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# A thermal bath induced new resonance on the linear and nonlinear spectra of a two-level system

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#### Abstract

Within the scope of generalized master equation we have shown the effect of bath with finite band width on the linear absorption and on the resonance fluorescence and absorption spectra of a driven two-state system. It is found that depending on the band width and temperature of the bath a reversible dynamics may set in even in the case of linear absorption induced by weak field. At higher temperature the bath induces new resonance on the spectral profiles. © 1999 Elsevier Science B.V. All rights reserved.

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# 1. Introduction

The study of dissipative dynamics of quantum systems within the framework of system-heat bath models has drawn wide attention for the last few decades [1–4]. Although the major works so far done in this direction are confined in the domain of weak coupling of the system and the bath within the Markov approximation, recent experiments involving ultrafast time scale [5] and correlated laser pulse with adjustable memory time [6], have given tremendous impetus to the study of system-heat bath model beyond the Markov approximation. The issue and relevance of non-Markovian relaxation processes have been addressed at length by many researchers. For instance, a cumulant expansion scheme [7] was considered by Villaeys et al. [8] to explain non-Lorentzian nature of optical absorption for the femtosecond transient processes. Gangopadhyay and Ray [9] constructed a system-operator concerned non-Markovian master equation for linear and nonlinear quantum optical model system in the weak coupling regime. Lewenstein et al. [10] observed a strong narrowing of the resonance fluorescence in a cavity with increasing driving field strength which they interpreted this as a dynamical decoupling of the atom from the vacuum field. A few other works [11] also deal with the effect of radiation–matter interaction in going beyond Markov approximation, where the memory time  $\tau$  is small but of the order of the decay time,  $\gamma^{-1}$ . Very recently, Brinati et al. [12] have found a non-Markovian effect at finite

temperature on the absorption lineshape function of a driven two-level system in the steady state regime. They have observed that the steady state absorption lineshape splits into twin peaks with increase in temperature.

An early thorough investigation on nonlinear optical phenomena with finite memory effect has been performed by Tsuensugu and Hanamura [13] by using Gaussian-Markovian frequency modulation of the two-level system. Using a Green's function approach, they have found the effect of memory on the coherent optical transient, emission and absorption spectra of the driven two-level system. Their studies were based on the stochastic Liouville equation which assumed a stochastic random modulation of the system frequency. Tanimura and Kubo [14] have shown that one can derive this equation of motion based on the system—bath Hamiltonian, which makes it possible to include temperature effects into the dynamics of the total system. Then Tanimura and Mukamel [15] further have generalized the equation of motion to study a system which consists of a two-level displaced harmonic oscillators system coupled to a heat bath and have studied the time dependent Stark effects.

Very recently the tremendous growth of experimental sophistication has revealed interesting behaviour in the decay of the excited atom confined in a cavity resonator or an optical band gap material whose modal density can be controlled and thus the rate and direction of the spontaneous emission can be designed appropriately [16]. Lewenstein et al. [10] studied the fate of the spontaneous emission of a coherently driven atom in a cavity considering a finite response time of the environment. This effect which is connected with non-Markovian features, is tested experimentally [17] quite recently, and interpreted theoretically [18] as a consequence of the renormalization of the cavity induced spontaneous emission by the strong driving field. In the same line Keitel et al. [19] have studied linewidth of the resonance fluorescence spectra in terms of the decay rate of the dressed coherencies and have predicted the sensitive influence of the density of modes with the possibility of suppression and enhancement of the fluorescence.

In this work we have shown the dynamical effect due to the bath with finite band width on the spontaneous emission of two state system both in the presence and absence of a strong driving field. In presence of an external driving field the evolution of the system with memory can lead the system to a different steady state where one can find the signature of the memory effect. However, a two-state system in a pure state will reach the thermal equilibrium in the long time limit by the effect of this bath despite of the fact that the dynamical evolution of the system will be different from that of the case with a broad band bath. By constructing an appropriate modified Bloch equation we have shown that if the modal density in the vicinity of the system frequency of interest is less than that of the vacuum space, the decay of the excited state will be retarded and if it is greater it will be accelerated. So with a prototype finite band width of modal density of the bath, we have studied the fate of the spontaneous emission from the two-level system in presence and in absence of a strong driving field. For this purpose we have studied the linear absorption by a transient weak light. We have also shown the effect of bath characteristics on the resonance fluorescence spectrum [20]a and probe absorption [20]b following a strong pump.

Although the spectroscopic studies on dissipative systems are done very extensively in the context of gas phase experiments, there are still many open questions to be answered if the optical system resides on the host material in condensed phases [21]. Then the system might be induced by various kind of noise processes, from very short to very long correlation time which can be mimicked by a colored bath. The system can reside on the appropriate material where the environment can be designed with our choice of control [16] over the spontaneous emission. In this model study we would like to understand the following points: (i) How the effect of band shape of the bath comes into play on the spectra in the linear and nonlinear regime of field strength. (ii) How the position or band center of the bath affect in the linear absorption. Thus one can find the origin of spectral shift and non-Lorentzian linear absorption which are commonly observed in the solution phase or in the bath with finite band width affect on the linear absorption and also in the strong field induced cases. (iv) To obtain the strong field induced effects we have made a critical comment on the density matrix theory of damping for a finite band width of the bath and on the applicability of Regression theorem.

The rest of the paper is organized as follows. In Section 2 we have constructed the master equation considering a finite bandwidth of the reservoir and the corresponding modified Bloch equation is presented. Section 3 is devoted to obtain the transient linear susceptibility. In Section 4 we have calculated the resonance fluorescence spectrum and weak field induced probe absorption of the driven system by the strong field. The paper is concluded in Section 5.

#### 2. Generalized master equation and modified Bloch equation

Let us consider a system described by a Hamiltonian  $H_s$ . We assume the reservoir Hamiltonian is  $H_R$  and the interaction between system and reservoir is V. So the total Hamiltonian can be written as

$$H_{\rm T} = H_{\rm s} + H_{\rm R} + V \equiv H_0 + V. \tag{2.1}$$

Generally, we write V as a sum of the products of the form

$$V = \hbar \sum_{i} Q_i F_i, \tag{2.2}$$

where  $Q_i$  and  $F_i$  are the system and reservoir operators, respectively, in the Schrödinger picture (S.P.).

