Proceedings of the Conference "From Quantum Optics to Photonics", Zakopane 2001

Population Inversion on Transition into the Ground State of Atoms Induced by Optical Excitation and Collisions

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Lasing on the sodium resonance transitions (D_1 and D_2 lines) at the superluminosity regime was observed upon the non-resonance excitation in the presence of a buffer gas. Dependences of the lasing intensity on the pump radiation intensity and its frequency detuning from the frequencies of resonance transitions were examined. It was found that under the specific experimental conditions (high buffer gas pressure, sufficiently high intensity of pump radiation) upon the large positive frequency detuning of pump radiation with respect to the frequency of resonance ("working") transition, contrary to ingrained conceptions, population inversion for the "working" transition is raised. That results in the observed phenomena.

PACS numbers: 42.50.Hz, 32.90.+a, 42.50.Ct

1. Introduction

Collision induced processes in a gas medium have a noticeable influence on the redistribution of the quasiresonance radiation. Study of these processes is important from the fundamental and practical point of view. In particular, it is to open new ways for lasing and to extend its frequency range. The opportunity of laser action on the transition into the ground state or into the lowest states that are extremely populated without perturbation is especially of interest. It is the opportunity that prospects the radiation with the shortest possible wavelength.

The possibility of laser action in alkali-metal vapors in mixture with buffer gas was demonstrated at the frequency corresponding to the D_1 line of potassium [1], sodium [2], and rubidium [3]. This phenomenon was observed upon resonance excitation at the frequency corresponding to the D_2 line and was induced by frequent collisions with buffer gas particles. The collisions should be frequent enough in order to establish Boltzmann distribution of population between fine-structure components ($P_{3/2}$ and $P_{1/2}$) during excitation pulse and radiative relaxation time. At these conditions population of the $P_{1/2}$ state becomes greater by Boltzmann factor than that of the $P_{3/2}$ state. An intensity of pump field was so high as to equalize populations of the ground state ($S_{1/2}$) and the resonant upper state ($P_{3/2}$). As a result, the population of level $P_{1/2}$ becomes greater (by the same Boltzmann factor) than the population of both the levels $P_{3/2}$ and $S_{1/2}$. In this way, the population inversion on 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 occurs. In the experiments [1, 2] lasing was observed at a superluminosity regime.

In the next work [4] we obtained laser action on the resonance transitions through appropriate polarization conditions and using the collisions with buffer gas particles. In this experiment we used a rather high power laser setup that allows us to vary experimental conditions within much more extended range. During these experiments we have unexpectedly found that the coherent radiation at the D_1 appears not only at the frequency of pump radiation tuned to the resonance with D_2 line but also at the frequency remarkably shifted (up to 250 cm^{-1}) to the high frequency side. Our paper presents the results of an experimental investigation of this effect and proposes its physical interpretation.

2. Experimental setup

The experimental setup is shown in Fig. 1. We used a tunable dye (R6G) laser (DL) pumped by second harmonic of Nd^{3+} YAG laser radiation. The DL has pulse power 15 kW at pulse duration of 5 ns, linear polarization, a tuning range from 575 to 595 nm and repetition frequency of 10 Hz. The spectrum of DL radiation consisted of power narrow laser line (with width of 0.3 cm^{-1}) and a broad band weak luminescence of the R6G dye. The spectral density of luminescence is

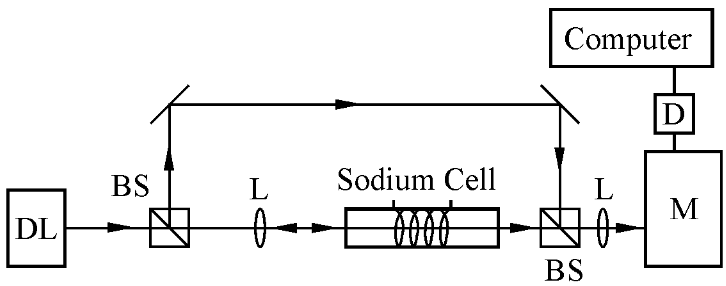

Fig. 1. Experimental setup. DL — pulsed dye laser, BS — beam splitter, L — lens, M — monochromator, D — photomultiplier.

by three orders of magnitude smaller than that for narrow laser line. The lens with focal length 55 cm focuses the laser radiation into the center of heated cell. The intensity near the beam waist is ≈ 10 MW/cm^2 and could be changed by neutral filters. The cell with sodium vapor has a 1.5 cm diameter and is 22 cm long with a heated zone of 5 cm in the central part. The sodium vapor density ($N \approx 10^{14}$ cm^{-3}) is controlled by varying the temperature, measured by thermocouple. The pressure of a buffer gas (helium) is varied from 10 to 800 Torr.

