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Power Spectrum of Light Scattered by Three-Level Systems*

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Abstract

The power spectrum of the light scattered by a three-level atom driven near resonance between the ground state ($|1\rangle$) and the second excited state ($|3\rangle$) by a monochromatic classical electric field is evaluated following the method developed by B. R. Mollow. The incident field is assumed to interact with the dipole moment between the states $|1\rangle$ and $|3\rangle$, while the emitted light is assumed to interact with that between the state $|3\rangle$ and the first excited state $|2\rangle$ of the atom. The atom is assumed to relax to equilibrium only via radiation damping for the sake of simplicity. The power spectrum of the scattered field is explicitly calculated from the two-time atomic dipole moment correlation function, which is evaluated on a Markoff-type assumption, and various limiting cases are discussed. In Appendix the case is considered in which radiative damping between the higher excited state and the ground state is also present.

§1. Introduction

B. R. Mollow discussed the emission spectrum from two-level systems under monochromatic incident light of arbitrary strength for the first time.[1),2)] As regards three-level systems T. Takagahara *et al.*[3)] made extensive studies for second order optical processes and T. Tokihiro and E. Hanamura[4)] treated the corresponding problem for the strong incident field using Langevin equations for the averaged atomic operators in the Heisenberg picture with phenomenological relaxation constants. In the present paper we evaluate the power spectrum of the radiation scattered by a three-level atom driven by a strong incident field, near the atomic resonance frequency between the ground state and the higher excited state, following

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the method developed by B. R. Mollow[1),2)] in the Schrödinger picture, in order to clarify some aspects of the problem more thoroughly. The atom is assumed to be isolated and fixed in position, and come into equilibrium with the driving field through the effect of radiation damping. Other relaxation processes are omitted from our analysis for the sake of simplicity except some relaxation processes between the first excited state and the ground state, which must be taken into account in order to maintain steady emission of light from the atom. In the limit of weak driving fields, the power spectrum consists almost entirely of the (broad) Raman part, while in the limit of strong driving fields the power spectrum of the scattered field has two peaks, the distance of the peaks increasing as the intensity of the driving field increases.

In the next section of this paper, the basic model is introduced. In § 3 explicit solutions are presented for the time evolution of the reduced density operator for the atom. In § 4 the power spectrum of the scattered field is given, and various limiting cases are discussed. In Appendix are given explicit solutions for the time evolution of the reduced density operator for the atom and the atomic correlation function for the case in which radiative damping between the higher excited state and the ground state is also taken into account.

§ 2. Driven Three-Level Atom in Resonant Approximation

We take as our basic model a fixed atom with three energy eigenstates: the ground state $|1\rangle$, the first excited state $|2\rangle$ and the second excited state $|3\rangle$ with energies $\hbar\omega_1$, $\hbar\omega_2$ and $\hbar\omega_3$ $(\omega_2<\omega_3)$ respectively. An arbitrary atomic operator may then be expressed as a linear combination of the nine basis operators $|i\rangle\langle j|$, (i, j=1, 2, 3). The electric dipole moment operator $\boldsymbol{d}(t)$ of the atom is assumed to have its matrix elements between the states $|1\rangle$ and $|3\rangle$ and between the states $|2\rangle$ and $|3\rangle$. It is thus expressed as

$$\boldsymbol{d}(t)=\boldsymbol{\mu}_{31}(|3\rangle\langle 1|)(t)+\boldsymbol{\mu}_{32}\,(|3\rangle\langle 2|)(t)+\text{h.c.}, \tag{2.1}$$

with

$$\boldsymbol{\mu}_{\text{ij}}\equiv\langle \text{i}|\boldsymbol{d}|\text{j}\rangle, \quad (\text{i, j}=1, 2, 3). \tag{2.2}$$

The electric field $\boldsymbol{E}(\boldsymbol{r}, t)$ at position $\boldsymbol{r}$ and time t is the sum of positive and negative frequency parts

$$\boldsymbol{E}(\boldsymbol{r}, t)=(1/2)^{1/2}[\boldsymbol{E}^{(+)}(\boldsymbol{r}, t)+\boldsymbol{E}^{(-)}(\boldsymbol{r}, t)], \tag{2.3}$$

with

$$\boldsymbol{E}^{(+)}(\boldsymbol{r}, t)=\mathcal{E}(\boldsymbol{r}, t)\boldsymbol{e}_0+\text{i}(\hbar/V)^{1/2}\sum_k \omega_k{}^{1/2}\boldsymbol{e}_k b_k(t)\exp(\text{i}\boldsymbol{k}\boldsymbol{r}), \tag{2.4}$$

where ω_k, $\boldsymbol{e}_k$ and b_k denote the frequency, polarization vector and annihilation operator respectively for the field mode $\boldsymbol{k}$, and $\mathcal{E}(\boldsymbol{r}, t)\boldsymbol{e}_0$ stands for the positive frequency part of the classical driving electric field. We assume further that the incident classical field oscillating harmonically near the atomic resonance frequency $\omega_{31}\equiv\omega_3-\omega_1$ interacts only with the dipole moment between the states $|3\rangle$ and $|1\rangle$, while the scattered quantum-mechanical electromagnetic field interacts only with the dipole moment between the states $|3\rangle$ and $|2\rangle$. (The interaction of the scattered field with the dipole moment between the states $|3\rangle$ and $|1\rangle$ is taken into account in the Appendix.) In order to ensure steady emission from the atom we are forced to take into account some relaxation processes between the states $|2\rangle$ and $|1\rangle$, possibly other than radiation damping. We denote this relaxation constant by γ.

