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Thermalized Liouville formalism applied to the onset and development of Rabi oscillations [*]

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Abstract

To describe the oscillations of the excitation energy between a small system, such as a nucleus, and a thermal bath having an excitation energy roughly tuned to the small system, a formalism is proposed where the motion of the density matrix is described in a Liouville space, with a metric adapted to the temperature of the bath. The obtained equation of motion is characterized by a memory kernel. A second order approximation of the kernel gives the onset of the oscillations, but is not absolutely coherent. An approximate calculation of the kernel, where the phase evolution is "motional-narrowed", is proposed, giving an answer satisfying physical conditions. Its low temperature limit is evaluated.

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1 Introduction

This work has been motivated to explain some nuclear physics results, either of the bound internal conversion type (BIC), or of its reciprocal known as nuclear excitation by electronic transition (NEET) [1]. Both are seen

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when a nuclear excited state and an electronic excited state, generally of the same atom, have excitation energies near one another. In the first of these effects, BIC, a nucleus, in an isomeric excited state, transits to its ground state but the emitted photon is not sent to the outside world – and when things are favorable, in the detector– , but is absorbed by the electronic cloud of the atom, and, of course, re-emitted further. This requires a high enough electronic excitation; this occurs when the atom has been stripped in order for the energy difference of electronic levels have the right order of magnitude. A typical case of this phenomenon is ^{125}Te for which a systematic study has been performed [2,3]. On the other hand, the NEET effect has probably been observed in ^{189}Os [4], and certainly in ^{197}Au, using synchrotron radiation to excite the electrons [5]. It is suspected that this process describes the excitation of ^{235}U to its isomeric state, when in a hot plasma, and that it is unavoidable in astrophysical systems [6].

A naive theory of those phenomena would use the Fermi Golden Rule, at least three times, to describe –in the case of the BIC effect– , firstly the nuclear transition, then the absorption of the emitted quantum by the electronic cloud, then the emission by these electrons of the photon to be detected in the experiments. But this is certainly wrong, because nothing has been detected in the intermediary processes, so the quantum coherencies are not destroyed and one must work with an amplitude as long as the excitation does not arrive at a detector. A related problem also is considered, the emission of a nucleus trapped in a small enough box [7].

In fact these phenomena are the beginning of a well known one, considered generally in the other limit, the quantum Rabi oscillations between two states, a phenomenon often found in coherent spectroscopy and in quantum electronics; the Jaynes-Cummings model is mostly used to describe it [8]. It has been used to describe many tests of quantum mechanical principles [9,10], including Schrödinger's cat experiments [11].

The method to be described here uses the Liouville space formalism, used very often in the theory of coherent spectroscopy, from nuclear magnetic resonance to nuclear angular correlations, as well as in the theory of irrreversible phenomena [12]. The first improvement, if it is one, is the introduction of thermal effects through an adapted metric in that space. This has been used to describe the onset of Rabi oscillations [13], the part of the phenomenon relevant to nuclear physics. It has been tempting to try to go further and to test the formalism in the very oscillating limit. This is not very simple [14], if one wishes to be rigorous, and only a rapid sketch of the method has been published [15]. But it is also possible to go more simply from the onset to the fully developed oscillations limit. The method is approximative, which is not a problem, but the approximations are not controlled, even if they look reasonable.

In the second part, the thermalized Liouville formalism is introduced, and in the third part the evolution equation of the density matrix in a small system is established; a simplified version of it is also given which allows us to keep only the diagonal elements, that is the populations, if at the initial time there is neither entanglement nor coherencies between partially occupied states. The fourth part describes an approximation to the second order in the coefficient coupling the small system and the thermalised reservoir. It shows that the origin of Rabi oscillations is not given by a condition on the strength of the coupling, like in the toy system analysis of Cohen-Tannoudji *et al.* [16], but grows gradually from the start, being more like a second order transition than a first order one; but, although giving hints for a better theory, the second order approximation gives unphysical results, and this fact has motivated further studies. Finally, in the fifth part, an approximate solution, valid and giving physically reasonable results is given, with calculations done only in the very low temperature limit, in order to retain simplicity.

2 A small system and a thermalized reservoir

Let us present the total Hamiltonian in the form

$$h = h_N^0 + h_R + h_{RN}^0 \tag{1}$$

where h_N^0 is the Hamiltonian of the small system, in the original problem of a nucleus, all of whose matrix elements are assumed to be known, h_R the Hamiltonian of the reservoir, and h_{RN}^0 is their coupling. We shall use a thermal mean field, that is

$$h_N = h_N^0 + Tr_R\left\{D_R^0 h_{RN}^0\right\} = h_N^0 + \langle h_{RN}^0\rangle. \tag{2}$$

D_R^0 being the thermal density operator of the environment, consisting normally of the electronic cloud and of the photon vacuum. With the associated modification

$$h_{RN} = h_{RN}^0 - \langle h_{RN}^0\rangle \tag{3}$$

one gets

$$\langle h_{RN}\rangle = Tr_R\left\{D_R^0 h_{RN}\right\} = 0. \tag{4}$$

We shall denote the states of the reservoir by μ, ν, while a, b, α, β will be used for the states of the small system, so that

$$D_R^0 = \sum_\mu |\mu\rangle p_\mu \langle\mu| \tag{5}$$

with

$$p_\mu = \frac{1}{Z_R} \exp\left(-\frac{E_\mu}{kT}\right). \tag{6}$$

The density matrix of the whole system will be denoted as $D(t)$, verifying the motion equation

$$i\frac{\mathrm{d}}{\mathrm{d}t} D(t) = \frac{1}{\hbar}[h, D(t)] = LD(t) \tag{7}$$

that is

$$D(t) = e^{-iLt} D(0) \tag{8}$$

in the Liouville formalism. The principles of this formalism where operators are taken as vectors of a new space, are included in an appendix for completeness.

