

## Second Order Optical Process of a Three-Level System in Contact with a Nearly Gaussian-Markoffian Noise Bath

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A general formulation is given for the second order optical process of a three-level system in contact with a heat bath consisting of harmonic oscillators with a proper frequency spectrum producing a Markoffian random perturbation. Elimination of the bath using the influence functional method of Feynman and Vernon yields a continuous fraction expression and the result affords a basis to clarify the relation between the stochastic and dynamical approaches to treat the partial destruction of the quantum coherence of the optical process. As an example, a three-level system with intermediate-state interaction is treated in some detail.

### §1. Introduction

In the present work, we study the second order optical process as an example of a quantum process in a dissipative environment. This study can also be extended to the tunneling problem, since by replacing photons with electrons, the optical system can be regarded as a kind of tunneling system. The interaction of the system with its environment destructs the coherence of the quantum process. In the second order optical process, there is only the Raman scattering where the quantum coherence of the whole system from the initial to the final states through the intermediate states is maintained if the natural radiative damping is the only mechanism acting in the intermediate states. When the random modulation from the environment comes into play to perturb the system, the coherence is disturbed and two types of incoherent processes appear; one is the luminescence in which the coherence of the process is completely lost and the other is the broadened Raman where the coherence is only partially lost. The physical essence of this problem lies in the way how the coherence is affected by the modulation from the environment.

In order to investigate this sort of problem,

two theoretical approaches have been widely used. The first is the dynamical approach in which the environment is explicitly represented by a dynamical system, e.g., by a set of oscillators, and their interaction with the system is treated in a dynamical way. The second is the stochastic approach in which the effect of environment is represented by a stochastic variable which perturbs the system.

The dynamical approach is microscopic and quantitative, but actual calculations are complicated and are not transparent. Analytical treatment is possible only for a weak and fast modulation case. The broadened-Raman emission, which appears in more ordinary conditions of modulation, cannot be treated in this approach, so that it is difficult to attain a deep insight. On the contrary, the stochastic approach can be used for a wide category of physical cases from a unified point of view. The formulation may be carried out analytically by non-perturbative methods. Thus this approach is particularly useful to treat this kind of problem. Since the stochastic approach is phenomenological by its nature, it is desirable to see how this approach is connected with a microscopic one.

In the first of the present series,<sup>1)</sup> we derived the equation of motion for a test system coupled to the nearly Gaussian-Markoffian heat bath from a dynamical point of view using the influence functional given by Feynman and

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Vernon, and evaluated its resolvent in a continued fractional expression. The result is equivalent to the stochastic Gaussian-Markoffian model except that our result includes the reaction effects of the bath. In the second paper,<sup>2)</sup> we derived a non-perturbative expression for two-time correlation functions of a system as an extension of ref. 1. We also pointed out that the higher order interaction with the environment gives rise to some special features for two-time correlation functions.

In this paper, we make a formulation of the second order optical process from the dynamical point of view equivalent to the stochastic Gaussian-Markoffian model as an extension and application of refs. 1 and 2. On the basis of this, we unify the stochastic and dynamical approaches. As an example, we discuss the power spectrum of scattered light from a three-level system interacting with the environment in its excited state (*intermediate state interaction*) which was studied in detail in the stochastic approach.<sup>3-5)</sup> We also present numerical analysis of the emission spectrum and discuss the reaction effects of the bath which is derived from our dynamical approach as the modification of the stochastic

theory. We also notice a mixing of the coherent and incoherent processes which produces the broadened Raman process.

This paper is organized in the following way. In the next section, we derive the formulation of the second order optical process of a three-level system. In §3 we apply our formulation to the intermediate state interaction model. Numerical results are also presented to show the reaction effects of the bath which were ignored in the previous stochastic treatments. The last section is devoted to the summary and conclusion.

## §2. Formulation

In order to formulate the problem in a general way, we introduce the *physical spectrum* proposed by Eberly and Wódkiewicz.<sup>6)</sup> This spectrum is based on the normalized counting rate of photodetector including the effect of filter or interferometer. The setting frequency and the spectral bandwidth of Fabry-Perot interferometer are respectively denoted by  $\omega$  and  $\Gamma_f$ . The emitted light is essentially defined by the oscillating dipole moment of the system. Then the physical spectrum at time  $t$  is expressed in the form

$$I(\omega, t) = 2\Gamma_f \int_0^t dt' \int_0^{t'} dt'' \exp [-(\Gamma_f - i\omega)(t - t')] \times \exp [-(\Gamma_f + i\omega)(t - t'')] \langle D_e^+(t') D_e^-(t'') \rangle, \quad (2.1)$$

where  $D_e^\pm(t)$  represent dipole operators of the atomic system to be observed and we assumed that the proportionality constant between the emitted light field and the dipole moment is included in these operators.

