

Effect of light pressure on transitions from excited states of the Ne atom

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Anomalous behavior of the saturated absorption resonance line shape on the $1s_5-2p_8(J=2-J=2)$ transition in the Ne atom is experimentally observed. The shape of the saturation resonance on transitions with degenerate excited states of atoms is numerically analyzed. The reasons and conditions for the anomalous behavior of the resonance line shape and the formation of its doublet spectral structure are established. The influence of the resonant light pressure force on the amplitude and the frequency properties of the saturated resonance is investigated. It is shown, that the asymmetry of the doublet splitting of the resonance is caused by the effect of resonant light pressure.

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I. INTRODUCTION

Action of the light pressure force under the resonant interaction of laser radiation with atoms results in change of both the spectral characteristics of resonances [1] and the macroscopical parameters of an ensemble [2,3]. Thus, owing to the requirement of cyclicity of the reemission process for the effective influence of a light field on macroscopical parameters of the ensemble, research is carried out with the atomic transitions from the ground state. As far as we know, the problem of the light pressure effect on transitions from the excited states of atoms has not yet been considered.

In the present work we report on detection of the light pressure effect at resonant interaction of light with excited states in the Ne atom exhibited in the line shape of the saturated absorption resonance of the atom. The experimentally observed features of the absorption spectrum are well agreed with outcomes of the model calculations made in view of the action of the light pressure force.

The feature of formation of the saturated absorption resonance form on the examined transition between degenerated states of atoms as the result of summation of four Lorentz-type contours of different widths and different values and signs of amplitudes makes it very sensitive to the change of the velocity distribution function of particles in a small range of speeds $v \sim \Gamma/\kappa$, which is exhibited in the spectrum of resonance at absorption of a small number of photons on transitions and from the excited states of atoms.

II. EXPERIMENTAL SETUP

Experiments were made on the $1s_5-2p_8(^3P_2-^3D_2)$ transition in the Ne²⁰ atom ($\lambda=633.4$ nm). Interest to the given transition is stipulated by the fact that because of a simple structure of energy levels and metastable lower state it represents an example of an optically orientable atomic system. Atoms under the action of the linear polarized pumping radiation are redistributed because of the spontaneous emission on the magnetic sublevels, occupying mainly a level with

$M=0$ of the lower state (scheme of transitions, Fig. 1).

A schematic of an experiment for observation of the saturated absorption resonance in the field of two opposite traveling optical waves in gas medium is presented in Fig. 2. We used a single-mode frequency-stabilized CW DCM dye laser pumped by the power Ar⁺ ion laser radiation [4]. The light beam of the CW dye laser was directed through the polarized prism and beam splitter into the discharge cell with pure Ne²⁰ gas. The gas pressure was about 2×10^{-2} Torr, the discharge current was about 15 mA, and the length of an absorbing layer was 5 cm. The power of a strong pumping field could be varied up to 30 mW, the width of laser radiation line was about 3 MHz. A weak probe field after the $\lambda/2$ plate had linear polarization orthogonal to the strong light field, a power of about 0.1 mW, and was directed opposite to a strong field at an angle of about 5×10^{-3} rad. The absorption line shape of a probe light field was recorded, varying the laser radiation frequency.

Absorption line shapes of the probe light field at various strong field intensities (saturation parameters \mathcal{R}) are shown in Fig. 3. Saturation parameter \mathcal{R} , depending on strong field intensity and atomic level decay rates, will be determined later, but here we point out, that its maximal experimental value reached ~ 500 . It is worthy to note, that the observed absorption spectrum of a probe light field in the presence of a strong counter-propagating light wave of linear orthogonal polarization represented, near the center of the atomic tran-

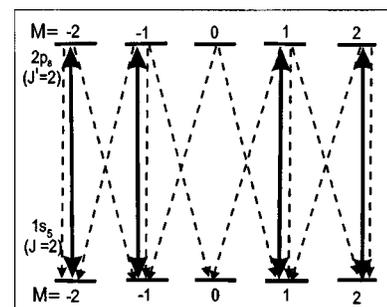


FIG. 1. Diagram of induced and spontaneous transitions between the degenerate lower (n) and upper (m) states of the Ne atom ($J_m = J_n = 2$).

