Nonlinear effects in resonant *SRS* line in a gas in the field of a standing wave

E. V. Baklanov, I. M. Beterov, B. Ya. Dubetskiĭ, and V. P. Chebotaev

Institute of Semiconductor Physics, Siberian Division, USSR Academy of Sciences
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We report experimental observation of a resonance of a new type in multiphoton transitions in a standing-wave field in a Raman gas laser. The theory shows that the resonance takes place in third-order perturbation theory, when the resonant *SRS* line is inhomogeneously broadened and can be regarded as an effect overlapping the dips in the velocity distribution of the nonlinear polarization in the field-induced standing wave.

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Interest has recently increased in resonances of multiphoton absorption in a gas in the field of a standing wave.[1] These resonances have properties of interest to spectroscopy and to various applications (see[2]). We report here observation of a new type of resonance, due to saturation of the resonant SRS line in the field of a standing wave as a result of multiphoton transitions.

It is known[3] that under the influence of a field of frequency ω at resonance with the $1\to 0$ transition there appear on the neighboring transition $0\to 2$ (Fig. 1) narrow lines of stimulted emission, with widths Γ_+ and Γ_-

$$\Gamma_+ = \gamma_1 + \gamma_2 + \left(\frac{\omega_{02}}{\omega_{01}} - 1\right)(\gamma_1 + \gamma_0);$$
$$\Gamma_- = \gamma_0 + \gamma_2 + (\omega_{02}/\omega_{01})(\gamma_1 + \gamma_0), \tag{1}$$

where γ_1, γ_0, and γ_2 are the damping rates of the corresponding levels. The index (+) corresponds to the case when the incident and scattered waves propagate in the same direction, and (−) to the opposite case. The resonances are located at the frequencies

$$\omega'_\pm = \omega_{02} \pm (\omega_{02}/\omega_{01})(\omega - \omega_{01}). \tag{2}$$

The resonance described in this paper takes place when the frequency difference $\omega' - \omega$ is equal to the frequency of the forbidden transition. Its width γ is equal to the width of the forbidden transition, $\gamma = \gamma_1 + \gamma_2$. It follows from our theoretical analysis that it occurs in third-order perturbation theory and can be regarded as the consequence of saturation effects in multiphoton transitions, when the SRS line is inhomogeneously broadened. This phenomenon is of interest for the stabilization of the frequency of gas lasers, for laser spectroscopy, and for the measurement of the frequencies of forbidden transitions.

The resonant dip at the center of the line is observed on the dependence of the generation power on the frequency of a neon laser at a wavelength 1.15 μ ($2s_2 - 2p_4$ transition) when pumped by a field at 1.5 μ on the neighboring transition $2s_2 - 2p_1$. Lasing in such a system and the waveform of the SRS line were investigated in[4,5]. It was shown in[5] that the line shape is determined mainly by two-quantum transitions. The gain line in the laser has a characteristic anisotropy. When ω is detuned from the line center and the lasing frequency is scanned, two peaks are observed, each corresponding to the gain line of one of the traveling waves on the $0\to 2$ transition (Fig. 2). Near the line center, the character of the generation changes. A shallow but sharply outlined dip appears in the generation line, with a width ~10 MHz close to the width of the two-photon transition $2p_1 - 2p_4$ (see Fig. 3). We note that the width of the Lamb dip on the 1.15 μ line is of the order of 45 MHz under these conditions. Let us explain the observed phenomenon and present the results of the calculation. The probability of the transition of the atom from level 1 to level 2 is given by

$$W_{1\to 2} = \frac{2|G|^2|G'|^2}{[(\gamma_1+\gamma_0)/2]^2+\Omega^2}\,\mathrm{Re}\left\{\frac{1}{\gamma_1}\frac{1}{[(\gamma_1+\gamma_2)/2]+i(\Omega'-\Omega)} + \frac{1}{\gamma_0}\frac{1}{[(\gamma_0+\gamma_2)/2]+i\Omega'} + \frac{1}{\{[(\gamma_1+\gamma_2)/2]+i(\Omega'-\Omega)\}\{[(\gamma_0+\gamma_2)/2]+i\Omega'\}}\right\}, \tag{3}$$

where $\Omega = \omega - \omega_{01}$, $\Omega' = \omega' - \omega_{02}$, $G = d_{01}E/(2\hbar)$, $G' = d_{02}E'/(2\hbar)$, and d_{01} and d_{02} are the matrix elements of the transitions $0\to 1$ and $2\to 0$. As already noted in[5], consideration of only two-quantum transitions under resonance conditions is possible when $\gamma_0 \gg \gamma_1$. In the general case, alongside the two quantum processes of the SRS type, there take place in the scheme under consideration also one-quantum stepwise transitions (the second term of (3)), and the emission line is determined by the interference of these processes. At $\Omega \sim 0$ and $\Omega' \sim 0$, and at $\gamma_0 \gg \gamma_1$, the second and third terms can be neglected, and the transition $1\to 2$ can be regarded as resonant SRS. The emission line of the individual atom is

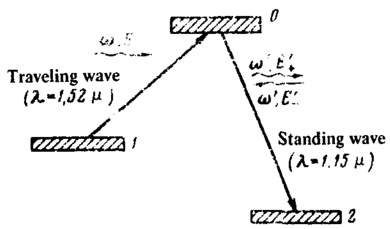

FIG. 1. Level scheme.

