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Near-IR lasing in caesium vapour

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Abstract. **Lasing in caesium vapour is obtained at $\lambda \sim 3$ μm (transitions $7P_{3/2} \rightarrow 7S_{1/2}$ and $7P_{1/2} \rightarrow 7S_{1/2}$). Longitudinal pumping is performed by the second harmonic of a parametric oscillator pumped by the second harmonic of a Nd^{3+}-doped garnet laser. The pump wavelength corresponds to the $6S_{1/2} \rightarrow 7P_{3/2}$ and $6S_{1/2} \rightarrow 7P_{1/2}$ transitions of caesium atoms. The pump radiation has a spectral width of 12 cm^{-1}, a pulse energy of 10 mJ, and a pulse repetition rate of 10 Hz. The IR laser pulse energy at a vapour cell temperature of 155 °C is 100 μJ. The efficiency of pump energy conversion to the laser radiation energy at $\lambda \sim 3$ μm is ∼1.5%.**

Keywords: *caesium vapour laser, near-IR region, longitudinal pumping.*

The concept of optically pumped lasers based on alkali-metal vapours was proposed as early as in 1958 [1]. This idea was practically implemented for the first time in 1962 [2] with resonance optical pumping of caesium vapours by one of the strong emission lines of helium atoms with $\lambda = 388.8$ nm. Rabinowitz et al. [2] obtained cw lasing on the $8^2P_{1/2} \rightarrow 8^2S_{1/2}$ transition of the caesium atom ($\lambda = 7.18$ μm), i.e., created the first optically pumped laser based on atomic transitions.

It is of practical interest to obtain efficient lasing on the $(n + 1)P \rightarrow (n + 1)S$ transitions of caesium or rubidium atoms at a wavelength near $\lambda = 3$ μm. Laser radiation in this spectral region is necessary, in particular, for optical location and diagnostics of pollutants in the atmosphere.

In [3–5], rubidium vapours were excited by a tunable dye laser ($\lambda = 421.7$ and 420.2 nm) to achieve cavity-free lasing. The pulsed laser energy achieved in [5] was ∼100 nJ, which corresponded to a pump energy conversion efficiency of $\sim 1.7 \times 10^{-4}$.

In the present work, we experimentally obtained three-micron cavity lasing in vapours of a caesium–helium mixture under pumping the $7P_{3/2}$ and $7P_{1/2}$ levels by blue radiation ($\lambda_p = 455.5$ and 459.5 nm, respectively). The energy conversion efficiency was ∼1.5%, which corresponds to a photon conversion efficiency of ∼10%.

As pump radiation, we used the second harmonic of an optical parametric oscillator based on a BBO crystal pumped by the second harmonic of a Nd^{3+}-doped garnet laser. The pump wavelength corresponded to the $6S_{1/2} \rightarrow 7P_{3/2}$ and $6S_{1/2} \rightarrow 7P_{1/2}$ transitions of caesium atoms, and the spectral width of the pump radiation was 0.25 nm (12 cm^{-1}). The temporal pulse profile corresponded to a Gaussian signal with a FWHM duration of ∼5 ns, and the transverse intensity profile of the pump beam ∼8 mm in diameter was close to a rectangular one. The energy of the blue and near-UV pump pulses did not exceed 10 mJ; the pulse repetition rate was 10 Hz.

A cylindrical cell 45 cm long was evacuated to a pressure of less than 10^{-6} Torr. We placed ∼2 g of caesium into the cell and added ^{4}He buffer gas to a pressure of ∼4 atm at room temperature. This ^{4}He pressure was used to increase the absorption linewidth of Cs atoms and match it to the pump radiation linewidth. The input and output windows were made of leucosapphire without antireflection coatings. The Fresnel losses at each window were ∼15% for both pump and IR radiation. Resistive heaters allowed us to set the temperature of the cell walls at up to 250 °C with an accuracy of ∼1%. To avoid metal condensation on the windows, they were heated by external heaters so that their temperature was about 20 °C higher than the temperature of the walls.

