J. Phys. B: At. Mol. Opt. Phys. **32** (1999) 649–661. Printed in the UK PII: S0953-4075(99)96658-0

Autler–Townes doublet probed by strong field

M G Stepanov†

Institute of Automation and Electrometry, Russian Academy of Sciences, Siberian Branch,
Novosibirsk 630090, Russia
and
Novosibirsk State University, Novosibirsk 630090, Russia

Received 10 August 1998, in final form 8 October 1998

Abstract. This paper deals with the Autler–Townes doublet structure. Applied driving and probing laser fields can have arbitrary intensities. An explanation is given of the broadening of doublet components with the growth of probing field intensity, which was observed in experiment. The effects of Doppler averaging are discussed.

1. Introduction

The spectroscopy of three-level systems occupies a highly important place in nonlinear spectroscopy. In addition to saturation or power broadening of resonance [1], new remarkable effects appear: field splitting [2] and truly coherent processes (such as Raman scattering or two-photon processes). The clearest issue is the absorption spectrum of three-level systems interacting with two monochromatic waves within perturbation theory in intensity of one of the waves—probe field spectrum [3–7]. Some attempts have been made to construct the theory where both waves are strong [8, 9]. In [8], the coherences in the equations for the density matrix were excluded, and equations containing only populations were derived. As a result, the probabilities of transitions induced by fields were renormalized and expressed in terms of populations only. Unfortunately, our intuition in predicting the behaviour of such systems is poor as yet; it is difficult to foresee the result without solving the equations. The computation of three-level systems with a great number of parameters (relaxation constants of levels, wave detunings and strengths) [9] is hard to analyse.

However, the power of light has been increased since early experiments, as one needs to get better light conversion or a more controllable set-up. In experiment [10], Λ-scheme interacting with two waves exhibits, at first sight, strange behaviour. When the intensity of the comparatively weak wave increases, the components of the Autler–Townes doublet broaden in its absorption spectrum; finally the doublet is transformed to a single line.

A three-level system with two strong fields can be a part of the scheme of four-wave resonant mixing. The study of this area is progressing rapidly, since it is hoped to obtain coherent CW short-wave radiation. To understand the whole picture it is useful to first consider the part of a system that interacts with strong fields.

The aim of this paper is to analyse qualitatively the effects arising from increasing the power of the weak wave in Λ-scheme. In what follows we discuss the general features of multi-level systems and the possibility of reducing them to simple two-level systems. We use

---

† E-mail address: `Stepanov@iae.nsk.su`

0953-4075/99/030649+13$19.50

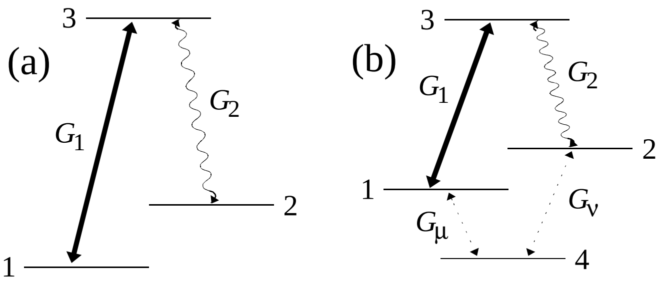

Figure 1. (*a*) Three-level system interacting with two electromagnetic waves and (*b*) its testing by two weak (probe) waves.

the probability amplitudes approach and the 'dressed' state concept (see [11,12]) to understand the spectra. The energies of 'dressed' states are identified with the poles of the Green function of master equations in ω-representation. We are not interested in intrinsic relaxation processes in an atomic system and this allows us to avoid introducing the density matrix. It reduces the system of N^2 equations for the elements of the density matrix to the system for N probability amplitudes (here $N = 3$).

This paper is organized as follows. In sections 2 and 3 the equations of motion and their Green function are described. In section 4 the energies of 'dressed' states are discussed. Section 5 is devoted to the calculation of the probe field spectrum in a four-level system—linear response of a three-level one; it shows the benefits of Green function. Section 6 presents the absorption spectra of the strong field. In section 7 we briefly consider the question of the Doppler broadening of spectra. In section 8 we finally summarize essential aspects of this paper.

2. Basic equations

Consider the atomic system consisting of three excited levels and its resonant interaction with two electromagnetic waves with frequencies ω_1, ω_2 (see figure 1). Hereafter we assume that the wave amplitudes are not changed essentially by interaction (optically thin media). Thus, we do not consider here the effects of light propagation. One can imagine that there is a thin layer with absorbing atoms and light travels through it almost without absorption. We also do not take into account the motion of atoms, i.e. we do not consider the effects connected with inhomogeneous broadening of absorption spectra. Then the time evolution of the atomic wavefunction is described by the Schrödinger equation ($\hbar = 1$)

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}|\psi\rangle = (\hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{\mathrm{int}})|\psi\rangle, \qquad |\psi\rangle = \sum_{i=1}^{3} a_i(t)|i\rangle, \tag{1}$$

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix} = \begin{pmatrix} E_1 - \frac{\mathrm{i}}{2}\Gamma_1 & 0 & -G_1^*\mathrm{e}^{\mathrm{i}\omega_1 t} \\ 0 & E_2 - \frac{\mathrm{i}}{2}\Gamma_2 & -G_2^*\mathrm{e}^{\mathrm{i}\omega_2 t} \\ -G_1\mathrm{e}^{-\mathrm{i}\omega_1 t} & -G_2\mathrm{e}^{-\mathrm{i}\omega_2 t} & E_3 - \frac{\mathrm{i}}{2}\Gamma_3 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix}. \tag{2}$$

Here $\hat{\mathcal{H}}_0$ is the atomic Hamiltonian, $\hat{\mathcal{H}}_{\mathrm{int}}$ describes the interaction with light; a_i, E_i, Γ_i are the probability amplitude, the energy, and the decay rate of state $|i\rangle$; G_i is the matrix element $\langle 3| - \hat{\mathcal{H}}_{\mathrm{int}}|i\rangle\mathrm{e}^{\mathrm{i}\omega_i t}$ of interaction. Equation (2) is written in the resonant (or rotating wave) approximation, which is valid if $G_i, \omega_i - E_3 + E_i \ll E_3 - E_i$, $i = 1, 2$. The frequency $\Omega_i = \omega_i - E_3 + E_i$ is the detuning of the wave i from resonance.

Since there is no closed cycle made up from wave fields (see figure 1(*a*)) one can bring (2) to the form without explicit dependence on time:

$$A_i = a_i e^{i(E_i-\Omega_i)t}, \quad i = 1, 2, \qquad A_3 = a_3 e^{iE_3 t}, \tag{3}$$

$$i\frac{d}{dt}\begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} = \begin{pmatrix} \Omega_1 - \frac{i}{2}\Gamma_1 & 0 & -G_1^* \\ 0 & \Omega_2 - \frac{i}{2}\Gamma_2 & -G_2^* \\ -G_1 & -G_2 & -\frac{i}{2}\Gamma_3 \end{pmatrix}\begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix}. \tag{4}$$

Moreover, fields G_i can be thought of as real. Nevertheless, sometimes we will write G_i^* instead of G_i. It helps to determine the interaction process which corresponds to the written expression.

