Spectroscopy of two-quantum transitions in a gas near resonances

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The shape of a resonant stimulated Raman scattering (SRS) line in a gas is investigated. Transitions in a three-level system in the presence of resonant monochromatic fields are classified within the framework of second-order perturbation theory on the basis of an analysis of the level relaxation constants. It is shown that for certain relations between the constants, the scattering line near resonance is determined by two-quantum processes. In the general case, the line shape reflects interference between two-quantum and one-quantum step-like transitions. The Doppler broadening of the forward and backward SCS resonance lines is determined. Experimental investigations of the resonance SCS line shape for 2s – 2p transitions in neon are described in detail. The line widths for forward and backward scattering and broadening induced by pressure and field are measured. Polarization measurements of the scattering line are carried out. The experimental results are compared with the theory. Various spectroscopic applications of two-quantum transitions are considered which pertain to the determination of the relaxation constants of certain levels in transitions between excited states and also to a study of collision broadening as well as to elucidation of the line structure concealed in ordinary conditions by considerable Doppler broadening.

INTRODUCTION

In the quantum theory of radiation[1-3], Raman (displacement) scattering takes place in second order of perturbation theory. It is usually regarded as a two-quantum process that arises via definite intermediate (virtual) states. Far from atomic or molecular resonance, in the region where matter is transparent, the cross section for Raman scattering is small, and it is practically unobservable in low-pressure gases. However, the presence of real levels should lead to a increase in the scattering cross section, just as the cross section for classical scattering increases near resonance (resonance fluorescence[4,5]). This makes it possible to perform experiments in low-pressure gases with gas lasers of low power but high frequency stability, and investigate the shape and the fine structure of the line with a resolution better than 10^{-8} in the optical and near infrared regions of the spectrum.

The presence of a real level and the damping of the states changes the general picture of the scattering. It is of interest in this connection to trace the variation of the kinetics of radiative transitions on going from the case of pure Raman scattering to scattering near a resonance. In the presence of real levels it becomes necessary to take into account not only the two-quantum transitions within the framework of second-order perturbation theory, but also single-quantum stepwise transitions[1)]. At exact resonance, these processes are indistinguishable and the scattering line shape is determined in the general case by the interference of two-quantum and single-quantum stepwise transitions.

A theoretical analysis of the line shape of resonance Raman scattering (stimulated or spontaneous) reduces to an investigation of the line shape of the emission of the weak signal in the presence of an optical field at a neighboring transition. These questions, with allowance for motion of the atoms, have recently attracted considerable attention[8-18]. As shown in these studies, the appearance of singularities of the resonant interaction of the optical field with a three-level system depends on the level configuration, populations, relaxation constants, etc. It is therefore no accident that different authors have focused their attention on different aspects of the problem, using different terminologies and interpretations of the phenomena predicted by the theory. Thus, Rautian et al.[9] interpreted the change of the spontaneous-emission line shape as a manifestation of interference of atomic states, while Holt[10] interpreted it as the result of frequency correlation in a two-quantum transition. Feld and Javan[11] attributed the changes in the emission-line shape to two-quantum transitions, and also to changes in the probability of single-quantum transitions from a common level in a strong resonant field. Stepwise single-quantum transitions were not considered. Popova et al.[12] proposed the following classification of the phenomena: establishment of a non-equilibrium velocity distribution of the atoms, nonlinear interference effect, and "splitting," i.e., the dynamic Stark effect. A similar classification was used by Hansch and Toschek[15].

From the results obtained in these papers, the most interesting and important for high-resolution spectroscopy are the difference between the line widths in forward and backward emission, and a strong narrowing of the emission line in one of the directions, which was predicted by Rautian et al.[9]. These phenomena were first observed experimentally in spontaneous emission[19,25] and in investigations of a three-level gas laser[20,21] and the line shape of stimulated emission[22-24]. There is no doubt that the line-width difference observed in a number of studies[19,22,25] is due to the presence of two-quantum transitions. Using the classical interpretation of Raman scattering, we can explain immediately the difference between the forward and backward scattering line widths. A transformation to the c.m.s. of the moving harmonic oscillator converts the frequency ω of the acting field into $\omega - \mathbf{k}\cdot\mathbf{v}$, where $\mathbf{k}$ is the wave vector and $\mathbf{v}$ is the velocity vector. The internal oscillations of the quantum system with frequency ω_{nl} modulate the stimulated oscillations and lead to the appearance of a combination frequency $\omega - \omega_{nl} - \mathbf{k}\cdot\mathbf{v}$ in the emission spectrum of the oscillator. Returning to the laboratory frame, we obtain the frequency $\omega - \omega_{nl} - (\mathbf{k} - \mathbf{k}_\mu)\cdot\mathbf{v}$, where $\mathbf{k}_\mu$ is the wave vector at the combination frequency. If we consider an ensemble of moving oscillators, we can readily establish that the cancellation of the Doppler shift leads to a narrowing of the Doppler line of the forward Raman scattering.

We show in the present paper that within the framework of second-order perturbation theory, the observed phenomena are a manifestation of two-quanta and single-quantum stepwise transitions as well as their interferences. This approach enables us to explain, from a unified point of view, the characteristic features of the emission of an ensemble of moving atoms in an external monochromatic field that is at resonance with an adjacent transition. In particular, we show that at exact resonance, under certain conditions, stimulated emission at the adjacent transition is due only to two-quantum transitions of the stimulated Raman scattering (SRS) type.

The experiments were performed in neon with the transitions $2s_2 - 2p_1$ and $2s_2 - 2p_4$, the choice of which satisfies in the main the requirements necessary for observation of resonant SRS. The results of the experimental investigations of the resonant SRS line shape in neon are in good agreement with the theory. The work concludes with an analysis of important spectroscopic applications of two-quantum transitions.

