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Theory of coherent, degenerate two-photon absorption and emission

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Abstract. The theory of the coherent, two-photon resonant interaction of a monochromatic field with N atoms is given. It is seen that the dynamics of the atom-field system can be completely determined when the field is "strong". Two specific examples are given: (i) two-photon absorption by atoms in ground state, and (ii) stimulated two-photon emission by fully excited atoms, assuming a coherent field in both cases. In case (ii), the field shows photon-antibunching after the decay of half of the atoms. The merits of our approach are shown by comparing with other treatments. Our results can also be applied to certain degenerate four-wave mixing processes which are described by a similar Hamiltonian.

Keywords. Two-photon absorption; two-photon emission; coherent two-photon processes; photon antibunching.

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1. Introduction

The interaction of radiation with matter exhibits a vast variety of phenomena. For an exact and complete description of this interaction, both the radiation field and the atoms (or molecules) must be treated quantum-mechanically. However, it is not possible to solve this problem exactly. One therefore uses semiclassical treatment in which radiation field is treated classically and the atoms are treated quantum-mechanically. The semi-classical treatment is justified if the radiation field is sufficiently strong *i.e.* the number of photons present at any time during the interaction is large compared to unity. This condition is generally satisfied in most of the interaction processes. However it does not hold in many cases *e.g.* in spontaneous emission etc., and therefore such cases cannot be described by the semiclassical theory. Further, semiclassical theory cannot give information about the statistical properties of the radiation field(s) after the interaction.

However, it is not possible to solve the problem of atom-field interaction exactly even in the semi-classical framework. One therefore uses the perturbation theory. In it, one assumes that the initial state of the atom(s) is not much changed due to the interaction with the field. However, this fundamental assumption is valid only for off-resonant processes or for weak fields. It breaks down in case of resonant interactions or when the field is very strong. Most of the treatments of atom-field interaction are based on the perturbation theory. Further, in perturbation theory, one assumes the field strength to be unchanged during the interaction process. This assumption is certainly not correct when the incident field is strongly absorbed. Therefore one must be careful when using the perturbation theory, particularly in nonlinear optics.

In a previous paper (Sunil Kumar and Mehta 1980a), we have given the quantum theory of the interaction of a single-mode resonant radiation field with N two-level atoms. In it, both the field and the atoms were treated quantum-mechanically without using the perturbation theory. In a subsequent paper (Sunil Kumar and Mehta 1981), we determined the time evolution of the statistical properties of the above system. Since the same Hamiltonian also describes the trilinear scattering processes *e.g.* parametric amplification, frequency conversion (up and down), spontaneous and stimulated Raman scattering, Brillouin scattering, etc, our results were applicable to such processes as well.

In this paper, we give the quantum theory of coherent, degenerate two-photon absorption and emission. Here the word 'coherent' implies the atomic coherence *i.e.* we shall treat the atoms collectively. It may be noted that in the usual theory of two-photon absorption, one does not consider the mutual interaction of the atoms *via* the common radiation field. In our treatment, this atomic coherence is taken into account. Further, the word 'degenerate' means that both the photons taking part in two-photon absorption and emission are of the same frequency. We shall not consider the non-degenerate case. We shall assume that there is no one-photon resonant intermediate atomic state between the initial and final atomic levels. Thus we shall not consider the step-wise two-photon transitions. Further, we shall neglect the relaxation of the atoms. Therefore, our treatment will be valid for interaction times small compared to the characteristic relaxation times of the atoms. Finally, we shall be interested in the intensity and statistical properties of the field and not in its frequency spectrum.

We shall see that this problem is mathematically identical to the problem of degenerate four-wave mixing processes satisfying the condition $\omega_1 = \omega_2 + 2\omega_3$ where ω_i's are the frequencies of the waves. Therefore our results could also be applied to this case.

The outline of the paper is as follows: In § 2, the Hamiltonian describing the system is given which also describes the degenerate four-wave mixing processes as mentioned above. In § 3, the equations of motion in Heisenberg picture are derived and the procedure to solve them is given. The explicit analytical solutions are given in § 4. In § 5, we consider two specific cases of interest in detail:
(i) two-photon absorption of a coherent field by an atomic system in ground state,
(ii) stimulated two-photon emission by fully excited atoms in the presence of a coherent field.
In § 6, we compare our method with the treatments of other authors.

2. The Hamiltonian for the system

We first consider the interaction of a single-mode radiation field with a single atom.

Consider a multi-level atom placed with a single-mode radiation field in a box of volume V. In the absence of interaction, the Hamiltonian describing the system is (Loudon 1973)

$$\hat{H}_0 = \sum_i \hbar\,\omega_i |i\rangle\langle i| + \hbar\omega\,\hat{a}^\dagger\hat{a} \tag{1}$$

where $\hbar\omega_i$ is the energy of the ith atomic level, $\hat{a}$ and $\hat{a}^\dagger$ are the annihilation, creation operators of the field of frequency ω. Here we have omitted the zero-point energy of the field which is constant and does not contribute to the atom field interaction. The interaction Hamiltonian is (in electric dipole approximation)

$$\hat{H}_I = -\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}, \tag{2}$$

where the electric dipole operator $\hat{\mathbf{d}}$ and electric field operator $\hat{\mathbf{E}}$ are given by (in the Schrödinger picture)

$$\hat{\mathbf{d}} = \sum_{ij} \hat{\mathbf{d}}_{ij}|i\rangle\langle j| \tag{3}$$

$$\hat{\mathbf{E}} = \left(\frac{\hbar\omega}{2\varepsilon_0 V}\right)^{\frac{1}{2}}(\hat{a}+\hat{a}^{\dagger})\hat{x}, \tag{4}$$

where $\mathbf{d}_{ij}$ is the matrix element of electric dipole moment operator between the states $|i\rangle$ and $|j\rangle$, ε_0 is the permittivity of vacuum and we have assumed the field to be plane-polarized in the direction $\hat{x}$.
Therefore

$$\hat{H}_I = -\left(\frac{\hbar\omega}{2\varepsilon_0 V}\right)^{\frac{1}{2}}\sum_{ij} d_{ij}|i\rangle\langle j|(\hat{a}+\hat{a}^{\dagger}), \tag{5}$$

where

$$d_{ij} = \mathbf{d}_{ij}\cdot\hat{x}. \tag{6}$$

The total Hamiltonian describing the system is then

$$\hat{H} = \hat{H}_0 + \hat{H}_I, \tag{7}$$

with $\hat{H}_0$, $\hat{H}_I$ given by (1) and (5).

