

Quantum Theory of Multiwave Mixing with a Local Field Correction

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(Received: July 18, 1997)

In this paper, we calculate the four coefficients for the quantum theory of multiwave mixing including a local-field correction resulting from dipole-dipole interactions. We make contact with the semiclassical calculations of probe absorption and four-wave-mixing coupling coefficients, and illustrate the effects of local field corrections on resonance-fluorescence and coupled-mode-fluorescence spectra. The method uses the hybrid quantum-Langevin-equation master-equation approach of An and Sargent.

I. INTRODUCTION

Previously An and Sargent[1] have used a combination of Langevin and master-equation techniques to derive the master equation for quantum theory of the multiwave mixing in a very efficient way. It provides an alternative to the pure density-matrix methods of an earlier paper[2] in their series on the quantum theory of multiwave mixing. It separates the purely quantum-field part from the essentially semiclassical role played by the medium. The coefficients in the master equation result from integrals over the double commutator in second-order perturbation theory and are Laplace transforms of two-time correlations of the medium spin operators. In turn, these transforms provide the most efficient way of solving the Bloch equations for the two-time correlations as given by the quantum-regression theorem. As such, the medium is described semiclassically, which is a substantial simplification over alternative methods. The technique is familiar in the calculation of resonance fluorescence. In addition, the method can reveal how population pulsations lead to the Rabi sidebands in the absorption and resonance-fluorescence spectra.

The effects of a local field correction (LFC) on the weak-field dielectric constant were first treated by Lorentz.[3] According to a prescription described by Lorentz, the local and macroscopic fields within an isotropic environment are related by

$$\vec{E}_{loc} = \vec{E} + \left(\frac{4\pi}{3}\right)\vec{P}, \quad (1)$$

where \vec{P} denotes the polarization of the material system. In this paper, we generalize the Lorentz correction and calculate the four coefficients for the quantum theory of multiwave mixing including LFC resulting from dipole-dipole interactions[4] by using the hybrid quantum-Langevin-equation master-equation approach of An and Sargent. We make contact with the semiclassical calculations of probe absorption and four-wave-mixing coupling coefficients, and illustrate the effects of LFC on resonance-fluorescence and coupled-mode-fluorescence spectra. We find that in addition to the real contribution derived in earlier work, the LFC inevitably contains an imaginary contribution, which can influence the shape of the multiwave mixing spectra.

II. QUANTUM LANGEVIN BLOCH EQUATIONS

We consider three modes of the electromagnetic field with frequencies ν_1, ν_2 and ν_3 that are arbitrarily detuned (consistent with the rotating-wave approximation) from the atomic resonance ω , ν_1 and ν_3 are placed symmetrically on opposite sides of ν_2 , that is, $\nu_3 = \nu_2 + (\nu_2 - \nu_1)$ as depicted in Fig. 1. Our Hamiltonian of the two-level atom-field system in the frame rotating at the pump frequency ν_2 is

$$H = \hbar\delta S_z - \hbar\Delta(a_1^\dagger a_1 - a_3^\dagger a_3) + \hbar[V_2 S_+ + g S_+(a_1 e^{\Delta t} U_1 + a_3 e^{-i\Delta t} U_3) + \text{adjoint}] \quad (2)$$

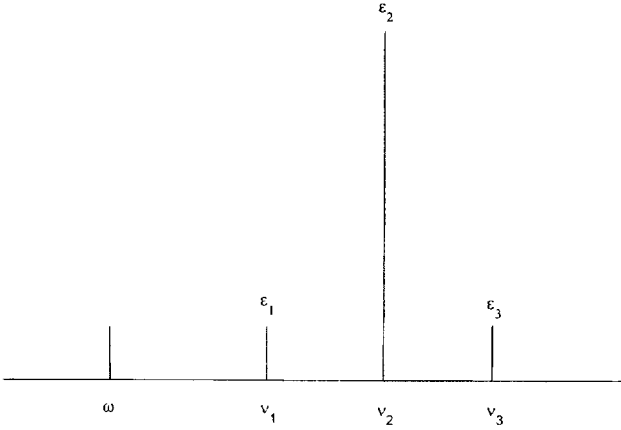


FIG. 1. Spectrum of three-wave fields. Waves with frequencies ν_1 and ν_2 are taken to be weak (nonsaturating), while the ν_2 wave is allowed to be arbitrarily intense.

