symbolically, on the example of two $3d'[5/2]^0_3$ and $3d'[5/2]^0_2$ levels, by the following scheme $Ne_a(3d'[5/2]^0_3+Ne_b(0) \rightarrow Ne_b(3d'[5/2]^0_2)+Ne_a(0)+\Delta E$. This reaction is undistinguishable from

$Ne_a(3d'[5/2]^0_3)+Ne_b(0) \rightarrow Ne_a(3d'[5/2]^0_2)+Ne_b(0)+\Delta E$, where Ne(0) denotes the Neon atom in the ground state. The excitation transfer may be also the result of collisions with Helium atoms in the ground state which are described by reaction

$$Ne(3d'[5/2]^0_3)+He(0) \rightarrow Ne(3d'[5/2]^0_2)+He(0)+\Delta E. \qquad \text{(III.1)}$$

As the number of Helium atoms exceeds strongly the number of Neon atoms, we should expect that the increase in population of the neighbouring levels during the laser oscillations is mainly caused by the collisions described by scheme (III.1). In the introduction we have mentioned that the reaction has the higher cross section if the resultant spin of atoms interacting in collisions is conserved. It follows, from the data presented in Table II that the Wigner spin rule is fulfiled only for two pairs of the levels $3d'[5/2]^0_3$, $3d'[3/2]^0_2$ and $3d'[5/2]^0_2$, $3d'[3/2]^0_1$ (taking into account first four levels). On the other hand, it is seen from the example of the 4*s* and 5*s* levels excitation by the resonant energy transfer from helium to neon atoms that the collisional cross section depends mainly on the distance between the levels, whereas for small distances the Wigner rule is not so important.

It is worthwhile to think over whether there is the state of detailed thermal equilibrium [5] because of small distance between the $3d'[5/2]^0_3$ and $3d'[5/2]^0_2$ levels. In order to achieve this kind of equilibrium, the atom-atom collisions should dominate the other relaxation processes i.e. atom-electron collisions or radiative transitions. Some temperture is established as a result of kinetic energy exchange between the atoms and of the collisions against the walls, however, it will not describe the population of excited states in general. In the case when the atomic collisions are the dominating mechanism of internal and kinetic energies exchange, the populations are given by Boltzmann distribution. It may happen that the atom-atom collisions are responsible for the relaxation of only few close-distant levels from the whole level diagram of excited atom. The detailed thermal equilibrium is related to such a situation.

The pressure of Neon were determined for which one can assume the detailed thermal equilibrium for the following pairs of levels [5, 6, 13]: $4s'[1/2]^0_1$, $4s'[1/2]^0_0$; $5s([3/2]^0_1$, $5s[3/2]^0_2$; $5s'[1/2]^0_1$, $5s'[1/2]^0_0$. It occurs that the detailed thermal equilibrium between the $4s'[1/2]^0_1$, $4s'[1/2]^0_0$ levels distant of 155 cm^{-1} from each other is obtained at the pressure of neon equal to 100 Tr, between the $5s[3/2]^0_1$, $5s[3/2]^0_2$ levels (distance of 85 cm^{-1}) at the

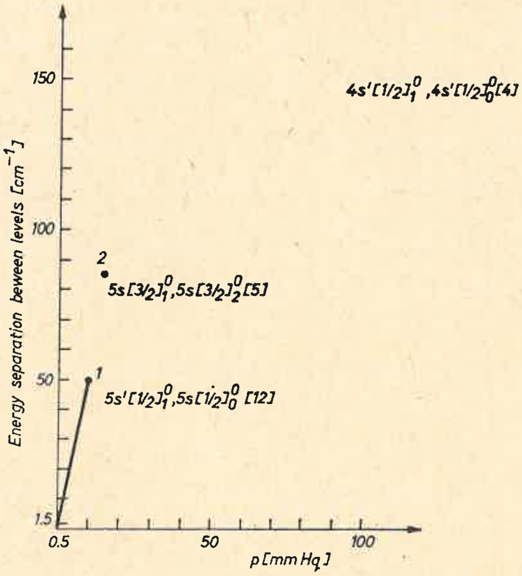


Fig. 2

pressure of 15 Tr and between the $5s'[1/2]^0_1$, $5s'[1/2]^0_0$ levels (distance of 50 cm^{-1}) at 10 Tr. The results are shown in Fig. 2 where the corresponding gas pressure is shown vs. distance between the levels.

