Quantum chaos in interactions of multilevel quantum systems with a coherent radiation field

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Studies on the dynamics of multilevel quantum systems in a coherent radiation field are reviewed. Major attention is paid to phenomena of quantum nonlinear resonance and the interaction of two quantum nonlinear resonances, and also to the analysis of the real physical systems where these phenomena are realized.

INTRODUCTION

Before we proceed to the main part of this article, we considered it necessary from the outset to indicate the set of problems that will be discussed in the article. This is all the more necessary since the title of the article contains a combination of the words "quantum" and "chaos," which today can be interpreted very broadly. Actually, up to now the theory of quantum chaos has linked a great many varied problems that bear little relation to one another at first glance. This and the search for a formal criterion of quantum chaos and its connection with the characteristics of classical chaos, the study of the structure and statistical properties of the energy spectrum of systems with broken integrals of motion, the comparison of the quantum-mechanical and the classical descriptions of the dynamics of nonautonomous systems, the theory of scattering by complex objects, and much else. (A brief review of some fundamental problems entering into the group of problems of quantum chaos, together with a wide selection of references, is contained in the recent review of Ref. 1.) The original linking element of all the approaches is, first, choosing systems that manifest to some degree a chaotic dynamics in the classical approach; second, conducting the analysis in the quasiclassical region of parameters. We note that a chaotic regime of motion can be manifested by the overwhelming majority of classical dynamic systems. Therefore it will not be wrong if we give a definition of the theory of quantum chaos as the field of contemporary physics that studies quantum systems in the quasiclassical region of parameters.

In the initial stage of the establishment of the theory of quantum chaos the chief aspect of the studies was a purely theoretical question: what is chaos in quantum systems? (In a more general formulation, this is a question of the relationship between classical and quantum mechanics. This question has not lost its pertinence even today.) However, in the course of time, when an understanding of the fundamental features of "chaos" in quantum systems has been attained (at least Hamiltonian systems), the applied aspect has begun to advance to first level-what new features can the theory of quantum chaos bring to the traditional fields of physics? What phenomena are to be explained in terms of the theory created afresh. The heightened interest in the applied aspect is assisted in no small degree by the enhanced possibilities of experimental physics that enable one to study quantum objects in the region of large quantum numbers and at high intensities of external fields.

of quantum chaos to the problem of the behavior of a quantum object under the action of coherent radiation. Here the formulation of the problem looks no more complex than the formulation of the problem of the behavior of a two- or three-level system under the action of a periodic perturbation: there is a system of levels and we are interested in the dynamics of the populations of the levels and the behavior of various observable quantities (e.g., the polarization), etc. However, rejection of the finite-level approximation (which is a necessary requirement in the analysis of the system in the quasiclassical region of parameters) enables one to study the dynamics more completely and to discover the specific regimes of motion, which, on the one hand, are analogous to the dynamics of a classical system, and on the other hand, have substantial differences owing to the quantum nature of the object.

This review is based on studies of authors on the stated theme in the past decade. Here one of our aims was to develop as much as possible a universal approach to analyzing systems in the quasiclassical region of parameters. Such a universal approach to analyzing systems in an external periodic field exists in classical mechanics and consists of three fundamental stages: transformation to "action-angle" variables, singling out of the nonlinear resonances of the system with the external field, and then taking account of the influence of the resonances on one another. Evidently, in the quantum analysis it is rational to follow this scheme, and the structure of the study reflects it. We note also that in classical mechanics one can distinguish two limiting cases: the existence in the system of a small number of resonances (in the limit only two), and of a large number, in the limit an infinite number of resonances. Along with their common features, these two limiting cases also have substantial differences. In particular, in the latter case the phenomenon of diffusion occurs, which is absent in the former case. A quantum analysis of the phenomenon of diffusion is contained in the review of Ref. 2. Running ahead, we note that these two limiting cases strongly differ also in the quantum approach.

In writing this work, we have put foremost consistency and integrity of presentation. For this reason we have tried to restrict the treatment to as few references as possible, although this involved a certain increase in the size of the review. We address the review to researchers working in the field of quantum optics and radiophysics. Therefore our fundamental aim in this study is the quantum analysis of concrete physical objects, while the question of the correspondence of the classical and quantum descriptions (as was already noted above, this aspect is necessarily present in

One can treat this study as the application of the theory

studies devoted to quantum chaos) is put on the second level.

Taking the occasion, we express our thanks to G. M. Zaslavskiĭ, together with whom we have obtained a large number of the results presented in the review, and also to B. V. Chirikov for constant attention to the work, for useful discussions, and critical remarks.

1. AN ISOLATED NONLINEAR RESONANCE

1.1. Classical nonlinear resonance

The concept of nonlinear resonance plays a fundamental role in modern classical mechanics.⁴⁻⁵ Rather than referring the reader to the cited books, we shall examine the phenomenon of nonlinear resonance using the very simple example of a plane rotator situated in the field of circularly polarized radiation

$$H = (\gamma I^2/2) + V \cos(\theta - \omega t). \tag{1.1}$$

In (1.1) *I* is the angular momentum, γ^{-1} is the moment of inertia, θ is the variable canonically conjugate to *I*, ω is the frequency of the external field, and $V \cos(\theta - \omega t)$ is the energy of interaction of the rotator with the external field. If we understand the rotator to be a two-dimensional model of a heteropolar molecule, then we have V = -Ed (*E* is the amplitude of the external field, and *d* is the dipole moment of the molecule), and $\theta - \omega t$ is the angle between the vectors of the dipole moment and the external field.

The free movement of the rotator amounts to rotation at the frequency $\Omega = \gamma I$. If the frequency of rotation of the rotator coincides approximately with that of the external field, then the rotator becomes entrained into nonlinear resonance, and its angular momentum I undergoes slow nutations. We can easily describe these slow motions if we transform to the variables $\Delta I = I - I^*$, where I^* is the center of the resonance, and $\Theta = \theta - \omega t$. As we can easily derive from (1.1), the variation of the variables ΔI and Θ is described by the effective Hamiltonian

$$H_{\rm eff} = [\gamma(\Delta I)^2/2] + V \cos \Theta. \tag{1.2}$$

The Hamiltonian (1.2) is commonly called the nonlinearresonance Hamiltonian. We note that formally the Hamiltonian of (1.2) coincides with the Hamiltonian of a mathematical pendulum in the field of the force of gravity. Figure 1 shows the phase portrait of the system of (1.2). The period of the motion of a point on the trajectory T (i.e., the period of nutation of the plane rotator) depends on the initial conditions. For the region close to the center of the resonance $(\Delta I \approx 0, \Theta = \pi)$, we can expand $\cos \Theta$ in (1.2) in a power



FIG. 1. Phase portrait of the system of Eq. (1.2) ($\gamma = 1$, V = 1).



FIG. 2. Eigenfrequency of a nonlinear resonance; $\Omega_{\rm ph} = (\gamma V)^{1/2}$, $J^{\bullet} = (8/\pi) (V/\tau)^{1/2}$.

series and $T \approx 2\pi/(\gamma V)^{1/2}$. The corresponding frequency is one of the two fundamental characteristics of the nonlinear resonance, and henceforth we shall call it the phase frequency and denote it by $\Omega_{\rm ph}$:

$$\Omega_{\rm ph} = (\gamma V)^{1/2}. \tag{1.3}$$

Figure 2 shows the dependence of the frequency of motion along the trajectory as a function of the variable $J = (1/2\pi) \oint \Delta I d\Theta$, which has the dimensions of action and is a characteristic of the trajectory in phase space. (Geometrically J is proportional to the area bounded by the trajectory.) As we approach the separatrix, which corresponds to the value $J^* = (8/\pi) (V/\gamma)^{1/2}$, $\Omega(J)$ approaches zero logarithmically. We note also that the second branch in Fig. 2 $(J > J^*)$ is doubly degenerate, which corresponds to the possibility of rotation of the pendulum in different directions.

Along with Ω_{ph} , another fundamental characteristic of the nonlinear resonance is its width

$$\delta I = 4(V/\gamma)^{1/2}.$$
 (1.4)

This corresponds to the dimension in terms of action of the region bounded by the separatrix. As applied to our example, δI reflects the maximum amplitude of nutation of the rotator. We note that, on passing through the separatrix, the amplitude of the nutations declines jumpwise by a factor of two.

1.2. Examples of isolated nonlinear resonance in different systems

We considered it expedient to present two more examples of nonlinear resonance to show its universal nature. In addition, the preceding example does not reflect the typical situation, since the original Hamiltonian is directly written in angular variables. As the second example let us examine the behavior of an electron in a circular orbit in a classical hydrogen atom under the action of a circularly polarized UHF field. In this example we found it necessary to transform preliminarily to action-angle variables. In action-angle variables the unperturbed Hamiltonian of the system has the form $H_0 = -e^4 m/2h^2 n^2 = -R/2n^2$ (*h* denotes Planck's constant \hbar). Here *n* is the dimensionless action (n = I/h), and m and e are the mass and charge of the electron, while R is Rydberg's constant. The position vector of the electron in the circular orbit is $r = (r(n)\cos\Theta)$, $r(n)\sin\Theta$ (as before we consider the two-dimensional case for simplicity). Here $r(n) = \alpha n^2$ is the distance from the nucleus to the electron $(\alpha = h^2/me^2)$, $\theta = \Omega(n)t$, and $\Omega(n) = R / n^3$ is the frequency of revolution of the electron. Then the Hamiltonian of the atom in the field of circularly



polarized radiation has the form

$$H = -(R/2n^2) - eEr(n)\cos(\theta - \omega t).$$

We shall find the center of the nonlinear resonance n^* from the condition $\Omega(n^*) = \omega$. Let us restrict the treatment to the case $n^* \ge 1$ (Rydberg atom). Let us expand $H_0(n)$ up to quadratic terms in the neighborhood of n^* , and keep only the zero-order term in the perturbation. Upon transforming to the variables $\Delta I = h(n - n^*)$, $\Theta = \theta - \omega t$, we arrive at the Hamiltonian (1.2), where $\gamma = -3R/g^2(n^*)^4$ and $V = -eE\alpha(n^*)^2$. When the condition $\delta I/hn^* \ll 1$ is satisfied, the role of the dropped terms is reduced to insignificant distortions of the phase curves in Fig. 1 and the $\Omega(J)$ relationship in Fig. 2. Yet if the latter condition is not satisfied, the nonlinear resonance has a somewhat more complex structure.

As the last example we shall examine a weakly nonlinear oscillator (a model of a vibrational degree of freedom of a molecule) under the action of the periodic perturbation

 $H = \Omega I - (\gamma I^2/2) + W I^{1/2} \cos \theta \cdot \cos \omega t .$

(Here we have $\omega \sim \Omega$, and we assume that *I* is not too large, $\gamma I \ll \Omega$). Using the resonance approximation, we transform the Hamiltonian to the form

 $H_{\rm eff} = (\Omega - \omega)I - (\gamma I^2/2) + (W/2)I^{1/2}\cos\Theta.$

Figure 3 qualitatively shows the phase portrait of this system. We see that the structure of the resonance is rather



FIG. 4. Dynamics of the populations of the levels of the system of Eq. (1.6). Values of the parameters: $\gamma = 1$, V = 1. $\omega = 0.1$, h = 1/5 ($\Omega_{\rm ph} = 1$).

FIG. 3. Structure of the nonlinear resonance in a molecule for different values of the detuning $\Omega - \omega$ ($W \sim \gamma$, $\Delta \approx 0$ (a), $\gamma/2$ (b), γ (c), and $3\gamma/2$ (d)).

complex and depends on the value of the detuning $\Delta = \Omega - \omega$. When the condition $\delta I > I^*$ is fulfilled (Fig. 3d), the structure of the resonance practically coincides with that shown in Fig. 1.

1.3. Quantum nonlinear resonance (QNR)

Let us proceed to study QNR, the concept of which was introduced in Refs. 6-9. Analogously to what we did in Sec. 1.1, we shall examine the phenomenon of QNR using the concrete example of the system of (1.1). The Schrödinger equation for a plane rotator has the form

$$i\hbar\partial\psi(\theta, t)/\partial\theta = H\psi(\theta, t),$$
 (1.5)

$$\hat{H} = -(\gamma h^2/2)\partial^2/\partial\theta^2 + V\cos(\theta - \omega t).$$
(1.6)

In the absence of a perturbation the system possesses a complete set of eigenfunctions $|n\rangle = (2\pi)^{-1/2} \exp(in\theta)$, which corresponds to free rotation of the rotator in one (positive *n*) or the other (negative *n*) direction. The external field causes transitions between adjacent states of the rotator, and one can ask the question of the dynamics of the populations of the levels $|c_n(t)|^2(\psi(0,t) = \Sigma c_n(t)|n\rangle)$.

Figure 4 shows the initial stage of the evolution of the system of (1.6), where as the initial condition we have chosen the population of the single level n^* whose transition frequency to the above-lying level is in resonance with the external field: $E_{n^*-1} - E_{n^*} = h\omega$, $E_n = \gamma h^2/2$. After the characteristic time $T = \pi/2\Omega_{\rm ph} \sim (V\gamma)^{-1/2}$, a certain finite number of levels becomes populated, and further transitions occur only inside this group of levels. The number of levels involved in the dynamics of the transitions depends on the location of the initially populated level with respect to the level n^* (the center of the resonance). Figure 5 shows the



FIG. 5. Mean population (over the period of the phase oscillations) of the levels of the systems of Eq. (1.6). The dots indicate the location of the initially populated level. The values of the parameters are the same as in Fig. 4. (Diagram from Ref. 10).

time-averaged population of the levels for different initial conditions (the location of the initially populated level is denoted by the dot). We see that, as we go from case (b) to (c), a "transition across the separatrix" occurs. The given numerical calculation illustrates one of the possibilities of "experimental" determination of the width of a QNR. Here, in the light of the correspondence principle, the width is determined by the formula

$$\delta n = \delta I/h = 4(V/\gamma)^{1/2}/h.$$
 (1.7)

Here δI is the width of the classical nonlinear resonance of (1.4). The given parameter plays an important role, and as will be shown below, determines the degree of classical character of the QNR. The greater the value of the parameter δn , the closer the dynamics of the QNR is to the dynamics of its classical analog. Conversely, as $\delta n \rightarrow 0$, the QNR degenerates into a two-level system, which has a specifically quantum dynamics.