The interaction Hamiltonian $\hat{H}_I$ includes the complete set of atomic states. Although the initial and final states of the atom and field satisfy the law of conservation of energy, the intermediate states contributing to the two-photon (and in general multi-photon) absorption and emission need not be energy-conserving. We therefore make a canonical transformation on the total Hamiltonian $\hat{H}$ such that the transformed Hamiltonian $\hat{K}$ has non-zero matrix elements between the initial and final states only (Heitler 1954). This type of canonical transformation is also used by Takatsuji (1975). However the interaction Hamiltonian is explicitly time-dependent in the studies of Heitler (1954) where interaction picture is used, and in the studies of Takatsuji (1975) where semiclassical treatment is given. Therefore the canonical transformation is also time-dependent and its determination involves multiple time-integrals. To simplify the calculations, we shall work in the Schrödinger picture in which the Hamiltonian $\hat{H}$ is time-independent and the determination of canonical transformation is easy. The equation of motion in Schrödinger picture is

$$i\hbar\frac{\partial}{\partial t}|\psi\rangle = \hat{H}|\psi\rangle, \tag{8}$$

where $|\psi\rangle$ is the wavefunction of the atom-field system. We make a canonical transformation on $|\psi\rangle$. Let $|\psi'\rangle$ be the transformed wavefunction given by

$$|\psi'\rangle = \hat{S}|\psi\rangle, \tag{9}$$

where the time-independent transformation matrix $\hat{S}$ is a unitary matrix, *i.e.*

$$\hat{S}^{\dagger}\hat{S} = 1. \tag{10}$$

The equation of motion of $|\psi'\rangle$ is easily obtained to be

$$i\hbar\frac{\partial}{\partial t}|\psi'\rangle = \hat{K}|\psi'\rangle, \tag{11}$$

where the transformed Hamiltonian

$$\hat{K} = \hat{S}\hat{H}\hat{S}^{-1}. \tag{12}$$

For almost all light beams, the interaction Hamiltonian $\hat{H}_I$ is small compared to the unperturbed Hamiltonian $\hat{H}_0$ since the electric field applied on the electron is small compared to the field due to the nucleus. Therefore one can expand $\hat{K}$ and $\hat{S}$ in a rapidly covergent power series in the perturbation as follows:

$$\hat{K} = \hat{K}_0 + \hat{K}_1 + \hat{K}_2 + \dots, \tag{13}$$

$$\hat{S} = 1 + \hat{S}_1 + \hat{S}_2 + \dots, \tag{14}$$

Then $$\hat{S}^{-1} = 1 - \hat{S}_1 + (\hat{S}_1^2 - \hat{S}_2) + \dots \tag{15}$$

Substituting (13)–(15) in (12) and comparing terms of the same order, we obtain

$$\hat{K}_0 = \hat{H}_0, \tag{16}$$

$$\hat{K}_1 = [\hat{S}_1, \hat{H}_0] + \hat{H}_I, \tag{17}$$

$$\hat{K}_2 = [S_2, H_0] + \hat{S}_1 H_I - \hat{K}_1\hat{S}_1, \tag{18}$$

and so on. The physical significance of the various terms in the expansion (13) is as follows. The zeroth-order term $\hat{K}_0$ is identical to the unperturbed Hamiltonian $\hat{H}_0$ and therefore it describes the system in the absence of interaction. The next term $\hat{K}_1$ is of first-order in the perturbation $\hat{H}_I$ and therefore describes atom-field interaction *via* absorption and emission of one photon since $\hat{H}_I$ is linear in $\hat{a}$, $\hat{a}^{\dagger}$. The term $\hat{K}_2$ is of second-order in $\hat{H}_I$ and hence describes interaction *via* two-photon processes. Since the perturbation is small, the nth term in the expansion (13) will be important only when the terms of order less than n (except $\hat{K}_0$) vanish. Further, if nth term is non-zero, then all the higher terms can be omitted since they are smaller. Thus it follows that to describe nth-order multiphoton processes, it is sufficient to consider the Hamiltonian

$$\hat{K} = \hat{K}_0 + \hat{K}_n, \tag{19}$$

with $$\hat{K}_i = 0, \qquad 1 \leqslant i \leqslant n-1. \tag{20}$$

Sometimes multiphoton processes of different orders may occur simultaneously. Then one must retain all the relevant terms in the expansion (13).

In case of two-photon absorption and emission, the Hamiltonian is

$$\hat{K} = \hat{K}_0 + \hat{K}_2, \tag{21}$$

with $$\hat{K}_1 = 0. \tag{22}$$

As mentioned before, we choose the transformation matrix $\hat{S}$ such that the transformed Hamiltonian $\hat{K}$ has non-zero matrix elements between only those two states, say $|a\rangle$

and $|b\rangle$, of the atom-field system for which energy is conserved.

Then $\quad \langle\alpha|\hat{K}_2|\beta\rangle = 0, \text{ if } |\alpha\rangle, |\beta\rangle \notin \{|a\rangle, |b\rangle\}$. (23)

