Spontaneous and Induced Emission of Soft Bosons: Exact Non-Markovian Solution

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An exact solution of the time-dependent problem has been obtained for a degenerate two-level system interacting with a harmonic boson field. Unlike the Weisskopf-Wigner solution this solution takes into account various many-boson processes and is non-Markovian. There is no exponential decay and the asymptotic behavior explicitly depends on the initial conditions. Both spontaneous and stimulated processes have been considered.

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In many applications it is assumed that the relaxation process can be described by the master equation

$$\dot{P}_{n} = -\sum_{k} \left(W_{nk} P_{n} - W_{kn} P_{k} \right).$$
⁽¹⁾

Here, P_n is the probability that the system is in the state *n* once W_{nk} is a probability of transition (per unit time) from state n to state k. This equation is derived in the Markovian approximation (or in the Weisskopf-Wigner approximation for spontaneous radiation¹). The necessary condition of this approximation is that the eigenfrequency ω_{mn} has to be much larger than the transition rate W_{mn} (see, e.g., Refs. 2 and 3). For spontaneous emission of phonons or photons, the transition rate W_{mn} is typically proportional to $|\omega_{mn}|^3$. This means that the condition

$$W_{mn} / \left| \omega_{mn} \right| \ll 1 \tag{2}$$

can be satisfied at very low frequencies, even for the degenerate levels with

$$\omega_{mn} = 0. \tag{3}$$

On the other hand, at very low frequencies the processes of induced emission and absorption of bosons may prevail over the spontaneous emission. The transition rate W_{nm} for typical one-boson processes has the form

$$W_{nm} = \frac{2\pi}{\hbar} \sum_{k} |B_{k}|^{2} \delta(E_{n} - E_{m} \pm \hbar \omega_{k}) (n_{k} + \frac{1}{2} \mp \frac{1}{2}), \qquad (4)$$

where B_k is the interaction parameter and n_k is the number of bosons in the kth mode. For thermal equilibrium, and at low frequencies, one gets

$$n_k \approx kT/\hbar\,\omega_k\,.\tag{5}$$

In this case expressions for W_{nm} are finite and proportional to T (see, e.g., the so-called direct process in spinlattice relaxation⁴).

In my previous paper,⁵ it was shown that a condition of type (2) is a necessary one, but it is not sufficient. The interaction between bosons and the subsystem of interest (say, a two-level system) may lead to the creation of exact nondissipative discrete states (such as "superconducting states"). In this case the Markovian-type master equation (1) cannot be derived. Therefore a new kind of necessary Markovian condition emerges: The exact spectrum of the whole system should not contain nondissipative discrete levels.

The concrete system which was considered in Ref. 5 and will be analyzed here comprises a two-level system described by the effective spin (r_1, r_2, r_3) which interacts with a harmonic boson field:

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$$H = (\frac{1}{2} + r_3)\hbar\omega_0 + \frac{1}{2}\sum_k [(p_k^2 + \omega_k^2 q_k^2) - \frac{1}{2}\hbar\omega_k] - r_1\sum_k \frac{2(2\omega_k)^{1/2}}{\sqrt{\hbar}} B_k q_k$$

= $(\frac{1}{2} + r_3)\hbar\omega_0 + \sum_k a_k^{\dagger} a_k \hbar\omega_k - (r_+ + r_-)\sum_k B_k (a_k^{\dagger} + a_k).$ (6)

The effective spin $\frac{1}{2}$ has the usual components in r_3 representation:

$$r_{1} = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}; \quad r_{2} = \frac{1}{2} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}; \quad r_{3} = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}; \quad r_{\pm} = r_{1} \pm ir_{2}.$$
(7)

The operators a_k^{\dagger} and a_k describe creation and annihilation operators of bosons (phonons, photons, magnons, etc.). In the earlier paper,⁵ an exact solution has been obtained for the truncated Hamiltonian, without $a^{\dagger}r_+$ -type terms (see also Davidson and Kozak⁶). This corresponds to the so-called rotating-wave approximation. Terms of r_+a^{\dagger} type

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have been treated in the perturbation approach. It has been shown that when ω_0 is smaller than a certain ω_c ,

$$\omega < \sum_{k} |B_{k}|^{2} / \hbar^{2} \omega_{k} = \omega_{c} , \qquad (8)$$

an exact discrete nondissipative level emerges and the usual Markovian treatment is not valid anymore. On the other hand, the case of very low frequencies cannot be treated by such an approach since many-boson processes cannot be neglected.