In spectroscopy, the observed quantities are the powers absorbed from the waves. A light field induces transitions between atomic levels, and absorption is connected with the change of levels population. The time derivative of population on level two is equal to

$$\frac{d}{dt}|A_2|^2 = -\Gamma_2|A_2|^2 - iG_2A_2A_3^* + iG_2^*A_2^*A_3. \tag{5}$$

The first term in the r.h.s. of (5) is due to the decay of the state $|2\rangle$. The other two terms are responsible for the absorption of the second wave.

The formulation of the problem of light absorption is the following: at the moment t_0 the atom is excited to the state $\boldsymbol{A} = \boldsymbol{A}_0$. Then the time evolution of $\boldsymbol{A}$ is governed by (4), and the power absorbed from the second wave by this atom is equal to

$$P(\boldsymbol{A}_0, \Omega_2, t_0) = \omega_2 \int_{t_0}^{\infty} dt\, (iG_2A_2A_3^* - iG_2^*A_2^*A_3). \tag{6}$$

Because equation (4) is time-independent, the power P does not depend on t_0. Then one can set $t_0 = 0$. In addition, P is a quadratic functional of $\boldsymbol{A}(t)$, and equation (4) is linear. Thus, P can be written as $P(\boldsymbol{A}_0, \Omega_2) = \omega_2 \boldsymbol{A}_0^\dagger \hat{\mathcal{P}}(\Omega_2)\boldsymbol{A}_0$, where $\cdot^\dagger$ is the Hermitian conjugation. The matrix $\hat{\mathcal{P}}$ contains complete information about the absorption of the second wave, i.e. using $\hat{\mathcal{P}}$ one can calculate P with any initial excitation $\boldsymbol{A}_0$. For example, $\mathcal{P}_{22}$ is equal to the probability of absorption of a photon from the second wave if the atom was initially excited to level two. Because P is real, the matrix $\hat{\mathcal{P}}$ is Hermitian.

3. Green function

Let us consider equation (4) with the initial condition $\boldsymbol{A}(t = 0) = \boldsymbol{A}_0$:

$$i\frac{d}{dt}\boldsymbol{A} = \hat{H}\boldsymbol{A} + i\delta(t)\boldsymbol{A}_0, \qquad \boldsymbol{A}(t) = \boldsymbol{0} \qquad \text{if } t < 0. \tag{7}$$

Since this equation is time-independent, it is convenient to consider its Fourier transform, i.e, its energy representation ($\hbar = 1$)

$$\boldsymbol{A}(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t}\boldsymbol{A}_\omega, \qquad \omega\boldsymbol{A}_\omega = \hat{H}\boldsymbol{A}_\omega + i\boldsymbol{A}_0. \tag{8}$$

It is a set of linear algebraic equations. Its solution is equal to

$$\boldsymbol{A}_\omega = i(\omega - \hat{H})^{-1}\boldsymbol{A}_0 = i\hat{\mathcal{D}}(\omega)\boldsymbol{A}_0. \tag{9}$$

The matrix $\hat{\mathcal{D}}(\omega)$ is the Green function of (4) in ω-representation; it is also the resolvent of the Hamiltonian $\hat{H}$. The matrix element $\mathcal{D}_{ij}(\omega)$ describes transitions $i \leftarrow j$ induced by fields. As a function of complex variable ω, the matrix $\hat{\mathcal{D}}(\omega)$ has poles at the eigenvalues λ_I, $I = 1, 2, 3$ of the Hamiltonian $\hat{H}$, i.e. at the quasi-energies of the stationary states of 'atom +

field' system ('dressed' states). The energy λ of a 'dressed' state is found from the equation $\mathrm{Det}\,(\lambda - \hat{H}) = 0$:

$$\begin{aligned}&\left(\lambda - \Omega_1 + \frac{\mathrm{i}}{2}\Gamma_1\right)\left(\lambda - \Omega_2 + \frac{\mathrm{i}}{2}\Gamma_2\right)\left(\lambda + \frac{\mathrm{i}}{2}\Gamma_3\right)\\ &\qquad = \left(\lambda - \Omega_1 + \frac{\mathrm{i}}{2}\Gamma_1\right)|G_2|^2 + \left(\lambda - \Omega_2 + \frac{\mathrm{i}}{2}\Gamma_2\right)|G_1|^2. \end{aligned}\tag{10}$$

Each λ_I, $I = 1, 2, 3$ has negative imaginary part $\mathrm{Im}\,\lambda_I < 0$ corresponding to the decay rate of the 'dressed' state $|I\rangle\rangle$. In other words, $\hat{\mathcal{D}}(\omega)$ has poles only in the lower half-plane.

If the fields G_1, G_2 are weak, then there is a hope that the evolution of the system is little different from the evolution without fields. One can use this 'bare' evolution as a starting point and expand the Green function $\hat{\mathcal{D}}(G_1, G_2)$ into a power series in the neighbourhood of $G_1 = 0$, $G_2 = 0$. We denote

$$\hat{E} = \begin{pmatrix} \Omega_1 - \frac{\mathrm{i}}{2}\Gamma_1 & 0 & 0 \\ 0 & \Omega_2 - \frac{\mathrm{i}}{2}\Gamma_2 & 0 \\ 0 & 0 & -\frac{\mathrm{i}}{2}\Gamma_3 \end{pmatrix}, \qquad \hat{G} = -\begin{pmatrix} 0 & 0 & G_1^* \\ 0 & 0 & G_2^* \\ G_1 & G_2 & 0 \end{pmatrix}. \tag{11}$$

Then $\hat{H} = \hat{E} + \hat{G}$. One can write the following expansion:

$$\hat{\mathcal{D}} = \hat{D} + \hat{D}\hat{G}\hat{D} + \hat{D}\hat{G}\hat{D}\hat{G}\hat{D} + \cdots = \hat{D} + \hat{D}\hat{G}\hat{\mathcal{D}}, \tag{12}$$

where the argument of $\hat{\mathcal{D}}$ and $\hat{D}$ is equal to ω, and $\hat{D}(\omega) = (\omega - \hat{E})^{-1}$. This series is an analogue of $(1-x)^{-1} = 1 + x + x^2 + \cdots$. 'Bare' Green function $\hat{D}(\omega)$ has poles at the energies of 'bare' states $\Omega_1 - \frac{\mathrm{i}}{2}\Gamma_1$, $\Omega_2 - \frac{\mathrm{i}}{2}\Gamma_2$, $-\frac{\mathrm{i}}{2}\Gamma_3$. The summation (12) shifts the poles.