where the Pauli spin operators are defined by

$$2S_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (3)$$

$$S_+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad (4)$$

$$S_- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \quad (5)$$

such that S_- flips the system from upper level to lower level, S_+ flips from lower to upper, the frequency differences $\delta = \omega - \nu_2$ and $\Delta = \nu_2 - \nu_1$, a_j is the annihilation operator for the j th field mode, the pump-mode interaction energy

$$V_2 = pE_2 U_2/2, \quad (6)$$

p is the electric-dipole matrix element, g is the atom-field coupling constant, E_2 is the pump-mode amplitude, and $U_j = U_j(r)$ is the spatial mode factor for mode j .

Introducing the noise operators, we easily find that the spin operators obey the quantum mechanical Langevin-Bloch equations.[1,5]

$$\dot{S}_+ = -(\gamma - i\delta)S_+ - 2iV_2^* S_z + F_+(t), \quad (7)$$

$$\dot{S}_z = -\Gamma(S_z + \frac{1}{2}) + iV_2^* S_- - iV_2 S_+ + F_z(t), \quad (8)$$

$$\dot{S}_- = -(\gamma + i\delta)S_- + 2iV_2 S_z + F_-(t), \quad (9)$$

where γ is the dipole decay constant ($\equiv 1/T_2$), F_α are Markovian noise operators, and Γ is the population difference decay constant ($\equiv 1/T_1$). In terms of the two-level density matrix, $\rho_{ab} = \langle S_- \rangle$ and $\rho_{aa} - \rho_{bb} =$

$2\langle S_z \rangle$.

The effects of LFC on the weak-field dielectric constant were first treated by Lorentz. Generalizing Eq.(1) to the system described by the Hamiltonian (2), we can write down the quantum Langevin Bloch equations including LFC in the form

$$\dot{S}_+ = -(\gamma - i\delta)S_+ - 2iV'^t S_z + F_+(t) \quad (10)$$

$$\dot{S}_z = -\Gamma(S_z + \frac{1}{2}) + iV'^t S_- - iV'^t S_+ + F_z(t) \quad (11)$$

$$\dot{S}_- = -(\gamma + i\delta)S_- + 2iV' S_z + F_-(t), \quad (12)$$

where the interaction energy V' including LFC is given, on the analogy of Eq.(1), by

$$V' = V_2 + \epsilon S_-, \quad (13)$$

and the complex constant ϵ determines the magnitude and phase of the local field correction. Since Eqs. (10) through (12) are nonlinear in the spin operators, we linearize about the semiclassical operating point by writing

$$S_\pm(t) = \langle S_\pm(t) \rangle + \delta S_\pm(t), \quad (14)$$

$$S_z(t) = \langle S_z(t) \rangle + \delta S_z(t), \quad (15)$$

Substituting these expressions into the equations of motion (10) through (12), and retaining terms linear in the fluctuation operators δS_i , we find the linearized Langevin Bloch equations

$$\frac{d\delta S_+}{dt} = -(\gamma - i\delta + 2i\epsilon^* \langle S_z \rangle) \delta S_+ - 2iV_2'^* \delta S_z + F_+(t) \quad (16)$$

$$\begin{aligned} \frac{d\delta S_z}{dt} &= -\Gamma \delta S_z + i[V_2'^* + \epsilon^* \langle S_+ \rangle] \delta S_- + i\epsilon^* \delta S_+ \langle S_- \rangle \\ &\quad - i[\delta V_2 + \epsilon \langle S_- \rangle] \delta S_+ - i\epsilon \delta S_- \langle S_+ \rangle + F_z(t) \\ &= -\Gamma \delta S_z + iV_2'^* \delta S_- - iV_2' \delta S_+ \\ &\quad + 2i\epsilon_i [\langle S_- \rangle \delta S_+ + \langle S_+ \rangle \delta S_-] + F_z(t) \end{aligned} \quad (17)$$

