Population dynamics in a quantum level-band-level system

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The dynamics of a level-band-level system is studied. The level populations are found as a function of the time, the strength of the interaction, the deviations from resonance, and the correlation properties of the matrix elements of the interaction operator. The resonance lines may become narrower as time elapses. The possible utilization of this effect in processes involving a resonant interaction of electromagnetic radiation with matter is discussed.

A quantum-mechanical level-band-level system arises in a natural way in several physical problems involving the interaction of electromagnetic radiation with matter. An example is the two-photon excitation of an electronic term of a molecule which occurs when the conditions for an intermediate resonance with states of a vibrational quasicontinuum are satisfied (Fig. 1). Another example is the application of electromagnetic radiation at two frequencies to an atom (Fig. 2), with the atom being excited into Rydberg or ionization states. One might also include here Raman scattering in intense fields (Fig. 3). From the quantum-mechanical standpoint, all these states correspond to the same level scheme (Fig. 4) in the quasienergy representation.

The behavior of a quantum level-band system[1] at times shorter than the state density g in the band (in a system of units with $\hbar = e = 1$) is well known. In this case the level population decays exponentially into the band. The behavior is well known at times greater than g (Ref. 2), at which revival processes[3,4]—inverse fluxes of population from the band to the level—come into play.

Level-band-level systems should be described in a similar way. One should bear in mind the important role which may be played and indeed is played by the correlation properties of the matrix elements of the operators representing the interaction of the level with the band. In the present paper we analyze the population dynamics in such a system under the assumption that only one level (the "lower level") is initially populated.

We intend to treat the problem in its simplest formulation, under the assumption that the interaction is turned on "instantaneously" at the time $\tau = 0$ and under the further assumption that this interaction does not give rise to additional transitions directly between states of the band. In other words, we are assuming that the operator structure of the perturbation causes transitions only between the levels and the band, while the states of the band interact with each other in such a way that there is no change in the positions of the levels in the band. Without any loss of generality, we can assume that the external fields are in the same direction. We will focus on finding the population of the "upper level" as a function of the time, the statistical properties of the matrix elements, and Δ_2 (the deviation from resonance).

We are interested in the course of the process both at relatively early times and in the limit $\tau \to \infty$. In the first of these cases, the solution of the problem is insensitive to the details of the spectrum, being universal in nature and being determined only by the gross characteristics of the system (in particular, the binary correlation function of the matrix elements of the interaction operator). In the second case, i.e., in the limit $\tau \to \infty$, in contrast, the behavior of the population is considerably more complex. In this second case the nature of the energy distribution of the levels becomes important. A natural way to study this asymptotic behavior is to ensemble-average,[5] which allows one to calculate average characteristics of the system which are insensitive to details of the spectrum. The actual form of the ensemble distribution function is of course determined by the particular problem.

We carry out an analytic solution of the problem of the level-band-level system in the asymptotic limit $\tau \to \infty$. We have managed to find this solution for the case in which the position of each of the levels of the band is statistically independent (the case of a so-called Poisson ensemble), with a model distribution function of the magnitude of the matrix elements of the interaction operator. A description of this sort is, strictly speaking, valid only for a system in which the band forms as a result of complex, multidimensional motions which do not fully interact with each other, as may be the case, for example, for a multidimensional vibrational

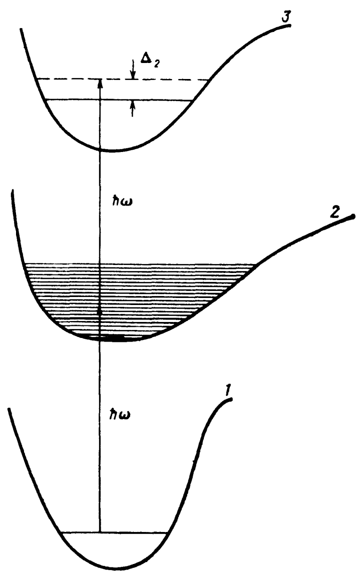

FIG. 1. Two-photon excitation of molecules.

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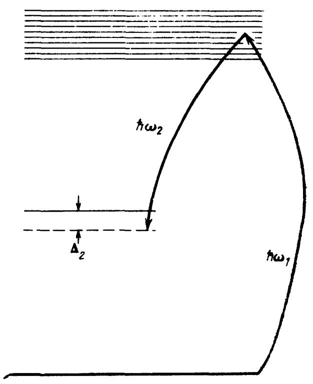

FIG. 2. Two-frequency action on an atom.

motion corresponding to an excited electronic term of a molecule. However, we believe that the results derived below are clearly of more general applicability, since—although we are not claiming a quantitative agreement—they do show the qualitative behavior of the population as a function of external parameters (the deviation from resonance and so forth), and they are pertinent to the description of the asymptotic behavior of other physical systems [Rydberg atoms, "destructive" quantum numbers (or integrals of motion), tunneling transmission, etc.].

It seems worthwhile to examine this question in more detail. The vibrational spectrum of a polyatomic molecule is a complex object. The energy position of each level is determined by a large number of parameters, so the specific relative positions of levels are quite different even in spectral regions lying quite close to each other. Under these conditions, even a relatively small change in the frequency of an electronic transition (caused by, for example, the Doppler effect) is capable of causing a substantial change in the particular picture of level positions near the resonance. Taking an average over such changes in frequency is of course equivalent to averaging over all possible realizations of the spectrum near the resonance. In other words, taking an average over one parameter is equivalent to taking an average over an ensemble of different realizations.

There is the important question of the nature of the ensemble probability distribution of such realizations, i.e., the distribution of the probability of realizations of different relative positions of the levels in different parts of the spectrum. One distinguishes between two limiting statistics: the case of a "hard" spectrum and the case of a "soft" spectrum. Hard spectra form as the result of an intense interaction, such that the pattern of level positions is formed as a result of a mutual repulsion of states. In such an interaction, the probability for neighboring levels to lie close together is low. Hard spectra are realized for motions which are fully stochastic.