Output radiation is focused by the lens onto the slit of the monochromator RAMANOR HG.2S (M) with an apparatus width about 0.5 cm^{-1}. Data from photomultiplier (D) connected with amplifier and integrator are registered with a computer. That allows us to store and average measured data. Monochromator is calibrated by sodium lamp. The lasing at the D_1 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) was inserted into the pump beam pathway, as shown in Fig. 1.

3. Experimental results

Firstly, the aim of our experiments was more detailed study of coherent radiation on the frequency of sodium D_1 line (transition $3P_{1/2}$–$3S_{1/2}$) in the forward and backward direction with respect to the pump radiation. The frequency of pump radiation was tuned in the area of sodium D lines.

It has been ascertained that in the absence of buffer gas there is no coherent radiation at the sodium D_1 line frequency in a broad range of other experimental conditions.

The coherent radiation at the sodium D_1 line frequency was observed at a buffer gas pressure $p_{\rm He} > 200$ Torr, sodium vapor density $N \approx 10^{14}$ cm^{-3}, pump radiation intensity $I_{\rm L} \approx 1$–2 MW/cm^2 while pump radiation frequency was varied in the region close to the D_2 line. Divergence of the output beam turns out to be similar to that of the pump beam. The coherent radiation at the sodium D_1 line frequency was observed not only in the direction of the pump field but also in the opposite direction.

At the conditions similar to those of [2] similar results were obtained. Namely, the threshold value of helium pressure is equal to 200 Torr. The lasing at the D_1 line steadily increases with helium pressure increasing up to 800 Torr. The coherent radiation at the D_1 line reaches its maximum when the pump radiation is tuned to the resonance with the frequency ω_{D_2} corresponding to sodium D_2 line ($3P_{3/2}$–$3S_{1/2}$ transition). The optimal sodium vapor density is equal to $\approx 2 \times 10^{14}$ cm^{-3} while the pump intensity is about 3 MW/cm^2.

Under the optimal conditions power of the lasing on the D_1 line was $\approx 3.5\%$ in comparison with that of pump radiation (forward direction case). Within the length of heated area $L \approx 5$ cm enhancement coefficient is $\alpha \approx 3 \div 3.5$ cm^{-1}. Therefore, factor αL reaches the value $15 \div 18$. Taking into account that broad band luminescence is used as a seed, it is the superluminosity regime. Appearance of the similar lasing in the backward direction confirms the superluminosity regime accomplishment. The intensity of the backward lasing is about several ten times smaller than for the forward one. It is due to the absence of any seed in this direction.

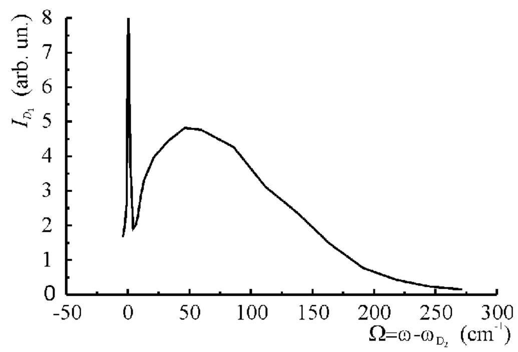

Fig. 2. Dependence of the sodium D_1 line lasing intensity I_{D_1} on pump radiation frequency detuning from D_2 line frequency $\Omega = \omega - \omega_{D_2}$, $I_L \approx 10$ MW/cm^2, $p_{He} =$ 600 Torr, $N \approx 2 \times 10^{14}$ cm^{-3}.