The non-zero matrix elements of $\hat{K}_2$ can easily be determined as follows. Taking the matrix element of (17) between the states $|\alpha\rangle$ and $|\beta\rangle$ and using (22), we obtain

$$\langle\alpha|\hat{S}_1|\beta\rangle = \frac{\langle\alpha|\hat{H}_I|\beta\rangle}{E_\beta - E_\alpha}, \tag{24}$$

where E_α, E_β are the energy of the atom-field system in the states $|\alpha\rangle$, $|\beta\rangle$ respectively. Note that for the energy conserving states $|a\rangle$ and $|b\rangle$

$$E_a = E_b. \tag{25}$$

We now take the matrix element of (18) between the states $|a\rangle$ and $|b\rangle$. Using (25), (24) and (22) we obtain

$$\begin{aligned}\langle\alpha|\hat{K}_2|\beta\rangle &= \langle\alpha|\hat{S}_1\hat{H}_I|\beta\rangle \\ &= \sum_\gamma \langle\alpha|\hat{S}_1|\gamma\rangle\langle\gamma|\hat{H}_I|\beta\rangle \\ &= \sum_\gamma \frac{\langle\alpha|\hat{H}_I|\gamma\rangle\langle\gamma|\hat{H}_I|\beta\rangle}{E_\gamma - E_\alpha}, \text{ if } |\alpha\rangle, |\beta\rangle \in \{|a\rangle, |b\rangle\}\end{aligned} \tag{26}$$

using the complete set $|\gamma\rangle$ of the atom-field system. Equations (23) and (26) completely determine $\hat{K}_2$. It is more useful to write the matrix elements of $\hat{K}_2$ explicitly in terms of the atomic and field parameters.
Let

$$\left.\begin{aligned}|a\rangle &= |1\rangle|\phi_1\rangle, \\ |b\rangle &= |2\rangle|\phi_2\rangle, \\ |\gamma\rangle &= |i\rangle|n\rangle,\end{aligned}\right\} \tag{27}$$

where $|\phi_1\rangle$ and $|\phi_2\rangle$ are the field states when the atom is in the upper state $|1\rangle$ and lower state $|2\rangle$ respectively. The states $|i\rangle$ and $|n\rangle$ form complete set for atom and field respectively i.e.

$$\left.\begin{aligned}\sum_i |i\rangle\langle i| &= 1 \\ \sum_n |n\rangle\langle n| &= 1.\end{aligned}\right\} \tag{28}$$

Then

$$\langle a|\hat{K}_2|a\rangle = \sum_\gamma \frac{\langle a|\hat{H}_I|\gamma\rangle\langle\gamma|\hat{H}_I|a\rangle}{E_\gamma - E_a}. \tag{29}$$

Since the interaction Hamiltonian $\hat{H}_I$ is linear in the annihilation and creation operators $\hat{a}$, $\hat{a}^\dagger$, in (29), only those intermediate states $|\gamma\rangle$ will contribute in which the number of photons is either one less or one more than in the initial state $|a\rangle$. Therefore

$$E_\gamma - E_a = E_i - E_1 \pm \hbar\omega. \tag{30}$$

Using (5), (27) and (30) in (29), we obtain

$$\langle a|\hat{K}_2|a\rangle = \frac{\hbar\omega}{2\varepsilon_0 V}\sum_{i,n}|d_{1i}|^2\left\{\frac{\langle\phi_1|\hat{a}|\hat{n}\rangle\langle n|\hat{a}^{\dagger}|\phi_1\rangle}{E_i - E_1 + \hbar\omega} + \frac{\langle\phi_1|\hat{a}^{\dagger}|n\rangle\langle n|\hat{a}|\phi_1\rangle}{E_i - E_1 - \hbar\omega}\right\}$$

$$= \frac{\hbar\omega}{2\varepsilon_0 V}\sum_{i}|d_{1i}|^2\left\{\frac{\langle\phi_1|\hat{a}\hat{a}^{\dagger}|\phi_1}{E_i - E_1 + \hbar\omega} + \frac{\langle\phi_1|\hat{a}^{\dagger}\hat{a}|\phi_1\rangle}{E_i - E_1 - \hbar\omega}\right\}$$

$$= \frac{\omega}{2\varepsilon_0 V}\sum_{i}|d_{1i}|^2\left\{\frac{n_1 + 1}{\omega_{i1} + \omega} + \frac{n_1}{\omega_{i1} - \omega}\right\}, \tag{31}$$

where $\hbar\omega_{ij} = E_i - E_j$ and n_1 is the average number of photons in the state $|\phi_1\rangle$. The matrix elements $\langle a|\hat{K}_2|a\rangle$ represents the transition from the state $|a\rangle$ to itself *via* the intermediate states $|\gamma\rangle$ *i.e.* the atom in the state $|1\rangle$ emits (absorbs) a photon to reach the intermediate state $|i\rangle$ and then absorbs (emits) it to return to the initial level $|1\rangle$. Similarly,

$$\langle b|\hat{K}_2|b\rangle = \frac{\omega}{2\varepsilon_0 V}\sum_{i}|d_{2i}|^2\left\{\frac{n_2 + 1}{\omega_{i2} + \omega} + \frac{n_2}{\omega_{i2} - \omega}\right\}, \tag{32}$$

where $n_2 = \langle\phi_2|\hat{a}^{\dagger}\hat{a}|\phi_2\rangle$ is the average number of photons in the state $|\phi_2\rangle$. These diagonal elements of $\hat{K}_2$ can be combined in the unperturbed Hamiltonian $\hat{H}_0$. The physical interpretation is that they are the energy shifts of the levels $|a\rangle$ and $|b\rangle$ of the atom-field system produced due to the virtual exchange of photons between atom and field. In the semiclassical limit ($n_1, n_2 \gg 1$), the energy shifts (31) and (32) reduce to those obtained by Takatsuji (1975). Equations (31) and (32) show that the energy shifts are not zero even in the absence of field (*i.e.* when $n_1 = 0$ or $n_2 = 0$). These vacuum energy shifts are due to the interaction of atom with vacuum fluctuations.

The off-diagonal matrix element $\langle a|\hat{K}_2|b\rangle$ represents the transition of the system from the state $|b\rangle$ to the state $|a\rangle$ *via* the intermediate states $|\gamma\rangle$. In other words, the atom in the lower state $|2\rangle$ absorbs a photon to reach the intermediate state $|i\rangle$ and then absorbs another photon to reach the upper state $|1\rangle$. Therefore, in (26) only those intermediate states $|\gamma\rangle$ will contribute which have one photon less than the initial state $|b\rangle$. Therefore,

$$\langle a|\hat{K}_2|b\rangle = \sum_{\gamma}\frac{\langle a|\hat{H}_I|\gamma\rangle\langle\gamma|\hat{H}_I|b\rangle}{E_\gamma - E_a}$$

$$= \frac{\hbar\omega}{2\varepsilon_0 V}\sum_{i,n}\frac{d_{1i}d_{i2}\langle\phi_1|\hat{a}|n\rangle\langle n|\hat{a}|\phi_2\rangle}{E_i - E_1 + \hbar\omega}$$

$$= \frac{\omega}{2\varepsilon_0 V}\sum_{i}\frac{d_{1i}d_{i2}\langle\phi_1|\hat{a}^2|\phi_2\rangle}{\omega_{i1} + \omega} \tag{33}$$

or

$$\langle 1|\hat{K}_2|2\rangle = \frac{\omega}{2\varepsilon_0 V}\sum_{i}\frac{d_{1i}d_{i2}}{\omega_{i1} + \omega}\hat{a}^2. \tag{34}$$

