Markovian and rotating-wave approximations versus exact solution of damped quantum oscillator

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The system of one harmonic oscillator interacting with a continuum is studied. The problem is solved for two cases: the exact (full) Hamiltonian and the truncated Hamiltonian [the rotating-wave approximation (RWA)]. Comparing these two solutions, we formulate the conditions of the applicability of the RWA. The case in which the solution can be treated as Markovian is also discussed. The conditions for the existence of discrete levels are obtained. It was shown that under certain conditions, nonoscillating terms in the solution, leading to the instability of the whole system, can exist. Such terms cannot be obtained within either the Markovian or the rotating-wave approximation. Two particular cases of coupling are proposed and the results of numerical simulations are discussed.

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I. INTRODUCTION

In many cases we need to describe physical systems that can be called "open" systems, that is, the interaction with the environment should be taken into account. One of the problems of this type is the effect of quantum damping, when a relatively simple system is coupled to a continuum: a large number of oscillators or a heat bath. The analysis of such problems is rather complicated from mathematical point of view and analytical results may be obtained only in the most simple situations, for example, for one discrete state, a two-level system, or a harmonic oscillator.

The traditional way to describe time development of a system is the master equation. Its principal feature is neglecting memory effects, that is, the behavior of a system is determined by its configuration only at the present time. Of course, this assumption does not always hold; the usual condition if its validity is the smallness of the characteristic correlation time in comparison with the relaxation time of the system (see, for example, [1,2]). It was discovered, however, that in some cases this condition is satisfied; nevertheless, memory effects are essential. This means that the Markovian approximation, which is essential for the derivation of the master equation, does not hold for such problems.

Another restriction of the applicability of the Markovian approximation is the following. The recently developed technique of ultrashort pulses allows us to obtain information about a system at very short times, less than the correlation time of the system. Obviously, the behavior of the system at such times cannot be described within the Markovian approximation. As a consequence, the problem of developing theoretical methods that do not use the Markovian approximation becomes actual.

The problem of one discrete state interacting with a continuum, which is closely connected with the phenomenon of autoionization, is well known. It was treated long ago by Rice [3] and later by Beutler [4] and Fano [5]. The energy spectrum of the combined system is usually continuous. However, under certain conditions the spectrum may also contain an isolated discrete level. Thus the system may remain on this level arbitrarily long. The existence of such states and their properties was discussed by Riess [6], Rosenfeld, Voigt, and Mead [7], and Gelbart and Jortner [8].

The second problem, the interaction between a two-level system and a continuum, is also of great interest [9,25,26]. It is well known that this problem is equivalent to the tunneling between two wells in a dissipative media. It was shown by Fain [9] that this system may also have undamped solutions. In the latter work the conditions for the existence of new discrete levels were obtained and it was shown that under these conditions tunneling friction vanishes. The phenomenon was called *supertunneling.*

One of the most recent reviews of the problem of spin 1/2 interacting with the harmonic solid was done by Leggett *et al.* [10]. However, the authors did not discuss the possibility of the existence of isolated modes.

The third problem of this type, which will be the subject of the present paper, is the description of a harmonic oscillator coupled to a continuum. Unlike most other problems dealing with memory effects, the problem of the harmonic oscillator has an exact solution. This enables us to use it as a test case that helps to evaluate the applicability of various approximations.

The problem of the harmonic oscillator was considered by many authors. The case of weak damping (small coupling) is well developed. The complete analysis was done, for example, by Louisell [11]. The equation derived by Louisell was used later in several papers [12–14], where it was applied to some particular cases. Most of the authors

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dealt with the Markovian approximation: the system was assumed to have no memory.

In a recent work of Harris [15] an exactly solvable model was proposed and the results are valid for an arbitrary value of the coupling coefficient. The author used a model based on the Hamiltonian of Unruh and Zurek [16], in which the coupling between a separate oscillator and the bath is assumed to be independent of frequency in the whole range of frequencies $(0, \infty)$, and obtained the exact solution, which gives strictly exponential relaxation of the system. But this model is not self-consistent since such coupling implies the infiniteness of the average of operators p^2 and q^2. To avoid this difficulty the author has to make a cutoff, that is, to introduce the upper frequency. But then the results can no longer be regarded as exact, but only as an approximation.

In the past few years a number of works dealing with the non-Markovian damping appeared, but all of them discuss only particular cases of coupling. One of the earliest works of this kind was performed by Süsse *et al.* [17], in which a specific form of frequency dependence of coupling coefficients was chosen in order to obtain analytical results. But that form of frequency dependence does not lead to local modes, so the authors limited themselves only to the effects of nonexponential relaxation.

It is thus very interesting to discuss the general case and stress the difference between the results obtained within and beyond the Markovian approximation. Deviations from the Markovian behavior cause nonexponential relaxation and under certain conditions new discrete states, resulting from interaction between the oscillator and the continuum, may appear.

In the above-listed works the analysis was performed within the rotating-wave approximation. This approximation was discussed, for example, by Fain [9], but exact restrictions of its validity have not been yet obtained. When one tries to avoid it, very serious difficulties arise. Even relatively simple systems, such as a harmonic oscillator coupled to a two- or three-level system, require very complicated mathematical analysis [18,19].

A comparison between the rotating-wave approximation and the exact solution for the harmonic oscillator was presented by Ondrechen, Nitzan, and Ratner [20] and later by Lindenberg and West [21]. But the authors did not obtain nondamped solutions and discussed only relaxation terms.

There is one classical problem that lies very close to ours. It is the oscillations in a harmonic-oscillator chain with an impurity [22]. The authors obtained the conditions for the local modes to appear, in the situation when one element of the chain is replaced by a lighter one, and proved that this leads to the violation of the ergodic properties of the system.

We are going to choose the coupling coefficients in the most general form and avoid using the rotating-wave approximation. We obtain the terms representing the relaxation of the system and discuss when they have exponential behavior. The conditions for the existence of local modes are also obtained. It is shown that nonoscillating terms, leading to the instability of the system, may appear. We show that these terms may exist even in the cases usually assumed to be pure Markovian. We discuss why such terms do not appear in a harmonic-oscillator chain with an impurity, although at a first sight it seems to be a particular case of our problem. As a result, we will be able to write more exact conditions for the validity of the Markovian approximation. In Sec. III the analysis is repeated within the rotating-wave approximation. This helps us to discuss the applicability of this approximation. In Appendix B we propose and discuss two particular cases that allow us to obtain analytical results. Finally, we prove a general assertion that within the exact treatment one cannot obtain a pure exponential relaxation of a quantum system.

II. GENERAL CASE: THE FULL HAMILTONIAN

We write the Hamiltonian of the system in the form

$$H = \omega_0 a^\dagger a + \sum_\nu \omega_\nu b_\nu^\dagger b_\nu + \sum_\nu \left(G_\nu b_\nu + G_\nu^* b_\nu^\dagger\right)(a + a^\dagger), \tag{1}$$

where the first two terms represent the uncoupled system and the environment and the last one their interaction. We can reduce differential equations for each of the operators $a, a^\dagger, b_\nu, b_\nu^\dagger$ to a system of algebraic equations by performing the Laplace transform, solve it, and then carry out the inverse Laplace transform. This derivation is presented in Appendix A and here we give only the results.

