PRAMANA
— journal of
physics

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Vol. 44, No. 5,
May 1995
pp. 471–479

Measurement of cross-section of excitation transfer from $3s_2$ to $4s_1$ and $5s_1$ energy levels of neon

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MS received 16 March 1994; revised 13 February 1995

Abstract. Collision induced non-radiative transitions in neon plasma have been studied using high intra-cavity radiation field of a 633 nm He–Ne laser. The transitions, induced from $3s_2$ energy level to $4s_1$ and $5s_1$ groups of energy levels, have been detected as changes in intensities of the spectral lines originating from these energy levels. From these intensity measurements, the quantities governing the transitions i.e. (i) S_3^e/S_{3RT}, the ratio of the probabilities of electronic deexcitation to the total radiative deexcitation of energy level 3 (ii) $\langle r_{23}^e v_e \rangle$, rate of excitation transfer per particle and (iii) S_{23}^e, the total probability for excitation transfer from level 2 to level 3 at a certain value of electron density have been calculated.

Keywords. Collisional cross-section; excitation transfer.

PACS No. 34·80

1. Introduction

It is known that the different constituents of a plasma viz. electrons, atoms in the ground and excited states and ions exchange their energy during collision. The excited atoms lose their energy by radiative as well as by non-radiative processes. Simultaneously, the population of excited atoms at a particular energy level is replenished by the processes of electron collision and absorption. To understand these processes and to derive the laws governing them, investigations are carried out by selectively exciting a particular energy level of a gas in a plasma using a laser radiation field.

The interaction of a strong laser radiation field with the excited atoms in a particular pair of energy levels, resonant with the incident laser frequency, leads to considerable changes in the population of atoms in these levels. These changes are communicated to other neighbouring energy levels which are connected to the particular pair of energy level by collision induced non-radiative and radiative transitions.

Useful information on interaction among these excited states has been obtained, by analysing the intensity changes in the spontaneous emission from these neighbouring energy levels as a result of interruption of laser radiation. By studying the dependence of these intensity changes on electron density and on pressure of the gas, it is possible to separate out the energy levels connected to electron-atom from those connected to atom–atom collision processes [1].

In an earlier work [2], only the rate of excitation transfer per particle, $\langle r_{23}^e v_e \rangle$, induced by electron-atom collision process from $3s_2$ level to only $4s_1'''$ and ($5s_1'''$ and $5s_1''$) levels of neon was determined.

We present here the data on (i) S_3^e/S_{3RT}, the ratio of the probabilities of electronic deexcitation to the total radiative deexcitation of energy level 3, (ii) $\langle r_{23}^e v_e \rangle$, the rate of excitation transfer per particle and (iii) S^e_{23}, the total probability for excitation transfer from level 2 to level 3 at a certain value of electron density obtained by electron-atom collision between the $3s_2$ level of neon and the close-by energy levels, $4s_1''''$, $4s_1'''$, ($5s_1''''$ and $5s_1''$) and ($5s_1'''$ and $5s_1'$) using the above technique.

We provide here the new data for the three quantities on excitation transfer from $3s_2$ to $4s_1$ and $5s_1$ groups of levels of neon and it also provides data on excitation transfer rate per particle in support of earlier work.

Experimental data on the transition between excited states are low compared to transitions from the ground state of the atoms. The great variety of practical problems calls for knowledge of a large number of quantities. Therefore maximum amount of experimental data on electron-atom collision processes is very essential.

2. Theory

Let us consider an atomic system with two quantum states represented by 2 and 1 having energies E_2 and E_1, with the frequency of radiation ν_{21} obeying the relation $E_2 - E_1 = h\nu_{21}$ as represented in figure 1(a). An incident laser radiation of frequency ν_{21} on such an atomic system will redistribute the population of atoms at the levels 2 and 1 and thus decreases the difference of the population between 2 and 1. If the incident radiation is periodically interrupted, the static population N_1 and N_2 will be modulated with amplitudes ΔN_1 and ΔN_2 with phases opposite to each other.

