ABSORPTION SPECTRUM OF A TWO-LEVEL ATOM IN A STRONG NONRESONANCE FIELD

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Within the framework of the perturbation theory developed on the basis of the Floquet–Lyapunov theorem the evolution operator of a two-level quantum system in a nonresonance monochromatic field is determined without the use of the rotating wave approximation. It is shown that matrix elements of the evolution operator which are linear combinations of the quasi-energy state wave functions make it possible to follow up the connections with the initial level populations of the system unperturbed by the field, in contrast to quasi-energy wave functions. Using the evolution operator obtained the problem of a stationary absorption spectrum of a weak monochromatic field resonance to a two-level atom has been solved. It is shown that, as a result of the action of a nonresonance perturbing field, the probe field absorption line strength decreases, and when recording the time-averaged signal at low frequencies of perturbing field the line profile contains narrow dips and peaks observable at microwave transitions.

1. INTRODUCTION

A periodic electromagnetic field, acting upon atoms and molecules changes the energy of their states, due to the dynamic Stark effect, and the spectra of resonance absorption and fluorescence vary qualitatively. In sufficiently intense external fields the distortions of absorption spectrum of a probe field used to probe an atom transition can be sensible even for nonresonance external fields. Such a situation may occur, for example, in microwave spectroscopy and in the method of laser Stark modulation spectroscopy,[1,2] where low-frequency radiowave fields are used.

Correct method for describing similar situation is the use of the formalizm of quasi-energy states (QES)[3–5] enabling one to calculate the spectrum of natural atom energies in the presence of a periodic field without the use of rotating wave approximation (RWA) inapplicable for low-frequency fields. The primary goal of this paper is to determine the influence of intense nonresonance (including low-frequency) electromagnetic field on the spectra of linear resonance absorption of monochromatic probe field of a dual-level quantum system based on a rigorous theory of perturbation for a nonresonance external field.[6]

2. WAVE FUNCTION OF A TWO-LEVEL SYSTEM IN A NONRESONANCE PERIODIC FIELD

Let us expand the wave function of a two-level system in a nonresonance periodic field over the eigenstate vectors

$$\Psi(t) = a_m(t)\,|m\rangle + a_n(t)\,|n\rangle\ . \tag{2.1}$$

By substituting (2.1) to the Schrödinger equation

$$i\,\dot\Psi(t) = (\hat H_0 - \hat d\,E\cos\omega t)\,\Psi(t)\ ;\quad \hbar \equiv 1\ , \tag{2.2}$$

where $\hat H_0$ is the Hamiltonian of an unperturbed system; $\hat d$ is the operator of dipole moment; E and ω are the electric field intensity and the light wave frequency, we derive an equation for the probability amplitudes a_m, a_n

$$\dot a_m = -\,i\,E_m\,a_m + i\,G\cos\omega t\ a_n\ ,$$

$$\dot a_n = -\,i\,E_n\,a_n + i\,G\cos\omega t\ a_m\ , \tag{2.3}$$

$$G = d_{mn}\,E\ ,\quad d_{mn} = \langle m|\,\hat d\,|\,n\rangle,$$

where E_m, E_n are the level energies.

Equation for the evolution operator (matriciant) of the system (2.3) is

$$\frac{\mathrm d}{\mathrm dt}\,\hat X(t) = \hat K_0\,\hat X(t) + G\,\hat D(t)\,\hat X(t)\ ; \tag{2.4}$$

$$\hat K_0 \equiv -\,i\,\hat H_0 = -\,i\begin{pmatrix} E_m & 0\\ 0 & E_n\end{pmatrix},\quad \hat D(t) = i\cos\omega t\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},$$

$$\hat{X}(0) = \hat{I}_2 \equiv \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} .$$

According to the Floquet–Lyapunov theorem[6]

$$\hat{X}(t, D) = \hat{F}(t, G)\, e^{\hat{K}(G)t} , \quad \hat{F}(t + 2\pi/\omega) = \hat{F}(t) . \qquad (2.5)$$