We shall also denote by σ the density matrix of the small system, and take an initial condition without any entanglement, that is

$$D(0) = \sigma(0) \otimes D_R^0. \tag{9}$$

So far our approach has been standard but we now shall use, in the Liouville space, a kind of metric adapted to the thermal state of the reservoir, and chiefly the associated projectors. One could define

$$(\mu a, \nu b|g|\mu' a', \nu' b') = p_\mu \delta_{\mu\mu'} \delta_{\nu\nu'} \delta_{aa'} \delta_{bb'} \tag{10}$$

giving the interior product of two vectors of this space, that is between two operators of the Hilbert space, as

$$(A \mid B) = Tr\left\{A^+ D_R^0 B\right\}. \tag{11}$$

With a non-Euclidian metric, one has two kinds of projectors, as it is possible to project either on vectors or on forms. Let us define

$$\Pi_{ab} = \sum_{\mu\mu'} |\mu a, \mu b)(\mu' a, \mu' b| p_{\mu'} \tag{12}$$

and, contravariantly

$$\widetilde{\Pi}_{ab} = \sum_{\mu\mu'} p_\mu \left|\mu a, \mu b)(\mu' a, \mu' b\right| . \tag{13}$$

It is now straightforward to check that those are true projectors, as

$$\Pi_{ab}\Pi_{cd} = \Pi_{ab}\delta_{ac}\delta_{bd} \tag{14}$$

and

$$\widetilde{\Pi}_{ab}\widetilde{\Pi}_{cd} = \widetilde{\Pi}_{ab}\delta_{ac}\delta_{bd} \tag{15}$$

are easily verified. Furthermore, denoting by L_N, L_R and L_{RN} the Liouvillian superoperators associated with h_N, h_R and h_{RN}, one has the following relations

$$(L_R + L_N)\widetilde{\Pi}_{ab} = \omega_{ab}\widetilde{\Pi}_{ab} \tag{16}$$

$$\widetilde{\Pi}_{ab}(L_R + L_N) = \omega_{ab}\widetilde{\Pi}_{ab} \tag{17}$$

and

$$\Pi_{ab}L_{RN} \mid \nu a, \nu b) = 0 \tag{18}$$

as a simple consequence of formulae (A2) and (A4).

Then, one defines

$$\Pi_D = \sum_{ab} \Pi_{ab} \tag{19}$$

and, of course,

$$\widetilde{\Pi}_D = \sum_{ab} \widetilde{\Pi}_{ab} \tag{20}$$

If Π or $\widetilde{\Pi}$ projector is diagonal in the small system Hamiltonian, one has, for instance,

$$(\nu a, \nu a \mid L\widetilde{\Pi}_{aa} = 0 \tag{21}$$

by a direct application of formulae (18) and (21) and as a direct consequence

$$\sum_\nu (\nu a, \nu a \mid L(1 - \widetilde{\Pi}_{aa}) = \sum_\nu (\nu a, \nu a \mid L_{RN} \tag{22}$$

which will be needed further.

3 The equations of motion

Applying the above notations and the Liouville formalism, to a non-entangled system, thermalized reservoir hypothesis (9), one can write at any positive time the matrix operator of the small system as

$$\sigma_{ab}(t) = \theta(t)\sum_{\mu\nu}\sum_{\alpha\beta}\left(\nu a, \nu b\left|e^{-iLt}p_\mu\right|\mu\alpha, \mu\beta\right)\sigma_{\alpha\beta}(0) \tag{23}$$

where $\theta(t)$ is the Heaviside function, and take its derivative

$$\frac{\mathrm{d}}{\mathrm{d}t}\sigma_{ab}(t) = \delta(t)\sigma_{ab}(t) - i\theta(t)\sum_{\mu\nu}\sum_{\alpha\beta}\left(\nu a, \nu b\left|Le^{-iLt}p_\mu\right|\mu\alpha, \mu\beta\right)\sigma_{\alpha\beta}(0). \tag{24}$$

To go further we use the formula

$$e^{-iLt} = e^{-iL\left(1-\widetilde{\Pi}_D\right)t} - i\int_0^t ds\, e^{-iL\left(1-\widetilde{\Pi}_D\right)s}L\widetilde{\Pi}_D e^{-iL(t-s)} \tag{25}$$

which is easily verified by the derivation over t of both terms and the fact that for $t = 0$, both terms are equal to 1.

Then one gets

$$\begin{aligned}
\tfrac{d}{dt}\sigma_{ab}(t) &= \delta(t)\sigma_{ab}(t) - i\theta(t)\sum_{\mu\nu}\sum_{\alpha\beta}\left(\nu a, \nu b\left|L\widetilde{\Pi}_{ab}e^{-iLt}p_\mu\right|\mu\alpha, \mu\beta\right)\sigma_{\alpha\beta}(0)\\
&-i\theta(t)\sum_{\mu\nu}\sum_{\alpha\beta}\left(\nu a, \nu b\left|L(1-\widetilde{\Pi}_{ab})e^{-iL(1-\widetilde{\Pi}_D)t}p_\mu\right|\mu\alpha, \mu\beta\right)\sigma_{\alpha\beta}(0)\\
&-i\theta(t)\sum_{\mu\nu}\sum_{\alpha\beta}(\nu a, \nu b\left|L(1-\widetilde{\Pi}_{ab})\int_0^t dse^{-iL(1-\widetilde{\Pi}_D)s}(-iL)\widetilde{\Pi}_D e^{-iL(t-s)}p_\mu\right|\\
&\qquad\qquad\mu\alpha, \mu\beta)\sigma_{\alpha\beta}(0)
\end{aligned} \tag{26}$$