Taking into consideration the above results and the following facts:

(a) the radiative lifetime of the $3d'[5/2]^0_3$, $3d'[5/2]^0_2$ levels are comparable with the radiative lifetimes of the $5s$ levels (the lifetimes of majority of levels mentioned before are given in Table III),

(b) we may state according to [5] that the pressure of pure neon and of the He—Ne

TABLE III

Symbol of the level in Racah notation	Radiative lifetime of atom [nsec]	References
$4s'[1/2]^0_1$	96	[14]
$4s'[1/2]^0_0$	160	
$5s'[1/2]^0_1$	23.1	[15]
$5s\ [3/2]^0_1$	19.5	
$3d'[5/2]^0_3$	18.7	[16]
$3d'[5/2]^0_2$	21.1	

mixture for which the thermalization between the levels takes place are more or less equal,

(c) when $\Delta E \to 0$ (ΔE — the distance between levels) the gas pressure related to thermal equilibrium $p \to 0$ since for $\Delta E = 0$ we consider the same level,

we may estimate the pressure value for which the thermal equilibrium will be reached between the $3d'[5/2]_3^0$ and $3d'[5/2]_2^0$ levels. Thanks to the conditions (a) and (c) we may fix the point $p = f(\Delta E)$ for the $3d'[5/2]_3^0$, $3d'[5/2]_2^0$ levels on the curve indicated by points 0, 1, 2 (cf. Fig. 2). The condition (b) allows to ascertain that the pressure taken from the curve will be also the thermalization pressure for the He—Ne mixture. The detailed thermal equilibrium between the $3d'[5/2]_3^0$, $3d'[5/2]_2^0$ levels takes place at the pressure of about 0.5 Tr as can be seen from the Fig. 2. In described experiment the pressure of He—Ne mixture was of about 2 Tr so we may with a good accuracy assume that a detailed thermal equilibrium occurs between the $3d'[5/2]_3^0$, and $3d'[5/2]_2^0$ levels.

Now, we will discuss the small increase in population of the $3d[5/2]_3^0$ and $3d[5/2]_2^0$ levels when the 7.6994 μm oscillation appears. These levels are coupled through radiative

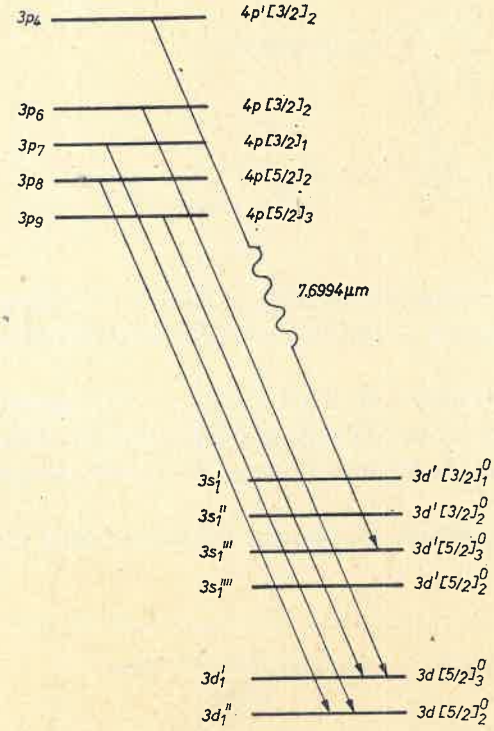


Fig. 3. Diagram of considered levels. The main transitions are stressed $4p \to \begin{matrix} 3d[5/2]_3^0 \\ 3d[5/2]_2^0 \end{matrix}$

transitions with the $4p$ levels (Fig. 3). Because the population of all $4p$ levels except the $4p[1/2]_1$ level (population change for this level is zero) decreases as a result of the 7.6994 μm action, we might expect the similar effect for populations of the $3d[5/2]_3^0$ and $3d[5/2]_2^0$

levels. The small increase in populations of the latter levels indicates that there is some other process opposite to the previous one. That is confirmed by the behaviour of these levels during the simultaneous generation of 7.6994 μm and 3.3913 μm in the case when the population change for the $4p'[3/2]_2$ is zero. In this time the population of all $4p$ levels except the $4p'[1/2]_0$ level did not change — Table I, column VI. In this case the population changes for the $3d[5/2]_3^0$ and $3d[5/2]_2^0$ levels increased slightly, hence they had the same character as the population changes for the $3d'[3/2]_1^0 \dots 3d'[5/2]_2^0$ levels. That is the evidence that these levels are coupled through collisions. We mentioned before in the case of the $5s$ levels that even for large distances between the levels excitation transfer may have high cross section if the Wigner rule is fulfiled. From the results presented in Table I and II we may suppose that the $3d[5/2]_3^0$ level is also excited by collisions of the atoms in the ground state with the atoms in the $3d'[5/2]_2^0$ state and the $3d[5/2]_2^0$ state with the atoms in the $3d'[5/2]_3^0$ state.

