

# Decay of an isolated level into a continuum corresponding to an infinite random walk

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The decay of a level into a continuum corresponding to an infinite random motion of a quantum-mechanical particle is analyzed. All the interference effects which result from returns of the particle to its initial state in the course of the random walk are taken into account exactly by an ensemble-average method, without any appeal to renormalization-group considerations or requirements of spatial uniformity "on the average." The level decay laws are found for long times for the cases of random walks which do and do not involve a return. The conditions for an "incomplete" decay of the level are determined. In other words, the conditions for the existence of a localized component of the population, consisting of the requirement that the random walk return and the requirement that the correlation time be finite, are determined. The time required for the population of the level to reach its asymptotic value is estimated. A possible mechanism for the formation of a "cold" ensemble during the excitation of polyatomic molecules by a resonant electromagnetic method is discussed. The process by which the motion becomes stochastic (by which the quantum numbers are destroyed) in quantum-mechanical systems of large dimensionality is also discussed.

## 1. STATEMENT OF THE PROBLEM AND BASIC RESULTS

The problem of the decay of an isolated level into a continuum is encountered in many fields of physics.<sup>1-7</sup> Although this problem does have a formal solution in quadratures, it is generally not possible to draw any really meaningful conclusions about the nature of the decay. Even the answer to the qualitative question of whether the level decays completely depends substantially on the details of the spectral density and the matrix elements which couple it with the continuum.

In this paper we wish to examine the problem—not of universal applicability but a fairly general particular case—of the decay of a level into a continuum, in which it is possible to reach some substantial conclusions. This analysis applies to systems in which an isolated level interacts directly with a relatively small fraction of the quantum-mechanical states of the continuum (a large finite or countable subset), which, not being eigenenergy states, interact in turn with all the other states and thereby indirectly with each other.

We will analyze the asymptotic behavior after a long time—longer than any of the temporal parameters in the problem. The asymptotic behavior of such a system is related to the well-known problem of localization<sup>1,8-10</sup> which arises in a study of one-electron states in solids with randomly distributed impurities. The approach taken in the present paper is significantly different from the approaches which are customarily taken in solid state physics to solve problems of this sort. We wish to formulate several rather general assertions regarding the nature of the motion in the system. Thereafter we will make no assumptions of the nature of the existence of a renormalization group. Our approach is thus valid for studying systems of arbitrary spatial dimensionality, and there is no requirement that the systems be spatially uniform "on the average."

The physical processes which can be described by the

level-continuum model (or the model of a level and a dense band) are extremely diverse. They include processes which correspond to the complete decay of states which are initially filled [spontaneous emission,<sup>2</sup> many-photon ionization of atoms,<sup>3,4</sup> and the formation of an absorption line during the interaction of electronic configurations in atoms<sup>5</sup> (autoionization)] and processes in which the decay of the states is incomplete (the excitation of low-lying levels of polyatomic molecules by a resonant field,<sup>6</sup> the formation of localized states in Anderson insulators,<sup>1</sup> etc.). Problems involving the destruction of quantum numbers and the appearance of quantum chaos, which are usually studied numerically for simple systems,<sup>7,11,12</sup> can also be reduced to level-continuum systems and studied analytically. This comment also applies to complex systems, if physically reasonable assumptions are made regarding the random walk in the stochastic layer.

The nature of the decay of a level into a dense band (continuum) of course depends on the statistics of the energy levels of the band and the size of the matrix elements of the operator representing the interaction of the level with the band. The statistics are in turn determined by the particular features of the dynamic process which resulted in the formation of the band. We believe that the most convenient way to incorporate the statistics is to use the method of ensemble averaging,<sup>13,14</sup> which can be used successfully in problems involving randomly inhomogeneous systems.<sup>15</sup> The analytic expressions can be made insensitive to the microstructure of the spectrum by appropriately choosing the ensemble of systems having identical average values of the characteristics important to the process under consideration, and then averaging the perturbation series for the level population over this ensemble. The procedure of ensemble-averaging is closed by a test to see whether the variances of these expressions are small.

In this paper we will apply the procedure of ensemble-averaging only to that fraction of the continuum states

which are directly coupled to the level. We assume that the distribution function of the values of the interaction matrix elements is given, and we assume that the energy position of each of the states selected is statistically independent of the positions of all of the other states selected. The interaction of the states with each other, mediated by all the other states of the dense band, is assumed to be complex and to have the characteristics of a random walk. The probability for the transfer of population from one state to another over a fixed time  $t$  is determined exclusively by the relative energy positions of the levels.

In other words, we are dealing with the dynamics of the filling of state 0 (which we will sometimes refer to below as the "ground state") in a system described by the Schrödinger equation

$$\begin{aligned} i\dot{\psi}_0 &= \sum_k V_{0k}\psi_k, \\ i\dot{\psi}_k &= \Delta_k\psi_k + V_{k0}\psi_0 + \sum_{\alpha} V_{k\alpha}\psi_{\alpha}, \\ i\dot{\psi}_{\alpha} &= \Delta_{\alpha}\psi_{\alpha} + \sum_k V_{\alpha k}\psi_k, \\ \psi_0(t=0) &= 1, \quad \hbar=1, \quad \int \psi_k(t) e^{-i\varepsilon t} dt = \psi_k(\varepsilon), \\ \int \psi_k^*(t) e^{-i\varepsilon t} dt &= \psi_k(\xi), \end{aligned} \quad (1)$$

with respect to which we adopt the following assumptions.

1. The energy position  $\Delta_k$  of each of the levels  $k$  is statistically independent of the positions of all the other levels of this set,  $\{k\}$  (we assume that it is equally probable over the interval from  $-\Gamma$  to  $\Gamma$ ; we will later take the limit  $\Gamma \rightarrow \infty$ ) and also statistically independent of the value of the matrix element  $V_{0k}$ .

2. The state density of the band,  $\{\alpha\}$ , is so high that the band can be regarded as infinitely dense.

3. The matrix elements  $V_{k\alpha}$  are random quantities, such that the only nonzero ensemble averages of sums are of the type  $\sum_{\alpha} V_{k\alpha} V_{\alpha k} X(\alpha)$ , where  $X(\alpha)$  is an arbitrary smooth function of the level energy, while averages of the type  $\sum_{\alpha} V_{k\alpha} V_{k'\alpha}^* X(\alpha)$  vanish.

4. The ensemble average of the quantity

$$\sum_{\alpha\alpha'} V_{k\alpha} V_{\alpha k'} V_{k\alpha'}^* V_{\alpha k'}^* (\varepsilon - \Delta_{\alpha})^{-1} (\xi - \Delta_{\alpha'})^{-1},$$

which is proportional to the population flux from one level ( $k$ ) to another ( $k'$ ), does not depend on the particular levels  $k$  and  $k'$  which it couples. It is equal to some function  $f(\varepsilon, \xi)$  which is identical for all pairs.

5. The ensemble average of the decay rate of the band levels  $\{k\}$  into the band ( $\alpha$ ), given by  $\sum_{\alpha} V_{k\alpha} V_{\alpha k} (\varepsilon - \Delta_{\alpha})^{-1}$ , does not depend on the index  $k$ . We denote this value by  $\gamma(\varepsilon)$ .

In other words, we have singled out from the entire dense band those states  $\{k\}$  which interact directly with level 0, and we diagonalize the Hamiltonian in terms of all the other states of the band. As a result of the latter procedure, we form a set of levels  $\{\alpha\}$ , which is related to the levels  $\{k\}$  by the random matrix elements  $V_{\alpha k}$ . The rate at which population flows from one level  $k$  to another level from the same set in the course of a process similar to a random walk is

determined by—only the function  $f(\varepsilon, \xi)$ , which is identical for all pairs. In other words, all the levels of band  $\{k\}$  are equivalent from the standpoint of the redistribution of population among states of the continuum.

