

METHODOLOGICAL NOTES

Scattering and emission of a quantum system in a strong electromagnetic wave¹⁾

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A method is considered for consistent analysis of a quantum-mechanical system situated in a potential that depends periodically on the time, for example, in the field of a strong classical electromagnetic wave. The emission of such a system is considered, and particularly the shift of the fundamental frequency ω' and the appearance of satellites $\omega' \pm h\omega$.

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1. INTRODUCTION

Interaction of a strong monochromatic electromagnetic wave with a quantum-mechanical system (atom or molecule) is of particular interest in connection with the development of masers and lasers.

Coherent scattering of a monochromatic wave, the dependence of the scattering cross section and of the refractive index on the amplitude, coherent generation of harmonics, incoherent emission at the natural frequencies of the atom, and the variation of these frequencies under the influence of waves—this is but a brief and incomplete list of the questions raised. These effects are considered in a large number of papers, of which we shall cite the most interesting ones^[1], in which references to earlier works are also given. Various methods were used: perturbation theory (with time-dependent potential), density-matrix (with allowance for dissipation), and others. A detailed and rigorous review of nonstationary perturbation theory was given recently by Langhoff et al.^[2]. Problems involved in the emission of perturbed states are not considered in^[2].

However, the most consistent, systematic, and at the same time simplest method is the use of quasienergy and quasienergy states (QES). Therefore by way of perfecting the method, and without claiming new results, we shall describe the quasienergy method. In this method, the influence of a classical electromagnetic wave on the atom is taken into account rigorously (at least in principle), and the spontaneous emission is regarded as a small dissipative perturbation. This method is exactly equivalent to the standard theory of the free atom, in which one first obtains exact solutions of the Schrödinger equation without taking the spontaneous emission into account, and a set of energy levels and natural (stationary) states is obtained. The spontaneous emission is regarded as a small perturbation of the system. In principle, when radiation is taken into ac-

count, only the lower state is exactly stationary. It is remarkable that in the presence of a strong wave, in the same approximation with allowance for the spontaneous processes, all the states are qualitatively identical in the sense that none of the QES is rigorously stationary. Let us recall the history of the quasienergy concept.

It is universally known that an electron situated in the spatially periodic field of a crystal lattice possesses a conserved quasimomentum p , $\psi(x+a) = e^{ipa}\psi(x)$, where a is the lattice constant and we have put $\hbar = 1$. Considering an electron (relativistic, obeying the Dirac equation) in the field of a strong wave, Nikishov and Ritus^[3] introduced the concept of four-dimensional quasimomentum. Its fourth component was called quasienergy.

Two papers^[4,5], published practically simultaneously in 1966, applied the quasienergy concept to an atomic system in the field of a wave (see also^[6,7]). Ritus^[4] considered a concrete method of obtaining the wave functions of QES. By definition, these functions satisfy the condition

$$\psi_h(t+T) = e^{-iF_h T} \psi_h(t). \quad (1)$$

If we separate the harmonic factor, we can write

$$\psi_h(t) = e^{-iF_h t} \varphi_h(t), \quad (2)$$

where $\varphi_k(t+T) = \varphi_k(t)$, so that $\varphi_k(t)$ is a strictly periodic (but not harmonic) function of the time²⁾. ψ_k and φ_k also depend on x .

The function ψ_i can be expanded in a Fourier series:

$$\psi_i = \sum_{n=-\infty}^{n=+\infty} c_{ink} \varphi_n(x) e^{-i(F_i + n\omega)t}, \quad \omega = 2\pi/T. \quad (3)$$

Ritus has constructed equations for the determination of the coefficient c_{ink} and of the quasienergy F_i itself.

Zel'dovich^[5] also introduced the concept of quasi-

energy in QES, did not consider the concrete method of calculating F_1 and ψ_1 , but instead considered in detail the question of the radiation by the system. It has been noted, in particular, that F_1 is defined in modulo ω , i.e., $F_1 = F_1 \pm \omega = F_1 \pm 2\omega$, so that we cannot say, for example, that $F_2 > F_1$, since it is always possible to choose³⁾ integers (not necessarily positive) m and n such that

$$F_2 + n\omega < F_1 + m\omega.$$

We thus have a democracy—all the QES spontaneously go over into one another and there are no energy hindrances, since the strong wave is a reservoir of energy. All that remain are hindrances of the type of parity. A transition between two specified states 2 and 1 gives not a single line, but a series, in accordance with the fact that the quasienergy is defined in modulo ω . However, since this modulus is the same for both states, the series is of the one-parameter type, and the frequency depends on $(n - m)$ but not on n and m separately.

Obviously, individual terms of the Fourier expansion give results that do not differ from the standard perturbation theory. The concept of quasienergy is useful but not essential. It would be difficult, however, to consider strong nonlinear effects and ignore the quasienergy.

Near resonance, i.e., at $E_2 - E_1 - n\omega \ll \omega$, the QES differ strongly from the stationary eigenstates of the unperturbed atom even at the relatively weak electromagnetic field of a maser or laser wave. It is precisely in this situation that the departure from the framework of perturbation theory, realized with the aid of the QES theory, is particularly fruitful. In a number of cases, the energy differences within a definite group of levels (two levels or more, but not a continuum!) are small, and the mixing of these levels with one another by a relatively weak field is of importance; remote levels and the ionization continuum are unaffected.

