物性研究 47−3（1986−12）

修士論文 （1985年度）

Destruction of Quantum Coherence

谷 村 吉 隆
Yoshitaka Tanimura
Department of Instrumentation, Graduate School of Engineering
Keio University

（1986年4月16日受理）

要旨

光と物質の相互作用による量子力学的干渉性の乱れについて研究を行った。論文は2つの部分から成り立っている。始めの部分は，その励起状態において外界とランダムな相互作用している多準位原子について考察した。3準位と4準位原子の2次光学過程の解を解析的に求めた。導出された結果は，ラマン，ラマン類似，ルミネッセンスに対応した3つの項に分類出来る事を示した。スペクトルの全強度に対する表式も導出し，議論を行った。2番目の部分では，まずコヒーレント，ノンコヒーレント部分を分離したスペクトルの一般式を導出した。次に真空場と相互作用している多数の2準位原子からなるマスター方程式をモローの方法を用いて導出しそのスペクトル表式を求めた。1つ及び2つの原子系に対する平衡状態の密度演算子とそのスペクトル解を解析的に求めた。2原子系の場合は配置による依存性があり，それについて議論を行った。

Destruction of Quantum Coherence

Quantum coherence of radiation-matter interaction is investigated. The thesis deals with two problems; First, a stochastic model of a multilevel atom interacting with its environment randomly in its excited states is considered. Analytical expressions of spectra of three- and four-level atoms in the second order optical process are obtained. It is shown that terms in the expressions can be classified into three types corresponding to the Raman, Raman-like and luminescence processes, respectively. The total intensity of emission spectrum is also calculated and discussed. Secondly, the general formula of power spectrum for the coherent and the incoherent parts is derived by using the probability conservation relation. The master equation for a cooperative two-level atomic system which interacts with the vacuum environment is studied by using Mollow's approach and its emission spectrum is calculated. For a single- and coupled-atomic system, the density operators of steady-state and the power spectra are calculated analytically. For the coupled-atomic system, the dependence on an arrangement of atoms is discussed.

谷村吉隆

CHAPTER I

Second Order Optical Process of Random Modulated Multi-Level Atom

§1. Introduction

The Raman scattering and luminescence are both second order optical processes and have been a subject of active study for a long time. The interaction of the system with its environment present in the intermediate states is called an *intermediate state interaction* (IMSI). In the Raman scattering, the quantum coherence is conserved, whereas it is interrupted in the luminescence by IMSI. Thus, a simple three-level atom gives only the Raman scattering if the natural radiative damping is the only mechanism acting in the excited state. The luminescence appears when IMSI destructs the quantum phase coherence in the excited state.[1)-9)] Kubo and his collaborators[10)-15)] have extensively studied various stochastic models, where the perturbation from the environment (reservoir) is regarded as a stochastic Markovian process, for the purpose of understanding the nature of IMSI in the second order optical process. Since the stochastic approach does not rely upon perturbative calculations, it is particularly useful to see how the coherent and non-coherent parts are dependent on the relevant parameters of the problem changing from one extreme to another. However, the results are usually still so much complicated that analytical expressions are not very transparent and one has to appeal to numerical treatments in order to get physical understanding.

The present chapter reports an analysis of three-level and four-level atoms modulated by the simplest two-state jump perturbation which allows straightforward algebraic treatments. This problem was treated by Takagawara some years ago, but his expression of the emission spectra was still complicated and he showed only some numerical examples. We found that the analytical expressions can be separated into a few terms which correspond to different processes with different character of coherence. Although we have not been able to discover the general principle of separation, the achieved separation is by itself very interesting and seems to throw a light into the question of coherent and incoherent parts of the second order process.

This chapter is organized in the following way. The next section is a brief summary of the stochastic theory of IMSI in the second order optical process. In section 3, we calculate the spectrum for a three-level atom with a diagonal modulation and discuss its features. In section 4, the spectrum for a four-level atom with an off-diagonal modulation is calculated and discussed for three different models. The last section is devoted to the summary and conclusion.

§2. Formulation

A formulation of IMSI in second order optical processes in the stochastic approach has been given by Kubo and his collaborators.[10)-15)] In this section, we present a derivation of the expression for emission spectra following the formulation recently described by Kubo in a conference report.[16)] A formula for the total emission intensity is also given.

We consider an atom system S interacting with a radiation field Φ and a reservoir R. The system S has three groups of quantum states, namely the initial state A, the intermediate state B_1, B_2, $\cdots$ and the final state C. Concerning the photon field Φ, the frequency of incident light is denoted by ν_1 and that of emitted light by ν_2. In the second order process, the atom in the state A absorbs a photon ν_1, transfers to one of the intermediate states (if there are more than one such states), and then transfers to the final state C emitting a photon ν_2. We ignore the non-resonant process in which the atom transfers to the intermediate states before absorbing ν_1. Therefore, the energies of the initial, intermediate and the final states of the system $S+\Phi$ are represented by

$$a = A + \nu_1, \qquad b_i = B_i \qquad \text{and} \qquad c = C + \nu_2, \tag{2.1}$$

where we put $\hbar=1$. The interaction of S with the incident light is denoted by V_1 and that with the emitted one by V_2. For simplicity we assume that the system $S+\Phi$ interacts with its reservoir R only in the intermediate states. The IMSI is

denoted by the Hamiltonian H_I. Thus, the Hamiltonian of the total system is written as

$$H_{tot} = H_{S+\Phi} + H_R + H_I + V_1 + V_2, \tag{2.2}$$

when S is in B's, and

$$H_{tot} = H_{S+\Phi} + H_R + V_1 + V_2, \tag{2.3}$$

when S is in A or C. Here, $H_{S+\Phi}$ and H_R denote the Hamiltonians of $S+\Phi$ and R without interaction, respectively.

The density matrix for the total system evolves in time following the equation

$$\dot{\rho}(t) = -i[\, H_{tot}, \rho(t)\,] \equiv -iH^{\times}_{tot}\rho(t), \tag{2.4}$$

where we have introduced the notation

$$O^{\times}X \equiv [\, O, X\,], \tag{2.5}$$

for a hyper-operator $O^{\times}$.

We assume the initial condition for eq.(2.4) as

$$\rho(0) = |a\rangle\langle a|\rho^0_R, \tag{2.6}$$

where ρ^0_R is the equilibrium density matrix of reservoir R satisfying

$$H^{\times}_R\rho^0_R = 0. \tag{2.7}$$

In a stochastic approach, the dynamical evolution of the reservoir R is replaced by a stochastic evolution. Let r denote a state of R and $P_r(t)$ the probability for finding R in the state r at time t. The stochastic evolution of $P_r(t)$ is assumed to follow the Markovian equation

$$\dot{P}_r(t) = -\sum_{r'}\Gamma(r,r')P_{r'}(t). \tag{2.8}$$

Thus, the stochastic operator Γ plays the role of $iH^{\times}_R$ in the foregoing treatment. The IMSI is represented by the interaction Hamiltonian $H_I(r)$ acting on S in the intermediate states when R is in the state r. The dynamical part of $H_I(r)$ is the adiabatic random modulation for each of the intermediate states. In addition to this, there may be off-diagonal (nonadiabatic) modulation between the intermediate

states. It has been shown many years ago by Kubo that the evolution eq.(2.4) is replaced by

$$\dot{\rho}(t) = -i(H^{\times}_{(r)} + V^{\times}_{1} + V^{\times}_{2})\rho(t) - \Gamma\rho(t), \tag{2.9}$$

where $H(r)$ is given by

$$H(r) = H_{S+\Phi} + H_{I}(t), \tag{2.10}$$

when S is in the intermediate states, and

$$H(r) = H_{S+\Phi}, \tag{2.11}$$

when S is in A or C. In eq.(2.9), the density matrix $\rho(t)$ is considered as a vector in the space of reservoir states. The component $\rho_{r}(t)$ is the density matrix of the system S+Φ, R being specified in the state r. The stochastic operator Γ operates on this vector, while the hyper-operators operates on each components of the vector. Hereafter, we denote a quantum state of S+Φ by a bras or kets such as $|a\rangle$, $|b_i\rangle$ and $|c\rangle$ or $\langle a|$, $\langle b_i|$ and $\langle c|$. Similarly state vectors of the reservoir are represented by round bras such as $(r|$, $|r)$, $(P|$ or $|P)$. In this notation

$$\Gamma(r,r') = (r|\,\Gamma\,|r') \tag{2.12}$$

in eq.(2.8) and $P_{r}(t) = (r|P_{(t)})$. The equilibrium state of R is written as $|0)$ or $(0|$ and satisfies

$$\Gamma|\,0) = 0 \qquad \text{and} \qquad (0|\,\Gamma = 0 \tag{2.13}$$

with the normalization

$$(0|0) = 1. \tag{2.14}$$

Then the initial condition (2.6) is written as

$$\rho(0) = |a\rangle\langle a| \cdot |0). \tag{2.15}$$

By solving eq.(2.9) with this condition, we obtain a transition probability P(c,t) that the system has reached the final state c at time t starting from the initial state a. This is given by

$$P(c,t) = (0|\langle c|\rho(t)|c\rangle. \tag{2.16}$$

Per unit time, the rate of emission (photon counting rate) W(c,t) is given by

$$W(c,t) = dP(c,t)/dt. \tag{2.17}$$

By introducing the Laplace transforms of $P(c,t)$ and $W(c,t)$ by

$$P[c,s] = \int_0^\infty dt\ e^{-st}P(c,t) \tag{2.18}$$

and

$$W[c,s] = \int_0^\infty dt\ e^{-st}W(c,t) = sP[c,s], \tag{2.19}$$

the CW response is written as

$$W(c,\infty) = \lim_{t\to\infty} W(c,t) = \lim_{s\to 0} sW[c,s]. \tag{2.20}$$

The total intensity of emission integrated over all values of ν_2 or c is calculated as

$$I = \int_{-\infty}^{\infty} dc\ W(c,\infty). \tag{2.21}$$

Based on eqs.(2.15)-(2.17), we can calculate the CW response in the second order optical process. We expand the formal solution of eq.(2.9) to the second order of V_2:

$$\rho_2(t) = \int_0^t dt' \int_0^{t'} dt\ e^{-i(t-t')(H_{SR}^{X}+V_1^{X})}(-iV_2^{X})e^{-i(t'-t'')(H_{SR}^{X}+V_1^{X})} (-iV_2^{X})e^{-it''(H_{SR}^{X}+V_1^{X})}\rho(0), \tag{2.22}$$

where

$$iH_{SR}^{X} = iH(_r^{X}) + \Gamma. \tag{2.23}$$

From eq.(2.16), the transition probability is given by

$$P(c,t) = \int_0^t dt_2 \int_0^{t_2} dt_1 \langle 0|\langle c|[\ e^{-i(t-t_2)(H_{SR}^{X}+V_1^{X})}V_2 e^{-i(t_2-t_1)(H_{SR}^{X}+V_1^{X})} \{e^{-it_1(H_{SR}^{X}+V_1^{X})}\rho(0)\}V_2 + e^{-i(t-t_2)(H_{SR}^{X}+V_1^{X})}\{e^{-i(t_2-t_1)(H_{SR}^{X}+V_1^{X})}V_2 e^{-it_1(H_{SR}^{X}+V_1^{X})}\rho(0)\}V_2\ ]|c\rangle. \tag{2.24}$$

The photon counting rate can be written as

$$W(c,t) = \int_0^t dt' \ (0|<c|V_2 e^{-i(t-t')(H-c+\Gamma+V_1)} \ e^{-it'(H_{SR}^{\times}+V_1^{\times})}\rho(0)V_2|c> + c.c. \tag{2.25}$$

Then the CW response eq.(2.20) is formally given by

$$W(c,\infty) = \lim_{s\to 0} (0|<c|V_2 \frac{1}{s+i(H-c)+\Gamma+iV_1}\rho^e V_2|c> + c.c. \tag{2.26}$$

where

$$\rho^e = \lim_{s\to 0} \frac{s}{s+i(H_{SR}^{\times}+V_1^{\times})}\rho(0), \tag{2.27}$$

represents the equilibrium-density operator.

We further expand eq.(2.26) to the second order in V_1. First we expand ρ^e. The zeroth order term does not contribute to eq.(2.26). The first and second order terms are given by

$$\begin{aligned}\rho_1^e &= \lim_{s\to 0} \frac{1}{s+iH_{SR}^{\times}} (-iV_1^{\times}) \frac{s}{s+iH_{SR}^{\times}}\rho(0) \\ &= \lim_{s\to 0} \frac{1}{s+iH_{SR}^{\times}}\rho(0)iV_1 + \cdots,\end{aligned} \tag{2.28}$$

$$\begin{aligned}\rho_2^e &= \lim_{s\to 0} \frac{1}{s+iH_{SR}^{\times}} (-iV_1^{\times}) \frac{1}{s+iH_{SR}^{\times}} (-iV_1^{\times}) \frac{s}{s+iH_{SR}^{\times}}\rho(0) \\ &= \lim_{s\to 0} \frac{1}{s+iH_{SR}^{\times}} [\{\frac{1}{s+iH^{\times}+\Gamma}V_1\rho(0)\}V_1 + V_1\{\frac{1}{s+iH^{\times}+\Gamma}\rho(0)V_1\}].\end{aligned} \tag{2.29}$$

where the irrelevant terms are omitted. The first order term eq.(2.28) has the matrix elements $<a|\rho_1^e|b_n>$, while ρ_2^e has elements only between the intermediate states. Next, we expand the first propagator in eq.(2.26) of right hand side in V_1. The zeroth order term combines with ρ_2^e and the first term with ρ_1^e. The final expression is given by[16)]

$$W(c,\infty) = \sum_{ijkl} \lim_{s\to 0} \Big\{ (0|\langle c|V_2|b_j\rangle\langle b_j|\frac{1}{s+\Gamma+\gamma_b+i(H-c)}|b_i\rangle\langle b_i|V_1|a\rangle$$

$$\times \frac{1}{s+\Gamma+i(a-c)}\langle c|V_2|b_l\rangle^*\langle b_l|\frac{1}{s+\Gamma+\gamma_b+i(H-a)}|b_k\rangle^*\langle b_k|V_1|a\rangle^*|0)\Big\} \quad (2.30.\mathrm{I})$$

$$+\sum_{ijkl}\sum_{nm}(0|\langle c|V_2|b_n\rangle\langle b_n|\frac{1}{s+\Gamma+\gamma_b+i(H-c)}|b_l\rangle\Big[\langle b_l b_m|\frac{1}{s+\Gamma+2\gamma_b+iH^{\times}}|b_j b_k\rangle$$

$$\times\langle b_j|\frac{1}{s+\Gamma+\gamma_b+i(H-a)}|b_i\rangle\langle b_i|V_1|a\rangle|0)\langle a|V_1|b_k\rangle\langle b_m|V_2|c\rangle \quad (2.30.\mathrm{II})$$

$$+\langle b_l b_m|\frac{1}{s+\Gamma+2\gamma_b+iH^{\times}}|b_k b_j\rangle\langle b_k|V_1|a\rangle\langle b_j|\frac{1}{s+\Gamma+\gamma_b+i(H-a)}|b_i\rangle^*$$

$$\times\langle b_i|V_1|a\rangle^*\langle b_m|V_2|c\rangle|0)\Big] \quad (2.30.\mathrm{III})$$

$$+\ \mathrm{c.c.}$$

Here, we have introduced the notation

$$\langle b_i b_j|A|b_k b_l\rangle = \langle b_i|\{A|b_k\rangle\langle b_l|\}|b_j\rangle, \quad (2.31)$$

for a hyper-operator A operating on $|b_k\rangle\langle b_l|$. In eq.(2.31), it is assumed that the natural radiation damping of the intermediate states is given by a phenomenological damping constant γ_b for the states B's. As we shall see in later sections, eq.(2.30.I) comes from ρ_1^e and gives rise to the Raman and broadened Raman parts. Whereas eqs.(2.30.II) and (2.30.III) are due to ρ_2^e and gives the luminescence part and at the same time compensates the negative contributions from ρ_1^e.

The total intensity of emission eq.(2.21) can also be calculated as

$$I = \int_{-\infty}^{\infty} dc\ (0|\langle c|V_2\frac{1}{s+i(H-c)+\Gamma+iV1}\rho^e V_2|c\rangle|0) + \mathrm{c.c.}$$

$$= 2\pi\sum_{ijkmn}(0|\langle c|V_2|b_n\rangle\langle b_n b_m|\frac{1}{2\gamma_b+iH^{\times}+\Gamma}|b_j b_k\rangle$$

$$\times\Big[\langle b_j|\frac{1}{\gamma b+i(H-a)}|b_i\rangle\langle b_i|V_1|a\rangle|0)\langle a|V_1|b_k\rangle\langle b_m|V_2|c\rangle$$

$$+\langle b_k|\frac{1}{\gamma b+i(H-a)}|b_i\rangle^*\langle b_j|V_1|a\rangle|0)\langle b_i|V_1|a\rangle^*\langle b_m|V_2|c\rangle\Big]. \quad (2.32)$$

§3. Three-Level Atom with Diagonal Modulation

In this section, we consider a three-level atom with a random level modulation of the intermediate state B. Then, the IMSI Hamiltonian (eqs.(2.10) and (2.11)) is written as

$$H(\Omega) = \begin{bmatrix} a & 0 & 0 \\ 0 & b+\Omega & 0 \\ 0 & 0 & c \end{bmatrix} \tag{3.1}$$

The modulation Ω is further assumed to take only two values $\Omega=\pm\Delta$ randomly with the average jumping rate $\gamma_m=1/\tau_m$(see Fig. 1). This is called a *two-state jump model*. The modulation Ω is expressed in a matrix form as

$$\Omega = \begin{pmatrix} \Delta & 0 \\ 0 & -\Delta \end{pmatrix}, \tag{3.2}$$

for the space of two-state. The time evolution of the random modulation is described by the operator

$$\Gamma = \tfrac{1}{2}\gamma_m \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix}. \tag{3.3}$$

The eigenvectors of Γ corresponding to the equilibrium state are given by

$$|0) = \tfrac{1}{2}\begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad \text{and} \quad (0| = (\,1,\ 1\,). \tag{3.4}$$

Other eigenvectors are

$$|1) = \tfrac{1}{2}\begin{pmatrix} 1 \\ -1 \end{pmatrix} \quad \text{and} \quad (1| = (\,1,\ -1\,). \tag{3.5}$$

These eigenvectors satisfy the following relations:

$$(0|\Omega|0) = (1|\Omega|1) = 0, \quad (0|\Omega|1) = (1|\Omega|0) = \Delta,$$
$$(0|\Gamma|0) = 0 \quad \text{and} \quad (1|\Gamma|1) = \gamma_m. \tag{3.6}$$

From eq.(2.30), the spectrum W is expressed as[17]

$$W = \lim_{s\to 0}\Big\{ (0|\frac{1}{\gamma_b+\Gamma-i(\omega_2-\Omega)}|0)\frac{1}{s+i(\omega_1-\omega_2)}(0|\frac{1}{\gamma_b+\Gamma+i(\omega_1-\Omega)}|0)$$
$$+(0|\frac{1}{\gamma_b+\Gamma-i(\omega_2-\Omega)}|1)\frac{1}{\gamma_m+i(\omega_1-\omega_2)}(1|\frac{1}{\gamma_b+\Gamma+i(\omega_1-\Omega)}|0)$$
$$+(0|\frac{1}{\gamma_b+\Gamma-i(\omega_2-\Omega)}|0)\frac{1}{2\gamma_b}[(0|\frac{1}{\gamma_b+\Gamma+i(\omega_1-\Omega)}|0)+c.c.]$$
$$+(0|\frac{1}{\gamma_b+\Gamma-i(\omega_2-\Omega)}|1)\frac{1}{2\gamma_b+\gamma_m}[(1|\frac{1}{\gamma_b+\Gamma+i(\omega_1-\Omega)}|0)+c.c.]\Big\}$$
$$+c.c. \qquad (3.7)$$

with

$$\omega_1 = a-b \qquad \text{and} \qquad \omega_2 = c-b. \qquad (3.8)$$

Here, we have set $\langle c|V_2|b\rangle=\langle b|V_2|c\rangle=1$, $\langle a|V_1|b\rangle=\langle b|V_1|a\rangle=1$.