Besides the dynamics of the populations, the question is important of the dynamics of the various observables. Below we shall mainly treat the dynamics of the induced polarization. The choice of this quantity is due to the fact that the Fourier spectrum of the induced polarization determines the emission spectrum of the system in the field of the electromagnetic wave. As the two mutually perpendicular components of the induced polarization we have

$$P_{x} = \langle \psi(\theta, t) | \cos \theta | \psi(\theta, t) \rangle, \quad P_{y} = \langle \psi(\theta, t) | \sin \theta | \psi(\theta, t) \rangle.$$
(1.8)

Let us make the following substitution in (1.5) and (1.6):

$$\psi(\theta, t) = \exp(-iE_n t/h)\phi(\Theta, t)\exp(in^{\bullet}\Theta), \ \Theta = \theta - \omega t. \ (1.9)$$

(This substitution is equivalent to transforming to the variables ΔI and Θ in the classical analysis.) Then for the function $\phi(\Theta,t)$ we have $ih\partial\phi/\partial t = \hat{H}_{eff}\phi$, where

$$\widehat{H}_{eff} = -(\gamma h^2/2)\partial^2/\partial\Theta^2 + V\cos\Theta - ih(\omega - \gamma hn^*)\partial/\partial\Theta .$$
(1.10)

Here we can neglect the last term, since according to the condition we have $\gamma hn^* \approx \omega$. Below we shall call the Hamiltonian of (1.10) the Hamiltonian of the QNR. In the new variables the formulas of (1.8) acquire the form

$$\begin{split} P_x &= p_c(t)\cos\omega t - p_s(t)\sin\omega t, \quad p_y = p_s(t)\cos\omega t + p_c(t)\sin\omega t, \\ p_c(t) &= \langle \phi(\Theta, t) | \cos\Theta | \phi(\Theta, t) \rangle, \\ p_s(t) &= \langle \phi(\Theta, t) | \sin\Theta | \phi(\Theta, t) \rangle. \end{split}$$

Figure 6 shows the evolution of $p_c(t)$ in the case of initial population of the single level n^* (i.e., $\phi(\Theta, 0) = (2\pi)^{-1/2}$, and $p_s(t)$ in this case equals zero). We see that, in the course of two or three periods of the phase frequency, $p_c(t)$ reaches the steady-state value $p_c(t) \approx -0.3$. We note that this value is not associated with the frequency and the amplitude of the external field, and as will be shown in the next section, is only a function of the initial state of the system.

In closing this section, let us consider the influence of detuning. In contrast to a two-level system, for QNR a variation of the external frequency does not lead to an appreciable change in the regime of dynamics of the QNR nor to a change in its fundamental parameters— Ω_{ph} and δn . Actually the condition of exact resonance has the form



FIG. 6. Dynamics of $p_c(t) = (\cos \Theta)$ for a QNR. Values of the parameters: $\gamma = 1$, V = 1, $\omega = 0$; at the initial instant of time a single level with $n^{\bullet} = 0$ was populated. The values of Planck's constant are: h = 1/8, 1/16, 1/32, and 1/64. The corresponding values of the quasiclassicity parameter are indicated in the diagram.

 $E_{n^{\bullet}+1} - E_{n^{\bullet}} = \gamma h(n^{\bullet} + 1/2) = \omega$. Hence we see that a change in the frequency by an amount greater than γh leads simply to a shift of the center of the resonance to another pair of levels. Variation of the frequency ω within the bounds of the nonlinearity γh is manifested only in some "subtle" effects at times substantially exceeding the characteristic time of the QNR ($\sim 1/\Omega_{\rm ph}$). Thus the concept of detuning is not very constructive, and is replaced by the concept "center of the QNR."

1.4. Correspondence times of classical and quantum dynamics

Usually in comparing classical and quantum dynamics one compares the trajectory of a classical particle and the trajectory of the center of a wave packet having well defined values of the variables. In the case of the system of (1.10) one can also construct a wave packet with well defined values of the action and the phase

$$\phi(\Theta, 0) = A \sum_{n} \exp[-2(n - n_0)^2 / \delta n] \exp[in(\Theta - \Theta_0)],$$

$$n_0 = I_0 / h. \qquad (1.12)$$

Here the center of the wave packet is given by the values of n_0 and Θ_0 , while the uncertainties in action and phase are, respectively, equal to $\langle \delta I^2 \rangle^{1/2} \sim h(\delta n)^{1/2} \propto h^{1/2}$, and $\langle \delta \Theta^2 \rangle \sim (\delta n)^{-1/2} \propto h^{1/2}$ (δn is defined in (1.7). Figure 7 shows the variation of the mean value $\langle \cos \Theta \rangle$ for different



FIG. 7. Dynamics of the quantum average $(\cos \Theta)$ in the case of an initial condition in the form of the wave packet of Eq. (1.12) with h = 1/128 and the dynamics of a classical particle (h = 0). The values are: $I_0 = 0$, $\Theta_0 = 1.57$; the values of the rest of the parameters are as before.

values of δn . We see that, with increase in the quasiclassicality parameter δn , the dynamics of the quantum averages tracks the dynamics of the classical particle for a longer time. We can show analytically that the correspondence time increases as $\delta n^{1/2}$

However, this simplified concept of the correspondence time, as we see it, is extremely incomplete. The point is that quantum mechanics is fundamentally probabilistic in character; for this reason one must compare the behavior of the quantum-mechanical average with the behavior of the average over an ensemble of classical particles. Here the classical ensemble is determined by the choice of the initial state of the quantum system. There are two limiting cases, which correspond to a choice of the initial state of the type of (1.12) and

$$\phi(\Theta, 0) = (2\pi)^{-1/2} \exp(in\Theta)_{\bullet}$$
(1.13)

(The value of the action is strictly determined, and the phase is completely indeterminate.) In the former case we must compare with the quantum packet a Gaussian packet of classical particles having a width of the order of $h^{1/2}$. In the latter case the classical ensemble amounts to an ensemble of particles uniformly distributed in phase, with the value of the action I = hn. From the standpoint of physical applications, the latter case is substantially more important, since it corresponds to population at the initial instant of time of a single level, which is easy to effect in a real experiment.

Figure 8 shows the dependence of the average value $\langle \langle \cos \Theta \rangle \rangle$ over an ensemble of classical particles uniformly distributed in phase at the initial instant of time with



FIG. 8. Dynamics of the average $p_c^{cl}(t) = \langle \langle \cos \Theta \rangle \rangle$ for the system of Eq. (1.2) over an ensemble of classical particles uniformly distributed in phase at the initial instant of time with $I_0 = 0$. The parameters are as before. (From Ref. 6).

I(0) = 0. The initial period of oscillations is determined by the frequency Ω_{ph} of (1.3). Subsequently, owing to the dependence of the frequency on the initial position of the particle, a "dephasing" of the motion of the individual particles occurs, and the value of $\langle \langle \cos \Theta \rangle \rangle$ reaches the steady-state value ≈ -0.3 . Graphs of the quantum-mechanical averages $\langle \phi(\Theta,t) | \cos \Theta | \phi(\Theta,t) \rangle$ for $\phi(\Theta,0) = (2\pi)^{-1/2}$ were shown in Fig. 6. Comparison of Fig. 8 with Fig. 6 shows that the curves rapidly converge to their classical limit with increase in the parameter δn . In the scheme of the numerical experiment described above, the fundamental question consists not in determining the time of strict correspondence in the behavior of the classical and quantum averages, but in determining the mean level of "quantum fluctuations." The question of the law according to which the level of quantum fluctuations declines with decreasing h (or, as is equivalent, with increase in the parameter δn) deserves exceptional attention (since a knowledge of the given relationship would enable one quantitatively to indicate the degree of applicability of the classical approach) and has been little studied up to the present. We only note that (as will be shown in Sec. 2.7) this law is not universal and depends on the type of motion (regular or chaotic) of the classical system.

1.5. Spectral properties of quantum nonlinear resonances

Let us proceed to discuss the spectral properties of QNRs. In studying the behavior of quantum systems under the action of a periodic perturbation, the fundamental role is played by the concept of the quasienergy functions (QFs) and the spectrum of quasienergies of the system, the quasienergy spectrum (QS). By definition the eigen-QF is the eigenfunction of the operator of evolution of the system in the period of the external field and has the form

$$\psi(\theta, t) = \exp(-i\lambda_k t/h)\psi_k(\theta, t). \tag{1.14}$$

Here we have $\psi_k(\theta, t + T) = \psi_k(\theta, t)$ (*T* is the period of the external field), and λ_k is the value of the quasienergy. We note that, as is implied by (1.14), the quasienergies are determined apart from a phase coefficient $\exp(il\omega t)$; *l* is an integer). The QFs form a complete basis, and the solution of the Schrödinger equation can be represented in the form of a linear combination of QFs:

$$\psi(\theta, t) = \sum_{k} \alpha_{k} \exp(-i\lambda_{k}t/h) \psi_{k}(\theta, t). \qquad (1.15)$$

Here α_k is determined from the initial conditions. The latter formula reflects the importance of the concept of the quasienergy states of the system. In essence, the quasienergy states determine the possible dynamics of the system and, if we know only the QS and the structure of the QFs, we can predict, at least qualitatively, the behavior of the different quantum-mechanical averages.

By using the substitution (1.9) one can easily show that, for the system being studied of a plane rotator, the QF is $\psi_k(\theta,t) \sim \phi_k(\theta - \omega t)$, where the $\phi_k(\Theta)$ are the eigenfunctions of the effective Hamiltonian of (1.10), while the eigenvalues of \hat{H}_{eff} determine the QS of the system. (We note here that formally the Hamiltonian of (1.10) with $\Delta = \gamma h n^* - \omega = 0$ coincides with the Hamiltonian of a quantum mathematical pendulum.) Figure 9 shows the QS of a system in the form of the eigenfrequencies (cf. Fig. 2). The value of $\Omega_k = (\lambda_{k+1} - \lambda_k)/h$ is plotted along the ver-



FIG. 9. Eigenfrequency of the QNR. The value of $\Omega_k = (\lambda_{k+1} - \lambda_k)/h$ is plotted along the *y* axis, where λ_k is the value of the quasienergy. Values of the parameters: V = 1, $\gamma = 1$, h = 1/20 ($\Omega_{\rm ph}$) = $(\gamma V)^{1/2}$, $k^* = (8/\pi h) (V/\gamma)^{1/2}$. (From Ref. 7.)

tical axis, and the number k of the quasienergy state along the horizontal axis. We see from the diagram that the spectrum of quasienergies has three branches. The first branch corresponds to a "vibrational regime" $(k < k^*)$, and the second and third $(k > k^*)$ correspond to a "rotational regime." Here we must make a slight remark on the "resonance" values of Δ . In particular, when $\Delta = 0$, owing to the symmetry properties of the Hamiltonian (1.10), its eigenfunctions are either even (even index k) or odd (odd index k) functions of Θ . Therefore, when $\Delta = 0$, the rotational states are a superposition of rotation in both directions. However, since the splitting of the levels is exponentially small, we can construct new functions

$$\widetilde{\varphi}_{k}(\Theta) = 2^{-1/2} (\phi_{2k}(\Theta) \pm i\phi_{2k+1}(\Theta)). \tag{1.16}$$

These have the asymptotic behavior $\tilde{\phi}_k(\Theta) = (2\pi)^{-1/2} \\ \times \exp(\pm ik\Theta)$ as $k \to \infty$ and are approximately eigenfunctions. This procedure is incorrect only for values of k close to $k^* = (3/\pi h) (V/\gamma)^{1/2}$. This region corresponds to the separatrix of the nonlinear resonance, where the difference of the classical from the quantum system is substantial. In particular, we see that the frequency of the quantum nonlinear resonance, in contrast to that of the classical resonance, does not take on a value of zero. States that belong to this region cannot be assigned to rotational nor to vibrational states. We can obtain an estimate for the width of this region (the number of quasienergy levels) (see the Appendix)

$$\Delta k \sim \ln \delta n. \tag{1.17}$$

Here δn is the width of the QNR.

Finally, we note another difference of the QNR from the classical nonlinear resonance, which involves the possibility of tunneling through the separatrix.⁷⁻⁹ This tunneling effect can be easily explained on the basis of Eq. (1.16). Let a group of levels below the separatrix of the QNR be populated at the initial instant of time (negative n, $|n| > \delta n/2$). Then, when t = 0, the expansion of the wave function is performed over the functions of (1.16) with negative k. Since the eigenvalue λ_{2k} of the function $\phi_{2k}(\Theta)$ does not coincide exactly with the eigenvalue of the function $\phi_{2k+1}(\Theta)$, a group of levels lying symmetrically (with respect to the center of the resonance) of the initially populated levels will become populated in the course of time. The characteristic time of this process is determined by the quantity $h/\Delta\lambda$, where $\Delta \lambda$ is the magnitude of the splitting of the levels λ_{2k} and λ_{2k-1} . This time greatly exceeds the characteristic time of the dynamics of the QNR ($\sim \Omega_{ph}^{-1}$). For this reason we shall in subsequent discussion set aside the tunneling effects as "second-order" effects.

1.6. The action-angle quantum-mechanical representation

In Sec. 1.3 we studied the QNR phenomenon using the example of a system whose Hamiltonian was directly written in terms of angle variables. If the original variables of the problem are not angles, then one must preliminarily transform to the action-angle quantum-mechanical representation.¹³ This representation is close to the energy representation, yet is more convenient for our purposes. Let us examine the transformation to the action-angle representation using the example of a one-dimensional system. Strictly speaking, the form of the representation depends on whether the spectrum of the system is semibounded (as, e.g., in the case of a linear oscillator, where the states are numbered by the index $n \ge 0$), or not $(-\infty < n < \infty)$. However, often in the case of a semibounded spectrum it is convenient to introduce fictional levels with a negative index n. An example of this approach is given in Ref. 14, which treats the behavior of a particle in a rectangular potential well under the action of an external periodic field. Therefore we shall consider below that the index n runs through values from $-\infty$ to ∞ .

The basis of the action-angle quantum-mechanical representation is two noncommuting operators \hat{I} and $\hat{\Lambda}$, which (in the energy representation) have the form

$$I_{n,m} = hn\delta_{n,m}, \quad \Lambda_{n,m} = \delta_{n+1,m},$$

$$[\hat{I}, \hat{\Lambda}] = -h\hat{\Lambda}, \quad \hat{\Lambda}\hat{\Lambda}^{+} = \hat{\Lambda}^{+}\hat{\Lambda} = \hat{1}.$$
 (1.18)

Then we can represent any operator \widehat{A} in the form

$$\widehat{A} = \sum_{l=0}^{\infty} \left[\widehat{\Lambda}^{l} a_{l}(\widehat{I}) + \text{H.c.} \right], \qquad (1.19)$$

Here the $a_i(\hat{I})$ are operator functions of \hat{I} . If now we perform an isomorphism of the Hilbert space of the system onto the space of periodic functions $|n\rangle \leftrightarrow (2\pi)^{-1/2} \exp(in\theta)$, then the operator \hat{A} turns out to be written in angle variables. Here we have $\hat{I} = -i\hbar\partial/\partial\theta$, $\hat{\Lambda} = \exp(i\theta)$.

