Generalized quasienergy states and spectrum of atoms in the field of intense multimode radiation

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(Submitted 27 June 1985)
Zh. Eksp. Teor. Fiz. **90**, 1154–1164 (April 1986)

A perturbation theory is developed for the generalized quasienergy that determines the spectrum of a quantum system in the presence of multimode radiation. Equations are obtained for the second-order additive Stark shift and for the fourth-order nonadditive shift in a bichromatic field. Nonresonant restructuring of the atomic spectrum in the field of two monochromatic waves of close frequency is investigated. It is shown that the spectrum acquires a satellite structure if the frequency difference is decreased to the level Stark shift. The intensities of the satellite lines are calculated.

§1. INTRODUCTION

The restructuring of an atomic structure in the presence of intense monochromatic radiation has been investigated in sufficient detail both theoretically and experimentally.[1,2] The theoretically most convenient and appropriate for the physical formulation of the problem is the semiclassical approach, in which the atom is described quantum-mechanically and the electromagnetic radiation classically.

In the case of monochromatic radiation, the Hamiltonian of an atom located in an electromagnetic field has a periodic dependence on the time. Shirley[3] has shown on the basis of the Floquet theorem that in this case the Schrodinger equation has solutions with a quasiperiodic time structure

$$|\psi_\varepsilon(t)\rangle=|U_\varepsilon(t)\rangle e^{-i\varepsilon t}, \tag{1}$$

$|U_\varepsilon(t+T)\rangle = |U_\varepsilon(t)\rangle$, where T is the period of the field. The solutions (1) were named quasienergy states,[4,5] and ε is called the quasienergy. The quasienergy determines the spectrum of a quantum system in an external monochromatic field.[3–6] It is most useful to separate beforehand the quasiperiodic structure of the Schrödinger-equation solutions both for the clarification of the qualitative features of the atom's behavior in the presence of monochromatic radiation, and for approximate (perturbation-theory) and numerical calculations of the spectrum and of the transition probabilities.[3,5,6,1] This is precisely why the method of quasienergy states (QES) has established itself as the most effective method of investigating the dynamics of quantum systems in an intense monochromatic field.

Shirley's method was recently generalized to include the multimode case (Ref. 7).[1)] Generalization of the Floquet theory enabled Ho and Chu[8] to study the behavior of the sublevels of a system in an intense bichromatic field outside the framework of the resonance approximation.

Most convenient for the investigation of multilevel systems in a multimode radiation field is, from our viewpoint, a generalization of the QES method on the basis of the "multitime formalism" (see, e.g., Ref. 10). Let the system Hamiltonian be

$$\hat{H}(q,t)=\hat{H}_0(q)+\sum_{i=1}^{s} V_i(q,t),$$

where $V_i(q,t+T_i) = V_i(q,t)$ and the periods T_i are generally speaking incommensurate, so that H is not a periodic function of the time, and the QES method is inapplicable. The entire reasoning that follows pertains to two fields V_1 and V_2 (generalization to more than two modes entails no difficulty). To elucidate the structure of the solutions of the Schrödinger equation

$$i\partial\Phi/\partial t=\hat{H}\Phi \tag{2}$$

it suffices to consider for the function $\tilde{\Phi}(q,t_1,t_2)$ and equation that depends on two temporal arguments:

$$i\partial\tilde{\Phi}/\partial t_1+i\partial\tilde{\Phi}/\partial t_2=\hat{H}(q,\ t_1,\ t_2)\Phi, \tag{3}$$

where

$$\hat{H}(q,\ t_1,\ t_2)=\hat{H}_0(q)+V_1(q,\ t_1)+V_2(q,\ t_2).$$

If we put $t_1 = t_2$, Eq. (2) becomes the Schrödinger equation (2) for the wave function

$$\Phi(q,\ t)=\tilde{\Phi}(q,\ t,\ t). \tag{4}$$

The operator

$$\mathscr{H}(q,\ t_1,\ t_2)=\hat{H}(q,\ t_1,\ t_2)-i\partial/\partial t_1-i\partial/\partial t_2 \tag{5}$$

commutes with the operator for the shift by T_1 and T_2 in the variables t_1 and t_2, respectively. Equation (3) has thus solutions with the following property:

$$\begin{aligned}&\tilde{\Phi}_{\varepsilon_1,\,\varepsilon_2}(q,\ t_1+T_1,\ t_2+T_2)\\&=\tilde{\Phi}_{\varepsilon_1,\,\varepsilon_2}(q,\ t_1,\ t_2)\exp[-i(\varepsilon_1T_1+\varepsilon_2T_2)\,].\end{aligned} \tag{6}$$

