

# Nonlinear Resonances Free of Field and Doppler Broadenings

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**Abstract**—For the case where the Rabi frequencies of the guiding fields are much larger than the relaxation constants but much smaller than the Doppler broadening, it is shown that resonances which are neither field nor Doppler broadened can appear in the absorption (or gain) spectrum of the probe field. A classification of four-level systems according to the number of resonances is made for cases where two strong fields interact either with opposite or adjoining transitions. The conditions under which the number of resonances reaches eight, while for stationary atoms the maximum number is four, are found. A method is proposed for calculating the number of resonances in a multilevel system with several strong fields using analysis of the extremum points of the frequency branches in the velocity–frequency plane. © 2000 MAIK “Nauka/Interperiodica”.

## 1. INTRODUCTION

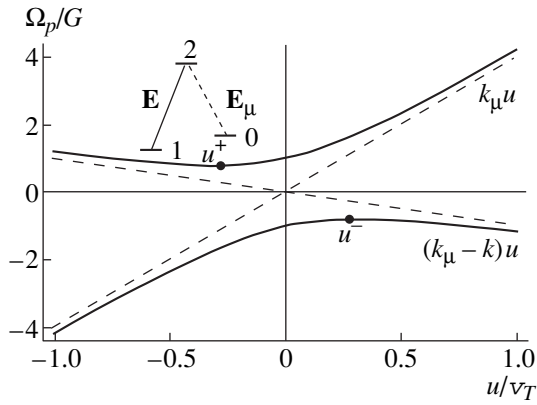
One of the basic problems of nonlinear optics is to develop methods for increasing the frequency of coherent radiation in gases. Among continuous-conversion schemes with an appreciable decrease of wavelength which have been developed, the three-level scheme of an anti-Stokes Raman laser [1, 2] and the four-level scheme of resonant four-wave mixing [3, 4] are of special interest. Doppler broadening decreases the conversion efficiency, but it can be compensated in many cases. It is important to know the resonance nonlinear susceptibility in gases in order to estimate the light conversion efficiency. Moving away from problems of propagation of an electromagnetic wave in a medium and making the assumption that the gas layer is optically thin, the calculation of the nonlinear susceptibility of  $n$ -level system reduces to solving an algebraic equation of degree  $n$  and averaging the absorbed power over the particle velocities [5]. Such averaging in calculation of the spectrum of the probe field in a three-level system interacting with a strong field on an adjoining transition was performed in the 1970s (see, for example, [6]). A characteristic feature of the spectrum found was that if the probe field was the longer wavelength field, then a doublet of narrow resonances remained after averaging.

In the absence of a strong field the resonance frequency is a linear function of the particle velocity because of the Doppler shift. To each group of particles moving with the same velocity there corresponds a unique resonance frequency. A strong field splits these resonances, and the magnitude of the splitting is also found to depend on the velocity. The split resonances are shifted from Bohr transition frequency to the Rabi frequencies, which become nonlinear functions of the velocity. Integration over the velocities signifies sum-

mation of “group spectra,” each of which is a response of a group of particles moving with a prescribed velocity. In [7] it was discovered that the velocity-averaged susceptibility grows rapidly when the detuning of the probe-field frequency is equal to the Rabi frequency  $\Omega_R$  at the extremum points of its velocity dependence. These spectral peaks are due to the fact that the susceptibility is proportional to the number of particles interacting resonantly with the field. The number of particles with the same shift of the resonance frequency is maximum at the extremal points. We shall call these extrema the turning points.

The contribution of an individual turning point to the probe-field spectrum in a three-level system has been calculated in [9] taking account of weak collisions with a change in the velocity. In [10] a scheme with resonant four-wave mixing with two strong fields on opposite transitions of a four-level system and two weak fields was examined. The asymptotic behavior of the velocity integral was found for the symmetric case of equal wave numbers of the weak fields and zero detunings of the strong fields. It was found that the number of peaks in the spectrum can increase compared with the case of stationary atoms. It was also shown that merging of the turning points leads to a large increase in the radiation conversion coefficient [11].

Numerical averaging over the velocities in a multilevel system (see, for example, [12, 13]) does not permit analyzing the dependence of the conversion coefficient on the relaxation constants, detunings, wave numbers, wave intensities, and spontaneous decay probability, since the problem is multiparametric. It is virtually impossible to obtain a general analytical expression for the work performed by the probe field in a multilevel system. The absorbed power can be written in a relatively compact form only for certain cases, spe-



**Fig. 1.** Frequency branches of a three-level system in the  $(u, \Omega_p)$  plane:  $k v_T = 5G$ ,  $k_\mu v_T = 4G$ ,  $\Omega = 0$ . Dashed straight lines—asymptotes of the frequency branches. The inset shows the level scheme; the solid line denotes the strong field  $\mathbf{E}$ ; the dashed line denotes the probe field  $\mathbf{E}_\mu$ . The turning points are shown on the frequency branches.

cifically, for a four-level system with strong fields on opposite transitions. In more complicated cases the problem reduces to solving a cubic or higher-degree equation. However, these difficult calculations can be avoided if one is interested only in the qualitative behavior of the spectrum of the probe field. The most informative part of the spectrum are the narrow resonance peaks. The problem becomes solvable if attention is confined only to the calculation of these peaks.

In the present paper we present a method for determining the number of narrow peaks in the probe-field spectrum for four-level systems with large Doppler and small homogeneous broadenings. For this the concept of frequency branches and turning points are presented in Section 2 for a three-level system interacting resonantly with one strong and one probe field. Four-level schemes with two strong fields on opposite or adjoining transitions in Section 3 are analyzed and the conditions for the existence of narrow resonances in the probe-field spectrum, when all waves are codirectional, are found. A classification of these schemes according to the number of peaks is presented. The results of the numerical calculation of a spectrum with eight peaks are presented in Section 4. The results obtained are discussed and compared with data obtained by other authors. Brief recommendations for checking the predicted effects experimentally are contained in the conclusions. An approximate expression for the work performed by the probe field in multilevel systems with large Doppler broadening is presented in the Appendix.

### 2. THREE-LEVEL SYSTEM

We shall consider a three-level system exposed to a strong electromagnetic wave  $\text{Re}\mathbf{E}\exp[i(kx - \omega t)]$  in resonance with a transition between levels 1 and 2. The energy, absorbed by such a system, of a probe wave

$\text{Re}\mathbf{E}_\mu\exp[i(k_\mu x - \omega_\mu t)]$  in resonance with a transition between levels 2 and 0 can be written as a sum of two terms resonant with respect to  $\omega_\mu$ . If the upper level is denoted by the index 2,  $E_2 > E_1$  and  $E_0$  (see inset in Fig. 1), then the expression for the work performed by the field  $P_\mu(\omega_\mu)$  per unit time [5] can be written as a sum of elementary fractions:

$$P_\mu(\omega_\mu) = 2\hbar\omega_{20}|G_\mu|^2 = \int_{-\infty}^{\infty} \frac{du}{2\Omega_R} \text{Re} \left[ \frac{\lambda^- N_{20}(u) + |G|^2 N_{21}(u)/(2\Omega_R - i\Gamma)}{\Gamma - i(\Omega'_\mu - \lambda^+)} - \frac{\lambda^+ N_{20}(u) - |G|^2 N_{21}(u)/(2\Omega_R + i\Gamma)}{\Gamma - i(\Omega'_\mu - \lambda^-)} \right], \quad (1)$$

where  $\omega_{20} = (E_2 - E_0)/\hbar$ ,  $\omega_{21} = (E_2 - E_1)/\hbar$  are the Bohr transition frequencies,