A different situation arises in the case in which states interact with each other only slightly. The spectrum formed in the process is essentially a superposition of independent

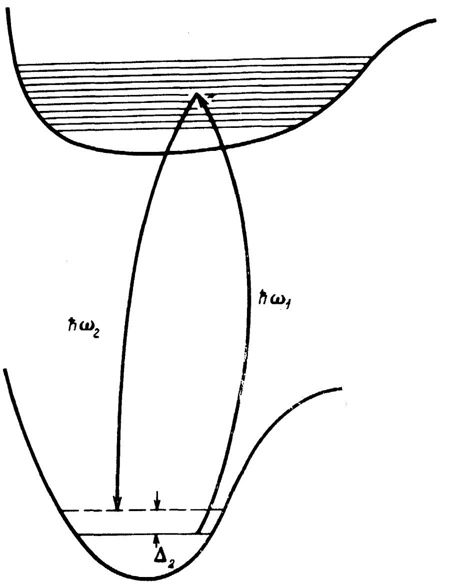

FIG. 3. Raman scattering ($\omega_{1,2}$ are the frequencies of the corresponding transitions).

spectra, and there can naturally be arbitrarily small differences in the distances between neighboring levels. The situation is typical of spectra formed as the result of a quantization of multidimensional motions which are independent in the different directions. This situation can be modeled by a factored ensemble distribution function which assumes a statistically independent position of each of the levels. This particular situation lends itself to a systematic analytic solution, and it is the subject of the present paper.

In the quasienergy representation, in the resonance approximation, the Schrödinger equation takes the following form after Fourier time transforms are taken:

$$
\begin{aligned}
\varepsilon\Psi_1 &= \frac{i}{2\pi} - \frac{\Delta_2}{2}\Psi_1 + E\Sigma\mu_{1n}\Psi_n,\\
\varepsilon\Psi_2 &= \frac{\Delta_2}{2}\Psi_2 + E\Sigma\mu_{2n}\Psi_n,\\
\varepsilon\Psi_n &= \Delta_n\Psi_n + E\mu_{n1}\Psi_1 + E\mu_{n2}\Psi_2.
\end{aligned}
\tag{1}
$$

Here $\Psi_{1,2}$ are the wave functions of the lower and upper levels; Ψ_n is the wave function of level n in the band; Δ_2, and Δ_n are the deviation of the upper level and that of level n in the band from resonance; E is the electric field; μ_{kn} and μ_{nl} are the matrix elements for dipole transitions ($k, l = 1, 2$); ε is the energy variable of the Ψ function; and the term $i/2\pi$ in the first equation in (1) means that only the lower level is initially filled. A solution of system (1) for Ψ_2 is

$$
\begin{aligned}
\Psi_2 = &-\frac{i}{2\pi}\left(E^2\Sigma\frac{\mu_{2n}\mu_{n1}}{\varepsilon-\Delta_n}\right)\left[\left(\varepsilon+\frac{\Delta_2}{2}-E^2\Sigma\frac{\mu_{1n}\mu_{n1}}{\varepsilon-\Delta_n}\right)\right.\\
&\times\left.\left(\varepsilon-\frac{\Delta_2}{2}-E^2\Sigma\frac{\mu_{2n}\mu_{n2}}{\varepsilon-\Delta_n}\right)-\left(E^2\Sigma\frac{\mu_{1n}\mu_{n2}}{\varepsilon-\Delta_n}\right)\left(E^2\Sigma\frac{\mu_{2n}\mu_{n1}}{\varepsilon-\Delta_n}\right)\right]^{-1}.
\end{aligned}
\tag{2}
$$

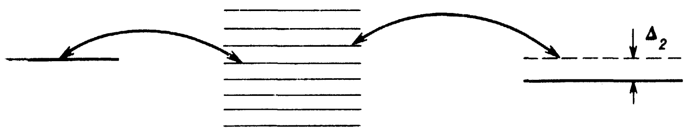

FIG. 4. Energy levels in the level-band-level system in the quasienergy representation.

From this point on the analysis depends on whether we are interested in times which are much shorter than the level density in the band ($\tau \ll g$) or much longer ($\tau \gg g$). In the early stage, by virtue of the uncertainty principle, $\Delta E \Delta \tau \sim 1$, the band can be treated as a continuum, and the summation in (2) can be replaced by an integration over the deviation from resonance. The result of this integration can be expressed in terms of the correlation functions of the matrix elements:

$$E^2 \sum \frac{\mu_{kn}\mu_{nl}}{\varepsilon - \Delta_n} \to 2\pi i g E^2 \langle \mu_k \mu_l \rangle, \qquad k, l = 1, 2.$$

After the roots of the equation quadratic in ε in the denominator in (2) are found in the particular case of a small deviation from resonance and a small difference between the correlation functions, we find, through the use of inverse Fourier transforms,

$$\Psi_2(\tau) \approx \frac{\langle \mu_1\mu_2 \rangle}{\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle} \Big\{ \exp[-2\pi g E^2 \tau (\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle)] - \exp\Big[-2\pi g E^2 \tau \frac{\langle \mu_1^2 \rangle \langle \mu_2^2 \rangle - \langle \mu_1 \mu_2 \rangle^2}{\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle} - \frac{\Delta_2^2 \tau}{2\pi g E^2 (\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle)} \Big] \Big\}. \tag{3}$$

We see that in the case $\langle \mu_1{}^1 \rangle \langle \mu_2{}^2 \rangle - \langle \mu_1 \mu_2 \rangle^2 \neq 0$, the population of the upper level reaches its maximum value $[\langle \mu_1 \mu_2 \rangle / (\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle)]^2$, in a time $\tau \sim [2\pi g E^2 (\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle)]^{-1}$ and then falls off as $\tau \to \infty$, under the condition $\Delta_2 = 0$, with a time scale $(\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle)[4g\pi E^2 (\langle \mu_1^2 \rangle \langle \mu_2^2 \rangle - \langle \mu_1 \mu_2 \rangle^2)]^{-1}$. For nonzero Δ_2, the upper level decays more rapidly. The frequency characteristic of the density of states of the upper level, $\rho_{22}(\Delta_2)$, is Gaussian with a time-dependent width $[4\pi g E^2 (\langle \mu_1^2 \rangle + \langle \mu_2^2 \rangle)/\tau]^{1/2}$. The formal replacement of the matrix elements of the dipole moment, $\mu_{2n} \to \mu_{2n} E_2/E$, solves the problem of the excitation of a system by two fields differing in intensity and the problem of two-photon excitation with different frequencies.

In the late stage, it is no longer valid to replace the summation over levels by an integration over the deviation from resonance. The temporal behavior of the system in this case depends on the details of the spectrum, the energy positions of each of the levels, and the size of the corresponding matrix elements. The behavior of the level population on the average over time is relatively insensitive to such details.[2] In this case we are justified in using an ensemble average,[4,6] in which the explicit expression for the quantity of interest here, $\rho_{22}(\tau, \{\Delta_n\}, \{\mu_{1n}\}, \{\mu_{2n}\})$, can be written in a finite analytic form after an average is taken over an ensemble with the distribution function $G(\{\Delta_n\}; \{\mu_{1n}\}, \{\mu_{2n}\}) \times \Pi d\Delta_n \Pi d\mu_{1n} \Pi d\mu_{2n}$, which determines the probability for the realization of a system with certain sets of values of the detuning Δ_n and with matrix elements μ_{1n}, μ_{2n}. The ensemble distribution functions depend on the nature of the processes which lead to the formation of the bands.

