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CROSSING OF QUASI-ENERGY LEVELS

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Study of the crossing of quasi-energy levels in a four-level system

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***Abstract*. It was shown previously that in taking into account only dipole transitions, the crossing of quasi-energy levels is possible in the system if any of the transitions forms a closed loop [1]. It followed herefrom that for the analysis of the crossing conditions, it is necessary to consider a system which has at least four levels. In this paper we show that we can uniquely specify which quasi-energy levels cross at the given values of the parameters of the atomic system and radiation field, without solving an algebraic quartic equation. It was found that the most suitable system for the implementation of the crossing is the group of energy levels $^5S_{1/2}$, $^5P_{1/2}$, $^5P_{3/2}$ and $^5D_{3/2}$ of a rubidium atom. The performed calculations of the laser field intensity and frequency values at which crossing takes place in this system show that they are easily attainable. It turned out that in this system there occur crossing of quasi-energy levels corresponding to the excited atomic levels.**

Keywords: *crossing of quasi-energy levels, four-level system.*

1. Introduction

It is known that the characteristics of transitions between states of a quantum system depend on the radiation parameters: pulse duration, carrier frequency detuning from the transition frequency, the width of the emission spectrum, etc. Interaction of laser radiation with a two-level system has been studied most thoroughly. If the radiation frequency is close to the frequencies of transitions between several levels, the two-level approximation is generally invalid. When the radiation parameters or parameters of a quantum system change adiabatically, in some cases we can restrict ourselves to finding the quasi-energy states of the system. However, if the adiabaticity condition is not fulfilled throughout this interaction process, it is also necessary to take into account non-adiabatic transitions [2, 3], which at the same time can be used for the selective population of levels, or population transfer between levels (see, for example, [4]).

Melikyan and Saakyan [1] determined the schemes of those transitions in a quantum system that permit the crossing of quasi-energy levels under the influence of the external field. The aim of this work is to find the algorithm for identification of crossing of quasi-energy levels in a four-level system with dipole transitions under the action of the laser field.

2. Determining the parameters at which crossing of the levels occurs

The concept of quasi-energy states of a quantum system in an external periodic field was first introduced in [5, 6]. These states appear under the condition that the spectral width of radiation (as well as the width of atomic energy levels) is much smaller than the resonance detuning. The amplitude of the wave field strength at the same time is considered an adiabatic function of time; therefore, atomic states without the field transform smoothly in quasi-energy states.

According to the Neumann – Wigner theorem [7, 8], the coincidence of the eigenvalues of a symmetric matrix can be achieved by changing, in general, at least two parameters. We choose the frequency and amplitude of the wave field strength as these parameters.

Following paper [4], we will show that the level crossing is possible in the case when the transitions form a closed loop under the influence of the external perturbations. The simplest system with the dipole transitions satisfying this condition is a four-level system (see Fig. 1).

Following Born and Fock [9] and assuming that the solutions of the stationary Schrödinger equation $H(s)\Psi_n(s) = \hbar\omega(s) \times \Psi_n(s)$ (here $s = t/T$, T is the characteristic time for changes in the Hamiltonian) are known at every instant of time, which is regarded as a parameter, while the solution of the nonstationary Schrödinger equation with the adiabatic Hamiltonian $H(t)$ is sought as a superposition of 'instant' eigenfunctions.

Therefore, we consider the N-level system in the field of a resonant linearly polarised wave with an adiabatically changing amplitude. The field will be described by the classical electric-field vector

$$\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}(s)\exp(\mathrm{i}\boldsymbol{kr} - \omega t) + \mathrm{c.\,c.} \quad (1)$$

The wave function of the system is found from the Schrödinger equation

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$$\mathrm{i}\hbar\frac{\partial\Phi}{\partial t}=(H_0+H_{\mathrm{int}})\Phi, \tag{2}$$

where H_0 is a Hamiltonian of an unperturbed system, and the interaction H_{int} in the dipole approximation has the from:

$$H_{\mathrm{int}}=-\frac{1}{2}E_{x,y}(s)(\boldsymbol{e}_{x,y}\hat{\boldsymbol{d}})\exp(-\mathrm{i}\omega t)+\mathrm{c.\,c.} \tag{3}$$

Taking as a reference point the energy of the lowest levels, the resonance condition is formulated as follows: for each mth level there is an integer k_m, such that the condition $\varepsilon_m=\omega_m-k_m\omega\ll\omega$, where ε_m is the detuning of the mth level, and ω_m is the frequency of the corresponding transitions. The wave function

$$\Phi=\sum_m c_m(t)\varphi_m\exp(-\mathrm{i}k_m\omega t), \tag{4}$$

where m are the eigenfunctions of the Hamiltonian H_0.