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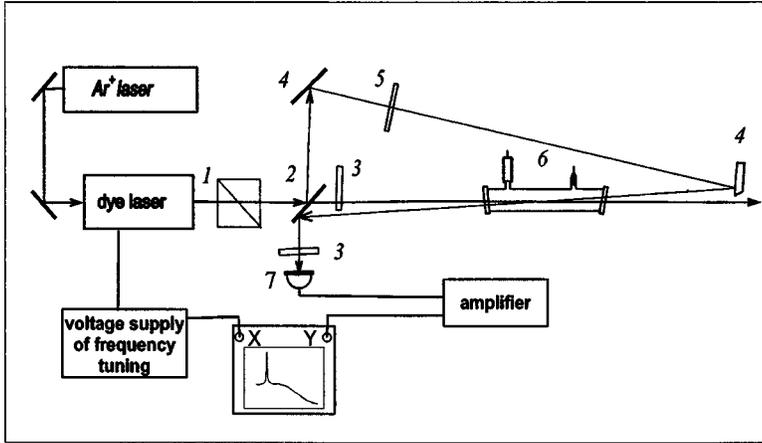


FIG. 2. Schematic diagram of the experimental apparatus: 1, polarized prism; 2, beam splitter; 3, beam absorbers; 4, mirrors; 5— $\lambda/2$ plate; 6, discharge cell, and 7, detector.

sition line, a narrow peak of absorption against the background with a wide base instead of a traditional Lamb-type dip. Increasing a strong field intensity (saturation parameter) raised the peak amplitude and varied the line shape of resonance. At certain saturation parameters of a strong field $\mathcal{R} (\mathcal{R} \geq 50)$ the peak of absorption split into two components with the different amplitudes, and the maximal value of splitting was about ~ 25 MHz at saturation parameter $\mathcal{R} = 500$. The spectral width of the absorption peak, measured at half of the peak amplitude, was about 14–70 MHz when saturation parameter \mathcal{R} varied between 50 and 500.

III. THEORETICAL MODEL

For interpretation of the observed features of the saturated absorption resonance spectrum, the theoretical model was advanced on the basis of the stationary equation for a density matrix in view of stimulated and spontaneous radiative processes on the given transition of the Ne atom and dynamics of the distribution function of particles on the magnetic sublevels of transition.

The problem of interaction of metastable states of atoms with laser radiation is complicated and essentially nonlinear, requiring concrete definition of the atomic level structure and properties of the resonance radiation. Therefore we shall further consider transitions between levels m and n in the Ne atom with values of the total momenta $J_n = J_m = 2$. Energy level diagrams and radiative processes are shown in Fig. 1. However, the results are also valid for transitions in other atoms with the same values of total angular momenta.

Let us consider a problem of the absorption spectrum of a probe field in the presence of radiation of a strong field of the same frequency and the opposite direction of propagation. The radiation of a strong field of strength \mathbf{E}_1 is supposed to be a plane monochromatic wave (of frequency ω and wave vector $\boldsymbol{\kappa}$), which is in resonance with the m - n atomic transition (transition frequency is ω_{mn}) with a linear polarization.

The radiation of a probe field of strength \mathbf{E}_2 is supposed to be a plane monochromatic wave (frequency $\omega_2 = \omega$ and wave vector $\boldsymbol{\kappa}_2 = -\boldsymbol{\kappa}$) with a circular σ^- polarization propagating in a direction opposite to that of the strong light wave. The case of a probe wave with a linear polarization orthogonal to the strong field similar to the conditions of our experi-

ment is also reduced to the considered problem.

We assume that the gas pressure is quite low; in this case, collisions can be neglected and the only relaxation mechanism is associated with spontaneous transitions in the atom. In case of the linear polarization of a strong field, we shall consider the problem in a coordinate system with the quantization axis z direction along the strong field strength \mathbf{E}_1 . In this coordinate system, the strong field induces transitions involving a change in the magnetic quantum number $\Delta M = 0$ (Fig. 1); spontaneous transitions in this case are observed with a change $\Delta M = 0, \pm 1$. In the given problem, the strong field sets a nonequilibrium population of magnetic sublevels, and an absorption coefficient of the probe field is determined by this nonequilibrium difference in the population.