To avoid any misunderstanding, we wish to stress that the interaction of levels  $\{k\}$  with continuum states  $\{\alpha\}$  generally does not have to lead to a complete and irreversible (exponential) decay of these levels. In other words, the interaction of some level with a dense set of other states can be described in by no means all situations by introducing a decay—a corresponding imaginary increment in the energy of this level. Under conditions such that this interaction is complicated, irregular, and even a discontinuous function of the energy of the state in the continuum, the dynamics of the decay of a noneigen state will generally not be exponential. In a problem of this sort, the particular function  $f(\varepsilon, \xi)$  is responsible for this irregularity, as it is for the nature of the decay process.

A distinction is drawn between two types of motions, depending on the nature of the behavior of the function  $f(\varepsilon, \xi)$  in the limit  $\varepsilon \rightarrow \xi$ . The motion is a "returning" motion if we have  $f \rightarrow \infty$  as  $\varepsilon \rightarrow \xi$ , or it is a "nonreturning" motion if we have  $f \rightarrow 0$  as  $\varepsilon \rightarrow \xi$ . For a nonreturning motion, a quantum-mechanical particle which is in one of the band levels  $\{k\}$  at the time  $t = 0$  will leave this level, and in the limit of large  $t$  it will not return to any of the other levels of this band. In this case, the decay of level of the band  $\{k\}$  to states of the band  $\{\alpha\}$  is irreversible. In the case of a returning random walk, we are dealing with a different situation. This type of motion corresponds to a repeated return of the quantum-mechanical particle to levels of band  $\{k\}$ ; i.e., the integral of the total population of all these levels over time diverges at the upper limit. If the probability for the particle to be in the states  $\{k\}$  is calculated by a path-sum method, the implication is that in the limit of interest here (long times) the situation is dominated by paths which undergo repeated self-intersections at the levels of the band  $\{k\}$ . We are actually talking about incorporating an interference among the wave functions which arise as a result of the repeated returns of the particles to the given group of states.

Incorporating the effect of path self-intersections is the basic problem in carrying out a summation. Although the topology of the Feynman diagrams which arise in the course of the calculations is considerably more complex than usual (trees, ladders, etc.), it nevertheless turns out that these diagrams can be summed. The procedure required here, which is based on certain methods of graph theory, is extremely laborious; we will present it in the following section of this paper. At this point we think it is worthwhile to preview the results which are found as a result of this summation and to list the characteristics of the system which are responsible for the decay of the ground state.

The most important results are two in number. First, a nonreturning random walk leads to a complete decay. Long times correspond to a decay law of the type  $\rho_0 \propto \exp(-\text{const } t^{1/2})$  (slower than exponential) for the population of state 0. The reason for this decay law is the existence in the selected ensemble of some improbable realizations of bands which do not have levels  $k$  which are sufficiently close to state 0. In this case, self-intersections of paths are inconsequential.

The second important result refers to the case of a re-

turn random walk. If a particle repeatedly returns to the levels of band  $\{k\}$ , all the paths will intersect repeatedly, thereby leading to a substantial change in the pattern as a result of interference effects. In such a situation, a return random walk leads to an incomplete decay of the level. In the limit of long times, an exponentially small fraction  $\exp\{-V_0^2 g^2 \text{const}\}$  of the population remains at this level; here  $g$  is the state density of band  $\{k\}$ , and  $V_0^2$  is the mean square value of the interaction matrix element  $|V_{0k}|^2$ .

An important role is played here by the characteristic correlation time of the random walk,  $\tau_c$ : the time over which the wave functions of the states of band  $\{k\}$  are changed by the nondiagonal matrix elements  $V_{k\alpha}$  of the Hamiltonian. Although this time does not appear explicitly in the result, it determines just when the asymptotic distribution of populations is established. This event occurs when the number of returns in the random walk exceeds the number of levels in band  $\{k\}$  which fall within a  $\tau_c^{-1}$  neighborhood of the energy of level 0. If the correlation time is exceedingly small,  $\tau_c \rightarrow 0$ , on the other hand, and the asymptotic value  $\rho$  is not reached over the time intervals of interest, then the random walk may be regarded as uncorrelated. Correspondingly, we would have  $f(\varepsilon, \xi) \propto f(\varepsilon - \xi)$ . As a result we have the intermediate asymptotic behavior  $\rho_0 \propto \exp(-\text{const } t^{2/3})$ . This functional dependence, as in the case of a nonreturning random walk, results from the influence of improbable realizations of the system. Consequently, the asymptotic time dependence of the population of level 0 is determined by two characteristics: the correlation time and whether the random walk is of a return nature.

## 2. CALCULATION OF THE DECAY PROBABILITY

It is convenient to seek a solution of Eq. (1) in the form of an infinite power series in the interaction  $V$ . In this case the probability amplitude for the filling of the ground state can be written as a sum over all possible closed paths which begin and end at level 0. Each path corresponds to a particular term in the series. A path is represented as a sequence of transitions between levels  $\{0, \{k\}, \{\alpha\}\}$ ; each of the levels met along the path corresponds to a factor  $\varepsilon^{-1}$ ,  $(\varepsilon - \Delta_k)^{-1}$ , or  $(\varepsilon - \Delta_\alpha)^{-1}$ , and each of the transitions corresponds to a factor  $V_{0k}$  or  $V_{k\alpha}$ , which is the probability amplitude for the given transition.

Over the long time intervals in which we are interested here, the system has time to undergo many transitions. The corresponding paths are thus long, with many self-intersections. If we represent each path by an oriented graph (or-graph), the graph will have a large number of parallel edges, since a given transition occurs repeatedly, or pairs of levels from the band  $\{k\}$  are connected by different nonintersecting paths. Since only the numbers of levels and transitions encountered along the path—not the particular order in which they occur—contribute to the series in a perturbation theory in  $V$ , many paths will have identical orgraphs and will thus contribute identically to the sum of the series. A summation over paths can then be replaced by a summation over different orgraphs with appropriate “statistical weights,” i.e., with the numbers of various possible circuits of the or-graph or “Eulerian paths.” An explicit expression is available<sup>16</sup> for a number of this sort:

$$\mathcal{E} = \left\{ \prod_j (d_j - 1)! \right\} \det \|p_{ij}\|, \quad (2)$$

where  $d_j$  is the multiplicity of node  $j$ , i.e., the number of edges which leave it (the number of times level  $j$  is encountered on the path), and  $\det \|p_{ij}\|$  is any of the minors of the connectedness matrix  $\|p_{ij}\|$ , which consists of the matrix elements  $p_{ij}$ , which (with  $i \neq j$ ) give the numbers of parallel edges connecting nodes  $i$  and  $j$  or (with  $i = j$ ) are assumed to be equal to the total number of edges which leave the node  $p_{ii} = \sum_j p_{ij}$ . For definiteness below, we will always deal with the minor which corresponds to the deletion of the row and the column which correspond to the ground state.

We are interested in the population, not the wave function, of state 0. To find the population we need to go through a completely analogous procedure—taking Fourier transforms, carrying out a series expansion in a perturbation theory in the interaction, and writing the series as a sum over orgraphs—for the complex-conjugate wave function, which we denote by  $\psi(\xi)$ , using a different variable  $\xi$ , which is conjugate of the time. We then need to multiply the resulting series term by term. Most of the terms in the series for the populations which is formed in this way vanish after we carry out the ensemble-averaging, by virtue of assumption 3. The only terms that are left are those for which the parts of the orgraphs which differ in topological structure from trees and which correspond to the levels of band  $\{k\}$  and  $\{\alpha\}$  and to transitions between them,  $V_{k\alpha}$ , are completely identical for the right-hand and left-hand brackets. If we also allow for the fact that the parts of the orgraphs which do have the topology of trees can be summed separately, with the result that we find a renormalization of the node factors [the level energies  $\Delta_k$  acquire imaginary increments  $\gamma(\varepsilon)$  and  $\gamma(\xi)$  which are positive for  $\xi$  and negative for  $\varepsilon$ ], then we can assert that the only terms of the series for the populations which are nonvanishing are those for which the orgraphs of the transitions between states of the  $\{k\}$  and  $\{\alpha\}$  bands are identical. By virtue of assumption 2, we can ignore self-intersection of the diagrams at the levels of band  $\{\alpha\}$ . By virtue of assumption 4, we can associate identical factors  $f(\varepsilon, \xi)$  with all coincident edges of the orgraphs for the right-hand and left-hand brackets connecting different levels of band  $\{k\}$  and passing through levels of  $\{\alpha\}$ .

