Non-Markovian resonance fluorescence

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Abstract We derive a general formula for the non-Markovian fluorescence spectrum of a multi-level system interacting with a bosonic environment. To this end, we apply linear-response theory to describe the dynamics of a detector monitoring the emission spectrum of a general multi-level system. The resultant emission lineshape function is directly related to the two-time correlation of system observables, which we derive using Nakajima-Zwanzig Generalized Master Equation without assuming a Markov approximation.

Keywords Generalized Master Equation · Non-Markovian · Quantum Optics · Resonance fluorescence

1 Introduction

Resonance fluorescence of a multi-level system, strongly driven by a laser field, has been an active area of research both theoretically and experimentally (Peng et al., 2019; Konthasinghe et al., 2019; Schll et al., 2019; Yang and An, 2016; Ulrich et al., 2011). Moreover, most of the theoretical work to compute the resonance fluorescence in the context of open quantum system, where system $S$ interacts with its environment $E$, has been developed in terms of a purely Markovian decay process. A Markovian decay process assumes that the correlation time is much smaller than the system decay time and it is not affected by the time scales close to system decay time. Once the theory has been relied on the assumption of a Markovian (history independent) decay process, the quantum regression theorem (QRT) can be applied to calculate the dynamics of system correlation function (Mollow, 1969; Swain, 1981; Lax, 2000, 1968). However, the dominant real physical processes in such interactions (e.g. nuclear spins, phonons) (Coish and Loss, 2004; Kumar, 2018; Krummheuer et al., 2002; Vagov et al., 2014; Weiler et al., 2012; Roy and Hughes, 2012) are known to follow a non-Markovian (history-dependent) dynamics. Therefore, the fluorescence lineshape predicted using a Markovian theory and applying QRT can overlook the correct physical process in the interaction and lead to incorrect results.

In this paper, we intend to perform a detailed theoretical analysis to compute a general formula for resonance fluorescence spectrum of a multi-level system undergoing a non-Markovian decay process. We have used the Nakajima-Zwanzig generalized master equation (GME) (Fick and Sauermann, 1990) to calculate the dynamics
of system’s two-time correlation function to all orders beyond Markov approximation, which is directly related to the resultant fluorescence spectrum (Mollow, 1969; de Vega and Alonso, 2008). Moreover, we discuss the presence of an additional non-vanishing term in the two-time correlation known as final term which does not allow us to describe the dynamics within QRT, since this will essentially lead to applying a Markovian approximation (Guarnieri et al., 2014; Ford and O’Connell, 1996). This final term can be neglected under Markovian assumption and then the well-known quantum regression theorem can be applied to compute the system correlation function (Swain, 1981).

In order to illustrate the theory, we have already discussed the fluorescence spectra of a laser driven two-level system, embedded in a cavity and coupled to a three-dimensional bath of acoustic phonons within non-Markovian regime in our previous paper (Kumar, 2018).

The structure of this paper is the following. In Sec. 2, we present a general Hamiltonian of a multi-level system which we want to discuss along with its environment. In Sec. 3, we discuss a theoretical setup and its general Hamiltonian in terms of system and environment observables. In Sec. 4, we discuss the linear response theory to describe the dynamics of emitted radiation in terms of system’s observables. In Sec. 5, we discuss and derive the Nakajima-Zwanzig GME to compute the two-time correlation and finally obtain the expression for fluorescence lineshape function. Sections 6 and 7 are devoted to discuss the results and conclusion, respectively.