Substitution of (2.5) to (2.4) leads to the equation for $\hat{F}$ and $\hat{K}_0$

$$\frac{d}{dt}\hat{F}(t,G) = \hat{K}_0 \hat{F}(t,G) - \hat{F}(t,G)\hat{K}(G) + G\hat{D}(t)\hat{F}(t,G). \qquad (2.6)$$

The perturbation theory relative small parameter G consists in the expansion

$$\hat{F}(t, G) = \hat{I}_2 + G\,\hat{F}_1(t) + G^2\,\hat{F}_2(t) + \dots ,$$

$$\hat{K}(G) = \hat{K}_0 + G\,\hat{K}_1 + G^2\,\hat{K}_2 + \dots , \qquad (2.7)$$

followed by seeking of the matrices $\hat{F}_i$, $\hat{K}_i$ by substituting (2.7) in (2.6) that gives rise to a system of matrix equations

$$\frac{d\hat{F}_1}{dt} = \hat{K}_0\,\hat{F}_1 - \hat{F}_1\,\hat{K}_0 - \hat{K}_1 + \hat{D} ,$$

$$\frac{d\hat{F}_2}{dt} = \hat{K}_0\,\hat{F}_2 - \hat{F}_2\,\hat{K}_0 - \hat{F}_1\,\hat{K}_1 - \hat{K}_2 + \hat{D}\,\hat{F}_1 , \qquad (2.8)$$

. . . .

By expanding the time periodic matrices $\hat{F}_i(t)$, $\hat{D}(t)$ in a Fourier series

$$\hat{F}_i(t) = \sum_{m=-\infty}^{\infty} \hat{F}_i^{(m)} e^{im\omega t} ,$$

$$\hat{D}(t) = \hat{D}^{(1)} e^{i\omega t} + \hat{D}^{(-1)} e^{-i\omega t} , \qquad (2.9)$$

$$\hat{D}^{(1)} = \hat{D}^{(-1)} = \frac{i}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} ,$$

as well as the Eq. (2.8), we find all matrices $\hat{F}_1^{(m)}$ from the first of transformed equations (2.8) for nonzero harmonics ($m \neq 0$). Further the matrix $F_1^{(0)}$ is determined from initial conditions (2.4). Substitution of the expansion (2.9) to the above conditions results in the expression

$$\hat{F}_i^{(0)} = -\sum_{\substack{m=-i \\ m \neq 0}}^{i} \hat{F}_i^{(m)} . \qquad (2.10)$$

Then from the first transformed Eq. (2.8) for $m = 0$ we find the matrix $\hat{K}_1$. Having determined $\hat{F}_1^{(m)}$ for all $m = -1, 0, 1$ we come to the second equation (2.8), and the procedure is repeated for the second and consequent orders of the perturbation theory.

As a result, correct to the second order of the perturbation theory of the field we obtain

$$\hat{K}(G) = -i\begin{pmatrix} E_m - G^2 k_2 & -G k_1 \\ -G k_1 & E_n + G^2 k_2 \end{pmatrix} , \qquad (2.11)$$

$$k_1 = \frac{\omega_0^2}{\omega_0^2 - \omega^2} , \quad k_2 = \frac{\omega_0 (\omega_0^2 + \omega^2)}{2(\omega_0^2 - \omega^2)^2} , \quad \omega_0 \equiv E_m - E_n .$$

$$\hat{F}(t, G) = \begin{pmatrix} 1 + \frac{1}{2}G^2 f_2 & G f_1 \\ -G f_1^* & 1 + \frac{1}{2}G^2 f_2^* \end{pmatrix} , \qquad (2.12)$$