It thus becomes possible to further simplify the structure of the perturbation series for calculating a population. Specifically, we can now eliminate the energy levels of band  $\{\alpha\}$  from consideration. The nodes of the orgraphs will then refer exclusively to the  $\{k\}$  band. Each edge connecting these nodes can be associated with a factor  $f(\varepsilon, \xi)$ . The matrix elements of the connectedness matrix  $p_{ij}$  now refer only to the number of parallel edges between nodes of band  $\{k\}$ , and a combinatorial factor compensates for the indistinguishability of the nodes of band  $\{\alpha\}$  which results from this procedure.

$$\left( \sum_{i,j \neq i} p_{ij} \right)! / \prod_{i,j \neq i} (p_{ij}!)$$

The difference between the number of Eulerian circuits for the right-hand and left-hand brackets now results exclusively from the difference between the orgraphs at the levels of band  $\{k\}$  and level 0. If we denote by  $m_{0k}$  the number of

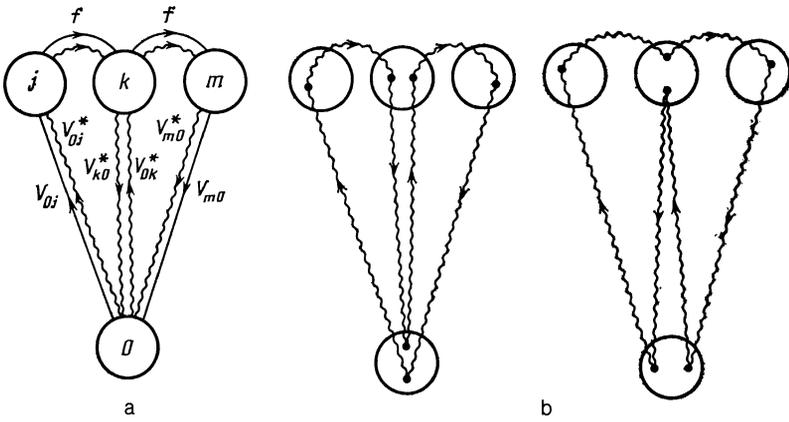


FIG. 1.

transitions  $V_{0k}$  which are encountered for the left-hand bracket, and by  $n_{0k}$  the corresponding number for the right-hand bracket, we can write the following expression for the population of the ground state:

$$\begin{aligned} \rho(\varepsilon, \xi) = & \frac{1}{4\pi^2 \varepsilon \xi} \sum_{\{p_{kj}\}} \sum_{\{n_{0k}\}} \sum_{\{m_{0k}\}} \left( \sum_k m_{0k} \right)! \\ & \cdot \left( \sum_k n_{0k} \right)! \left( \sum_{k, j \neq k} p_{kj} \right)! \det \| p_{kj} \\ & - \delta_{kj} \left( \sum_{k'} p_{k'j} - m_{0j} \right) \| \det \| p_{kj} \\ & - \delta_{kj} \left( \sum_{k'} p_{k'j} - n_{0j} \right) \| \prod_{k, j \neq k} \left\{ \frac{f(\varepsilon, \xi)^{p_{kj}}}{p_{kj}!} \right\} \\ & \cdot \prod_k \left\{ \frac{(V_{0k})^{n_{0k}} (V_{k0})^{n_{0k} - \sum_j (p_{kj} - p_{jk})}}{\varepsilon^{n_{0k}} [\varepsilon - \Delta_k - i\gamma(\varepsilon)]^{n_{0k} + \sum_j p_{kj}}} \right. \\ & \left. \cdot \frac{(V_{k0})^{m_{0k}} (V_{0k})^{m_{0k} - \sum_j (p_{kj} - p_{jk})}}{\xi^{m_{0k}} [\xi - \Delta_k - i\gamma(\xi)]^{m_{0k} + \sum_j p_{kj}}} \right\}, \quad (3) \end{aligned}$$

where a summation over  $\{p_{ij}\}$  means a summation over all of the  $p_{ij}$ , each of which takes on values from 0 to  $\infty$ . Similar comments apply to  $n$  and  $m$ .

As an example, Fig. 1 (a) shows a graph representation of one of the terms of the series for the population (straight lines correspond to the right-hand bracket, and wavy lines to the left-hand bracket). Corresponding to the orgraph is a term of the following form in the series for the population:

$$\begin{aligned} & \varepsilon^{-2} \xi^{-2} (\varepsilon - \Delta_j - i\gamma)^{-1} (\varepsilon - \Delta_k - i\gamma)^{-1} (\varepsilon - \Delta_m - i\gamma)^{-1} (\xi - \Delta_j + i\gamma)^{-1}, \\ & (\xi - \Delta_k + i\gamma)^{-2} (\xi - \Delta_m + i\gamma)^{-1} V_{0j} V_{0j}^* V_{m0} V_{m0}^* V_{0k} V_{0k}^* [f(\varepsilon, \xi)]^2. \quad (4) \end{aligned}$$

Here  $p_{jk} = 1$ ;  $p_{km} = 1$ ; the number of Eulerian paths for the right-hand bracket is equal to 1; and that for the left-hand bracket is equal 2. Figure 1 (b) shows Eulerian paths for the left-hand bracket.

For convenience in the calculations, we will make one more change in the order of the summation. The reason is that there are many different levels in band  $\{k\}$ , and a path of finite length is incapable of reaching all of them. This assertion means that among the factors in expression (3) there

are many 1s (zeroth powers). We eliminate them from consideration. To do this, we choose exclusively those levels through which the paths of the given orgraph pass, and we call the set the "carrier of the orgraph." We carry out a summation over all orgraphs with a given carrier, and we then carry out a summation over all carriers. With the summation in this order, the value of  $n_{0k} + m_{0k} + \sum_j p_{kj}$  is a natural number for any node  $k$ . Expression (3) can then be put in the form

$$\rho(\varepsilon, \xi) = \sum_{\{X\}} \frac{1}{4\pi^2 \varepsilon \xi} \sum_{\{p_{kj}\}} \sum_{\{n_{0k}\}} \sum_{\{m_{0k}\}} (\dots), \quad (5)$$

where the sum over  $\{X\}$  means a summation over all possible carriers.