In the present paper we use the following ensemble distribution function:

$$G = \prod \{ [\Gamma^{-1}\theta(\Delta_n + \Gamma/2)\theta(\Delta_n - \Gamma/2)] [((1-\alpha)\delta(\mu_{1n} - \mu_{2n}) + \alpha\delta(\mu_{1n} + \mu_{2n}))\mu_{1n} \exp(-\mu_{1n}{}^2/\mu^2)^{\prime}/2\mu^2] \}, \qquad \Gamma \to \infty \tag{4}$$

[$\theta(x)$ is the unit step function, Γ is the width of the band, and μ^2 is the mean square of μ_{1n}^2 and μ_{2n}^2]. We are accordingly adopting the following assumptions: (1) The energy position of each of the band levels has a statistically independent distribution[1] over the wide (in the limiting case, infinite) interval from $-\Gamma/2$ to $\Gamma/2$. (2) The values of the matrix elements of the dipole moments μ_{1n} and μ_{2n} have a distribution which is statistically independent (independent of the energy position of the level). We are assuming that μ_{1n} and μ_{2n} are highly correlated with each other, always have identical magnitudes, differ in sign with a probability α, have the same sign with a probability $1 - \alpha$.

We use expression (2) and write Ψ_2 in a form convenient for subsequent averaging over the ensemble:

$$\Psi_2(\varepsilon) = \frac{1}{4\pi^3} \frac{\partial}{\partial \chi}\Big|_{\chi=0} \int \exp\Big\{ i\varepsilon(x^2+y^2+z^2+t^2) - \frac{i}{2}\Delta_2(x^2+y^2-z^2-t^2) - iE^2 \sum (\varepsilon - \Delta_n)^{-1} [\mu_{1n}{}^2(x^2+y^2) + \mu_{2n}{}^2(z^2+t^2) + 2\mu_{1n}\mu_{2n}(xz+yt+\chi)] \Big\} dx\,dy\,dz\,dt. \tag{5}$$

Here we have used the relation

$$(ab-c^2)^{-1} = -\pi^{-2} \int \exp[ia(x^2+y^2) + ib(z^2+t^2) - 2ic(xz+yt)]\,dx\,dy\,dz\,dt.$$

We denote by Q_n the expression in square brackets in (5). The ensemble average of the population of the upper level is found by multiplying (5) by its complex conjugate and taking an average with the distribution function (4). The variables $x, \ldots, t$ in the integral representation of the function Ψ_2^* analogous to (5) are denoted by $x', \ldots, t'$, respectively. We replace χ by the variable ψ:

$$\rho_{22}(\tau) = \int \Psi_2(\varepsilon)\Psi_2^*(\xi) e^{i\tau(\varepsilon-\xi)} d\varepsilon\, d\xi\, G \prod (d\mu_{1n}\, d\mu_{2n}\, d\Delta_n).$$

Since the ensemble distribution function, expression (5) for the population Ψ_2, and the corresponding expression for Ψ_2^* all factor, i.e., all break up into a product of functions for individual levels, the procedure of taking an average can be carried out as follows. Each of the n cofactors corresponding to a given level,

$$a_n = \int \exp\Big(-i\frac{E^2 Q_n}{\varepsilon - \Delta_n} + i\frac{E^2 Q_n'}{\xi - \Delta_n} \Big) \times \gamma(\Delta_n) g(\mu_{1n}, \mu_{2n})\, d\mu_{1n}\, d\mu_{2n}\, d\Delta_n, \tag{6}$$

differs only slightly from unity by virtue of our assumption $\Gamma \to \infty$, and we can use the expression

$$a_n = 1 + \Gamma^{-1} \int_{-\Gamma/2}^{\Gamma/2} [e^{(\ldots)} - 1]\, d\Delta_n\, g(\mu_{1n}, \mu_{2n})\, d\mu_{1n}\, d\mu_{2n} = \exp\Big\{ \Gamma^{-1} \int_{-\Gamma/2}^{\Gamma/2} [e^{(\ldots)} - 1] g(\mu_{1n}, \mu_{2n})\, d\mu_{1n}\, d\mu_{2n}\, d\Delta_n \Big\}, \tag{7}$$

where the ellipsis (...) in parentheses means the argument of the exponential function in (6). The expression which we have found does not depend on n. Consequently, the population, which is proportional to the product of factors α_n, contains a factor

$$\exp\Big\{ g \int_{-\Gamma/2}^{\Gamma/2} [e^{(\ldots)} - 1] g(\mu_1, \mu_2)\, d\mu_1\, d\mu_2\, d\Delta \Big\}, \tag{8}$$

where $g = N/\Gamma$ is the density of all the levels in the band (N is the total number of levels), and $g(\mu_1, \mu_2)$ in (6)–(8) represents the expression in the second set of square brackets in (4). In the course of the transformation, the contour (C) of the integration over $d\Delta$ undergoes changes. By subtracting one from the exponential function, we can get rid of the singularity at infinity. As a result, C becomes a figure-eight circumventing the essential singularities $\Delta = \varepsilon$ and $\Delta = \xi$. Integrating over $d\mu_1$, $d\mu_2$, and $d\Delta$; making the change of variables

$$\zeta=(\varepsilon-\xi)/E\mu,\quad \eta=(\varepsilon+\xi)/E\mu,\quad \Delta'=\Delta_2/E\mu,$$
$$g'=\pi g E\mu,\quad \tau'=\tau E\mu,\quad \chi\to\chi\zeta/2E\mu,\quad \psi\to\psi\zeta/2E\mu,$$

and introducing the two-dimensional vectors

$$\mathbf{x}=(\zeta/E\mu)^{1/2}\{x+z,\ y+t\},\quad \mathbf{y}=(\zeta/E\mu)^{1/2}\{x-z,\ y-t\},$$
$$\mathbf{x}'=(\zeta/E\mu)^{1/2}\{x'+z',\ y'+t'\},\quad \mathbf{y}'=(\zeta/E\mu)^{1/2}\{x'-z',\ y'-t'\},$$

