Excitation of sodium atoms by 330-nm laser pulses

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Abstract. The evolution of populations of Na($4P$) and Na($3P_{1/2}$) atoms in a dense sodium vapour irradiated by nanosecond laser pulses tuned near the $3S \rightarrow 4P$ transition was investigated. It was observed that the population of Na($4P$) atoms remained high only within the laser pulse, in spite of the relatively long lifetime of the $4P$ level (110 ns). The $3P_{1/2}$ level, which is populated as a result of cascade transitions from the higher levels, reached the highest population several nanoseconds after the laser shot. The fast population changes are explained by cascade-stimulated transitions between the excited atomic levels.

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Irradiation of atoms by resonant light changes the population of atomic levels. As far as low-density atomic gas and weak excitation are concerned, transitions between the levels are caused by the light absorption and spontaneous emission. Thus the population of the excited levels can be determined by means of a simple rate-equation model. An example of the evolution of the Na($4P$), Na($3D$), Na($4S$), and Na($3P$) populations, in a low-density sodium vapour initially excited to the $4P_{1/2}$ level, is shown in Fig. 1b.

In dense vapours various processes influence the population of atomic levels. One of the commonly known effects is the resonance radiation trapping [1]. As was shown recently [2–5] the mechanism is nonlinear when the excitation is strong. Then, different collisional processes have to be taken into account [6, 7], among them electronic and ionic impacts should be considered, because the vapour is usually ionized. Moreover, at higher densities and stronger excitations, various stimulated radiative processes can occur [8–19]. A variety of mechanisms arising in such a medium makes the theoretical description of the population evolution quite difficult.

We have focused our attention on the populations of the atomic levels in a dense sodium vapour ($N \approx 10^{14}$ cm^{-3}) excited by a strong laser pulse tuned near the $3S$–$4P$ transition ($\lambda = 330$ nm). Dense atomic and molecular vapours excited to
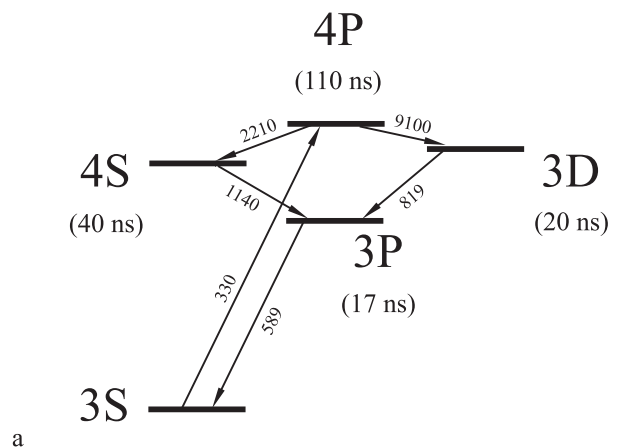
a
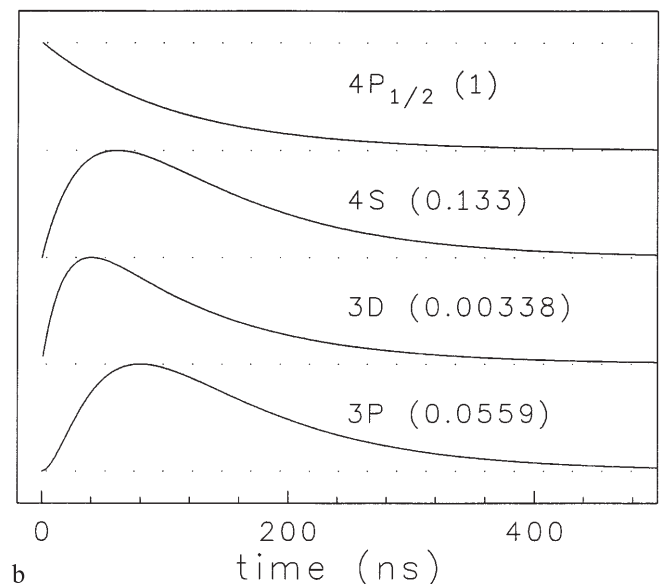
b

