Statistics of energy spectra

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When there is a stochastic disruption of the integrals of motion of a system, the corresponding quantum numbers disappear. The energy spectrum of the system becomes quasirandom. In the present paper, the quantization rules and the distribution of distances between pairs of adjacent levels are studied for this case. The probability for the appearance of very closely spaced levels is governed by a critical index which is expressed in terms of the Kolmogorov entropy for the given system (i.e., in terms of the growth rate for the instability of the classical trajectories in the corresponding phase space). Various physical situations are discussed in which a random spectral structure can arise. The capabilities of a quasiclassical analysis in the case of a stochastic disruption of the integrals of motion are discussed.

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INTRODUCTION

There is now a huge literature on the nucleation of stochastic properties in classical dynamic systems with few degrees of freedom. In contrast, work on this question for quantum systems is only beginning. The interest in this question stems from not only practical applications and new phenomena but also the need to develop a special quantum-mechanical description of systems which exhibit a stochastic instability. The present review will be restricted to that part of the problem which is associated with the structure of the energy spectrum, i.e., the quantization rules. The purpose here is to review systematically the results which have been established in various branches of physics and to draw attention to these results.

An approach which has long been used in a variety of physical problems is to transform from a determinate description of certain properties of the system to a statistical description of these properties in cases in which small changes in parameters lead to such pronounced changes in these properties that a detailed description of these properties would become meaningless. The idea of introducing a statistical description of the energy spectrum of a complicated system can be credited to Wigner, Landau, and Smorodinskiĭ, who studied excited states of heavy nuclei. These ideas are formally embodied in the theories of Wigner, Porter and Rosenzweig, and Dyson. In these papers, a certain hypothesis is introduced regarding the nature of the statistical ensemble of energy levels of the system. This approach sidesteps at least two questions of fundamental importance: *When* and *why* should a statistical description of the spectrum of the system arise? Research over the past decade has not only answered

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these questions but has also led to important changes in the theoretical analysis of the characteristics of the level distribution. At the same time, it has become possible to extend substantially the class of physical objects in which a statistical spectral structure should arise under certain conditions. To review this research is the purpose of the present paper.

1. ORIGINS OF THE PROBLEM

Our problem arose at the intersection of several directions in research on the spectral properties of very complicated systems. We shall specify below what is meant by a "complicated" system or a "complicated" spectrum of a system, but no matter how hard we try to complicate the spectral structure it is not a simple matter to imagine which fundamental characteristics of the motion of the system should be associated with the spectral structure. Perhaps for this reason it would be useful to examine those logical ideas which underlie the theory of the energy spectrum of "complicated" quantum systems.

(a) Einstein's comment

We should begin the historical review of the question with Einstein's 1917 paper[1] on quasiclassical quantization rules.

The quantization rules (the Bohr–Sommerfeld rules)

$$\frac{1}{2\pi}\oint p_i\,dq_i = n_i\hbar \qquad (i=1,\ 2,\ \ldots,\ M), \tag{1}$$

where M is the number of degrees of freedom, were known at the time. Einstein found the rules in (1) to be unsatisfactory because they were applicable only if the variables in the system could be completely separated, since only in this case was it possible to choose p_i and q_i such that the expressions $\oint p_i dq_i$ would be invariants of the motion. These expressions must be invariants, since the quantum numbers must be integrals of motion. Since this property of separation of variables was totally unrelated to the quantum problem proper, Einstein proposed a different quantization method:

$$S_k \equiv \frac{1}{2\pi}\int\limits_{C_k}\sum_{i=1}^{M} p_i\,dq_i = n_k\hbar \qquad (k=1,\ 2,\ \ldots,\ M), \tag{2}$$

where the contours C_k will be specified below. The quantity $\sum_{i=1}^{M} p_i dq_i$ is known to be one of the Poincaré integral invariants.[2] Consequently, S_k is an invariant. Then if there exist precisely M independent, single-valued integrals of motion (i.e., if the number of such integrals of motion is precisely equal to the number of degrees of freedom), then the expression $\sum_{i=1}^{M} p_i dq_i$ is a total differential. A proof can be found in Einstein's paper[1] or, in more modern form, in Arnol'd's book.[3] Then with a complete set of independent, single-valued integrals of motion, the quantity S_k depends on only the positions of the ends of the integration contour and is independent of the shape of this contour. If we consider only closed contours C_k, then we have $S_k = 0$ in those cases in which the contour C_k can be shrunk to a point. Nonvanishing values of S_k can be found only if the space containing the C_k is multiply connected, and the phase space of the system is precisely of this nature. Let us examine the motion in this space in more detail.

If the variables can be separated, then there exist M integrals of motion, as which we can choose the actions

$$I_k = \frac{1}{2\pi}\oint p_k\,dq_k \qquad (k=1,\ 2,\ \ldots,\ M).$$

In this case we have

$$H = \sum_{k=1}^{M} H_k(I_k),$$

and the equations of motion are

$$\dot{I}_k = -\frac{\partial H}{\partial \vartheta_k} = 0, \quad \dot{\vartheta}_k = \frac{\partial H}{\partial I_k} = \frac{\partial H_k}{\partial I_k} = \omega_k(I_k) \qquad (k=1,\ 2,\ \ldots,\ M).$$

More generally, there are M integrals of motion $(F_1, F_2, \ldots, F_M)$, but the variables cannot be separated. In this case we can use (2) to determine M new integrals of motion $(S_1, S_2, \ldots, S_M)$ and write

$$F_i = F_i(S_1,\ S_2,\ \ldots,\ S_M) \quad (i=1,\ 2,\ \ldots,\ M).$$

This transformation exists; furthermore, the variables (S_k, ϑ_k) are canonically conjugate pairs,[3]

$$\begin{aligned} &H = H(S_1,\ S_2,\ \ldots,\ S_M),\\ &\dot{S}_k = -\frac{\partial H}{\partial \vartheta_k} = 0; \quad \dot{\vartheta}_k = \frac{\partial H}{\partial S_k} = \omega_k(S_1,\ S_2,\ \ldots,\ S_M). \end{aligned} \tag{3}$$

It follows from (3) that the motion occurs along an M-dimensional torus with frequencies ω_k. In particular, in the case $M=2$, the trajectory is wound around a two-dimensional torus (Fig. 1). On this torus we can construct two closed basis contours (Fig. 2), which can be neither shrunk to a point nor reduced to each other. We call such contours "irreducible." On an M-dimensional torus it is possible to construct precisely M irreducible contours C_k $(k=1,2,\ldots,M)$. These contours should also be chosen for (2), so that we have M equations of the type

$$S_k(F_1,\ F_2,\ \ldots,\ F_M) = n_k\hbar \qquad (k=1,\ 2,\ \ldots,\ M), \tag{4}$$

which determine the quantization rules on the action in the case of nonseparable variables.

Einstein's quantization rules were subsequently derived and refined through nonquasiclassical corrections in papers by Keller,[4] Balian and Bloch,[5,6] Berry and Mount,[7] and, in most rigorous form, Maslov[1)] (Refs. 8 and 9). All these refinements deal with the case in which the number of integrals of motion is equal to the number of degrees of freedom.

Einstein concluded his paper[1] with the comment that there might be cases in which the number of integrals of motion was less than the number of degrees of freedom, as, for example, in the three-body problem (Einstein cited Poincaré's results). What should the

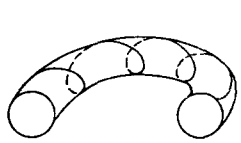

FIG. 1. Invariant torus of a system with two degrees of freedom.

[1)]There is an elementary derivation of quantization rule (4) in Ref. 29.

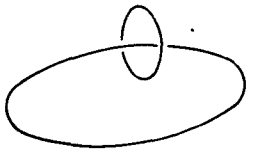

FIG. 2. Irreducible contours in the case of two degrees of freedom.

quantization roles be in this case? Many years were to pass before information was acquired on those properties of dynamic systems which were necessary for answering this question.

(b) Stochastic violation of the integrals of motion

When we say that a trajectory in phase space is wound around a torus, we of course mean that the torus itself is an invariant. If a system has invariant tori, then the incorporation of an additional interaction between different degrees of freedom can lead to a disruption of the invariant tori. The Kolmogorov–Arnol'd–Moser (KAM) theory[10-12] led to the result that sufficiently small perturbations could conserve invariant tori.

If the perturbations are large, the invariant tori are generally disrupted, and the disruption occurs (in apparently all cases) in a stochastic manner. This assertion requires a rather detailed explanation. Let us imagine a phase volume in which a system can execute a finite motion (such a volume is shown schematically in Fig. 3). If the initial conditions fall in certain regions of the phase space (as marked by the heavy curves in Fig. 3), then the trajectories in these regions are closed. Correspondingly, there is a nontrivial integral of motion (for example, the area bounded by this trajectory). Outside these regions, which are referred to as "stability islands," the motion is random; by this we mean that the motion has two properties: It is ergodic and it is mixed (i.e., there is an uncoupling of the correlations between certain variables).