We can also show that in the quasiclassical approach the functions $a_i(I)$ are the coefficients of the expansion of the classical variable $A(I,\theta)$ in a Fourier series. The latter property is extremely convenient, since often the analysis of the problem is conducted in the quasiclassical region. In this case we simply replace the variable I in the classical Hamiltonian with the operator $\hat{I} = -i\hbar\partial/\partial\theta$. The error in this procedure will be of the order of h. As an example let us present the form of the Hamiltonian for a linear oscillator in the field of a periodic wave in the action-angle representation

$$H = h(\Omega(-i\partial/\partial\theta) + 1/2) + (W/2)\cos\omega t \cdot [\exp(-i\theta)(-ih\partial/\partial\theta)^{1/2} + c.c.].$$

We can easily see that in the region of large quantum numbers neither the 1/2 in the first term nor the sequence order of the operators in the second term plays a role, and the given Hamiltonian can be obtained by quantization of the classical Hamiltonian $H = \Omega I + W I^{1/2} \cos \theta \cdot \cos \omega t$.

In closing this section let us consider the phase (or angle) operator. The phase operator is defined as $\hat{\theta} = \theta$. How-

ever, this operator is not defined over the entire space of periodic functions, but only over a dense subset consisting of functions that take on a zero value for $\theta = 0$ or $\theta = 2\pi$ (Ref. 15) (we can easily see that in this subset $[\hat{\theta}, \hat{I}] = -i\hbar$). However, for us these mathematical subtleties are not essential, since the phase operator is not needed in real physical problems.

1.7. QNR in systems having no classical analog

In this section we wish to show that the concept of QNR can be substantially expanded, and this allows us to use the concept of QNR to analyze quantum systems having no classical analog. The fundamental idea consists in the following. As was already noted above, the dynamics of quantum systems in an external periodic field is determined by the quasienergy states of the system. Therefore systems having the same structure of the QS should behave similarly. The defining features of the QS of a QNP are the presence of two branches (one of which is doubly degenerate), separated by a "separatrix." Precisely this property determines all the features of the QNR.

As the first example let us examine a two-level system, which we shall associate with two vibrational (or electronic) levels of a molecule, under the action of a linearly polarized resonance field.¹⁶ If the molecule is fixed (i.e., has no possibility of rotating), then the QS is defined trivially and has the form

$$\lambda_{1,2} = (h\Delta\omega/2) \pm [(Ed/2)^2 \cos^2\theta + (h\Delta\omega/2)^2]^{1/2}.$$
 (1.20)

Here $\lambda_{1,2}$ are the two quasienergy levels, $\Delta \omega$ is the magnitude of the detuning from resonance, E is the amplitude of the external field, d is the value of the matrix element of the transition, and θ is the angle between the polarization vector of the field and the vector of the dipole moment of the transition. Yet if the molecule exists in free space, the structure of the QS is considerably more complex. In analyzing the problem we shall restrict the treatment to the two-dimensional case and the two-level approximation in the vibrational degree of freedom. In this case the wave function of the molecule can be represented in the form

$$\Psi(\theta, t) = \begin{pmatrix} \psi_2(\theta, t) \\ \psi_1(\theta, t) \end{pmatrix}, \quad \psi_\alpha(\theta, t) = (2\pi)^{-1/2} \sum_l c_{l,\alpha}(t) \exp(il\theta).$$
(1.21)

Here $c_{l,\alpha}(t)$ is the amplitude of the populations of the vibrational-rotational level corresponding to the energy $E = E_{\alpha} + Bl^2$ ($\alpha = 1,2$; and B is the rotational constant of the molecule). The Hamiltonian of the system has the form

$$\widehat{H} = \begin{pmatrix} E_2 & 0\\ 0 & E_1 \end{pmatrix} - B \frac{\partial^2}{\partial \theta^2} \begin{pmatrix} 1 & 0\\ 0 & 1 \end{pmatrix} - E \cos \omega t \begin{pmatrix} 0 & d\\ d & 0 \end{pmatrix} \cos \theta, \quad (1.22)$$

and one assumes that $(E_2 - E_1)/h - \omega = \Delta \omega \ll \omega$. (As an illustration Fig. 10a shows the structure of the eigenenergy spectrum of the system and the transitions caused by the external field.) Omitting the mathematical part (see Appendix II), we shall formulate only the end result. The eigenquasienergy states of the system have the form

$$\Psi_{k}(\theta, t) = \exp(-i\lambda_{k}t/h) \begin{pmatrix} \exp(-i\omega t) & 0\\ 0 & 1 \end{pmatrix} \begin{pmatrix} \phi_{2}^{(k)}(\theta)\\ \phi_{1}^{(k)}(\theta) \end{pmatrix} \quad (1.23)$$



FIG. 10. a—Structure of the energy levels and transitions of the system of Eq. (1.22). b—Structure of the transitions in a semiconductor in the field of a standing wave.

Here we find the values of the quasienergies λ_k from the equations for the eigenvalues of the following effective Hamiltonians:

$$H_{\text{eff}}^{(1,2)} = -B\partial^2/\partial\theta^2 + U_{1,2}(\theta), \qquad (1.24)$$

 $U_{1,2}(\theta) = h\Delta\omega/2 \pm [(Ed/2)^2 \cos^2\theta + (h\Delta\omega/2)^2]^{1/2}.$

The simplest cases are those of exact resonance

$$\hat{H}_{\text{eff}}^{(1,2)} = -B\partial^2/\partial\theta^2 \pm (Ed/2)\cos\theta \qquad (1.25)$$

and that of detuning large in comparison with the Rabi frequency

$$\hat{H}_{\text{eff}}^{(2)} = h\Delta\omega - B\partial^2/\partial\theta^2 + (E^2d^2/4h\Delta\omega)\cos^2\theta,$$

$$\hat{H}_{\text{eff}}^{(1)} = -B\partial^2/\partial\theta^2 - (E^2d^2/4h\Delta\omega)\cos^2\theta.$$
(1.26)

Below we shall return to discussing the dynamics of the system (1.22) in these two limiting cases. Here we shall only note that, as we see from the last formulas, the structure of the QS of a rotating two-level molecule resembles the structure of the QS of the QNR of (1.10). Thus we expect to see in the dynamics of the system (1.22) features characteristic of a QNR. To avoid misunderstanding, we note again that in this review we are treating only the case of a strong field, and Eq. (1.24) is presented precisely for this case. More concretely, the depth of the potential well $U_{1,2}(\theta)$ in (1.24) is assumed to be much larger than the rotational constant of the molecule (i.e., a large number of levels is involved in the dynamics of the transitions).

As the second example let us examine the QS of an electron in a semiconductor in the field of a standing electromagnetic wave¹⁷

$$\hat{H} = \hat{H}_0 - (e/mc)A(\mathbf{r}, t)\hat{\mathbf{p}}.$$

Here \hat{H}_0 determines the band structure of the semiconductor $\varepsilon_{\alpha}(\mathbf{k}), \mathbf{A}(\mathbf{r}, t)$ is the vector potential: $\mathbf{A} = (\mathbf{E}c/\omega)\cos(q\mathbf{r})\cos\omega t$. This problem is interesting also in that, despite the three-dimensional formulation, it reduces to one-dimensional without any additional approximations. Actually, let the wave propagate along the *x* axis with polarization of the vector \mathbf{E} along the *z* axis. Then we can easily convince ourselves that the external field "couples" states with the wave vectors $\mathbf{k} = (k_x, k_y, k_z)$ and $\mathbf{k}' = (k_x \pm q, k_y, k_z)$ (see Fig. 10b). Thus the values of the projection of the quasimomentum vector k_y and k_z do not vary and one can treat them as parameters of the problem. Correspondingly, k_x is denoted as k in Fig. 10b, while

 $\varepsilon_{\alpha}(k) = \varepsilon_{\alpha}(k,k_{y},k_{z})$. We see from the diagram that the structure of the transitions in the semiconductor is analogous to the structure of the transitions in the previous example. The role of the rotational constant is played by the quantity $\gamma = h^2 q^2 / 2m_{\alpha}$, where m_{α} is the effective mass of the electron in the band. Using the same mathematical method as in the previous example, we can show that the QS of the electron amounts to $2\pi/aq$ (*a* is the lattice period) of the subbands along the direction of k_x and has a structure similar to that of the QNR. In particular, in the case $-m_1 = m_2 = m^*$ with exact equality of the frequency of the external field to the energy gap, the effective Hamiltonian determining the location of the boundaries of the subbands has the form

$$\hat{H}_{\rm eff} = 4\gamma^2 (-i\partial/\partial\theta)^4 + (E^2 d^2/2) \cos^2\theta.$$

Here d denotes the magnitude of the matrix element of the transition between the bands $(d \le eh/m\omega a)$. For the classical analog of $\hat{H}_{\rm eff}$, $(-ih\partial/\partial\theta \rightarrow \hat{I})$ defines the concept of the separatrix, and hence one can introduce the concept of the width of the resonance. As the last formula implies, the width of the resonance in a semiconductor, just as in the case of an ordinary nonlinear resonance, is proportional to the square root of the energy of interaction of the system with the field.

1.8. QNR and the orientation of molecules by an infrared field

This section can be considered as an example of using the theory of QNR presented above in analyzing a concrete physical phenomenon: the orientation of molecules by an external high-frequency field. This phenomenon is widely known in liquids and dense gases, where relaxation processes are substantial. Below we shall show on the basis of the model of (1.22) that orientation is possible even in the absence of relaxation processes. In this case the macroscopic orientation of a molecular gas is a consequence of the nontrivial dynamics of the transitions, which can be described within the framework of QNR concepts.¹⁸

Let us examine the case of a detuning $|Ed/h\Delta\omega| = \varepsilon \ll 1$ that is large in comparison with the Rabi frequency. In this case the QS of the system is determined by the equation for the eigenvalues of the effective Hamiltonians of (1.26). The quasienergy eigenfunctions have the form (1.23), whereby (see Appendix I) the QFs corresponding to the Hamiltonian with index 1 have the property $\phi_1^{(k)}(\theta) \sim 1, \phi_2^{(k)}(\theta) \sim \varepsilon$. Conversely, for the QFs corresponding to the Hamiltonian with index 2, we have $\phi_2^{(k)}(\theta) \sim 1, \phi_1^{(k)}(\theta) \sim \varepsilon$. Let the system exist at the initial instant of time in the ground vibrational state, i.e., $\psi_2(\theta,0) = 0$. Then the expansion of the solution over the basis of the QFs (see Eq. (1.15)) occurs mainly over the QFs of the Hamiltonian I. Relying only on this fact, we can make the following statement: in the process of time evolution $\psi(\theta, t)$ is of the order of ε , i.e., the population of the upper vibrational level remains negligibly small; $\psi_1(\theta, t)$ can be represented as the sum of slowly and rapidly (with the characteristic frequency $\Delta \omega$) varying terms. Here in absolute magnitude the rapidly varying term is of the order of ε ; the slowly varying part $\psi_1(\theta, t)$ obeys the Schrödinger equation

$$i\hbar\partial\widetilde{\psi}_1(\theta, t)/\partial t = H_{\text{eff}}^{(1)}\widetilde{\psi}_1(\theta, t)$$
 (1.27)

with the Hamiltonian of (1.26); the induced polarization vector of the system is parallel to the external field vector and, apart from terms of the order of ε^2 , has the form

$$P(t) = -d(Ed/h\Delta\omega)\langle \tilde{\psi}_{1}(\theta, t) | \cos^{2}\theta | \tilde{\psi}_{1}(\theta, t) \rangle \cos \omega t$$

$$\equiv p(t)\cos \omega t. \qquad (1.28)$$

Evidently the factor $\langle \tilde{\psi}_1 | \cos^2 \theta | \tilde{\psi}_1 \rangle \equiv | \cos^2 \theta \rangle$ in (1.28) characterizes the mean orientation of the molecules. A uniform angular distribution of the molecules corresponds to the value $(\cos^2 \theta) = 1/2$. A value above 1/2 corresponds to predominant orientation of the molecules along the field, and below 1/2-transverse to the vector E. By the substitution $\theta = 2\Theta$ the Hamiltonian $\hat{H}_{eff}^{(1)}$ is transformed into the Hamiltonian of the nonlinear resonance, while the value of $(\cos^2 \theta)$ is determined by the value of the quantum-mechan-QNR $(\cos^2\theta)$ ical average $(\cos \Theta)$ for the $= (1 + \langle \cos \Theta \rangle)/2$. Since, in the process of evolution of the system of (1.10), $\langle \cos \Theta \rangle$ acquires values differing from zero (see Fig. 6), a predominant orientation of the molecules in the molecular gas will arise under the action of the infrared field that depends on the sign of the detuning $\Delta \omega$, either across or along the direction of polarization of the vector E.

An additional study was performed in Ref. 18 of the process of orientation in the case of a pulsed laser regime (in this case the amplitude of the field E in Eqs. (1.27) and (1.28) should be replaced by the envelope of E(t). It turned out that, when the condition $\tau^{-1} \gg \Omega_{\rm ph}$ is satisfied (τ is the duration of the pulse, and $\Omega_{\rm ph}$ is the phase frequency of the QNR), the orientation of the molecules occurs more efficiently. We note that, since the mean orientation of the molecules enters into the formula for the induced polarization, the orientation effect is reflected in the polarization properties of the medium. Figure 11 shows a graph of the normalized envelope $\tilde{p}(t) = p(t)/(d^2 E_{\text{max}}/h\Delta\omega)$ in the case of a Gaussian form of the pulse $E(t) = E_{\text{max}} \exp[-(t/\tau)^2]$ for two values of the detuning equal in magnitude and opposite in sign. We see from the diagram that the orientation effect is very significant, and can alter the polarization properties of the medium by an order of magnitude. In turn, the change in the polarization properties of the medium at the instant of passage of the electromagnetic field pulse through it will be reflected in various physical processes. In particular, Ref. 19



FIG. 11. Dynamics of the normalized envelope of the polarization. Values of the parameters: $B = 0.00125 \text{ cm}^{-1}$, energy of interaction with the field $(Ed)^2/4h\Delta\omega = \pm 2 \text{ cm}^{-1}$, pulse duration $\tau = 3 \times 10^{-10}$ s. The period of oscillation of the curve *I* is determined by the period of the phase oscillations. (From Ref. 13.)

experimentally studied the influence of the dynamic orientation of the molecules on the process of nonlinear-optical frequency shift in naphthalene vapor.