We now make use of assumption 1: In our ensemble, each level  $k$  is distributed at random in the band, and it can take on values  $\Delta_k$  from  $-\infty$  to  $\infty$  with equal probabilities. We average the terms of series (3), noting that we have

$$\int_{-\infty}^{\infty} \frac{1}{(a-\Delta)^m} \frac{1}{(b-\Delta)^n} g d\Delta = \frac{2\pi i g}{(a-b)^{m+n-1}} \frac{(n+m-2)!}{(n-1)!(m-1)!} \quad (6)$$

for  $n, m > 0$ . In the cases  $n = 0, m = 1$ , and  $n = 1, m = 0$  the integral is equal to  $\pm ig\pi$  (depending on the sign of the imaginary part of  $a$  and  $b$ ). Using the integral representation of the factorials

$$\left( \sum_j X_j \right)! = \int_0^{\infty} \prod_j \{\sigma^x\} e^{-\sigma} d\sigma, \quad (7)$$

we find the following expression for the population of the ground state:

$$\begin{aligned} \rho = & \frac{1}{4\pi^2 \varepsilon \xi} \sum_{\{X\}} \sum_{\{n_{0k}\}} \sum_{\{m_{0k}\}} \sum_{\{p_{kj}\}} \int_0^{\infty} d\lambda \int_0^{\infty} d\sigma \int_0^{\infty} d\tau \\ & \times \exp(-\lambda - \sigma - \tau) \prod_{k, j \neq k} \left\{ \left[ \frac{-f(\varepsilon, \xi) \tau}{(\varepsilon - \xi - 2i\gamma)^2} \right]^{p_{kj}} \frac{1}{p_{kj}} \right\} \det \| p_{kj} \\ & - \delta_{kj} \left( \sum_l p_{kl} - m_{0k} \right) \| \det \| p_{kj} - \delta_{kj} \left( \sum_l p_{kl} - n_{0k} \right) \| \prod_k \\ & \times \left\{ - \int_0^{\infty} d\tau_k \frac{\exp(-\tau_k)}{\tau_k^2} 2\pi i g_k (\varepsilon - \xi - 2\gamma) \right. \\ & \left. \times \left( \frac{-|V_{0k}|^2 \lambda \tau_k}{\varepsilon (\varepsilon - \xi - 2i\gamma)} \right)^{n_{0k}} \left( \frac{|V_{0k}|^2 \sigma \tau_k}{\xi (\varepsilon - \xi - 2i\gamma)} \right)^{m_{0k}} \right\} \end{aligned}$$

$$\times |V_{0k}|^2 \sum_j (p_{kj} - p_{jk}) \tau_k^2 \sum_j p_{kj} [n_{k0}! m_{k0}! [n_{k0} + \sum_j (p_{kj} - p_{jk})]^{-1}]^{-1}, \quad (8)$$

where  $2\gamma = \gamma(\varepsilon) + \gamma(\xi)$ .

The idea of the following transformations is to put the population of the ground state in the form of a product of factors each of which depends on only the parameters corresponding to one level. An averaging is then carried out over these parameters. In this approach, the primary difficulty stems from the presence of determinants which depend on the indices of many levels in expression (8). The levels can be split, however, by using generating functions. The actual procedure, in the form in which we have managed to carry it out, is extremely involved, and we do not have space here to reproduce it in detail. It is summarized in the Appendix, where the appropriate notation is also introduced. The final expression is

$$\rho = -[4\pi^3 (\varepsilon - i\pi g \langle V^2 \rangle) (\xi + i\pi g \langle V^2 \rangle)]^{-1} \cdot \int_0^\infty \int_0^\infty \int_0^\infty d\lambda d\sigma d\tau \int_{-\infty}^\infty \int_{-\infty}^\infty dx dy \int_0^\infty du \cdot \int dv (4\pi uv)^{-1} \exp(-\lambda - \sigma - \tau - x^2 - y^2) \cdot [(1-u^{-1})(1-v^{-1})(1-A_2)^{-1} \cdot \exp(A_1 - A_2) + |A_3|^2 (1-A_2)^{-2} \exp(A_1 - A_2)], \quad (9)$$

where we are using the notation

$$A_1(k) = \int \frac{g(V) dV}{2\pi i} \int_{C(\mu)} dv v^{-2} \int_{-\pi}^\pi \int_{-\pi}^\pi [f\tau v^2 (1+u) - \frac{\partial}{\partial s} - \frac{\partial}{\partial R}] [f\tau v^2 (1+v) - \frac{\partial}{\partial s} - \frac{\partial}{\partial L}] e^{\Lambda(k)} d\theta d\theta |_{s,R,L=0},$$

$$A_2(k) = - \int \frac{g(V) dV}{2\pi i} \int_{C(\mu)} dv \int_{-\pi}^\pi \int_{-\pi}^\pi f\tau e^{\Lambda(k)} d\theta d\theta |_{s,R,L=0},$$

$$A_3(k) = - \int \frac{g(V) dV}{2\pi i} \int_{C(\mu)} dv \int_{-\pi}^\pi \int_{-\pi}^\pi d\theta d\theta f\tau \cdot \exp[\Lambda(k) + i(\theta + \bar{\theta})] |_{s,R,L=0},$$

$$\Lambda(k) = -(\varepsilon - \xi - 2i\gamma) v e^{-s/2} + 2(|V|^2 v)^{1/2} \cdot \left[ \frac{-\lambda \exp(4-s/2)}{\xi + i\pi g \langle V^2 \rangle} \right]^{1/2} \cdot \cos\left(\theta - i\frac{L}{2} + i\frac{s}{4}\right) + \left(\frac{\sigma \exp(R-s/2)}{e - i\pi g \langle V^2 \rangle}\right)^{1/2} \cdot \cos\left(\bar{\theta} - i\frac{R}{2} + i\frac{s}{4}\right) + 2i(f\tau)^{1/2} v [x \cos(\theta + \bar{\theta}) + y \sin(\theta + \bar{\theta})] + f\tau v^2. \quad (10)$$

Here  $\Phi$  is a contour around the origin, and  $C(\mu)$  is a contour consisting of a ray which starts from the origin and goes off

to infinity at an angle  $\mu$  from the real axis. The phase  $\mu$  is chosen in such a way that all the integrals converge. The quantity  $g(V)$  is the spectral density of those levels for which the matrix element of the interaction with the ground state lies in a  $dV$  interval around  $V$ .

Expression (9), along with expressions (10), describes the behavior of the population of a state associated with a band of levels between which the transitions described by the transfer function  $f(\varepsilon, \xi)$  occur as a result of a mediated interaction through a dense band. Although this point cannot be seen directly from the expressions written here, the population of the ground state remains equal to unity at all times in the case  $V \equiv 0$ . To verify this physically obvious fact, it is necessary to carry out several transformations which consist basically of using the relations found for the Bessel functions after integrating over  $d\theta$  and  $d\bar{\theta}$ , introducing the new variable  $J = A_1 - A_2$ , and integrating by parts.

For the transformations below, we make use of the specific functional form of  $g(V)$ , which makes it possible to substantially simplify the expressions derived above:

$$g(V) = (2gV/V_0^2) \exp(-V^2/V_0^2), \quad (11)$$

where  $V_0 \equiv \langle V^2 \rangle^{1/2}$  is the mean square matrix element of the transition operator, and  $g$  is the spectral density of all levels. Noting that  $V$  and  $\theta - \bar{\theta}$  can be treated as polar coordinates under integration, carrying out the corresponding integrations in terms of the equivalent Cartesian coordinates  $V \cos(\theta - \bar{\theta})$  and  $V \sin(\theta - \bar{\theta})$ , changing the order of the integration over  $d(\theta + \bar{\theta})$  and the differentiation, introducing the change of variables  $v \rightarrow \nu \exp(s/2)$  and then differentiating with respect to  $ds$ ,  $dL$ , and  $dR$ , we find an expression for the population. After the terms in the relation for the quantity  $J = A_1 - A_2$  which are proportional to  $\nu^{-1}$  are eliminated through the use of recurrence relations for the Bessel functions; after we use the identity

$$\exp(f\tau v^2) = \pi^{-1/2} \int_{-\infty}^\infty \exp[(f\tau)^{1/2} \nu Y - Y^2] dY; \quad (12)$$

and after we introduce the variables

$$Z = (\varepsilon - \xi - 2i\gamma) + V_0^2 [\sigma (\varepsilon - i\pi g V_0^2)^{-1} - \lambda (\xi + i\pi g V_0^2)^{-1}] + 2(f\tau)^{1/2} Y, \\ r^2 = 4 \{ V_0^2 [-\lambda \sigma (\varepsilon - i\pi g V_0^2)^{-1} (\xi + i\pi g V_0^2)^{-1}]^{1/2} + i(f\tau)^{1/2} x \}^2 - 4f\tau y^2, \quad \varphi = \arctg(y/x) \quad (13)$$

