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Resonances of electromagnetically induced transparency and absorption in a light field of elliptically polarised waves

D.V. Kovalenko, M.Yu. Basalaev, V.I. Yudin, A.V. Taichenachev

Abstract. **Electromagnetically induced transparency (EIT) and electromagnetically induced absorption (EIA) resonances excited by a strong two-frequency field are considered for various values of the total angular momenta of the ground (F_g) and excited (F_e) states at a degenerated optical closed transition $F_g \to F_e$. The light field is formed by two co-propagating waves with arbitrary elliptical polarisations. The process of spontaneous transfer of anisotropy from the excited state to the ground state is shown to determine the formation of the EIA resonance at the transition $F_g = F \to F_e = F + 1$. The results obtained generalise the classification of transitions into 'bright' ($F_g = F \to F_e = F + 1$) and 'dark' ($F_g = F \to F_e = F$ and $F_g = F \to F_e = F - 1$) transitions with respect to the direction of a subnatural resonance.**

Keywords: *electromagnetically induced transparency, electromagnetically induced absorption, anisotropy transfer, low-frequency Zeeman coherence.*

1. Introduction

In modern laser spectroscopy, the nonlinear interference effects associated with atomic coherence attract considerable interest. An example of these effects is the resonances of electromagnetically induced transparency (EIT) [1] and electromagnetically induced absorption (EIA) [2]. The first type of resonance is due to coherent population trapping (CPT) [3–5], when the electromagnetic field does not interact with the atomic medium; in this case, a long-lived coherent ('dark') state is formed and an ultra-narrow dip is observed in the absorption signal. In turn, the physical cause of the EIA resonance, which is opposite in sign to the EIT resonance, is the spontaneous transfer of anisotropy (including the low-frequency Zeeman coherence) from the excited state of the atom to the ground state [6]. The main feature of such resonances is their width, which can be much less than the natural width and reach hundreds or even units of hertz [7–9]. Due to this, they find many significant applications in quantum metrology [10–12], nonlinear optics [13, 14], optical communications [15], etc.

At present, various experimental [2, 16, 17] and theoretical [18–21] studies have made it possible to develop a classification of atomic dipole transitions with allowance for the sign of resonance (EIT or EIA) in the regime of weak saturation of the atomic transition. 'Dark' transitions are transitions of the type $F_g = F \to F_e = F$ and $F_g = F \to F_e = F - 1$ (where F_g and F_e are the total angular momenta of the atom in the ground and excited states, respectively), at which EIT resonances are observed. 'Bright' transitions are transitions of the type $F_g = F \to F_e = F + 1$, where EIA is formed. In particular, Lazebnyi et al [22] theoretically justified this classification in the framework of the perturbation theory for a two-frequency configuration of two co-propagating waves with arbitrary elliptical polarisations.

In this paper, we generalise the results obtained in [22] to the case of a strong field when the perturbation theory is not valid. To this end, we consider a model of an atomic medium with and without anisotropy transfer. It is shown that this problem can be reduced to equations for a density matrix with coefficients that periodically depend on time. Using the method of constructing a dynamic steady state [23], we calculate the periodic absorption signal for various parameters of the light field and angular momenta F_g and F_e. As a result, the previously established classification of dipole transitions by the sign of subnatural resonance is confirmed regardless of the intensities of light waves.

2. Theory and calculations

We consider the interaction of an elliptically polarised bichromatic field

$$\boldsymbol{E}(t) = E_1 \boldsymbol{e}_1 \mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2 \boldsymbol{e}_2 \mathrm{e}^{-\mathrm{i}\omega_2 t} + \text{c.c.} \tag{1}$$

with an atomic medium in which the ground (g) and excited (e) states that are degenerated with respect to the projections of the total angular momentum form a closed optical dipole transition $F_g \to F_e$ (Fig. 1). Here, $E_{1,2}$ and $\omega_{1,2}$ are the scalar amplitudes and frequencies of light waves, respectively. The unit complex vectors $\boldsymbol{e}_1$ and $\boldsymbol{e}_2$ of elliptic polarisation can be represented in a cyclic basis:

$$\boldsymbol{e}_j = \sum_{q=0,\pm 1} e_j^{(q)} \boldsymbol{e}_q \ \ (j = 1,2), \tag{2}$$

where $\boldsymbol{e}_{\pm 1} = \mp(\boldsymbol{e}_x \pm \mathrm{i}\boldsymbol{e}_y)/\sqrt{2}$; $\boldsymbol{e}_0 = \boldsymbol{e}_z$ are the unit vectors of the cyclic basis; and $e_j^{(q)}$ are the contravariant components of

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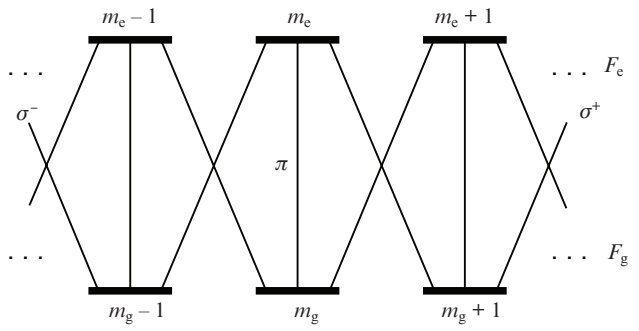

Figure 1. Diagram of atomic energy levels degenerated with respect to the projections of the total angular momenta of the ground (F_g) and excited (F_e) states on the quantization axis z. Lines denote the light-induced transitions of σ^+-, σ^-- and π-types.

the unit polarisation vector of the jth wave. Let us direct the x axis along the principal axis of the polarisation ellipse of the wave $\boldsymbol{E}_1$; then for unit polarisation vectors (2) we have

$$\begin{aligned} \boldsymbol{e}_1 &= -\sin(\varepsilon_1 - \pi/4)\boldsymbol{e}_{-1} - \cos(\varepsilon_1 - \pi/4)\boldsymbol{e}_{+1}, \\ \boldsymbol{e}_2 &= -\sin(\varepsilon_2 - \pi/4)\mathrm{e}^{\mathrm{i}\phi}\boldsymbol{e}_{-1} - \cos(\varepsilon_2 - \pi/4)\mathrm{e}^{-\mathrm{i}\phi}\boldsymbol{e}_{+1}. \end{aligned} \tag{3}$$

Here, ϕ is the angle between the main axes of the polarisation ellipses (Fig. 2); and the ellipticity parameter ε is defined in the interval $-\pi/4 \leqslant \varepsilon \leqslant \pi/4$, with $|\tan(\varepsilon)|$ being the ratio of the semiaxes of the ellipse, and the sign ε defining the direction of rotation of the electric component of the light field. In particular, $\varepsilon = \pm\pi/4$ и $\varepsilon = 0$ correspond to circular (right- and left-hand) and linear polarisations, respectively.

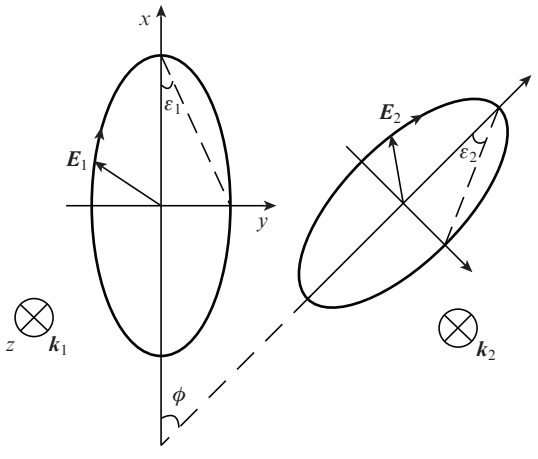

Figure 2. Mutual orientation of polarisation ellipses of the waves; $\boldsymbol{k}_1 = \boldsymbol{k}_2$ are the wave vectors of the waves, ϕ is the angle between the principal axes of the ellipses, and $\varepsilon_{1,2}$ are the ellipticity parameters.

