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TRANSPARENCY AND RATE EQUATIONS FOR 3391 nm TRANSITION IN NEON

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Population inversion in the 3391 nm transition in a He–Ne discharge was experimentally measured with transparency and rate equations methods. Experiment shows large differences between the results obtained with the two methods.

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1. Introduction

The population ratio of a pair of atomic levels is the fundamental parameter which informs about the ability of lasing on an optical transition between energy levels in an excited atomic ensemble. Thus, it is very important in laser techniques to have a simple and credible experimental method of measuring the population ratio of the levels.

Hitherto the only method fulfilling these criteria seemed to be the so-called transparency method applied to the experiment by Hänsch and Toschek [1] and also by others [2–8]; on transparency for two transitions with a common level see Ref. [9]. However, quite recently, Kuppens et al. have published a paper in which they present, among other things, a method for measuring population ratio on the basis of rate equations of the level populations [10]. An almost identical method was presented and applied by Johnston to investigations of an Ar^{+} ion laser [11, 12].

In this paper we present both methods in detail and confront them in an experiment on a He–Ne tube in which the stimulated oscillations at 3391 nm are developed without a resonator.

2. The transparency method

The experimental realization of the transparency method is illustrated in Fig. 1. A probe cell PC, containing the investigated medium, is illuminated by a strong radiation beam LB at a wavelength corresponding to an atomic transition

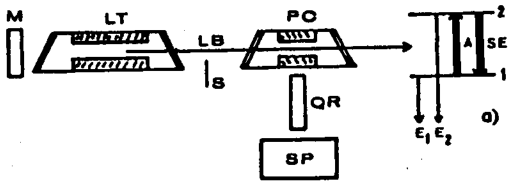

Fig. 1. Experimental setup for transparency investigations: laser tube LT and mirror M form a system generating a coherent laser beam LB. QR is quartz glass rod, SP is a grating spectrophotometer, S is a screen for stopping LB. (a) Scheme of levels interacting with LB. SE is a number of stimulated by LB emission transitions, A is a number of stimulated absorption transitions, E_2 and E_1 are intensities of spontaneously emitted lines starting from the levels 2 and 1, respectively.

between the investigated upper and lower levels, 2 and 1, respectively, as shown in Fig. 1a.

Let N_2 and N_1 be the populations of the levels defined as numbers of atoms excited to the levels per unit volume of the medium.

Optical properties of the medium are radically different for the following possible physical situations: (a) $N_2/g_2 > N_1/g_1$, (b) $N_2/g_2 < N_1/g_1$ and (c) $N_2/g_2 = N_1/g_1$, where g_2 and g_1 are statistical weights of the levels 2 and 1, respectively, Fig. 1a.

In (a) the LB induces more stimulated emission transitions SE than absorptions A. The LB is gained, and its switching on decreases and increases populations of levels 2 and 1, respectively.

In (b) the LB induces more transitions A than SE and the medium absorbs the LB. In this case, switching on the LB, inversely to the former situation, increases the population of level 2 and decreases that of level 1.

Finally, in case (c) the number of transitions SE is equal to the number of absorptions A, and the medium neither gains nor absorbs the LB. We say that the medium is transparent to the LB. Switching on the LB does not induce any changes in the level populations, both for level 2 and 1. These null changes are the best sign that the transparency of the medium has been attained, and that the population of level 2 divided by g_2 is equal to the population of level 1 divided by g_1.

In an experiment of the type shown in Fig. 1, the population changes may be monitored as changes in intensities of the spectral lines E_2, E_1 originating from the levels and emitted in a transversal direction to LB. Hence, in the transparent state of the medium we may write

$$\frac{E_2^\infty}{E_1^\infty} = \frac{c_2}{c_1}\frac{N_2^\infty}{N_1^\infty} = \frac{c_2}{c_1}\frac{g_2}{g_1}, \tag{1}$$

where c_2, c_1 are some functions of: the wavelengths of the spectral lines used in measurements, the detector sensitivities at these wavelengths, and geometrical configuration of the spectral and detection system. The superscript ∞ denotes the transparency state of the medium and suggests that transparency is also obtained when intensity of LB tends to infinity.