Fig. 1. a Simplified Grotrian diagram of sodium levels with transition of interest. **b** Results of calculations of spontaneous relaxation of the atomic levels population in a low-density sodium vapour initially excited to the $4P_{1/2}$ level. The excitation process is considered negligibly short and localized at zero on the time scale. The numbers in brackets denote the highest relative population of the levels

higher levels are often considered as potential new sources of laser radiation. They are also used as efficient sources of low-temperature electrodeless plasma, which usually occurs as the result of collisions [6, 7]. Therefore, the understanding of processes that take place in such a medium is important in order to recognize the energy transfer mechanisms from light to the excited atoms ensemble, and then to describe the conversion to the energy of other excited atoms or molecules, or to the energy of plasma.

Excitation of dense sodium vapour by 330-nm laser pulses was studied earlier by means of a numerical model [20], but only the collisional energy transfer and the linear radiation imprisonment were taken into account. Recently, new radiative phenomena were observed in such a medium. Han, Lu, and Ma [21] and then Stacewicz, Kotowski, and Wiewiór [22] observed a generation of radiation at wavelengths close to the atomic lines corresponding to the cascade transitions between the excited levels: 2210 nm ($4P \rightarrow 4S$), 9100 nm ($4P \rightarrow 3D$) 1140 nm ($4S \rightarrow 3P$), 819 nm ($3D \rightarrow 3P$), and 589 nm ($3P \rightarrow 3S$). The emission was generated in both forward and backward directions with respect to the laser beam and usually was emitted in cones or collimated beams. More-detailed investigations of this phenomenon were carried out by Domiaty et al. [23] and Stacewicz and Kozłowvski [24]. It was found that the generation was due to several processes such as stimulated electronic Raman scattering (SERS), hyper Raman scattering (HRS), four wave mixing (FWM), and amplified spontaneous emission (ASE). These processes influence each other. Their contribution to the generation was discussed in detail in previous papers [23–25]. On the whole, ASE occurs for all the lines, especially when the laser is tuned close to the $3S \rightarrow 4P$ transition. SERS dominates at 9100 and 2210 nm. For the lower transitions at 1140 and 819 nm (see Fig. 1a) the emission arises from HRS with an ASE contribution, whereas FWM (responsible for the conical geometry of radiation emitted in the forward direction with respect to the laser beam) occurs mainly at 589 nm. Since the emission is generated at wavelengths close to the resonance wavelengths, the process is also affected by the absorption [25]. The duration times of generated pulses do not exceed several nanoseconds. The power of the pulses can even reach several kW, which means that they can influence the atomic population much more efficiently than spontaneous transitions or inelastic collisions.

In such a medium the population of excited atoms cannot be studied by the usual method, i.e. by the analysis of fluorescence on lines originating from the levels of interest. As was shown in our previous paper [24], the generation occurring at these wavelengths (9100, 2210, 1140, and 819 nm) provides strong peaks of scattered radiation, which disturb the fluorescence signals. The fluorescence at the $3P \rightarrow 3S$ transition (589 nm) is strongly affected by the radiation trapping. Therefore in our experiment the evolution of the population of atomic levels was studied using two other methods: absorption and photoionization from the excited states.