$$N_{ij}(u) = \frac{1}{\sqrt{\pi} v_T} (N_i - N_j) \exp\left(-\frac{u^2}{v_T^2}\right)$$

is the equilibrium distribution of the population difference with temperature  $T = m v_T^2/2$ ,  $m$  is the mass of the particles,  $v_T$  is the thermal velocity,  $u$  is the projection of the particle velocity on the direction of propagation of a strong wave, and  $N_j$  is the population of the  $j$ th level,  $G = \mathbf{E} \cdot \mathbf{d}_{21}/2\hbar$  and  $G_\mu = \mathbf{E}_\mu \cdot \mathbf{d}_{20}/2\hbar$  are the Rabi frequencies, which are proportional to the field amplitudes,  $\Omega_\mu = \omega_\mu - \omega_{20}$  and  $\Omega = \omega - \omega_{21}$  are the detunings of the frequencies of the electromagnetic fields from the Bohr transition frequencies, and  $\Gamma$  are the relaxation constants, which we have chosen, for simplicity, to be the same for all levels and which we assume to be small. The quantities  $\lambda^\pm$  are related with the splitting of the energy of level 2 by a strong electromagnetic field (so-called energies of dressed states):

$$\lambda^\pm = \frac{\Omega'}{2} \pm \Omega_R, \quad \Omega_R = \sqrt{\frac{\Omega'^2}{4} + |G|^2},$$

where  $\Omega_R$  is the generalized Rabi frequency of the strong field taking account of the Doppler shift  $\Omega' = \Omega - ku$  and  $\Omega'_\mu = \Omega_\mu - k_\mu u$ . Both terms in the integrand in Eq. (1) describe a resonance of the probe field with a transition between one of the dressed states of the subsystem 1–2 and the state 0.

We are interested in the qualitative form of the spectrum of the probe field in nondegenerate systems with large Doppler broadening and located in strong fields. We shall consider the situation where the Rabi frequency is much larger than the relaxation constants but much smaller than the Doppler width of a line:

$$\Gamma \ll G \ll k v_T. \quad (2)$$

The work performed by the probe field in resonance with a transition between two levels, which belong to different (not coupled with one another by strong fields) subsystems of a multilevel system, can always be written as a sum of resonant, with respect to  $\Omega_\mu$ , terms, similarly to the expression (1) (see Appendix). Consequently, it is convenient to start the explanation of the turning-point method, used below for analyzing the spectrum in multilevel systems, for a three-level system for which the work performed by the field can be calculated explicitly.

Since the conditions of a maximum of the integrand in Eq. (1),

$$\Omega_\mu = \Omega_p^\pm(u) \equiv \lambda^\pm(u) + k_\mu u, \quad (3)$$

depend on the particle velocity  $u$ , in the general case velocity averaging decreases the amplitude  $P_\mu$  by the factor  $\Gamma/kv_T$ . The decrease in the work performed by the field is due to the fact that only a negligible fraction of particles ( $\sim \Gamma/kv_T$ ) interacts resonantly with the probe field. The only exception is the case where the condition (3) holds simultaneously with the condition for an extremum of the function

$$\frac{d\Omega_p^\pm}{du} = k_\mu - \frac{k}{2} \pm \frac{k(ku_p - \Omega)}{2\sqrt{(\Omega - ku_p)^2 + 4|G|^2}} = 0, \quad (4)$$

which means that when the particle velocity varies near  $u_p$  the condition (3) continues to hold within the terms which are quadratic in the deviation  $(u - u_p)$ . The number of points interacting with the field increases rapidly and hence sharp peaks should be observed in the spectrum of the probe field  $P_\mu(\omega_\mu)$  at  $\Omega_\mu = \Omega_p^\pm(u_p^\pm)$ . From Eq. (4) we find the velocity

$$ku_p^\pm = \Omega \pm |G| \frac{k - 2k_\mu}{\sqrt{k_\mu(k - k_\mu)}}, \quad (5)$$

and substituting it into Eq. (3) we find the position of the resonances in the spectrum [6, 7]

$$\Omega_\mu^\pm = \Omega_p^\pm(u_p^\pm) = \frac{k_\mu}{k} \Omega \pm 2|G| \frac{\sqrt{(k - k_\mu)k_\mu}}{k}. \quad (6)$$

In multilevel systems the functions  $\Omega_p(u)$ , which determine the value of the resonance frequency as a function of the particle velocity, have a form which makes it impossible to solve Eqs. (3) and (4) analytically. We shall formulate a graphical method for calculating the peaks in the spectrum of the probe field. We plot in the  $(u, \Omega_p)$  plane the curves  $\Omega_p^\pm(u)$  (3), which we call frequency branches (Fig. 1). The condition (4) gives in the plane  $(u_p^\pm, \Omega_p^\pm(u_p^\pm))$  the ‘‘turning points’’ where the frequency branch possesses an extremum. Thus, in order to determine the position of resonances in the probe-field spectrum, it is sufficient to represent

the frequency branches  $\Omega_p(u)$  in the  $(u, \Omega_p)$  plane and find the turning points where  $d\Omega_p/du = 0$ . For a three-level system there are only two frequency branches,  $\Omega_p^\pm(u)$ , each of which is a branch of a hyperbola, and consequently there can be no more than one turning point on each of them. It is evident from Fig. 1 that resonances in the spectrum which are associated with the turning points can be observed only if the slopes of the asymptotes of the frequency branches,  $\Omega_p(u) \sim k_\mu u$  and  $\Omega_p(u) \sim (k_\mu - k)u$ , have different signs, i.e.,  $k_\mu(k_\mu - k) < 0$ . This result also follows from the expression (6), which gives a real frequency only for  $k_\mu(k - k_\mu) > 0$ . The asymptotic values of the slopes of the frequency branches are related with the relative arrangement of the energy levels in the three-level system:

$$k_\mu \propto \frac{\omega_{20}}{c} = \frac{E_2 - E_0}{\hbar c}, \quad k_\mu - k \propto \frac{\omega_{10}}{c} = \frac{E_1 - E_0}{\hbar c}.$$

Analyzing the expression (1) for the probe-field spectrum for an arbitrary scheme of levels, it is evident that the slopes of the asymptotes have different signs if

$$(E_2 - E_0)(E_1 - E_0) < 0. \quad (7)$$

In other words, the energy  $E_0$  must fall between the levels  $E_1$  and  $E_2$ : these are the so-called Stokes  $\Lambda$  and  $V$  schemes. If the condition (7) does not hold (anti-Stokes schemes), then the expression (6), determining the position of the turning points, is imaginary and therefore there will be no narrow resonances in the spectrum. The conditions for the existence of narrow resonances when electromagnetic waves propagate opposite to one another can be found similarly:

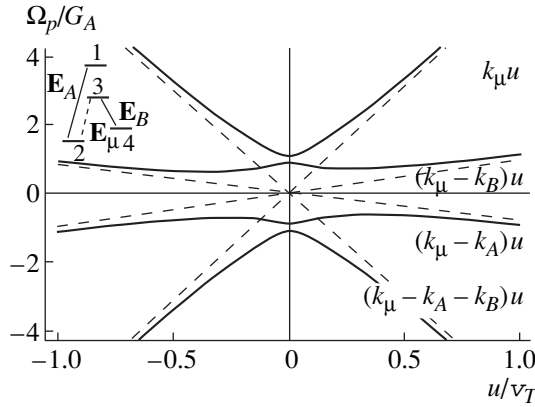
$$k_\mu(k_\mu + k) < 0, \quad |2E_0 - 3E_2 + E_1| < |E_2 - E_1|.$$