To the contrary, in the case of optical transitions in an atom far from resonance, there appears, simultaneously with nonlinear effects, a strong ionization of the atom, and the observation of nonlinear effects becomes difficult. In this case the quasienergy spectrum turns out to be continuous, and the discrete states have a complex quasienergy the imaginary part of which characterizes the probability of ionization of the atom in a given state by the wave. Among the cases in which one can hope for a useful application of quasienergy are nearby levels of atoms with molecules resulting from spin-orbit or hyperfine splitting; the system of degenerate levels of the hydrogen atom and the almost degenerate hydrogenlike high levels of atoms and ions; rotational levels of dipolar molecules, particularly those split by the magnetic field; level pairs produced in the presence of two equivalent states with low probability of spontaneous transition between them⁴⁾. Some of the foregoing examples are considered in greater detail at the end of the article.

2. EVOLUTION OF SYSTEM

We consider in greater detail the analogy between the stationary states of an unperturbed system and the QES of a system in a periodic field, and the application of these concepts to the problem of the evolution of a system.

In both cases, the Schrödinger equation holds:

$$i\partial\Psi/\partial t = \mathcal{H}\Psi, \quad (4)$$

and in the unperturbed case $\mathcal{H} = \mathcal{H}_0(x, \partial/\partial x)$ does not depend on the time. Thus, we have before us a linear partial differential equation. No one, however, even in possession of a superpowerful electronic computer, will start to solve this equation numerically⁵⁾, by a difference method, by finding the increments

$$\Psi(x, t + \Delta t) = \Psi(x, t) - i\mathcal{H}_0\Psi(x, t)\Delta t. \quad (5)$$

Instead, the solution is broken up into several stages:

1) we find the eigenstates, i.e., particular solutions of the type

$$\psi_k = \varphi_k(x) e^{-iE_k t}; \quad (6)$$

2) We represent the arbitrary initial state $\Psi(x, t_0)$ as a superposition of eigenstates, i.e., we find c_k in the expression

$$\Psi(x, t_0) = \sum_k c_k \varphi_k(x) e^{-iE_k t_0}; \quad (7)$$

3) The solution of the evolution problem, i.e., of calculating the value of $\Psi(x, t)$ at an arbitrary instant of time, is then written out immediately:

$$\Psi(x, t) = \sum_k c_k \varphi_k(x) e^{-iE_k t}. \quad (8)$$

This procedure is universally known. It is presented here only to demonstrate the complete analogy with quasienergy theory. Let $\mathcal{H} = \mathcal{H}(x, \partial/\partial x, t)$ contain the time t in explicit form. Then there are no solutions of the type (6). However, in the case of a periodic dependence $\psi(t + T) = \psi(t)$ we can find QES, i.e., solutions that reproduce themselves periodically. Thus, in the case of a periodic $\mathcal{H}(t)$, the first stage consists of finding solutions of the type

$$\psi_k = \varphi_k(x, t) e^{-iF_k t}, \quad (9)$$

where φ_k are not constant but depend on the time periodically. The second stage consists of expanding an arbitrary function $\Psi(x, t_0)$, specified at the instant of time t_0 :

$$\Psi(x, t_0) = \sum_k c_k \varphi_k(x, t_0) e^{-iF_k t_0}. \quad (10)$$

Obtaining the coefficient c_k , we construct a solution that satisfies both the Schrödinger equation and the initial condition at the instant t_0 :

$$\Psi(x, t) = \sum_k c_k \varphi_k(x, t) e^{-iF_k t} = \sum_k c_k \psi_k. \quad (11)$$

The analogy is thus complete. The coefficients of the various ψ_k are strictly constant and the general character of the solution is directly evident.

A feature common to the solution with the time-independent Hamiltonian \mathcal{H}_0 and with the periodic Hamiltonian $\mathcal{H}(t)$ is that the spontaneous emission (transitions from one state to another) are disregarded for the time being. The principal basis in the case of \mathcal{H}_0 is the existence of a complete orthonormal set of functions $\varphi_k(x)$, which have a solution of the eigenvalue equation

$$\mathcal{H}_0 \varphi_k(x) = E_k \varphi_k(x).$$

In the case of a periodic potential, the mathematical formulation of the problem of finding the QES functions $\varphi_k(x, t)$ is at first glance entirely different, and it cannot be reduced to the problem with time eliminated. It turns out, however, that the QES also form a complete orthonormal system.

