Lasing on the D_2 line of sodium in a helium atmosphere due to optical pumping on the D_1 line (up-conversion)

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A method is proposed to produce population inversion on transitions involving the ground state of atoms. The method is realized experimentally with sodium atoms. Lasing at the frequency corresponding to the sodium D_2 line is achieved in the presence of pump radiation resonant to the D_1 line with helium as a buffer gas.

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In recent years a growing interest has been attracted to new schemes of laser action in gases. Especially attractive is an opportunity to achive lasing from highly excited levels into the ground state of atoms and molecules because these transitions have the shortest wavelength. On the other hand, the ground electronic state has the highest population that essentially complicates the problem. There are two basic ways to achieve indicated lasing: to create real inversion on transition into the ground state and to use a different kind of coherent effects. Thus the greatest interest is attributed to the opportunity of lasing with a smaller wavelength than that for a pumping radiation.

There are numerous works investigating the amplification of radiation in alkali-metal vapors on the transition into the ground state in the V scheme. Some of them exploit effects of coherence to suppress absorption on the transition from the ground state to the appropriate upper level while the upper level is slightly populated by various methods, such as discharge current [1] and collisions with buffer-gas particles [2]. Another way is connected with the process of parametric generation on the basis of four-wave mixing [3–6].

In this paper we aim to draw attention to the essential expansion of opportunities to create inversion and reach laser action on transitions into the ground state. The method is based on manipulation with the polarization of pump waves and a specific collisional population transfer mechanism.

In earlier papers [7,8] the possibility of laser action was demonstrated at the frequency corresponding to the D_1 line in the presence of a strong pump field tuned in resonance with the D_2 line in alkali-metal vapors. The effect is generated by collisions with buffer-gas particles. The collisions should be frequent enough in order to establish a Boltzmann distribution of population between fine-structure components ($P_{3/2}$ and $P_{1/2}$) during pump pulse action. At these conditions magnetic sublevel populations of the $P_{1/2}$ state appear by the Boltzmann factor higher than that of the $P_{3/2}$ state. A strong pump field is used to equalize magnetic sublevel populations of the ground state $S_{1/2}$ and the resonant upper state $P_{3/2}$. As a result, the population of level $P_{1/2}$ appears to be higher than the population of level $S_{1/2}$. Thus, the population inversion on the transition from the upper state $P_{1/2}$

into the ground state $S_{1/2}$ is achieved, and laser action at the frequency of the D_1 line remains possible. In Refs. [7,8] lasing reached the superluminosity regime.

In correspondence with the nature of the process the generated frequency in Refs. [7,8] was lower compared to the pump frequency (down-conversion). This naturally leads to the question whether up-conversion is possible, i.e., to achieve laser action (on the D_2 line) with a frequency higher than that for the pump field (resonant to the D_1 line)? The present paper gives a positive answer. Such an opportunity appears if one combines the above-mentioned collisional processes with specific polarization effects. Let us prove this statement. Consider an interaction of the pump pulse (specifically polarized) with a gas of atoms in a mixture with a buffer gas at a high pressure. Let us take for definitness sodium atoms with a corresponding level scheme, see Fig. 1. The pump pulse with a carrier frequency resonant to the D_1 line has the following temporal and polarization structure. It consists of a long low-intensity circularly polarized prepulse and a much more intensive and short main pulse with orthogonal circular polarization. The prepulse is used for optical orientation of sodium atoms in the ground state. From this one can set the requirement on its duration (it has to be longer compared to upper state relaxation time) and intensity limit (it may be low, but enough for optical orientation only). After the end of the prepulse almost all the population is optically pumped into one of the magnetic sublevels of the ground state [its population is shown in Fig. 1(a) by symbolic column]. The main pulse may be shorter than the relaxation time of the excited levels. It transfers the population from the ground state onto the sublevel of the excited level $P_{1/2}$ that is initially empty [the corresponding transition is
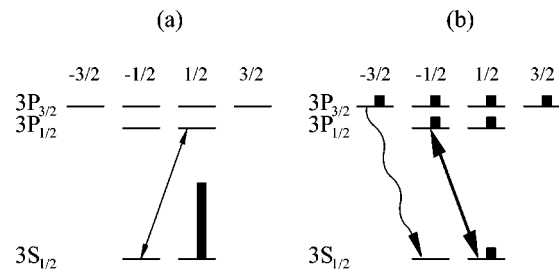

FIG. 1. Relevant energy levels of Na and optical transitions (quantization axis is collinear to wave vectors).