The shape of the spectrum near the resonance  $\Omega_\mu^\pm$  can be found in the approximation (2), using only the fact that the first derivative of  $\Omega_p^\pm(u)$  vanishes at the point  $u = u_p$ . Neglecting all terms which are small to the extent that  $G/kv_T$  and  $\Gamma/G$  are small, we obtain

$$P_\mu(\Omega_\mu) \approx 2\pi\hbar\omega_{20}|G_\mu|^2 \times \left[ \frac{\Gamma^+(\Omega_\mu)}{2\Omega_R(u_p^+)} \left( \lambda^-(u_p^+)N_{20}(u_p^+) + \frac{|G|^2 N_{21}(u_p^+)}{2\Omega_R(u_p^+)} \right) - \frac{\Gamma(\Omega_\mu)}{2\Omega_R(u_p^-)} \left( \lambda^+(u_p^-)N_{20}(u_p^-) - \frac{|G|^2 N_{21}(u_p^-)}{2\Omega_R(u_p^-)} \right) \right], \quad (8)$$

where

$$I^\pm(\Omega_\mu) = \frac{1}{\pi} \text{Re} \int_{-\infty}^{\infty} \frac{du}{\Gamma - i(\Omega_\mu - k_\mu u - \lambda^\pm(u))}. \quad (9)$$



**Fig. 2.** Frequency branches of a four-level system in the  $(u, \Omega_p)$  plane:  $k_A v_T = 7G_A$ ,  $k_B v_T = 5G_A$ ,  $k_\mu v_T = 6G_A$ ,  $G_B = 0.1G_A$ ,  $\Omega_A = \Omega_B = 0$ . Dashed straight lines—asymptotes of the frequency branches. The inset shows the four-level system with two strong fields on opposite transitions; the solid lines denote the strong fields  $\mathbf{E}_A$  and  $\mathbf{E}_B$ ; the dashed lines denote the probe field  $\mathbf{E}_\mu$ .

The derivation of the expression (8) used the fact that the width of the integrand in Eq. (1) of order  $\Gamma$  is determined by the denominator, and consequently the remaining expression can be removed from the integral at the points  $u = u_p^+$  or  $u = u_p^-$  in accordance with the indices + or – in Eq. (9).

Expanding the function  $\Omega_p^\pm(u)$  in the power series in  $u - u_p^\pm$  near the turning points  $u = u_p^\pm$  and retaining quadratic terms

$$\Omega_p^\pm = \Omega_\mu^\pm + a^\pm (u - u_p^\pm)^2 + \dots, \quad (10)$$

where the letters  $a^\pm \neq 0$  denote second derivatives at the turning points

$$a^\pm = \left. \frac{1}{2} \frac{d^2 \Omega_p^\pm}{du^2} \right|_{u=u_p^\pm} = \pm \frac{k_\mu^{3/2} (k - k_\mu)^{3/2}}{|G|k},$$

we obtain the profile of an isolated peak, which was first found in [6]:<sup>1</sup>

$$I^\pm(\Omega_\mu) = \text{Re} \sqrt{\frac{\pm k |G|}{k_\mu^{3/2} (k - k_\mu)^{3/2} (\Omega_\mu - \Omega_\mu^\pm + i\Gamma)}}. \quad (11)$$

The spectral peaks given by Eq. (11) have a width of the order of  $\Gamma$ , their amplitude is proportional to  $\sqrt{|G|/\Gamma}$ , and they have a characteristic asymmetric shape.

As  $k_\mu \rightarrow k$  the formula (11) becomes inapplicable. Consequently, the condition (2) must be replaced by a more stringent condition  $G\sqrt{k_\mu(k - k_\mu)}/k \gg \Gamma$ , which

<sup>1</sup> The Eq. (54) of [6] contains a misprint: instead of 1/2 the exponent is written as 2.

means a large field-induced splitting (6) compared with the relaxation constant. The condition that the field splitting is small compared with the Doppler width (2) is also replaced by a stronger condition, following from Eq. (5). The velocities at the turning points  $u_p^\pm$  must be less than the thermal velocity  $v_T$ :  $|u_p^\pm| \ll v_T$ .

### 3. FOUR-LEVEL SYSTEM

#### 3.1. Four-Level System 2 + 2

Let us consider a four-level system interacting with two strong electromagnetic fields,  $G_A$  and  $G_B$ . If the fields are in resonance with opposite transitions, then the system can be represented in the form of two independent two-level subsystems (we shall call such a system 2 + 2). However, if the fields interact with neighboring transitions, then the system consists of a three-level system and an individual level (we shall call such a system 3 + 1). We begin by analyzing the first case (see inset in Fig. 2). The work performed by the probe field  $\text{Re} \mathbf{E}_\mu \exp[i(\omega_\mu t - k_\mu x)]$  in resonance with the transition 2–3 can be written as a sum

$$P_\mu(\Omega_\mu) = \sum_{I, J=1, 2} P^{IJ}(\Omega_\mu), \quad (12)$$

where

$$P^{IJ}(\Omega_\mu) = \text{Re} \int_{-\infty}^{\infty} \frac{C^{IJ}(u) du}{\Gamma - i(\Omega_\mu - k_\mu u - \lambda_A^I(u) + \lambda_B^J(u))},$$

$$\lambda_A^{1,2} = \frac{1}{2}(\Omega_A - k_A u \pm \sqrt{(\Omega_A - k_A u)^2 + 4|G_A|^2}), \quad (13)$$

$$\lambda_B^{1,2} = -\frac{1}{2}(\Omega_B - k_B u \pm \sqrt{(\Omega_B - k_B u)^2 + 4|G_B|^2}),$$

and  $\lambda_A^I$  and  $\lambda_B^J$  are the energies of the “dressed” states of independent two-level systems. The designations  $I$  and  $J$  of the frequency branches are introduced in the Appendix. The derivation of the formula (12) and the determination of the coefficients  $C^{IJ}$  are also presented in the Appendix.

To find the number of resonances in the probe-field spectrum, we represent the frequency branches in the  $(u, \Omega_p)$  plane, for example, at  $\Omega_A = \Omega_B = 0$  (Fig. 2). Since the position of the resonances in the spectrum is determined by the values of the extrema of the frequency branches, we shall analyze the condition for vanishing of the derivatives:

$$2 \frac{d\Omega_p^{IJ}(u)}{du} = \Delta k + \kappa^{IJ}(u) = 0, \quad (14)$$

$$\begin{aligned} \kappa^{12}(u) &= -\kappa^{21}(u) \\ &= \frac{k_B(\Omega_B - k_B u)}{\sqrt{(\Omega_B - k_B u)^2 + 4|G_B|^2}} + \frac{k_A(\Omega_A - k_A u)}{\sqrt{(\Omega_A - k_A u)^2 + 4|G_A|^2}}, \\ \kappa^{11}(u) &= -\kappa^{22}(u) \\ &= \frac{k_B(\Omega_B - k_B u)}{\sqrt{(\Omega_B - k_B u)^2 + 4|G_B|^2}} - \frac{k_A(\Omega_A - k_A u)}{\sqrt{(\Omega_A - k_A u)^2 + 4|G_A|^2}}, \end{aligned}$$

where  $\Delta k = 2k_u - k_A - k_B$ . The values of the function  $\kappa^{IJ}(u)$  are limited by the region  $|\kappa^{IJ}(u)| < k_A + k_B$ , and consequently Eq. (14) does not possess any solutions for  $|\Delta k| > k_A + k_B$ . The function  $\kappa^{12}(u)$  (and also the function  $-\kappa^{21}(u)$ ) is monotonic and in the limit  $u \rightarrow \pm\infty$  approaches the asymptotic values  $\mp(k_A + k_B)$ . Consequently, when

$$|\Delta k| < k_A + k_B \quad (15)$$

the frequency branches  $\Omega_p^{12}$  and  $\Omega_p^{21}$  each possess a turning point. The function  $\kappa^{11}(u)$  (just as  $-\kappa^{22}(u)$ ) approaches  $\pm(k_A - k_B)$  in the limit  $u \rightarrow \pm\infty$  but it is not monotonic. It possesses two extrema, which can be found from the condition  $d^2\Omega_p^{11}/du^2 = d\kappa^{11}/du = 0$ :

$$u_{\pm} = \frac{\Omega_A}{k_A} - \frac{\gamma k_B}{k_A} \Delta \pm \sqrt{\Delta^2 + Q}. \quad (16)$$