Figure 1 shows a fragment of the energy level diagram of the caesium atom taken from [6]. The gap between the $7P_{3/2}$ and $7P_{1/2}$ levels is 181 cm^{-1} [6, 7], and the rate of collisional mixing of these levels at the ^{4}He pressure $p \approx 4$ atm should be rather high. Therefore, lasing in the mixture with helium can occur at the direct $7P_{3/2} \rightarrow 7S_{1/2}$ and $7P_{1/2} \rightarrow 7S_{1/2}$ transitions with $\lambda = 2.93$ and 3.10 μm, respectively.

The concentration of caesium atoms was calibrated by measuring the cell transmission at the wavelength of an additional diode laser ($\lambda = 852.32$ nm), which corresponds to the

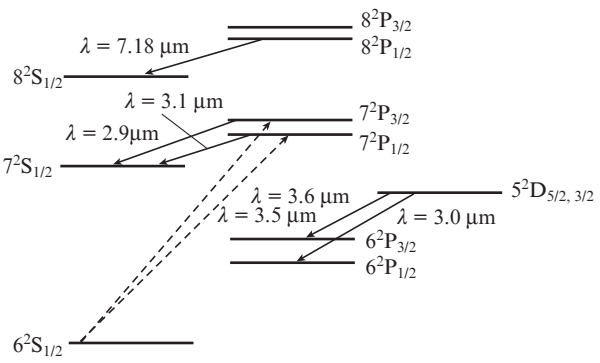

Figure 1. Fragment of the energy level diagram of the caesium atom. The dashed lines show pump radiation exciting the $7P_{3/2}$ and $7P_{1/2}$ terms.

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long-wavelength wing of the $6S_{1/2} \rightarrow 6P_{3/2}$ absorption transition line ($\lambda = 852$ nm). In the temperature range of 60–80 °C, the measured concentration of caesium atoms n_{Cs} corresponded to the calculated values, namely, to $n_{Cs} = 4 \times 10^{12}$ cm^{-3} at 80 °C. Thus, according to theoretical estimates, the concentration $n_{Cs} \sim 10^{15}$ cm^{-3} needed for IR lasing should be achieved at temperatures of 150–160 °C [8].

Figure 2 shows the temperature dependences of the cell transmission coefficients $K_{tr} = W_{out}/W_{in}$ for wavelengths used to pump the $7P_{3/2}$ and $7P_{1/2}$ levels ($\lambda_p = 455.5$ and 459.5 nm, respectively), which were measured with calorimeters (W_{out} is the pump energy passed through the cells with Cs vapours and W_{in} is the energy incident on the cell). One can see from Fig. 2 that the working temperature range in this experiment is, as expected, 100–150 °C.

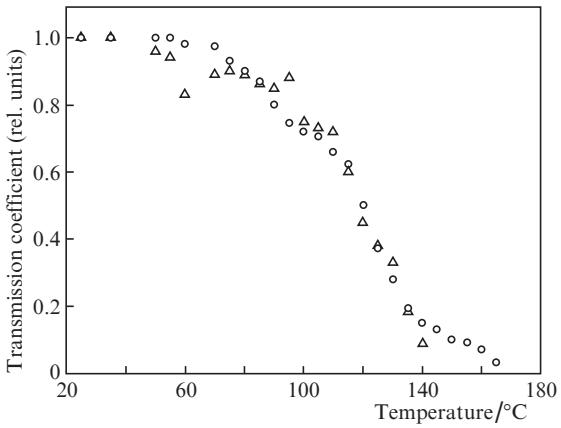

Figure 2. Temperature dependences of the cell transmission coefficients for radiation pumping the (o) $7P_{3/2}$ ($\lambda_p = 455.5$ nm) and (Δ) $7P_{1/2}$ ($\lambda_p = 459.5$ nm) levels.

The system of recording IR radiation at the cell output consisted of detectors operating in the range of 2–20 μm and a monochromator recording the signal within a band of 2–9 μm. One of the detectors measured the total lasing energy summed over all emission lines; the second detector was placed behind the monochromator and, hence, measured the energy emitted at a particular transition.

To obtain lasing on the IR transition of the caesium atom, a highly reflecting cavity mirror in front of the cell was aligned normally to the pump beam, while the plane-parallel output window of the cell served as an output mirror. When the cell was heated to ~60 °C, the IR detectors recorded a signal whose fluctuations caused by the pump energy instability (~5%) exceeded 100%, which is characteristic for near-threshold lasing. With a further increase in the cell temperature, the IR signal (output energy) fluctuations were stabilised at a level of 10%–12%. A misalignment of the highly reflecting mirror of the cavity at a cell temperature of ~150 °C led to a more than tenfold decrease in the output signal. Therefore, cavity lasing in caesium vapours is evident.