The atomic medium is supposed to be sufficiently rarefied, which allows us to neglect the effects of interatomic interaction and solve the problem in the single-atom approximation. For a mathematical description of the interaction of atoms with an electromagnetic field, we will use the standard formalism of the density matrix $\hat{\rho}$, which saisfies the following equation:

$$\frac{\partial}{\partial t}\hat{\rho} = -\frac{\mathrm{i}}{\hbar}[(\hat{H}_0 + \hat{V}), \hat{\rho}] + \hat{\Gamma}\{\hat{\rho}\}, \tag{4}$$

where

$$\hat{H}_0 = \sum_{j=\mathrm{g,e}}\sum_{m_j}\mathcal{E}_j |F_j, m_j\rangle\langle F_j, m_j| \tag{5}$$

is the Hamiltonian of an unperturbed atom in the basis of Zeeman states $|F, m\rangle$; $\mathcal{E}_j$ is the energy of the jth state; m_j are the projections of the jth angular momentum F_j onto the quantisation axis z, running through the values $m_j = -F_j, -F_j + 1, \ldots, F_j$; $\hat{\Gamma}\{\hat{\rho}\}$ is the operator describing relaxation processes (spontaneous, collisional, time-of-flight, etc.); and $\hat{V} = -(\hat{\boldsymbol{d}}\cdot\boldsymbol{E})$ is the operator of interaction of atoms with the field ($\hat{\boldsymbol{d}}$ is the vector operator of the electric dipole moment), which in the rotating wave approximation is defined as

$$\hat{V} = \hbar R_1 \hat{V}_1(t) + \hbar R_2 \hat{V}_2(t) + \text{h.c.} \tag{6}$$

Here, $R_{1,2} = -dE_{1,2}/\hbar$ are the Rabi frequencies (d is the reduced matrix element of the dipole moment $\hat{\boldsymbol{d}}$); h.c. is the Hermitian conjugation;

$$\hat{V}_{1,2}(t) = \hat{V}_{1,2}\mathrm{e}^{-\mathrm{i}\omega_{1,2}t} \tag{7}$$

are the dimensionless interaction operators; and

$$\hat{V}_j = \hat{\boldsymbol{T}}\cdot e_j = \sum_{q=0,\pm1}\hat{T}_q e_j^{(q)} \ (j = 1, 2). \tag{8}$$

The cyclic components of the vector operator $\hat{\boldsymbol{T}}$ are expressed in terms of 3jm symbols:

$$\hat{T}_q = \sum_{\{m\}}(-1)^{F_e - m_e}\begin{pmatrix} F_e & 1 & F_g \\ -m_e & q & m_g \end{pmatrix}|F_e, m_e\rangle\langle F_g, m_g|. \tag{9}$$

We separate the density matrix $\hat{\rho}$ into four blocks,

$$\hat{\rho} = \hat{\rho}^{gg} + \hat{\rho}^{ee} + \hat{\rho}^{eg} + \hat{\rho}^{ge}, \tag{10}$$

where each block is a matrix

$$\hat{\rho}^{ab} = \sum_{m_a, m_b}\rho^{ab}_{m_a,m_b}|F_a, m_a\rangle\langle F_b, m_b|. \tag{11}$$

Since the density matrix is Hermitian, $\hat{\rho}^{gg\dagger} = \hat{\rho}^{gg}$, $\hat{\rho}^{ee\dagger} = \hat{\rho}^{ee}$, $\hat{\rho}^{eg\dagger} = \hat{\rho}^{ge}$. The diagonal matrix blocks $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$ describe the populations of atomic states and low-frequency (Zeeman) coherences, and the off-diagonal matrix blocks $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$ correspond to optical coherences.

