Fluorescence into flat and structured radiation continua: An atomic density matrix without a master equation.

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We investigate an atomic Λ-system with one transition coupled to a laser field and a flat continuum of vacuum modes and the other transition coupled to field modes near the edge of a photonic band gap. The system requires simultaneous treatment of Markovian and non-Markovian dissipation processes, but the photonic band gap-continuum cannot be eliminated within a density matrix treatment. Instead we propose a formalism based on Monte-Carlo wavefunctions, and we present results relevant to the experimental characterization of a structured continuum.

With the advent of photonic band gap (PBG) materials and dispersive media, the mode structure of the electromagnetic field can be tailored in a controllable fashion providing for instance band gaps or defect modes of various forms\textsuperscript{[1,2]}. The rapidly varying mode structure in the radiation reservoir invalidates the Born-Markov approximations normally employed for a simple quantum system like an atom when this is located inside a PBG-material with transition frequency near the edge of the gap. The reservoir degrees of freedom are thus not easily eliminated to derive a master equation for the reduced system dynamics. The main body\textsuperscript{[1,2]} of theoretical works on atomic interactions within PBG materials has therefore addressed the unitary dynamics in terms of the complete atom(s)+field wavefunctions. In this paper, we address a Λ-system with one laser-driven transition experiencing a flat vacuum without structure and the frequency of the other transition near the edge of a PBG. A unitary wavefunction dynamics is incompatible with the treatment of the atomic fluorescence on the “free”-space transition, and we must seek a way to apply the simple Markovian properties of this process in the solution of the complete problem. A formulation in terms of Monte-Carlo wavefunctions (MCWF) turns out to be particularly useful for this purpose. This method then also suggests itself as a means of solving other problems emerging in the overlapping domain of quantum optics, semiconductors and nano-structures where dissipation of Markovian and non-Markovian character may co-exist. Furthermore, our work establishes an application of the MCWF treatment which goes beyond its conventional correspondence with Born-Markov master equations; at no point does such a master equation appear in this work.

The Λ-system is interesting from an experimental point of view since atoms may be present in their ground state in the dielectric host, and the dynamics of the interaction with the field modes in the vicinity of the gap may be studied when the laser excitation on the “free”-space transition is turned on. In partial analogy with the shelving scheme technique we note that the fluorescence signal on the “free”-space transition may serve as to probe details of the interaction between the atom and the field modes in the PBG material.

In a PBG one finds a modified dispersion relation for the photons in the radiation reservoir. The methods presented in the following apply for any mode structure and dispersion relation, but to illustrate the method we employ the isotropic model introduced by John and Wang\textsuperscript{[3–6]}\textsuperscript{[3–6]}. Their dispersion relation for a periodic array of dielectric scatterers of radius \(a\) and index of refaction \(n\), reads (with a separation of \(b = 2an\) between the scatterers):

\[
\omega_k = \frac{c}{4na} \arccos \left[ \frac{4n \cos(2ka(1 + n)) + (1 - n)^2}{(1 + n)^2} \right]
\]

which leads to a gap centered at the frequency \(\omega_0 = \frac{\pi c}{4na}\). With \(n = 1.082\) the gap width \(\Delta \omega\) is 0.05\(\omega_0\) and the upper band edge frequency \(\omega_e\) is given by \(\omega_e = \omega_0 + \frac{\hbar \Delta \omega}{2}\). In the vicinity of the band edge we have (effective mass approximation)

\[
\omega_k = \omega_e + A(k - k_0)^2,
\]

where \(A = -2ac / \sin(4na\omega_e/c)\) and \(k_0 = \pi / (2a(n + 1))\).

We consider a three-level atom with two lower levels \(|a\rangle\) and \(|c\rangle\) coupled by the electric dipole coupling to a common excited level \(|b\rangle\), see fig. 4. On the \(|a\rangle \leftrightarrow |b\rangle\) transition we apply a laser field, and the atom may decay by spontaneous emission due to the coupling to a flat radiation reservoir. The transition \(|b\rangle \rightarrow |c\rangle\) is accompanied by the emission of a photon with frequency in the vicinity of the photonic band gap edge, and this atomic transition is significantly modified by the presence of the dielectric host.