Substituting (4) in Schrödinger equation (2) and taking into account the orthonormal function φ_m for the coefficients $c_m(t)$, we obtain a system of differential equations:

$$\mathrm{i}\hbar\frac{\partial c_m}{\partial t}=\hbar\varepsilon_m c_m+\sum_n c_n\{V_{mn}\exp[\mathrm{i}\omega(k_m-k_n-1)t]$$
$$+V^*_{mn}\exp[\mathrm{i}\omega(k_m-k_n+1)t]\}. \tag{5}$$

Using the resonance approximation (rapidly oscillating terms are neglected) with the linear polarisation of the wave field (V_{mn} can be considered real), for the amplitudes c_m we finally obtain the system of equations:

$$\mathrm{i}\hbar\frac{\partial c_m}{\partial t}=\sum_n c_n(V_{mn}+\hbar\varepsilon_m\delta_{mn}). \tag{6}$$

Here δ_{mn} is the Kronecker symbol.

Introducing the one-column matrix $\hat{c}$ with elements $c_1,c_2,...,c_N$, and the matrix $\hat{W}$ with elements $W_{mn}=V_{mn}(s)+\varepsilon_m\delta_{mn}$, equation (6) can be written in the matrix form:

$$\mathrm{i}\hbar\frac{\partial\hat{c}}{\partial t}=\hat{W}(s)\hat{c}. \tag{7}$$

One can see that equation (7) formally coincides with the Schrödinger equation with an adiabatically changing Hamiltonian. As we have already mentioned, in solving the Schrödinger equation with this Hamiltonian, the value of s is considered as a parameter, and 'instant' eigenfunctions and eigenvalues are found.

This approximation is also called adiabatic-following approximation. By substituting $\hat{c}(t)=\hat{b}(t)\times\exp(-\mathrm{i}\lambda t)$, we will write (7) in the stationary form:

$$\sum_n[\hbar(\lambda-\varepsilon_m)\delta_{mn}-V_{mn}]\,b_n=0. \tag{8}$$

Here λ is the quasi-energy [5, 6], and its value is determined from the nontriviality condition of the solutions of matrix equation (8) for b_n. The nontriviality condition means that the determinant $D=\|\hbar(\lambda-\varepsilon_m)\delta_{mn}-V_{mn}\|$ must be zero. This equation of degree N in λ has, generally speaking, N different real roots. The crossing of two quasi-levels means that the multiplicity of one of the roots of equation $D(\lambda)=0$ is equal to two. If there is a double root of the equation $D(\lambda)=0$, the matrix rank is reduced to two at $\lambda=\lambda_{\mathrm{cross}}$. In other words, if $\lambda=\lambda_{\mathrm{cross}}$, all the first minors of the determinant $D(\lambda)$ vanish.

Using the method, based on the analysis of the rank of the determinant $D(\lambda)$, we will show by the example of a four-level atom how to find the conditions for crossing the quasi-levels, as well as will try to answer the question which levels cross. To this end, we consider a four-level system, shown below in Fig. 1.

The determinant of the system has the form

$$D(\lambda)=\begin{vmatrix}\lambda & -V_{12} & 0 & -V_{14}\\ -V_{21} & \lambda-\varepsilon_2 & -V_{23} & 0\\ 0 & -V_{32} & \lambda-\varepsilon_3 & -V_{34}\\ -V_{41} & 0 & -V_{43} & \lambda-\varepsilon_4\end{vmatrix}. \tag{9}$$

Here we consider the matrix elements to be real quantities, referring to the linear polarisation of the external radiation field.

First, we determine the minimum number of minors whose equality to zero means that all other minors vanish. Due to the symmetry of the matrix, only 10 of the 16 minors should be considered. We will consider the minors of the diagonal elements and the elements that are above the diagonal. We will take, for example, the minors of the elements of the last column. Using the property of determinants, which consists in the fact that $\sum_n a_{nj}A_{ni}=0$ at $i\neq j$ (A_{ni} are the cofactors of the elements a_{ni}), we find that three minors in last column are linearly independent, i.e., for example, it follows from the condition $M_{44}=M_{34}=M_{24}=0$ that $M_{14}=0$. Continuing this argument, we see that two minors from the third column and one minor from the second column are linearly independent, while the minors from the first column are automatically set to zero. Thus, we conclude that for all the principal minors of a symmetric matrix to be equal to zero, it is sufficient to equate six out of 16 minors to zero.