1. LINE SHAPE OF STIMULATED SCATTERING NEAR RESONANCE

We consider the emission of an atom in an external field that contains two monochromatic components with frequencies ω and ω_μ and amplitudes $\mathbf{E}$ and $\mathbf{E}_\mu$, in a scheme corresponding to Raman scattering ($n \to m \to l$, Fig. 1). The system of equations for the probability amplitudes takes the form

$$\begin{aligned} \dot{a}_m + \gamma_m a_m &= iVe^{-i\Omega t}a_n + iV_\mu e^{-i\Omega_\mu t}a_l, \\ \dot{a}_n + \gamma_n a_n &= iV^* e^{i\Omega t}a_m, \\ \dot{a}_l + \gamma_l a_l &= iV_\mu^* e^{i\Omega_\mu t}a_m, \end{aligned} \quad (1)$$

where

$$\Omega = \omega - \omega_{mn}, \quad \Omega_\mu = \omega_\mu - \omega_{ml}, \quad V = \mathbf{p}_{mn}\mathbf{E}/2\hbar, \quad V_\mu = \mathbf{p}_{ml}\mathbf{E}_\mu/2\hbar,$$

$\mathbf{p}_{i,j}$ are the dipole-moment matrix elements. The scheme of the Raman scattering corresponds to a situation in which the atom is excited to a state n at the initial instant $t = 0$, i.e., $a_n(0) = 1$. We are interested in the total probability of finding the atom in the state l. This probability is equal to

$$w_\mu = 2\gamma_l \int_0^\infty |a_l(t)|^2 dt. \quad (2)$$

We shall solve the system (1) by successive approximations in V and V_μ, and confine ourselves to second-order perturbation-theory effects. If the atom is excited to the state n at the instant $t = 0$, i.e., $a_n(0) = 1$, $a_m(0) = 0$, and $a_l(0) = 0$, we obtain in first-order perturbation theory[2)]

$$a_m^{(1)}(t) = \frac{iV}{\gamma_n - \gamma_m + i\Omega}[\exp\{-\gamma_m t\} - \exp\{-\gamma_n t + i\Omega t\}]. \quad (3)$$

The probability amplitude $a_m(t)$ consists of two terms,

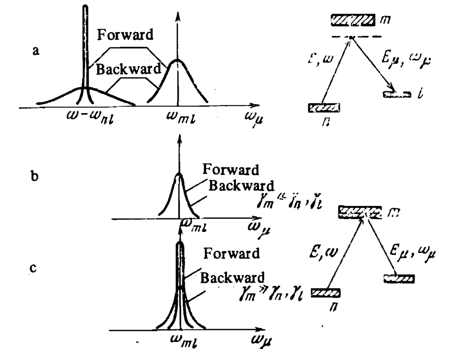

FIG. 1. Radiative processes in three-level system near resonance: a—Doppler-broadened line shape of Raman scattering and of single-quantum stepwise transition with the field strongly off-resonance ($\Omega \gg kv$); b—Doppler-broadened line of single-quantum stepwise transition at exact resonance ($\Omega \approx 0$); c—Doppler-brandened shape of resonant Raman scattering ($\Omega \approx 0$).

one of which attenuates during the lifetime of the atom in the state m, while the second oscillates at a frequency Ω and attenuates during the lifetime of the atom in the state n.

Second-order perturbation theory yields

$$a_l^{(2)}(t) = \frac{VV_\mu}{\gamma_n - \gamma_m + i\Omega}\left[\frac{\exp\{-\gamma_m t + i\Omega_\mu t\} - \exp\{-\gamma_l t\}}{\gamma_l - \gamma_m + i\Omega_\mu} - \frac{\exp\{-\gamma_n t + i(\Omega_\mu - \Omega)t\} - \exp\{-\gamma_l t\}}{\gamma_l - \gamma_n + i(\Omega_\mu - \Omega)}\right]. \quad (4)$$

The amplitude $a_l^{(2)}(t)$ is a sum of two terms, the first of which is interpreted as the amplitude of the probability in a single-quantum stepwise transition. Its form does not differ from the case when the atom is excited at the initial instant of time to the state m. The second term contains information both on the initial state n and on the quantum absorbed in the transition $m \to n$, and corresponds to the probability amplitude of the two-quantum transition.

Substituting (4) in (2), we obtain after transformation the following expression for the probability of stimulated emission at the detuning frequency Ω_μ:

$$w_\mu = \frac{|V|^2}{(\gamma_n - \gamma_m)^2 + \Omega^2}\left\{\frac{\gamma_m + \gamma_l}{\gamma_m}\frac{|V_\mu|^2}{\Omega_\mu^2 + (\gamma_m + \gamma_l)^2} + \frac{\gamma_n + \gamma_l}{\gamma_n} \times \frac{|V_\mu|^2}{(\Omega_\mu - \Omega)^2 + (\gamma_n + \gamma_l)^2} - 2|V_\mu|^2 \operatorname{Re}\frac{1}{\gamma_m + \gamma_n - i\Omega}\left[\frac{1}{\gamma_m + \gamma_l - i\Omega_\mu} + \frac{1}{\gamma_n + \gamma_l + i(\Omega_\mu - \Omega)}\right]\right\}. \quad (5)$$

The probability w_μ consists of three terms. The first term in the curly brackets is analogous to the expression for the line of the single-quantum stepwise transition, and the second is analogous to the expression for the two-quantum transition line. The third term can be interpreted as a result of the interference of the stepwise and two-quantum transitions, since we are adding not the total probabilities of two processes, but their amplitudes.

Ordinary Raman scattering corresponds to the case of strong deviation from resonance $\Omega \gg \Gamma$. The emission line consists of two individual components, one of which corresponds to a stepwise transition and the other to an ordinary two-quantum transition of the Raman-scattering type for the considered level scheme. When considering the emission spectrum near the frequencies $\Omega_\mu \sim \Omega$ and $\Omega_\mu \sim 0$ the interference term is immaterial. The stepwise-transition line has a width that coincides with the natural width of the $m \to l$ transition, and the line width of the Raman scattering turns out to equal $\gamma_n + \gamma_l$, i.e., it corresponds to the transition $n \to l$, which is forbidden in the single-quantum limit. The limiting transition to the classical Raman scattering $\gamma_n, \gamma_l \to 0$ corresponds to a scattering line with zero width, and calls for an analysis of not the emission probability but of the transition rate.