The system under observation is composed of the main system **A** and the heat bath **B**. We denote a state of **A** by coherent states  $\phi$  and  $\phi^*$  and **B** by a  $c$ -number coordinate  $X$ . The variable  $X$  is assumed to have the Gaussian property. Furthermore, the interaction between **A** and **B** is assumed to have  $H_I = V(\phi, \phi^*)X$ , where  $V(\phi, \phi^*)$  is an operator of **A**. As was shown in refs. 1 and 2, we can express such a system in a functional integral form. By performing the integration over the bath, we find the physical spectrum eq. (2.1) to be

$$I(\omega, t) = \text{tr} \{ \xi(t) \}, \quad (2.2)$$

where the operator  $\xi(t)$  is defined by

$$\xi(t) = \iint N^{-1} d\phi_f^* d\phi_f \iint N^{-1} d\phi_f'^* d\phi_f' |\phi_f\rangle \xi'(\phi_f^*, \phi_f'; t) \langle \phi_f' |, \quad (2.3)$$

with the element

$$\xi'(\phi_f^*, \phi_f'; t) = \text{T} \left( 2\Gamma_f \int_0^t dt' \int_0^{t'} dt'' \int D_f[\phi^*(\tau)\phi(\tau)] \int D_f[\phi'^*(\tau)\phi'(\tau)] \right)$$

$$\begin{aligned} & \times \exp [(i/\hbar)S_A(\phi^*, \phi; t)]F(\phi^*, \phi, \phi'^*, \phi'; t) \exp [-(i/\hbar)S_A^*(\phi'^*, \phi'; t)] \\ & \times e^{-(\Gamma_f-i\omega)(t-t')} e^{-(\Gamma_f+i\omega)(t-t'')} D_c^+(\phi'^*, \phi', t') D_c^-(\phi^*, \phi, t''). \end{aligned} \quad (2.4)$$

In the above,

$$\begin{aligned} S_A(\phi^*, \phi; t) &= \lim_{M \rightarrow \infty} \sum_{k=1}^M \varepsilon [i\hbar \dot{\phi}_k^*(\phi_k - \phi_{k-1}) / \varepsilon - H_A(\phi_k^*, \phi_{k-1})] \\ &\equiv \int_0^t d\tau \left[ i\hbar \dot{\phi}^*(\tau) \dot{\phi}(\tau) - H_A(\phi^*(\tau), \phi(\tau)) \right] \end{aligned} \quad (2.5)$$

is the Schrödinger action of  $\mathbf{A}$ ,  $D_c^\pm(\phi^*, \phi, t')$  are the dipole elements at time  $t'$  and

$$\int D_f[\phi^*(\tau)\phi(\tau)] \equiv \lim_{M \rightarrow \infty} \prod_{k=1}^{M-1} \int N^{-1} d\phi_k^* d\phi_k \quad (2.6)$$

denotes integration over the measure of the paths of  $\{\phi^*(\tau), \phi(\tau)\}$  over the time interval  $(0, t)$  for the fixed initial state  $\phi(0) = \phi_0$  and the final state  $\phi^*(t) = \phi_f^*$  with the normalization constant  $N$ . The valuables  $\phi^*(t)$ ,  $\phi(t)$  etc. represent complex numbers for a Boson system and are Grassmann numbers for a Fermion system in which case the symbol  $T$  takes care of their proper ordering in time. The effect of the bath is described by the Feynman-Vernon influence functional<sup>7)</sup>

$$\begin{aligned} F(\phi^*, \phi, \phi'^*, \phi'; t) &= \exp \left\{ (-i\Delta)^2 \int_0^{t'} d\tau' \int_0^{\tau'} d\tau e^{-\gamma(\tau'-\tau)} V^\times(\phi^*, \phi, \phi'^*, \phi'; \tau') \right. \\ & \quad \left. \times \left( V^\times(\phi^*, \phi, \phi'^*, \phi'; \tau) - i \frac{\beta\hbar\gamma}{2} V^\circ(\phi^*, \phi, \phi'^*, \phi'; \tau) \right) \right\}, \end{aligned} \quad (2.7)$$

where we set

$$\begin{aligned} V^\times(\phi^*, \phi, \phi'^*, \phi'; t) &= V(\phi^*(t), \phi(t)) - V(\phi'^*(t), \phi'(t)), \\ V^\circ(\phi^*, \phi, \phi'^*, \phi'; t) &= V(\phi^*(t), \phi(t)) + V(\phi'^*(t), \phi'(t)). \end{aligned} \quad (2.8)$$

To derive this particular form, we assumed that the bath is initially at the inverse temperature  $\beta = 1/k_B T$  and shows the Debye relaxation with the damping constant  $\gamma$  for an external pulsive excitation. The coupling strength is denoted by the dimensionless constant  $\Delta$ .