In the resonant approximation we are thus left with the following interaction Hamiltonian:

$$H_{\rm I}(t)=-\hbar[(|3\rangle\langle 1|)(t)\lambda\mathcal{E}(0, t)+(|3\rangle\langle 2|)(t){\rm i}\sum_{\boldsymbol{k}} g_{\boldsymbol{k}} b_{\boldsymbol{k}}(t)]+{\rm h.\,c.}, \tag{2.5}$$

with

$$\lambda\equiv(1/2)^{1/2}(\boldsymbol{\mu}_{31}\boldsymbol{e}_0)/\hbar \tag{2.6}$$

and

$$g_{\boldsymbol{k}}\equiv(\boldsymbol{\mu}_{32}\boldsymbol{e}_{\boldsymbol{k}})(\omega_{\boldsymbol{k}}/2\hbar V)^{1/2}. \tag{2.7}$$

The total Hamiltonian $H(t)$ of the system is

$$H(t)=H_0(t)+H_{\rm I}(t), \tag{2.8}$$

with

$$H_0(t)=\sum_{\rm j=1}^{3}\hbar\omega_{\rm j}(|{\rm j}\rangle\langle {\rm j}|)(t)+\sum_{\boldsymbol{k}}\hbar\omega_{\boldsymbol{k}} b^{\dagger}_{\boldsymbol{k}}(t) b_{\boldsymbol{k}}(t). \tag{2.9}$$

By integrating the equations of motion for the operators $b_{\boldsymbol{k}}(t)$ and $b^{\dagger}_{\boldsymbol{k}}(t)$, we get in the scattering region

$$\boldsymbol{E}^{(+)}(\boldsymbol{r}, t)=\boldsymbol{\varphi}(\boldsymbol{r})(|2\rangle\langle 3|)(t-r/c)+\boldsymbol{E}_{\rm f}^{(+)}(\boldsymbol{r}, t), \tag{2.10}$$

where

$$\boldsymbol{\varphi}(\boldsymbol{r})\equiv-(2^{1/2}\omega_{32}^2/4\pi c^2 r^3)(\boldsymbol{\mu}_{32}\times\boldsymbol{r})\times\boldsymbol{r}, \tag{2.11}$$

with

$$\omega_{32}\equiv\omega_3-\omega_2 \tag{2.12}$$

and the freely propagating field operator $\boldsymbol{E}_{\rm f}^{(+)}(\boldsymbol{r}, t)$ consists of a linear combination of annihilation operators $b_{\boldsymbol{k}}$. The first order field correlation function at $\boldsymbol{r}'=\boldsymbol{r}$ is

$$\begin{aligned} G^{(1)}{}_{\rm jk}(\boldsymbol{r}, t'\,;\ \boldsymbol{r}, t)&=\langle E^{(-)}{}_{\rm j}(\boldsymbol{r}, t')E^{(+)}{}_{\rm k}(\boldsymbol{r}, t)\rangle \\ &=\varphi_{\rm j}(\boldsymbol{r})\varphi_{\rm k}(\boldsymbol{r})g_{32;23}(t-t'),\quad ({\rm j, k=x, y, z}), \end{aligned} \tag{2.13}$$

where the atomic correlation function is defined by

$$g_{ij;kl}(t-t')=\langle(|i\rangle\langle j|)(t')(|k\rangle\langle l|)(t)\rangle,\quad (i, j, k, l=1, 2, 3). \tag{2.14}$$

The power spectrum of the scattered field is given by

$$I(\nu;\ \boldsymbol{r})\equiv\int_{-\infty}^{\infty}d\tau\ \exp(i\nu\tau)\sum_{j}G^{(1)}{}_{jj}(\boldsymbol{r}, 0\ ;\ \boldsymbol{r}, \tau)$$

$$=|\varphi(\boldsymbol{r})|^2\tilde{g}_{32;23}(\nu), \tag{2.15}$$

with

$$\tilde{g}_{ij;kl}(\nu)\equiv\int_{-\infty}^{\infty}d\tau\ \exp(i\nu\tau)g_{ij;kl}(\tau),\quad (i, j, k, l=1, 2, 3). \tag{2.16}$$

The total intensity of the scattered field is

$$I_{tot}(\boldsymbol{r})=\int I(\nu;\ \boldsymbol{r})d\nu=2\pi\langle\boldsymbol{E}^{(-)}(\boldsymbol{r}, t)\boldsymbol{E}^{(+)}(\boldsymbol{r}, t)\rangle$$

$$=2\pi|\varphi(\boldsymbol{r})|^2 g_{32;23}(0). \tag{2.17}$$

§ 3. Reduced Density Operator for the Atom

The Schrödinger density operator $\rho(t)$ for the joint system of field modes and atom is approximated on a Markoff assumption by the expression

$$\rho(t)=|0\rangle_{FF}\langle 0|\rho_a(t), \tag{3.1}$$

where the vacuum state $|0\rangle_F$ for the field is defined by

$$b_k|0\rangle_F=0. \tag{3.2}$$

The reduced density operator for the atom is defined by

$$\rho_a(t)=\mathrm{Tr}_F\rho(t), \tag{3.3}$$

where Tr_F means trace with respect to the fixed states of the field. In terms of the time development operator $U(t, t')$ for the Hamiltonian $H_0+H_I(t)$ in the Schrödinger picture, the reduced density oparator for the atom at time t is given by the relation

$$\rho_a(t)=\mathrm{Tr}_F[U(t, t')|0\rangle_{FF}\langle 0|\rho_a(t')U^{-1}(t, t')]. \tag{3.4}$$