We separate the temporal components of $\tilde{\Phi}$:

$$\tilde{\Phi}_{\varepsilon_1,\,\varepsilon_2}(q,\ t_1,\ t_2)=U_{\varepsilon_1,\,\varepsilon_2}(q,\ t_1,\ t_2)\exp[-i(\varepsilon_1t_1+\varepsilon_2t_2)\,], \tag{7}$$

where, as follows from (6),

$$U_{\varepsilon_1,\,\varepsilon_2}(q,\ t_1+T_1,\ t_2+T_2)=U_{\varepsilon_1,\,\varepsilon_2}(q,\ t_1,\ t_2),$$

so that this function can be expanded in a double Fourier

0038-5646/86/040672-06$04.00

series:

$$U_{\varepsilon_1,\varepsilon_2}(q,t_1,t_2)=\sum_{n_1,n_2} v_{n_1,n_2}(q)\exp[i(n_1\omega_1t_1+n_2\omega_2t_2)],$$

where $\omega_1=2\pi/T_1,\omega_2=2\pi/T_2$.

It can be concluded on the basis of (4) and (7) that there exist Schrödinger-equation solutions with the following structure:

$$\Phi_{\varepsilon=\varepsilon_1+\varepsilon_2}(q,\ t)=U_\varepsilon(q,\ t)e^{-i\varepsilon t}, \tag{8}$$

where the function $U_\varepsilon(q,t)$ can be expanded in a series

$$U_\varepsilon(q,t)=\sum_{n_1,n_2} v_{n_1,n_2}(q)\exp[i(n_1\omega_1+n_2\omega_2)t]. \tag{9}$$

The state described by the wave function Φ_ε (8) will be called the generalized QES, and ε the generalized quasienergy. We show in the next section that ε is real (we do not consider decaying QES here). In full analogy with the single-mode case, the generalized quasienergy determines the spectrum of a quantum system in a multimode radiation field. Thus, in a bichromatic field the system emits spontaneously at the frequencies

$$\Omega=\varepsilon_i-\varepsilon_f+k_1\omega_1+k_2\omega_2, \tag{10}$$

where k_1 and k_2 are integers.

Note that in the case of commensurate frequencies the generalized QES are transformed into the QES (1). Equation (9) describes then the structure of the periodic part of the QES function somewhat more specifically.

The "multitime formalism," naturally, leads to the same structure (8) of the Schrödinger-equation solutions as the approach based on the generalization of the Shirley method,[7] but in the approximate calculation of the spectrum it dispenses with an analysis of the Floquet Hamiltonian with its cumbersome structure,[3,7] and permits development, for the generalized quasienergy, of a perturbation theory (PT) close in form to the stationary PT.[2)]

We develop in this paper a PT for the generalized quasienergy, and investigate, for the multimode case, the nonresonant atomic-spectrum restructuring that occurs when the two radiation-mode frequencies come close together.

§2. PERTURBATION THEORY FOR GENERALIZED QUASIENERGY. SHIFT OF ISOLATED LEVEL

We return to the "two-time formalism." Substituting the function $\tilde{\Phi}_{\varepsilon_1,\varepsilon_2}$ (7) in Eq. (3) we obtain an equation for U:

$$\hat{\mathscr{H}}U_\varepsilon=\varepsilon U_\varepsilon, \tag{11}$$

where the operator $\hat{\mathscr{H}}$ is defined in (5). Equation (11) shows that the function U is determined by the generalized quasienergy $\varepsilon=\varepsilon_1+\varepsilon_2$.

We generalize the method proposed in Ref. 11 for QES. To this end, we introduce the Hilbert space $Q^{\oplus}\tau_1{}^{\oplus}\tau_2$ of functions that depend on the spatial variable q and on the temporal variables t_1 and t_2, and is periodic in t_1 and t_2 with periods T_1 and T_2. We define the scalar product of the elements of this space as follows:

$$(U_1|U_2)=(T_1T_2)^{-1}\int_0^{T_1}dt_1\int_0^{T_2}dt_2\int dq\,U_1^*U_2$$
$$=(T_1T_2)^{-1}\int_0^{T_1}dt_1\int_0^{T_2}dt_2\langle U_1|U_2\rangle. \tag{12}$$