The Laplace transform of eq. (2.2) can be performed by deriving simultaneous differential equations of eq. (2.4). The procedure is the same as that was done in ref. 2. For the time-independent Hamiltonian  $H_A(t) = H_0$ , the result is simply given by

$$\begin{aligned} I[\omega, s] &= \frac{2\Gamma_f}{s+2\Gamma_f} \text{tr} \left\{ D_c^+ Z_0[s+\Gamma_f+i\omega] \left( D_c^- Z_0[s] \rho(0) \right) \right. \\ & \quad \left. + \sum_{n=1}^{\infty} n! (i\Delta)^{2n} D_c^+ Z_0[s+\Gamma_f+i\omega] \left( \prod_{\alpha=1}^n (V^\times Z_\alpha[s+\Gamma_f+i\omega]) \right) \right. \\ & \quad \left. \times \left[ D_c^- \left( \prod_{\beta=1}^n (Z_{n-\beta+1}[s] \Theta) \right) Z_0[s] \rho(0) \right] \right\} + \text{c.c.}, \end{aligned} \quad (2.9)$$

where  $\rho(0)$  represents the initial condition of  $\mathbf{A}$  and we define

$$\begin{aligned} Z_\alpha[s] &= \frac{1}{s + \alpha\gamma - (i\hbar)^{-1} H_0^\times + V^\times \frac{1}{s + (\alpha+1)\gamma - (i\hbar)^{-1} H_0^\times + V^\times} \frac{1}{s + (\alpha+2)\gamma - (i\hbar)^{-1} H_0^\times + V^\times} \Theta} \\ & \quad \frac{1}{s + (\alpha+1)\gamma - (i\hbar)^{-1} H_0^\times + V^\times \frac{1}{s + (\alpha+2)\gamma - (i\hbar)^{-1} H_0^\times + V^\times} \frac{1}{s + (\alpha+3)\gamma - (i\hbar)^{-1} H_0^\times + V^\times} \Theta} \end{aligned} \quad (2.10)$$

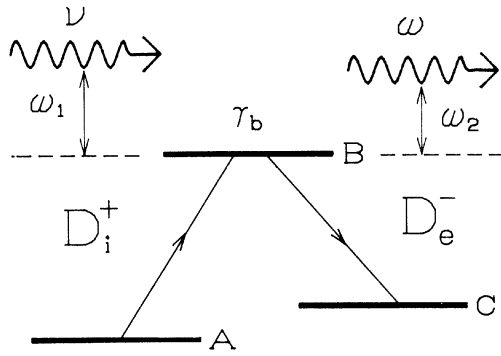


Fig. 1. Energy level scheme of a second order optical process of a three-level system.

In the above we set

$$\Theta \equiv V^\times - i \frac{\beta \hbar \gamma}{2} V^\circ. \tag{2.11}$$

The notations  $\times$  and  $\circ$  for hyper operators are defined by

$$\begin{aligned} F^\circ G &\equiv FG + GF, \\ F^\times G &\equiv FG - GF, \end{aligned} \tag{2.12}$$

for any operators  $F$  and  $G$ . As was discussed in ref. 2, the second term of eq. (2.9) represents the effect of the higher-order of the system-bath interaction on the correlation functions.

Consider the second order optical process

of a three-level system interacting with the bath. We assume that the main system  $A$  is composed of the three-level system described by a ground state  $|A\rangle$ , an intermediate state  $|B\rangle$  and a final state  $|C\rangle$ , and the interaction between these states and  $c$ -number electric field. The Hamiltonian of  $A$  is given by

$$H_A(t) = H_0 + H_R(t), \tag{2.13}$$

where  $H_0$  is for the three-level system and  $H_R(t)$  is the interaction between the three-level system and the electric field of radiation. We assume  $H_R(t)$  in the rotating wave approximation

$$H_R(t) = i\hbar(D_i^+ e^{-i\nu t} + D_i^- e^{i\nu t}), \tag{2.14}$$

where  $\nu$  represents the frequency of incident field and  $D_i^\pm$  are the dipole operators of the three-level system. We assumed that the proportionality constant of this interaction is included in these operators. When the interaction  $H_R(t)$  is weak, the weight  $\exp[(i/\hbar)S_A(Q; t, 0)]$  and its conjugate can be expanded by  $(i/\hbar)^{-1}H_R(t)$ . By retaining relevant terms corresponding to the transition from  $|A\rangle$  to  $|C\rangle$  through  $|B\rangle$ , the physical spectrum of the second order optical process can be written as

$$\begin{aligned} \xi'(\phi_f^*, \phi_f'; t') = & T \left( 2\Gamma_f \int_0^{t'} dt' \int_0^{t'} dt'' \int_0^{t''} dt_1 \int_0^{t''} dt_2 \int D[Q(\tau)] \int D[Q'(\tau)] \exp[(i/\hbar)S_0(Q; t)] \right. \\ & \times F(Q, Q'; t) \exp[-(i/\hbar)S_0^*(Q'; t)] e^{-(\Gamma_f - i\omega)(t-t')} e^{-(\Gamma_f + i\omega)(t-t'')} e^{i\nu(t_1 - t_2)} \\ & \left. \times D_e^+(Q'(t')) D_e^-(Q(t'')) D_i^-(Q'(t_1)) D_i^+(Q(t_2)) \right), \end{aligned} \tag{2.15}$$

where we simply write  $Q(\tau)$  or  $Q'(\tau)$  for  $(\phi^*(\tau), \phi(\tau))$  or  $(\phi'^*(\tau), \phi'(\tau))$  and  $S_0(Q; t)$  represents the Schrödinger action of the Hamiltonian  $H_0$ . In order to perform the

