FLUORESCENCE OF SODIUM VAPOUR EXCITED BY 330 nm LASER PULSES

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We report results of investigation on the fluorescence induced in sodium vapour by pulses of laser light tuned near the $3S \rightarrow 4P$ transition (330 nm). The fluorescence was observed in perpendicular and parallel directions in respect to the laser beam. A depletion of fluorescence intensity corresponding to the $5D \rightarrow 3P$ transition (498 nm) was observed along the laser beam. We explain this effect as a result of reabsorption that is caused by intense population of the $3P$ level due to cascade stimulated processes. The most intense depletion occurs about 20 ns after the laser pulse and it corresponds to the moment of maximal population of the $3P$ level.

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1. Introduction

When resonant laser pulses tuned to higher lying levels irradiate atomic vapour, a broad spectrum of fluorescence occurs. It is a well-known optical phenomenon that the fluorescence spectrum contains lines starting either from the levels, which are directly excited by the laser light, or from the lower lying levels which are populated due to cascade spontaneous transitions. The shapes of the fluorescence pulses as well as their intensities at wavelengths of interest can be easily found by solving the rate equations involving the respective Einstein coefficients [1]. This model concerns however only the low atomic concentrations. At the number densities higher than 10^{11} cm^{-3}, population of the levels can be changed significantly due to various processes. One of them, which can occur in such circumstances [1–6], is a contribution of stimulated transitions. The other important factor is the influence of electrons which usually are produced in dense

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vapours [7–10]. The atomic collisions as well as the radiative and impact processes leading to the dimers dissociation and the dimers fluorescence becomes important [11, 12]. Finally the radiation reabsorption can modify the fluorescence characteristics [13–17]. In this paper we report investigations on the fluorescence that occurs in dense sodium vapour under $3S \rightarrow 4P$ excitation.

2. Experimental setup

The experimental setup (Fig. 1) was already presented in details in our previous papers [1, 3]. Briefly, the sodium vapour was stored in a cross-heatpipe which temperature was regulated by a PID controller in the range of 300–400°C. It corresponds to the sodium vapour concentration of 10^{14}–5×10^{15} cm^{-3} [18]. About 10 Torr of helium as a buffer gas was added. A length of the vapour column was equal to 10 cm. The exciting 330 nm light pulses were generated by frequency doubling of pulses from a dye laser working on DCM. The ultraviolet pulses reached energies of about 0.4 mJ, their spectral linewidth was about 0.01 nm, while their temporal FWHM was about 7 ns. The laser beam was directed into the cell by means of a dielectric mirror. The mirror was fully reflecting at 330 nm, but it transmitted well the light in the visible range. The fluorescence of the excited sodium vapour was observed both in parallel and perpendicular directions in respect to the laser beam. The light emitted perpendicularly was focused on a slit of