In this Letter, I present exact solutions for the system described by the Hamiltonian (6) when $\omega_0 \rightarrow 0$.

Exact probability amplitudes.—The eigenfunctions and eigenenergies of Hamiltonian (6) (with $\omega_0 = 0$) can be written as

$$\Psi'_{\pm 1/2,\{n\}} = \phi'_{\pm 1/2} \prod_{k} \phi_{n_{k}} (q_{k} - q_{1k,2k}^{0});$$

$$E'_{\pm 1/2,\{n\}} = \sum_{k} n_{k} \hbar \omega_{k} - \sum_{k} \frac{|B_{k}|^{2}}{\hbar \omega_{k}}.$$
(9)

Here $\phi_n(q)$ are eigenfunctions of the harmonic oscillator

energy; $\phi'_{\pm 1/2}$ are eigenfunctions of the operator r_1 ,

$$r_{1}\phi'_{\pm 1/2} = \pm \frac{1}{2}\phi'_{\pm 1/2}, \quad \phi'_{1/2} = \frac{1}{\sqrt{2}}(\phi_{1/2} - \phi_{-1/2}),$$

$$\phi'_{-1/2} = -\frac{1}{\sqrt{2}}(\phi_{1/2} - \phi_{-1/2});$$
(10)

 $\phi \pm \frac{1}{2}$ are eigenfunctions of r_3 ; and

$$q_{1k}^{0} = -q_{2k}^{0} = (2/\hbar\omega_k)^{1/2} B_k .$$
⁽¹¹⁾

Following usual procedures, we can find the probability amplitudes c_m for the unperturbed states

$$\Psi(t) = \sum_{m} c_m(t) \Psi_m e^{-(i/\hbar)E_m t}.$$
(12)

It is assumed that at t=0 the system is in one of the unperturbed states Ψ_0 . Then, one easily gets

$$c_{m}(t)e^{-(i/\hbar)E_{m}t} = (\Psi_{m}, \Psi(t))$$
$$= \sum_{n} (\Psi_{m}, \Psi_{n}')(\Psi_{n}', \Psi_{0})e^{-(i/\hbar)E_{n}'t}.$$
 (13)

We assume that initially (t=0) the system was in the state

$$\Psi_0 = \phi_{1/2,\{N\}} = \phi_{1/2} \prod_k \phi_{N_k}(q_k); \quad E_0 = \sum_k N_k \hbar \omega_k , \quad (14)$$

with spin $r_3 = \frac{1}{2}$. Then we get

$$c_{\pm 1/2,\{n\}} \exp\left[i\sum_{k} (N_{k} - n_{k})\omega_{k}t\right] = \frac{1}{2} \langle N | \exp\left[-\frac{i}{\hbar}\sum_{k} q_{1k}^{0}p_{k}(t)\right] \exp\left[\frac{i}{\hbar}\sum_{k} q_{1k}^{0}p_{k}\right]$$
$$\pm \exp\left[-\frac{i}{\hbar}\sum_{k} q_{2k}^{0}p_{k}(t)\right] \exp\left[\frac{i}{\hbar}\sum_{k} q_{2k}^{0}p_{k}\right] | n \rangle, \quad (15)$$

where

$$|N\rangle = \prod_{k} \phi_{N_{k}}(q_{k}); \quad |n\rangle = \prod_{k} \phi_{n_{k}}(q_{k})$$
(16)

and

 $p_k(t) = p_k \cos(\omega_k t) - \omega_k q_k \sin(\omega_k t).$

When ω_k forms a quasicontinuum, and $q_{1k,2k}^0 \propto N^{-1/2}$ (with $N \rightarrow \infty$, when approaching the continuum), we obtain for $c_{1/2,\{N\}}$

$$c_{1/2,\{N\}} = \exp\left(-\sum_{k} \frac{|B_{k}|^{2}}{\hbar^{2} \omega_{k}^{2}} \{(2N_{k}+1)[1-\cos(\omega_{k}t)] + i\sin(\omega_{k}t)\}\right).$$
(18)