we find the following expression for the population of level 2:

$$\rho_{22}(\tau')=\frac{1}{2^7\pi^6}\frac{\partial^2}{\partial\chi\,\partial\psi}\int \exp\Big\{\frac{i}{4}\zeta(\eta+\zeta)(\mathbf{x}^2+\mathbf{y}^2)+i\zeta\tau$$
$$-\frac{i}{4}\zeta(\eta-\zeta)(\mathbf{x}'^2+\mathbf{y}'^2)+\frac{i}{2}\zeta\Delta'(\mathbf{x}\mathbf{y}-\mathbf{x}'\mathbf{y}')$$
$$-\alpha g'\zeta\frac{(\mathbf{x}^2-\mathbf{x}'^2+\chi-\psi)^2-i(\mathbf{x}^2+\mathbf{x}'^2+\chi+\psi)}{[(\mathbf{x}^2-\mathbf{x}'^2+\chi-\psi)^2-2i(\mathbf{x}^2+\mathbf{x}'^2+\chi+\psi)-1]^{1/2}}$$
$$-(1-\alpha)g'\zeta\frac{(\mathbf{y}^2-\mathbf{y}'^2-\chi+\psi)^2-i(\mathbf{y}^2+\mathbf{y}'^2-\chi-\psi)}{[(\mathbf{y}^2-\mathbf{y}'^2-\chi+\psi)^2-2i(\mathbf{y}^2+\mathbf{y}'^2-\chi-\psi)-1]^{1/2}}\Big\}$$
$$\times d^2x\,d^2y\,d^2x'\,d^2y'\,d\eta\,d\zeta\,\zeta^2. \tag{9}$$

Let us use expression (9) in the asymptotic limit $\tau\to\infty$ to study the behavior of the population of the upper level as a function of the deviation from resonance, Δ_2, and as a function of the parameter α, which is the probability for "noncoincidence" of the signs of matrix elements μ_{1n} and μ_{2n} of the transition dipole moment. We assume that the number of levels in the band which are in resonance is large: $\pi g E\mu \gg 1$. Near the resonance ($\Delta_2 \lesssim E\mu$), for the case of fully correlated matrix elements of the dipole moment ($\alpha = 0$), the population of the upper level is 1/4 (see Appendix 1). If the correlation is not complete ($\alpha \ll 1$, but $\pi\alpha g E\mu \gg 1$), the population falls to a value $(2\alpha\pi g E\mu)^{-2}$. At deviations $|\Delta_2| > E\mu$ with $\alpha = 0$ (see Appendices 2 and 3), the population of the upper level is $4/(3\pi g E\mu)^2$. For $0<\alpha\ll 1$, with $\alpha\pi g E\mu \gg 1$, the population is $(2\pi\alpha g E\mu)^{-2}$. With $\alpha = 0$ and far from the resonance ($|\Delta_2| > E\mu$), the population of level 2 decreases with increasing deviation from resonance, in proportion to $(2E\mu/\Delta_2)^2$. With $\alpha\neq 0$ and $\alpha\pi g E\mu \gg 1$, this regime sets in at $|\Delta_2| > \alpha g E\mu$.

Figure 5 shows the population of the upper level as a function of the time and the deviation from resonance. Curve 1 corresponds to the case of fully correlated matrix elements for the (lower level)-band and band-(upper level) transitions. Over times $\tau \sim (8\pi g E^2\mu^2)^{-1}$ the population increases from zero to its maximum value. The spectrum of the population (the population as a function of Δ_2) then becomes Gaussian with a time-dependent width $(8\pi g E^2\mu^2/\tau)^{1/2}$. In other words, as time elapses the spectrum starts to become narrower, without a decrease in height. It reaches a steady state over a time[2] $\tau\sim g$. Curve 2 in Fig. 5 corresponds to the dynamics of the excitation with slightly uncorrelated matrix elements, $1\gg\alpha\gg(\pi g E\mu)^{-1}$. In the early stage, $\tau\ll g$, the line of the spectral characteristic not only contracts but also shrinks in amplitude, and at times $\tau\gtrsim g$ the spectrum becomes broader because the resonance is "smoothed out."

It should be kept in mind that after a time $\tau\sim(\pi g E^2\mu^2)^{-1}$ half the population is localized in states of the band, and at τ in the interval $(E^2\mu^2 g)^{-1}\ll\tau\ll g$ this is true of an ever-increasing fraction of the population, while there is an ever-decreasing fraction in level 2. However, for a system with a deviation $\Delta_2\neq 0$ from strict resonance the fraction of the population which is in level 2 falls off exponentially. In other words, the selective filling of level 2 becomes more pronounced, in accordance with $\exp(-\Delta_2^2\tau/gE^2\mu^2)$, while the absolute value of the "yield" falls off as $\exp(-E^2\mu^2\alpha g\tau)$. This behavior might be utilized for isotope separation: Level 1 would be the ground state of atoms having an isotopic shift, and level 2 an excited state of these atoms. The band would be the fairly closely spaced Rydberg states, which would be coupled with levels 1 and 2 by lasing transitions. The weak dependence of the transition matrix element on the index of the Rydberg level suggests that the correlation function α would be approximately zero and that this method, supplemented with a process for calculating the number of atoms in state 2, might allow a high population (yield) $\rho_{22} = 1/4$ to be combined with a high selectivity of the process.

Let us summarize the results of this study.

1. The population of the upper state increases to its

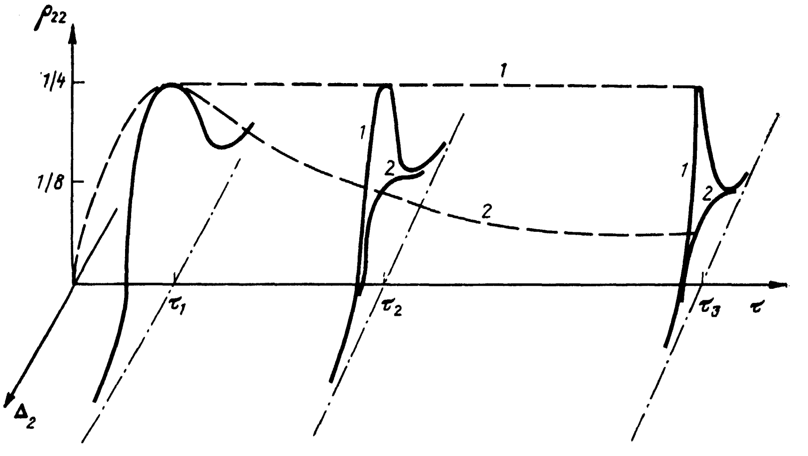

FIG. 5. Population of the upper level, ρ_{22}, as a function of the time τ and of the deviation from resonance, Δ_2. 1—$\alpha = 0$; 2—$\alpha\neq 0$. Dashed lines) $\Delta_2 = 0$; dot-dashed lines) sections at identical times $\tau_1\sim(8\pi g E^2\mu^2)^{-1}$, $\tau_1<\tau_2<g$, $\tau_3\gtrsim g$.

maximum value over a comparatively short time $\tau_1 \sim (8\pi g E^2 \mu^2)$.