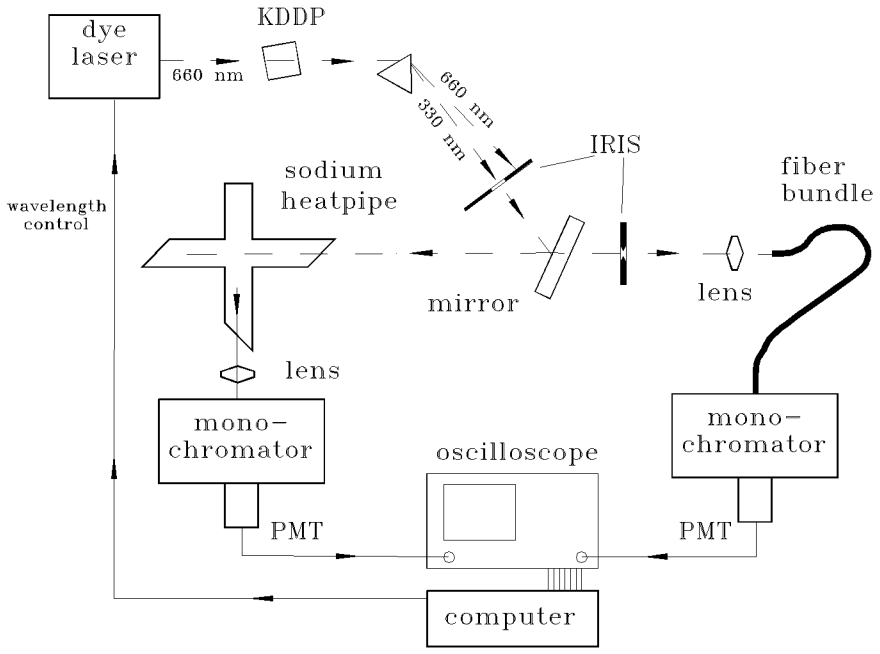

Fig. 1. Experimental setup.

a monochromator (SPM-2). Longitudinal observation of the fluorescence was done through the mirror in the backward direction. In order to register only the light emitted from the laser irradiated region and along the laser beam the observation angle was reduced by means of an iris. The light was coupled to the slit of the second monochromator (GDM 1000) by means of a fiber bundle. The fluorescence was detected by a digital oscilloscope (HP 54540) connected to a computer. The temporal resolution of the system reached about 2 ns.

3. Perpendicularly observed fluorescence

Grotrian diagram of sodium with transitions of interest is shown in Fig. 2 while Fig. 3 presents normalized signals of the fluorescence registered perpendicularly in respect to the laser beam at different wavelengths. Besides the signal at

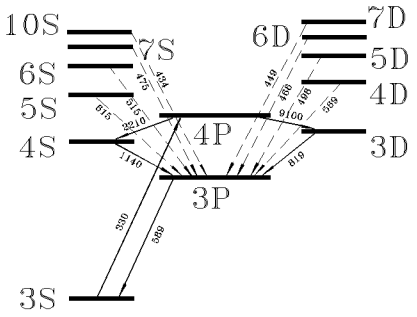

Fig. 2. Simplified Grotrian diagram of sodium. Continuous lines show either the laser excitation or the stimulated transitions, while the dashed lines present the fluorescence transitions.

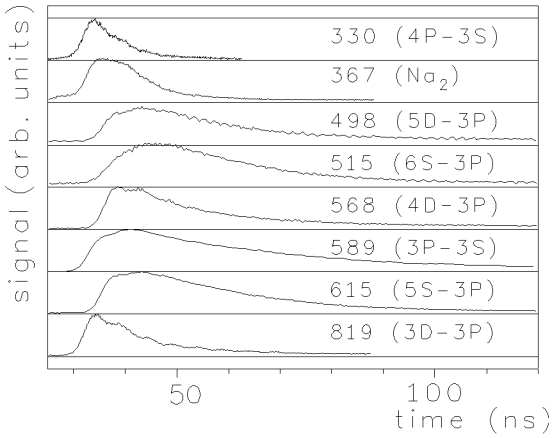

Fig. 3. Fluorescence pulses registered perpendicularly to the laser beam for the laser wavelength of $\lambda_{EX} = 330.4$ nm.

330 nm ($4P \rightarrow 3S$) originating from the level that was directly excited by the laser light, one can select the signals at 819 and 589 nm starting from the levels involved in the cascade stimulated process ($3D \rightarrow 3P$ and $3P \rightarrow 3S$, respectively) [1–3]. There are also the signals corresponding to transitions from the levels lying higher than the $4P$ level. One observed the fluorescence of Na_2 molecules (367 nm) as well.

The signal at 330 nm consisted mainly of the laser radiation scattered in the sodium vapour on atoms, molecules, and clusters [19, 20]. Similarly the signal at 819 nm contained a strong contribution of light generated in the vapour by stimulated emission between $3D \rightarrow 3P$ levels. Since both the laser pulse (330 nm) and the stimulated emission pulse (819 nm) lasted only several nanoseconds [1, 3], the observed signals reached their maximum significantly faster than the signals at other wavelengths. However the decay time of the 330 and 819 nm signals was much longer than the duration time of the laser pulse, which suggests a contribution of the spontaneous emission as well. The emission at 589 nm was excited by the stimulated cascade population of $3P$ level. In this signal there was also a contribution of the light generated on this line due to stimulated emission. However the 589 nm signal changed much slower than the signals at 330 and 819 nm. It was caused by the imprisonment of the resonance radiation in the dense sodium vapour [14–16].