At resonance, the interpretation of the observed emission spectrum becomes much more difficult, since when interference is taken into account an emission line can no longer be ascribed uniquely to one of the processes. The concepts of stepwise and two-quantum transitions, of course, remain in force, since we remain, as before, within the framework of second-order perturbation theory, but a fundamental fact is that in no case can we indicate the channel through which the atom went from the state n to the state l. However, even under conditions of exact resonance, at a definite ratio of γ_m and γ_n the line shape can be ascribed exclusively either to single-quantum stepwise or two-quantum transitions.

Let $\gamma_m/\gamma_n \ll 1$, i.e., let the lifetime of the atom at the common level be large. Then the first term in (5), which is proportional[3)] to $(1/\gamma_m)^2$, turns out to be predominant. The contributions of the second term $\sim(1/\gamma_n)^2$ and of the interference term $\sim 1/\gamma_m\gamma_n$ can be neglected. In this case the scattering line is a stepwise-transition line. Physically this is easily understood. The change of the population of the level m in the field is larger the smaller γ_m/γ_n. The emission probability has the form of the product of the probabilities of the single-quantum transitions $n \to m$ and $m \to l$:

$$w_\mu \approx \frac{|V|^2}{\Omega^2+\gamma_n^2}\frac{\gamma_m+\gamma_l}{\gamma_m}\frac{|V_\mu|^2}{\Omega_\mu^2+(\gamma_m+\gamma_l)^2}. \qquad (6)$$

If $\gamma_n/\gamma_m \ll 1$, then the common level m decays very rapidly. The principal contribution is that of two-quantum transitions (the second term of (5)), and the emission-line shape takes the form

$$w_\mu \approx \frac{|V|^2}{\Omega^2+\gamma_m^2}\frac{\gamma_n+\gamma_l}{\gamma_n}\frac{|V_\mu|^2}{(\Omega_\mu-\Omega)^2+(\gamma_n+\gamma_l)^2}. \qquad (7)$$

In this case the population of the level m is low and we neglect the single-quantum stepwise transitions. (If we draw an analogy with the harmonic oscillator, this means that we have neglected the free oscillations of the oscillator in comparison with the forced oscillations, the damping of which is determined by the time of action of the driving force—in our case, by the lifetime of the atom at the level n.)

When γ_m and γ_n turn out to be comparable, both processes are essential and it is necessary to take into account all three terms in the emission line shape. The interpretation of the various terms of formula (5) becomes difficult, since the emission-line shape is influenced not only by the interference between the stepwise and two-quantum processes, but also by change in the amplitude of one process when the other is taken into account, a change described in (5) by the factor preceding the curly brackets.

The emission line of moving atoms can be obtained by introducing the Doppler-shift frequencies $\Omega \to \Omega - \mathbf{k}\cdot\mathbf{v}$ and $\Omega_\mu \to \Omega_\mu - \mathbf{k}_\mu\cdot\mathbf{v}$, with subsequent averaging over the velocities. Here $\mathbf{k}$ and $\mathbf{k}_\mu$ are the wave vectors of the strong and weak fields, respectively. For simplicity, we consider collinear beams. In the case $\gamma_n \gg \gamma_m$, averaging of w_μ over the velocities yields the following emission line:

$$\langle w_\mu\rangle^{(\pm)} \approx \frac{|VV_\mu|^2}{\gamma_m}\frac{\pi^{1/2}}{\gamma_n k\bar{v}}\exp\left[-\left(\frac{\Omega}{k\bar{v}}\right)^2\right]\frac{\Gamma_0}{(\Omega_\mu \pm k_\mu\Omega/k)^2+\Gamma_0^2} \qquad (8)$$
$$\Gamma_0 \approx \gamma_m+\gamma_l+k_\mu\gamma_n/k,$$

i.e., an isotropic line and one coinciding with the line obtained from an analysis of population effects only (Bennett "holes"[26]).

At $\gamma_m \gg \gamma_n$ and $k_\mu \approx k$, the emission line shape for parallel propagation of the waves has a common form that coincides with the case of large detuning[7]

$$\langle w_\mu\rangle^{(-)}_{\text{scat}} \approx \frac{|VV_\mu|^2}{\gamma_m}\frac{\pi^{1/2}}{\gamma_n k\bar{v}}\exp\left[-\left(\frac{\Omega}{k\bar{v}}\right)^2\right]\frac{\gamma_n+\gamma_l}{(\Omega_\mu-\Omega)^2+(\gamma_n+\gamma_l)^2}. \qquad (9)$$

We can estimate the increase of the cross section of the resonant Raman scattering in comparison with the nonresonant case:

$$\frac{\sigma_{\text{res}}}{\sigma_\Omega} = \sqrt{\pi}\frac{\Omega^2}{\gamma_m k\bar{v}}, \qquad (10)$$

where $|\Omega| \gg k\bar{v} \gg \gamma_m$.

In the general case at $\Omega \sim k\bar{v}$ and arbitrary k_μ and k, averaging yields

$$\langle w_\mu\rangle^{(-)}_{\text{scat}} \approx \frac{|VV_\mu|^2}{\gamma_m}\frac{\pi^{1/2}}{\gamma_n k\bar{v}}\exp\left[-\left(\frac{\Omega}{k\bar{v}}\right)^2\right]\frac{\Gamma_-}{(\Omega_\mu - k_\mu\Omega/k)^2+\Gamma_-^2} \qquad (11)$$
$$\Gamma_- \approx \gamma_n+\gamma_l+(k_\mu/k-1)\gamma_m \quad \text{at} \quad k_\mu > k,$$
$$\Gamma_- = \gamma_n+\gamma_l+(1-k_\mu/k)\gamma_m \quad \text{at} \quad k_\mu < k.$$

The backward scattering line is given by

$$\langle w_\mu\rangle^{(+)}_{\text{scat}} \approx \frac{|VV_\mu|^2}{\gamma_m}\frac{\pi^{1/2}}{\gamma_n k\bar{v}}\exp\left[-\left(\frac{\Omega}{kv}\right)^2\right]\frac{\Gamma_+}{(\Omega_\mu + k_\mu\Omega/k)^2+\Gamma_+^2}, \qquad (12)$$
$$\Gamma_+ \approx \gamma_n+\gamma_l+(k_\mu/k+1)\gamma_m \equiv \Gamma_0.$$

The expressions (8), (11), and (12) obtained for the line shape of stimulated emission in an external field are illustrated in Fig. 1, which also shows the case of strong deviation from resonance[7].