$$f_j = \sum_{m=-j}^{j} f_j^{(m)} e^{im\omega t} , \; j = 1, 2 ,$$

$$f_1^{(\pm 1)} = \frac{1}{2(\omega_0 \pm \omega)} , \quad f_1^{(0)} = -\frac{\omega_0}{\omega_0^2 - \omega^2} ,$$

$$f_1^{(\pm 2)} = \mp \frac{1}{4\omega(\omega_0 \mp \omega)} ,$$

$$f_2^{(\pm 1)} = \frac{\omega_0}{(\omega_0^2 - \omega^2)(\omega_0 \pm \omega)} , \quad f_2^{(0)} = -\frac{3\omega_0^2 + \omega^2}{2(\omega_0^2 - \omega^2)^2} .$$

Denominators in Eqs. (2.11) and (2.12) contain the resonant factors $(\omega_0 - \omega)$. The necessary condition of the small values of terms of the expansion (2.7) imposing a limitation on the proximity of the frequency to the first order resonance $\omega_0 \approx \omega$, is

$$G \ll 2|\omega_0 - \omega| . \qquad (2.13)$$

Inversion of the matrix exponent $\exp[\hat{K}(G)t]$ in Eq. (2.5) is performed with the use of Sylvester theorem[6]

$$\exp[\hat{K}(G)t] = \begin{pmatrix} (1-c)e_m + c\,e_n & -d(e_m - e_n) \\ -d(e_m - e_n) & (1-c)e_n + c\,e_m \end{pmatrix} , \qquad (2.14)$$

$$c = G^2 \frac{\omega_0^2}{(\omega_0^2 - \omega^2)^2} , \quad d = G\frac{\omega_0}{\omega_0^2 - \omega^2} ,$$

$$e_{m,n} = \exp[-i(E_{m,n} \pm \Delta)t] , \quad \Delta = G^2 \frac{\omega_0}{2(\omega_0^2 - \omega^2)} .$$

Having multiplied, in Eq. (2.5), the matrices (2.12) and (2.14), we obtain the final solution of (2.4) correct to the second order of the perturbation theory of the field G

$$X_{11} = \left(1 + \frac{1}{4}G^2 \varphi_{e2}\right) e_m + \frac{1}{2}G^2 \varphi_{e1}\varphi_{o1}\, e_n ,$$

$$X_{12} = - G\,\varphi_{e1}\, e_m + \frac{1}{2} G\,\varphi_{o1}\, e_n \,,$$

$$X_{21} = - \frac{1}{2} G\,\varphi_{o1}^*\, e_m + G\,\varphi_{e1}\, e_n \,, \qquad (2.15)$$

$$X_{22} = \frac{1}{2} G^2\,\varphi_{e1}\,\varphi_{o1}^*\, e_m + \left(1 + \frac{1}{4} G^2\,\varphi_{e2}^*\right) e_n \,,$$

$$\varphi_{e1} = \frac{\omega_0}{\omega_0^2 - \omega^2}\,, \quad \varphi_{o1} = \frac{e^{-i\,\omega t}}{\omega_0 - \omega} + \frac{e^{i\,\omega t}}{\omega_0 + \omega}\,,$$

$$\varphi_{e2} = \frac{e^{-2i\omega t}}{2\omega(\omega_0 + \omega)} - \frac{3\,\omega_0^2 + \omega^2}{(\omega_0^2 - \omega^2)^2} - \frac{e^{2i\omega t}}{2\omega(\omega_0 - \omega)}\,.$$

The matrix $\hat{X}$ (2.15) is a unitary matrix correct to G^2 and enables the transition from eigenstates $|m\rangle$, $|n\rangle$ of the unperturbed system to the interaction representation

$$\begin{pmatrix} |u\rangle \\ |l\rangle \end{pmatrix} = \begin{pmatrix} X_{11} & X_{12} \\ X_{21} & X_{22} \end{pmatrix} \begin{pmatrix} |m\rangle \\ |n\rangle \end{pmatrix}, \qquad (2.16)$$

where the states $|u\rangle$ and $|l\rangle$ correspond to the population of the states m and n at $t = 0$.