Using the higher pump intensity (up to $I_L \approx 10$ MW/cm^2) the following features are observed (Fig. 2). The sharp peak $I_{D_1}(\Omega)$ is observed when the pump radiation frequency detuning $\Omega = \omega - \omega_{D_2} \approx 0$ is close to zero. The increase in the detuning Ω leads first to decrease in the intensity of generated radiation I_{D_1}, but further increase in the detuning gives rise to unexpected increase in the generated radiation intensity until the detuning value about $\Omega \approx 60$ cm^{-1}. With next increase in Ω the value I_{D_1} decreases but intensity of the lasing on the D_1 line has noticeable value till pump radiation frequency detuning of $\Omega \approx 250$ cm^{-1}. At the same time, when pump radiation has a large detuning in addition to the coherent radiation at the frequency of D_1 line a similar radiation appears at the frequency corresponding to the D_2 line. These generated radiations are observed both in the direction of the pump radiation and in the opposite one. The rate of spatial coherence of generated radiation was examined. It was found that angular spread of the lasing beam turns out to be similar to that of the pump beam. The intensity of generated radiation is strongly depending on the intensity of pump radiation I_L. With I_L decreasing twice the region of detuning Ω with noticeable intensity of generated radiation decreases twice while the maximal lasing intensity decreases 5 times. Lasing on sodium D lines disappears at $I_L \approx 3$ MW/cm^2. The important point is that coherent radiation on the resonance transitions is absent when frequency of pump radiation is smaller than frequency corresponding to the sodium D_1 line. Electronic Raman scattering and three photon lines were not observed in our experiments.

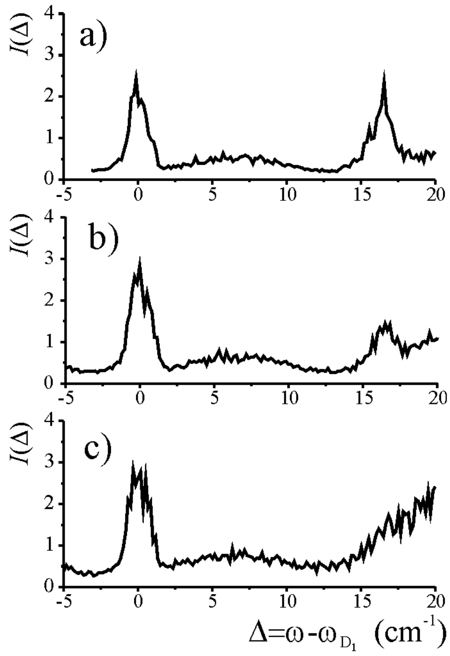

Fig. 3. Spectrograms of generated radiation ($I(\Delta)$) in arbitrary units for the different pump radiation frequency detuning $\Omega = \omega - \omega_{D_2}$, (a) $\Omega \approx 60$ cm^{-1}, (b) $\Omega \approx 33.5$ cm^{-1}, (c) $\Omega \approx 20$ cm^{-1}, $I_{\rm L} \approx 10$ MW/cm^2, $p_{\rm He} = 600$ Torr, $N \approx 2 \times 10^{14}$ cm^{-3}.

Figure 3 represents the characteristic spectra of the generated radiation. Here x-axis corresponds to the frequency of generated radiation and zero value corresponds to the sodium D_1 line. There is coherent radiation both on the D_1 line and D_2 line with approximately equal intensities (two peaks about $\Delta = 0$ and $\Delta = 17.2$ cm^{-1}) at $\Omega = 60$ cm^{-1} (Fig. 3a).

Non-zero signal between peaks is due to weak broadband luminescence of the R6G dye. While the frequency of the pump radiation approaches to the frequency of D_2 line the coherent radiation corresponding to this line disappears. Therefore only lasing on the D_1 line is observed (Fig. 3c). Further decrease in the frequency of the pump radiation down to $\Omega = -5.5$ cm^{-1} leads to disappearance of the lasing on the D_1 line as well. Increase in the signal at a high frequency side (Fig. 3c) is due to parasitic light scattering of the pump radiation.

4. Population inversion in two-level system under the continuous excitation

Usual theoretical models do not account for this novel result of observation of coherent radiation on the resonance lines of sodium with a quite non-resonance excitation. Therefore this result needs a particular analysis.