Atomic levels denoted by 3 and 0 close to 2 and 1 respectively as shown in figure 1(b) may also show modulation in their population as a consequence of transfer of excitation energy from levels 2 and 1 either due to non-radiative transitions involving collisions, or radiative transitions. The magnitude of these modulations at 0 and 3, in general will be determined by the efficiency of excitation transfer from levels 1 and 2 to

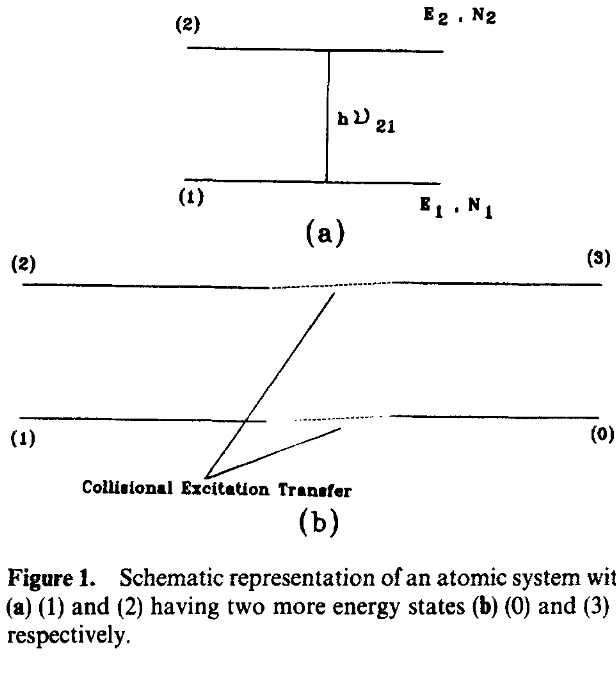

Figure 1. Schematic representation of an atomic system with two quantum states (**a**) (1) and (2) having two more energy states (**b**) (0) and (3) close to each of them respectively.

these levels. The measurements of the amplitude and phases of modulation at levels 0 and 3 provide information on the rates of collisional non-radiative transitions as well as on radiative transitions. The collisional excitation transfer may involve either atom–atom collision or electron-atom collision. Level 3 can be considered free from the cascade radiative transitions from higher energy levels, at least as a result of incident laser radiation. But level 0 will certainly be affected by the cascade radiative transitions from level 2.

Equation 1 gives the relations derived by Khaikin [2] for the excitation transfer process from levels 2 to 3 caused by electron-atom collisions only.

$$\frac{\Delta I_2}{\Delta I_3}=\frac{S_{3RT}}{\langle r_{23}^e v_e\rangle}\frac{A_2 v_2}{A_3 v_3}\frac{1}{n_e}+\frac{\langle r_3^e v_e\rangle}{\langle r_{23}^e v_e\rangle}\frac{A_2 v_2}{A_3 v_3} \tag{1}$$

where ΔI_2 and ΔI_3 are the intensity changes of spectral lines originating from levels 2 and 3 respectively, S_{3RT}, is the probability of radiative deexcitation from level 3 to other lower energy levels; $\langle r_{23}^e v_e\rangle$, is the rate of excitation transfer from levels 2 to 3, per particle induced by electron collision; v_e, is the average value of electron velocity; A_2 and v_2 and A_3 and v_3, are the Einstein coefficients for spontaneous emission from levels 2 and 3 respectively to some other lower level and the corresponding frequency of emission; n_e, is the electron density; and r_3^e, is the effective cross-section of the non-radiative deexcitation rate of levels 3 to 2 and all other levels by electron collision.

Equation (1) reduces to the form

$$y=ax+b \tag{2}$$

where

$$y=\frac{\Delta I_2}{\Delta I_3}, \tag{2a}$$

$$a=\frac{S_{3RT}}{\langle r_{23}^e v_e\rangle}\frac{A_2 v_2}{A_3 v_3}, \tag{2b}$$

$$x=1/n_e, \tag{2c}$$

$$b=\frac{\langle r_3^e v_e\rangle}{\langle r_{23}^e v_e\rangle}\frac{A_2 v_2}{A_3 v_3}. \tag{2d}$$

From (2a–2d), we see that by measuring the modulation in intensities of the spectral line originating from levels 2 and 3 at different values of electron density, we can plot a linear relation between $\Delta I_2/\Delta I_3$ and $1/n_e$. From this linear plot, we determine: S_3^e/S_{3RT}, $\langle r_{23}^e v_e\rangle$ and S_{23}^e.