A more graphic interpretation of the onset of stochastic conditions is usually associated with an instability of the phase-space trajectories with respect to an arbitrarily small perturbation in the initial conditions. In this instability, the distance between trajectories increases exponentially over time:

$$D = D_0 \exp(ht). \tag{5}$$

Because of this behavior of the trajectories, a droplet in the phase space (Fig. 4a) rapidly assumes a very complicated shape (Fig. 4b) and uniformly covers the phase space. The quantity h in Eq. (5) is called the

FIG. 3. Partitioning of the phase space into stability islands and a "stochastic sea."

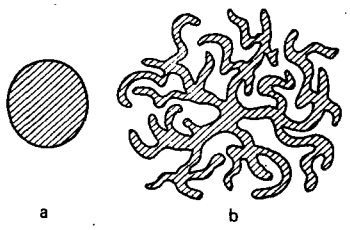

FIG. 4. Spreading of a phase droplet in stochastically unstable motion.

"Kolmogorov entropy" (more precisely, the Kolmogorov entropy[13,14] is an average of h over the phase space, multiplied by a constant of the order of unity[15]). An exact equation for the Kolmogorov entropy was derived by Sinai.[6] Dynamic systems exhibiting the mixing property (i.e., $h>0$) are called "K systems."[2)] In a real situation, the K system is somewhat of an abstraction. In the typical problems which arise in physics, there are always stability islands on which $h=0$. These islands may be arbitrarily small and may "fill" the entire phase space in a very complicated way. Their measure, however, may be very small in comparison with the measure of the region of stochastic motion, and it is in this sense that the term "stochastic system" is used below. Ordinarily, the transition from dynamic or regular motion in a system to stochastic motion is governed by the critical values of certain parameters; most commonly, the parameter is the energy of the system (see, for example, the review in Ref. 16). Let us examine some examples of the onset of stochastic conditions.

Example 1. The Hénon–Heiles Model.[17] The Hamiltonian of a two-particle system is

$$H=\tfrac{1}{2}(\dot{x}^2+\dot{y}^2)+\tfrac{1}{2}(x^2+y^2)+x^2y-\tfrac{1}{3}y^3. \tag{6}$$

Hénon and Heiles[17] have carried out a numerical analysis of the motion described by Eq. (6), working in the following manner: Points of the trajectory corresponding to a certain energy $H=E$ are noted on the $(\dot{y},y)$ plane with $x=0$. For sufficiently small values of E, these points are grouped in a family of closed curves ($E=0.0833$ in Fig. 5), and this situation corresponds to the existence of an additional integral of motion (other

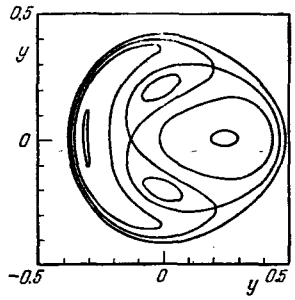

FIG. 5. Phase trajectories in the Hénon–Heiles model (the stable energy region).

[2)]Strictly speaking, the definition of K systems given by Kolmogorov[13] also includes the requirement of a certain dimensionality of the mixing of trajectories in different parts of phase space. In the physics literature, however, the concept of K systems is used in a wider sense, and it presupposes only the existence of a mixing of trajectories, i.e., a local instability of these trajectories.[16]

than the energy). If $E>1/12$, some of the closed curves begin to break up. Figure 6 shows the trajectories for the case $E=0.125$. Parts of the trajectories become stochastic, but the region of stability islands is still quite large. With a further increase in E, the islands "melt," and at $E=0.1667$ nearly the entire phase volume is in the region of stochastic motion.

Example 2. The Sinaĭ billiard-ball system. Hopf[18] has shown that the particle motion in a space with a negative curvature exhibits mixing. Krylov generalized this result to prove the stochastic nature of the motion of hard spheres which collide by absolutely elastic collisions.[19] Sinaĭ[20,21] carried out a rigorous analysis of billiard-ball systems. Some examples of billiard-ball systems in which the particle motion is stochastic are shown in Figs. 7a (for a "star-shaped" billiard table) and 7b (for a "caterpillar-shaped" table). An originally parallel beam of trajectories in such a system rapidly diverges. The divergence in phase space is described by a law which satisfies condition (5).

Example 3. Gliding electrons. The motion of electrons which are "jumping" on a metal surface and whose trajectories are twisted by a magnetic field directed parallel to the metal surface is stochastic if the surface has a negative curvature and if the electron energy is above a certain critical value (Fig. 8; the heavy curve is the surface, and the magnetic field is perpendicular to the plane of the figure). This system is analogous to the caterpillar-shaped billiard table. The criterion for stochastic motion is[22]

$$R>\frac{1}{2}\frac{a^2}{b}\equiv R_0, \tag{7}$$

where R is the electron gyroradius, a is the period of the surface inhomogeneity, and b is the inhomogeneity height ($b\ll a$). Condition (7) can be rewritten

$$E>E_0=\frac{1}{2}mR_0^2\Omega^2=\frac{1}{8}\frac{m\Omega^2a^4}{b^2}, \tag{8}$$

where E is the energy, E_0 is the critical energy (the boundary of the stochastic region), and Ω is the cyclotron frequency.

Examples 1 and 3 lead us to some unexpected questions. Up to certain values of the energy, the quantization of the corresponding Hamiltonians can be reduced to certain standard systems. But how are we to quantize the same Hamiltonian at energies above the critical energy E_0? There is only a single integral of motion remaining in this region (the energy), so that we are immediately led back to Einstein's comment, but now with a completely different meaning: What should we find for the spectrum of a system in which some of the integrals of motion are disrupted in a stochastic manner? An analogous question arises in the determination of the oscillation spectrum of membranes or resonators which are shaped as in Fig. 7. Since there exists an isomorphism of trajectories for example 2, and for examples 1 and 3 in the case[3] $E>E_0$, the problem formulated above reduces to the problem of quantizing the Sinaĭ billiard-ball system.

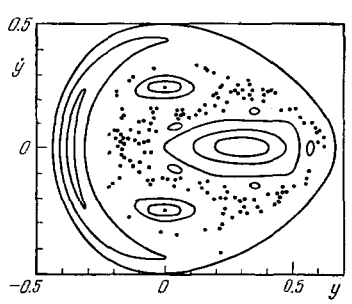

FIG. 6. Beginning of the stochastic disruption of trajectories in the Hénon–Heiles model.

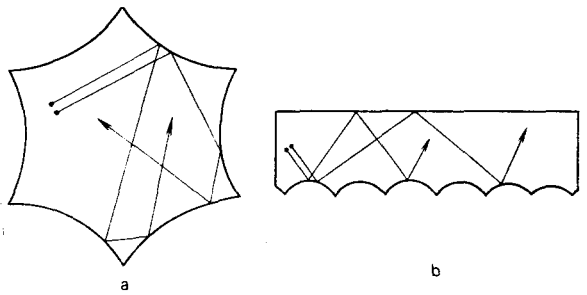

FIG. 7. Examples of negative-curvature billiard-ball systems. a) "Star"; b) "Caterpillar."

(c) Dyson's theory

In this section we take up another question, which developed independently of those discussed above, but which bear directly on the problem formulated above, as will become clear.

At a high excitation energy of a heavy nucleus, the energy-level structure becomes distorted, and the distortion is so pronounced and irregular that a statistical description becomes a better description of the actual situation. In introducing this idea, Wigner[23] and Landau and Smorodinskiĭ[24] pointed out yet another fundamental consequence of the unusual approach of treating the energy as a random variable: For levels of a given symmetry, the probability $P(E;\Delta E)$ for finding two adjacent levels near energy E, within a separation ΔE, must approach zero in the limit $\Delta E\to 0$ (the "level repulsion principle"). This principle means that, because of the strong correlation between adjacent levels, the distribution of distances between levels cannot correspond to ordinary probability distributions, e.g., the Poisson distribution $\exp(-|\Delta E|)$ or the Gaussian distribution $\exp[-(\Delta E)^2]$.

Wigner, Porter, and Dyson derived a statistical theory for the level distribution. Most of the work in this field (through 1965) is collected in Ref. 25, and it is analyzed in the excellent review by Porter.[26] Just as a certain hypothesis regarding the statistical ensemble of states is introduced in statistical mechanics, it is hypothesized in the statistical theory of the spectrum that there is an equivalence between the distribution of lev-

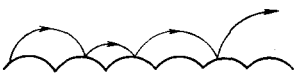

FIG. 8. Trajectories of electrons which are being twisted by a magnetic field near an inhomogeneous reflecting surface.