The diagonal element $\mathcal{D}_{ii}$ is the Green function of an atom in the state $|i\rangle$. It also has poles at λ_I. There is an admixture of all dressed states $|I\rangle\rangle$ in $|i\rangle$. In the limit of weak fields $\mathcal{D}_{ii}$ has an obvious pole. If the fields induce transitions between the states $|i\rangle$ and $|j\rangle$, then the atom in the state $|i\rangle$ lives in the state $|j\rangle$ for some time. Then the pole corresponding to $|j\rangle$ appears in its Green function. When the fields become stronger these poles shift.

Now we can write a simple expression for the matrix $\hat{\mathcal{P}}(\Omega_2)$ describing the absorption of the second wave.

$$P(\boldsymbol{A}_0, \Omega_2) = \omega_2 \int_0^\infty \mathrm{d}t\, \boldsymbol{A}^\dagger(t)\hat{P}\boldsymbol{A}(t), \qquad \mathrm{i}\hat{P} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & G_2^* \\ 0 & -G_2 & 0 \end{pmatrix}. \tag{13}$$

Vector $\boldsymbol{A}(t)$ can be rewritten in terms of the Green function. We obtain

$$\begin{aligned}\hat{\mathcal{P}}(\Omega_2) &= \int_0^\infty \mathrm{d}t \int \frac{\mathrm{d}\omega_1 \mathrm{d}\omega_2}{(2\pi)^2} \mathrm{e}^{\mathrm{i}(\omega_1 - \omega_2)t} \hat{\mathcal{D}}^\dagger(\omega_1)\hat{P}\hat{\mathcal{D}}(\omega_2) \\ &= \mathrm{i}\int \frac{\mathrm{d}\omega_1 \mathrm{d}\omega_2}{(2\pi)^2} \frac{\hat{\mathcal{D}}^\dagger(\omega_1)\hat{P}\hat{\mathcal{D}}(\omega_2)}{\omega_2 - \omega_1 - \mathrm{i}0} = \int \frac{\mathrm{d}\omega}{2\pi} \hat{\mathcal{D}}^\dagger(\omega)\hat{P}\hat{\mathcal{D}}(\omega). \end{aligned}\tag{14}$$

The correction i0 in (14) is due to causality. The physical determination of the integral over t consists in multiplying by a factor $\mathrm{e}^{-\varepsilon t}$ with $\varepsilon \to +0$. Since ω (as well as Ω_1 and Ω_2) is real on the contour of integration, in the integral we can replace $\hat{\mathcal{D}}^\dagger(\omega)$ by $\hat{\mathcal{D}}^c(\omega)$. The operation $\cdot^c$ is defined as follows: $\cdot^c \overset{\text{def}}{=} \cdot^\dagger$, but during this operation the frequencies ω, Ω_i, $i = 1, 2$ are thought to be real, i.e. they need not be complex conjugated. The operation $\cdot^c$ changes only the sign of Γ. Thereafter the integrand in (14) is an analytic function of ω, Ω_i, i.e. it does not depend on ω^*, Ω_i^*. Moreover, it is rational; one can apply residue theory to calculate the integral. In its turn, $\hat{\mathcal{P}}(\Omega_2)$ is an analytic function of Ω_2 and the poles of $\hat{\mathcal{P}}$ correspond to the resonances in the second wave absorption spectrum. The real part of the pole position gives the frequency of the resonance, the imaginary part represents the resonance width.

The matrix $\hat{H}$ has right and left eigenvectors: $\hat{H}|I\rangle\rangle = \lambda_I |I\rangle\rangle$, $\langle\langle I|\hat{H} = \langle\langle I|\lambda_I$. They can be normalized so that $\langle\langle I|J\rangle\rangle = \delta_{IJ}$. Then

$$\hat{H} = \sum_{I=1}^{3} |I\rangle\rangle \lambda_I \langle\langle I|, \qquad \hat{\mathcal{D}}(\omega) = \sum_{I=1}^{3} |I\rangle\rangle \frac{1}{\omega - \lambda_I} \langle\langle I|. \tag{15}$$

If $\hat{\mathcal{P}}(\Omega_2)$ has a pole at $\Omega_2 = \Omega$, then the integral (14) for $\Omega_2 = \Omega$ is divergent. The contour of integration bypasses the poles of $\hat{\mathcal{D}}(\omega)$ and $\hat{\mathcal{D}}^c(\omega)$ in different directions. Then the integral (14) may be divergent only if $\hat{\mathcal{D}}$ and $\hat{\mathcal{D}}^c$ have at least one common pole $\lambda_I = \lambda_J^c$. When λ_I and λ_J^c merge together they squeeze the integration contour, and the integral (14) acquires singularity. Time evolution of the dressed state $|I\rangle\rangle$ is described by the exponent $\mathrm{e}^{-\mathrm{i}\lambda_I t}$. The case where λ_I is close to λ_J^c corresponds to the resonance between the waves and the dressed states $|I\rangle\rangle$, $|J\rangle\rangle$. The absorption matrix $\hat{\mathcal{P}}(\Omega_2)$ contains resonance denominators of the form $\lambda_I - \lambda_J^c$.

4. Dressed states

Here, for simplicity, we neglect relaxation constants. In fact, the resonance widths under power broadening depend on the ratio of Γ even when the latter are small. Therefore, the contents of this paragraph are useful for the case of equal relaxation constants (or, maybe, when they are comparable to each other) or for finding resonance positions in the absorption spectrum of auxiliary probe field. The energies of the dressed states satisfy algebraic equation (10). Let us write it in the form

$$[\lambda(\lambda - \Omega_1) - |G_1|^2](\lambda - \Omega_2) = (\lambda - \Omega_1)|G_2|^2. \tag{16}$$

If we consider the energy levels of the system ‘atom + field’, then we will see the triples of close levels. For weak fields these levels are: $|3\rangle|n_1, n_2\rangle$ (atomic state $|3\rangle$, n_1 quanta in the first field, and n_2 quanta in the second one), $|1\rangle|n_1 + 1, n_2\rangle$, and $|2\rangle|n_1, n_2 + 1\rangle$. If we consider the system within perturbation theory, we use the atomic basis of states. They are close in energy; in the resonant approximation the field-induced transitions occur only inside each triple. The ‘dressed’ energy levels are found from the secular equation for each triple (10). In the resonant approximation the triples are independent.

The resonances in the spectra occur when two eigenvalues λ are close to each other. The minimal distance between them gives the order of resonance width. The general idea is that one can consider two close λ separately from others and write the quadratic secular equation for them. These two eigenvalues are pushed apart by the intermixing—the general scenario due to the Hermiticity of interaction.

When $G_2 \ll G_1, \Omega_{1,2}$, λ is found from the condition of vanishing of the l.h.s. in (16):

$$\lambda_{1,2} = (\,\Omega_1 \pm (\Omega_1^2 + 4|G_1|^2)^{1/2}/2, \qquad \lambda_3 = \Omega_2. \tag{17}$$

First two dressed states result from $|1\rangle$ and $|3\rangle$ by the first wave field splitting. The third one corresponds to $|2\rangle$. The first wave makes the distance between λ_1 and λ_2 not less than $2|G_1|$. Then the resonances in the first wave absorption spectrum undergo power broadening—their width becomes proportional to $|G_1|$.