Neglecting the zero-point energies of the field modes, and setting the atomic energy levels to the values \(0, \hbar \omega_b\) and \(\hbar \omega_e\) respectively, we write the Hamiltonian for the system (\(\hbar = 1\)):
\[ H = \omega_b \sigma_{bb} + \omega_c \sigma_{cc} + \sum_{\lambda} \omega_\lambda a_\lambda \dagger a_\lambda + \sum_{\lambda} \omega_\lambda b_\lambda \dagger b_\lambda + V \] (2)

where the interaction term in the rotating wave approximation is given by

\[ \begin{align*}
V &= i \sum_{\lambda} g_\lambda (a_\lambda \dagger \sigma_{ab} - a_\lambda \sigma_{ba}) + i \sum_{\lambda} g_\lambda (b_\lambda \dagger \sigma_{cb} - b_\lambda \sigma_{bc}) \\
&\quad + i \gamma (\sigma_{ba} - \sigma_{ab} e^{i \omega_c t})
\end{align*} \tag{3} \]

where \( \sigma_{ij} \) denote atomic dyadic operators \(|i\rangle \langle j|\) with \( i, j \in \{a, b, c\}\); \( a_\lambda, b_\lambda \) are the field annihilation operators of the flat vacuum and PBG vacuum, respectively, and the laser field is represented by a semiclassical c-number field. We assume that the coupling to the flat continuum may be treated by perturbation theory in the usual way, \textit{i.e.} an energy shift (Lamb shift) and a decay rate \( \gamma \) may be attributed to the excited state \(|b\rangle\). The Lamb shift is assumed incorporated in the atomic energy \( \omega_b \) in eq. (2), and the decay rate describes an incoherent transition mechanism by which atoms in the excited state \(|b\rangle\) decay to the ground state \(|a\rangle\). We shall incorporate the decay mechanism by an effective non-hermitian Hamiltonian \( H_{\text{eff}} \). First, we identify the wavefunction evolution governed by this Hamiltonian, and next, by appealing to the Monte Carlo wavefunction formalism we shall obtain the exact evolution of the atomic system.

We apply the resolvent operator defined as

\[ G(z) = \frac{1}{z - H_{\text{eff}}} \]

where \( z \) is a complex Laplace variable and \( H_{\text{eff}} \) is the effective non-hermitian Hamiltonian of the system given by eqs. (2) and (3), but with the sums over \( a_\lambda \) operators suppressed, and the replacement \( \omega_b \to \omega_b - i \gamma / 2 \).

With the system initially in state \( a \), the resolvent operator equations read

\[ \begin{align*}
(z - 0)G_{aa}(z) &= 1 + V_{ab} G_{ba}(z + \omega_L) \\
(z - \omega_\lambda)G_{ca}(z) &= V_{bc} G_{ba}(z) \\
(z - \omega_b + i \gamma / 2)G_{ba}(z) &= V_{ba} G_{aa}(z - \omega_L) + \sum_\lambda V_{bc_\lambda} G_{c_\lambda a}(z)
\end{align*} \tag{4} \]

where \( V_{ab} = g_L \) and the amplitudes \( G_{c_\lambda a}(z) \) pertain to the PBG-continuum states \(|c_\lambda \rangle \otimes |1_\lambda \rangle\). Using eq. (4) in the summation over continuum modes and turning the summation into an integral, we get

\[ (z - \omega_b + i \gamma / 2)G_{ba}(z) = V_{ba} G_{aa}(z - \omega_L) - iCG_{ba}(z) / \sqrt{z - \omega_c} \]

where the effective dipole coupling to the mode structure is given by \( C = d^2 k_0^2 \omega_c / (4 \pi \epsilon_0 \sqrt{\lambda}) \) \( \text{[4]} \), with \( d \) the atomic dipole moment on the \( b \leftrightarrow c \) transition.

Solving these coupled, algebraic equations for \( G_{aa} \) and \( G_{ba} \), we find

\[ \begin{align*}
G_{aa}(z) &= \frac{(z - \omega_b + i \frac{1}{2} \gamma) + i C / \sqrt{z - \omega_c}}{(z - \omega_L)((z - \omega_b + i \frac{1}{2} \gamma) + i C / \sqrt{z - \omega_c}) - |V_{ab}|^2} \\
G_{ba}(z) &= \frac{V_{ba}}{(z - \omega_L)((z - \omega_b + i \frac{1}{2} \gamma) + i C / \sqrt{z - \omega_c}) - |V_{ba}|^2}
\end{align*} \tag{5, 6} \]