2. THE INTERACTION OF TWO NONLINEAR RESONANCES 2.1. Onset of chaos in the interaction of two classical nonlinear resonances

In Secs. 1.1 and 1.2 we examined the phenomenon of nonlinear resonance in classical systems. Here we deliberately chose as examples systems in which only one nonlinear resonance is possible. This situation is not typical. As a rule, in a nonlinear system under the action of an external field several nonlinear resonances arise together. Here the mutual influence of the resonances on one another leads to the appearance of regions of chaotic motion.³⁻⁵ We note that two nonlinear resonances already suffice for the appearance of a chaotic component.

As a very simple system in which the phenomenon of interaction of two nonlinear resonances takes place, let us study a plane rotator under the action of the linearly polarized field

$$H = (\gamma I^2/2) + 2V \cos\theta \cdot \cos\omega t.$$
(2.1)

Here, as before, we have 2V = -Ed, and θ is the angle between the field vector and the vector d. This model is a twodimensional idealization of a polar molecule in a UHF field. Upon resolving the linearly polarized field into two circularly polarized components $2V\cos\theta\cos$ $\times \omega t = V \times [\cos(\theta - \omega t) + \cos(\theta + \omega t)]$, we find that two nonlinear resonances occur in the system at the values $I_{1,2}^*$ $= \pm \omega/\gamma$ with the width δI determined by Eq. (1.4): $\Delta I = 4(V\gamma)^{1/2}$. The mutual influence of the resonances on one another is characterized by the Chirikov parameter of overlap of the nonlinear resonances, which is the ratio of the sum of the half-widths of the resonances to the distance between them:

$$K = \left[(\delta I_1/2) + (\delta I_2/2) \right] / \left| I_2^* - I_1^* \right| = 2(\gamma V)^{1/2} / \omega. \quad (2.2)$$

Figure 12 shows the Poincaré cross section for the system of (1.2) for different values of the parameter K. The cross section is constructed by numerical integration of the equations of motion over the period of the external field. We note that the system being studied with one and one-half degrees of freedom can be put into correspondence with a system with two degrees of freedom

$$H_{\text{eff}} = \omega J + (\gamma I^2/2) + 2V \cos \theta \cdot \cos \phi.$$
(2.3)

Here $\phi = \omega t$ is the phase of the field, and J is the canonically conjugate variable to ϕ . H_{eff} determines the quasienergy of the system, which is an integral of the motion. This situation has been used for monitoring the accuracy of calculation.

We see from Fig. 12 that for small values of K the chief consequence of the influence of the nonlinear resonances on one another is the appearance of secondary nonlinear resonances and thin stochastic layers in the vicinity of the separatrices of the nonlinear resonances. With increasing K the thickness of the stochastic layers increases, and at $K \approx 4$ already the entire central region of phase space is occupied by the stochastic component. If the initial conditions I(0), $\theta(0)$ belong to this region, then the trajectory of the system



FIG. 12. Structure of the phase space of the system of Eq. (2.1) for different values of the parameter K. The dots correspond to values of I(t), $\theta(t)$ for t = Tn; T is the period of the external field, and n is an integer. Values of the parameters: V = 1, $\gamma = 1$; a: $\omega = 3.5$ (K = 0.57), 46 trajectories of duration <100 T; b: $\omega = 1.5$ (K = 1.33), one trajectory of duration 200 periods; c: $\omega = 0.45$ (K = 4.44), one trajectory of duration 2000 T. Breakdown of the last invariant curve separating the resonances occurs at K = 0.97.

i(t), $\theta(t)$ amounts to a random process. The measure of the "chaotic character" of a trajectory is the Lyapunov index of exponential instability σ ,³ which determines the rate of divergence of two trajectories that are close at t = 0:

$$d(t) \sim d(0)\exp(\sigma t) \tag{2.4}$$

(d(t)) is the distance in phase space between the two closelying trajectories). For the values of the parameters that we shall use below ($\gamma = 1$, V = 0.5, $\omega = 0.3$; K = 4.71), numerical experiment yields the mean value $\sigma = 0.1$. Thus, when the initial conditions differ by 0.01, two trajectories diverge in phase even after two or three periods of the external field.

In closing this section we note that for the system being studied the chosen value $K \approx 4$ corresponds to the greatest degree of chaos. This value of the parameter K corresponds to the case of coincidence in order of magnitude of the intrinsic frequency of the nonlinear resonance of (1.13) with the frequency of the external field. A further increase in the value of K has the result that the two resonances "merge" into one, and the dynamics of the system is again close to regular (in this case the intrinsic frequency substantially exceeds the frequency of the external field and the adiabatic theory of perturbations is applicable³).



FIG. 13. Dynamics of the average value $p_c^{\rm cl}(t) = \langle \cos \theta \rangle$ for the system of Eq. (2.1) over an ensemble of particles uniformly distributed in phase at the initial instant of time with I(0) = 0. Values of the parameters: V = 0.5, $\gamma = 1$, $\omega = 0.3$ (K = 4.71). (From Ref. 11.)

2.2. Dynamics of the classical averages

Below, for purposes of comparison with the results of a quantum-mechanical analysis of the system of (2.1), we shall need information on the behavior of the averages. Let us restrict the treatment to the case of developed chaos, when $K \approx 4$. Figure 13 shows the behavior of $p_c^{cl}(t)$ $= \langle \langle \cos \theta \rangle \rangle$ for the value K = 4.71 (V = 0.5, $\gamma = 1$, $\omega = 0.3$; cf. Fig. 8, which corresponds to the choice $\omega = 0$). As the ensemble over which the averaging was performed, we chose, just as in the case of Fig. 8, an ensemble of particles uniformly distributed in phase with I = 0 at the initial instant of time. Since we associate the system of (2.1) with a dipole molecule, then evidently $p_{c}^{cl}(t)$ characterizes the macroscopic orientation of the molecular gas. We see from Fig. 13 that, in the course of 1-2 periods of the external field, $p_{\rm c}^{\rm cl}(t)$ reaches a steady-state regime, which amounts to periodic oscillations at the frequency of the external field. The initial stage (up to times t = 30 in Fig. 13) corresponds to a transition period during which the particles (representatives of the ensemble) uniformly fill all the phase volume accessible to them owing to the exponential instability. From this instant on, the average over the ensemble equals the average over the phase volume of the chaotic component

$$p_{c}^{cl}(t) = \langle \langle \cos \theta \rangle \rangle = \int_{G(t)} d\theta dI \cos \theta$$

(The volume of the region G(t) is constant.) Further oscillations of $p_c^{cl}(t)$ involve the change in the form of the phase space G(t) accessible to the particles, which, as we see from Fig. 14, oscillates at the frequency ω .

We see by comparing Figs. 8 and 13 that, both in the case of an isolated resonance and in that of a system of two interacting resonances, the behavior of the averages over the course of a relatively short time amounts to a regular process. However, there is a substantial difference. In the case of the integrable system of (1.2) the form of the curve strongly depends on the form of the ensemble (the initial conditions). In the case of the nonintegrable system of (2.1) the form of the curve is universal and does not depend on the initial conditions (with the proviso that each representative of the ensemble belongs to the chaotic component). This universality of the behavior of $p_c^{cl}(t)$ is introduced by the chaotic character of the motion of the individual particles. Consequently mixing occurs in the system, and the average over the ensemble of any quantity equals the average over the phase volume and does not depend on the choice of ensemble.

2.3. Interaction of QNRs

A system of two interacting resonances is one of the simplest models of classical stochasticity. Hence it seems natural to use it to study quantum chaos. We should note that this model differs in principle from the other popular model of quantum chaos-a quantum rotator under the action of δ -pulses²⁰ (see also the references in Ref. 2). The latter model is the simplest representative of a class of systems having an unbounded volume of the chaotic component of phase space. In these systems the phenomenon of diffusion occurs, and in going to the quantum-mechanical description the main effect is a decline in diffusion. To explain the phenomenon of the decline in diffusion an analogy has been adduced between the localization of the QFs of the quantum rotator and the Anderson localization of electrons in a solid.²¹ This approach has proved very fruitful, and has enabled understanding the features of diffusion in other quantum systems having an unbounded chaotic phase volume (in particular, in the hydrogen atom under the action of microwave radiation²).

The system of two interacting resonances being discussed represents a class of systems having a finite volume of chaotic phase space. Here the phenomenon of diffusion is absent and one needs other methods of analysis. Evidently the quantum analog of the system (2.1) has the form

$$H = -(\gamma h^2/2)\partial^2/\partial\theta^2 + 2V\cos\theta \cdot \cos\omega t$$
(2.5)

(We note that, when $\omega = 0$, Eq. (2.5) transforms into the Hamiltonian of an isolated QNR of (1.10).) We shall examine the properties of the system of (2.5) from three different



FIG. 14. Poincaré cross section of the system of Eq. (2.1) at different instants of time: a: $t_n = Tn$; b: $t_n = (T/4) + Tn$; c: $t_n = (T/2) + Tn$. The values of the parameters are as before. The diagram reflects the form of the phase volume of the system of Eq. (2.3) that is filled with the chaotic component (the variable J can be excluded from the treatment owing to the conservation of quasienergy).



FIG. 15. Mean population of the levels of the system of Eq. (2.5). The dots indicate the location of the levels having transitions in resonance with the external field (centers of the QNR). Values of the parameters: V = 1, $\gamma = 1$, h = 1/10; a: $\omega = 3.5$ (K = 0.57), b: $\omega = 3$ (K = 0.67), c: $\omega = 2.5$ (K = 0.8), d: $\omega = 2$ (K = 1). (From Ref. 10.)

standpoints. From the standpoint of the dynamics of the populations of the levels, this involves the structures of the eigen-QFs and the behavior of the quantum-mechanical averages (the observables). As an observable quantity we shall consider, as in Sec. 1, the induced polarization (see (1.8)), normalized to the quantity d:

$$P_{\mathbf{x}} = \langle \psi(\theta, t) | \cos \theta | \psi(\theta, t) \rangle, \quad P_{\mathbf{y}} = \langle \psi | \sin \theta | \psi \rangle.$$
(2.6)

Figure 15 shows the time average of the population of the levels of the system for different values of the quantity ω . At the initial instant of time the ground state of the system was populated. The dots in the diagram denote the levels such that the transitions from them are in resonance with the external field (i.e., the centers of the resonances). We recall that the external field gives rise to transitions only between adjacent levels. Case *a* corresponds to capture of the system into a secondary resonance (see Fig. 12), which can be treated as being isolated, while case b corresponds to overlapped resonances. In this case, in a time of the order of the period of the phase oscillations, a finite number of levels ($\sim \delta n$) becomes populated, and thenceforth the populations of these levels fluctuate without any visible regularities. We note that in the quantum case the overlap of nonlinear resonances occurs somewhat earlier (for a lower value of the Chirikov parameter of (2.2)) than in the classical case. This involves the fact that one must take account of the finite width of the transition region of the QNR in the vicinity of the separatrix, which at low values of the parameter of quasiclassicity introduces substantial corrections to the critical value K.

2.4. Structure of the QFs. Chaotic delocalization

Let us examine the structure of the eigenfunctions of the operator of evolution of the system in the period of the external field

$$\widehat{U}(\theta) = \widehat{T} \exp[-(i/h) \int_{0}^{T} \widehat{H}(t) dt]$$
(2.7)

as a function of the value of the parameter K. An important characteristic of a QF is its degree of delocalization, which we take to mean the number of effectively nonzero coefficients in the expansion of the QF in the unperturbed basis:

$$\psi_k(\theta) = \sum_n c_n |n\rangle.$$

As a graphic characteristic of the degree of delocalization, the concept of "dispersion" D was proposed:²²

$$D = \sum_{n} |n - \overline{n}|^{2} |c_{n}|^{2}, \quad \overline{n} = \sum_{n} n |c_{n}|^{2}.$$

Figure 16 shows the "dispersion" of the QFs as a function of its "center of gravity" \bar{n} . Each point corresponds to one QF. We note that in Fig. 16 only the QFs are represented that have odd symmetry, i.e., half of all the QFs. In this case the expansion is performed over the functions $|n\rangle = \pi^{-1/2} \sin(n\theta)$. The diagram for the even QFs has a similar form.

We see from the diagram that, when $K < K_{cr}$ ($K_{cr} \sim 1$), the points mainly lie on three branches. This structure of the diagram reflects the structure of the QFs of isolated QNRs. The middle branch corresponds to QFs corresponding to the vibrational regime (see Sec. 1.5), and the upper and lower branches to QFs corresponding to the rotational regime. Here the most delocalized functions are those which correspond to the region of the separatrices of the nonlinear resonances. We note also the presence of secondary QNRs, which look like excursions on the lower branch. An analytical method was proposed¹² for calculating the QFs of the secondary QNRs, and it was shown that the structure of the



FIG. 16. Relationship between the "dispersion" and the "center of gravity" of the eigenfunctions of the operator of Eq. (2.7). Values of the parameters: V = 1, $\gamma = 1$, h = 1/10; a: $\omega = 3.5$ (K = 0.57)-resonances not overlapping; b: $\omega = 0.45$ (K = 4.4)-strong overlap of the resonances. (From Ref. 26.)

secondary QNRs resembles that of the primary ones. In this sense the quantum system copies the features of the classical system (see Fig. 12). However, while in the classical case secondary nonlinear resonances exist of any order, in the quantum case the maximum order of a secondary QNR is determined by the condition $\delta I^{(k)} \sim h (\delta I^{(k)})$ is the width of a secondary classical resonance of the k th order). (A more detailed study of the self-similarity properties of quantum systems in the subcritical region of the parameter K is contained in Refs. 23–25.)

With increase in the overlap parameter k, the QFs belonging to the lower slope become delocalized, and when $K \approx 4$, a large group of QFs has a high degree of delocalization. This group of functions corresponds to the chaotic component of the phase space of the system of interacting classical resonances. We see from the diagram that this group lacks any regular structure, and the points, at least externally, lie at random. Correspondingly the QS also has no regular structure. In this case, to describe the QS it is expedient to introduce the statistical methods of analysis. (A corresponding statistical ensemble can be chosen by weakly varying the parameters of the system.) A statistical analysis of the QFs and the QS has been performed in Ref. 26. Here we shall only present the histogram of the distribution of states between adjacent quasienergy levels for the "chaotic" group of functions (Fig. 17). As is known, for completely chaotic systems (like billiards with negative curvature) the statistics of the distribution of states between adjacent levels is close to the Wigner-Dyson function, but for completely integrable systems it is close to the Poisson function (see Refs. 1 and 2). As we see from Fig. 17, in the case being discussed the statistics is intermediate between the Wigner-Dyson and the Poisson statistics.