Straightforward but a litte cumbersome arguments show that the second term of the right-hand side is equal to $-i\omega_{ab}\sigma_{ab}$ while the third one vanishes. As to the integral of the fourth term, the expansion of $\widetilde{\Pi}_D$ results in the shape (23) of $\sigma(t)$, and gives, without any approximation,

$$\frac{d}{dt}\sigma_{ab}(t) = \delta(t)\sigma_{ab}(t) - i\omega_{ab}\sigma_{ab} - i\theta(t)\sum_{\alpha\beta}\int_0^t dsK_{ab}^{\alpha\beta}(s)\,\sigma_{\alpha\beta}(t-s) \tag{27}$$

where

$$K_{ab}^{\alpha\beta}(t) = -i\theta(t)\sum_{\mu\nu}\left(\nu a, \nu b\left|L_{RN}e^{-iL\left(1-\widetilde{\Pi}_D\right)t}L_{RN}p_\mu\right|\mu\alpha, \mu\beta\right). \tag{28}$$

These equations will be transformed by a Laplace or one-sided Fourier transform in a solvable algebraic system, with initial values of $\sigma(0)$ as its right hand side. If this density matrix has no off-diagonal elements, the $\sigma_{ab}(z)$ can be eliminated, but there is still a faster way to do that. We remark that formula (26) was derived without any hypothesis about $\widetilde{\Pi}_D$, so we replace it by $\widetilde{\Pi}_{Dd}$ which is this sum of projectors reduced to the diagonal ones

$$\widetilde{\Pi}_{Dd} = \sum_a \widetilde{\Pi}_{aa}. \tag{29}$$

If we write the above evolution equation for the diagonal terms σ_{aa} using $\widetilde{\Pi}_{Dd}$, the second term of the right hand side will be suppressed, as ω_{aa} is obviously 0. The fourth term involves a restriction of K to diagonal elements. But the third term is not automatically vanishing, if the off-diagonal initial terms are not 0; this is the way an initial coherence influences the story of the matrix elements, showing that if it exists, the full equations (27) and (28) will be needed. To distinguish the new kernel from the old one, we shall denote it by only two indices $K_a^\alpha(t)$.

The new equation is thus now

$$\frac{d}{dt}\sigma_{aa}(t) = \delta(t)\sigma_{aa}(t) - i\theta(t)\sum_b \int_0^t ds K_a^b(s)\,\sigma_{bb}(t-s) \tag{30}$$

where

$$K_a^b(t) = -i\theta(t)\sum_{\mu\nu}\left(\nu a, \nu a \left| L_{RN} e^{-iL\left(1-\widetilde{\Pi}_{Dd}\right)t} L_{RN} p_\mu \right| \mu b, \mu b\right). \tag{31}$$

And now, as has been already said above, we shall transform the evolution equations into algebraic ones, by the transforms

$$\widetilde{\sigma}_{ab}(z) = \int_0^\infty dt\, e^{izt}\sigma_{ab}(t) = \int_{-\infty}^\infty dt\, e^{izt}\sigma_{ab}(t) \tag{32}$$

$$\widetilde{K}_b^a(z) = \int_0^\infty dt\, e^{izt} K_b^a(t) = \int_{-\infty}^\infty dt\, e^{izt} K_b^a(t). \tag{33}$$

The transformed evolution equation to be used further is thus the restricted one

$$z\widetilde{\sigma}_{aa}(z) - \sum_b K_a^b(z)\,\widetilde{\sigma}_{bb}(z) = i\sigma_{aa}(0) \tag{34}$$

with K_a^b defined by

$$\widetilde{K}_a^b(z) = \sum_{\mu\nu} \left(\nu a, \nu a \left| L_{RN} \frac{1}{z - L\left(1 - \widetilde{\Pi}_{Dd}\right)} L_{RN} p_\mu \right| \mu b, \mu b \right) \tag{35}$$

which is the Fourier transform of

$$K_a^b(t) = -i\theta(t) \sum_{\mu\nu} \left(\nu a, \nu a \left| L_{RN} e^{-iL\left(1-\widetilde{\Pi}_{Dd}\right)t} L_{RN} p_\mu \right| \mu b, \mu b \right). \tag{36}$$

4 The second order approximation

The simplest attempt to get an approximate solution of the motion equations is to compute the kernel K to the second order in h_{RN} or, equivalently, in L_{RN}. It will not allow us to go far in the description of Rabi oscillations, as one can presume that any transfer of the excitation from the small system to the reservoir or back will imply a new h_{RN} or L_{RN} factor. Nevertheless, one see that it is useful, showing the onset of those oscilations, and giving hints for further motion. We shall start with the full expression of $K(t)$, where the L_{RN} term has been omitted in the exponent expanded in series. Thus

$$\begin{aligned} K_{ab2}^{\alpha\beta}(t) &= -i\theta(t) \sum_{\mu\nu} \left(\nu a, \nu b \left| L_{RN} \sum_{n=0}^{\infty} \frac{(-i)^n}{n!} (L_R + L_N)^n \right. \right. \\ &\qquad \left. \left. \left(1 - \widetilde{\Pi}_D\right) t^n L_{RN} p_\mu \right| \mu\alpha, \mu\beta \right) \end{aligned} \tag{37}$$

where one has taken into account the facts that the projectors $\widetilde{\Pi}_{ab}$ commute with $(L_R + L_N)$, and that $\left(1 - \widetilde{\Pi}_D\right)$ being a projector, is equal to its nth power. By using then the expansion (A.2) of the Liouville superoperators, one thus gets

$$K_{ab2}^{\alpha\beta}(t) = -i\theta(t) \sum_{\mu\nu} \left(\nu a, \nu b \left| L_{RN} e^{-i(L_R+L_N)t} L_{RN} p_\mu \right| \mu\alpha, \mu\beta \right) \tag{38}$$

and

$$\widetilde{K}_{ab2}^{\alpha\beta}(z) = \sum \mu\nu \left(\nu a, \nu b \left| L_{RN} \frac{1}{z - (L_R + L_N)} L_{RN} p_\mu \right| \mu\alpha, \mu\beta \right). \tag{39}$$