Then, substituting expressions (5) and (6) into equation (4) and factoring fast time oscillations at the frequency of one of the waves in the optical coherences (for example, at ω_1)

$$\hat{\rho}^{eg} = \hat{\tilde{\rho}}^{eg}\mathrm{e}^{-\mathrm{i}\omega_1 t}, \quad \hat{\rho}^{ge} = \hat{\tilde{\rho}}^{ge}\mathrm{e}^{\mathrm{i}\omega_1 t}, \tag{12}$$

we obtain the system of equations:

$$\left(\frac{\partial}{\partial t} + \gamma_{\mathrm{opt}} + \Gamma_0 - \mathrm{i}\delta\right)\hat{\tilde{\rho}}^{eg} = -\mathrm{i}R_1(\hat{V}_1\hat{\rho}^{gg} - \hat{\rho}^{ee}\hat{V}_1) -$$

$$
\begin{aligned}
&-\mathrm{i}R_2\mathrm{e}^{-\mathrm{i}\Delta t}(\hat{V}_2\hat{\rho}^{\mathrm{gg}}-\hat{\rho}^{\mathrm{ee}}\hat{V}_2),\\
&\left(\frac{\partial}{\partial t}+\gamma_{\mathrm{opt}}+\Gamma_0+\mathrm{i}\delta\right)\hat{\tilde{\rho}}^{\mathrm{ge}}=-\mathrm{i}R_1(\hat{V}_1^{\dagger}\hat{\rho}^{ee}-\hat{\rho}^{\mathrm{gg}}\hat{V}_1^{\dagger})\\
&-\mathrm{i}R_2\mathrm{e}^{\mathrm{i}\Delta t}(\hat{V}_2^{\dagger}\hat{\rho}^{\mathrm{ee}}-\hat{\rho}^{\mathrm{gg}}\hat{V}_2^{\dagger}),\\
&\left(\frac{\partial}{\partial t}+\gamma_{\mathrm{sp}}+\Gamma_0\right)\hat{\rho}^{\mathrm{ee}}=-\mathrm{i}R_1(\hat{V}_1\hat{\tilde{\rho}}^{\mathrm{ge}}-\hat{\tilde{\rho}}^{\mathrm{eg}}\hat{V}_1^{\dagger})\\
&-\mathrm{i}R_2(\mathrm{e}^{-\mathrm{i}\Delta t}\hat{V}_2\hat{\tilde{\rho}}^{\mathrm{ge}}-\mathrm{e}^{\mathrm{i}\Delta t}\hat{\tilde{\rho}}^{\mathrm{eg}}\hat{V}_2^{\dagger}),\\
&\left(\frac{\partial}{\partial t}+\Gamma_0\right)\hat{\rho}^{\mathrm{gg}}-\Gamma_0\hat{\rho}^{\mathrm{gg}}(0)=\hat{\gamma}\{\hat{\rho}^{\mathrm{ee}}\}-\mathrm{i}R_1(\hat{V}_1^{\dagger}\hat{\tilde{\rho}}^{\mathrm{eg}}-\hat{\tilde{\rho}}^{\mathrm{ge}}\hat{V}_1)\\
&-\mathrm{i}R_2(\mathrm{e}^{\mathrm{i}\Delta t}\hat{V}_2^{\dagger}\hat{\tilde{\rho}}^{\mathrm{eg}}-\mathrm{e}^{-\mathrm{i}\Delta t}\hat{\tilde{\rho}}^{\mathrm{ge}}\hat{V}_2).
\end{aligned}
\tag{13}
$$