2.5. Structure of the QFs. Two-dimensional delocalization

The chaotic delocalization of QFs discussed above is an important manifestation of chaos in a quantum system. However, the analysis of QFs presented above is not complete. Actually, in calculating the QFs on the basis of the evolution operator of (2.7), we lose information on the "temporal" dependence of the QFs. We can obtain this information as follows. Let us represent the eigen-QFs in the form of a double Fourier series

$$\psi_k(\theta, t) = (2\pi)^{-1} \sum_n \left[\sum_m c_{n,m}^{(k)} \exp(im\omega t) \right] \exp(in\theta).$$
(2.8)



FIG. 17. Distribution of the distance s between adjacent quasienergy levels for chaotic states. The quantity Δ corresponds to the mean distance between levels, $\Delta = h\omega/N$ (N is the number of levels). Values of the parameters: V = 1, $\gamma = 1$, h = 1/20, 0.45 < ω < 0.46 (K = 4.4). The smooth curve corresponds to a Poisson distribution. (From Ref. 26.)

Upon substituting the quasienergy solution $\psi(\theta,t) = \exp(-i\lambda_k t/h)\psi_k(\theta,t)$ into the Schrödinger equation with the Hamiltonian of (2.5), we obtain the following twoindex equation for the value of the quasienergy λ_k :

$$[(\gamma h^2 n^2/2) + h\omega m]c_{n,m}^{(k)} + (V/4) \sum_{+,-} c_{n\pm 1,m\pm 1}^{(k)} = \lambda_k c_{n,m}^{(k)}. (2.9)$$

We note that Eq. (2.9) can also be treated as the equation for the eigenvalues of the effective Hamiltonian \tilde{H}_{eff} of (2.3), where $I \rightarrow -ih\partial /\partial \theta$, $J \rightarrow -ih\partial /\partial \phi$, $\phi = \omega t$. Equation (2.9) was solved numerically, and the accuracy of calculation was monitored on the basis of the orthogonality of the QFs. The characteristic structure of the QFs of a system of two interacting QNRs is reflected in Fig. 18b, which gives the values of the coefficients $c_{n,m}$. We see from the diagram that, besides the delocalization "along the n axis," a delocalization occurs "along the m axis," i.e., the sum over m in (2.8) contains a large number of terms. One can obtain an estimate of the degree of delocalization over m (the number of terms δm in the sum over m in (2.8) on the basis of a quasiclassical approach. As Fig. 14 implies, in a chaotic regime the quantity I varies in the interval $\delta I \approx 4 (V/\gamma)^{1/2}$. Since the quasienergy of the system of (2.3) is a conserved quantity, the variation of the variable I is accompanied by a variation in the variable J in the interval

$$\delta J \approx \gamma \delta I^2 / 2\omega. \tag{2.10}$$

The developed stochasticity corresponds to a choice of the external frequency ω of the order of the phase frequency of the nonlinear resonance $\Omega_{\rm ph} = (\gamma v)^{1/2}$. Taking this remark into account, we have the following value for the region of variation of J:

$$\delta J \approx \delta I^2/2(\gamma V)^{1/2} = 4(V/\gamma)^{1/2} = \delta I.$$

Upon quantizing the variables I and J we obtain the characteristic degree of delocalization of the QFs: $\delta m \sim \delta n \sim \delta I / h$.

The delocalization of the QFs along the *m* axis is a strong manifestation of chaos in the quantum system being discussed and leads, as will be shown in the next section, to important physical consequences. We note that, in the case of a regular regime of motion, delocalization along the *m* axis is absent: as an illustration Fig. 18a shows the structure of the QFs of the isolated QNR of (1.6). We see that only the coefficients $c_{n,m}$ with n + m = const differ from zero. That is, the sum over *m* in (2.8) contains only one term (one can easily obtain this result analytically from (1.9). In the case $K \leq 1$ (two resonances, weak interaction), the structure of the QFs is close to that presented in Fig. 18a (elements weakly differing from zero appear on two adjacent diagonals), and can be calculated by perturbation theory.

2.6. Spectrum of the induced polarization

Let us proceed to discuss the physical consequences of two-dimensional delocalization. We shall examine the quantum-mechanical average of an arbitrary operator: $\langle A(t) \rangle = \langle \psi(\theta,t) | \hat{A} | \psi(\theta,t) \rangle$. In view of (1.15), the following expansion is valid

$$\langle A(t) \rangle = \sum a_k^* a_l^* \exp[i(\lambda_l - \lambda_k)t/h] \langle \psi_l(\theta, t) | A | \psi_k(\theta, t) \rangle. \quad (2.11)$$

Let us estimate the number of lines N in the frequency spectrum of the variable $\langle A(t) \rangle$. Let the initial state of the system



FIG. 18. Structure of the QFs of an isolated QNR (a) and a system of two interacting QNRs (b) Values of the parameters: V = 2, $\gamma = 1$, $\omega = 1$. (From Ref. 28.)

correspond to the ground state. Then, evidently, the number of nonzero coefficients a_k is determined by the degree of delocalization δn . Thus the spectrum of $\langle A(t) \rangle$ will contain $\sim \delta n^2/2$ lines at frequencies $(\lambda_i - \lambda_k)/h$. Now let us take account of the fact that the functions $\psi_k(\theta,t)$ are periodic functions of the time. Consequently every line is "converted" into a series of lines separated by an amount that is a multiple of ω . However, while in a chaotic regime the spectrum $\langle \psi_l(\theta,t) | \hat{A} | \psi_k(\theta,t) \rangle$ consists of the order of δm lines, in the regular case we have only several lines and their number *does not increase* with increase in the quasiclassicity parameter δn . To summarize what we have presented above, we obtain the following estimate for the number of lines N in the frequency spectrum:

$$N \sim \delta n^{\alpha}. \tag{2.12}$$

Here $\alpha = 2$ for the case of regular dynamics and $\alpha = 3$ for chaotic dynamics. We note that the presented estimate is an upper bound, and for a more exact estimate of the quantity N we need further studies. However, it is clear that in a chaotic regime the value of α is always larger than in a regular regime.

Figure 19a shows the Fourier spectrum of $\overline{A}(\omega)$ $(\overline{A}(\omega) = \langle A(t) \rangle \exp(i\omega t)$ the bar indicating averaging over time) of the observable $\hat{A} = \cos \theta$ for the system of (2.5) with the following values of the parameters: $\gamma = 1$, V = 0.5, $\omega = 0.3$ (K = 4.71), and h = 1/16, with the ground state $(\cos \theta)$ is the normalized induced polarization along the direction of the external field: $P_x(t) = p_c(t)$. (We note that in the case of a symmetrical initial condition $P_{\nu}(t) \equiv 0$). As we have already noted above, the spectrum of the induced polarization determines the emission spectrum of the system and can be studied experimentally. Figure 19b presents the frequency spectrum of $p_{c}(t)$ for the case of a regular regime of motion, which corresponds to the choice $\omega = 0$. The time of averaging was chosen equal to T = 840. By comparing Figs. 19a and 19b we see that in the regular case the spectrum of $p_{c}(t)$ consists of a small number of well resolved lines. In the chaotic case the number of lines is considerably larger. This effect is a consequence of two-dimensional delocalization and is one of the more marked manifestations of chaos in quantum mechanics for systems with a bounded volume of the chaotic component. We should note that, in the case under discussion (a molecule in a UHF field), one needs a high degree of resolution to detect this effect. With a low degree of resolution (short averaging times in the numerical experiment), the qualitative difference between the two cases is lost. In Sec. 2.9 we shall discuss the behavior of a molecule in an infrared field, where the effect being discussed is manifested considerably more markedly and the difference in the spectrum of the induced polarization for the chaotic and regular cases remains on the qualitative level, even at a low degree of resolution.

populated at the initial instant of time. According to (2.6),



FIG. 19. Spectrum of the induced polarization in the cases of chaotic (a) and regular (b) dynamics of the system of Eq. (2.5). Values of the parameters: V = 0.5, $\gamma = 1$, h = 1/16; a: $\omega = 0.3$ (K = 4.71), b: $\omega = 0$. Averaging time: T = 840. (Here and below the spectrum is given only for positive values of w, while for negative w the graph is symmetrical to that given.) (From Ref. 11.)

2.7. Correspondence times

We shall begin the discussion of the problem of the correspondence times with the question of the connection between the dynamics of the wave packet of (1.12) and the dynamics of an individual classical particle. One can easily show that in this case the correspondence times are logarithmically small. Actually, we shall set the quantum packet of (1.12) in correspondence with a packet of classical particles with a width in terms of action of the order of $h(\delta n)^{1/2}$ and of phase of the order of $\delta n^{-1/2}$. In view of the local instability of (2.4), the rough dimension of the packet will increase as $\exp(\sigma t)$, and in the course of the characteristic time

$$\tau \sim \ln(2\pi/\delta n^{-1/2})/\sigma \propto \ln(1/h)$$
 (2.13)

the packet will "spread" in phase by an amount of the order of 2π . Thus in the course of the time τ neither the dynamics of the averages over the classical ensemble nor, all the more, the dynamics of the quantum averages will follow the trajectory of a classical particle that corresponds at the initial instant of time with the center of the packet. One can obtain the time of (2.13) also in a more rigorous way by starting directly with the quasiclassical representation of the wave function.²⁹ To illustrate what we have presented above, Fig. 20 shows graphs of $\cos \theta(t)$ for a classical particle with the initial conditions $I(0), \theta(0) = 3.14$ (this trajectory is chaotic with a value of the parameter of local instability $\sigma = 0.4$) tion of the form (1.12) for h = 1/128. We see that a correspondence time is practically absent. We recall that, in the regular case (see Fig. 7) for the same values of Planck's constant and parameters of the system, the correspondence time is rather large. In closing this paragraph, we wish to draw attention to Ref. 30, which was devoted to comparing the evolution of the Wigner function and the roughly approximated Wigner function with the evolution of the classical distribution function specifically for the case of an initial condition in the form of a Gaussian packet with a width $\sim h^{1/2}$.

and of the quantum average $\langle \cos \theta \rangle$ for an initial wave func-



FIG. 20. Dynamics of the quantum average $\langle \cos \theta \rangle$ of the system of Eq. (2.5) $(V = 0.5, \gamma = 1, \omega = 0.3, h = 1/128)$ in the case in which the initial condition is chosen in the form of the wave packet of Eq. (1.12) with $I_0 = 0, \theta_0 = 3.14$ (1) and the value of $\cos \theta(t)$ for a classical particle with these same initial conditions (2) (the given trajectory is chaotic with $\sigma = 0.4$).



FIG. 21. Dynamics of the average $p_c(t) = \langle \cos \theta \rangle$ of the quantum system of Eq. (2.5) in the case of an initial condition in the form of population of a single level with n = 0. The value of h is 1/16 with the other parameters as before. (From Ref. 11.)

Let us proceed to the case of an initial condition of the form of (1.13). Despite the fact that in a chaotic regime there is no correspondence between the behavior of an individual classical particle and a quantum packet, there is correspondence for the dynamics of the averages, and in the quantum case the graph of the quantum-mechanical average $\langle \cos \theta \rangle$ repeats in general features the graph of the average over the classical ensemble (see Fig. 13), even for small values of the quasiclassicality parameter (Fig. 21). As has already been noted in Sec. 1.1, in this case the question of the correspondence times transforms into the question of the level of quantum fluctuations. Figure 22 shows the variation of the mean level of fluctuations

$$S^{2} = (\langle \cos \theta \rangle - \langle \langle \cos \theta \rangle \rangle)^{2}$$

(here $\langle \cos \theta \rangle$ is the quantum-mechanical average, and $\langle \langle \cos \theta \rangle \rangle$ is the classical average) and the values of Planck's constant $h = (1/2)^M$ for regular and chaotic dynamics. We see that in the regular case the convergence is considerably better. At present we cannot indicate with assurance the law according to which the decrease in the level of fluctuations occurs with decreasing h. This question needs further study.

In closing this section we note another approach to the question of the correspondence of classical and quantummechanical description of a dynamic system, based on comparing the spectra of the observables. As one can easily show, in the classical case the spectrum is continuous. In particular, the spectrum of the process shown in Fig. 8 is determined by the relationship

$$p_{\rm c}^{\rm cl}(w) = \sum_{k=0}^{\infty} \int_{0}^{J^*} b_k(J) \delta(w - k\Omega(J)) \mathrm{d}J. \qquad (2.14)$$

Here the $b_k(J)$ are certain functions of $J, J^* = \delta J/2\pi$, and the graph of $\Omega(J)$ is shown in Fig. 2. (In deriving (2.14) we transformed from integrating over the initial phase θ_0 to integrating over J and took account of the fact that $\cos \theta(t, \theta_0)$ is a periodic process with the frequency $\Omega(J)$.) In the quantum case the frequency spectrum of the observable $p_{c}(w)$ is discrete. However, at a finite degree of resolution (finite observation times), the spectrum of an observable quantity is a convolution of the discrete spectrum and an apparatus function F(w) and also is continuous (in a numerical experiment F(w) is determined by the time of observation T: $|F(w)| = 2 \sin(wT/2)/wT)$. Thus we can study the law according to which the "quantum" spectrum approaches the "classical" as a function of the value of Planck's constant and the time of observation. Studies of this type are of considerable interest, especially for interpreting experimental results from the analysis of the frequency spectra of chaotic systems.

2.8. A dipole molecule in a UHF field

In the previous sections our fundamental model was a plane rotator under the action of a periodic, circularly or linearly polarized, field. Evidently this system is a two-dimensional idealization of a dipole molecule in a UHF field. In this section we shall show that this idealization in the case of a linearly polarized field is completely regular and that transition to the three-dimensional case does not entail substantial changes in the theory.

Let us direct the z axis parallel to the polarization vector of the external field. Then the Hamiltonian of the system has the form

$$\widehat{H} = \widehat{H}_0 - Ed\cos\theta \cdot \cos\omega t.$$
(2.15)

Here \hat{H}_0 corresponds to free rotation of the molecule. The eigenfunctions of \hat{H}_0 are the spherical functions $\Phi_{l,m}$, which correspond to the eigenvalue $E_l = Bl(l + 1)$ (B is the rotational constant of the molecule), and $|m| \leq l$. Let us represent the wave function of the system in the form of an expansion in spherical functions

$$\psi(t) = \sum_{l,m} a_l^{(m)}(t) \Phi_{l,m}.$$
 (2.16)

Upon substituting (2.16) into (2.15), we obtain the following system of equations for the amplitudes of the populations of the levels:

$$ih\partial a_{l}^{(m)}/\partial t = E_{l}a_{l}^{(m)} - (Ed/2)(V_{l}^{(m)}a_{l-1}^{(m)} + V_{l+1}^{(m)}a_{l+1}^{(m)})\cos\omega t,$$

$$V_{l}^{(m)} = [(l-m)(l+m)/(2l-1)(2l+1)]^{1/2}.$$
(2.17)



FIG. 22. Mean level of quantum fluctuations as a function of $h = (1/2)^{M}$ in the case of regular (1) and chaotic (2) dynamics of the system. Values of the parameters as before, time of averaging T = 50.