2 Model Hamiltonian

We start with the Hamiltonian of a general multi-level system interacting with radiation modes of the electromagnetic field, which can be written as system \( H_S \), field \( H_R \), and interaction \( H_{SR} \) in terms of the standard Jaynes-Cummings model within a rotating-wave approximation,

\[
H = H_0 + H_{SR} \\
H_0 = H_S + H_R \\
H_R = \sum_k \hbar \omega_k a_k^\dagger a_k \\
H_{SR} = \sum_k \hbar g_k (\sigma_+ a_k + \sigma_- a_k^\dagger),
\]

where \( \sigma_+ = |a\rangle\langle b| \) and \( \sigma_- = |b\rangle\langle a| \) are the raising and lowering operators between a selected excited state \( |a\rangle \) and ground state \( |b\rangle \) in the Hilbert space of the system and \( a_k, a_k^\dagger \) are the annihilation and creation operators in the Hilbert space of a set of electromagnetic modes coupled to the system. The coupling to the radiation modes is given by the coupling constant \( g_k \) to a mode of frequency \( \omega_k \). The coupling constant \( g_k \) is related to the electric-dipole transition matrix element \( |\rho_{ab}| \) through Eq. (4).

3 Fluorescence spectra

We theoretically describe a realistic detector with the well-known gedanken spectrum analyzer (Scully and Zubairy, 1997; Cohen-Tannoudji and Grynberg, 2004) which measures the scattered fluorescence light from the emitting system. We assume that the radiation field emitted by the system is detected by a two-level (detector) atom, with transition frequency \( \omega_\alpha - \omega_\beta = \omega_0 \). The detector atom has sharp levels \( |\alpha\rangle \) and \( |\beta\rangle \) separated by energy \( \omega_0 \) and is initially prepared in the ground state \( |\beta\rangle \) and is placed inside a shutter, that only opens during a certain observation time \( T \), during which it receives the emitted radiation and may be excited to the upper level \( |\alpha\rangle \). The excitation rate of the detector sets the detector response profile, centered at frequency \( \omega_0 \) with
bandwidth $\Delta \omega_B$. The detector bandwidth is assumed to be small compared to the typical feature size in the fluorescence spectrum. The detector atom has a Hamiltonian of the form,

$$H_D = \frac{\hbar \omega_0}{2} (|\alpha\rangle \langle \alpha| - |\beta\rangle \langle \beta|).$$  \hspace{1cm} (5)$$

The total Hamiltonian (for system, field and detector) has the form,

$$H_T = H + H_D + H_{DR}. \hspace{1cm} (6)$$

The coupling between detector atom $H_D$ and the radiation field, included in the Hamiltonian $H_T$, is given by the Hamiltonian

$$H_{DR} = \sum_k \hbar g_k^D (|\alpha\rangle \langle \beta| a_k + |\beta\rangle \langle \alpha| a_k^\dagger + |\alpha\rangle \langle \beta| a_k + |\beta\rangle \langle \alpha| a_k), \hspace{1cm} (7)$$

where $g_k^D$ is the coupling of the detector to field mode $k$ and the detector coupling Hamiltonian $H_{DR}$, in the interaction picture with respect to the Hamiltonian $H_0' = H + H_D$, is given by

$$H_{DR}^I(t) = e^{iH_0't/\hbar} H_{DR} e^{-iH_0't/\hbar}, \hspace{1cm} (8)$$

we adopt the rotating wave approximation which is equivalent to dropping the energy non-conserving terms, to write

$$H_{DR}^I(t) = \sum_k \hbar g_k^D \left( \sigma_{\alpha\beta} a_k(t) e^{i\omega_0 t} + \sigma_{\beta\alpha} a_k^\dagger(t) e^{-i\omega_0 t} \right), \hspace{1cm} (9)$$

Here, $\sigma_{\alpha\beta} = |\alpha\rangle \langle \beta|, \sigma_{\beta\alpha} = |\beta\rangle \langle \alpha|$ and $a_k(t)$ is in the interaction picture with $H_0'$, and we assume that the electric field is linearly polarized along the $x$-axis. The positive-frequency part of the electric field is defined by

$$E^+(t) = \sum_k \varepsilon_k a_k(t), \hspace{1cm} (10)$$

the negative-frequency part of the electric field is $E^-(t) = [E^+(t)]^\dagger$ and the detector coupling $g_k^D$ is related to the electric-dipole transition matrix element $|\rho_{\alpha\beta}|$ through (Scully and Zubairy, 1997),