Laplace transform of eq. (2.15), we rearrange the time ordering of this as Fig. 2. Then, for example, (I) of Fig. 2 can be expressed as the form

$$\begin{aligned} \xi'(\phi_f^*, \phi_f'; t) = & T \left( 2\Gamma_f \int_0^t dt' \int_0^{t'} dt_1 \int_0^{t_1} dt'' \int_0^{t''} dt_2 \int D[Q(\tau)] \int D[Q'(\tau)] \right. \\ & \times \exp[(i/\hbar)S_0(Q; t)] F(Q, Q'; t) \exp[-(i/\hbar)S_0^*(Q'; t)] \\ & \times e^{-2\Gamma_f(t-t')} D_e^+(Q'(t')) e^{-(\Gamma_f + i\omega)(t'-t_1)} D_i^-(Q'(t_1)) \\ & \left. \times e^{-(\Gamma_f + i(\omega - \nu))(t_1 - t'')} D_e^-(Q(t'')) e^{i\nu(t'' - t_2)} D_i^+(Q(t_2)) \right). \end{aligned} \tag{2.16}$$

The Laplace transformation of elements (I), (II) and (III) are calculated from the Laplace transformation of functions  $t-t'$ ,  $t'-t_1$ , etc. of integrand. Each Laplace transform can be

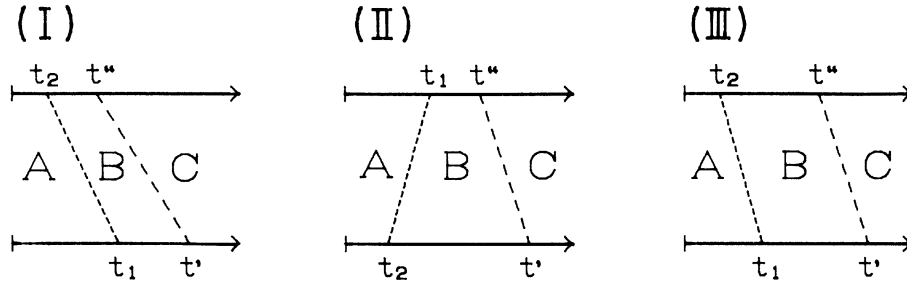


Fig. 2. Decomposed propagator of a second order optical process.

evaluated in parallel to eq. (2.9). The final result is given in the operator form as

$$I[\omega, s] = I_I[\omega, s] + I_{II}[\omega, s] + I_{III}[\omega, s] + \text{c.c.}, \tag{2.17}$$

where

$$I_I[\omega, s] = |\langle C | D_c^- | B \rangle \langle B | D_i^+ | A \rangle|^2 \frac{2\Gamma_f}{s + 2\Gamma_f} \sum_{jkl} \langle\langle CB | R_{1j}[s + \Gamma_f + i\omega] | CB \rangle\rangle \times \langle\langle CA | R_{jk}[s + \Gamma_f + i(\omega - \nu)] | CA \rangle\rangle \langle\langle BA | R_{kl}[s - i\nu] | BA \rangle\rangle \langle\langle AA | R_{l1}[s] | AA \rangle\rangle, \tag{2.18}$$

$$I_{II}[\omega, s] = |\langle C | D_c^- | B \rangle \langle B | D_i^+ | A \rangle|^2 \frac{2\Gamma_f}{s + 2\Gamma_f} \sum_{jkl} \langle\langle CB | R_{1j}[s + \Gamma_f + i\omega] | CB \rangle\rangle \langle\langle BB | R_{jk}[s] | BB \rangle\rangle \times \langle\langle AB | R_{kl}[s + i\nu] | AB \rangle\rangle \langle\langle AA | R_{l1}[s] | AA \rangle\rangle, \tag{2.19}$$

and

$$I_{III}[\omega, s] = |\langle C | D_c^- | B \rangle \langle B | D_i^+ | A \rangle|^2 \frac{2\Gamma_f}{s + 2\Gamma_f} \sum_{jkl} \langle\langle CB | R_{1j}[s + \Gamma_f + i\omega] | CB \rangle\rangle \langle\langle BB | R_{jk}[s] | BB \rangle\rangle \times \langle\langle BA | R_{kl}[s - i\nu] | BA \rangle\rangle \langle\langle AA | R_{l1}[s] | AA \rangle\rangle. \tag{2.20}$$