FIG. 2. Waveform of generation line at a pump-wave frequency detuning $\Omega = 50$ MHz.

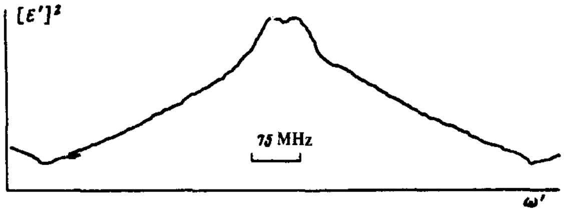

FIG. 3. Resonant dip on the frequency dependence of the generation power when the pump wave is tuned to the line center (the broad part of the line is due to diffusion of the excitation in velocity space as a result of dragging of the resonant radiation.[4]

of width $\gamma = \gamma_1 + \gamma_2$, much smaller than for the emission of the ensemble of atoms. At $\omega_{02}/\omega_{01} > 1$, the forward and backward SRS lines thus experience strong inhomogeneous broadening.

The transition probability for atoms having a projection of the velocity v in the standing-wave field is given by

$$W_{1\to 2}(v) = \frac{2|G|^2\,|G'|^2}{\left(\frac{\gamma_1+\gamma_0}{2}\right)^2 + \left(\Omega - \frac{\omega}{c}v\right)^2} \times \operatorname{Re}\sum_{+,-}\left[\frac{1}{\gamma_1}\;\frac{1}{\frac{\gamma_1+\gamma_2}{2} + i\left(\Omega' \mp \frac{\omega'}{c}v - \Omega + \frac{\omega}{c}v\right)}\right]. \quad (4)$$

Depending on the velocity of the atoms, the probability of the two-quantum transition has two sharp maxima. Accordingly, two peaks appear at the level 2 in the velocity distribution of the atoms, with width $\Delta v \sim (\gamma/\omega)c$. From (4) we easily obtain the velocities that are at resonance with the two-quantum process:

$$v_\pm = \frac{\Omega' - \Omega}{\pm\omega' - \omega}\,c. \quad (5)$$

The SRS saturation effect is obviously due to the inverse transition from the level 2 and is determined by the change of the population of level 2. At $\Omega' - \Omega > \gamma$, the two oppositely traveling waves interact with different atoms. At $\Omega' - \Omega \lesssim \gamma$ both waves interact with the same atoms, i.e., the nonlinear polarization induced in the medium by one wave contributes to the absorption of the other wave on the $0 - 2$ transition.

A rigorous theoretical analysis in third-order perturbation theory yields the following expression for the gain α in the $0 \to 2$ transition:

$$\alpha = \alpha_0\left\{1 - \kappa\left[\phi(\Omega) + \frac{\eta^2 - 1}{4\eta^2}\,\frac{\gamma_2\Gamma(\gamma\Gamma + \Omega(\Omega' - \Omega))}{(\Gamma^2 + \Omega^2)(\gamma^2 + (\Omega' - \Omega)^2)}\right]\right\} \quad (6)$$

where $\phi(\Omega)$ is a smooth function of Ω and has a maximum of unity at $\Omega = 0$,

$$\kappa = \frac{4|G'|^2}{\gamma_2\Gamma}, \quad \Gamma = \frac{\gamma_0}{2},$$

$$\alpha_0 = \frac{8\pi^{3/2} N |d_{02}|^2 G^2 \eta^2}{\hbar\bar{v}(\eta^2 - 1)(\Gamma^2 + \Omega^2)}$$

$\eta = \omega'/\omega > 1$, and $\bar{v}$ is the average thermal velocity. Expression (6) describes the dip as a function of the difference $\Omega' - \Omega$ between the detunings. Its amplitude depends on the relaxation constants and on the frequency detuning Ω. At $\Omega \gg \Gamma$ the resonant dip with width γ vanishes. Nor does it appear at $\omega' \leq \omega$, since there is no overlap of the regions of the interacting atoms at any value of Ω or Ω'. In addition, at $\omega' \approx \omega$ the forward SRS line is homogeneously broadened.

Analysis shows that in the case of two standing waves in each transition the described dip in the SRS line will be observed at all ω' and ω. Taking into account the progress attained in the development of tunable lasers, we can conclude that in practice, in any transition in the optical band, it is possible to obtain lasing via a three-level scheme. Therefore the phenomenon described here can play an important role in precision spectroscopy of optically-forbidden transitions. It is possible to use this method, as well as two-photon absorption resonance, in experiments on the exact measurement of the frequency of the $1S - 2S$ transition of the hydrogen atom, and of the frequencies of vibrational-rotational transitions that are inactive in the infrared region of the spectrum. In some cases, in comparison with two-photon absorption resonance, the described phenomenon can offer advantages, for there is no need for high-power radiation sources under conditions of exact resonance.

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