Similarly

$$\langle 2|\hat{K}_2|1\rangle = \frac{\omega}{2\varepsilon_0 V}\sum_{i}\frac{d_{2i}d_{i1}}{\omega_{i2} - \omega}\hat{a}^{\dagger 2}. \tag{35}$$

From (21), (16), (1), (23), (31), (32), (34) and (35), the total Hamiltonian $\hat{K}$ is given by

$$\hat{K} = \hbar\omega'_1|1\rangle\langle 1| + \hbar\,\omega'_2|2\rangle\langle 2| + \hbar\,\omega\,\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\hbar g\{|1\rangle\langle 2|\hat{a}^2 + |2\rangle\langle 1|\hat{a}^{\dagger 2}\}, \tag{36}$$

where the atomic frequencies ω'_1 and ω'_2 include the energy shifts (given by (31) and (32)) and

$$\tfrac{1}{2}\hbar g = \frac{\omega}{2\varepsilon_0 V}\sum_i \frac{d_{1i}\,d_{i2}}{\omega_{i1} + \omega} \tag{37}$$

gives the coupling parameter g. For brevity, we shall omit the primes in ω'_1 and ω'_2. If we we choose the zero-level of energy midway between the two atomic levels so that

$$\omega_1 = +\omega, \qquad \omega_2 = -\omega, \tag{38}$$

then (36) can be written as

$$\hat{K} = \hbar\omega\hat{\sigma}_z + \hbar\omega\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\hbar g(\hat{\sigma}_+\hat{a}^2 + \hat{\sigma}_-\hat{a}^{\dagger 2}) \tag{39}$$

where $\hat{\sigma}$'s are the Pauli spin matrices.

If there are N atoms interacting coherently with the field, then the Hamiltonian is given by

$$\begin{aligned}\hat{K} &= \hbar\omega\sum_i(\hat{\sigma}_z)_i + \hbar\omega\,\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\hbar g\sum_i\{(\hat{\sigma}_+)_i\,\hat{a}^2 \\ &\quad \exp(2i\mathbf{k}\cdot\mathbf{R}_i) + (\hat{\sigma}_-)_i\,\hat{a}^{\dagger 2}\exp(-2i\mathbf{k}\cdot\mathbf{R}_i)\} \\ &= 2\hbar\omega\,\hat{J}_z + \hbar\omega\hat{a}^\dagger\hat{a} + \hbar g(\hat{J}_+\hat{a}^2 + \hat{J}_-\hat{a}^{\dagger 2}),\end{aligned} \tag{40}$$

where $\hat{\sigma}_i$ are the spin matrices for the ith atom, $\mathbf{R}_i$ is the position (of the nucleus) of the ith atom and

$$\left.\begin{aligned}\hat{J}_z &= \tfrac{1}{2}\sum_i(\hat{\sigma}_z)_i \\ \hat{J}_\pm &= \tfrac{1}{2}\sum_i(\hat{\sigma}_\pm)_i\exp(\pm 2i\mathbf{k}\cdot\mathbf{R}_i)\end{aligned}\right\} \tag{41}$$

are the collective atomic operators. Equation (40) gives the required Hamiltonian for a resonant two-photon interaction between N atoms and a monochromatic field.

Using the Schwinger's representation of angular momentum operators in terms of boson operators (Sunil Kumar and Mehta 1980a)

$$\left.\begin{aligned}&\hat{J}_+ = \hat{a}_1^\dagger\hat{a}_2, \quad \hat{J}_- = \hat{a}_1\,\hat{a}_2^\dagger \\ &\hat{J}_z = \tfrac{1}{2}(\hat{a}_1^\dagger\hat{a}_1 - \hat{a}_2^\dagger\hat{a}_2)\end{aligned}\right\} \tag{42}$$

the Hamiltonian (40) can be written as

$$\hat{K} = \hbar\omega(\hat{a}_1^\dagger\hat{a}_1 - \hat{a}_2^\dagger\hat{a}_2) + \hbar\omega\,\hat{a}^\dagger\hat{a} + \hbar g(\hat{a}_1^\dagger\hat{a}_2\hat{a}^2 + \hat{a}_1\,\hat{a}_2^\dagger\hat{a}^{\dagger 2}). \tag{43}$$

We put it in a more symmetric form as follows:

$$\hat{H} = \sum_{i=1}^{3}\hbar\omega_i\hat{a}_i^\dagger\hat{a}_i + \hbar g(\hat{a}_1^\dagger\hat{a}_2\,\hat{a}_3^2 + \hat{a}_1\,\hat{a}_2^\dagger\hat{a}_3^{\dagger 2}), \tag{44}$$

where we have again used the more familiar symbol $\hat{H}$ for the Hamiltonian with the understanding that it now represents the transformed Hamiltonian, and have used the subscript 3 to denote the field parameters and

$$\omega_1 = -\omega_2 = \omega_3 = \omega. \tag{45}$$

As explained in (Sunil Kumar and Mehta 1980a) the operators $\hat{a}_1, \hat{a}_1^\dagger$ and $\hat{a}_2, \hat{a}_2^\dagger$ can be interpreted as the annihilation and creation operators for the upper and lower atomic levels $|1\rangle$ and $|2\rangle$ respectively. The Hamiltonian (44) also describes degenerate four-wave mixing processes satisfying the condition

$$\omega_1 - \omega_2 - 2\omega_3 = 0 \tag{46}$$

(note that this condition is satisfied in the special case (45)). A general theory of nonlinear optical processes (including degenerate four-wave mixing) is given by Hanna *et al* (1979). To make our results applicable to the degenerate four-wave mixing processes also, we shall consider the Hamiltonian (44) with the condition (46). To determine the time-evolution of the system described by the Hamiltonian (44), we shall work in the Heisenberg picture.