The spectrum W can be rearranged into a sum of three terms as (see Appendix)

$$W = W_R + W_{BR} + W_L, \qquad (3.9)$$

with

$$W_R = 2\pi\cdot\delta(\omega_1-\omega_2)\frac{1}{(\gamma_b+\gamma(\omega_1))^2+(\omega_1-\delta(\omega_1))^2}, \qquad (3.10)$$

$$W_{BR} = 2\frac{\gamma_m}{\gamma_m^2+(\omega_1-\omega_2)^2}\cdot\frac{1}{(\gamma_b+\gamma_m)^2+\omega_1^2}\cdot\frac{1}{(\gamma_b+\gamma_m)^2+\omega_2^2}$$
$$\cdot\frac{\Delta^2\{[-\omega_1\cdot\omega_2+(\gamma_b+\gamma_m)^2+\Delta^2]^2+(\gamma_b+\gamma_m)^2\cdot(\omega_1+\omega_2)^2\}}{[(\gamma_b+\gamma(\omega_1))^2+(\omega_1-\delta(\omega_1))^2][(\gamma_b+\gamma(\omega_2))^2+(\omega_2-\delta(\omega_2))^2]} \qquad (3.11)$$

and

$$W_L = 2\left\{W_{a\to b}\frac{\gamma(\omega_1)}{\gamma_b+\gamma(\omega_1)}\cdot\frac{\gamma_m}{\gamma_b+\gamma_m}\right\}\frac{\gamma_b+\gamma_m}{\gamma_b\cdot\gamma_m}\left\{\frac{\gamma_m}{\gamma_b+\gamma_m}\cdot\frac{\gamma(\omega_2)}{\gamma_b+\gamma(\omega_2)}W_{b\to c}\right\}. \qquad (3.12)$$

Here,

$$\gamma(\omega) = \frac{(\gamma_b+\gamma_m)\Delta^2}{(\gamma_b+\gamma_m)^2+\omega^2} \qquad \text{and} \qquad \delta(\omega) = \frac{\omega\Delta^2}{(\gamma_b+\gamma_m)^2+\omega^2} \qquad (3.13)$$

can be interpreted as shifts of the damping rate and the frequency, which are induced by the modulation, respectively. In eq.(3.12) the factors $W_{a\to b}$ and $W_{b\to c}$ represent the absorption and emission rate, which are given by

$$W_{a\to b} = \mathrm{Re}\left(0\left|\frac{1}{\gamma_b+\Gamma+i(\omega_1-\Omega)}\right|0\right) = \frac{\gamma_b+\gamma(\omega_1)}{(\gamma_b+\gamma(\omega_1))^2+(\omega_1-\delta(\omega_1))^2} \tag{3.14}$$

and

$$W_{b\to c} = \frac{\gamma_b+\gamma(\omega_2)}{(\gamma_b+\gamma(\omega_2))^2+(\omega_2-\delta(\omega_2))^2}\,, \tag{3.15}$$

respectively.

From eq.(2.32), the total intensity of emission is calculated as

$$I = 2\pi\cdot W_{a\to b}\frac{1}{\gamma_b}. \tag{3.16}$$

The first term eq.(3.10) represents a pure Raman process. It is composed of a δ-function at $\omega_1=\omega_2$ and the intensity has the resonance form in which the damping and the frequency are modified by the corresponding shifts eq.(3.13) with $\omega=\omega_1$. The second term (3.11) seems to represent the Raman-like process, whereas the third term (3.12) the luminescence.

In the static limit of $\gamma_m\to 0$, the third term (3.12) vanishes and the second term becomes

$$W_{BR} = 2\pi\cdot\delta(\omega_1-\omega_2)\frac{\Delta^2}{[\gamma_b^2+(\omega_1+\Delta)^2][\gamma_b^2+(\omega_1-\Delta)^2]}. \tag{3.17}$$

Then, we have

$$W = W_R + W_{BR} = 2\pi\cdot\delta(\omega_1-\omega_2)\cdot\tfrac{1}{2}\left\{\frac{1}{\gamma_b^2+(\omega_1+\Delta)^2}+\frac{1}{\gamma_b^2+(\omega_1-\Delta)^2}\right\}. \tag{3.18}$$

This is an average of spectra of two pure Raman processes through the intermediate state $B+\Delta$ and $B-\Delta$. Thus, the second term can be interpreted as a part of Raman process in this limit.

For a finite value of γ_m, the luminescence appears and the Raman line of the second term (3.11) is broadened to a Lorentzian form as the result of the first factor of the right hand side. This broading is caused by the nonstatic IMSI, through which the reservoir exchanges energy with the radiation. Thus, we regard it as a Raman-like process and call it a broadened Raman term. The last three

factors of eq.(3.11) determine the intensity. Because the Raman line is broadened, these factors form peaks at frequencies where ω_2 conincides with the energy difference between either of excited states and the final state. It seems to be natural that these peaks are caused by the resonant enhancement of the Raman component. In eq.(3.17), though the last factor also has the maxima at the positions of resonance, this term does not show additional peaks because of the δ-function.

In the motional narrowing limit of $\gamma_m >> 1$ with $\Delta^2/\gamma_m \to \gamma'$, $\delta(\omega)$ vanishes and $\gamma(\omega)$ becomes γ'. Then eq.(3.11) becomes

$$W_{BR} = 2 \cdot W'_{a\to b} \frac{\gamma'}{\gamma_b+\gamma'} \frac{1}{\gamma_b} \frac{\gamma_b}{\gamma_b+\gamma'} W'_{b\to c}, \tag{3.19}$$

where

$$W'_{a\to b} = \frac{\gamma_b+\gamma'}{(\gamma_b+\gamma')^2+\omega_1^2} \quad \text{and} \quad W'_{b\to c} = \frac{\gamma_b+\gamma'}{(\gamma_b+\gamma')^2+\omega_2^2}. \tag{3.20}$$

In this limit, each term of W can be expressed as a product of transition rates and branching factors (See Fig. 2). Thus we have

$$W = 2\pi \cdot \delta(\omega_1-\omega_2) \frac{1}{(\gamma_b+\gamma')^2+\omega_1^2} + 2 \cdot W'_{a\to b} \frac{\gamma'}{\gamma_b(\gamma_b+\gamma')} W'_{b\to c}. \tag{3.21}$$

The pure Raman term W_R becomes the first term of eq.(3.21). The second term represents the luminescence and consists of three factors. The first and final factors correspond to the transition rates from A to B and from B to C by absorption and emission of light, respectively. The second factor is composed of the damping factor $1/\gamma_b$ and a branching ratio $\gamma'/(\gamma_b+\gamma')$ between the Raman and luminescence term in the state B. It is seen that W_{BR} becomes a part of luminescence in this limit. Thus, W_{BR} has both features of the Raman and luminescence processes.

For a finite value of γ_m, the luminescence term eq.(3.12) is also composed of three factors. From the observation in the motional narrowing limit (See Fig. 2), we expect that these factors may correspond to transition rates and branching

ratios. On the contrary, as the quantum coherence of photon is partially maintained, W_{BR} can not be expressed by three factors as eq.(3.19). Thus, the straightforward interpretation based on the branching ratios is not applicable for a finite value of γ_m. If we integrate each term of W eq.(3.9) over ω_2, W_R, W_{BR} and W_L merge into a single term as eq.(3.16), which is a product of the transition rate from the state a to b and the damping rate in the state b.

In the fast modulation limit $\gamma_m \to \infty$, W_{BR} and W_L vanishe. Then, we have

$$W = 2\pi \cdot \delta(\omega_1 - \omega_2) \frac{1}{\gamma_b^2 + \omega_1^2}, \tag{3.22}$$

which is equal to the spectrum in the absence of modulation.

Figures 3 and 4 show the spectral distributions of W_{BR} and W_L normalized by the total intensity eq.(3.14) for a set of parameters with $\omega_1=0.15$, $\Delta=0.5$ and $\gamma_b=0.01$. In Fig. 3, W_{BR} shows a sharp peak at the Raman position $\omega_1=\omega_2$ for $\gamma_m=0.01$. Because of resonance, this term also shows additional peaks at frequencies $\omega_2=\pm\Delta$. The Raman peak declines rather quickly as γ_m increases. Already for $\gamma_m=0.1$, the Raman peak becomes almost unnoticeable. In Fig. 3, the luminescence peaks at $\omega_2 = 0.5$ are distinct for small values of γ_m. When γ_m becomes larger, the additional peaks of W_{BR} and two peaks of W_L merge into a single peak and are motionally narrowed.

§4. Four-Level Atom with Off-Diagonal Modulation

Next we consider a four-level atom with intermediate states B_1 and B_2. The IMSI from the reservoir is assumed to give rise to an off-diagonal interaction between B_1 and B_2. In this case, IMSI Hamiltonian is written as

$$H(\Omega) = \begin{bmatrix} a & 0 & 0 & 0 \\ 0 & b_1 & \Omega & 0 \\ 0 & \Omega & b_2 & 0 \\ 0 & 0 & 0 & c \end{bmatrix}. \tag{4.1}$$

Here, the modulation Ω is assumed to be the same two state jump process as in §3.

The interaction V_1 connects A and B_1. Concerning the interaction V_2 between B's and C, we consider the following three models: V_2 connects C with (I) B_2, (II) B_1, and (III) both B_1 and B_2 (See Fig. 5).

4.1 Model I

From eq.(2.32), W is given by[17]

$$\begin{aligned} W = \lim_{s\to 0} \Bigg(& (0|\langle b_2|\frac{1}{\gamma_b+\Gamma+i(H-c)}|b_1\rangle\frac{1}{s+\Gamma+i(a-c)}\langle b_2|\frac{1}{\gamma_b+\Gamma+i(a-H)}|b_1\rangle|0) \\ & + \sum_{ij=1}^{2}(0|\langle b_2|\frac{1}{\gamma_b+\Gamma+i(H-c)}|b_i\rangle \\ & \times[\langle b_i b_2|\frac{1}{2\gamma_b+\Gamma+iH^{\times}}|b_1 b_j\rangle\langle b_j|\frac{1}{\gamma_b+\Gamma+i(a-H)}|b_1\rangle|0) \\ & + \langle b_i b_2|\frac{1}{2\gamma_b+\Gamma+iH^{\times}}|b_j b_1\rangle\langle b_j|\frac{1}{\gamma_b+\Gamma+i(H-a)}|b_1\rangle|0)]\Bigg) \\ & + \text{c.c.} \end{aligned} \tag{4.2}$$

Where we set $V_1=V_2=1$. Here, the Hamiltonian H of intermediate state is assume to be

$$H = \begin{bmatrix} b_1 & \Omega \\ \Omega & b_2 \end{bmatrix} \tag{4.3}$$

with its quantum states

$$\langle b_1| = [\,1,\,0\,], \qquad \langle b_2| = [\,0,\,1\,]. \tag{4.4}$$

In eq.(4.2), the same notation as eqs.(3.2)-(3.5) are used for stochastic operators and vectors.

The calculation of eq,(4.2) is elementary but tedious. We set

$$\omega_1 = a - \bar{b}, \quad \omega_2 = c - \bar{b}, \quad \omega_0 = b_1 - b_2, \qquad \bar{b} = (b_1+b_2)/2 ,$$

$$\omega_{i\pm} = \omega_i \pm \omega /2. \tag{4.5}$$

Then the final result is written as,

$$W = W_{BR} + W_{L1}, \tag{4.6}$$

where

$$W_{BR} = 2\frac{\gamma_m}{\gamma_m^2+(\omega_1-\omega_2)^2}\frac{1}{(\gamma_b+\gamma_m)^2+\omega_1^2}\cdot\frac{1}{(\gamma_b+\gamma_m)^2+\omega_2^2}$$
$$\cdot\frac{\Delta^2\{[-\omega_{1+}\cdot\omega_{2-}+(\gamma_b+\gamma_m)^2+\Delta^2]^2+(\gamma_b+\gamma_-)^2\cdot(\omega_1+\omega_2)^2\}}{[(\gamma_b+\gamma(\omega_{1+}))^2+(\omega_{1-}-\delta(\omega_{1+}))^2][(\gamma_b+\gamma(\omega_{2-}))^2+(\omega_{2+}-\delta(\omega_{2-}))^2]} \tag{4.7}$$

and

$$W_{L1} = 2\left[W_{a\to b1}\frac{\gamma(\omega_{1+})}{\gamma_b+\gamma(\omega_{1+})}\frac{\gamma_m}{\gamma_b+\gamma_m}\right]\cdot\frac{\gamma_{12}}{\gamma_b(\gamma_b+2\gamma_{12})}\left[\frac{\gamma_m}{\gamma_b+\gamma_m}\frac{\gamma(\omega_{2-})}{\gamma_b+\gamma(\omega_{2-})}W_{b2\to c}\right], \tag{4.8}$$

with

$$W_{a\to b1} = \frac{\gamma_b+\gamma(\omega_{1+})}{(\gamma+\gamma(\omega_{1+}))^2+(\omega_{1-}-\delta(\omega_{1+}))^2} \tag{4.9}$$

and

$$W_{b2\to c} = \frac{\gamma_b+\gamma(\omega_{2-})}{(\gamma+\gamma(\omega_{2-}))^2+(\omega_{2+}-\delta(\omega_{2-}))^2}. \tag{4.10}$$

Here, $\delta(\omega)$ and $\gamma(\omega)$ are defined by eq.(3.13) and γ_{12} is given by

$$\gamma_{12} = \frac{(2\gamma_b+\gamma_m)\Delta^2}{(2\gamma_b+\gamma_m)^2+\omega_0^2}. \tag{4.11}$$

The middle factor of eq.(4.8) coincides with the factor corresponding to the transition rate from B_1 to B_2, which is given by

$$W_{b1\to b2} = (0|\langle b_2 b_2|\frac{1}{2\gamma_b+\Gamma+iH^{\times}}|b_1 b_1\rangle|0) + c.c. = \frac{\gamma_{12}}{\gamma_b(\gamma_b+2\gamma_{12})}. \tag{4.12}$$

In eq.(4.6), the pure Raman process cannot appear, because levels B_1 and B_2 are connected by the modulation Ω and the quantum coherence of photons is dis-

turbed by them. But the broadened Raman process which maintains the quantum coherence not perfectly but partially still exists. In the static limit $\gamma_m \to 0$, its coherence recovers perfectly and W_{BR} becomes

$$W_{BR} = 2\pi \cdot \delta(\omega_1 - \omega_2) \frac{\Delta^2}{[\gamma_b^2 + (\omega_1 + \sqrt{\Delta^2 + \omega_0^2/4})^2][\gamma_b^2 + (\omega_1 - \sqrt{\Delta^2 + \omega_0^2/4})^2]}. \quad (4.13)$$

The right hand side of this equation has the form corresponding to the resonance Raman intensity which shows maxima at two eigenstates of eq.(4.3).

In the motional narrowing limit $\gamma_m >> 1$ with $\Delta^2/\gamma_m \to \gamma'$, the quantum coherence in W_{BR} is completely disturbed by the modulation and W_{BR} is given by

$$W_{BR} = 2 \cdot W'_{a\to b1} \cdot \frac{\gamma'}{\gamma_b + \gamma'} \cdot \frac{1}{\gamma_b} \cdot \frac{\gamma_b}{\gamma_b + \gamma'} \cdot W'_{b2\to c}. \quad (4.14)$$

Here, $W'_{a\to b1}$ and $W'_{b2\to c}$ are expressed as

$$W'_{a\to b1} = \frac{\gamma_b + \gamma'}{(\gamma_b + \gamma')^2 + \omega_{1-}^2} \quad \text{and} \quad W'_{b2\to c} = \frac{\gamma_b + \gamma'}{(\gamma_b + \gamma')^2 + \omega_{2+}^2}. \quad (4.15)$$

Then, $W = W_{BR} + W_{L1}$ is written as

$$W = W^{\bullet}_{a\to b1} \frac{\gamma'}{\gamma_b(\gamma_b + 2\gamma')} W^{\bullet}_{b2\to c}. \quad (4.16)$$

Here, the first and last factors correspond to the absorption process from A to B_1 and emission from B_2 to C, respectively. The middle factor represents the transition process from B_1 to B_2, because γ_{12} in eq.(4.8) becomes γ' in this limits. Thus, eq.(4.16) corresponds to the luminescence process, which represents the transition $A \to B_1 \to B_2 \to C$, and has no such branching as in eq.(3.21).