For the operator $a(t)$, we get the expression

$$\langle a(t)\rangle = I(t) + \sum_k e^{iz_k t}\mathrm{Res}(z_k), \tag{2}$$

where z_k are local modes and $I(t)$ denotes the contour integral (A12) of Appendix A

$$I(t) = \frac{1}{\pi}\int_{\omega_1}^{\omega_2} \varphi(y)\frac{e^{iyt}(\omega_0 - y)^2 - e^{-iyt}(\omega_0 + y)^2}{\left[\omega_0^2 - y^2 - 2\omega_0 F(y)\right]^2 + 4\omega_0^2\varphi^2(y)}\langle a(0)\rangle\, dy$$

$$+\frac{1}{\pi}\int_{\omega_1}^{\omega_2} \varphi(y)\frac{\left(e^{iyt} - e^{-iyt}\right)\left(\omega_0^2 - y^2\right)}{\left[\omega_0^2 - y^2 - 2\omega_0 F(y)\right]^2 + 4\omega_0^2\varphi^2(y)}\langle a^\dagger(0)\rangle\, dy, \tag{3}$$

where

$$F(y) = -\mathrm{P}\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \left[\frac{1}{y-\omega} - \frac{1}{y+\omega}\right] d\omega, \qquad (4)$$

$$\varphi(y) = \pi |G(y)|^2 \chi_y(\omega_1,\omega_2) - \pi |G(-y)|^2 \chi_y(-\omega_1,-\omega_2), \qquad (5)$$

$$|G(\omega_\nu)|^2 = |G_\nu|^2 \rho(\omega_\nu). \qquad (6)$$

The P in (4) denotes the principal value. Here the functions $F(y)$ and $\varphi(y)$ are real, $F(y)$ is symmetric, $\varphi(y)$ is antisymmetric, and $\varphi(y) > 0$ for $y > 0$.

We first consider this term in the Markovian approximation. This case corresponds to weak coupling, that is, $F(y)$ and $\varphi(y)$ are assumed to be small and independent of y in the vicinity of ω_0. Under this condition we can expand the integration to the whole real axes and use the residue theorem. The zeros of the dominator can be easily found

$$y = \pm\sqrt{\omega_0^2 - 2\omega_0 F \pm 2i\omega_0\varphi} \approx \pm(\omega_0 - F) \pm i\varphi.$$

Hence we obtain the time dependence in the form $e^{\pm i(\omega_0 - F)t - \varphi t}$, the well-known result for the Markovian approximation.

In Appendix B we present some examples and calculate the value of this integral. We will see under what conditions $I(t)$ has a Markovian type of relaxation.

We now proceed to the second term in (2), the residues. It turns out (see Appendix A) that if poles exist, they are either pure real or pure imaginary. Let us consider these two cases separately.

If the poles have no real part, we get the modes without damping, that is, isolated modes that may lie only in the intervals $|y| < \omega_1$ or $|y| > \omega_2$ (note that ω_1 may be zero and ω_2 infinity; in that case isolated modes cannot exist).

Isolated modes exist if the following conditions are satisfied:

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{1}{\omega} d\omega < \omega_0/4,$$

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \left[\frac{1}{\omega-\omega_1} - \frac{1}{\omega+\omega_1}\right] d\omega > (\omega_0^2 - \omega_1^2)/2\omega_0$$

for the pole in the region $|y| < \omega_1$, (7)

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \left[\frac{1}{\omega_2-\omega} - \frac{1}{\omega_2+\omega}\right] d\omega > (\omega_2^2 - \omega_0^2)/2\omega_0$$

for the pole in the region $|y| > \omega_2$. (8)

In that case the oscillating terms corresponding to the local modes have no damping. Hence $\langle a(t)\rangle$ has no limit as $t \to \infty$; this result could not be obtained within the Markovian approximation.

The contribution to the sum of residues can also be made by the poles with zero frequency. In that case we obtain two solutions equal in absolute value and different in sign. The condition for these poles to exist can be written as

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{1}{\omega} d\omega > \omega_0/4. \qquad (9)$$

Comparing this result with (7) we see that the nonoscillating term due to this pole appears immediately after the disappearance of the isolated mode.

We can now describe the whole dynamics of the poles. When the coupling is not strong enough, none of the conditions is satisfied and the poles do not exist. As the coupling increases, two poles appear at the points $\pm i\omega_1$ and then approach the origin. The poles reach the origin when inequality (9) [or the first inequality in (7)] turns into equality. After this, the poles begin to diverge from the origin along the real axes. For such strength of coupling we obtain two nonoscillating solution: one of them is decreasing and the other is increasing.

Before we continue, we would like to say some more words about these nonoscillating terms because they look rather unusual. Instability of this kind can be obtained in much simpler cases, even in the problem of two interacting harmonic oscillators. This problem is, of course, classical, discussed in every book on mechanics (see, for example, Ref. [24]). Nevertheless, the possibility of the existence of such solutions is not described there. Let us look how the instability can be obtained.

If we consider two identical coupled oscillators with the Hamiltonian

$$H = \frac{1}{2}(p_x^2 + p_y^2) + \frac{\omega_0^2}{2}(x^2 + y^2) + \alpha xy,$$

we will immediately obtain the new frequencies of this system

$$\omega_1^2 = \omega_0^2 - \alpha, \qquad \omega_2^2 = \omega_0^2 + \alpha.$$

Obviously, if the coupling constant is sufficiently large, ω_1^2 will be negative, that is, we will obtain a nonoscillating solution. It should be noted that this solution *does not contradict* the law of energy conservation. The oscillator takes the energy from the potential energy: the situation when one of the frequencies becomes imaginary corresponds to the case when the potential energy is no longer described by a positive definite form. This means that the initial equilibrium point is no longer the point of a stable equilibrium and the infinitely increasing solution is restricted only by anharmonic terms.

When we consider a large number of oscillators, the situation is almost the same, but with one difference: in this configuration each oscillator of the thermal bath interacts with only one singled out oscillator with frequency ω_0. This means that the frequency shift of each of oscillators in the continuum is too small to give instability (its magnitude is proportional to $1/n$, where n is the number of oscillators in the bath).

The singled out oscillator, on the contrary, is coupled to the whole continuum and due to this its frequency

may become imaginary. Physically, this means that only anharmonic terms, even if they are very small, make the motion of the oscillator finite.

Compare the results with that for the harmonic-oscillator chain with an impurity [22]. In the latter case the nonoscillating terms do not appear. This difference can be explained in the following way. If we look at the Hamiltonian of our problem (1), we will see that it is possible to vary the coupling strength, while the frequency ω_0 remains constant. Hence inequality (9) can be satisfied if coupling is sufficiently strong.