When Ω_2, i.e. λ_3 is close to λ_1, we can consider λ_1 and λ_3 separately from λ_2. Denoting $\Omega_2 = \lambda_1 + \Omega$, $\lambda = \lambda_1 + \Lambda$, we obtain

$$M_{1,2} = \frac{1}{2}\left(1 \mp \frac{\Omega_1}{(\Omega_1^2 + 4|G_1|^2)^{1/2}}\right), \qquad \Lambda(\Lambda - \Omega) \simeq M_1|G_2|^2. \tag{18}$$

The energies λ_1 and λ_3 are split by the second wave, whose intensity is multiplied by the factor M_1. The case $\lambda_3 \simeq \lambda_2$ is handled in a similar way: one needs only to substitute λ_2 and

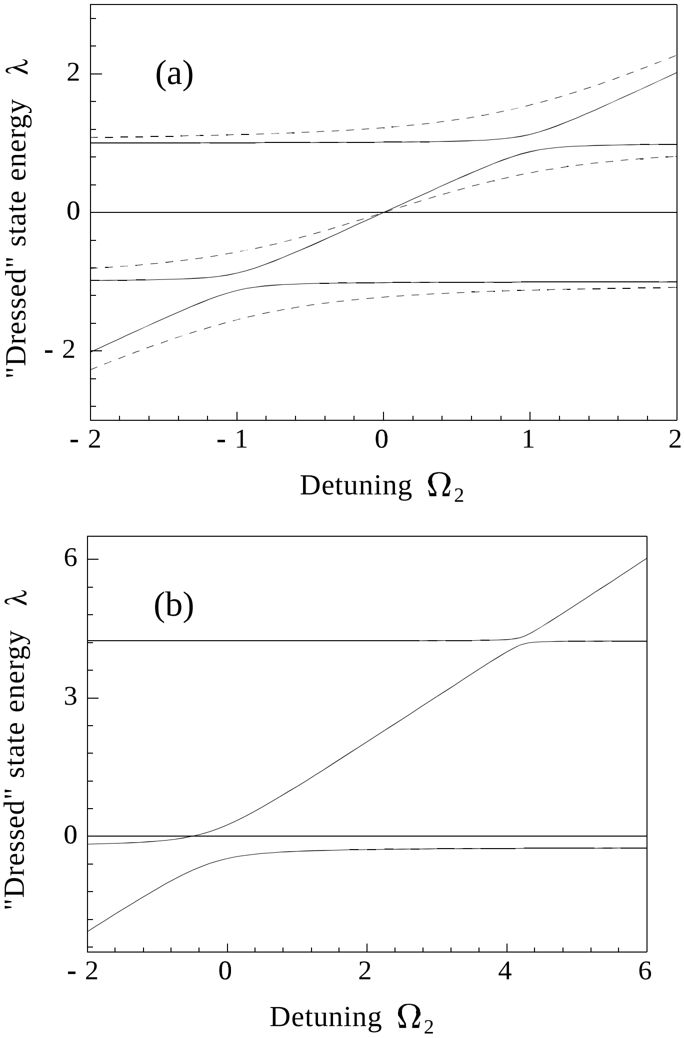

Figure 2. Dressed state energies $\lambda(\Omega_2)$, $G_1 = 1$. One can see the asymptotics $\lambda_{1,2}$, λ_3 (17). When $\lambda_3 \simeq \lambda_{1,2}$ the energies λ repel each other with the rate $(M_{1,2})^{1/2}|G_2|$ (see (18)). (*a*) $\Omega_1 = 0$, $G_2^2 = \frac{1}{32}$ (solid curve), $G_2^2 = \frac{1}{2}$ (dashed curve). (*b*) $\Omega_1 = 4$, $G_2^2 = \frac{1}{8}$. When $\Omega_1 \neq 0$ we have $M_1 \neq M_2$, the splitting by the second wave is asymmetric.

M_2 for λ_1 and M_1. The coefficients $M_{1,2}$ have been called 'memory factors' or 'correlation factors' [5, 13]. One can diagonalize the Hamiltonian $\hat{H}$ neglecting the second field G_2 and turn from bare states $|1\rangle$ and $|3\rangle$ to the states dressed by the first field having energies $\lambda_{1,2}$. Then $\sqrt{M_{1,2}}|G_2|$ are simply the matrix elements of interaction between these dressed states and bare state $|2\rangle$, induced by the second field. It is well known that while the second wave is weak (probe), its absorption spectrum $P(\Omega_2)$ has two resonance lines (Autler–Townes doublet) [2]. In this case their width is determined by the relaxation constants. From the behaviour of the dressed states we conclude that these two lines will undergo power broadening by the second field if it becomes stronger.

5. Probe field spectrum

We consider the scheme illustrated in figure 1(*b*) in the case where only the fourth level is excited (i.e. $\boldsymbol{A}_0 = (0, 0, 0, 1)$). We assume the fields G_μ and G_ν to be weak. Thus, only the first order of perturbation theory in $G_{\mu,\nu}$ is needed. In the main order, $A_4 = \exp\left(-\Gamma_4 t/2 + \mathrm{i}(\Omega_1 + \Omega_\mu)t\right) \cdot \theta(t)$, where $A_4 = a_4 \exp\left(\mathrm{i}(E_4 - \Omega_1 - \Omega_\mu)t\right)$. The probability amplitudes A_i, $i = 1, 2, 3$ have the form

$$\begin{aligned} A_i(t) &= \int \mathrm{d}t_0 \left(G_\mu \mathcal{D}_{i1}(t-t_0) + G_\nu \mathrm{e}^{-\mathrm{i}\varepsilon t} \mathcal{D}_{i2}(t-t_0)\right) A_4(t_0) \\ &= \int \frac{\mathrm{d}\omega}{2\pi} \mathrm{e}^{-\mathrm{i}\omega t} \left(\frac{G_\mu \mathcal{D}_{i1}(\omega)}{\Gamma_4/2 + \mathrm{i}(\Omega_1 + \Omega_\mu - \omega)} + \frac{G_\nu \mathcal{D}_{i2}(\omega)}{\Gamma_4/2 + \mathrm{i}(\Omega_2 + \Omega_\nu - \omega)} \right), \end{aligned} \tag{19}$$

where $\varepsilon = \Omega_2 + \Omega_\nu - \Omega_1 - \Omega_\mu$ and $\hat{\mathcal{D}}(t) = \int \mathrm{d}\omega\, \mathrm{e}^{-\mathrm{i}\omega t} \hat{\mathcal{D}}(\omega)/2\pi$. The power absorbed from the field G_μ is equal to