In practice, as a rule, a periodic potential consists of a time-independent Hamiltonian \mathcal{H}_0 and a periodic part

$\mathcal{H}'(t)$, $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}'$. The problem, however, is to determine the exact solutions of the Schrödinger equation for $\mathcal{H}(t)$, unlike in perturbation theory, in which \mathcal{H}' would be regarded as small. Nonetheless, we can trace the correspondence between the solutions for \mathcal{H}_0 and the solutions for $\mathcal{H}(t)$. We introduce formally for this purpose a parameter α , define $\mathcal{H} = \mathcal{H}_0 + \alpha\mathcal{H}'$, and trace the variation of the solutions when α varies from 0 to 1. Each eigenfunction \mathcal{H}_0 turns out to be set in correspondence to one quasienergy solution. If we consider a closed group of states \mathcal{H}_0 , which are "intmixed" with one another by the "perturbation" $\alpha\mathcal{H}'$, then it is clear that the number of QES does not differ from the initial number of states \mathcal{H}_0 .

In this case we can also indicate an explicit algorithm for finding the wave functions of the QES and the values of the quasienergy. We shall consider below concretely the simplest example of a group consisting of two states.

We proceed from the mathematical problem of states that depend on the parameter α in the Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \alpha\mathcal{H}'(t)$ to the physical problem of smoothly turning on a periodic potential. Let $\alpha = \alpha(t)$ increase slowly (during a time much longer than the period T , which is assumed constant) from 0 to 1. If the system is in a specified k -th state of \mathcal{H}_0 at the initial instant of time and at $\alpha = 0$, then when α grows slowly the system goes over into a pure k -th QES state⁶⁾.

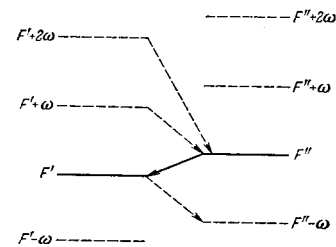
Thus, the QES have a clear-cut physical meaning, that of states obtained when a periodic perturbation \mathcal{H}' is turned on smoothly. Moreover, we have obtained a non-obvious theorem: from an eigenstate of \mathcal{H}_0 we obtain, by smoothly turning on \mathcal{H}' , a state that has a periodicity property with period T equal to the period of \mathcal{H}' (see the review^[2]). If we use this property, we can obtain the desired solution of the Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}'$ even without considering the process of turning on \mathcal{H}' .

These statements are undoubtedly correct in the case of a finite group of states and under the additional condition that there be no exact resonance: $F_n - F_k \neq m\omega$, where m is an integer. When the continuum plays a role, an additional investigation is necessary. Exact resonance exists apparently only for noninteracting levels, just as terms of like symmetry do not intersect in the stationary theory.

3. RADIATION OF THE EIGENSTATES OF $\mathcal{H}(t)$

In the preceding section we have emphasized the similarity of the QES to the ordinary eigenstates of \mathcal{H}_0 , and in particular the complete analogy in the solution of the problem of the evolution of an arbitrary initial state. One should see, however, not only the similarity but also the difference between the QES and stationary states. This difference is most strikingly pronounced when one considers the radiation of the system itself, radiation not included in the Hamiltonians \mathcal{H}_0 and \mathcal{H}' .

In the case of stationary states, radiation takes place only on going from an upper state to a lower one. Yet a QES radiates by itself (a given n -th QES, without transition to others, $n \pm 1$, $n \pm 2$, ...). The quantity $\int \psi_n^* \times \psi_n dV$ is not constant, but varies in time with the same period as the wave. From the condition of periodicity of $\psi_n(x, t)$ it follows that an electromagnetic wave is radiated and has the same period as the exciting wave. The radiated wave, however, is not harmonic! Its Fourier expansion contains a principal component with frequency ω and



small harmonic components with frequencies $n\omega$ that are multiples of ω .

Obviously, this radiation must be classified as coherent scattering; the radiation of a given QES (without transition to another!) contributes to the real part of the refractive index of a medium filled with the considered atoms. We can calculate the nonlinear polarizability of the medium and the change in the waveform of the wave as a result of the harmonics.

Transitions between different QES are similar to transitions from an excited stationary state to the ground state (or from one excited state to another). Such transitions occur in accordance with the laws of probability and are accompanied by emission of photons with a frequency characteristic of the atom, i.e., different from the wave frequency.

We have already noted in Sec. 1 (Introduction) that in the presence of two QES ψ_n and ψ_k it is impossible to say which of them has the larger and which the smaller quasienergy (see the figure). Spontaneous transitions go in both directions: $\psi_n \rightarrow \psi_k$ with emission $\omega' = F_n - F_k + p\omega$, and $\psi_k \rightarrow \psi_n$ with emission $\omega'' = F_k - F_n + q\omega$, where p and q are integers⁷⁾ such that $\omega' > 0$ and $\omega'' > 0$. In its aggregate, the transition $n \rightarrow k \rightarrow n$ is accompanied by emission of two quanta, such that $\omega' + \omega'' = (p + q)\omega$, so that the energy taken from the field of the strong wave (from $\mathcal{H}'(t)$) is equal to an integer multiple of the frequency ω of the strong wave, as expected.

We assume that the transition probability is small and we disregard the natural widths of the quasienergy levels. However, the probability ratio of the transitions $\psi_n \rightarrow \psi_k$ and $\psi_k \rightarrow \psi_n$ is significant, since it governs the stationary number of atoms in different states⁸⁾, and consequently also the characteristics of the radiation and the refractive index, averaged over the ensemble of the atoms.