We see from (2.17) that the magnetic quantum number does not vary in the process of interaction of the system with the field and is a parameter of the problem. Taking this circumstance into account, we can rewrite the system of equations (2.17) in a "prettier" form. Let us denote $c_n(t) = a_{n+m-1}^{(m)}(t)$. Then the index n = l - m + 1 runs through values from 1 to ∞ , and Eq. (2.17) acquires the form

$$i\hbar\partial c_n/\partial t = \widetilde{E}_n c - (Ed/2)(\widetilde{V}_n c_{n-1} + \widetilde{V}_{n+1} c_{n+1})\cos\omega t,$$
 (2.18)

$$\widetilde{V}_n = [(n-1)(n+2m-1)/(2n+2m-1)(2n+2m+1)]^{1/2},$$

$$\widetilde{E}_n = B(n+m+1)(n+m).$$

Now we see that the system of (2.18) is equivalent to the system of equations for the amplitudes of the populations of the plane rotator of (2.5) with the additional condition that we are treating only the solutions odd in θ $(\psi(\theta,t) = \pi^{-1/2} \sum_{n=1}^{\infty} c_n(t) \sin(n\theta))$:

$$i\hbar\partial c_n/\partial t = Bn^2 c_n - (Ed/2)(V_n c_{n-1} + V_{n+1} c_{n+1})\cos\omega t,$$

 $V_1 = 0, \quad V_n = 1/2 \ (n > 1), \quad B = \gamma h^2/2.$
(2.19)

Thus the rotating polar molecule interacting with the resonance UHF field realizes two QNRs with the overlap parameter of the resonances $K = \delta n/n^*$, where δn is the width of the QNR, $\delta n = 2(Ed/B)^{1/2}$, and n^* is the center of the QNR, $n^* = (\omega - B)2B$. We obtain the greatest coincidence with a plane rotator, as we should expect, in the case m = 0. $\widetilde{E}_n = Bn(n-1)$ Then we have and $\tilde{V}_n = [(n-1)n/(2n-1)(2n+1)]^{1/2}$ instead of E_n $=Bn^2$ and $V_n = 1/2$ for the plane rotator. These differences introduce only small quantitative divergences into the dynamics of the systems (2.15) and (2.19). In the general case the parameter that determines the degree of divergence is the ratio of the distance to the center of the QNR n^* to the value of the magnetic quantum number m. Below we shall assume that $n^*/m \ge 1$, so that we can use a two-dimensional model to analyze the rotational dynamics of the molecule.

Let us take up the characteristic values of the parameters. The fundamental condition for applicability of the theory presented in the previous sections is the condition $\delta n \ge 1$, which in the notation used in this section acquires the form $2(Ed/B)^{1/2}$. Moreover, it is desirable that the chosen molecule can be treated as a rotator (rather than a top), and that conditions are satisfied that hinder the excitation of the vibrational degree of freedom. All these conditions can be satisfied for heavy polar diatomic molecules, e.g., for the molecule CsI. The molecule CsI is a rotator and has the following values of the parameters: B = 708 MHz (2.36×10^{-2}) cm⁻¹), d = 12.1 D. The energy of the first vibrational level corresponds to the energy of the 71st rotational level, which imposes the condition $\delta n < 140$. If we assume that E = 3kV/cm (10 CGS units), we have a value of the energy of interaction of the system with the field $Ed = 0.61 \text{ cm}^{-1}$ and a value of the quasiclassicity parameter $\delta n = 10$. The frequency of the external agent ω is assumed to be of the order of $\Omega_{\rm ph} = (BEd)^{1/2} = 0.12$ cm⁻¹ = 3.5 GHz. As the object of observation, it is apparently expedient to use a beam of molecules preliminarily cooled to energies lower than the energy of interaction of the system with the field. For the example being discussed this yields a temperature ~ 1 K.

The "acceptability" of molecules of the type of CsI for studies of chaotic rotational dynamics is discussed in greater detail in Ref. 31.

To avoid misunderstanding we note that we are presenting here an estimate of a diatomic molecule only for reasons of simplicity and in no way do we reject the possibility of using a polyatomic molecule as the object for a real experiment. This is all the more so since the condition $2(Ed/B)^{1/2} \ge 1$ is fulfilled for it at a considerably lower amplitude of the external field.

2.9. A diatomic molecule under the action of an infrared field

In this section we shall examine the phenomenon of interaction of two QNRs in a diatomic molecule under the action of an infrared field.³² We shall start the analysis of the problem with the classical approach and restrict the treatment of the rotational degree of freedom to the two-dimensional approximation. In the classical approach a diatomic molecule in a Σ state can be correlated with a system of two point masses with an interaction potential U(r) having a minimum at $r = r_0$. In action-angle variables the unperturbed motion of the molecule corresponds to the Hamiltonian $H_0 = H_0(I, L^2)$, where the quantity I characterizes the vibrational motion of the nuclei (I = 0 corresponds to absence of vibration), and the variable L coincides with the angular momentum. We shall denote the conjugate variables to I and L, respectively, by ϕ and θ (θ is the angle between the x axis and the axis of the molecule). Below we shall restrict the treatment to the case of small values of I and L $(H_0(I,L^2) \equiv E \ll |U(r_0)|)$. Then, upon expanding H_0 up to quadratic terms, we have

$$H_0 = h\Omega n - xh\Omega n^2 + Bl^2, \qquad (2.20)$$

$$r = r_0 + (2hn/\Omega M)^{1/2} \cos \phi.$$
 (2.21)

In (2.20) and (2.21) we have transformed to the dimensionless actions n = I/h and l = L/h and used the standard notation for the frequency of harmonic vibrations $\Omega = dH_0/dI$, for the anharmonicity $x = h(d^2H_0/dI^2)/\Omega$, the moment of inertia $B = h^2 dH_0/dL^2$, and the reduced mass M.

,

Let us study the process of interaction of the molecule with the linearly polarized field E(t) with polarization along the x axis. We shall assume the dipole moment of the molecule to be $\mathbf{d} = e_{\text{eff}}\mathbf{r}$, where the effective charge of the atoms is $e_{\text{eff}} = d_0/r_0$. Then the Hamiltonian of interaction of the molecule with the radiation has the form $H_{\text{int}} =$ $- (\text{Ed}) = -Ee_{\text{eff}}r\cos\theta\cdot\cos\omega t$. Upon substituting r from (2.21) and keeping only the terms responsible for the resonance interaction, we have

$$H_{\rm int} = W n^{1/2} \cos \theta \cdot \cos(\phi - \omega t), \quad W = -e_{\rm eff} E (2h/\Omega M)^{1/2}, \quad (2.22)$$

Here W is the interaction parameter of the infrared radiation with the molecule. If we characterize the field intensity by the intensity of the radiation, then, e.g., for the parameters of the GeO molecule $h\omega = 985.8 \text{ cm}^{-1}$, M = 13.1 atomic weight units, and radiation intensity 25 GW/cm², we have $W = 10.72 \text{ cm}^{-1}$.

We shall represent the linearly polarized field as the sum of two circularly polarized components: $Wn \cos \theta$ $\times \cos(\phi - \omega t) = (W/2) n \times [\cos(\phi - \omega t - \theta) + \cos(\phi - \omega t + \theta)]$. Each term in the latter formula leads to the



appearance in the system of a nonlinear resonance, which we shall treat in somewhat greater detail. Let us study the term containing the phase $\phi - \omega t + \theta$. We shall perform a canonical substitution of variables $(n,l) \rightarrow (n,k)$, $(\phi,\theta) \rightarrow (\overline{\Theta}, \overline{\Theta})$, where k = l - n, $\overline{\Theta} = \phi - \omega t + \theta$ is the slow phase, and $\overline{\Theta}$ is the conjugate variable to k. Then the Hamiltonian $H_0 + H_{int}$ acquires the form

$$H = \Delta(k)n - \gamma n^2 + Bk^2 + (W/2)n^{1/2}\cos\Theta = H_{\text{eff}}(n,\Theta) + Bk^2,$$

$$\Delta(k) = h(\Omega - \omega) + 2Bk, \quad \gamma = xh\Omega - B.$$
(2.23)

We see from (2.23) that the quantity k is an integral of the motion, and thus the problem has been reduced to analyzing a one-dimensional system having the Hamiltonian $H_{\text{eff}}(n,\Theta)$, where the quantity k plays the role of a parameter.

The phase portrait of the system (2.23) was reproduced in Fig. 3. As we see from the diagram, at values of the parameter Δ above a certain critical value (k = -11 for the chosen values of the parameters), a "principal" and an "associated" nonlinear resonance exist in the system. However, at values of the radiation intensity below 10^{11} W/cm², the real dimensions of the associated resonance for most diatomic molecules are small, and we can neglect its influence on the dynamics of the system. In the case in which the system is captured into the principal nonlinear resonance, the motion of the molecule amounts to periodic changes in the vibrational state n(t) (which are accompanied by a change in the rotational state l(t) = n(t) + k at the characteristic frequency $\Omega_{\rm ph} \sim 10^{12} \, {\rm s}^{-1}$. The maximum and minimum possible values of n are determined here by the location of the separatrix of the principal nonlinear resonance. The location of the separatrix for the chosen parameters of the system is shown in Fig. 23 by the dashed lines. In the same diagram the solid lines indicate the location of the center of the principal resonance (upper curve), and also the location of the hyperbolic point of the principal resonance and the center of the associated resonance. Thus the region of influence of the nonlinear resonance due to the left-polarized component is bounded by the dashed lines in Fig. 23. The region of influence of the nonlinear resonance due to the right-polarized component will lie symmetrically (with respect to the coordinate axes) to that shown in the diagram. We can easily see that the nonlinear resonances have a common region of influence in the vicinity of l = 0; according to what we have presented earlier, we expect chaotic dynamics in this region. As an illustration, Fig. 23 shows the projections of the trajectory of a GeO molecule for two initial conditions: a) n(0) = l(0) = 4, $\Theta(0) = \phi(0) = 0;$ b) n(0) = 5, $l(0) = 15, \Theta(0) = \phi(0) = 0$. We see that in the latter case

FIG. 23. Examples of chaotic (1) and regular (2) trajectories of a GeO molecule in a linearly polarized infrared field of high intensity (25 GW/cm²). Duration of trajectories: 10^{-10} s and 10^{-11} s, respectively. The diagram also shows the location of the separatrix (dashed lines) and the center (upper solid curve) of the principal nonlinear resonance due to the leftpolarized component of the external field. (From Ref. 32.)

the quantity k is approximately conserved $(k \approx 10)$. For the initial conditions belonging to the common region of influence of the nonlinear resonances (case a), the trajectory is chaotic. The duration of the trajectories on the real time scale amounts to 10^{-10} and 10^{-11} s, respectively.

Let us proceed to the quantum-mechanical analysis of the system. As before, we shall restrict the treatment to small values of the quantum numbers n and l. Then the Hamiltonian of the molecule under the action of linearly polarized radiation in the action-angle representation has the form

$$\hat{H} = h\Omega(\hat{J} + (1/2) - x\hat{J}^2) + B\hat{I}^2 + [Ed_0 + (W/2)(\hat{J}^{1/2}\exp(i\phi) + \exp(-i\phi)\hat{J}^{1/2})]\cos\theta \cdot \cos\omega t.$$
(2.24)

Here we have $J = -i\partial/\partial\phi$, $I = -i\partial/\partial\theta$, and the value of W is determined by Eq. (2.22). We shall seek the eigenquasienergy solutions of the system in the form

$$\psi(t) = \exp\left[-i(\lambda_k t/h + \Omega t/2)\right] \sum_{n,l} c_{n,l} \exp(in\omega t) |n, l\rangle. \quad (2.25)$$

Substituting (2.25) into (2.24) and neglecting the nonresonance terms, we obtain the following two-index equation for the eigenvalues of the quasienergy:

$$[h(\Omega - \omega)n - xh\Omega n^{2} + Bl^{2}]c_{n,l}^{(k)} + (W/4) \sum_{+,-} [(n+1)^{1/2}c_{n+1,l+1}^{(k)} + n^{1/2}c_{n-1,l+1}^{(k)}] = \lambda_{k}c_{n,l}^{(k)}.$$
(2.26)

In comparing the latter formula with Eq. (2.9) we see that the system being studied is equivalent in many ways to the system of two interacting resonances of (2.5). In particular, the QFs of the molecule for a circularly polarized field (in the summations in (2.26) one term each is present; the first sign corresponds to a left-polarized field, and the second to right-polarized) and the QFs for a linearly polarized field will have a qualitative difference analogous to that shown in Fig. 18. This qualitative difference will be manifested in two main effects.

Let the system exist in the ground state at the initial instant of time. Then in the case of a circularly (left) polarized field, only the levels with n = l will be populated. For the chosen values of the parameters in the numerical experiment, 6–7 vibrational-rotational levels were excited, which agrees satisfactorily with the width of the classical nonlinear resonance (see Fig. 23). In the case of linearly polarized radiation, $\delta n \sim 7-8$ levels in *n* and $\delta l \sim 21-25$ levels in *l* were involved in the dynamics of the system, i.e., in all, of the order of 200 vibrational-rotational levels. Thus one of the



effects of overlap of two QNRs is a substantial increase in the number of levels involved in the dynamics of the transitions: $\delta N \sim \delta n \cdot \delta l$.

The second effect involves the number of lines in the induced-polarization spectrum. Let us represent the highfrequency component of the polarization of the molecule in the form

$$P_{x}(t) = p_{c}^{(x)}(t)\cos\omega t + p_{s}^{(x)}(t)\sin\omega t, \qquad (2.27)$$

$$P_{v}(t) = p_{c}^{(y)}(t)\cos\omega t + p_{s}^{(y)}(t)\sin\omega t.$$

Here we have $p_c^{(x,y)}(t) = (1/T) \oint \langle \psi(t) | \hat{d}_{x,y} | \psi(t) \rangle \cos \omega t \, dt$, where T is the period of the external field. Since the emission intensity of the oscillating dipole is proportional to the second derivative of the polarization, the frequency spectrum of the quantities $p_{c,s}(t)$ determines the spectral composition of the high-frequency radiation of the molecule. We note that, under the initial condition that the ground state is populated, owing to the symmetry of the solution, we have $p_{c,s}^{(y)} \equiv 0$ for a linearly polarized field, while for a circularly polarized field we have $p_s^{(y)} = -p_c^{(x)}, p_c^{(y)} = -p_s^{(x)}$. Figure 24 shows the frequency spectrum of the dimensionless envelope $\tilde{p}_{c}(t)$ $(|\tilde{p}_{c}(t)| \leq 1)$ in the case of linearly (a) and circularly (b) polarized radiation (the spectrum of $\tilde{p}_s(t)$ has an analogous form). We see that in the regular case the spectrum consists of a small number of well resolved lines. In the regime of quantum chaos the spectrum is very complex and close to the spectrum of a random process. This effect is fully analogous to that discussed in Sec. 2.6, and is a consequence of the two-dimensional delocalization of the QFs of the system.

