INVESTIGATIONS OF POPULATIONS FOR $5s'[1/2]^0_1$ AND $4p'[3/2]_2$ NEON LEVELS INTERACTING WITH LASER RADIATION AT 633 AND 3391 nm

BY L. LIS

Institute of Physics, Polish Academy of Sciences, Warsaw*

(Received April 6, 1978; revised version received February 2, 1979)

Populations of the $5s'[1/2]^0_1$ and $4p'[3/2]_2$ neon levels, interacting with simultaneous laser radiations at 633 and 3391 nm, were investigated as functions of neon pressure. The experiment showed that under such conditions an additional population transfer to the $4p'[3/2]_2$ level, not coming from the $5s'[1/2]^0_1$ level, appeared. A share of this population transfer was estimated by means of population balance equations.

The problem of resonant interaction between two competitive laser radiations and excited neon atoms was discussed by Brunner, Hertz et al. in Refs [1–2]. In this paper we shall reconsider a question, in our opinion, very close to that of the quoted references. To formulate this question let us assume three levels of the multilevel neon system, and two strong radiations, at 633 and 3391 nm, inducing optical transitions between these levels (see Fig. 3a). Next, take the level $4p'[3/2]_2$ and ask — "can the changes in populations of this level, induced by 3391 laser radiation, be defined by changes in the population of the $5s'[1/2]^0_1$ level". This question was experimentally resolved by the method of changes in populations induced by two simultaneously acting laser radiations. While the radiation at 3391 nm was interrupted, two of the levels, $5s'[1/2]^0_1$ and $4p'[3/2]_2$, were investigated; intensity of another beam of radiation at 633 nm was held constant.

1. Experiment

As has been said the experiment was carried out for two laser radiations and neon levels $5s'[1/2]^0_1$ and $4p'[3/2]_2$. The laser beam at 633 nm crossed the one at 3391 nm at a right angle inside a probing cell with neon discharge, see Fig. 1. Such a setting for the beams was necessary to avoid a Doppler profile perturbation of the investigated levels by a mode

* Address: Instytut Fizyki PAN, Al. Lotników 32/46, 02-668 Warszawa, Poland.

structure of the pumping laser radiation at 633 nm. Intensities of spectral lines at 543 and 342 nm, which reflect populations of the levels, were measured perpendicularly to both laser beams with a grating monochromator and recording system. The population ratios used in the experiment were found by the plasma transparency method [3–4].

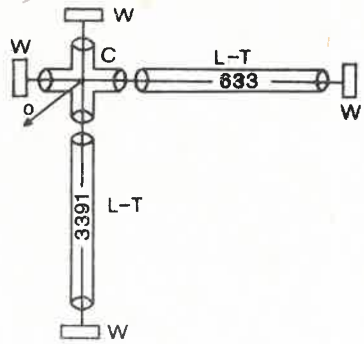

Fig. 1. Scheme of the experimental apparatus: L—T — laser tubes, C — probing cell, W — mirrors O — observation direction. On the diagram prisms inserted into laser cavities are not showed

2. *Results*

We begin with the following observation. The laser radiation at 633 nm increases the $5s'[1/2]_1^0$ level population to such a degree that population inversion on the 3391 transition appears even for a pure neon discharge within the wide pressure range. The changes induced by the 3391 laser radiation, when the one at 633 nm is present, are drawn on Fig. 2. They

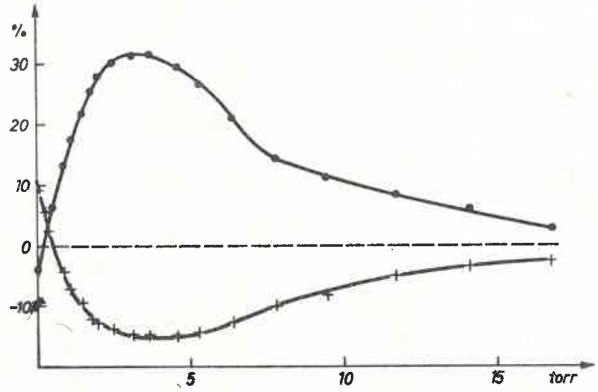

Fig. 2. Plot of percentage changes in intensities of spectral lines, $5s'[1/2]_1^0 \rightarrow 3p[1/2]_1$ — 543 nm, crosses, and $4p'[3/2]_2 \rightarrow 3s[3/2]_1^0$ — 342 nm, dots, with neon pressure. The changes at 633 laser power were obtained from interrupting the 3391 laser radiation