The joint density operator  $\kappa$  for the system and the reservoir obeys the Liouville-von Neumann equation in the interaction picture (I.P.) as

$$\frac{\partial \kappa}{\partial t} = -\frac{i}{\hbar} [V(t), \kappa].$$
(2.3)

Following the standard procedure [4,22] of averaging over the bath variables, we obtain the equation of motion of the reduced density of the system in S.P. as

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\frac{i}{\hbar} \left[ H_{\mathrm{s}}, \rho \right] - \sum_{i,j} \int_{0}^{t} \mathrm{d}t' \left\{ \left[ Q_{i} Q_{j}' \rho'(t') - Q_{i}' \rho'(t') Q_{i} \right] \langle F_{i} F_{j}' \rangle \right\}_{\mathrm{R}} - \left[ Q_{i} \rho'(t') Q_{j}' - \rho'(t') Q_{j}' Q_{i} \right] \langle F_{j}' F_{i} \rangle_{\mathrm{R}} \right\}$$
(2.4)

where

$$\left(Q_{j}\rho(t')\right)' = \exp\left[-\frac{i}{\hbar}H_{s}(t-t')\right]Q_{j}\rho(t')\exp\left[\frac{i}{\hbar}H_{s}(t-t')\right],$$
(2.5)

$$\left(F_{j}\right)' = \exp\left[-\frac{i}{\hbar}H_{R}(t-t')\right]F_{j}\exp\left[\frac{i}{\hbar}H_{R}(t-t')\right],$$
(2.6)

where we have made only one approximation that the joint density is always factorizable, i.e,

$$\kappa(t) = s(t)f(t), \tag{2.7}$$

where s(t) and f(t) are the system and bath density operators respectively in I.P. and then we assume the bath is in thermal equilibrium at all time. Eq. (2.4) is the generalized master equation derived by many authors using different techniques [22].

In what follows we consider a two-level system coupled to a reservoir in thermal equilibrium at some temperature T, and driven by a classical monochromatic light field. The system Hamiltonian is expressed by the Pauli operators as

$$H_{\rm s} = \frac{\hbar\,\omega_0}{2}\,\sigma_z.\tag{2.8}$$

The heat bath Hamiltonian and the system-bath interaction are given by

$$H_{\rm R} = \sum_{j} \hbar \,\omega_j b_j^{\dagger} b_j, \tag{2.9}$$

$$V = \hbar \sum_{j} g_{j} \Big( \sigma_{+} b_{j} + \sigma_{-} b_{j}^{\dagger} \Big), \qquad (2.10)$$

respectively, where  $b_j$  ( $b_j^{\dagger}$ ) is the annihilation (creation) operator of the bath mode with frequency  $\omega_j$  and  $g_j$  is the coupling strength between system and the bath modes. We assume the laser field interaction with the amplitude  $E_0$  and frequency  $\omega$  in the rotating wave approximation (RWA) form as

$$V_{\text{ext}}(t) = \hbar \left[ E_0 \mathrm{e}^{-i\omega t} \sigma_+ + E_0 \mathrm{e}^{i\omega t} \sigma_- \right].$$
(2.11)

So the reduced density operator equation of motion becomes,

$$\frac{d\rho}{dt} = -\frac{i\omega_{0}}{2} [\sigma_{z},\rho] - i [E_{0}e^{-i\omega t}\sigma_{+} + E_{0}e^{i\omega t}\sigma_{-},\rho] 
- \sum_{i,j} \int_{0}^{t} dt' \left\langle \left[\sigma_{+}\sigma_{-}\rho'(t') - \sigma_{-}\rho'(t')\sigma_{+}\right] \left\langle \sum_{j} |g_{j}|^{2}b_{j}b_{j}^{\dagger}e^{i(\omega_{0}-\omega_{j})(t-t')} \right\rangle_{R} 
- \left[\sigma_{+}\rho'(t')\sigma_{-}-\rho'(t')\sigma_{-}\sigma_{+}\right] \left\langle \sum_{j} |g_{j}|^{2}b_{j}^{\dagger}b_{j}e^{i(\omega_{0}-\omega_{j})(t-t')} \right\rangle_{R} 
+ \left[\sigma_{-}\sigma_{+}\rho'(t') - \sigma_{+}\rho'(t')\sigma_{-}\right] \left\langle \sum_{j} |g_{j}|^{2}b_{j}^{\dagger}b_{j}e^{-i(\omega_{0}-\omega_{j})(t-t')} \right\rangle_{R} 
- \left[\sigma_{-}\rho'(t')\sigma_{+}-\rho'(t')\sigma_{+}\sigma_{-}\right] \left\langle \sum_{j} |g_{j}|^{2}b_{j}b_{j}^{\dagger}e^{-i(\omega_{0}-\omega_{j})(t-t')} \right\rangle_{R} \right\}$$
(2.12)

where

$$\rho'(t') = e^{-i\omega_0\sigma_z(t-t')}\rho(t')e^{i\omega_0\sigma_z(t-t')}.$$
(2.13)

The Bloch equations then can be obtained as

$$\langle \dot{\sigma}_{+} \rangle = i \omega_0 \langle \sigma_{+} \rangle - i E^*(t) \langle \sigma_z \rangle - \int_0^t \mathrm{d}t' \langle \sigma_{+}(t') \rangle Q_{12}(t-t') \mathrm{e}^{i \omega_0(t-t')}, \qquad (2.14a)$$

$$\langle \dot{\sigma}_{z} \rangle = -2i \left[ E(t) \langle \sigma_{+} \rangle - E^{*}(t) \langle \sigma_{-} \rangle \right] - \int_{0}^{t} \mathrm{d}t' \left[ Q_{0}(t-t') + \langle \sigma_{z}(t') \rangle Q_{z}(t-t') \right], \tag{2.14b}$$

where

$$\begin{aligned} Q_{12}(t-t') &= \sum_{j} |g_{j}|^{2} (1+2\bar{n}(\omega_{j})) e^{i\omega_{j}(t-t')}, \\ Q_{z}(t-t') &= 2\sum_{j} |g_{j}|^{2} (1+2\bar{n}(\omega_{j})) \cos((\omega_{0}-\omega_{j})(t-t')), \\ Q_{0}(t-t') &= 2\sum_{j} |g_{j}|^{2} \cos((\omega_{0}-\omega_{j})(t-t')), \end{aligned}$$

with

$$\bar{n}(\omega_j) = \frac{1}{e^{\hbar \omega_j / kT} - 1}.$$
(2.15)

Now invoking a slowly varying envelope approximation,  $\langle \sigma_+(t) \rangle = S_+(t)e^{i\omega t}$ ,  $E(t) = \frac{E_0}{2}e^{i\omega t}$  and  $\langle \sigma_z(t) \rangle = S_+(t)$  one finds

$$\dot{S}_{+}(t) = i\Delta S_{+}(t) + \frac{i}{2}E_{0}S_{z}(t) - \int_{0}^{t}S_{+}(t')Q_{12}'(t-t')\,\mathrm{d}t', \qquad (2.16a)$$

and

$$\dot{S}_{z}(t) = iE_{0}[S_{+}(t) - S_{-}(t)] - \int_{0}^{t} [Q_{0}(t-t') + S_{z}(t')Q_{z}(t-t')] dt', \qquad (2.16b)$$

where

$$Q'_{12}(t-t') = \sum_{j} |g_{j}|^{2} (1+2\bar{n}(\omega_{j})) e^{-i(\omega-\omega_{j})(t-t')}$$
(2.17)

with  $\Delta = \omega_0 - \omega$ .