1 Experiment

The experimental setup is presented Fig. 2. The sodium vapour was stored in a heatpipe at a temperature of 350 °C.
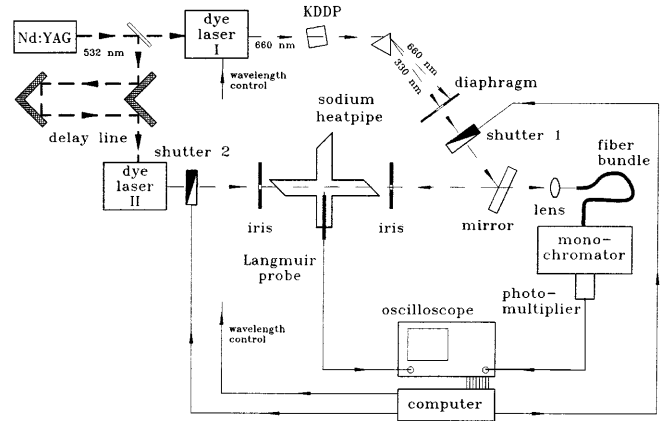

Fig. 2. The experimental setup

Helium at a pressure of about 10 Torr was added as a buffer gas. The length of the vapour column was 10 cm.

The exciting 330-nm light pulses were achieved by frequency-doubling of pulses from a dye laser working on DCM. The energy of the ultraviolet pulses reached about 0.4 mJ, their spectral linewidth was about 0.01 nm, and their FWHM time was about 7 ns. The radiation from the second dye laser was used as a probe beam. In order to achieve broad and flat spectra this laser was tuned by a prism and a mirror, and an uncoated glass wedge was used for the output. The spectral linewidth of the probe laser was about several Å and because of a low Q factor of its cavity no mode structure was observed in the laser spectrum. A perfect collinearity of counterpropagating laser beams was achieved in the heatpipe by means of two iris diaphragms. The exciting beam diameter inside the heatpipe was about 3 mm. In order to penetrate only the well-excited region of the vapour the probe beam diameter was about 1 mm, and the diameter of the exciting beam was about 3 mm. Both dye lasers were pumped by the second-harmonic of the Nd:YAG laser. Temporal coincidence of the exciting and probe pulses was controlled by means of an optical delay line installed at the part of the Nd:YAG laser beam pumping the probe laser.

The absorption method consists in the well-known fact that the total energy absorbed at a resonance line is proportional to the product of the atomic concentration N_i in the initial level and the oscillator strength f_{ik} of the transition. We used this method for the investigations of the Na($3P_{1/2}$) concentration. The probe laser was tuned to the 568-nm line corresponding to the $3P_{1/2} \rightarrow 4D_{3/2}$ transition. To avoid disturbance of the Na($3P_{1/2}$) atomic population, the energy of the probe pulse applied was as low as 20 nJ. The light transmitted through the heatpipe was collected by a fiber bundle attached to a monochromator tuned to the absorption line. The monochromator reduced the effective linewidth of the probe laser to a value of about 60 GHz, allowing us to determine a definite absorption of the light pulse within a narrow atomic line. The signal was detected by a photomultiplier and then analyzed by means of a fast digital oscilloscope controlled by a computer.

In order to determine the absorption caused by the excited atoms and to exclude losses of the light caused by the heatpipe window and other optical elements the measurements were done alternately when the vapour was illuminated

by the exciting ultraviolet beam and then when the 330-nm light producing the excited atoms was stopped by shutter number 1. Then the total absorption coefficient was found from

$$A = \frac{\Phi_0 - \Phi_T}{\Phi_0} \, , \qquad (1)$$

where Φ_T denotes the energy of the probe laser pulse transmitted through the cell in the presence of the ultraviolet beam and Φ_0 corresponds to the probe pulse energy when the ultraviolet beam was switched off. Alternatively, the absorption coefficient can be calculated by using the Lambert–Beer law:

$$A = 1 - \int P(\nu) e^{k(\nu)l} \, d\nu \, , \qquad (2)$$

where $P(\nu)$ describes the effective (monochromator) probe line profile, the length of the cell, and $k(\nu)$ the spectral absorption coefficient, which was calculated by taking both Doppler broadening and Lorenz broadening (Voigt profile) into account.