Of particular interest is the question of inversion. In a static field (\mathcal{H}_0) it is possible to choose an inverted initial state, i.e., to specify initial concentrations $N_k > N_n$ at $E_k > E_n$. By creating a resonator tuned to the frequency $\omega_{kn} = E_k - E_n$, we obtain a coherent ("laser") pulse. In the absence of the resonator, we obtain a spontaneous emission of frequency ω_{kn} . In the course of time, however, all the atoms go over by radiation to the lower energy state. In the field \mathcal{H}_0 without energy pumping, the excited states, and particularly the inverted state of an aggregate of atoms exist only for a limited time.

An entirely different case is that of a system situated in a periodic field $\mathcal{H}(t)$ in the quasienergy situation. It was noted above that in such a system there are, in principle, spontaneous transitions for all states into all states. As $t \rightarrow \infty$, a definite distribution of the atoms with respect to the QES is established and is character-

ized by a set of numbers N_k , where k is the number of the QES. All these numbers differ from zero at $t \rightarrow \infty$.

However, if there are spontaneous optical transitions between each pair of levels in both directions, $k \neq n$, and furthermore with different frequencies ω' and ω'' , the situation is reversed for one of these transitions, say for $k \rightarrow n$, $\omega'' = F_k - F_n + q\omega$ (see above), if $N_k > N_n$. Let us recall the meaning of inversion. We have considered so far only spontaneous emission at the frequency ω'' . However, if there is spontaneous emission, then there also exists a proportional induced emission, and there is also absorption of the radiation in the inverse transition. The concentration relation $N_k > N_n$ causes the induced emission to be stronger than the absorption. Thus, although the spontaneous emission at the frequency ω'' may turn out to be weaker than the spontaneous emission of the other lines, say ω' , nevertheless the emission at ω'' can be enhanced to the level of a laser pulse by means of a suitable resonator. In practice one should nevertheless expect the lowest QES to be the most populated in the stationary state. More accurately speaking, in accordance with all the rules of the game, this QES should be given the expanded title "QES obtained adiabatically from the lower level of the time-independent \mathcal{H}_0 ."

If $\mathcal{H}'(t)$ is small, then the high-frequency components in this state (remote satellites with large p) are correspondingly small, the probability of spontaneous transition of the "former lower" state into one of the "former upper" states is small, and consequently high and difficult requirements are imposed on the Q of the resonator at the frequency ω'' that ensures generation.

We have not considered here a large number of questions of importance in the practical calculation of the generation (for example, the Doppler line broadening, the kinetics of accumulation of the inverted population, etc.). Within the framework of a theoretical article on quantum-mechanical systems in a periodic field, we must confine ourselves to the indication that generation of coherent radiation at a frequency different from the pump frequency is possible in principle.

The greater part of the general considerations given above has been advanced earlier^[1,5].

4. TWO-LEVEL SYSTEM

Let us show, following^[4], how quasienergy considerations are realized in the simplest case of a two-level system. We disregard spontaneous emission in this section. The wave function and the evolution of this system are determined completely by two amplitudes, $a(t)$ and $b(t)$:

$$\Psi(x, t) = a(t) \varphi(x) + b(t) \chi(x), \quad (12)$$

where φ and χ are the spatial parts of the stationary states of the unperturbed system with energies A and B . Thus, for an unperturbed system we have

$$\Psi_0 = a_0 e^{-iAt} \varphi(x) + b_0 e^{-iBt} \chi(x). \quad (13)$$

We assume that φ and χ have different parity; in the dipole approximation, the interaction with the total energy E is given by the matrix element

$$M = eE \int \varphi(x) x \chi(x) dx, \quad (14)$$

by virtue of which, the final Schrödinger equation in the presence of a field is

$$\begin{aligned} i da/dt &= Aa + 2V \cos \omega t \cdot b, \\ i db/dt &= 2V \cos \omega t \cdot a + Bb; \end{aligned} \quad (15)$$

we have put here $\hbar = 1$, V is proportional to the amplitude of the wave and to the matrix element, the number 2 has been separated for convenience, and ω is the wave frequency.

We write down directly the solution with quasienergy F in the form of Fourier series, i.e.,

$$\begin{aligned} a(t) &= \alpha_0 e^{-iFt} + \alpha_2 e^{-i(F+2\omega)t} + \alpha_{-2} e^{-i(F-2\omega)t} + \dots, \\ b(t) &= \beta_1 e^{-i(F+\omega)t} + \beta_{-1} e^{-i(F-\omega)t} + \beta_3 e^{-i(F+3\omega)t} + \dots \end{aligned} \quad (16)$$

Obviously, these series satisfy identically the condition $a(t+T) = e^{-iFT} a(t)$, and analogously for b , since $T = 2\pi/\omega$. It is convenient to write $2 \cos \omega t = e^{i\omega t} + e^{-i\omega t}$, and this makes obvious the rules of parity of the numbers for the Fourier series for a and b .