and

$$\sigma' = \xi [\xi (\varepsilon - i\pi g V_0^2)^{-1} - \lambda (\xi + i\pi g V_0^2)^{-1}], \quad \xi = \varepsilon - \xi, \quad \eta = \varepsilon + \xi,$$

$$z = \sigma (\varepsilon - i\pi g V_0^2)^{-1} + \lambda (\xi + i\pi g V_0^2)^{-1}; \quad (14)$$

this expression for the population takes the form

$$\rho = \frac{1}{2\pi^2 \xi} \int d\sigma' dz d\eta d\tau dx dy [(1-A_L)(1-A_R)(1-A_2)^{-1} - A_{RL}(1-A_2)^{-1} - |A_3|^2 (1-A_2)^{-2}] \exp(J - \sigma' - iz\eta - x^2 - y^2), \quad (15)$$

where

$$\begin{aligned}
J &= \frac{1}{2} \pi^{1/2} g \int_{-\infty}^{\infty} \{ -V_0^4 (\sigma'^2 \xi^{-2} + z^2) + [V_0^2 (\sigma'^2 \xi^{-2} - z^2)]^{1/2} \\
&\quad - 2i(f\tau)^{1/2} x \}^2 - 4f\tau y^2 \\
&\quad + V_0^2 \sigma' \xi^{-1} [\xi/2 - i\gamma + (f\tau)^{1/2} Y] \} \{ [V_0^2 (\sigma'^2 \xi^{-2} - z^2)]^{1/2} \\
&\quad - 2i(f\tau)^{1/2} x \}^2 \\
&\quad - [\xi - 2i\gamma + V_0^2 \sigma' \xi^{-1} + 2(f\tau)^{1/2} Y]^2 \}^{-1/2} \\
&\quad \exp(-Y^2) dY, \\
A_2 &= -\frac{gf\tau}{2i\pi^{1/2}} \int_{-\infty}^{\infty} (Z^2 - r^2)^{-1/2} \exp(-Y^2) dY, \\
A_3 &= -e^{i\varphi} \frac{gf\tau}{2i\pi^{1/2}} \int_{-\infty}^{\infty} [Zr^{-1} (Z^2 - r^2)^{-1/2} - r^{-1}] \exp(-Y^2) dY, \\
A_{R,L} &= i\pi^{1/2} gf\tau \int_{-\infty}^{\infty} \left[ \frac{V_0^2}{2} \left( \frac{\sigma'}{\xi} \pm z \right) Z (Z^2 - r^2)^{-1/2} - r^2 (Z^2 - r^2)^{-1/2} \right] \\
&\quad \times \exp(-Y^2) dY, \\
A_{RL} &= \frac{\pi^{1/2} gf^2 \tau^2}{2i} \int_{-\infty}^{\infty} [2 + 3r^2 (Z^2 - r^2)^{-1}] (Z^2 - r^2)^{-1/2} \\
&\quad \times \exp(-Y^2) dY. \tag{16}
\end{aligned}$$

We consider the following cases in more detail.

**a)** We assume  $f \equiv 0$  and  $\gamma \equiv 0$ . In other words, we assume that there is no interaction between band  $\{k\}$  and band  $\{\alpha\}$ . We then have  $A_2 = A_3 = A_{R,L} = A_{RL} = 0$ , and the expression for the population of the ground state becomes the same as that found in Ref. 6 for a level-band system. Under the condition  $V_0 g \ll 1$ , there is essentially no decay of the level, while at  $V_0^2 g^2 \gg 1$  a steady-state population  $\rho_0 \sim (V_0 g)^{-2}$  is reached after a long time.

**b)** The case of a nonreturning random walk ( $\gamma \neq 0$ , and  $f \rightarrow 0$  in the limit  $\xi \rightarrow 0$ ) corresponds to

$$A_R = A_L = A_{R,L} = A_2 = A_3 = 0, \quad J = -1/2 \pi g (V_0^2 \sigma' i \gamma / \xi)^{1/2}, \tag{17}$$

from which we find

$$\rho_0 = \frac{1}{\pi \xi} \int d\sigma' \exp \left\{ -\sigma' - \frac{\pi g}{2} \left( \frac{V_0 \sigma' i \gamma}{\xi} \right)^{1/2} \right\}, \tag{18}$$

and, at long times,

$$\rho_0 \sim (t \gamma g^2 V_0^2)^{1/2} \exp \{ -1/2 (t \gamma g^2 V_0^2)^{1/2} \}. \tag{19}$$

**c)** The case of an uncorrelated return random walk, i.e., the case  $\gamma \neq 0$ ,  $f(\xi, \eta) \rightarrow 2\pi i f(\xi)$ , corresponds to

$$\begin{aligned}
A_{R,L} &= 1/2 i \pi g [V_0^2 \sigma' \xi^{-1} (f\tau)^{1/2}]^{1/2} c_{R,L}(x), \\
A_{RL} &= 1/2 i \pi g [V_0^2 \sigma' \xi^{-1} (f\tau)^{1/2}]^{-1/2} c_{RL}(x), \tag{20}
\end{aligned}$$

$$\begin{aligned}
A_2 &= e^{-i\varphi} A_3 = 1/2 i g f \tau [V_0^2 \sigma' \xi^{-1} (f\tau)^{1/2}]^{-1/2} c_2(x), \\
J &= 1/2 \pi g [V_0^2 \sigma' \xi^{-1} (f\tau)^{1/2}]^{1/2} c_f(x),
\end{aligned}$$

where  $c_R(x)$ ,  $c_L(x)$ ,  $c_{RL}(x)$ ,  $c_2(x)$ ,  $c_f(x)$  are functions of the integration variable  $x$ , which are of order unity. Using the expressions given above; carrying out the integration  $d\sigma'$

(the  $1 - A_2$  pole) and over  $d\tau$  by the method of steepest descent, in which we make use of the small values of the quantities  $V_0^{-2} g^{-2}$ ,  $f^{-1}$ , and  $\xi$ ; taking a Gaussian integral over  $dy$  and a steepest-descent integral over  $dx$ , we find the following expression for the principal component of the population after a long time:

$$\rho_0(t) = \frac{(g V_0^2 t)^{1/4}}{g^2 f (g^{-1/2} V^{-1/2} t^{-1/2})} \exp[-\text{const}(g V_0^2 t)^{1/2}]. \tag{21}$$

**d)** The case of a returning random walk with a finite correlation time  $\tau_c$  leads to the following expressions for  $f$  and  $\gamma$  in the asymptotic expressions for large values of  $\eta$ , i.e., at  $\eta \gg \tau_c^{-1}$ :

$$\begin{aligned}
f(\xi, \eta) &\sim f(\xi) / \tau_c \eta^2, \\
\gamma(\varepsilon) + \gamma(\xi) &= \gamma(\xi, \eta) = \gamma(\xi, 0) / \tau_c^2 \eta^2. \tag{22}
\end{aligned}$$