As we have not employed Markov approximation thus we need to supply the band shape function of the bath to carry out the integration in Eq. (2.16). In the literature [7–11], most of the workers have introduced the memory effect through the kernels  $Q_{+,-,z}(t)$  as proportional to some time dependent functions. For example, in Refs. [8,10,11] the kernels are taken as of the form  $e^{-|(t-t')|/\tau}$ . But in this way of introducing time dependent memory kernels lacks the knowledge of temperature dependence of the non-Markovian decay rate. An equivalent way of choosing exponential time decay in the bath correlation function is to use a distribution function  $P(\omega')$  with a finite width. We use Lorentzian distribution function of  $P(\omega')$  as

$$P(\omega') = \frac{P_0}{1 + (\omega' - \omega_{\rm B})^2 \tau^2},$$
(2.18)

where  $\tau$  is the characteristic time of the reservoir or the inverse of the reservoir bandwidth and  $\omega_B$  is the center frequency of the band.  $\tau$  is normally of the order of  $\gamma^{-1}$  but  $\gamma\tau$  is always less than 1.  $P_0$  is a dimensionless constant quantity. The bandshape function we have chosen here is a prototype shape function for finite width of the vacuum band and thus will be suffice to obtain the qualitative idea about the effect of any finite band width of the bath. As any arbitrary distribution function may not satisfy the physical requirement of applying the regression theorem, this function guarantees the attainment of thermal equilibrium in the long time limit of the initial pure two-state system and also the problem can be handled analytically.

Using the bath distribution function we can calculate the following quantities

$$Q_{12}'(t-t') = \frac{\gamma}{2\tau} (1+2\bar{n}(\omega_0)) e^{(-i\Delta_b - \frac{1}{\tau})(t-t')}, \qquad (2.19)$$

$$Q_{z}(t-t') = \frac{\gamma}{\tau} (1+2\bar{n}(\omega_{0})) \cos(\Delta_{c}(t-t')) e^{-\frac{t-t'}{\tau}}$$
(2.20)

and

$$A(t) \equiv \int_{0}^{t} Q_{0}(t-t') dt' = \gamma \frac{1}{\Delta_{c}^{2} \tau^{2} + 1} \left[ 1 - e^{-t/\tau} (\cos \Delta_{c} t - \Delta_{c} \tau \sin \Delta_{c} t) \right],$$
(2.21)

where  $\Delta_{\rm b} = \omega - \omega_{\rm B}$  and  $\Delta_{\rm c} = \Delta + \Delta_{\rm b}$  and  $\gamma = 2\pi |g(\omega_0)|^2 P_0$ .

Thus the modified Bloch equation reduces to

$$\dot{S}_{+}(t) = i\Delta S_{+}(t) + \frac{i}{2}E_{0}S_{z}(t) - \frac{\gamma}{2\tau}(1 + 2\bar{n}(\omega_{0}))\int_{0}^{t}S_{+}(t')e^{(-i\Delta_{b} - \frac{1}{\tau})(t-t')},$$
(2.22a)

and

$$\dot{S}_{z}(t) = iE_{0}[S_{+}(t) - S_{-}(t)] - A(t) - \frac{\gamma}{\tau}(1 + 2\bar{n}(\omega_{0}))\int_{0}^{t}S_{z}(t')\cos[\Delta_{c}(t-t')]e^{-\frac{t-t'}{\tau}}dt'.$$
 (2.22b)

Thus we find that at the Markovian limit i.e, when  $\tau \rightarrow 0$ , we recover the usual Bloch equation. For example, one can show that

$$\lim_{\tau \to 0} \int_0^t S_i(t') \frac{1}{\tau} e^{-(t-t')/\tau} dt' = S_i(t)$$
(2.23)

and  $\lim_{\tau \to 0} A(t) = \gamma$  where  $S_i$  is any one of  $S_+$ ,  $S_-$  or  $S_z$ .

The modified Bloch equation as given in Eq. (2.22) is an important result in this work because depending on the band center, width and temperature of the bath the system dynamics can change immensely from the conventional Bloch equation and can lead to different steady state. Note that to derive Eqs. (2.22) we have considered  $\overline{n}(\omega') = \overline{n}(\omega_0)$  in order to obtain the qualitative results of non-Markovian effects. A more accurate estimation can be given with a Taylor series expansion of  $\overline{n}(\omega')$  around  $\overline{n}(\omega_0)$ . But for the rest of the paper we consider the usual approximation of  $\overline{n}(\omega') = \overline{n}(\omega_0)$ .

# 3. Transient absorption lineshape

In the ultrafast dephasing processes observed experimentally [5] take place on a time scale where the system evolves with memory and thus a delta correlated bath, often used theoretically, is no longer valid. The non-Lorentzian absorption profile [8], obtained experimentally in the solution phase or in the condensed phase, are fitted normally with some memory function, but these methods are not always physically transparent. Here in the following discussion we will be able to find the physical origin of the spectral shift and asymmetry along with the fact that at higher temperature how the new resonances appear due to finite band width of the bath even in the case of linear absorption. Our aim here is to calculate the transient susceptibility which is again related to the polarization in the linear response term as follows:

$$P(t) = \int_0^t \chi(t - t') E(t') dt'.$$
(3.1)

Microscopically P(t) is defined as

$$P(t) = \text{Tr}[\rho(t)\mu] \equiv \rho_{21}\mu_{12} + \rho_{12}\mu_{21}, \qquad (3.2)$$

where  $\mu$  is the transition dipole matrix element.

Taking the Laplace transform of Eq. (2.22a) one obtains

$$\bar{S}_{+}(p) = \left[S_{+}(0) + \frac{iE_{0}}{2}\bar{S}_{z}\right] \left[p - i\Delta + \frac{\gamma(1+2\bar{n})}{2(p\tau + i\Delta_{\rm B}\tau + 1)}\right]^{-1}$$
(3.3)

where

$$\overline{S}_{+}(p) = \int_{0}^{\infty} \mathrm{e}^{-pt} S_{+}(t) \,\mathrm{d}t$$

Assuming initially the system is in the ground state and thus we obtain,

$$S_{+}(t) = \frac{iE_{0}}{2}L^{-1} \left[ \frac{\bar{S}_{z}(p)}{p - i\Delta + \gamma(1 + 2\bar{n})/2(p\tau - i\Delta_{\rm B}\tau + 1)} \right]$$
(3.4)

and thus we find

$$\langle \sigma_{+}(t) \rangle = \frac{iE_{0}}{2} \mathrm{e}^{i\omega t} \int_{0}^{t} (A \mathrm{e}^{\lambda_{1}t'} + A' \mathrm{e}^{\lambda_{2}t'}) \langle \sigma_{z}(t-t') \rangle \mathrm{d}t'.$$
(3.5)

Therefore, the imaginary part of the susceptibility is given by

$$\chi''(\omega) = -\frac{|\mu_{21}|^2}{3\hbar} \operatorname{Re}\left(\int_0^t (Ae^{\lambda_1 t'} + A'e^{\lambda_2 t'}) \langle \sigma_z(t-t') \rangle dt'\right), \tag{3.6}$$

where

$$A = \frac{\lambda_1 + i\,\varDelta_{\rm B} + \frac{1}{\tau}}{\lambda_1 - \lambda_2}\,, \quad A' = \frac{\lambda_2 + i\,\varDelta_{\rm B} + \frac{1}{\tau}}{\lambda_2 - \lambda_1}\,,$$

and the factor  $\frac{1}{3}$  accounts for the isotropic absorption.