3. The Heisenberg equations of motion

The equations of motion of the annihilation operators $\hat{a}_i(t)$ of the three modes $(i = 1, 2, 3)$ are

$$\left.\begin{aligned} i\frac{d\hat{a}_1}{dt} &= \frac{1}{\hbar}[\hat{a}_1, \hat{H}] = \omega_1\hat{a}_1 + g\hat{a}_2\hat{a}_3^2 \\ i\frac{d\hat{a}_2}{dt} &= \frac{1}{\hbar}[\hat{a}_2, \hat{H}] = \omega_2\hat{a}_2 + g\hat{a}_1\hat{a}_3^{\dagger 2} \\ i\frac{d\hat{a}_3}{dt} &= \frac{1}{\hbar}[\hat{a}_3, \hat{H}] = \omega_3\hat{a}_3 + 2g\hat{a}_1\hat{a}_2^\dagger\hat{a}_3^\dagger. \end{aligned}\right\} \tag{47}$$

It is not possible to solve the coupled operator equations (47). We shall therefore use the same method as in (Sunil Kumar and Mehta 1980a) to solve the problem. The outline of this method is as follows: we first obtain the equations of motion of the number operators $\hat{N}_i(t) = \hat{a}_i^\dagger(t)\,\hat{a}_i(t), i = 1, 2, 3$ which can be decoupled. One of these operator equations is then converted into c-number equation assuming that the corresponding mode i is "strong". This c-number equation is easily integrated to give the mode population $N_i(t)$. One now replaces the annihilation and creation operators of the ith mode, a_i and $\hat{a}_i^\dagger$, by their classical analogs in the 3-mode Hamiltonian (44), which then reduces to a 2-mode Hamiltonian. The time-evolution of these two modes is rather easily determined. Since the three modes are correlated, the statistics of the strong mode i can be determined from the statistics of the other two modes. Finally the self-consistency of the method is checked by verifying whether the final results satisfy the conditions for the *ith* mode to be "strong". Here we note that if in the 3-mode Hamiltonian (44), either mode 1 or 2 is treated classically, the reduced 2-mode Hamiltonian contains $\hat{a}_3^2$ and $\hat{a}_3^{\dagger 2}$. It is not possible to solve the equations of motion with such a nonlinear Hamiltonian. Therefore, we shall consider only the case when mode 3 is "strong".

The equation of motion of the number operator $\hat{N}_3(t)$ is

$$i\hbar \frac{d\hat{N}_3}{dt} = [\hat{N}_3, \hat{H}]$$
$$= 2\hbar g[-\hat{a}_1^\dagger \hat{a}_2 \hat{a}_3^2 + \hat{a}_1 a_2^\dagger a_3^{\dagger 2}]. \tag{48}$$

Differentiating (48) and using (47), we obtain

$$\frac{d^2\hat{N}_3}{dt^2} = 4g^2[\hat{N}_1(\hat{N}_2+1)(4\hat{N}_3+2) + (\hat{N}_1 - \hat{N}_2)(\hat{N}_3^2 - \hat{N}_3)]$$
$$= -2g^2[4\hat{N}_3^3 - 3(2\hat{B} - 2\hat{A} - 1)\hat{N}_3^2 + 2\{\hat{B}(\hat{B} - 2\hat{A} - 2) + 1\}\hat{N}_3 + \hat{B}(\hat{B} - 2\hat{A} - 2)], \tag{49}$$

where

$$\hat{A} = \hat{N}_1 + \hat{N}_2, \; \hat{B} = 2\hat{N}_1 + \hat{N}_3 \tag{50}$$

are constants of motion since

$$[\hat{A}, \hat{H}] = 0 = [\hat{B}, \hat{H}], \tag{51}$$

or physically since the annihilation (creation) of a particle in mode 1 is accompanied with the creation (annihilation) of one particle in mode 2 and two particles in mode 3 (see the Hamiltonian (44)).

Taking average of (49) in the initial state of the system, and assuming

$$\left.\begin{aligned} &\frac{\langle \hat{N}_3^3 \rangle - \langle N_3 \rangle^3}{\langle \hat{N}_3 \rangle^3} \ll 1 \\ &\frac{\langle \hat{X}\hat{N}_3^2 \rangle - \langle \hat{X} \rangle \langle \hat{N}_3 \rangle^2}{\langle \hat{X} \rangle \langle \hat{N}_3 \rangle^2} \ll 1 \\ &\frac{\langle \hat{B}\hat{X}\hat{N}_3 \rangle - \langle \hat{B} \rangle \langle \hat{X} \rangle \langle \hat{N}_3 \rangle}{\langle \hat{B} \rangle \langle \hat{X} \rangle \langle \hat{N}_3 \rangle} \ll 1 \\ &B - 2A \gg 1, \end{aligned}\right\} \tag{52}$$

where $\hat{X} = \hat{A}$, $\hat{B}$ and 1, we obtain

$$\frac{d^2 N_3}{dt^2} = -2g^2[4N_3^3 - 3(2B - 2A - 1)N_3^2 + 2\{B(B - 2A - 2) + 1\}N_3 + B(B - 2A - 2)] \tag{53}$$

where $N_3 = \langle \hat{N}_3 \rangle$. Note that the four conditions (52) are necessary to factorize the averages of the corresponding four terms in (49).

Further, since the minimum possible value of N_3 is $(B - 2A)$ (see (50)), the last of the conditions (52) implies that

$$N_3(t) \gg 1, \text{ for all } t. \tag{54}$$

We shall call the mode 3 as "strong" mode if the conditions (52) are satisfied since the operator equation of motion (49) of mode 3 can then be treated as a c-number equation (53). We now determine the time-evolution of the system by following the procedure outlined above.