The matrix under study, apart from symmetry, has one more property, because of which the number of linearly independent minors decreases, namely: each column and each row has an element equal to zero. Continuing the argument, we conclude that it is sufficient to equate to zero two minors from the last column, one minor from the third column and then all the other minors will be also equal to zero.

Let us equate to zero the minors $M_{13}=M_{23}=M_{24}$. Then, for unknown quantities of the intensity, quasi-energy and frequency of crossing we obtain:

$$E^2_{\mathrm{cross}}=\frac{\lambda(\lambda-\varepsilon_4)d_{32}\hbar^2}{d_{41}(d_{12}d_{34}-d_{32}d_{41})}, \tag{10}$$

$$\lambda_{\mathrm{cross}}=\frac{d_{12}d_{41}}{d_{12}d_{41}+d_{23}d_{34}}(\omega_3-2\omega_{\mathrm{cross}}), \tag{11}$$

$$\omega_{\mathrm{cross}}=\frac{d_{12}d_{41}\omega_3}{d_{12}d_{41}-d_{32}d_{34}}$$
$$-\frac{(d_{12}d_{41}+d_{32}d_{43})(d_{12}d_{32}\omega_4+d_{14}d_{34}\omega_2)}{(d_{12}d_{41}-d_{32}d_{34})(d_{12}d_{32}+d_{14}d_{34})}. \tag{12}$$

Note that expressions (10), (11), (12) depend on the signs of d_{ij}. If the system is such that the calculations by formulas

(10)–(12) yield $E^2_{\rm cross} > 0$ and $\omega_{\rm cross} > 0$ then the crossing of quasi-energies for such a system is possible. In addition, to have the initial resonance approximation unviolated, there should be fulfilled the conditions of smallness compared with the atomic fields, as well as smallness of detunings compared to $\omega_{\rm cross}$.

From the condition of equality of the minors M_{13}, M_{23}, M_{24} to zero follows the equality of all other minors of matrix (9) to zero. Converse is also true: if we equate to zero the other three minors, the minors M_{13}, M_{23}, M_{24} will also be zero; hence, conditions (10)–(12) are the only conditions under which the crossing of the quasi-levels is possible. In other words, only one crossing of quasi-levels can occur in the four-level system.

3. Identification of crossing levels

In the case of the adiabatic switching of the external field, each atomic level undergoes a transition to the quasi-energy level; therefore, we can say that there is a unique mutual correspondence between the atomic and quasi-energy levels. From the viewpoint of nonlinear spectroscopy it is desirable to know in advance exactly to what atomic levels there correspond the crossing quasi-energy levels. To answer this question, we consider the equation $D(\lambda) = 0$. The dependence of the quasi-energy branches on the field strength is given by a formula for the roots of the quartic equation. However, as seen from the equation itself, the inverse dependence, i.e., the dependence of the intensity on the quasi-energy is obtained by solving a biquadratic equation that is much more convenient and does not impose any restrictions on the consideration. The biquadratic equation has the form

$$aE^4 + bE^2 + c = 0, \tag{13}$$

where the coefficients a, b, c are the functions of λ and of the parameter of the problem

$$a = (d_{41}d_{21} - d_{23}d_{43})^2, \tag{14}$$

$$b = \lambda(\lambda - \varepsilon_2)d_{43}^2 - \lambda(\lambda - \varepsilon_4)d_{23}^2 + (\lambda - \varepsilon_4)(\lambda - \varepsilon_3)d_{12}^2 - (\lambda - \varepsilon_2)(\lambda - \varepsilon_3)d_{41}^2, \tag{15}$$

$$c = \lambda(\lambda - \varepsilon_2)(\lambda - \varepsilon_3)(\lambda - \varepsilon_4). \tag{16}$$

Equation (13) has, with respect to $E^2 = I$, two solutions:

$$I_{1,2} = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a}, \tag{17}$$

i.e., there are two branches of the intensity dependence of λ. For each value of the intensity, there are four quasi-energy values, the quasi-energies coinciding with detunings ε_i at $I = 0$, and with increasing intensity they move along the branches $I(\lambda)$. To elucidate the trajectories of the levels we can consider the asymptote I at $\lambda \to \infty$:

$$\lim_{\lambda\to\infty} I_{1,2} = \frac{\lambda^2(d_{12}^2 + d_{14}^2 + d_{23}^2 + d_{43}^2 \pm \sqrt{G})}{2a}. \tag{18}$$

Here

$$G = \lim_{\lambda\to\infty} \frac{b^2 - 4ac}{\lambda^2},$$

where it is necessary to fulfill the condition $d_{12}^2 + d_{14}^2 + d_{14}^2 + d_{23}^2 + d_{43}^2 > \sqrt{G}$. However, given the large number of parameters, it is more convenient to directly construct plots for two branches (17) at $\omega = \omega_{\rm cross}$, from which we will get the answer which of the levels cross.