On the other hand, the Hamiltonian used by Cukier and Mazur [22] is

$$H = \frac{1}{2}P^2 + \frac{1}{2}\Omega^2 Q^2 + \frac{1}{2}\sum_{k=1}^{2N}\left(p_k^2 + \omega_k^2 q_k^2\right) + \sum_{k=1}^{2N}\varepsilon_k q_k Q,$$

where p_k and q_k denote the momenta and coordinates of the equal mass particles and P and Q the momentum and coordinate of the impurity,

$$\omega_k^2 = \omega_0^2 \sin^2\frac{k\pi}{2(2N+1)}, \quad \omega_0^2 = \frac{4\alpha}{m}, \quad k = 1, 2, \ldots, 2N,$$

where m is the mass of the equal mass particle, α is the force constant, the interaction coefficients

$$\varepsilon_k = \begin{cases} -\frac{\mu^{\frac{1}{2}}}{2}\left(\frac{2}{2N+1}\right)^{\frac{1}{2}} \sin\frac{k\pi}{2N+1}, & k = 1, 3, \ldots, 2N-1 \\ 0, & k = 2, 4, \ldots, 2N, \end{cases}$$

the frequency

$$\Omega^2 = \mu\omega_0^2/2,$$

and μ is ratio of the masses of the regular particle and the impurity. This ratio plays the role of the function $G(\omega)$ in our problem. Varying the impurity mass corresponds to varying the coupling strength for the Hamiltonian (1).

If we write an inequality analogous to (9), then both the right- and left-hand sides will be proportional to coupling. It turns out that a pole equal to zero (corresponding to the translational motion of the chain as a whole) always exist; hence this inequality turns into an equality for all values of coupling: if we increase the latter, the two sides of (9) increase together and the condition of the existence of a nonoscillating solution can never be satisfied. This problem was also mentioned by Ullersma [23], the author did not discuss the consequences of the case when the Hamiltonian is not positive definite.

Now we have all the information that enables us to write down the conditions under which our solution can be assumed to be Markovian.

(i) The relaxation term $I(t)$ should have exponential behavior. It takes place in the well known case of weak coupling when the following condition is satisfied:

$$|G(\omega_0)|^2/\omega_0 \ll 1. \tag{10}$$

This inequality is usually referred to as the only condition of the validity of the Markovian approximation, although in the general case this is not so.

(ii) The discrete levels and nonoscillating terms should be absent. The solution can be called Markovian only if the second inequality in (7) and the inequality (8) are not satisfied. As shown in Appendix B, the discrete level above ω_2 usually appears when the lower one already exists. Hence the condition of the absence of isolated modes is

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{\omega\, d\omega}{\omega^2 - \omega_1^2} < (\omega_0^2 - \omega_1^2)/4\omega_0. \tag{11}$$

If this inequality is satisfied but the two sides of it are of the same order, the relaxation is still nonexponential because the domain that gives the main contribution to integral (A12) lies too close to the bounds of the continuum. Hence we should rewrite it as

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{\omega\, d\omega}{\omega^2 - \omega_1^2} \ll (\omega_0^2 - \omega_1^2)/4\omega_0. \tag{12}$$

This inequality is independent of (10). The ratio $|G(\omega)|^2/\omega_0$ may be small; nevertheless, the pole exists, and we cannot use the Markovian approximation. Some examples illustrating this assertion are given in Appendix B.

Moreover, a general statement that a quantum system cannot possess a pure exponential decay may be formulated. The exponential decay may be obtained only within certain approximations. In Appendix C this assertion is discussed in detail and its proof is given.

We also want to note the the above results are temperature independent. The initial temperature of the bath will appear in the formulas when we consider the relaxation of higher powers of the operator a: $aa^\dagger$, a^2, and so on.

In the next section we discuss the rotating-wave approximation and stress the difference between the general solution obtained above and the solution written within this approximation.

III. ROTATING-WAVE APPROXIMATION

In this section we repeat the above analysis within the rotating-wave approximation (RWA). A comparison of the results will help us to formulate the conditions of its applicability. Besides, this section is interesting from the following point of view: within the rotating-wave approximation the problem of an oscillator coupled to a continuum is identical to a two-level system coupled to a continuum, discussed by many authors (see, for example, Davidson and Kozak [26] (and references therein) or Fain [9]). Indeed, suppose that the oscillator is on its second level. If we consider an interaction with a continuum with the initial number of bosons equal to zero (for example, the problem of spontaneous emission of bosons), the only permitted transition is to the ground state. This means that all upper levels do not affect the behavior of system. In other words, the dynamics of our system coincides with that of a two-level system. If we add counterrotating terms to the Hamiltonian, these two problems become different: the oscillator can now rise to

higher levels and the behavior of these two systems will be different.

We thus see that within RWA two different problems have one and the same truncated Hamiltonian. Consequently, the analysis presented is useful not only for considering the harmonic oscillator, but it can also describe two-level system. (It is known that at the present time the latter problem cannot be solved without the rotating-wave approximation.)

This part of our work has something in common with the works of Davidson and Kozak, who used the rotating-wave approximation to study the relaxation and the lineshape of the emitted photon of a two-level system interacting with a continuous spectrum of radiation [25,26]. In Ref. [26] the authors studied the applicability of the Wigner-Weisskopf approximation and showed that for small values of coupling coefficient it gives the same results as the RWA. The behavior of isolated modes and deviations from the exponential decay within the RWA were discussed in [25].

We follow now the technique, presented in Sec. II. Within the RWA the Hamiltonian (1) is reduced to

$$H = \omega_0 a^\dagger a + \sum_\nu \omega_\nu b_\nu^\dagger b_\nu + \sum_\nu G_\nu \left(b_\nu a^\dagger + b_\nu^\dagger a\right). \quad (13)$$

The integral part of the solution is given by

$$I(t) = \frac{1}{\pi}\int_{-\omega_2}^{-\omega_1} \varphi_{\mathrm{RWA}}(y) \times \frac{e^{iyt}}{\left[\omega_0 + y - F_{\mathrm{RWA}}(y)\right]^2 + \left[\varphi_{\mathrm{RWA}}(y)\right]^2}\langle a(0)\rangle\, dy, \quad (14)$$

where

$$F_{\mathrm{RWA}}(y) = -\mathrm{P}\int_{\omega_1}^{\omega_2} G^2(\omega)\frac{d\omega}{y+\omega},$$
$$\varphi_{\mathrm{RWA}}(y) = G^2(-y)\chi_y(-\omega_1,-\omega_2) \quad (15)$$

and the poles are the solutions of the equation

$$s + i\omega_0 + iF_{\mathrm{RWA}}(y) + \mathrm{sgn}(x)\varphi_{\mathrm{RWA}}(y) = 0 \quad (16)$$

or

$$F_{\mathrm{RWA}}(y) = -(\omega_0 + y),$$
$$\varphi_{\mathrm{RWA}}(y) = -|x|. \quad (17)$$

Since $\varphi_{\mathrm{RWA}}(y) \geq 0$ for all y, it follows that there are no poles if $y \in (-\omega_2, -\omega_1)$; otherwise they have no real part. We represent this equation graphically in Fig. 1.