$$\frac{d\delta S_-}{dt} = -(\gamma + i\delta - 2i\epsilon \langle S_z \rangle) \delta S_- + 2iV_2' \delta S_z + F_-(t), \quad (18)$$

where the semiclassical pump interaction energy V_2' is given by

$$V_2' = V_2 + \epsilon \langle S_- \rangle. \quad (19)$$

The semiclassical steady-state values are given by

$$\langle S_- \rangle = 2iV_2 \langle S_z \rangle D_2' \quad (20)$$

$$\langle S_z \rangle = -\frac{1}{2} \frac{1}{1 + I'_2 L'_2} \quad (21)$$

where the renormalized complex denominator

$$D'_2 = \frac{1}{\gamma + i(\delta - 2\epsilon \langle S_z \rangle)}, \quad (22)$$

the renormalized saturator intensity I'_2 is given by

$$I'_2 = \frac{4|V'_2|}{\Gamma\gamma}, \quad (23)$$

and the modified saturator dimensionless Lorentzian is given by

$$L'_2 = \frac{\gamma^2}{(\gamma + \epsilon_i \langle S_z \rangle)^2 + (\omega - \nu_2 - \epsilon_r \langle S_z \rangle)^2}. \quad (24)$$

Note that Eq. (21) is a recursive formula for $\langle S_z \rangle$, which converges by iteration for sufficiently small $|\epsilon|$.

The equations of motion (16) through (18) can be written in the matrix form

$$\frac{d}{dt} \delta S_i(t) = \sum_k B_{ik} \delta S_j(t) + F_i(t), \quad (25)$$

where $i, k = (+, z, -)$ and the matrix B is given by

$$B = \begin{pmatrix} -\gamma - i\delta - 2i\epsilon^* \langle S_z \rangle & -2iV'_2{}^* & 0 \\ -iV_2 + 2\epsilon_i \langle S_- \rangle & -\Gamma & iV_2^* + 2\epsilon_i \langle S_+ \rangle \\ 0 & 2iV'_2 & -\gamma - i\delta + 2i\epsilon \langle S_z \rangle \end{pmatrix} \quad (26)$$

To evaluate the quantum multiwave-mixing coefficients, we need the Laplace transforms of various two-

time correlation functions $\langle \delta S_i(\tau) \delta S_j(0) \rangle$ of the fluctuations $\delta S_i(\tau)$ of the atomic operators about their semiclassical mean values. Eq.(25) gives for the evolution of the two-time correlation functions

$$\begin{aligned} \frac{d}{d\tau} \langle \delta S_i(\tau) \delta S_j(0) \rangle &= \langle (d\delta S_j(\tau)/d\tau) \delta S_j(0) \rangle \\ &= \sum_k B_{ik} \langle \delta S_k(\tau) \delta S_j(0) \rangle + \langle F_i(\tau) \delta S_j(0) \rangle. \end{aligned} \quad (27)$$

Since the noise operator $F_i(\tau)$ can not influence $\delta S_j(0)$ for $\tau > 0$, we find

$$\frac{d}{d\tau} \langle \delta S_i(\tau) S_j(0) \rangle = \sum_k B_{ik} \langle \delta S_k(\tau) \delta S_j(0) \rangle, \text{ for } \tau > 0 \quad (28)$$

that is, the two-time correlation functions $\langle \delta S_i(\tau) \delta S_j(0) \rangle$ obey the same equations of motion as the single time functions $\langle \delta S_i(\tau) \rangle$. This is a statement of the quantum regression theorem for the $\delta S_i(\tau)$. [5] Taking the Laplace transform of the differential Eq.(28), we find the algebraic matrix equation