$$g_k^D = \frac{|\rho_{\alpha\beta}| \varepsilon_k}{\hbar}. \hspace{1cm} (11)$$

In the long-wavelength limit, we take the dipole element $\rho_{\alpha\beta} \approx \rho_{\alpha\beta}^k$ (independent of $k$). From Eqs. (9) and (11), we find the detector coupling Hamiltonian

$$H_{DR}^I(t) = \rho_{\alpha\beta} \sigma_{\alpha\beta} E^+(t) e^{i\omega_0 t} + \rho_{\beta\alpha} \sigma_{\beta\alpha} E^-(t) e^{-i\omega_0 t}, \hspace{1cm} (12)$$

where $\rho_{\alpha\beta} = e\langle \alpha | x | \beta \rangle = \rho_{\alpha\beta}^*$. From here and what follows, for the simplicity we will use $\hbar = 1$. 
4 Linear response theory

Linear response theory (Bruus and Flensberg, 2004) states that the response to a weak external perturbation is proportional to the perturbation, and therefore all we need to understand is the proportionality constant. We consider that the detector is weakly coupled to the scattered emitted field, which allows us to do the linear response on the detector coupling. With this setup, the idea is to calculate the spectrum of light emitted by the system in the presence of a detector coupled to the emitted field at some point in time $t > 0$. This spectrum is defined in terms of the probability to excite the detector atom, $P(\omega_0, t)$ at time $t$. The probability of exciting the detector atom to excited level $|\alpha\rangle$ is found by calculating the expectation value of the projection operator $|\alpha\rangle\langle\alpha|$, which can be evaluated as

$$P(\omega_0, t) = \langle \psi_I(t)|\alpha\rangle\langle\alpha|\psi_I(t)\rangle,$$

(13)

where $|\psi(t)\rangle$ is the interaction-picture state, and is given by

$$|\psi_I(T)\rangle = U_I(T)|\psi_I(0)\rangle,$$

(14)

$U_I(T)$ is the time-evolution operator given by

$$U_I(T) = \mathcal{T}\exp\{-i\int_0^T dt H^{DR}_I(t)\} = 1 - i \int_0^T dt_1 H^{DR}_I(t_1) \frac{1}{2} T \int_0^T dt_1 \int_0^T dt_2 H^{DR}_I(t_1) H^{DR}_I(t_2) + \ldots$$

(16)

We assume that the detector atom is initially in the ground state $|\beta\rangle$ and that the initial state of the system, radiation modes, and detector is given by the product state $|\psi_{SR}(0)\rangle \otimes |\beta\rangle$, where $|\psi_{SR}(0)\rangle$ is an eigenstate of the Hamiltonian $H$, including the coupling between system and the radiation modes. Suppose now that at some time $t=0$, an external perturbation is applied, driving the system out of equilibrium. The perturbation is described by the term $H^{DR}_I(t)$. Now we wish to find the expectation value of the operator $|\alpha\rangle\langle\alpha|$ at time $T > 0$. In order to do so, we must find the time evolution of the state $|\psi_I(T)\rangle$ in the interaction picture with the interaction picture Hamiltonian $H^{DR}_I(t)$. To linear order in $H^{DR}_I$, we obtain an expression for the state in the interaction picture up to first order in the perturbation

$$|\psi_I(T)\rangle \approx \left[1 - i \int_0^T dt_1 H^{DR}_I(t_1)\right] |\psi_{SR}(0)\rangle.$$

(17)

Going back to the Schrödinger picture, we have

$$|\psi(T)\rangle = e^{-i H^0 t} \left[1 - i \int_0^T dt_1 H^{DR}_I(t_1)\right] |\psi_{SR}(0)\rangle.$$

(18)

Therefore the excitation probability, in the Schrödinger picture, takes the form

$$P(\omega_0, t = T) = \langle \psi(0)|U^\dagger_I(T)e^{i H^0 T} |\alpha\rangle\langle\alpha|e^{-i H^0 T} U_I(T)|\psi(0)\rangle = \langle \psi(0)|U^\dagger_I(T)|\alpha\rangle\langle\alpha|U_I(T)|\psi(0)\rangle,$$