Similar formulae can be given for K^b_a, in its time or frequency z shape, and, due to vanishing of the Π projectors, at this order of approximation K^{bb}_{aa2} and K^b_{a2} are identical. We can then use formula (A2) to transform (38) into

$$K^{\alpha\beta}_{ab2}(t) = -\frac{i}{\hbar^2}\theta\left(t\right)e^{-i\omega_{ab}t}\sum_{\mu\nu}\left\langle \nu a\left|\left[h_{RN}\left(t\right),\left[h_{RN},|\mu\alpha\,\rangle p_\mu\langle\,\mu\beta|\right]\right]\right|\nu b\right\rangle . \tag{40}$$

Using this expansion, it is straightforward to check that

$$K^a_{a2}(t) = -\sum_{b\neq a}K^a_{b2}(t) \tag{41}$$

a relation which must be valid in any consistent approximation for K as well as for K_2. To go further, we consider the most general coupling between the small system and the reservoir, writing

$$h_{RN} = \int d\eta N\left(\eta\right)R\left(\eta\right) \tag{42}$$

where $N\left(\eta\right)$ are operators in the small system Hilbert space and $R\left(\eta\right)$ are those in the environment Hilbert space. Introducing this expansion in (38), one gets

$$\begin{aligned}\underset{b\,\neq\,a}{K^a_{b2}(t)} \quad &= \quad \frac{i}{\hbar^2}\theta(t)\int\int d\eta d\eta'\left\langle a\left|N\left(\eta\right)\right|b\right\rangle\left\langle b\left|N\left(\eta'\right)\right|a\right\rangle \\ &\sum_{\mu\nu}p_\mu\left[\begin{array}{c}\left\langle\nu\left|R\left(\eta,t\right)\right|\mu\right\rangle\left\langle\mu\left|R\left(\eta'\right)\right|\nu\right\rangle e^{-i\omega_{ba}t} \\ +\left\langle\nu\left|R\left(\eta\right)\right|\mu\right\rangle\left\langle\mu\left|R\left(\eta',t\right)\right|\nu\right\rangle e^{-i\omega_{ab}t}\end{array}\right]\end{aligned} \tag{43}$$

where $\left\langle\nu\left|R\left(\eta,t\right)\right|\mu\right\rangle$ denotes $\left\langle v\left|R\left(\eta\right)\right|\mu\right\rangle e^{-i\omega_{\mu\nu}t}$. We shall now introduce the quantum correlation function of the reservoir

$$\Phi\left(\eta,\eta',t\right) = \frac{1}{\hbar}Tr\left\{D^0_R R\left(\eta,t\right)R\left(\eta'\right)\right\} \tag{44}$$

and thus get

$$\begin{aligned}\underset{b\,\neq\,a}{K^a_{b2}(t)} \quad &= \quad \frac{i}{\hbar}\theta(t)\int\int d\eta d\eta'\left\langle a\left|N\left(\eta\right)\right|b\right\rangle\left\langle b\left|N\left(\eta'\right)\right|a\right\rangle \\ &\left[\Phi\left(\eta,\eta',t\right)e^{-i\omega_{ba}t}+\Phi\left(\eta,\eta',-t\right)e^{-i\omega_{ab}t}\right].\end{aligned} \tag{45}$$

We shall use mainly the fluctuation spectrum, that is the Fourier transform of the correlation function,

$$\begin{aligned}\widetilde{\Phi}\left(\eta,\eta',\omega\right) &= \int dte^{i\omega t}\Phi\left(\eta,\eta',t\right)\\ &= 2\pi\sum_{\mu\nu}p_\nu\left\langle\nu\left|R\left(\eta\right)\right|\mu\right\rangle\left\langle\mu\left|R\left(\eta'\right)\right|\nu\right\rangle\delta\left(\hbar\omega-E_\mu+E_\nu\right)\end{aligned}\tag{46}$$

This fluctuation spectrum is related to the imaginary part of the susceptibility of the reservoir

$$\chi''\left(\eta,\eta',t\right)=\frac{1}{2\hbar}Tr\left\{D_R^0\left[R\left(\eta,t\right),R\left(\eta'\right)\right]\right\}\tag{47}$$

by the usual relation

$$\widetilde{\Phi}\left(\eta,\eta',\omega\right)=\frac{1}{1-e^{-\frac{\hbar\omega}{kT}}}\chi''\left(\eta,\eta',\omega\right)\tag{48}$$

which is the adapted shape of the fluctuation dissipation theorem [17]; equally, one has the relation

$$\widetilde{\Phi}\left(\eta,\eta',-\omega\right)=e^{-\frac{\hbar\omega}{kT}}\widetilde{\Phi}\left(\eta,\eta',\omega\right)\tag{49}$$

given by Ayant [18], and by Kubo and Tomita [19], which insures thermodynamical equilibrium.

Using the correlation spectrum of the reservoir the expression (45) for $\widetilde{K}_{a2}^{b}$, one gets, for distinct a and b,

$$\begin{aligned}\widetilde{K}_{a2}^{b}\left(z\right) &= \frac{1}{2\pi\hbar}\int\int d\eta d\eta'\left\langle a\left|N\left(\eta\right)\right|b\right\rangle\left\langle b\left|N\left(\eta'\right)\right|a\right\rangle\\ &\qquad\int d\omega\widetilde{\Phi}\left(\eta,\eta',\omega\right)\frac{z+i\epsilon}{\left(\omega-\omega_{ab}\right)^2-\left(z+i\varepsilon\right)^2}\end{aligned}\tag{50}$$

and, from that expression, using (49), one can see that, if $b\neq a$,

$$\widetilde{K}_{a2}^{b}(0)=e^{-(\hbar\omega_{ab}/kT)}\widetilde{K}_{b2}^{a}(0).\tag{51}$$