4. The time evolution of the system

We first solve (53) and determine $N_3(t)$. Multiplying (53) by $2\dfrac{\mathrm{d}N_3}{\mathrm{d}t}$ and integrating,

$$\left(\frac{\mathrm{d}N_3}{\mathrm{d}t}\right)^2 = -4g^2[N_3^4-(2B-2A-1)N_3^3+\{B(B-2A-2)+1\}N_3^2 + B(B-2A-2)N_3]+C, \tag{55}$$

where C is the constant of integration. Its value can be determined from the initial conditions by using (48) and (55). For simplicity, we assume that initially at $t = 0$, at least one of the three modes is in a number state so that from (48),

$$(\mathrm{d}N_3/\mathrm{d}t)_{t=0} = 0. \tag{56}$$

Also, let $N_3\,(t = 0) = N_{30}$. (57)

Using the initial conditions (56) and (57) in (55), the value of C can be determined. Then (55) becomes

$$\begin{aligned}\left(\frac{\mathrm{d}N_3}{\mathrm{d}t}\right)^2 &= -4g^2[(N_3^4-N_{30}^4)-(2B-2A-1)(N_3^3-N_{30}^3)\\ &\quad+\{B(B-2A-2)+1\}(N_3^2-N_{30}^2)+B(B-2A-2)(N_3-N_{30})]\\ &= -4g^2(N_3-N_{30})(N_3-\alpha)(N_3-\beta)(N_3-\gamma),\end{aligned} \tag{58}$$

where α, β, γ are the roots of the cubic

$$f(N_3) = N_3^3+aN_3^2+bN_3+c \tag{59}$$

where

$$a = N_{30}-(2B-2A-1),\ b = aN_{30}+B(B-2A-2)+1,$$

and $$c = bN_{30}+B(B-2A-2). \tag{60}$$

Equations (60) can also be put in the form

$$\left.\begin{aligned} a &= -(B-N_{30})-(B-2A-1),\\ b &= -(B-N_{30})\{N_{30}-(B-2A)+1\}-(B-1),\\ c &= -(B-N_{30})\{N_{30}-(B-2A)+1\}\,N_{30}-B\{N_{30}-(B-2A)+1\}\\ &\quad -(B-N_{30}),\end{aligned}\right\} \tag{61}$$

which shows that

$$a, b, c < 0, \tag{62}$$

since $B \geqslant N_{30} \geqslant B-2A$
and $B \gg 1,\ B-2A \gg 1$ (63)

from (50) and (52).

The solution of (58) is in terms of the elliptic functions. The exact form of the solution depends on whether α, β, γ are real or complex and if real, whether greater or less than N_{30}. Therefore, we shall first determine the nature of the roots α, β, γ.

We note that in the cubic expression (59), there is only one change of sign in the coefficients (see (62)). Therefore from the theory of algebric equations, at the most one root, say α is positive. Further, it is easily verified that

$$B+1 > \alpha > 2B-2A-N_{30}, \tag{64}$$

since

$$\begin{aligned} f(N_3 = B+1) &= N_{30}^3 - (B-2A-2)N_{30}^2 - (B-2A-2)N_{30} + 2A + 3 \\ &= N_{30}(N_{30}+1)\{N_{30}-(B-2A)\} + N_{30}^2 + 3N_{30} \\ &\quad + 2A + 3 \\ &> 0, \end{aligned} \tag{65}$$

and

$$\begin{aligned} f(N_3 = 2B-2A-N_{30}) &= N_{30}^2(2B-2A+1) - N_{30}(2B-2A)(2B-2A+1) \\ &\quad + \{B^2 - 2A(B+1)\}(2B-2A+1) \\ &= -(2B-2A+1)(B-N_{30})\{N_{30}-(B-2A)\} - 2A(2B-2A+1) < 0, \end{aligned} \tag{66}$$

using (63).

The other two roots β and γ are either both negative or complex conjugates of each other. If they are both negative, then

$$(\beta-\gamma)^2 > 0, \tag{67}$$

and if they are complex conjugates, then

$$(\beta-\gamma)^2 = (2i \operatorname{Im} \beta)^2 < 0. \tag{68}$$

We show that inequality (68) holds for all values of N_{30}. To prove it, we note that (see (59) and (62))

$$\left.\begin{aligned} \alpha+\beta+\gamma &= -a > 0 \\ \alpha\beta+\beta\gamma+\gamma\alpha &= b < 0 \\ \alpha\beta\gamma &= -c > 0 \end{aligned}\right\} \tag{69}$$

Since $\alpha > (2B-2A-N_{30}) > 0$ (see (64)), therefore $(\beta+\gamma)$ is always negative and $\beta\gamma$ is always positive (see (69) and (62)). Further from (69), as α increases, $(\beta+\gamma)$ decreases *i.e.* $|\beta+\gamma|$ increases and $\beta\gamma$ decreases, so that $(\beta-\gamma)^2 = (\beta+\gamma)^2 - 4\beta\gamma$ increases. Hence if the inequality (68) holds for the upper limit of α, then it will hold for all lower values of α. For the upper limit of α *i.e.* for $\alpha = B+1$ (see (64)), it is easily seen that

$$(\beta-\gamma)^2 = -(3N_{30}+B-2A)(N_{30}-(B-2A)) - 4N_{30} - 8 < 0. \tag{70}$$

Hence β, γ are complex conjugates *i.e.* $\gamma = \beta^*$.

Therefore, we can write (58) as

$$\left(\frac{dN_3}{dt}\right) = -4g^2(N_3-N_{30})(N_3-\alpha)(N_3-\beta)(N_3-\beta^*) \tag{71}$$

or

$$2gt = \int_{N_{30}}^{N_3} \frac{dN_3}{\{-(N_3-N_{30})(N_3-\alpha)(N_3-\beta)(N_3-\beta^*)\}^{1/2}} \tag{72}$$

To evaluate the above integral, we note that from (71), $dN_3/dt = 0$ for $N_3 = N_{30}$ and α. Hence N_{30}, α are the extremum values of N_3. For $\alpha \geqslant N_3(t) \geqslant N_{30}$, (72) gives (Byrd and Friedman, 1971, p. 133)

$$N_3(t) = \frac{N_{30}P\{1+cn(\tau,k)\} + \alpha Q\{1-cn(\tau,k)\}}{P\{1+cn(\tau,k)\} + Q\{1-cn(\tau,k)\}}, \tag{73}$$

where

$$\left.\begin{aligned} P &= \{(\alpha-\beta)(\alpha-\beta^*)\}^{1/2} \\ Q &= \{(N_{30}-\beta)(N_{30}-\beta^*)\}^{1/2} \\ \tau &= 2(PQ)^{1/2}\, gt \\ k^2 &= \{(\alpha-N_{30})^2-(P-Q)^2\}/(4PQ). \end{aligned}\right\} \tag{74}$$