Equation (11) can be regarded as the equation for the eigenvalues of the Hermitian operator $\hat{\mathscr{H}}$ acting in $Q^{\oplus}\tau_1{}^{\oplus}\tau_2$. Hermiticity of means that the generalized quasienergy is real, that the eigenvectors $|U_\varepsilon)$ corresponding to the different eigenvalues are orthogonal, and that the set of eigenvectors is complete. With allowance for the definition (12) of the scalar product, we write the orthonormalization condition

$$(U_{\varepsilon_m}|U_{\varepsilon_n})=\delta_{m,n}.$$

The completeness condition is

$$\sum_\varepsilon|U_\varepsilon)(U_\varepsilon|=I.$$

We note that from the completeness of the set U_ε in $Q^{\oplus}\tau_1\tau_2$ follows also completeness of the set of these functions in Q space. Therefore any solution of the Schrödinger equation (2) can be represented by a series in the wave functions of the generalized QES $\Phi_\varepsilon(B)$:

$$\psi(q,t)=\sum_\varepsilon c_\varepsilon\Phi_\varepsilon(q,t).$$

The generalized quasienergy, just as the usual one, is not uniquely defined. Thus, if $U_{\varepsilon n}$ and ε_n are the eigenfunction and eigenvalue of the operator $\hat{\mathscr{H}}$ (5) that satisfy Eq. (11), then

$$U_{\varepsilon_{n,k1,k2}}(q,t)=U_{\varepsilon_n}(q,t)\exp[i(k_1\omega_1t_1+k_2\omega_2t_2)], \tag{13}$$
$$\varepsilon_{n,\,k1,\,k2}=\varepsilon_n+k_1\omega_1+k_2\omega_2$$

also satisfy this equation. The wave function (8) corresponding to the generalized QES is not altered here.

The PT for the eigenfunctions and eigenvalues of the operator $\hat{\mathscr{H}}$ (5) in the space $Q^{\oplus}\tau_1{}^{\oplus}\tau_2$ is constructed in the usual manner. The operator is represented in the form

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}_0+V,$$

where

$$\hat{\mathscr{H}}_0=\hat{H}_0(q)-i\partial/\partial t_1-i\partial/\partial t_2$$

[see (3)], and the perturbation operator V is of the form

$$V(q,\,t_1,\,t_2)=V_1(q,\,t_1)+V_2(q,\,t_2).$$

The solution of the unperturbed problem

$$\hat{\mathscr{H}}_0U_n^{(0)}=\varepsilon_n^{(0)}\,U_n^{(0)}$$

is assumed known. In the standard formulas of the stationary PT, the matrix elements of the perturbation operator V should be taken in our case to mean

$$V_{nm}=(U_n^{(0)}|V|U_m^{(0)})$$

in accordance with the definition of the scalar product (12). The indicated ambiguity of the generalized quasienergy (13) leads to an additional quasidegeneracy of the eigenvalues of the operator $\hat{\mathscr{H}}_0$ in the case of resonance between the atomic levels and the external fields. Thus, if

$$\varepsilon_n^{(0)} \approx \varepsilon_m^{(0)} + \omega_1 + \omega_2,$$

we get

$$\varepsilon_{n,1,1}^{(0)} \approx \varepsilon_m^{(0)}.$$

Thus, in the approach developed here the resonances can be considered by PT for nearby levels, in full analogy with the single mode case, when the QES method is applicable,[12]

A PT correction, of any order, to a nondegenerate eigenvalue of the operator $\hat{\mathscr{H}}_0$ can be easily found by iterating the equations for the eigenvalue ε_n (see, e.g., Ref. 13):

$$\varepsilon_n = \varepsilon_n^{(0)} + \sum_{k=0}^{\infty} (U_n^{(0)} | V \{G^r_{\varepsilon_n^{(0)}} (\varepsilon_n^{(0)} - \varepsilon_n + V)\}^k | U_n^{(0)}).$$

Here

$$G^r_{\varepsilon_n^{(0)}} = \sum_m{}' \frac{|U_m^{(0)})(U_m^{(0)}|}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}}, \quad \sum_m{}' \equiv \sum_{m \neq n} \tag{14}$$

is the reduced Green's function. We shall assume below that

$$\varepsilon_n = \varepsilon_n^{(0)} + \varepsilon_n^{(2)} + \varepsilon_n^{(4)} + \ldots,$$

inasmuch as in an atom, as in any system with inversion center, there are generally speaking no level shifts of odd order in the field. For $\varepsilon_n^{(2)}$ we obtain the known PT expression

$$\varepsilon_n^{(2)} = (U_n^{(0)} | V G^r_{\varepsilon_n^{(0)}} V | U_n^{(0)}) = \overline{\sum_{m_s}}{}' \frac{|V_{nm}|^2}{\varepsilon_n^{(0)} - \varepsilon_m^{(0)}}. \tag{15}$$