Here

$$\begin{aligned} \kappa^{11}(u_{\pm}) &= -\kappa^{22}(u_{\pm}) \\ &= \frac{k_A k_B \Delta (1 - \gamma) \pm (k_A^2 - \gamma k_B^2) \sqrt{\Delta^2 + Q}}{\sqrt{\Delta^2 (k_A^2 + \gamma^2 k_B^2) + M \mp 2\gamma k_A k_B \Delta \sqrt{\Delta^2 + Q}}}, \end{aligned} \quad (17)$$

where

$$\begin{aligned} \Delta &= \gamma \frac{k_A \Omega_B - k_B \Omega_A}{k_A^2 - \gamma^2 k_B^2}, \quad M = 4\gamma^2 \frac{|G_B|^2 k_A^2 - |G_A|^2 k_B^2}{k_A^2 - \gamma^2 k_B^2}, \\ Q &= 4 \frac{\gamma^2 |G_B|^2 - |G_A|^2}{k_A^2 - \gamma^2 k_B^2}, \quad \gamma = \left( \frac{|G_A| k_A}{|G_B| k_B} \right)^{2/3}. \end{aligned}$$

Hence it follows that the frequency branch  $\Omega_p^{11}(u)$  (or the branch  $\Omega_p^{22}(u)$ ) possesses a turning point for  $u < u_{-}$ , if  $\Delta k$  lies in the interval between  $k_A - k_B$  and  $\kappa^{11}(u_{-})$  (or between  $k_B - k_A$  and  $\kappa^{22}(u_{-})$ ). If  $\Delta k$  falls between  $\kappa^{11}(u_{-})$  and  $\kappa^{11}(u_{+})$  (or between  $\kappa^{22}(u_{-})$  and  $\kappa^{22}(u_{+})$ ), then the frequency branch  $\Omega_p^{11}(u)$  ( $\Omega_p^{22}(u)$ ) possesses an extremum in the range  $u_{-} < u < u_{+}$ . Finally, for  $\Delta k$  between  $\kappa^{11}(u_{+})$  and  $k_B - k_A$  (or between  $\kappa^{22}(u_{+})$  and  $k_A - k_B$ ) a turning point arises  $u > u_{+}$ .

The ranges of  $\Delta k$  enumerated above can intersect, so that each frequency branch,  $\Omega_p^{11}(u)$  and  $\Omega_p^{22}(u)$ , can

have up to three turning points. Thus, the total number of turning points on all frequency branches and hence the maximum number of resonances in the spectrum of the probe field can reach 8. For  $\Delta^2 + Q < 0$  the expression (16) is complex, i.e., the functions  $\kappa^{11}(u)$  and  $\kappa^{22}(u)$  become monotonic. In this case the frequency branches  $\Omega_p^{11}(u)$  and  $\Omega_p^{22}(u)$  each contain one extremum for  $|\Delta k| < |k_A - k_B|$ .

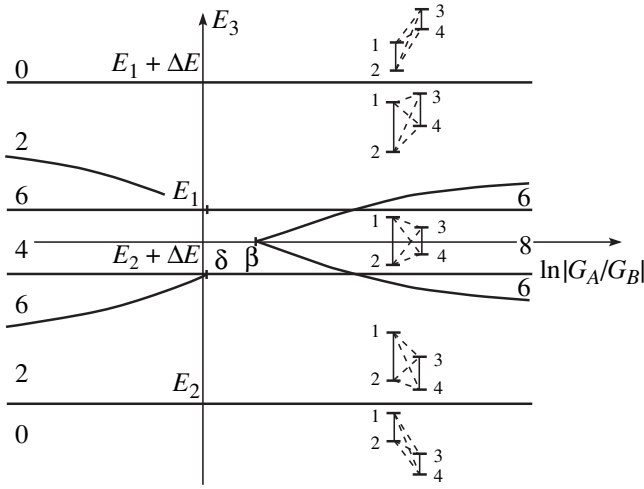
We shall investigate two limiting cases in greater detail:  $\Delta^2 \gg |Q|$ —the difference of the detunings of strong fields is large compared with their Rabi frequencies—and the opposite limit  $\Delta^2 \ll |Q|$ . In the limit  $|Q| \ll \Delta^2$  it follows from the expression (17) and Eq. (14) that for  $|\Delta k| < |k_A - k_B|$  the frequency branches  $\Omega_p^{11}(u)$  and  $\Omega_p^{22}(u)$  each possess one extremum. In the interval  $|k_A - k_B| < |\Delta k| < k_A + k_B$  either  $\Omega_p^{11}(u)$  or  $\Omega_p^{22}(u)$  possesses two extrema. Thus, in the limit of a large difference of the detunings  $\Delta\Omega = \Omega_A - \Omega_B k_A/k_B$  for strong fields the probe-field spectrum contains four resonances if the inequality (15) is satisfied. This condition can be formulated in an invariant form which is valid for any level scheme: if the energy of at least one level of the second subsystem lies between the energies of the levels of the first subsystem, then the number of resonances is 4. In the opposite case there will be no narrow resonances.

When the detunings of strong fields are in resonance with one another,  $\Delta = 0$ , the expressions (16) and (17) simplify substantially:

$$\begin{aligned} u_{\pm} &= \frac{\Omega_A}{k_A} \\ &\pm 2 \frac{\sqrt{(|G_B| k_A^2)^{2/3} - (|G_A| k_B^2)^{2/3}}}{\sqrt{(|G_B|^2 k_A)^{2/3} - (|G_A|^2 k_B)^{2/3}}} \left( \frac{|G_A G_B|}{k_A k_B} \right)^{2/3}, \\ \kappa^{11}(u_{\pm}) &= -\kappa^{22}(u_{\pm}) = \pm \kappa, \\ \kappa &= \left( (|G_B| k_A^2)^{2/3} - (|G_A| k_B^2)^{2/3} \right) \\ &\times \sqrt{\frac{(|G_B| k_A^2)^{2/3} - (|G_A| k_B^2)^{2/3}}{|G_B|^2 k_A^2 - |G_A|^2 k_B^2}}. \end{aligned} \quad (18)$$