3. Evaluation of the ratio of the $3d'[5/2]_3^0$ ($3d'[5/2]_2^0$) level decay constant related to the non-radiative transitions $3d'[5/2]_3^0 \to 3d'[5/2]_2^0$ ($3d'[5/2]_2^0 \to 3d'[5/2]_3^0$) and the decay constant connected with all the other transitions

The ratios mentioned in the title may be calculated owing to the existence of thermal equilibrium between the $3d'[5/2]_3^0$ and $3d'[5/2]_2^0$ levels. The level symbols, important for the further consideration, are shown in Fig. 4.

Let us consider the balance equations describing population of the level 0 for the cases denoted in Fig. 4 as (a), (b), (c): the case (a) — there is no laser action in the system

$$R_0+\gamma_{60}N_6^a+\gamma_{40}N_4^a+\tilde{\gamma}_{10}N_1^a = \tilde{\gamma}_0 N_0^a+\tilde{\gamma}_{01}N_0^a, \tag{III.2}$$

the case (b) — there is a laser action $8 \to 6$

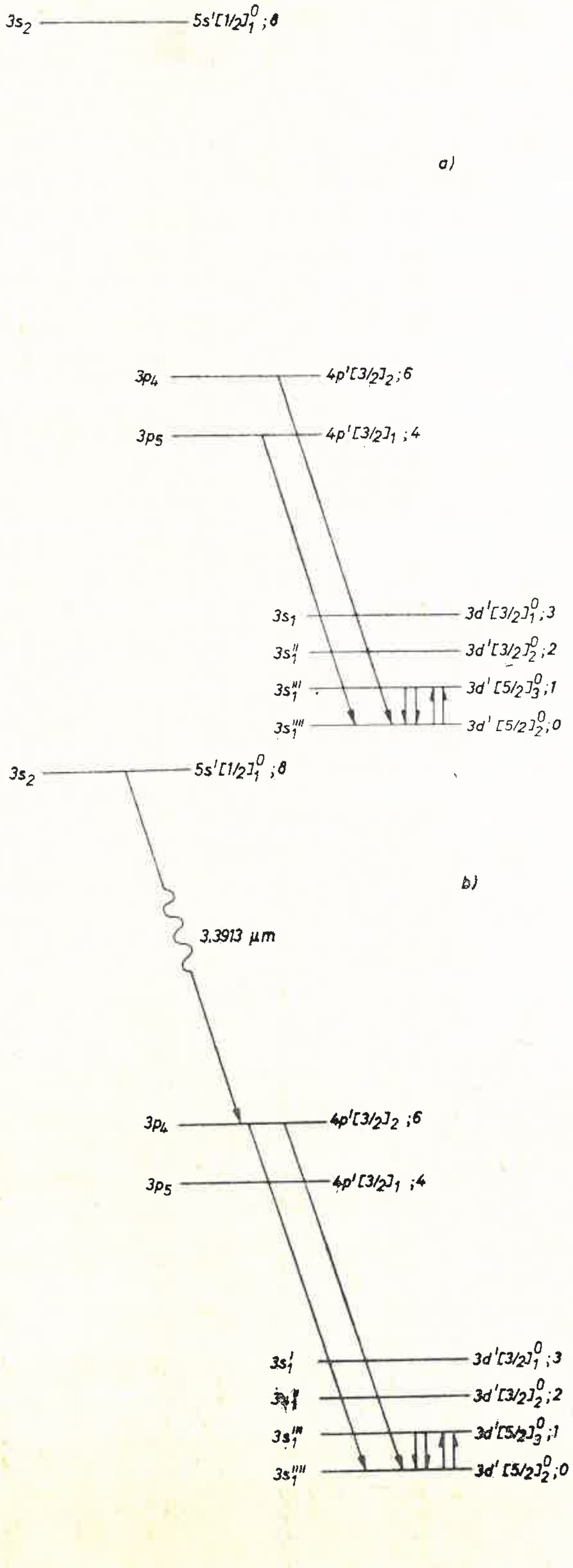
$$R_0+\gamma_{60}N_{6k}^b+\gamma_{40}N_{4k}^b+\tilde{\gamma}_{10}N_{1k}^b = \tilde{\gamma}_0 N_{0k}^b+\tilde{\gamma}_{01}N_{0k}^b, \tag{III.3}$$

the case (c) — there is a cascade laser action $8 \to 6 \to 1$

$$R_0+\gamma_{60}N_{6k}^c+\gamma_{40}N_{4k}^c+\tilde{\gamma}_{10}N_{1k}^c = \tilde{\gamma}_0 N_{0k}^c+\tilde{\gamma}_{01}N_{0k}^c. \tag{III.4}$$