The fourth order correction $\varepsilon_n^{(4)}$, recognizing that $\varepsilon_n^{(1)}$ vanishes, takes the form

$$\varepsilon_n^{(4)} = (U_n^{(0)} | V G^r_{\varepsilon_n^{(0)}} V G^r_{\varepsilon_n^{(0)}} V G^r_{\varepsilon_n^{(0)}} V | U_n^{(0)})$$
$$+ (U_n^{(0)} | V G^r_{\varepsilon_n^{(0)}} V | U_n^{(0)}) (U_n^{(0)} | V (\partial G^r_{\varepsilon_n^{(0)}} / \partial \varepsilon_n^{(0)}) V | U_n^{(0)}), \tag{16}$$

where account is taken of the identity

$$G^r_{\varepsilon_n^{(0)}} G^r_{\varepsilon_n^{(0)}} = \sum_m{}' \frac{|U_m^{(0)})(U_m^{(0)}|}{(\varepsilon_n^{(0)} - \varepsilon_m^{(0)})^2} = - \frac{\partial G^r_{\varepsilon_n^{(0)}}}{\partial \varepsilon_n^{(0)}},$$

that follows from the definition (14) of the reduced Green's function and from the fact that $|U_m^{(0)})$ is orthonormalized.

Let the atom be acted upon by two monochromatic waves:

$$\mathbf{F}_j = \operatorname{Re} \mathbf{F}_{0j} e^{-i\omega_j t}, \quad j = 1, 2.$$

The interaction operator of the jth radiation mode takes in the dipole approximation the form

$$V_j(q, t) = V_j^+ e^{i\omega_j t} + V_j^- e^{-i\omega_j t},$$

where

$$V_j^- = -\tfrac{1}{2} \mathbf{F}_{0j} \mathbf{d}, \quad V_j^+ = (V_j^-)^+,$$

d is the atom dipole-moment operator. In the case of an isolated atomic level, the shift of the generalized quasienergy can be determined by PT for nondegenerate levels. One can regard as isolated an atomic level with zero angular momentum and far enough from the other levels, or a level with nonzero angular momentum if the two waves are polarized in the same plane (the angular-momentum component along the common polarization direction is preserved, or else the two waves propagate in the same direction and have the same circular polarization (the component of the atom angular momentum along the propagation direction is preserved). It is assumed here that the electromagnetic field is not at resonance with the atomic level (nor are there any resonances with the combination frequencies).

To find the shift of the generalized quasienergy we write down the operator of the atom interaction with the bichromatic field in the "two-time formalism":

$$V(q, t_1, t_2) = \sum_{j=1}^{2} (V_j^+ e^{i\omega_j t_j} + V_j^- e^{-i\omega_j t_j}). \tag{17}$$

Let us specify the unperturbed function $U^{(0)}$ in greater detail. Let φ^n be the wave function of the atom,

$$\hat{H}_0 \varphi_n = E_n \varphi_n,$$

and E_n the energy of the corresponding atomic state. We introduce the notation: $U_{n,0,0}^{(0)} \equiv \varphi_n$, $\varepsilon_n^{(0)} \equiv E_n$, and

$$U_{n,k_1,k_2}^{(0)} = U_{n,0,0}^{(0)} \exp[i(k_1 \omega_1 t_1 + k_2 \omega_2 t_2)],$$

$$\varepsilon_{n,k_1,k_2}^{(0)} = \varepsilon_n^{(0)} + k_1 \omega_1 + k_2 \omega_2$$

in accord with the ambiguity of the definition of the quasienergy (13). For the shift (15) of second order in the field we obtain

$$\varepsilon_n^{(2)} = \sum_{m,k_1,k_2}{}' \frac{(U_{n,0,0}^{(0)} | V | U_{m,k_1,k_2}^{(0)}) (U_{m,k_1,k_2}^{(0)} | V | U_{n,0,0}^{(0)})}{E_n - E_m - k_1 \omega_1 - k_2 \omega_2}.$$

Substituting here the perturbation V in the form (17) and taking the definition (12) of the scalar product into account, we obtain ultimately

$$\varepsilon_n^{(2)} = \varepsilon_n^{(2)}(\omega_1) + \varepsilon_n^{(2)}(\omega_2). \tag{18}$$

Here

$$\varepsilon_n^{(2)}(\omega_i) = \langle \varphi_n | V_i^- g_{E_n - \omega_i} V_i^+ | \varphi_n \rangle + \langle \varphi_n | V_i^+ g_{E_n + \omega_i} V_i^- | \varphi_n \rangle \tag{19}$$

is the quadratic Stark shift of the nth level in the ith field,[1] and

$$g_E = (E - \hat{H}_0)^{-1}$$

is the Green's function of the atom. The Stark shift is thus additive in second order in the field. This result is obviously valid for an arbitrary number of modes.