The extremal values of the function  $\kappa^{11}(u_{\pm})$  depend on the ratios of the Rabi frequencies of strong fields:  $|G_A/G_B|$ . For definiteness we shall assume  $k_A > k_B$ . For  $|G_A/G_B| < \sqrt{k_A/k_B}$  we find  $\kappa^{11}(u_{-}) < k_B - k_A$  and  $\kappa^{11}(u_{+}) > k_A - k_B$ . Recall that as  $u \rightarrow \pm\infty$  the function  $\kappa^{11}(u)$  approaches the asymptotic values  $\pm(k_A - k_B)$ , respectively. Consequently, the equation  $\Delta k = \kappa^{11}(u)$  possesses two solutions with

$$k_A - k_B < |\Delta k| < \kappa, \quad |G_A/G_B| < \sqrt{k_A/k_B} \quad (19)$$



**Fig. 3.** The parameter ranges corresponding to different numbers of nonlinear resonances in the probe field spectrum in the  $(\ln|G_A/G_B|, E_3)$  plane for four-level systems with strong fields on opposite transitions, shown in the insets (the dashed lines denote examples of the arrangement of the probe field).  $\Omega_A/k_A = \Omega_B/k_B$ ,  $E_1 - E_2 > \Delta E \equiv E_3 - E_4$ ,  $\beta = (1/2)\ln[(E_1 - E_2)/\Delta E]$ ,  $\delta = 2\ln[(E_1 - E_2)/\Delta E]$ . The large numbers denote the number of resonances in the corresponding regions separated by the heavy lines.

or one solution if

$$|\Delta k| < k_A - k_B. \quad (20)$$

Similarly, the frequency branch  $\Omega_p^{22}$  possesses two extrema if the condition (19) is satisfied and one solution if the inequality (20) is satisfied.

For  $\sqrt{k_A/k_B} < |G_A/G_B| < k_A^2/k_B^2$  the expression (18) is complex, so that  $\kappa^{11}(u)$  and  $\kappa^{22}(u)$  are monotonic functions and hence the frequency branches  $\Omega_p^{11}(u)$  and  $\Omega_p^{22}(u)$  each possess one extremum, while the condition (20) is satisfied. Finally, for  $k_A^2/k_B^2 < G_A/G_B$  we find  $\kappa^{11}(u_-) > 0$  and  $\kappa^{11}(u_+) < 0$ , so that the frequency branches  $\Omega_p^{11}(u)$  and  $\Omega_p^{22}(u)$  each possess three turning points for

$$|\Delta k| < \min(|\kappa|, k_A - k_B), \quad |G_A/G_B| > k_A^2/k_B^2,$$

and two turning points for

$$k_A - k_B < |\Delta k| < |\kappa|,$$

or one turning point each if

$$|\kappa| < |\Delta k| < k_A - k_B.$$

Figure 3 illustrates the usual rearrangement of the probe-field spectrum as a function of the parameters of the four-level system in the plane  $(\ln|G_A/G_B|, E_3)$  for  $\Omega_A k_B = \Omega_B k_A$ . Here we employ the following notations:  $E_1$  and  $E_2$  are the energies of the upper and lower levels

of the transition in resonance with the field  $G_A$ ,  $E_3$  and  $E_4$  are the energies of the upper and lower levels of the transition in resonance with the field  $G_B$ . For definiteness we assume  $\Delta E = E_3 - E_4 < E_1 - E_2$ . In this case the expression  $\Delta k = \kappa$  for the boundaries between the regions with a different number of resonances can be written in a form valid for an arbitrary level scheme:

$$|E_4 + E_3 - E_1 - E_2| = \frac{(|G_B|^{2/3}(E_1 - E_2)^{4/3} - |G_A|^{2/3}(E_3 - E_4)^{4/3})^{3/2}}{\sqrt{|G_B|^2(E_1 - E_2)^2 - |G_A|^2(E_3 - E_4)^2}}. \quad (21)$$

The level schemes are shown in the insets. The large numbers denote the number of narrow resonances in the corresponding regions of the parameters. Thus, the number of resonances in the probe-field spectrum depends on the level scheme and on the relative detuning of the frequencies and intensities of the strong fields, and in general it can assume the values 0, 2, 4, 6, and 8. An odd number occurs in degenerate cases, specifically, if the condition (21) is satisfied.

### 3.2. Four-Level System 3 + 1

We now consider a four-wave system interacting with two strong fields ( $\mathbf{E}_1$  and  $\mathbf{E}_2$ ) on neighboring transitions; see inset in Fig. 4. In zeroth order perturbation theory in the probe field this four-level system decomposes into three- and one-level subsystems. The energies of the dressed states  $\lambda_A^I$ ,  $I = 1, 2, 3$ , of the three-level system interacting resonantly with two electromagnetic fields ( $\mathbf{E}_1$  and  $\mathbf{E}_2$ ), can be determined in principle using the Cardano formula from the equation

$$\lambda_A^I(\lambda_A^I + \Omega_1^I)(\lambda_A^I + \Omega_2^I) - |G_1|^2(\lambda_A^I + \Omega_2^I) - |G_2|^2(\lambda_A^I + \Omega_1^I) = 0, \quad (22)$$

where  $\Omega_{1,2}^I = \Omega_{1,2} - k_{1,2}u$  are the detunings of the fields taking account of the Doppler shift and  $G_{1,2} = \mathbf{E}_{1,2} \mathbf{d}_{1,2} / 2\hbar$  is the Rabi frequency of the fields. The energy of a dressed state of the subsystem consisting of one level is  $\lambda_B = 0$ . To find the number of turning points we shall investigate the frequency branches  $\Omega_p^I = k_\mu u + \lambda_B - \lambda_A^I = k_\mu u - \lambda_A^I$  (see Appendix). Substituting  $\lambda_A^I = k_\mu u - \Omega_p^I$  into Eq. (22) we obtain a cubic polynomial of  $\Omega_p^I$  and  $u$ , implicitly giving the form of the frequency branches,

$$P_3(\Omega_p^I, u) = \Omega_p^I(\Omega_p^I - \Omega_1^I)(\Omega_p^I - \Omega_2^I) - (\Omega_p^I - \Omega_2^I)|G_1|^2 - (\Omega_p^I - \Omega_1^I)|G_2|^2 = 0. \quad (23)$$

If  $G_1, G_2 = 0$ , then all three frequency branches are linear functions of the velocity ( $I = 1, 2, 3$ ):

$$\begin{aligned}\Omega_p^I(u) &= k_\mu u, & \Omega_1 + (k_\mu - k_1)u, \\ & & \Omega_2 + (k_\mu - k_2)u,\end{aligned}$$

which intersect at the points  $u_1^* = \Omega_1/k_1$ ,  $u_2^* = \Omega_2/k_2$ , and  $u_3^* = (\Omega_1 - \Omega_2)/(k_1 - k_2)$ . When  $G_1, G_2 \neq 0$  the frequency branches  $\Omega_p^I(u)$  do not intersect, but rather they repel one another by an amount of the order of  $G_1$  and  $G_2$ . If the difference of the detunings of the strong fields is large,  $\Delta\Omega = |\Omega_1 - \Omega_2 k_1/k_2| \gg G_1, G_2$ , then near the points  $u_v^*$ ,  $v = 1, 2, 3$  the frequency branches pass smoothly from one linear asymptote to another. If  $u_v^*$  is a point of intersection of the asymptotes, whose slopes have different signs, then there are two turning points near this point, similarly to the case of a three-level system (Fig. 4). The maximum number of such points of intersection is 2, so that the maximum number of extrema of the function  $\Omega_p^I$  is 4. However, if the slopes of all three asymptotes have the same sign, then all frequency branches are monotonic functions and there are no narrow resonances in the probe-field spectrum. The conditions found can be formulated in terms of the energy: if the energy level of the second subsystem (in this case this is an individual level) lies outside the levels of the three-level subsystem, then there are no narrow resonances in the probe-field spectrum and conversely, if a solitary level lies between the levels of the three-level system, the maximum possible number of resonances is 4.