Appearance of the enhanced coherent radiation both on the transition $3P_{1/2}$–$3S_{1/2}$ and $3P_{3/2}$–$3S_{1/2}$ with extremely non-resonance excitation allows to suppose that this phenomenon is associated with a simple two-level system. Taking into account high buffer gas pressure used in the experiments pulsed behavior of pump radiation does not play a principal role: during the pulse action population distribution typical of continuous radiation is established.

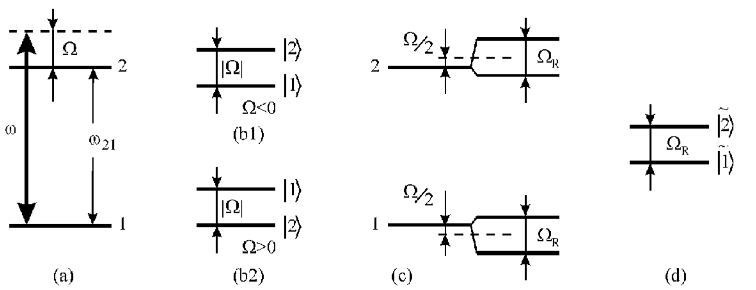

Fig. 4. Energetic level diagrams for different bases described in the text.

In this way, the problem is as follows. A gas of two-level atoms in mixture with buffer gas of high pressure is affected by continuous laser radiation with frequency far from resonance value. Collisions of atoms with buffer gas particles are elastic, i.e. do not cause the transition between upper and lower levels (2 and 1 in Fig. 4). The question is: is it possible to observe the amplification of radiation at the resonance frequency ω_{21} of the atomic transition? According to standard theory (see for example [5]) population difference under the specified conditions is (Doppler broadening at the high buffer gas pressure is insignificant):

$$N_1 - N_2 = \frac{N}{1+\kappa}, \quad \Omega = \omega - \omega_{21}, \quad \kappa = \frac{4|G|^2 \Gamma}{\Omega^2 \Gamma_2}, \quad G \equiv \frac{d_{21}\mathcal{E}_0}{2\hbar}, \tag{1}$$

where Γ_2 is the constant of radiative decay of excited level, Γ is the collision halfwidth of the absorption line, N is the concentration of resonance atoms, N_1, N_2 is the population of the levels 1 and 2, $\mathcal{E}_0$ is the amplitude of electric field of pump radiation, d_{21} is the dipole moment matrix element for the transition 2–1. Condition $|\Omega| \gg \Gamma$ is used in (1). According to (1) and to ingrained conceptions continuous radiation cannot create population inversion and is able only to equalize populations of levels 1 and 2. In principle, in the presence of nonlinear interference phenomena there are some spectral regions in which amplification on weak radiation without population inversion is possible [6]. However, under the conditions of our experiments these phenomena do not appear. Under the specific conditions (high buffer gas pressure, high intensity of pump radiation and quite non-resonance conditions) Eq. (1) turns out not to be valid and is needed to be corrected.

First of all, it is to be noted that at the high buffer gas pressure effective saturation factor κ from (1) can be high even if $|\Omega| \gg \Gamma, |G|$. It is due to large ratio Γ/Γ_2 (collisions do not cause the decay of level 2, but increase the Γ value). Absorption line possesses a great field broadening despite that value $|G|$ can be not so large with respect to Γ.

Let us consider the influence of "elastic" (with respect to the optical transition 2–1) collisions on the optical transitions and populations of levels 1 and 2. Invalidity of standard consideration is easily to show in the framework of compound systems [7]. Therefore, we will follow this approach [7].

Let us specify Hamiltonian

$$\boldsymbol{H} = \boldsymbol{H}_{\mathcal{A}} + \boldsymbol{H}_{\mathcal{E}} + \boldsymbol{V}_{\mathcal{A}\mathcal{E}} + \boldsymbol{U}(t). \tag{2}$$

Here $\boldsymbol{H}_{\mathcal{A}}$ — Hamiltonian of considered two-level atoms; $\boldsymbol{H}_{\mathcal{E}}$ — Hamiltonian of the radiation field; $\boldsymbol{V}_{\mathcal{A}\mathcal{E}}$, $\boldsymbol{U}(t)$ — operators of interaction of particles $\mathcal{A}$ with radiation and buffer particles. Operator $\boldsymbol{U}(t)$ is a series of separated pulses randomly distributed in timescale, but time dependence of each pulse is fixed. This approach of $\boldsymbol{U}(t)$ corresponds to quasi-classical scattering.