$$\begin{aligned} P_\mu(\Omega_\mu) &= 2\omega_\mu \operatorname{Re} \mathrm{i} G_\mu^* \int_0^\infty \mathrm{d}t\, A_4^* A_1 = 2\omega_\mu \operatorname{Re}\, \mathrm{i} G_\mu^* \int \frac{\mathrm{d}\omega}{2\pi} \frac{1}{\Gamma_4/2 - \mathrm{i}(\Omega_1 + \Omega_\mu - \omega)} \\ &\quad \times \left(\frac{G_\mu \mathcal{D}_{11}(\omega)}{\Gamma_4/2 + \mathrm{i}(\Omega_1 + \Omega_\mu - \omega)} + \frac{G_\nu \mathcal{D}_{12}(\omega)}{\Gamma_4/2 + \mathrm{i}(\Omega_2 + \Omega_\nu - \omega)} \right) \\ &= 2\omega_\mu \operatorname{Re} \mathrm{i} G_\mu^* \left(\frac{G_\mu \mathcal{D}_{11}(\omega_*)}{\Gamma_4} + \frac{G_\nu \mathcal{D}_{12}(\omega_*)}{\Gamma_4 + \mathrm{i}\varepsilon} \right), \end{aligned} \tag{20}$$

where $\omega_* = \Omega_1 + \Omega_\mu + \mathrm{i}\Gamma_4/2$. The expression for the absorption $P_\mu(\Omega_\mu)$ linearly depends on the Green functions $\mathcal{D}_{11}$ and $\mathcal{D}_{12}$, since we considered the linear response of the three-level system. Note that if the initial excitation goes to levels 1, 2, 3, then the result will contain the product of two Green functions. If one tries to calculate the power absorbed from the field G_μ (20) using (14), where two Green functions and the matrix $\hat{P}$ are taken for the four-level system, then one of these Green functions ($\hat{\mathcal{D}}$ or $\hat{\mathcal{D}}^c$) will be equal to $\mathcal{D}_{44} \simeq D_{44}$.

The second term in brackets in (20) corresponds to the parametric process. There is a closed cycle made up from fields μ-1-2-ν with the whole detuning ε. This contribution to the spectrum $P_\mu(\varepsilon)$ has the width Γ_4. However, this is only the gain in the thin media approximation (or the parametric instability increment). If the parametric process leads to instability, then the emission spectrum of thick media has different width, which can be much smaller.

6. Spectrum $\hat{\mathcal{P}}(\Omega_2)$

Here we consider the case of small equal relaxation constants $\Gamma_1 = \Gamma_2 = \Gamma_3 \to 0$. We focus only on the diagonal elements of the matrix $\hat{\mathcal{P}}$, which give the absorption power when the initial excitation goes to the real atomic levels. For real G_j, $j = 1, 2$, the components $\langle i|I\rangle\rangle$ of the eigenvectors of $\hat{H}$ are also real. From (14) and (15) immediately follows

$$\mathcal{P}_{ii}(\Omega_2) = 2G_2 \sum_{I \neq J} \frac{\langle i|I\rangle\rangle \langle\langle I|2\rangle \langle 3|J\rangle\rangle \langle\langle J|i\rangle}{\lambda_J - \lambda_I}. \tag{21}$$

As a function of λ, (21) is a rational expression symmetric with respect to permutations. Then it can be easily expressed in terms of the coefficients of (10), i.e. wave detunings and intensities. Note that $\operatorname{Tr} \hat{\mathcal{P}}$ is equal to zero, since $\sum_i \langle i|I\rangle\rangle \langle\langle J|i\rangle = \delta_{IJ}$ (in the case of equal populations of levels the interaction is absent). The power absorbed from the second wave when the excitation goes to level two is given by

$$\mathcal{P}_{22} = 2G_2^2 P/Q, \qquad I_1 = G_1^2, \qquad I_1 = G_1^2, \qquad I_{12} = G_1^2 + G_2^2, \tag{22}$$

$$P = (4I_{12} + \Omega_1^2)(I_{12} + (\Omega_1 - \Omega_2)^2) - I_1\Omega_2^2 - I_2[3I_{12} + 7\Omega_1^2 - 10\Omega_1\Omega_2 + 4\Omega_2^2], \tag{23}$$

$$\begin{aligned} Q = {} & (4I_{12} + \Omega_1^2)(I_{12} + (\Omega_1 - \Omega_2)\Omega_2)^2 + I_2(\Omega_1 - \Omega_2) \\ & \times[2(\Omega_1 - 2\Omega_2)(9I_{12} + (\Omega_1 + \Omega_2)(2\Omega_1 - \Omega_2)) - 27I_2(\Omega_1 - \Omega_2)]. \end{aligned} \tag{24}$$

Consider the simple case $\Omega_1 = 0$. One can measure frequency in $|G_1|$ units, i.e. set $|G_1| = 1$ and obtain

$$\mathcal{P}_{22}(\Omega_2) = 2G_2^2 \frac{G_2^4 + 5G_2^2 + 4 + 3\Omega_2^2}{4(G_2^2 + 1)^3 + (G_2^4 + 20G_2^2 - 8)\Omega_2^2 + 4\Omega_2^4}. \tag{25}$$

The poles in the spectrum are situated at the points

$$\Omega_{2*} = \pm\left(1 - \frac{5G_2^2}{2} - \frac{G_2^4}{8} \pm \frac{G_2(G_2^2 - 8)^{3/2}}{8}\right)^{1/2}, \tag{26}$$

(for the poles positions in other simple cases see appendix B). The asymptotics for small and large G_2 are the following:

$$\Omega_{2*} = \pm\begin{cases} 1 \pm \sqrt{2}\mathrm{i}G_2 - G_2^2/4, & G_2 \ll 1, \\ 2\mathrm{i}G_2, \mathrm{i}G_2^2/2, & G_2 \gg 1. \end{cases} \tag{27}$$

The poles $\Omega_{2*} = \pm \mathrm{i}G_2^2/2$ for $G_2 \gg 1$ are inessential, since their contribution to the spectrum is negligible. The positions of the poles $\Omega_{2*} = \pm 2\mathrm{i}G_2$ correspond to the usual power broadening by the second wave or to saturation [1]. The resonance positions $\Omega_{2*} \simeq \pm 1$ for $G_2 \ll 1$ are the positions of Autler–Townes doublet components [2]. The addition $\sqrt{2}\mathrm{i}G_2$ is the power broadening of these components by the second field. The term $-G_2^2/4$ is the nonlinear shift of the resonance produced by this field (see appendix A).

When $G_2^2 = 8$ the poles in the spectrum $P(\Omega)$ merge together: $\Omega_{2*} = \pm 3^{3/2}\mathrm{i}$, $\mathcal{P}_{22} = 4(104 + 3\Omega_2^2)/(27 + \Omega_2^2)^2$ (see figure 4(*a*)). At $G_2 = \sqrt{8}$ all dressed state energies are equal to each other: $\lambda_{1,2,3} = \Omega_{2*}/3 = \pm\sqrt{3}\mathrm{i}$. This situation seems to be destroyed when $\Omega_1 \neq 0$ (see figure 4(*b*)) or relaxation constants are not negligible.