The observed narrowing of the emission line in the case of parallel propagation of the waves is due to cancellation of the Doppler shift and takes place only in the expression which we connect with the two-quantum transition. Therefore, while in the general case the emission of individual atoms could not be ascribed to any one particular process, the difference between the forward and backward line widths is undoubtedly due to the presence of two-quantum transitions and is most significant when it is precisely these transitions which determine the emission-line shape. It is therefore no accident that the difference between Γ_+ and Γ_- contains only the width of the common level.

In connection with the considered picture of the transitions, it is of interest to discuss specifically the case when the excited atoms are at a common level m and $\gamma_m \gg \gamma_n$. Then, as shown in a number of papers [15,16,23], at equal populations of the levels m and n, changes occur in the line shape of the signal at the transition $m \to l$ in the presence of a field at the transition $m \to n$, provided only $k > k_\mu$. The effect was maximal at $k_\mu = k/2$, and was attributed within the framework of second-order perturbation theory to the dynamic Stark effect in a rapidly alternating field, i.e., to the splitting effect. The dependence of the effect on the ratio of the wave numbers k_μ and k, which was presented above, was not explained.

Without stopping to analyze this phenomenon in detail, we note that in this case the onset of the second-order effect can be attributed to interference of the single-quantum transition $m \to l$ with the two-quantum transition $n \to m \to l$ that results from a single-quantum transition from the level m to a long-lived level n under the influence of the external field. It is easy to establish that this interference is essential for one atom, if the time-oscillating factors in the probability amplitudes, equal to $\exp(i\Omega_\mu t)$ and $\exp[i(\Omega - \Omega_\mu)t]$ for the single-quantum and two-quantum transitions, respectively, have identical or nearly equal frequencies. Changing over to the Doppler-shifted frequencies, we obtain the condition under which the interference makes a maximum contribution to the resonance:

$$-k_\mu v \approx (k_\mu - k)v, \quad k_\mu \approx k/2. \tag{13}$$

At $k_\mu > k$, the condition for the interference between the single-quantum and two-quantum processes is not satisfied. Thus, we obtain on the basis of simple considerations the result revealed by analysis of the line shape[15,16], which has not previously been clearly explained.

The foregoing singularities of the line shape can be used extensively in spectroscopic research. In essence, we can speak of qualitatively new ways of investigating the widths and shifts of lines of optically forbidden transitions, of relaxation constants of individual levels, of the diffusion of excitation over magnetic sublevels in velocity space, and of spectroscopy within the natural line width. We shall describe below some results of experimental investigations in which the spectroscopy of two-quantum transitions in a gas is used.

2. EXPERIMENTAL INVESTIGATIONS OF THE LINE SHAPE OF STIMULATED RAMAN (SHIFTED) RESONANT SCATTERING IN NEON

a) Choice of transitions. The experiment aimed at observing the line shape of resonant SRS consisted of investigating the amplification line width at $\lambda = 1.15\ \mu$ (the transition $2s_2 - 2p_4$ in Paschen notation) in the presence of a strong monochromatic field at the adjacent transition $2s_2 - 2p_1$ ($\lambda = 1.52\ \mu$).

This system of transitions was chosen from the following considerations: the common level $2s_2$ is resonant, i.e., it is coupled to the ground state by a strong optical transition. As a result, the lifetime $\tau = 1/2\gamma_{2s_2}$ is much shofter than the lifetime of the level $2p_1$ or $2p_4$. The wavelengths of the transitions $2s_2 - 2p_4$ and $2s_2 - 2p_1$ are close enough so that we can expect the cancellation of the Doppler shifts in the two-quantum transition to be sufficiently effective and the contribution of the Doppler broadening to be small. An important circumstance from the point of view of verifying the theory was that numerous recent investigations have led to accumulation of appreciable material on the relaxation times of the 2s and 2p levels of neon, and finally, inasmuch as in the present case $k_\mu > k$, the effect of interference of single-quantum and two-quantum processes should not come into play in the weak-field approximation if the population of the $2s_2$ level is finite. All this enables us to regard the experiment as a direct observation of the line of stimulated resonant Raman scattering.

The lifetimes of the 2p levels of Ne were measured by Bennett and Kindlmann[28]. These measurements yielded the values $2\gamma_{2p_1} = 6.95 \times 10^7\ \text{sec}^{-1}$ and $2\gamma_{2p_4} = 5.23 \times 10^7\ \text{sec}^{-1}$. For the $2s_2$ level we use the result of the calculation of the Lamb hole for a 1.15 μ laser operating in pure Ne[21]. This calculation yields a value $2\gamma_{2s_2} = 1.6 \times 10^8\ \text{sec}^{-1}$. The width of the forbidden $2p_1 - 2p_4$ transition turns out to be $2(\gamma_{2p_1} + \gamma_{2p_4})/2\pi = 19$ MHz. The width of the peak in the velocity distribution at the $2s_2$ level, in $\Delta v/k$ units, is $2(\gamma_{2s_2} + \gamma_{2p_1})/2\pi = 36.8$ MHz. As already indicated, the width $2\Gamma_-$ of the forward scattering line is equal to the width of the two-quantum transition, which is made up of the width of the forbidden $2p_1 - 2p_4$ transition and the Doppler part $2(k_\mu/k - 1)(\gamma_{2s_2} + \gamma_{2p_1})$. We thus get 30.8 MHz for $2\Gamma_-$. The backward scattering line width $2\Gamma_0$ is equal to the width of the stepwise transition. It is governed by the contribution of the nonequilibrium velocity distribution of the atoms, which is equal to $(1/2\pi)(2\gamma_{2s_2} + 2\gamma_{2p_1})k_\mu/k = 48.6$ MHz, and by the $2s_2 - 2p_4$ transition. For $2\Gamma_0$ we obtain 82.6 MHz.