[3] Strictly speaking, this isomorphism exists within an error corresponding to the effect of the stability islands.

els E_k and eigenvalues λ_k of the ensemble of random matrices of a certain symmetry (we shall refer to this hypothesis below as the "λ–E equivalence hypothesis"). Wigner[23] and Porter and Rosenzweig[26] dealt with a Gaussian ensemble of random matrices. The work in this field was completed formally by Dyson,[27] who treated ensembles of random matrices—orthogonal, unitary, and simplicial. The basic result of that research was the following equation for the probability distribution for the distances ΔE between adjacent levels in the part of the spectrum near energies E:

$$P(E;\ \Delta E) = A\ |\ \Delta E\ |^{\alpha} \exp\left[-B\ (\Delta E)^2\right]. \tag{9}$$

Here A and B are certain functions which vary slowly with E, and the critical index α is 1, 2, or 4, depending on the type of symmetry of the system. Wigner's theory leads to an equation similar to (9), but with slightly different values of A and B. In the limit $\Delta E \to 0$ we have

$$P(E;\ \Delta E) \sim |\ \Delta E\ |^{\alpha}, \tag{10}$$

and this structure of P is the most important aspect of the situation, since it determines the level repulsion law.

Admittedly, the λ–E equivalence hypothesis is not obvious. The basic argument for this hypothesis stems from the circumstance that the eigenvalue distribution of the ensemble of random matrices is characterized by a "repulsion," i.e., by the same property as should be exhibited by the level distribution. However, there is a fundamental question which remains unresolved: Just which properties of the interactions in the system should lead to the random spectral structure? It is pertinent to note here that at the time Wigner's and Dyson's papers were appearing the concept of the "complexity" of an interaction was at an extremely naive level. In particular, it was assumed that this was a characteristic property of systems having many degrees of freedom. It is now clear (see below) that the spectrum of a system may exhibit statistical properties even if there are only two (!) degrees of freedom. Consequently, there are at least two questions which arise in connection with the hypothesis of λ–E equivalence:

1. How can the function $P(E;\Delta E)$ be derived from first principles, i.e., from the equations of motion, and how do we determine whether distributions of the type in (9) and (10) actually exist?

2. Does the critical index α in the repulsion law actually depend on only the symmetry of the system? (Is it independent of the particular properties of the system?)

(d) Disordered systems

To answer the first of these questions, we must at least know those physical factors which can lead to a random distribution of levels. During the period of Wigner's and Dyson's work, the lack of information on these factors was dealt with by introducing the concept (also somewhat vague) of the existence of an "interaction black box." In this situation it was natural to turn to a system for which certain quantities have a known random distribution. The first attempt of this type was made by Pokrovskiĭ[28] for a one-dimensional disordered system.

The Hamiltonian of the disordered system,

$$H = \frac{p^2}{2m} + V(x),$$

contains the potential $V(x)$ which is some random process. For example, it may be

$$V(x) = V_0 \sum_k \delta(x - x_k),$$

where x_k have a Poisson distribution. One of the central problems in research on disordered systems is that of determining the density of states[62-64] $\rho(E)$. The distribution of distances between levels $P(E;\Delta E)$ is a more subtle characteristic, and in the one-dimensional case with $V_0 > 0$ Pogrovskiĭ found

$$P(E;\ \Delta E) \propto \exp\left[-\frac{\text{const}}{(\Delta E)^2}\right], \tag{11}$$

i.e., an exponential level repulsion law. This result is meaningful only for sufficiently high energies. At high values of E, the random potential $V(x)$ is a perturbation, so that the energy spectrum should be similar to the unperturbed spectrum [with the maximum (infinite) repulsion] in which the adjacent levels are separated by a distance $\sim 1/L$ (L is the dimension of the system). Molchanov has derived the distribution of level separations for the same system, but for small values of E.

Let us assume that ΔE is the distance between adjacent levels near the energy E. Then there exists a limit

$$\lim_{L\to\infty} (L\Delta E).$$

Molchanov's result reduces to

$$P(E;\ L\,\Delta E > \delta) \propto \exp\left[-\rho(E)\,\delta\right],$$

which corresponds to a Poisson level distribution. The absence of repulsion at low energies can be understood on the basis of the following qualitative considerations, which are also credited to Molchanov.

In the one-dimensional case, the entire energy spectrum of a disordered system is a point spectrum, and the states are localized. Lifshits[62] and Mott[63] began the discussion of these properties of the spectrum, and rigorous proof was first found in Ref. 66. It follows from the localization of states that the wave functions of the states fall off exponentially in the limits[67] $x \to \pm\infty$. It follows further that all the states have a certain localization region Δx in space. Here it is important to note that the localization property is a property exhibited by all states (with a unit probability). At small values of E,

$$\Delta x \sim \frac{2\pi\hbar}{\sqrt{2mE}},$$

while we have $\Delta x \to \infty$ in the limit $E \to 0$. The characteristic width of the potential well in which the state is localized is thus large at small values of E and corresponds to an improbable fluctuation of the potential $V(x)$ during which a large "hole" of width $\sim \Delta x$ is formed.[62] The effective shape of the corresponding potential was found in Ref. 65. The appearance of two such holes with only a small separation along the x axis is improbable,

so that the regions in which the levels are localized are relatively far apart along the x axis and essentially do not overlap. It follows that there is no interaction between localized states at small values of E, so that there is no repulsion.

The nature of the level distribution in disordered two-dimensional and three-dimensional systems and the case of intermediate energies in one-dimensional systems are clearly interesting problems, which have not yet been solved, but even with the information available we can see that the relationship between the resulting distribution and an equation like (9) is too weak. In comparison with (9), the level repulsion in a one-dimensional disordered system is too weak at low energies and too strong at high energies. This conclusion is not surprising, since the dynamic properties of a one-dimensional system are fundamentally different from those of a system of strongly interacting particles.

2. SPECTRAL STRUCTURE UPON A VIOLATION OF THE INTEGRALS OF MOTION

(a) Formulation of the response

The preceding chapter contains most of the information required not only to understand the formulation of the problem but also to resolve it. An equation for $P(E;\Delta E)$ in the stochastic region was derived by Zaslavskiĭ and Filonenko[22] for the gliding-electron model for two asymptotic regions: $\Delta E \ll \varepsilon$ and $\Delta E \gg \varepsilon$, where ε is the average distance between levels. It was later shown[9] that a universal equation holds in the limit $\Delta E \to 0$.

Let us assume $E > E_0$ and that the energy integral is the only single-valued integral of motion; all the other integrals are stochastically violated. Then the level repulsion law becomes[29]

$$P(E;\ \Delta E) \sim \text{const} \cdot |\Delta E|^{\text{const}/h} \qquad (\Delta E \ll \varepsilon), \tag{12}$$

where all the constants are function of E, and $h = h(E)$ has the same meaning as in Eq. (5). In other words, this is the Kolmogorov entropy or the effective growth rate for the instability of the trajectories in phase space. The law in (12) has the same structure (i.e., a power-law variation with $|\Delta E|$) as the distribution in the Dyson theory, (10), but the critical index here is governed by the value of h, which characterizes the properties of the local instability of the system.[4)]

Equation (12) determines the distribution of distances between levels in the case $\Delta E \ll \varepsilon$. A distribution was derived in Ref. 22 for the opposite limit, $\Delta E \gg \varepsilon$ [see Example 3 in section 2(a)]:

$$P(E;\ \Delta E) \propto \exp\left[-\text{const} \cdot \left(\frac{\Delta E}{\varepsilon}\right)^2\right],$$

i.e., a Gaussian law. This equation was derived on the basis of quite general considerations, so it, like Eq. (12), is presumably a universal equation for the case of a stochastic disruption of the integrals of motion.

[4)]According to (5), the quantity h corresponds to the growth rate for the local instability and can easily be estimated in order of magnitude for real systems by linearizing the equation of motion.[16]

Equation (12) will be derived in the following section; here we will examine in more detail the general structure of the level distribution.

(b) Characteristics of the spectral structure

Since the nature of the quantization and the energy-level distribution itself are strongly affected by the properties of the trajectories of the classical particles, we are led to the concept[30] of a classification of levels in various series. Each series of levels is associated with a certain class of classical trajectories. The different series of levels may overlap each other in a very complicated manner, so that even if it is possible to determine all the series of the energy spectrum it may be a rather complicated problem to arrange the levels in order of increasing energy.

Percival[31] suggested the name "regular spectrum" for those levels which correspond to nominally periodic motions of the classical particle, while those levels corresponding to the stochastic motion would constitute the "irregular spectrum." For the classical trajectories corresponding to Fig. 3, for example, it is possible to distinguish one series corresponding to an irregular spectrum and three series of regular components, which correspond to motion on different stability islands.

Since the subject of the present review is the irregular series of the energy spectrum, we should take special note of the relationship between the level statistics and an ensemble in ordinary statistical physics. The level system of an irregular spectrum cannot be the same representative of an ensemble as, for example, any state of a many-body system. This circumstance was pointed out by Dyson.[27] The exact description is discarded, not for the system of levels, but for a system in which the nature of the interactions is very complicated and whose spectrum is to be determined. We shall see below that excited molecules constitute a system of this type and that an exact determination of the states of such a system is just as meaningless as the determination of the coordinates of a large number of particles. The energy spectrum of excited molecules is a subtle characteristic of a system, and a probabilistic description of the states of a system automatically leads to the appearance of probabilistic properties in the spectrum. For example, for the star-shaped billiard table in Fig. 7a, a statistical ensemble might be formed by stars with a small scatter in geometric characteristics. The general nature of the trajectories in such systems is independent of the detailed properties of this scatter, so that the spectral structure is also independent of this scatter. Then each particular star geometry can serve as a representative of an ensemble which generates a certain system of levels. This method of introducing a representative of an ensemble of levels corresponds precisely to a real physical situation, although it is apparently not the only possible method. For example, in the excitation of molecules or nuclei, an ensemble may be formed as the result of a small spread of the excited states of different molecules or nuclei.