We restrict our investigation to a two-levels system in the high temperature regime, when the energy difference between the excitation energies $E_e-E_g=\hbar\omega_N$ is much larger than kT. If we start from $\sigma_{ee}(0)=1$ all the other matrix elements of σ vanishing, the evolution equations, reduced to those of the diagonal elements of the density matrix, are

$$z\widetilde{\sigma}_{ee}\left(z\right)-\widetilde{K}_{e}^{e}\left(z\right)\widetilde{\sigma}_{ee}\left(z\right)-\widetilde{K}_{e}^{g}\left(z\right)\widetilde{\sigma}_{gg}\left(z\right)=i \tag{52}$$

$$z\widetilde{\sigma}_{gg}\left(z\right)-\widetilde{K}_{g}^{e}\left(z\right)\widetilde{\sigma}_{ee}\left(z\right)-\widetilde{K}_{g}^{g}\left(z\right)\widetilde{\sigma}_{gg}\left(z\right)=0. \tag{53}$$

In those equations, only two K coefficients are independent, as seen from (41) within the second order approximation, but anyway physically obvious. Eliminating σ_{gg}, yields

$$\widetilde{\sigma}_{ee}\left(z\right)=\frac{i}{\widetilde{K}_{e}^{e}\left(z\right)+\widetilde{K}_{g}^{g}\left(z\right)}\left[\frac{\widetilde{K}_{e}^{e}\left(z\right)}{z-\widetilde{K}_{e}^{e}\left(z\right)+\widetilde{K}_{g}^{g}\left(z\right)}+\frac{\widetilde{K}_{g}^{g}\left(z\right)}{z}\right]. \tag{54}$$

The pertinent range of z used to describe the Rabi oscillations is many orders of magnitude less than ω_{eg}, the energy difference of the two states, divided by $\hbar$. So one can, using (41) and (51), neglect K_{g}^{g}, which gives the approximate formula

$$\widetilde{\sigma}_{ee}\left(z\right)=\frac{i}{z-\widetilde{K}_{e}^{e}\left(z\right)} \tag{55}$$

to be used from now onwards.

The fluctuation spectrum $\widetilde{\Phi}$ of the reservoir may have a slow variation but in the presence of the Rabi oscillations, there should be states of the reservoir with an energy difference of the order of $\hbar\omega_N$ and creating a resonance at a frequency which will be called ω_e, introducing a factor

$$\left(1+\left(\frac{\omega-\omega_e}{\zeta}\right)^2\right)^{-1} \tag{56}$$

and an element of integration $\zeta^{-1}d\omega$. We will take this resonance into account more precisely by neglecting the remaining part of the spectrum and writing

$$\frac{1}{\hbar}\int\int d\eta d\eta'\left\langle a\left|N\left(\eta\right)\right|b\right\rangle\left\langle b\left|N\left(\eta'\right)\right|a\right\rangle\widetilde{\Phi}\left(\eta,\eta',\omega\right)=\Gamma\frac{\zeta}{\left(\omega-\omega_e\right)^2+\zeta^2} \tag{57}$$

and compute the value of $\widetilde{K}_{e2}^{e}\left(z\right)$ using (50) and (57). So

$$\widetilde{K}_{e2}^{e}\left(z\right)=-\widetilde{K}_{g2}^{e}\left(z\right)=-\frac{\Gamma}{\pi}\int_{-\infty}^{\infty}d\omega\frac{\zeta}{\left(\omega-\omega_e\right)^2+\zeta^2}\frac{z+i\epsilon}{\left(\omega-\omega_{ab}\right)^2-\left(z+i\varepsilon\right)^2} \tag{58}$$

which is evaluated by closing the contour in the upper half plane. One thus gets

$$\widetilde{K}_{e2}^{e}(z)=\Gamma\frac{z+i\zeta}{(z+i\zeta)^2+(\omega_e-\omega_N)^2} \tag{59}$$

or, with the notation

$$\omega_{e,N}^2=(\omega_e-\omega_N)^2+\zeta^2 \tag{60}$$

$$\widetilde{K}_{e2}^{e}(z)=\Gamma\frac{z+i\zeta}{z^2+2i\zeta z-\omega_{e,N}^2}. \tag{61}$$

One has now for the population of the excited state the Fourier transform of

$$\widetilde{\sigma}_{ee}(z)=i\frac{z^2+2i\zeta z-\omega_{e,N}^2}{z^3+2i\zeta z^2-\left(\Gamma+\omega_{e,N}^2\right)z-i\Gamma\zeta}. \tag{62}$$

The denominator of this expression is a third order polynomial and hence has three poles and three frequencies which appear in the time variation of the population of the excited state. To find them, we shall solve the cubic equation in X, defined by

$$z=iX. \tag{63}$$

It is

$$X^3+2\zeta X^2+\left(\Gamma+\omega_{eN}^2\right)X+\zeta\Gamma=0 \tag{64}$$

whose coefficients are all positive, so there is one real root and two complex conjugate ones. If one notes that ζ is a small parameter, smaller than ω_{eN} by definition, which is smaller than $\sqrt{\Gamma}$ in the range of values where the Rabi oscillations are efficient, the roots are easily evaluated. At $\zeta=0$ the roots are

$$X_1=0 \quad X_2=i\sqrt{\Gamma+\omega_{eN}^2} \quad X_3=-i\sqrt{\Gamma+\omega_{eN}^2} \tag{65}$$

and at first order in ζ

$$X_1=-\frac{\Gamma}{\Gamma+\omega_{eN}^2}\zeta \tag{66}$$

$$X_2=-\frac{\frac{1}{2}\Gamma+\omega_{eN}^2}{\Gamma+\omega_{eN}^2}\zeta+i\sqrt{\Gamma+\omega_{eN}^2} \tag{67}$$

$$X_3=-\frac{\frac{1}{2}\Gamma+\omega_{eN}^2}{\Gamma+\omega_{eN}^2}\zeta-i\sqrt{\Gamma+\omega_{eN}^2}. \tag{68}$$

Then we can, after some algebra, come back to the time expansion of σ_{ee}, that is

$$\sigma_{ee}(t) = \frac{\omega_{eN}^2}{\Gamma + \omega_{eN}^2} e^{-\frac{\Gamma}{\Gamma+\omega_{eN}^2}\zeta t} + \frac{\Gamma}{\Gamma + \omega_{eN}^2} e^{-\frac{\frac{1}{2}\Gamma+\omega_{eN}^2}{\Gamma+\omega_{eN}^2}\zeta t} \cos\left(\sqrt{\Gamma + \omega_{eN}^2} t - \varphi\right) \tag{69}$$

where φ is an irrelevant phase coefficient.