2. At times greater than τ_1, at resonance ($\Delta_2 = 0$), with a complete correlation of the matrix elements for the level-band and band-level transitions ($\mu_{1n} = \mu_{2n}$), the population of the upper state remains $\rho_{22} = 1/4$. This conclusion follows from the symmetry of the problem. If there is a partial correlation, the population of the upper state remains finite at $\tau \gg \tau_1$, although it does fall off with increasing number of "decorrelated" levels which have reached resonance. The asymptotic value of the population of the upper level is zero only if there is no correlation at all. It is this particular case which was studied in Ref. 7.

3. When there is a deviation from resonance ($\Delta_2 \neq 0$), the population at each fixed time falls off with the deviation from resonance in a Gaussian fashion with a half-width inversely proportional to the square root of the interaction time. As a result, the spectrum becomes narrower as time elapses.

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APPENDIX 1

Let us evaluate the integral in expression (9) in the case $\Delta' = 0$ in the limit of an infinitely long time ($\tau' \to \infty$). For this purpose we switch to new integration variables:

$$\mathbf{x}^2 = z-\chi, \quad \mathbf{x}'^2 = z'-\psi, \quad \mathbf{y}^2 = t+\chi, \quad \mathbf{y}'^2 = t'+\psi.$$

Correspondingly, the measure of the integration, $d^2x d^2x' d^2y d^2y'$, becomes $\pi^4 dz dz' dt dt'$. In this notation, $\rho_{22}(\tau')$ becomes

$$\rho_{22}(\tau') = \frac{1}{2^7\pi^2}\frac{\partial^2}{\partial\chi\,\partial\psi}\int_\chi^\infty dz \int_\psi^\infty dz' \int_{-\chi}^\infty dt \int_{-\psi}^\infty dt' \int d\zeta\,\zeta^2\, d\eta$$
$$\times\exp\Big\{i\zeta\tau' + \frac{i}{4}\zeta^2(z+z'+t+t') + \frac{i}{4}\zeta\eta(z+t-z'-t')$$
$$-\alpha g'\zeta\frac{(z-z')^2-i(z+z')}{[(z-z')^2-2i(z+z')-1]^{1/2}}$$
$$-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')'}{[(t-t')^2-2i(t+t')-1]^{1/2}}\Big\}. \qquad \text{(A1)}$$

We integrate over $d\eta$ in this expression, obtaining a factor $8\pi\delta(z+t-z'-t')/\zeta$, as a result. We then differentiate with respect to the parameters χ and ψ:

$$\rho_{22}(\tau') = \frac{1}{16\pi}\int dt\, dt'\, d\zeta\,\zeta\,\delta(t-t')$$
$$\times\exp\Big[\frac{i}{2}\zeta^2 t + i\zeta\tau' - (1-\alpha)g'\zeta\frac{-2it}{(-4it-1)^{1/2}}\Big]$$
$$+\frac{1}{16\pi}\int dz\,dz'\,d\zeta\,\zeta\,\delta(z-z')$$
$$\times\exp\Big[\frac{i}{2}\zeta^2 z + i\zeta\tau' - \alpha g'\zeta\frac{-2iz}{(-4iz-1)^{1/2}}\Big]$$
$$+\frac{1}{16}\int dz\,dt'\,d\zeta\,\zeta\,\delta(z-t')\exp\Big[\frac{i}{2}\zeta^2 z + i\zeta\tau' - g'\zeta z\Big]$$
$$+\frac{1}{16\pi}\int dz'\,dt\,d\zeta\,\zeta\,\delta(z'-t)\exp\Big[\frac{i}{2}\zeta^2 t + i\zeta\tau' - g'\zeta t\Big]. \qquad \text{(A2)}$$

The last two terms in (A2) make vanishingly small contributions at long times. In the first two terms we change the integration variables z, $t = ir^2/\zeta$ and, using the asymptotic expression

$$\exp(i\zeta\tau')/\zeta = 2\pi i\delta(\zeta) + O(1/\tau'), \qquad \text{(A3)}$$

we integrate over $d\zeta$:

$$\rho_{22}(\infty) = \frac{1}{4}\int_0^\infty dr\, r\exp\Big(-\frac{r^2}{2}\Big)$$
$$\times\{\exp[-(1-\alpha)g'r] - \exp(-\alpha g'r)\}. \qquad \text{(A4)}$$

We thus see that we have $\rho_{22}(\infty) = (2\alpha g')^{-2}$ for $1 \ll \alpha g' \ll (1-\alpha)g'$ and $\rho_{22}(\infty) = 1/4$ for $\alpha g' \ll 1 \ll (1-\alpha)g'$.

APPENDIX 2

Let us evaluate the integral in (9) in the limit $\alpha = 0$. Evaluating the Gaussian integrals over d^2x and d^2x', we switch to the new integration variables t and t' which were introduced in Appendix 1:

$$\rho_{22}(\tau') = \frac{1}{8\pi^2}\frac{\partial^2}{\partial\chi\,\partial\psi}\int_{-\chi}^\infty dt\int_{-\psi}^\infty dt'\exp\Big\{\frac{i}{4}\zeta\Big(\eta+\zeta-\frac{\Delta'^2}{\eta+\zeta}\Big)(t+\chi)$$
$$-\frac{i}{4}\zeta\Big(\eta-\zeta-\frac{\Delta'^2}{\eta-\zeta}\Big)(t'+\psi) + i\zeta\tau'$$
$$-g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\Big\}.$$