The total intensity of emission is calculated as

$$I = 2\pi \cdot \left\{ W_{a\to b1} \frac{\gamma(\omega_{1+})}{\gamma_b + \gamma(\omega_{1+})} \frac{\gamma_m}{\gamma_b + \gamma_m} \right\} \cdot \left\{ \frac{\gamma_b + \gamma_m}{\gamma_b \cdot \gamma_m} - \frac{\gamma_{12}}{\gamma_b(\gamma_b + 2\gamma_{12})} \right\}. \quad (4.17)$$

4.2 Model II

This calculation can be done in the same way as in the subsection 4.1. The final results is given by

$$W = W_R + W_{L2}, \quad (4.18)$$

where

$$W_R = 2\pi \cdot \delta(\omega_1 - \omega_2) \frac{1}{(\gamma_b + \gamma(\omega_{1+}))^2 + (\omega_{1-} - \delta(\omega_{1+}))^2}, \tag{4.19}$$

and

$$W_{L2} = 2 \left\{ W_{a\to b1} \frac{\gamma(\omega_{1+})}{\gamma_b + \gamma(\omega_{1+})} \frac{\gamma_m}{\gamma_b + \gamma_m} \right\} \cdot \left\{ \frac{\gamma_b + \gamma_m}{\gamma_b \cdot \gamma_m} - \frac{\gamma_{12}}{\gamma_b(\gamma_b + 2\gamma_{12})} \right\}$$

$$\times \left\{ \frac{\gamma_m}{\gamma_b + \gamma_m} \frac{\gamma(\omega_{2+})}{\gamma_b + \gamma(\omega_{2+})} W_{b1\to c} \right\}. \tag{4.20}$$

Here, $W_{b1\to c}$ is defined by

$$W_{b1\to c} = \frac{\gamma_b + \gamma(\omega_{2+})}{(\gamma_b + \gamma(\omega_{2+}))^2 + (\omega_{2-} - \delta(\omega_{2+}))^2}. \tag{4.21}$$

The factors $(\gamma_b + \gamma_m)/(\gamma_b \cdot \gamma_m)$ and $\gamma_{12}/\gamma_b(\gamma_b + 2\gamma_{12})$ appeared in the middle part of eq.(4.20) coincide with middle part of eq.(3.12) and eq.(4.12), respectively.

In the static limit, W_{L2} vanishes and W is given by

$$W = 2\pi \cdot \delta(\omega_1 - \omega_2) \frac{\gamma_b^2 + \omega_{1+}^2}{[\gamma_b^2 + (\omega_1 + \sqrt{\Delta^2 + \omega_0^2/4})^2][\gamma_b^2 + (\omega_1 - \sqrt{\Delta^2 + \omega_0^2/4})^2]}. \tag{4.22}$$

This equation is similar to eq.(4.13).

In the motional narrowing limit, W becomes

$$W = 2\pi \cdot \delta(\omega_1 - \omega_2) \frac{1}{(\gamma_b + \gamma')^2 + \omega_{1-}^2} + 2 \cdot W'_{a\to b1} \frac{\gamma'}{\gamma_b + \gamma'} \left[\frac{1}{\gamma_b} - \frac{\gamma'}{\gamma_b(\gamma_b + 2\gamma')} \right] \frac{\gamma'}{\gamma_b + \gamma'} W'_{b1\to c} \tag{4.23}$$

where

$$W'_{b1\to c} = \frac{\gamma_b + \gamma'}{(\gamma_b + \gamma')^2 + \omega_{2-}^2}. \tag{4.24}$$

For the general value of γ_m, the total intensity of emission is written as

$$I = 2\pi \cdot W_{a\to b1} \left\{ \frac{1}{\gamma_b + \gamma(\omega_{1+})} + \frac{\gamma(\omega_{1+})}{\gamma_b + \gamma(\omega_{1+})} \cdot \frac{\gamma_m}{\gamma_b + \gamma_m} \cdot \frac{\gamma_{12}}{\gamma_b(\gamma_b + 2\gamma_{12})} \right\}. \tag{4.25}$$

4.3 Model III

The result for this model is obtained by only adding the result of model one and two. Then, W is written by using eqs.(4.7), (4.8), (4.19) and (4.20) as

$$W = W_R + W_{BR} + W_{L1} + W_{L2}. \tag{4.26}$$

The total intensity of emission is written as

$$I = 2\pi \cdot W_{a\to b1} \frac{1}{\gamma_b}. \tag{4.27}$$

This intensity is composed of the factor corresponding to the transition from A to B and the damping factor in B levels as eq.(3.16).

In the static limit, $W=W_R+W_{BR}$ is given by

$$W = 2\pi \cdot \delta(\omega_1-\omega_2)\tfrac{1}{2}\left[\frac{1}{\gamma_b{}^2+(\omega_1+\sqrt{\Delta^2+\omega_0{}^2/4})^2} + \frac{1}{\gamma_b{}^2+(\omega_1-\sqrt{\Delta^2+\omega_0{}^2/4})^2} \right]. \tag{4.28}$$

This equation contains two terms factors corresponding to two eigenstates.

In the motional narrowing limit, W becomes

$$W = 2\pi \cdot \delta(\omega_1-\omega_2) \frac{1}{(\gamma_b+\gamma')^2+\omega_{1-}^2} + 2\cdot W'_{a\to b1}\left\{ \frac{\gamma'}{\gamma_b+\gamma'} \left[\frac{1}{\gamma_b} - \frac{\gamma'}{\gamma_b(\gamma_b+2\gamma')} \right] \frac{\gamma'}{\gamma_b+\gamma'} \cdot W'_{b1\to c} + \frac{\gamma'}{\gamma_b(\gamma_b+2\gamma')} \cdot \frac{\gamma'}{\gamma_b+\gamma'} W'_{b2\to c} \right\}. \tag{4.29}$$

Each term of this equation is composed of the factors corresponding to transitions $A\to B_1$, $B_1\to B_2$, $B_2\to C$, and branching factors as shown in Fig. 6.

For a finite value of γ_m, it seems that each term of eq.(4.26) can be interpreted in the same way as in the motional narrowing limit. However, as there is the quantum coherence in W_{BR}, the simple branching scheme is not applicable.

If we put $\omega_0=0$, the solution eq.(4.26) agree with eq.(3.9). This is easily understood as follows: The eigenstates of H (eq.(4.3)) are given by $|b_\pm\rangle = (|b_1\rangle \pm |b_2\rangle)/\sqrt{2}$ with eigenvalues $b\pm\Delta$. Though V_1 connects $|a\rangle$ with both $|b_+\rangle$ and $|b_-\rangle$, V_2 connects $|c\rangle$ with only $|b_+\rangle$. Thus, the present model reduces to the model discussed in §3 and eq.(4.26) reduce to eq.(3.9).

Figures 7, 8 and 9 illustrate the spectral distributions of W_{BR}, W_{L1} and W_{L2}

normalized by the total intensity eq.(4.27), respectively. We set $\omega_1=0.15$, $\Delta=0.5$, and $\gamma_b=0.01$ as in Figs. 3 and 4. The excited states are separated by $\omega_0=0.3$. For small values of γ_m, W_{L1} and W_{L2} show two peaks at frequencies of two eigenstates of eq.(4.3), respectively. In Fig. 8, the left peak of W_{L2}, which corresponds to the frequency of eigen state near B_2, is higher than the right, because B_2 and C are connected directly by V_2 in the model one. Similarly, the right peak of W_{L2} is higher than the left in Fig. 9. When γ_m becomes larger, two peaks of W_{L1} merge into a single peak at the frequency of $\omega_2=\omega_0/2$ and these of W_{L2} at $\omega_2=-\omega_0/2$, as they are motionally narrowed.

§5. Conclusion

We have shown that each term appeared in the expression of spectrum can be classified into three types. These three types of terms seem to have meaning as physical processes. One of them corresponds to the pure Raman process, which maintains the quantum coherence. Another one is regarded as the luminescence process, where the quantum coherence is perfectly disturbed by the random modulation of IMSI. This type of term can be expressed as a product of three factors, which seemingly correspond to the absorption, relaxation and emission processes, respectively. The third type of term shows the broadened Raman peaks and can be interpreted as the Raman-like process. This term becomes a part of the Raman term in the static limit of Random modulation, but a part of the luminescence term in the motional narrowing limit. It seems to be a reason of these mixing features that the quantum coherence is destroyed only partially by IMSI in this process. The present classification of terms has been derivedby the rearrangement of the complicated formula and is not based on the direct calculation of each physical process. Thus, the correspondence between three types of terms and physical processes such as the Raman and luminescence processes is not very trans-

parent so long as we are concerned with CW response. It will be clearer, if transient responses are treated along the same line. We leave it for future study.

We have also calculated the total intensity of emission and have shown that it is expressed by the transition rate from the initial state to intermediate state.

Appendix

The terms in eq.(3.7) can be calculated by using the following formula

$$(0|\frac{1}{\gamma_b+\Gamma+i(\omega-\Omega)}|0) = (0|\frac{1}{\gamma_b+\Gamma+i\omega}|0)+(0|\frac{1}{\gamma_b+\Gamma+i\omega}|0)(0|i\Omega|1)(1|\frac{1}{\gamma_b+\Gamma+i\omega}|1)$$

$$\times(1|i\Omega|0)(0|\frac{1}{\gamma_b+\Gamma+i\omega}|0) + \cdots$$

$$= (0|\frac{1}{\gamma_b+\Gamma+i\omega-(0|i\Omega|1)(1|\frac{1}{\gamma_b+\Gamma+i\omega}|1)(1|i\Omega|0)}|0)$$

$$= \frac{1}{\gamma_b+\gamma(\omega)+i(\omega-\delta(\omega))} \qquad \text{(A.1)}$$

and

$$(0|\frac{1}{\gamma_b+\Gamma+i(\omega-\Omega)}|1) = (0|\frac{1}{\gamma_b+\Gamma+i(\omega-\Omega)}|0)(0|i\Omega|1)(1|\frac{1}{\gamma_b+\Gamma+i\omega}|1)$$

$$= \frac{1}{\gamma_b+\gamma(\omega)+i(\omega-\delta(\omega))}\cdot\frac{i\Delta}{\gamma_b+\gamma_m+i\omega} \cdot \qquad \text{(A.2)}$$

with

$$\gamma(\omega)=\frac{(\gamma_b+\gamma_m)\Delta^2}{(\gamma_b+\gamma_m)^2+\omega^2} \quad \text{and} \quad \delta(\omega)=\frac{\omega\Delta^2}{(\gamma_b+\gamma_m)^2+\omega^2} \qquad \text{(A.3)}$$

References

1) L. Huber, Phys. Rev. 158, 843 (1967).

2) L. Huber, Phys. Rev. 170, 418 (1967).

3) L. Huber, Phys. Rev. 178, 93 (1967).

4) A. Nitzan and J. Jortner, J. Chem. Phys. 57, 2870 (1972).

5) S. Mukamel, A. Ben-Reuven and J. Jortner, Phys. Rev. A12, 947 (1975).

6) S. Mukamel and A. Nitzan, J. Chem. Phys. 66, 2462 (1977).

7) S. Mukamel, J. Chem. Phys. 71, 2884 (1979).

8) Y. Toyozawa, J. Phys. Soc. Jpn. 41, 400 (1976).

9) A. Kotani and Y. Toyozawa, J. Phys. Soc. Jpn. 41, 1699 (1976).

10) R. Kubo, T. Takagawara and E. Hanamura, Physics of Highly Excited states (Proc. of Oji seminer, Springer Verlag Hedelberg, 304 (1976).

11) T. Takagawara, E. Hanamura and R. Kubo, J. Phys. Soc. Jpn. 43, 802 (1977).

12) T. Takagawara, E. Hanamura and R. Kubo, J. Phys. Soc. Jpn. 43, 811 (1977).

13) T. Takagawara, E. Hanamura and R. Kubo, J. Phys. Soc. Jpn. 43, 1522 (1977).

14) T. Takagawara, E. Hanamura and R. Kubo, J. Phys. Soc. Jpn. 44, 728 (1978).

15) T. Takagawara, E. Hanamura and R. Kubo, J. Phys. Soc. Jpn. 44, 742 (1978).

16) R. Kubo, Pure & Appl. Chem. 57, 201 (1985).

17) T. Takagawara, Dr. Thesis, Dep. Physics., University of Tokyo, (1976).

18) Y. Tanimura, H. Takano and R. Kubo, in preparation.

Figure Captions

Fig. 1. A three-level atom with a diagonal modulation.

Fig. 2. Branching features of the spectral of a three-level atom in the motional narrowing limit. The factors $W'_{a\to b}$ and $W'_{b\to c}$ are defined by eq.(3.20).

Fig. 3. Emission spectra of the broadened Raman term of a three-level atom with a diagonal modulation.

Fig. 4. Emission spectra of the luminescence term of a three-level atom with a diagonal modulation.

Fig. 5. A four-level atom with an off-diagonal modulation. The interaction V_1 connects A and B_1. Three models are considered for V_2 as follows: V_2 connects C with B_2 (I), B_1 (II) and both B_1 and B_2 (I)+(II).

Fig. 6. Branching features of the spectrum of a four-level atom in the motional narrowing limit. The factors $W'_{a\to b1}$, $W'_{b1\to c}$ and $W'_{b2\to c}$ are defined by eqs.(4.15) and (4.24).

Fig. 7. Emission spectra of the broadened Raman term of a four-level atom with an off-diagonal modulation.

Fig. 8. Emission spectra of the luminescence term 1 of a four-level atom with an off-diagonal modulation.

Fig. 9. Emission spectra of the luminescence term 2 of a four-level atom with an off-diagonal modulation.

FIG. 1

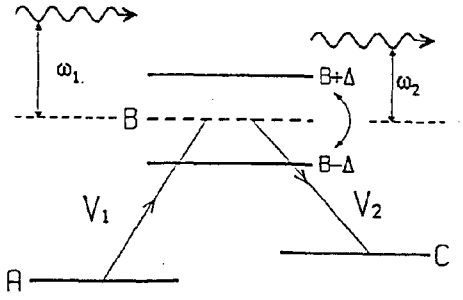


FIG. 2

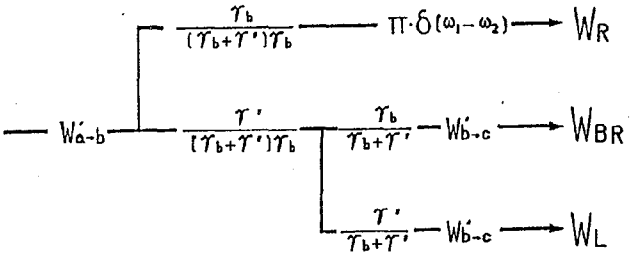


FIG. 3

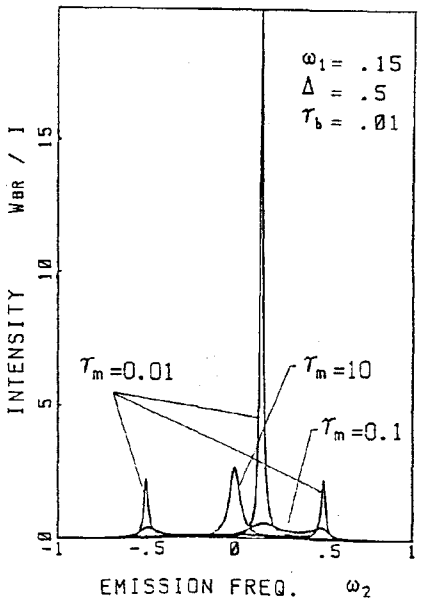


FIG. 4

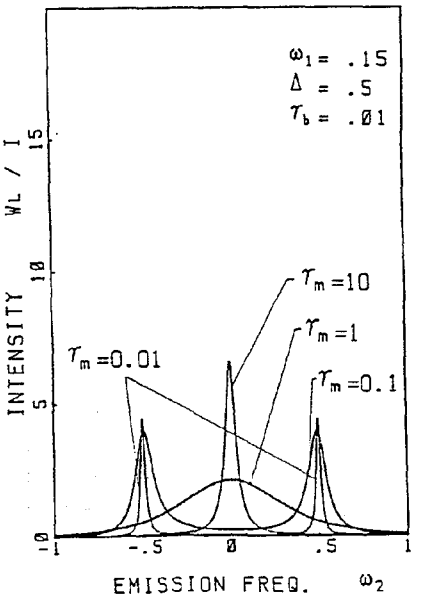


FIG. 5

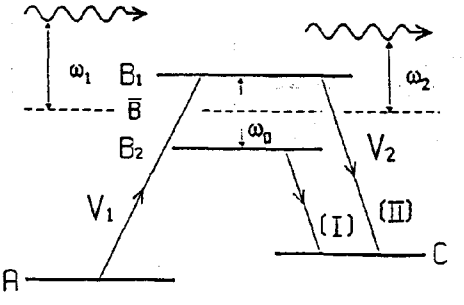


FIG. 6

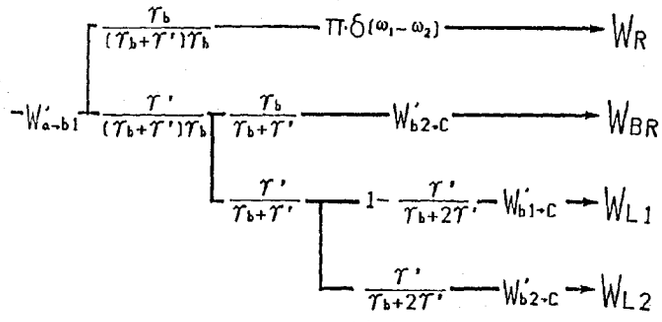


FIG. 7

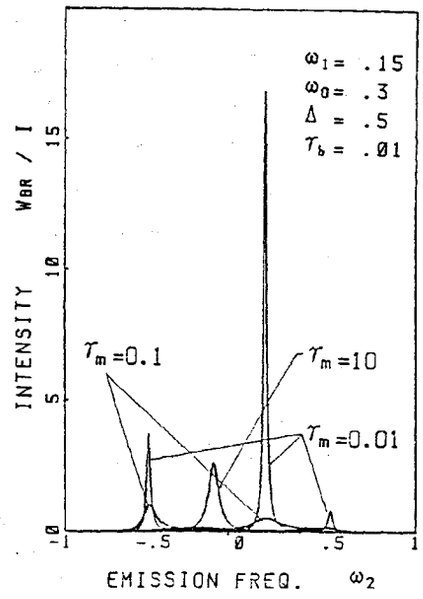


FIG. 8

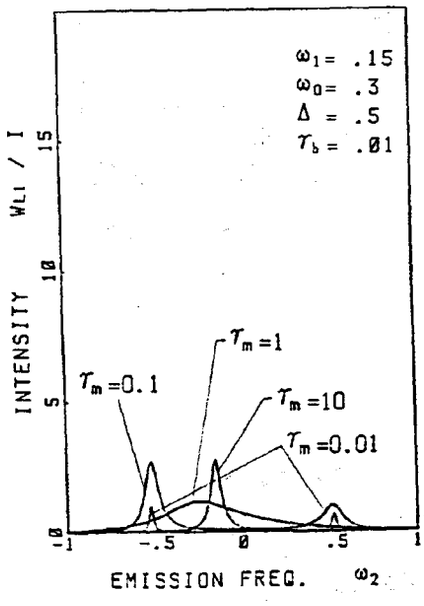
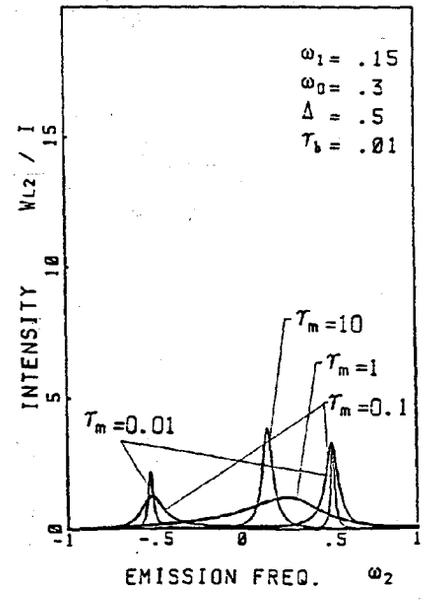