Within the framework of resonance two-level approach operator $\boldsymbol{H}_{\mathcal{A}}$ is referred to atomic states 1 and 2 (Fig. 4a). Operator $\boldsymbol{H}_{\mathcal{A}} + \boldsymbol{H}_{\mathcal{E}}$ is referred to states depicted in Fig. 4b1,b2

$$|1\rangle \equiv |1, n\rangle, \quad |2\rangle \equiv |2, n-1\rangle, \tag{3}$$

where n is the number of photons in radiation field. These states are states of compound system (atom + field) without interaction between them. Their relative positions in energy scale depend on the sign of Ω: with negative Ω state $|1\rangle$ possesses a lower energy than $|2\rangle$ state, with positive Ω the case is opposite.

Operator $\boldsymbol{H}_{\mathcal{A}} + \boldsymbol{V}_{\mathcal{A}\mathcal{E}}$ does not possess stationary states, but its states (quasi--energy levels) are frequently used to illustrate dynamic Stark effect. Corresponding diagram is shown in Fig. 4c. Finally, operator $\boldsymbol{H}_{\mathcal{A}} + \boldsymbol{H}_{\mathcal{E}} + \boldsymbol{V}_{\mathcal{A}\mathcal{E}}$ is referred to states $|\tilde{1}\rangle$, $|\tilde{2}\rangle$ (Fig. 4d). In the absence of other perturbations these states are stationary and sometimes called "dressed" states. In particular, in the basis of $|1\rangle$ and $|2\rangle$ states arbitrary wave function can be expressed as

$$\Psi = \mathrm{e}^{-\mathrm{i}\frac{E_1}{\hbar}t - \mathrm{i}n\omega t}\left(a_1|1\rangle + a_2|2\rangle \mathrm{e}^{\mathrm{i}\Omega t}\right), \tag{4}$$

where a_1 and a_2 — corresponding amplitudes of the states. The Schrödinger equation with Hamiltonian (2) gives

$$\frac{\mathrm{d}}{\mathrm{d}t}a_1 = iG^*\mathrm{e}^{\mathrm{i}\Omega t}a_2 - \frac{\mathrm{i}}{\hbar}U_{11}a_1, \quad \frac{\mathrm{d}}{\mathrm{d}t}a_2 = iG\mathrm{e}^{-\mathrm{i}\Omega t}a_1 - \frac{\mathrm{i}}{\hbar}U_{22}a_2,$$

$$U_{ii} = \langle i|\boldsymbol{U}|i\rangle. \tag{5}$$

Here, only diagonal matrix elements of operator of interaction with buffer particles are not equal to zero. In the absence of interaction $\boldsymbol{U}$ solution of (5) is well known

$$a_1^0 = \mathrm{e}^{\mathrm{i}\frac{\Omega}{2}t}\left(A_1\mathrm{e}^{\mathrm{i}\frac{\Omega_R}{2}t} + A_2\mathrm{e}^{-\mathrm{i}\frac{\Omega_R}{2}t}\right),$$

$$a_2^0 = \mathrm{e}^{-\mathrm{i}\frac{\Omega}{2}t}\left(A_1\frac{2G}{\Omega_R-\Omega}\mathrm{e}^{\mathrm{i}\frac{\Omega_R}{2}t} - A_2\frac{2G}{\Omega_R+\Omega}\mathrm{e}^{-\mathrm{i}\frac{\Omega_R}{2}t}\right),$$

$$\Omega_R \equiv \sqrt{\Omega^2+4|G|^2}. \tag{6}$$

Here A_1 and A_2 — arbitrary coefficients determined by initial conditions. Corresponding wave function Ψ^0 can be expressed as

$$\Psi^0 = \mathrm{e}^{-\mathrm{i}\frac{E_1}{\hbar}t - \mathrm{i}n\omega t + \mathrm{i}\frac{\Omega}{2}t}\left(\tilde{a}_1^0|\tilde{1}\rangle\mathrm{e}^{\mathrm{i}\frac{\Omega_R}{2}t} + \tilde{a}_2^0|\tilde{2}\rangle\mathrm{e}^{-\mathrm{i}\frac{\Omega_R}{2}t}\right),$$

$$\tilde{a}_1^0 = \frac{A_1}{b_1}, \quad \tilde{a}_2^0 = \frac{A_2}{b_2^*}, \quad b_1 = \frac{1}{\sqrt{2}}\sqrt{1-\frac{\Omega}{\Omega_R}}, \quad b_2 = \frac{G}{|G|}\frac{1}{\sqrt{1+\frac{\Omega}{\Omega_R}}}. \tag{7}$$