The system of the stationary kinetic equations for a density matrix on the $^3P_2 - ^3D_2$ transition of the Ne atom in the strong light field, according to [5], has the following form:

$$\Gamma_n \rho_M = -W_M (\rho_M - r_M) + \sum_{M'} A_{MM'} r_{M'} + Q_M \quad (1)$$

$$\Gamma_m r_M = W_M (\rho_M - r_M) + q_M. \quad (2)$$

Here, ρ_M and r_M are the diagonal elements of the density matrix describing the population of the lower and upper magnetic sublevels, $|M|, |M'| \leq 2$, $W_M = (2\Gamma |G_M|^2) / (\Gamma^2 + \Omega^2)$ is the probability of induced $nM \leftrightarrow mM'$ transitions; $\Omega_1 = \Omega - \kappa v$, $\Omega = \omega - \omega_{mn}$ being the detuning of the strong light field frequency from the atomic resonant transition frequency with allowance for the Doppler shift; v is the atom velocity; $\Gamma = (\Gamma_m + \Gamma_n) / 2$ is the transition width; $G_M = (dE / 2\sqrt{3}\hbar) (-1)^{J_n - M} \langle J_n M J_n - M | 10 \rangle$ is the parameter of interaction of atom on magnetic sublevel with the strong field. Γ_m, Γ_n, q_M , and Q_M are the rates of relaxation and excitation of sublevels mM and nM ; d is the reduced dipole moment of the m - n transition, E is the strength of the strong light field; and the expression within the angle brackets is the factor of vector summation. The rates of spontaneous transitions $mM - nM$ are determined by the Einstein coefficients A_{mn} and the factors of vector summation.

Absorption coefficient of the counter-propagating probe light wave with circular polarization per atom (all subse-

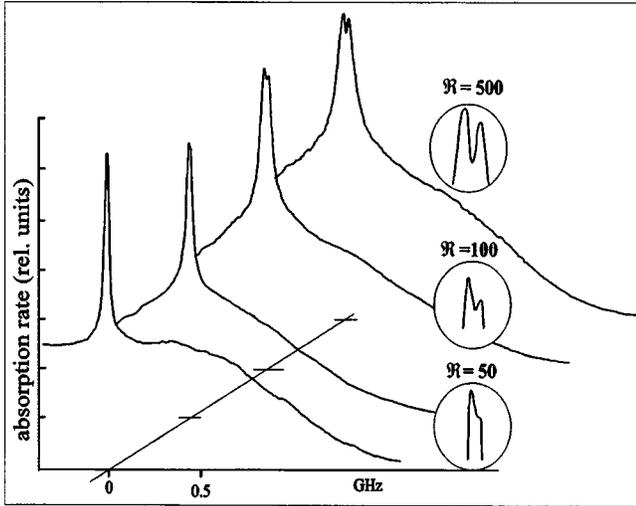


FIG. 3. Experimentally observed absorption line shape of a probe light field for various values of saturation parameters \mathcal{R} of the strong field. The peak structures with more high resolution are shown in separated fragments.

quent results are given for the σ^- polarization) was defined as

$$K_s^- = K_0 \sum_{-2 \leq M \leq 2} |d_{MM-1}|^2 \left\langle \frac{\Gamma^2}{\Gamma^2 + (\Omega + \kappa v)^2} (\rho_M - r_{M-1}) \right\rangle \quad (3)$$

where $K_0 = 4\pi\omega_{mn}/(c\hbar\Gamma)$, d_{MM-1} is the dipole moment of transition between the magnetic sublevels and the angle brackets indicate averaging over velocities. Averaging in (3) was carried out with an equilibrium (Maxwellian) and non-equilibrium velocity distribution functions of particles.

The system of Eqs. (1) and (2) together with Eq. (3) was solved numerically on a grid with steps of the relative frequency detuning of $\Omega/\kappa v_i$ and the relative particle velocity v/v_i equal to 10^{-3} , where v_i is the most probable velocity of an ensemble. In averaging, all particles with velocities $|v/v_i| \leq 3$ were taken into account. It can be seen that the number of discarded particles is exponentially small. The following values of the atomic transition parameters were taken for calculations [4]: the probability of spontaneous transition was $A_{mn} = 1.36 \times 10^7 \text{ s}^{-1}$; the rate of the lower-level decay $\Gamma_n = 10^5 \text{ s}^{-1}$; the rate of the upper-level decay Γ_m was defined in terms of the branching parameter $\alpha = A_{mn}/\Gamma_m$, thus the value of α being varied in the range 0.1–1. The strong field-saturation parameter, defined as $\mathcal{R} = |dE|^2/6\hbar^2\Gamma\Gamma_n$, was varied in the range 0.5– 10^4 . Excitation rates Q_M and q_M determine the population of states of the Ne atom in the absence of a strong field. Since the population of the lower n state in a gas discharge is considerably higher than the population of the upper m state, we set $q_M = 0$ in numerical calculations and assumed that the pumping of the lower state is the same to all sublevels with a rate $Q_M = \Gamma_n$, although the population of sublevels with $|M|=2$ under the discharge cell conditions is slightly higher than the population of the remaining sublevels [6].