Now, for a given state of the medium, absorption or gain, we may write

$$\frac{N_2}{N_1} = \frac{E_2}{E_1}\frac{E_1^\infty}{E_2^\infty}\frac{g_2}{g_1}. \tag{2}$$

In an experimental system of the type as in Fig. 1 we change conditions of the medium excitation in such a manner as to obtain the null changes of the spectral lines monitored perpendicularly to LB when it is switched on and off. The ratio E_2^∞/E_1^∞ can then be measured and formula (2) may be used. More detailed considerations on the transparency method can be found in Ref. [3].

3. Rate equations

The second method of experimental determination of the population ratio is presented in Ref. [10]. The method also makes use of the side-light monitoring.

In order to present the working principle of the method let us consider population balance equations or rate equations for the levels of the atomic medium at the output end of the discharge as in Fig. 2. For our two levels, in stationary conditions, we may write

$$\gamma_2 N_2 = \lambda_2 - R_s, \tag{3a}$$

$$\gamma_1 N_1 = \lambda_1 + \gamma_{21} N_2 + R_s, \tag{3b}$$

where γ_2, γ_1, λ_2, λ_1 are decay rates and excitation velocities of levels 2 and 1, respectively, γ_{21} is the spontaneous rate of transition between the levels, and R_s is the difference between the velocity of stimulated emission SE and absorption A.

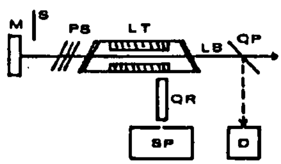

Fig. 2. Scheme of experimental setup for measurements of population ratios of levels 2 and 1. PS are soda glass plates, QP is a quartz splitting plate, D is a power meter system of LB. The remaining symbols are as in Fig. 1.

The general problem associated with the rate equations lies in proper definition of R_s, in particular for a transition with a mixed broadening, with homogeneous and inhomogeneous components. In this paper, following Kuppens et al. [10], we assume

$$R_s = \frac{\sigma I}{h\nu}\left(\frac{N_2}{g_2} - \frac{N_1}{g_1}\right), \tag{4}$$

where I is the intensity of the radiation of LB in the volume of the medium in which N_2 and N_1 are to be measured, σ is the stimulated emission cross-section, ν is the frequency of the LB radiation, and h is Planck's constant. Solving Eqs. (3) with (4) yields

$$n_i = \frac{N_i}{N_i^0} = \frac{1 + k_i I/I_s}{1 + I/I_s} \tag{5}$$

for $i = 1, 2$, where

$$k_1 = a\frac{\gamma_1\gamma_2 g_1}{\lambda_1\gamma_2 + \lambda_2\gamma_{21}}, \quad k_2 = a\frac{\gamma_2 g_2}{\lambda_2},$$

$$a = \frac{\lambda_1 + \lambda_2}{\gamma_2 g_2 - \gamma_{21} g_2 + \gamma_1 g_1}, \quad I_s = a\frac{h\nu\gamma_1\gamma_2\, g_1 g_2}{\sigma(\lambda_1 + \lambda_2)},$$

and superscript "0" is used to denote quantities for $I = 0$ and $R_s = 0$. The formula (5), derived by Kuppens et al. [10] and, in my opinion, also by Johnston [11], is the basis of the rate equations method (RE). If we are able to measure n_i and n_i' for the different values of I and I', respectively, we may compute k_i as follows:

$$k_i = \frac{(1 - n_i')n_i I - (1 - n_i)n_i' I'}{(1 - n_i')I - (1 - n_i)I'}, \tag{6}$$

and with the absolute value of I we obtain

$$I_s^i = I\frac{k_i - n_i}{n_i - 1}. \tag{7}$$

Let us notice that in formulas (6) and (7) we may put in place of n_i and n_i' respective intensity ratios of the spectral lines E_i/E_i^0 and E_i'/E_i^0 measured in the side-light spectrum.

The superscript i in (7) denotes the level whose population was used to compute the saturation intensity I_s. Naturally, according to the "theory" presented here there should be no difference between I_s^1 and I_s^2, however, experiment does not fully confirm the above conclusion. The values of k_1 and k_2 with $N_2^\infty/N_1^\infty = g_2/g_1$ then yield

$$\frac{N_2^0}{N_1^0} = \frac{k_1 g_2}{k_2 g_1}. \tag{8}$$

Also the ratio of the relative changes in the level populations δN_2 and δN_1 induced while LB is switched on and off may be given as follows:

$$\frac{\delta N_2}{\delta N_1} = \frac{1 - k_2}{k_1 - 1}, \tag{9}$$

where $\delta N_2 = 1 - N_2/N_2^0$ and $\delta N_1 = N_1/N_1^0 - 1$.