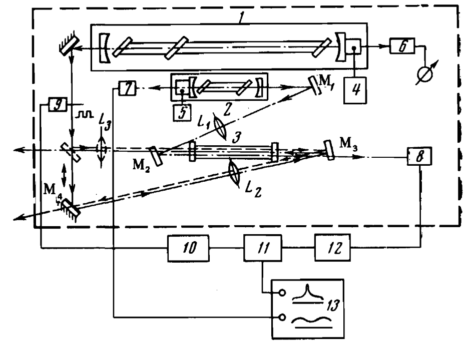

FIG. 2. Experimental setup for the observation of the line shape of resonant forward ($\mathbf{k} \cdot \mathbf{k}\mu < 0$) (dashed lines) and backward ($\mathbf{k} \cdot \mathbf{k}\mu < 0$) (solid SRS in neon). 1 – He-Ne laser ($\lambda = 1.52\mu$; strong field); 2 – He-Ne laser ($\lambda = 1.15\mu$, weak field); 3 – discharge cell; 4,5 – units to control the piezoceramics; 6, 7, 8 – photodiodes; 9 – mechanical modulator; 10 – acoustic oscillator; 11–phasesensitive detector; 12 – selective amplifier; 13 – automatic two-channel recorder.

We note that the line width of the two-quantum transition is 104 MHz in the case of backward scattering. This difference is due to stepwise single-quantum transitions. In our case $\gamma_m/\gamma_n \approx 3$ and the stepwise transitions cannot be fully neglected. Nonetheless, the differences between the forward and backward scattering lines are quite noticeable. Moreover, the forward scattering line at $\lambda = 1.15\ \mu$ turns out to be narrower than the natural width of this line.

b) Description of experimental setup. In contrast to our earlier line-shape investigations, where the principal attention was focused on the investigation of the dragging of the resonant radiation[24], in this paper we are interested mainly in the shapes of the narrow resonances. Therefore both the optical and the electronic systems were significantly modified. All the measurements were aimed at obtaining a large signal/noise ratio and at increasing the measurement accuracy. The experimental setup is shown in Fig. 2. Radiation from a high-power single-frequency laser 1, operating at the 1.52 μ line, was directed into a discharge tube 3 (length 50 cm, diameter 2 mm) filled with the pure isotope Ne^{20}. To measure the shape of the gain line or the absorption at the $2s_2 - 2p_4$ transition, a weak field with $\lambda = 1.15\ \mu$ from a short scanned single-mode helium-neon laser 2 was introduced into the cell 3. The mutual direction of propagation of the strong and weak fields could be altered by varying the position of the mirror

M_4. The dashed lines in the figure show the path of the rays in the case of parallel propagation of the strong and weak fields. In the case of antiparallel propagation of the beams, the interference mirror M_3, which had a reflection maximum in the region of $\lambda = 1.52\ \mu$, directed the strong-field radiation into the cell, and in the case of parallel propagation of the beams this mirror served as a filter that passed only the radiation at $\lambda = 1.15\ \mu$ to the photo-receiver. The mirror transmission at this wavelength was 60%. The system of mirrors M_1 and M_2 with reflection maxima at $\lambda = 1.15\ \mu$, ensured the admission of the weak-field radiation to the cell and filtering of the $\lambda = 1.52\ \mu$ radiation for the purpose of eliminating the coupling between lasers 1 and 2.

The matching of the wave fronts was carried out with the aid of lenses, which simultaneously focused the strong field in order to obtain a maximum saturation parameter in the cell. It should be indicated that failure of the wave fronts to coincide has a twofold influence, since it affects the magnitude of the signal as well as the line shape. The entire theory was constructed for plane traveling monochromatic waves in the case of both the strong and the weak fields. In real systems we deal, as a rule, with a spherical wave and with beams that are bounded by apertures. The transverse variation of the field can be disregarded for a single atom, when the moving atom traverses during its lifetime a distance in which the field remains practically unchanged. The atom is in a field of constant amplitude[4)]. At $d \approx 0.2$ cm and $\overline{v}/d\Gamma \approx 5 \times 10^{-3} \ll 1$ we can neglect the phase and amplitude modulations resulting from the finite dimensions and curvature of the beam. The influence of the sphericity on the gain line shape can be disregarded if full matching of the wave fronts is ensured. For the optical system used in the experiment, the influence of these factors could make a contribution no larger than 1 MHz to the width. The strong field was modulated with an electromechanical chopper at a frequency of approximately 40 Hz. The depth of modulation was 100%. As a result of the nonlinearity of the field interaction in cell 3, a modulation with the same frequency, 40 Hz, appeared in the radiation at $1.15\ \mu$ after it had passed through the cell. This radiation was registered with photoreceiver 8. The alternating signal from the photoreceiver was amplified with a U2-6 selective amplifier and fed to the input of a KZ-2 synchronous detector. The reference voltage was the signal from an acoustic oscillator that set the modulation frequency of the strong field. The signal from the output of the synchronous detector was fed to one of the channels of an automatic two-channel recorder 13. The time constant of the recording system was $\tau \leq 0.1$ sec.

The weak-field emission frequency was scanned slowly by applying a sawtooth voltage to a piezoceramic to which one of the mirrors of the resonator of laser 2 was fastened. The distance between the mirrors of this laser was 12.3 cm (the region of weak-field frequency scanning was 1220 MHz). The emission of laser 2 was fed to photoreceiver 7 (type FD-3), which was operated in the photovoltage-sensitivity regime, and the output of the receiver was fed to the second channel of the automatic recorder 13. Thus, the second channel of the automatic recorder registered the change of the amplitude of the weak field when its frequency was scanned. The alternating signal at the output of the synchronous detector was proportional to the weak field. It is therefore clear that the ratio of the first spectrogram of the automatic recorder 13 to the second is proportional to the alternating part of the absorption coefficient at the $\lambda = 1.15\ \mu$ line of neon in tube 3, and that this part is due to the strong external field $\lambda = 1.52\ \mu$.