In figure 3 one can see that the power broadening of Autler–Townes doublet components is proportional to the amplitude of the second wave. When $\Omega_1 \neq 0$ the broadening is asymmetric because the rates of interaction between level two and the states dressed by the first field are not equal to each other ($M_1 \neq M_2$). When the second field becomes stronger the resonances come closer to each other. When $G_2 \gg G_1, \Omega_1$ the poles in the spectrum are situated at the points

$$\Omega_{2*} = \pm 2\mathrm{i}G_2, \qquad \Omega_{2*} = -\frac{G_2^2}{\Omega_1 \pm 2\mathrm{i}}. \tag{28}$$

The first pair of poles corresponds to the simple resonance between levels two and three taking into account the saturation caused by the second field. If we decrease the intensity of the second wave, then this resonance will be transformed to the component of Autler–Townes doublet $\Omega_2 = (\Omega_1 - (\Omega_1^2 + 4|G_1|^2)^{1/2})/2$. The second pair of poles corresponds to the other component of the doublet (if the first field is weak, then this line is the resonance between the virtual level and level two, i.e. two-photon resonance). The position of this resonance for $G_2 \gg G_1, \Omega_1$ (the real part of the pole position) has the sign opposite to that of Ω_1. Note that in figure 3(*b*) the narrower part of the spectrum goes to the left. For extremely high values of G_2 the second part of the spectrum becomes wider than the first one (the width becomes proportional to G_2^2, see dots in figure 4(*b*)).

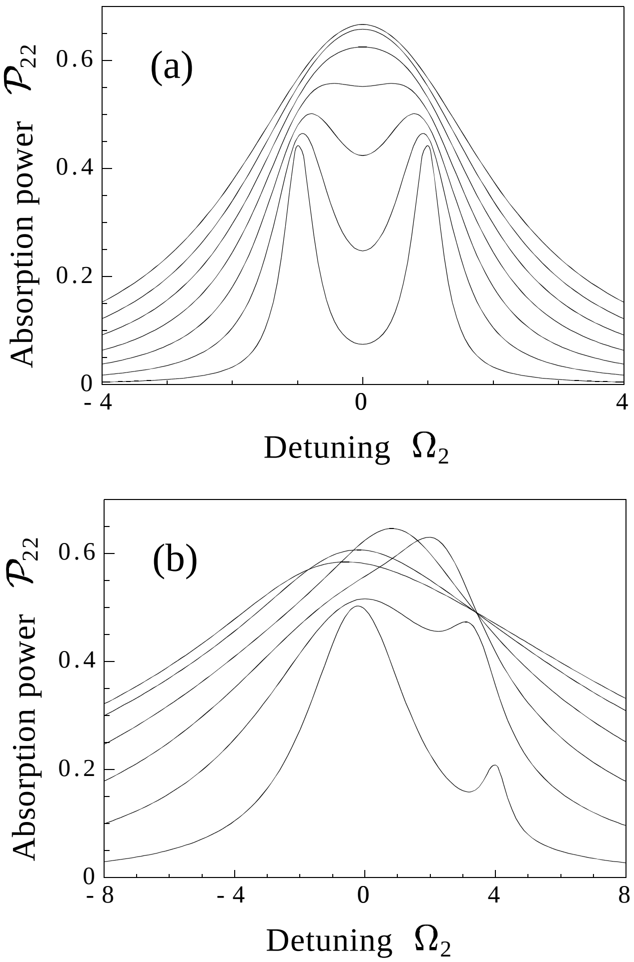

Figure 3. Absorption power $\mathcal{P}_{22}(\Omega_2, G_2)$ at $G_1 = 1$, $\Omega_1 = 0$ (*a*) and $\Omega_1 = 4$ (*b*). The amplitude of the second field G_2 varies from 0.2 to 1.4 with the step 0.2 (*a*), and from 0.5 to 3 with the step 0.5 (*b*).

The fact that the width of one of the two resonances is proportional to G_2^2 for $G_2 \gg G_1, \Omega_1$ corresponds to the following: the second field has the amplitude G_2, which is much greater than the distance $(\Omega_1^2 + 4|G_1|^2)^{1/2}$ between the states dressed by the first field. Then the second field feels these two states as only one state; it cannot see the energy structures with resolution higher than $|G_2|$. As the system becomes similar to an ordinary two-level system, only one resonance with the power width $2|G_2|$ should remain. There are different ways to kill the second resonance: to increase its width or to decrease its amplitude, both ways being used here. Mathematically, the fourth-order algebraic equation for Ω_{2*} is transformed to the second-order algebraic equation, i.e. the coefficients of Ω_{2*}^4 and Ω_{2*}^3 are comparatively small. It is well known that the algebraic equation with small leading coefficient ϵ has at least one root which is large in parameter $1/\epsilon$.

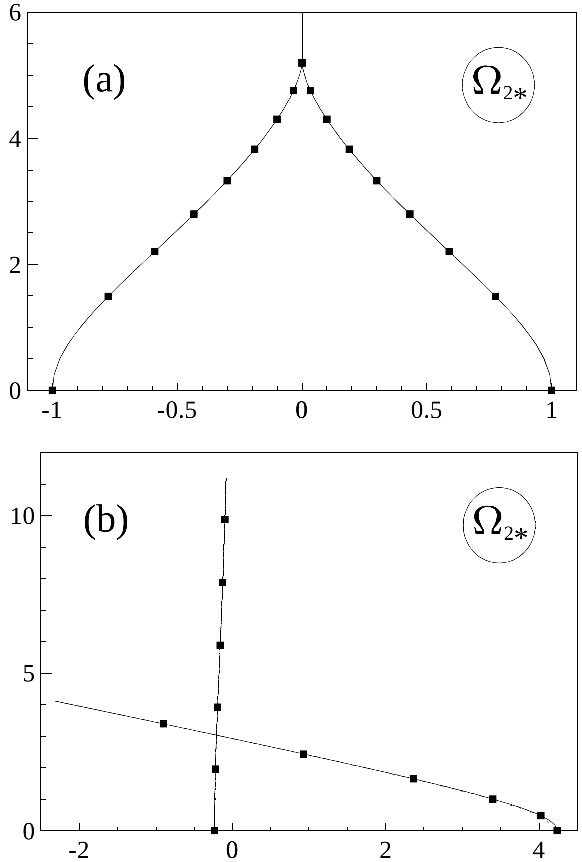

Figure 4. Pole positions $\Omega_{2*} = (\mathrm{Re}\,\Omega_{2*}, \mathrm{Im}\,\Omega_{2*})$ for different values of G_2, $G_1 = 1$, $\Omega_1 = 0$ (*a*) and $\Omega_1 = 4$ (*b*). The dots in (*a*) correspond to $G_2 = \sqrt{n}$, $n = 0, 1, \ldots, 8$, and in (*b*) to $G_2 = 0, 1, \ldots, 5$.