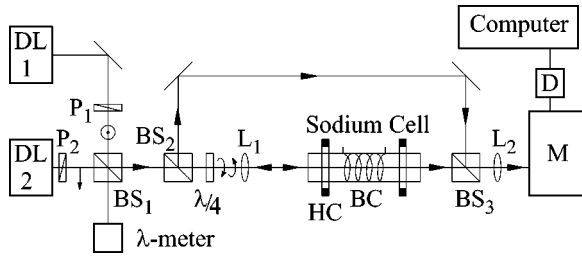*Electronic address: fractal@iae.nsk.su

FIG. 2. Schematic of the experiment to observe superluminosity on the sodium D_2 line. DL, dye lasers; P, polarizers; L, lenses; HC, Helmholtz coils; BC, bifilar heater coil; BS, beam splitters; M, monochromator; D, detector.

FIG. 3. Output lasing signal around the D_2-line frequency (ω_{D_2}) viewed from opposite directions. The forward signal on the D_2 line (a) is 80 times higher then the backward signal (b). Curves 1 [in (a) and (b)] show the output signal of the D_2 line with D_1 pumping for 810 Torr helium pressure, $T = 495$ K, $B = 80$ G. Curves 2 [in (a) and (b)] show output signal of the D_2 line at the same condition but for $B = 0$. Pump beam detunings from the D_1 resonance are equal to zero.

shown in Fig. 1(b) by a solid arrow]. The intensity of the main pulse must be high enough to maintain the equality of populations for the coupled sublevels. Furthermore, at high buffer-gas pressure collisions are frequent enough to mix the states $P_{1/2}$ and $P_{3/2}$ and their magnetic sublevels as well. As a result the population is distributed between the sublevels almost equally, as shown in Fig. 1(b) by small columns. The sublevel $M = -1/2$ of the ground state is almost empty (remember that it is pumped out by the prepulse), whereas other sublevels of the ground state and excited states are populated almost equally (the population difference by the Boltzmann factor between $P_{1/2}$ and $P_{3/2}$ states is insignificant here and may be neglected). One can see that population inversion between some upper magnetic sublevels (including those for the $P_{3/2}$ level) and the sublevel $M = -1/2$ of the ground state is created. Hence, necessary conditions for laser action, specifically from the level $P_{3/2}$ to the ground state $S_{1/2}$, are met.

Thus, we have shown that there exist conditions for producing laser action on the D_2 line with pulsed excitation resonant to the D_1 line. Taking into account partial oscillator strengths, we see that the highest gain is achieved on the transition $P_{3/2}(M = -3/2) \rightarrow S_{1/2}(M = -1/2)$, as shown in Fig. 1(b) by a wavy arrow. It means that the generated wave has presumably the same polarization as the main pump pulse. To avoid misunderstanding, we should remind the reader that collisions with particles of a nonmagnetic buffer gas (noble gas as an example) mix magnetic sublevels of the ground state in alkali metals very weakly, so we neglect this factor with confidence.

On the basis of the above treatment, we have experimentally realized such a scheme with population inversion on transition $3P_{3/2}$-$3S_{1/2}$ of sodium. A general schematic of our experimental setup is shown in Fig. 2. In the experiment it was easier and more convenient to use cw radiation instead of the prepulse. In this case it maintains the orientation of the ground state between pump pulses, coming from another laser source. We used a cw dye laser DL1 with linear vertical polarization (of ≈ 3-GHz spectral linewidth) and pulsed dye laser DL2 with horizontal polarization (of 10-GHz linewidth and ≈ 5-ns pulse duration). In the spectrum of the pulsed laser together with narrow-band laser radiation, a broad band luminescence of the R6G dye is present, but its spectral density is by 3 orders of magnitude weaker.