The time necessary to plot one order was approximately 40 sec. This called for high stability of the strong-field frequency. Short-duration frequency deviations were averaged during registration. We used passive stabilization of the length of the resonator 1. To this end, the experimental setup was placed on a massive steel plate, which was located on an insulated concrete foundation. The error in the determination of the width due to the strong-field frequency drift during the time of plotting of the narrow structure of the line shape was of the order of 1 MHz, and this error was largely eliminated over many recordings made at different scanning directions. The relative short-duration laser stability was 10^{-9}.

In conclusion, let us discuss the method of eliminating the "underlining" resulting from the dragging of the resonant radiation. This was necessary to improve the measurement accuracy. The presence of the "underlining" complicated the analysis of the shapes of the narrow resonances (see Fig. 3). Since the shape of the "underlining" coincides with the absorption line[24], we could choose the amplitude and phase of the discharge-current modulation in such a way that the signal making up the "underlining" was completely cancelled out by the signal produced as a result of the modulation of the current, in the entire range of variation of the weak-field frequency. In practice, the selection was carried out in such a way that the amplitude of the alternating signal was equal to zero in the case of a deviation $\Omega_\mu \gg \Gamma_-, \Gamma_0$ from the narrow peak. This indeed signified a cancellation of the Doppler "underlining".

3. EXPERIMENTAL RESULTS AND DISCUSSION

a) Anisotropy of emission-line shape. Comparison with theory. Figure 3 shows the recorded stimulated emission (absorption) line shape for weak- and strong-field waves traveling in parallel (a) or antiparallel (b) directions. The figure shows simultaneously the

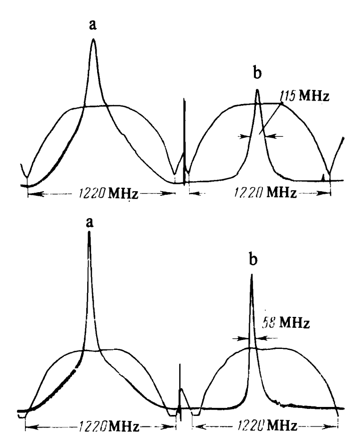

FIG. 3. Line shape of resonant SRS in neon: a — without cancellation of the Doppler "underlining"; b — pure scattering line. Upper trace backward scattering (P_{Ne} = 0.5 Torr, discharge current I_{dis} = 17 mA), lower trace — forward scattering (p_{Ne} = 0.9 Torr, I_{dis} = 15 mA).

variation of the weak-signal power. The gain line has a rather complicated shape. It constitutes a narrow peak against the background of a much broader "underlining". The "underlining," which has a width equal to the Doppler width and is connected with the equilibrium distribution with respect to the velocities, can be attributed only to the diffusion of the excitation in velocity space. This phenomenon was considered in detail earlier[24]. We shall dwell here on an investigation of the sharp structure that is produced in the emission line in a strong field.

The experimental curves clearly show the anisotropy of the line shape. The character of the anisotropy coincides qualitatively with that predicted by the theory. For a weak wave propagating in the same direction as the strong field ($\mathbf{k}\cdot\mathbf{k}_\mu > 0$), the emission line has a width of 58 MHz at a pressure 0.9 mm Hg, which is approximately half the value that follows from consideration of only population-saturation effects. For oppositely traveling waves ($\mathbf{k}\cdot\mathbf{k}_\mu < 0$) the width of the emission peak (115 MHz) is approximately equal to the sum of the Lorentz width at the $2s_2 - 2p_4$ transition and the width in the velocity distribution of the atoms. The amplitudes of the signals also differ (qualitatively they can be compared relative to the amplitude of the equilibrium "underlining").

To compare the experimental and theoretical results, we have investigated the dependence of the width on the pressure and on the intensity of the external field. With increasing field intensity at the 1.52 μ wavelength, we observed a certain broadening of the narrow resonances in the forward and backward scattering lines. Spectroscopic analysis is simplest when the fields are weak. In this case there is no need to take into account the transverse and longitudinal inhomogeneities of the fields. In our experiments, at the highest intensity of the strong field, the saturation parameter did not exceed $\kappa \sim 0.5$. The high signal/noise ratio made it possible to perform the measurements in a wide range of fields and to register reliably the broadening of the narrow resonances by the field. In the case of very weak fields, the signal decreases and the measurement accuracy decreases. Therefore the values of the widths corresponding to the case $\kappa \ll 1$ were obtained principally by extrapolating the dependence of the width on the field to zero field at each value of the pressure. Since all the measurements were made at saturation parameters no larger than 0.5, we have naturally used linear extrapolation. The measurement results obtained in different fields are shown in Fig. 4.

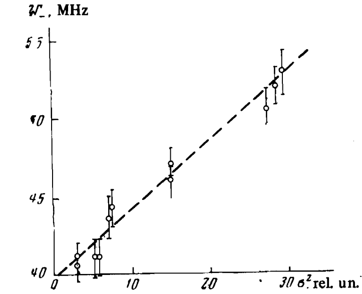

FIG. 4. Dependence of the line width of resonant forward SRS on the pump-field intensity (p_{Ne} = 0.5 mm Hg, I_{dis} = 15 mA).

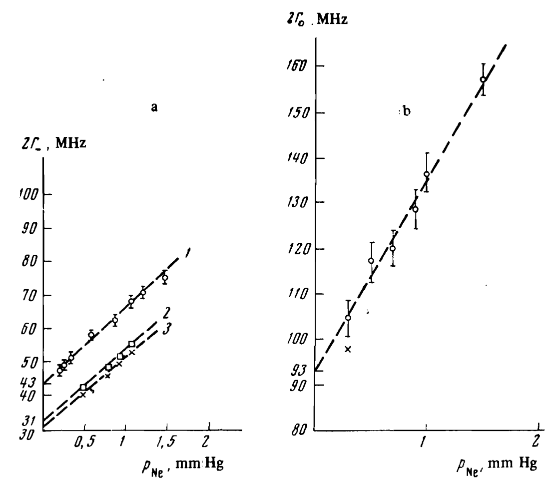

FIG. 5. Dependence of the line widths of resonant forward (a) and backward (b) SRS on the neon pressure in the cell. Curves 1, 2, and 3 were obtained at generation power values W = 1.2, 0.24, and 0 mW.