7. Doppler broadening of spectra

One can take into account the Doppler broadening of absorption spectra by substituting $\Omega_1 - \boldsymbol{k}_1\boldsymbol{v}$ and $\Omega_2 - \boldsymbol{k}_2\boldsymbol{v}$ for Ω_1 and Ω_2 and integrating the result (22) with some distribution of particles $\hat{\rho}(\boldsymbol{v})$. Consider the case of copropagating waves, where only a longitudinal projection $v_{||}$ of velocity is needed. When the Doppler width is infinite, the answer for the spectrum is the integral of the rational function $p_4(v_{||})/p_6(v_{||})$ over velocity, where $p_n(x)$ is the nth degree polynomial. Some simplification takes place when $k_1 = k_2$ or $k_i = 0$ for some $i = 1, 2$. In the general case the expressions for velocity-averaged $\hat{\mathcal{P}}(\Omega_2)$ are difficult to derive and analyse. We will discuss the effects of the Doppler broadening only qualitatively.

When the field G_2 is weak and $k_2 < k_1$, the velocity-averaged spectrum $\hat{\mathcal{P}}(\Omega_2)$ has two narrow asymmetric resonance lines coming from turning points [14] of frequency branches

$$\Omega_{2*}(v_{||}) \simeq k_2 v_{||} + (\Omega_1 - k_1 v_{||} \pm ((\Omega_1 - k_1 v_{||})^2 + 4|G_1|^2)^{1/2})/2. \qquad (29)$$

The turning point is the extreme point of velocity dependence of resonance frequencies Ω_{2*}, i.e. at the turning point v_{tp} we have $(\mathrm{d}\Omega_{2*}/\mathrm{d}v_{||})|_{v_{||}=v_{\mathrm{tp}}} = 0$. The spectra of this type are well known (see, e.g., [15]). For the universal shape of asymmetric line due to the turning point

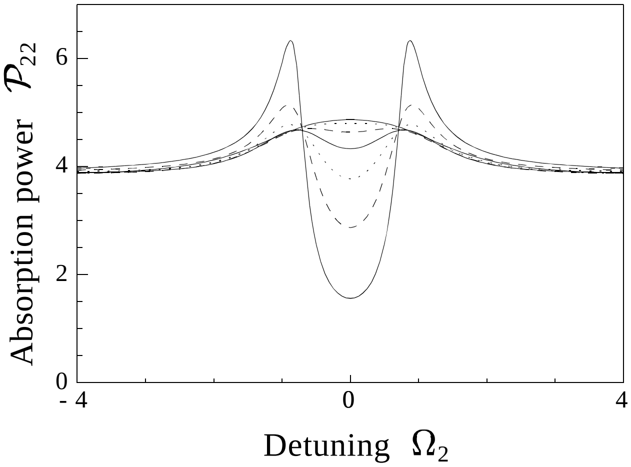

Figure 5. Velocity-averaged spectrum $\mathcal{P}_{22}(\Omega_2)$ (numerical calculations), $G_1 = 1$, $\Omega_1 = 0$, $k_1 = 1$, $k_2 = 0.8$, G_2 varies from 0.2 to 1.4 with the step 0.2. Uniform velocity distribution or infinite Doppler width.

see [16]. When $G_2 \ll \Gamma$, the characteristic width of this resonance line is of order Γ. When $G_2 \gg \Gamma$, the resonance line width is determined by the power broadening by the second wave. Nevertheless, while $G_2 \ll G_1$ one can use the expression (29) (where the influence of G_2 on the resonance position is neglected) for Ω_{2*}. The rough feature of the spectrum—two narrow asymmetric resonance lines—remains, but the width of these lines depends on G_2 (see figure 5).

When Ω_1 and G_1 are compared to the Doppler width, an 'isolated peak' appears in the spectrum. Its width depends on Ω_1, G_1 (see [14, 17]) and is larger than G_2 in the general case. The dependence of the width on Ω_1 and G_1 due to inhomogeneous broadening is not a subject of this paper, but at certain conditions the width decreases and becomes a value of order G_2.

When $G_2 \sim G_1 \gg \Gamma$, the absorption spectrum does not contain narrow lines, i.e. all resonance widths are of order $|G_1|$ or $|G_2|$.

If $k_2 > k_1$, then the frequency branches (29) have no turning points, and the absorption spectrum $\hat{\mathcal{P}}(\Omega_2)$ has no narrow resonance lines even when the second wave is weak. If we increase the intensity G_2^2, the narrow lines will not appear.

8. Conclusions

Let us summarize the description of the three-level system resonantly interacting with two strong monochromatic waves.

The expression for the spectrum contains resonant denominators $\lambda_I - \lambda_J^*$, where λ_I is the energy of the Ith 'dressed' state. For an N-level atomic system the dressed state energies satisfy the algebraic equation of Nth order which is the secular equation for N levels of the system 'atom + field' that are close in energy. When two dressed states are close to each other and all other states are far from them in energy, the secular equation can be reduced to the quadratic secular equation for these two states. The part of the atomic system consisting of these two dressed states behaves as an ordinary two-level system.

If some connected component of strong fields covers the whole atomic system with N states, then all N 'dressed' states will be far from each other. As a consequence, all resonances in the spectra will be wide. An atomic system can be covered by strong fields, but the strong fields can be disconnected (e.g. fields 1 and ν in figure 1(*b*) are strong). Then the matrix elements between the states dressed by field 1 and by field ν will be small and resonances will be narrow.

When the second wave is weak we can think that it couples level 2 (see figure 1) and levels 1, 3 split by the first wave. The distance between the split levels is much greater than the amplitude of the second wave; these two transitions cannot be resonant simultaneously. Each transition can be treated as a two-level system. The effects of power broadening by the second wave appear. When the second wave is very strong, only the two-level system coupled by it remains and Autler–Townes doublet becomes blurred.

Indeed, the power width in a two-level system depends on the ratio of relaxation constants Γ of levels even when the latter are small. Thus, the power width of Autler–Townes component, due to the intensity of the second wave, should also depend on the ratio of Γs. The expressions will be similar to the well-known ones for a two-level system: one should take Γ_2 and the decay rate of the state 'dressed' by the first field.

In experiment [10] the first wave detuning Ω_1 was equal to zero and two components had the same power width. It is interesting to observe the asymmetric broadening when $\Omega_1 \neq 0$.

When the second wave is generated in the media, the self-consistent problem taking into account energy losses should be solved. The doublet gives two frequencies of generation, which merge together at a certain rate of reflection losses.

Acknowledgments

The author is grateful to E V Podivilov and S A Babin for useful discussions. This paper was partially supported by the Soros Foundation (gr. a98-674), by INTAS (gr. 96-0457) within the program of ICFPM, and by RFBR (gr. 96-02-19052).