3. PROBABILITY FOR THE APPEARANCE OF CLOSELY SPACED LEVELS

(a) Assumptions

In this section we shall determine the probability $P(E;\Delta E)$ for the appearance of a level with an energy $E+\Delta E$ in the interval dE if we know that there is an eigenstate with an energy E in the interval dE. We shall be interested in the asymptotic behavior in the limit $\Delta E \to 0$. This problem was solved in Ref. 22 for the case of gliding electrons through the use of the quasiclassical asymptotic behavior for the wave functions. It was shown in Ref. 19 that the results of Ref. 22 are much more general in nature. The derivation below basically follows Ref. 29.

Let us specify some general conditions under which $P(E;\Delta E)$ is determined. We shall assume below that all the integrals of motion are stochastically violated, except for the energy interval. In other words, the system is isomorphic with the Sinaĭ billiard system. Furthermore, we assume for simplicity that the stochastic trajectories cover the region of motion in a fairly uniform manner (the distribution function is approximately constant). For the star-shaped or caterpillar-shaped billiard tables, this assumption means, for example, that there are many arcs with a large curvature. Finally, we are interested only in the quasiclassical asymptotic behavior, for very large eigenenergies E (the energy levels are numbered in order of increasing eigenvalue E).

(b) Quantization rules

The direct method for analyzing the energy eigenvalues E_k involves a determination of the response function $g(E)$, given by

$$g(E)=\int dqG(q,\ q,\ E)=\sum_k \frac{1}{E-E_k}, \tag{13}$$

where $G(q'',q',E)$ is the Green's function of the stationary Schrödinger equation, and q represents the set of coordinates. In the quasiclassical approximation, G is

$$G(q'',\ q',\ E)=A\exp\left\{i\left[\frac{1}{\hbar}S(q'',\ q',\ E)+\psi\right]\right\}, \tag{14}$$

where the action S is given by

$$S(q'',\ q',\ E)=\sum_{i=1}^{M}\int_{q'}^{q''} dq_i p_i(q,\ E), \tag{15}$$

and the factor A and the phase ψ can be determined. Since A and ψ are slowly varying functions of the coordinates and make small corrections to the quantization rules, we shall restrict this discussion to the approximation in which ψ and the change in A are negligible. This assumption is always valid in the limit $\hbar \to 0$.

We can simplify Eq. (13) by using the condition $S/\hbar \gg 1$ and making use of the fact that there is a rapidly oscillating function in the integral in (13). The condition for an extremum of $S(q,q,E)$ is

$$\frac{\partial S(q,\ q,\ E)}{\partial q_i}=\left[\frac{\partial S(q'',\ q',\ E)}{\partial q_i''}+\frac{\partial S(q'',\ q',\ E)}{\partial q_i'}\right]_{q_i''=q_i'=q_i} = p_i''-p_i'=0 \qquad (i=1,\ 2,\ \ldots,\ M),$$

where p_i' and p_i'' are the momenta at the initial and final points, respectively, of the trajectory. The equality of these momenta means that at the extremum the action $S(q,q,E)$ is

$$S_e(q)=\sum_{i=1}^{M}\oint_q dq_i p_i(q,\ E),$$

where the integration is carried out over a closed contour in phase space which passes through the point q on the surface corresponding to the point E. It follows that $g(E)$ can be written as

$$g(E)\approx A_1\int dq\exp\left[\frac{i}{\hbar}S_e(q)\right], \tag{16}$$

where A_1 is some new constant.

Since the trajectories of a classical particle constitute a random process, it is meaningless to speak in terms of closed trajectories with a measure of zero. We shall instead discuss trajectories which emerge from a small volume $\Delta\Gamma$ of phase space and which return to the same volume. This procedure corresponds to a "coarsening" operation similar to that used in statistical mechanics. Then in (16) we make the substitution

$$\exp\left\{\frac{i}{\hbar}S_e(q)\right\}\to\frac{1}{\Delta\Gamma}\sum_{C(\Delta\Gamma)}\exp\left[\frac{i}{\hbar}S_e(q)\right], \tag{17}$$

where the sum is over all the contours $C(\Delta\Gamma)$ which begin and end in the coarsening volume $\Delta\Gamma$. Since the particle distribution function is homogeneous, the right side of (17) is independent of q, so that (16) can be rewritten as

$$g(E)\approx A_1 V\frac{1}{\Delta\Gamma}\sum_{C(\Delta\Gamma)}\exp\left(\frac{i}{\hbar}S_e\right), \tag{18}$$

where V is the volume. The set of trajectories which emerge from $\Delta\Gamma$ and which return to $\Delta\Gamma$ consists of (a) those trajectories which leave $\Delta\Gamma$ and which return to $\Delta\Gamma$ after a certain time, (b) those trajectories which execute two such cycles, etc., up to an infinite number of cycles. We denote by

$$S^{(m)}(E)=\sum_{i=1}^{M}\int_{C_m(\Delta\Gamma)} p_i\,dq_i \tag{19}$$

the action in the m-th cycle, along the contours $C_m(\Delta\Gamma)$. Then (18) can be rewritten as

$$g(E)=A_1V\left\{1+\frac{1}{\mu\Delta\Gamma}\sum_{C_1(\Delta\Gamma)}\exp\left[\frac{i}{\hbar}S^{(1)}(E)\right]+\frac{1}{\mu\Delta\Gamma}\sum_{C_1(\Delta\Gamma)}\frac{1}{\mu\Delta\Gamma}\sum_{C_2(\Delta\Gamma)}\times\exp\left(\frac{i}{\hbar}[S^{(1)}(E)+S^{(2)}(E)]\right)+\ldots+\frac{1}{\mu\Delta\Gamma}\sum_{C_1\Delta(\Gamma)}\frac{1}{\mu\Delta\Gamma}\times\sum_{C_2(\Delta\Gamma)}\ldots\frac{1}{\mu\Delta\Gamma}\sum_{C_\infty(\Delta\Gamma)}\exp\left(\frac{i}{\hbar}[S^{(1)}(E)+S^{(2)}(E)+\ldots+S^{(\infty)}(E)]\right)\right\}. \tag{20}$$

Here the factor μ, whose dimensions correspond to the reciprocal of an action, is proportional to the density of these cyclic trajectories in phase space. It is chosen to satisfy the normalization condition

$$\sum_{C_m(\Delta\Gamma)}1=\mu\Delta\Gamma \qquad (m=1,\ 2,\ \ldots,\ \infty).$$

By virtue of the law of large numbers, the set of all the cyclic trajectories, for example, $C_1(\Delta\Gamma)$, is no different from any set $C_m(\Delta\Gamma)$; i.e., all the beams of trajectories $C_m(\Delta\Gamma)$ for an arbitrary m-th cycle are equivalent to each other in the statistical sense. Then Eq.

(20) can be rewritten as

$$\bar{g}(E)=1+\frac{1}{\mu\Delta\Gamma}\sum_{C_1(\Delta\Gamma)}\exp\left[\frac{i}{\hbar}S^{(1)}(E)\right]\bar{g}(E), \tag{21}$$

where we have introduced the normalized response function $\bar{g}=g/A_1V$ for convenience. It follows from (21) that the poles of the response function are determined from

$$\frac{1}{\mu\Delta\Gamma}\sum_{C_1(\Delta\Gamma)}\exp\left[\frac{i}{\hbar}S^{(1)}(E)\right]=1. \tag{22}$$

Equation (22) gives us new quantization rules in the quasiclassical limit for the case in which all the integrals of motion except the energy are stochastically violated. To study this equation we shall make use of more-detailed information on the action $S^{(1)}(E)$ of a particle which has executed a single closed cycle, and we shall also be somewhat more specific about the coarsening operation.