Here, $\Delta \equiv \omega_2 - \omega_1$; γ_{sp} is the rate of radiation decay of the excited state; Γ_0 is the constant responsible for the time of flight or diffusion relaxation in the ground state to the initial (isotropic) distribution $\hat{\rho}^{\mathrm{gg}}(0) = \hat{1}^{\mathrm{gg}} \cdot \mathrm{Tr}\{\hat{\rho}\}/(2F_{\mathrm{g}}+1)$ in the absence of a light field; $\hat{1}^{\mathrm{gg}}$ is the identity matrix of dimension $(2F_{\mathrm{g}}+1)\times(2F_{\mathrm{g}}+1)$; $\mathrm{Tr}\{...\}$ is the operation of computing the trace of the matrix; γ_{opt} is the relaxation rate of optical coherences; $\delta \equiv \omega_1 - \omega_0$ is the detuning of the frequency ω_1 of one of the waves of the light field from the transition frequency ω_0; and $\hat{\gamma}\{\hat{\rho}^{\mathrm{ee}}\}$ is an operator describing the arrival of atoms from an excited level to the ground level. In the standard spontaneous relaxation model with anisotropy transfer taken into account, we have

$$
\hat{\gamma}\{\hat{\rho}^{\mathrm{ee}}\} = \gamma_{\mathrm{sp}}(2F_{\mathrm{e}}+1)\sum_{q=0,\pm1}\hat{T}_q^{\dagger}\hat{\rho}^{\mathrm{ee}}\hat{T}_q. \tag{14}
$$

In the model without anisotropy transfer, another expression holds:

$$
\hat{\gamma}\{\hat{\rho}^{\mathrm{ee}}\} = \gamma_{\mathrm{sp}}\frac{\hat{1}^{\mathrm{gg}}\cdot\mathrm{Tr}\{\hat{\rho}^{\mathrm{ee}}\}}{2F_{\mathrm{g}}+1}. \tag{15}
$$

Note that for the cyclic transition $F_{\mathrm{g}} \to F_{\mathrm{e}}$, the total population at the ground and excited levels is conserved:

$$
\mathrm{Tr}\{\hat{\rho}^{\mathrm{gg}}\} + \mathrm{Tr}\{\hat{\rho}^{\mathrm{ee}}\} = 1. \tag{16}
$$

One can see that the right-hand sides of equations (13) are periodic functions of time with a period $T = 2\pi/|\Delta|$. To calculate the time dependence of the density matrix described by these equations, we used our method of constructing a dynamic steady state, described in detail in [23]. The essence of this method is as follows. First, the system of differential equations for the density matrix (13) is presented in the vector form:

$$
\partial_t\boldsymbol{\rho}(t) = \hat{L}(t)\boldsymbol{\rho}(t), \quad \mathrm{Tr}\{\hat{\rho}(t)\} = \sum_j \rho_{jj}(t) = 1. \tag{17}
$$

Here, the column vector $\boldsymbol{\rho}(t)$ is formed from the matrix elements $\rho_{jk}(t)$, and the linear operator $\hat{L}(t)$ is constructed from the coefficients of the dynamic system of equations (13). If for some moment of time t_1 there exists a vector $\boldsymbol{\rho}(t_1)$, then, in accordance with equation (13), for another moment of time, t_2, we can write:

$$
\boldsymbol{\rho}(t_2) = \hat{A}(t_2, t_1)\boldsymbol{\rho}(t_1), \tag{18}
$$

where the two-dimensional evolution operator $\hat{A}(t_2, t_1)$ is found from the matrix dependence $\hat{L}(t)$. If the periodicity condition is true, $\hat{L}(t+T) = \hat{L}(t)$, for arbitrary t_2 and t_1 we have a relation:

$$
\hat{A}(t_2+T,\ t_1+T) = \hat{A}(t_2,\ t_1). \tag{19}
$$

In our work [23], we rigorously proved the existence of a periodic solution $\boldsymbol{\rho}(t+T) = \boldsymbol{\rho}(t)$ for an arbitrary periodically excited quantum system. Due to relaxation processes, this state is realised as asymptotic ($t \to +\infty$) regardless of the initial conditions. Thus, periodicity is the main property of a dynamic steady state, which satisfies the equation:

$$
\boldsymbol{\rho}(t) = \hat{A}(t+T, t)\boldsymbol{\rho}(t), \quad \mathrm{Tr}\{\hat{\rho}(t)\} = \sum_j \rho_{jj}(t) = 1. \tag{20}
$$