The fourth-order shift in the field is determined by the general formula (16), which contains a non-diagram term with the derivative of the Green's function. We represent the generalized-quasienergy shift $\varepsilon_n^{(4)}$ in the form

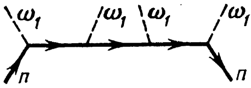
$$\varepsilon_n^{(4)} = \varepsilon_n^{(4)}(\omega_1) + \varepsilon_n^{(4)}(\omega_2) + \varepsilon_n^{(4)}(\omega_1, \omega_2). \tag{20}$$

Here $\varepsilon_n^{(4)}(\omega_1)$ and $\varepsilon_n^{(4)}(\omega_2)$ are the fourth-order level shifts in each of the fields, and are expressed in terms of the dynamic hyperpolarizability of the atom,[1] while $\varepsilon_n^{(4)}(\omega_1, \omega_2)$ contains an effect of interference of two fields. The term $\varepsilon_n^{(4)}(\omega_1, \omega_2)$ (20) appears because combined four-photon transitions (two photons ω_1 and two photons ω_2) are possible without a change of the states of the atom and the field. It is these transitions which lead to a level shift in accordance with the adiabatic theorem.[13] The interference effect due to three fields should obviously appear in the sixth-order shift, etc.

After substituting the perturbation V (17), the nondiagram term in (16) takes the form

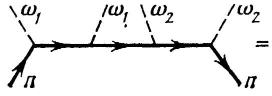
$$\varepsilon_n^{(2)}\sum_{i=1}^{2}\left(\langle\varphi_n|V_i^-(\partial g_{E_n-\omega_i}/\partial E_n)V_i^+|\varphi_n\rangle\right.$$
$$\left.+\langle\varphi_n|V_i^+(\partial g_{E_n+\omega_i}/\partial E_n)V_i^-|\varphi_n\rangle\right), \qquad (21)$$

where the second order shift $\varepsilon_n^{(2)}$ is defined by Eq. (18). Contributions to $\varepsilon_n^{(4)}(\omega_1)$ are made only by the four terms obtained on expansion of the product (21). In addition, the contribution to from the first diagram term of (16) is made by 4!/(2!2!) = 6 diagrams:



$$=\langle n|V_1^+g_{E_n+\omega_1}V_1^-g_{E_n}V_1^+g_{E_n+\omega_1}V_1^-|n\rangle$$

and by those obtained from it upon nonequivalent permutations of the photon lines. The structure of $\varepsilon_n^{(4)}(\omega_2)$ in (20) is similar. Contributions to $\varepsilon_n^{(4)}(\omega_1,\omega_2)$ are made by the remaining eight terms of (21), and also by the diagram



$$=\langle n|V_2^+g_{E_n+\omega_2}V_2^-g_{E_n}V_1^+g_{E_n+\omega_1}V_1^-|n\rangle$$

and by 23 more diagrams obtained from it by permutation of the photon lines. By g_{E_n} is meant here the reduced Green's function of the atom:

$$g_{E_n}=\sum_m{}'\frac{|\varphi_m\rangle\langle\varphi_m|}{E_n-E_m}=\lim_{E\to E_n}\left\{(E-\hat{H}_0)^{-1}-\frac{|\varphi_n\rangle\langle\varphi_n|}{E-E_n}\right\}.$$

§3. RESTRUCTURING OF ATOMIC SPECTRUM ON MUTUAL APPROACH OF TWO MODE FREQUENCIES

In the limit of equal frequencies $\omega_1=\omega_2=\omega_3$ the usual QES is applicable.[1] The second-order quasienergy shift if field is determined by Eq. (19) with the substitutions $\omega_i\to\omega, V_i^\pm\to V_1^\pm+V_2^\pm$:

$$\varepsilon_n^{(2)}=\varepsilon_n^{(2)}(\omega_1)|_{\omega_1=\omega}+\varepsilon_n^{(2)}(\omega_2)|_{\omega_2=\omega}+2\operatorname{Re}W_n(\omega,\omega), \qquad (22)$$

where

$$W_n(\omega_1,\omega_2)=\langle\varphi_n|V_1^+g_{E_n+\omega_2}V_2^-|\varphi_n\rangle+\langle\varphi_n|V_2^-g_{E_n-\omega_1}V_1^+|\varphi_n\rangle. \qquad (23)$$

The generalized quasienergy should go over into the quasienergy value at $\omega_1=\omega_2$, but (8) does not go over into (22) in this case. Equation (18) for the shift of the generalized quasienergy in a bichromatic field no longer holds when the frequencies come closer together, and this in turn points to the onset of singularities in the atom's spectrum.