(19)

here we have used $[H^0, |\alpha\rangle\langle\alpha|] = 0$. By substituting the values of $H^{DR}_I(t_1)$ and then $|\psi(T)\rangle$ in equations (17) and (13), respectively, the resulting expression for the excitation probability in the interaction picture is calculated as,

$$P(\omega_0, T) = |\rho_{\alpha\beta}|^2 \int_0^T dt_1 \int_0^T dt_2 \langle E^-(t_1) E^+(t_2)\rangle e^{i \omega_0 (t_1 - t_2)}.$$

(20)

Here, we have generalized to a mixed-state initial condition and the average $\langle \ldots \rangle = \text{Tr}\{\ldots \rho(0)\}$, where $\rho(0)$ is the total density matrix of the scattering system plus detector and the radiation field at time $T = 0$ is not
a product state. From Eq. (20), it follows that the excitation probability, \( P(\omega_0, T) \), of the detector atom is proportional to the two-time correlation function of the field operators, i.e., \( \langle E^-(t_1)E^+(t_2) \rangle \). We write the probability of excitation in terms of observables of the emitting system. We start with the Heisenberg equation of motion for \( a_k(t) \) with the Hamiltonian \( H \) given by Eq. (2), i.e.,

\[
\frac{da_k(t)}{dt} = -i[a_k(t), H],
\]

we obtain an equation for \( a_k(t) \)

\[
\dot{a}_k(t) = -i\left(\omega_k a_k(t) + g_k \sigma_-(t)\right).
\]

After integrating the above equation starting from time \( t = t_0 < 0 \), we obtain the expression

\[
a_k(t) = a_k(0)e^{-i\omega_k t} - i \int_0^t d\tau g_k \sigma_-(\tau)e^{-i\omega_k(t-\tau)}.
\]

We insert the above expression for \( a_k(t) \) into the positive-frequency part of the electric field operator, giving

\[
E^+(t) = \sum_k \varepsilon_k a_k^\dagger(0)e^{i\omega_k t} + i \int_0^t d\tau g_k \sigma_+(\tau)e^{i\omega_k(t-\tau)}.
\]

Similarly, the negative frequency part of the electric field operator is

\[
E^-(t) = \sum_k \varepsilon_k a_k^\dagger(0)e^{-i\omega_k t} + i \int_0^t d\tau g_k \sigma_-(\tau)e^{-i\omega_k(t-\tau)}.
\]

Replacing equations (24) and (25) in eqn. (20), we get (neglecting terms \( \sim \langle a_k a_k^\dagger \rangle, \langle a_k \sigma_+ \rangle \) and \( \sigma_- a_k^\dagger \))

\[
P(\omega_0, T) = \int_0^T dt_1 \int_0^T dt_2 e^{-i\omega_0(t_1-t_2)} \times \left\{ \int_0^{t_1} d\tau_1 \int_0^{t_2} d\tau S^*(t_1 - \tau')S(t_2 - \tau)\langle \sigma_+(\tau')\sigma_-(\tau) \rangle \right\},
\]

where \( S(t - \tau) \) is the detector response function

\[
S(t - \tau) = \sum_k g_k g_k^D e^{-i\omega_k(t-\tau)}
\]

\[
= \alpha \sum_k g_k^2 e^{-i\omega_k(t-\tau)}
\]

here we have used \( g_k^D = |\rho_{k\beta}| \varepsilon_k \) and \( \alpha = \frac{g_k^D}{|\rho_{k\beta}|} \) is the ratio of the detector and system coupling strengths. The detector response function \( S(t) \) typically decays rapidly with a decay time \( \tau_{cd} \), which is given by the inverse of the detector bandwidth, \( \Delta\omega_B \).