Two turning points can merge as the difference  $\Delta\Omega$  of the detunings of strong fields decreases. The condition for merging of two extrema of the function  $\Omega_p^I$  is determined by solving simultaneously the equations

$$P_3(\Omega_\mu, u) = 0, \quad \frac{dP_3(\Omega_\mu, u)}{du} = 0, \quad \frac{d^2P_3(\Omega_\mu, u)}{du^2} = 0.$$

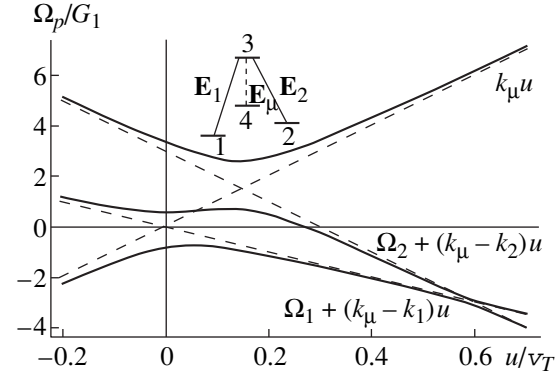
Solving the system and eliminating  $\Omega_\mu$  and  $u$  we obtain an expression for the boundary separating the region of parameters for which 4 or 2 resonances exist for a level scheme shown in the inset in Fig. 4:

$$\Delta\Omega^2 = \frac{|G_1|^2(1+z)}{k_2^2 k_\mu(k_1 - k_\mu)} \quad (24)$$

$$\times [2k_2 k_\mu - k_1(k_2 + k_\mu) + (2k_1 k_\mu - k_2(k_1 + k_\mu))z]^2,$$

where

$$z^3 = \frac{|G_2|^2 k_1 - k_\mu}{|G_1|^2 k_2 - k_\mu}.$$



**Fig. 4.** Frequency branches of a four-wave system in the  $(u, \Omega_p)$  plane:  $k_1 v_T = 20G_1$ ,  $k_2 v_T = 15G_1$ ,  $k_\mu v_T = 10G_1$ ,  $G_2 = 0.7G_1$ ,  $\Omega_1 = 3G_1$ , and  $\Omega_2 = 0$ . Dashed straight lines— asymptotes of the frequency branches. The inset shows a four-level system with two strong fields on adjoining transitions; the solid lines denote the strong fields  $E_1$  and  $E_2$ ; the dashed line denotes the probe field  $E_\mu$ .

The equation (24) possesses a real solution only if

$$|G_1|^2 \frac{k_\mu}{k_1 - k_\mu} > |G_2|^2 \frac{k_\mu}{k_\mu - k_2}. \quad (25)$$

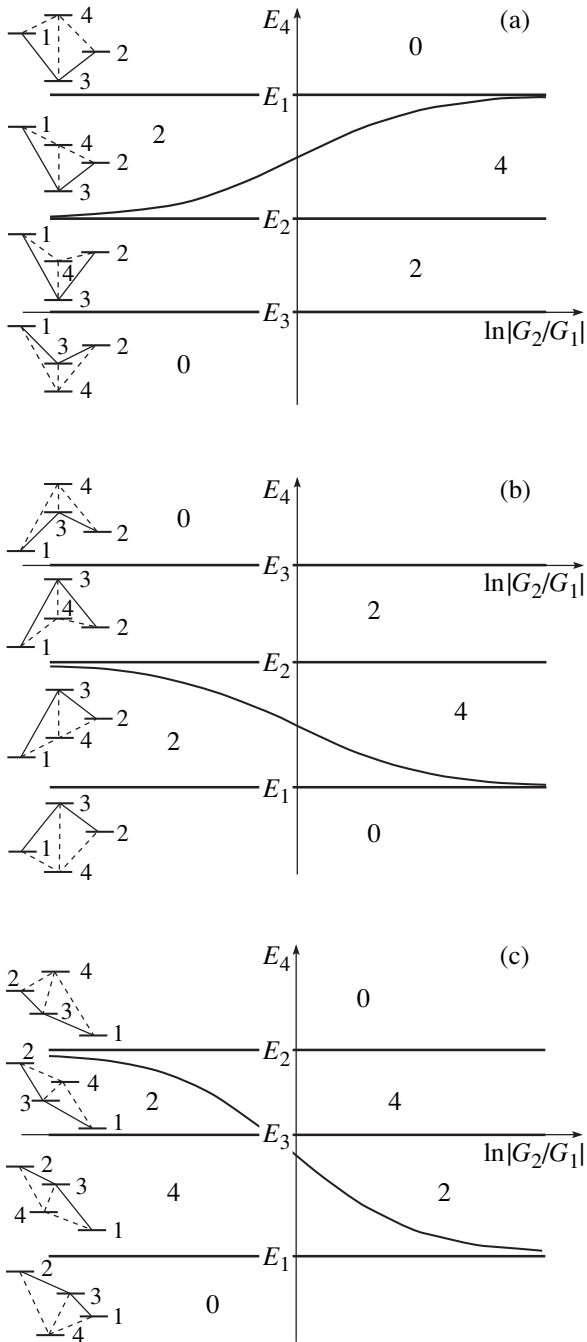
A classification of four-level systems with two strong fields on neighboring transitions with respect to the number of resonances with  $\Delta\Omega = 0$  is presented in the parameter plane  $(\ln|G_2/G_1|, E_4)$  in Fig. 5. For an arbitrary level scheme we introduce the following notations:  $E_1$  and  $E_3$  are the energies of the levels of the transition in resonance with the field  $G_1$ ;  $E_2$  and  $E_3$  are the energies of the levels of a transition in resonance with the field  $G_2$ ; the probe field is in resonance with the transition between level 4 and one of the levels which are coupled by strong fields. For definiteness we assume  $|E_1 - E_3| > |E_2 - E_3|$ . Then the condition (25), determining the region of parameters for which two resonances exist, can be written in a form valid for an arbitrary level scheme:

$$|G_1|^2 \frac{E_3 - E_4}{E_4 - E_1} + |G_2|^2 \frac{E_3 - E_4}{E_4 - E_2} > 0.$$

The level schemes are shown in the insets in Fig. 5. The large numbers in Fig. 5 denote the number of resonances in the corresponding regions of the parameters. Figures 5a, 5b, and 5c refer to the schemes in which a pair of strong fields forms. Respectively, V,  $\Lambda$ , and the cascade  $\Xi$  configurations.

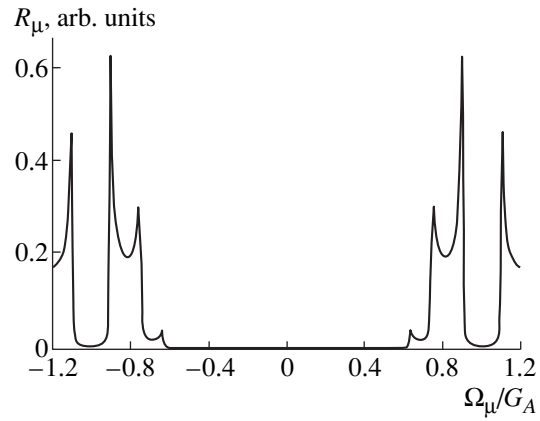
#### 4. DISCUSSION

In summary, it is found that a four-wave system behaves similarly to a three-level system. A pair of narrow resonances appears in the probe-field spectrum of a three-level system (Fig. 1) if the level which is coupled with the system by the probe field falls between



**Fig. 5.** The parameter regions corresponding to different numbers of nonlinear resonances in the probe-field spectrum in the  $(\ln|G_2/G_1|, E_4)$  plane with  $\Omega_1/k_1 = \Omega_2/k_2$  for four-level systems with strong fields on adjoining transitions shown in the insets (the dashed lines show the examples of the arrangement of the probe field). The large numbers denote the number of resonances in the corresponding regions separated by the heavy curves: (a)  $V$  schemes; (b)  $\Lambda$  schemes; (c)  $\Xi$  schemes.