Figure 5 shows the measurement results, obtained at different pressures, for the broadening of the resonant Raman forward (a) and backward (b) scattering lines in neon. The crosses mark values obtained by extrapolation to zero field. The widths extrapolated to field and zero pressure were respectively $2\Gamma_0 = 86 \pm 3$ MHz and $2\Gamma_- = 31.5 \pm 2$ MHz. These results are in good agreement with the calculated values. Consequently, one can speak of a quantitative confirmation of the theory[11-15]. The observed narrowing of the line (by a factor 2.7) is an experimental confirmation of the existence of two-quantum processes near resonance in a gas, and the possibility of observing them in relatively weak fields.

b) Measurements of the effective collision cross sections. The influence of collisions on the line shape of stimulated emission or absorption in a three-level system was investigated by Rautian and Feoktistov[29]. They analyzed the model of strong collisions (as a result of collision, the velocity of the atom lies in the average thermal range) and collisions leading to phase randomization (the Weisskopf broadening mechanism). It turned out that the collisions become manifest in different ways in the forward and backward scattering line shapes. The backward scattering line retains a Lorentz shape. Collisions lead only to a broadening of the line. The line width can in this case be obtained by adding the impact terms to the radiative widths:

$$\Gamma_0 = \Gamma_{ml} + k_\mu \Gamma_{mn} / k; \quad \Gamma_{ml} = \gamma_m + \gamma_l + \delta_{ml} + \nu, \quad \Gamma_{mn} = \gamma_m + \gamma_n + \delta_{mn} + \nu, \tag{14}$$

where ν is the frequency of the strong collisions and δ is the Weisskopf part of the broadening. The forward scattering line retains a Lorentz shape only in the case of strong collisions. In the presence of phase randomization in collisions, the line is the sum of two Lorentz contours with widths Γ_- and Γ_0, and with an amplitude ratio $\Gamma_0(\Gamma_0 - \Gamma_- - \Gamma_m)/\Gamma_-\Gamma_m$[24]. Here Γ_-, Γ_0, and Γ_m include allowance for the collisions

$$\Gamma_- = \Gamma_{nl} + (k_\mu / k - 1)\Gamma_{mn}, \quad \Gamma_{nl} = \gamma_n + \gamma_l + \delta_{nl}. \tag{15}$$

The generally complicated line shape makes it difficult to reduce the experimental curves, particularly when the collision mechanism is not known.

Our investigations of the broadening of the 1.15 μ line in pure neon[24] have shown that an appreciable contribution (on the order of 50%) to the line broadening on the 2s – 2p transitions of neon is made by strong collisions in resonant dipole-dipole interaction. Using the results of these measurements, we have estimated the possible value of the "underlining" with width Γ_0 in the forward scattering line. At a pressure of 1 Torr, the amplitude of the "underlining" should not exceed 10% of the amplitude of the peak with width Γ_-, i.e., the forward line shape can be regarded, with a certain degree of approximation, as a Lorentz shape with width Γ_-. Consequently, data on the broadening of the scattering line contours (Fig. 5) can be used directly to determine the collision cross sections.

The measured value of the forward scattering line broadening turned out to be $2\partial\Gamma_-/\partial p = 17 \pm 2$ MHz/Torr, and the broadening of the backward scattering line was $2\partial\Gamma_0/\partial p = 46 \pm 3$ MHz/Torr. If we use the data on the 1.15 μ line broadening obtained from a reduction of the Lamb dip in a pure Ne laser[24], namely $\partial\Gamma_\mu/\partial p = 8.8 \pm 0.8$ MHz/Torr, then we can determine the broadening of the 1.52 μ line of Ne from the value of $\partial\Gamma_0/\partial p$. It turns out to be equal to $\partial\Gamma/\partial p = 10.8 \pm 2.1$ MHz/Torr. This value agrees sufficiently well with the value obtained earlier[30] in investigations of the broadening of the generation-power peak in a laser with nonlinear absorption, $\partial\Gamma/\partial p = 12.1 \pm 5$ MHz/Torr[5)].

Using the quantities $\partial\Gamma_-/\partial p$ and $\partial\Gamma/\partial p$, we obtain for the forbidden transition $2p_1 \rightarrow 2p_4$ a broadening equal to 5.1 ± 2.6 MHz/Torr. The contributions made by the $2s_2$ level to the broadening of the 1.15 μ and 1.52 μ lines are apparently the same. Therefore the difference $\partial\Gamma/\partial p - \partial\Gamma_\mu/\partial p = 2 \pm 1.8$ MHz/Torr gives the difference between the broadenings of the levels $2p_1$ and $2p_4$. We obtain, as a result, a broadening of 1.5 MHz/Torr for the $2p_4$ level and 3.6 MHz/Torr for the $2p_1$ level. The double resonance method was recently used to measure the cross section for the relaxation of the magnetic moment of the $2p_9$ level from the same electronic configuration for collisions in Ne[33], and a value $\sigma_{\text{Ne-Ne}} = (10.7 \pm 0.3) \times 10^{-15}$ cm^2 was obtained. The cross sections for the broadening of the levels $2p_1$ and $2p_4$, according to the results of our measurements, are 3.6×10^{-15} and 8.6×10^{-15} cm^2, respectively; it is easily seen that the obtained values are of the same order of magnitude.

