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Generation of Radiation on Yellow and Infrared Lines in Sodium Vapour Excited to the 4*P* Level

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Abstract. Generation of the radiation pulses on resonant (589 nm) as well as infrared (2210 nm and 1140 nm) lines was observed in sodium vapour excited by laser light tuned to the 3*S*–4*P* transition.

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1 Introduction

The use of alkali metal vapour as a lasing medium is of broad interest. Already in the fundamental work of Schawlow and Townes [1] a possibility of generation on cascade transitions between atomic states in potassium vapour excited to the 5*P* level was considered. During the next years lasing was attained in almost all alkali vapours. As far as sodium is concerned the infrared laser emission from two-photon excited atoms was observed by Gross et al. [2] and by Müller and Hertel [3]. Collision assisted generation from sodium atoms was investigated by Müller et al. [4] and Krökel et al. [5]. Recently, other emission processes such as wave-mixing [6–10] or hyper Raman mechanisms [11] were observed. In the present paper we report results of an experiment where the generation of the radiation on resonant ($\lambda = 589$ nm) as well as on infrared ($\lambda = 2210$ nm and 1140 nm) lines in sodium vapour resembling the scheme of Schawlow and Townes [1] was observed.

2 Experiment

The experimental setup is presented in Fig. 1. A pyrex cell of 3 cm length (along the laser beam) containing a drop of sodium was placed in an oven the temperature of which was electronically controlled with a precision of 1° C and varied between 270–350° C. The sodium vapour number densities were determined using Nesmeyanov formula [12]. In order to protect the cell walls against the corrosive alkali vapour influence a borax layer was used [13]. No buffer gas was applied.

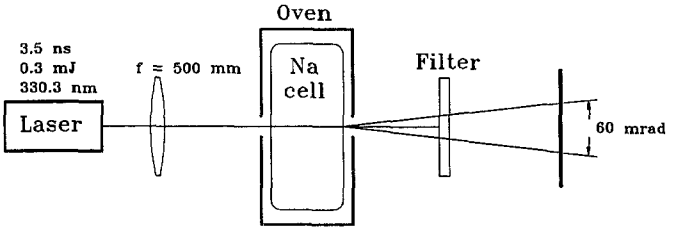

Fig. 1. Experimental setup

The exciting light pulses were generated in the system of a dye laser working on DCM, pumped by the second-harmonic of a Nd : YAG laser ($\lambda = 512$ nm). In order to obtain the ultraviolet radiation corresponding to 3*S*–4*P* transition ($\lambda = 330.2$ nm) the output pulses of the dye laser were frequency doubled. Finally, the light pulses of about 3.5 ns FWHM, 0.3 mJ, and 0.02 nm spectral width were generated. The laser light was focused by means of a 50 cm focal length lens forming a beam of the intensity of 10 MW/cm^2 inside the cell.

As it is commonly known, in dense atomic vapour irradiated by resonant laser light a very efficient collisional ionization occurs [14, 15]. We have used the ionization signal generated by this process as a simple and precise tuning indicator [16, 17]. In order to measure this signal two electrodes 2 cm apart working as Langmuir probes [18] were installed in the cell.

The radiation emitted from the cell was spectrally analyzed by optical filters. Its energy was measured by a pyroelectric energy meter (Laser Precision Corp., model RJ7200) with a flat response in 0.2–16 μm spectral range. The time resolved measurements were performed using a fast silicone photodiode and a real time oscilloscope (Tektronix model 2445B).

3 Results and Discussion

Under the irradiation of the sodium cell by the pulses tuned to the 3*S*–4*P* transition a strong yellow emission at 589 nm was observed. It was generated for

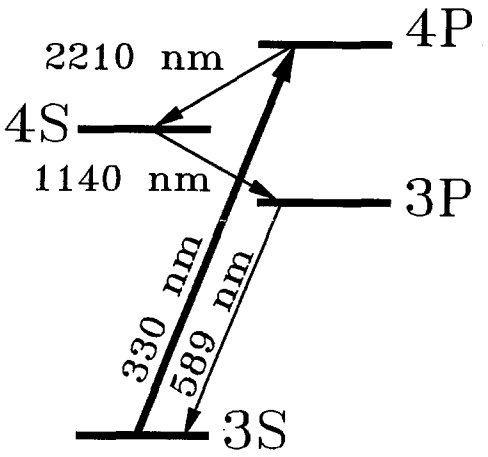

Fig. 2. Simplified Grotrian diagram of atomic sodium levels with observed transitions