In closing this section we shall take up the question of the validity of the two-dimensional approximation. In the case of a linearly polarized field the three-dimensional model has an additional integral of the motion (the projection of the angular momentum on the direction of polarization of the field). This, as was shown in the previous section, ensures the adequacy of the two-dimensional model of the real situation. The case of a circularly polarized field for the three-dimensional model is more complex: the system does not possess an additional global integral of the motion and in the general case one cannot reduce the two-dimensional model to a three-dimensional one (however, one can do this in certain special cases³²). The dynamics of a molecule in a circularly polarized field requires additional study.

2.10. A "rotating" two-level system

It was shown in the previous section that a regime of quantum chaos can occur in a molecule in the case of linearly

FIG. 24. Frequency spectrum of the envelope $\bar{p}_c^{(x)}(t)$ of the induced polarization in the GeO molecule in the case of a circularly (a; regular regime) and linearly (b; chaotic regime) polarized field. (From Ref. 32.)

polarized light. Here the condition was assumed to be satisfied that the interaction energy of the system with the field substantially exceeds the anharmonicity constant, $W > xh\Omega$. However, for most diatomic molecules the fulfillment of this condition requires very high intensities of laser radiation, which presents certain difficulties from the experimental standpoint. It was shown in that section that a chaotic regime of motion can be obtained also at substantially smaller amplitudes of the external field $W < xh\Omega$ (however, W > B) under the condition that the external field is modulated at the frequency v: $E(t) = E \cos vt \cdot \cos \omega t$, while the carrier frequency ω is in resonance with a vibrational transition (one can achieve the modulation by using two lasers with slightly differing frequencies). The condition $W < xh\Omega$ allows one to restrict the treatment in analyzing the problem to two vibrational levels. Thus we arrive at a two-level system with account taken of the "fine structure of the levels" due to the rotational degree of freedom (see Sec. 1.7).

According to (1.24) the system of equations in the wave functions $\psi_{1,2}(\theta,t)$ for two vibrational levels has the form

$$i\hbar\partial\psi_2/\partial t = (E_2 - B\partial^2/\partial\theta^2)\psi_2 - dE(t)\cos\omega t \cdot \cos\theta \cdot \psi_1, (2.28)$$

$$i\hbar\partial\psi_1/\partial t = (E_1 - B\partial^2/\partial\theta^2)\psi_1 - dE(t)\cos\omega t \cdot \cos\theta \cdot \psi_2$$

Here $E(t) = E \cos \nu t$, and *d* is the value of the matrix element of the transition. Let us introduce the function $\Phi(\theta,t) = \exp(i\omega t)\psi_2(\theta,t) + \psi_1(\theta,t)$. Then, in the resonance approximation (in the approximation of a rotating wave) and under the condition of exact resonance $E_2 - E_1 = h\omega$, we obtain the following equation for $\Phi(\theta,t)$:

$$th\partial\Phi/\partial t = \hat{H}_{eff}\Phi, \quad \hat{H}_{eff} = -B\partial^2/\partial\theta^2 - (Ed/2)\cos\theta\cdot\cos\nu t.$$
(2.29)

The latter is fully equivalent to the Schrödinger equation for a system of two interacting QNRs (the model of a molecule in a UHF field). The absence of modulation corresponds to an isolated QNR. The function $\Phi(\theta,t)$ fully describes the dynamics of the system under study. In particular, we have the following for the value of the induced polarization:

$$P(t) = d(p_c(t)\cos\omega t + p_s(t)\sin\omega t), \qquad (2.30)$$

$$p_{c}(t) = (1/2) \sum_{l} (b_{l+1}^{*} b_{l} + \text{c.c.}),$$

$$p_{s}(t) = (1/2i) \sum_{l \text{ odd}} (b_{l+1}^{*} b_{l} + b_{l+1}^{*} b_{l}).$$

Here $b_i(t)$ are the coefficients of the Fourier expansion of



FIG. 25. Population of the upper vibrational level of the system of Eq. (2.28) in the cases of a regular (a) and a chaotic (b) regime. Values of the parameters as in Fig. 26.

 $\Phi(\theta,t)$. Before we proceed to the comparative analysis of the behavior of the envelopes in the cases of regular ($\nu = 0$) and chaotic ($\nu \neq 0, K > 1$) dynamics of the system, we note that the characteristic frequencies $p_c(t)$ and $p_s(t)$ can differ by several orders of magnitude. Actually, as we see from (2.30), $p_c(t)$ corresponds to polarization of the system with

the effective Hamiltonian of (2.29): $p_c(t) = \langle \Phi | \cos \theta | \Phi \rangle$. Thus, according to the results of Sec. 1.8, the characteristic frequency $p_c(t)$ is determined by the frequency of the phase oscillations $\Omega_{\rm ph} = (BEd)^{1/2}/h$. This frequency corresponds to the slow motion of the system in the rotational degree of freedom. On the other hand, the characteristic fre-



FIG. 26. Spectrum of the envelope of $p_s(t)$ for the system of Eq. (2.28) in the cases of regular (a) and chaotic (b) dynamics of the system. (The spectrum of $p_c(t)$ is shown in Fig. 19.) Values of the parameters: $\gamma = 2B/h^2 = 1, V = -Ed/2 = 1.a: v = 0, b: v = 0.3$. Averaging time T = 210. (From Ref. 11.)



FIG. 27. Autocorrelation function of the process $p_s(t)$. Parameters as before. (From Ref. 11.)

quency $p_s(t)$ is determined by the Rabi frequency: $\Omega_{\rm R} = Ed/h$. This fact is associated with the circumstance that the envelope for sin ωt determines the absorption law of the energy of the system (2.28). (The energy of the system is proportional to the population of the upper level. A graph of the population of the upper level for chaotic and regular regimes of motion is shown in Fig. 25.) Thus, $p_s(t)$ reflects the fast motion of the system in the vibrational degree of freedom. Since we are studying the case $\delta n = 2(Ed/B)^{1/2} \ge 1$, we have

$$\Omega_{\rm R} = Ed/h = \Omega_{\rm ph} \delta n/2 \gg \Omega_{\rm ph} \left(\Omega_{\rm ph} = (BEd)^{1/2}/h \right).$$

As was already noted in Sec. 2.6, the fundamental manifestation of chaos in the system of (2.29) is the "complexity" of the frequency spectrum of the quantum-mechanical averages. The spectrum of the envelope of $p_{c}(t)$ in the cases of regular (v = 0) and chaotic (v = 0.3, K = 4.71) dynamics was shown in Fig. 19. Figure 26 shows the spectrum of the envelope of $p_s(t)$. As we see by comparing Figs. 19 and 26, for $p_s(t)$ the qualitative difference between the regular and chaotic regimes is considerably stronger. While in the regular case the spectrum of $p_{s}(w)$ consists of a relatively small number of lines and possesses the characteristic structure of the spectrum of a quasiperiodic process, in the chaotic case the spectrum of $p_s(w)$ is close to the spectrum of a random process. The fact that we can treat the process $p_s(t)$ as a random process for the system being analyzed when K > 1 is confirmed also by the behavior of the autocorrelation function $R(\tau) = p_s(t + \tau)p_s(t)$ (Fig. 27).

The system studied in this section, as we see it, seems to be the most acceptable object for experimental study of quantum chaos in systems having a bounded volume of the chaotic component. As compared with the diatomic molecule (Sec. 2.9), here, first, one requires a considerably smaller intensity of the external field, and second, this system enables one to treat both the chaotic and the regular regimes of motion. The advantage in comparison with the dipole molecule in a UHF field consists in the fact that, instead of (or alongside) $p_c(t)$, we can analyze another characteristic— $p_s(t)$. The advantage of measuring the quantity $p_s(t)$ lies in the fact that, as the amplitude E of the external field increases, the frequency interval $p_s(w)$ increases in proportion to E (rather than $E^{1/2}$ in the case of $p_c(w)$). Therefore the problem of resolving the frequency lines for studying the relationship (2.12) is considerably simplified.

CONCLUSION

In this article we have examined from the classical and quantum standpoints the dynamics of an isolated nonlinear resonance and a system of two interacting resonances. The latter system manifests a chaotic regime of motion in the classical treatment, which engenders interest in the dynamics of this system in the quantum case. As we have noted above, this system is a representative of a class of systems having a bounded volume of the chaotic component. In this it differs in principle from the models of quantum chaos most studied up to now-a quantum rotator under the action of a periodic sequence of δ -pulses and a hydrogen atom in a UHF field (see Ref. 2 and the references given there). (In the latter systems in the classical limit the phenomenon of diffusion can occur, and the efforts of the authors are directed mainly at explaining the features of diffusion in the quantum case.)

We can arbitrarily combine the questions dealt with in this review into three main groups: 1) comparison of the classical and quantum dynamics of the systems being studied; 2) the fundamental manifestation of chaos in the quantum approach; 3) application of the theory to analyze concrete physical objects. Let us start with the last point. We examined the phenomenon of QNR and the interaction of QNRs that arise in molecules under the action of UHF and infrared fields of high intensity (not below 1 MW/cm²). Of course, these systems do not exhaust all the systems in which QNR can occur and the interaction of QNRs. We restricted the treatment to analyzing the chosen systems, since for them the phenomenon of QNR and the interaction of two QNRs are manifested in the most "standard" form.

The question of the fundamental manifestations of chaos in the quantum case should be divided into two parts. They are the "theoretical" manifestations of chaos, which include the general analysis of the solution of Schrödinger's equation, the structure of the QFs, etc., and the "practical" manifestations of chaos, i.e., the manifestations that can be detected experimentally. Drawing an analogy with dynamic systems having an unbounded chaotic component of phase space, we can give a brief answer to the question that we have posed as follows. For the latter systems the fundamental manifestation of chaos is the relative (in comparison with the regular case) delocalization of the QFs, while the practical manifestation is the phenomenon of diffusion with subsequent saturation due to quantum effects. For the systems discussed in this study having a bounded chaotic component (in which the phenomenon of diffusion is practically absent), the fundamental theoretical manifestation of chaos is the two-dimensional delocalization of the QFs, while the practical manifestation is the "complexity" of the spectrum of the intrinsic radiation of the system. In particular, this is manifested in differing laws of increase of the number of lines with increase in the value of the quasiclassicity parameter (with increased amplitude of the external field) for the regular and chaotic regimes of motion and with the quasirandomness of the behavior of the quantum-mechanical averages. We note that in this sense the dynamics of the quantum system is "more random" than that of the classical system, since the dynamics of the classical averages (as was shown in Sec. 2.2) amounts to a periodic process.

As regards the latter question of the correspondence between classical and quantum dynamics, for the systems discussed here this question is too complex to offer an exhaustive answer at present. Therefore, on the theoretical level we have restricted the treatment only to formulating more distinctly the problem for further study. However, the presented numerical calculations of the dynamics of the system of ONR can be useful on the practical level, since they allow one to point out the concrete values of the parameters for which the dynamics of the system (e.g., a molecule in a UHF field) is close to classical and the approaches of classical mechanics can be employed. We note also that we have discussed the problem of correspondence only as applied to the observable quantities and left aside the analysis of the structure of the phase space of the quantum system (i.e., the analysis of the wave function in the Husimi representation or in the representation of coherent states) as being an unobservable characteristic of the system, at least at present.

Finally we wish to take up another question that we have not yet touched upon in this review. This is the question of taking account of the influence of relaxation processes. Taking account of relaxation processes for the systems being discussed constitutes an extremely complex and at the same time important problem. The complexity arises from the large number of levels (up to several hundred) participating in the dynamics of the system, which requires taking nontraditional approaches and methods of analysis. (In Appendix III we discuss one of these possible methods.) Moreover, preliminary studies of model quasiclassical systems show that in a certain sense dissipation restores classical features in the dynamics of a quantum object.³³⁻³⁶ That is, the dynamics of a quantum system with dissipation is closer to classical than in the absence of dissipation. Therefore the question of taking account of the influence of relaxation processes on the dynamics of multilevel quantum systems is interesting also from the fundamental standpoint of correspondence of classical and quantum mechanics. We hope to return to this problem in the future.

APPENDICES

Ι.

Let us use the analogy between the Hamiltonian of the QNR at $\Delta = 0$ ($\hat{H}_{eff} = -(\gamma h^2/2)\partial^2/\partial\Theta^2 + V\cos\Theta$) and the Hamiltonian of a particle of mass γ^{-1} in the periodic potential $V \cos \Theta$. Then the physical reason for absence of a sharp boundary of the transition from a vibrational to a rotational regime consists in the effect of tunneling. Being exponentially small for low energy values, the effect of tunneling becomes appreciable as λ_k approaches the value of the energy at the separatrix. In this case the eigenfunctions corresponding to the values of λ_k ($\lambda_{\min} < \lambda_k < V$) cannot be treated as wave functions of vibrations of the particle in an isolated potential well. Analogously, upon approach to the separatrix from above, the effect of reflection over the well leads to splitting of the levels λ_{2k} and λ_{2k+1} , which becomes appreciable when $\lambda < \lambda_{max}$. To characterize quantitatively the width of the transition region $\lambda_{\min} < \lambda < \lambda_{\max}$, we shall assume λ_{max} equal to the value beyond which the magnitude of the splitting of the levels does not exceed a preassigned small number ε (the magnitude of ε can be determined by the degree of accuracy of observation, $\varepsilon \sim h/t$). We note that the splitting of the levels λ_{2k} and λ_{2k+1} coincides with the value of the width of the forbidden band for a particle in a periodic potential. Analogously, as the value of λ_{\min} we shall choose a value below which the width of the allowed bands does not exceed the value of ε (as is known, the width of the allowed band is determined by the magnitude of the tunneling effect).