This equation can be used to construct a universal computational algorithm (without applying the Floquet or Fourier formalism). Indeed, we consider some chosen time interval $[t_0, t_0+T]$, which can be divided into N small subintervals between points $t_n = t_0 + n\tau$ ($n = 0, 1, \ldots, N$), where $\tau = T/N$ is the duration of the subintervals. We approximate the dependence $\hat{L}(t)$ by the step function, where the matrix $\hat{L}(t)$ has a constant value $\hat{L}(t_{n-1})$ inside the subinterval $(t_{n-1}, t_n]$. In this case, the vector $\boldsymbol{\rho}(t_0)$ at the initial point t_0 is determined by equation (20) with the evolution operator $\hat{A}(t_0+T, t_0)$ in the form of a sequential product of matrix exponentials:

$$
\hat{A}(t_0+T, t_0) \approx \prod_{n=1}^{n=N}\mathrm{e}^{\tau\hat{L}(t_{n-1})} = \mathrm{e}^{\tau\hat{L}(t_{n-1})}\times\cdots\times\mathrm{e}^{\tau\hat{L}(t_1)}\times\mathrm{e}^{\tau\hat{L}(t_0)}. \tag{21}
$$

The vectors $\boldsymbol{\rho}(t_n)$ at other points of the interval $[t_0, t_0+T]$ are determined through the recurrence relation

$$
\boldsymbol{\rho}(t_n) = \mathrm{e}^{\tau\hat{L}(t_{n-1})}\boldsymbol{\rho}(t_n-1). \tag{22}
$$

As a spectroscopic signal, we consider the light field absorption, which in the approximation of an optically thin medium is determined by the total population of the excited level:

$$
S = \mathrm{Tr}\{\hat{\rho}^{\mathrm{ee}}\}. \tag{23}
$$

Then, signal (23) should be averaged over the period T:

$$
\langle S\rangle_T = \frac{1}{T}\int_0^T S(t)\,\mathrm{d}t. \tag{24}
$$

In this work, we study the spectroscopic signal $\langle S\rangle_T$ for a closed dipole transition as a function of the values of F_{g} and F_{e}. The absorption signal (24) is a function of the frequency difference of the waves Δ, and the subnatural resonance structure appears near $\Delta = 0$. Numerical calculations were performed for various elliptical polarisations of the waves (including linear and circular) and under the condition $R_1^2 + R_2^2 \gg \gamma_{\mathrm{sp}}\Gamma_0$, i.e. for a sufficiently high field intensity, when the perturbation theory [22] is no longer valid. We considered a model with spontaneous transfer of anisotropy (14) (Fig. 3) and without it (15) (Fig. 4).

Figures 3a and 3b show the dependences of the signal $\langle S\rangle_T$ on Δ for particular cases of transitions $F_{\mathrm{g}} = F \to F_{\mathrm{e}} = F+1$.