$$(sI - B) \begin{pmatrix} \delta S_{+j}(s) \\ \delta S_{zj}(s) \\ \delta S_{-j}(s) \end{pmatrix} = \begin{pmatrix} \langle \delta S_{+}(0) \delta S_j(0) \rangle \\ \langle \delta S_z(0) \delta S_j(0) \rangle \\ \langle \delta S_{-}(0) \delta S_j(0) \rangle \end{pmatrix}, \quad (29)$$

where I is the identity matrix, the Laplace transform $\delta S_{ij}(s)$ is given by

$$\delta S_{ij}(s) = \int_0^\infty dt e^{-st} \langle \delta S_i(\tau) \delta S_j(0) \rangle, \quad (30)$$

the matrix

$$sI - B = \begin{pmatrix} \gamma - i\delta + 2i\epsilon^* \langle S_z \rangle + s & 2iV'_2{}^* & 0 \\ iV_2 - 2\epsilon_i \langle S_- \rangle & \Gamma + s & -iV_2^* - 2\epsilon_i \langle S_+ \rangle \\ 0 & -2iV'_2 & \gamma + i\delta - 2i\epsilon \langle S_z \rangle - s \end{pmatrix} = \begin{pmatrix} v_1^* & 2iV'_2{}^* & 0 \\ iV_2'' & v_2^* & -iV_2''^* \\ 0 & -2iV'_2 & v_3 \end{pmatrix}, \quad (31)$$

the complex frequencies v_i are defined by [note that $s = -i\Delta = -i(\nu_2 - \nu_1)$]

$$v'_n = \gamma + i\delta - 2i\epsilon \langle S_z \rangle + (n-2)s \quad (32)$$

$$v = \Gamma - s = \Gamma + i\Delta, \quad (33)$$

and the modified interaction energy V_2'' is given by

$$V_2'' = V_2 + 2i\epsilon_i \langle S_- \rangle = V_2' - \epsilon^* \langle S_- \rangle. \quad (34)$$

The solutions for the fluctuation spectra $\delta S_{ij}(s)$ are given by the method of determinants using appropriate initial conditions. All solutions require the determinant of the matrix $sI - B$, which is given by

$$D'(s) = v^* v_3' v_1'^* + 2V_2' V_2''^* v_1'^* + 2V_2'^* V_2'' v_3' \quad (35)$$

The initial conditions on the RHS of Eq.(29) are given by the general expression

$$\langle \delta S_i(0) \delta S_j(0) \rangle = \langle S_i(0) S_j(0) \rangle - \langle S_i(0) \rangle \langle S_j(0) \rangle. \quad (36)$$

III. CALCULATION OF THE QUANTUM MULTI-WAVE MIXING COEFFICIENTS

The four quantum multiwave-mixing coefficients A_1, B_1, C_1 , and D_1 appear in the dual-sidemode reduced-density-operator master equation

$$\begin{aligned} \dot{\rho} = & -A_1(\rho a_1 a_1^\dagger - a_1^\dagger \rho a_1) - B_1(a_1^\dagger a_1 \rho - a_1 \rho a_1^\dagger) \\ & + D_1(\rho a_3^\dagger a_1^\dagger - a_1^\dagger \rho a_3^\dagger) + C_1(a_1^\dagger a_3^\dagger \rho - a_3^\dagger \rho a_1^\dagger) \\ & + (\text{same with } 1 \rightarrow 3) + \text{adjoint}. \end{aligned} \quad (37)$$

and are given in general by the Laplace transforms

$$\langle \delta S_z(0) \delta S_-(0) \rangle = -\langle S_a \rangle \langle S_- \rangle \quad (43)$$

$$A_1 = [g^2 \delta S_{+-}(s)]^*, \quad (38)$$

$$\langle \delta S_-(0) \delta S_-(0) \rangle = -\langle S_- \rangle^2. \quad (44)$$