In this expression we differentiate with respect to the parameters ψ and χ:

$$\rho_{22}(\tau') = \frac{1}{8\pi^2}\int\frac{\exp(i\zeta\tau')}{\eta^2-\zeta^2}d\eta\,d\zeta$$
$$-\frac{i}{2^5\pi^2}\int dt'\frac{d\eta\,d\zeta}{\eta^2-\zeta^2}\zeta\Big(\eta-\zeta-\frac{\Delta'^2}{\eta-\zeta}\Big)$$
$$\times\exp\Big[-\frac{i}{4}\zeta\Big(\eta-\zeta-\frac{\Delta'^2}{\eta-\zeta}\Big)t' + i\zeta\tau' - g'\zeta t'\Big]$$
$$+\frac{i}{2^5\pi^2}\int dt\frac{d\eta\,d\zeta}{\eta^2-\zeta^2}\zeta\cdot$$
$$\times\Big(\eta+\zeta-\frac{\Delta'^2}{\eta+\zeta}\Big)\exp\Big[\frac{i}{4}\zeta\Big(\eta+\zeta-\frac{\Delta'^2}{\eta+\zeta}\Big)t + i\zeta\tau' - g'\zeta t\Big]$$
$$+\frac{1}{2^7\pi^2}\int dt\,dt'\frac{d\eta\,d\zeta}{\eta^2-\zeta^2}\zeta^2\Big(\eta+\zeta-\frac{\Delta'^2}{\eta+\zeta}\Big)\Big(\eta-\zeta-\frac{\Delta'^2}{\eta-\zeta}\Big)$$
$$\times\exp\Big\{\frac{i}{4}\zeta\Big(\eta+\zeta-\frac{\Delta'^2}{\eta+\zeta}\Big)t - \frac{i}{4}\zeta\Big(\eta-\zeta-\frac{\Delta'^2}{\eta-\zeta}\Big)t'$$
$$+i\zeta\tau' - g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\Big\}. \qquad \text{(A5)}$$

The first term in (A5) is 1/4. In the second term we integrate first over dt' and then over $d\zeta$ and $d\eta$:

$$-\frac{1}{8\pi^2}\int\frac{d\eta\,d\zeta}{\eta^2-\zeta^2}\frac{{}^1/_4 i[(\eta-\zeta)^2-\Delta'^2]\exp(i\zeta\tau')}{g'(\eta-\zeta)+{}^1/_4 i[(\eta-\zeta)^2-\Delta'^2]} = -\frac{1}{4}+\ldots, \qquad \text{(A6)}$$

where the ellipsis (...) represents vanishingly small terms in the limit $\tau' \to \infty$. In the limit of long times the third term in (A5) also gives us a value of $-1/4$. The range of integration in the fourth term in (A5) can be broken up in a natural way into two subregions over the variables ζ and η. The contribution to the integral from the first subregion, $\zeta=0=\eta$, is 1/4. In the second subregion, $\zeta\neq 0\neq\eta$, we switch to new integration variables in accordance with

$$0\leqslant\theta\leqslant\pi,\quad t-t'=r\cos\theta/\zeta,\quad t+t'=ir^2\sin^2\theta/2\zeta^2.$$

The measure of the integration, $dtdt'$, in these new variables is $-ir^2\sin\theta dr d\theta/2\zeta^3$. The expression for the population is

$$\rho_{22}(\tau')=-\frac{i}{2^8\pi^2}\int dr\,r^2\,d\theta\sin\theta\frac{d\eta\,d\zeta}{\zeta(\eta^2-\zeta^2)}$$
$$\times\left(\eta+\zeta-\frac{\Delta'^2}{\eta+\zeta}\right)\left(\eta-\zeta-\frac{\Delta'^2}{\eta-\zeta}\right)$$
$$\times\exp\left[-\frac{r^2}{8}\sin^2\theta\left(1+\frac{\Delta'^2}{\eta^2-\zeta^2}\right)\right.$$
$$+i\zeta\tau'-\frac{i}{4}r\eta\left(1-\frac{\Delta'^2}{\eta^2-\zeta^2}\right)\cos\theta$$
$$\left.-g'r\left(\cos^2\theta+\frac{1}{2}\sin^2\theta\right)\left(1-\frac{\zeta^2}{r^2}\right)^{-1/2}\right].\qquad(A7)$$

Using the asymptotic expression (A3) in (A6), we carry out the integration over $d\zeta$. We then ignore the term in the argument of the exponential function which is quadratic in r by virtue of the relation $g'\gg 1$, and we integrate over dr:

$$\rho_{22}(\infty)=\frac{1}{2^6\pi}\int d\eta\,d\theta\left(1-\frac{\Delta'^2}{\eta^2}\right)^2$$
$$\times\sin\theta\left]g'\left(\cos^2\theta+\frac{1}{2}\sin^2\theta\right)\right.$$
$$\left.-\frac{i}{4}\left(\eta-\frac{\Delta'^2}{\eta}\right)\cos\theta\right]^{-3}\qquad(A8)$$

In this expression we use the approximation $1/2\sin^2\theta+\cos^2\theta=3/4$ and then integrate over the angle $d\theta$:

$$\rho_{22}(\infty)=\frac{3g'}{2^5\pi}\int d\eta\left(1-\frac{\Delta'^2}{\eta^2}\right)^2$$
$$\times\left[\left(\frac{3}{4}g'\right)^2+\frac{1}{16}\left(\eta-\frac{\Delta'^2}{\eta}\right)^2\right]^{-2}.\qquad(A9)$$

For "small" deviations from resonance, $1\ll|\Delta'|\ll g'$, for which large values of η are important in (A9), the population is $\rho_{22}(\infty)=4/9g'^2$. For large values of the deviation from resonance, $|\Delta'|\gg g'\gg 1$, at which small values of η are important in (A9), the population is $\rho_{22}(\infty)=4/\Delta'^2$.

APPENDIX 3

In this section we evaluate the integral in (9) for long times under the conditions $\alpha g'\gg 1$ and $|\Delta'|\gg 1$. For this purpose we switch to variables z, z', t, t', as in Appendix 1, and we introduce the two angles φ and φ':

$$\mathbf{xy}=[(z-\chi)(t+\chi)]^{1/2}\cos\varphi,\quad \mathbf{x'y'}=[(z'-\psi)(t'+\psi)]^{1/2}\cos\varphi'.$$

The measure of the integration in these variables is $dzdz'dtdt'd\varphi d\varphi'\pi^2/4$. After integrating over $d\eta$, we rewrite (9) in the new notation:

$$\rho_{22}(\tau')=\frac{1}{2^6\pi^3}\frac{\partial^2}{\partial\chi\,\partial\psi}\int_\chi^\infty dz\int_\psi^\infty dz'\int_{-\chi}^\infty dt\int_{-\psi}^\infty dt'$$
$$\times\int d\zeta\,\zeta\,d\varphi\,d\varphi'\,\delta(z+t-z'-t')$$
$$\cdot\exp\left\{i\zeta\tau'+\frac{i}{4}\zeta^2(z+t+z'+t')+\frac{i}{2}\zeta\Delta'[((z-\chi)(t+\chi))^{1/2}\right.$$
$$\times\cos\varphi-((z'-\psi)(t'+\psi))^{1/2}\cos\varphi']$$
$$-\alpha g'\zeta\frac{(z-z')^2-i(z+z')}{[(z-z')^2-2i(z+z')-1]^{1/2}}$$
$$\left.-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\right\}.$$