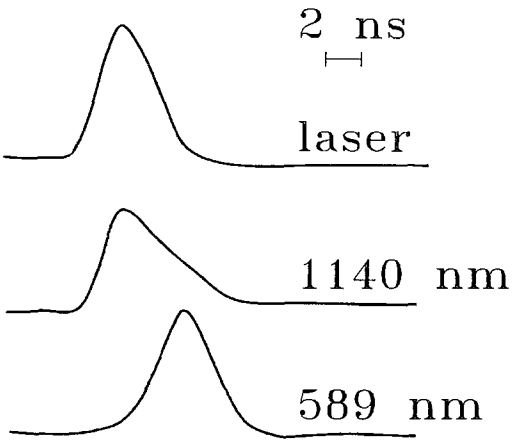

Fig. 3. Oscilloscope traces of exciting laser and emitted radiation pulses

the atomic concentration range from 5×10^{13} cm^{-3} to 5×10^{15} cm^{-3}. It occurred in the case of excitation of any fine structure resonance line but when the laser was tuned to the $3S$–$4P_{3/2}$ transition the emission efficiency was about 3 times higher than for the $3S$–$4P_{1/2}$ excitation. As is shown in Fig. 1, the radiation has a conical geometry and is emitted mainly in the forward direction along the excited volume, where the reabsorption on D-lines is minimum. We have also detected emission on 1140 nm and 2210 nm lines. The maximum pulse energy generated on all three lines reached about 1.2 μJ.

We have attributed the observed phenomenon to a cascade generation between $4P$–$4S$, $4S$–$3P$, and $3P$–$3S$ levels. Figure 2 presents the simplified Grotrian diagram of a sodium atom with the transitions involved. The pumping laser light saturates the $3S$–$4P$ transition within several picoseconds [19]. As a consequence, an inversion of population between $4P$ and $4S$ levels is created leading to the avalanche stimulated emission on the 2210 nm line. Since the $4S$ level is efficiently populated, the transition to the $3P$ level generates 1140 nm radiation. Redistribution of the population creates an inversion between the $3P$ and the $3S$ ground state and the generation of the 589 nm line takes place. We have not observed the cascade emission between $4P$–$3D$–$3P$ levels ($\lambda=9100$ nm and 819 nm, respectively). It is caused by a very small probability for the $4P$–$3D$ transition.

Figure 3 presents the temporal shapes of pulses of the exciting laser and the emitted radiation[1]. Unfortunately we could not record the time resolved signals on the 2210 nm line. The pulse of 1140 nm light of 4 ns FWHM is slightly longer than the pumping one. The peak of the 589 nm radiation signal is delayed about 3 ns with respect to the peak of the laser pulse because the inversion between $3P$ and $3S$ levels builds up as a consequence of two consecutive transitions ($4P$–$4S$ and $4S$–$3P$). Short delay times of the output pulses confirm that on the contrary to the emission observed by Müller et al. [4], the origin of this phenomenon is a radiative rather than

[1] The results presented in this paper concern the case of $3S$–$4P_{3/2}$ excitation

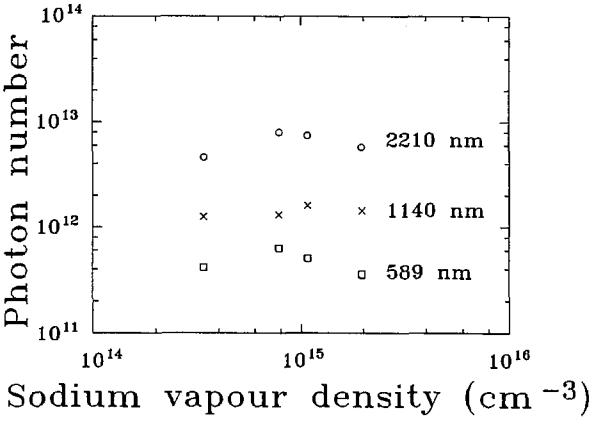

Fig. 4. Photon number per pulse vs sodium concentration

a collisional one. The duration times of the observed pulses are several times shorter than the natural lifetimes of any excited levels involved (110 ns, 38 ns, and 16.4 ns for $4P$, $4S$, and $3P$ states, respectively). It also confirms the mechanism suggested above.