The above expression for the population of the excited state shows that the tendency to have Rabi oscillations is starting at the lower level, that it behaves continuously with the coupling not as a first order transition in the toy model of Cohen-Tannoudji. But it is clear that the above expression has a major defect. If $\omega_{eN}^2 \geq \Gamma$, the coupling between the states of the small system and of the reservoir is smaller than the detuning or than the linewidth of the resonance, or than both of them. But in the opposite case of a strong coupling, if $\omega_{eN}^2 \leq \Gamma$, the cosine oscillations will cause the population of the excited state e to oscillate with a negative inferior limit, the decay times of the exponential not having had the time to have an effect.

5 Beyond the second order approximation

The above approach has a non-surprising difficulty: as it has been already said, many L_{RN} must be accounted for in the calculation if one wants to describe many oscilations. Describing the origin of the problem in another way, we note that the denominator the expression (39) of $\widetilde{K_2}(z)$ is $z - (L_R + L_N)$, and if the detuning is vanishing, and the width of the Lorenztian function is very small, there is a divergency in the second order approximation. Thus we have to get rid of this non-physical divergency.

In spite of this difficulty, two methods can be applied to solve the problem. The first one has been proposed by J.J. Niez [15]. Its principle is to take out of the set of states of the reservoir those two which transition between each other creates the resonance described above by a Lorentzian shape. Even if the electronic system includes a harmonic oscillator with many pairs of states whose energy difference is comparable with $\hbar\omega_N$, the matrix elements between those pairs are proportional to $\sqrt{n+1}$, so that they give different Rabi frequencies and only one pair is significant, if n is small enough; this has been used to prove the quantification of the electromagnetic field [10].

The calculation made by J.J. Niez is not as simple as it seems: taking two states out of the reservoir to introduce them in the nucleus Hamiltonian, will leave one with a reservoir capable mainly of damping and all the interesting dynamics will now be in the small system; then, as usual the new reservoir-system coupling will be treated at the second order, and even a Markovian approximation could be valuable. Furthermore, only the supervectors of the Liouville space coupling the upper electronic state with the lower nuclear one, or reciprocally, are to be considered; this is equivalent to the rotating frame approximation.

Nevertheless, there are some technical difficulties in this program, and we propose now an approximate solution, simple enough at least in the very low temperature regime. It could be extended to a higher temperatures but we shall not do it here, as this would make the calculations more complicated, but it will be clear that such an extension is not too difficult to perform.

If one wants to compute $\widetilde{K_e^e}(z)$without being restricted to the second order in h_{RN} or L_{RN} , one must evaluate $\exp\left(-iL\left(1-\widetilde{\Pi}_{Dd}\right)t\right)$to introduce it in (36) . This could be evaluated by a method analogous to a path -integral calculation, by writing

$$\exp\left(-iL\left(1-\widetilde{\Pi}_{Dd}\right)t\right)=\left(1-iL\left(1-\widetilde{\Pi}_{Dd}\right)\frac{t}{n}\right)^{n} \tag{70}$$

with n becoming infinite. At every time point, four parameters of the small system are to be known, the amplitudes of the four components of σ. To each of them a distribution between the states μ of the reservoir, and the corresponding amplitudes will describe the instantaneous wave function, and the values of the matrix elements of the whole density matrix D. The total amplitude attached to any of the four components $(a\left|\sigma\right|b)$ is the sum of the components attached to $(\mu a\left|D\right|\nu b)$ for all μ and ν, with phase factors integral along the path of the "local" values of L_R+L_N, the "local" values of L_{RN} giving the rate of transfer between the components.

In fact, the implied Liouville superoperator is not L but

$$L'=L\left(1-\widetilde{\Pi}_{Dd}\right) \tag{71}$$

whose matrix elements are given by

$$\begin{aligned}\left(\nu\alpha,\nu'\alpha'\left|L\left(1-\widetilde{\Pi}_{Dd}\right)\right|\mu\beta,\mu'\beta'\right)&=\left(\omega_{\alpha\alpha'}+\omega_{\nu\nu'}\right)\delta_{\mu\nu}\delta_{\mu'\nu'}\delta_{\alpha\beta}\delta_{\alpha'\beta'}\\&+\frac{1}{\hbar}\left[\left\langle\nu\alpha\left|h_{RN}\right|\mu\beta\right\rangle\delta_{\mu'\nu'}\delta_{\alpha'\beta'}-\left\langle\mu'\beta'\left|h_{RN}\right|\nu'\alpha'\right\rangle\delta_{\mu\nu}\delta_{\alpha\beta}\right]\\&-\frac{1}{\hbar}\left(p_{\nu'}\delta_{\beta\alpha'}-p_{\nu}\delta_{\beta'\alpha}\right)\left\langle\nu\alpha\left|h_{RN}\right|\nu'\alpha'\right\rangle\delta_{\mu\mu'}\delta_{\beta\beta'}\end{aligned}$$