Substituting the series into the equation, we obtain an infinite system of coupled equations for the coefficients. Equating the determinant to zero, we obtain two eigenvalues F' and F'' , and then the coefficients $\alpha'_n, \beta'_n, \alpha''_n$ and β''_n corresponding to these two solutions. We write down this system, or more accurately its first two equations:

$$F\alpha_0 = A\alpha_0 + V(\beta_1 + \beta_{-1}), \quad (F + \omega)\beta_1 = B\beta_1 + V(\alpha_0 + \alpha_2). \quad (17)$$

Let

$$B > A, \quad \omega - (B - A) = \delta \quad (\delta < \omega); \quad V < B - A.$$

Then a reasonable iteration yields $F = A + \epsilon$ ($\epsilon < \omega$),

$$\begin{aligned} \epsilon\alpha_0 &= V\beta_1, \quad (\delta + \epsilon)\beta_1 = V\alpha_0, \quad \epsilon(\delta + \epsilon) = V^2, \\ \epsilon', \epsilon'' &= -(\delta/2) \pm [(\delta^2/4) + V^2]^{1/2}. \end{aligned} \quad (18)$$

When the wave is adiabatically turned on (by increasing V from $V = 0$ at $\omega = \text{const}$), the first state, corresponding to the solution ϵ' (+) is obtained from the lower state:

$$V \rightarrow 0, \quad F' = A + (V^2/\delta), \quad \alpha'_0 = 1 - (V^2/2\delta^2), \quad \beta'_1 = V/\delta. \quad (19)$$

The second state ϵ'' also pertains to the upper unperturbed state:

$$\begin{aligned} V \rightarrow 0, \quad F'' &= A - \delta - (V^2/\delta) = B - \omega - (V^2/\delta), \\ \alpha''_0 &= -V/\delta, \quad \beta''_1 = 1 - (V^2/2\delta^2). \end{aligned} \quad (20)$$

At first glance it seems strange that the quasienergy does not tend to the energy of the unperturbed upper state B as $V \rightarrow 0$. But we see here precisely a manifestation of the fact that F is defined in modulo ω . In the second solution, the principal term at $V \rightarrow 0$ is $\psi'' = \beta''_1 e^{-i(F+\omega)t} \chi - \chi e^{iBt}$, as expected.

At $V \sim \delta$, the coefficients α_0 and β_1 are of the same order; in particular, at $V \gg \delta$ we have

$$\begin{aligned} F' &= A + V, \quad \psi' = e^{-i(A+V)t} (\varphi + e^{-i\omega t} \chi), \\ F'' &= A - V, \quad \psi'' = e^{-i(B-V)t} (\varphi e^{i\omega t} - \chi). \end{aligned} \quad (21)$$

In a typical "resonant" case, the ratio of V and δ can be arbitrary, $V \gtrsim \delta$, but both V and δ are small in comparison with $B - A$. It is then easy to verify that the discarded terms of the series ($\alpha_{\pm 2, \pm 4}, \beta_{-1, \pm 3}, \dots$) constitute an expansion in powers of the perturbation, for example,

$$\alpha'_{\pm 2} \sim V^2/(B-A)^2; \quad \beta'_{-1}, \beta'_{\pm 3} \sim V^3/(B-A)^3; \quad \alpha''_{\pm 4} \sim V^4/(B-A)^4 \text{ etc.}$$

At fixed V/δ , we can also speak of expansion in powers of the "degree of resonance" δ/ω . The denominators of the expressions for the higher-order terms will contain $B - A + \omega, B - A + 2\omega, \dots$

The iteration and the entire quasienergy approach are good in the case when the natural width of the upper stationary state γ_0 is small (the probability of the spontaneous transition $B \rightarrow A$ is small): $\gamma_0 = W_{BA} \ll B - A$; then there is a region

$$B - A \sim \omega \gg \delta \gg \gamma_0.$$

Even the first approximation described above (with α_0 and β_1 retained) contains nontrivial results. Each of the two states F' and F'' has a variable dipole moment proportional to $\alpha_0 \beta_1'$ and $\alpha_0'' \beta_1''$, respectively, varying with time at the frequency ω . When $V < \delta$, the coherent radiation is proportional to V^2/δ^2 , corresponding to a scattering cross section that does not depend on the amplitude and depends in Lorentz fashion on the difference $\delta = \omega - (B - A) = \omega - \omega_0$. When $V > \delta$, saturation sets in: the radiation tends to a constant limit, by virtue of which the cross section decreases like V^{-2} . Thus, V plays the role of the width in the shape of the scattering resonance; the stronger the wave, the larger the frequency interval in which the cross section has a plateau.

Let us dwell briefly on the history of the problem. By the method of adiabatic perturbation theory, without explicitly using the concepts of quasienergy and QES, the two-level system was considered in a paper by Poplar^[9]. In terms of the quasienergy, the question was considered in^[10], where both scattering and harmonics were considered. For an account of the quantized character of the electromagnetic field in this case, see^[11].