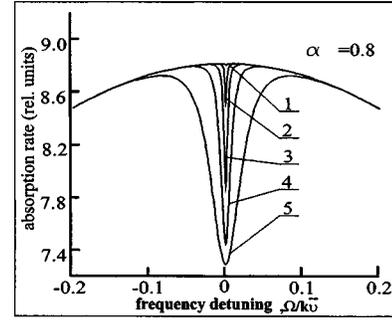


FIG. 4. Absorption line shape of the probe light field for various saturation parameters \mathcal{R} of the strong light field, calculated with the equilibrium velocity distribution function of particles and branching parameter $\alpha = 0.8$: $\mathcal{R} = 1$ (1), 10 (2), 10^2 (3), 10^3 (4), 10^4 (5).

IV. SINGULARITIES IN THE ABSORPTION SPECTRUM OF THE PROBE FIELD ON THE $J_n=2 \rightarrow J_m=2$ TRANSITION OF THE Ne ATOM

Numerical research on the given model allowed one to reveal the reasons for unusual behavior of saturated absorption resonance in an atomic system with degenerate levels, including the reason of a doublet splitting of the resonance peak.

Transition $J_m=2 \rightarrow J_n=2$ is characterized by the following peculiarities: the dipole moment and, hence, the probability of a spontaneous transition between magnetic sublevels with $M=0$ are equal to zero ($d_{00}=0, A_{00}=0$), whereas the values of the dipole moments and spontaneous transition probabilities between other sublevels differ considerably. In particular, the ratio of the maximal probability A_{22} to the minimal probability A_{11} is equal to 4. This leads to an elevated population of the lower state n with $M=0$ (because of the optical pumping and absence of an induced transition from this sublevel in a strong field) and, it is essential, in different field action (broadening) of a strong field on the line shape of a probe light field on transitions between individual magnetic sublevels. This is manifested in the complex form of the resulting absorption coefficient spectrum in the vicinity of the center of a transition line.

The absorption spectrum of a probe field on transition between degenerate states of atom Ne, introduced in Fig. 1, is determined by the result of summation of four Doppler contours with Lorentz-type structures near the center of a line transition reflecting modification of the magnetic sublevel populations of the lower atom state under the action of a strong field and having different widths and different amplitudes (both in value and in sign).

It was found that the action of a strong field reduced to the following dynamics in the velocity distribution of particles on magnetic sublevels of the lower state: (i) the formation of a dip in the velocity particle distribution function (Bennett hole) on the sublevels with $M = \pm 2$ for any values of saturation parameter \mathcal{R} ; (ii) the emergence of a small-amplitude peak on sublevels with $M = \pm 1$ for values of parameter $\mathcal{R} \leq 1$, that converted for values $\mathcal{R} > 1$ into the dip with the line width much smaller than that observed on sublevels with $M = \pm 2$; (iii) the formation of a peak on the sublevel with

10^4 (6).

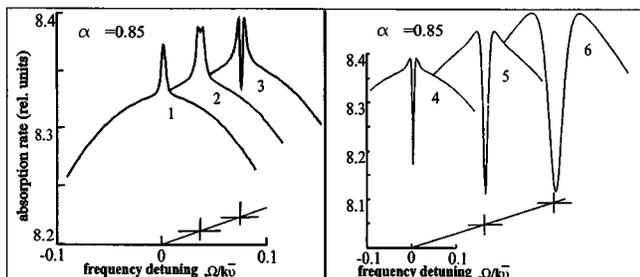


FIG. 5. Absorption line shape of the probe light field for various saturation parameters \mathcal{R} of the strong light field, calculated with the equilibrium velocity distribution function of particles and branching parameter $\alpha=0.85$: $\mathcal{R}=1$ (1), 5 (2), 10 (3), 10^2 (4), 10^3 (5), 10^4 (6).