$$k(\nu) = \frac{A_{if} c^2 g_f N_i}{2\pi^{\frac{3}{2}} \gamma g_i V_p \nu_0^2} \int_{-\infty}^{\infty} \frac{e^{-V^2/V_p^2} (\gamma/2)^2 \, dV}{4\pi^2 [\nu - \nu_0 (1 + V/c)]^2 + (\gamma/2)^2} \, . \qquad (3)$$

Here A_{if} denotes the spontaneous emission coefficient g_i and g_f the statistical weights of the initial and final levels, and V the atomic velocity. $V_p = \sqrt{2k_B T/M}$ is the average speed of atoms (k_B Boltzmann constant, T vapour temperature, M atomic mass). ν_0 is the frequency of the transition, and c the speed of light. In the heatpipe, the linewidth γ was determined mainly by resonance broadening (γ_R) and by pressure broadening (γ_{He}) : $\gamma = \gamma_R + \gamma_{He}$ [27, 28]. Finally the concentration of atoms N_i in the initial state was found by solving the system equations (1–3). The standard deviation for this method does not exceed 50%.

The absorption method is not convenient for the determination of the population of levels higher than $3P$ (for example, $4P$), because either the lines starting from such levels belong to the infrared range and their generation by dye lasers is difficult, or their oscillator strengths are too small to observe the losses of the energy of the probe pulses transmitted through the heatpipe. In these cases the photoionization of the excited atoms can be used. In our experiment an efficient stepwise ionization of excited atoms by 330-nm laser light was observed as a current peak on the Langmuir probe. When an additional probe light pulse was introduced to the cell, there was a rise in the photoelectron signal due to an increase in the ionizing light intensity. For these measurements the energies of probe pulses of about 0.2 mJ were applied.

Figure 3 presents a typical signal induced on the Langmuir probe by a 330-nm pulse. The probe was connected to a 50-Ω input on the digital oscilloscope. No bias voltage was used. A positive pulse of several ns duration time was caused by the photoelectric effect on the electrode surface by the scattered laser light[1]. This pulse was well correlated with the

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[1] The threshold wavelength for the photoelectric effect in sodium is about 528 nm. The threshold wavelengths for photoionization of Na(4P), Na(3D), and Na(4S) are equal to 895, 815, and 636 nm.
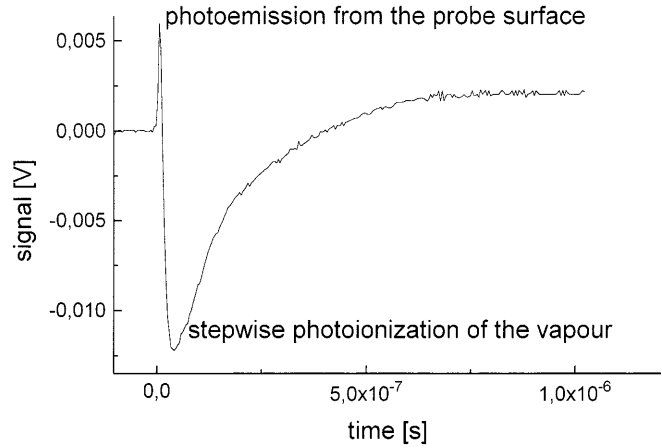

Fig. 3. A typical photoionization signal observed by means of the plasma probe in sodium vapour excited by 330-nm laser pulses

laser shot. The negative peak of current which occurred afterwards (its amplitude we denote by I_0) was a matter for measurement, because it followed from a flux of electrons generated in the vapour by stepwise photoionization. This peak disappeared when the ultraviolet laser was tuned far from the $3S \rightarrow 4P$ resonance, i.e. when there was no production of excited atoms. The peak delay (about 40 ns, with respect to the laser shot) corresponded to the time taken for the diffusion of the plasma cloud from the photoionization region to the probe; it depended on the distance between the probe and the laser beam. When both the probe and the exciting pulses were applied we observed an increase in the peak amplitude (to the value denoted by I_P). Since the wavelength of the probe light (850 nm) exceeded only the photoionization threshold of the $4P$ level, the increase of the photoionization signal,