The formulas (8) and (9) allow us to obtain (see Ref. [12]):

$$\frac{\gamma_2 - \gamma_{21}}{\gamma_1} = \frac{k_2(k_1 - 1)g_1}{k_1(1 - k_2)g_2}. \tag{10}$$

As can be seen, the coefficients k_i play a very important role in computing the atomic ensemble parameters which has stimulated our interest in the RE method.

4. General remarks on the experiment

Both methods, the transparency method and the rate equations method, were confronted in experiments on the neon transition at 3391 nm which occurs between the $5s'[1/2]_1^0$ (2) and $4p'[3/2]_2$ (1) levels (Racah notation) [13]. Experiments were performed with and on a ^{3}He–^{20}Ne discharge tube with a flat gold mirror at one of the tube ends. The tube, which had a discharge region 150 cm in length and internal diameter of 0.3 cm, generated 12 mW of output power at 3391 nm.

For transparency investigations we used a short ^{3}He–^{20}Ne probe cell (PC) with discharge length of 5.5 cm. The cell was made of the same type of pyrex capillary as the long discharge tube. In this way, both tubes had the same inside and outside diameters.

The PC, with Brewster's windows, was filled with ^{3}He and ^{20}Ne isotopes at 1 : 5 ratio of partial pressure and at total pressure of 1.5 Tr.

Arbitrary populations of levels 2 and 1 were measured through intensities of the 543 and 341.79 nm (hereafter abbreviated to 342), respectively, which were monitored in the side-light spectrum [14]. Instead of the 543 line we could also use with success other lines starting from level 2 because these lines are well separated from the others.

However, we deal with a different situation in the case of level 1. Here each line is accompanied by a satellite line separated by about 0.01 nm, which strongly disturbs the measurements. These satellite lines start from the $4p'[1/2]_1$ neon level which is distant from the investigated $4p'[3/2]_2$ by about 1 cm^{-1} [15]. Our choice of the 342 line results from the relatively small intensity of its satellite.

To measure intensities of the lines we used a grating spectrophotometer with spectral resolution of about 0.1 nm. The side light of the laser tube or the probe cell was transmitted to the spectrophotometer entrance slit with a quartz glass rod of 0.3 cm diameter. The use of such a rod, as a light guide, avoided the chromatic aberration problems which are encountered with a lens or lens system. Thus, with the use of the rod the intensity ratio of the lines is the same for different distances of the rod entrance from the discharge tube and also for different distances of the rod exit from the entrance slit of the spectrophotometer, naturally for a sensible range of distances.

However, the main problem is with measurements of the 342 line since this line undergoes strong absorption in the tube wall which may be an obstacle in the transparency method. Hence for the reliable results, the probe cell and the laser tube have to be made of the same sort of glass and the tubes have to be of the same wall thickness. It was determined experimentally that a change in wall thickness of 0.01 cm changes the intensity ratio of the lines by 1.5%. Naturally, this problem does not apply to the rate equations method which does not require a separate probe cell.

5. Experiments and results

5.1. Transparency of probe cell

Transparency of the PC was tested in an experimental setup as in Fig. 1. The setup constituted the long tube, the probe cell, and side-light detecting system. The long tube, enhanced by the mirror M, sent the 3391 nm beam (LB) into the probe cell PC. Opaque screen S was used for switching LB on and off.

In Fig. 3 we present results of measurements of intensity ratio E_2/E_1 versus the probe cell current. The plots A and B were obtained when the LB passed and did not pass through the cell, respectively. As can be seen, the transparency of the discharge in PC is attained at a current of 22.5 mA, and below and above this current value the discharge in the PC shows some small gain and some small absorption, respectively.

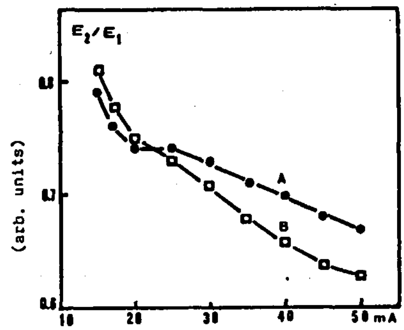

Fig. 3. Intensity ratios E_2/E_1 measured in the side-light spectrum of the *PC* versus its discharge current. Plots *A* and *B* are for *LB* passing and not passing through *PC*, respectively.