\[
\tau_{cd} = \frac{1}{\Delta\omega_B}
\]

whereas \( \langle \sigma_+(\tau')\sigma_-(\tau) \rangle \) evolves on a typical time scale \( T_{sys} \), where, by assumption, \( T_{sys} \gg \tau_{cd} \). We apply change of variables in the Eq. (26) as \( \tau' = t_1 - \tau' \) and \( \tau = t_2 - \tau \) to write

\[
P(\omega_0, T) = \int_0^T dt_1 \int_0^T dt_2 e^{-i\omega_0(t_1-t_2)} \times \left\{ \int_0^{t_1} d\tau_1 \int_0^{t_2} d\tau S^*(\tau')S(\tau)\langle \sigma_+(t_1 - \tau')\sigma_-(t_2 - \tau) \rangle \right\}.
\]
When \( \langle \sigma_+ (t_1 - \tau') \sigma_- (t_2 - \tau) \rangle \) is a slow varying function where \( S(\tau) \) is finite, we approximate: 

\[
\langle \sigma_+ (t_1 - \tau') \sigma_- (t_2 - \tau) \rangle \approx \langle \sigma_+ (t_1) \sigma_- (t_2) \rangle
\]

and extend the upper limit of integrations to \( t_{1,2} \to \infty \) for \( t_{1,2} \gg \tau_{cd} \), we obtain

\[
P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_0^T dt_2 e^{-i \omega_0 (t_1 - t_2)} \times \left\{ \langle \sigma_+ (t_1) \sigma_- (t_2) \rangle \right\},
\]

where \( \bar{I} = \int_0^\infty d\tau S(\tau) \propto g g^D r_{cd} \). Rewriting Eq. (31)

\[
P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_0^T dt_2 e^{-i \omega_0 (t_1 - t_2)} \times \left\{ \text{Tr} \{ \sigma_- (t_2 - t_1) \rho(t_1) \sigma_+ \} \right\},
\]

here we have used the cyclic property of trace. We apply change of variables in the above equation, \( x = t_2 - t_1 \), to write

\[
P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_{-t_1}^{T-t_1} dx \ e^{i \omega_0 x} \times \left\{ \text{Tr} \{ \sigma_- (x) \rho(t_1) \sigma_+ \} \right\}.
\]

Here, we will work in the regime where the observation time \( T \) is much larger than the system evolution time \( T_{sys} \). In this limit, we take \( T - t_1 \to \infty \) and \( -t_1 \to -\infty \), therefore

\[
P(\omega_0, T) \approx |\bar{I}|^2 \int_0^T dt_1 \int_{-\infty}^{\infty} dx \ e^{i \omega_0 x} \text{Tr} \{ \sigma_- (x) \rho(t_1) \sigma_+ \}.
\]

The contribution of the \( x \)-integration from the boundaries is small in the parameter \( \frac{T - t_1}{\tau_{cd}} \to 0 \)

\[
P(\omega_0, T) \approx |\bar{I}|^2 \int_{-\infty}^{\infty} dx \ e^{i \omega_0 x} \text{Tr} \{ \sigma_- (x) \int_0^T dt_1 \rho(t_1) \sigma_+ \} = T |\bar{I}|^2 \int_{-\infty}^{\infty} dx \ e^{i \omega_0 x} \text{Tr} \{ \sigma_- (x) \rho_T \sigma_+ \}.
\]

where the time-averaged density matrix is

\[
\rho_T = \frac{1}{T} \int_0^T dt_1 \rho(t_1).
\]

4.1 Fluorescence spectrum in the stationary regime

The fluorescence spectrum \( F(\omega_0) \) in terms of the two-time correlation function of system observables and in the stationary limit has the form

\[
F(\omega_0) = \lim_{T \to \infty} \frac{1}{T} P(\omega_0, T) = \lim_{T \to \infty} \frac{1}{T} |\bar{I}|^2 |\tilde{f}_s|^2 \int_{-\infty}^{\infty} dt \ e^{i \omega_0 t} \text{Tr} \{ \sigma_- (t) \bar{\rho} \sigma_+ \}
\]

\[
= \frac{|S(0)|^2}{2} \int_{-\infty}^{\infty} dt \ e^{i \omega_0 t} \langle \sigma_- (t) \sigma_+ \rangle
\]