(c) Stochastic instability in phase space

In all the billiard-ball examples in Figs. 7 and 8, the particle trajectory can be characterized by a discrete time transformation $\hat{T}$, which determines the relationship between the dynamic variables after (or before) two consecutive collisions of the particle with the walls. This relationship can be written as

$$(p_{n+1},\ q_{n+1})=\hat{T}\,(p_n,\ q_n)$$

or

$$p_{n+1}=f_1(p_n,\ q_n);\quad q_{n+1}=f_2(p_n,\ q_n),$$

where f_1 and f_2 are certain functions which are defined such that $\hat{T}$ preserves the measure (the corresponding phase volume). In general, it is useful to single out the rapidly changing variables. Then we can assert that this system is a K system, i.e., that if there is a local (stochastic) instability in the sense of (5) in the phase space, then there always exists at least one variable along which there is a rapid mixing. Furthermore, it is precisely the instability with respect to small perturbations of this variable which makes the system a K system.[15,16,32] Let us illustrate this assertion in the example of gliding electrons (Fig. 8). We denote by x_n the coordinate of the particle upon the n-th collision with the surface, while φ_n is the angle made by the trajectory with the x axis at the point of the n-th collision. We further assume that the surface arcs have a chord a and a height $b \ll a$. Then the transformation $\hat{T}$ is[22]

$$\begin{aligned}\xi_{n+1}&=\left\{\xi_n+2\frac{R}{a}\sin\varphi_{n+1}\right\},\\ \varphi_{n+1}&=\varphi_n+\varepsilon\chi(\xi_n),\qquad \varepsilon=\frac{b}{a}\ll 1,\end{aligned} \tag{23}$$

where the braces denote the fractional part of the argument, $\xi_n\equiv\{x_n/a\}$, and $\chi(\xi)$ is a function which is determined by the shape of the surface arc and which is normalized to satisfy the condition $\max\chi\sim 1$ (in the case $\varepsilon\ll 1$, this function is generally a parabola). It can be seen from (23) that the quantity ξ is a rapidly changing variable and the condition for a local instability of this variable is

$$\begin{aligned}K&=\left|\frac{\delta\xi_{n+1}}{\delta\xi_n}\right|\sim 2\varepsilon\frac{R}{a}\chi'\gg 1;\\ \chi'(\xi)&=\frac{d\chi(\xi)}{d\xi}\end{aligned} \tag{24}$$

[inequality (7) follows from (24) in the case $\chi'\sim 1$]. We now assume that $\delta\xi_0$ is the perturbation of the initial condition ξ_0. Then

$$\delta\xi_n\sim K\delta\xi_{n-1}\sim K^2\delta\xi_{n-2}\sim\ldots\sim K^n\delta\xi_0,$$

or

$$\delta\xi_n\sim e^{nh}\delta\xi_0,\quad h=\ln K. \tag{25}$$

In the general (multidimensional) case, the relationship between the Kolmogorov entropy and the trajectory extension coefficient K may differ from (25) (Refs. 61, 15, 13, and 33), but h will remain proportional to $\ln K$. The initial perturbation $\delta\xi_0$ thus grows exponentially over time until $\delta\xi_{N_0}$ reaches a value ~1 at the time N_0. This time is given by

$$N_0=\frac{\ln(1/|\delta\xi_0|)}{\ln K}=\frac{1}{h}\ln\frac{1}{|\delta\xi_0|}, \tag{26}$$

After the time defined by (26), two trajectories whose initial conditions differ by a small amount $\delta\xi_0$ diverge to the extent that they become statistically independent.

This situation is a general situation in the sense that in systems which stochastically violated integrals of motion it is possible to determine the growth rate of the local instability, $\ln K$, and use it to write the time N_0 over which a small perturbation of the initial condition leads to a statistical independence of trajectories. This comment will be amplified in more detailed in the following section.

A stochastic instability of trajectories in phase space distinguishes a set of random trajectories from the set of periodic trajectories in the derivation of the quantization rules. Specifically, the quasiclassical approximation in (14) for the Green's function is determined for a narrow beam of trajectories which lie near a certain (basis) classical trajectory. It would be meaningless to choose a periodic trajectory as the basis trajectory, since those trajectories which lie in a small neighborhood of it will experience a stochastic instability and will diverge exponentially rapidly.

(d) Level distribution

Let us analyze the quantization rule in (22). We first note that the region $\Delta\Gamma$ is assumed to be quite small (it may be smaller than $\hbar$). Just how small $\Delta\Gamma$ is can be determined on the basis of the following considerations. As $\Delta\Gamma$ becomes smaller, there is an increase in $\tau_{\Delta\Gamma}$, which is the time required for the return to $\Delta\Gamma$. The time $\tau_{\Delta\Gamma}$ must be large in comparison with the relaxation time of the particle distribution function. In this case the substitution in (17) leads to only a slight coarsening of the trajectory, without substantially affecting its stochastic properties. In other words, all trajectories which enter $C(\Delta\Gamma)$ are concentrated near some "average" trajectory, which differs only very slightly from the "typical" stochastic trajectory of the particle. Then condition (22) can be replaced by

$$\exp\left[\frac{i}{\hbar}S_{\Delta\Gamma}(E)\right]=1, \tag{27}$$

where $S_{\Delta\Gamma}(E)$ is the action of a particle which has undergone one "approximate" cycle. The concept of an approximate cycle is introduced in such a manner that if a phase sphere of volume $\Delta\Gamma$ is placed at the end of the particle's trajectory, then the beginning of the trajectory should fall within this sphere.

We denote by $P(E)$ the probability that a level with energy E is an eigenlevel. Then it follows from (27) that

$$P(E) = \sum_{m=0}^{\infty} p_m(E), \tag{28}$$

that $p_m(E)$ is the probability that $S(E)$ takes on the value $2\pi m\hbar$ where m is an integer. Since the trajectory is long (since $\Delta\Gamma$ is small), we can use the law of large numbers. Then a plot of $p_m(E)$ against m would reveal a sharp peak (Fig. 9) at some value $m_0(E) \gg 1$. Then it follows by definition that this peak corresponds to the trajectory for which the action is

$$\bar{S}_{\Delta\Gamma}(E) = 2\pi\hbar m_0(E). \tag{29}$$

We call the trajectory with the action $\bar{S}_{\Delta\Gamma}$ the "typical trajectory" for the level E.

We now assume that the energy $E+\Delta E$ corresponds to the level nearest E. After similar arguments, we find the following for the typical trajectory with the energy $E+\Delta E$:

$$\bar{S}_{\Delta\Gamma}(E+\Delta E) = 2\pi\hbar m_1(E), \tag{30}$$

where the integer[1] is not very different from m_0. The basic consequence of (29) and (30) is

$$|\bar{S}_{\Delta\Gamma}(E+\Delta E) - \bar{S}_{\Delta\Gamma}(E)| = 2\pi\hbar|m_1 - m_0| \geqslant 2\pi\hbar; \tag{31}$$

i.e., the difference between the action values for the two typical trajectories corresponding to two adjacent levels, separated by ΔE, cannot be less than $2\pi\hbar$. A further study shows that the asymptotic behavior of $P(E;\Delta E)$ in the limit $\Delta E \to 0$ can be found from this inequality. The shape of the trajectory varies with E as a parameter. We fix the typical trajectory corresponding to energy E, and we perturb the parameter E on this trajectory by a small amount ΔE. The typical trajectory corresponds to a certain (typical) cycle time $\tau_{\Delta\Gamma}$, so that the following question arises: How are we to obtain a finite change in the action, $\geqslant 2\pi\hbar$, over a finite time interval for an arbitrarily small perturbation of the parameter of the trajectory ($\Delta E \to 0$)? This would clearly be impossible in the case of stable trajectories. In particular, it can already be seen why level repulsion should be stronger in the stable case (i.e., when there exists a complete set of integrals of motion) than in the unstable case. Furthermore, as the instability of the trajectory with respect to small perturbations of its parameters becomes stronger the level repulsion will become weaker.

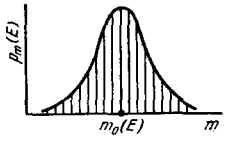

FIG. 9. Distribution of the probabilities for various values of the action for a random trajectory.

In Section 3c we mentioned that in the case of a stochastic instability one of the variables (ξ) changes very substantially between two successive collisions of a particle with the wall of the billiard table. This fact can be seen, for example, from the typical equations in (23). The extension parameter K is a function of the energy [the gyroradius R in Eq. (23) is a function of the energy], so that a perturbation of E by an amount ΔE leads to a perturbation of the initial condition by an amount

$$\delta\xi_0 \sim \frac{dK(E)}{dE}\Delta E, \tag{32}$$

as can be seen from Eq. (23). We now note that the action on a trajectory with an energy $E+\Delta E$ can never differ by a nonvanishing amount $\geqslant 2\pi\hbar$ from the action on the trajectory with energy E, provided that these trajectories do not become statistically independent. Over time intervals shorter than the scale time for the stochastic instability, the change in the action due to the perturbation is small, and inequality (31) cannot be satisfied in the limit $\Delta E \to 0$. On the other hand, the action values for statistically independent trajectories may differ by an arbitrary amount with a nonvanishing probability.