In Fig. 4 the number of photons generated on each line per exciting pulse as a function of the atomic concentration is presented. The highest photon fluxes were emitted on the 2210 nm line while for 1140 nm and 589 nm the photon numbers were about 4 and 12 times lower, respectively. As it could be expected, the most efficient lasing between $4P$ and $4S$ levels occurs because the $4P$ state is directly populated by the pump laser pulse. For the other transitions the generation efficiency decreases due to the competitive processes such as, for example, the spontaneous emission or reabsorption. The decrease in the generation efficiency for vapour densities higher than 10^{15} cm^{-3} results from the fact that the exciting laser pulse energy is too low to saturate the resonant transition along the cell.

In Fig. 5 the dependence of the output pulse energy on 589 nm vs the input pulse energy is presented. A fairly good linear fit of the experimental points confirms a simple mechanism of the observed phenomenon presented above.

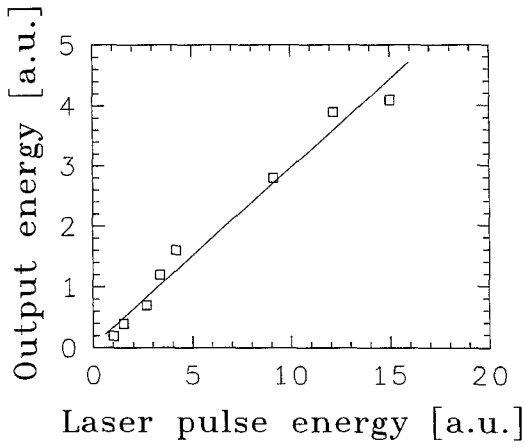

Fig. 5. 589 nm output pulse energy vs pumping laser pulse energy

As mentioned above, in resonantly excited dense atomic vapour a very efficient collisional ionization process occurs. In the theoretical model it was predicted [20] that in sodium vapour such an ionization should be more efficient in the case of the excitation to the $4P$ level ($\lambda = 330.3$ nm) than to the $3P$ level ($\lambda = 589$ nm). However, our recent measurements show that for the vapour densities higher than 10^{14} cm^{-3} the electron concentrations achieved due to the ultraviolet excitation are lower than that registered in the case of the yellow pumping light [21]. We attribute the discrepancy between the theoretical and experimental results to the phenomenon of the generation described above. For the ionization process the ensemble of excited atoms plays the role of an energy reservoir. In collisions – mainly superelastic ones between electrons and excited atoms – the energy stored in the atoms is converted into the plasma energy. In the case of $3S$–$4P$ excitation, the stimulated emission efficiently depopulates excited levels and consequently the energy is dissipated in the radiative processes. This conclusion is further confirmed by the fact that the number of photons generated per pulse (see Fig. 4) is comparable with the number of atoms in the volume irradiated by the pumping light.

The origin of the conical geometry of the observed emission is not understood in detail yet. We have considered several mechanisms especially those initiating the avalanche process such as, for example, resonantly enhanced four-wave mixing [8] but so far none of them gives a satisfactory explanation. The wave-mixing process should lead to the equal number of photons for each wavelength emitted from the cell (see Fig. 4) as well as to similar shapes of the pulses for each transition (see Fig. 3). However, it is quite possible that this process initiates the stimulated emission. The experiments investigating this mechanism are in progress.

In conclusion, we have observed the generation of radiation pulses on the atomic lines in sodium vapour excited to $4P$ state. It results from the cascade transitions between lower lying states.

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