Assuming the probe absorption is sufficiently weak and transient we consider  $\langle \sigma_z(t-t') \rangle = -1$  and thus we obtain

$$\chi''(\omega) = -\frac{|\mu_{21}|^2}{3\hbar} \operatorname{Re}\left\{A\frac{e^{\lambda_1 t} - 1}{\lambda_1} + A'\frac{e^{\lambda_2 t} - 1}{\lambda_2}\right\},\tag{3.7}$$

where  $\lambda_{1(2)}$  are given by

$$\lambda_{1(2)} = -\frac{1}{2\tau} + \frac{i\Delta_x}{2} \pm \frac{1}{2}\sqrt{1/\tau^2 - \Delta_x^2 - 2i\Delta_x/\tau - 4\Gamma' + 4i\Delta/\tau},$$
(3.8)

with  $\Gamma' = \frac{\gamma}{2\tau}(1+2\bar{n}) + \Delta\Delta_{\rm B}$ ,  $\Delta_{\rm B} = \omega - \omega_{\rm B}$  and  $\Delta_x = \Delta - \Delta_{\rm B}$ . For time long as compared to any other time scale we find

$$\chi''(\omega) = -\frac{|\mu_{21}|^2}{3\hbar} \operatorname{Re}\left\{\frac{A}{\lambda_1} + \frac{A'}{\lambda_2}\right\}.$$
(3.9)

Finally, apart from some proportionality factor of  $\chi''(\omega)$ , the bandshape function can be given by

$$B = \frac{\frac{\gamma}{2}(1+2\bar{n})}{\Delta^{4}\tau^{2} - 2\,\Delta^{3}\tau^{2}\Delta_{d} + \Delta^{2}\left(1+\Delta_{d}^{2}\tau^{2} - \frac{\gamma\tau}{2}(1+2\bar{n})\right) + \Delta\gamma(1+2\bar{n})\,\Delta_{d}\tau + \frac{\gamma^{2}}{4}(1+2\bar{n})^{2}},\qquad(3.10)$$

where

$$\Delta_{\rm d} = \omega_0 - \omega_{\rm B}. \tag{3.11}$$

For  $\Delta_{\rm d} = 0$ , one obtains,

$$B = \frac{\frac{\gamma}{2}(1+2\bar{n})}{\Delta^4 \tau^2 + \Delta^2 \left(1 - \frac{\gamma\tau}{2}(1+2\bar{n})\right) + \frac{\gamma^2}{4}(1+2\bar{n})^2},$$
(3.12)



Fig. 1. (a) Variation of bandshape function  $B\gamma/\text{modal}$  density of the bath  $P(\omega - \omega_0)$  is shown against detuning  $\Delta/\gamma \equiv (\omega - \omega_0)/\gamma$  with  $\bar{n} = 0$ ,  $\Delta_d/\gamma = 0$  for two different values of  $k = \tau\gamma = 0.0$ , 0.5. (Both scales are arbitrary). (b) Same as in (a) with  $\bar{n} = 0$ ,  $k = \tau\gamma = 0.4$  for different values of  $\Delta_d/\gamma = -1.0$ , 0.0 and 1.0. (c) Same as in (a) with  $\bar{n} = 2$  and  $\Delta_d/\gamma = 0$  for different values of  $k = \tau\gamma = 0.0$ , 0.2 and 0.5. (d) Same as in (a) with  $k = \tau\gamma = 0.5$  and  $\Delta_d/\gamma = 0$  for different values of  $\bar{n} = 0.0$ , 0.5 and 2.0. (e) Same as in (a) with  $\bar{n} = 2.0$  and  $k = \tau\gamma = 0.5$  for different values of  $\Delta_d/\gamma = -1.0$ , 0.0 and 1.0.

and if  $\tau \to 0$ ,

$$B = \frac{\frac{\gamma}{2}(1+2\bar{n})}{\Delta^2 + \frac{\gamma^2}{4}(1+2\bar{n})^2}.$$
(3.13)

To analyze the absorption lineshape we have plotted the bandshape with the probe detuning  $\Delta$  for the different parameters of the vacuum band. We are considering three parameters of the bath,  $\tau$ , temperature,  $\bar{n}$ and  $\Delta_d = \omega_0 - \omega_{\rm B}$ , i.e, detuning of the center of the reservoir band from the system resonance frequency. We consider very small  $\Delta_d$  which is of the order of  $\gamma$  or less. In Fig. 1(a) we show that at zero temperature and  $\Delta_{\rm d}=0$  the absorption lineshape becomes narrower with increase in  $\tau$  but there is no shift in the peak. When  $\Delta_d \neq 0$  and for nonzero value of  $\tau$  we find a spectral shift depending on the sign of  $\Delta_d$ . This is shown in Fig. 1(b). Along with the shift we find the line shape is non-Lorentzian. This is because with the increase in detuning  $\Delta_{\rm d}$  the number density of bath modes in the vicinity of the frequency of two-level system is less than in the case of  $\tau = 0$  i.e., infinite band width of the bath. That means the system finds less bath modes to dispense energy to the bath and also to be dephased by the bath modes. Therefore, the decay rate decreases which makes the lineshape narrower and the two-state system follows the shape of the vacuum band when it is decaying and the Lorentzian absorption band becomes asymmetric. If the bath mode density near  $\omega_0$  is very small, the system will be decoupled from the bath and will show a delta function peak. In Fig. 1(c) and (d) as  $\Delta_d = 0$  there is no spectral shift. In these cases at finite band width of the bath, the absorption line shape splits into twin peaks. As in the case of linear absorption we consider only weak probe field situation, so there is no saturation unlike the case in Ref. [12]. Therefore a narrow band of vacuum with high thermal average photon number acts as an another coherent field and we can interpret this phenomenon as thermal bath induced Rabi splitting. If we move the vacuum band from the line center of the two-state system, i.e.  $\Delta_d \neq 0$  we find the splitting becomes asymmetric which is shown in Fig. 1(e). The reason of the asymmetry in splitting is as follows. The splitting means the system assumes two characteristic frequencies instead of its original resonance frequency  $\omega_0$  and these two frequencies experience different bath mode density in their neighbourhood and thus the decay rate of each individual peaks are different and are of different heights. In all Figs. 1(a-e) we have drawn the modal density of the bath for the corresponding values of  $\tau$  for comparison.

