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INVESTIGATIONS OF ELECTRON IMPACT INDUCED EXCITATIONS FROM $3P$ TO HIGHER LYING LEVELS OF SODIUM

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Results of measurements of the rate constants for the electron impact induced transitions between $3P$ and higher excited $3D$, $4P$, and $5S$ levels are presented. The experimental data were corrected for the radiation imprisonment using a new model of this effect.

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1. Introduction

Electron impact induced transitions between excited atomic levels play an important role in many phenomena, especially in astrophysics, plasma physics, and spectroscopy. Nevertheless, only few experimental studies on these topics have been performed [1–4] and the data for these kind of collisions are available primarily from theoretical approaches [5–10]. A review of these results was given by Andersen et al. [11].

In our two recent papers [12, 13] a new method of investigation of electron impact induced transitions between excited atomic levels was presented. Briefly, the experiment was performed in a cell of diameter $2a = 32$ mm, containing a mixture of caesium and sodium vapour (Fig. 1). The sodium atoms whose concentration was of the order of 10^{11} cm^{-3} [14] were the subject of investigations. The caesium vapour with density of about three order of magnitude higher served as an electron source.

The electrons were produced using a method discovered by McIlrath and Lucatorto [15, 16] and then developed by several laboratories [17–20]. The method consists in the strong excitation of gaseous medium by laser pulse. The energy stored in excited atoms is transferred to the plasma mainly by superelastic collisions between electrons and the laser excited atoms. In our experiment the vapour was ionized using dye laser pulses tuned to 455 nm caesium resonance line. The

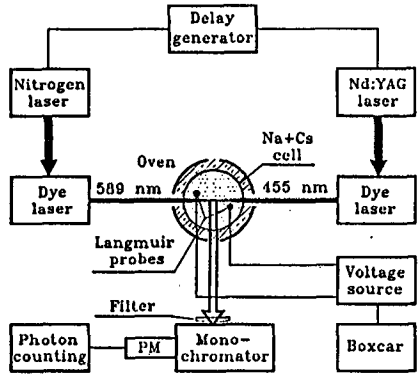

Fig. 1. Experimental setup.

pulse FWHM time was about 3.5 ns and its energy reached 3.5 mJ. Under such conditions the electron concentration of the order of 10^{12} cm^{-3} was achieved. The electron energy and temperature were determined by means of the double Langmuir probes method [21].

Pulses of a second dye laser, tuned to the D_2 sodium line ($\lambda = 589$ nm), were used to produce the Na($3P$) atoms. In order to prepare the electron gas with a Maxwellian energy distribution [22] these pulses were delayed by $t_D = 6$ μs with respect to the ionizing pulses, using a delay generator. Dye laser beams entered the cell collinearly. The diameter of each beam was about 8 mm. The energy of 589 nm pulse was about 0.3 mJ and its FWHM time was about 7 ns.

Due to the collisions between Na($3P$) atoms and electrons higher lying states nl were populated. The rate constants for $3P$–nl electron impact induced excitations were determined by measuring a fluorescence signal on the atomic lines originating from these nl levels. The respective sodium lines were selected using a set of interference and colour glass filters, as well as a prismatic monochromator. The fluorescence signal appearing per one laser pulse was integrated by a gated photon counting system (SRS 400).

2. The evolution of plasma

The mechanism of the alkali vapour ionization by nanosecond resonant laser pulses was previously investigated [23, 24]. It was estimated that this process takes place within several hundreds of nanoseconds. Since the ionization efficiency is approximately proportional to the third power of the excited Cs atoms concentration, the plasma production occurs mainly in the region of the vapour illuminated by the 455 nm laser pulse. Therefore, for the initial distribution of electrons we assume a step function as in Fig. 2.