Our study of the radiation spectrum using an IR monochromator showed that pumping of caesium vapours to the $7P_{3/2}$ level ($\lambda_p = 455.5$ nm) leads to lasing at four IR lines (Fig. 3) with wavelengths of 2.933, 3.013, 3.096, and 3.495 μm. Pumping of the $7P_{1/2}$ level ($\lambda_p = 459.5$ nm) results in three IR laser lines with wavelengths of 3.013, 3.096, and 3.495 μm. Note that this pumping does not excite the laser line with $\lambda =$ 2.933 μm, which corresponds to the $7P_{3/2} \rightarrow 7S_{1/2}$ transition in the case of pumping with the shorter wavelength ($\lambda_p =$ 455.5 nm, $6S_{1/2} \rightarrow 7P_{1/2}$). At the same time, the presence of the laser line with $\lambda = 3.495$ μm indicates that the $7P_{3/2} \rightarrow 5D_{5/2}$ transition is excited by both pumping wavelengths. This means that the emission spectrum of caesium atoms should exhibit intense luminescence at $\lambda \sim 1.4$ μm. However, since the $5D_{5/2}$ level lifetime considerably exceeds the lifetime of the $7P_{3/2}$ level (1300 ns versus 135 ns [9]), lasing at this wavelength ($\lambda \sim 1.4$ μm) is impossible.

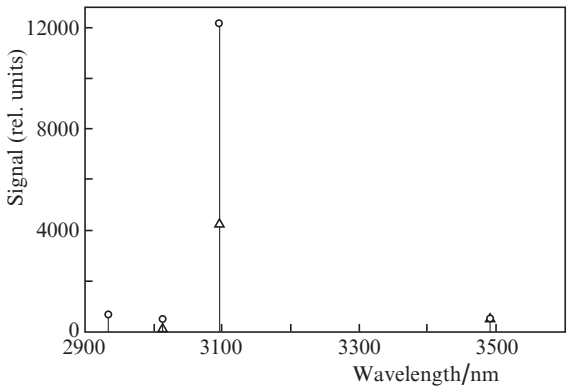

Figure 3. Lasing spectra of caesium atoms upon pumping to the (o) $7P_{3/2}$ and (Δ) $7P_{1/2}$ levels at a temperature of 140 °C.

Comparison of the observed laser wavelengths with the data of [6, 9] shows that the experimentally measured wavelengths correspond to the following transitions in the caesium atom:

$$\lambda = 2.933\ \mu m - 7P_{3/2} \rightarrow 7S_{1/2},$$
$$\lambda = 3.013\ \mu m - 5D_{3/2} \rightarrow 6P_{1/2},$$
$$\lambda = 3.096\ \mu m - 7P_{1/2} \rightarrow 7S_{1/2},$$
$$\lambda = 3.495\ \mu m - 5D_{5/2} \rightarrow 6P_{3/2}.$$

The accuracy of our experimental values was determined by the characteristics of the used monochromator to be ~1 nm, which means that some discrepancy between the measured wavelengths and their reference values can be caused by inaccuracies in the calibration of the used monochromator and in its alignment to the line peak, as well as by a shift of the laser levels due to the interaction of caesium atoms with the buffer gas.

As is seen from Fig. 3, the maximum output energy is obtained at $\lambda \sim 3.096$ μm (transition $7P_{1/2} \rightarrow 7S_{1/2}$) for both pump wavelengths. This is quite natural because lasing from the $7P_{1/2}$ level begins either in the case of its direct population or due to a rather fast mixing of the $7P_{3/2}$ and $7P_{1/2}$ levels as a result of collisions. The fact that the IR radiation intensity in the case of the caesium atom excitation to the $7P_{3/2}$ level is approximately threefold higher (Fig. 3) than upon excitation to the $7P_{1/2}$ level is probably related to the specific features of the excited levels.