In the present model for the atom, it obeys the differential equation

$$\frac{d}{dt}\rho_a(t)=\kappa|2\rangle\langle 3|\rho_a(t)|3\rangle\langle 2|$$

$$-(1/2)\kappa[|3\rangle\langle 3|\rho_a(t)+\rho_a(t)|3\rangle\langle 3|]$$

$$+\gamma|1\rangle\langle 2|\rho_a(t)|2\rangle\langle 1|$$

$$-(1/2)\gamma[|2\rangle\langle 2|\rho_a(t)+\rho_a(t)|2\rangle\langle 2|]$$
$$+i[-\sum_{j=1}^{3}\omega_j|j\rangle\langle j|+\lambda\mathcal{E}(0,t)|3\rangle\langle 1|$$
$$+\lambda^*\mathcal{E}^*(0,t)|1\rangle\langle 3|,\ \ \rho_a(t)], \tag{3.5}$$

where κ is the natural decay rate between the states $|3\rangle$ and $|2\rangle$ of the atom,

$$\kappa=|\mu_{32}|^2\omega_{32}^2/(3\pi\hbar c^3). \tag{3.6}$$

The density operator $\rho_a(t)$ may be expressed in terms of the basis operators as

$$\rho_a(t)=\sum_{l,m=1}^{3}C_{lm}(t)|l\rangle\langle m|, \tag{3.7}$$

where

$$C_{lm}(t)=\langle l|\rho_a(t)|m\rangle=\mathrm{Tr}[\rho(t)|m\rangle\langle l|]=C_{ml}(t)^*. \tag{3.8}$$

Substitution of eq. (3.7) into eq. (3.5) leads to the differential equations for $C_{lm}(t)$ or a column vector $X(t)$ defined by the expression

$$X(t)={}^t[C_{11}(t),\ C_{12}(t),\ C_{13}(t),\ C_{21}(t),\ C_{22}(t),\ C_{23}(t),\ C_{31}(t),\ C_{32}(t),\ C_{33}(t)]. \tag{3.9}$$

We shall be interested in the case in which the driving field oscillates harmonically at a frequency ω which is assumed to lie near the atomic resonance frequency ω_{31}, so that

$$\mathcal{E}(0,t)=\mathcal{E}_0\exp(-i\omega t), \tag{3.10}$$

where $\mathcal{E}_0$ is a complex constant. In that case we define a new column vector $X'(t)$, the corresponding elements of which being denoted by $C'_{lm}(t)$, by the following relation

$$X'(t)=B(t)X(t), \tag{3.11}$$

where $B(t)$ is the following 9×9 diagonal matrix

$$B(t)=\begin{pmatrix} 1 &&&&&&&& \\ & 1 &&&&&&& \\ && e^{-i\omega t} &&&&&& \\ &&& 1 &&&&& \\ &&&& 1 &&&& \\ &&&&& e^{-i\omega t} &&& \\ &&&&&& e^{i\omega t} && \\ &&&&&&& e^{i\omega t} & \\ &&&&&&&& 1 \end{pmatrix}, \tag{3.12}$$

all the other elements being zero.
Then the vector $X'(t)$ satisfies the differential equation

$$\frac{d}{dt}X'(t)=DX'(t), \tag{3.13}$$

where D is a 9×9 matrix independent of time. The expression for the matrix D is

$$D=\begin{pmatrix} 0 & 0 & -ie & 0 & \gamma & 0 & ie^* & 0 & 0 \\ 0 & a^* & 0 & 0 & 0 & 0 & 0 & ie^* & 0 \\ -ie^* & 0 & b & 0 & 0 & 0 & 0 & 0 & ie^* \\ 0 & 0 & 0 & a & 0 & -ie & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\gamma & 0 & 0 & 0 & \kappa \\ 0 & 0 & 0 & -ie^* & 0 & c & 0 & 0 & 0 \\ ie & 0 & 0 & 0 & 0 & 0 & b^* & 0 & -ie \\ 0 & ie & 0 & 0 & 0 & 0 & 0 & c^* & 0 \\ 0 & 0 & ie & 0 & 0 & 0 & -ie^* & 0 & -\kappa \end{pmatrix}, \tag{3.14}$$

where

$$a\equiv-\gamma/2-i\omega_{21}, \tag{3.15a}$$

$$b\equiv-\kappa/2-i\Delta\omega_1, \tag{3.15b}$$

$$c\equiv-(\gamma+\kappa)/2-i\Delta\omega_2, \tag{3.15c}$$

$$e\equiv\lambda\mathcal{E}_0, \tag{3.15d}$$

$$\Delta\omega_1\equiv\omega-\omega_{31}, \tag{3.15e}$$

$$\Delta\omega_2\equiv\omega-\omega_{32} \tag{3.15f}$$

and

$$\omega_{ij}\equiv\omega_i-\omega_j,\quad (i,j=1,2,3). \tag{3.15g}$$

The solution for $X'(t'+\tau)$ takes the form

$$X'(t'+\tau)=\mathscr{U}'(\tau;t')\,X'(t'), \tag{3.16}$$

where $\mathscr{U}'(\tau;t')$ is a 9×9 matrix satisfying the initial condition

$$\mathscr{U}'(0;t')=I, \tag{3.17}$$

I standing for the unit matrix. Similarly we get

$$X(t'+\tau)=\mathscr{U}(\tau;t')X(t'), \tag{3.18}$$

with

$$\mathscr{U}(\tau;\ t')=B^{-1}(t'+\tau)\mathscr{U}'(\tau;t')B(t') \tag{3.19}$$

and

$$\mathscr{U}(0;t')=\mathrm{I}. \tag{3.20}$$

By integrating eq. (3.13) the Laplace transform of the matrix $\mathscr{U}'(\tau;t')$ is found to be

$$\hat{\mathscr{U}}'(s)\equiv\int_0^\infty e^{-s\tau}\ \mathscr{U}'(\tau;t')d\tau=(s\mathrm{I}-D)^{-1}, \tag{3.21}$$

which is independent of t'.