Appendix A. Resonances in probe field spectrum

Consider the n-level system with dressed energies $\lambda_1, \lambda_2, \ldots, \lambda_n$ and probe field spectrum, where the probe field G with detuning Ω resonantly interacts with level $|0\rangle$ and the n-level system. The dressed energies λ of the whole system can be found from the Hamiltonian:

$$\hat{H} = \begin{pmatrix} \Omega & \sqrt{M_1}G & \sqrt{M_2}G & \ldots & \sqrt{M_n}G \\ \sqrt{M_1}G & \lambda_1 & 0 & \ldots & 0 \\ \sqrt{M_2}G & 0 & \lambda_2 & \ldots & 0 \\ \ldots & \ldots & \ldots & \ldots & \ldots \\ \sqrt{M_n}G & 0 & 0 & \ldots & \lambda_n \end{pmatrix}, \tag{A.1}$$

$$\lambda - \Omega = \left(\frac{M_1}{\lambda - \lambda_1} + \frac{M_2}{\lambda - \lambda_2} + \cdots + \frac{M_n}{\lambda - \lambda_n} \right) G^2. \tag{A.2}$$

The resonances are situated at $\Omega \simeq \lambda_i$, $i = 1, 2, \ldots, n$. When Ω is close to λ_1 the equation (A.2) has multiple roots if

$$\Omega = \lambda_1 \pm 2\mathrm{i}\sqrt{M_1}G - \left(\frac{M_2}{\lambda_1 - \lambda_2} + \ldots + \frac{M_n}{\lambda_1 - \lambda_n} \right) G^2 + \mathrm{O}(G^3). \tag{A.3}$$

The Green function $\mathcal{D}_{00}(\omega)$ has the form

$$\mathcal{D}_{00}^{-1}(\omega) = ((\omega - \hat{H})_{00}^{-1})^{-1} = \omega - \Omega - \sum_i \frac{M_i G^2}{\omega - \lambda_i}. \tag{A.4}$$

The mass operator $\omega - \Omega - \mathcal{D}_{00}^{-1}$ is the sum of loops through levels $i = 1, 2, \ldots, n$. When $\Omega \simeq \lambda_1$ the loop $i = 1$ is much greater than the others. This loop gives the power broadening of the resonance $\Omega \simeq \lambda_1$. Other loops renormalize the detuning, which gives the shift of the resonance. Note that the shift is of order $G^2/(\lambda_1 - \lambda_i)$, which is much smaller than the width of the resonance $\sim G$. The shift of the resonance in (A.3) can be important only when the field G is not very weak. Then it gives a qualitatively correct answer. When the probe field G is weak enough, the shift can be neglected and the resonances $\Omega \simeq \lambda_i$ live independently.

Appendix B. Resonance positions in spectra

Although it is not very difficult to calculate the integral (14), some properties of $\hat{\mathcal{P}}(\Omega_2)$ can be elucidated by simple algebraic manipulations. It should be noticed that the same result (also without integration) can be obtained using the density matrix formalism.

The resonance position $\Omega_2 = \Omega$ in the second wave absorption spectrum can be found from the condition that one pole of $\hat{\mathcal{D}}$ coincides with at least one pole of $\hat{\mathcal{D}}^c$. Consider the following combination of eigenvalues:

$$\mathcal{Z} = \prod_{I,J} (\lambda_I - \lambda_J^c) = \mathrm{Det}\, f_{\hat{H}}(\hat{H}^c) = -\mathrm{Det}\, f_{\hat{H}^c}(\hat{H}), \tag{B.1}$$

where $f_{\hat{A}}(\lambda) = \mathrm{Det}\,(\hat{A} - \lambda)$. The resonance occurs when $\mathcal{Z} = 0$. As a function of λs and λ^c, $\mathcal{Z}$ is symmetric with respect to permutations, so it can be easily expressed in terms of detunings, fields, and relaxation constants. In the general case the expression for $\mathcal{Z}$ is bulky, hence we will consider some particular cases. We assume that $\Gamma_1 = \Gamma_2 = \Gamma_3 = \Gamma$; $\Omega_1 = \Omega_2 = \Omega$ (B.3), $\Omega_1 = 0$ (B.4), $\Omega_1 = \Gamma = 0$ (B.5):

$$\mathcal{Z} = -\Gamma^3 Z, \qquad I_1 = G_1^2, \qquad I_1 = G_1^2, \qquad I_{12} = G_1^2 + G_2^2, \tag{B.2}$$

$$Z = (\Omega^2 + 4I_{12} + \Gamma^2)(\Gamma^2\Omega^2 + (I_{12} + \Gamma^2)^2), \tag{B.3}$$

$$Z = (4I_{12} + \Gamma^2)((\Omega_2^2 - I_{12} - \Gamma^2)^2 + 4\Gamma^2\Omega_2^2) - I_2\Omega_2^2(4\Omega_2^2 - 36I_1 - 9I_2), \tag{B.4}$$

$$Z = 4I_{12}(\Omega_2^2 - I_{12})^2 - I_2\Omega_2^2(4\Omega_2^2 - 36I_1 - 9I_2). \tag{B.5}$$

References

[1] Karplus R and Schwinger I A 1948 *Phys. Rev.* **73** 1020

[2] Autler S H and Townes C H 1955 *Phys. Rev.* **100** 703

[3] Feld M S and Javan A 1968 *Phys. Rev. Lett.* **20** 578

[4] Feld M S and Javan A 1969 *Phys. Rev.* **177** 540

[5] Popova T Ya, Popov A K, Rautian S G and Feoktistov A A 1969 *Zh. Eksp. Teor. Fiz.* **57** 444 (in Russian)

[6] Vasilenko L S, Chebotaev V P and Shishaev A V 1970 *Pis. Zh. Eksp. Teor. Fiz.* **12** 161 (Engl. transl. 1970 *JETP Lett.* **12** 113)

[7] Hänsch Th and Toschek P 1970 *Z. Phys.* **236** 213

[8] Salomaa R 1977 *J. Phys. B: At. Mol. Phys.* **10** 3005

[9] Popov A K, Kuchin V M and Myslivets S A 1998 *Zh. Eksp. Teor. Fiz.* **113** 445 (Engl. transl. 1998 *JETP* **86** 244)

[10] Wellegehausen B 1982 *ACS Symposium Series* **179** (Washington, DC: American Chemical Society) p 461

[11] Cohen-Tannoudji C 1977 *Frontiers in Laser Spectroscopy* (Amsterdam: North-Holland)

[12] Berman P R and Salomaa R 1982 *Phys. Rev.* A **25** 2667

[13] Rautian S G and Shalagin A M 1991 *Kinetic Problems of Nonlinear Spectroscopy* (Amsterdam: Elsevier)

[14] Bykova O G, Lebedeva V V, Bykova N G and Petukhov A V 1982 *Opt. Spektrosk.* **53** 171 (in Russian)

[15] Beterov I M and Chebotaev V P 1974 *Progress in Quantum Electronics* vol 3, part 1, ed J H Sanders and S Stenholm (Oxford: Pergamon) pp 1–106

[16] Shapiro D A and Stepanov M G 1998 *Pis. Zh. Eksp. Teor. Fiz.* **68** 27 (Engl. transl. 1998 *JETP Lett.* **68** 29)

[17] Janik G, Nagourney W and Dehmelt H 1985 *J. Opt. Soc. Am.* B **2** 1251