Quasi-energies of the system are the factors $E_m + \Delta$ and $E_n - \Delta$ in the exponents e_m and e_n (2.14), (2.15). From Eqs. (2.16) and (2.15) it follows that the mathematically rigorous solutions for the states $|u\rangle$ and $|l\rangle$, having a directly seen relation to the initial populations of the system unperturbed by the field, are linear combinations of QES.

3. SPECTRUM OF A PROBE FIELD ABSORPTION BY A TWO-LEVEL ATOM DISTURBED BY AN EXTERNAL NONRESONANCE FIELD

To calculate the probe field absorption spectrum we use the equation for the medium density matrix in the model of relaxation constants. For simplicity of the calculations all the constants are assumed to be equal and the rotating wave approximation is used for the probe field

$$\dot{\hat{\rho}} + \gamma\,\hat{\rho} = \left[\hat{K}_0 + V\hat{D}_\mu(t) + G\hat{D}(t),\, \hat{\rho}\right] + \gamma\,\hat{Q}\,,$$

$$\hat{D}_\mu = i \begin{pmatrix} 0 & e_m \\ e_\mu^* & 0 \end{pmatrix}, \quad \hat{Q} = \begin{pmatrix} \rho_m^0 & 0 \\ 0 & \rho_n^0 \end{pmatrix}, \qquad (3.1)$$

$$e_\mu = \exp(-\,i\,\omega_\mu\, t)\,, \quad V = d_{mn}\, E_\mu / 2\,,$$

where E_μ and ω_μ are the amplitude and the frequency of the probe field; ρ_m^0 and ρ_n^0 are the equilibrium populations of the levels m and n in the absence of fields.

Transition to the interaction representation (2.16) removes the explicit form of the disturbing field G from Eqs. (3.1), which after specific definition with the use of Eq. (2.15) takes the form

$$\dot{\rho} + \left(\gamma - \frac{4\,i\,V\,G\,\omega_0}{\omega_0^2 - \omega^2}\cos\delta t\right)\rho =$$

$$= i\,V\,n\,e^{-i\delta t}\left[1 - G^2 f(t)\right] + \gamma n_0 \frac{G\,\omega_0}{\omega_0^2 - \omega^2}\,, \qquad (3.2)$$

$$\dot{n} + \gamma\,n = 4\,V\,\mathrm{Re}\,i\,e^{-i\delta t}\,\rho\left[1 - G^2 f^*(t)\right] +$$

$$+ \gamma\,n_0\left[1 - G^2\varphi(t)\right],$$

$$f(t) = \frac{e^{-2i\omega t}}{4\omega(\omega - \omega_0)} + \frac{3\,\omega_0^2 + \omega^2}{2(\omega_0^2 - \omega^2)^2} - \frac{e^{2i\omega t}}{4\omega(\omega_0 + \omega)} +$$

$$+ \frac{\omega_0^2}{(\omega_0^2 - \omega^2)^2}\, e^{2i\delta t},$$

$$\varphi(t) = \frac{1}{2(\omega_0^2 - \omega^2)}\left(e^{-2i\omega t} + 2\,\frac{3\,\omega_0^2 + \omega^2}{\omega_0^2 - \omega^2} + e^{2i\omega t}\right),$$

where $\rho = \rho_{ul}$ is the off-diagonal element of the density matrix in ul-representation (2.16); $n = \rho_{ll} - \rho_{uu}$ is the difference between populations disturbed by the field G of the states $|u\rangle$ and $|l\rangle$; $\delta = \omega_\mu - \omega_0 - 2\Delta$ is detuning of the probe field frequency from the transition resonance frequency disturbed by the field G system; the value Δ is determined in Eqs. (2.14), $n_0 = \rho_n^0 - \rho_m^0$ is the equilibrium difference in population of the levels (in the absence of the fields).