Relaxation via spontaneous emission of bosons: $N_k = 0$.—In this case, the wave function of the system can be written in the form

$$\Psi(t) = c_{1/2,0}(t)\Psi_{1/2,0} + \sum_{k} c_{-1/2,k}\Psi_{-1/2,k}e^{-i\omega_{k}t} + \sum_{k_{1},k_{2}} c_{1/2,k_{1}k_{2}}\Psi_{1/2,k_{1}k_{2}}\exp[-i(\omega_{k_{1}}+\omega_{k_{2}})t] + \cdots,$$
(19)

where

$$\Psi_{1/2,0} = \phi_{1/2} \prod_{k} \phi_0(q_k), \quad \Psi_{-1/2,k} = \phi_{-1/2} \phi_1(q_k) \prod_{k' (\neq k)} \phi_0(q_{k'}), \quad \Psi_{1/2,k_1k_2} = \phi_{1/2} \phi_1(k_1) \phi_1(k_2) \prod_{k' (\neq k_1,k_2)} \phi_0(q_k), \quad \text{etc.}$$
(20)

The probability amplitudes appearing in (19) can be easily found from (15) and (18),

$$c_{1/2,0} = \exp\left[1 - \sum_{k} \frac{|B_{k}|^{2}}{\hbar^{2} \omega_{k}^{2}} (1 - e^{-i\omega_{k}t})\right], \quad c_{-1/2,k_{1}...k_{2n+1}} \exp\left[-i\sum_{j=1}^{2n+1} \omega_{k_{j}t}\right] = c_{1/2,0}(t) \prod_{j=1}^{2n+1} \frac{B_{k_{j}}}{\hbar \omega_{k_{j}}} [1 - \exp(-i\omega_{k_{j}}t)],$$

$$c_{1/2,k_{1}...k_{2n}} \exp\left[-i\sum_{j=1}^{2n} \omega_{k_{j}}t\right] = c_{1/2,0}(t) \prod_{j=1}^{2n} \frac{B_{k_{j}}}{\hbar \omega_{k_{j}}} [1 - \exp(-i\omega_{k_{j}}t)].$$
(21)

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Thus we have found all the probability amplitudes for various multiboson processes appearing in (19). The probabilities for the system to have spins $\pm \frac{1}{2}$ can also be easily found:

$$P_{1/2} = |c_{1/2,0}|^{2} + \sum_{k_{1}k_{2}} |c_{1/2,k_{1}k_{2}}|^{2} + \dots = e^{-\Sigma(t)} \cosh\Sigma(t) ,$$

$$P_{-1/2} = \sum_{k} |c_{-1/2,k}|^{2} + \sum_{k_{1}k_{2}k_{3}} |c_{-1/2,k_{1}k_{2}k_{3}}|^{2} + \dots = e^{-\Sigma(t)} \sinh\Sigma(t) ,$$
ere
$$(22)$$

where

$$\Sigma(t) = 2\sum_{k} \frac{|B_k|^2}{\hbar \omega_k} [1 - \cos(\omega_k t)].$$
(23)

We can find the asymptotic values of $P_{\pm 1/2}$ if we note that

$$\lim_{t \to \infty} \Sigma(t) = 2 \sum \frac{|B_k|^2}{\hbar^2 \omega_k^2} = \Sigma_{\infty}.$$
 (24)

For the weak interaction,

$$\Sigma \ll 1, P_{1/2}(\infty) \approx 1 - \Sigma_{\infty}, P_{-1/2} \approx \Sigma_{\infty},$$
 (25)

while for the strong interaction,

$$\Sigma \gg 1, P_{1/2}(\infty) = P_{-1/2}(\infty) = \frac{1}{2}$$
 (26)

Relaxation via induced emission (absorption) of bo-

sons:
$$N_K \gg 1$$
.— When $N_k \neq 0$ (induced emission, absorption) the expression for $c_{1/2N}$ is given by (18), and in the case

$$N_k \gg 1 , \tag{27}$$

this expression obtains the form

$$c_{1/2,N} = \exp[-\frac{1}{2} \Sigma_{N}(t)];$$

$$\Sigma_{N}(t) = 4 \sum_{k} \frac{|B_{k}|^{2} N_{k}}{(\hbar \omega_{k})^{2}} [1 - \cos(\omega_{k} t)].$$
(28)