For $N_{30} \geqslant N_3(t) \geqslant \alpha$, we write

$$\int_{N_{30}}^{N_3} = \int_{\alpha}^{N_3} - \int_{\alpha}^{N_{30}} \tag{75}$$

and evaluate each integral as before. The result obtained is again (73). Thus $N_3(t)$ is given by (73) for $\alpha > N_{30}$ and $\alpha < N_{30}$. In the limiting case $\alpha = N_{30}$, (73) reduces to

$$N_3(t) = N_{30}. \tag{76}$$

which shows that population of the mode 3 remains constant in time. However, as we shall see later, the results obtained in this case are not self-consistent. Hence, we shall exclude this case. We now determine the time-evolution of the modes 1 and 2. Since $N_3(t) \gg 1$, for all t (see (54)), we can replace the operators $\hat{a}_3, \hat{a}_3^\dagger$ in the Hamiltonian (44) by their classical analogs:

$$\begin{aligned} \hat{a}_3(t) &\to a_3(t) = \{N_3(t)\}^{1/2}\exp[-i\{\omega_3 t+\phi_3(t)\}] \\ \hat{a}_3^\dagger(t) &\to a_3^*(t) = \{N_3(t)\}^{1/2}\exp[i\{\omega_3 t+\phi_3(t)\}] \end{aligned} \tag{77}$$

where $\phi_3(t)$ is expected to be a slowly varying function of t, and can be determined as follows. From the last of equations (47).

$$\hat{a}_3^\dagger \frac{\mathrm{d}\hat{a}_3}{\mathrm{d}t} - \frac{\mathrm{d}\hat{a}_3^\dagger}{\mathrm{d}t}\hat{a}_3 = -2i\omega_3\hat{N}_3 - \frac{2i}{\hbar}\hat{H}_I, \tag{78}$$

where

$$\hat{H}_I = \hbar g(\hat{a}_1\hat{a}_2^\dagger\hat{a}_3^{\dagger 2} + \hat{a}_1^\dagger\hat{a}_2\hat{a}_3^2), \tag{79}$$

is the interaction Hamiltonian and is a constant of motion since

$$[\hat{H}_I, \hat{H}] = 0. \tag{80}$$

The classical analog of (78) is obtained by using (77):

$$a_3^* \frac{\mathrm{d}a_3}{\mathrm{d}t} - \frac{\mathrm{d}a_3^*}{\mathrm{d}t}a_3 = -2i\,N_3(t)(\omega_3+\dot{\phi}_3). \tag{81}$$

Comparing (78) and (81),

$$N_3(t)\dot{\phi}_3(t) = \frac{1}{\hbar}\langle\hat{H}_I\rangle. \tag{82}$$

Since $N_3(t)$ is known and $\langle\hat{H}_I\rangle$ is constant, $\phi_3(t)$ can be determined.

We now use (77) in the Hamiltonian (44) to obtain

$$\begin{aligned} \hat{H} \to \hat{H}_3 = {} & \hbar\omega_1\hat{a}_1^\dagger\hat{a}_1 + \hbar\omega_2\hat{a}_2^\dagger\hat{a}_2 + \hbar\omega_3 N_3(t) \\ & + \hbar g\, N_3(t)\{\hat{a}_1^\dagger\,\hat{a}_2\exp[-2i(\omega_3 t+\phi_3)] \\ & + \hat{a}_1\,\hat{a}_2^\dagger\exp[2i(\omega_3 t+\phi_3)]\}. \end{aligned} \tag{83}$$

The equations of motion corresponding to the Hamiltonian (83) can exactly be solved if the condition

$$\dot{\phi}_3(t)/N_3(t) = \text{constant} \tag{84}$$

is satisfied (Lu 1973; Sunil Kumar and Mehta 1980b). Equations (82) and (84) can be consistent only if

$$\langle \hat{H}_I \rangle = 0. \tag{85}$$

Since at least one of the modes is assumed to be in a number state (see (56)), the condition (85) is satisfied. Then from (82),

$$\phi_3(t) = 0 \tag{86}$$

since the constant of integration can be set to be zero without any loss in generality. Then

$$\begin{aligned}\hat{H}_3 &= \hbar\omega_1 \hat{a}_1^\dagger a_1 + \hbar\omega_2 \hat{a}_2^\dagger \hat{a}_2 + \hbar\omega_3 N_3(t) \\ &\quad + \hbar g N_3(t)\{\hat{a}_1^\dagger a_2 \exp(-2i\omega_3 t) + \hat{a}_1 \hat{a}_2^\dagger \exp(2i\omega_3 t)\}.\end{aligned} \tag{87}$$

The equations of motion of the operators $\hat{a}_1(t)$ and $\hat{a}_2(t)$ for the Hamiltonian (87) can easily be solved to give (Sunil Kumar and Mehta 1980a)

$$\begin{aligned}\hat{a}_1(t) &= \exp(-i\omega_1 t)\{\hat{a}_1(0)\cos\theta_3(t) - i\hat{a}_2(0)\sin\theta_3(t)\} \\ \hat{a}_2(t) &= \exp(-i\omega_2 t)\{\hat{a}_2(0)\cos\theta_3(t) - i\hat{a}_1(0)\sin\theta_3(t)\}\end{aligned} \tag{88}$$

where

$$\theta_3(t) = g\int_0^t N_3(t')\,\mathrm{d}t' \tag{89}$$

The integral of (89) can easily be found (Byrd and Friedman 1971, p. 216) to give

$$\begin{aligned}\theta_3(t) &= \frac{(PN_{30} - Q\alpha)\tau}{2(PQ)^{1/2}(P-Q)} + \frac{(\alpha - N_{30})(P+Q)}{4(PQ)^{1/2}(P-Q)}\pi\left(\phi, -\frac{(P-Q)^2}{4PQ}, k\right) \\ &\quad - \tfrac{1}{2}\tan^{-1}\left(\frac{\alpha - N_{30}}{2(PQ)^{1/2}}\, sd\,(\tau, k)\right),\end{aligned} \tag{90}$$

where p, Q, τ, k are given by (74),

$$\cos\phi = cn\,(\tau, k) \tag{91}$$

and $\pi\,(\phi, \alpha^2, k)$ is the elliptic integral of the third kind given by (Byrd and Friedman 1971, p. 223)

$$\pi(\phi, \alpha^2, k) = \int_0^\phi \frac{\mathrm{d}\theta}{(1-\alpha^2\sin^2\theta)(1-k^2\sin^2\theta)^{1/2}}. \tag{92}$$