New wave functions $|\tilde{1}\rangle$ and $|\tilde{2}\rangle$ are introduced here. These wave functions form orthonormal basis and relate to $|1\rangle$ and $|2\rangle$ by

$$|\tilde{1}\rangle = b_1|1\rangle + b_2|2\rangle, \quad |\tilde{2}\rangle = b_2^*|1\rangle - b_1|2\rangle, \quad |b_1|^2+|b_2|^2 = 1. \tag{8}$$

The matrix of inverse transformation is equal to the transformation matrix (8). The states $|\tilde{1}\rangle$ and $|\tilde{2}\rangle$ are stationary according to (7), and their energetic levels correspond to Fig. 4d. In this way, these states are stationary states of compound system with Hamiltonian $\boldsymbol{H}_{\mathcal{A}} + \boldsymbol{H}_{\mathcal{E}} + \boldsymbol{V}_{\mathcal{A}\mathcal{E}}$. The state $|\tilde{1}\rangle$ is lower than state $|\tilde{2}\rangle$ in energy scale independently of the sign of Ω. Wave function corresponding to arbitrary Hamiltonian can be decomposed in basis of wave functions of compound system. Let us introduce probability amplitudes $\tilde{a}_1$ and $\tilde{a}_2$ instead of $\tilde{a}_1^0$ and $\tilde{a}_2^0$. Let us consider the equations based on the Schrödinger equation with whole Hamiltonian (2):

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\tilde{a}_1 = \tilde{U}_{12}\tilde{a}_2\mathrm{e}^{-\mathrm{i}\Omega_R t} + \tilde{U}_{11}\tilde{a}_1,$$

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\tilde{a}_2 = \tilde{U}_{21}\tilde{a}_1\mathrm{e}^{\mathrm{i}\Omega_R t} + \tilde{U}_{22}\tilde{a}_2, \quad \tilde{U}_{ij} = \langle\tilde{i}|\boldsymbol{U}|\tilde{j}\rangle. \tag{9}$$

Taking into account relations (8) for matrix elements of $\tilde{U}_{ij}$ we obtain the following equations:

$$\tilde{U}_{21} = \frac{G}{\Omega_R}\Delta U, \quad \tilde{U}_{12} = \tilde{U}_{21}^*, \quad \tilde{U}_{11} = \overline{U} - \frac{\Omega}{2\Omega_R}\Delta U, \quad \tilde{U}_{22} = \overline{U} + \frac{\Omega}{2\Omega_R}\Delta U,$$

$$\overline{U} = \frac{U_{11}+U_{22}}{2}, \quad \Delta U = U_{11} - U_{22}. \tag{10}$$

When operator $\boldsymbol{U}$ corresponds to interaction including collisions with buffer gas particles, interpretation of the obtained result is as follows. In the basis of unperturbed atomic states $\mathcal{A}$ (Fig. 4a) in the absence of radiation, collisions do not cause the transitions between states 1 and 2, i.e. collisions are "elastic". With vanishing radiation intensity collisions remain elastic in the basis of $|\tilde{1}\rangle$ and $|\tilde{2}\rangle$ states (see (9) and (10)). Non-vanishing intensity of radiation leads to appearance of collisional transitions between states $|\tilde{1}\rangle$ and $|\tilde{2}\rangle$ ($\tilde{U}_{21} \neq 0$). Therefore, collisions are inelastic with energy gap $\hbar\Omega_R$. Elastic channel of scattering is also changed. It is to be noted that matrix element $\tilde{U}_{21}$ of interaction potential causing the inelastic transition contains both properties of initial collisional operator and properties of radiation. It means that the quantum of radiation field participates in the relevant collision event. These collisions are named optical collisions [7]. According to [7] optical collisions are the main way of changes of energy of radiation under the condition $|\Omega| \gg \Gamma$, in contrast to condition $|\Omega| \lesssim \Gamma$ when changes of radiation energy occurs mainly during particle free path. In other words, radiative processes that change atomic state under the condition $|\Omega| \gg \Gamma$ take place mainly during the collisions. It is the most important point for further consideration.