Figure 4 shows the experimental temperature dependences of the laser pulse energy for the strongest IR lines, at $\lambda =$ 3.096 μm (transition $7P_{1/2} \rightarrow 7S_{1/2}$) and $\lambda = 3.013$ μm (transition $5D_{3/2} \rightarrow 6P_{1/2}$), as well as the temperature dependence of the total energy of all lines (also in relative units). It is necessary to note that the energies at $\lambda = 3.096$ and 3.013 μm were

determined at identical alignments of the monochromator and identical attenuating filters in front of the IR detector recording the signal at the exit from the monochromator. Therefore, it is possible to compare these energies (Figs 4a and 4b) for different pump wavelengths ($6S_{1/2} \rightarrow 7P_{3/2}$ and $6S_{1/2} \rightarrow 7P_{1/2}$). However, the total energy signal was recorded by another detector and, hence, its signal cannot be compared with the signals at individual lines.

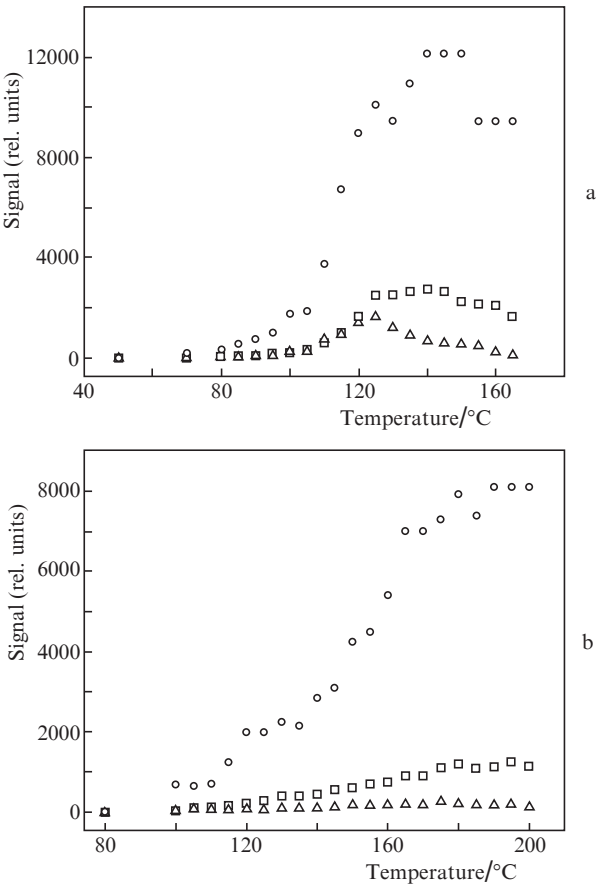

Figure 4. Temperature dependences of the laser energy at wavelengths of (○) 3.096 and (△) 3.013 μm and (□) summed over the spectrum in the case of pumping to the (a) $7P_{3/2}$ and (b) $7P_{1/2}$ levels.

Our experiments showed that the highest pulse energy of the three-micron radiation was obtained in the case of excitation of the $7P_{3/2}$ level (λ_p = 455.5 nm). Figure 5 presents the spectra of pump radiation (pumping to the $7P_{3/2}$ level) incident on the cell and passed through it, which are recorded at a temperature of 150 °C. One can see that 84.5 % of the pump energy is absorbed along the entire path (45 cm). This value is, in fact, the quantum efficiency, which determines the maximum possible IR lasing efficiency. Direct measurement of the pump energy absorbed in the cell by measuring the transmission coefficient with the use of calorimeters yielded the same value (see Fig. 2). Therefore, the energy spent on the level excitation is almost completely transferred to the laser energy. Note that the spectra of the incident and passed pump radiation were recorded using similar but different spectrometers. The absolute wavelength calibration of the spectrometer was performed using a He–Ne laser before each measurement only for the incident pump radiation. The spectrometer recording the passed radiation was calibrated at the beginning of measurements, and the wavelength fitting of the measured spectrum of the passed pump radiation was performed by the wings of the spectra of the incident and passed beams. The pump pulse energy in all conducted experiments was approximately the same and equal to 8–10 mJ. The laser energy measured upon pumping of each level was 100 μJ. Therefore, the energy efficiency was 1 %–1.5 %, which corresponds to the experimentally obtained quantum efficiency with allowance for the ratio of the pump and laser wavelengths. The really measured energy efficiency is considerably lower than the calculated one, which is related to the existence of unaccounted losses in the laser cavity at the lasing wavelength. Optimisation of the laser scheme by, for example, selecting the output mirror, which could increase the output energy by several times, was not performed.