S_3^e/S_{3RT}, the ratio of the probabilities of electronic deexcitation to the total radiative deexcitation of energy level 3 is obtained from (2b) and (2d) as follows:

$$\frac{S_3^e}{S_{3RT}}=\frac{\langle r_3^e v_e\rangle}{S_{3RT}}n_e=b/a\,n_e \tag{3}$$

where S_3^e is the total non-radiative rate of levels 3 to 2 and all other levels by electron collision, a is the slope of linear plot between $\Delta I_2/\Delta I_3$ and $1/n_e$ and b is the intercept on $\Delta I_2/\Delta I_3$ axis.

$\langle r^e_{23} v_e \rangle$, the rate of excitation transfer per particle is obtained by rewriting (2b) as:

$$\langle r^e_{23} v_e \rangle = \frac{A_2 v_2}{A_3 v_3} \frac{S_{3RT}}{a}, \tag{4}$$

where A_2 is the transition probability of the spectral line originating from level 2; S_{3RT}/A_3 is the ratio of the probability of radiative deexcitation from level 3 to other lower energy levels to the transition probability of the spectral line originating from level 3. These have been taken from theoretical data of Khaikin [2] and Inatsugu and Holmes [4].

S^e_{23}, the total probability for excitation transfer from levels 2 to 3 at a certain value of electron density n_e is obtained as follows:

$$S^e_{23} = \langle r^e_{23} v_e \rangle \times n_e \tag{5}$$

where $\langle r^e_{23} v_e \rangle$ is given by (4).

3. Experimental set-up

The experimental arrangement is shown in figure 2. NPL developed Brewster angle He–Ne laser, of 700 mm length and 1000 mm cavity length was used to generate high intra-cavity laser power by using both the mirrors with reflectivity of 99·8%. This intra-cavity laser radiation passes through a 100 mm long discharge plasma cell placed in a tandem with the laser plasma tube. The laser is operated by a constant current power supply and the laser output power stability is better than 0·1%. As the detecting technique uses a lock-in amplifier, it was observed that 0·1% power stability of the laser power is sufficient for the present study. There is a provision to vary the electric

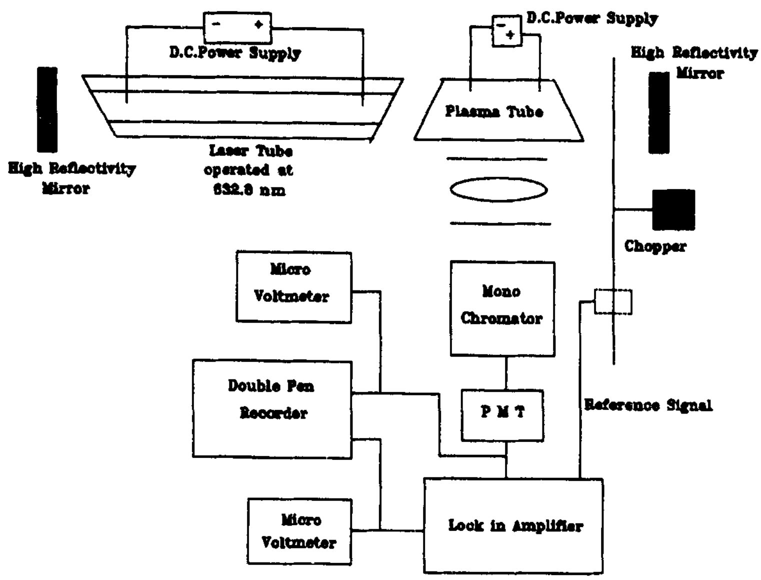

Figure 2. Schematic representation of the experimental set-up.

discharge current through the He–Ne mixture in this cell. Intensity of the spectral lines emitted through the side walls of the discharge cell have been studied using the usual combination of monochromator, a photomultiplier and a microvoltmeter. As the magnitude of the intensity changes induced by laser radiation is very small, a chopper and lock-in amplifier were also used to detect them. The chopping frequency used in this experiment was 34 Hz.

As the response of the detecting system varies with the spectral range, the whole system was calibrated using a light source whose spectral radiant emittance was calibrated against a PTB standard lamp. For determining the electron density value in a He–Ne plasma obtained in a tube diameter of 3 mm at a pressure of 1·2 torr, values obtained by earlier workers [2, 3] based on radio frequency technique were used. These values show that the electron density is linearly dependent on the range of discharge current considered (10 to 100 mA) and its value is $5 \times 10^{10}/cm^3$ at 10 mA under the above mentioned conditions.