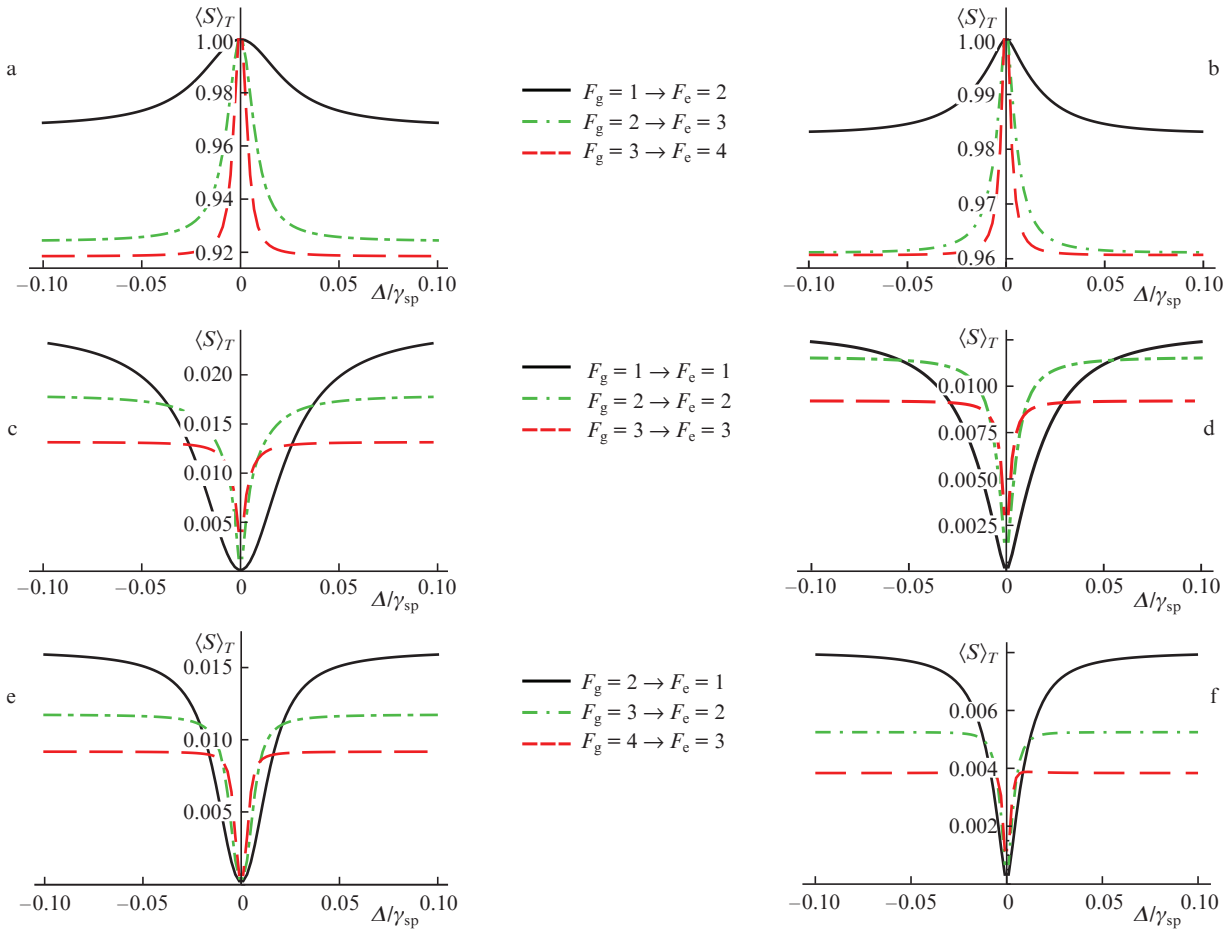

Figure 3. Dependences of the absorption signal $\langle S\rangle_T$ on the wave frequency difference $\Delta \equiv \omega_2 - \omega_1$ (in units of γ_{sp}) in a model with spontaneous transfer of anisotropy from the excited state to the ground state for special cases of the transitions (a, b) $F_g = F \to F_e = F + 1$ (the curves are normalised to the value of the signal near $\Delta = 0$), (c, d) $F_g = F \to F_e = F$, and (e, f) $F_g = F \to F_e = F - 1$ for (a, c, e) $\varepsilon_1 = \pi/4$, $\varepsilon_2 = -\pi/4$ and (b, d, f) $\varepsilon_1 = \pi/8$, $\varepsilon_2 = -\pi/8$. Other model parameters include $R_1 = R_2 = 2\gamma_{sp}$, $\gamma_{opt} = 50\gamma_{sp}$, $\Gamma_0 = 10^{-4}\gamma_{sp}$, $\phi = 0$.