According to (26) and (32), the dimensionless time (the number of collisions with the billiard-table walls) over which an energy perturbation ΔE leads to statistically independent trajectories is

$$N_0 = \left(\ln\frac{1}{|\Delta E|} + \ln\frac{1}{dK/dE}\right)(\ln K)^{-1} \xrightarrow[\Delta E\to 0]{} \left(\ln\frac{1}{|\Delta E|}\right)[\ln K(E)]^{-1}. \tag{33}$$

For a typical trajectory with energy E, on the other hand, there exist a typical time and thus a typical number of collisions $\bar{N} = \bar{N}(E)$. According to (33), the quantity N_0 increases in the limit $\Delta E \to 0$, while $\bar{N}(E)$ does not change. Then at sufficiently small values of ΔE the following inequality always holds:

$$N_0 \gg \bar{N}. \tag{34}$$

A trajectory with an energy $E+\Delta E$ can thus become statistically independent of a trajectory with an energy E if the particle with the energy $E+\Delta E$ undergoes N_0 collisions, where N_0 is very large in comparison with $\bar{N}$, which corresponds to the maximum of the distribution $P(N)$ of the number of collisions over the stochastic trajectory. The probability $P(N_0)$, however, is the probability for a very rare fluctuation, so that

$$P(N_0) \propto \exp(-\text{const}\, N_0). \tag{35}$$

Since the appearance of $\geqslant N_0$ collisions on a trajectory with an energy $E+\Delta E$ implies that inequality (31) can be satisfied with a nonzero probability, then we have $P(E;\Delta E) \propto P(N_0)$ or, according to (35),

$$P(E;\ \Delta E) \propto \exp(-\text{const}\, N_0). \tag{36}$$

Substitution of (33) into (36) leads to the response

$$P(E;\ \Delta E) \propto |\Delta E|^{\text{const}/\ln K(E)} = |\Delta E|^{\text{const}/h(E)}, \tag{37}$$

which is given in (12). The constant ("const") in (37) can depend on E and is independent of ΔE, within a smaller error. To calculate this constant we need not

only methods that are more rigorous than those described here but also a more detailed model. The result found in Ref. 22 for the case of gliding electrons is const $\approx 1/2$.

Equation (37) has a simple physical interpretation. As K increases, the exponent in (37) decreases, and the level repulsion becomes weaker. In the limit $K \to \infty$, the probability $P(E;\Delta E)$ becomes independent of ΔE, and the repulsion disappears. The reason is that, as K increases, the trajectories become stochastic more rapidly; i.e., the local instability becomes stronger, and the correlation between the eigenvalues is weakened. In the limit in which the time required for the trajectories to become stochastic approaches zero ($K \to \infty$), the level correlation vanishes.

(e) Analysis of experimental data; critical exponents

The first experimental data on the analysis of the distribution of level separations were obtained by Gurevich and Pevzner[34] from data on the nuclei In^{113}, In^{115}, Cs^{133}, Tb^{159}, Ho^{165}, Tm^{169}, Hf^{177}, Hf^{179}, Ta^{181}, U^{235}, and U^{238}. They found the histogram shown in Fig. 10, which clearly exhibits level repulsion (the quantity plotted along the abscissa is $\Delta E/\langle\Delta E\rangle$, while the number of cases is plotted along the ordinate). An analogous curve was reported by Porter and Rosenzweig[35] for a group of elements in the osmium region (Hf, Ta, W, Re, Os, and Ir; Fig. 11, where the curve is the Wigner distribution) for the electronic levels of excited neutral atoms. A statistical level distribution should arise here because of the strong spin-orbit interaction, which violates the quantum numbers L and S. Data on the statistics of atomic levels are also given in Refs. 36 and 37. In Figs. 10 and 11 we see excellent confirmation of the level repulsion principle. This characteristic of the distribution, however, is too coarse for our purposes. Finer characteristics are the critical exponent α in Eq. (10) and the exponent β in the expression

$$\frac{dP(E;\Delta E)}{d\Delta E} \propto |\Delta E|^{\beta} \qquad (\Delta E \to 0).$$

According to (37),

$$\alpha = \frac{\text{const}}{h(E)}, \quad \beta = \frac{\text{const}}{h(E)} - 1 = \alpha - 1.$$

It follows that β becomes negative in all cases or at least beginning at certain values of E (since h increases with increasing E), and we have $dP(E;\Delta E)/d\Delta E \to \infty$ in the limit $\Delta E \to 0$. This behavior of the distribution $P(E;\Delta E)$ near $\Delta E = 0$ is not found in the Wigner–Dyson

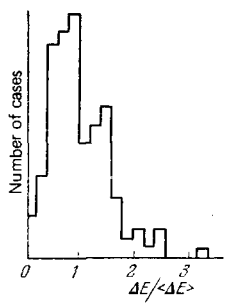

FIG. 10. Histogram of the distribution of distances between levels according to the data of Ref. 34.

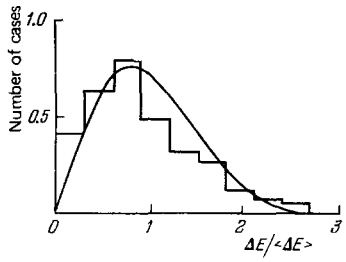

FIG. 11. Histogram of the distribution of distances between levels according to the data of Ref. 35.

distribution, since in it the values are

$$\alpha = 1, 2, 4, \quad \beta = 0, 1, 3 \geqslant 0.$$

It is not a simple matter to determine the critical exponents α and β experimentally, but we can determine the actual level repulsion law and the reasons for it. Although the behavior of the histograms in Figs. 10 and 11 implies $\beta \neq 0, 1, 3$, we cannot draw any definite conclusions[5)] because of the relatively large error of these histograms near $\Delta E = 0$.

4. SOME APPLICATIONS

The structure of an irregular spectrum has now attracted the interest of many investigators and is the subject of a lively discussion in the literature (see, for example, the reviews of the quasiclassical quantization rules[7,38]; papers on the spectra of excited molecules[39–41] and the motion of an electron in an anisotropic field[42,43]; and papers dealing with the structure of the wave function[44]). Let us examine some of these questions in more detail.

(a) Gliding electrons

The problem of electrons which are gliding along the surface of a metal in a magnetic field has been studied in several papers[45–47] in connection with the problem of surface electronic levels of metals. The periodic negative-curvature corrugation of the surface (Fig. 7b) introduced in Ref. 22 could approximate the periodic crystalline structure of a metal. The resulting irregularity of the surface-electron spectrum should be manifested in the following manner: When a weak external field at a low frequency ω is imposed on the surface electrons, they should absorb energy intensely if ω is equal to the separation of adjacent surface levels. The number of such transitions, however, must be proportional to $P(E;\Delta E = \hbar\omega)$, so that the intensity of the ab-

[5)] While this paper was being prepared for publication, McDonald and Kaufman[76] published a numerical analysis of the oscillation spectrum for a "stadium-shaped" billiard system. The motion of the particles in such a system exhibits mixing, as was shown by Bunimovich.[68] The distribution of the distances between levels found in Ref. 76 exhibits a clearly expressed repulsion, in accordance with the theory in Chapter 3. Shown for comparison in Ref. 76 is the distribution of distances between levels in the case of the integrable problem of the oscillations of a circular disk, in which there is no repulsion. G. Casati has graciously furnished information on analogous numerical results for the stadium-shaped billiard system.

sorption of energy from the external field should be, according to (37),

$$I(\omega) \propto \omega^{\text{const}/\ln K}. \quad (38)$$

Since the stochastic behavior appears in the spectrum, [according to (8)] only if the magnetic field is below a certain critical level, a decrease in the magnetic field should be accompanied by a sharp change in the frequency characteristic of the absorption line.

(b) Excited molecules

Highly excited molecules or molecules in a predissociation state constitute one of the best applications of the theory of the statistical level distribution, whereby this theory can be used to solve several fundamental questions. On the one hand, the structure of the vibrational spectra of the molecules is strongly affected by the number of integrals of motion of the given molecule. On the other hand, the rate of monomolecular reactions is essentially determined by the internal states of the molecules, especially in cases in which the internal relaxation time of the molecules is comparable with the scale times for the interaction or excitation of the molecules. These circumstances were understood a long time ago,[40,48] and they have received extensive study, primarily through numerical calculations by Rice *et al.*[40,41] More attention has been drawn to the spectra of excited molecules recently because of the collisionless dissociation of molecules which occurs when a molecule is subjected to an intense laser pulse. Although the treatment in the first papers on excited molecules was somewhat simple, Oxtoby and Rice[41] correctly noted the relationship between the spectral properties of the molecule and whether there was a stochastic instability in the molecule. Pomphrey's numerical calculations[74] for the Hénon–Heiles model in (6) showed that strong fluctuations appear in the distances between the quantum energy levels at the transition from the region of regular motion to the region of stochastic motion.

We can go through some very simple arguments to illustrate the spectral changes in the case of intense excitation of molecules.[29] Krylov has shown[19] that a gas of particles which collide as hard spheres exhibits mixing properties if

$$K = \frac{R}{2a} \gg 1, \quad (39)$$

where a is the sphere radius, and R is the characteristic distance between the centers of the spheres. In a more general case, a could be understood as the characteristic dimension of the scattering potential, for example, of an atom in a molecule. Then the quantity R, which again determines the distance between the centers of the interacting regions, is a function of the energy of the molecule. From experiment we know that the quantity $R = R(E)$ increases with increasing energy of the molecules. This fact means that condition (39) can be used to determine the critical energy E_0,

$$R(E_0) \approx 2a,$$

at which all (or some) of the integrals of motion disappear, except for the energy integral. The stable case corresponds to the inequality $K < 1$ or $R < 2a$, i.e., a "closed packing" of spheres of interaction. In this case we know that it is possible to introduce a self-consistent potential, and we arrive at a single-particle problem (the independent-particle model in nuclear theory) which is well defined (with all the necessary quantum numbers). Condition (39), in contrast, corresponds to the inequality $R > 2a$. The system becomes "loose," and it is in this case that the statistical level distribution should arise.