The onset of these singularities is easy to trace quantitatively by changing to a quantum description of the electromagnetic field. When the frequencies come close together, the energy of the state "atom + n_1 photons ω_1 + n_2 photons ω_2" becomes close to the energy of the states "atom + (n_1+k) photons ω_1 + (n_2-k) photons ω_2." Since the energies are close, even a weak interaction of the field with the atom causes photons to be transferred from one mode to the other. The result is a restructuring of the atomic spectrum.

If the generalized quasienergy is determined by the PT developed in §2, the case of close frequencies must be considered specially, for in this case unperturbed eigenvalues of the operator $\hat{\mathscr{H}}_0$ come close together. Indeed, since the definition (13) is ambiguous, the quasienergies that come close together are

$$\varepsilon_{n,k,-k}^{(0)}=\varepsilon_n^{(0)}+k\delta\equiv\varepsilon_{n,k}^{(0)},$$

where $\varepsilon_n^{(0)}=E_n$ is the energy of the atomic state, $\delta=\omega_1-\omega_2$, $k=0,\pm1,\pm2,\ldots$. The corresponding eigenfunctions of the operator take the form

$$U_{n,k,-k}^{(0)}=\varphi_n(q)\exp[ik(\omega_1t_1-\omega_2t_2)]\equiv U_{n,k}^{(0)}.$$

Thus, as the frequency difference decreases, PT for close levels must be used to determine the generalized quasienergy. We note that a similar problem arose when the static limit of the quasienergy was considered.[1]

According to the PT for close levels, we seek the eigenfunction of the operator $\hat{\mathscr{H}}$ in the form

$$U_{\varepsilon_n}(q,t_1,t_2)=\sum_{k=-\infty}^{\infty}a_kU_{n,k}^{(0)}$$
$$=\varphi_n(q)\sum_{k=-\infty}^{\infty}a_k\exp[ik(\omega_1t_1-\omega_2t_2)], \qquad (24)$$

where the coefficients a_k satisfy the system of equations

$$[\varepsilon_n-\varepsilon_{n,k}^{(0)}-W_{kk}]a_k=\sum_{k'\neq k}W_{kk'}a_{k'}. \qquad (25)$$

The interaction V (17) does not mix the functions $U_{n,k}^{(0)}$ in first order. In second order we have

$$W_{kk'}=\sum_{m,k_1,k_2}{}'\frac{(U_{n,k}^{(0)}|V|U_{m,k_1,k_2}^{(0)})(U_{m,k_1,k_2}^{(0)}|V|U_{n,k'}^{(0)})}{E_n+k\delta-E_m-k_1\omega_1-k_2\omega_2}. \qquad (26)$$

The sum Σ' does not include the terms with $(m,k_1,k_2)=(n,k,-k)$.

The integration with respect to t_1 and t_2 in the calculation of the matrix elements of the operator V in (26) leaves only the following nonzero values

$$W_{kk}=\varepsilon_n^{(2)}(\omega_1)+\varepsilon_n^{(2)}(\omega_2)\equiv v_n,$$

$W_{kk-1}=W_n$ and $W_{kk+1}=W_n^*$, where $\varepsilon_n^{(2)}(\omega_i)$ and W_n are defined by Eqs. (19) and (23). We noted that without loss of accuracy we can substitute $\omega_1\leftrightarrow\omega_2$ in (23).

The system (25) for the coefficients a_k becomes much simpler:

$$(\varepsilon_n-E_n-k\delta-v_n)a_k-W_na_{k-1}-W_n^*a_{k+1}=0. \qquad (27)$$

According to (24), the almost periodic part (9) of the wave function of the generalized QES (8) takes in our case the form

$$U_{\varepsilon_n}(q,t)=U_{\varepsilon_n}(q,t,t)=\varphi_n(q)f(t),$$

where

$$f(t)=\sum_k a_k \exp(ik\delta t), \quad f(t+2\pi/\delta)=f(t).$$

It is easy to verify that the system (27) is equivalent to a differential equation for the function $f(t)$

$$idf/dt+[\varepsilon_n-E_n-v_n-2|W_n|\cos(\delta t+\alpha_n)]f=0,$$

where $\alpha_n = \arg W_n$. This equation has a solution that satisfies the condition $|f| = 1$, in the form

$$f(t)=\exp\{i[(\varepsilon_n-E_n-v_n)t$$
$$-2|W_n|\delta^{-1}(\sin(\delta t+\alpha_n)-\sin\alpha_n)]\}.$$

From the condition that $f(t)$ be periodic we obtain the generalized quasienergy ε_n:

$$\varepsilon_n=E_n+v_n=E_n+\varepsilon_n^{(2)}(\omega_1)+\varepsilon_n^{(2)}(\omega_2). \tag{28}$$