In order to get the power spectrum of the scattered field in the steady situation, we use for $C_{lm}(t')$ in the expression for the power spectrum their asymptotic values $C'_{lm}(t'\to\infty)\equiv C'^{\infty}_{lm}$. Taking the Laplace transform of eq. (3.16) after setting $t'=0$, we get

$$\hat{X}'(s)=\hat{\mathscr{U}}'(s)X'(0)=(s\mathrm{I}-D)^{-1}X'(0). \tag{3.22}$$

The asymptotic values can be obtained by the relation

$$C'^{\infty}_{lm}=\lim_{s\to 0}s\hat{C}'_{lm}(s). \tag{3.23}$$

In evaluating these asymptotic values we may choose the initial state $X'(0)$ in eq. (3.22) arbitrarily as long as the normalization condition for the atomic density operator $\rho_a(t)$ is satisfied, namely

$$\sum_i C'_{ii}(0)=1.$$

Thus we may take $X'(0)$, for example, as

$$X'(0)={}^t[1,\ 0,\ 0,\ 0,\ 0,\ 0,\ 0,\ 0,\ 0]. \tag{3.24}$$

In this way we get

$$C'^{\infty}_{13}=i\gamma b^*e^*/[\gamma|b|^2+(2\gamma+\kappa)|e|^2]$$
$$=-2i\lambda^*\mathcal{E}_0^*\gamma(\kappa-2i\Delta\omega_1)/\{\gamma[\kappa^2+4(\Delta\omega_1)^2]+(2\gamma+\kappa)\Omega^2\}, \tag{3.25}$$

$$C'^{\infty}_{23}=0 \tag{3.26}$$

and

$$C'^{\infty}_{33}=\gamma|e|^2/[\gamma|b|^2+(2\gamma+\kappa)|e|^2]$$
$$=\gamma\Omega^2/\{\gamma[\kappa^2+4(\Delta\omega_1)^2]+(2\gamma+\kappa)\Omega^2\}, \tag{3.27}$$

where the Rabi frequency Ω is defined by

$$\Omega \equiv 2|\lambda||\mathcal{E}_0|. \tag{3.28}$$

The matrix elements of $\hat{\mathscr{U}}'(s)$ we need are

$$\hat{\mathscr{U}}'_{32;12}(s) = \mathrm{i}e/f(s) = \mathrm{i}\lambda\mathcal{E}_0/f(s) \tag{3.29}$$

and

$$\hat{\mathscr{U}}'_{32;32}(s) = (s-a^*)/f(s) = (s+\gamma/2-\mathrm{i}\omega_{21})/f(s), \tag{3.30}$$

where

$$\begin{aligned} f(s) &\equiv (s-a^*)(s-c^*)+|e|^2 \\ &= (s+\gamma/2-\mathrm{i}\omega_{21})[s+(1/2)(\gamma+\kappa)-\mathrm{i}\Delta\omega_2]+\Omega^2/4 \\ &= s^2+[(2\gamma+\kappa)/2-\mathrm{i}(\omega_{21}+\Delta\omega_2)]s \\ &\quad +\gamma(\gamma+\kappa)/4-\omega_{21}\Delta\omega_2+\Omega^2/4 \\ &\quad -\mathrm{i}(1/2)[\gamma\Delta\omega_2+(\gamma+\kappa)\omega_{21}]. \end{aligned} \tag{3.31}$$

The total intensity given by eq. (2.17) is thus

$$\begin{aligned} I_{\mathrm{tot}}(\boldsymbol{r}) &= 2\pi|\varphi(\boldsymbol{r})|^2 g_{32;23}(0) = 2\pi|\varphi(\boldsymbol{r})|^2\, C'^{\infty}{}_{33} \\ &= 2\pi|\varphi(\boldsymbol{r})|^2\gamma\Omega^2/\{\gamma[\kappa^2+4(\Delta\omega_1)^2]+(2\gamma+\kappa)\Omega^2\}. \end{aligned} \tag{3.32}$$

The intensity of the coherently scattered light is

$$\begin{aligned} I_{\mathrm{coh}}(\boldsymbol{r}) &= 2\pi\langle \boldsymbol{E}^{(-)}(\boldsymbol{r},t)\rangle\langle \boldsymbol{E}^{(+)}(\boldsymbol{r},t)\rangle \\ &= 2\pi|\varphi(\boldsymbol{r})|^2|C'^{\infty}{}_{23}|^2 = 0. \end{aligned} \tag{3.33}$$

§ 4. Power Spectrum of Scattered Field

The two-time atomic correlation function $g_{\mathrm{ij;kl}}(t-t')$ defined by eq. (2.14) can be expressed as

$$\begin{aligned} & g_{\mathrm{ij;kl}}(t-t') \\ &\quad = \mathrm{Tr}[|0\rangle_{\mathrm{FF}}\langle 0|\rho_{\mathrm{a}}(t')|\mathrm{i}\rangle\langle \mathrm{j}|U^{-1}(t,t')|\mathrm{k}\rangle\langle \mathrm{l}|U(t,t')], \end{aligned} \tag{4.1}$$

while the expectation value $C_{\mathrm{lk}}(t)$ of atomic operator at a given time defined by eq. (3.8) can be written in the form

$$C_{\mathrm{lk}}(t) = \mathrm{Tr}[|0\rangle_{\mathrm{FF}}\langle 0|\rho_{\mathrm{a}}(t')U^{-1}(t,t')|\mathrm{k}\rangle\langle \mathrm{l}|U(t,t')]. \tag{4.2}$$

Comparison of eqs. (4.1) and (4.2) shows that the expression for $g_{\mathrm{ij;kl}}(t-t')$ is obtained by substituting $\rho_{\mathrm{a}}(t')|\mathrm{i}\rangle\langle \mathrm{j}|$ for $\rho_{\mathrm{a}}(t')$ in the expression for $C_{\mathrm{lk}}(t)$. If we do these procedures in the expression

$$C_{1k}(t)=\sum_{m,n=1}^{3}\mathscr{U}_{1k;mn}(\tau;t')C_{mn}(t'), \tag{4.3}$$

we get

$$g_{ij;kl}(t-t')=\sum_{m=1}^{3}\mathscr{U}_{lk;mj}(\tau;t')C_{mi}(t'). \tag{4.4}$$