# 4. Spectra of the driven two-state system by a strong field

Here we find out the effect of the bath with finite band width on the fluorescence and absorption spectra of the driven system. For this we need to obtain the steady state quantities of the system. Here we assume the reservoir band center is on resonance with  $\omega_0$ , i.e,  $\Delta_d = 0$ . Taking the Laplace transform of Eqs. (2.22) one arrives at,

$$p\bar{S}_{+}(p) - S_{0} = \bar{S}_{+}(p) + \frac{i}{2}E_{0}\bar{S}_{z}(p) - \bar{S}_{+}(p)\bar{Q}_{+}, \qquad (4.1a)$$

$$p\bar{S}_{z}(p) - S_{z} = iE_{0}(\bar{S}_{+}(p) - \bar{S}_{-}(p)) - \gamma\left(\frac{1}{p} - \frac{1}{p+1/\tau}\right) - \bar{S}_{z}(p)\bar{Q}_{z},$$
(4.1b)

where

$$\overline{Q}_{+}(p) = \int_{0}^{\infty} e^{-pt} Q'_{12}(t) dt, \quad \left(\overline{Q}_{-}(p)\right)^{*} = \overline{Q}_{+}(p)$$
(4.2)

with  $\overline{Q}_{+}(p)$  and  $\overline{Q}_{z}(p)$  being

$$\overline{Q}_{+}(p) = \frac{\gamma}{2} (1 + 2\overline{n}(\omega_0)) \frac{-i}{\Delta \tau - i(1 + p\tau)}, \qquad (4.3)$$

$$\overline{Q}_{z}(p) = 2p\gamma (1 + 2\overline{n}(\omega_{0})) \frac{1}{(1 + p\tau)}.$$
(4.4)

Thus the steady state solution of Bloch equation reduces to

$$S_{z}(t \to \infty) = -\frac{\left[\Delta(1+\Omega^{2}) + \frac{\gamma\Omega}{2}(1+2\bar{n})\right]^{2} + \frac{\gamma^{2}}{4}(1+2\bar{n})^{2}}{\frac{1}{2}E_{0}^{2}(1+2\bar{n})(1+\Omega^{2}) + (1+2\bar{n})\left\{\left[\Delta(1+\Omega^{2}) + \frac{\gamma\Omega}{2}(1+2\bar{n})\right]^{2} + \frac{\gamma^{2}}{4}(1+2\bar{n})^{2}\right\}}$$

$$(4.5)$$

and

$$S_{+}(t \to \infty) = \frac{iE_{0}/2}{-i\Delta + \frac{\gamma}{2}(1+2\bar{n})\frac{1-i\Omega}{1+\Omega^{2}}}S_{z}(t \to \infty), \qquad (4.6)$$

where  $\Omega = \Delta \tau$ .

This is interesting that the non-Markovian feature, i.e, the reservoir characteristic time  $\tau$  appears in the steady state solution of the Bloch equations. For nonzero detuning the steady state values are different from the usual broad band bath case and steady state observables will depend on  $\tau$ . The details of steady state behaviour are discussed in Ref. [12]. Now we are interested about the dynamical features due to the modified Bloch equation. At finite  $\tau$ , the decay becomes slower due to longer correlation of the heat bath noise. For this purpose we study the strong field induced resonance fluorescence [20]a and probe absorption of the driven system [20]b unlike the ordinary probe absorption to include the effect of saturation.

# 4.1. Resonance fluorescence spectrum

In order to calculate the strong field resonance fluorescence spectra [20]a  $S(\nu - \omega_0)$ , i.e.,

$$S(\nu - \omega_0) = \operatorname{Re}\left[\lim_{t \to \infty} \int_0^\infty e^{-i(\nu - \omega_0)t'} \langle \sigma_+(t + t') \sigma_-(t) \rangle \,\mathrm{d}t'\right],\tag{4.7}$$

we follow the standard prescription [20] of using regression hypothesis. The validity of regression hypothesis and a brief comment on quantum Markov process for the finite bandwidth of the reservoir is given in the Appendix. Here we assume all detuning parameters are zero, i.e,  $\Delta = 0$  and  $\Delta_d = 0$  to make life simple and without loosing any extra understanding. That means here we consider the vacuum band is centered exactly on the resonance frequency of the two-state system.

Calculating the Laplace transform of the quantity

$$G(t') = \lim_{t \to \infty} \langle \sigma_+(t+t') \sigma_-(t) \rangle, \tag{4.8}$$

we obtain

$$G(p) = L(G(t')) = \frac{f(p)}{pF(p)},$$
(4.9)

where

$$F(p) = \frac{1}{(1+p\tau)^2} \Big( \tau p^2 + p + \frac{\gamma}{2} (1+2\bar{n}) \Big) \Big[ \tau p^3 + p^2 + p \Big( E_0^2 \tau + 3\gamma (1+2\bar{n})/2 \Big) + E_0^2 \Big].$$
(4.10)

From this F(p) one can still find the poles of Mollow spectrum [20]a at  $\tau \to 0$ . But it can be approximately factorized as follows

$$F(p) = \frac{\tau^2}{(1+p\tau)^2} (p-\lambda_+)(p-\lambda_-)(p-\Lambda_0)(p-\Lambda_+)(p-\Lambda_-).$$
(4.11)

f(p) can be expressed as

$$f(p) = \frac{1}{2} \left( S_z(t \to \infty) + 1 \right) p \left\{ \left[ p + \overline{Q}_-(p) \right] \left[ p + \overline{Q}_z(0) \right] + E_0^2 / 2 \right\} - S_-(t \to \infty) \left( p + \frac{\gamma}{p\tau + 1} \right) \frac{iE_0}{2} \left( p + \overline{Q}_-(0) \right).$$
(4.12)

Now taking  $\gamma \ll E_0$ , F(p) can be factorized approximately and the roots of p for F(p) = 0, i.e.,  $\lambda_{\pm}$ ,  $\Lambda_0$  and  $\Lambda_+$  can be given as

$$\lambda_{\pm} = -\frac{1}{2\tau} \Big[ 1 \mp \{ 1 - 2k(1 + 2\bar{n}) \}^{1/2} \Big], \quad \Lambda_0 = -\frac{1}{\tau} + E_0^2 \tau + \frac{3}{2} \gamma (1 + 2\bar{n})$$

and

$$\Lambda_{\pm} = a \pm ib \tag{4.13}$$

where

$$a = -\frac{1}{2} \left( E_0^2 \tau + \frac{3}{2} \gamma (1 + 2\bar{n}) \right), \quad b = \sqrt{E_0^2 - a^2}$$

and  $k = \gamma \tau$ . Here  $\lambda_{\pm}$  are exact roots but  $\Lambda_0$  and  $\Lambda_{\pm}$  are approximate roots. For numerical calculation of the spectrum we have calculated the poles  $\Lambda_0$  and  $\Lambda_{\pm}$  by using a polynomial root finding algorithm.