5. SPONTANEOUS TRANSITIONS

We turn now to spontaneous transitions. The probability of the transition $F'' \rightarrow F'$ is proportional to $|\alpha_0' \beta_1''|^2$. As $V \rightarrow 0$ we obtain the maximum probability, equal to $\gamma_0 = W'_{BA}$, of the unperturbed system. The probability of the inverse "unnatural" transition $F' \rightarrow F''$ is proportional to $|\beta_1' \alpha_0''|^2 \sim V^4/\delta^4$ when $V < \delta$. The radiated frequencies are

$$\begin{aligned} \omega''' &= F'' - F' + \omega = B - A - (2V^2/\delta) = \omega - \delta - (2V^2/\delta), \\ \omega'' &= F' - F'' + \omega = 2\omega - (B - A) + (2V^2/\delta) = \omega + \delta + (2V^2/\delta). \end{aligned} \quad (22)$$

The complete cycle $F' \rightarrow F'' \rightarrow F'$ reduces to the radiation of ω'' and ω''' with selection of two quanta of energy 2ω from the classical field of the wave. We note that the width of the spectral line of the spontaneous transitions does not become larger than γ_0 for any value of V , regardless of the broadening of the coherent-scattering resonance. When it comes to spontaneous emission, it is the line shift and not the line width that depends on the wave amplitude (on V).

In the stationary state, the ratio of the number of atoms in F'' to the number of F' is the inverse of the ratio of the probabilities. At $V < \delta$ we have $n''/n' \sim V^4/\delta^4$. When $V > \delta$, however, n' and n'' become comparable. These two states makes contributions of opposite sign to the real part of the forward scattering amplitude, i.e., to the refractive index. Therefore with increasing V the refractive index decreases like V^{-4} , i.e., more rapidly than the scattering cross section.

An examination of the next terms of the expansion $\alpha_{\pm 2, \pm 4}, \beta_{-1, \pm 3}, \dots$ makes it possible to calculate the coherent emission of the harmonics by each of the states. It is easy to verify here that only odd harmonics, 3ω and 5ω , proportional to V^6 and V^{10} at small V are produced. In addition, it is possible to find the probability

of the spontaneous emission $\pm(F'' - F') + n\omega$ with large n in transitions from one QES to another.

The system of equations written out above can also be used to determine the probability of overtone excitation. Let us specify, for example, $3\omega = B - A - \delta$ with $\delta \ll \omega$; we then obtain a resonant solution with large $\alpha_0, \beta_1, \alpha_2$, and β_3 , and with a resonance condition $\delta \sim V^3/\omega^2$.

It is probable that the method developed above will also be useful for multilevel atomic-molecular systems. If the unperturbed system has a continuous spectrum (in addition to a discrete one), the quasienergy becomes complex, and this describes multiphoton ionization. Finally, in addition to spontaneous emission at frequencies different from ω , we can also consider the induced process, i.e., generation at frequencies ω' and ω'' such that $\omega' + \omega'' = n\omega$.

6. QUASIENERGY AND LINEAR STARK EFFECT

Kovarskiĭ and Perel'man^[10] applied the quasienergy concept to a consideration of excited states of hydrogen. The "random" degeneracy, for example of 2S and 2P levels, makes the excited states of the hydrogen atom with $n \geq 2$ particularly sensitive to the action of an electric field⁹⁾. As is well known, it is precisely in such atoms that the linear Stark effect takes place. From the levels 2S and 2P with $m = 0$ we can construct linear combination with definite values of the dipole moment $\varphi_+ = (2S + 2P)/\sqrt{2}$ and $\varphi_- = (2S - 2P)/\sqrt{2}$. In this basis, the electric field yields diagonal matrix elements; in other words, the field does not cause transitions from φ_+ to φ_- or from φ_- to φ_+ . The QES wave functions are therefore particularly simple in this case:

$$\begin{aligned} \psi_+(x, t) &= (1/\sqrt{2}) e^{i\varphi_+ \cos \omega t} (2S + 2P), \\ \psi_-(x, t) &= (1/\sqrt{2}) e^{-i\varphi_- \cos \omega t} (2S - 2P). \end{aligned} \quad (23)$$

The expansion of the function $\exp(i\mu \cos \omega t)$ in a Fourier series is universally known, namely, the coefficient of $\cos(n\omega t)$ is the Bessel function $J_n(\mu)$. We consider now the absorption spectrum of a normal hydrogen atom near the line Ly_α , i.e., the transition $1S \rightarrow (2S, 2P)$. If the atoms are in the field of a strong wave, the Ly_α line splits and satellites $\omega_n = \omega_0 \pm n\omega$ appear, where ω_0 is the frequency of the unperturbed Ly_α line. The amplitude of the satellite depends on the amplitude of the wave in which the atoms are situated, like $J_n^2(\mu)$, where $\mu = eEa_0 \cdot \sqrt{3}/\omega$, E is the field of the wave, and $ea_0 \cdot \sqrt{3}$ is the dipole moment of the hydrogen atom in the state $2S \pm 2P$. It is curious, therefore, that the amplitude of the satellite depends periodically on the amplitude of the wave. The higher the number n of the satellite, the larger the field necessary to obtain the maximum amplitude of the given satellite (the functions J_n behave in this manner).

This example demonstrates not only the strength but also the difficulty of the quasienergy approach. The point is that two QES are degenerate here for precisely the same reason that made it so much easier to find these states. Therefore an exact analysis of the absorption probability in the vicinity of Ly_α as a function of the angle, of the polarization, and of the satellite number calls for allowance for the phase relations between two QES. This analysis is beyond the scope of the present article.