For instance, one has

$$\begin{aligned}(\nu e, \nu' e\,|L'|\,\mu e, \mu' e) = \omega_{\mu\mu'}\delta_{\mu\nu}\delta_{\mu'\nu'} + \tfrac{1}{\hbar}\left[\langle \nu e\,|h_{RN}|\,\mu e\rangle\,\delta_{\mu'\nu'}\right. \\ \left. - \langle \mu' e\,|h_{RN}|\,\nu' e\rangle\,\delta_{\mu\nu}\right] + \tfrac{1}{\hbar}\left(p_\nu - p_{\nu'}\right)\langle \nu e\,|h_{RN}|\,\nu' e\rangle\,\delta_{\mu\mu'}\end{aligned} \tag{72}$$

$$\left(\nu e, \nu' e\,\left|L'\right|\,\mu e, \mu' g\right) = -\frac{1}{\hbar}\left\langle \mu' g\,|h_{RN}|\,\nu' e\right\rangle\delta_{\mu\nu} \tag{73}$$

or

$$\left(\nu e, \nu' g\,\left|L'\right|\,\mu e, \mu' e\right) = -\frac{1}{\hbar}\left\langle \mu' e\,|h_{RN}|\,\nu' g\right\rangle\delta_{\mu\nu} + \frac{1}{\hbar}p_\nu\left\langle \nu e\,|h_{RN}|\,\nu' g\right\rangle\delta_{\mu\mu'} \tag{74}$$

These are typical expressions. We are interested in their mean values when summing over the reservoir indices. For (72), $\omega_{\mu\mu'}$ will oscillate around zero, the pertinent energies of the reservoir when associated with the nuclear excited state being distributed at random.The matrix elements of h_{RN} are diagonal in the nuclear Hilbert space; if they are also diagonal in the reservoir Hilbert space, they will vanish according to (4) but even when they are non diagonal, their mean values probably vanish and we shall neglect them; so the mean value of (72) will be taken as zero.

In the case of (73), we have a matrix element coupling the transition of the small system to the transition of the reservoir, or, generally, states coupled by the Lorentz distribution of (57), and we shall denote the mean value of those as γ, whose square will be Γ of (57). Here the mean value of (73) will thus be $-\gamma$.

The case of (74) is more tricky. The states of the reservoir coupled to the small system excited state will normally be occupied, especially in the low temperature limit, while those coupled to the nuclear ground state will be empty. Here ν is associated with e, so p_ν is close to one and the second term will thus compensate for the first one. This is due to the presence of the projector $\widetilde{\Pi}_{Dd}$ in the expression of L'; this will happen each time the right nuclear indices of the supermatrix element will be ee, and also for the case where the left indices will be ge, instead of eg. For gg, it is zero anywhere, only one transition being implied by each supermatrix element.

If we imagine the path integral alluded to above as the propagation of values of the four matrix elements of σ, the oscillations of their mean values due to the reservoir indices will be taken as giving, on average, vanishing result and thus neglected. This is a kind of "motional narrowing approximation" of the propagation and we shall make it, considering only the mean value of L', to calculate K_e^e. If we had forgotten the thermalized projector

$\widetilde{\Pi}_{Dd}$, we would have used the full matrix of the mean value of L, that is

$$L = \begin{pmatrix} 0 & -\gamma & \gamma & 0 \\ -\gamma & \delta\omega & 0 & \gamma \\ \gamma & 0 & -\delta\omega & -\gamma \\ 0 & \gamma & -\gamma & 0 \end{pmatrix} \tag{75}$$

where the labels of rows and columns are in the order ee, eg, ge and gg. By taking the thermalized projector into account, the matrix is reduced to

$$L' \simeq \begin{pmatrix} \delta\omega & 0 & \gamma \\ 0 & -\delta\omega & -\gamma \\ \gamma & -\gamma & 0 \end{pmatrix} \tag{76}$$

where $\hbar\delta\omega$ is the difference between the nuclear and the electronic excitation energies and $\delta\omega$ is the detuning pulsation

$$\delta\omega = \omega_N - \omega_e \tag{77}$$

while the ee element is vanishing. Nevertheless, in the second order approximation, the width of the Lorentzian distribution of reservoir energy differences coupled to the small system transition, is seen to be taken into account by the replacement of z by $z+i\zeta$ in the expression of $\widetilde{K}$. In any case, this results from integrating the $(z-L')^{-1}$ factor in the (35) expansion, as in (59). We shall adopt a strategy to compute not $K(t)$, but $\widetilde{K}(z)$, by a direct application of (35) reduced to the only nuclear Liouville space. As L' has been shown to be thus a third order matrix, it is not a problem to invert it and to compute $(z-L')^{-1}$ in its eigenbasis, and to make a simple transformation of the axis to get the final result.

The three eigenvalues are

$$\lambda_1 = 0 \quad \lambda_2 = \sqrt{2\gamma^2 + \delta\omega^2} \quad \lambda_3 = -\sqrt{2\gamma^2 + \delta\omega^2} \tag{78}$$

and the corresponding eigenvectors are easily evaluated in the eg, ge and gg basis. It is a simple and short algebraic calculation to obtain that

$$\widetilde{K_e}^{e}(z) = 2\Gamma \frac{z + i\zeta}{z^2 + 2i\zeta z - \left(2\Gamma + \omega_{eN}^2\right)} \tag{79}$$

which is a formula with the same structure as (61), but without the possibility of the invariant term in the denominator to vanish at the resonance if its width goes to zero. Introducing this expression in (55), and proceeding as above, one gets

$$\sigma_{ee}(t) = \frac{2\Gamma + \omega_{eN}^2}{4\Gamma + \omega_{eN}^2} e^{-\frac{2\Gamma}{4\Gamma+\omega_{eN}^2}\zeta t} + \frac{2\Gamma}{4\Gamma + \omega_{eN}^2} e^{-\frac{3\Gamma+\omega_{eN}^2}{4\Gamma+\omega_{eN}^2}\zeta t} \cos\left(\sqrt{4\Gamma + \omega_{eN}^2}t - \varphi\zeta\right) \tag{80}$$

where ζ has been shown to be proportional to the phase shift.

In this expression one can see that, when $\delta\omega = 0$ at the resonance, and the width ζ vanishes, the occupation number of the nuclear excited state oscillates between one and zero, which is reasonable, as compared to its negative value in the second order approximation. On the other hand, if we had forgotten the thermalized projector $\widetilde{\Pi}_{Dd}$, we could have made the calculation equally easy since a fourth order matrix being as simple to diagonalize as a third order one. We would then have obtained a similar formula, but the coefficient of the cosine would have been

$$\frac{\Gamma}{3\Gamma + \omega_{eN}^2} \tag{81}$$

and a full development of the Rabi oscillations would have been missed.