4. Specific example – rubidium atom

Now we will use the obtained results to study the rubidium atom Rb^{85}_{37}. We will take the system of levels that satisfies the condition of cyclicity, namely, the levels whose scheme is shown in Fig. 1. Here all the states are degenerate, the ground state with a zero orbital angular momentum being doubly degenerate. We believe that the external radiation field is linearly polarised. Then, according to the selection rules in the dipole approximation, the field relates sublevels corresponding to the zero projection of the orbital angular momentum and the same spin projection. As a result, from the entire system of four degenerate levels we can single out two independent subsystems of interacting sublevels – four sublevels in each subsystem, as the ground state is doubly degenerate. One can see from Fig. 1 that the energies of the levels are as follows (the energy of the 5s level is taken as a reference point): $5\mathrm{p}_{1/2}$ – $\omega_2 = 12578$ cm^{-1}, $5\mathrm{p}_{3/2}$ – $\omega_4 = 12816$ cm^{-1} and $5\mathrm{d}_{3/2}$ – $\omega_3 = 25700$ cm^{-1}.

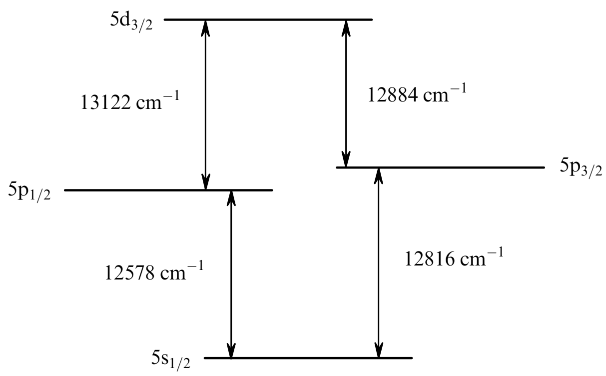

Figure 1. Scheme of transitions in a rubidium atom.

The oscillator strengths of transitions, defined as $f_{ij} = 2m\omega_{ij}|\boldsymbol{d}_{ij}|^2/\hbar e^2$ (the values are taken from [10]) are as follows:

$$f_{12} = 0.32, \quad f_{13} = 0.67, \quad f_{24} = 0.0425, \quad f_{34} = 0.0043;$$

they are used to find the matrix elements.

Substituting these data into (12), we obtain $\omega_{\rm cros} = 2.32226 \times 10^{15}$ s^{-1}. This value is then substituted into (11), whence we find $\lambda_{\rm cross} = -2.66885 \times 10^{13}$ s^{-1}. Finally, substituting $\omega_{\rm cros}$ and $\lambda_{\rm cross}$ into (10), we obtain $E_{\rm cross} = 2.39 \times 10^5$ V cm^{-1} .

As seen from Fig. 2, the equation $D(\lambda) = 0$ has four roots for all E, except $E = E_{\rm cross}$. At this value of the intensity, two of the four roots merge, so that at point $\lambda_{\rm cross}$ the dependence $D(\lambda)$ is tangent to the straight line $D(\lambda) = 0$.

Substituting $\omega_{\rm cross}$ into the formulas for calculating the detunings before switching on the field, we get $\varepsilon_2 = -6.22595 \times 10^{13}$ s^{-1}, $\varepsilon_3 = -2.68871 \times 10^{13}$ s^{-1}, $\varepsilon_4 = -1.95455 \times 10^{13}$ s^{-1}, i.e., $\varepsilon_2 < \varepsilon_3 < \varepsilon_4 < 0$. The corresponding dependences for $\omega = \omega_{\rm cross}$, $\omega < \omega_{\rm cross}$ and $\omega > \omega_{\rm cross}$