Similarly to I, N_i^a-denotes population of the "i" level without the laser actions, N_{ik}^c — population in the presence of the cascade $8 \to 6 \to 1$ denoted by "k", N_{ik}^b — in the presence of the laser action $8 \to 6$, i.e. the upper transition in the cascade "k". The other symbols denote: γ_{60}, γ_{40} — transition probabilities of spontaneous emission between the levels $6 \to 0$ and $4 \to 0$ respectively, $\tilde{\gamma}_{10}$ — the decay constant of the "1" level population connected with nonradiative transitions from the level "1" to "0", $\tilde{\gamma}_{01}$ — the decay constant of the "0" level population connected with the opposite process, $\tilde{\gamma}_i$ — the decay constant of the "i" level population connected with the radiative and nonradiative transitions (excluding $1 \to 0$).

We assumed that in the equations (III.2)—(III.4) the only important transitions, for the 0 level population, are the radiative transitions $6 \to 0$ and $4 \to 0$. We made this assumption according to the transition probabilities [11] and the population changes for


3s2
5s'[1/2]^0_1 ;8
a)
3p4
4p'[3/2]_2 ;6
3p5
4p'[3/2]_1 ;4
3s1
3d'[3/2]^0_1 ;3
3s1''
3d'[3/2]^0_2 ;2
3s1'''
3d'[5/2]^0_3 ;1
3s1''''
3d'[5/2]^0_2 ;0
3s2
5s'[1/2]^0_1 ;8
b)
3.3913 µm
3p4
4p'[3/2]_2 ;6
3p5
4p'[3/2]_1 ;4
3s1'
3d'[3/2]^0_1 ;3
3s1''
3d'[3/2]^0_2 ;2
3s1'''
3d'[5/2]^0_3 ;1
3s1''''
3d'[5/2]^0_2 ;0

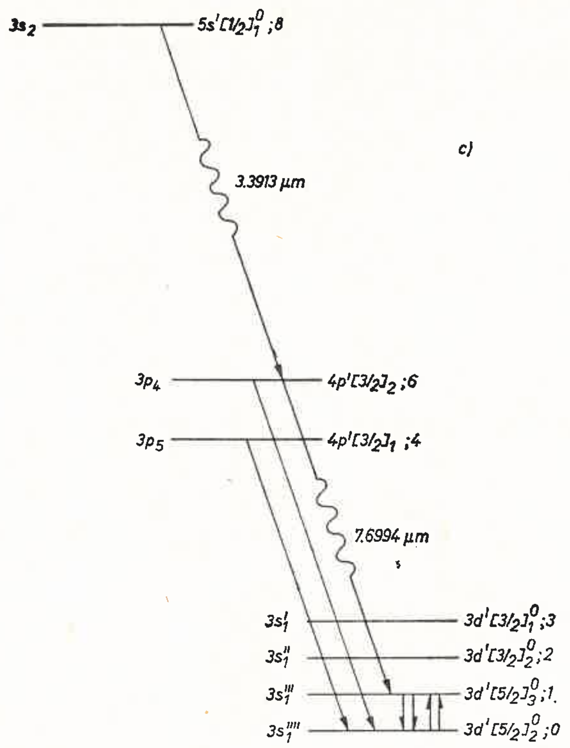


Fig. 4a, b, c. Solid, wavy and double lines denote spontaneous, stimulated and nonradiative transitions respectively

the $4p$ levels caused by the switching on the considered laser transitions (Table I). For the detailed equilibrium we can write the following equations [5] for the $\tilde{\gamma}_{10}$ and $\tilde{\gamma}_{01}$ constants and for populations N_1^a, N_0^a

$$\frac{\tilde{\gamma}_{10}}{\tilde{\gamma}_{01}} = \frac{g_0}{g_1} \exp\left(\frac{E_1 - E_0}{kT}\right) \tag{III.5}$$

$$\frac{N_1^a}{N_0^a} = \frac{g_1}{g_0} \exp\left(-\frac{E_1 - E_0}{kT}\right). \tag{III.6}$$