The coefficient a_k is expressed in terms of the Bessel function J_k:

$$a_k=(-1)^k J_k(2|W_n|\delta^{-1})\exp[i(k\alpha_n+2|W_n|\delta^{-1}\sin\alpha_n)]. \tag{29}$$

Thus, when the frequencies of two radiation modes come closer the wave function of the generalized QES that corresponds to the generalized quasienergy ε_n (28) acquires the structure

$$\Phi_{\varepsilon n}(q,t)=\varphi_n(q)f(t)e^{-i\varepsilon_n t}=\varphi_n(q)\exp\{-i[\varepsilon_n t$$
$$+2|W_n|\delta^{-1}(\sin(\delta t+\alpha_n)-\sin\alpha_n)]\}. \tag{30}$$

We investigate now the limiting cases.

1. Distant frequencies: $|W_n|\ll|\delta|$. From 30 we get

$$\Phi_{\varepsilon n}(q,t)=\varphi_n(q)e^{-i\varepsilon_n t}.$$

In this case there is no mixing of the harmonics of the generalized QES, i.e., $a_k\approx\delta_{k,0}$ in (24), as follows also from the explicit expression (29) for the coefficient a_k. The shift of the generalized quasienergy (28) is determined by the additive Stark shift (18) in the bichromatic field.

2. Very close frequencies: $|W|\ll|\delta|$. Taking the limit in (30) we obtain

$$\Phi_{\varepsilon n}(q,t)=\varphi_n(q)\exp[-i(\varepsilon_n+2|W_n|\cos\alpha_n)t]. \tag{31}$$

Note that at the phase chosen here for the function Φ_{ε_n}, the limiting transition of (30) in (31) is valid only if $|\delta|t\ll1$. This seemingly strange condition has in fact the following meaning. In the limit $|W_n|\gg|\delta|$, Φ_{ε_n} (30) is a rapidly oscillating function of t, except for short time intervals $\tau\ll|\delta|^{-1}$ which are close to $t=0$, $2\pi/|\delta|$, $4\pi/|\delta|$,...at the chosen phase of the function Φ_{ε_n} (at the chosen time origin). Φ_{ε_n} is substantially different from zero only in these time intervals, and it just here that (30) goes over into (31) (generally speaking, apart from the phase). The role of the generalized quasienergy is now assumed in fact by the quantity

$$\tilde{\varepsilon}_n=\varepsilon_n+2|W_n|\cos\alpha_n=E_n+\varepsilon_n^{(2)}(\omega_1)+\varepsilon_n^{(2)}(\omega_2)+2\,\mathrm{Re}\,W_n, \tag{32}$$

so that the correct value of the level shift (22) is obtained at $\omega_1=\omega_2$.

The criterion for the proximity of the frequencies is the condition

$$|\delta|\sim|W_n|. \tag{33}$$

The matrix element $|W_n|$ (23) is of the order of the Stark shift of the level. It can therefore be stated that frequencies are close if the different δ between them is of the order of the Stark shift of the level.

Let us examine in greater detail the singularities of the emission or absorption spectrum of an atom acted upon by bichromatic laser radiation with frequencies that are close in the sense of (33). The expression obtained for the wave function (30) indicates that the atom emission spectrum corresponding to the transition $n\rightarrow n'$ acquires, besides the fundamental line determined by the difference $\Omega_0=\varepsilon_n-\varepsilon_{n'}$, between the generalized quasienergies, also satellites having the frequencies

$$\Omega_\nu=\varepsilon_n-\varepsilon_{n'}-\nu\delta, \quad \nu=\pm1,\pm2,\dots . \tag{34}$$

Equation (34) corresponds to the general expression (10) with $k_2=\nu$. A satellite structure appears also in the absorption spectrum corresponding to the transition $n'\rightarrow n$.

The intensity I_ν of the νth satellites is determined by the squared modulus of the νth harmonic of the function

$$\exp\{2i\delta^{-1}[|W_{n'}|\sin(\delta t+\alpha_{n'})-|W_n|\sin(\delta t+\alpha_n)]\}.$$

Thus,

$$I_\nu\sim J_\nu^2[2\delta^{-1}(|W_n|^2+|W_{n'}|^2-2|W_nW_{n'}|\cos(\alpha_n-\alpha_{n'}))^{1/2}]. \tag{35}$$

The proportionality coefficient common to all the satellites is equal to the atom-emission intensity in the absence of the laser field.