FIG. 9



CHAPTER II

Emission Spectrum of Two-Level Atoms

§1. Introduction

In studying the radiation-matter interaction, one sometimes assumes a system of two-level atoms (molecule) as the simplest model, which has attracted an enormous amount of interest.[1),2)] However, the first theoretical satisfactory description was achieved relatively recently by Mollow.[3)] His paper deals with a single two-level atom driven by a monochromatic electric field. The atom comes into equilibrium with the field through the effect of a "natural radiation damping" induced by the coupling between the atom and the radiation field at zero-temperature. He derived a "master equation" for the reduced density operator of atomic system and calculated its motion. In a subsequent paper,[4)] Mollow investigated the power spectrum of resonant fluorescence for the same model and found the so-called "dynamical Stark effect".[5)]

The master equation for the collective two-level atomic system was first given by Lehmberg[6)-8)] (see also Ref. 9). An interaction between atoms through a bath causes "cooperative dampings"[10)-15)] and "cooperative Lamb shifts"[16)] which depend on the arrangement of atoms. As shown by Dicke,[17)] a system of N two-level atoms may be treated equivalently as a system with angular momentum $J \leqslant N/2$. He used this feature and investigated a spontaneous emission (no driving field) of a collective two-level atomic system with use of the perturbation theory. The main result is the prediction of the "supperradiance" effect.[18)-28)] Concerning this problem, a considerable amount of work on the spontaneous emission has been done with the use of the master equation approach.[29)-41)] All of these calculations are based on the assumption that atoms are distributed over a small region (small-sample model). In this case, cooperative damping agrees with the natural radiation damping and the master equation becomes quite simple.

The emission spectrum of collective atomic system which is driven by the monochromatic electric field was first given by Agarwal et al.[42),43)] They used a small sample model and obtained numerical results for the cases of two and

three atoms. In the subsequent study, Agarwal et al. calculated the same model of two atoms analytically and showed the existence of additional sidebands of spectrum.[44] Several investigations have also been done on this subject.[45)-48)] However, most of the treatments have used the small-sample model and the master equation which includes the cooperative effects more precisely has not been studied.

The main purpose of this chapter is to investigate the power spectrum of cooperative atoms which may be distributed over a region large compared with the resonant wavelength.

This chapter is organized in the following way. In the next section, a general formulation of the coherent and incoherent spectrum is developed. In §3, we derive a master equation by an approach different from that of Lehmberg. Section 4 gives a cooperative emission spectrum. In §5, practical calculations for the models of single atom and two atoms are given. The last section is devoted to the conclusion and brief summary.

§2. Coherent and Incoherent Parts of Spectrum

We consider a system, which is denoted by the Hamiltonian H and its density operator $\rho(t)$. The equation of motion is written as

$$\dot{\rho}(t) = -i\mathcal{L}\rho(t), \tag{2.1}$$

where we have put $\hbar=1$. The quantal Liouville operator $i\mathcal{L}$ is defined by

$$i\mathcal{L}X = i[\,H, X\,] \tag{2.2}$$

for an operator X. The density operator satisfies the probability conservation relation

$$tr\{\rho(t)\} = const, \tag{2.3}$$

since

$$tr\{\dot{\rho}(t)\} = -itr\{H\rho(t)-\rho(t)H\} = 0. \tag{2.4}$$

As is well known, a power spectrum is expressed by the Fourier-Laplace transform of the correlation function of a physical variable, for example, a dipole moment of an atom, or a magnetic moment of a spin. We put its creation and annihilation operator D^+ and D^-, then

$$I(\nu) = \frac{1}{\pi} Re\left\{ \lim_{\varepsilon\to 0} \int_0^\infty dt\ e^{-st} <D^+(t_0)D^-(t_0+t)>|_{s=\ i\nu+\varepsilon} \right\}, \tag{2.5}$$

where $D^{\pm}(t)$ are the Heisenberg operators and their time evolution is determined by the Hamiltonian. We introduce the unitary time-evolution operator $U(t_2,t_1)$ which is defined by the relations

$$\frac{d}{dt_2}U(t_2,t_1) = -iH\,U(t_2,t_1) \tag{2.6}$$

and

$$U(t_1,t_1) = 1. \tag{2.7}$$

Its solution is expressed as

$$U(t_2,t_1) = \exp[-iH(t_2-t_1)]. \tag{2.8}$$

The density operator for the system at time t_2 is then given by

$$\rho(t_2) = U(t_2,t_1)\rho(t_1)U(t_1,t_2). \tag{2.9}$$

This is equivalent to the formal solution of the Liouville equation (2.1)

$$\rho(t_2) = \exp[-i\mathcal{L}(t_2-t_1)]\rho(t_1). \tag{2.10}$$

The Heisenberg operator $D(t)$ is also expressed by

$$D(t_2) = U(t_1,t_2)D(t_1)U(t_2,t_1), \tag{2.11}$$

with

$$D(0) = D. \tag{2.12}$$

The correlation function appearing in eq.(2.5) is now written as [49]

$$\begin{aligned} <D^+(t)D^-(t')> &= Tr\{U(0,t)D^+U(t,0)U(0,t')D^-U(t',0)\rho(0)\} \\ &= Tr\{D^-U(t',t)\rho(t)D^+U(t,t')\} \\ &= Tr\{D^-\exp[-i\mathcal{L}(t'-t)](\rho(t)D^+)\}. \end{aligned} \tag{2.13}$$

The power spectrum eq.(2.5) is then give by

$$I(\nu) = \frac{1}{\pi}\mathrm{Re}\left\{ \lim_{\varepsilon\to 0} \mathrm{tr}\{D^{-}(s+i\mathcal{L})^{-1}(\rho^{e}D^{+})\}\Big|_{s=\, i\nu+\varepsilon}\right\}, \tag{2.14}$$

where we have replaced the density operator to the equilibrium one, assuming that after a long time the system has reached the equilibrium state uniquely. The equilibrium density operator satisfies the relation

$$-i\mathcal{L}\rho^{e} = 0 \tag{2.15}$$

and is expressed in the Laplace transform as

$$\rho^{e} = \lim_{t_0\to\infty} \rho(t_0) = \lim_{s\to 0} s\rho[s], \tag{2.16}$$

where

$$\rho[s] = \int_0^{\infty} dt\; e^{-st}\rho(t). \tag{2.17}$$

The evaluation of the hyper-operator $i\mathcal{L}$ is given in Appendix A.

Note that eq.(2.14) can be interpreted as the time evolution of the system. The state of the system is initially in the equilibrium, then the creation operator D^{+} makes it nonequilibrium (excited) one. That state evolves in time followed by the resolvent $(s+i\mathcal{L})^{-1}$. Finally, the annihilation operator D^{-} returns the evolved excitation to the equilibrium.

Now, we derive a general formula of power spectrum for the coherent and incoherent scattering. We put constant in eq.(2.3) is unity. Then,

$$\mathrm{tr}\{\rho(t_0)\} = 1. \tag{2.18}$$

We assume that this equation is only one restriction relation between the elements of the density operator. Let us consider the projection operator Q which projects the density operator onto the equilibrium. We define Q by

$$Q = \rho^{e}\cdot\mathrm{tr}\{ \tag{2.19}$$

We also define another projection operator Q' by

$$Q' = (1-Q) = 1 - \rho^{e}\mathrm{tr}\{ \tag{2.20}$$

It can be shown that Q and Q' satisfy the following relations

$$Q^2 = Q, \tag{2.21}$$

$$Q'^2 = Q', \tag{2.22}$$

and

$$QQ' = Q'Q = 0. \tag{2.23}$$

From eq.(2.15), Q and Q' also satisfy

$$Qi\mathcal{L} = i\mathcal{L}Q = 0 \tag{2.24}$$

and

$$Q'i\mathcal{L} = i\mathcal{L}Q' = i\mathcal{L}. \tag{2.25}$$

The density operator is decomposed into two components

$$\rho(t) = \rho^e + Q'\rho(t). \tag{2.26}$$

By using eqs.(2.24) and (2.26), the Laplace transform of eq.(2.1) is expressed as

$$\begin{aligned}\rho[s] &= (s+i\mathcal{L})^{-1}\rho(t_0) \\ &= \frac{1}{s}\rho^e + (s+i\mathcal{L})^{-1}Q'\rho(t_0).\end{aligned} \tag{2.27}$$

Then we can write the resolvent $(s+i\mathcal{L})^{-1}$ as

$$(s+i\mathcal{L})^{-1} = \frac{1}{s}\mathcal{Y} + \mathcal{G}[s], \tag{2.28}$$

where

$$\mathcal{Y} = \rho^e \mathrm{tr}\{ \tag{2.29}$$

and

$$\mathcal{G}[s] = (s+i\mathcal{L})^{-1}Q'. \tag{2.30}$$

Equation (2.30) satisfies

$$\lim_{s\to 0} s\,\mathcal{G}[s] = 0. \tag{2.31}$$

therefore, the operator $\mathcal{G}[s]$ has no element proportional to 1/s.

The formal solution of eq.(2.1) is then given by

$$\rho(t) = \rho^e + \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} ds\, e^{st}\mathcal{G}[s]\rho(t_0), \tag{2.32}$$

where c is taken to the right of all singularities in integral. This solution has separated the equilibrium part from the unsteady-state one. The operator $\mathcal{G}[s]$ denotes the time evolution of the unsteady-state part of $\rho(t_0)$ and when the density operator coincides with the equilibrium one, the second term becomes zero. Hereafter, we call $\mathcal{G}[s]$ the "unsteady-evolution operator".

Above discussion also allows us to separate the power spectrum into two parts. From eqs.(2.28)-(2.30), we may write eq.(2.14) as the form

$$I(\nu) = \delta(\nu) I^{coh} + I^{incoh}_{(\nu)}, \tag{2.33}$$

where

$$I^{coh} = tr\{D^- \rho^e\} \cdot tr\{\rho^e D^+\}, \tag{2.34}$$

and

$$I^{incoh}_{(\nu)} = \frac{1}{\pi} \mathrm{Re}\ tr\{D^- \mathcal{G}[s](\rho^e D^+)\}\Big|_{s=i\nu}. \tag{2.35}$$

The first term eq.(2.34) corresponds to the coherent part of the power spectrum and is proportional to the square of the absolute value of the induced dipole moment. The second term eq.(2.35) is the incoherent part. Its form represents the time evolution of the system as like as eq.(2.14), but the evolution is now governed by the "unsteady-evolution operator" $\mathcal{G}[s]$.[51] The equilibrium density operator and hyper-operators eqs.(2.29) and (2.30) are evaluated in Appendix B.

§3. Master Equation for Two-Level Atoms

We derive a master equation describing the dynamics of two-level atoms which are interacting with a vacuum bath and classical (c-number) electric field by using Mollow's method.[3] This equation has been obtained by Lehmberg[7] by integrating the Heisenberg equation of motion directly. Our derivation is essentially similar to Agarwal.[38]

3.1 Hamiltonian

Let us consider a system A of N two-level atoms at positions $\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_N$ The excited and ground states of j-th atom which are separated by energy ω_0 (here after we put $\hbar=1$) are denoted by $|1\rangle_j$ and $|0\rangle_j$, respectively. For these states, annihilation and creation operators are written as

$$a_j = |0\rangle_{jj}\langle 1|, \tag{3.1}$$

and

$$a_j^+ = |1\rangle_{jj}\langle 0|. \tag{3.2}$$

These atoms are supposed to be driven by classical (c-number) external field resonant or quasi-resonant with the atomic frequency ω_0. At the position $\mathbf{r}_j$, the field is given by

$$\mathbf{E}_j(t) = \mathcal{E}[e^{i(\mathbf{k}\cdot\mathbf{r}_j-\omega t)} + e^{i(-\mathbf{k}\cdot\mathbf{r}_j+\omega t)}]\mathbf{e}_0/\sqrt{2}, \tag{3.3}$$

where ω, $\mathbf{k}$, $\mathcal{E}$ and $\mathbf{e}_0$ are the frequency, wave number vector, field amplitude and polarization vector, respectively. Atoms also interact with each other through dipole-dipole interaction. The dipole moment operator for the j-th atom may be expressed in terms of the dipole matrix element $\mathbf{d}_j = {}_j\langle 1|\boldsymbol{\mu}|0\rangle_j$ as

$$\boldsymbol{\mu}_j = \mathbf{d}_j(a_j^+ + a_j), \tag{3.4}$$

in which we have assumed that with proper choice of wave function $\mathbf{d}_j$ can be made real.

The Hamiltonian for the system A is given by

$$H_A(t) = H_S + H_d + H_E(t), \tag{3.5}$$

where H_s is the atomic energy

$$H_S = \sum_j \omega_0 a_j^+ a_j , \tag{3.6}$$

and H_d is the dipole-dipole interaction

$$H_d = \sum_{jk} V_{jk} a_j^+ a_k , \qquad (j \neq k) \tag{3.7}$$

with

$$V_{jk} = \frac{1}{r_{jk}^3} [(\mathbf{d}_j \cdot \mathbf{d}_k) r_{jk}^2 - 3(\mathbf{d}_j \cdot \mathbf{r}_{jk})(\mathbf{d}_k \cdot \mathbf{r}_{jk})] . \qquad (\mathbf{r}_{jk} = \mathbf{r}_j - \mathbf{r}_k) \tag{3.8}$$

The interaction between electric field and atoms is expressed by the "rotating-wave approximation" (RWA) form as

$$H_E(t) = - \sum_j (R_j e^{i\omega t} a_j + R_j^* e^{-i\omega t} a_j^+) , \tag{3.9}$$

where we define the complex Rabi frequencies as

$$R_j = \frac{1}{\sqrt{2}} \mathcal{E} (\mathbf{d}_j \cdot \mathbf{e}_0) e^{i\mathbf{k}\cdot\mathbf{r}_j} . \tag{3.10}$$

Now, we assume that the system A of atoms are coupled to a bath system B. The energy of bath system is then given by

$$H_B = \sum_q \omega_q b_q^+ b_q . \tag{3.11}$$

where annihilation and creation operators b_q and b_q^+ satisfy the commutation relations

$$[\, b_q , b_{q'}^+ \,] = \delta_{qq'} . \tag{3.12}$$

For the combined system A+B, the Hamiltonian is now written as

$$H(t) = H_0(t) + H_I , \tag{3.13}$$

where $H_0(t)$ is the unperturbed Hamiltonian with respect to the bath

$$H_0(t) = H_A(t) + H_B , \tag{3.14}$$

and interaction H_I is assumed to be have RWA form

$$H_I = -i \sum_{jq} (g_{jq} a_j^+ b_q - g_{jq}^* a_j b_q^+) , \tag{3.15}$$

in which g_q's denote coupling constants.