The further expansion of plasma is caused by the ambipolar diffusion [25]. The evolution of the electron concentration is described by equation

$$\frac{\delta e(r,t)}{\delta t} = D_A \nabla^2 e(r,t), \tag{1}$$

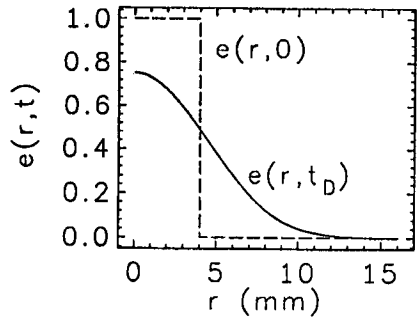

Fig. 2. Distribution of electrons in sodium cell for $t_D = 6$ μs. The dashed line shows initial distribution of electrons produced by laser pulse.

where D_A denotes the ambipolar diffusion coefficient. The dimensionless function $e(r,t)$ describes the distribution of the electron concentration $N_e(r,t) = N_{e\,max}e(r,t)$, where $N_{e\,max}$ is the initial electron concentration at the centre of the cell. After sufficiently long time, the $e(r,t)$ time dependent part can be expressed by a single exponential function, characterized by a decay constant τ. Using the Langmuir probes, it was found that $\tau \approx 100$ μs. This means that the diffusion is very slow in comparison with the submicrosecond process of the plasma production. Since τ is related to the D_A [25], by solving Eq. (1) one can find electron distribution for the delay time $t_D = 6$ μs, i.e. for the time when the Na($3P$) atoms are produced by the 589 nm pulse and the collisional experiment starts. The evaluated $e(r,t_D)$ function is shown in Fig. 2.

3. The radiation imprisonment

Production of the Na(nl) atoms depends on the electron and Na($3P$) atoms distribution. Since the measurements were performed in the optically dense vapours, in order to find the Na($3P$) atoms distribution, a diffusion of the resonance radiation must be taken into account.

In order to analyze the radiation trapping the Compton–Milne [26, 27] or the Holstein–Biberman [28–30] theories are usually used. However, both models describing the radiation imprisonment in weakly excited media assume constant absorption coefficient. This is only roughly satisfactory for the case when high intensity resonance light is used. A saturation of the optical transition, which occurs for such circumstances, leads to substantial reduction of the absorption coefficient, the phenomenon known as the optical bleaching. As we stated elsewhere [31], due to this effect the decay of the excited atoms is faster for the strong excitation than for a weak one. In this experiment resonant radiation power density was about 80 kW/cm^2 with the spectral linewidth about 0.01 nm, then the $3S$–$3P$ transition was fully saturated within the whole laser pulse [32, 33], i.e. for the time interval of 14 ns.

In order to describe the radiation trapping for such conditions we use a modified Holstein–Biberman theory. This approach is based on the integro-differential

rate equation of the balance between spontaneous depopulation and radiative excitation. For the $3S$–$3P$ transition in sodium

$$\frac{\delta N_{3P}(r,t)}{\delta t} = -A_{3P}N_{3P}(r,t) + A_{3P}\int G(r,r',t)N_{3P}(r',t)\mathrm{d}^3r', \tag{2}$$

where $N_{3P}(r,t)$ is the excited atoms concentration and A_{3P} — the rate of the spontaneous emission. We assume that during the 589 nm pulse, in the region of the cell which is irradiated by the light beam, $N_{3P} = Ng_{3P}/(g_{3S}+g_{3P})$, where N denotes the vapour number density and g_{3S}, g_{3P} — the statistic weights of ground and excited states, respectively.

In Eq. (2) the integration is taken over all the vapour volume. $G(r,r',t)$ denotes the probability that the photon emitted from a volume element around a point r' will be reabsorbed in r:

$$G(r,r',t) = -\frac{1}{4\pi\rho^2}\left.\frac{\delta T(\rho,t)}{\delta\rho}\right|_r, \tag{3}$$

where $\rho = |r-r'|$. The integral transmission coefficient $T(\rho,t)$ was found from

$$T(\rho,t) = \int P(\nu)T'(\rho,\nu,t)\mathrm{d}\nu, \tag{4}$$

where $P(\nu)$ is the normalized line shape. The frequency dependent transmission coefficient $T'(\rho,\nu,t)$ was obtained by solving the equation

$$\frac{\delta T'(\rho',\nu,t)}{\delta\rho'} = -T'(\rho',\nu,t)k(r'',\nu,t), \tag{5}$$

where $k(r'',\nu,t)$ is the absorption coefficient, r'' denotes a point on the photon trajectory and $\rho' = |r'-r''|$. We assume straight line trajectories of light, neglecting the effects caused by gradients of the refractive index.