$M=0$ for any parameters \mathcal{R} . It is just the result of summation of the given four contours with different amplitudes and widths that determines a complicated character of the line shape of saturated absorption resonance, including the doublet structure in the resonance peak discovered in our experiment.

It was also found that the line shape of the resonance depended on the branching parameter α determined as $\alpha = A_{mn}/\Gamma_m$. For values of $\alpha < 0.85$, the resonance line shape near the centerline of atomic transition is a conventional saturated absorption resonance in the form of a Lorentzian dip against the background of a broad Doppler absorption contour with a width and amplitude depending on the intensity (saturation parameter) of the strong field (Fig. 4). But for values $0.85 \leq \alpha \leq 1$ the resonance form changed: instead of the traditional dip the peak of absorption with a spectral width and an amplitude depending on the intensity of the strong field is formed.

In the vicinity of $\alpha \approx 0.85$, the field dependencies of the saturated absorption resonance spectrum from the saturation parameter \mathcal{R} of a strong field are complex (Fig. 5). For values of saturation parameter $\mathcal{R} \leq 1$ the peak of a small amplitude is observed (Fig. 5, curve 1), but with the increase of the strong field intensity for values of $\mathcal{R} \leq 10$ this peak splits into two components (curve 2), while for values of $\mathcal{R} \geq 10$ a dip with a width and amplitude determined by the intensity of the strong field is formed (curves 3–6). In this case, the maximum amplitude of the dip considerably exceeds the amplitude of the peak.

In the range of the branching parameter $0.9 \leq \alpha \leq 0.95$ and for saturation parameters $\mathcal{R} \geq 50$ the resonance peak

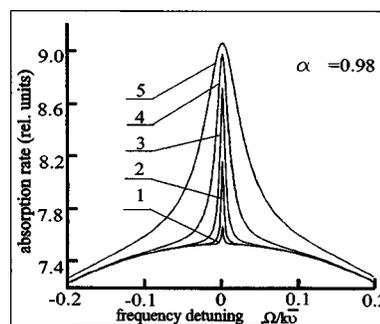


FIG. 7. Absorption line shape of the probe light field for various saturation parameters \mathcal{R} of the strong light field, calculated with the equilibrium velocity distribution function of particles and branching parameter $\alpha=0.98$: $\mathcal{R}=1$ (1), 10 (2), 10^2 (3), 10^3 (4), 10^4 (5).

splits into two components. The characteristic absorption line shapes of the probe light field for a number of values of saturation parameter of the strong field and for the value of branching parameter $\alpha=0.9$ are shown in Fig. 6(a). As follows from Fig. 6(a), the amplitude of the absorption peak weakly depends on the intensity of a strong field, whereas the width of the peak and the value of the doublet splitting hardly depend on this quantity in the examined range of parameters \mathcal{R} .

For values of $\alpha > 0.95$, the doublet structure at the centerline of the probe light field is not manifested, and the resultant spectrum has the form of an absorption peak with the ordinary Doppler lining (Fig. 7). Amplitude and width of the peak are determined by the intensity of the strong field.

Comparison of the experimentally observed relations of an absorption spectrum of the probe field shown in Fig. 3 and the presented results of model calculations shows good quantitative agreement between the experimental and computational data, obtained at values for branching parameter $\alpha \sim 0.9$ [Fig. 6(a)]. According to the experimental data, the maximal value of frequency between peaks of the doublet structure splitting was about ~ 25 MHz for a value of saturation parameter of the strong field $\mathcal{R} \sim 500$, whereas the width of the absorption peak varied between 14–70 MHz upon a change in the saturation parameter \mathcal{R} in the range 50–500.

As follows from the results of calculations, variation of the saturation parameter of strong field in the range of $\mathcal{R} = 10^2$ – 10^4 leads to the variation of the half-amplitude width of the absorption resonance peak in the range 12–95 MHz, whereas the frequency value between peaks of the doublet