Apparently the simplest consequence of a disruption of the integrals of motion for highly excited molecules is that the absorption spectra should be anomalous at low frequencies. The spectrum can easily be determined on the basis of the same considerations as in section 4a for gliding electrons, i.e., the intensity of the field absorption at the frequency ω should behave as in (38).

(c) Predissociation state in the formation of molecular bonds

In the formation of molecular bonds, in particular, the hydrogen bond, a predissociation state arises. A study of the infrared spectra during the formation of the bond reveals a pronounced broadening. A quantum-mechanical theory for the corresponding line width was first derived by Stepanov.[49] This theory was subsequently developed by Stepanov and many other investigators (see, for example, the analysis in Refs. 50 and 51). Until now, however, the line widths predicted by the theory have been almost two orders of magnitude (!) smaller than the observed width for the hydrogen bond.

It is interesting to consider the following hypothesis: As the atomic groups forming the molecule approach each other, a perturbation acting on a hydrogen ion in one of the groups disrupts its integrals of motion (or, equivalently, disrupts the quantum numbers which determine the motion of the hydrogen ion). A stochastic instability arises for the hydrogen ion and leads to the anomalous width of the corresponding vibrational spectra. The stochastic instability is one of the strongest of the known dynamic instabilities[16] and can lead to anomalously pronounced broadening of the vibrational lines. In this connection it is interesting to note that a stochastic instability in a two-particle system with an exponential interaction potential (similar to the Morse potential) was studied by Casati and Ford[52] in connection with problems of a different type. The resulting picture in the phase plane is similar to Figs. 5 and 6. It can thus definitely be asserted that there is a stochastic instability of the hydrogen bond when the distance between the groups forming the molecule is less than a certain critical value.

(d) Stochastic violation of a bound state of atoms with a field[53]

The Hamiltonian describing the bound states of two-level atoms with a field (in the Dicke model)

$$H = H_0 + H_r + H_{ar}, \qquad H_0 = \hbar\omega\,(c^+c + R_z);$$
$$H_r = \hbar\omega\Lambda\,(R_+c + R_-c^+), \quad H_{ar} = \hbar\omega\Lambda\,(R_+c^+ + R_-c); \tag{40}$$
$$\Lambda = \sqrt{\frac{16\pi\rho\mu^2}{\hbar\omega}},$$

where c^+ and c are the photon creation and annihilation operators, $r_{\nu i}(\nu = z, +, -)$ are the energy spin components of atom i, $R_\nu = \sum_{i=1}^{N} r_{\nu i}$, ω is the field frequency (assumed equal to the frequency of the atomic transition), Λ is the dimensionless constant of the interaction of the atoms and the field, ρ is the density of atoms, and μ is their dipole moment. The Hamiltonian component H_r describes the resonant interaction and satisfies the commutation relation

$$[H_0,\ H_r] = 0. \tag{41}$$

The nonresonant part, H_{ar}, is usually ignored. In this case, two conservation laws follow from (40) and (41):

$$[H,\ H_0] = 0, \quad [H,\ H_r] = 0. \tag{42}$$

These laws can be used to express the energy of the system as a function of two quantum numbers. This problem has been studied by many investigators, and questions associated with the spectrum in the resonant approximation ($H_{ar} = 0$) are discussed in detail in the review by Stenholm.[54]

The total Hamiltonian in (41) leads to the following system of equations in the semiclassical approximation:

$$\ddot{\mathscr{E}} + \omega^2\mathscr{E} = \tfrac{1}{2}\omega^2\Lambda m,$$
$$\ddot{m} + \omega m = -\tfrac{1}{2}\omega^2\Lambda\mathscr{E}\sqrt{r^2 - m^2 - \frac{\dot{m}^2}{\omega^2}}, \tag{43}$$

where $\mathscr{E}$ is the dimensionless field, m is the transverse projection of the energy spin per atom, and r is a constant of the motion (the "cooperation number"). The energy integral of system (43) is

$$E = \frac{1}{\omega^2}\dot{\mathscr{E}}^2 + \mathscr{E}^2 + \sqrt{r^2 - m^2 - \frac{\dot{m}^2}{\omega^2}} - \Lambda\mathscr{E}m.$$

The quantity

$$n = \sqrt{r^2 - m^2 - \frac{\dot{m}^2}{\omega^2}}$$

corresponds to the difference between the populations of the atomic levels, while

$$y = \mathscr{E}^2 + \frac{\dot{\mathscr{E}}^2}{\omega^2}$$

is the number of photons per atom. System (43) was studied in Ref. 53, where it was shown that in the case $\Lambda < 1$ the system remains basically stable. As before, two integrals of motion remain, corresponding to two quantum numbers. In the case $\Lambda > 1$, however, the situation is markedly different. The trajectories become stochastic and nearly fill the phase space. In other words, in the case of an interaction with $\Lambda > 1$ in a closed system consisting of an atom and a field, a statistical level distribution should arise. Figure 12 shows an example of a disrupted trajectory in the (n, y) plane ($E = 1$, $\Lambda = 1.8$). In the case $H_{ar} = 0$, we have

$$n + y = \text{const},$$

according to (42), and the straight line in Fig. 12 corresponds to this conservation law. Figure 13, also plotted from the data of Ref. 53, shows the distance D as a function of the dimensionless time $\tau = \omega t$ for two very nearly equal initial conditions: curve 1) $\Lambda = 0.9$, $E = 1$; 2) $\Lambda = 3$, $E = 1$. This figure demonstrates the evolution of the local instability. From the figure we can immediately determine the instability growth rate and the associated line width γ. In the case $\Lambda = 3$ (curve 2 in Fig. 13), we find $\gamma \sim 0.5\omega$ (cf. the comment regarding the line width in Section 4c.

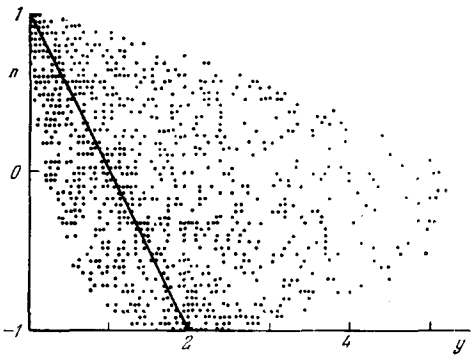

FIG. 12. Stochastic trajectory filling the phase space (for the system consisting of atoms and a radiation field).

We wish to emphasize a feature which distinguishes this system from those discussed above: The stochastic disruption of the quantum numbers occurs along the interaction-constant scale, rather than along the energy scale.

The question discussed in this section may be related to the problem of multiple production of particles. The condition for the occurrence of a stochastic situation in the collision of particles at very high energies has been discussed by Chernavskiĭ[55] and Feĭnberg.[56]

(e) Is it possible to hear the shape of a drum?

This question is usually understood as having the following meaning: Can the shape of a resonator be determined from its spectral characteristics? The answer is known to be no,[3] since quite different resonator shapes may produce identical spectra.

The appearance of a stochastic component in the eigenvalue spectrum in the quantization of K systems leads to another explanation for the negative answer to this question: Two isomorphic K systems have an identical spectrum (identical in the statistical sense) but may have wildly different shapes.

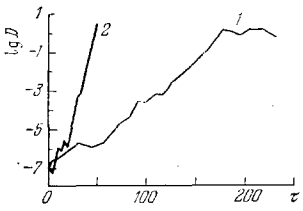

FIG. 13. Evolution of a local instability in a system consisting of atoms and a radiation field.

5. SPECTRUM OF QUASIENERGY LEVELS

When a periodic perturbation acts on quantum system, we can introduce the concept of quasienergy, as the concept of quasimomentum is introduced, as was first shown by Zel'dovich.[69,70] Let us assume that the Hamiltonian

$$H = H_0(p, x) + V(x, t)$$

has the periodic property

$$V(x, t + T) = V(x, t).$$

Then the wave functions can be chosen such that

$$\psi(x, t + T) = e^{(i/\hbar)\mathcal{E}T}\psi(x, t),$$

where $\mathcal{E}$ is the "quasienergy." States with a certain quasienergy play the same role in the time-dependent case as states with a definite energy play in the steady-state case. In the classical theory, however, periodic perturbations can lead to a stochastic motion if the perturbation is strong enough. The result is a stochastic disruption of the energy integral (the only integral). What should happen to the quasienergy spectrum if the corresponding classical Hamiltonian describes motion with mixing in the case $\hbar = 0$? At present we have no answer to this question. The information which is available and the difficulties which arise in this problem are so interesting that we will discuss them separately.