The conditions for existence of the poles are:

$$D \equiv F_{\mathrm{RWA}}(-\omega_2) > \omega_2 - \omega_0 \text{ in the region } y < -\omega_2,$$
$$E \equiv F_{\mathrm{RWA}}(-\omega_1) < \omega_1 - \omega_0 \text{ in the region } y > -\omega_1, \quad (18)$$

or, using the expression for $F_{\mathrm{RWA}}(y)$,

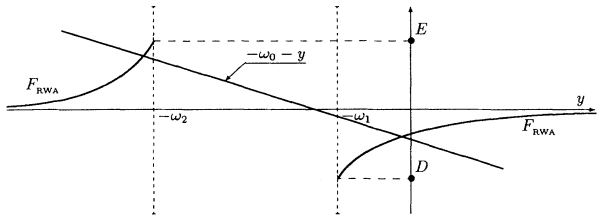

FIG. 1. Graphic solution of the secular equation in order to obtain isolated modes. The rotating-wave approximation.

$$\mathrm{P}\int_{\omega_1}^{\omega_2} G^2(\omega)\frac{d\omega}{\omega_2 - \omega} > \omega_2 - \omega_0 \text{ in the region } y < -\omega_2,$$
$$\mathrm{P}\int_{\omega_1}^{\omega_2} G^2(\omega)\frac{d\omega}{\omega_1 - \omega} < \omega_1 - \omega_0 \text{ in the region } y > -\omega_1 \quad (19)$$

At this stage we can stress the difference between the exact and the RWA solutions. First of all, we see that in the former case the pole in the region $|y| < \omega_1$ disappears as $\omega_1 \to 0$, whereas within the RWA it may not (see Fig. 1). The presence or absence of an isolated mode is a property that has a strong influence on the behavior of the system, so we can use conditions (19) as the conditions of applicability of the RWA. If they predict isolated modes, while the exact conditions (7) and (8) do not (or vice versa), it is obvious that we have to use the full Hamiltonian. If, for example, $\omega_1 = 0$, we cannot use the RWA if

$$\mathrm{P}\int_0^{\omega_2} G^2(\omega)\frac{d\omega}{\omega} \gtrsim \omega_0, \quad (20)$$

that is, when there exists a pole below zero.

If $\omega_1 \neq 0$ and coupling is sufficiently strong, condition (19) always predicts a pole, while the exact analysis leads to the result [Eq. (7)] that there exists a maximal boundary value of the coupling and the pole exists only in the case when coupling does not exceed it. So the following restriction appears: the RWA cannot be valid when conditions (19) predict a pole while (7) and (8) do not and vice versa.

The second difference between the RWA and the full Hamiltonian is the existence of the nonoscillating terms that can be obtained only within the exact consideration. These terms exist if [see (9)]:

$$P\int_{\omega_1}^{\omega_2} G^2(\omega)\frac{d\omega}{\omega} > \omega_0/4. \quad (21)$$

Hence, if the above inequality is satisfied, the RWA is also not valid.

IV. CONCLUSION

The conventional way to describe relaxation processes, such as the interaction between a simple system and a continuum, is based on the master equation. This equation is derived by using the Markovian approximation.

The commonly accepted conditions of its validity are the smallness of damping in comparison with the oscillator frequency [see inequality (10)]. The problem of the harmonic oscillator coupled to the continuum is one of the simplest models that can help us test the Markovian approximation because it is possible to obtain for it an exact solution.

We showed that the usual exponential relaxation takes place only for weak coupling and only in the case when the strength of coupling vanishes near the boundaries of the continuum. The usual condition of the validity of the Markovian approximation relates only to the behavior of the relaxation term $I(t)$, but it turns out that the solution may contain terms without damping and even nonoscillating terms with exponential increase. The latter leads to instability of the whole system. Obviously, in that case the behavior changes dramatically. We have seen that these parts of the solution may exist even for extremely small values of G^2/ω_0.

We have introduced the additional condition (12). When this condition is not satisfied, we cannot treat the behavior of the system as Markovian; both conditions (10) and (12) should be satisfied if we want to obtain the Markovian behavior of the system. Moreover, if (12) is satisfied but the two sides of it are of the same order, the relaxation is still not exponential.

The second important point of the present work is the comparison between the solution obtained within the rotating-wave approximation and the exact one. We have formulated the conditions under which these two solutions are close, that is, the rotating-wave approximation is valid. The application of this approximation to our problem is of independent interest. We showed that within the RWA the problem under consideration is identical to that of a two-level system coupled to a continuum. Hence the results obtained in Sec. III are also valid for the two-level system.

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APPENDIX A: FORMAL SOLUTION FOR THE FULL HAMILTONIAN

We write the full Hamiltonian of the system in the form:

$$H = \omega_0 a^\dagger a + \sum_\nu \omega_\nu b_\nu^\dagger b_\nu + \sum_\nu \left(G_\nu b_\nu + G_\nu^* b_\nu^\dagger\right)(a + a^\dagger), \tag{A1}$$

where the first two terms represent the uncoupled system and the environment and the last one their interaction. Solving differential equations for each of the operators $a, a^\dagger, b_\nu, b_\nu^\dagger$ by performing the Laplace transform, we obtain

$$\begin{aligned} a(t) &= \frac{1}{2\pi i}\int_{\varepsilon-i\infty}^{\varepsilon+i\infty} e^{st} a(s)\, ds \\ &= \frac{1}{2\pi i}\int_{\varepsilon-i\infty}^{\varepsilon+i\infty} e^{st}\Bigg(U_1(s)a(0) + U_2(s)a^\dagger(0) \\ &\quad + \sum_\nu \left[V_{1\nu}(s)b_\nu(0) + V_{2\nu}(s)b_\nu^\dagger(0)\right]\Bigg) ds, \end{aligned} \tag{A2}$$

where

$$U_1(s) = \frac{s - i\omega_0 + R(s)}{s^2 + \omega_0^2 + 2i\omega_0 R(s)}, \tag{A3}$$

$$U_2(s) = \frac{R(s)}{s^2 + \omega_0^2 + 2i\omega_0 R(s)}, \tag{A4}$$

$$V_{1\nu}(s) = \frac{s - i\omega_0}{s^2 + \omega_0^2 + 2i\omega_0 R(s)} \frac{iG_\nu}{s + i\omega_\nu}, \tag{A5}$$

$$V_{2\nu}(s) = \frac{s - i\omega_0}{s^2 + \omega_0^2 + 2i\omega_0 R(s)} \frac{iG_\nu^*}{s - i\omega_\nu}, \tag{A6}$$

$$R(s) = -\sum_\nu |G_\nu|^2 \left[\frac{1}{s + i\omega_\nu} - \frac{1}{s - i\omega_\nu}\right]. \tag{A7}$$

The integration path lies to the right of all singularities of the integrand.

To obtain these singularities and use the residue theorem we have to consider function $R(s)$ in detail. First of all we rewrite it as an integral, setting $s = x + iy$:

$$\begin{aligned} R(x + iy) = -i\int_{\omega_1}^{\omega_2} &|G(\omega)|^2 \\ &\times \left[\frac{1}{y - ix - \omega} - \frac{1}{y - ix + \omega}\right] d\omega, \end{aligned} \tag{A8}$$

where $|G(\omega_\nu)|^2 = |G_\nu|^2 \rho(\omega_\nu)$.