$$B_1 = g^2 \delta S_{-+}(s^*), \quad (39)$$

$$C_1 = -g^2 \delta S_{--}(s^*), \quad (40)$$

$$D_1 = -[g^2 \delta S_{++}(s)]^*. \quad (41)$$

They have the physical meanings that $A_1 + c.c$ gives the resonance fluorescence spectrum,[6] $B_1 - A_1$ gives the semiclassical probe absorption coefficient,[7] $C_1 + C_3$ gives the coupled-mode fluorescence spectrum, and $D_1 - C_1$ gives the semiclassical four-wave mixing coupling coefficient.[2] To calculate A_1 and C_1 , we solve for $\delta S_{+-}(s)$ and $\delta S_{--}(s)$ by applying the method of determinants to Eqs.(29) and (36) with $\delta S_j = \delta S_-$, and for B_1 and D_1 we solve these equation with $\delta S_j = \delta S_+$. Eq.(36) gives the initial conditions for $\delta S_j = \delta S_-$ as

$$\langle \delta S_+(0) \delta S_-(0) \rangle = \langle S_a \rangle - |\langle S_- \rangle|^2 \quad (42)$$

The resonance fluorescence coefficient $A_1 = [g^2 \delta S_{+-}(s)]^*$, where $D'(s)S_{+-}(s)$ is given by the determinant of the matrix

$$\begin{pmatrix} \langle S_a \rangle - |\langle S_- \rangle|^2 & 2iV_2'^* & 0 \\ -\langle S_a \rangle \langle S_- \rangle & v^* & -iV_2''^* \\ -\langle S_- \rangle^2 & -2iV_2' & v_3' \end{pmatrix}$$

that is,

$$D'(s)\delta S_{+-}(s) = (\langle S_a \rangle - |\langle S_- \rangle|^2)[v^*v_3' + 2V_2''^*V_2'] - 2iV_2'^*[-\langle S_a \rangle \langle S_- \rangle v_3' - iV_2''^* \langle S_- \rangle^2]. \quad (45)$$

For vanishing local-field correction ($\epsilon = 0$), we have

$$D(s)\delta S_{+-}(s) = (\langle S_- \rangle - |\langle S_- \rangle|^2)[v^*v_3 + 2|V_2|^2] + 2iV_2'^* \langle S_- \rangle [\langle S_a \rangle v_3 + iV_2^* \langle S_- \rangle]. \quad (46)$$

Substituting Eq.(45) into Eq.(38), we have the resonance-fluorescence coefficient

$$A_1 = g^2 \frac{(\langle S_a \rangle - |\langle S_- \rangle|^2)[vv_3'^* + 2V_2''V_2'^*] - 2V_2'[i\langle S_a \rangle \langle S_+ \rangle v_3'^* + V_2'' \langle S_+ \rangle^2]}{vv_3'^*v_1' + 2V_2'^*V_2v_1' + 2V_2'V_2''^*v_3'^*}. \quad (47)$$

The coupled-mode fluorescence coefficient C_1 is given by $-g^2 \delta S_{--}(s^*)$, where $D'(s)\delta S_{--}(s)$ is given by determinant of the matrix

$$\begin{pmatrix} v_1'^* & 2iV_2'^* & \langle S_a \rangle - |\langle S_- \rangle|^2 \\ iV_2''^* & v^* & -\langle S_a \rangle \langle S_- \rangle \\ 0 & -2iV_2' & -\langle S_- \rangle^2 \end{pmatrix}$$

This gives

$$D'(s)\delta S_{--}(s) = -\langle S_- \rangle^2[v^*v_1'^* + 2V_2'^*V_2''] + 2V_2'[-i\langle S_a \rangle \langle S_- \rangle v_1'^* + V_2''(\langle S_a \rangle - |\langle S_- \rangle|^2)]. \quad (48)$$

For $\epsilon = 0$, we have

$$D(s)\delta S_{--}(s) = -\langle S_- \rangle^2[v^*v_1^* + 2|V_2|^2] + 2V_2[-i\langle S_a \rangle \langle S_- \rangle v_1^* + V_2(\langle S_a \rangle - |\langle S_- \rangle|^2)]. \quad (49)$$