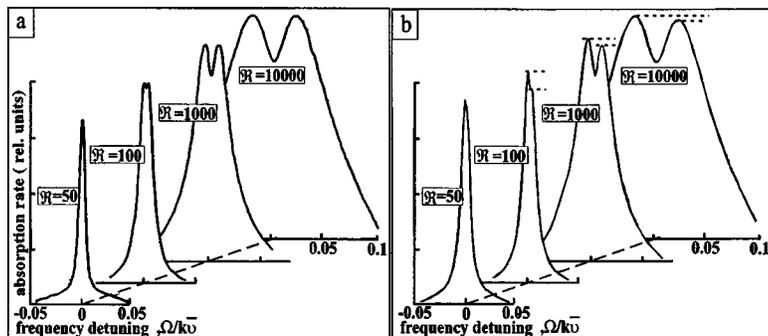


FIG. 6. Absorption line shape of the probe light field for various saturation parameters \mathcal{R} of the strong light field, calculated with the equilibrium (a) and nonequilibrium (b) velocity distribution function of particles. Branching parameter is $\alpha=0.9$.

structure varies in this case from 3 to 33 MHz. It should be emphasized, that model calculations with the equilibrium distribution of particles on magnetic sublevels lead to a symmetric splitting of the peak of the saturated absorption resonance line [Fig. 6(a)] whereas asymmetric splitting of the resonance line was observed in experiments (Fig. 3).

Thus we shall point out that the value of the branching parameter according to the data [4] on the rates of spontaneous decay channels of the $2p_8$ level of the Ne atom is $\alpha = 0.336$. In accordance with the results described above the absorption spectrum of the probe light field for this value of α must have a dip similar to that shown in Fig. 4. However, in experiment an absorption peak was observed, indicating the emergence (under conditions of our experiment) of physical processes considerably increasing the contribution from the transition on which the strong field is acting and reducing the contribution from other relaxation channels of the upper level. The nature of these processes still remains unclear.

A strong dependence of the saturated absorption resonance line shape on the branching parameter α has the following physical nature. Parameter α determines the fraction of the “useful” decay rate of the upper level m via m - n channel of the total decay rate Γ_m . Action of the optical pumping by the strong light field on the studied atomic transition with the degenerated levels leads to a higher population of the lower sublevel with $M=0$, investments of the other sublevels depend as $\alpha^{|M|}$.

For small values of α ($\alpha < 0.85$) the fraction of particles reaching the sublevel with $M=0$ of the lower state as a result of optical pumping is relatively small (particles are mainly pumped to the third levels via the channels $n \rightarrow m \rightarrow j \neq n$). For this reason, the resultant contour of the probe field absorption line is determined by the Bennett structures in the population of the lower sublevels with $M = \pm 1, \pm 2$ which leads to the traditional Lamb-type dip near the centerline of the atomic transition. With increasing α , the fraction of particles reaching the sublevel with $M=0$ as a result of optical pumping increases, which leads to an increase in the contribution of the given sublevel to the shape of the absorption coefficient of the probe field in the form of a peak at the centerline (for $\alpha \sim 1$, this contribution becomes decisive, Fig. 7). In a certain range of α values ($\alpha \sim 0.85$ – 0.9), the amplitudes of peak and dips of the Bennett structures on magnetic sublevels are almost identical, while their widths differ substantially. This leads to a complex dependence of the total absorption coefficient of the probe light field. Figures 5 and 6(a) reflect the dynamic of the saturated absorption resonance spectrum for a varying intensity of the strong light field in the given range of α .

V. LIGHT PRESSURE EFFECT ON THE $J_n=2 \rightarrow J_m=2$ TRANSITION OF THE Ne ATOM

In connection with the above circumstance, studies of influence of the light pressure force caused by a strong field on the line shape of the saturated absorption resonance were made. The technique, developed by us earlier [7,8], was used as the basis for these calculations.

In contrast to transitions from the ground state, the action of light pressure force for transitions from excited states of atoms exhibit peculiarities associated with the following factors.

(i) a finite lifetime of the lower state and existence of several decay channels for the upper state. In this case, the time t_r of the resonant interaction of an atom with the strong field is determined by the spontaneous transition probability and the branching parameter α as $t_r = A_{mn}^{-1} / (1 - \alpha)$. The interaction time t_r is found to be the same for all atoms in the ensemble in contrast to the case of interaction of light with atoms in the ground state, when the time t_r is determined by the time of flight of the atom through a light beam.

(ii) A difference in the values of the dipole moment and probabilities of transitions between degenerated sublevels with different values of magnetic quantum number. As result, action of the light pressure caused by a strong field produces different effects on the particle distribution on these magnetic sublevels. The analysis shows that the maximum effect of the strong field is manifested in the particle distribution over the sublevels with $M = \pm 2$.