Thus, an investigation of the forward and backward scattering line broadening has made it possible to determine the broadening cross section not only for spectral lines but also for individual levels. It should be pointed out, however, that this information is obtained relatively simply if the model of the collisions realized in the experiments is known.

c) Measurement of the width of the common level. The investigation of two-quantum transitions in a three-level system opens the possibility for measuring the width of the common level[22]. In the absence of collisions, at $k_\mu > k$ and $\kappa \ll 1$ we have, in accordance with (11) and (12),

$$\Gamma_0 - \Gamma_- = 2\gamma_m. \qquad (16)$$

Thus, measurement of the difference of the line widths in forward and backward scattering gives directly the value of the width of the common level in the absence of any other information on the relaxation. In the presence of only quenching or "strong" collisions, which are due, for example, to resonant transfer of excitation, the relation (16) can be expressed in the form

$$\Gamma_0 - \Gamma_- = 2(\gamma_m + \nu), \qquad (17)$$

where ν is the frequency of the collisions of the atom

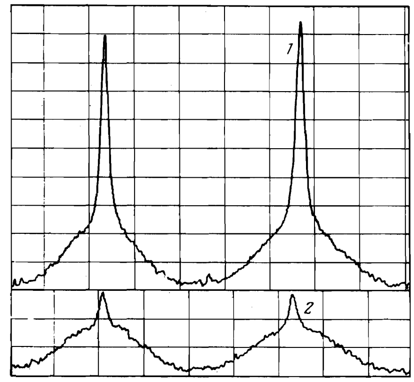

FIG. 6. Plot of the line shape of the resonant SRS in neon for circularly polarized waves. Curves 1 and 2 correspond to orthogonal circular polarizations of the pump field.

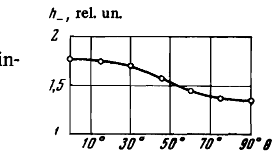

FIG. 7. Polarization anisotropy of the intensity of resonant SRS in neon.

or molecule in the state m. Measurement of the difference of the widths as a function of the pressure makes it possible in this case to measure the cross section for the broadening of the common level.

We have used the experimental results described above to determine the width of the level $2s_2$. Extrapolation to zero values of the pressure and the field yields $2\gamma_m = \Gamma_0 - \Gamma_- = 27 \pm 5$ MHz, i.e., a spontaneous lifetime $\tau = 1/2\gamma_m = (6.1 \pm 1) \times 10^{-9}$ sec, which is in good agreement with the results of direct measurements in which use is made of the technique of delayed coincidences and of registration in the region of the vacuum ultraviolet, for example $\tau = (7.78 \pm 0.8) \times 10^{-9}$ sec[34]. We note that the measurements are more accurate the larger the width of the common level, i.e., the measurement accuracy increases in the investigation of short-lived levels.

d) Polarization investigations. The investigation cannot be complete if the polarization properties of the scattering line are not considered. The experiments have shown that the observed sharp structure is exceedingly sensitive to polarization of both the external field and of the weak sounding signal. Thus, Fig. 6 shows the line shape of the scattering at $\mathbf{k} \cdot \mathbf{k}_\mu > 0$ for a circularly polarized wave at $\lambda = 1.52\ \mu$. The weak field at 1.15 μ also has circular polarization, which in one case is orthogonal to the direction of rotation of the plane of polarization of the strong field (curve 1), while in the other case the directions coincide (curve 2). The intensity ratio at the maximum of the scattering line is 1 : 6, which agrees (within the limits of measurement accuracy) with the intensity ratio of the Zeeman components calculated with the aid of 3j-symbols on the transition $J = 1 \rightarrow J' = 2$ and from the sublevel $M = \pm 1$.

Investigations were also made of the degree of polarization of the scattering line as a function of the azimuthal angle between the polarization planes of the weak and strong fields (Fig. 7). The observed maximum degree of polarization of the scattering line is 25%. The polarization anisotropy of the sharp structure of the line is described with sufficient accuracy by the ratio

$$G_0 / G_0 = 1 - 0.25 \sin^2 \theta, \quad (18)$$

where G_0 is the gain for coinciding linear polarizations.

In the investigated pressure range (0.3–1.5 Torr), the polarization properties of the scattering line remained practically unchanged. This indicates that the reorientation of the magnetic moments of an atom is accompanied by a simultaneous change in its velocity in its own gas under conditions of resonant exchange of excitation. In the case of "weak" collisions, the relaxation of the orientation should lead to a depolarization of the sharp structure of the line.

CONCLUSION

The investigations of the line shape of stimulated resonant Raman scattering in neon on the transitions $2s_2 - 2p$ have shown that the use of two-quantum transitions is highly effective in high-resolution spectroscopy in gases. The extension of the employed methods to include other atomic and molecular systems can yield important information on the width of the lines and individual levels, on collision relaxation, etc. The narrowing of the line in resonant forward Raman scattering in comparison not only with Doppler width but also with the natural width of the transition, which is accompanied by an increase in the gain, can be used to produce a special type of lasers that are pumped with monochromatic radiation, namely resonant Raman lasers. The improved resolution in the investigations of the line structure, as compared with population effects, can be used in a number of cases to determine atomic and molecular constants, nuclear moments, etc., to register and measure weak electric and magnetic fields, and finally, for spectroscopy not only within the Doppler width but also within the natural width.

[1)] The difference between these elementary processes lies in the fact that, unlike the stepwise transition, the probability of the two-quantum transition cannot be formally represented in the form of a product of the probabilities of absorption and emission of individual protons [6,7].

[2)] The applicability of perturbation theory is determined by the necessary (but generally not sufficient) condition $|a_m^{(1)}| << 1$. If the relaxation constants γ_m and γ_n are close to each other and $\Omega = 0$, then, expanding the indeterminate form in (3), we obtain at $\gamma_m = \gamma_n = \gamma_0$ the condition $|V|/\gamma_0 << 1$ in a form that coincides with that used by Lamb for a two-level system in the theory of the gas laser [27]. Popova et al. [7], who analyzed resonant radiative processes in a three-level system, used the smallness condition $|V| << |\Omega - i\gamma|$, where $\gamma = \gamma_n - \gamma_m$. At $\Omega = 0$ and $\gamma_n = \gamma_m$, this condition becomes meaningless.

[3)] The analysis has been carried out for the case $\gamma_l << \gamma_m$. Analogous results are obtained when $\gamma_l >> \gamma_m, \gamma_n$.

[4)] However, atoms in different parts of the volume interact with fields of different amplitude. On averaging over the volume in the case of strong fields, this influences the emission-line shape.

[5)] We call attention to the strong difference between the data obtained here on the broadening of the 1.15 and 1.52 μ lines and the results of Szoke and Javan [31], where the measurements were made with an He-Ne mixture. This difference was already discussed in detail [32] and was attributed to the specific influence of the elastic scattering of the neon atoms in collisions with neon atoms in the He-Ne mixture.

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