(37)

where \( \bar{\rho} = \lim_{T \to \infty} \rho_T \). Taking the complex conjugate of the expression \( F(\omega_0) \) to obtain

\[
F^*(\omega_0) = |S(0)|^2 \int_{-\infty}^{\infty} dx \ e^{-i \omega_0 x} \langle \sigma_- (x) \sigma_+ \rangle^\dagger,
\]

(38)
where
\[(\sigma_- x \sigma_+)^1 = (\sigma_- \sigma_+),\] (39)
and therefore,
\[F^*(\omega_0) = |S(0)|^2 \int_{-\infty}^{\infty} dx e^{-i\omega_0 x} (\sigma_- \sigma_+).\] (40)
Performing a change of variables \(x = -t\) and for stationary conditions, we have
\[F^*(\omega_0) = |S(0)|^2 \int_{-\infty}^{\infty} dt e^{i\omega_0 t} (\sigma_- \sigma_+).\] (41)
By comparing the equations (41) and (37) one can see that \(F^*(\omega_0) = F(\omega_0)\), which means that \(F(\omega_0)\) is a real function. The Fourier transform appearing in Eq. (37) can then be written as a Laplace transform
\[F(\omega_0) = |S^*(0)|^2 2\text{Re} \int_{0}^{\infty} dt e^{i\omega_0 t} \text{Tr}\{\sigma_- \rho \sigma_+\},\] (42)
where we used the cyclicity of trace in the last step, a factor of two comes from the change in the interval of integration and the operator \(\Omega(t)\) is given by the expression\(^1\)
\[\Omega(t) = e^{-iHt} \rho \sigma_+ e^{iHt}.\] (43)
The above operator contains all the information needed for the fluorescence spectrum but it is defined in the Hilbert space of the entire world, i.e. the system and the reservoir. In a sense it contains too much information so we would like to find a new operator defined only in the system Hilbert space. For this purpose, we introduce a projection method which will be discussed in the next chapter.

5 Nakajima-Zwanzig generalized master equation

In order to compute the spectrum given by equation (42), we write the equation for the dynamics of the operator \(\Omega(t)\) and find its Laplace transform. Equation (42) is analogous to the expression for the single time expectation value,
\[\langle \sigma_- (t) \rangle = \text{Tr}\{\sigma_- \rho(t)\},\] (44)
with \(\rho(t)\) replaced by \(\Omega(t)\) (Swain, 1981). Thus, we first write an equation of motion for the dynamics of the reduced density matrix, then the equation for the dynamics of \(\Omega(t)\) can be derived in the same way.

The radiation field and system are decoupled for times \(t < t_0\) and are prepared independently in the states described by density matrices \(\rho_R(0)\) and \(\rho_S(0)\), respectively. At time \(t = t_0\), when the radiation field and system are brought into contact, the state of the entire system is described by the full density matrix \(\rho(t_0)\):
\[\rho(t_0) = \rho_S(t_0) \otimes \rho_R(t_0).\] (45)
\(^1\) Since \(\sigma_+\) and \(\sigma_-\) are operators in the system Hilbert space and \([H_D, H] = 0\), the evolution of \(\Omega(t)\) is determined by the Hamiltonian of the emitting system and radiation field, \(H\), in the absence of the detector.
To evaluate the dynamics of the reduced density operator, we introduce a projection superoperator \( P \), defined by its action on an arbitrary operator \( \mathcal{O} \): \( \mathcal{O} P = \rho_R(t_0) \text{Tr}_R \mathcal{O} \). \( P \) is chosen to preserve all system expectation values:

\[
\langle \mathcal{O}_S \rangle(t) = \text{Tr}\{ \mathcal{O}_S \rho(t) \} = \text{Tr}\{ \mathcal{O}_S P \rho(t) \} = \text{Tr}_S\{ \mathcal{O}_S \rho_S(t) \} \text{Tr}_R \mathcal{O} \rho_R(t) = \text{Tr}_S\{ \mathcal{O}_S \rho_S(t) \} \text{Tr}_R \mathcal{O} \rho_R(t_0) = \text{Tr}_S\{ \mathcal{O}_S \rho_S(t_0) \} \]