3.2 Derivation of Master Equation

The density matrix for the A+B system is denoted by $\rho(t)$, which follows the equation of motion

$$\dot{\rho}(t) = -i\mathcal{L}_H(t)\rho(t), \tag{3.16}$$

where $\mathcal{L}_H(t)$ is the quantal Liouville operator acting on a Hilbert space operator X as commutator with the Hamiltonian operator:

$$-i\mathcal{L}_H(t)X = -i[\, H_0(t)+H_I,\ X\,]. \tag{3.17}$$

Here, we introduce an unperturbed time evolution operator $U_0(t)$, which is defined by

$$\dot{U}_0(t) = -iH_0(t)U_0(t), \tag{3.18}$$

and

$$U_0(t) = 1. \tag{3.19}$$

Its solution is written as

$$U_0(t) = \exp_{\leftarrow}[-i\int_0^t H_0(\tau)d\tau] \tag{3.20}$$

in the ordered exponential form. Then, the interaction picture operators $\rho'(t)$ and $H_I(t)$ are defined as

$$\rho'(t) = \bar{U_0}(t)\rho(t)U_0(t), \tag{3.21}$$

and

$$H_I(t) = U_0(t)H_I\bar{U_0}(t). \tag{3.22}$$

In this picture, eq.(3.16) becomes

$$\dot{\rho}'(t) = -i\mathcal{L}_I(t)\rho'(t), \tag{3.23}$$

or equivalently

$$\rho'(t) = \rho'(t') - i\int_{t'}^{t} dt_1\, \mathcal{L}_I(t_1)\rho'(t_1), \tag{3.24}$$

where

$$\mathcal{L}_I(t)X = [\, H_I(t),\ X\,]. \tag{3.25}$$

The difference

$$\Delta\rho'(t) = \rho'(t+\Delta t) - \rho'(t), \tag{3.26}$$

can be calculated from eq.(3.24) by iterating itself

$$\Delta\rho'(t) = -i\int_t^{t+\Delta t} dt_1\, \mathcal{L}_I(t_1)\rho'(t) - \int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2\, \mathcal{L}_I(t_1)(\mathcal{L}_I(t_2)\rho'(t)) + \cdots. \tag{3.27}$$

where Δt is a time smaller than the time scale characterizing the system. Let us assume that the bath B is initially at zero temperature and atoms are in an arbitrary mixed state. The initial density operator for the system is then given by

$$\rho(0) = \rho'(0) = \rho_A'(0)|0\rangle_{BB}\langle 0|. \tag{3.28}$$

We further assume that the excitations which are induced in the bath by its interaction with the atoms remain so small in the characteristic time of the atom system. Then, the bath may be approximated at any time by its initial state, and the density operator for system of atoms and bath at time t is written as

$$\rho'(t) = \rho_A'(t)|0\rangle_{BB}\langle 0|. \tag{3.29}$$

The reduced density operator ρ_A for the system A is given by

$$\rho_A'(t) = \mathrm{Tr}_B\{\rho'(t)\}. \tag{3.30}$$

The relation between the Schrödinger picture and the interaction picture is now expressed as

$$\rho_A'(t) = U_0'(t)\rho_A(t)U_0'^{-}(t), \tag{3.31}$$

where

$$U_0'(t) = \exp_{\leftarrow}\left[-i\int_0^t H_A(\tau)d\tau\right]. \tag{3.32}$$

From eq.(3.27), keeping terms up to the second order in $\mathcal{L}_I(t)$, we obtain

$$\Delta\rho_A'(t) = -i\mathrm{Tr}_B\left\{\int_t^{t+\Delta t} dt_1\, \mathcal{L}_I(t_1)\rho_A'(t)\right\} - \mathrm{Tr}_B\left\{\int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2\, \mathcal{L}_I(t_1)(\mathcal{L}_I(t_2)\rho_A'(t))\right\}. \tag{3.33}$$

In the right hand side of this equation, terms involving

$$Tr_B\{b_q|0\rangle_{BB}\langle 0|\}, \quad Tr_B\{b_q|0\rangle_{BB}\langle 0|b_q^+\}, \cdots\cdots, \tag{3.34}$$

and

$$Tr_B\{b_q^+|0\rangle_{BB}\langle 0|b_{q'}\}, \quad Tr_B\{b_q b_{q'}^+|0\rangle_{BB}\langle 0|\}, \cdots\cdots, \tag{3.35}$$

(where $q\neq q'$) vanish by taking the trace over the bath states. Thus, we have

$$\Delta\rho_A'(t) = \Delta t\sum_{jkq} g_{jq}^* g_{kq}\{\int_{-\infty}^{\infty} d\tau e^{i(\omega_q-\omega_0)\tau} a_j\rho_A'(t)a_k^+ - \int_0^{\infty} d\tau\,[e^{-i(\omega_q-\omega_0)\tau}a_j^+a_k\rho_A'(t) + e^{i(\omega_q-\omega_0)\tau}\rho_A'(t)a_j a_k^+]\}, \tag{3.36}$$

where we have evaluated the double integral of the second term of eq.(2.33) as (see Appendix C)

$$\int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2\, \mathcal{L}_I(t_1)(\mathcal{L}_I(t_2)\rho_A'(t)) = \Delta t\int_0^{\infty} d\tau\, \mathcal{L}_I(t+\tau)(\mathcal{L}_I(t)\rho_A'(t)). \tag{3.37}$$

We will now take the limit $L^3\to\infty$ so that the summation over q should be replaced by an integral over the continuum of modes, i.e.

$$\sum_q \to (\frac{L}{2\pi c})^3\int d\omega\int d\Omega\, \omega^2, \tag{3.38}$$

where $\omega=ck$ and $d\Omega$ denote a solid angle. We introduce

$$N(\omega) = \frac{\omega^2}{2\pi c^3}, \tag{3.39}$$

and

$$g^2(\omega, r_{jk}) = \frac{L^3}{4\pi^2}\int d\Omega\, g_{jq}^* g_{kq}. \tag{3.40}$$

Then dividing eq.(2.36) by Δt, we get the "coarse-grained" time derivation of ρ_A in the form

$$\dot{\rho}_A(t) = \sum_{jk}\{K_{jk}^0 a_j\rho_A(t)a_k^+ - (K_{jk}+i\delta_{jk})a_j^+a_k\rho_A(t) - (K_{jk}-i\delta_{jk})\rho_A(t)a_j^+a_k\}. \tag{3.41}$$

Here, we put

$$K_{jk}^0 = \int d\omega\int_{-\infty}^{\infty} d\tau\, N(\omega)g^2(\omega, r_{jk})e^{-i(\omega-\omega_0)\tau} \tag{3.42}$$

and

$$K_{jk}+i\delta_{jk} = \int d\omega \int_0^\infty d\tau\, N(\omega) g^2(\omega, r_{jk}) e^{-i(\omega-\omega_0)\tau} . \tag{3.43}$$

More explicitly, these parameters are then given by

$$\tfrac{1}{2}K^0_{jk} = K_{jk} = \pi N(\omega) g^2(\omega_0, r_{jk}) , \tag{3.44}$$

and

$$\delta_{jk} = -P\int d\omega \frac{N(\omega) g^2(\omega, r_{jk})}{\omega - \omega_0} . \tag{3.45}$$

Transforming eq.(3.41) to the Schrödinger picture, we have

$$\begin{aligned}\dot{\rho}_A(t) = \sum_{jk} (&K^0_{jk} a_j \rho_A(t) a_k^+ - K_{jk}\{ a_j^+ a_k, \rho_A(t) \} - i\delta_{jk}[a_j^+ a_k, \rho_A(t)]) \\ &+ i\sum_j [R_j e^{i\omega t} a_j + R_j^* e^{-i\omega t} a_j^+, \rho_A(t)] - i\sum_{jk} V_{jk}[a_j^+ a_k, \rho_A(t)] \\ &- i\sum_j \omega_0 [a_j^+ a_j, \rho_A(t)] ,\end{aligned} \tag{3.46}$$

where $\{\}$ denotes an anticommutator.

In the following, we consider that the system B represents the electromagnetic field into which atoms radiate (vacuum field). The coupling constants are now given by

$$g_{jk\lambda} = (2\pi c k/L^3)^{\frac{1}{2}} (\mathbf{d}_j \cdot \mathbf{e}_{k\lambda}) e^{i\mathbf{k}\mathbf{r}_j} . \tag{3.47}$$

Here, L^3 is the volume in which the field is enclosed and will eventually be taken to infinity, and $\mathbf{e}_{k\lambda}$ is the polarization vector possessing the property

$$\sum_\lambda \mathbf{e}^\alpha_{k\lambda} \cdot \mathbf{e}^\beta_{k\lambda} = \delta_{\alpha\beta} - \hat{\mathbf{k}}_\alpha \cdot \hat{\mathbf{k}}_\beta \qquad (\alpha, \beta = x, y, z) , \tag{3.48}$$

where $\hat{\mathbf{k}}$ is the unit vector in the direction $\mathbf{k}$.

Then, the damping factors K^0_{jk} and K_{jk}, are calculated as (see Appendix D)

$$\kappa = \tfrac{1}{2}K^0_{jj} = K_{jj} = 2d^2\omega_0^3/(3\pi c^3) , \tag{3.49}$$

and

$$\gamma_{jk} = \tfrac{1}{2}K^0_{jk} = K_{jk} = \kappa W_{jk}(\omega_0) \qquad (j \neq k) , \tag{3.50}$$

Here, W_{jk} is defined by

$$W_{jk}(\omega) = \frac{3}{2}\left\{A_{jk}\frac{\sin(\omega r_{jk}/c)}{(\omega r_{jk}/c)} + B_{jk}\left[\frac{\cos(\omega r_{jk}/c)}{(\omega r_{jk}/c)^2} - \frac{\sin(\omega r_{jk}/c)}{(\omega r_{jk}/c)^3}\right]\right\}, \tag{3.51}$$

with

$$A_{jk} = [(\mathbf{d}_j\cdot\mathbf{d}_k) - (\mathbf{d}_j\cdot\mathbf{r}_{jk})(\mathbf{d}_k\cdot\mathbf{r}_{jk})/r_{jk}^2]/d^2, \tag{3.52}$$

and

$$B_{jk} = [(\mathbf{d}_j\cdot\mathbf{d}_k) - 3(\mathbf{d}_j\cdot\mathbf{r}_{jk})(\mathbf{d}_k\cdot\mathbf{r}_{jk})/r_{jk}^2]/d^2. \tag{3.53}$$

In eqs.(3.49)-(3.53), It is assumed that $d = |\mathbf{d}_i|$. The constant κ is responsible for the "natural radiation damping" and the γ_{jk} the "cooperative damping" which are due to the presence of other atoms.[10)-15)] From eqs.(2.45), we may be evaluated the "cooperative frequency shift" $i\delta_{jk}$.[16)] Using a contour integral, we obtain

$$\delta_{jk} = \kappa V_{jk}(\omega_0), \qquad (j\neq k) \tag{3.54}$$

where

$$V_{jk}(\omega) = \frac{3}{2}\left\{A_{jk}\frac{\cos(\omega r_{jk}/c)}{(\omega r_{jk}/c)} - B_{jk}\left[\frac{\sin(\omega r_{jk}/c)}{(\omega r_{jk}/c)^2} + \frac{\cos(\omega r_{jk}/c)}{(\omega r_{jk}/c)^3}\right]\right\}. \tag{3.55}$$

The "Lamb shift" $i\delta_{jj}$ would diverge logarithmically were it not for the frequency cut off. It's value is not shown here, as it can be renormalized into ω_0. The master equation (3.46) now becomes

$$\dot{\rho}_A(t) = -i\mathcal{L}_A(t)\rho_A(t), \tag{3.56}$$

where

$$\begin{aligned} -i\mathcal{L}_A(t)X = &\sum_j (2\kappa a_j X a_j^+ - \kappa\{a_j^+a_j, X\} - i\omega_0[a_j^+a_j, X]) \\ &+ \sum_{jk} (2\gamma_{jk} a_j X a_k^+ - \gamma_{jk}\{a_j^+a_k, X\} - i\Omega_{jk}[a_j^+a_k, X]) \\ &+ i\sum_j [R_j e^{i\omega t}a_j + R_j^* e^{-i\omega t}a_j^+, X]. \qquad (j\neq k) \end{aligned} \tag{3.57}$$

In eq.(3.57), we put

$$\omega_0 + \delta_{jj} \to \omega_0 \qquad \text{and} \qquad \delta_{jk} + V_{jk} \to \Omega_{jk}. \tag{3.58}$$

§4. Power Spectrum of Emission Field

4.1 Correlation function

For a general field operator A, let A^+ denote the positive frequency parts and A^- the negative frequency part, that is

$$A = A^+ + A^- . \tag{4.1}$$

Quantum electrodynamics tell us that an electric field operator $E_{(r,t)}$, which is the solution to the Maxwell equations, is expressed in the Heisenberg picture as[38),50)]

$$E^{\pm}(r,t) = E_f^{\pm}(r,t) + \nabla\times\nabla\times Z^{\pm}(r,t), \tag{4.2}$$

where $E_f^{\pm}$ is the homogeneous solution and $Z^{\pm}$ is the Hertz dipole operator

$$Z^{+}(r,t) = \sum_{j=1}^{n} d_j a_j(t-|r-r_j|/c)/|r-r_j| . \tag{4.3}$$

In the scattering region $|r|>>|r_j|$, the distance $|r-r_j|$ may be written as

$$|r - r_j| = r - \hat{r}\cdot r_j , \tag{4.4}$$

where $r=r\hat{r}$. We must take care to approximate the operator $a_j(t)$, as it has the rapidly varying phase factor. To separate out the unperturbed time dependence (rapidly varying part), We write

$$a_j(t) = a'_j(t)e^{-i\omega_0 t} . \tag{4.5}$$

This separation allows us to make the approximation.

$$a'_j(t-\hat{r}\cdot(r-r_j)/c) = a'_j(t-r/c) . \tag{4.6}$$

Then we may write

$$a_j(t-|r-r_j|/c) = a_j(t-r/c)e^{i\omega_0 r\cdot r_j/c} . \tag{4.7}$$

The Hertz dipole operator is now given by

$$Z^{+}(r,t) = \sum_{j=1}^{n} d_j e^{i\omega_0 r\cdot r_j/c} a_j(t-r/c)/r, \tag{4.8}$$

and the emission field eq.(4.2) may be expressed in the scattering region as[26]

$$\mathbf{E}^{+}(\mathbf{r},t) = \mathbf{E}_{f}^{+}(\mathbf{r},t) - \sum_{j=1}^{n} \frac{\omega_0}{4\pi c r}[(\mathbf{d}_j \times \mathbf{r}) \times \mathbf{r}] e^{i\omega_0 \mathbf{r}\cdot\mathbf{r}_j/c} a_j(t-r/c). \qquad (4.9)$$

Since the field is initially in the vacuum state, the homogeneous field operator $\mathbf{E}_{f}^{\pm}(\mathbf{r},t)$ will not contribute to the normally ordered field correlation function:

$$G(t,t') = \langle \mathbf{E}^{-}(\mathbf{r},t)\cdot\mathbf{E}^{+}(\mathbf{r},t')\rangle = \frac{\omega_0}{16\pi^2 c^2 r^2}\langle \mathbf{D}^{+}(t-r/c)\cdot\mathbf{D}^{-}(t'-r/c)\rangle, \qquad (4.10)$$

where

$$\mathbf{D}^{-}(t) = \sum_{j=1}^{n} \mathbf{d}_j e^{i\omega_0 \mathbf{r}\cdot\mathbf{r}_j/c} a_j(t). \qquad (4.11)$$

We will assume that the atomic system is stationary and the correlation function is invariant under the transition of time, that is,

$$G(t,t') = \langle \mathbf{D}^{+}(t)\mathbf{D}^{-}(t')\rangle, \qquad (4.12)$$

where the proportional constant is omitted.

4.2 Power spectrum of emission field

The Liouville operator of system is given by eq.(3.5). We may impose the time dependence of Liouvillian on the density operator with use of the time evolution operators

$$U_E(t) = \exp[-i\omega t \textstyle\sum_j a_j^{+} a_j], \qquad (4.13)$$

and

$$U_E^{-}(t) = \exp[i\omega t \textstyle\sum_j a_j^{+} a_j]. \qquad (4.14)$$

These operators satisfy the relations

$$U_E(t) a_j U_E^{-}(t) = a_j e^{-i\omega t}, \qquad (4.15)$$

$$U_E(t) a_j^{+} U_E^{-}(t) = a_j^{+} e^{i\omega t}. \qquad (4.16)$$

In obtaining eqs.(4.15) and (4.16) we have used the formula

$$e^{L}Ae^{-L} = A + [\,L, A\,] + \frac{1}{2!}[\,L, [\,L, A\,]\,] + \cdots\cdots, \tag{4.17}$$

(where L and A are operator) and the anticomutation relation

$$[\,a_j, a_k\,] = 0 \quad (j \neq k) \tag{4.18}$$

Then by multiplying both sides of eq.(3.56) by $U_{E(t)}$ from the left and $U_{E}^{-}(t)$ from the right, we have (ignoring suffix A)

$$\dot{\tilde{\rho}}(t) = -i\tilde{\mathcal{L}}\tilde{\rho}(t), \tag{4.19}$$

where

$$\tilde{\rho}(t) = U_{E}(t)\rho(t)U_{E}^{-}(t), \tag{4.20}$$

$$\begin{aligned} -i\tilde{\mathcal{L}}X = &\sum_{j} (2\kappa a_j X a_j^{+} - \kappa\{\, a_j^{+}a_j, X\,\} + i(\omega-\omega_0)[\, a_j^{+}a_j, X\,]) \\ &+ \sum_{jk} (2\gamma_{jk} a_j X a_k^{+} - \gamma_{jk}\{\, a_j^{+}a_k, X\,\} - i\Omega_{jk}[\, a_j^{+}a_k, X\,]) \\ &+ i\sum_{j} [\, R_j a_j + R_j^{*} a_j^{+}, X\,]. \qquad (j \neq k) \end{aligned} \tag{4.21}$$

With use of eqs.(4.13) and (4.14), the correlation function eq.(2.14) can be written as

$$\begin{aligned} \langle D^{+}(t)D^{-}(t')\rangle &= \mathrm{tr}\{D^{-}U(t',t)U_{E}^{-}(t)U_{E}(t)\rho(t)U_{E}^{-}(t)U_{E}(t)D^{+}U(t,t')\} \\ &= \mathrm{tr}\{D^{-}U(t',t)U_{E}^{-}(t)\tilde{\rho}(t)D^{+}U_{E}(t)U(t,t')\}e^{+i\omega t} \\ &= \mathrm{tr}\{D^{-}\exp[-i\tilde{\mathcal{L}}(t'-t)](\tilde{\rho}(t)D^{+})\}e^{+i\omega t}. \end{aligned} \tag{4.22}$$

Then the power spectrum is now calculated from eq.(2.14) by the simple replacement $i\nu \to -i(\omega-\nu)$ as

$$I(\nu) = \frac{1}{\pi}\mathrm{Re}\left[\lim_{\varepsilon\to 0} \mathrm{tr}\{D^{-}(s+i\tilde{\mathcal{L}})^{-1}(\tilde{\rho}^{e}D^{+})\}\Big|_{s=-i(\omega-\nu)+\varepsilon}\right]. \tag{4.23}$$

We may also calculate the emission spectrum of the coherent and incoherent part from eq.(2.34) by the replacement $i\nu \to -i(\omega-\nu)$, as equation (4.19) satisfy the probability conservation relation eq.(2.3).

§5. Single- and Two-Atom

5.1 Single-atom case (Mollow's model)

For an atom which is separated enough from others, the cooperative effects are not present and eq.(4.19) reduces in this case (ignoring the suffix j, k) to

$$\dot{\tilde{\rho}}(t) = -i\tilde{\mathcal{L}}\tilde{\rho}(t), \tag{5.1}$$

where

$$-i\tilde{\mathcal{L}}X = 2\varkappa aXa^{+} - \kappa\{ a^{+}a, X\} + i(\omega-\omega_0)[a^{+}a, X] + i[R(a+a^{+}), X]. \tag{5.2}$$

Here, we have assumed that the phase R can be made real.