Substitution Eq.(48) into Eq.(40) and noting that $v_1(s^*) = v_3(s)$, $v_3(s^*) = v_1$, $v(s^*) = v^*(s)$ and $D'(s^*) = [D'(s)]^*$, we have the coupled-mode fluorescence coefficient

$$C_1 = g^2 \frac{\langle S_- \rangle^2 [vv_3'^* + 2V_2''V_2'^*] + 2V_2'[iv_3'^* \langle S_- \rangle \langle S_a \rangle - V_2''(\langle S_a \rangle - |\langle S_- \rangle|^2)]}{vv_3'^*v_1' + 2V_2'^*V_2''v_3' + 2V_2'V_2''^*v_3'^*}. \quad (50)$$

For the B_1 and D_1 coefficients, we choose $\delta S_j(0)$ in Eqs.(29) and (36) to be $\delta S_+(0)$. Eq.(36) gives the corresponding initial conditions

$$\langle \delta S_+(0) \delta S_+(0) \rangle = -\langle S_+ \rangle^2 \quad (51)$$

$$\langle \delta S_z(0) \delta S_+(0) \rangle = \langle S_+ \rangle \langle S_b \rangle \quad (52)$$

$$\langle \delta S_-(0) \delta S_+(0) \rangle = \langle S_b \rangle - |\langle S_- \rangle|^2. \quad (53)$$

Hence to find $\delta S_{--}(s)$ and $\delta S_{-+}(s)$, in Eqs.(45) and (49) we replace the initial conditions given by Eqs. (42) through (44) by those given by Eqs. (51) through (52), respectively, e.g., $\langle \delta S_+(0) \delta S_-(0) \rangle = \langle S_a \rangle - |\langle S_- \rangle|^2$ is replaced by $\langle \delta S_+(0) \delta S_+(0) \rangle = -\langle S_+ \rangle^2$. This gives

$$D'(s)\delta S_{++}(s) = -\langle S_+ \rangle^2[v^*v_3' + 2V_2''^*V_2'] + 2V_2'^*[-i\langle S_b \rangle \langle S_+ \rangle v_3' + V_2''^*(\langle S_b \rangle - |\langle S_- \rangle|^2)]. \quad (54)$$

$$D'(s)\delta S_{-+}(s) = (\langle S_b \rangle - |\langle S_- \rangle|^2)[v^*v'_1 + 2V_2'^*V_2''] + 2V_2'[2\langle S_b \rangle\langle S_+ \rangle v_1'^* + V_2''\langle S_+ \rangle^2]. \quad (55)$$

Substituting Eqs.(55) and (54) into Eqs.(39) and (41), respectively, we have

$$B_1 = g^2 \frac{(\langle S_b \rangle - |\langle S_- \rangle|^2)[vv_3'^* + 2V_2'^*V_2''] + 2V_2'[2\langle S_b \rangle\langle S_+ \rangle v_3'^* - V_2''\langle S_+ \rangle^2]}{vv_3'^*v'_1 + 2V_2'^*V_2''v'_1 + 2V_2'V_2''^*v_3'^*} \quad (56)$$

$$D_1 = g^2 \frac{\langle S_- \rangle^2[vv_3'^* + 2V_2'^*V_2''] - 2V_2'[2v_3'^*\langle S_- \rangle\langle S_b \rangle + V_2''(\langle S_b \rangle - |\langle S_- \rangle|^2)]}{vv_3'^*v'_1 + 2V_2'^*V_2''v'_1 + 2V_2'V_2''^*v_3'^*} \quad (57)$$

To calculate the semiclassical absorption coefficient $\alpha_1 = B_1 - A_1$, we calculate the difference

$$B_1 - A_1 = g^2\delta S_{-+}(s^*) - g^2[\delta S_{-+}(s)]^*$$

Noting that $D'(s^*) = [D'(s)]^*$, we have

$$\frac{B_1 - A_1}{g^2} = \frac{-2\langle S_z \rangle[vv_3'^* + 2V_2'^*V_2''] + 2iV_2'\langle S_+ \rangle v_3'^*}{vv_3'^*v'_1 + 2V_2'^*V_2''v'_1 + 2V_2'V_2''^*v_3'^*} \quad (58)$$