In a weak field ($|G| \ll |\Omega|$) radiative processes $|\Omega| \gg \Gamma$ are clearly demonstrated in the basis of unperturbed atom (Fig. 5a). If atom is in the state 1 then during collision atom can absorb photon and became in the state 2. In this situation, energy excess (or deficit) $\hbar\Omega$ is compensated by energy of translational movement of collided particles (Fig. 5a). The case of induced emission is similar when atom was primarily in the state 2 (Fig. 5b).

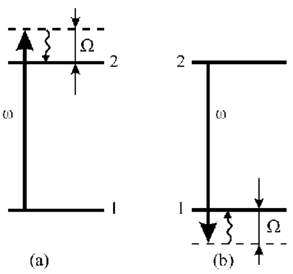

Fig. 5. Optical collisions in the basis of unperturbed atomic states. (a) Act of photon absorption $\hbar\omega$, (b) act of induced emission of photon.

In strong field really stationary states are $|\tilde{1}\rangle$ and $|\tilde{2}\rangle$ states of compound system. For these states collisions and interaction with radiation are characterized by interaction operator $\boldsymbol{U}$ with matrix elements (10). Kinetic equation with collision integral for the density matrix of this "dressed" atom can be found in a regular way. Behavior of act of collision is determined by the operator $\boldsymbol{U}$ including both elastic and inelastic channels of scattering. Naturally, all general conclusions of kinetic theory are valid in the considered case. In particular, general conclusion of thermodynamics concerning the Boltzmann distribution establishment in consequence of collisions is also valid. For our case it leads to population of state $|\tilde{2}\rangle$ being less than population of $|\tilde{1}\rangle$ by the Boltzmann factor $\exp(-\hbar\Omega_R/k_{\mathrm{B}}T)$.

Let us return to Eq. (8) that relates states $|\tilde{i}\rangle$ and $|i\rangle$. With positive detuning and $4|G|^2 \ll \Omega^2$ we have

$$|\tilde{1}\rangle \simeq |2\rangle\,, \quad |\tilde{2}\rangle \simeq \frac{G}{|G|}|1\rangle. \tag{11}$$

Thus upper level of compound system corresponds to lower atomic level while lower level of compound system corresponds to upper atomic level. Therefore, with large positive pump radiation frequency detuning ($|\Omega| \gg |G|, \Gamma$) population of upper atomic level $|2\rangle$ becomes greater than population of lower atomic level $|1\rangle$ by Boltzmann factor $\exp(\hbar\Omega/k_{\rm B}T)$ due to optical collisions

$$|a_2|^2 = \exp(\hbar\Omega/k_{\rm B}T)|a_1|^2. \tag{12}$$

In the opposite case ($\Omega < 0, |\Omega| \gg |G|, \Gamma$)

$$|\tilde{1}\rangle \simeq |1\rangle, \quad |\tilde{2}\rangle \simeq -|2\rangle. \tag{13}$$

In this case population of lower atomic level is greater than upper one. Equation (12) is valid both for positive and negative detunings Ω. When gas (thermostat) temperature is high, populations of states $|1\rangle$ and $|2\rangle$ become equal. Results specific for widely used standard approach turn out to be valid only under the condition

$$\hbar|\Omega| \ll k_{\rm B}T. \tag{14}$$

Results of paper [7] and previous works concerning optical and radiative collisions are also limited by the condition (14).

The obtained result can be clearly demonstrated in the basis of unperturbed atomic states (Fig. 5). With large positive detuning of pump radiation Ω during the absorption act, energy excess is transferred to the environment while in order to allow atom to pass from state 2 to state 1 during the act of induced emission energy deficit must be taken from environment. The number of particles that allow induced emission is by factor $\exp(-\hbar\Omega/k_{\rm B}T)$ smaller than the number of particles that participate in the absorption process. As a result population of state 2 becomes greater than population of state 1, i.e. population inversion appears.