4. Observations

Figure 3 gives some of the energy levels of neon. The 633 nm laser radiation occurs due to transitions of the atoms from $3s_2$ to $2p_4$ states. At the time the laser radiation field is interrupted by the chopper, the stimulation of the atoms in the $3s_2$ state to deexcite to $2p_4$ is stopped and therefore the population of the atoms in the $3s_2$ state increases. It has been experimentally observed that all the spectral lines originating from the $3s_2$ level have shown an increase in intensity. At the same time, the population of the atoms in

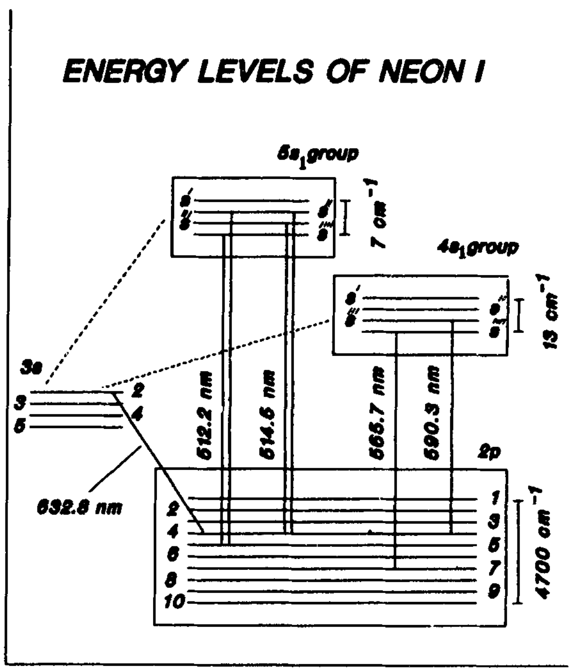

Figure 3. Schematic representation of some of the energy levels of NeI. The energy spacing is only representative and not as per actual values.

the $2p_4$ state has decreased. The percentage change in the intensity of the lines originating from these two levels is about 20. This change in $3s_2$ level population gets communicated to adjoining energy levels and for those within $3500\,cm^{-1}$ a modulation up to 5% is produced.

If the change in the intensity of a particular line originating from a particular energy level x is denoted as ΔI_x and the change in the intensity of a line from $3s_2$ level as ΔI_{3s_2}, then $\Delta I_x/\Delta I_{3s_2}$ is a measure of the magnitude of the population change that is getting communicated to the level x due to the population change in the level $3s_2$.

Out of the large number of energy levels the $4s_1$ and $5s_1$ groups were selected for detailed study. In the case of $4s_1$ group, only the levels $4s_1''''$ and $4s_1'''$ were connected to $3s_2$ by electron-atom collision process, giving rise to radiations of 565·7 nm and 590·2 nm respectively. However, it was not possible to spectrally isolate a spectral line which would represent the sub-levels of $5s_1$ group; the lines at 512·2 nm and 514·5 nm were taken to represent the two sub-levels ($5s_1''''$ and $5s_1'$) and ($5s_1'''$ and $5s_1''$) respectively. The intensity change $\Delta I_{543\cdot 4}$ of line 543·4 nm originating from $3s_2$ to $2p_{10}$ level has been taken as modulation at $3s_2$ level.

The ratio of the changes in intensity of the line 543·4 nm to the lines under study has been obtained at different values of electron density n_e. A graph between $I_{543\cdot 4}/\Delta I_x$ and $1/n_e$, where x corresponds to the spectral lines under study, gives a linear plot from which the values of quantities a and b in (3) and (4) have been obtained. Such plots obtained for the spectral lines 565·7 nm, 590·2 nm, 512·2 nm and 514·5 nm are given in figures 4a–d respectively.

5. Results

The values of S_3^e/S_{3RT}, $\langle r_{23}^e v_e\rangle$ and S_{23}^e defined in the text, obtained for the above given energy levels $4s_1''''$, $4s_1'''$, ($5s_1''''$ and $5s_1'$) and ($5s_1'''$ and $5s_1''$) of neon are given in table 1. It