the levels coupled by the strong field. The Autler-Townes doublet in the anti-Stokes component vanishes in the  $V$  or  $\Lambda$  scheme with codirected waves. Similarly, in a four-level system with strong fields on neighboring



**Fig. 6.** The work performed by the probe field on the transition 2–3 as a function of the detuning  $\Omega_\mu$  for the scheme shown in the inset in Fig. 2:  $G_B = 0.1G_A$ ,  $k_A v_T = 7G_A$ ,  $k_\mu v_T = 5.8G_A$ ,  $k_B v_T = 5G_A$ , and  $\Gamma = 0.001G_A$ .

transitions the maximum number of narrow peaks in the spectrum (up to four) is possible when a level coupled by the probe field with a three-level subsystem falls between its levels. For stationary atoms the number of resonances is three. Thermal motion increases this number to four. If the strong fields are applied on opposite transitions (see inset in Fig. 3), then the number of peaks is also sensitive to the relative arrangement of the two-level subsystems. There are no resonances if the subsystems lie at a different height on the energy scale. There will be more peaks if the intervals of the energies of the subsystems overlap. The number of resonances is maximum when both levels of one subsystem fall between the levels of another subsystem. In the latter case the number of peaks in the spectrum reaches 8, while for stationary atoms only four peaks are possible in a four-level system.

An example of a probe-field spectrum with the maximum number of peaks is presented in Fig. 6. It was obtained numerically using a simple program which inverts the matrix of the system using Gauss' method and integrates over velocities using Simpson's rule. In previous works on the experimental and theoretical investigation of the spectra of multilevel systems, the number of peaks observed was smaller probably because the parameter range that had to be reached in order for the maximum number to be obtained was quite narrow.

We shall now indicate some recent works devoted to three- and four-level systems with large Doppler broadening and interacting with strong fields. A four-wave mixing scheme was studied in [14]. The energies of dressed states, differing from the frequency branches by an added linear function of the velocity, were found. The dependences of the mixing ratio on the ratio of the Rabi frequencies were obtained. In [15] and [16], which are devoted to coherent effects in Doppler-broadened medium, cascade and  $V$ -like three-level sys-



tems, respectively, were studied. In these works the frequency branches with no turning points and with two turning points were represented, and the group spectra were calculated. A numerical calculation gave two resonant peaks in accordance with the concepts expounded above.

In [11] a four-level system with two strong fields on opposite transitions was studied. A so-called down-conversion scheme, shown in the inset in Fig. 2, was analyzed. Only six peaks were found in the calculation, though there were eight turning points, which is explained by the symmetry of the system studied ( $k_\mu = (1/2)(k_A + k_B)$ ,  $\Omega_A = \Omega_B = 0$ ). Two pairs of turning points contributed to the same peaks.

The effect of nonlinear interference processes on Doppler-broadened transitions in the scheme shown in Fig. 3 (second scheme at the top) was investigated in [12]. In this scheme six peaks can in principle be observed, and up to five peaks were found numerically. A “ladder” four-level scheme with two strong fields on adjoining transitions was investigated in [17]. The directions of propagation of the waves on successive transitions of a cascade alternated, so that the scheme reduces to the one shown in Fig. 5b (third scheme from the top). Up to four resonant peaks can exist in this scheme. However, the authors confine their attention to the case where the Rabi frequency of the strong field is much larger than the Doppler width. Then the neighborhood of only one turning point contributes to the probe-field spectrum, so that only one peak is found in this work. Narrow peaks in a degenerate four-wave mixing in Rb vapors were found in the experimental work [18]. The Doppler shifts were compensated, so that up to three peaks were observed.

## 5. CONCLUSIONS

The multippeak structures predicted for the probe-field spectrum are not confirmed experimentally. It would be especially interesting to check the degenerate cases where turning points merge, leading to a sharp increase of the nonlinear susceptibility. Considering the sensitivity of the number of peaks to the parameters of the problem, the experiment should be designed in accordance with the recommendations presented: (1) the lasers should be stable enough so that random frequency drifts do not destroy the assumption that the relaxation constants are small; (2) the medium should be rarefied, so that collisions do not increase the relaxation; (3) the angular momenta of the levels and the polarization of the fields must be chosen so that the effect does not disappear on averaging over the projections of the angular momenta; and (4) the medium should be thin compared with the absorption length of strong fields, which can smear the peaks.

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## APPENDIX

Let us consider a  $n$ -level system interacting resonantly with several monochromatic electromagnetic waves. The evolution of the atomic wave functions satisfies the Schrödinger equation

$$i\frac{d\mathbf{a}}{dt} = \hat{\mathcal{H}}\mathbf{a}, \quad \hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{\text{int}}(t), \quad (26)$$

where  $\mathbf{a} = (a_1, \dots, a_n)^T$  is a column vector of the probability amplitudes of the states  $|i\rangle$ ,  $i = 1, \dots, n$ ;  $\hat{\mathcal{H}}_0$  is the atomic Hamiltonian, and  $\hat{\mathcal{H}}_{\text{int}}(t)$  describes the interaction of an atom with the electromagnetic field.

In the present work we shall confine our attention to interaction schemes for which Eq. (26) can be put into a form with a time-independent Hamiltonian  $\hat{H}$  using a unitary transformation

$$A_j = a_j \exp(i\delta_j t), \quad j = 1, \dots, n.$$

A sufficient condition for the existence of such a transformation is simultaneous satisfaction of the relations  $\delta_j - \delta_i = \omega_{ij}$ , where  $\omega_{ij}$  is the frequency of the wave in resonance with the transition  $i \rightarrow j$ , and  $E_i < E_j$ . Let us consider a collection of levels coupled with one another by strong fields whose Rabi frequencies are much larger than the relaxation constants of the levels,  $G \gg \Gamma$ . The energies of the dressed states of such a subsystem “repel” one another by an amount of the order of the Rabi frequencies  $G$ . To obtain narrow spectral structures the probe field must couple levels belonging to two different subsystems.

Let us consider the absorption spectrum  $P_\mu(\Omega_\mu)$  of a probe field with frequency  $\omega_\mu$ , which couples the levels  $i$  and  $j$  from the first and second subsystems, respectively, where  $\Omega_\mu = \omega_\mu - (E_j - E_i)/\hbar$  is the detuning of the probe wave from resonance and  $E_i < E_j$ . The Hamiltonian  $\hat{H}$  can be written in the form

$$\hat{H} = \begin{pmatrix} \hat{H}_A & \delta\hat{H} \\ \delta\hat{H}^\dagger & \hat{H}_B + \Omega_\mu \hat{I} \end{pmatrix}, \quad (27)$$

where  $\hat{I}$  is a unit matrix and  $\delta\hat{H}$  is a small perturbation proportional to the amplitude of the probe field. The matrices  $\hat{H}_A$  and  $\hat{H}_B$  do not contain the parameters of the weak field. Neglecting the amplitude of the weak

field, the secular equation  $|\hat{H} - \Lambda\hat{I}| = 0$  separates into two equations

$$|\hat{H}_A - \Lambda\hat{I}| = 0, \quad |\hat{H}_B + \Omega_\mu\hat{I} - \Lambda\hat{I}| = 0.$$

We denote the normalized eigenvectors and eigennumbers of these two subsystems as  $\mathbf{A}^I, \lambda_A^I$ , and  $\mathbf{B}^J, \Omega_\mu + \lambda_B^J$ , respectively, where the superscripts enumerate the eigenvectors and the subscripts denote the subsystem.