5. Discussion

The carried out analysis is made neglecting the relaxation of levels 1 and 2 in order to clearly show the appearance of population inversion. Taking into account relaxation time $1/\Gamma_2$ of level 2 (level 1 is considered as ground state) the condition to observe population inversion is that during relaxation time a large number of optical collisions should take place. The rate of optical transitions caused by optical collisions is $2|G|^2\Gamma_{\rm OC}/\Omega^2$ [7], where $\Gamma_{\rm OC}$ — rate of phase relaxation upon the optical collisions. To estimate it one can assume that $\Gamma_{\rm OC} \approx \Gamma$, where Γ — collisional broadening. For the phenomena to appear significantly the following condition should be accomplished:

$$\frac{2\,|G|^2\,\Gamma}{\Omega^2 \Gamma_2} \gg 1, \tag{15}$$

that means high value of the saturation factor of Eq. (1).

Under our experimental conditions with helium pressure $p_{\mathrm{He}} = 600$ Torr using data of sodium D_1 line broadening in helium atmosphere [8] one can calculate $\Gamma \approx 10^4$ MHz. The value of Γ_2 is equal to the first Einstein coefficient for the transition $P_{1/2}$–$S_{1/2}$ (see for example [9]) $\Gamma_2 = 10$ MHz. Therefore, in our experimental conditions $\Gamma/\Gamma_2 \approx 10^3$. To estimate value $|G|$ one can use equation [5] for the transition $P_{1/2}$–$S_{1/2}$ and linear polarization of radiation

$$|G| = 0.334 \times 10^{-2}\sqrt{\frac{1}{3}\bar{\lambda}\bar{S}f}\ [\mathrm{cm}^{-1}],$$

where $f = 0.33$ — oscillator strength of the transition $S_{1/2} \rightarrow P_{1/2}$, $\overline{\lambda}$ — wavelength of radiation in μm, $\overline{S}$ — density of energy flow W/cm^2. With Poynting vector $\overline{S} = 10^7$ one obtains $|G| \approx 3$ cm^{-1}. That results in effective saturation factor $\kappa \approx 10$ (1) at $\Omega = 60$ cm^{-1} (detuning value at that phenomenon is maximal). Thus, conditions to observe population inversion in our experiments were fulfilled.

Taking into account that with negative detuning amplified radiation at the frequency corresponding to resonance transition was not observed we can assert that experiments have shown the phenomena discussed in the previous section.

6. Conclusion

This work presents theoretical explanation and experimental confirmation of new phenomenon existence, namely, appearance of population inversion in two-level system under the absorption of non-resonance continuous laser radiation. The phenomenon appears upon high buffer gas pressure and is caused by the so-called optical collisions. Revealed phenomenon points out to limit of widely used physical approaches based on the quantum-kinetic equations for density matrix (see for example [10–12]). Derivation of these equations was made under the assumption that radiation does not participate in the process of collision, while the interaction with radiation takes place only during particle free path. This approach is valid while detuning of radiation frequency with respect to the resonance frequency is not essentially large compared to collisional broadening. Otherwise one should take into account that the time of establishment of "balance" between radiation and quantum system is about $\sim |\Omega|^{-1}$, and to the moment of collision the particle does not remain in the pure atomic state. This fact was taken into account in the theory of optical collisions (see [7] and citation therein), however in these works rather obvious fact of the Boltzmann distribution establishment over the states of compound system, as well as population inversion appearance was not observed. Account of participation of radiation in collisional process should lead to correction of usual kinetic equations for density matrix, based on unperturbed atomic state basis. It is our aim for the next work. First steps in this direction were made in works of Pestov and Rautian [12] (see also [5]). In this work relevant equation was derived in operator form. Further aim is to apply it to specific physical objectives. It is possible that some objectives needs kinetic equations for density matrix based on compound system states basis.

The observed phenomena, as it seems from our point of view, are useful for practical applications as a possibility to achieve coherent radiation within new spectral areas. If one takes into account multiphoton excitation to the upper energy states, it allows one to obtain lasing in short wavelength area using population inversion appearing with respect to the ground state.

Acknowledgments

Authors are grateful to M.G. Stepanov and E.V. Podivilov for the fruitful discussion. This work was supported by Russian Foundation for Basic Research (project 01-02-17433), program of Minnauki "Physics of quantum and wave processes".

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