Therefore, the incoherent part of the spectrum can be written as

$$S(\nu - \omega_{0}) = -\operatorname{Re}\left\{\frac{C_{\Lambda_{0}}}{i(\omega_{0} - \nu) + \Lambda_{0}} + \frac{C_{\Lambda_{+}}}{i(\omega_{0} - \nu) + \Lambda_{+}} + \frac{C_{\Lambda_{-}}}{i(\omega_{0} - \nu) + \Lambda_{-}} + \frac{C_{\lambda_{+}}}{i(\omega_{0} - \nu) + \lambda_{+}} + \frac{C_{\lambda_{-}}}{i(\omega_{0} - \nu) + \lambda_{-}}\right\},$$
(4.14)

where the C-values are the residues of f(p)/pF(p) at five simple poles of F(p).

To demonstrate the dependence of the effect of tailored reservoir on the fluorescence spectra, we take  $\tau$  and  $\bar{n}$  as parameters. In Fig. 2(a) we plot the fluorescence spectra and the modal density of the reservoir with detuning  $\omega_0 - \omega$ . For  $\tau = 0$ , i.e, for infinitely broadband reservoir, the fluorescence spectrum shows the well known Mollow triplet [20]a. For finite bandwidth of the bath, i.e, for nonzero  $\tau$ , the central peak stays as same profile, however, the side peaks become narrower. The reason is the following. Under the strong driving field, dipole moment of the two-state system oscillates at three different dressed frequencies,  $\omega_0$  and  $\omega_0 \pm \Omega_R$ , where  $\Omega_R$  is the Rabi frequency induced by the strong laser field. The width of these peaks mainly depend on the spontaneous decay rates induced by vacuum radiation field. When the value of  $\tau$  increases, the density of bath



Fig. 2. (a). Fluorescence spectrum  $S(\nu - \omega_0)\gamma/\text{modal}$  density of the bath  $P(\nu - \omega_0)$  versus frequency  $(\nu - \omega_0)/\gamma$  is plotted, with  $E_0/\gamma = 10.0$  and  $\overline{n} = 0.0$  for three different values of  $k = \gamma \tau = 0.0,0.05$  and 0.1. (Both scales are arbitrary). (b). Same as in Fig. 2(a) with k = 0.05 for three different values of  $\overline{n} = 0.0, 1.0$  and 2.0.

modes decreases near the side peak frequencies and thus the width of the side peaks decrease sharply. However, the mode density of the reservoir near the central peak does not change much. This is why the width of the central peak is not affected. The small shift in side peaks is due to the fact that the Rabi frequency is detuned from the band center of the bath and can be explained as in the case of linear absorption.

In Fig. 2(b) we show the fluorescence spectra with increase in temperature but for small value of  $\tau$ , i.e,  $\gamma \tau = 0.05$ . With increase in temperature, the peaks are all broadened almost as in the case of broadband reservoir. However, the side peaks are shifted outward slightly. That means the Rabi frequencies increase slightly and this is due to the finite correlation of the bath.

In Fig. 3(a) we show the spectra at fixed nonzero temperature for different values of  $\tau$ . With moderate values of  $\tau$ , i.e, when the density of bath modes become very low near the range of Rabi frequencies, the width of the side peaks decrease sharply. In such ranges of temperature and  $\tau$ , the width of the central peak increases and if  $\tau$  is larger than this the central peak disintegrates into twin peaks. The splitting in the central peak for different temperatures is shown in Figs. 3(a) and 3(b). This splitting of central peak for narrow bandwidth of the bath is induced by thermal quanta of the bath. We find this feature for the central peak only because the vacuum band shape we have chosen is centered around the system resonance and thus the vacuum modes are present very little near the frequency range of  $\omega_0 \pm \Omega_R$  to dispense energy to the bath and the strong field induced Rabi



Fig. 3. (a) Same as in Fig. 2(a) with  $\overline{n} = 1.0$  for three different values of k = 0.05, 0.2 and 0.4. (b) Same as in Fig. 2(a) with k = 0.4 for three different values of  $\overline{n} = 0.5$ , 1.0 and 2.0.

sidebands are decoupled from the bath. If we would assume the maximum of the reservoir band near the side peaks, we could have found the reversible dynamical feature through the side peaks also.

The splitting of the central peak at higher temperature  $(\bar{n})$  and for shorter bandwidth of the bath can be traced from the poles near the resonance frequency of the system, i.e.,  $p = \lambda_{\pm} = -\frac{1}{2\tau} [1 \mp \{1 - 2k(1 + 2\bar{n})\}^{1/2}]$ . For low values of  $\tau$  and  $\bar{n}$ ,  $1 - 2k(1 + 2\bar{n})$  is positive and the two Lorentzians are merged together. When  $\tau$  and  $\bar{n}$ are of moderate values,  $\lambda_{\pm}$  become complex and the central peak splits up into two. One can evaluate the separation between the peaks as  $S = \frac{1}{\sqrt{2k}} [2k(2\bar{n}+1)-1]^{1/2}$  and this can be described as a thermal bath induced Rabi splitting. S increases with decrease in bandwidth of the bath and with increase in temperature. In effect, a colored bath with very short bandwidth with large thermal photon number acts as an another coherent driving field.

#### 4.2. Absorption spectrum

The absorption spectrum of a weak field probing a strongly driven two-level system has been calculated by Mollow [20]b through the correlation function by the density matrix approach which we will adopt here. The detailed theoretical and experimental comparison can be found in the Ref. [20]c. Here we just show the dependence of the finite bandwidth and temperature of the bath on the absorption profile.