show a maximum and minimum (for the $4p'[3/2]_2$ and $5s'[1/2]_1^0$ levels, respectively) at 3 torr, where the 633 laser radiation increases the population ratio of the investigated levels $5s'[1/2]_1^0$ and $4p'[3/2]_2$ to unity, whereas, without any laser radiation this ratio (at the same pressure as above) is 0.18. Below 3 torr the changes go to zero, but what is very strange, the zero-changes for each of the levels are attained at different neon pressures.

In the experimental situation without laser radiation at 633 nm when plasma transparency at 3391 nm is reached (due to the addition of helium), in accordance with the balance equations as in Refs [5–6], changes of the levels vanish simultaneously.

As is seen, in this experiment population balance as is Refs [5–6], taking into account only energy transfer between the investigated levels, is not fulfilled. Let us notice (see Fig. 2) that there is a pressure range for which the changes have this same plus sign, and here we are not able to state whether the plasma absorbs or gains the 3391 radiation. It seems that at this point where we have no changes in population of the $5s'[1/2]_1^0$ level, with good accuracy, transparency of the plasma to the 3391 transition is obtained.

The effect can be explained by assuming an additional population transfer, induced by both laser radiations, to the $4p'[3/2]_2$ level and not coming from the $5s'[1/2]_1^0$ one.

3. *Discussion*

In further discussion the levels $5s'[1/2]_1^0$ and $4p'[3/2]_2$ will be called 2 and 3 as in Fig. 3. We can distinguish four situations: a) without any laser radiation, b) with laser radiation at 633 nm, c) with laser radiation at 3391 nm, d) with both 633 and 3391 laser radiations.

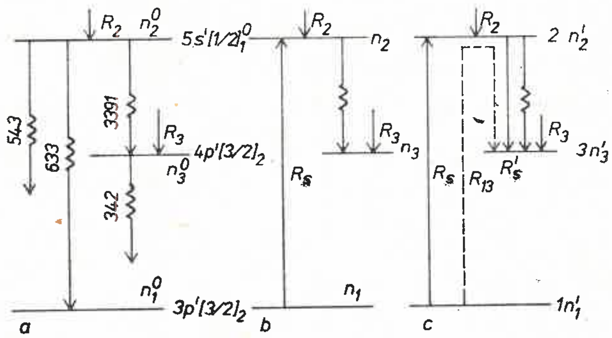

Fig. 3. Scheme of neon levels and optical transitions discussed in this work. a — situation without laser radiation, n^0 — symbols of level populations, b — situation with laser radiation at 633 nm, n, R_s — populations and laser pumping rate, c — situation with both laser radiations at 633 and 3391 nm, n', R_s' — populations and laser pumping rate between 2 and 3 levels

The next discussion will be concerned with the effect of transition from b) to d) situation. In the situation b) the transitions 1–2 are strongly induced with a rate R_s. Turning the 3391 laser radiation on, besides the induced processes 2–3, the additional population transfer to the level $3(R_{13})$ appears. Using the equations of population balance, holding R_s constant we obtain

$$\alpha(R_s) = \frac{n_3'-n_3}{n_2-n_2'} = \frac{\gamma_2^*-\gamma_{23}^*}{\gamma_3^*} + \frac{R_{13}}{(n_2-n_2')\gamma_3^*}, \tag{1}$$

where n, n' are populations in situations b) and d) respectively, γ_2^*, γ_3^*, γ_{23}^* — effective decay rates and transition probability. The collisions, which transfer the population from levels 2 and 3 to the others, causes γ^* and $\alpha(R_s)$ to be functions of neon pressure. Evidently we must assume $R_{13} = 0$ for $R_s = 0$ and we have

$$\alpha(R_s)-\alpha(0) = \frac{R_{13}}{(n_2-n_2')\gamma_3^*}\,, \tag{2}$$

where $\alpha(0) = \dfrac{n_3^0-n_3''}{n_2''-n_2^0}$ and n^0, n'' are populations for a) and c) situations, respectively.