For a low density sodium vapour the atomic line shape is well determined by the Doppler–Gauss profile [34]. Then the emission line is described by

$$P(\nu) = \frac{\lambda_0}{\sqrt{\pi}v_0}\exp\left[-\frac{(\nu-\nu_0)^2}{\nu_0^2}\frac{c^2}{v_0^2}\right] \tag{6}$$

and the absorption coefficient

$$k(r,\nu,t) = \frac{\lambda_0^3 A_{3P}}{8\pi^{3/2}v_0} \times\left[\frac{g_{3P}}{g_{3S}}N - \frac{g_{3P}+g_{3S}}{g_{3S}}N_{3P}(r,t)\right]\exp\left[-\frac{(\nu-\nu_0)^2}{\nu_0^2}\frac{c^2}{v_0^2}\right], \tag{7}$$

where ν_0 is the frequency at the centre of the line, λ_0 — the respective wavelength (589 nm), c — the light velocity and $v_0 = \sqrt{2k_BT_g/M}$ denotes the mean atomic velocity (k_B is the Boltzmann constant, T_g — the absolute vapour temperature and M — the atomic mass).

The problem was solved numerically by means of SGI IRIS 4D/35 computer. Results of calculations, i.e. the normalized distribution of the Na($3P$) atoms denoted as $d(r,t) = N_{3P}(r,t)(g_{3P}+g_{3S})/Ng_{3P}$ for $N = 5\times10^{11}$ cm^{-3} is presented in Fig. 3. A flat part of this function corresponds to the optical saturation which takes place in the region irradiated by 589 nm laser pulse. Due to the diffusion of

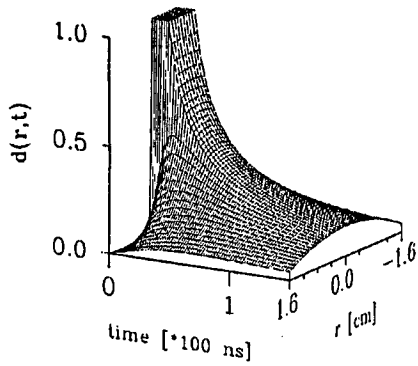

Fig. 3. Evolution of Na($3P$) atoms distribution.

radiation the vapour is excited also in the region which was not illuminated. As it was shown in the previous paper [31], after sufficiently long time, excitation decay can be described by a single exponential function characterized by a time constant τ^*_{3P}. This parameter is known as the radiation imprisonment time [28, 29]. In our case the value of τ^*_{3P} even as high as 200 ns was measured, while the natural lifetime of the sodium $3P$ state is only 16.7 ns.

4. The experimental signal

Collisions between electrons and the Na($3P$) atoms lead to the excitation of higher lying nl levels. Evolution of the Na(nl) atoms concentration is described by a following rate equation:

$$\frac{\mathrm{d}N_{nl}(r,t)}{\mathrm{d}t} = -A_{nl}N_{nl}(r,t) + k_{3P-nl}(T_\mathrm{e})N_\mathrm{e}(r,t_\mathrm{D})N_{3P}(r,t), \tag{8}$$

where A_{nl} denotes the rate for the nl-level depopulation due to the spontaneous emission and $k_{3P-nl}(T_\mathrm{e})$ — the quantity which ought to be evaluated, i.e the efficiency of the electron impact induced $3P$–nl excitations (T_e is the electron temperature).

It was assumed that the changes of the electron temperature, as well as the changes of the electron distribution are very slow with respect to the rates of the atomic transitions, then the values of T_e and $N_\mathrm{e}(r,t_\mathrm{D})$ do not depend on time. We neglect collisional population of the nl levels from other states than $3P$, because the efficiencies of these transitions are more than one order of magnitude smaller than the $k_{3P-nl}(T_\mathrm{e})$ value [13]. The collisional depopulation of nl levels is also neglected.