Specifically large values of  $\eta$  are responsible for the incomplete decay of state 0. After a long time, we should retain in expression (13) for the quantity  $Z$  only terms of order  $V_0^2 \sigma'$  and of order  $V_0^2 \sigma' \xi^{-1} (f\tau)^{1/2}$ . Since in the limit  $\xi \rightarrow 0$  we assume  $f(\xi) \xi \rightarrow 0$ , we have  $A_2 \rightarrow A_3 \rightarrow A_{R,L} \rightarrow 0$ , and the integral over  $d\sigma'$  is evaluated by the method of steepest descent. The saddle-point value  $\sigma'$  is such that in the limit  $\xi \rightarrow 0$  both  $A_R$  and  $A_L$  tend toward zero. After the change of variables

$$\eta = (f(\xi) \tau / \tau_c \xi^2)^{1/2} \eta', \quad z = (\xi^2 \tau_c / f(\xi) \tau)^{1/2} z' \tag{23}$$

and an integration over  $d\tau$ , we find

$$\begin{aligned}
\rho_0 &\sim \int \frac{V_0 g}{\xi} U(\eta'; x) \\
&\quad \times \exp \{ -i \eta' z' - V_0^2 g^2 \Psi(\eta'; x) \} d\eta' dz' dx,
\end{aligned}$$

where  $\Psi(\eta'; x)$  and  $U(\eta'; x)$  are functions which are of order unity for arguments of order unity. Evaluating the integral over  $dz'$  and the integrals over  $d\eta'$  and  $dx$  by the method of steepest descent ( $V_0^2 g^2 \gg 1$ ), we find

$$\rho_0 \sim \xi^{-1} g^{-1} V_0^{-1} \exp(-V_0^2 g^2 \text{const}), \tag{24}$$

which yields, in the limit  $t \rightarrow \infty$ ,

$$\rho_0 \sim g^{-1} V_0^{-1} \exp(-V_0^2 g^2 \text{const}). \tag{25}$$

The correlation time  $\tau_c$  does not appear in the result for the population. It determines not the steady-state value  $\rho_0$  but the time which is required to reach a steady state. Specifically, since the saddle-point value is  $\eta' \sim 1$ , we have  $\eta \sim [f(\xi) \tau_0^{-1} \xi^{-2}]^{1/2}$ , so satisfaction of the condition  $\eta \gg \tau_c^{-1}$  requires

$$\xi^{-2} f(\xi) \gg \tau_c^{-1}. \tag{26}$$

Using  $\xi \sim t^{-1}$ , we find from this expression an estimate of the time required for the population to reach its asymptotic value.

### 3. DISCUSSION OF RESULTS

Limiting cases **b** and **c**, which correspond to a nonreturning random walk and an uncorrelated returning random walk, can be given a graphic interpretation on the basis of the idea that the spectra for Poisson and Dyson ensembles differ in "hardness." For this purpose we need to examine the

probability for returns of a random walk to the levels of band  $\{k\}$  as an effective interaction between them. A returning random walk in the asymptotic behavior at large  $t$  then corresponds to a strong interaction, while a nonreturning random walk corresponds to asymptotically noninteracting states.

The temporal behavior of the population of the ground state,  $\rho_0$ , as  $t \rightarrow \infty$  determines those realizations of the band which correspond to the slowest decay, i.e., bands which do not have levels which lie close to state 0 along the energy scale. The probability that there will be no levels of band  $\{k\}$  in a  $\Delta$  neighborhood of this state under the condition  $g\Delta \gg 1$  is of order  $\exp(-g\Delta)$  for a Poisson (noninteracting) ensemble and of order  $\exp(-g^2\Delta^2)$  (to within the coefficient of the exponential function) for Dyson ensembles. The rate of the decay of the population of level 0 to states of band  $\{\alpha\}$  through a level of band  $\{k\}$ , tuned an amount  $\Delta$  away from resonance, is determined by the composite matrix element  $W$ , which can be estimated to be

$$W(\Delta) \sim V_0^2 \gamma / \Delta^2. \quad (27)$$

The rate of the decay through this and all other levels of band  $\{k\}$ , detuned by a greater amount, is

$$W = \int_{|h| > \Delta} W(h) g dh \sim V_0^2 \gamma g |\Delta|^{-1}. \quad (28)$$

This is the decay rate of level 0 in the case in which the nearest of the states in the realization of the band  $\{k\}$  lies a distance  $\Delta$  away along the energy scale. We can thus write

$$\rho_0 \sim \exp(-V_0^2 \gamma g t / |\Delta|). \quad (29)$$

Taking an average of this quantity over the probability distribution for the formation of an energy gap of size  $\Delta$  in a Poisson ensemble  $\exp(-g\Delta)$ , we find

$$\begin{aligned} \rho_0 &= \int \exp\left(-\frac{V_0^2 \gamma g^2 t}{|\Delta| g} - g|\Delta|\right) g d\Delta \\ &\sim (V_0^2 \gamma g^2 t)^{-1/2} \exp[-2(V_0^2 \gamma g^2 t)^{1/2}], \end{aligned} \quad (30)$$

which agrees with expression (19). Taking an average of expression (29) over a Dyson ensemble,  $(g\Delta)^\alpha \exp(-g^2\Delta^2)$ , we find

$$\rho_0 \sim t^{\alpha/3} \exp[-\text{const}(\gamma V_0^2 g^2 t)^{3/2}], \quad (31)$$

which agrees to within the coefficient of the exponential function with expression (21). If the function  $f$  in the asymptotic region of small  $\xi$  is represented in the form  $\xi^{-\beta}$ , then there is complete agreement between expressions (21) and (31) under the conditions  $\alpha = 7 - \beta$ , and  $\gamma g \sim 1$ . Since we have  $1 > \beta > 0$  for a returning random walk, we find that the quantity  $\alpha$  lies in the interval  $7 > \alpha > 6$ . In other words, the interaction of states as the result of an uncorrelated returning random walk leads to the formation of ensembles which are harder than ordinary Dyson ensembles. Here, however, we are talking about that hardness which determines the coefficient of the exponential function in the asymptotic expression for the case of large separations between levels. Furthermore, since we are dealing with a repulsion of decaying levels in this example, it is totally meaningless to talk about their relative positions at distances smaller than the decay rate.

We should emphasize that the results found for cases **b** and **c** indicate that after long times the ground-state population is not self-averaging. It is dominated by systems which lead to a slow decay of the level and which are encountered only rarely in the ensemble. Consequently, the results found here cannot be used to describe any single system, and validity of the model requires the physical existence of an ensemble of different systems.

The situation is quite different in limiting case **d**, in which the wave functions have a finite correlation time, and the level decay is incomplete. The reason is that the asymptotic expression for its population should obviously be a continuous function of the microscopic parameters of the system (the extent to which the levels are "detuned," the matrix elements of the transition operator, etc.). Consequently, if the mean value of the population of state 0 is finite in the limit  $t \rightarrow \infty$ , then it must also be finite for the overwhelming majority of possible specific realizations of the system. The exceptional case comprise a set of measure zero. In other words, a finite value of the correlation time leads to the existence of a localized component of the population.

The role played by the requirement that the correlation time be finite can be understood by noting that only when this requirement is met does there exist a nonvanishing population flux from a state of band  $\{k\}$  to level 0. Specifically, if the quantity  $\sum_k V_{0k} \psi_k(t)$  (the flux of probability amplitude) varies irregularly with a typical correlation time  $\tau_c$ , its time integral can be estimated from

$$\begin{aligned} \sum_k \int_0^{t \gg \tau_c} V_{0k} \psi_k(t) dt &\sim \left(\frac{t}{\tau_c}\right)^{1/2} \sum_k \int_0^{\tau_c} V_{0k} \psi_k(t) dt \\ &\sim \sum_k \langle V_{0k} | \psi_k \rangle (\tau_c t)^{1/2}, \end{aligned} \quad (32)$$

which corresponds to diffusion population fluxes  $\sim V_0^2 \tau_c \sum_k \rho_k$ .