Expressions for the probability amplitudes $c_{1/2,N_k} \pm 1...$ can be found in the same way as expressions (21). It is easy to find that in the case (27), when one can neglect unity in comparison with N_k , we get

$$|c_{-1/2,N_{k_{1}}\pm 1...N_{k_{2n+1}}\pm 1}|^{2} = |c_{1/2,N}|^{2} \prod_{j=1}^{2n+1} \frac{2|B_{k_{j}}|^{2}N_{k_{j}}}{(\hbar\omega_{k_{j}})^{2}} [1 - \cos(\omega_{k_{j}}t)],$$

$$|c_{1/2,N_{k_{1}}\pm 1...N_{k_{2n}}\pm 1}|^{2} = |c_{1/2,N}|^{2} \prod_{j=1}^{2n} \frac{2|B_{k_{j}}|^{2}N_{k_{j}}}{(\hbar\omega_{k_{j}})^{2}} [1 - \cos(\omega_{k_{j}}t)].$$
(29)

We also obtain expressions similar to (22),

$$P_{1/2}(t) = e^{-\Sigma_N(t)} \cosh \Sigma_N(t), \quad P_{-1/2}(t) = e^{-\Sigma_N(t)} \sinh \Sigma_N(t)$$

where Σ_N is given by Eq. (28). Again, for large N when

$$\Sigma_N(\infty) = 4\sum_k \frac{|B_k|^2 N_k}{(\hbar \omega_k)^2} \gg 1, \qquad (31)$$

we get asymptotically

$$P_{1/2}^{\infty} = P_{-1/2}^{\infty} = \frac{1}{2} . \tag{32}$$

In the general case, the asymptotic values essentially depend on the initial condition $P_{1/2}(0)$. Thus for the weak interaction $[\Sigma_N(\infty) \ll 1]$,

$$P_{1/2}^{\infty} \approx 1 - \Sigma_N(\infty), \quad P_{-1/2}^{\infty} \approx \Sigma_N(\infty).$$
 (33)

Conclusions. - In case of the degenerate two-level system interacting with a harmonic boson field, we have obtained an exact solution of the time-dependent problem. Unlike previous exact solutions^{5,6} it does not employ a truncated Hamiltonian corresponding to the rotatingwave approximation. The probabilities of various many-boson processes have also been obtained. The solution behaves in an explicitly non-Markovian way. Expressions (22) and (30) do not decay exponentially like Weisskopf-Wigner solutions, or solutions of the master equation (1). Another striking difference with a conventional behavior is an explicit dependence on the initial condition. As mentioned, it is connected with the existence of the discrete nondissipative levels in the exact energy spectrum of the system. In the present model these levels are doubly degenerate, with the eigenenergy

$$E_{1/2,0} = E_{-1/2,0} = -\sum_{k} \frac{|B_{k}|^{2}}{\hbar \omega_{k}},$$

which follows from (9).

Indeed, all the implications of this exact solution are also valid for nonvanishing but small energy differences $\hbar \omega_0$. This can be verified by a comparison with the corresponding solutions⁵ in the region of their overlap.

I believe that possible experimental applications exist in the field of radiationless transitions, such as spinlattice relaxation.⁴

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(30)

85287-1604.

 1 V. Weisskopf and E. Wigner, Z. Phys. **63**, 54 (1930), and **65**, 18 (1930).

²B. Fain, Physica (Amsterdam) **101A**, 67 (1980).

³B. Fain, *Theory of Rate Process in Condensed Media* (Springer-Verlag, Berlin, 1980).

⁴Spin-Lattice Relaxation in Ionic Solids, edited by A. A. Manenkov and R. Orbach (Harper and Row, New York, 1966).

⁵B. Fain, Phys. Rev. A **37**, 547 (1988).

 6 R. Davidson and J. J. Kozak, J. Math. Phys. **26**, 56 (1985), and references therein.