In such conditions the solution of Eq. (8) is expressed by

$$N_{nl}(r,t) = k_{3P-nl}(T_\mathrm{e})\frac{g_{3P}}{g_{3P}+g_{3S}}NN_{\mathrm{e}\max}e(r,t_\mathrm{D})\mathrm{e}^{-A_{nl}t}\int_0^t d(r,t')\mathrm{e}^{A_{nl}t'}\mathrm{d}t'. \tag{9}$$

The total number of photons n_f registered by the optical system on the wavelength λ corresponding to the transition between nl and lower lying $n'l'$ levels is given by

$$n_f = \Phi_\lambda A_{nl-n'l'} \int_0^\infty dt \int_V d^3V N_{nl}(r,t), \tag{10}$$

where Φ_λ denotes the quantum efficiency of the optical system, $A_{nl-n'l'}$ — the spontaneous emission rate for the transition of interest, and V — the volume of the cell. In the experiment the integration of photons was performed over the finite time interval $T = 1$ μs. As it was shown earlier [13], for $T > 4\tau^*_{3P}$ the error caused by finite integration time is negligible.

In Eq. (10), A_{nl}, $A_{nl-n'l'}$, g_{3S}, g_{3P} are the atomic constants [35, 36]. Values of the optical efficiency Φ_λ and the electron temperature T_e were measured in the experiment. However, the determination of the electron $N_{e\,max}$ and atomic concentrations N can be affected by a large error. Therefore, these quantities were eliminated from the final calculations by means of the reference signal n_{589}, i.e. the number of 589 nm photons appearing per one ionizing pulse due to the electron impact $3P$–$3S$ excitations in sodium

$$n_{589} = \Phi_{589} T k_{3S-3P}(T_e) N N_{e\,max} \int_V d^3V e(r, t_D), \tag{11}$$

where Φ_{589} denotes the quantum efficiency of the optical system for the sodium D lines. The rate constants $k_{3S-3P}(T_e)$ for the collisional population of Na($3P$) atoms were calculated using the experimental data obtained by Enemark and Gallagher [37]. For the reference signal measurements only the dye laser producing the plasma was switched on. The photons appearing on 589 nm line were integrated during the same time $T = 1$ μs as in the case of the n_f measurements.

From Eqs. (10) and (11) one can find the $k_{3S-nl}(T_e)$ constant

$$k_{3P-nl}(T_e) = k_{3S-3P}(T_e) \frac{T}{\tau^*_{3P}} \frac{g_{3S} + g_{3P}}{g_{3P}} \frac{\Phi_{589} n_\lambda}{\Phi_\lambda n_{589}} \frac{A_{nl}}{A_{nl-n'l'}} X, \tag{12}$$

where the coefficient X describing the influence of the radiation and electron diffusion processes is expressed by

$$X = \int_V d^3V e(r, t_D) \left[\frac{A_{nl}}{\tau^*_{3P}} \int d^3V e(r, t_D) \int_0^\infty dt\, e^{-A_{nl}t} \times \int_0^t d(r, t') e^{A_{nl}t'} dt' \right]^{-1}. \tag{13}$$

In our previous papers [12, 13], where the model of the weak excitation for the radiation diffusion was assumed, the value of this parameter $X = 1.37$ was used. In the analysis presented here, when the optical bleaching as well as the plasma diffusion are taken into account, X was estimated to be equal to 2.14.

The stability of the X parameter was checked for the vapour densities corresponding to the temperature range used in our experiment, i.e. between 130 and 180°C. It was found that the X does not vary more than about 5%. X is also not sensitive to the changes of A_{nl}, t_D and the ambipolar diffusion coefficient.