The vectors eqs.(A.3) and (A.5) are defined by

$$\Phi = [\ |1><1|\ ,\ |0><0|\ ,\ |0><1|\ ,\ |1><0|\ ], \tag{5.3}$$

and

$$\Phi^{\dagger} = \begin{bmatrix} |1><1| \\ |0><0| \\ |1><0| \\ |0><1| \end{bmatrix}. \tag{5.4}$$

The operator eq.(4.20) is written as

$$\tilde{\rho}(t) = \Phi\cdot\tilde{P}(t), \tag{5.5}$$

where

$$\tilde{P}(t) = \begin{bmatrix} p_1 \\ p_2 \\ p_3 e^{-i\omega t} \\ p_4 e^{i\omega t} \end{bmatrix}. \tag{5.6}$$

From eqs.(A.7) and (A.8), the master equation (5.2) is expressed in the matrix form as

$$\dot{\tilde{P}}(t) = -i\tilde{L}\,\tilde{P}(t). \tag{5.7}$$

where

$$i\tilde{L} = \begin{bmatrix} K & iE \\ iE^{t} & W \end{bmatrix}, \tag{5.8}$$

$$K = \begin{bmatrix} 2\varkappa & 0 \\ -2\varkappa & 0 \end{bmatrix}, \quad W = \begin{bmatrix} z & 0 \\ 0 & z^{*} \end{bmatrix}, \quad E = R\begin{bmatrix} -1 & 1 \\ 1 & -1 \end{bmatrix}. \tag{5.9}$$

with

$$z = \kappa + i(\omega-\omega_0), \tag{5.10}$$

$$z^{*} = \kappa - i(\omega-\omega_0). \tag{5.11}$$

Then, the matrix appearing in the Laplace transform of eq.(5.7) can be written as

$$(S+i\tilde{L})^{-1} = \begin{bmatrix} \frac{1}{G[s]} & -\frac{1}{G[s]}\, iE\frac{1}{S+W} \\ \frac{1}{S+W}\, iE^{t}\frac{1}{G[s]} & \frac{1}{S+W} - \frac{1}{S+W}E^{t}\frac{1}{G[s]}E\frac{1}{S+W} \end{bmatrix}, \tag{5.12}$$

where the fractional expressions stand for the inverse matrices. In this equation, we define

$$\frac{1}{G[s]} = \frac{1}{S + K + E^{t}\frac{1}{S+W}E} = \frac{1}{S + K + g(s)\begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix}}, \tag{5.13}$$

where g(s) represents to the effect of the driving field,

$$g(s) = R^{2}\left(\frac{1}{s+z} + \frac{1}{s+z}^{*}\right). \tag{5.14}$$

The equation (5.1) satisfies the conservation relation

$$p_1 + p_2 = \text{const}, \tag{5.15}$$

and we can remove the one element p_2. Then, eqs.(B.16), (B.20), (B.30) are given by

$$P(t) = \begin{bmatrix} p_1 \\ p_3 \\ p_4 \end{bmatrix}, \quad Y = \begin{bmatrix} 0 \\ -iR \\ iR \end{bmatrix}, \quad X = \begin{bmatrix} 1 & 0 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{5.16}$$

The reduced matrix is now written as

$$i\tilde{L}' = \begin{bmatrix} \kappa & iE' \\ 2iE^{t\prime} & W \end{bmatrix}, \tag{5.17}$$

where

$$E' = [\ -R,\ R\ ]. \tag{5.18}$$

The similar manipulation of eq.(5.12) permits us to write down the inverse matrix in the form

$$(S+i\tilde{L}')^{-1} = \frac{1}{G[s]} \begin{bmatrix} 1 & Rh(s) & Rh(s)^* \\ 2Rh(s) & h^2(s)[2R^2+(s+2\kappa)(s+z^*)] & 2R^2h^2(s) \\ 2Rh(s)^* & 2R^2h^2(s) & h^2(s)[2R^2+(s+2\kappa)(s+z)] \end{bmatrix}, \tag{5.19}$$

where

$$G[s] = \frac{1}{s + 2\kappa + 2R^2(h(s)+h(s)^*)}, \tag{5.20}$$

and

$$h(s) = i/(s+z). \tag{5.21}$$

The elements of the steady-state density operator are then calculated from eq.(B.29) as

$$P^e = \begin{bmatrix} \tilde{p}_1^e \\ \tilde{p}_2^e \\ \tilde{p}_3^e e^{i\omega t} \\ \tilde{p}_4^e e^{-i\omega t} \end{bmatrix} = \frac{1}{G[0]} \begin{bmatrix} g(0) \\ 2\kappa+g(0) \\ -2iR\, e^{i\omega t}/z \\ 2iR\, e^{-i\omega t}/z^* \end{bmatrix}. \tag{5.22}$$

The dipole operator D and D^+ are now given by

$$D = da, \qquad \text{and} \qquad D^+ = da^+. \tag{5.23}$$

As shown in §4, the power spectrum is calculated from eq.(B.31) with the replacement $i\nu \to -i(\omega-\nu)$. The matrix eq.(B.34) is calculated as

$$H[s] = \frac{h^2(s)}{G[s]}\left[\, 2R^2+(s+2\kappa)(s+z),\ 0,\ \frac{g(0)}{G[s]}R\{2(s+z^*)R^2-(s+2\kappa)(s+z)\},\ 0\,\right] \tag{5.24}$$

and we have

$$H[s]\cdot\tilde{P}^e = p_1^{e\,2}h^2(s)[2(s+2\kappa)^2+4R^2]/G[s]. \tag{5.25}$$

Then the power spectrum is finally given by

$$I[\nu] = \delta(\nu-\omega)I^{coh} + I^{incoh}_{[\nu]}, \tag{5.26}$$

where

$$I^{coh} = \frac{4R^2\kappa^2}{z^2}\frac{1}{(G[0])^2}, \tag{5.27}$$

$$I^{incoh}_{[\nu]} = \frac{1}{\pi}\left[\mathrm{Re}\{\, p_1^{e\,2}h^2(s)\frac{2(s+2\kappa)^2+4R^2}{G[s]}\Big|_{s=i(\omega-\nu)}\}\right]. \tag{5.28}$$

5.2 Two-atom case

We now consider two atoms 1 and 2 with the excitation energies, ω_1 and ω_2, measured from the ground state. The distance between two atoms is r and we express the cooperative damping eq.(3.50) and shifts eq.(6.54) as γ and Ω, respectively. The master equation (4.21) is given by

$$\dot{\tilde{\rho}}(t) = -i\tilde{L}\tilde{\rho}(t) \tag{5.29}$$

where

$$\begin{aligned} -i\tilde{L}X = {} & 2\kappa(a_1Xa_1^+ + a_2Xa_2^+) - \kappa\{\, a_1^+a_1 + a_2^+a_2,\ X\,\} \\ & + i(\omega-\omega_1)[\, a_1^+a_1,\ X\,] + i(\omega-\omega_2)[\, a_2^+a_2,\ X\,] \\ & + 2\gamma(a_1Xa_2^+ + a_2Xa_1^+) - \gamma\{\, a_2^+a_1 + a_1^+a_2,\ X\,\} + i\Omega[\, a_2^+a_1 + a_1^+a_2,\ X\,] \\ & + i[\, R_1a_1 + R_2a_2 + R_1^*a_1^+ + R_2^*a_2^+,\ X\,]. \end{aligned} \tag{5.30}$$

The dipole operators of system are now expressed as

$$D^{+} = d_1 a_1 + d_2 a_2 e^{-ikr} a_2, \qquad D^{-} = d_1 a_1 + d_2 e^{ikr} a_2. \tag{5.31}$$

It is helpful to note the formal identity between states of two-level atomic system and those for a single system having an angular moment.[17] We can choose the base of atomic system as the triplet states

$$|1\rangle = |0\rangle_1 |0\rangle_2, \tag{5.32}$$

$$|2\rangle = \frac{1}{\sqrt{2}}(|1\rangle_1 |0\rangle_2 + |0\rangle_1 |1\rangle_2), \tag{5.33}$$

$$|3\rangle = |1\rangle_1 |1\rangle_2, \tag{5.34}$$

and the one singlet state

$$|4\rangle = \frac{1}{\sqrt{2}}(|1\rangle_1 |0\rangle_2 - |0\rangle_1 |1\rangle_2). \tag{5.35}$$

The density operator eq.(4.20) is expanded as

$$\tilde{\rho}(t) = \Phi \cdot \tilde{P}(t), \tag{5.36}$$

where

$$\Phi = [\ \Phi_A,\ \Phi_B,\ \Phi_B^*,\ \Phi_C,\ \Phi_C^*\ ], \tag{5.37}$$

with

$$\Phi_A = [\ |1\rangle\langle 1|,\ |2\rangle\langle 2|,\ |3\rangle\langle 3|,\ |4\rangle\langle 4|\ ], \tag{5.38}$$

$$\Phi_B = [\ |1\rangle\langle 2|,\ |2\rangle\langle 3|,\ |1\rangle\langle 3|\ ], \tag{5.39}$$

$$\Phi_C = [\ |1\rangle\langle 4|,\ |4\rangle\langle 3|,\ |4\rangle\langle 2|\ ], \tag{5.40}$$

and

$$\tilde{P}(t) = \begin{bmatrix} P_A \\ \tilde{P}_B \\ \tilde{P}_B^* \\ \tilde{P}_C \\ \tilde{P}_C^* \end{bmatrix}, \tag{5.41}$$

with

$$\mathbf{P}_A = \begin{bmatrix} p_1 \\ p_2 \\ p_3 \\ p_4 \end{bmatrix}, \qquad \tilde{\mathbf{P}}_B = \begin{bmatrix} p_5 e^{-i\omega t} \\ p_6 e^{-i\omega t} \\ p_7 e^{-2i\omega t} \end{bmatrix}, \qquad \tilde{\mathbf{P}}_C = \begin{bmatrix} p_8 e^{-i\omega t} \\ p_9 e^{-i\omega t} \\ p_{10} \end{bmatrix}. \tag{5.42}$$

The matrix form of the Liouvillian is given in Appendix E. The energy levels and damping rates of the system is illustrated by Fig. 2.

Equation (5.29) satisfy the probability conservation relation

$$p_1 + p_2 + p_3 + p_4 = \text{const}, \tag{5.43}$$

and we can remove the one element p_4 from $\mathbf{P}(t)$. Then we have

$$(\mathbf{S}+i\tilde{\mathbf{L}}')^{-1} = \begin{bmatrix} \mathbf{S}+\mathbf{A}' & i\mathbf{E}' \\ i\mathbf{E}''^{t} & \mathbf{S}+\mathbf{B} \end{bmatrix}^{-1}, \tag{5.44}$$

where

$$\mathbf{A}' = \begin{bmatrix} \mathbf{K}' & i\mathbf{E}'_A & -i\mathbf{E}'^{*}_A \\ i\mathbf{E}'^{\dagger}_A & \mathbf{W}_A & \mathbf{0} \\ -i\mathbf{E}'^{t}_A & \mathbf{0} & \mathbf{W}^{*}_A \end{bmatrix}, \qquad i\mathbf{E}' = \begin{bmatrix} i\mathbf{E}'_1 & -i\mathbf{E}'^{*}_1 \\ i\mathbf{E}_2 & i\mathbf{E}^{*}_3 \\ -i\mathbf{E}_3 & -i\mathbf{E}^{*}_2 \end{bmatrix}, \tag{5.45}$$

and

$$i\mathbf{E}''^{t} = \begin{bmatrix} i\mathbf{E}''_1 & i\mathbf{E}^{\dagger}_2 & -i\mathbf{E}^{\dagger}_3 \\ -i\mathbf{E}''^{*}_1 & i\mathbf{E}^{t}_3 & -i\mathbf{E}^{t}_2 \end{bmatrix}, \qquad \mathbf{B} = \begin{bmatrix} \mathbf{W}_B & i\mathbf{E}_B \\ i\mathbf{E}^{\dagger}_B & \mathbf{W}^{*}_B \end{bmatrix}. \tag{5.46}$$

The matrix elements in eqs.(5.45) and (5.46) are also matrices and are given in Appendicies E and F.

Here, we consider the case

$$\mathbf{d} = \mathbf{d}_1 = \mathbf{d}_2 \qquad \text{and} \qquad \omega_1 = \omega_2 . \tag{5.47}$$

This assumption reduces to

$$R_- = 0, \qquad \text{and} \qquad \omega_- = 0. \tag{5.48}$$

The inverse matrix eq.(5.44) then becomes

$$(S+i\tilde{L}')^{-1} = \begin{bmatrix} (S+A')^{-1} & 0 \\ 0 & (S+B)^{-1} \end{bmatrix}. \tag{5.49}$$

The inverse matrix $(S+iA)^{-1}$ is given by

$$(S+iA')^{-1} = \begin{bmatrix} \frac{1}{G[s]} & -\frac{1}{G[s]} iE_\alpha \frac{1}{S+W} \\ \frac{1}{S+W} iE_\alpha^t \frac{1}{G[s]} & \frac{1}{S+W} - \frac{1}{S+W} E_\alpha^t \frac{1}{G[s]} E_\alpha \frac{1}{S+W} \end{bmatrix}, \tag{5.50}$$

where

$$G[s]^{-1} = \frac{1}{S+K+g[s]}, \qquad \frac{1}{S+W} = \begin{bmatrix} \frac{1}{S+W_A} & 0 \\ 0 & \frac{1}{S+W_A^*} \end{bmatrix}, \tag{5.51}$$

$$E_\alpha = [\, iE_A, \; -iE_A^* \,].$$

The matrix $g[s]$ is defined by

$$g[s] = E_\alpha (S+W)^{-1} E_\alpha^t = E_A (S+W_A)^{-1} E_A^t + c.c. \tag{5.52}$$

which corresponds to the effect of driving force. More explicitly, $g[s]$ is given by

$$g[s] = \begin{bmatrix} g_1+g_e & -g_1 & -g_e \\ -g_1-g_3 & g_1+g_2+g_3 & -g_2 \\ g_3-g_e & -g_2-g_3 & g_2+g_e \end{bmatrix}, \tag{5.53}$$

where

$$g_1 = (s+b)(s+c)R_+^2/\det 1 + c.c. \tag{5.54}$$

$$g_2 = (s+a)(s+c)R_+^2/\det 1 + c.c. \tag{5.55}$$

$$g_3 = -2(\kappa+\gamma)(s+c)R_+^2/\det 1 + c.c. \tag{5.56}$$

$$g_e = R_+^*/\det 1 + c.c. \tag{5.57}$$

and

$$\det 1 = s^3 + s^2(a+b+c) + s(ab+bc+ac+2R_+^2) + c(R_+^2+ab). \tag{5.58}$$

The variables a, b, c are given in Appendix E. In the complex conjugate appearing in eqs.(5.54)-(5.57), we have assumed that the parameter s is a real constant. The connection of levels are illustrated in Fig. 2. The calculation of eq.(5.50) can be done by the manipulation of 3x3 matrixes. Several elements are calculated as

$$F_{13} = G_{13}^{-1}[s] = \{[s+2(\kappa+\gamma)]g_e + A\}/\det G, \tag{5.59}$$

$$F_{23} = G_{23}^{-1}[s] = [(s+4\kappa)g_2 + 2(\kappa+\gamma)g_e + A]/\det G, \tag{5.60}$$

$$F_{33} = G_{33}^{-1}[s] = [s^2 + s(6\kappa+2\gamma+2g_1+g_2+g_3+g_e) + 8\kappa(\kappa+\gamma) + 4\kappa(g_1+g_2+g_3) + 2(\kappa+\gamma)g_e + A]/\det G, \tag{5.61}$$

where

$$\det G = s^3 + s^2(8\kappa+2g_1+2g_2+g_3+g_e) + s[4(5\kappa^2-\gamma^2)-2(3\kappa-\gamma)(2g_2+g_3)+4(2\kappa-\gamma)g_1+12\kappa g_e+A] + 8(\kappa-\gamma)[2\kappa(\kappa+\gamma)+\kappa(g_1+2g_2+g_3)+2(\kappa+\gamma)g_e+A], \tag{5.62}$$

and

$$A = g_1 g_2 + g_e(g_1+g_2+g_3). \tag{5.63}$$

By using these elements, another elements of $\mathbf{F}$ are written as

$$F_{43} = \{[(s+b)(s+c)+R_+^2]F_a + R_+^2 F_b\}\, iR_+/\det 1, \tag{5.64}$$

$$F_{53} = \{[(s+c)(s+a)+R_+^2]F_b + [2(\kappa+\gamma)(s+c)+R_+^2]F_a\}\, iR_+/\det 1, \tag{5.65}$$

$$F_{63} = \{[s-2(\kappa+\gamma)+b]F_a - (s+a)F_b\}R_+^2/\det 1, \tag{5.66}$$

where

$$F_a = F_{23} - F_{13} = [(s+4\kappa)g_2 - sg_e]/\det G, \tag{5.67}$$

$$F_b = F_{33} - F_{23} = [s^2+s(6\kappa+2\gamma+2g_1+g_3)+8\kappa(\kappa+\gamma)+4\kappa(g_1+g_3)]/\det G. \tag{5.68}$$

The state populations in the equilibrium are calculated from eq.(B.29) as

$$p_1^e = \{ -(\kappa+\gamma)g_e^0 + A^0 \} / B, \tag{5.69}$$

$$p_2^e = \{ -4\kappa g_2^0 - 2(\kappa+\gamma)g_e^0 + A^0 \} / B, \tag{5.70}$$

$$p_3^e = \{ 4\kappa(2\kappa - g_1^0 - g_2^0 - g_3^0) - 2(\kappa+\gamma)g_e^0 + A^0 \} / B, \tag{5.71}$$

$$p_4^e = 1 - p_1 - p_2 - p_3, \tag{5.72}$$

where we put

$$g_i^0 = g_i|_{s=0}, \qquad A^0 = A|_{s=0}, \tag{5.73}$$

and

$$B = 8\{ 2\kappa(\kappa+\gamma) + \kappa(g_1^0 + 2g_2^0 + g_3^0) + 2(\kappa+\gamma) \} . \tag{5.74}$$

Figures 3-5 illustrate the distance dependence of the state populations eqs.-(5.69)-(5.72). As cooperative shift will diverge logarithmically, elements quickly change in the very short distance. Figure 3 corresponds to the weak field case. In this case, the damping processes are important and the population of upper-level of the triplet state decreases. The field becomes stronger, the population of upper-level increases. In Fig. 4, all elements vary with distance. This is caused by cooperative damping and shift (see Fig. 1). However, in the very strong field case Fig. 5, the driving effects becomes important and the distance dependence declines.