It turns out that $R(s)$ has branch cuts on the imaginary axes from ω_1 to ω_2 and from $-\omega_2$ to $-\omega_1$ (in the case $\omega_1 = 0$ and $\omega_2 = \infty$, the branch cut covers the whole axes). Hence

$$\lim_{x\to 0} R(x + iy) = iF(y) + \mathrm{sgn}(x)\varphi(y), \tag{A9}$$

where

$$F(y) = -\mathrm{P}\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \left[\frac{1}{y - \omega} - \frac{1}{y + \omega}\right] d\omega, \tag{A10}$$

$$\varphi(y) = \pi\,|G(y)|^2\,\chi_y(\omega_1, \omega_2) - \pi\,|G(-y)|^2\,\chi_y(-\omega_1, -\omega_2), \tag{A11}$$

and the crossed integral sign in (A10) denotes the principal part. Here the functions $F(y)$ and $\varphi(y)$ are real, $F(y)$ is symmetric, $\varphi(y)$ is antisymmetric, and $\varphi(y) > 0$ for $y > 0$.

Summarizing, we can say that the integral (A2) resides

in two different parts: the integral round branch cuts [denote it by $I(t)$] and, if the integrand has poles, the sum of residues.

1. Relaxation part of the solution

First consider the integral round the branch cuts $I(t)$. We consider the average value $\langle a(t)\rangle$,

$$\langle a(t)\rangle = \frac{1}{2\pi i}\int_{\varepsilon-i\infty}^{\varepsilon+i\infty} e^{st}\left[U_1(s)\langle a(0)\rangle + U_2(s)\langle a^\dagger(0)\rangle\right] ds.$$

[We have rewritten (A2) for the average values, assuming that initially the thermal bath was in equilibrium; hence $\langle b(0)\rangle = \langle b^\dagger(0)\rangle = 0$.]

The integrand has two branch cuts: one from $-\omega_2$ to $-\omega_1$ and the second from ω_1 to ω_2. Using (A3) and (A4) and the notation (A10) and (A11), we arrive at

$$I(t) = \frac{1}{\pi}\int_{\omega_1}^{\omega_2} \varphi(y)\frac{e^{iyt}(\omega_0-y)^2 - e^{-iyt}(\omega_0+y)^2}{\left[\omega_0^2-y^2-2\omega_0 F(y)\right]^2 + 4\omega_0^2\varphi^2(y)}\langle a(0)\rangle\, dy$$
$$+\frac{1}{\pi}\int_{\omega_1}^{\omega_2} \varphi(y)\frac{\left(e^{iyt}-e^{-iyt}\right)(\omega_0^2-y^2)}{\left[\omega_0^2-y^2-2\omega_0 F(y)\right]^2 + 4\omega_0^2\varphi^2(y)}\langle a^\dagger(0)\rangle\, dy. \tag{A12}$$

2. Discrete levels

Now we consider under what conditions functions U_i and V_i have poles and calculate residues in these points. The poles can be found from the equation

$$s^2 + \omega^2 + 2i\omega_0 R(s) = 0.$$

Separating the real and imaginary parts in $R(s)$, we obtain

$$R(x+iy) = xyF_1(x,y) + iF_2(x,y). \tag{A13}$$

It can be easily seen that $F_1(x,y) > 0$ for any x, y; $F_2(x,y) > 0$ for $y^2 < \omega_1^2$ and $F_2(x,y) < 0$ for $y^2 > \omega_2^2 + x^2$. Then,

$$x^2 - y^2 + \omega_0^2 - 2\omega_0 F_2(x,y) = 0, \tag{A14}$$
$$xy[1+\omega_0 F_1(x,y)] = 0. \tag{A15}$$

Since $F_1(x,y) > 0$, there are only two possibilities to satisfy equation (A15): $x = 0$ or $y = 0$. Hence, if poles exist, they are either pure real or pure imaginary. Let us consider these two cases separately.

3. The solution x=0: Poles without damping

As we have already seen, $\lim_{x\to 0}[xF_2(x)]$ is nonzero if $|y| \in (\omega_1, \omega_2)$, that is, Eq. (A15) cannot be satisfied for such y. If $|y| < \omega_1$ or $|y| > \omega_2$, we may omit the sign of principal value, since no singularities remain now on the integrating path, and rewrite (A14) substituting (A10),

$$\omega_0^2 - y^2 = 2\omega_0 \int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{2\omega\, d\omega}{\omega^2 - y^2}. \tag{A16}$$

If this equation has solutions, we obtain isolated modes, that is, oscillating terms without damping. Although we do not know the exact values of function $G(\omega)$, some general results can be obtained.

We can see from Eq. (A10) that $R(y)$ is a monotone function for $|y| < \omega_1$ and $|y| > \omega_2$. Its approximate behavior is presented in Fig. 2, where the left-hand side of (A16), the function $f(t) = (\omega_0^2 - y^2)/2\omega_0$, is also displayed. The intersection points of these two curves give us the locations of isolated modes and we can now write down the conditions of their existence.

It is easy to show that $A \equiv F(0) > 0$, $B \equiv F(\omega_1) > 0$, $C \equiv F(\omega_2) < 0$, and $F(\infty) = 0$. [B or C or both can go to infinity. This depends on the behavior of $G(\omega)$ near the points $y = \omega_1$ and $y = \omega_2$. If, for example, $G(\omega_1 + 0)$ and $G(\omega_2 - 0)$ tend to zero, B and C are finite.]

Hence isolated modes exist, provided the following conditions are satisfied:

$$A < f(0), B > f(\omega_1)$$

for the pole in the region $|y| < \omega_1$,

$$C < f(\omega_2) \quad \text{for the pole in the region } |y| > \omega_2,$$

or, using the expressions for A, B, C, and $f(\omega)$

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{1}{\omega}\, d\omega < \omega_0/4,$$
$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \left[\frac{1}{\omega-\omega_1} - \frac{1}{\omega+\omega_1}\right] d\omega > (\omega_0^2 - \omega_1^2)/2\omega_0$$

for the pole in the region $|y| < \omega_1$, (A17)

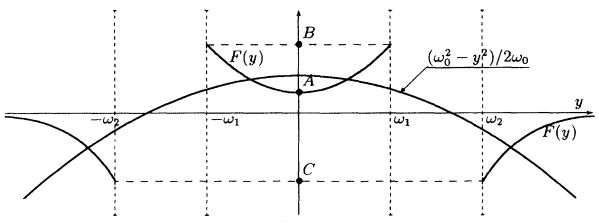

FIG. 2. Graphic solution of the secular equation in order to obtain isolated modes. The general case.

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \left[\frac{1}{\omega_2-\omega}-\frac{1}{\omega_2+\omega}\right] d\omega > (\omega_2^2-\omega_0^2)/2\omega_0$$

for the pole in the region $|y| > \omega_2$. (A18)

If isolated modes exist, the oscillating terms corresponding to them have no damping. As a consequence, $\langle a(t)\rangle$ has no limit as $t \to \infty$; this result could not be obtained within the Markovian approximation.

4. The solution y=0: Poles with zero frequency

In this case we perform similar operations and obtain the following equation for the poles:

$$\omega_0^2 + x^2 = 2\omega_0 \int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{2\omega\, d\omega}{\omega^2+x^2}. \quad \text{(A19)}$$

Similar to the previous analysis we can formulate the conditions of existence of the poles. It turns out that there are only two solutions of this equation, equal in absolute value and different in sign [since (A19) is a quadratic equation in x]. As the left-hand side of (A19) increases in x and the right-hand side decreases, we obtain the condition for the intersection to take place

$$\int_{\omega_1}^{\omega_2} |G(\omega)|^2 \frac{1}{\omega}\, d\omega > \omega_0/4. \quad \text{(A20)}$$

Comparing this result with (A17), we see that nonoscillating term due to this pole appears immediately after the disappearance of the isolated mode.