In particular we get

$$g_{32;23}(\tau;t')=\sum_{m=1}^{3}\mathscr{U}_{32;m2}(\tau;t')C_{m3}(t')$$

$$=\exp(-i\omega\tau)\sum_{m=1}^{3}\mathscr{U}'_{32;m2}(\tau)C'_{m3}(t'), \tag{4.5}$$

and its Laplace transform

$$\hat{g}_{32;23}(s)=\hat{\mathscr{U}}'_{32;12}(s+i\omega)C'^{\infty}_{13}+\hat{\mathscr{U}}'_{32;32}(s+i\omega)C'^{\infty}_{33}. \tag{4.6}$$

Since $\hat{g}_{32;23}(s)$ is regular on the imaginary axis of the s plane, the spectral correlation function $\tilde{g}_{32;23}(\nu)$ defined by eq. (2.16) can be evaluated as

$$\tilde{g}_{32;23}(\nu)=2\mathrm{Re}[\hat{g}_{32;23}(-i\nu)]=C'^{\infty}_{33}$$
$$\times\{\gamma(\omega_{32}-\nu)^2+(\gamma+\kappa)[\gamma(\gamma+\kappa)+\Omega^2]/4\}/|f(i(\omega-\nu))|^2. \tag{4.7}$$

The two roots of the equation $f(s)=0$ are given by

$$s_{\pm}=-(2\gamma+\kappa)/4+i(\omega_{21}+\Delta\omega_2)/2\pm\sqrt{R}/2, \tag{4.8}$$

with

$$R\equiv\kappa^2/4-(\Delta\omega_1)^2-\Omega^2-i\kappa\Delta\omega_1. \tag{4.9}$$

Let us first consider the case in which the incident field is exactly on resonance with the atomic frequency ω_{31} ($\Delta\omega_1=0$). The two roots s_+ and s_- are then

$$s_{\pm}=-(2\gamma+\kappa)/4+i\omega_{21}\pm\sqrt{D}/2 \tag{4.10}$$

With

$$D\equiv\kappa^2/4-\Omega^2. \tag{4.11}$$

If $D>0$, then $\tilde{g}(\nu)$ is given by

$$\tilde{g}(\nu)=\{\gamma\Omega^2/[\gamma\kappa^2+(2\gamma+\kappa)\Omega^2]\}$$
$$\times\Big[\frac{A_+}{(\nu-\omega_{32})^2+(\gamma+\kappa/2-\sqrt{D})^2/4}$$
$$+\frac{A_-}{(\nu-\omega_{32})^2+(\gamma+\kappa/2+\sqrt{D})^2/4}\Big], \tag{4.12}$$

where

$$A_{+}=(\gamma\kappa+2\Omega^{2}+2\gamma\sqrt{D})/(4\sqrt{D}) \tag{4.13}$$

and

$$A_{-}=-(\gamma\kappa+2\Omega^{2}-2\gamma\sqrt{D})/(4\sqrt{D}). \tag{4.14}$$

The spectrum consists of luminescence centered at $\nu=\omega_{32}$. If the incident field is weak enough so that the Rabi frequency is very small compared to the natural decay rate κ and the phenomenological relaxation constant γ, eq. (4.12) reduces to

$$\tilde{g}_{32;23}(\nu)=\frac{\gamma\Omega^{2}}{\kappa^{2}[(\nu-\omega_{32})^{2}+\gamma^{2}/4]} \quad \text{for } \Delta\omega_{1}=0 \text{ and } \Omega\ll\gamma,\kappa, \tag{4.15}$$

that is, the spectrum consists of the (broad) Raman scattering part with the width $\gamma/2$. If $D<0$, on the other hand, the two roots s_{+} and s_{-} are given by

$$s_{\pm}=-(2\gamma+\kappa)/4+i\omega_{21}\pm i(1/2)\sqrt{D'} \tag{4.16}$$

with

$$D'\equiv-D=\Omega^{2}-\kappa^{2}/4. \tag{4.17}$$

In this case we have

$$\begin{aligned}\tilde{g}_{32;23}(\nu)=&(1/2)C'^{\infty}{}_{33}\\ &\times\left[\frac{(\gamma+\kappa)-\kappa(\omega_{32}-\nu)/\sqrt{D'}}{(\omega_{32}-\nu-\sqrt{D'}/2)^{2}+(2\gamma+\kappa)^{2}/16}\right.\\ &\left.+\frac{(\gamma+\kappa)+\kappa(\omega_{32}-\nu)/\sqrt{D'}}{(\omega_{32}-\nu+\sqrt{D'}/2)^{2}+(2\gamma+\kappa)^{2}/16}\right],\end{aligned} \tag{4.18}$$

that is, the power spectrum consists of a superposition of two Lorentzians centered at $\nu=\omega_{32}\pm\sqrt{D'}/2$ with the same width $(2\gamma+\kappa)/4$.

For $\Delta\omega_{1}\neq0$ we give results for $\tilde{g}_{32;23}(\nu)$ in two limiting cases of interest: that of very low and that of very high incident field intensity. If the incident field is weak enough so that the Rabi frequency is much smaller than both the parameters γ and κ, we have

$$\tilde{g}_{32;23}(\nu)=\{\gamma\Omega^{2}/[\kappa^{2}+4(\Delta\omega_{1})^{2}]\}\frac{1}{(\omega-\omega_{21}-\nu)^{2}+\gamma^{2}/4} \quad \text{for } \Omega\ll\gamma,\kappa. \tag{4.19}$$

In this case the spectrum consists wholly of the (broad) Raman scattering part: the Lorentzian centered at $\nu=\omega-\omega_{21}$ with the width $\gamma/2$.