From the equations (III.2)—(III.4) and taking into account relations (III.5) and (III.6) we get

$$\frac{\gamma_{60}}{\tilde{\gamma}_0}\left(\frac{N_{6k}^b - N_6^a}{N_6^a}\right)\frac{N_6^a}{N_0^a} + \frac{\gamma_{40}}{\tilde{\gamma}_0}\left(\frac{N_{4k}^b - N_4^a}{N_4^a}\right)\frac{N_4^a}{N_0^a} - \left(\frac{N_{0k}^b - N_0^a}{N_0^a}\right)$$

$$= \frac{\tilde{\gamma}_{01}}{\tilde{\gamma}_0}\left(\frac{N_{0k}^b - N_0^a}{N_0^a} - \frac{N_{1k}^b - N_1^a}{N_1^a}\right) \tag{III.7}$$

TABLE IV

Investigated level:		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]
in Racah notation	„i" according to scheme in Fig. 4	3.3913 μm $(N^{b}_{ik}-N^{a}_{i})/N^{a}_{i}$	7.6994 μm in presence of 3.3913 μm $(N^{c}_{ik}-N^{a}_{i})/N^{a}_{i}$	8.0066 μm in presence of 3.3913 μm $(N^{c}_{im}-N^{a}_{i})/N^{a}_{i}$	
$4p'[3/2]_2$	6	+117.0	+38.1	+105.1	3418* $4p'[3/2]_2 \to 3s\ [3/2]^0_1$ 3418* $4p'[3/2]_1 \to 3s\ [3/2]^0_1$
$4p'[3/2]_1$	4	+80.0	+21.9		3600 $4p'[3/2]_1 \to 3s'[1/2]^0_1$
$3d'[5/2]^0_3$	1	+10.6	+68.1	+16.1	7943* $3d'[5/2]^0_3 \to 3p\ [5/2]_2$ 7944* $3d'[5/2]^0_2 \to 3p\ [5/2]_2$
$3d'[5/2]^0_2$	0	+12.8	+34.3	+28.8	8591 $3d'[5/2]^0_2 \to 3p'[3/2]_1$

* The asterisk indicates the line used as an indicator of the population change, overlapping with the neighbouring line.

and

$$\frac{\gamma_{60}}{\tilde{\gamma}_0}\left(\frac{N^{c}_{6k}-N^{a}_{6}}{N^{a}_{6}}\right)\frac{N^{a}_{6}}{N^{a}_{0}}+\frac{\gamma_{40}}{\tilde{\gamma}_0}\left(\frac{N^{c}_{4k}-N^{a}_{4}}{N^{a}_{4}}\right)\frac{N^{a}_{4}}{N^{a}_{0}}-\left(\frac{N^{c}_{0k}-N^{a}_{0}}{N^{a}_{0}}\right)$$

$$=\frac{\tilde{\gamma}_{01}}{\tilde{\gamma}_0}\left(\frac{N^{c}_{0k}-N^{a}_{0}}{N^{a}_{0}}-\frac{N^{c}_{1k}-N^{a}_{1}}{N^{a}_{1}}\right). \qquad \text{(III.8)}$$

From the equations (III.7) and (III.8) the ratio $\tilde{\gamma}_{01}/\tilde{\gamma}_0$ is obtained. Following numerical values are assumed to calculate this ratio

— the relative population changes of levels: from Table IV,

— $\gamma_{60} = 0.161\times10^5\ \text{sec}^{-1}$, $\gamma_{40} = 2.109\times10^5\ \text{sec}^{-1}$ from [11],

— $N^{a}_{6}/N^{a}_{0} = 1.22$, calculated from the relations (I.13) and (III.6),

— $N^{a}_{4}/N^{a}_{0} \geqslant g_4/g_0 = 3/5$, calculated from the threshold condition for the laser action in the transition $4\to0$ (8.0066 μm).

The ratio of decay constants fulfils the inequality

$$\tilde{\gamma}_{01}/\tilde{\gamma}_0 \geqslant 0.89, \qquad \text{(III.9)}$$

because the ratio N^{a}_{4}/N^{a}_{0} have to be bigger than g_4/g_0. In the same way the ratio of decay constant of the "1" level related to nonradiative transitions $1\to0$ and the decay constant of this level connected with all other transitions (more details in Appendix) is

$$\tilde{\gamma}_{10}/\tilde{\gamma}_1 \approx 0.62. \qquad \text{(III.10)}$$

We will get the ratio of decay constants for 0 and 1 levels dividing equations (III.10) and (III.9) by sides and taking into account that $\tilde{\gamma}_{01}/\tilde{\gamma}_{10} \approx 1.4$ (III.5)