One can see that these resonances are directed upwards, i.e., the EIA effect takes place; therefore, the transitions $F_g = F \to F_e = F + 1$ are 'bright' ones. Figures 3c, 3d, and 3e, 3f show similar dependences for particular cases of transitions $F_g = F \to F_e = F$ and $F_g = F \to F_e = F - 1$, respectively. In this case, the resonances are directed downward, and the EIT effect is manifested, and therefore these transitions are 'dark' ones. Figure 4 shows the subnatural resonances that are formed at the transitions $F_g = 1 \to F_e = 2$ (Fig. 4a), $F_g = 2 \to F_e = 3$ (Fig. 4b) and $F_g = 3 \to F_e = 4$ (Fig. 4c) without taking into account the spontaneous transfer of anisotropy. However, here the resonances are already directed downward (in contrast to the results in Figs 3a and 3b), i.e., EIT resonances are formed. Thus, the ellipticity of the waves does not affect the sign of the subnatural resonance in the two-frequency configuration at high wave intensities, and this sign is determined only by the angular momenta F_g and F_e. In this case, the formation of the EIA resonance is associated with the spontaneous transfer of anisotropy from the excited state to the ground state.

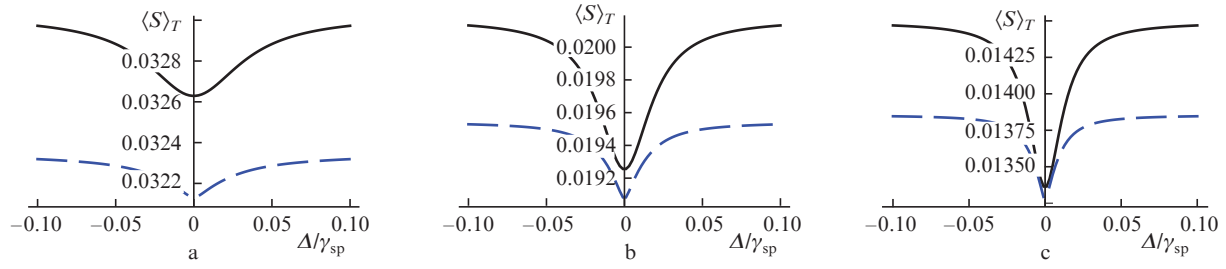

Figure 4. Dependences of the absorption signal $\langle S\rangle_T$ on the wave frequency difference $\Delta \equiv \omega_2 - \omega_1$ (in units of γ_{sp}) in a model without spontaneous transfer of anisotropy from the excited state to the ground state for the transitions (a) $F_g = 1 \to F_e = 2$, (b) $F_g = 2 \to F_e = 3$, and (c) $F_g = 3 \to F_e = 4$ for $\varepsilon_1 = \pi/4$, $\varepsilon_2 = -\pi/4$ (solid curves) and $\varepsilon_1 = \pi/8$, $\varepsilon_2 = -\pi/8$ (dashed curves). Other model parameters include $R_1 = R_2 = 2\gamma_{sp}$, $\gamma_{opt} = 50\gamma_{sp}$, $\Gamma_0 = 10^{-4}\gamma_{sp}$, $\phi = 0$.

3. Conclusions

We have studied theoretically the sign of subnatural resonance as function of the values of the total angular momenta of the ground (F_g) and excited (F_e) states of the closed dipole transition outside the framework of the perturbation theory. As a model, we have considered the interaction between the two-frequency field of two collinear waves with arbitrary elliptical polarisation and a two-level atomic system degenerated with respect to the projections of the total angular momentum. It has been shown that the type of resonance (EIT or EIA) is independent of the parameters of ellipticity and wave intensity. In this case, the EIA resonance is formed due to the spontaneous transfer of anisotropy from the excited state of the atom to the ground state. As a result, we have generalized the previous classification of cyclic dipole transitions in the direction of subnatural resonance for a strong light field. 'Bright' transitions are the transitions $F_g = F \to F_e = F + 1$, at which it is possible to observe the EIA resonance. In turn, the 'dark' transitions include the transitions $F_g = F \to F_e = F$ and $F_g = F \to F_e = F - 1$ for which the EIT resonance is observed.

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