If the incident field is intense enough so that Ω is much greater than γ and κ, the two roots of $f(s)=0$ are approximately given by

$$s_+=-\sigma_++i(1/2)(\omega_{21}+\Delta\omega_2+\Omega') \tag{4.20a}$$

and

$$s_-=-\sigma_-+i(1/2)(\omega_{21}+\Delta\omega_2-\Omega') \tag{4.20b}$$

with

$$\sigma_+=(1/4)(2\gamma+\kappa+\kappa\Delta\omega_1/\Omega') \tag{4.21a}$$

and

$$\sigma_-=(1/4)(2\gamma+\kappa-\kappa\Delta\omega_1/\Omega'), \tag{4.21b}$$

where the parameter Ω' is defined by

$$\Omega'^2\equiv\Omega^2+(\Delta\omega_1)^2. \tag{4.22}$$

The spectral correlation function $\tilde{g}_{32;23}(\nu)$ in this case is then given by

$$\begin{aligned}\tilde{g}_{32;23}(\nu)=&\frac{\gamma\Omega^2}{\Omega'^2[(2\gamma+\kappa)\Omega^2+4\gamma(\Delta\omega_1)^2]}\\ &\times\left[\frac{B_+-\kappa\Omega^2(\nu_+-\nu)/(2\Omega')}{(\nu_+-\nu)^2+\sigma_+^2}\right.\\ &\left.+\frac{B_-+\kappa\Omega^2(\nu_--\nu)/(2\Omega')}{(\nu_--\nu)^2+\sigma_-^2}\right]\\ &\qquad\text{for } \Omega\gg\gamma,\kappa,\end{aligned} \tag{4.23}$$

where

$$\nu_+\equiv\omega_{32}+\Delta\omega_1/2-\Omega'/2, \tag{4.24a}$$

$$\nu_-\equiv\omega_{32}+\Delta\omega_1/2+\Omega'/2 \tag{4.24b}$$

and

$$B_+\equiv(2\gamma+\kappa)\Omega^2/4+\gamma(\Delta\omega_1)^2/2-\gamma\Delta\omega_1\Omega'/2 \tag{4.25a}$$

and

$$B_-\equiv(2\gamma+\kappa)\Omega^2/4+\gamma(\Delta\omega_1)^2/2+\gamma\Delta\omega_1\Omega'/2. \tag{4.25b}$$

If $\Delta\omega_1>0$, i.e. $\omega_{32}<\omega-\omega_{21}$, then ν_+ and ν_- approach ω_{32} and $\omega-\omega_{21}$ respectively, as the intensity of the incident field is reduced. On the other hand, if $\Delta\omega_1<0$, i.e. $\omega-\omega_{21}<\omega_{32}$, then ν_+ and ν_- approach $\omega-\omega_{21}$ and ω_{32} respectively, as Ω is reduced.

In the limit of very intensive incident fields, Ω is much greater than γ, κ and $|\Delta\omega_1|$, and we find that eqs. (4.23)-(4.25) reduce to

$$\tilde{g}_{32;23}(\nu)=\frac{\gamma}{4}\left[\frac{1}{(\nu_+-\nu)^2+(2\gamma+\kappa)^2/16}+\frac{1}{(\nu_--\nu)^2+(2\gamma+\kappa)^2/16}\right] \qquad (4.26)$$

with

$$\nu_+=\omega_{32}+\Delta\omega_1/2-\Omega/2 \qquad (4.27a)$$

and

$$\nu_-=\omega_{32}+\Delta\omega_1/2+\Omega/2. \qquad (4.27b)$$

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References

1) B. R. Mollow and M. M. Miller (1969) Ann. Phys. **52**, p. 464
2) B. R. Mollow (1969) Phys. Rev. **188**, p. 1969
3) T. Takagahara, E. Hanamura and R. Kubo (1977) J. Phys. Soc. Japan, **43**, p. 802, p. 811, p. 1522; (1978) ibid., **44**, p. 728, p. 742
4) T. Tokihiro and E. Hanamura (1980) Suppl. Progr. Theor. Phys. No. 69, p. 451

Appendix

In this appendix we give explicit expressions for the atomic correlation function for the case in which the radiative damping between the higher excited state $|3\rangle$ and the ground state $|1\rangle$ is taken into account. We denote this natural decay rate by κ_1 and that between the states $|3\rangle$ and $|2\rangle$ which we have so far written as κ by κ_2.

The positive frequency part of the scattered field is

$$\boldsymbol{E}^{(+)}(\boldsymbol{r},t)=\varphi_{31}(\boldsymbol{r})(|1\rangle\langle 3|)(t-r/c)$$
$$+\varphi_{32}(\boldsymbol{r})(|2\rangle\langle 3|)(t-r/c)+\boldsymbol{E}_f{}^{(+)}(\boldsymbol{r},t) \tag{A.1}$$

with obvious notations.

The differential equation for the atomic density operator $\rho_a(t)$ reads

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\rho_a(t)=&\kappa_1|1\rangle\langle 3|\rho_a(t)|3\rangle\langle 1|\\ &+\kappa_2|2\rangle\langle 3|\rho_a(t)|3\rangle\langle 2|\\ &-(1/2)(\kappa_1+\kappa_2)[|3\rangle\langle 3|\rho_a(t)+\rho_a(t)|3\rangle\langle 3|]\\ &+\gamma|1\rangle\langle 2|\rho_a(t)|2\rangle\langle 1|\\ &-(1/2)\gamma[|2\rangle\langle 2|\rho_a(t)+\rho_a(t)|2\rangle\langle 2|]\\ &+\mathrm{i}[-\sum_{j=1}^{3}\omega_j|j\rangle\langle j|+\lambda\mathcal{E}(0,t)|3\rangle\langle 1|\\ &+\lambda^*\mathcal{E}^*(0,t)|1\rangle\langle 3|,\ \rho_a(t)].\end{aligned} \tag{A.2}$$