and satisfies \( P^2 = P \). For factorized initial conditions [Eq. (45)], \( P \rho(t_0) = \rho(t_0) \), which is a sufficient condition to rewrite the von-Neumann equation

\[
\dot{\rho}(t) = -i[H, \rho(t)] = -iL \rho(t)
\]

in the form of the exact Nakajima-Zwanzig generalized master equation (GME) (Fick and Sauermann, 1990), where \( L \) is the full Liouvillian, defined as \( L_\alpha \mathcal{O} = [H_\alpha, \mathcal{O}] \) and \( \alpha = S, R, 0, SR \). Multiplying equation (48) by \( P \) on both sides, we get

\[
P \dot{\rho}(t) = -iPL \rho(t).
\]

Introducing the complement of \( P \): \( Q = 1 - P \) and using \( P + Q = 1 \) we obtain

\[
P \dot{\rho}(t) = -iPLP \rho(t) - iPLQ \rho(t).
\]

To write the above equation in terms of \( P \rho(t) \) alone, we multiply equation (48) by \( Q \), to get

\[
Q \dot{\rho}(t) = -iQLP \rho(t) - iQLQ \rho(t).
\]

We solve the above equation for \( Q \rho(t) \) using the separation of variables method

\[
Q \dot{\rho}(t) + iQLQ \rho(t) = -iQLP \rho(t).
\]

Multiplying the above equation by \( e^{iQLt} \) on both sides gives

\[
e^{iQLt} Q \dot{\rho}(t) + i e^{iQLt} QLQ \rho(t) = -i e^{iQLt} QLP \rho(t) \quad \frac{d}{dt} e^{iQLt} Q \rho(t) = -i e^{iQLt} QLP \rho(t).
\]

After integrating the above equation and assuming \( Q \rho(t_0) = 0 \), i.e., assuming that the so-called final part of \( \rho(t_0) \) is zero, we obtain

\[
Q \rho(t) = -i \int_{t_0}^t dt' e^{-iQL(t-t')} QLP \rho(t').
\]

Substituting equation (54) into equation (50), we obtain the standard form of the Nakajima-Zwanzig generalized master equation (Fick and Sauermann, 1990)

\[
P \dot{\rho}(t) = -iPLP \rho(t) - i \int_{t_0}^t dt' \Sigma(t-t') P \rho(t'),
\]

\[
\Sigma(t) = -iPLQ e^{-iQLt} QLP.
\]
where $\Sigma(t)$ is the self-energy superoperator. We can derive an equation of motion for $\Omega(t)$ analogous to the equation for $\rho(t)$ [Eq. (55)]. However, an additional term appears because $Q\Omega(0) \neq Q\bar{\rho}\sigma_+ \neq 0$, see Eq. (54). The resulting GME for $P\Omega(t)$ is then

$$P\Omega(t) = -iP\Omega(t) - i \int_0^t dt' \Sigma(t-t')\Omega(t') - iPLQe^{-iQLt}Q\Omega(0),$$

where $\Sigma(t)$ is defined in Eq. (56) and the last term in the above equation contains $Q\Omega(0)$, i.e. the final part of $\Omega(0)$ is non-zero. This last term accounts for conditions that accumulate between the system and radiation modes in the time interval $t \in [t_0, 0]$ for $t_0 < 0$. The long-time average value, is defined as

$$\bar{\rho} = \lim_{T \to \infty} \frac{1}{T} \int_0^T dt \rho(t) = \lim_{s \to 0} s\rho(s).$$