Generally, the velocity distribution function of particles over sublevels in the field of a high-intensity light wave was determined from the solution of the Fokker-Planck equation having the following form under the condition when the Doppler frequency shift of a moving atom caused by the recoil effect $\kappa v_r = \hbar \kappa^2 / m$ is small in comparison to a homogeneous atom transition width ($\kappa v_r \ll \Gamma$) [2,3]:

$$df(v_z, t) / dt + \partial[A_z f(v_z, t)] / \partial v_z + \partial^2[C_{zz} f(v_z, t)] / \partial v_z^2 = 0. \quad (4)$$

Here, the factor A_z is determined by the light pressure force as

$$A_z = \pm \Gamma v_z \mathcal{R} / [1 + \mathcal{R} + (\Omega - \kappa v_z)^2 / \Gamma^2] \quad (5)$$

whereas the quantity C_{zz} , determining particle diffusion in the velocity space, has the form

$$C_{zz} = 0.5 \Gamma v_r^2 \mathcal{R} / [1 + \mathcal{R} + (\Omega - \kappa v_z)^2 / \Gamma^2]. \quad (6)$$

The plus and minus signs correspond to particles moving parallel and antiparallel to the wave vector κ of the strong light field, respectively.

The Fokker-Planck equation (4) was solved numerically on a two-dimensional grid [600, 1000] with a step of relative velocity variation of $\kappa \Delta v_z / \Gamma = 0.5 \div 0.1$ and a step of relative time variation ($\tau_r = \kappa v_r t$) $\Delta \tau = 0.1 \div 0.05$. In model calculations we used the following values of atomic parameters: the most probable velocity $v_r \approx 7 \times 10^2 \Gamma / \kappa$ at a gas temperature of $T = 300$ K, the radiation wavelength $\lambda \approx 633$ nm, the recoil frequency $\Delta \omega_r = \kappa v_r = 3.3 \times 10^5 \text{ s}^{-1}$, the relative recoil frequency $\Delta \omega_r / \Gamma \sim 5 \times 10^{-2}$, and the values of saturation parameter \mathcal{R} varied in the range of 0.1 – 10^4 . The relative interaction time of the atom with the strong field (during which the velocity distribution function of particles changes) was $\tau_r \approx 0, 5$ – 1 for values of branching parameter $\alpha \sim 0.9$ – $0, 95$. Note that at these values of α the doublet structure of the saturated absorption resonance line shape is formed.

Figure 6(b) presents results of calculations of line-shape behavior of the absorption spectrum of the probe light field near the center of the transition line of the Ne atom with nonequilibrium velocity distribution of particles depending on the saturation parameter \mathcal{R} of a strong field. It is evident from comparison of introduced relations Figs 6(a) and 6(b), that the action of light pressure force is manifested in the asymmetry of the absorption peak for saturation parameters $\mathcal{R} \leq 50$ and in the amplitude ratio of the peak-splitting doublet for $\mathcal{R} \geq 100$. It is worthy to note a good quantitative agreement between the experimental and computational data on the relative change of amplitudes of the doublet structure: the maximal experimental value was approximately equal to 8%, while the calculated values were 5–9%.

As follows from [7], the influence of light pressure force on the spectral reflectance of the saturated absorption resonance (the shape of resonance and the position of its minimum relative to the atomic transition frequency) is maximal for values of saturation parameter $\mathcal{R} = 1-2$. However, the peculiarity of formation of the doublet spectral structure on transitions with degenerate states of atom as a result of subtraction of several contours (realization of the difference scheme of observation on atomic level) makes it very sensitive to the velocity distribution function of particles interact-

ing with a strong field and permits one to observe an action of light pressure force in the nonoptimal-in-effect region of \mathcal{R} values.

VI. CONCLUSION

In conclusion it should be noted that the mechanism of the effective increase of branching parameter α for a block of closely located $1 S_i$ levels in the Ne atom is still obscured. The main reason for this increase is, apparently, electron collisions, resulting in intermixing level population inside $1 S_i$ states of atom.

The presented results testify to the essential influence of the light pressure force on properties of the saturated absorption resonance on transitions from excited atomic states. The value of the effect can be significant for light atoms. The latter circumstance is especially important for interpretation of results of precision metrological measurements on the basis of the saturated absorption method, including the Rydberg constant [9].

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