We would also like to call attention to the circumstance that the correlation time  $\tau_c$  does not appear in the expression for the asymptotic value of the population. This time determines not the steady-state value  $\rho_0$  but the time which is required to reach the steady state. Condition (26), which is a necessary condition here, means that the quantity  $gf(\xi)\xi^{-2}$  exceeds the number  $(g\tau_c^{-1})$  of levels which fall in a  $\tau_c^{-1}$  neighborhood of the resonance. We note that the variable  $\xi$  is related to the time by  $\xi \sim t^{-1}$ . We also note that the function  $f(\xi)$  signifies the Fourier transform of the probability for observing a particle at time  $t$  in some level  $k$  of band  $\{k\}$  under the condition that at  $t = 0$  the particle was at a level  $k' \neq k$  of this band and first returned to it at the time  $t$ . We then see that the quantity  $\xi^{-2} f(\xi) g$  is an order-of-magnitude estimate of the time integral of the total flux of the population which returns to band  $\{k\}$  by the time  $t \sim \xi^{-1}$ . When the value of this quantity per state of band  $\{k\}$  participating in the process (there are  $g\tau_c^{-1}$  such states) becomes of order unity, the decay of the ground state is terminated. The return fluxes of population from the band to the level stabilize its average population.

This statement means that a necessary condition for the termination of the decay is that the quantum-mechanical particle described by the Schrödinger equation (1) must be

in each state of band  $\{k\}$  which satisfies the resonance condition with a probability close to unity. It can also be asserted that the steady-state value of the level population is reached after the quantum-mechanical particle which was originally localized in a phase volume  $\mathcal{V} \sim (2\pi\hbar)^S$  of one state of the band goes, in the course of the random walk, into a phase volume corresponding to another state of the band. The time required for this event is essentially the Poincaré recurrence time for the minimum (consistent with quantum mechanics) phase volumes.

There is yet another interesting circumstance here. In a quantum-mechanical system consisting of an isolated level and a discrete band,<sup>17-19</sup> the band may be thought of as a continuum with smoothly varying parameters—the square amplitude of the transition probability and the state density—at times  $t < g$ , when the typical distance between levels,  $\delta \sim g^{-1}$ , is smaller than the uncertainty in their energy position,  $\sim t^{-1}$ . For such a system, the wave function of the ground state is

$$\psi_0(t) = \exp(-V^2 g t \text{ const}). \quad (33)$$

By a time  $t \sim g$ , i.e., by the limiting time for which the analysis is valid, the population of the level is

$$\rho_0 \propto \exp(-V_0^2 g^2 \text{ const}), \quad (34)$$

which agrees to within the coefficient of the exponential function with (25). At times  $t \gg g$ , “revivals”—return probability fluxes from the band to the level—begin to play an important role. These revivals subsequently lead to an increase in the population of the level, to a value  $g^{-2} V_0^{-2}$ , corresponding to the principle of detailed balance. Specifically, if  $t \gg g$ , and if the phase shift between neighboring states satisfies  $|\Delta_k - \Delta_{k-1}| t \gg 1$ , these population fluxes may be regarded as random, rapidly oscillating fluxes, equal on the average to  $g^{-1}$ . Equating the forward and return fluxes,  $\rho_0 V_0^2 g = g^{-1}$ , we find  $\rho_0 \sim V_0^{-2} g^{-2}$ . In other words, the population of the level is on the order of  $N^{-1}$ , where  $N = g(V^2 g)$  is the number of states of the band which have reached resonance.

The presence of an interaction between the levels of band  $\{k\}$ , mediated through the states of band  $\{\alpha\}$  in the course of the random walk, apparently has the consequence that the phases of the wave functions of levels  $k$  are interacting—locked together or correlated. The phase shift between neighboring levels, on the other hand, is not a random quantity. Accordingly, that linear combination  $\sum V_{\alpha k} \psi_k(t)$  of the wave functions of band  $\{k\}$  which determines the probability amplitude flux to the ground state can no longer be estimated from the wave functions of band  $\{k\}$  which determines the probability amplitude flux to the ground state can no longer be estimated from  $V_{\psi\Sigma} N^{1/2} V_0 / N^{1/2} \sim V_0$ , as in the summation of  $N$  randomly oriented vectors each of length  $V_0 / N^{1/2}$ . The uniform arrangement of the phases of the  $\psi$  functions on a circle leads to the estimate  $V_{\psi\Sigma} \sim V_0 \exp(-N)$ , which agrees to within the coefficient of the exponential function with expression (34) and which leads to expression (25). The phase capture suppresses the “revival” process.

In conclusion we would like to discuss two physical problems which can be solved through the use of the model system which we have been discussing here. The first of these

problems concerns the dynamics of the filling of the low-lying levels of polyatomic molecules in an electromagnetic field which is resonant with one of the vibrational modes. We are interested in the mechanism for the formation of a so-called cold ensemble, i.e., a significant fraction of the molecules which, despite the existence of an external field, either are not excited or are excited only slightly. This analysis can be carried out on the basis of the model of a multilevel band-type system.<sup>20</sup> The role of the factor which forms the ensemble of systems is played by the rotational motion of the molecule as a whole, which, by virtue of the thermal distribution of the molecules among rotational states and by virtue of the vibrational-rotational interaction, can lead to irregularities in the vibrational spectrum and in the matrix of the dipole-moment operator. At times shorter than the Poincaré time, such a system can be described by balance equations, and the average populations of the levels which have reached resonance decay in accordance with a random-walk law. Limiting  $\mathbf{d}$  makes it possible to describe the behavior of a system at times longer than the Poincaré recurrence time. State 0 is understood in this case as the only level which is occupied at  $t = 0$  (and which corresponds to the vibrational ground state); the states  $\{k\}$  are understood as levels which are dipole-accessible from this state; and the function  $f$  is understood as representing those transitions which, under the influence of the radiation, couple levels  $k$  through higher-lying states. If the spectral width of the absorption band is finite, so that its inverse—the correlation time of the random walk—is also finite, the decay by a random-walk law will come to a halt, the population distribution will reach a steady state, and a level which initially had a population  $\rho = 1$  will be populated only slightly:  $\rho \propto \exp(-V_0^2 g^2 \text{ const})$ . A situation of this sort corresponds to quantum-mechanical steady states localized within a region  $\mathcal{N} \propto \exp(V_0^2 g^2 \text{ const})$  in terms of band indices.

The existence of localized states can explain the formation of a cold ensemble during the infrared excitation of a system of small polyatomic molecules.<sup>21</sup> Specifically, if the density of quantum states in the region of the low-lying vibrational levels and the spectral widths of the bands are small, the time required for dissociation of the molecule,  $t_D$ , may be much longer than the Poincaré recurrence time for the low-lying levels,  $t_p$ . There will thus exist a time interval  $t_D \gg t \gg t_p$  within there are localized states in the low-lying levels. Because of the rapid growth of the number density of quantum levels with increasing energy, the population distributions corresponding to these localized states decay rapidly with increasing index of the excited level.

The second problem concerns the disruption of integrals of motion (quantum numbers) in nonlinear physical systems when they are subjected to a perturbation. In classical mechanics, a resonance between the periodic motions corresponding to these integrals can be achieved by appropriately choosing various values of the integrals of motion which are conserved with any prescribed accuracy in the unperturbed system. In systems with three or more dimensions, this situation can be arranged at essentially any point in the space of the action variables; i.e., the grid of resonances is dense everywhere (Ref. 22, for example). For this reason, if a perturbing interaction is not degenerate because of some symmetry, it will lead to the complete destruction of all the integrals in motion other than the energy, and it will

lead to the appearance of a stochastic motion over the entire constant-energy surface.

In the quantum-mechanical case, the situation is more complicated. On the one hand, because of the discrete nature of the spectrum it is not always possible to satisfy the resonance conditions. On the other hand, even in the absence of an intermediate resonance there may be an effective tunneling interaction. To what extent the integrals of motion are violated in the process can be determined by solving the problem discussed above. For this purpose, we can take state 0 to be any state of the unperturbed Hamiltonian which corresponds to a completely integrable motion and which can therefore be described by a set of quantum numbers  $\{n_i\}$ . A perturbation  $V$  of a sufficiently simple structure gives rise to probability amplitudes for transitions from this state to other eigenenergy states of the unperturbed Hamiltonian with quantum numbers  $\{n'_i\}$  which differ from  $\{n_i\}$  by a relatively small change in the values of  $n_i$  in a relatively small number of positions. The set of these states, along with those which are reached in higher-order perturbations in  $V$  as a result of tunneling through greatly "detuned" levels, should be treated as a band of levels  $\{k\}$ .