The weak probe field absorption spectrum of the driven two-state system can be calculated [20]b,c from

$$I(\omega_p - \omega_0) = \left[\lim_{t \to \infty} \int_0^\infty e^{-i(\omega_p - \omega_0)t'} \langle \left[\sigma_-(t + t'), \sigma_+(t)\right] \rangle dt'\right],\tag{4.15}$$

where the real and imaginary parts of  $I(\omega_p - \omega_0)$  give the absorption and dispersion spectrum, respectively. Here  $\omega_p$  is the frequency of the weak probe field. We also assume  $\Delta_d = 0$  as in the case of resonance fluorescence. By performing Laplace transform of the quantity

$$g(t') = \lim_{t \to \infty} \langle \left[ \sigma_+(t+t'), \sigma_-(t) \right] \rangle, \tag{4.16}$$

we obtain

$$g(p) = L(g(t')) = \frac{(f_2(p) - f_1(p))}{pF(p)},$$
(4.17)

where F(p) is given in Eq. (4.11) and

$$f_{1}(p) = \frac{1}{2} (S_{z}(t \to \infty) + 1) p \left\{ \left[ p + \overline{Q}_{-}(p) \right] \left[ p + \overline{Q}_{z}(0) \right] + E_{0}^{2}/2 \right\} + S_{+}(t \to \infty) (p + \gamma) \frac{iE_{0}}{2} (p + \overline{Q}_{-}),$$
(4.18)

and

$$f_{2}(p) = \frac{1}{2} (1 - S_{z}(t \to \infty)) p \left\{ \left[ p + \overline{Q}_{-}(p) \right] \left[ p + \overline{Q}_{z}(0) \right] + E_{0}^{2}/2 \right\} - S_{+}(t \to \infty) (p - \gamma) \frac{iE_{0}}{2} (p + \overline{Q}_{-}).$$
(4.19)

We thus obtain

$$I(\omega_{p} - \omega_{0}) = -\left[\frac{C_{A_{0}}'}{i(\omega_{0} - \omega_{p}) + \Lambda_{0}} + \frac{C_{A_{+}}'}{i(\omega_{0} - \omega_{p}) + \Lambda_{+}} + \frac{C_{A_{-}}'}{i(\omega_{0} - \omega_{p}) + \Lambda_{-}} + \frac{C_{A_{+}}'}{i(\omega_{0} - \omega_{p}) + \lambda_{+}} + \frac{C_{A_{-}}'}{i(\omega_{0} - \omega_{p}) + \lambda_{-}}\right],$$
(4.20)

where C'-values are the residues of  $(f_2(p) - f_1(p))/pF(p)$  at the five poles of F(p) which are discussed earlier for the case of emission spectrum. The absorptive part of the spectrum is the real part of the expression  $I(\omega_p - \omega_0)$ .

In Fig. 4 we show the absorption profile to find the effect of k and  $\bar{n}$ . Here also we set  $E_0/\gamma = 10.0$ . In Fig. 4(a) we take  $\bar{n} = 0$  and for different values of k. At zero temperature and for finite bandwidth of the bath, the profile is similar to the absorption calculated by Mollow [20]b,c, i.e, a gain in the absorption profile is observed. The effect of k is not so drastic near the center frequency but the Rabi sidebands affected by shifting outward and become narrower with increase in k. This shift is analogous to the shift in linear absorption in the case when the band center of the bath is detuned from the system frequency. In the strongly driven case, as is here, system dipole has three dressed frequencies. Among them the Rabi side band peaks get affected strongly because of the Rabi sideband modes are detuned from the center of the reservoir band and the bath mode density is also very low in the vicinity of these frequencies and is shifted.



Fig. 4. (a) Absorption spectrum Re[ $I(\omega_p - \omega_0)\gamma$ ] versus frequency  $(\omega_p - \omega_0)/\gamma$  is plotted, with  $E_0/\gamma = 10.0$  and  $\overline{n} = 0.0$  for three different values of k = 0.0, 0.05 and 0.1. (Both scales are arbitrary). (b). Same as in (a) with k = 0.4 for  $\overline{n} = 0.5$  and 2.0.

In Fig. 4(b) we show the absorption profile at k = 0.4 for different  $\bar{n} = 0.5$  and 2.0 as a typical case to demonstrate. At finite temperature, the effect of  $\tau$  and  $\bar{n}$  on the absorption profile are not so drastic as in the case of fluorescence near the center frequency. This is because the absorption intensity near the two-state resonance is usually very low and thus it is difficult to detect the splitting due to increase in temperature and decrease in bandwidth of the bath.

# 5. Conclusion

We consider the system-heat bath model in the resonant interaction form for the bath with a finite bandwidth to investigate the decaying dynamics of the system within one approximation that the system-bath joint density is always factorizable. Due to the evolution of the reduced system with memory, a strong driving field leads to a qualitatively different dynamics. We have given the modified Bloch equation for the Lorentzian bath mode distribution as a prototype function for finite bandwidth of the bath. In the limit of infinitely broadband vacuum, it reduces to the usual Bloch equation. The decay dynamics is studied through the transient linear absorption, resonance fluorescence spectra and absorption spectra of the driven system. In the linear absorption we find the physical origin of spectral shift and non-Lorentzian lineshape which are usually observed in the solution phase or in the condensed phase. At higher temperature and narrow bandwidth of the bath we find the splitting in the absorption profile.

We have found a dramatic modification of the Mollow resonance fluorescence spectrum. At higher temperature and for narrow bandwidth of the bath, a thermal bath induced splitting emerges on the central peak of the profile of Mollow triplet. When the bath mode density decreases near the range of Rabi frequencies, the peak width of the Mollow sidebands decrease very sharply. In the absorption profile of the driven system we have found the signature of the bath through the sidebands. In the gain part of the profile the effect is smaller and thermal bath induced splitting in the central peak is hard to observe.

We have analyzed the linear and nonlinear spectra purely from the dynamical point of view by assuming the system–reservoir interaction is characterized by a finite bandwidth of the bath. Appendix A is added to comment on the naming of the dynamics as non-Markovian and on the physical validity of the application of regression theorem for arbitrary bandshape function of the bath. To understand the physical origin of the splitting on the spectral profiles a heuristic argument is presented in Appendix B.

Although the Bloch equation picture is very general to describe many phenomena from NMR to Quantum Optics in gas phase systems, however, a single impurity molecule in condensed phase with a realistic host system demands a modification of Bloch equation to simulate the effect of the environment. For example, very recently a pump-probe experiment [21]b with a new host-guest system, e.g, benzanthracene in a napthalene crystal, showed hyper Raman and sub-harmonic resonance on the profile of fluorescence which is explained by modified Bloch equation. These modifications are needed not only just to satisfy the experimental data but also to understand the effect of various noise processes induced by the host matrix from very short to very long correlation time [23]. In this work we considered a model colored noise environment through a Lorentzian bandwidth of the bath, which is a prototype of a finite bandwidth of bath, to understand dynamical modification on the spectra. We hope a suitable host matrix can be designed to act as an environment with a finite response time on the guest molecule to show the proposed effect to occur.

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# Appendix A. A few comments on regression hypothesis and Markov property in the density matrix theory of damping

First of all we show here that the approximation we have made in Eq. (2.7) i.e.,

$$\kappa(t) = s(t)f(t) \equiv s(t)f(0) \tag{A.1}$$

satisfy regression theorem [24,25]. This means the two-time correlation function evolves in the same way as the expectation value.