Furthermore for $\epsilon = 0$, we have

$$\begin{aligned} \frac{B_1 - A_1}{g^2} &= \frac{-2\langle S_z \rangle[vv_3'^* + 2|V_2|^2] + 2iV_2\langle S_+ \rangle v_3'^*}{vv_3'^*v'_1 + 2|V_2|^2(v_1 + v_3'^*)} \\ &= -2\langle S_z \rangle D_1 \frac{v + 2|V_2|^2(D_3^* - D_2^*)}{v + 2|V_2|^2(D_1 + D_3^*)} \\ &= -2\langle S_z \rangle D_1 \left[1 - \frac{2|V_2|^2(D_1 + D_2^*)}{v + 2|V_2|^2(D_1 + D_3^*)} \right] \end{aligned} \quad (59)$$

where the complex denominator

$$D_n = \frac{1}{v_n}. \quad (60)$$

Eq.(59) agrees with the standard semiclassical probe-absorption coefficient given by Eq.(8.15) of Ref. 5. Similarly subtracting Eq.(57) from (50), we have the

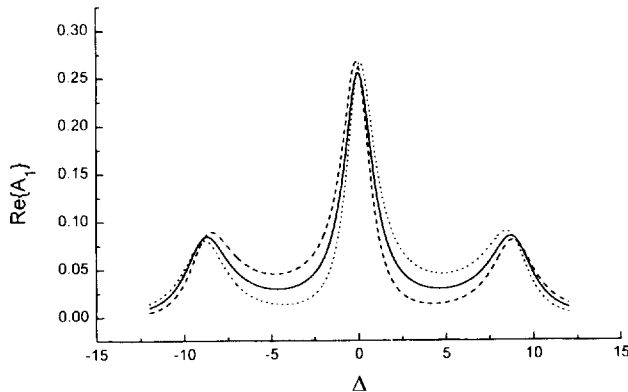


FIG. 2. Real part of A_1 versus the pump-probe frequency Δ for $\epsilon_i = 0$ and $\epsilon_r = -5$ (dashed line), 0 (solid line), and 5 (dotted line) with $\delta = 0, g = 1$, and $I_2 = 40$. All frequencies are given in unit of γ .

semiclassical coupling coefficient

$$\frac{C_1 - D_1}{g^2} = -\frac{4V_2'\langle S_z \rangle[V_2v_3'^*D_2' + V_2'']}{vv_3'^*v'_1 + 2V_2'^*V_2''v'_1 + 2V_2'V_2''^*v_3'^*} \quad (61)$$

For $\epsilon = 0$, this reduces to

$$\frac{C_1 - D_1}{g^2} = -\frac{4D_1V_2^2\langle S_z \rangle(D_2 + D_3^*)}{v + 2|V_2|^2(D_1 + D_3^*)}, \quad (62)$$

which agrees with Eq.(9.21) of Ref. 5.

Figs. 2 and 3 illustrate the resonance-fluorescence spectrum $A_1 + c.c.$ versus the pump-probe frequency

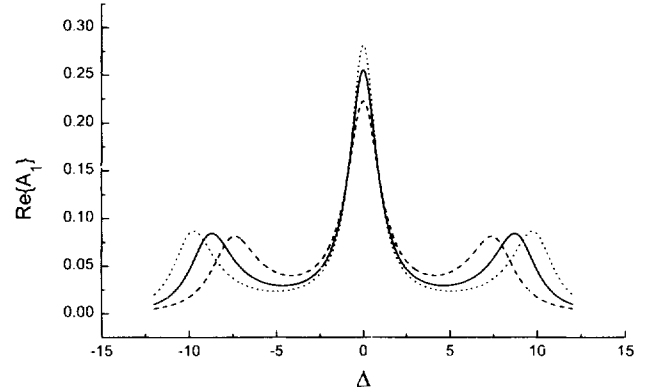


FIG. 3. Real part of A_1 versus the pump-probe frequency Δ for $\epsilon_r = 0$ and $\epsilon_i = -5$ (dashed line), 0 (solid line), and 5 (dotted line). Other parameters are the same as in Fig. 2.