$$\tilde{\gamma}_1/\tilde{\gamma}_0 \geqslant 1.03. \qquad \text{(III.11)}$$

It follows that

$$\frac{\tilde{\gamma}_1 N^{a}_{1}}{\tilde{\gamma}_0 N^{a}_{0}} \geqslant 1.44, \qquad (N^{a}_{1}/N^{a}_{0} \approx 1.4 - \text{(III.6)}),$$

the number of transitions from the $3d'[5/2]^{0}_{3}$ is larger than the number of transitions from the $3d'[5/2]^{0}_{2}$ level.

The constants $\tilde{\gamma}_0$ and $\tilde{\gamma}_1$ have, in the balance equations (III.2) and (A.1) — Appendix, the form

$$\tilde{\gamma}_0 = \gamma_{0p}+\tilde{\gamma}_{02}+\tilde{\gamma}_{03}-\tilde{\gamma}_{20}N^{a}_{2}/N^{a}_{0}-\tilde{\gamma}_{30}N^{a}_{3}/N^{a}_{0},$$

$$\tilde{\gamma}_1 = \gamma_{1p}+\tilde{\gamma}_{12}+\tilde{\gamma}_{13}-\tilde{\gamma}_{21}N^{a}_{2}/N^{a}_{1}-\tilde{\gamma}_{31}N^{a}_{3}/N^{a}_{1},$$

where γ_{ip} is the decay constant of the level "i" connected with radiative transitions, $\tilde{\gamma}_{ij}$ is the decay constant of the level "i" connected with nonradiative transitions $i\to j$. If there was the detailed thermal equilibrium between the levels 0 and 2, 0 and 3, and between levels 1 and 2, 1 and 3 then in such a situation the ratio of constants $\tilde{\gamma}_0$ and $\tilde{\gamma}_1$ would be totally determined by the ratio of radiative decay constants for the levels 0 and 1. From the data presented in Table III we would obtain for this ratio the value

$$\tilde{\gamma}_0/\tilde{\gamma}_1 = \gamma_{0p}/\gamma_{1p} \approx 1.13$$

4. *Analysis of population changes for the 3p levels*

As it was underlined in [3], [17] the population changes for the $3p$ levels caused by the oscillation 3.3913 μm are the resultants of the changes transferred from the $4p$ levels by the radiative cascade transitions through the intermediate levels $4s$ and $3d$ and from the $5s$ levels by the direct transitions.

Weaver and Freiberg [3] calculated, assuming the 100% increase in the $4p'[3/2]_2$ level population as a result of the 3.3913 μm generation, among the other values the population changes for the first four levels from the $3d$ group — Table V.[1] The authors state

TABLE V

Symbol of the level in Racah notation	Change of the level population (in per cent) of population change for the $4p'[3/2]_2$ level:	
	[3]	presented work
$3d'[3/2]^0_1$	+0.04	~ 9.8
$3d'[3/2]^0_2$	+0.27	~ 9.0
$3d'[5/2]^0_3$	+2.56	~ 9.1
$3d'[5/2]^0_2$	+0.17	~10.9

that the excitation transfer to the $3p$ levels in transitions $4p \to 3d \to 3p$ may be completely neglected because the population changes for the levels (Table V) are very small, and that the only transition $4p'[3/2]_2 \to 3d'[5/2]^0_3 \to 3p'[3/2]_2$ has bigger contribution. This kind of interpretation raises some doubts when we take into account the experimental facts (among the other found in present work). Such a situation would be in fact true if we ignored the radiative transitions $4p \to 3d$ and exchange collisions between the $3d$ levels. However, it would not correspond with the reality, especially because of the strong transitions $4p'[1/2]_1$, $4p'[3/2]_1 \to 3d$. It should be reminded that the $4p'[1/2]_1$, $4p'[3/2]_1$ levels are very strongly coupled with the $4p'[3/2]_2$ level through the nonradiative transitions (Table I, column III). This problem is discussed in details by Lis [9, 7]. Such an interpretation could not be valid also because of excitation exchange process which causes that all the $3d'[3/2]^0_1 \ldots 3d'[5/2]^0_2$ levels have almost identical population changes during the 3.3913 μm oscillation. Due to this fact the population changes for these (3d levels)

[1] The calculation was performed using the transition probabilities evaluated from the wave functions [18–20] obtained with the assumption of $j-l$ coupling. In this coupling the potential has the Coulomb form introduced in [21].

are slightly larger than the values expected from Weaver's and Freiberg's calculation — Table V.