$$E = I_P - I_0 \qquad (4)$$

was proportional to the concentration of Na(4P) atoms. We assume that the population of atoms excited higher than the Na(4P) level is neglible. Peaks I_0 and I_P were registered for closed and opened position of the shutter number 2. Since the spectra for the stepwise photoionization are not well known [29] and the parameters of the probe laser beam inside the heatpipe can not be determined precisely, this method allows us to find only the relative concentrations of the excited atoms. The standard deviation for this method is also about 50%.

Using both methods the evolution of the excited levels population was studied by changing the delay between the exciting and the probe pulses. The temporal resolution of these measurements was limited by the laser pulse length.

2 Results and discussion

In Fig. 4 the Na(4P) concentration as a function of the exciting laser wavelength is presented. This spectrum was compared with the excitation spectrum for the total (forward and backward with respect to the laser beam) stimulated emission at 2210 nm. The $4P \rightarrow 4S$ and $4P \rightarrow 3D$ transitions
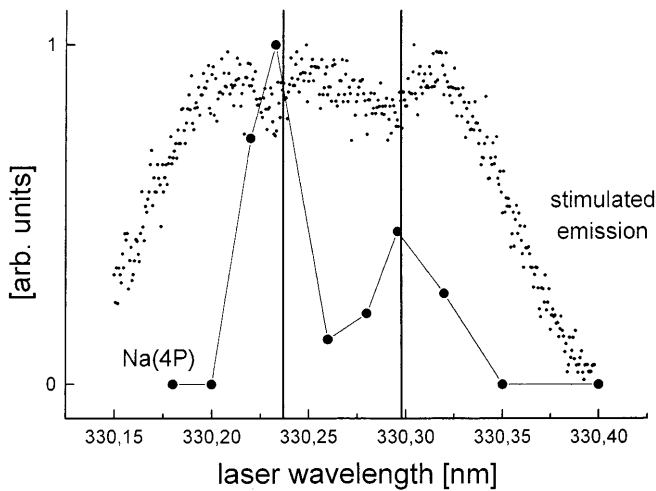
Fig. 4. Excitation spectrum of the stimulated emission at 2210 nm (*dots*) and for the Na(4*P*) population. The two vertical lines represent the wavelengths of the $3S \rightarrow 4P$ doublet

Fig. 6. Population of Na($3P_{1/2}$) atoms as a function of the existing laser wavelength compared with the excitation spectrum of the stimulated emission

initialize the stimulated process but the contribution of the second channel is relatively very weak [24]. Therefore the emission spectrum at 2210 nm can be considered as the spectrum for stimulated process induced by 330-nm laser pulses. A high population of the Na(4*P*) atoms was achieved only for the laser wavelengths localized near the resonance lines. As far as the stimulated emission is concerned, its spectrum is broad and flat. This is because the emission at 2210 nm is not due solely to ASE arising from the high population of the 4*P* level. This generation is also caused by SERS [21–25], which is responsible for the broad wings of the excitation spectrum of the 2210-nm, emission. However SERS does not lead to the Na(4*P*) population.