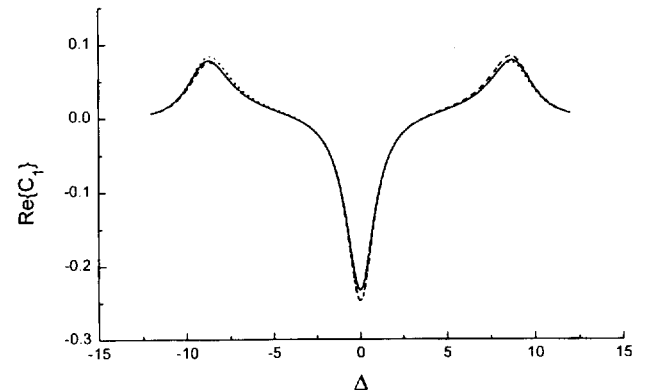
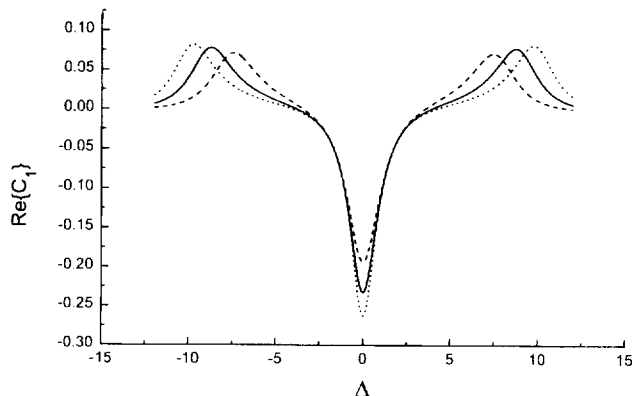


FIG. 4. Real part of C_1 corresponding to Fig. 2.

FIG. 5. Real part of C_1 corresponding to Fig. 3.

Δ for various cases of LFC parameter ϵ . In Fig.2 the dashed line, solid line, and dotted line plot the resonance fluorescence spectrum for $\epsilon_r = -5, 0,$ and $5,$ respectively, when $\epsilon_i = 0$. We see that the resonance fluorescence spectrum shows asymmetric shapes with respect to Δ as ϵ_r varies. On the other hand, Fig. 3 plots the resonance fluorescence spectrum for $\epsilon_i = -5, 0,$ and 5 when $\epsilon_r = 0$. In this case the spectrum shows symmetric shapes as ϵ_i varies. We also find that the width of the peak in the spectrum becomes narrower as ϵ_i increases from negative to positive values. Figs. 4 and 5 plot the coupled mode coefficient $C_1 + c.c.$ for the same parameters as in Figs. 2 and 3, showing behavior similar to that of the resonance fluorescence spectrum.

IV. CONCLUSIONS

We have shown that an approach using the quantum Langevin Bloch equation together with the quantum regression theorem derives straightforwardly the

quantum multiwave mixing coefficients including LFC. We have checked that without LFC (i.e., $\epsilon = 0$) these coefficients reduce to the standard multiwave mixing coefficients. By plotting the coefficients we have seen that the spectrum of the coefficients shows symmetrical or asymmetrical behavior, depending on the phase of ϵ (i.e., $\epsilon_r = 0$ or $\epsilon_i = 0$). We expect that the changes in the spectral shapes will provide one possible way of measuring the effect of local field corrections. Although we did not try to understand the microscopic origin of the LFC parameter ϵ in this paper, we hope to discuss about it in detail in a future publication.

ACKNOWLEDGMENTS

This work was supported by the Basic Science Institute Program, Ministry of Education of Korea project BSRI '96 - 2415 and in part by KOSEF grant (1996) 95-0701-02-01-3.

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