Now we have all the information about the behavior of the integrand in (A2) and can calculate this integral by the residue theorem. As already said, our solution consists of two parts and we can write it down in the most general form

$$\langle a(t)\rangle = I(t) + \sum_k e^{iy_k t}\mathrm{Res}(y_k) + e^{x_0 t}\mathrm{Res}(x_0) + e^{-x_0 t}\mathrm{Res}(-x_0).$$

APPENDIX B: EXACTLY SOLVABLE MODELS

In this section we apply the method presented above to a number of simple systems [that is, special cases of the function $G(y)$] to illustrate the results. As a first example we consider a constant coupling G inside the interval (ω_1,ω_2) and zero outside. The functions $F(y)$ and $\varphi(y)$ in that case take the form

$$F(y) = -G^2 \ln\left|\frac{y^2-\omega_1^2}{y^2-\omega_2^2}\right|,$$
$$\varphi(y) = \begin{cases} G^2 & \text{for } y\in(\omega_1,\omega_2) \\ -G^2 & \text{for } y\in(-\omega_2,-\omega_1) \\ 0 & \text{otherwise.}\end{cases} \quad \text{(B1)}$$

We can see that $F(\omega_1) = +\infty$ and $F(\omega_2) = -\infty$. So the condition of the existence of the isolate mode below ω_1 is

$$G < G_0 = \sqrt{\frac{\omega_0}{4\ln|\omega_2/\omega_1|}} \quad \text{(B2)}$$

and the mode above ω_2 exists always. When the coupling constant G exceeds G_0, the isolated mode turns into two nonoscillating terms, as described above. It follows that if $\omega_1 \to 0$ then $G_0 \to 0$, that is, there always exists the nonoscillating solution.

As a second example we take the parabolic profile of coupling that has a maximum between ω_1 and ω_2, vanishes at ω_1 and ω_2, and identically equals zero for $y < \omega_1$ and $y > \omega_2$. Inside the interval (ω_1,ω_2) we describe this profile as

$$|G(w)|^2 = 4G^2 \frac{(\omega_2-\omega)(\omega-\omega_1)}{(\omega_2-\omega_0)(\omega_0-\omega_1)}.$$

[The constant factor $(\omega_2-\omega_0)(\omega_0-\omega_1)$ was added to normalize the expression: the value of $G(\omega)$ at the point $\omega=\omega_0$ is now equal to G.]

Functions $F(y)$ and $\varphi(y)$ take the form

$$F(y) = -\frac{G^2}{(\omega_2-\omega_0)(\omega_0-\omega_1)} \times\left(\omega_1^2-\omega_2^2+(y-\omega_1)(y-\omega_2)\ln\left|\frac{\omega_2-y}{\omega_1-y}\right| +(y+\omega_1)(y+\omega_2)\ln\left|\frac{\omega_2+y}{\omega_1+y}\right|\right), \quad \text{(B3)}$$

$$\varphi(y) = \begin{cases} 4G^2(\omega_2-y)(y-\omega_1)/(\omega_2-\omega_0)(\omega_0-\omega_1) \\ \quad \text{for } y\in(\omega_1,\omega_2) \\ 4G^2(\omega_2+y)(y+\omega_1)/(\omega_2-\omega_0)(\omega_0-\omega_1) \\ \quad \text{for } y\in(-\omega_2,-\omega_1) \\ 0 \quad \text{otherwise.}\end{cases} \quad \text{(B4)}$$

In the latter case, because of the continuity of $G(\omega)$, the function $F(y)$ is finite at the points $y=\omega_1$ and $y=\omega_2$. Consequently, unlike the previous example, the poles do not exist if coupling is weak and appear only when it exceeds a certain value defined by Eqs. (7) and (8).

We would like to mention here once again the review of Leggett *et al.* [10]. The authors considered several cases of frequency dependence of the coupling strength. They introduced the spectral function $J(\omega)$ [an analog of the function $F(\omega)$ in our paper] and assumed its frequency dependence in the form $J(\omega) = A\omega^s e^{-\omega/\omega_c}$. The case $s=1$ was called the "Ohmic" case. In these terms the two cases considered by us are the following. The case of constant profile of function $G(\omega)$ corresponds to $s=-1$ and that of parabolic profile to $s=0$.

We performed numerical simulation and obtained the location of the poles for these two particular cases as a function of the coupling constant G. The results are presented in Fig. 3. We choose the following parameters: $\omega_2 = 1.5\omega_0$ and $\omega_1 = 0.5\omega_0$ [Fig. 3(a)] and $\omega_1 = 0.9\omega_0$ [Fig. 3(b)]. The continuous line represents the case of

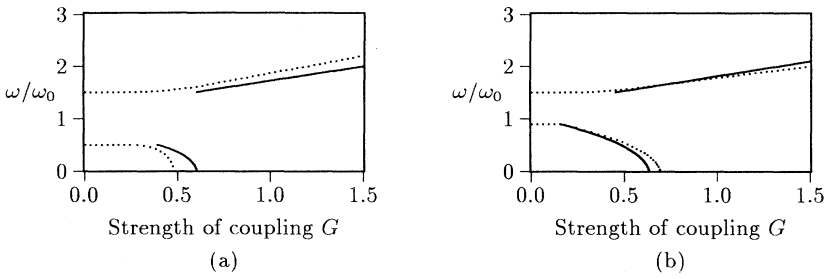

FIG. 3. Frequencies of the isolated modes as a function of coupling coefficient. (a) The case $\omega_1 = 0.5$. (b) The case $\omega_1 = 0.9$. The continuous line represents the parabolic coupling and the dashed line the constant coupling.

the constant coupling and the dashed one the parabolic coupling.

The relaxation term $I(t)$ can be calculated numerically by formula (A12). We have calculated this term as a function of t using different values of the parameters. The initial values were chosen to be $\langle a(0)\rangle = \langle a^\dagger(0)\rangle = 1$; ω_1, ω_2, and G were varied. The absolute value of $I(t)$ as a function of t is presented in Figs. 4(a)–4(d). Figs. 4(a) and 4(b) relate to constant coupling and Figs. 4(c) and 4(d) to parabolic one.

Figures 4(a) and 4(c) were calculated for $\omega_1 = 0.5\omega_0$ and $\omega_2 = 2.0\omega_0$ and the set of curves on each graph relates to different values of coupling constant G: $G = 0.1, 0.3, 0.7$. Figures 4(b) and 4(d) were calculated for $\omega_1 = 0.1\omega_0$, $\omega_2 = 5.0\omega_0$, and $G = 0.1, 0.3, 0.5$. The continuous lines represent the solution for the full Hamiltonian, and the dashed one for the RWA.

In all the figures we can see the oscillations of $\langle a(t)\rangle$ as a function of time. These oscillations come from the finiteness of the upper frequency ω_2 and are in good agreement with the results of Khalfin [28], where they were discussed.

We know that in the Markovian case relaxation is exponential. Our results show that deviations from this law of relaxation appear even for relatively weak coupling, when the isolated modes do not yet exist (see also Appendix C).