5.2. Population in the long tube

With E_2^∞/E_1^∞ measured in the *PC* we can evaluate the population ratio at any point of the discharge along the long tube, by measuring intensities of the 543 and 342 lines E_2 and E_1, respectively.

However, in the experiment we measured the line intensities at the output end of the discharge as in Fig. 2. This was because for the above part of the discharge, the radiation power which interacts with the neon atoms equals the output power of the tube and the obtained results may be analyzed both with the transparency and with the rate equations methods simultaneously.

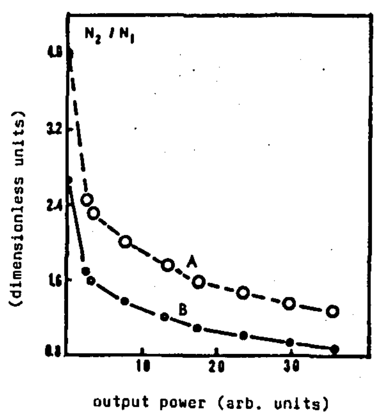

Fig. 4. Population ratio N_2/N_1 at the discharge as in Fig. 2 versus the output power of the generating system. Plots *A* and *B* are obtained with transparency and rate equations, respectively.

The measurements were performed at the discharge current of 32 mA as a function of the output of the tube. Different values of the output powers were obtained by putting various numbers of soda glass plates between the mirror M and the tube. In this way, we could change the output power from 12 to 1 mW. The lowest output power value was generated by the tube without the mirror M.

It is a well-known experimental fact that any stray reflection of the radiation into the tube changes its generation. In order to minimize such reflections we measured only a small part of the output, that reflected by a quartz glass plate QP, Fig. 2. In this way, perturbation of the tube generation by the dispersed beam from the power meter element was avoided or at least strongly discriminated.

The zero-power radiation in the discharge part under investigations, was obtained when the mirror M was removed and placed at the right end of the tube in such a way that it reflected the generated radiation back into the tube. In the discharge part to be investigated, the 3391 radiation of the tube becomes negligibly small and the level populations are very close to N_2^0 and N_1^0 [16].

In Fig. 4, plot A, we present experimental results of the population ratios versus the output power of the tube.

5.3. Rate equations measurements

In the rate equations method we present the measurements in the form of n_2 and n_1 versus output power of the tube, see Fig. 5. With n_i and n_i' measured at values of the output power of 36 and 18 of arbitrary units, respectively, we obtain $k_1 = 2.3$ and $k_2 = 0.5$ and, in consequence, other parameters which are listed in Table. In the table there are also given results of k_i and I_s collected from Ref. [10].

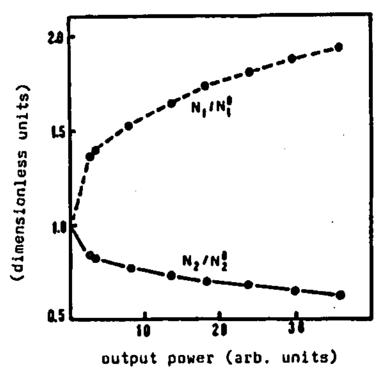

Fig. 5. Relative populations N_i/N_i^0 for the levels 2 and 1 versus output power of the system as in Fig. 2.

The results from Fig. 5 may be plotted as a function of $z = 1/(1 + I/I_s)$. Such a coordinate transformation, assuming $I_s = 13$ arb. units, gives the results of n_i arranged along two straight lines as in Fig. 6.

Population ratio N_2/N_1, obtained with the rate equations method, as a function of the tube output is shown in Fig. 4, plot B.

TABLE

RE and transparency method results.

	Our RE results	RE results from Ref. [10]
k_1	2.3 ± 0.05	2.3 ± 0.1
k_2	0.5 ± 0.02	0.43 ± 0.04
I_s^1	4.3 mW	360 μW
I_s^2	3.7 mW	440 μW
I_s^a	4.0 mW	400 μW
	Derived results from RE	Transparency method
N_2^0/N_1^0	2.7 ± 0.2	4.0 ± 0.2
$\delta N_2/\delta N_1$	2.6 ± 0.2	2.2 ± 0.1 (direct measur.)
$(\gamma_2 - \gamma_{21})/\gamma_1$	1.0 ± 0.1	0.55 ± 0.05

I_s^a — average saturation intensity.