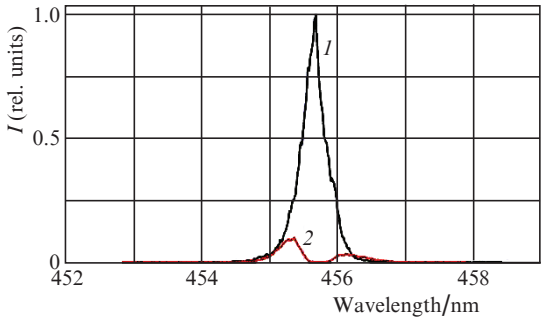

Figure 5. Intensities I of the spectra of pump radiation (*1*) incident on the cell and (*2*) passed through it. Cell temperature is ~150 °C, λ_p = 455.5 nm.

Figure 6 shows the experimental and calculated shapes of IR laser pulses at a temperature of the cell walls of 155 °C. The experimental oscillograms are obtained using a cooled fast (response time ~1 ns) IR diode.

The theoretical study of lasing was based on the kinetic 3D model of a laser with longitudinal end pumping in the ray optics approximation of pump radiation and laser radiation transfer. The model includes the kinetics of the 6S, 6P, 7S and 7P levels of the caesium atom, photoionisation, Penning ionisation, and recombination and takes into account the experimental shape of the pump pulse and its spectral distribution. The nonsteady-state mutually consistent problem is solved using splitting with respect to physical processes. First, pump rates are found; next, the level population kinetic equations are solved without allowance for stimulated processes; then, the laser gain is calculated; and the change in populations under action of laser radiation at the end of the time step is found.

One can see from Fig. 6 that the laser pulse has a multipeak structure. This is probably caused by a short (~5 ns) pump pulse duration and a rather long cell (time of passage through the cell is ~1.4 ns). A positive inversion on the laser transition under these conditions can be formed not over the entire active medium length, which may lead to the appearance of spikes in the laser pulse. Note that the negative part of the signal on the experimental oscillogram is obviously related to the insufficiently wide band of the photodiode–oscilloscope system and, therefore, the laser pulse duration should be noticeably shorter than the response time of the photodiode detector (the oscilloscope bandwidth is no less than

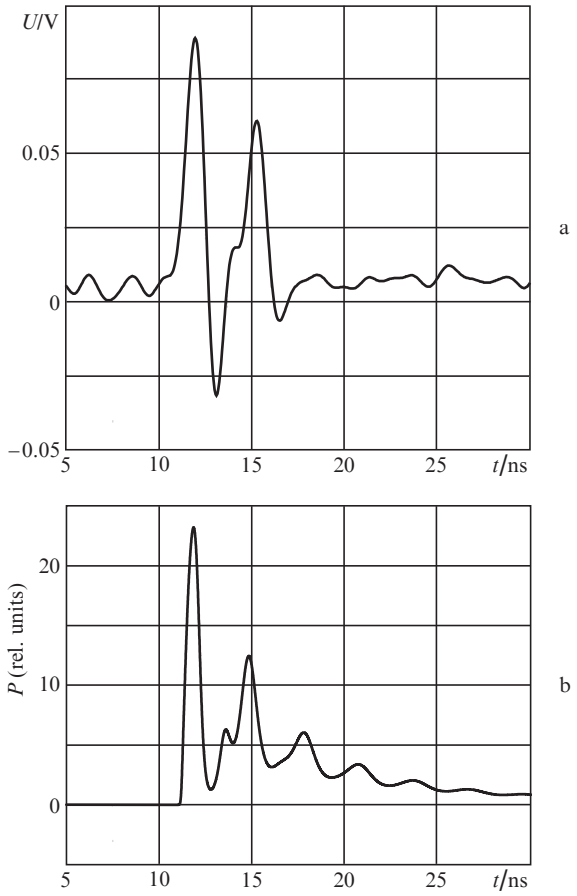

Figure 6. (a) Experimental and (b) calculated IR laser pulses at a temperature of cell walls of 155 °C.

1 GHz). As is seen from Fig. 6, the calculated shape of the laser pulse qualitatively coincides with the experimental shape and the calculated laser pulse energy (105 μJ) is quite close to the measured value.

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