This is equivalent to the differential equation for $X'(t)$

$$\frac{\mathrm{d}}{\mathrm{d}t}X'(t)=DX'(t), \tag{A.3}$$

where the time-independent matrix D is given by

$$D=\begin{pmatrix} 0 & 0 & -\mathrm{i}e & 0 & \gamma & 0 & \mathrm{i}e^* & 0 & \kappa_1\\ 0 & a^* & 0 & 0 & 0 & 0 & 0 & \mathrm{i}e^* & 0\\ -\mathrm{i}e^* & 0 & b' & 0 & 0 & 0 & 0 & 0 & \mathrm{i}e^*\\ 0 & 0 & 0 & a & 0 & -\mathrm{i}e & 0 & 0 & 0\\ 0 & 0 & 0 & 0 & -\gamma & 0 & 0 & 0 & \kappa_2\\ 0 & 0 & 0 & -\mathrm{i}e^* & 0 & c' & 0 & 0 & 0\\ \mathrm{i}e & 0 & 0 & 0 & 0 & 0 & b'^* & 0 & -\mathrm{i}e\\ 0 & \mathrm{i}e & 0 & 0 & 0 & 0 & 0 & c'^* & 0\\ 0 & 0 & \mathrm{i}e & 0 & 0 & 0 & -\mathrm{i}e^* & 0 & -\kappa_1-\kappa_2 \end{pmatrix}, \tag{A.4}$$

where a and e are given respectively by eq. (3.15a) and eq. (3.15d) and

$$b' \equiv -(\kappa_1+\kappa_2)/2-i\Delta\omega_1 \tag{A.5a}$$

and

$$c' \equiv -(\gamma+\kappa_1+\kappa_2)/2-i\Delta\omega_2. \tag{A.5b}$$

This expression for D for the present model is readily obtained from the corresponding expression for D in §3 given by eq. (3.14) simply by replacing κ, b and c with $\kappa_1+\kappa_2$, b' and c' respectively, except that zero of the (11, 33) element and κ of the (22, 33) element of the matrix D defined by eq. (3.14) must be replaced with κ_1 and κ_2 respectively.

The first-order field correlation function at $\boldsymbol{r}'=\boldsymbol{r}$ is

$$\begin{aligned} G^{(1)}{}_{jk}(\boldsymbol{r},t';\ \boldsymbol{r},t) &= \varphi_{31,j}(\boldsymbol{r})\varphi_{31,k}(\boldsymbol{r})g_{31;13}(t-t') \\ &+\varphi_{32,j}(\boldsymbol{r})\varphi_{32,k}(\boldsymbol{r})g_{32;23}(t-t') \\ &+\varphi_{31,j}(\boldsymbol{r})\varphi_{32,k}(\boldsymbol{r})g_{31;23}(t-t') \\ &+\varphi_{32,j}(\boldsymbol{r})\varphi_{31,k}(\boldsymbol{r})g_{32;13}(t-t'),\quad (j,k=x,y,z). \end{aligned} \tag{A.6}$$

The asymptotic values of the expectation values of the atomic density operator are

$$\begin{aligned} C'^{\infty}{}_{13} &= 4i\gamma b'^{*}e^{*}/[4\gamma|b'|^2+(2\gamma+\kappa_2)\Omega^2] \\ &= -2i\lambda^{*}\mathcal{E}_0{}^{*}(\kappa_1+\kappa_2-2i\Delta\omega_1)/ \\ &\quad \{\gamma[(\kappa_1+\kappa_2)^2+4(\Delta\omega_1)^2]+(2\gamma+\kappa_2)\Omega^2\}, \end{aligned} \tag{A.7a}$$

$$C'^{\infty}{}_{23}=0 \tag{A.7b}$$

and

$$\begin{aligned} C'^{\infty}{}_{33} &= \gamma\Omega^2/[4\gamma|b'|^2+(2\gamma+\kappa_2)\Omega^2] \\ &= \gamma\Omega^2/\{\gamma[(\kappa_1+\kappa_2)^2+4(\Delta\omega_1)^2]+(2\gamma+\kappa_2)\Omega^2\}. \end{aligned} \tag{A.7c}$$

With use of eq. (4.4) the atomic correlation functions are expressed as

$$\begin{aligned} g_{31;13}(\tau) &= \sum_{m=1}^{3}\mathscr{U}_{31;m1}(\tau;\ t'\to\infty)C^{\infty}{}_{m3} \\ &= \exp(-i\omega\tau)\sum_{m=1}^{3}\mathscr{U}'_{31;m1}(\tau)C'^{\infty}{}_{m3}, \end{aligned} \tag{A.8a}$$

$$\begin{aligned} g_{32;23}(\tau) &= \sum_{m=1}^{3}\mathscr{U}_{32;m2}(\tau;\ t'\to\infty)C^{\infty}{}_{m3} \\ &= \exp(-i\omega\tau)\sum_{m=1}^{3}\mathscr{U}'_{32;m2}(\tau)C'^{\infty}{}_{m3}, \end{aligned} \tag{A.8b}$$

$$g_{31;23}(\tau)=\sum_{m=1}^{3}\mathscr{U}_{32;m1}(\tau;\ t'\to\infty)C^{\infty}{}_{m3}$$

$$=\exp(-i\omega\tau)\sum_{m=1}^{3}\mathscr{U}'{}_{32;m1}(\tau)C'{}^{\infty}{}_{m3} \tag{A.8c}$$

and

$$g_{32;13}(\tau)=\sum_{m=1}^{3}\mathscr{U}_{31;m2}(\tau;\ t'\to\infty)C^{\infty}{}_{m3}$$

$$=\exp(-i\omega\tau)\sum_{m=1}^{3}\mathscr{U}'{}_{31;m2}(\tau)C'{}^{\infty}{}_{m3}. \tag{A.8d}$$