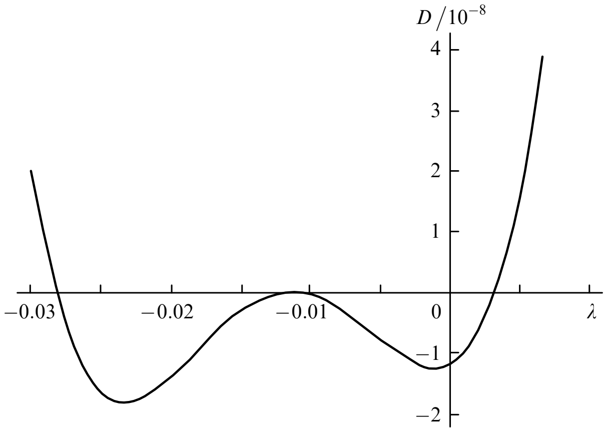

Figure 2. Dependence of D on λ (see the text).

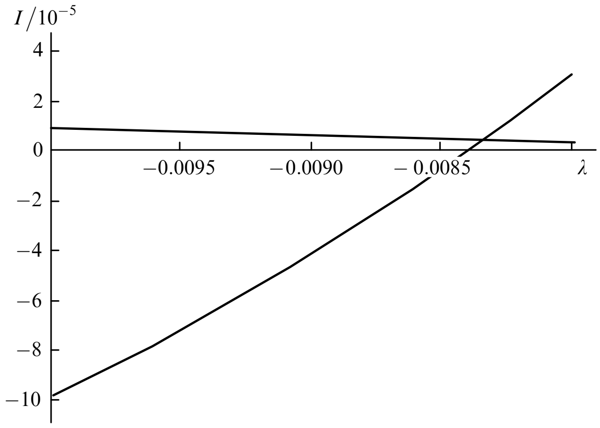

Figure 3. Dependence of I on λ at $\omega = \omega_{\text{cross}}$.

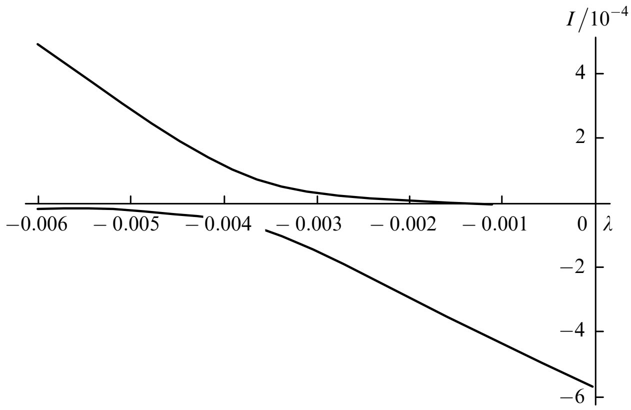

Figure 4. Dependence of I on λ at $\omega < \omega_{\text{cross}}$.

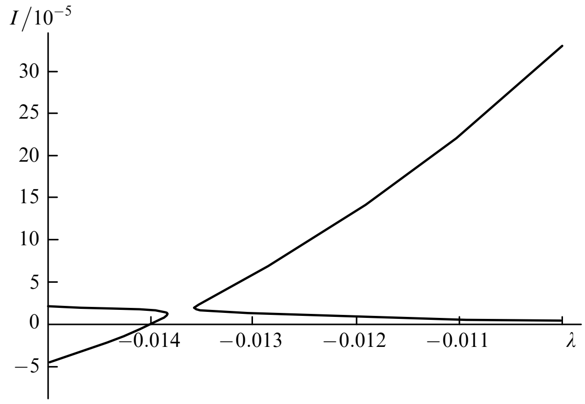

Figure 5. Dependence of I on λ at $\omega > \omega_{\text{cross}}$.

are shown in Figs 3 – 5. Comparing the positions of the quasi-energies at a zero intensity, i.e. the resonance detunings, and their positions when the field is switched on, we find that in this system there occurs crossing of the levels corresponding to the detunings ε_3 and ε_4. In this case, these levels correspond to excited atomic states, which hinders observation of nonadiabatic transitions between them.

5. Conclusions

In this paper we have found the conditions under which the crossing of quasi-energy levels of a four-level atom is possible, and have given a method for identifying the crossing levels. As a concrete example, we consider a rubidium atom for which we have found the appropriate wave frequencies and intensities; it is shown that crossing of only those quasi-energy levels is possible, which are formed from the atomic levels of the first excited states. In the case of a greater number of levels, the formulas become cumbersome, and the corresponding equations should be solved numerically. The results obtained can be used to carry out experiments on population transfer between the excited levels.

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