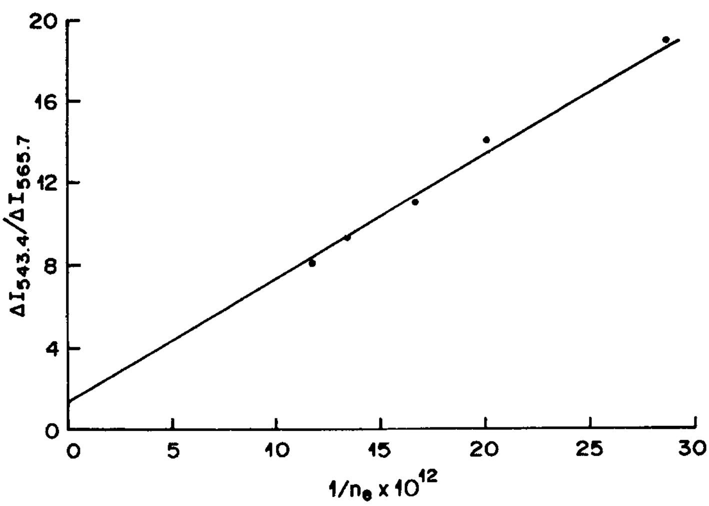

Figure 4(a). Variation of the ratio of intensity changes of two spectral lines at 543·4 and 565·7 nm, representing the two states of neon, $3s_2$ and $4s_1''''$ respectively, with respect to the reciprocal of electron density of the plasma in the cell.

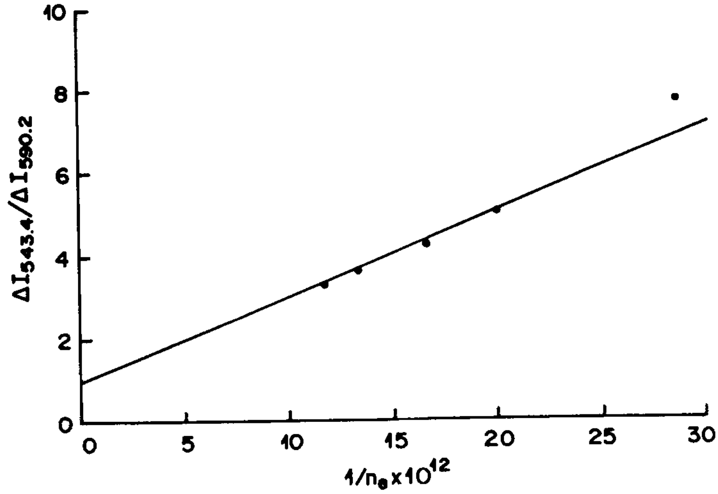

Figure 4(b). Variation of the ratio of intensity changes of two spectral lines at 543·4 and 590·2 nm, representing the two states of neon, $3s_2$ and $4s_1'''$ respectively, with respect to the reciprocal of electron density of the plasma in the cell.

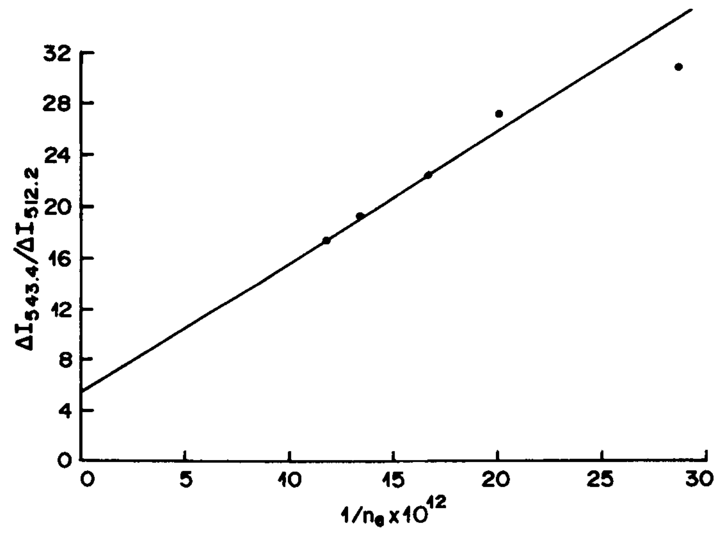

Figure 4(c). Variation of the ratio of intensity changes of two spectral lines at 543·4 and 512·2 nm, representing the two states of neon, $3s_2$ and ($5s_1''''$ and $5s_1''$) respectively, with respect to the reciprocal of electron density of the plasma in the cell.

can be noted that the values of the three quantities for $4s_1'''$ and ($5s_1''''$ and $5s_1''$) energy levels are being reported for the first time.

In table 1 the values of Khaikin [2] for one quantity $\langle r_{23}^e v_e \rangle$ for levels $4s_1'''$ and ($5s_1''''$ and $5s_1''$) are given for comparison. It can be seen that the values obtained by us agree well with this.