In this expression we differentiate with respect to ψ and χ:

$$\rho_{22}(\tau')=\frac{1}{2^6\pi^3}\int dt\,dt'\,d\zeta\,\zeta\,d\varphi\,d\varphi'\,\delta(t-t')\exp\left[\frac{i}{2}\zeta^2t+i\zeta\tau'\right.$$
$$\left.-(1-\alpha)g'\zeta\frac{-2it}{(-4it-1)^{3/2}}\right]+\frac{1}{2^6\pi^3}\int dz\,dz'\,d\zeta\,\zeta\,d\varphi\,d\varphi'\,\delta(z-z')$$
$$\times\exp\left[\frac{i}{2}\zeta^2z+i\zeta\tau'-\alpha g'\zeta\frac{-2iz}{(-4iz-1)^{1/2}}\right]$$
$$-\frac{1}{2^6\pi^3}\int dz\,dt'\,d\zeta\,\zeta\,d\varphi\,d\varphi'\,\delta(z-t')$$
$$\times\exp\left[\frac{i}{2}\zeta^2z+i\zeta\tau'-g'\zeta z\right]-\frac{1}{2^6\pi^3}\int dz'\,dt\,d\zeta\,\zeta\,d\varphi\,d\varphi'\,\delta(z'-t)$$
$$\times\exp\left[\frac{i}{2}\zeta^2t+i\zeta\tau'-g'\zeta t\right]$$
$$+\frac{\Delta'^2}{2^{10}\pi^3}\int dz\,dz'\,dt\,dt'\frac{(z-t)(z'-t')}{(zz'tt')^{1/2}}d\zeta\,\zeta^3\,d\varphi$$
$$\times\cos\varphi\,d\varphi'\cos\varphi'\,\delta(z+t-z'-t')\exp\left[\frac{i}{4}\zeta^2(z+t-z'-t')+i\zeta\tau'\right.$$
$$+\frac{i}{2}\zeta\Delta'[(zt)^{1/2}\cos\varphi-(z't')^{1/2}\cos\varphi']$$
$$-\alpha g'\zeta\frac{(z-z')^2-i(z+z')}{[(z-z')^2-2i(z+z')-1]^{1/2}}$$
$$\left.-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t-t')-1]^{1/2}}\right]$$
$$-\frac{i\Delta'}{2^8\pi^3}\int dz\,dt\,dt'\frac{z-t}{(zt)^{1/2}}$$
$$\times d\zeta\,\zeta^2\,d\varphi\cos\varphi\,d\varphi'\,\delta(z+t-t')\exp\left[\frac{i}{4}\zeta^2(z+t+t')+i\zeta\tau'\right.$$
$$+\frac{i}{2}\zeta\Delta'(zt)^{1/2}\cos\varphi-\alpha g'\zeta z$$
$$\left.-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\right]$$
$$+\frac{i\Delta'}{2^8\pi^3}\int dz\,dz'\,dt\frac{z-t}{(zt)^{1/2}}d\zeta\,\zeta^2\,d\varphi\cos\varphi\,d\varphi'\,\delta(z+t-z')$$
$$\times\exp\left[\frac{i}{4}\zeta^2(z+t+z')+i\zeta\tau'+\frac{i}{2}\zeta\Delta'(zt)^{1/2}\cos\varphi\right.$$
$$\left.-\alpha g'\zeta\frac{(z-z')^2-i(z+z')}{[(z-z')^2-2i(z+z')-1]^{1/2}}-(1-\alpha)g'\zeta t\right]$$
$$+\frac{i\Delta'}{2^8\pi^3}\int dz'\,dt\,dt'$$
$$\times\frac{z'-t'}{(z't')^{1/2}}d\varphi\,d\varphi'\cos\varphi'\,d\zeta\,\zeta^2\delta(t-z'-t')\exp\left[\frac{i}{4}\zeta^2(t+z'+t')\right.$$

$$+i\zeta\tau'-\frac{i}{2}\zeta\Delta'(z't')^{1/2}\cos\varphi'-\alpha g'\zeta z'$$

$$-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\Bigg]$$

$$-\frac{i\Delta'}{2^8\pi^3}\int dz'\,dz\,dt'\,\frac{z'-t'}{(z't')^{1/2}}d\zeta\,\zeta^2\,d\varphi\,d\varphi'\cos\varphi'\delta(z-z'-t')$$

$$\times\exp\left[\frac{i}{4}\zeta^2(t'+z'+z)+i\zeta\tau'-\frac{i}{2}\zeta\Delta'(z't')^{1/2}\cos\varphi'\right.$$

$$\left.-\alpha g\zeta\frac{(z-z')^2-i(z+z')}{[(z-z')^2-2i(z+z')-1]^{1/2}}-(1-\alpha)g'\zeta t'\right].\qquad(\mathrm{A}10)$$

The third and fourth terms in (A10) are vanishingly small. The sixth and eighth are equal to the first, with the opposite sign, while the seventh and ninth are equal to the second, again with the opposite sign. We will demonstrate this point using the sixth term as an example. Since we have $z\sim\zeta^{-1}$ and $t\sim\zeta^{-2}$, the term $\alpha g'\zeta z$ in the argument of the exponential function is negligible in comparison with $i\zeta\Delta'(zt)^{1/2}\cos\varphi/2$, while the term $-(1/4)i\zeta^2 z$ is negligible in comparison with $i\zeta^2(t+t')/4$. Replacing $z+t-t'$ by $t-t'$ in the argument of the δ-function, and replacing $(z-t)(zt)^{-1/2}$ by $(t/z)^{1/2}$ in the coefficient of the exponential function, we integrate over dz:

$$-\frac{1}{2^6\pi^3}\int dt\,dt'\,d\zeta\,\zeta^2\delta(t-t')d\varphi\,d\varphi'\exp\left\{\frac{i}{4}\zeta^2(t+t')\right.$$

$$\left.+i\zeta\tau'-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\right\}.\qquad(\mathrm{A}11)$$