Peculiarities of Eqs. (3.2) are the transfer of the result of perturbation by the field G to the time depending coefficients containing the oscillating exponents with the periods determined by the frequency ω and the detuning δ as well as the appearance of a continuous in time "pumping" of the polarization, proportional to the first power of G (the latter term of the first equation (3.2)), and periodic in time Bloch-Siegert shear[7] (the second term in brackets of the left-hand side of the first equation (3.2)) due to the interference of the fields V and G. Note that because of simplifying assumptions concerning the equality of all the system relaxation constants after the transfer to ul-representation the relaxation part remains invariant. It can be shown that inequality of relaxation constants results in their and the field G time dependence in ul-representation as well as in the appearance of cross-relaxation terms, proportional to differences in these constants, which additionally couple the equations for level populations and atom polarization.

The action of the probe field, to which the absorption factor of test field is proportional, in the used representation of interaction is of the form

$$P = 2\,V\,\mathrm{Re}\,ie_\mu^*\left[X_{11}\,X_{22}^*\,\rho + X_{21}\,X_{12}^*\,\rho^* - X_{11}\,X_{12}^*\,n\right]. \quad (3.4)$$

Stationary solution (3.2) and (3.4) in weak fields V is

$$P = -\frac{2n_0\, V^2 \gamma}{\gamma^2 + \delta^2}\left[1 - 2\, G^2 \frac{2\omega_0^2 + \omega^2}{(\omega_0^2 - \omega^2)^2}\right] -$$

$$- n_0\, VG \left[\frac{2\omega_0 \sin \delta t}{\omega_0^2 - \omega^2} - \frac{\sin(\delta + \omega)t}{\omega_0 - \omega} - \frac{\sin(\delta - \omega)t}{\omega_0 + \omega}\right] -$$

$$- 2\, n_0\, V^2 G^2 \,\mathrm{Re} \sum_{k=-2}^{2} \left(A_k + \frac{B_k}{\gamma + i\,\delta}\, e^{2i\delta t}\right) e^{ik\omega t}, \quad (3.5)$$

$$A_0 = 0\,, \quad A_{\pm 1} = \frac{1}{\gamma - i\delta}\, \frac{\omega_0}{(\omega_0^2 - \omega^2)(\omega_0 \mp \omega)}\,,$$

$$A_{\pm 2} = \mp \frac{1}{\gamma - i\delta}\, \frac{1}{4\omega(\omega_0 \mp \omega)} \pm \frac{1}{\gamma - i(\delta \mp 2\omega)}\, \frac{1}{4\omega(\omega_0 \pm \omega)} -$$

$$- \frac{\gamma}{(\gamma \pm 2i\omega)\,[\gamma - i(\delta \mp 2\omega)]}\, \frac{1}{2(\omega_0^2 - \omega^2)}\,,$$

$$B_0 = \frac{3\,\omega_0^2 - \omega^2}{2(\omega_0^2 - \omega^2)^2}\,, \quad B_{\pm 1} = -(\gamma - i\delta)\, A_{\pm 1}\,,$$

$$B_{\pm 2} = \frac{1}{4(\omega_0 \mp \omega)^2}\,.$$

Note that the constant component of the absorption coefficient (the first line (3.5)) tends to uniformly decrease with the probe field frequency, that can be interpreted as an additional broadening of the transition levels due to their spectral “blurring” owing to the dynamic Stark effect appearing in the second order perturbation theory in G. The rate of the decrease is determined by the external field intensity and the values of ω_0, $|\omega_0 - \omega|$. The estimates show that such a decrease may be quite noticeable, though still being within the limits of the perturbation theory, for example, for transitions in the microwave region as well as for higher-frequency transitions (including optical ones) at $|\omega_0 - \omega| \ll \omega_0$.