Pulses of the fluorescence at 498 ($5D \rightarrow 3P$), 515 ($6S \rightarrow 3P$), 569 ($4D \rightarrow 3P$), and 615 nm ($5S \rightarrow 3P$) nm originate from the atomic levels lying higher than the $4P$ level. These levels were populated due to electron impact. The most important were collisions of electrons with Na($4P$) atoms because of the reduced energy threshold for excitation of highly lying states. The risetimes of these signals were of about 10^{-8} s. As it was stated in our previous paper [1] within this time a high density of Na($4P$) atoms was also present in the cell. Maxima of the signals at 498, 515, 569, and 615 nm were reached several nanoseconds later than the maxima corresponding to the pulses at 330 or 819 nm. The decay times of the

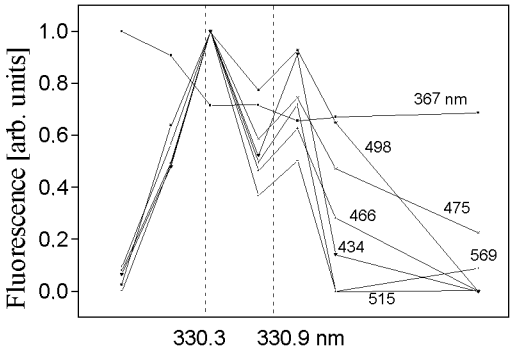

Fig. 4. Excitation spectra of the fluorescence at respective wavelengths. Dashed lines correspond to the $3S \rightarrow 4P$ doublet.

fluorescence pulses at these wavelengths were shorter than the natural lifetimes of the upper levels of the respective transitions (τ_{5S} = 75 ns, τ_{6S} = 150 ns, τ_{4D} = 54 ns, τ_{5D} = 110 ns). This shortening was mainly caused by the electron impact deexcitations [10]. For this reason the pulses of fluorescence from highly excited levels are approximately similar to each other. Besides the lines mentioned above the fluorescence at 434 nm ($10S \rightarrow 3P$), 449 nm ($7D \rightarrow 3P$), 466 nm ($6D \rightarrow 3P$), and 475 nm ($7S \rightarrow 3P$) was also observed.

The 367 nm fluorescence pulse corresponds to the transition between $C^1\Pi_u \rightarrow X^1\Sigma_g^+$ levels of the sodium molecule. A fast intensity rise of this pulse suggests the direct excitation of Na_2 by the laser light at 330 nm rather than the impact mechanism. This is also confirmed by the excitation spectra (Fig. 4). The atomic fluorescence occurred when the laser is tuned near the $3S \rightarrow 4P$ resonance lines, while the molecular fluorescence does not manifest such behavior.

4. Longitudinally observed fluorescence

Registration of the fluorescence on wavelengths corresponding to cascades from the $4P$ level could not be done along the laser beam because at these lines the signals are strongly affected by the stimulated emission. For this reason the measurements were performed only for transitions starting from the levels lying higher than the $4P$ level. However for the parallel observation in respect to the laser beam a new phenomenon occurred in comparison with the perpendicular observation. As it is seen in Fig. 5 on the rising slope of the fluorescence pulse at the 498 nm ($5D \rightarrow 3P$) after an initial increase in the signal a depletion of the intensity appears. Its duration time was of about 20 ns. Such decrease in the fluorescence was not observed perpendicularly to the laser beam. This effect occurred only when the laser was tuned to one of the components of the resonance

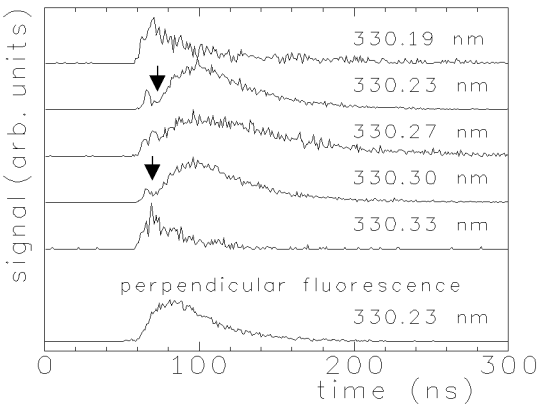

Fig. 5. Pulses of the fluorescence at 498 nm observed parallelly in respect to the laser beam at different wavelengths of excitation. Arrows show the depletion on the rising slope of the pulses.