5. Results and discussion

Figure 4a shows the dependence of the rate constant for $3P$–$4P$ impact induced excitation on the electron temperature. The Na($4P$) atoms were detected by registration of the fluorescence on 330 nm line corresponding to the $4P$–$3S$ transition (Fig. 5). These data were presented also previously [12, 13], but without

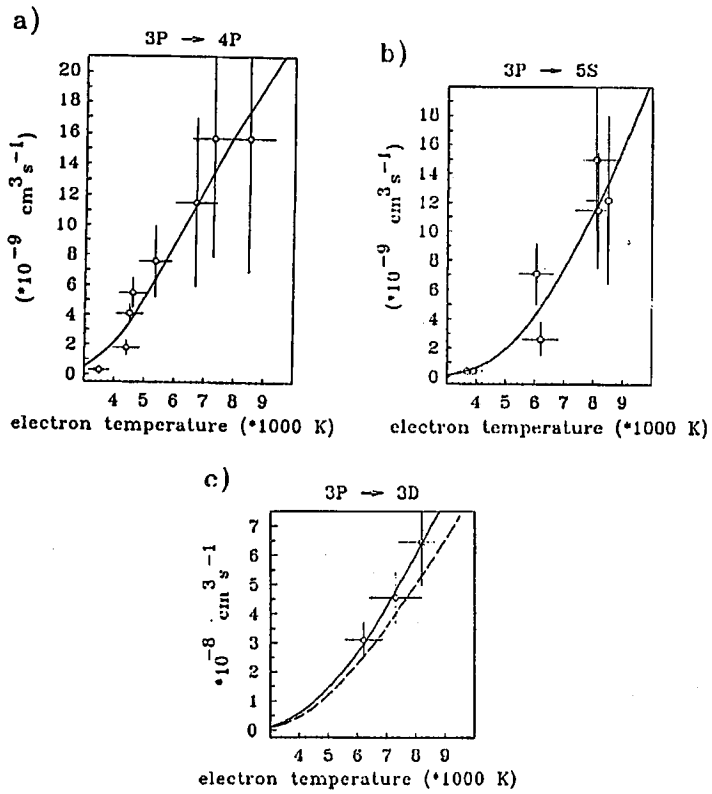

Fig. 4. Rate constants for electron impact induced $3P$–nl transitions in sodium as a function of electron temperature. The solid lines represent efficiencies calculated using Gryziński's approximation corrected by the factor of 1.5 (a) and 1.3 (b). The dashed line in part (c) presents results of Stumpf and Gallagher [38].

proper correction for the radiation and electron diffusion.

The rate constant for $3P$–$5S$ excitation is shown in Fig. 4b. For these measurements the fluorescence on the line 615 nm corresponding to $5S$–$3P$ optical transition was registered.

The efficiency of the collisionally induced transitions between $3P$ and $3D$ levels was measured on 819 nm sodium line (n_{S19}). However, k_{3P-3D} was determined in a slightly different way than in the above two cases, because as a reference signal, instead of 589 nm fluorescence, a number of photons n_R appearing on line

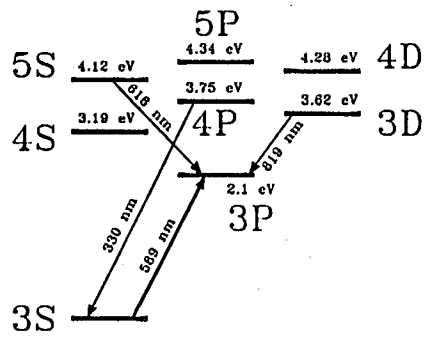

Fig. 5. Simplified diagram of sodium levels with observed transitions.

819 nm due to $3S$–$3D$ electron impact excitation was used. Similarly as in other cases, for the reference signal measurements the 589 nm dye laser was switched off. Since both signals were observed on the same 819 nm line, for this case the quantum efficiencies of the optical system were eliminated from the final equation for the rate constant

$$k_{3P-3D}(T_e) = k_{3S-3D}(T_e)\frac{T}{\tau^*_{3P}}\frac{g_{3S}+g_{3P}}{g_{3P}}\frac{n_{819}}{n_R}X. \tag{14}$$

The rate constant $k_{3S-3D}(T_e)$ for the reference transition was calculated using the cross-section given by Stumpf and Gallagher [38]. The excitation of 3D level from the ground state does not influence the n_{819} signal, because for $T_e < 9000$ K the k_{3S-3D} constant is smaller than $0.02k_{3P-3D}$.