The problem of the excitation of the atom was considered earlier^[1, 13] without the use of the quasienergy

concept. It is of interest to consider also the rotational levels of a dipolar molecule of the HCl type or an atomic system with nonzero angular momentum, but without random degeneracy. At $l \neq 0$ there are $2l + 1$ sublevels. From this we can make up linear combinations with a definite quadrupole moment that interacts with the gradient of the electric field of the strong wave. On the other hand, in $l \rightarrow l'$ transitions with different parity, there is a dipole matrix element, and when the strong-wave frequency is close to resonance, the sublevels of two systems, l and l' , are mixed in the QES. The rules for mixing (selection) depend on the polarization of the strong wave, for example $m' = m \pm 1$ for plane polarization and $m' = m + 1$ for circular polarization¹⁰⁾.

In the case of a diatomic amplitude with constant moment of inertia, there are whole-number relations between the energies of the successive levels, and the resonance for one transition ($0 - 1$) coincides with the resonance of the higher orders for the next transitions, accurate to within the centrifugal deformation of the molecule.

The situation with a magnetic dipole in a magnetic field, where the equidistance and the final number of levels combine, was considered in detail in the theory of magnetic resonance. Even this short list demonstrates the large size of the class of phenomena to which the quasienergy theory can be applied.

7. NUMERICAL ESTIMATES

At what value of the field and at what power of the laser beam do the characteristic phenomena described above become manifest? Phenomena in weak fields can be described with the aid of quasienergy, but they are also successfully described by stationary perturbation theory—a circumstance noted on the very first page of this review. The advantages of the quasienergy approach appear in fields such that the first nonvanishing approximation of perturbation theory is adequate. Let us estimate the required field.

We start with the simplest case (see Sec. 6 above and^[10]), of the system of ($2S - 2P$) levels of the hydrogen atom. In this case a) the unperturbed Hamiltonian \mathcal{H}_0 is degenerate, b) the perturbation is factorized, $\mathcal{H}' = V(x) \cos \omega t$, c) it is possible to select the eigenstates of \mathcal{H}_0 and $V(x)$ simultaneously, d) the QES are also factorized as a result, i.e., they take the form $\varphi(x)f(t)$ where, however, $f(t)$ is not a harmonic function:

$$f \sim \exp(-i\gamma\omega \int \cos \omega t dt). \quad (24)$$

Obviously, the condition that the result be nontrivial is $\gamma \gtrsim 1$. The dimensionless criterion γ is equal in this case to

$$\gamma = M_{2S,2P} / \hbar\omega = \sqrt{3} ea_0 E / \hbar\omega, \quad (25)$$

where M is the matrix element of the transition in the field E , and a_0 is the Bohr radius ($a_0 = \hbar^2 / me^2$).

The condition $\gamma = 1$ gives the field amplitude $E = m\omega / h\sqrt{3}$. Let us find the corresponding power of an ideally focused laser beam. We specify the focal-spot area λ^2 , where λ is the wavelength; we obtain

$$W = E^2 \lambda^2 c / 8\pi = (\pi/6) mc^2 (mc^2 / \hbar) e^2 / \hbar c = 2 \cdot 10^{12} \text{ erg/sec} = 2 \cdot 10^5 \text{ W}.$$

This value can be regarded as rather modest in comparison with the presently attainable powers. It should be borne in mind, however, that it pertains to a focused

beam. Spectroscopic experiments call for a field that fills a sufficient volume. However, consideration of experimental devices is not the task of the present article.

If we are dealing with electron spin flip, then the matrix element of the interaction is equal to $M = \mu H$; it is smaller than in the preceding case by a factor $(\hbar c / e^2) \sqrt{3} \approx 240$. Accordingly, the required power is 6×10^4 times larger, $\sim 10^{10}$ W. Realization of experiments with this power is a difficult task.

Finally, a typical task is the excitation of a nondegenerate state by radiation close to resonance. In this case we cannot confine ourselves to the first term of the perturbation-theory series, when

$$\gamma' \sim M / \hbar |\omega - \omega_0| \sim 1. \quad (26)$$

For an allowed transition we have $M \sim ea_0 E$, and the condition $\gamma' = 1$ gives for a beam with cross-section area S a power value

$$W_S = 6 \cdot 10^5 (S/\lambda^2) |\omega - \omega_0| \omega^2 \text{ W}. \quad (27)$$

8. CONCLUDING REMARKS

It must be emphasized that the majority of the results were obtained earlier by other methods. We note below the corresponding investigations and the results. The quasienergy method developed above, however, seems to be the most adequate and economical even where the results are known. New ways of obtaining these results will be of methodological interest.

The task of the present methodological article is to demonstrate, with a very simple example, the method of operating with quasienergy and the benefits of this concept, especially in the simplest resonant case. The described simple methods can be useful in problems connected with astrophysical maser radiation, and in the theory of light transmission through rarefied media (see the note in^[15]). The author did not intend to review fully the entire literature. Concerning more complicated problems, we confine ourselves to indicating the references^[10, 13, 16, 17]. We shall discuss briefly only the principal question, namely, do states with definite real quasienergy always exist for a system situated in a periodic field?