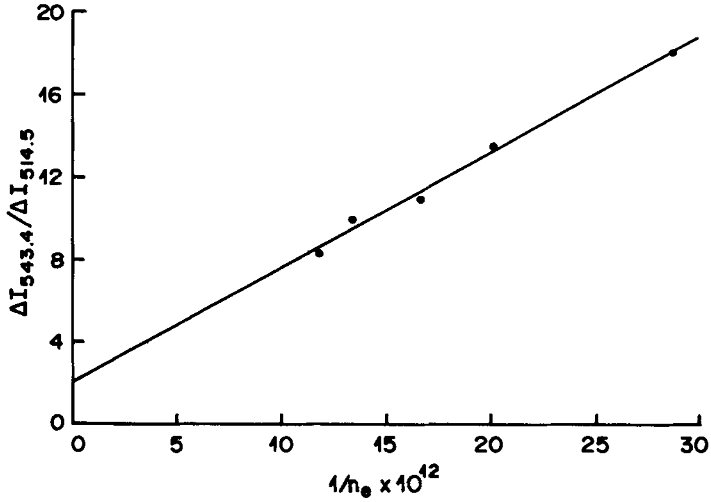

Figure 4(d). Variation of the ratio of intensity changes of two spectral lines at 543·4 and 514·5 nm, representing the two states of neon, $3s_2$ and ($5s_1'''$ and $5s_1''$) respectively, with respect to the reciprocal of electron density of the plasma in the cell.

Table 1. Values of quantities governing the laser induced transitions from intensity measurements.

Energy level (P.N.)* with spectral line (nm)	S_3^e/S_{3RT} at 10 mA	$\langle r_{23}^e v_e \rangle$ cm^3/s ($\times 10^{-8}$)	$S_{23}^e s$ at $n_e = 5 \times 10^{10}$ ($\times 10^7$)	$\langle r_{23}^e v_e \rangle$** cm^3/s ($\times 10^{-8}$)
$4s_1''''$(565·7)	0·11	1420	0·07	—
$4s_1'''$(590·2)	0·19	2150	0·11	2200
$5s_1''''$ and $5s_1''$ (512·2)	0·27	390	0·02	—
$5s_1'''$ and $5s_1''$ (514·5)	0·18	410	0·02	370

*P.N. represents Paschen Notation; **From Khaikin [2]

6. Conclusions and discussion

(1) The values of excitation transfer rate per particle $\langle r_{23}^e v_e \rangle$, obtained experimentally, is quite high ($\approx 10^{-5}$) compared to the typical values of $\langle rv \rangle$ from ground state which are of the order of 10^{-8} cm^3/s.

This is attributed to the fact that (i) the geometrical cross-section of the excited atoms in $4s_1$ and $5s_1$ groups of energy levels exceed by about 2 orders of magnitude to those of the ground state atoms and (ii) because of the small energy discrepancy between the participating levels, this process has a resonance nature and cross-section for such processes high.

(2) The probability of radiative deexcitation of the level $3s_2$ of neon is $2{\cdot}1 \times 10^7$. Now as per table 1 the probability of excitation transfer S_{23}^e from $3s_2$ to say $4s_1$ by electron

collision is $\approx 10^6$. Even if we add all the probabilities of excitation transfer from $3s_2$ level to all the neighbouring energy levels by electron collision, it may come up to 2×10^6.

It may be thus concluded that the electron impact deexcitation of $3s_2$ level at the usual operating current (10 mA) for the He–Ne laser is about an order of magnitude less than the total radiative transition from it.

(3) From the values of probabilities of deexcitation by electron impact to that by radiative process S_3^e/S_{3RT}, for energy levels $4s_1''''$ and $4s_1'''$ and for group $5s_1$, we see that the radiation transition probability exceeds by an order of magnitude to the electron collision deexcitation probability.

Thus it can be seen that, whereas, Khaikin's data on non-radiative transitions between $3s_2$ and the adjoining levels of $4s_1$ and $5s_1$ groups of energy levels is for only one quantity, i.e. $\langle r_{23}^e v_e \rangle$ and for unresolved overlap transitions, our data are for all the three quantities defined in the text belonging to resolved transitions from $4s_1''''$, $4s_1'''$, ($5s_1''''$ and $5s_1''$) and ($5s_1'''$ and $5s_1'$) groups of energy levels.

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