In the above we denote the density elements in the hyper-operator form as

$$|AB\rangle\rangle = |A\rangle\langle B|, \tag{2.21}$$

$$\langle\langle AB | \rho(t) = \langle A | \rho(t) | B \rangle,$$

then the initial condition is written as  $\rho(0) = |AA\rangle\rangle$ . The elements  $R_{jk}[s]$  are defined by using eq. (2.10) as

$$R_{jk}[s] = \sum_{q=1}^{\min(j,k)} \left\{ (-i\Delta)^{j+k-2q} \frac{(j-1)!}{(q-1)!} \left[ \prod_{\alpha=q}^{j-1} (Z_{j-\alpha+q-1}[s]\Theta) \right] Z_{q-1}[s] \left[ \prod_{\beta=q}^{k-1} (V^\times Z_\beta[s]) \right] \right\}, \tag{2.22}$$

where  $\min(j, k)$  takes smaller value of  $j$  or  $k$  and we set

$$\prod_{\alpha=q}^{k'} (Z_{k-\alpha+q}[s+i\omega]\Theta) = \prod_{\beta=q}^{k'} (V^\times Z_\beta[s]) = 1. \quad (\text{for } k' < q) \tag{2.23}$$

By using eq. (2.17), the continuous wave (CW) response spectrum is given by

$$I(\omega) = \lim_{s \rightarrow 0} sI[\omega, s]. \tag{2.24}$$

### §3. Intermediate States Interaction

The interaction of the system with the bath when it is the intermediate states is called an in-

intermediate state interaction. We are concerned with the question how the quantum coherence is affected by the bath interaction and how it is manifested in optical response. This subject has been extensively studied from the stochastic approach.<sup>3-5)</sup> Here we discuss this by using the formalism of §2 derived from the dynamical point of view.

We write the Hamiltonian of the three-level system as the form

$$H_0 = \hbar(\omega_A |A\rangle\langle A| + \omega_B |B\rangle\langle B| + \omega_C |C\rangle\langle C|). \quad (3.1)$$

The intermediate state interaction is expressed by

$$V = |B\rangle\langle B|. \quad (3.2)$$

We further assumed that the natural radiation damping of the intermediate state is given by a phenomenological damping constant  $\gamma_b$  for the state  $|B\rangle$ . Then, from eqs. (2.17)–(2.20), we can evaluate the emission spectrum for the CW response, which is equivalent to the stochastic result<sup>3-5)</sup> except for including the spectral bandwidth of interferometer  $\Gamma_f$  and the reaction effects of the bath with the factor  $\delta$ . Corresponding to eq. (2.9), we may divide this result into two terms as

$$I(\omega) = I_0 + I_1, \quad (3.3)$$

where

$$I_0 = |\langle C | D_c^- | B \rangle \langle B | D_i^+ | A \rangle|^2 \left[ 2 \cdot \text{Re} \left( S_0^+ [\Gamma_f + \gamma_b + i\omega_2] \frac{1}{\Gamma_f - i(\omega_1 - \omega_2)} S_0^- [\gamma_b - i\omega_1] \right) + 2 \cdot \text{Re} (S_0^+ [\Gamma_f + \gamma_b + i\omega_2]) \frac{1}{\gamma_b} \text{Re} (S_0^- [\gamma_b - i\omega_1]) \right], \quad (3.4)$$

and

$$I_1 = |\langle C | D_c^- | B \rangle \langle B | D_i^+ | A \rangle|^2 \left[ 2 \sum_{n=1}^{\infty} \text{Re} \left\{ n! (i\Delta)^n \left( \prod_{\alpha=0}^n S_{\alpha}^+ [\Gamma_f + \gamma_b + i\omega_2] \right) \frac{1}{\Gamma_f + n\gamma - i(\omega_1 - \omega_2)} \right. \right. \\ \left. \left. \times (-\Delta\delta - i\Delta)^n \left( \prod_{\beta=0}^n S_{\beta}^- [\gamma_b - i\omega_1] \right) \right\} + 4 \sum_{n=1}^{\infty} \sum_{m=0}^{\infty} \text{Re} \left\{ (i\Delta)^n \left( \prod_{\alpha=0}^n S_{\alpha}^+ [\Gamma_f + \gamma_b + i\omega_2] \right) \right. \right. \\ \left. \left. \times (-2\delta\Delta)^m (n+m)! \left( \prod_{j=n}^{n+m} \frac{1}{j\gamma + 2\gamma_b} \right) \text{Re} \left\{ (-\Delta\delta - i\Delta)^n \left( \prod_{\beta=0}^n S_{\beta}^- [\gamma_b - i\omega_1] \right) \right\} \right\} \right]. \quad (3.5)$$

In the above,  $S_{\alpha}^{\pm}[s]$  are defined by

$$S_{\alpha}^{\pm}[s] = \frac{1}{s + \alpha\gamma + \frac{(\alpha+1)\Delta^2(1 \pm i\delta)}{s + (\alpha+1)\gamma + \frac{(\alpha+2)\Delta^2(1 \pm i\delta)}{s + (\alpha+2)\gamma + \dots}}}, \quad (3.6)$$

and we set

$$\omega_1 = \nu + \omega_A - \omega_B, \quad (3.7)$$

$$\omega_2 = \omega + \omega_C - \omega_B, \quad (3.8)$$

$$\delta = \beta\hbar\gamma/2. \quad (3.9)$$

The main difference between our formalism and the stochastic one is that now we take into account the reaction effects of the bath with factor  $\delta$ . The resolvent  $S_0^{\pm}[s]$  is equivalent to  $Z_0[s]$  of eq. (2.10), thereby, eq. (3.4) may be identified with the first term of eq. (2.9), whereas eq. (3.5) the second. Then the effects of the higher-order interaction between the system and the bath corresponds to eq. (3.5). In the following, we discuss how the reaction effects of the bath changes the previous stochastic results of ref. 3 and how higher-order interaction between the system and

the bath affects in the emission spectrum.