Any action on the quantum mechanical system can be regarded as a unitary operator. We consider the action during one period. The quasienergy is the result of diagonalization of the operator of single-period action. In a finite-dimensional system (for example, in a two-level system), diagonalization is always possible. The unitarity of the operator ensures reality of the quasienergy. In an infinite-dimensional system, however, diagonalization is not always possible. Perelomov and Popov (see^[17]) have constructed an interesting example by considering a harmonic oscillator with frequency ω_0 and a perturbation of the type $Vx^2 \cos \omega t$. At values of ω close to parametric resonance ($\omega \approx 2\omega_0$), the energy of the oscillator increases without limit from any initial state, meaning that there are no QES that repeat with a period $2\pi/\omega$.

Thus, the very existence of quasienergies is not trivial. In real systems, the quasienergy concept is approximate; it exists to the extent to which one can neglect multiphoton ionization. But one need not fear the approximation once the character of the approximation is understood. It is appropriate to refer here to the re-

markable article by Fock^[18] concerning approximate solutions in physics.

LIST OF SYMBOLS

- $\hbar \equiv 1$ —Planck's constant;
 \mathcal{H}_0 —time-independent Hamiltonian;
 $\mathcal{H}(t)$ —Hamiltonian that depends periodically on time;
 $\mathcal{H}'(t)$ —time-dependent part of $\mathcal{H}(t)$, $\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}'(t)$;
 α —parameter describing the turning-on of the perturbation, $\mathcal{H} = \mathcal{H}_0 + \alpha\mathcal{H}'$, $0 \leq \alpha \leq 1$;
 T —period, $\mathcal{H}(t + T) \equiv \mathcal{H}(t)$;
 ω —circular frequency of $\mathcal{H}(t)$ and $\mathcal{H}'(t)$, so that $\omega = 2\pi/T$;
 E_k —energy of the k -th eigenvalue of \mathcal{H}_0 ;
 $\varphi_k(x)$ —corresponding eigenfunction, $\mathcal{H}_0\varphi_k = E_k\varphi_k$
 $\varphi_k(x)\exp(-iE_k t)$ —solution of Schrödinger equation for \mathcal{H}_0 ;
 F_k —quasienergy of k -th quasienergy state of $\mathcal{H}(t)$;
 $\psi_k(x, t) = \varphi_k(x, t)\exp(-iF_k t)$ —solution of the Schrödinger equation for $\mathcal{H}(t)$;
 $\varphi_k(x, t)$ —the corresponding eigenfunction, $\varphi_k(x, t + T) \equiv \varphi_k(x, t)$;
 $F_{kp} = F_k + p\omega$ —satellites of the quasienergy of the k -th state, p is an integer (positive, negative, or zero);
 $\omega' = \omega_{knr} = F_{kp} - F_{nq}$ —frequency of transition from k -th to n -th state of the quasienergy as a result of the satellites kp and nq , $p - q = r$;
 $\omega' = F_k - F_n + r\omega > 0$ —condition of spontaneous radiation, $\omega' > 0$;
 N_k —concentration of particles in the k -th state, cm^{-3} ;
 $N_k > N_n$ —inversion condition at the frequencies $\omega' = \omega_{knr}$ for all r such that $\omega' > 0$.

¹Reported at Conference on strong electromagnetic waves (Balaton, Hungary, September 1972).

²We call attention to the fact that $F_k T$ is a number in the exponential of (1) but is a linear function of the time in the exponential of (2).

³However, such a change in the definition of F_i simultaneously changes the corresponding function φ_i , which nevertheless remains periodic. On the other hand, we shall note below the possibility of obtaining QES by a smooth transition from a rigorous stationary state for a time-independent Hamiltonian \mathcal{H}_0 . In the latter case, the energy of the k -th state E_k is uniquely defined. During the course of a smooth transition one can see which of the values $F_k + n\omega$ goes over into E_k ; this value of F_k can naturally be called the principal value, and the remaining, which differ by an integer multiple of ω , can be called satellites.

⁴We note also the application of quasienergy in the analysis of an electron acted upon by two fields, a constant magnetic field and a wave field [8].

⁵The discussion that follows pertains to a situation in which the principal role is played by discrete states. In a continuous spectrum, particularly for a free electron, direct methods can be effective.

⁶In the case of instantaneous switching ($\alpha = 0$, $t < t_1$, $\alpha = 1$, $t > t_1$) we would have to expand $\varphi_k(x)$ in terms of $\varphi_n(x, t_1)$, and then the coeffi-

cients c_{nk} with $n \neq k$ are not equal to zero in principle.

⁷Not necessarily positive. It is also possible for one of them to vanish.

⁸Without taking the spontaneous transitions into account, this quantity will depend on the initial conditions and on the manner in which the wave is turned on.

⁹The influence of random degeneracy in the problem of multiphoton ionization of the atom was considered by Keldysh [12].

¹⁰The case of an arbitrary number of degenerate states was considered by Kovarskii and Perel'man [14].

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