The dynamics of the system is obtained by inverting the amplitudes to time domain by means of the inversion integral for the time evolution operator,

\[ U(t) = \frac{1}{2 \pi i} \int_{-\infty + i \epsilon}^{-\infty - i \epsilon} dz G(z) e^{-izt} \]

where \( \epsilon \) is an infinitesimal small positive quantity. Due to the high order of the polynomial of \( z \) in the denominator and the presence of the square root terms in \( \text{[4]} \) and \( \text{[3]} \), it is not easy to apply the residue theorem and to obtain the amplitudes in time domain analytically. Instead we compute the two inversion integrals numerically. This integration is straightforward, and the calculation yields for example the populations of the initial ground state \(|a\rangle\) and of the excited atomic state \(|b\rangle\) as function of time, \( \pi^0_a(t) = |U_{aa}(t)|^2 \), \( \pi^0_b(t) = |U_{ba}(t)|^2 \). We keep track of the norm \( P(t) \) of the wavefunction, noting that it changes only due to the imaginary part of the excited state energy, and hence \( \frac{d P}{dt} \big|_{\text{loss}} = -\gamma \pi^0_b(t) \), which in integrated form reads

\[ P(t) = 1 - \gamma \int_0^t dt' \pi^0_b(t') \]

In fig. \( \text{[3]} \) we show an example of the relevant time dependent quantities \( P(t) \), \( \pi^0_a(t) \) and \( \pi^0_b(t) \). From the figure, it is evident that the populations in the states \(|a\rangle\) and \(|b\rangle\) approach zero after a transient evolution. There is, however, a substantial part of the population \( (P(\infty) \sim 20\%) \) which is not lost by fluorescence on the free-space transition. This population is transferred to the atomic state \(|c\rangle\) associated with the PBG-continuum, and at any time we have \( \pi^0_c(t) = P(t) - \pi^0_a(t) - \pi^0_b(t) \).

The spontaneous decay on the \(|b\rangle \to |a\rangle\) transition was treated only as a loss mechanism for the excited state amplitude, but the atoms are incoherently fed back in the ground state \(|a\rangle\), and from here they are re-excited by the laser. In a density matrix formulation, when tracing over the resulting different photon number states of the flat reservoir, the elimination of the PBG-modes would be exceedingly difficult, if possible at all.

It has been shown that dissipative problems in quantum mechanics may be solved by stochastic wavefunction equations as an alternative to master equations \( \text{[5]} \text{[3]} \). In the “quantum jump” scheme, one propagates state vectors according to a non-hermitian Hamiltonian, and at certain instants of time, chosen according to a random process, this propagation is interrupted by quantum jump projections of the state vectors (see \( \text{[3]} \) for a recent review). In the formulations of the method so far, the continuously propagated state vector is described as
the solution of a Schrödinger equation, but, the atomic populations \( \pi^0_0(t) \) identified after elimination of the PBG reservoir above may be applied just as well for the construction of the atomic density matrix.

The function \( P(t) \) is the norm of the no-jump state vector and consequently the probability that a photon has not been registered in the flat reservoir at time \( t \).

The ensemble averaged populations can be found by solution of integral equations: The population of an atomic state \( \pi_{ci}(t) \) is a sum of a term representing the population given that the atom has not decayed and a term representing the population given that the latest jump occurred at time \( t' \),

\[
\pi_{ci}(t) = \pi^0_{ci}(t) + \int_0^t dt' \gamma \pi_{bi}(t') \pi^0_{ci}(t - t').
\]

Note that \( \pi^0_{ci}(t) = \pi^0_0(t)/P(t) \cdot P(t) \) provides the given normalized population with the appropriate no-jump weight-factor. Eq. (8) must be solved for \( \pi_{ci}(t) \) first, e.g. by a Laplace transform: \( \pi_0(z) = \pi^0_0(z)/(1 - \gamma \pi_0^0(z)) \), and one may subsequently obtain the other populations (see also [10]).

The populations can of course also be found by simulations. In a single trajectory one considers the normalized populations \( \pi_i(t) = \pi^0_i(t)/P(t) \) (and other density matrix elements if necessary), until a jump occurs when \( P(t) \) equals a random number \( \varepsilon \) chosen uniformly on the interval between zero and unity. The quantum jump puts the atom in the state \( |a\rangle \), and from here the evolution starts over again. In fig. 3 we plot \( \pi_a(t) \) and \( \pi_c(t) \) obtained by an average of \( 10^3 \) stochastic wavefunctions for the same parameters as used in fig. 2.

Let us comment on the atomic dynamics obtained in fig. 3. After an initial transient evolution, the populations \( \pi_{a,b}(t) \) approach zero in a non-exponential way. The fluorescence signal on the \( |b\rangle \rightarrow |a\rangle \) transition thus vanishes as opposed to the case of a two-level atom in free space. In the simulations we note that no jump will occur if the random number \( \varepsilon \) is smaller than \( P(\infty) \).