It is interesting to know whether the isolated modes appear for the parameters used in Fig. 4. Substituting expressions for the function $F(y)$ into (7) and (8) we obtain the conditions for coupling coefficient G. These results are summarized in Table I.

The presence of isolated modes can be easily seen by examining the starting points of the curves $I(t)$. The difference between $I(0)$ and $\langle a(0)\rangle$ relates to the total amplitude of the isolated modes [see Eq. (2)] and this can be a direct test for their existence.

As we see, for the case of constant coupling the isolated modes exist always, even for extremely small values of G^2/ω_0. This fact forces us to use the exact theory rather than the RWA, although it is commonly accepted that the validity of this approximation breaks down only for a strong interaction.

We want to add some words about the applicability of these two models used for numerical analysis. As known from the general theory, the profile of coupling multiplied by the density of states has some universal features [27]. It may have singularities in first derivative, which are the points at which the first derivative tends to infinity. Moreover, the Van Hove theorems (see, for example,

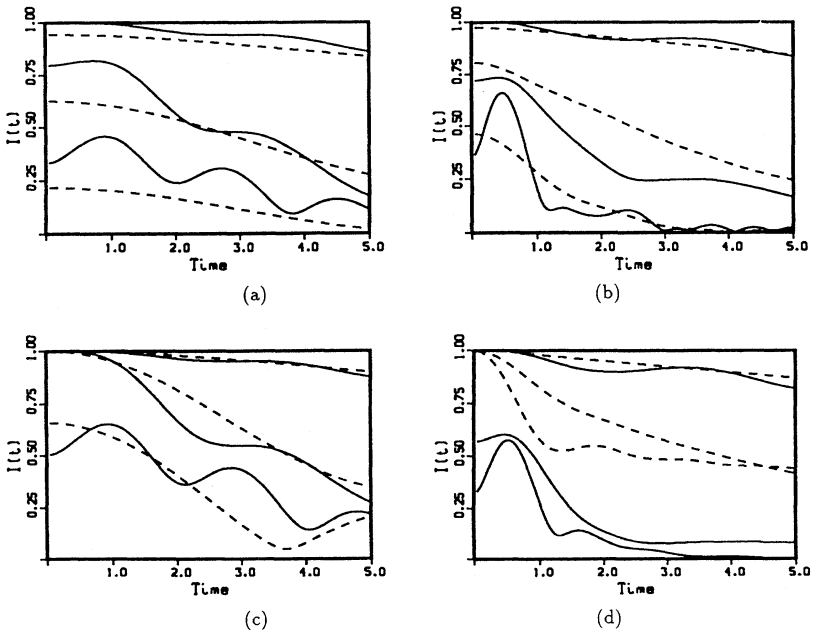

FIG. 4. Time dependence of the relaxation term $I(t)$. (a) Constant coupling, $\omega_1 = 0.5\omega_0$, $\omega_2 = 2.0\omega_0$. (b) Constant coupling, $\omega_1 = 0.1\omega_0$, $\omega_2 = 5.0\omega_0$. (c) Parabolic coupling, $\omega_1 = 0.5\omega_0$, $\omega_2 = 2.0\omega_0$. (d) Parabolic coupling, $\omega_1 = 0.1\omega_0$, $\omega_2 = 5.0\omega_0$. The solid lines represent the solution obtain for the full Hamiltonian and the dashed lines represent the solution obtained within the RWA. The coupling coefficient was $G = 0.1, 0.3, 0.7$ for (a) and (c) and $G = 0.1, 0.3, 0.5$ for (b) and (d).

TABLE I. Interval of coupling strength (the ratio G^2/ω_0) inside which the local modes or nonoscillating terms exist. The values of the parameters correspond to the cases presented in Fig. 4.

Parameters	Pole below ω_1	Pole above ω_2
constant coupling, $\omega_1 = 0.5,\ \omega_2 = 2.0$	$0 - 0.42$	$0 - \infty$
constant coupling, $\omega_1 = 0.1,\ \omega_2 = 5.0$	$0 - 0.25$	$0 - \infty$
parabolic coupling, $\omega_1 = 0.5,\ \omega_2 = 2.0$	$0.13 - 0.50$	$0.89 - \infty$
parabolic coupling, $\omega_1 = 0.1,\ \omega_2 = 5.0$	$0.28 - 0.29$	$2.15 - \infty$

Ref. [27]) state that singular points of that type exist always. For example, $G(\omega) \to 0$ and $|G'(\omega)| \to \infty$ at the boundaries of the continuum. Neither of our models satisfies these conditions. So the following questions arise. To what extent we may trust our model results? Do the features obtained have universal meaning or are they only artifacts of the concrete model?

First we consider the behavior of the profile near boundary points, that is, ω_1 and ω_2. Within the parabolic model, $G(\omega) \to 0$ as $\omega \to \omega_1, \omega_2$ and $G'(\omega)$ remains finite. Within the model of constant coupling, $G'(\omega)$ becomes infinite at these points; moreover, $G(\omega)$ is not a continuous function there. So the real profile is somewhere between these two models. We may expect that the behavior of the real system will also be between these models. As one can see by comparing Figs. 4(a) and 4(c) [and 4(b) and 4(d), respectively], there is no essential difference between them. This fact can be an indirect confirmation of the reliability of our results. Speaking about singularities inside the profile, one can expect that they will not affect much the results, as in all equations $G(\omega)$ stands only under the integral sign, and the singularity of the first derivative is too weak to influence appreciably the integral value.

APPENDIX C: IMPOSSIBILITY OF EXPONENTIAL RELAXATION

Any physical system is subject to quite general limitations on its relaxation behavior. Such limitations were considered long ago by Khalfin [28]. However, his results are applicable only to pure states whereas the relaxation in most realizable systems is described by mixed states. We will consider here the general case, including mixed states. We show that strictly exponential relaxation cannot be realized in physical systems. The principal corollary of this assertion is that any model in which such a relaxation is obtained as an *exact solution* is not physical. The result derived here can be used as a general test for verifying the physical feasibility of relaxation theories and models. Of course, this does not prevent one from getting exponential decay as an approximate description of relaxation (see, for example, [29], Chap. 8).

All the results presented here are based only on the semifiniteness of the energy distribution density $w(E)$, that is, that the energy spectrum is bounded below by some value E_1, which, without losing the generality, may be set zero. It is obvious that any real physical system possesses this property. We will show that the diagonal elements of the density matrix, that is, the probability for a system to be found in some definite state, cannot follow the exponential law of relaxation for all times. We will also investigate the decay of the mean value of quantum mechanical operators and show that they possess the same property, provided they are either nonnegative or nonpositive definite.