The experimental error was analyzed in detail previously [13]. It was found that for electron temperatures lower than 4000 K the error is not greater than 20%. Such an error follows from the application of the experimental data to Eqs. (12) or (14) respectively. For higher temperatures the fast increase in the plasma concentration is observed and the population of the nl level can be significantly modified due to secondary inelastic collisions of the Na(nl) atoms with electrons. Cascade transitions to nl level from higher excited states also increase the uncertainty of the k_{3P-nl} constant determination. In order to evaluate the error caused by these effects we used the Gryziński approximation for the respective cross-sections [5, 6], because no appropriate experimental data is available.

The effects mentioned above can significantly magnify the uncertainty of determination of the k_{3P-4P} rate constant. The $4P$ level can be efficiently populated through the cascade transitions from higher states, mainly from $5S$ and $4D$, because the oscillator strengths for the transitions from these levels to the $4P$ one are rather large [36] and for high electron temperatures the rates for the impact excitation of the $5S$ and $4D$ states are comparable with the k_{3P-4P}. Also depopulation of the $4P$ level due to secondary collisions with electrons is characterized by a large rate, especially for the $4P$–$3D$ transition, because the $4P$ level lies only 0.136 eV above the $3D$ one. This magnifies the standard deviation of k_{3P-4P} to about 70% at 9000 K.

As far as the k_{3P-3D} rate is concerned, the error caused by the spontaneous emission from higher lying states to $3D$ one is negligible. For electron temperatures below 10^4 K, among P levels only the $4P$ one is efficiently populated, but it is coupled with $3D$ via transition characterized by a very small oscillator strength. Also the secondary $4P$–$3D$ inelastic collisions do not change the $3D$ level population stronger than of about 10%, even at the highest electron densities (about 2×10^{12} cm^{-3}, as it was observed for 8500 K). The depopulation of this level due to collisions of the second kind does not exceed 1%. Finally, we evaluate that for the electron temperature of 9000 K the experimental error of the k_{3P-3D} constant rises to about 30%.

The superelastic collisions can significantly reduce the population of the $5S$ level — even up to 40% for the electron concentration of 2×10^{12} cm^{-3}. The collisionally induced and the radiative transitions from higher excited states (mainly from the $5P$ state) can populate this level up to 30%, then we evaluate that for the temperature of 9000 K the standard deviation of the k_{3P-5S} constant can reach even 50%.

The standard deviation of the electron temperature determination is about 10%.

In our experiments inelastic collisions between electrons and atoms take place at very low electron energies, practically only several eV above the threshold. For such low energies the best evaluation of the cross-sections is achieved using close coupling approximation [39]. However, in many experimental works simpler formulas are used. These approaches, based on the Born approximation (Johnson and Hinnov [40], Vajnstein et al. [41]) or the classical approximation (Gryziński [5, 6]), agree with the experimental results usually within the factor of 2. Since for the formulas given in Refs. [40, 41] one needs to fit several parameters, we compared our results with the rate constants calculated using simpler expressions of Gryziński. These approaches are presented in Fig. 4a–c by solid lines. In order to achieve the best fit to the experimental data, the Gryziński cross-sections were multiplied by the factor of 1.5 for the $3P$–$4P$ transition and by the factor of 1.3 for the $3P$–$5S$ transition. For the k_{3P-3D} constant quite good agreement of the experimental results with the Gryziński model without any correction was achieved.

6. Conclusion

Using a new method, we measured the rate constants for the electron impact induced transitions from $3P$ to $3D$, $4P$ and $5S$ levels of sodium. The rates for $3P \rightarrow 4P$ and $3P \rightarrow 5S$ excitations were determined for the first time. The results for $3P \rightarrow 3D$ transitions can be compared with the rate constants calculated using the cross-sections measured by Stumpf and Gallagher [38] (in Fig. 4c presented by a dashed line). A discrepancy between these results is smaller than the standard deviation.

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