doublet: $3S_{1/2} \rightarrow 4P_{3/2}$ ($\lambda_{EX} = 330.23$ nm) or $3S_{1/2} \rightarrow 4P_{1/2}$ ($\lambda_{EX} = 330.30$ nm). This is clearly seen when the laser line is localized either between the resonance lines $\lambda_{EX} = 330.27$ nm or at wings of the $3S \rightarrow 4P$ transition (330.19, 330.33 nm). At wings of the excitation line the plasma generation was less efficient and one also observed a significant shortening of the fluorescence pulse.

We explain the depletion of the fluorescence by reabsorption of the spectral line. This effect was caused by the efficient population of the Na($3P$) atoms due to cascade stimulated transitions from the laser pumped $4P$ level via $4S$ or $3D$ levels (see Fig. 2). The production of the Na($3P$) atoms was the most efficient when the laser was tuned to the $3S \rightarrow 4P$ transition. The high density of Na($3P$) atoms caused the reabsorption of the 498 nm light which leaded to the decrease in the fluorescence signal. The duration time of the depletion coincides well with the presence of the high density of the Na($3P$) atoms in the cell. The minimum of the fluorescence corresponds well to the maximum of the Na($3P$) number density that was found in our previous experiment using the pump-probe laser methods [1].

During the depletion the resonant light flux emitted from the excited volume was diminished in the optically thick medium. The integral brightness of the light flux K', leaving the source, whose length is equal to l, is described by the brightness K_1 of the primary source of radiation (that is not affected by the reabsorption). It is related to the unity of length of the lighting column by means of the formula [15]

$$K' = K_1 l S(\chi_0 l),$$

where

$$S(\chi_0 l) = \int [1 - \exp(-\chi(\nu) l)] \mathrm{d}\nu$$

is a function describing the absorption line shape $\chi(\nu)$, while $\chi_0 \propto f_{ik} \lambda_{ki}^2 N_i / \Delta\lambda$ is the absorption coefficient at the line centre, f_{ik} is the oscillator strength, λ denotes the light wavelength, and $\Delta\lambda$ — the line width. The N_i describes the lower level population. When the lower level number density N_i increases then the optical thickness of the medium $\chi_0 l$ increases as well and the line brightness K' decreases.

It should be pointed out however that the fluorescence depletion could be registered only when the geometry of observation is carefully reduced to the volume that is strongly excited by the laser light. Only in such a case the 498 nm radiation passes through the vapour region where due to the cascade population of the $3P$ level for several nanoseconds the high optical thickness is produced. At any other angle of observation (as for the perpendicular direction) the effect vanishes, because the 498 nm light crosses the vapour region where the number density of Na($3P$) atoms is relatively low, so the optical thickness is negligible.

We suppose that in principle a similar effect could be also observed for the fluorescence signals at other lines corresponding to transitions from the higher excited levels to the $3P$ state. However we have not registered it for any other sodium line. The depletion can be observed only when the rising slope of the fluorescence pulse is well registered. For observations at 615 or 569 nm we were not able to eliminate a strong radiation emitted at 589 nm. The lines leading from the levels lying higher than $5D$ level were so weak that during initial several tens of nanoseconds, when the depletion occurs, the longitudinal fluorescence was not distinguishable from the noise level.

5. Conclusion

The excitation of dense atomic vapour by intense laser light tuned to an excited level may lead to fluorescence at various atomic lines. In sodium excited to the $4P$ level we registered the fluorescence for several transitions both perpendicularly and longitudinally in respect to the laser beam. For the longitudinal observation of the fluorescence pulse at 498 nm ($5D$–$3P$) one observes a depletion which occurs at the moment of the maximum population on the Na($3P$) level. The depletion is caused by reabsorption of the radiation at this wavelength.

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