From eqs.(5.64)-(5.66), the other elements are given by

$$\tilde{p}_5^e = 4i\kappa R\{ (bc+R^2)g_2^0 + [2(\kappa+\gamma) + g_1^0 + g_3^0]R^2 \} / B, \tag{5.75}$$

$$\tilde{p}_6^e = 4i\kappa R\{ (ac+R^2)[2(\kappa+\gamma) + g_1^0 + g_3^0] + [2(\kappa+\gamma) + R^2]g_2^0 \} / B, \tag{5.76}$$

$$\tilde{p}_7^e = 4\kappa\{ [b - 2(\kappa+\gamma)]g_2^0 - a[2(\kappa+\gamma) + g_1^0 + g_3^0] \} / B. \tag{5.77}$$

The phases of the dipole operator eq.(5.31) depends on the position of the detector. We choose these as

$$D^+ = d(a_1^+ + a_2^+), \qquad D^- = d(a_1 + a_2). \tag{5.78}$$

which satisfy the following relations:

$$D^+|0\rangle = \sqrt{2}d|1\rangle, \qquad D^-|2\rangle = \sqrt{2}d|1\rangle, \tag{5.79}$$

$$D^+|1\rangle = \sqrt{2}d|0\rangle, \qquad D^-|3\rangle = \sqrt{2}d|2\rangle, \tag{5.80}$$

$$D^+|2\rangle = D^+|3\rangle = 0, \qquad D^-|0\rangle = D^-|3\rangle = 0. \tag{5.81}$$

Then from eqs.(B.31)-(B.33), the power spectrum is finally given by [52]

$$I[\nu] = \delta(\nu-\omega) I^{coh} + I^{incoh}[\nu], \tag{5.82}$$

where

$$I^{coh} = 2d^2 |\tilde{p}_5+\tilde{p}_6|^2, \tag{5.83}$$

and

$$I^{incoh}[\nu] = 2d^2 \mathrm{Re}\{ p_2(F_{78}+F_{79}) + p_3(F_{88}+F_{89}) + \tilde{p}_5(F_{18}+F_{19}-\alpha) + \tilde{p}_6(F_{28}+F_{29}-\alpha) + \tilde{p}_7(F_{48}+F_{49}) + \tilde{p}_9(F_{98}+F_{99}) \}|_{S=i(\nu-\omega)}, \tag{5.84}$$

with

$$\alpha = \sum_{i=1}^{9} \tilde{p}_i (F_{i8}+F_{i9}). \tag{5.85}$$

§6. Conclusion

In this chapter, we have investigated the cooperative feature of two-level atoms. In §1, the general formulation of coherent and incoherent parts of spectrum is presented. From this formulation, we see that the amplitude of coherent part is proportional to the square of the transition amplitude from the equilibrium state, and the incoherent part is governed by the unsteady-state time evolution operator. In sections 3 and 4, we study the dynamics of two-level atomic system and its emission spectrum. Section 5 is devoted to the calculation of the steady-state density operators and the emission spectra for one and two atoms system. In this section, we show that the results can be written in relatively compact forms. In the two atom case, the distance dependence of state populations was discussed. The present work is the first step of this problem. More detailed studies of power spectra are being carried out.

Although we limit here to a relatively simple problem, the method here employed can be used for more complicated problem. It is interesting to investigate such cases that the atoms are modulated by reservoir independently or the system consists of three or more atoms. We leave these for future study.

Appendix A

Let us evaluate the hyper-operator $(s+i\mathcal{L})^{-1}$. We suppose that the system consists of the n states and the density operator is expanded as

$$\rho(t) = p_1|1><1| + p_2|2><2| + \cdots + p_n|n><n| + p_{n+1}|1><2| + p_{n+2}|1><3| + \cdots + p_{n^2-1}|n><n-2| + p_{n^2}|n><n-1|, \quad (A.1)$$

where p_j are time dependent coefficients. Now, we introduce an operator $\mathbf{f}$ and its inverse $\mathbf{f}^{-1}$. With use of these operators, the density operator in eq.(A.1) is transformed into an n^2-dimensional vector as

$$\mathbf{f}\rho(t) = \mathbf{P}(t) = \begin{bmatrix} p_1 \\ p_2 \\ p_3 \\ \cdot \\ \cdot \\ \cdot \\ p_{n^2} \end{bmatrix} \quad (A.2)$$

and

$$\mathbf{f}^{-1}\mathbf{P}(t) = p_1|1><1| + p_2|2><2| + \cdots = \rho(t). \quad (A.3)$$

These operators are expressed by the vectors

$$\Phi = [\ |1><1|, |2><2|, \cdots, |n><n|, |1><2|, |1><3|, \cdots\cdots\ ] \quad (A.4)$$

and

$$\Phi^{\dagger} = \begin{bmatrix} |1><1| \\ |2><2| \\ \cdot \\ \cdot \\ \cdot \\ |n><n| \\ |2><1| \\ |3><1| \\ \cdot \\ \cdot \\ \cdot \end{bmatrix} \quad (A.5)$$

as

$$\mathbf{f}\rho(t) = tr\{\Phi^{\dagger}\rho(t)\} = \mathbf{P}(t) \quad (A.6)$$

and

$$\mathbf{f}^{-1}\mathbf{P}(t) = \Phi \cdot \mathbf{P}(t) = \rho(t). \tag{A.7}$$

The equation of motion (2.1) is now written as

$$\mathbf{f}\dot{\rho}(t) = -i\mathbf{L}\mathbf{f}\rho(t), \tag{A.8}$$

where

$$i\mathbf{L} = \mathbf{f}\, i\mathcal{L}\, \mathbf{f}^{-1} \tag{A.9}$$

is the $n^2 \times n^2$ matrix corresponding to the Liouvillian and its elements are expressed by the elements of $\Phi^{\dagger}$ and Φ as

$$iL_{jk} = (\mathbf{f}\, i\mathcal{L}\, \mathbf{f}^{-1})_{jk} = tr\{\Phi_j^{\dagger}\, i\mathcal{L}\Phi_k\}. \tag{A.10}$$

The Laplace transform of eq.(2.1) is then given by

$$\rho[s] = \mathbf{f}^{-1}(\mathbf{S}+i\mathbf{L})^{-1}\mathbf{f}\rho(t_0), \tag{A.11}$$

where $\mathbf{S}$ is the unit matrix multiplied by the constant s. The power spectrum eq.(2.5) is written as

$$\begin{aligned} I(\nu) &= \frac{1}{\pi}\mathrm{Re}\left[\lim_{\varepsilon\to 0} tr\{D^{-}\mathbf{f}^{-1}(\mathbf{S}+i\mathbf{L})^{-1}\mathbf{f}(\rho^{e}D^{+})\}\big|_{s=\, i\nu+\varepsilon}\right] \\ &= \frac{1}{\pi}\mathrm{Re}\left[\lim_{\varepsilon\to 0} tr\{D^{+}\Phi\}\,(\mathbf{S}+i\mathbf{L})^{-1}\, tr\{\Phi^{\dagger}\rho^{e}D^{+}\}\big|_{s=\, i\nu+\varepsilon}\right], \end{aligned} \tag{A.12}$$

in which the equilibrium density operator is given by

$$\rho^{e} = \lim_{s\to 0} \mathbf{f}^{-1}\, s(\mathbf{S}+i\mathbf{L})^{-1}\, \mathbf{f}(\rho(t_0)). \tag{A.13}$$

Appendix B

From eq.(A.1), the state conservation relation (2.3) is written as

$$tr\{\rho(t)\} = \sum_{j=1}^{n} p_j = \text{const.} \tag{B.1}$$

To evaluate the equilibrium density operator and hyper-operator eq.(2.31) and (2.32), we consider the projection operator R which removes the one state proportional to $|\alpha><\alpha|$ $(1 \leq \alpha \leq n)$. Then,

$$R\rho(t) = \rho'(t), \tag{B.2}$$

where

$$\rho'(t) = p_1|1><1| + p_2|2><2| + \cdots + p_{\alpha-1}|\alpha-1><\alpha-1| + p_{\alpha+1}|\alpha+1><\alpha+1| + \cdots\cdots + p_n|n><n| + \cdots\cdots. \tag{B.3}$$

From eq.(B,1), the removed state is expressed by

$$p_\alpha|\alpha><\alpha| = (-\sum_{j\neq\alpha}^{n} p_j + \text{const})|\alpha><\alpha|, \tag{B.4}$$

where

$$\text{const} = tr\{\rho(t_0)\}. \tag{B.5}$$

The first term in the bracket on the right hand side of eq.(B.4) is expressed as

$$-\sum_{j\neq\alpha}^{n} p_j = tr\{\rho'(t)\}. \tag{B.6}$$

Then we may write

$$\rho(t) = \rho'(t) + p_\alpha|\alpha><\alpha| = Q_\alpha\rho'(t) + |\alpha><\alpha|\text{const}, \tag{B.7}$$

where

$$Q_\alpha = 1 - |\alpha><\alpha|\, tr\{\ . \tag{B.8}$$

This equation also means that the reduced density operator can be recovered by the operator I as

$$I\rho'(t) = \rho(t), \tag{B.9}$$

Where

$$IX = Q_\alpha X + |\alpha><\alpha|\text{const}. \tag{B.10}$$

By multiplying the both sides of eq.(2.7) by R, we have

$$\dot{\rho}'(t) = -Ri\mathcal{L}\rho(t). \tag{B.11}$$

From eq.(B.7), it can be written as

$$\dot{\rho}'(t) = -Ri\mathcal{L}Q_\alpha\rho'(t) - Ri\mathcal{L}|\alpha><\alpha|\text{const}. \tag{B.12}$$

We now employ the matrix representation. Let us consider the operator $\mathbf{f}'$ which removes the one element $p_\alpha|\alpha><\alpha|$ from $\rho(t)$ and transform it into vector space. The operator $\mathbf{f}'$ and its inverse $\mathbf{f}'^{-1}$ are defined by the vector

$$\Phi' = [\ |1><1|, \cdots, |\alpha-1><\alpha-1|, |\alpha+1><\alpha+1|, \cdots, |1><2|, |1><3|, \cdots\cdot\ ], \tag{B.13}$$

is the form

$$\mathbf{f}'\rho(t) = tr\{\Phi'^{\dagger}\rho(t)\} = \mathbf{P}'(t) \tag{B.14}$$

and

$$\mathbf{f}'^{-1}\mathbf{P}'(t) = \Phi'\cdot\mathbf{P}'(t) = \rho'(t), \tag{B.15}$$

where

$$\mathbf{P}'(t) = \begin{bmatrix} p_1 \\ p_2 \\ \cdot \\ \cdot \\ p_{\alpha-1} \\ p_{\alpha+1} \\ \cdot \\ \cdot \end{bmatrix}. \tag{B.16}$$

is a vector with n^2-1 components and $\rho'(t)$ is given in eq.(B.3). The projection operator R is then expressed as

$$R = \mathbf{f}'^{-1}\mathbf{f}'. \tag{B.17}$$

Then, from eqs.(B.14) and (B.17), eq.(B.12) can be written in the matrix form as

$$\dot{\mathbf{P}}'(t) = -i\mathbf{L}'\mathbf{P}'(t) - \mathbf{Y}\,\text{const}, \tag{B.18}$$

where

$$i\mathbf{L}' = \mathbf{f}'i\mathcal{L}Q_\alpha\mathbf{f}'^{-1}, \tag{B.19}$$

$$\mathbf{Y} = \mathbf{f}'i\mathcal{L}|\alpha><\alpha|. \tag{B.20}$$

The Laplace transform of eq.(B.18) is given by

$$\mathbf{P}'[s] = (S+i\mathbf{L}')^{-1}\mathbf{P}'(t_0) - \frac{1}{s}(S+i\mathbf{L})^{-1}\mathbf{Y}\ \text{const.} \tag{B.21}$$

By eqs.(B.5), (B.15) and operator I, we have

$$\rho[s] = I\mathbf{f}'^{-1}\left\{(S+i\mathbf{L}')^{-1}\mathbf{f}' - \frac{1}{s}(S+i\mathbf{L}')^{-1}\mathbf{Y}\mathrm{tr}\{\right\}\rho(t_0). \tag{B.22}$$

Note that iL of eq.(A. 8) is the $n^2 \times n^2$ matrix, , iL' is $(n^2-1)\times(n^2-1)$. As we have assumed that there is the one relation eq.(2.3) or eq.(B.1), the number of independent variables of $\mathbf{P}(t)$ is n^2-1. It means that the rank of iL and iL' are n^2-1 and

$$\det|i\mathbf{L}| = 0, \tag{B.23}$$

but

$$\det|i\mathbf{L}'| \neq 0. \tag{B.24}$$

Because of this, the inverse matrix $(S+i\mathbf{L})^{-1}$ has no element proportional to 1/s. Therefore, the equilibrium density operator eq.(2.15) is given by

$$\rho^e = \lim_{s\to 0} s\rho[s] = -I\mathbf{f}'^{-1}(i\mathbf{L}')^{-1}\mathbf{Y}\ \text{const.} \tag{B.25}$$

With use of this equation, eq.(B.22) can be written as

$$\rho[s] = (\frac{1}{s}y + \mathcal{G}[s])\rho(t_0), \tag{B.26}$$

where

$$y = \frac{1}{\text{const}}\rho^e\mathrm{tr}\{ \tag{B.27}$$

and

$$\mathcal{G}[s] = I\mathbf{f}'^{-1}\left\{(S+i\mathbf{L}')^{-1}\mathbf{f}'(1 - \rho^e\mathrm{tr}\{\ )\right\}. \tag{B.28}$$

In the matrix form, the equilibrium density operator eq.(B.25) is expressed by

$$P^e = -X(iL')^{-1}Y + tr\{\Phi^\dagger|\alpha><\alpha|\}, \tag{B.29}$$

where

$$X = fQ_\alpha f'^{-1} \tag{B.30}$$

is the $n^2 x (n^2-1)$ matrix. For the power spectrum, several manipulations reduce eqs.(2.34)-(2.36) to the form

$$I(\nu) = \delta(\nu) I^{coh} + I^{incoh}_{(\nu)}, \tag{B.31}$$

where

$$I^{coh} = tr\{\Phi D^-\}\cdot P^e \times tr\{\Phi D^+\}\cdot P^e, \tag{B.32}$$

and

$$I^{incoh}_{(\nu)} = \frac{1}{\pi} Re\left[tr\{H[s]\cdot P^e\}|_{s=i\nu}\right], \tag{B.33}$$

with

$$H[s] = tr\{D^-\Phi'\}(S+iL')^{-1}(tr\{\Phi'^\dagger \Phi D^+\} - P'^e tr\{\Phi D^+\}). \tag{B.34}$$

Here, we have used

$$tr\{D^-\Phi X\} = tr\{D^-\Phi'\}, \qquad tr\{D^-\Phi Y\} = 0. \tag{B.35}$$

Appendix C

Let us consider the integral

$$I = \int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2 \, \mathcal{L}_I(t_1)\left(\mathcal{L}_I(t_2)\rho_A(t)\right). \tag{C.1}$$

This integral can be written more conveniently by making changes in the variables as

$$\int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2 \, F(t_1,t_2) = \int_t^{t+\Delta t} dt_2 \int_{t_2}^{t+\Delta t} dt_1 \, F(t_1,t_2), \tag{C.2}$$

since both integration are carried out over the same area in the t_1-t_2 plane. We further set

$$\tau' = t_2 - t, \quad \tau'' = t_1 - t_2 = t_1 - t - \tau', \tag{C.3}$$

in term of which eq.(C.2) becomes

$$\int_t^{\Delta t} d\tau' \int_0^{\Delta t-\tau'} d\tau'' \, F(t+\tau'+\tau'', t+\tau'). \tag{C.4}$$

Then eq.(C.1) is written as

$$I = \int_0^{\Delta t} d\tau' \int_0^{\Delta t-\tau'} d\tau'' \, \mathcal{L}_I(t+\tau'+\tau'')\left(\mathcal{L}_I(t+\tau')\rho_A(t)\right). \tag{C.5}$$

$\mathcal{L}_I(t+\tau'+\tau'')$ and $\mathcal{L}_I(t+\tau')$ have nonzero correlation only for small τ''. Hence the integral over τ'' can be extended to infinity without change in value. Furthermore the stationarity of the random processes make it possible to make $\Delta t \to 0$ in the limit for τ'. Therefore, by replacing τ'' to τ, we have

$$I = \Delta t \int_0^{\infty} dt \, \mathcal{L}_I(t+\tau)\left(\mathcal{L}_I(t)\rho_A(t)\right). \tag{C.6}$$

Appendix D

The function eq.(3.40) is given by

$$g^2(ck, r_{jk}) = \int d\Omega \sum_{\lambda} g^*_{jk\lambda} g_{kk\lambda}$$

$$= \frac{ck}{2\pi} \int d\Omega \sum_{\lambda} [\mathbf{d}_j \cdot \mathbf{e}_{k\lambda}][\mathbf{d}_k \cdot \mathbf{e}_{k\lambda}] e^{i\mathbf{k}\cdot\mathbf{r}_{jk}}. \tag{D.1}$$

By choosing $\mathbf{r}_{jk}$ along the polar axis, as shown in Fig. 6, and using eq.(3.48), we have

$$g^2(ck, r_{jk}) = \frac{ck}{2\pi} \int d\phi d\theta \sin\theta [\mathbf{d}_j \cdot \mathbf{d}_k - \mathbf{d}_j \cdot \mathbf{d}_k \cos\theta_j \cos\theta_k] e^{i\mathbf{k}\cdot\mathbf{r}_{jk}\cos\theta} \tag{D.2}$$