Here, the Laplace transform is defined as $F(s) = \int_0^\infty dt e^{-st} f(t)$. Inserting this definition into Eq. (54), we find (assuming that $\frac{1}{0^++iQL}$ exists),

$$Q\bar{\rho} = -\frac{1}{0^++iQL}QLP\bar{\rho}.$$

Here, $0^+$ is a positive infinitesimal. We want to write $Q\Omega(0) = Q\bar{\rho}\sigma_+$ in terms of $P\bar{\rho}\sigma_+$. To this end, substituting for $Q\bar{\rho}$ in equation (57), we have

$$P\Omega(t) = -iP\Omega(t) - i \int_0^t dt' \Sigma(t-t')\Omega(t') - PLQe^{-iQLt}\left(\frac{1}{0^++iQL}\right)QLP\bar{\rho}\sigma_+.$$

The above equation is identical to equation (55) except for the last part, the so-called final part, which is expressed as

$$\Phi(t) = -PLQe^{-iQLt}\left(\frac{1}{0^++iQL}\right)QLP\bar{\rho}\sigma_+.$$

The expression for the fluorescence spectrum given in equation (42) is written in terms of the trace of the system operator $\sigma_-$ as $\text{Tr}\{\sigma_- \Omega(t)\}$.

Using the properties of the projection superoperator, we have

$$\text{Tr}\{\sigma_- \Omega(t)\} = \text{Tr}\{\sigma_- P\Omega(t)\} = \text{Tr}_S\{\sigma_- \Omega_S(t)\},$$

where in the last step we have defined

$$\Omega_S(t) = \text{Tr}_R\{\Omega(t)\}.$$

Therefore, the expression for the fluorescence spectrum given in Eq. (42) takes the form

$$F(\omega_0) = 2|S^*(0)|^2 \text{Re} \int_0^\infty dt e^{i\omega_0 t} \text{Tr}_S\{\sigma_- \Omega_S(t)\}.$$ 

For completeness, we substitute the expression for the Laplace transform $P\Omega(s)$ in the equation for the fluorescence spectrum to obtain an expression for the lineshape function as

$$F(\omega_0) = 2|S^*(0)|^2 \text{Re} \left[ \text{Tr}\left\{\sigma_- \frac{1}{s+iPLP + i\Sigma(s)}(\mathbb{1} + \Phi(s)) P\bar{\rho}\sigma_+\right\}\right]_{s=-i\omega_0}$$

where the Laplace transforms of the self-energy and the final part are given, respectively, below

$$\Sigma(s) = -iPLQ\frac{1}{-i\omega_0 + iQL}QLP$$

(66)
\[ \Phi(s = -i\omega_0) = -PLQ \left( \frac{1}{-\omega_0 + iQL} \right) \left( \frac{1}{0^+ + iQL} \right) QLP, \]  
\[ \bar{\rho} = \lim_{s \to 0} s - iPLP + i\Sigma(s) \rho(t_0). \]

We have derived a formula which is valid for studying the fluorescence spectrum of a general system undergoing non-Markovian dynamics.

6 Results and discussion

In this section, we analyze and discuss the results obtained in the previous sections. The self-energy and final terms given by Eqs. (66) and (67), respectively can be expanded to the all powers of perturbation/interaction (e.g. nuclear spins, phonons etc) present in the problem. Similarly, the stationary density matrix in Eq. (68) can be found after expanding the self-energy superoperator in the powers of perturbation Liouvillian. After solving for a specific interaction, the Eq. (65) will give rise to a final expression for the lineshape function of a multi-level system within non-Markovian interaction (Kumar, 2018).

7 Conclusion

In the present paper, we have given a general analytical formula for the dynamics (fluorescence lineshape) of a multi-level system interacting with its environment via a non-Markovian interaction. We have also shown that quantum regression theorem can not be used to describe the dynamics of two-time correlation because of a non-zero final term. The self-energy and final term superoperators can be expanded in all powers of perturbation without applying the Born-approximation in terms of coupling to the environment. We have tried to keep the formulae as general as possible and not made any assumptions about the system or the environment, so that this theory can be applied to any quantum-optical system (not limited to a two-level system) or the environment (nuclear spins or phonons). Furthermore, for a Markovian type interaction above formulae can be reduced and used to study the systems with Markovian interactions and vanishing final term.

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