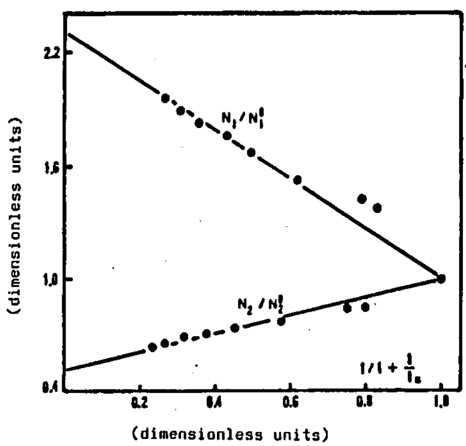

Fig. 6. Results as in Fig. 5 plotted against $z = 1/(1 + I/I_s)$ under assumption of $I_s = 13$ arb. units.

5.4. Discussion

Starting the discussion, let us digress from the main subject of the paper and let us look at the results in the table collected from Ref. [10]. Naturally, it is impossible to compare the results of Ref. [10] with ours because of different conditions of the He–Ne discharge. For instance, the authors used helium and neon isotopes in natural abundance and filled with them a laser tube with internal diameter of capillary of 0.1 cm (0.3 cm in our case).

However, in spite of the above differences in the discharge conditions, we notice that our results obtained with the rate equations are very close to those in Ref. [10]. Next, let us notice that our I_s divided by the cross-section area of the discharge is nearly the same as that from Ref. [10].

Now, let us turn to the main problem of this paper which is encountered on confronting our experimental results obtained with the rate equations with those from the transparency method. As is seen, the values of N_2/N_1 from transparency method are about 1.5 times bigger than those obtained with the rate equations, see Fig. 4, plots A and B, respectively. This difference which is outside the experimental error seems to be the result of a systematic error either of the rate equations or transparency method.

In searching for such an error, let us first reconsider the transparency method. In this method the main problem is to obtain the null changes in the level populations, irrespective of the spectrum of the LB, provided the spectrum is within the frequency range of the inhomogeneous broadening of the transition, or better, close to its centre. This spectrum may be, for instance, in single mode, although multimode radiation is more suitable in the experiment.

Let us now discuss a possible error of the method which may be caused by the self-absorption effect on the 342 nm line. Analogous self-absorption on the 609.6 nm line has been discussed and accounted for in the transparency experiments by Inatsugu et al. in Ref. [3]. We say analogous, since both lines, 342 and 609.6 nm, terminate on the same metastable neon level $3s[3/2]_1^0$.

The self-absorption, increasing with increase in neon contamination, is larger in the transparent than in the gaining He–Ne mixture. Hence, E_1^{∞}/E_1 measured in the experiment is smaller than it would be if the self-absorption was absent. The obtained value of N_2/N_1 is therefore an underestimate which suggests that the self-absorption effect is not able to explain the observed difference between the measured population ratios.

As for the rate equations, let us mention that following Ref. [12], Eqs. (5) are derived and strictly correct "for the case of a single-frequency field saturating a homogeneously broadened transition". However, it seems sensible that the equations are, in some degree, valid for single-mode field saturating an inhomogeneously broadened transition, in particular for sufficiently large powers of the field. This is well supported by the results in Fig. 6 where experimentally measured n_2 and n_1, expressed as a function of $z = 1/(1 + I/I_s)$, are arranged along straight lines. Only two pairs of points, those for small values of the output powers, are not on the straight lines and this means that Eqs. (5) are not valid for these output powers.

We recall that according to the literature [16, 17] the 3391 nm radiation of a He–Ne long tube is generated within a small frequency range around the centre of the inhomogeneous Doppler broadening of the transition. In such a case, intensities of the lines 543 and 342 nm of the side-light spectrum are the result of spontaneous emission from not only the atoms in resonance with the radiation field LB but also the atoms out of the resonance, but which are also inside the Doppler profile of the transition. Thus, if the spectrum of the radiation field LB does not fill the whole Doppler width of the transition, the error of n_i measured in the side light may be significant.

Also the difference between values of I_s obtained on the basis of the upper and the lower level suggests that the rate equations model is not quite adequate to the experiment.

In spite of the above objections we are convinced that the RE model may be applied to laser measurements, although only in clearly defined physical situations.

6. Conclusions

The rate equations and the transparency method are very attractive in experimental measurements of the population ratios in laser systems.

However, in view of the fact that the transparency method has been successfully verified by Inatsugu et al. [3, 4], one may conclude that the "Doppler profile" problem may be the main reason for possible error of the rate equations method.

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