1. Relaxation of the density matrix

We first study the behavior of the density matrix. Let u, v be the index of the dynamic system and α the index of the thermal bath. Then the density matrix of the subsystem is

$$\rho_{uv} = \sum_{\alpha} \rho_{u\alpha;v\alpha}, \tag{C1}$$

where $\rho_{u\alpha;v\beta}$ is the density matrix of the whole system. Thus the density matrix ρ_{mn} generally describes the mixed states even if $\rho_{m\alpha;n\beta}$ corresponds to the pure states. We consider a system described by the Hamiltonian

$$H = H_0 + V, \tag{C2}$$

where H_0 is the unperturbed Hamiltonian and V is the interaction energy. The density matrix ρ in the H_0 representation can be written as (see, for example, Ref. [9])

$$\rho_{uv} = \sum_{u',v',L,M} S_{uL} S^*_{u'L} \rho_{u'v'}(0) S_{v'M} S^*_{vM} e^{-i\omega'_{LM} t}, \tag{C3}$$

where S is a unitary matrix that connects the density matrix in the H_0 representation with the density matrix in the H representation, $\hbar\omega'_{LM} = E'_L - E'_M$, E'_L and E'_M are the eigenvalues of the Hamiltonian H, and $\rho_{u'v'}(0)$ is the unperturbed density matrix.

As shown below, we need for our consideration only the diagonal elements of ρ, which represent the probability for the system to be found in a correspondent state:

$$\rho_{uu} = \sum_{u',v',L,M} S_{uL} S^*_{u'L}\, \rho_{u'v'}(0)\, S_{v'M} S^*_{uM}\, e^{-i\omega'_{LM} t}. \tag{C4}$$

Let us transform the initial density matrix $\rho_{u'v'}(0)$ to the diagonal form. It can be done by a unitary matrix T: $\tilde{\rho}(0) = T^* \rho(0) T$, where $\tilde{\rho}_{kl}(0) = \tilde{\rho}_{kk}(0)\delta_{kl}$. Then $\rho(0) = T\tilde{\rho}(0)T^*$ or $\rho_{u'v'}(0) = \sum_k T_{u'k}\tilde{\rho}_{kk}(0)T^*_{kv'}$. Substituting this into (C4), we obtain

$$\rho_{uu} = \sum_{u',v',L,M,k} S_{uL} S^*_{u'L} T_{u'k}\, \widetilde{\rho}_{kk}(0)\, S_{v'M} S^*_{uM} T^*_{kv'} \times e^{-i\omega'_{LM} t}. \tag{C5}$$

Separating the summations with respect to k; u', L; and v', M, we get

$$\rho_{uu} = \sum_k \widetilde{\rho}_{kk}(0) \left[\left(\sum_{u',L} S_{uL} S^*_{u'L} T_{u'k} e^{-i\omega'_L t} \right) \times \left(\sum_{v',M} S_{v'M} S^*_{uM} T^*_{kv'} e^{i\omega'_M t} \right) \right]. \tag{C6}$$

In the last sum we replace v' by u' and M by L and notice that the expression in the second set of large parentheses is the complex conjugate of the expression in the first set of large parentheses. Denoting the latter by $F_{uk}(t)$ we can write

$$\rho_{uu} = \sum_k \widetilde{\rho}_{kk}(0) \left| F_{uk}(t) \right|^2. \tag{C7}$$

We need to prove now that ρ_{uu}, the probability for the system to be found in the state u, tends to zero as $t \to \infty$ more slowly than the exponential function. This means that we should evaluate ρ_{uu} from below.

Since all the values of $\widetilde{\rho}_{kk}(0)$ that figure in expression (C7) are positive, we can always find an integer N such that $0 < \lambda \equiv \widetilde{\rho}_{NN}(0) \le \widetilde{\rho}_{kk}(0)$ for any k. Hence

$$\rho_{uu}(t) \ge \lambda \sum_k \left| F_{uk}(t) \right|^2. \tag{C8}$$

Let us now analyze $F_{uk}(t)$ for a fixed k. Denoting $\sum_{u'} S^*_{u'L} T_{u'k}$ by $\widetilde{S}_{Lk}$ we obtain

$$F_{uk}(t) = \sum_L S_{uL} \widetilde{S}_{Lk} e^{-i\omega'_L t}. \tag{C9}$$

Passing from the sum to the integral we can finally write

$$F_{uk}(t) = \int_0^\infty w_{uk}(\omega) e^{-i\omega t}\, d\omega, \tag{C10}$$

where $w_{uk}(\omega) = S_u(\omega) \widetilde{S}_k(\omega)$.

It follows from the theory of the Fourier series that the Fourier transform of the function with a bounded carrier cannot be an exponential function. Indeed, by the theorem of Paley and Wiener [30] and since the function $w_{uk}(\omega)$ is zero for negative ω, its Fourier transform $F_{uk}(t)$ should satisfy the inequality

$$\int_{-\infty}^{\infty} \frac{\left| \ln \left| F_{uk}(t) \right| \right|}{1 + t^2} dt < \infty. \tag{C11}$$

One of the principal corollaries of this condition is that $|F_{uk}(t)|$ cannot obey the exponential law of relaxation for all times. Indeed, if $|F_{uk}(t)|$ is an exponential function for $t \to \infty$, then integral (C11) diverges. This means that $|F_{uk}(t)|$ should vanish more slowly than the exponential function, at least as $e^{-\gamma t/(\ln t)^\alpha}$, $\alpha > 0$. Returning to (C8), we can see that $\rho_{uu}(t)$ possesses the same property. This means that pure exponential decay for a real physical system can never take place.

2. Relaxation of operators

The mean value of a quantum mechanical operator A can be written as

$$\langle A(t) \rangle = \mathrm{Tr}\left[A\rho(t) \right] = \sum_{k,n} A_{k,n} \rho_{nk}(t). \tag{C12}$$

Since A is independent of time, we can always transform it to a diagonal form. This gives us the possibility not to take into consideration off-diagonal elements of $\rho(t)$. Thus, assuming A to be diagonal, we obtain

$$\langle A(t) \rangle = \sum_k A_{kk} \rho_{kk}(t). \tag{C13}$$

As A is now in the diagonal representation, the numbers A_{kk} are its eigenvalues λ_k. Assume now that A is a non-negative definite operator. Then all its eigenvalues are non-negative and we can use the same reasoning as in the preceding section. Let λ be the minimal nonzero eigenvalue of A: $\lambda_k \ge \lambda$. Then

$$\langle A(t) \rangle \ge \lambda \sum_k \rho_{kk}(t). \tag{C14}$$

Using the above results concerning the time dependence of ρ_{kk}, we come to the conclusion that a quantum mechanical operator cannot obey the exponential law of relaxation, provided it is non-negative definite. (The same assertion obviously holds for nonpositive definite operators.)

We cannot prove the same property for operators that are neither non-negative nor nonpositive definite. Indeed, in this case not all λ_k are of the same sign and (C13) is not a series of only positive or only negative numbers. Hence it may vanish faster than each of its terms, and nothing can be said about the behavior of such operators. Although the requirement that the operators should be either non-negative or nonpositive definite is rather strong, many quantum mechanical operators satisfy it. For example, it is so for the operator of energy, population n, angular momentum L^2, and the squares of all the other operators (such as p^2, q^2, etc.).

We want to recall here that we deal with the exact solution and all the statements of this reasoning relate to this case only. Obviously, an exponential relaxation may be obtained as an approximate solution of a problem. The result obtained here can serve as a criterion for the validity of results in the relaxation theory. The situation may be compared to the usage of thermodynamical prohibitions (like the impossibility of achieving temperature equal to the absolute zero). On the other hand, our result does not say anything about quantitative deviation from the exponential behavior in any specific model.

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