(a) Mappings for quantum systems

A typical example, and the most convenient example for constructing discrete mappings, is a nonlinear oscillator which is acted on by a periodic train of δ-function pulses:

$$V(x, t) = U(x)\sum_{n=-\infty}^{\infty}\delta(t - nT). \tag{44}$$

We write

$$\psi_n = \psi(x, t = nT + 0),$$

i.e., the wave function immediately after the n-th pulse. It is not difficult to derive the recurrence relation[71,72]

$$\psi_{n+1} = e^{-(i/\hbar)U(x)}e^{-(i/\hbar)H_0T}\psi_n \equiv \hat{T}_q\psi_n. \tag{45}$$

We thus have

$$\hat{T}_q\psi_n = \exp\left(\frac{i}{\hbar}\mathcal{E}T\right)\psi_n,$$

and the determination of the quasienergies reduces to the determination of the eigenvalues of the mapping $\hat{T}_q$.

In the classical case, on the other hand, it is possible to construct a mapping $\hat{T}$ for the variables (p, x) which determine the state of the system:

$$(p_{n+1}, x_{n+1}) = \hat{T}(p_n, x_n).$$

The mapping $\hat{T}$ for the perturbation potential in (44) has been analyzed in many papers.[15,16,32,72] The nature of the motion governed by the transformation $\hat{T}$ depends on the parameter

$$K = \text{const}\cdot\alpha\varepsilon\omega T, \tag{46}$$

where the constant is a number on the order of unity; α is a dimensionless nonlinearity parameter, given by

$$\alpha = \frac{I}{\omega(I)}\frac{d\omega(I)}{dI},$$

$\omega(I)$ is the nonlinear frequency, governed by the unperturbed Hamiltonian $H_0(p, x)$; and ε is the dimensionless perturbation parameter ($\varepsilon \sim U/H_0T$). The parameter K has the same meaning as in (24). In the case $K < 1$, the motion is conditionally periodic (stable), but under the condition

$$K = \text{const}\cdot\alpha\varepsilon\omega T > 1 \tag{47}$$

a stochastic instability occurs, causing the classical system to become a K system.

We now assume that condition (47) holds. What properties will the quantum system have? In other words, if the mapping $\hat{T}$ corresponds to a K system, what will the mapping $\hat{T}_q$ be? This is simply a different formulation of the question asked at the beginning of this section.

The evolution of coherent states under condition (47) was studied in Ref. 60, where it was shown that the eigenvalues of the creation and annihilation operators exhibit the mixing property in the quasiclassical approximation. The analysis of Ref. 60, however, holds for only a finite time

$$t < t_h \propto t_0\frac{I}{\hbar} \tag{48}$$

where t_0 is some characteristic time of the problem. An analogous result was found in Ref. 7 through a numerical analysis involving calculations of the expected values $\langle x\rangle$ and $\langle p\rangle$. Their time evolution corresponds to the evolution of the variables (x, p) of the equivalent classical system for only a finite time. Here again, as so many times before, we must examine the distinction between the quantum and classical approaches.

(b) Two cases of wave-packet spreading

Since in quantum mechanics we are always dealing with a wave packet rather than a separate trajectory of a system, it is useful to discuss the evolution in phase space of some volume filled by classical particles. If these particles are linear oscillators, all with identical frequencies, then the cell which they fill in phase space moves without any deformation of its boundaries. In quantum mechanics, this circumstance corresponds to the possibility of constructing a nonspreading wave packet (a so-called coherent state). In the nonlinear case, we have a different situation. The oscillator frequencies depend on the oscillator energies, so that different parts of the cell in phase space move at different velocities. As a result, there is a distortion of the cell boundaries, as shown schematically in Fig. 14 ($t_2 > t_1 > t_0$). Not only is there a spreading of the phase-space cell; we also see a steepening of its "front" and the appearance of an ambiguity in the shape of the cell boundary. This picture is analogous to the steepening of the profile of a nonlinear wave in hydrodynamics (Fig. 15, $t_2 > t_1 > t_0$). In quantum mechanics, this circumstance corresponds to the absence of nonspreading coherent states in the nonlinear case.

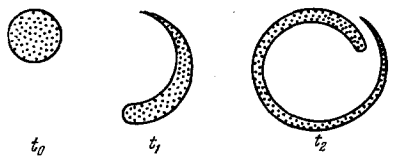

FIG. 14. Spreading of a phase-space droplet of an ensemble of non-interaction nonlinear oscillators.

This spreading of the wave packets in the nonlinear case is a slow process. The situation is different in the case of K systems, in which the spreading of the phase-space cell and the deformation of the boundaries of this cell occur exponentially rapidly because of mixing. In this case, the concept of a wave beam, even a very narrow beam ($\sim\hbar$), rapidly becomes meaningless. The occurrence of a local instability renders the concept of a quasiclassical beam indefinite over very long periods of time. We are thus running into some fundamental difficulties in the quasiclassical description of K systems.

In this connection we should discuss two numerical results of a study carried out for systems with the potential in (44) (at present, these two results are the only ones available). As mentioned earlier, it was shown in Ref. 61 that the observed expectation values $\langle x\rangle$ and $\langle p\rangle$ vary with the time in accordance with the classical theory, for sufficiently short times. It also follows from the results of Ref. 73 that, for the same (short) times for which the numerical analysis was carried out, the behavior of the probability amplitude agrees well with the data from classical dynamics.

It follows that at present we cannot use the quasiclassical approach in its present form for unbounded times. Then we immediately draw the further conclusion that we cannot speak of a quasienergy spectrum of K systems, either discrete or continuous.

6. CONCLUSION

The questions discussed in this review show that we are dealing with new phenomena, about which little is known, either theoretically or experimentally. The basic purpose of this review has thus been to draw attention to these questions and to point out relationships between certain problems which appear at first glance to be unrelated. We would now like to list some other unresolved questions.

(a) Quantization of stochastically unstable systems

The question of the energy spectrum of systems with violated integrals of motion is a particular case of a more general question: What are the quantum properties of such systems?

For a long time now we have been accustomed to expecting a rather reliable quasiclassical approximation in the limit $\hbar\to 0$. In Keller's expression, this approximation consists of describing a certain method for pinning some wave "flesh" onto a classical "skeleton." These questions have now been discussed in many papers (see the reviews in Refs. 7, 9, and 57). Is the ordinary quasiclassical method retained for describing systems with violated integrals of motion? There are reasons for doubting that the answer to this question is yes, as we shall now see.

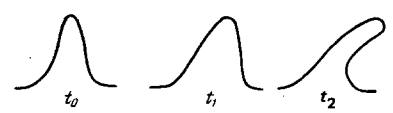

FIG. 15. Formation of a multivalued profile of a nonlinear wave.

(b) Wave functions

The first paper in which wave functions derived in the quasiclassical approximation were subjected to a critical analysis was that by Arnol'd.[58] Studying a special example, Arnol'd raised the hypothesis that "quasimodes", rather than modes, exist in the quasiclassical approximation (see also Ref. 3). The meaning here is that as time elapses the wave function becomes progressively less similar to an oscillation (of, say, the plane-wave type), and it spreads quite rapidly and converts into a quasimode. Such functions satisfy the equation quite accurately, but they may be very different from the eigenfunctions.

The question of the wave-function structure in the case of a stochastically unstable Hamiltonian becomes particularly important when we note that the possibility that there are no modes at all in this case is not ruled out. The reason lies in the local instability of the trajectories of the particle, which leads to an automatic spreading of the wave "flesh." This property of wave functions was discovered in Ref. 22, and in Ref. 60 it was shown that wave packets which are initially coherent undergo an exponential spreading. The convenient introduction of quasimodes could apparently simplify the analysis of stochastic quantum systems.

The structure of the wave functions of the quasiclassical approximation in the case of regular and irregular spectra has also been discussed by Berry.[44,59]

(c) Correspondence principle

A study of stochastic properties in quantum systems shows that certain difficulties arise in the ordinary quasiclassical approach. At this point we cannot state whether these difficulties are fundamental or technical in nature. On the other hand, the correspondence principle gives us some alternatives: Either classical K systems are too crude a description of reality, and there is no such thing as stochastic behavior in the quantum world, or we are not able to take the limit properly in the quasiclassical case for K systems. The latter possibility brings back Einstein's critical comments regarding the classical limit in quantum mechanics, and we would like to end this review with his words:

"We now ask: Does quantum mechanics (with the limitations on accuracy which follow from it) incorporate that real description of the motion of macroscopic objects which is given by classical mechanics?"[75]

For a long time now I have been able to discuss the questions covered in this review with V. I. Arnol'd, I. M. Lifshits, Ya. G. Sinaĭ, and B. V. Chirikov. I received much assistance from V. F. Shabanov in analyzing the material in Section 4c. G. P. Berman, I. A. Malkin, and L. A. Pastur made several useful comments regarding the original version of the manuscript. S. A. Molchanov drew my attention to some results reported in Section 1a before their publication elsewhere. To all these people I express my sincere gratitude.

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