Let the angular coordinates of $\mathbf{d}_i$ be ϕ_i' and θ_i', then we have

$$\cos\theta_i = \cos\theta\cos\theta_i' + \sin\theta\sin\theta_i'\cos(\phi - \phi_i'). \tag{D.3}$$

from $\mathbf{d}_i \cdot \mathbf{k}$. Substituting this equation into eq.(B.2) and integrating over θ and ϕ, we have

$$g^2(\omega, r_{jk}) = \frac{4\omega d^2}{3\pi c^3} W_{jk}(\omega) \tag{D.4}$$

where

$$W_{jk}(\omega) = \frac{3}{2}\left\{ A_{jk} \frac{\sin(\omega r_{jk}/c)}{(\omega r_{jk}/c)} + B_{jk}\left[\frac{\cos(\omega r_{jk}/c)}{(\omega r_{jk}/c)^2} - \frac{\sin(\omega r_{jk}/c)}{(\omega r_{jk}/c)^3} \right] \right\}, \tag{D.5}$$

$$A_{jk} = [(\mathbf{d}_j \cdot \mathbf{d}_k) - (\mathbf{d}_j \cdot \mathbf{r}_{jk})(\mathbf{d}_k \cdot \mathbf{r}_{jk})/r_{jk}^2]/d^2, \tag{D.6}$$

$$B_{jk} = [(\mathbf{d}_j \cdot \mathbf{d}_k) - 3(\mathbf{d}_j \cdot \mathbf{r}_{jk})(\mathbf{d}_k \cdot \mathbf{r}_{jk})/r_{jk}^2]/d^2. \tag{D.7}$$

In this equation, it is assumed that $d = |\mathbf{d}_j|$. In the limiting case $\mathbf{r}_{jk} \to 0$, W_{jk} becomes unity and the natural radiation damping is then given by

$$\kappa = K_{jj} = 2d^2\omega_0^3/(3\pi c^3). \tag{D.8}$$

With use of this constant, the cooperative damping γ_{jk} is expressed as

$$\gamma_{jk} = K_{jk} = \kappa W_{jk}(\omega_0). \qquad (j \neq k) \tag{D.9}$$

Appendix E

From eq.(A.9), the Liouvillian eq.(5.30) is expressed in the matrix form as

$$i\tilde{L} = \begin{bmatrix} K & iE_A & -iE_A^* & iE_1 & -iE_1^* \\ iE_A^+ & W_A & 0 & iE_2 & iE_3^* \\ -iE_A^t & 0 & W_A^* & -iE_3 & -iE_2^* \\ iE_1^+ & iE_2^+ & -iE_3^+ & W_B & iE_B \\ -iE_1^t & iE_3^t & -iE_2^t & iE_B^+ & W_B^* \end{bmatrix}, \tag{E.1}$$

where

$$K = \begin{bmatrix} 4\kappa & 0 & 0 & 0 \\ -2(\kappa+\gamma) & 2(\kappa+\gamma) & 0 & 0 \\ 0 & -2(\kappa+\gamma) & 0 & -2(\kappa-\gamma) \\ -2(\kappa-\gamma) & 0 & 0 & 2(\kappa-\gamma) \end{bmatrix}, \tag{E.2}$$

$$W_A = \begin{bmatrix} a & 0 & iR_+ \\ -2(\kappa+\gamma) & b & -iR_+^* \\ iR_+ & -iR_+ & c^+ \end{bmatrix}, \qquad iE_A = iR_+ \cdot \begin{bmatrix} 1 & 0 & 0 \\ -1 & 1 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \tag{E.3}$$

$$W_B = \begin{bmatrix} d & 0 & 0 \\ 2(\kappa-\gamma) & e & iR_+ \\ 0 & iR_+ & f^+ \end{bmatrix}, \qquad iE_B = iR_+ \cdot \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix} \tag{E.4}$$

and

$$iE_1 = i\begin{bmatrix} -R_- & 0 & 0 \\ 0 & 0 & \omega_- \\ 0 & -R_- & 0 \\ R_- & R_- & -\omega_- \end{bmatrix}, iE_2 = i\begin{bmatrix} -\omega_- & 0 & R_- \\ 0 & \omega_- & 0 \\ R_- & R_- & 0 \end{bmatrix}, iE_3 = i\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -R_- \\ 0 & 0 & 0 \end{bmatrix}. \tag{E.5}$$

with

$$a = 3\kappa + \gamma - i(\omega_+ + \Omega), \quad b = \kappa + \gamma - i(\omega_+ - \Omega), \quad c = 2\kappa - 2i\omega_+, \tag{E.6}$$

$$d = 3\kappa - \gamma - i(\omega_+ - \Omega), \quad e = \kappa - \gamma + i(\omega_+ + \Omega), \quad f = 2\kappa - 2i\Omega, \tag{E.7}$$

and

$$R_+ = R_1 + R_2, \quad R_- = R_1 - R_2, \quad \omega_+ = \omega_1 + \omega_2, \quad \omega_- = \omega_1 - \omega_2. \tag{E.8}$$

Appendix F

The matrix eq.(B.19) is now given by

$$i\tilde{L}' = \begin{bmatrix} K' & iE'_A & -iE'^{*}_A & iE'_1 & -iE'^{*}_1 \\ iE'^{\dagger}_A & W_A & 0 & iE_2 & iE^{*}_3 \\ -iE'^{t}_A & 0 & W^{*}_A & -iE_3 & -iE^{*}_2 \\ iE''_1 & iE^{\dagger}_2 & -iE^{\dagger}_3 & W_B & iE_B \\ -iE''^{*}_1 & iE^{t}_3 & -iE^{t}_2 & iE^{\dagger}_B & W^{*}_B \end{bmatrix}, \tag{F.1}$$

where

$$K' = \begin{bmatrix} 4K & 0 & 0 \\ -2(K+\gamma) & 2(K+\gamma) & 0 \\ 2(K-\gamma) & -4\gamma & 2(K-\gamma) \end{bmatrix}, \tag{F.2}$$

$$iE'_A = iR_+ \cdot \begin{bmatrix} 1 & 0 & 0 \\ -1 & 1 & 0 \\ 0 & -1 & 0 \end{bmatrix}, \qquad iE''_1 = i \begin{bmatrix} -2R_- & -R_- & -R_- \\ -R_- & -R_- & -2R_- \\ \omega_- & 2\omega_- & \omega_- \end{bmatrix} \tag{F.3}$$

and the other elements are given in Appendix E.

References

1) J. H. Eberly and L. Allen, "Optical Resonance and Two-Level Atoms", (John Wiley & Sons, New York, 1974)

2) M. Sargent, M. O. Scully, and W. E. Lamb, Jr, "Laser Physics", (Addison-Wesley, Massachusetts, 1974)

3) B. R. Mollow and M. M. Miller, Ann. Phys. 52, 464 (1969).

4) B. R. Mollow, Phys. Rev. 188, 1969 (1969).

5) R. Bonifacio (Ed.), "Dissipative Systems in Quantum Optics", Topics in Current Physics 27, (Springer, Berlin-Heidelberg-New York, 1982)

6) R. H. Lehmberg, Phys. Rev. 181, 32 (1969).

7) R. H. Lehmberg, Phys. Rev. A2, 883 (1970).

8) R. H. Lehmberg, Phys. Rev. A2, 889 (1970).

9) A. A. Belavin, B. Ya. Zel'dovich, A. M. Perelomov and V. S. Popov, Sov. Phys. JETP 29, 264 (1969).

10) M. J. Stephen, J. Chem. Phys. 40, 699 (1964).

11) D. A. Hutchinson and H. F. Hameka, J. Chem. Phys. 41, 2006 (1964)

12) E. A. Power, J. Chem. Phys. 46, 4297 (1967).

13) C. S. Chang and P. Stehle, Phys. Rev. A4, 630 (1971).

14) P. W. Milonni and P. L. Knight, Phys. Rev. A10, 1096 (1974).

15) P. W. Milonni and P. L. Knight, Phys. Rev. A11, 1090 (1975).

16) F. T. Arecchi and D. M. Kim, Opt. Commum. 2, 324 (1970).

17) R. H. Dicke, Phys. Rev. 93, 99 (1954).

18) M. Tavis and F. W. Cummings, Phys. Rev. 170, 379 (1968).

19) M. Dillard and H. R. Robl, Phys. Rev. 184, 312 (1969).

20) R. Bonifacio, D. M. Kim and M. O. Scully, Phys. Rev. 187, 441 (1969).

21) D. F. Walls and R. Barakat, Phys. Rev. A1, 446 (1970).

22) R. Bonifacio and G. Preparata, Phys. Rev. A2, 336 (1970).

23) D. Dialetis, Phys. Rev. A2, 599 (1970).

24) F. T. Arecchi and E. Courtens, Phys. Rev. A2, 1730 (1970).

25) J. H. Eberly and N. E. Rehler, Phys. Rev. A2, 1607 (1970).

26) N. E. Rehler and J. H. Eberly, Phys. Rev. A3, 1735 (1971).

27) J. H. Eberly, American J. Phys. 40, 1347 (1972).

28) V. Degiorgio, Opt. Commum. 2, 362 (1971).

29) G. S. Agarwal, Phys. Rev. A2, 2038 (1970).

30) G. S. Agarwal, Phys. Rev. A3, 1783 (1971).

31) G. S. Agarwal, Phys. Rev. A3, 1791 (1971).

32) G. S. Agarwal, Phys. Rev. A4, 1778 (1971).

33) R. Bonifacio, P. Schwendimann and F. Haake, Phys. Rev. A4, 302 (1971).

34) R. Bonifacio, P. Schwendimann and F. Haake, Phys. Rev. A4, 854(1971).

35) A. M. Ponte and A. Tallet, Phys. Rev. A4, 1319 (1971).

36) V. Degiorgio and F. Ghielmetti, Phys. Rev. A4, 241 (1971).

37) L. Mandel and L. Wolf (Eds.), "Proc. Third Rochester Conference on Coherence and Quantum Optics", (New York, Plenum, 1973)

38) G. S. Agarwal, "Quantum Optics", Springer Tracts in Modern Physics 70, (Springer, Berlin-Heidelberg-New York, 1974).

39) R. Bonifacio and L. A. Lugiato, Phys. Rev. A11, 1507 (1975).

40) R. Bonifacio and L. A. Lugiato, Phys. Rev. A12, 587 (1975).

41) G. S. Agarwal, Phys. Rev. A12, 1475 (1975).

42) G. S. Agarwal, A. C. Brown, L. M. Narducci, G. Vetri, Phys. Rev. A15, 1613 (1977).

43) G. S. Agarwal, D. H. Feng, L. M. Narducci, R. Gilmore and R. A. Tuft, Phys. Rev. A20, 2040 (1979).

44) G. S. Agarwal, R. Saxena, L. M. Narducci, D. H. Feng and R. Gilmore, Phys. Rev. A21, 257 (1980).

45) I. R. Senitzky, Phys. Rev. Lett. 40, 1334 (1978).

46) C. Mavroyannis, Phys. Rev. A18, 185 (1978).

47) A. S. Jahangir Amin and J. G. Cordes, Phys. Rev. A18, 1298 (1978).

48) H. J. Carmichael, J. Phys. B13, 3551 (1980).

49) T. Chikama, K. Saikawa and R. Kubo, J. Phys. Soc. Jpn. 43, 811 (1984).

50) R. Loudon, "The Quantum Theory of Light", (Oxford Univ. Press, Oxford, 1973)

51) Y. Tanimura, to be appear.

Acknowledgments

The author wishes to express his gratitude to professor R. Kubo for leading him to this problem and various instructive discussions. He also grateful to Dr. Takano for valuable discussions and critical reading of the manuscript.

Figure Captions

Fig. 1. The distance dependence of the cooperative damping and frequency shift normarized by natural radiation damping on the interatomic spacing **r** for dipole operator elements **d** perpendicular to **r** (A_{12}=1, B_{12}=1).

Fig. 2. Energy-level diagram for the triplet and singlet states. The effect of field connects the three triplet states.

Fig. 3. Dependence of state-populations on interatomic spacing for two atoms. We put A=1 and B=1 as like as Fig. 1. Here, the Rabi frequency is small compare with the damping.

Fig. 4. Same as Fig. 3 with the Rabi frequency comparable to the damping.

Fig. 5. Same as Fig. 3 with the Rabi frequency much larger than the damping.

Fig. 6. Geometrical relations for the computations of cooperative damping.

FIG. 1

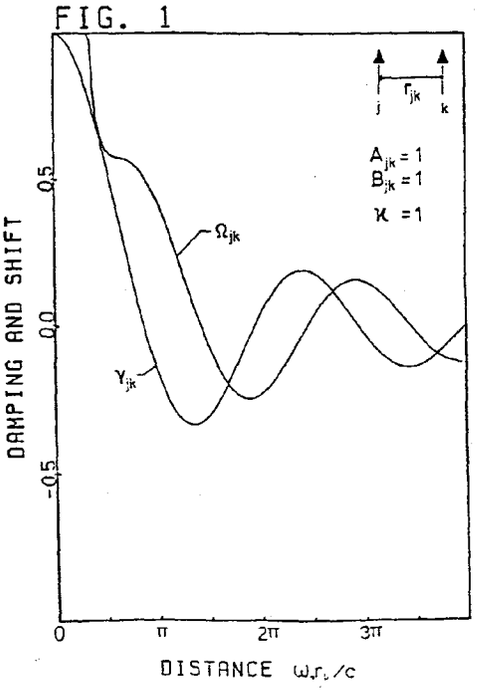


FIG. 2

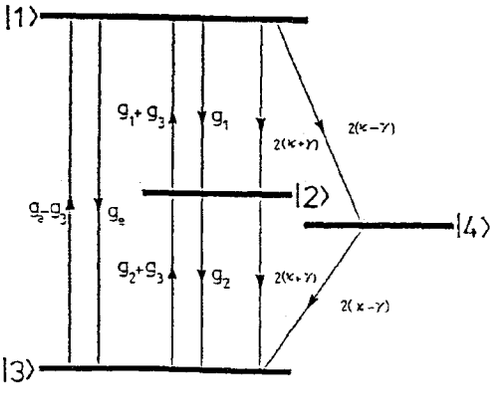


FIG. 3

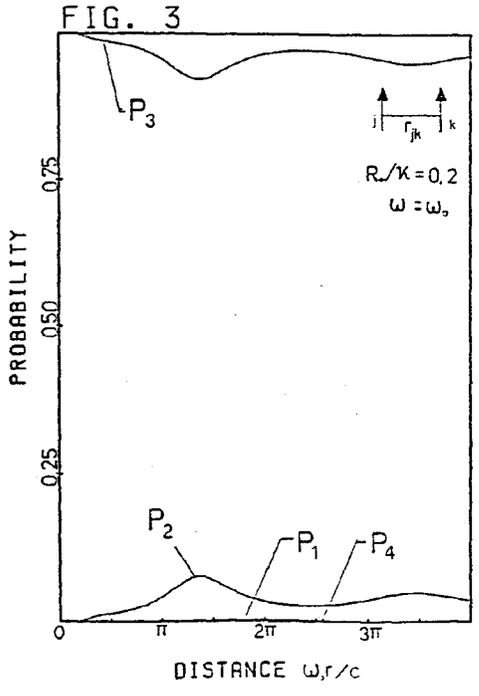


FIG. 4

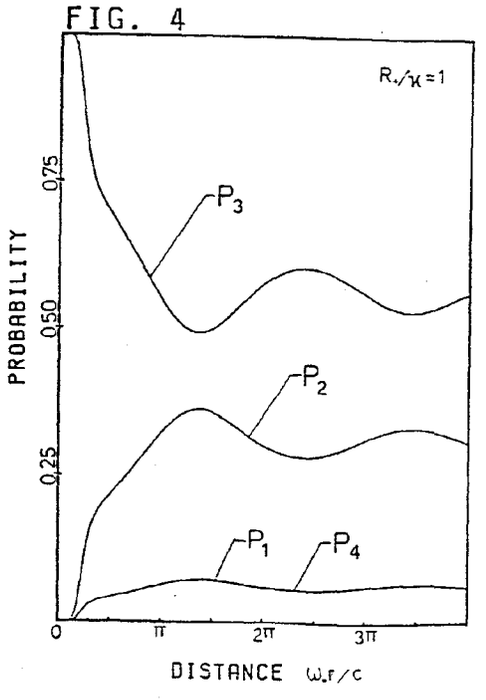


FIG. 5

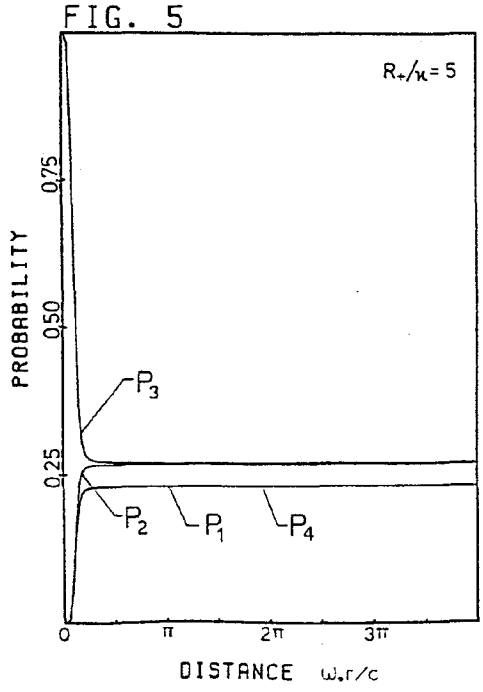
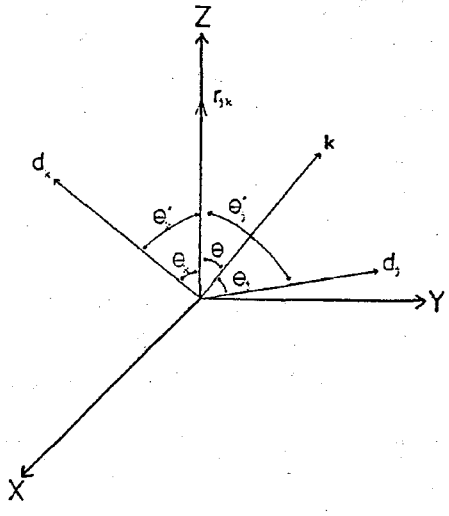

R+/κ = 5
PROBABILITY
0,75
0,50
0,25
P3
P2
P1
P4
0
π
2π
3π
DISTANCE ω₀r/c

FIG. 6


Z
k
X
Y