Let us estimate the value of λ_{\min} . As is known, the relative width of the allowed bands is determined by the quantity $2D^{1/2}/\pi (D \leq 1)$, where D is the penetrability coefficient of the potential barrier separating two potential wells of the potential $V \cos \Theta$:

$$D = \exp\{-(8/h) \int_{0}^{\theta} [(V \cos \Theta - \lambda)/\gamma]^{1/2} d\Theta\},\$$

$$\theta = \arccos(V/\lambda).$$
(I.1)

Since we are assuming λ to be close to V, the integral in (I.1) is approximately equal to $(\pi/2)(V/\gamma)^{1/2}[1-(\lambda/V)]$. Upon assuming that $\varepsilon = 2D^{1/2}/\pi$, we obtain

$$1 - \lambda_{\min} / V \sim |\ln \varepsilon| / \delta n. \tag{I.2}$$

Let us estimate the number Δk of levels lying in the transition region $\lambda_{\min} < \lambda < V$. We shall use the quasiclassical quantization rule, and then we have

$$\Delta k \approx h^{-1} \int_{\lambda_{\rm gab}}^{V} d\lambda / \Omega(\lambda). \tag{I.3}$$

Substituting into (I.3) the value of $\Omega(\lambda)$:³

$$\Omega^{-1}(\lambda) = 2 \ln [4\sqrt{2}(1-\lambda/V)^{-1/2}]/\pi(V\gamma)^{1/2}, \qquad (I.4)$$

we have

$$\Delta k = \left| \ln \varepsilon \right| \left[\ln(32\delta n / |\ln \varepsilon|) + 1/2 \right] / 8 \sim \ln(\delta n). \quad (I.5)$$

The estimate of λ_{\max} and the number of levels lying in the transition region $V < \lambda < \lambda_{\max}$ can be performed analogously.

Upon substituting (1.23) into the Schrödinger equation with the Hamiltonian of (1.22) and using the resonance approximation, we obtain the following equation in the eigenvalues of the quasienergy λ :

$$(\Delta\omega\delta_{\alpha,2} + Bl^2)b_{l,\alpha} - (Ed/4)(b_{l+1,\beta} + b_{l-1,\beta}) = \lambda b_{l,\alpha}; \quad (\text{II.1})$$

Here the $b_{l,\alpha}$ are the coefficients of the Fourier expansion of $\phi_{\alpha}(\theta)$. Let us define the vector **a** as follows:

$$a_l = b_{l,1}, l \text{ even }; a_l = b_{l,2}, l \text{ odd }.$$
 (II.2)

Then Eq. (II.1) acquires the form

$$[\Delta \omega \cdot \chi(l) + Bl^2]a_l - (Ed/4)(a_{l+1} + a_{l-1}) = \lambda a_{l^*} \quad (II.3)$$

Here $\chi(l) = 0$ for l even, $\chi(l) = 1$ for l odd. We note that Eq. (II.3) enables us to determine only half of the QFs. The second half of the QFs corresponds to the other symmetry and corresponds to a choice of the vector **a** in the form

$$a_l = b_{l,2}, l \text{ even }; \quad a_l = b_{l,1}, l \text{ odd }.$$
 (II.4)

In this case we again arrive at Eq. (II.3), where $\chi(l)$ is redefined according to the rule: $\chi(l) = 0$ for l odd, $\chi(l) = 1$ for l even.

Let us proceed to analyze Eq. (II.3). We shall derive from this exact equation in all the components of the vector **a** an approximate equation in the even components. For the odd components we shall use the identity

$$a_{l} = Ed(a_{l+1} + a_{l-1})/4(\lambda - h\Delta\omega - Bl^{2}).$$
(II.5)

Upon substituting into (II.3) we have

$$Bl^{2}a_{l} + (Ed/4)^{2}$$

$$\times \left[\frac{a_{l+2} + a_{l}}{\lambda - h\Delta\omega - B(l+1)^{2}} + \frac{a_{l} + a_{l-2}}{\lambda - h\Delta\omega - B(l-1)^{2}}\right] = \lambda a_{l}$$
(II.6)

Assuming approximately that $(l+1)^2 \approx (l-1)^2 \approx l^2$ and introducing the arbitrary function $\Phi(\theta) = (2\pi)^{-1/2} \Sigma a_l \times \exp(il\theta)$, we obtain

$$\begin{aligned} [\lambda - B\partial^2/\partial\theta^2 + h\Delta\omega/2]^2 \Phi(\theta) \\ &= [(Ed/2)^2 \cos^2\theta + (h\Delta\omega/2)^2] \Phi(\theta). \end{aligned} \tag{II.7}$$

We note that, if the vector **a** is defined according to Eq. (II.4), then the equation for the function $\Phi(\theta)$ in the approximation being used will have the same form. Thus (II.7) determines the complete set of QFs.

We are interested in the case in which the amplitude of the external field is rather large. In this case we can seek the solution of (II.7) in the quasiclassical approximation

$$\Phi(\theta) \sim \exp[i\hbar^{-1}\int I(\theta,\lambda)d\theta]).$$
(II.8)

Here we have $I(\theta,\lambda) = [\lambda - U_{1,2}(\theta)]^{1/2}$, $U_{1,2}(\theta) = h\Delta\omega/2 \pm [(Ed/2)^2 \cos^2 \theta + (h\Delta\omega/2)^2]^{1/2}$. The eigenvalues λ correspond to the quantized values of the action $J(\lambda) = (2\pi)^{-1/2} \int I(\theta,\lambda) d\theta$. Finally we note that in the quasiclassical approach the eigenfunctions of Eq. (II.7) coincide with those of the effective Hamiltonians of (1.24). Thus one can formulate the final answer in simplified form as was done in Sec. 1.7.

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ili. Taking account of relaxation processes

In this section we shall treat the dynamics of the quantum systems being discussed in the presence of strong relaxation processes. The condition of strong relaxation means that the steady-state response of the system to a periodic agent also amounts to a periodic process at the same frequency (in moderate dissipation in a classical system (and a quantum system³³) a more complex regime of motion can occur, due to the appearance in the system of a strange attractor). In particular, the induced polarization of the system as $t \to \infty$ can be represented in the form of a Fourier series

$$P(t) = d\sum_{k} [\chi_{k}^{c} \cos(k\omega t) + \chi_{k}^{s} \sin(k\omega t)]. \qquad (\text{III.1})$$

Here ω is the frequency of the external field. Thus, in the case of strong dissipation the problem reduces to studying the dependence of the susceptibilities χ_k^{cs} on the parameters of the system.

Further, one conducts the analysis within the framework of the following model. We assume that the dynamics of the system is described by the density matrix $\hat{\rho}(t)$, which obeys the equation

$$i\hbar\partial\hat{\rho}/\partial t = [\hat{H},\hat{\rho}] + \Gamma(\hat{\rho}), \quad \hat{H} = \hat{H}_0 + \hat{H}_{int}(t), \quad (III.2)$$

$$\Gamma(\hat{\rho}) = -i\hbar\eta(\hat{\rho} - \hat{\rho}_0), \quad \hat{\rho}_0 \sim \exp(-\hat{H}_0/k_BT) \quad (\text{III.3})$$

 $(\eta$ is the rate of relaxation in the equilibrium state ρ_0). We have chosen a very simple form for the relaxation term, since for the given $\Gamma(\hat{\rho})$ the solution of (III.2) can be found exactly.³⁷ Of course, there is a question of the correctness of the given form of the relaxation term, especially for high values of the external field. However, we shall not discuss here the degree of applicability of this model and shall assume that Eqs. (III.2) and (III.3), at least qualitatively, correctly describe the dynamics of the quantum system with account taken of the influence of the heat bath (e.g., molecules in the equilibrium medium of a buffer gas) for arbitrary values of the amplitude of the external field.

According to Ref. 37 the established regime of motion is described within the framework of the model of (III.2) and (III.3) by the density matrix

$$\hat{\rho}(t) = \hat{\rho}_0 + \sum_{k,l} r_{k,l} \hat{R}^{(k,l)}(t), \qquad (\text{III.4})$$

$$r_{k,l}(t) = \exp[i(\lambda_l - \lambda_k)th^{-1} - \eta t]$$

$$\times (1/i) \int dt \operatorname{Tr}\{[\hat{H}_{int}(t), \hat{\rho}_0] \hat{R}^{(k,l)}(t)\} \exp[-i(\lambda_l - \lambda_k)th^{-1} + \eta t]\}$$
(III.5)

In (III.4) we have introduced the concept of the quasienergy matrix of the system. In the coordinate representation we have $R^{(k,l)}(x',x|t) = \psi_k(x',t)\psi_l^*(x,t)$, where $\psi_k(x,t)$ is the QF of the Hamiltonian \hat{H} corresponding to the quasienergy value λ_k , and x is the set of coordinates of the system. We note that the coefficients $r_{k,l}$ and the matrices $\hat{R}^{(k,l)}$ are periodic functions of the time. Thus the expansion (III.1) is valid for the polarization of the system ($P(t) = \text{Tr}(\hat{d}\hat{\rho})$, where \hat{d} is the dipole-moment operator). Equations (III.4) and (III.5) imply that the properties of the QFs and QS of

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the system. We present below the results of calculating the susceptibility for the model of the vibrational degree of freedom of a molecule in an infrared field (see Sec. 1.2):

$$H = h\Omega(\hat{a}^{+}\hat{a} + 1/2) - \gamma h^{2}(\hat{a}^{+}\hat{a})^{2} - (Ed/2)[\hat{a}^{+}\exp(-i\omega t) + \hat{a}\exp(i\omega t)] (III.6)$$

and the model of the rotational degree of freedom of a molecule in a UHF field (Sec. 2.3):

$$\widehat{H} = -(\gamma h^2/2)\partial^2/\partial\theta^2 - Ed\cos\theta \cdot \cos\omega t, \qquad (\text{III.7})$$

the structure of the QFs of which has been discussed in detail in the review.

Using the properties of the QFs of the completely integrable system of (III.6), we can show that in Eq. (III.1) only the $\chi_1^{(c,s)}$ differ from zero. In particular, χ_1^s , which determines the form of the absorption line of the system, is found by the formula

$$\begin{split} \chi_{1}^{8} &= (Ed/2h) \sum_{l > k} f_{k,l}(T) f_{k,l}^{\prime} \{ \eta / [(\lambda_{l} - \lambda_{k})^{2} / h^{2} + \eta^{2}] \}, \quad (\text{III.8}) \\ f_{k,l}(T) &= \sum_{n} n^{1/2} (\rho_{n}^{(0)} - \rho_{n-1}^{(0)}) (R_{n-1,n}^{(k,l)} - R_{n,n-1}^{(k,l)}), \\ f_{k,l}^{\prime} &= \sum_{n} n^{1/2} (R_{n-1,n}^{(k,l)} - R_{n,n-1}^{(k,l)}). \end{split}$$

Here the $R_{n,m}^{(k,l)}$ are the elements of the quasienergy matrix. It is convenient to render the values of the quasienergy dimensionless by using the value of the nonlinearity γ . Then Eqs. (III.8) and (III.6) imply that χ_1^s is a function only of the dimensionless magnitude of the perturbation

FIG. 28. Form of the absorption line for the model of the vibrational degree of freedom of the molecule of Eq. (III.6): $\Delta = (\Omega - \omega)/\gamma h$ is the dimensionless detuning. The value of the dimensionless perturbation parameter $W = -Ed/\gamma h^2$ is: a: W = 0.1, b: W = 10. Parameters of the heat bath: $\tilde{\eta} = \eta/\gamma h = 0.1$, $h\Omega/k_B T = 1$ (From Ref. 37.)

 $W = -Ed/\gamma h^2,$ the dimensionless detuning $\Delta = (\Omega - \omega)/\gamma h$, and the dimensionless rate of relaxation $\tilde{\eta} = \eta / \gamma h$. A graph of χ_1^s (*W*, Δ) as a function of Δ is shown in Fig. 28 for two values of the parameter W: a) W = 0.2; b) W = 10. Figure 28a serves as a test; the peaks correspond to the transitions 0-1, 1-2, etc., while the width of the peaks is determined by the value of $\tilde{\eta}$, which was chosen equal to 0.1. Case (b) reflects the characteristic form of a ONR line. The form of the absorption line can be explained on the basis of the properties of the QFs and QS of the quantum nonlinear resonance. The spectrum of quasienergies of the system (III.6) is shown in Fig. 29 (the value of the quasienergies is plotted in units of γh^2). The several first lines with practically equal splitting correspond to the QFs of the "center" of the QNR. The degree of delocalization of these QFs is determined by the width of the QNR, while the distance between the quasienergy levels is determined by the phase frequency of the ONR. These states form a broad, smooth curve (see Fig. 28b) with a characteristic width in dimensioned variables of the order of $\gamma h \delta n \approx \Omega_{\rm ph}$. The quasicrossings of the lines in Fig. 29 correspond to one-photon, two-photon, etc., resonances. These quasienergy states determine the resonance peaks on the right-hand slope of the line. With increasing intensity of the external field, the relative height of the peaks diminishes. In closing this paragraph we note that, in contrast to the pure quantum case W = 0.1 (curve in Fig. 28a), the width of the QNR lines does not depend on the value of η if $\eta < \Omega_{\rm ph}$.

Let us proceed to discuss the system of (III.7). As we have already noted repeatedly, the fundamental qualitative difference of the QFs of a nonintegrable from an integrable system is the property of two-dimensional delocalization



FIG. 29. Spectrum of quasienergies of the system of Eq. (III.6) for W = 10 as a function of Δ . The magnitude of the splitting of the several first levels is determined by the value of the phase frequency of the QNR. (From Ref. 37.)



FIG. 30. Values of the coefficients χ_k^c for the system of Eq. (III.7) as functions of the magnitude of the perturbation V = -Ed/h. Values of the parameters: $\gamma h = 1$, $\omega = 2$, $\eta = 0.1$, $k_B T = 0.5$. (From Ref. 28.)

(see Sec. 2.5). As applied to the problem discussed in this section, this property is reflected in the anomalously large number of effectively nonzero terms in Eq. (III.1). This number is determined approximately by the degree of delocalization of the QFs along the m axis. Figure 30 shows the results of numerical calculation of χ_k^c for the following values of the parameters of the system (III.7): $\gamma h = 1, \omega = 2$, 1 < V < 16 (V = Ed/h) and parameters of the heat bath: $\eta = 0.1, k_{\rm B}T = 0.5$. Upon varying V in the chosen interval, the overlap parameter of the resonances K of (2.2) varies in the range K = 0.7-2.8. We see from Fig. 30 that, when K > 1(V>2), a large number of harmonics is generated (owing to the symmetry properties, the even harmonics are absent). We also see that the concept of the number of harmonics is well defined.

Apparently the generation of a large number of harmonics is one of the most marked manifestations of quantum chaos. We can propose the following experimental scheme to verify this effect. We fix the amplitude of the external field at a value that ensures the fulfillment of the condition of quasiclassicality $\delta n \ge 1$, and vary the frequency so as to alter the stochasticity parameter K. When $K \ll 1$ we shall observe the generation of only the first harmonic. As the frequency of the field is reduced, we expect at $K \approx 1$ a rapid growth in the number of generated harmonics. We should expect the maximum number of harmonics at $K \approx 4.5$. This value of K corresponds to the "most chaotic" regime if we examine the system of (III.7) from the standpoint of classical mechanics, and correspondingly, to the greatest degree of two-dimensional delocalization of the QFs in the quantum-mechanical case.

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