After passing the quarter-wave ($\lambda/4$) plate the beam of the cw laser becomes clockwise polarized, and the beam of
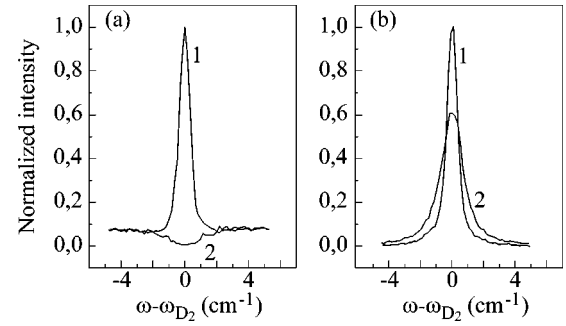
the pulsed laser is counterclockwise polarized. The cw radiation prepares the medium by converting it into the $S_{1/2}(M = 1/2)$ state, and high-power pulses are used for populating upper levels. Both beams propagate in the same direction and are focused by the lens L_1 with focal length $F = 55$ cm into the cell with a sodium-helium mixture. The intensity near the beam waist is ≈ 60 W/cm^2 for the cw laser, and up to 6 MW/cm^2 for the pulsed laser. The cell has a 1.5-cm diameter and is 22-cm long with a heated zone (BC) of 4.5 cm in the central part. The sodium vapor density is controlled by varying the temperature, measured by thermocouple. The cell is placed between the Helmholz coils (HC) that provide an external longitudinal magnetic field to eliminate the deorientating effect of the transverse component of the laboratory field. The magnitude of the external field up to $B \approx 80$ G is available. Output radiation is focused by the lens L_2 onto the slit of the monochromator RAMANOR HG.2S (M) with an apparatus width of about 0.5 cm^{-1}. Data from the photomultiplier D connected to the amplifier and integrator are registered with a computer. That allows us to store and average measured data. The generation at the D_2-line frequency was measured in the direction of the pump beam as well as in the opposite direction. For this purpose the beam splitter (BS_2) was inserted into the pump beam pathway, as shown in Fig. 2.

First of all, it has been ascertained that in the absence of the external magnetic field and at low buffer-gas (helium) pressure there is no coherent radiation at the D_2-line frequency in a broad range of other experimental parameters. After the external magnetic field $B > 0.5$ G is applied, an intense coherent radiation at the D_2-line frequency appears at a helium pressure higher than 200 Torr with cw and pulse lasers are tuned to exact resonance with the D_1 line, see curve 1 in Fig. 3(a). Divergence of the output beam appears to be no more than that of the pump beam. Registered spectral width is about 0.7 cm^{-1}, being close to the apparatus width. The radiation at the D_2-line frequency has nearly the same polarization as the strong pulsed field. Curve 2 in Fig. 3(a) illustrates the measurement without magnetic field (B

=0). In this case only the absorption line is observed because broad band R6G dye luminescence is absorbed in optically thick media. The coherent radiation at the D_2-line frequency is observed both in the direction of the pump beam and in the opposite direction [Fig. 3(b)]. The spectral width for the forward and backward output radiations is nearly the same. In contrast to the forward radiation, the backward one is also observable in the absence of the external magnetic field. Note that the intensity of the forward output radiaton is 80 times as high as that of the backward radiation (in the presence of the magnetic field). This fact can be explained by the effect of dye luminescence that serves as a seed leading to amplification in its direction.

It is interesting to note that the backward generation occurs in the absence of the longitudinal magnetic field, i.e., when the optical orientation is noticeably destroyed. We suppose that this happens due to a sufficient intensity of the cw radiation that is able to transfer the residual population from sublevel $S_{1/2}$ $M=-1/2$ into excited states, thus helping to create inversion on the operating transition. The absence of forward generation under identical conditions is most likely explained as follows. The cw radiation gets absorbed propagating along the heated zone. Because of this, the inversion condition is no longer valid in the output part of the zone. Hence, the generated radiation coming forward is absorbed in the output part.