For example, for a four-wave system (see inset in Fig. 2) the Hamiltonian has the form (27), where

$$\hat{H}_A = \begin{pmatrix} \Omega_A & -G_A \\ -G_A^* & 0 \end{pmatrix}, \quad (28)$$

$$\hat{H}_B = \begin{pmatrix} 0 & -G_B \\ -G_B^* & -\Omega_B \end{pmatrix}, \quad \delta\hat{H} = \begin{pmatrix} 0 & 0 \\ -G_\mu^* & 0 \end{pmatrix}.$$

The eigenvalues of the matrices  $\hat{H}_A$  and  $\hat{H}_B$  can be written in the form

$$\lambda_A^{1,2} = \frac{1}{2}(\Omega_A \pm \sqrt{\Omega_A^2 + 4|G_A|^2}), \quad (29)$$

$$\lambda_B^{1,2} = -\frac{1}{2}(\Omega_B \pm \sqrt{\Omega_B^2 + 4|G_B|^2}).$$

Let one level in the subsystem, described by the operator  $\hat{H}_A$ , be occupied initially. Then the time dependence of the wave function  $\mathbf{A}(t)$  can be represented in the form

$$\mathbf{A}(t) = \sum_{I=1}^{n_A} \mathbf{A}^I \exp(-i\lambda_A^I t) (\mathbf{A}^I, \mathbf{A}^0), \quad (30)$$

where

$$(\mathbf{X}, \mathbf{Y}) = \sum_{v=1}^n X_v^* Y_v$$

is the scalar product of the vectors  $\mathbf{X}$  and  $\mathbf{Y}$ ,  $\mathbf{A}^0 = \mathbf{A}(t)|_{t=0}$  is the initial wave function normalized to one, and  $n_A$  is the number of levels in the subsystem  $A$ . Substituting the solution obtained into the equation for  $\mathbf{B}$  in first-order perturbation theory in the amplitude of the probe field we obtain

$$i\frac{d\mathbf{B}}{dt} = \delta\hat{H}^\dagger \mathbf{A} + (\hat{H}_B + \Omega_\mu\hat{I})\mathbf{B}. \quad (31)$$

Solving the system of equations we obtain

$$\mathbf{B} = -\sum_{I=1}^{n_A} \sum_{J=1}^{n_B} \mathbf{B}^J \exp[-i(\lambda_B^J + \Omega_\mu)t] \\ \times (\mathbf{B}^J, (\lambda_A^I - (\Omega_\mu + \hat{H}_B))^{-1} \delta\hat{H}^\dagger \mathbf{A}^I) (\mathbf{A}^I, \mathbf{A}^0) \\ + \sum_{I=1}^{n_A} \exp(-i\lambda_A^I t) [(\lambda_A^I - (\Omega_\mu + \hat{H}_B))^{-1} \delta\hat{H}^\dagger \mathbf{A}^I] (\mathbf{A}^I, \mathbf{A}^0),$$

where  $n_B$  is the number of levels in the subsystem  $B$ .

The work performed by the probe field can be calculated using the formula (see, for example, [5])

$$R_\mu(\Omega_\mu) = -2\hbar\omega_\mu \text{Re} \left[ iG_\mu^* \int_0^\infty dt e^{-\Gamma t} A_i^*(t) B_j(t) \right] \\ = -2\hbar\omega_\mu \text{Re} \left[ G_\mu^* \sum_{I,I'=1}^{n_A} \sum_{J=1}^{n_B} B_j^J (A_i^I)^* (\mathbf{A}^0, \mathbf{A}^I) (\mathbf{A}^I, \mathbf{A}^0) \right. \\ \times (\mathbf{B}^J, \delta\hat{H}^\dagger \mathbf{A}^I) [\Gamma + i(\Omega_\mu - \lambda_A^I + \lambda_B^J)]^{-1} \\ \left. \times [\Gamma - i(\lambda_A^I - \lambda_A^{I'})]^{-1} \right].$$

In this sum the terms with  $I = I'$  contain the small parameter  $\Gamma$  in the denominator, since the difference  $\lambda_A^I - \lambda_A^{I'}$  vanishes. The other terms in the sum (for  $I \neq I'$ ) can be neglected, since they are small to the extent that the parameter  $\Gamma/G$  is small. In this approximation the power absorption wave per unit volume  $P_\mu(\Omega_\mu) = \Gamma N R_\mu(\Omega_\mu)$  can be written in the form

$$P_\mu(\Omega_\mu) = 2\hbar\omega_\mu \\ \times \text{Re} \sum_{I=1}^{n_A} \sum_{J=1}^{n_B} \frac{N |G_\mu|^2 |B_j^J A_i^I (\mathbf{A}^0, \mathbf{A}^I)|^2}{\Gamma + i(\Omega_\mu - \lambda_A^I + \lambda_B^J)}, \quad (32)$$

where  $N$  is the equilibrium population.

To take account of the Doppler effect for particles moving with velocity  $u$ , the detuning  $\Omega_\nu$  of all fields must be replaced by  $\Omega_\nu - k_\nu u$  and the expression obtained must be averaged over the velocities. We shall now use the graphical method, formulated in Section 2, to calculate the peaks in the probe-field spectrum. The condition  $\Delta^{IJ}(u, \Omega_\mu) = 0$ , where

$$\Delta^{IJ}(u, \Omega_\mu) = \Omega_\mu - k_\mu u - \lambda_A^I(u) + \lambda_B^J(u) \quad (33)$$

determines the frequency branches  $\Omega_\mu = \Omega_p^{IJ}(u) = k_\mu u + \lambda_A^J(u) - \lambda_B^I(u)$ . The conditions for an extremum of the frequency branches give the turning points  $(u, \Omega_p)$  in the  $(u_p^{IJ}, \Omega_p^{IJ}(u_p^{IJ}))$  plane. For detunings  $\Omega_\mu = \Omega_p^{IJ}(u_p^{IJ})$  narrow resonances are observed in the probe-field spectrum. The number of such resonances is determined by

the number of turning points. In the limit of small  $\Gamma$  the contribution of each turning point to the work performed by the field can be studied independently:

$$P_{\mu}(\Omega_{\mu}) = \sum_I^{n_A} \sum_J^{n_B} P^{IJ}(\Omega_{\mu}), \tag{34}$$

$$P^{IJ}(\Omega_{\mu}) = \text{Re} \int_{-\infty}^{\infty} \frac{C^{IJ}(u) du}{\Gamma + i[\Omega_{\mu} - k_{\mu}u - \lambda_A^I(u) + \lambda_B^J(u)]},$$

where  $C^{IJ}(u)$  is the numerator in the expression (32). Since there can be several turning points for a fixed frequency branch, we shall enumerate them with the index  $m$ . Then  $P^{IJ}(\Omega_{\mu})$  can be written approximately as a sum over all turning points:

$$P^{IJ}(\Omega_{\mu}) \approx \pi \sum_m C^{IJ}((u_p^{IJ})_m) \times \text{Re} \sqrt{\frac{1}{a_m^{IJ}[\Omega_{\mu} - (\Omega_p^{IJ}((u_p^{IJ})_m) + i\Gamma)]}},$$

$$a_m^{IJ} = \frac{1}{2} \frac{d^2 \Omega_p^{IJ}(u)}{du^2} \Big|_{u=(u_p^{IJ})_m}.$$

The total number of narrow resonances in the spectrum can be determined by summing over the frequency branches and over the turning points of each branch.

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