The interference component of the field energy, proportional to the product of amplitudes of disturbing and probe fields (second line in (3.5)) is due to the joint action of constant pumping of polarization and Bloch-Siegert dynamic shift (see the discussion of (3.2)) and oscillates at the frequencies $|\delta|$ and $|\delta \pm \omega|$. The square in disturbing field amplitude addition to the absorption coefficient contains the frequencies ω, 2ω, $|2\delta \pm \omega|$ and $|2\delta \pm 2\omega|$.

Experimental recording of the absorption line contour of a probe field is coupled with the signal averaging over time or with separating out of any of the characteristic frequencies of oscillations $P(t)$. In the case of simple averaging over the interval $[-T/2, T/2]$ for mean value of the field energy we have from Eq. (3.5)

$$\bar{P} = -\frac{2n_0\, V^2 \gamma}{\gamma^2 + \delta^2}\left\{1 - 2G^2 \frac{2\,\omega_0^2 + \omega^2}{(\omega_0^2 - \omega^2)^2} + \frac{2G^2}{T}\left[\frac{3\,\omega_0^2 - \omega^2}{4(\omega_0^2 - \omega^2)^2} \times\right.\right.$$

$$\times \frac{\sin T\delta}{\delta} - \frac{\omega_0 \sin T(\delta + \omega/2)}{2(\omega_0^2 - \omega^2)(\omega_0 - \omega)(\delta + \omega/2)} -$$

$$- \frac{\omega_0 \sin T(\delta - \omega/2)}{2(\omega_0^2 - \omega^2)\,(\omega_0 + \omega)\,(\delta - \omega/2)} +$$

$$\left.\left. + \frac{\sin T(\delta + \omega)}{8(\omega_0 - \omega)^2(\delta + \omega)} + \frac{\sin T(\delta - \omega)}{8(\omega_0 + \omega)^2(\delta - \omega)}\right]\right\}. \quad (3.6)$$

Because of the averaging the interference component P does not contribute to P. It is evident from Eq. (3.6) that at a relatively large values of T the line contour averaged over time contains narrow resonances whose maximum relative amplitude is determined by the value of the second-order terms of perturbation theory with respect to the external field. Resonance amplitudes at the frequency detuning $\delta = 0$ and $\delta = \pm\,\omega$ are positive and at $\delta = \pm\,\omega/2$ they are negative. In the case of $\omega << \omega_0$ relative amplitude of the constant addition in P is $I_c = -4G^2/\omega_0^2$. Taking its modulus to be unity, we have for resonance amplitudes: $I_0 = 3/8$; $I_{\pm\omega/2} = -1/4$ and $I_{\pm\omega} = 1/16$. For the quasi-resonance $\omega_0 - \omega \equiv \Delta_0 << \omega_0$ we obtain $I_c = -3/2(G^2/\Delta_0^2) \Rightarrow -1$; $I_0 = 1/6$; $I_{-\omega/2} = -1.3$, $I_{\omega/2} = -\Delta_0/(6\omega_0)$; $I_{-\omega} = 1/6$ and $I_\omega = \Delta_0^2/(24\omega_0^2)$. Thus, in this case the resonances at positive frequency detuning $\delta = \omega/2$ and $\delta = \omega$ have been essentially suppressed.

At small frequencies $\omega \le \gamma$ the resonances are within the limits of the line contour $P(\delta)$. If $\omega \ge (4 - 5)\,\gamma$, then resonances manifest themselves as the line satellites whose amplitudes are discriminated additionally by the shape factor $\gamma/(\gamma^2 + \delta^2)$. The latter fact hinders experimental recording of satellites in case of the quasi-resonance $\omega_0 - \omega \ll \omega_0$ at high frequencies ω_0. Thus, the low-frequency transitions are optimal for observing the resonances.

Among other important for the experiment peculiarities of the resonance absorption of a weak field in the presence of a strong nonresonance radiation it is also worth noting the above discussed effective decrease of the line intensity, by the factor $1 - I_c$, and its shear to the lower frequencies -2Δ.

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