Formula (1) shows that the changes in population of the 2 and 3 levels do not attain zero-value simultaneously if only $R_{13} \neq 0$. In Fig. 4 experimental values of $\alpha^*(R_s)$ and $\alpha^*(0)$ ($\alpha^* = -\alpha$) are given as functions of neon pressure.

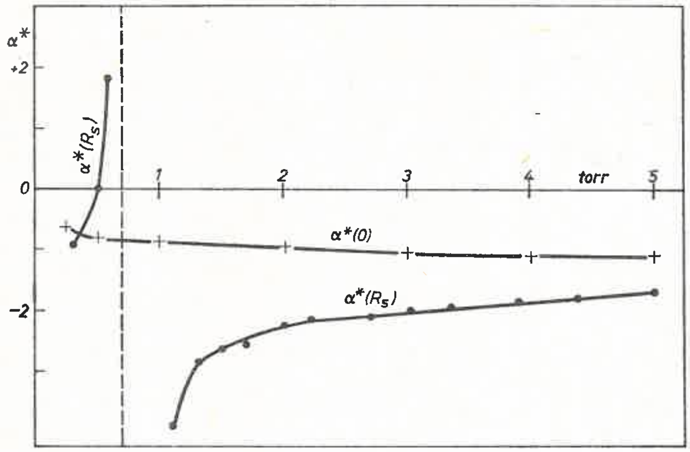

Fig. 4. Variation of $\alpha^*(R_s)$ and $\alpha^*(0)$ with neon pressure; vertical dashed line is asymptote $n_2-n_2' = 0$ at 0.7 torr of neon pressure

A share of R_{13} in respect to R_s can be computed from (2). Because $R_s = \gamma_2^*(n_2-n_2^0)$, see Ref. [5], so we have

$$\frac{R_{13}}{R_s} = \frac{\gamma_3^*}{\gamma_2^*}[\alpha(R_s)-\alpha(0)]\,\frac{n_2-n_2'}{n_2-n_2^0} \quad \text{for} \quad n_2-n_2' \neq 0$$

and

$$\frac{\gamma_3^*}{\gamma_2^*}\,\frac{n_3'-n_3}{n_2-n_2^0} \quad \text{for} \quad n_2-n_2' = 0.$$

For example, at 5 torr of neon pressure $\alpha(R_s)-\alpha(0) = 0.6$ and $(n_2-n_2') : (n_2-n_2^0) = 0.2$. Assuming $\gamma_2^* \approx \gamma_3^*$ we have $R_{13} : R_s = 0.12$. Analogous calculations for the transparent plasma give 0.10.

Up to now we have not said from which level the additional population transfer is coming. It seems that it may only be from the $3p'[3/2]_2$ level. The minus changes in population of this level were observed when 3391 laser radiation was interrupted, while 633 laser radiation was held constant. Would this effect be caused by processes described in Ref. [7], pages 377–391?

One could ascribe the effect to changes in R_1, R_2, R_3, R_s caused by switching the 3391 laser radiation on. However, this is not the case when the plasma transparency at 3391 nm is reached, because then all these changes should vanish as well.

Experimental errors did not exceed $\pm 0.5\%$ for population changes, $\pm 2.5\%$ for α and ± 0.1 torr for neon pressures.

4. Conclusion

Summarizing we may answer the question formulated at the beginning of the work. The results show that under our experimental conditions changes in population of the $5s'[1/2]_1^0$ level are not sufficient for describing the changes in population of the $4p'[3/2]_2$ level. This is because of the population flow R_{13} (not coming from the $5s'[1/2]_1^0$ level) which only appears when both laser radiations simultaneously act on excited neon atoms.

REFERENCES

[1] W. Brunner, H. Paul, G. Richter, *Ann. Phys.* (Germany) **22**, 119 (1969).
[2] J. Hertz, K. Hoffmann, *Ann. Phys.* (Germany) **22**, 134 (1969).
[3] T. Hänsch, P. Toschek, *Phys. Lett.* **20**, 273 (1966).
[4] T. V. Bychkova, V. G. Kirpilenko, S. G. Rautian, A. S. Khaikin, *Opt. Spektrosk.* **22**, 678 (1967).
[5] T. F. Johnston, *Appl. Phys. Lett.* **4**, 161 (1970).
[6] L. Lis, *Acta Phys. Pol.* **A42**, 307 (1972).
[7] V. M. Fain, *Fotony i Nelineinye Sredy*, Sovetskoe Radio, Moskva 1972.