We turn now to the fifth term in (A10). We integrate over $d\varphi$ and $d\varphi'$ by the method of steepest descent. After a long time, the nonvanishing constituent term (which we denote by A_5) of the fifth term in (A10) is

$$A_5=\frac{\Delta'}{2^7\pi^3}\int dt\,dt'\,dz\,dz'(z-t)(z'-t')(zz'tt')^{-3/4}d\zeta\,\zeta^2\cdot$$

$$\times\delta(z+t-z'-t')\exp\left\{i\zeta\tau'+\frac{1}{4}\zeta^2(z+t+z'+t')\right.$$

$$-\frac{i}{2}\zeta\Delta'[(zt)^{1/2}-(z't')^{1/2}]$$

$$-\alpha g'\zeta\frac{(z-z')^2-i(z+z')}{[(z-z')^2-2i(z+z')-1]^{1/2}}$$

$$\left.-(1-\alpha)g'\zeta\frac{(t-t')^2-i(t+t')}{[(t-t')^2-2i(t+t')-1]^{1/2}}\right\}.\qquad(\mathrm{A}12)$$

In (A12) we consider the limiting case $1\ll|\Delta'|\ll\alpha g'\ll(1-\alpha)g'$. For this purpose we switch to the new integration variables

$$z-z'=u/\zeta,\quad z+z'=v/\zeta^2,\quad t-t'=u'/\zeta,\quad t+t'=v'/\zeta^2.$$

Correspondingly, the measure of the integration, $dzdz'dtdt'$, becomes $dudvdu'dv'/4\zeta^6$. Using the asymptotic expression (A3), we can then integrate over $d\zeta$:

$$A_5=-\frac{i\Delta'}{2^7\pi}\int du\,dv\,du'\,dv'\frac{(v-v')^2}{(vv')^{3/2}}\delta(u+u')$$

$$\times\exp\left[\frac{i}{4}(v+v')\right.$$

$$\left.+\frac{i}{4}\Delta'u\frac{v-v'}{(vv')^{1/2}}-\alpha g'\frac{u^2-iv}{(u^2-2iv)^{1/2}}-(1-\alpha)g'\frac{u^2-iv'}{(u^2-2iv')^{1/2}}\right].\qquad(\mathrm{A}13)$$

The integral over du' in this expression can be eliminated with the help of the δ-function. Since we have $u^2\sim iv'$, we switch from the variable u to $\eta'=u(iv')^{1/2}$. After this substitution, we retain only the terms which are linear in η'. We are then in a position to integrate over $d\eta'$:

$$A_5=-\frac{i\Delta'}{2^7\pi}\int d\eta'\,dv\,dv'(v-v')^2(vv')^{-3/2}\exp\left[\frac{i}{4}(v+v')\right.$$

$$+\frac{i}{4}\Delta'(iv')^{1/2}\frac{v-v'}{(vv')^{1/2}}-\alpha g'\frac{i(v'-v)}{[i(v'-2v)]^{1/2}}-2i(1-\alpha)g'\eta'$$

$$\left.+\frac{i}{4}\Delta'\frac{v-v'}{(vv')^{1/2}}\eta'\right]=-\frac{i\Delta'}{2^6}\int dv'\,dv(v'-v)^2(vv')^{-3/2}$$

$$\times\delta\left(\Delta'\frac{v-v'}{4(vv')^{1/2}}-2(1-\alpha)g'\right)$$

$$\times\exp\left[\frac{i}{4}(v+v')+\frac{i}{4}\Delta'(iv')^{1/2}\frac{v-v'}{(v-v')^{1/2}}\right.$$

$$\left.-\alpha g'\frac{i(v'-v)}{[i(v'-2v)]^{1/2}}\right].\qquad(\mathrm{A}14)$$

Using $v\gg v'$, we eliminate the integration of dv in (A14) with the help of the δ-function:

$$A_5=-\frac{i}{8}\left[\frac{8(1-\alpha)g'}{\Delta'}\right]^2\int dv'$$

$$\times\exp\left\{\frac{i}{4}v'\left[1+\left(\frac{8(1-\alpha)g'}{\Delta'}\right)^2\right]\right.$$

$$\left.-2g'(1-\alpha)(iv')^{1/2}-\alpha g\frac{8(1-\alpha)g'}{\Delta'}\left(-\frac{iv'}{2}\right)^{1/2}\right\}.\qquad(\mathrm{A}15)$$

In the latter case, we use the change of variables $v'=2ir^2$:

$$A_5=\frac{1}{2}\left[\frac{8(1-\alpha)g'}{\Delta'}\right]^2\int_0^\infty dr\,r\exp\left\{-\frac{r^2}{2}\left[1+\left(\frac{8(1-\alpha)g'}{\Delta'}\right)^2\right]\right.$$

$$\left.-r\left[2^{3/2}(1-\alpha)g+\frac{8(1-\alpha)\alpha g'^2}{\Delta'}\right]\right\}.\qquad(\mathrm{A}16)$$

We thus have $\rho_{22}(\infty)=(2\alpha g')^{-2}$ under the condition $|\Delta'|\ll\alpha g'$.

We now consider the next limiting case: $\alpha g'\ll|\Delta'|\ll(1-\alpha)g'$. In this case we must add to (A13) a term

$$B_5=\frac{\Delta'}{2^7\pi^2}\int dz\,dz'(zz')^{1/4}(tt')^{-1/4}dt\,dt'\,d\zeta\,\zeta^2\delta(z-z')$$

$$\times\exp\left\{i\zeta\tau'-\frac{i}{2}\zeta\Delta'[(zt)^{1/2}-(z't')^{1/2}]-\alpha g'\zeta\frac{-2iz}{(-4iz-1)^{1/2}}\right\},\qquad(\mathrm{A}17)$$

which is found from (A12) in the region of "small" values of t and t'. We change integration variables in (A17):

$$t\to x=\zeta\Delta'(zt)^{1/2}/2,\quad t'\to x'=\zeta\Delta'(z't')^{1/2}/2.$$

Evaluating the Gaussian integral over the variables x and x' over the half-interval, we find an expression which is the same as the second term in (A10). In this limiting case we thus have $\rho_{22}(\infty)=4/\Delta'^2$, as follows from (A16).

[1)] A statistically independent distribution of the energy positions of each of the levels—a Poisson ensemble—arises if the system is not fully stochastic—if not all the degrees of freedom have been drawn into in the chaotic motion.

[2)] In contradiction of the result of Ref. 7, where there was no nonvanishing population of levels at large values of τ.

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