At t = 0, we have assumed

$$\kappa(0) = s(0)f(0). \tag{A.2}$$

The reduced system and reservoir density operators are defined by

$$s(t) = \operatorname{Tr}_{B}\{\kappa(t)\}, \quad f(t) = \operatorname{Tr}_{s}\{\kappa(t)\}, \quad (A.3)$$

respectively. In terms of  $\kappa(t)$ , the average of a system operator B, is given by

$$\langle B(t) \rangle = \operatorname{Tr}_{s} \{ \operatorname{Tr}_{B} \{ B(0) \kappa(t) \} \} = \operatorname{Tr}_{s} \{ B(0) \operatorname{Tr}_{B} \{ U(t) \kappa(0) U^{-1}(t) \} \} = \operatorname{Tr}_{s} \{ B(0) s(t) \},$$
(A.4)

where U is the total system–bath evolution operator. The two-time correlation function  $\langle A(t)B(t')\rangle$  is given by

$$\langle A(t)B(t')\rangle = \operatorname{Tr}_{s}\{\operatorname{Tr}_{B}\{A(t)B(t')\kappa(0)\}\} = \operatorname{Tr}_{s}\{\operatorname{Tr}_{B}\{U^{-1}(t)A(0)U(t)U^{-1}(t')B(0)U(t')\kappa(0)\}\}$$

$$= \operatorname{Tr}_{s}\{A(0)U(t-t')B(0)U(t')\kappa(0)U^{-1}(t')U^{-1}(t-t')\}\}$$

$$= \operatorname{Tr}_{s}\{A(0)\operatorname{Tr}_{B}\{U(t-t')B(0)\kappa(t')U^{-1}(t-t')\}\},$$
(A.5)

where A is also a system operator. Here we should notice that the Eq. (A.5) is an exact expression.

Comparing Eqs. (A.4) and (A.5), we can find that both s(t) and the function

$$\Omega(t,t') = \text{Tr}_{B} \{ U(t-t') B(0) \kappa(t') U^{-1}(t-t') \}$$
(A.6)

appear to evolve in the similar way but with subtle difference. Note that the initial time for the evolution of s(t) is t = 0 when the system-bath correlation vanishes. Without loss of generality one is interested in evaluating the correlation function  $\langle A(t)B(t')\rangle$  for the case  $t \ge t'$ . Thus one can assume  $\Omega(t,t') = 0$ , for  $t \le t'$ . So the initial time for the evolution of  $\Omega(t,t')$  is t = t' and thus a system-bath correlation will exist at t = t'. This correlation at the initial time in  $\Omega(t,t')$ , gives some extra correction factor to the evolution of  $\Omega(t,t')$  thereby causing  $\Omega(t,t')$  to evolve differently from s(t). This extra factor will be destroyed provided  $\kappa(t')$  factorizes for all time, i.e,

$$\kappa(t') = s(t')f(0). \tag{A.7}$$

This extra factor is calculated by Swain [24]. Thus when (A.7) is satisfied, the two-time correlation function like (A.5) evolves in the same way as single time expectation value (A.4), i.e., quantum regression theorem holds.

Here we would like to comment on the Markov property in the context of density matrix theory of damping. By using the Langevin description of Heisenberg's equation of motion, Lax [25] has proved that the Markov property implies the regression theorem as well as the converse. In that derivation Markov property means the Langevin force at time t be uncorrelated to any information at earlier times. In terms of density matrix theory he [25] has proved that the regression theorem can be taken as a definition of Markov property by which quantum dynamics corresponds to classical dynamics. Thus according to Lax, in the density matrix theory Markov property is a derived condition when the regression theorem holds good. This precisely means correlation between system and bath does not evolve in time.

Actually the memory can come in the picture in two different ways: (i) one way is that the joint system–bath density evolves with memory, which means when the factorization like (A.7) is not valid. This should be called as non-Markovian in the sense of Lax. (ii) Other way depends on the factorization assumption, Eq. (A7), which means the reduced system density can evolve with memory. This is Markovian in the sense of Lax, because regression hypothesis holds. In our work we follow the second condition but referred as non-Markovian to keep track with the earlier literature [8–12]. In the text we have named the master equation for the reduced density as a system operator concerned non-Markovian equation which evolves in time with memory and clearly needs a shape function of the vacuum band. Thus one can conclude that the regression theorem or Markov property of Lax has more general validity than the standard Born-Markov theory (Louisell [3]) and can cover the case when the reduced system evolves with time convolution. Since the factorization assumption is independent of bandwidth or memory function, the physical validity of the application of regression theorem lies on the choice of the memory function or the bandwidth of the bath. One possible way is to choose the memory function so that the dynamics should reach the appropriate thermal equilibrium in the long time limit.

#### Appendix B. Origin of splitting on the spectral profiles: a heuristic argument

To understand the thermal splitting and to identify the source of eigenvalues  $\lambda_{\pm}$  we consider the evolution of coherence term in Eq. (2.22a), for  $\Delta = 0$  and  $\Delta_{\rm b} = 0$  as

$$\dot{S}_{+}(t) = \frac{i}{2} E_0 S_z(t) - \frac{\gamma}{2} (1 + 2\bar{n}(\omega_0)) \int_0^t S_{+}(t') e^{-(t-t')/\tau} dt',$$
(B.1)

which might be written as the pair of coupled equations

$$\dot{S}_{+}(t) = \frac{i}{2} E_0 S_z(t) - \frac{\gamma}{2} (1 + 2\bar{n}(\omega_0)) \alpha(t), \quad \dot{\alpha}(t) = -\frac{1}{\tau} \alpha(t) + S_{+}(t), \quad (B.2)$$

with initial condition  $\alpha(0) = 0$ . If one wishes to, one may regard the introduction of  $\alpha(t)$  as simply a mathematical device, but in fact this variable has the physical significance of a field amplitude. Now write  $S_+(t) = X(t) + iY(t)$  and  $\alpha(t) = x(t) + iy(t)$ . Then the equations for the real parts are

$$\dot{X}(t) = -\frac{\gamma}{2} (1 + 2\bar{n}(\omega_0)) x(t), \quad \dot{x}(t) = -\frac{1}{\tau} x(t) + X(t).$$
(B.3)

The eigenvalues derived from these equations are

$$\lambda_{\pm} = -\frac{1}{2\tau} \Big[ 1 \mp \sqrt{1 - 2\gamma\tau (1 + 2\bar{n}(\omega_0))} \Big], \tag{B.4}$$

which can be accounted for the splitting. The coupling between x(t) and X(t) that allows the eigenvalues to be complex is, indeed, of the form as that which causes the vacuum field Rabi splitting in the cavity where x(t) is the real part of the amplitude of the cavity field and X(t) the real part of the atomic polarization [20]c.

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