Figure 5 presents the temporal evolution of the Na(4*P*) atoms. Their population remains high only within the laser pulse and decays with a time constant of about 10 ns, which is much shorter than the natural lifetime of the 4*P* level (110 ns). The phenomenon is caused by stimulated depopulation. The maximum number of photons (6×10^{13}) generated at 2210 nm per pulse, when the laser was tuned to the $3S_{1/2} \rightarrow 3P_{3/2}$ transition, was comparable to the number of
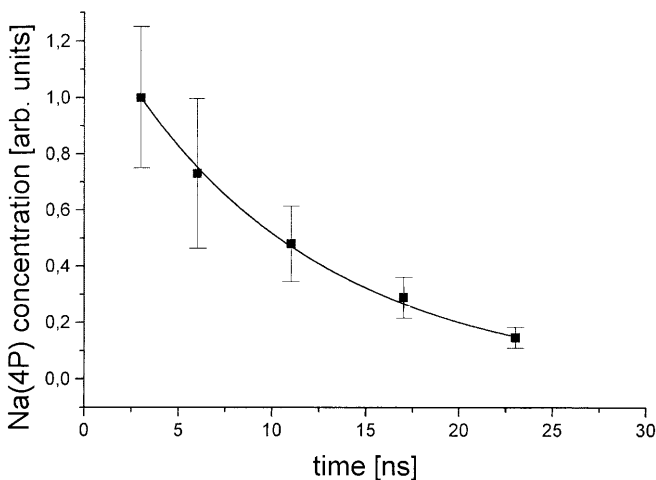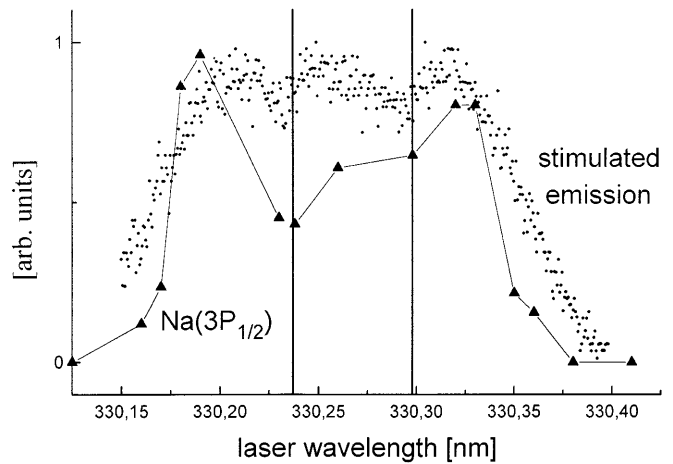
atoms contained in the vapour volume interacting with the light beam.

In Fig. 6 the excitation spectrum of Na($3P_{1/2}$) atoms is presented. For this level the spectrum is almost as broad as the excitation spectrum for the stimulated emission, because the Na(3*P*) atoms can be produced not only by ASE cascades from the 4*P* level, but also by HRS or a combination of SERS and ASE. The maximum value of the Na($3P_{1/2}$) concentration reached 9×10^{13} cm^{-3}. The temporal behavior of the Na($3P_{1/2}$) concentration (Fig. 7) shows that this level reaches the population maximum within approximately 20 ns from the beginning of the exciting laser pulse. The evolution of this population, which is due to the stimulated mechanism, is much faster than could be anticipated from the spontaneous emission (Fig. 1).

When the probe laser was tuned to 589 nm we observed amplification of the light beam. This amplification as a function of the exciting laser wavelength is shown in Fig. 8. As was mentioned above the amplification at 589 nm occurs as a result of ASE and FWM. The efficiency of other phenomena, such as the secondary nonlinear processes starting from the excited levels. is negligible [24]. Since in our experiment
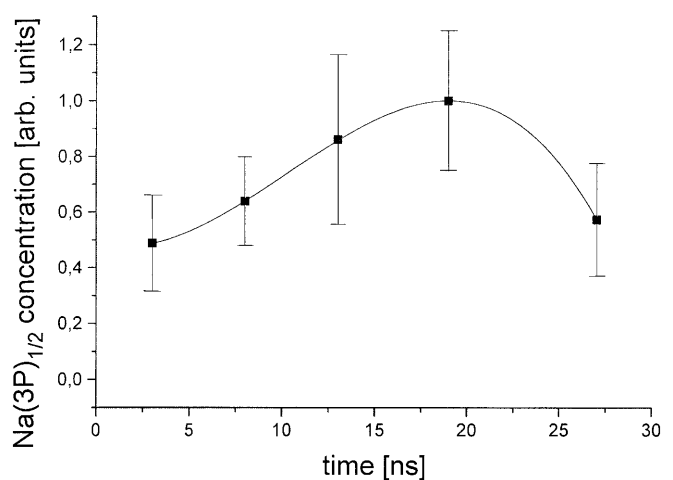