The power spectrum of the scattered field defined by eq. (2.15) is

$$I(\nu;\ \boldsymbol{r})=|\varphi_{31}(\boldsymbol{r})|^2\tilde{g}_{31;13}(\nu)+|\varphi_{32}(\boldsymbol{r})|^2\tilde{g}_{32;23}(\nu)$$
$$+(\varphi_{31}(\boldsymbol{r})\varphi_{32}(\boldsymbol{r}))[\tilde{g}_{31;23}(\nu)+\tilde{g}_{32;13}(\nu)]. \tag{A.9}$$

The total intensity of the scattered field $I_{\mathrm{tot}}(\boldsymbol{r})$ is given by

$$(1/2\pi)I_{\mathrm{tot}}(\boldsymbol{r})=|\varphi_{31}(\boldsymbol{r})|^2\,g_{31;13}(0)+|\varphi_{32}(\boldsymbol{r})|^2\,g_{32;23}(0)$$
$$+(\varphi_{31}(\boldsymbol{r})\varphi_{32}(\boldsymbol{r}))[g_{31;23}(0)+g_{32;13}(0)]$$
$$=(|\varphi_{31}(\boldsymbol{r})|^2+|\varphi_{32}(\boldsymbol{r})|^2)\,C'{}^{\infty}{}_{33}. \tag{A.10}$$

The intensity of the coherently scattered field $I_{\mathrm{coh}}(\boldsymbol{r})$ is given by

$$(1/2\pi)I_{\mathrm{coh}}(\boldsymbol{r})=|\varphi_{31}(\boldsymbol{r})|^2\,|C'{}^{\infty}{}_{13}|^2. \tag{A.11}$$

In the limit of the weak incident field, we have

$$I_{\mathrm{coh}}(\boldsymbol{r})/I_{\mathrm{tot}}(\boldsymbol{r})=|\varphi_{31}(\boldsymbol{r})|^2/(|\varphi_{31}(\boldsymbol{r})|^2+|\varphi_{32}(\boldsymbol{r})|^2). \tag{A.12}$$

The Laplace transforms of the atomic correlation functions are evaluated as

$$\hat{g}_{31;13}(s)=\hat{\mathscr{U}}'{}_{31;11}(s+i\omega)C'{}^{\infty}{}_{13}+\hat{\mathscr{U}}'{}_{31;31}(s+i\omega)C'{}^{\infty}{}_{33}, \tag{A.13a}$$

$$\hat{g}_{32;23}(s)=\hat{\mathscr{U}}'{}_{32;12}(s+i\omega)C'{}^{\infty}{}_{13}+\hat{\mathscr{U}}'{}_{32;32}(s+i\omega)C'{}^{\infty}{}_{33}, \tag{A.13b}$$

and

$$\hat{g}_{31;23}(s)=\hat{g}_{32;13}(s)=0, \tag{A.13c}$$

where

$$\hat{\mathscr{U}}'{}_{31;11}(s)=ie(s+\gamma)(s+\kappa_1+\kappa_2)(s-b')/[sf_1(s)]$$
$$=i\lambda\mathcal{E}_0(s+\gamma)(s+\kappa_1+\kappa_2)(s+\kappa_1/2+\kappa_2/2+i\Delta\omega_2)/[sf_1(s)] \tag{A.14}$$

and

$$\hat{\mathscr{U}}'_{31;31}(s)=[(s+\gamma)(s+\kappa_1+\kappa_2)(s-b')+(2s+2\gamma+\kappa_2)|e|^2]/f_1(s)$$
$$=[(s+\gamma)(s+\kappa_1+\kappa_2)(s+\kappa_1/2+\kappa_2/2+\mathrm{i}\Delta\omega_1)$$
$$+(2s+2\gamma+\kappa_2)\Omega^2/4]/f_1(s) \qquad \text{(A.15)}$$

with

$$f_1(s)\equiv(s+\gamma)(s+\kappa_1+\kappa_2)|b'|^2+(2s+\kappa_1+\kappa_2)(2s+2\gamma+\kappa_2)|e|^2$$
$$=(s+\gamma)(s+\kappa_1+\kappa_2)[(s+\kappa_1/2+\kappa_2/2)^2+(\Delta\omega_1)^2]$$
$$+(2s+\kappa_1+\kappa_2)(2s+2\gamma+\kappa_2)\Omega^2/4, \qquad \text{(A.16)}$$

and

$$\hat{\mathscr{U}}'_{32;12}(s)=\mathrm{i}e/f_2(s)=\mathrm{i}\lambda\mathcal{E}_0/f_2(s) \qquad \text{(A.17)}$$

and

$$\hat{\mathscr{U}}'_{32;32}(s)=(s-a^*)/f_2(s)=(s+\gamma/2-\mathrm{i}\omega_{21})/f_2(s) \qquad \text{(A.18)}$$

with

$$f_2(s)\equiv(s-a^*)(s-c'^*)+|e|^2$$
$$=[(2s+\gamma-2\mathrm{i}\omega_{21})(2s+\kappa_1+\kappa_2+\gamma-2\mathrm{i}\Delta\omega_2)+\Omega^2]/4. \qquad \text{(A.19)}$$

The spectral correlation function $\tilde{g}_{32;32}(\nu)$ which results from the transition between the atomic states $|3\rangle$ and $|2\rangle$ is given by

$$\tilde{g}_{32;32}(\nu)=C'^{\infty}_{33}\{\gamma(\omega_{32}-\nu)^2+(\gamma+\kappa_1+\kappa_2)$$
$$\times[\gamma(\gamma+\kappa_1+\kappa_2)+\Omega^2]/4\}/|f_2(\mathrm{i}(\omega-\nu))|^2. \qquad \text{(A.20)}$$

This expression is obtained from the corresponding expression given by eq. (4.7) for the case in which the parameter κ_1 is absent simply by replacing κ with $\kappa_1+\kappa_2$ except for the factor C'^{∞}_{33}, for which eq. (A.7c) must be used instead of eq. (3.27). Therefore the various expressions for the power spectrum given in § 4 can readily be translated into the corresponding expressions for the present case in which the relaxation constant κ_1 is taken into account.