If, on the other hand, we also know that this interaction is capable of leading to the formation of a stochastic layer (states for which the nondiagonal terms in  $V$  are greater than the energy differences) for at least a relatively small fraction of the eigenenergy states, and if the random walk corresponding to this layer is a returning walk, then we can choose as band  $\{\alpha\}$  the set of eigenenergy states which are formed in the layer when  $V$  is taken into account. If the correlation time of the random walks (the reciprocal of the width of the stochastic layer in energy space) is finite, the state does not decay completely—only to a magnitude  $\exp(-g^2V^2)$ . A nonreturning walk and an infinitely short correlation time may lead to the complete violation of the integrals of motion (quantum numbers). It can also be assumed that approximately  $\exp(-g^2V^2) \cdot 100\%$  of the total number of quantum states are states which have not decayed and which are described by the previous quantum numbers. A fraction of undecayed states of this magnitude corresponds to realizations of the  $\{\alpha\}$  band which have no levels in a  $V$  neighborhood of the resonance. The quantity  $\exp(-g^2V^2)$  under the condition  $gV \gg 1$  describes the probability for such a realization in Dyson ensembles.

We wish to thank N. V. Karlov for a discussion of these results.

## APPENDIX

Let us go through the procedure for deriving (9) from expression (8). Using standard relations of the type  $x = (\partial/\partial y)_{y=0} \exp(xy)$  we introduce variables  $L_k, R_k, \alpha_{kj}$ , and  $s_k$ , which are the adjoints of  $n_{k0}, m_{k0}, p_{kj}$  and  $\sum_j p_{kj}$ , respectively, in the determinants. In this case the determinants become differential operators of the type

$$\det \left\| \frac{\partial}{\partial \alpha_{kj}} + \delta_{kj} \left( \frac{\partial}{\partial s_k} + \frac{\partial}{\partial R_k} \right) \right\|,$$

and they act on the expression as a whole. We then carry out a summation<sup>1)</sup> over  $n_{k0}$  and  $m_{k0}$ . As a result, Bessel functions arise. These functions can be expressed by means of the variables  $\theta_k$  and  $\vartheta_k$  in terms of the corresponding standard Sommerfeld integral representations. We then carry out a summation over  $p_{jk}$ , and as a result we find an exponential function of argument  $f\tau$ :

$$\sum_{i,k \neq j} \nu_k \nu_j \exp(i\theta_k + i\vartheta_k - i\theta_j - i\vartheta_j + \alpha_{jk}),$$

where  $\nu_k = \tau_k (\varepsilon - \xi - 2\gamma)^{-1} \exp(s_k/2)$ .

After the order of the summation and the integration is changed, this exponential function is acted upon by two differential determinant operators. As a result, the first determinant acquires, in place of the arguments  $\partial/\partial \alpha_{kj}$ , arguments  $(\dots) \exp \alpha_{kj}$  and itself becomes the object acted upon by the second determinant. The action of the second determinant on the exponential function leads to the appearance of the same arguments as in the first case, along with which we should retain the operator part  $\partial/\partial \alpha_{kj}$ . After this procedure is carried out, the variables  $\alpha_{kj}$  in the argument of this exponential function, "carried through" the differentiation operators  $\partial/\partial \alpha_{kj}$ , are assumed to be zero, and they become a bilinear form of variables corresponding to different levels:  $\sum_{j,k \neq j} a_k a_j^*$ . For the exponential function of this bilinear form there exists a two-dimensional integral representation  $[\int dx dy \exp(-x^2 - y^2) (\dots)]$  of the quantity  $\exp(xA + yB + C)$ , which is multiplicative in terms of the variables with different indices, where

$$A \sim \sum_k \operatorname{Re} a_k, \quad B \sim \sum_k \operatorname{Im} a_k, \quad C \sim \sum_k |a_k|^2.$$

In this step, the only quantity which is not multiplicative in terms of the variables corresponding to different levels is the product of the determinants. Since the order of the derivatives with respect to each of the  $\alpha_{kj}$  is no higher than the first, it can be written in the form

$$\det \left\| -f\tau \nu_k \nu_j + \exp[i(\theta_k + \vartheta_k - \theta_j - \vartheta_j)] \frac{\partial}{\partial \alpha_{kj}} \right. \\ \left. + \delta_{kj} \left( \frac{\partial}{\partial s_k} + \frac{\partial}{\partial R_k} \right) \right\| \det \left\| -f\tau \nu_j \nu_k (1 - \alpha_{jk}) + \delta_{jk} \left( \frac{\partial}{\partial s_k} + \frac{\partial}{\partial L_k} \right) \right\|.$$

Expanding the determinant of the sum of the two matrices in minors, we have

$$\sum_{\substack{(C) \subset (X) \\ (C') \subset (X)}} \det \left\| -f\tau \nu_j \nu_k - \delta_{kj} \left( \frac{\partial}{\partial s_k} + \frac{\partial}{\partial R_k} \right) \right\| \det \left\| -f\tau \nu_k \nu_j + \delta_{kj} \left( \frac{\partial}{\partial s_k} + \frac{\partial}{\partial L_k} \right) \right\|_{k \in (C), j \in (C')} \det \left\| \frac{\partial}{\partial \alpha_{kj}} \right\| \det \|\alpha_{kj}\|_{k \in (X)/(C), j \in (X)/(C')}$$

where  $\{C\}$  is the set of rows, and  $\{C'\}$  is the set of columns (which have identical numbers of elements). The first two determinants, however, are nonzero only if  $\{C\}$  and  $\{C'\}$  differ by no more than a single element. The product of the last two determinants is a numerical factor, equal to the number of nonzero terms in the determinant with a zero mean diagonal or a zero diagonal nearest the mean diagonal. The nonzero determinants can be written in multiplicative form:

$$\det\| -f\tau v_k v_j - \delta_{kj} \hat{c}_k \|_{k,j \in \{C\}}$$

$$= \frac{1}{2\pi i} \int_{\Phi} (-u^{-2} + u^{-1}) \prod_{k \in \{C\}} [f\tau v_k^2 (1+u) - \hat{c}_k] du,$$

$$\det\| -f\tau v_k v_j - \delta_{kj} \hat{c}_k \|_{k \in \{C\} \cup m, j \in \{C\} \cup n} = -f\tau v_m v_n \prod_{k \in \{C\}} (f\tau v_k^2 - \hat{c}_k),$$

where contour  $\Phi$  circumvents point 0 in the positive direction.

Since a summation is carried out over all possible carriers, a summation over minors can also be incorporated in it. The reason is that the minors differ from diagonal minors by no more than a single element. The transformation from a summation over carriers to an integration over the parameters of the levels and a summation over repeated levels with identical parameters is then made. In other words, we carry out a chain of transformations of the type

$$\sum_{\{X\}} \prod_{k \in \{X\}} a(V_k, g_k)$$

$$\rightarrow \prod_{\substack{V_k \in \{V\} \\ g_k \in \{g\}}} \sum_{n(V_k, g_k)=0}^{\infty} \frac{[N(V_k, g_k) a(V_k, g_k)]^{n(V_k, g_k)}}{n(V_k, g_k)!}$$

$$\rightarrow \exp \int N(V, g) a(V, g) dV dg,$$

where  $N(V_k, g_k)$  is the fraction of levels with parameters  $V_k$  and  $g_k$ , and  $n(V_k, g_k)$  is the number of their repetitions.

<sup>1)</sup> When the effect of the parts of the diagrams which have a tree topology is taken into account (this effect reduces to a renormalization,<sup>18</sup> and it generates imaginary increments in  $\varepsilon$  and  $\xi$ ).

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