The output intensity of generated radiation under fixed other conditions reaches its maximum when frequencies of both lasers (cw and pulsed) ω_L are tuned in resonance with the D_1 line of frequency ω_{D_1}. Detuning from the exact resonance $|\Omega|=|\omega_L-\omega_{D_1}|\approx 4$ cm^{-1} leads to the disappearance of the output signal. Appearing at a helium pressure of 200 Torr, the intensity at the D_2-line frequency rises monotonically with increasing pressure up to 810 Torr, the highest available in the experiment. The maximum of output intensity measured as a function of sodium vapor density is reached at $N\approx 8\times 10^{12}$ cm^{-3}. Under these conditions the cell transmission is $\approx 90\%$ for the pulse radiation, and $\approx 80\%$ for the cw laser beam.

Starting at an external magnetic field strength B as low as 0.5 G (comparable to the laboratory field), the output signal grows almost linearly with increasing field up to $B\approx 5$ G when it saturates. Note that an application of the external magnetic field considerably helps to orientate sodium atoms by circularly polarized cw radiation. The luminescence intensity is attenuated by a factor of $3\div 4$ after the field is applied, which means the portion of oriented atoms reaches more than 80%.

When the pump intensity is attenuated down to $1.5\div 2$ MW/cm^2 the output intensity does not change significantly. Further attenuation of the pump radiation leads to a smooth decrease in the generated power while its spectral width remains constant. At the same time, attenuation of the cw laser power leads to an abrupt fall in the generated power at the D_2-line frequency. At optimal conditions ($\Omega=0$, $N\approx 10^{13}$ cm^{-3}, helium pressure ≈ 810 Torr, $B\approx 5$ G) the output intensity of generated radiaton reaches $1.5\div 2\%$ of the absorbed pump intensity (of the pulsed laser). The intensity of
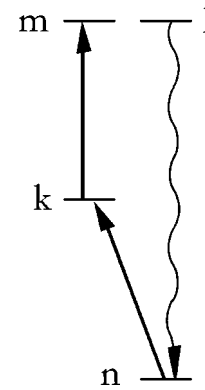

FIG. 4. Elucidation of the posibility of short-wave lasing.

the generated radiation inside the cell is estimated to be ~ 5 kW/cm^2, corresponding to strong saturation, $æ \gg 1$.[1] This suggests that the generation occurs in the superluminosity regime (at least in the forward direction), i.e., the generated wave is capable of utilizing a considerable portion of inversion on the operating transition.

We also have considered parametric processes such as wave mixing as an alternative interpretation of our experimental results. It is well known that wave-mixing processes are suppressed at the resonance conditions due to absorption [9], while our signal conversely has a maximum at the frequency resonant to the D_2 line and vastly decays with detuning from the resonance, and, moreover, the signal generated in the wave mixing in the backward direction need not be present due to phase mismatching [5,4]. Thus our original interpretation presented above seems to be more realistic and reasonable.

Our work is based on the composition of well-known phenomena (laser excitation, optical pumping, collisional population transfer), but we need to stress that only the appropriate combination of the phenomena mentioned above lead to the results that were obvious beforehand.

Thus we have shown in the present work that the proposed combination of polarization and collisional transfer mechanisms opens new opportunities for resonant radiative processes. In view of the development of the general method applied here for a special case, we propose a variant of the scheme that allows one to achieve the generation of violet and UV radiation on transitions into the ground state. For this purpose, instead of a single-photon process a two-step or two-photon excitation into higher-lying states can be explored (see Fig. 4). In this case, radiative transition from the excited state (m) into the ground state is parity forbidden. However, if there is another close (within kT range) level l of different parity, coupled radiatively with the ground state n, the developed method remains applicable. Using high-pressure buffer gas an efficient collisional mixing of upper

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levels m and l can be provided. Then, due to the Boltzmann factor (if level l is lower than level m) and high intensity pump waves, a population inversion on transition l-n can be achieved, thus resulting in laser action in a short-wave range. In case the available intensity and the Boltzmann factor are not enough for generation, or level l lies higher than level m, it is possible to prepare the system using the same polarization technique: optical orientation of the ground state n by resonant radiation (at first-step transition) opens an addi-tional opportunity to build up population inversion between corresponding sublevels of transition $l-n$.

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