6 Conclusions

The above calculations have shown that the thermalized Liouville formalism allows us to describe the phenomenon of the Rabi oscillations for all values of the detuning and coupling. The introduction of the effect of the reservoir by considering only the fluctuation function through the fluctuation dissipation theorem, is felt by the small system like a spring, and which is enough to induce oscillations to avoid the construction of a mechanical model of the electronic states on speaking terms with the nucleus, to keep the language of the initial problem. Nevertheless, the approximations, while reasonable, are not controlled, and further work is probably needed in that direction. Nevertheless, in the frequently used model of Caldeira and Leggett [20] , a general thermal bath is represented by a set of harmonic oscillators, which means that all the information about it is given by its spectrum, which is the $\Phi\left(\omega\right)$ used above.

A point to be noted is that at $t = 0$ the formulae for the time variation of population of the nuclear excited state start with a finite derivative, and not analytically. This is due to the use of a full Lorenztian shape of the spectrum of the reservoir. Such a shape is unphysical, it should have been truncated for high frequencies, in order to have finite moments [21, 22]: only then, could an analytical beginning of $\sigma(t)$ have been obtained.

Another defect of the method is that the reservoir is assumed to have been thermalized before the starting point of the analysis, and any further evolution due to the extracouplings of the reservoir seen by the small system with the outside world is not taken into account. This defect is a generic one in the theory of susceptibilities, but in the analysis of a phenomenon as sensitive as Rabi oscillations, it may be more important. Besides, in a method with uncontrolled approximations, it is difficult to say something pertinent on this problem. As shown, for instance, by Tolkunov and Privman [23], the short time problems may be related not only to the finite moments condition, but also to the behaviour of the "internal" bath dynamics and rethermalization.

Furthermore, the above calculations are restricted, in fact, to the resonant part of the reservoir response function. The effects of the remaining parts of the reservoir have already been studied mainly for a purely bosonic environment and a two-level system in the second order or Born approximation. It has been shown that some decoherence is created by the environment, but the calculations have been done in the case where a free oscillation would exist in the small system even if it were isolated [24], or in the special case of an adiabatic coupling [25]. In a realistic calculation, all the effects suggested in those publications should be taken into account as well as those studied above; but a generalization of their methods, using a second order approximation for the non-resonant part of the spectrum and the motional-narrowing one for the resonant part, would not be a simple task.

The most of this work has been done in collaboration with Dr. J.J. Niez or at least in close relation with him. I am indebted to him for having submitted the problem from his analysis of experimental data, and for many stimulating discussions. Professor Privman, who was kind enough to send us one related article of his before publication, is thanked. Professor Vagner who gave me the occasion to finish and publish this work is also thanked for his efficiency. J. Pearce is thanked for careful philological help.

Appendix

The "Liouville formalism" is based on the remark that the commutator $[A, B]$ of two operators is a linear function of each of them. So one of the operators, say B, can be considered as a vector having as coordinates all its matrix elements and the commutator will be given by the action of an adapted operator. The vector space of the operators is the direct product of the Hilbert space by its dual, and operators in this space are often spoken of as “superoperators”. We shall use the notations

$$\left\langle m \left| Op \right| m' \right\rangle = \left(mm' \mid Op \right) \tag{A1}$$

where one sees the matrix elements to become the components of a vector. In the Liouville space, we shall use round brackets instead of the angular ones in the Hilbert space; otherwise, we keep a Dirac notation. Formula (A1) implies that the unit ket-bra $|m >< m'|$operator of the Hilbert space, corresponds to the unit vector $\mid mm\prime)$ in the Liouville space.

The important correspondance is that the commutator $[h, Op]$ corresponds to the Liouvillle ket $\widehat{h} \mid Op)$ where $\widehat{h}$ is the superoperator associated with the Hilbert operator h. One can see easily that the $\widehat{h}$ matrix elements are related to the h ones by

$$\left(mn \left| \widehat{h} \right| m'n' \right) = \left\langle m \left| h \right| m' \right\rangle \delta_{nn'} - \left\langle n' \left| h \right| n \right\rangle \delta_{mm'} \tag{A2}$$

Consequently, if the basis used is a diagonal one for h, the associated one is diagonal for $\widehat{h}$, and

$$\langle m \left| h \right| m \rangle = E_m \tag{A3}$$

corresponds to

$$\left(mn \left| \widehat{h} \right| m'n' \right) = \left(E_m - E_n \right) \delta_{mm'} \delta_{nn'} = \hbar \omega_{mn} \delta_{mm'} \delta_{nn'} \tag{A4}$$

The eigenvalues of the Liouville operator, or superoperator, are the frequencies of the transitions observed in a spectroscopic study of the system; this explains the use of this formalism in spectroscopic theory. To simplify, we denote the Liouville commutators divided by $\hbar$, associated with the hamiltonian h_i,

$$L_i = \frac{\widehat{h_i}}{\hbar} \tag{A5}$$

With this formalism the evolution equation for an operator

$$i\hbar \frac{\mathrm{d}}{\mathrm{d}t} Op(t) = [h, Op(t)] \tag{A6}$$

is replaced by

$$i \frac{\mathrm{d}}{\mathrm{d}t} \mid Op(t)) = L \mid Op(t)) \tag{A7}$$

and its solution

$$Op(t) = e^{-i\hat{h}t/\hbar} Op(0) e^{iht/\hbar} \tag{A8}$$

by

$$\mid Op(t)) = e^{-iLt} \mid Op(0)) \tag{A9}$$

Considering that all the methods used in quantum mechnics are mathematical technics used to solve problems in vectorial spaces, they can, without conceptual difficulties, be adapted to problems in the Liouville space.

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