If the two waves polarized in one plane either have the same propagation direction or the same right (left) circular polarization, W_n is a real number. Equation (35) for the intensity of the satellites takes in this case the simpler form

$$I_\nu\sim J_\nu^2[2\delta^{-1}(W_{n'}-W_n)].$$

From the known property of Bessel functions of integer index, $J_{-\nu}=(-1)^\nu J_\nu$, it follows that the intensity distribution in the satellite spectrum is symmetric about the fundamental line. In addition the combined intensity $\Sigma_\nu I_\nu$ of the fundamental lines and of the satellites is equal to the emission intensity in the absence of the laser field, as follows from the identity

$$\sum_{\nu=-\infty}^{\infty} J_\nu^2=1.$$

The intensity of the satellites in the absorption spectrum is also given by (35).

Figure 1 shows the atom-spectrum satellite structure produced when the frequencies of two emission modes come close together. It follows from (35) that the intensity of the satellites depends on the frequency difference δ and on the intensity, propagation direction, and polarization of each of the modes. In the limit of distant frequencies

$$|\delta|\gg|W_n|, \ |W_{n'}|,$$

the satellite structure of the spectrum vanishes ($I_\nu/$

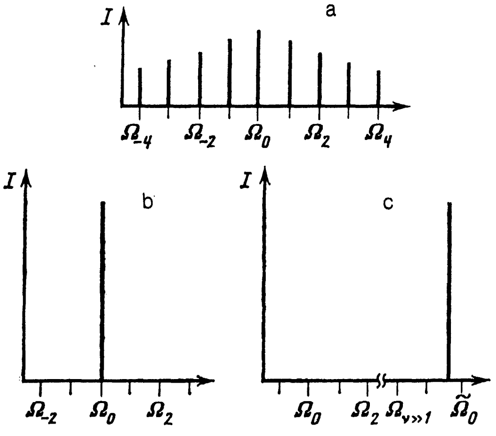


FIG. 1. Structure of the spectrum of an atom in a bichromatic field: a—satellites $\Omega_\nu = \Omega_0 - \nu\delta$ in the case of close frequencies $|\delta| \sim |W_n|, |W_{n'}|$; b—distant frequencies, $|\delta| \gg |W_n|, |W_{n'}|$; c—very close frequencies, $|\delta| \ll |W_n|, |W_{n'}|$.

$I_0 \sim |W/\delta|^{2|\nu|}$, where $|W| = \max(|W_n|, |W_{n'}|)$) leaving radiation at the fundamental frequency $\Omega_0 = \varepsilon_n - \varepsilon_{n'}$, determined by the difference between the generalized quasienergies (28). In the limit of very close frequencies, $|\delta| \ll |W_n|, |W_{n'}|$, the intensity of the satellites and of the fundamental lines decreases (like $|\delta/W|$, at $|\nu| \ll |W/\delta|$). What is left, generally speaking is only radiation at the frequency

$$\tilde{\Omega}_0 = \tilde{\varepsilon}_n - \tilde{\varepsilon}_{n'} = \Omega_0 + 2\,\mathrm{Re}(W_n - W_{n'})$$

[see (32)], which is produced in transitions between states described by the wave functions (31). It follows from the foregoing analysis of the transition from (30) to (31), this radiation occurs during time intervals $\tau \ll |\delta|^{-1}$ and its intensity is determined by the squared modulus not of one harmonic but of an entire group of terms with $\nu \sim (\Omega_0 - \tilde{\Omega}_0)\delta^{-1}$ in the Fourier expansion of the transition amplitude.

We stress in particular that the onset of the spectrum satellite structure is a nonresonant effect. The frequency of the laser radiation can be arbitrary, all that is necessary is that the frequencies of the two modes be close in the sense of (33). As the frequency approaches resonance with the atomic transition, increases take place in the Stark shift of the level and in the matrix element W_n (23), so that the condition (33) becomes satisfied at a larger frequency difference δ.

We note also that the necessary frequency difference can be attained via the Doppler effect by irradiating an atom beam or a gas by colliding (generally, crossed) electromagnetic waves of equal frequency.

Spectrum singularities similar to those considered here occur obviously also when an atom is irradiated by more than two monochromatic waves of close frequency. The satellite structure of the spectrum turns out here to be more elaborate. It includes lines with frequencies $\Omega_0 + \nu_{ij}\delta_{ij}$, as well as lines with frequencies $\Omega_0 + \nu_{12}\delta_{12} + \nu_{13}\delta_{13} + \dots$, where $\delta_{ij} = \omega_i - \omega_j$, $\nu_{ij} = 0, \pm 1, \pm 2, \dots$. An analytic expression cannot be obtained for the satellite intensities even in the case of three waves, but in principle it can be calculated numerically.

[1)] Note that the structure of Schrödinger-equation solutions in the multimode case was indicated already by Shirley[3] in an analysis of the transition from the quantum description to the semiclassical one.

[2)] A stationary PT for the quasienergy was developed in Ref. 11.

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Translated by J. G. Adashko