Fig. 5. The Na(4*P*) population as a function of time

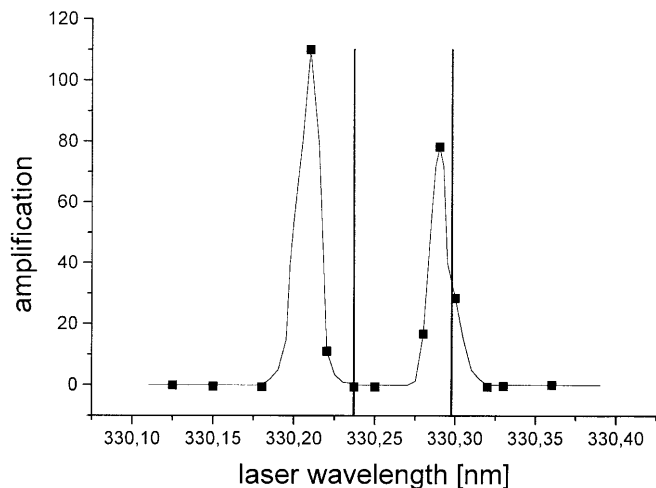**Fig. 7.** The Na($3P_{1/2}$) population as a function of time

Fig. 8. Amplification of 589-nm as a function of the exciting laser wavelength

the probe beam propagates in the opposite direction to the exciting beam, the phase matching conditions for the FWM can not be fulfilled and this amplification can arise only as a result of "ordinary" stimulated emission. Therefore the amplification coefficient is a measure of the population inversion between the $3P$ and $3S$ levels. The amplification, which can reach a factor of 110, arises only when there is temporal coincidence between the exciting and the probe pulse and when the ultraviolet laser is well tuned to the $3S \rightarrow 4P$ transition. This is in good agreement with our observations of ASE in the previous experiment [24]. Because ASE depopulates the $3P$ level efficiently, the maximum concentration of the Na($3P_{1/2}$) atoms occurs with a several-nanosecond delay with respect to the laser pulse (see Fig. 7), when the generation at the yellow line disappears.

The other effect, which can be responsible for the amplification, is lasing without inversion (LWI) [30]. It was observed by Zibrov et al. [31]. In our experiment, amplification due to the inversion of population and amplification and due to LWI are indistinguishable. However the amplification due to LWI does not usually exceed a factor of 3, and is observed in a narrow spectral range, limited by the atomic linewidth. Therefore it seems that LWI does not contribute significantly to the amplification of a spectrally broad probe laser beam.

The maximum of the amplification spectrum is shifted to the short wavelengths with respect to the $3S \rightarrow 4P$ resonance lines. This is probably caused by self-focusing of the exciting laser light, which was hard to avoid in our experiment despite the use of beams of a relatively large diameter. As was observed in the experiments of Grischkovsky [32] the self-focusing takes place on the blue wing of the atomic line. In our experiment the self-focusing led to a rise in the laser intensity, which increased the efficiency of the nonlinear processes populating the $3P$ level. The blueshift was not observed for the Na($4P$) excitation spectrum (Fig. 4), since these atoms were populated due to linear excitation.

3 Conclusion

We investigated the population of the $4P$ and the $3P$ levels in a dense sodium vapour excited by laser pulses tuned near the $3S \rightarrow 4P$ transition. In these circumstances the Na($4P$) atoms are intensively depopulated as a result of cascade-stimulated emission. The same cascades populate the Na($3P$) atoms so efficiently, that the inversion of population between the $3P$ and the $3S$ levels can occur, which is manifested by light amplification at the 589-nm line.

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