The transitions from the $3d'[3/2]^0_1$, $3d'[3/2]^0_2$, $3d'[5/2]^0_2$ levels should be taken into account according to the above, except the $4p'[3/2]_2 \rightarrow 3d'[5/2]^0_3 \rightarrow 3p$ transition, since they may also contribute to the population changes for some $3p$ levels. Let us see on which levels from the $3p$ group these transitions may have an influence. To solve this problem, the 7.6994 μm laser action was excited in the presence of cooperating 3.3913 μm action which caused a strong population increase in the $3d'[5/2]^0_3$ and neighbouring levels. We observed then not too strong, but distinct increase in population of the $3p'[1/2]_1$, $3p(1/2]_0$, $3p'[3/2]_2$ levels which according to [11] is mainly due to transitions

$$3d'[3/2]^0_2 \rightarrow 3p'[1/2]_1$$

$$3d'[3/2]^0_1 \rightarrow 3p[1/2]_0$$

$$3d'[5/2]^0_3 \rightarrow 3p'[3/2]_2.$$

Let us notice the levels in interest are coupled with the $5s$ and $4p$ levels and there is a decrease in population of these levels when we switch on the 7.6994 μm generation.

The levels belonging to the $3d'$ group changed the sign in the similar way after switching on the 5.403 μm oscillation. Let us notice, however, that the population change in percents for the $3p[1/2]_0$ level is bigger than for the $3p'[3/2]_2$ level in this case. While during the 7.6994 μm oscillation this relation is opposite. It confirms the fact that the $3p[1/2]_0$ level is mainly coupled with the $3d'[3/2]^0_1$ level, and $3p'[3/2]_2$ with $3d'[5/2]^0_3$ level.

We also observed the increase in population of the $3p'|1/2]_0$ and $3p(3/2]_1$ levels during the oscillation 5.403 μm that indicated their connection with the $3d'[3/2]^0_1$ level.

APPENDIX

Determination of the $\tilde{\gamma}_{10}/\tilde{\gamma}_1$ ratio

Let us consider the balance equations describing the population of the level 1 in the stationary state for the following cases (in accordance to the Fig. 5a, b, c)
the case (a) — there is no laser action in the system

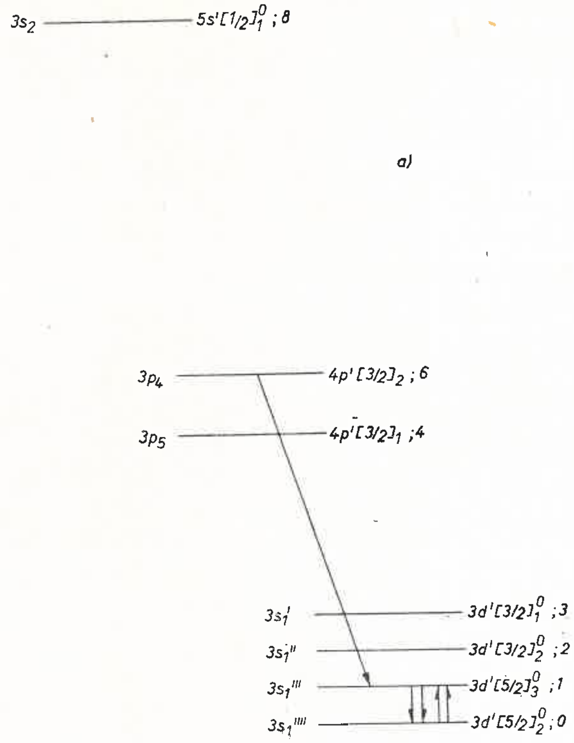
$$R_1+\gamma_{61}N^a_6+\tilde{\gamma}_{01}N^a_0 = \tilde{\gamma}_1 N^a_1+\tilde{\gamma}_{10}N^a_1, \tag{A.1}$$