When this value is non-zero, each realization only exhibits a limited number of jumps since eventually the value chosen for \( \varepsilon \) will be smaller than \( P(\infty) \). The probability of having exactly \( k \) photon emissions (jumps) in a given simulated trajectory is \((1 - P(\infty))^k P(\infty)\), and the mean number of photons emitted per atom is \( P(\infty)^{-1} - 1 \). The amplitude \( U_{ci,a}(t) \) is the inverse Laplace transform of \( G_{ci,a} = V_{ci,b}G_{ba}(z - \omega_\lambda) \). In the long time-limit only the pole \( z = \omega_\lambda \) contributes and thus \( U_{ci,a}(t) = V_{ci,b}G_{ba}(\omega_\lambda)e^{-i\omega_\lambda t} \) and since \( P(\infty) = \pi^0_0(\infty) \) we find

\[
P(\infty) = \sum_\lambda |V_{ci,b}G_{ba}(\omega_\lambda)|^2
\]

The summation can be turned into an integral, which is calculated numerically. For a \( \Lambda \)-system in free space with a branching of the decay from the upper state, there will also be a finite number of fluorescence photons emitted on the laser driven transition, corresponding to \( P(\infty) = \gamma'/[(\gamma + \gamma') \cdot \omega_\lambda] \) with \( \gamma, \gamma' \) being the decay rates of state \( |b\rangle \) to the states \( |a\rangle \) and \( |c\rangle \) respectively, and the total number of fluorescence photons on the \( |b\rangle \rightarrow |a\rangle \) transition is thus independent of the parameters of the driving field. This is different in our case, as shown in fig. 3, where \( P(\infty) \) and the mean number of fluorescence photons emitted on the free space transition are plotted as functions of the laser detuning from the PBG edge for different choices of the laser coupling. For a rather weak laser coupling, the transition to the atomic state \( |c\rangle \) is a Raman-process which is strongly suppressed when the laser is tuned below the band gap edge since there are then no resonant PBG-modes for the Stokes photon. A stronger laser coupling leads to an Autler-Townes splitting of level \( |b\rangle \) and population is then transferred to the PBG-continuum by a higher order process, removing the step-like character of \( P(\infty) \). The fluorescence signal may hence probe details of the PBG structure.

In conclusion we have demonstrated a technique for the solution of a problem, for which a Born-Markov master equation does not exist. We have in a parallel study derived a reduced master equation for the \( \Lambda \)-system applying only the Born-approximation. The numerical solution of the resulting non-Markovian master equation yields a very poor agreement with the exact results presented here thus invalidating the use of the Born-approximation. These results will be presented elsewhere. The specific form of the structured continuum is not essential for our approach, but it is important that only one photon states of the PBG-continuum appear (a possible slow decay from state \( |c\rangle \) back to \( |a\rangle \) can only be treated if we may assume that the photon in the PBG continuum escapes before the atom is reexcited to level \( |b\rangle \)). The simulations and the analytical expression (8) are simplified by the fact that all jumps put the atom in the same state. Our formalism, however, is perfectly capable of treating more general systems with branching of the decay from state \( |b\rangle \) to multiple states \( |a_j\rangle \). This implies that the index \( a \) is replaced by the set of indices \( a_j \) with the corresponding enlargement of the set of equations (3) and (8). The no-jump evolution and the associated decay function, following a jump to a given level \( a_j \), are then readily computed.

The MCWF technique has been applied to non-Markovian problems through the solution of Markovian master equations for enlarged model systems. Our situation, however, is different, since without ever having a master equation we have, by recourse to conditioned wavefunction dynamics, been able to obtain the atomic density matrix. We anticipate that by combination of the ideas in ref. [17] and in this paper, a wide class of non-Markovian problems may become tractable.
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FIG. 1. Level scheme

\[ \begin{align*}
|a\rangle & \quad \text{---} \quad |c\rangle \\
\omega_e & \quad \text{---} \quad |b\rangle \\
\omega_L & \quad \text{---} \quad \text{--- }
\end{align*} \]

FIG. 2. The populations \( \pi^0_a(t) \) (dashed line), \( \pi^0_b(t) \) (solid line) and the norm \( P(t) \) (dotted line) are plotted as functions of time. The parameters chosen are: \( C^{2/3} = \gamma/3 \), \( V_{ab} = \gamma \), \( \omega_e = \omega_b \).

FIG. 3. The populations \( \pi_a(t) \) (dashed line) and \( \pi_b(t) \) (solid line) are plotted as functions of time. The parameters chosen are the same as in fig. 2. The curves are averaged over \( 10^4 \) trajectories.

FIG. 4. Solid line is \( P(\infty) \) for \( V_{ab} = \gamma/2 \) and dashed line the number of fluorescence photons. The dot-dashed line is \( P(\infty) \) for \( V_{ab} = 3\gamma \) and the long-dashed line the number of fluorescence photons. The parameters chosen: \( C^{2/3} = \gamma \), \( \omega_b = \omega_e \).