First we discuss the reaction effects of the bath. Hereafter we put  $\langle C|D_e^-|B\rangle = \langle B|D_i^+|A\rangle = 1$  for simplicity. Equation (3.3) with eqs. (3.4) and (3.5) contain the coherent and incoherent processes. By putting  $\Gamma_f \rightarrow 0$ , a part of eq. (3.4) corresponds to the coherent process becomes the delta-function form as

$$2\pi \cdot \delta(\omega_1 - \omega_2) \cdot |S_0^-[\gamma_b - i\omega_1]|^2. \quad (3.10)$$

This is called as the pure Raman component. The amplitude of this becomes the Lorentzian in the motional narrowing limit  $\gamma \gg \gamma_b, \omega_1, \Delta$  with  $\Delta^2/\gamma \rightarrow \gamma'^{1)}$

$$2\pi \cdot \delta(\omega_1 - \omega_2) \frac{1}{\{\gamma_b + \text{Re}(\eta)\}^2 + \{\omega_1 - \text{Im}(\eta)\}^2}, \quad (3.11)$$

where

$$\eta = \frac{\gamma'(1 - i\beta\hbar\gamma/2)}{-2i\beta\hbar\gamma'/2} \cdot \quad (3.12)$$

$$1 + \frac{-3i\beta\hbar\gamma'/2}{2 + \frac{3 + \dots}{3 + \dots}}$$

In contrast, the amplitude of eq. (3.10) becomes the Gaussian like form in the slow modulation limit  $\gamma \ll \omega_1$

$$2\pi \cdot \delta(\omega_1 - \omega_2) \cdot \frac{2}{\Delta^2} \exp[(\gamma_b^2 - \omega_1^2)/\Delta^2] \cdot |\text{Erfc}[(\gamma_b - i\omega_1)/\sqrt{2}\Delta]|^2, \quad (3.13)$$

where  $\text{Erfc}[\ ]$  is the error function. In this case, the reaction effects of the bath disappear.

Equation (3.5) and a remaining term of eq. (3.4) correspond to the incoherent process. For a finite value of  $\gamma$ , there is no transparent

expression available for the incoherent component, but numerical calculation can be made. Figure 3 shows the emission spectra of the incoherent component for different values of  $\gamma$  fixing the other parameters as  $\Gamma_f = 1.0 \times 10^{-4}$ ,  $\gamma_b = 1.0$ ,  $\Delta = 3.0$  and  $\omega_1 = 10$ . In each figures, the solid line and the dashed line, respectively, denote the spectrum for  $\hbar\beta = 0.02$  and  $\hbar\beta = 0.0$ . Then the dashed lines of  $\hbar\beta = 0.0$  correspond to the previous stochastic results. In Fig. 3(a) of fast modulation, the Lorentzian like peak of the luminescence appears about at the atomic resonant position. The reaction effects of the bath shifts the luminescence peak to the lower frequencies. This can be seen from the analytical expression of the luminescence in the motional narrowing limit which has discussed in eq. (3.11). In this limit, the luminescence component can be expressed in the Lorentzian form as

$$2 \frac{\gamma_b + \text{Re}(\eta)}{\{\gamma_b + \text{Re}(\eta)\}^2 + \{\omega_2 - \text{Im}(\eta)\}^2} \frac{\text{Re}(\eta)}{\gamma_b(\gamma_b + \text{Re}(\eta))} \frac{\gamma_b + \text{Re}(\eta)}{\{\gamma_b + \text{Re}(\eta)\}^2 + \{\omega_1 - \text{Im}(\eta)\}^2}. \quad (3.14)$$

The real part of  $\eta$  works upon eq. (3.14) as the damping, whereas the imaginary part the frequency shift. Thereby, the reaction effects of the bath with terms  $\delta$  in eq. (3.12) induce the frequency shift of the luminescence peak. In Fig. 3(b), the incoherent element is seen to have peaks not only at the luminescence position about  $\omega_2 = 0$  but also at the Raman position  $\omega_2 = 10$ . This peak comes from the factor  $1/\{\Gamma_f + n\gamma - i(\omega_1 - \omega_2)\}$  of eq. (3.5) and is called as the broadened Raman peak which was discussed in the stochastic approach.<sup>3-5)</sup> The position of the luminescence is shifted by

the reaction effect of the bath, but the broadened Raman peak is not changed. In Fig. 3(c), the broadened Raman becomes larger whereas the luminescence becomes smaller. In Fig. 3(d), the luminescence peak is unnoticeable and the broadened Raman peak in a Lorentzian form with the width about  $\gamma$  seen at the Raman position. The reaction effects of the bath do not play any important role and difference of two lines is unnoticeable. In the static limit of  $\gamma \rightarrow 0$ , the broadened Raman components can be analytically evaluated in the delta-function form as