the case (b) — there is the laser action $8 \rightarrow 6$

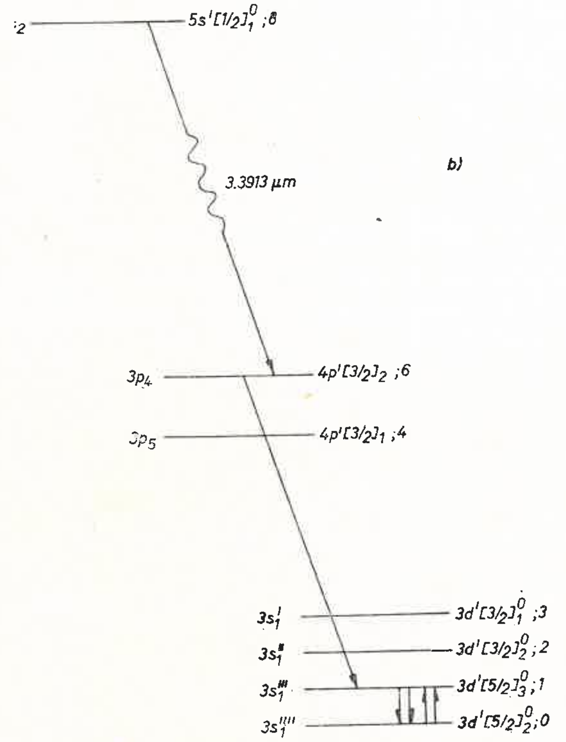
$$R_1+\gamma_{61}N^b_{6k}+\tilde{\gamma}_{01}N^b_{0k} = \tilde{\gamma}_1 N^b_{1k}+\tilde{\gamma}_{10}N^b_{1k}, \tag{A.2}$$

the case (c) — there are the laser actions $8 \rightarrow 6$ and $4 \rightarrow 0$. Level population in this case will be denoted by N^c_{im}.

$$R_1+\gamma_{61}N^c_{6m}+\tilde{\gamma}_{01}N^c_{0m} = \tilde{\gamma}_1 N^c_{1m}+\tilde{\gamma}_{10}N^c_{1m} \tag{A.3}$$


3s2
5s'[1/2]$_1^0$; 8
3p4
4p'[3/2]2 ; 6
3p5
4p'[3/2]1 ; 4
3s1'
3d'[3/2]$_1^0$; 3
3s1''
3d'[3/2]$_2^0$; 2
3s1'''
3d'[5/2]$_3^0$; 1
3s1''''
3d'[5/2]$_2^0$; 0

a)


5s'[1/2]$_1^0$; 8
3.3913 μm
3p4
4p'[3/2]2 ; 6
3p5
4p'[3/2]1 ; 4
3s1'
3d'[3/2]$_1^0$; 3
3s1''
3d'[3/2]$_2^0$; 2
3s1'''
3d'[5/2]$_3^0$; 1
3s1''''
3d'[5/2]$_2^0$; 0

b)

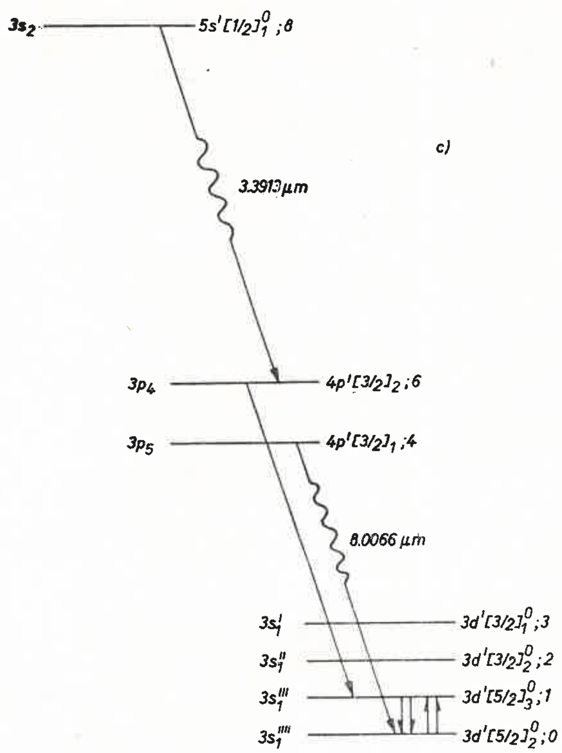


Fig. 5a, b, c. Solid, wavy and double lines denote spontaneous, stimulated and nonradiative transitions respectively

The radiative transition 6 → 1 is included to above equations. This transition, among the all possible $4p \rightarrow 3d'[5/2]_3^0$ transitions is distinguished because of a big transition probability [11]. We obtained from the equations (A.1)—(A.3), (III.5) and (III.6) the relation similar to (III.7) and (III. 8). In order to obtain the ratio $\tilde{\gamma}_{10}/\tilde{\gamma}_1$ we put into the appropriate relations the numerical data of relative population changes for considered levels (Table IV). We assumed for the ratio N_6^a/N_1^a the value taken from equation (II.2).

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