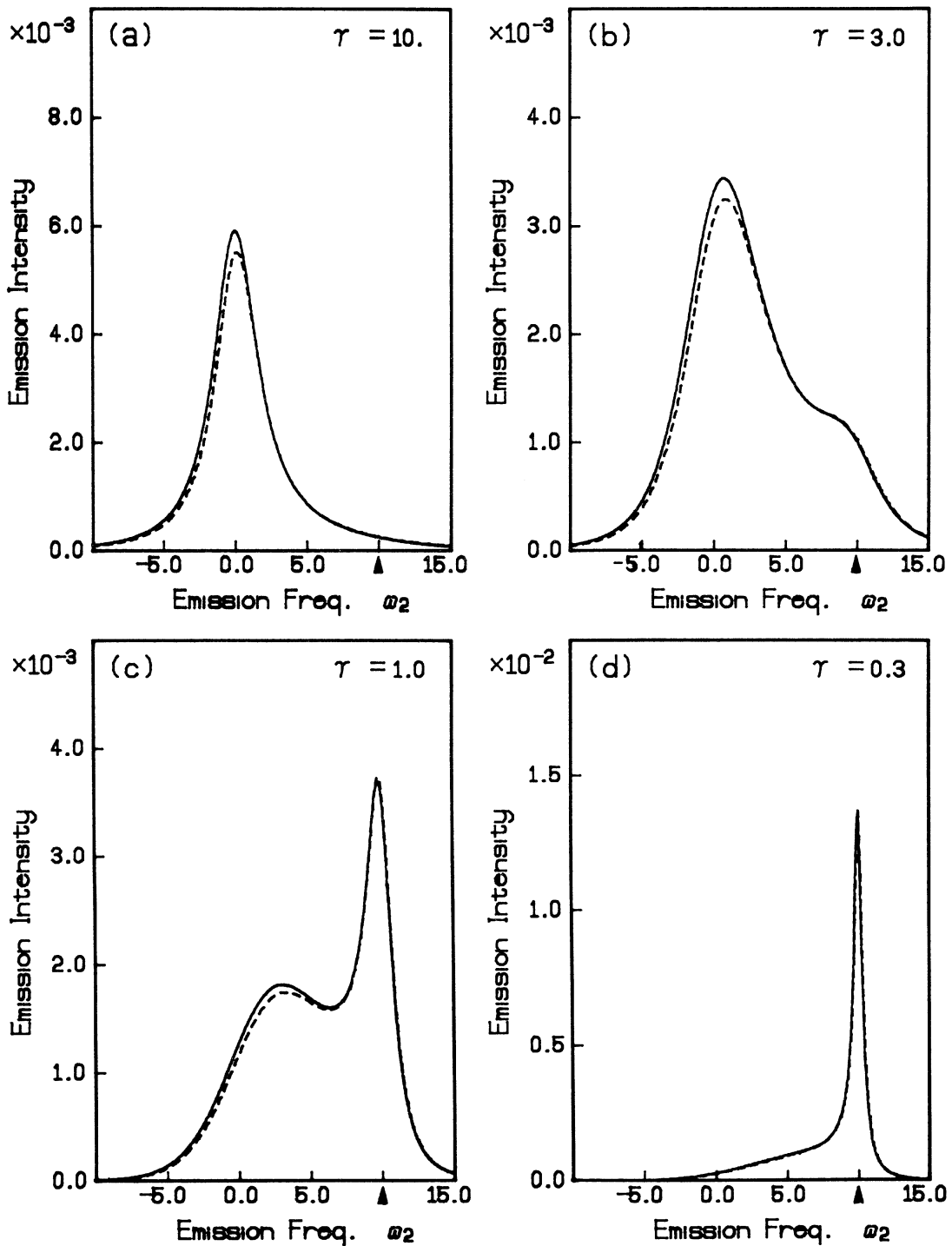


Fig. 3. Emission spectra of a three-level system, respectively, for (a)  $\gamma=10$ , (b)  $\gamma=3.0$ , (c)  $\gamma=1.0$  and (d)  $\gamma=0.3$ . The solid lines and the dashed lines are calculated from eq. (3.3) but the later is neglecting the reaction effects of the bath. The arrows indicate the position of the incident frequency  $\omega_1$ . The  $\delta$ -shaped pure coherent components are not shown here.

$$2\pi \cdot \delta(\omega_1 - \omega_2) \sum_{n=1}^{\infty} n! \Delta^{2n} \left| \prod_{\beta=0}^n S_{\beta}^{-} [\gamma_{\beta} - i\omega_1] \right|^2. \tag{3.15}$$

In this case, we can not distinguish this elements from the pure Raman component eq. (3.10) from its shape and, in this sense, we

may say that the broadened Raman element has a mixed character of coherent and incoherent processes.

Next we discuss the effects of the higher-order interaction with the bath on the power spectrum. Figure 4 shows the components  $I_0$  of eq. (3.4) and  $I_1$  of eq. (3.5) with parameter



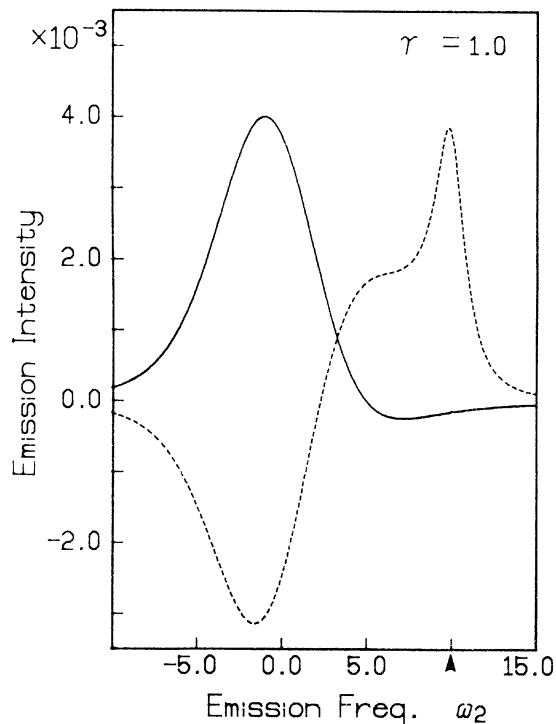


Fig. 4. The components  $I_0$  and  $I_1$  of emission spectrum for  $\gamma=1.0$ . The solid line is  $I_0$  and the dashed is  $I_1$ . The pure Raman component is not shown here and the total emission spectrum is given in Fig. 3(c).

$\gamma=1.0$ . The parameters are chosen in the same as solid line of Fig. 3(b) thereby the total spectrum of Fig. 4 agree with this. As seen in this figure, the component  $I_1$  not only presents the broadened Raman peak but also partially cancels the luminescence peak of  $I_0$ . This cancellation is particularly important for the strong interaction case. Figure 5 is same as Fig. 4, except that  $\gamma=0.3$ . In this case, the negative contribution of  $I_1$  about at the resonant position is almost equal to the luminescence element of  $I_0$  and the total spectrum which is shown in Fig. 3(d) only shows the broadened Raman peak.

#### §4. Summary

We have formulated the second order optical process on the basis of the physical spectrum using the functional integral method. As an application, the CW emission spectrum of the three-level system with the intermediate state interaction is calculated including the reaction effects of the bath. It was shown that the position and the line shape of the luminescence peak is modified by the reaction effects of the bath compared with the result

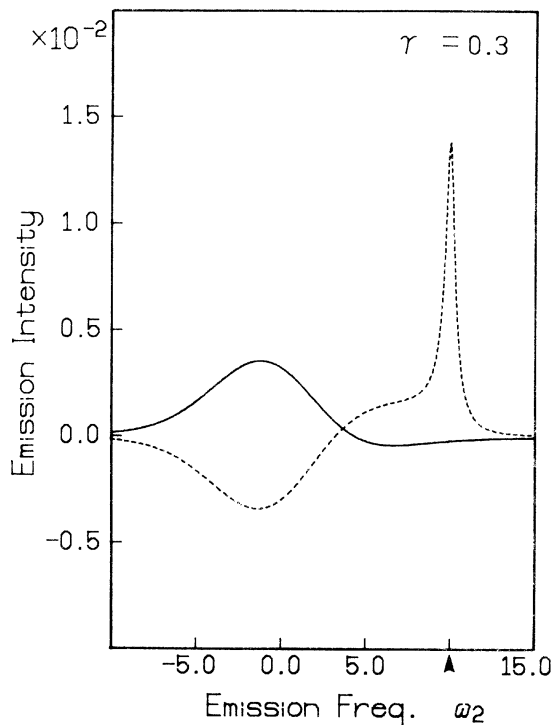


Fig. 5. Same as Fig. 4 except  $\gamma=0.3$ . The total emission spectrum is given in Fig. 3(d).

calculated from the stochastic approach, whereas the peak of the coherent and incoherent mixing process called as the broadened Raman peak is not affected by this effect.

We have also discussed the effects of the higher-order interaction with the bath which has been neglected in the master equation approach. We have shown that the higher-order interaction between the system and the bath causes the broadened Raman peak and the negative contribution of the luminescence. These effects become important particularly for the strong interaction case.

We limited ourselves here to CW response of the second order optical process. Extension of the analysis to the cases of the higher order optical process is interesting. The research of the time dependent spectrum of the pulsive excitation is also necessary to discuss the coherent and incoherent component more detail. We will discuss the detail in the forthcoming papers.

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