Here α in the argument of $\pi\,(\phi, \alpha^2, k)$ should not be confused with the root α of (59). From (88), we note that for $N_3(t) = N_{30}, N_1(t)$ is a function of time which is not consistent with the requirement that $B = 2N_1 + N_3$ must be a constant of motion (see (50)). Therefore we exclude this case. It is seen from (88) that all the dynamical and statistical properties of the modes 1 and 2 are exchanged in time since

$$\hat{a}_2(t+T) \equiv \hat{a}_1(t) \tag{93}$$

except for an insignificant change in phase, where T is the quarter period of oscillations and is given by

$$\theta_3(t+T) = \theta_3(t) + \frac{\pi}{2}. \tag{94}$$

The statistical properties of the modes 1 and 2 (*e.g.* normal-ordered moments, the density matrix etc) at any time t can easily be found for their various initial states (*e.g.* chaotic, coherent, mixture of chaotic and coherent, number states) (Sunil Kumar and Mehta 1981). The statistical properties of the "strong" mode 3 can then be determined from those of the modes 1 and 2 by using the relations (50).

5. Two specific examples

We now consider two specific cases of interest as examples: (i) two-photon absorption by atoms in ground state, and (ii) stimulated two-photon emission by fully excited atoms. In both cases, the initial field is assumed to be a coherent field.

5.1 *Two-photon absorption by atoms in ground state*

In this case, the modes 1 and 2 (corresponding to the excited and ground state atoms respectively) are in the number states $|0\rangle$ and $|N_{20}\rangle$ respectively. The mode 3 corresponding to the field is in a coherent state $|z\rangle$. Therefore,

$$N_{10} = 0, \quad N_{20} = A, \quad N_{30} = |z|^2 = B, \tag{95}$$

using the relations (50). Also from (48),

$$(\mathrm{d}N_3/\mathrm{d}t)_{t=0} = 0. \tag{96}$$

We first determine the roots α, β of the function (see (59) and (61))

$$f(N_3) = N_3^3 - (B-2A-1)N_3^2 - (B-1)N_3 - B(2A+1). \tag{97}$$

We note that

$$f(B-2A) = -4A(B-A) - 2A < 0, \tag{98}$$

$$f(B-2A+1) = 2(B-2A)^2 - B^2 + 3B - 10A + 3 > 0,$$
$$\text{if } B > 2(2+\sqrt{2})A \approx 7A. \tag{99}$$

Therefore,

$$(B-2A) < \alpha < (B-2A+1), \text{ if } B > 7A. \tag{100}$$

Since $B - 2A \gg 1$ (see (52)), therefore we can put

$$\alpha \approx B - 2A, \text{ if } B > 7A. \tag{101}$$

For $B > 7A$, α differs from $(B-2A)$ by a number of the order of unity. Further, from (69), (97) and (101),

$$\left.\begin{aligned}\beta+\beta^* &= 2\,\mathrm{Re}(\beta) \approx -1\\ \beta\beta^* &= |\beta|^2 \approx \frac{2AB}{B-2A}\end{aligned}\right\} \tag{102}$$

which determine β. It is easily seen that

$$2A < |\beta|^2 < 3A < \alpha \tag{103}$$

since $B > 7A$. The mode population $N_3(t)$ is then given by (73) with (see (74))

$$\left.\begin{aligned}&P \approx B-2A\\ &Q \approx B\\ &\tau \approx 2\{B(B-2A)\}^{1/2}\,gt\\ &k^2 \ll 1\end{aligned}\right\} \tag{104}$$

Since

$$cn(\tau, k) \approx \cos(\tau), \text{ if } k \ll 1 \tag{105}$$

therefore (73) gives

$$N_3(t) = \frac{B(B-2A)}{(B-A)-A\cos\tau}. \tag{106}$$

Equation (106) shows that the population of mode 3 oscillates periodically between B and $(B-2A)$.

The time-evolution of the modes 1 and 2 is given by (88) with $\theta_3(t)$ given by (see (89) and (106))

$$\tan\,\theta_3(t) = \left(\frac{B}{B-2A}\right)^{1/2}\tan\left(\frac{\tau}{2}\right). \tag{107}$$

In particular, the populations of the modes 1 and 2 are given by

$$\left.\begin{aligned}N_1(t) &= A\sin^2\theta_3(t)\\ N_2(t) &= A\cos^2\theta_3(t)\end{aligned}\right\}. \tag{108}$$

It is easy to verify that (108) and (106) satisfy the energy conservation relations (50) which proves the self-consistency of the results. We finally show that our results satisfy the conditions (52) for the mode 3 to be "strong". The various averages in (52) can be determined by using the relation

$$\hat{N}_3(t) = \hat{B} - 2\hat{N}_1(t) \tag{109}$$

and (88). The final results are

$$\frac{\langle \hat{B}\hat{N}_3\rangle - BN_3}{BN_3} = \frac{1}{N_3} \ll 1, \tag{110a}$$

$$\frac{\langle \hat{B}\hat{A}\hat{N}_3\rangle - BAN_3}{BAN_3} = \frac{1}{N_3} \ll 1, \tag{110b}$$

$$\frac{\langle \hat{B}^2\hat{N}_3\rangle - B^2N_3}{B^2N_3} = \frac{2B+N_3+1}{BN_3} \ll 1, \tag{110c}$$

$$\frac{\langle \hat{N}_3^2 \rangle - N_3^2}{N_3^2} = \frac{B + A\sin^2(2\theta_3)}{N_3^2} \ll 1, \tag{110d}$$

$$\frac{\langle \hat{B}\hat{N}_3^2 \rangle - BN_3^2}{BN_3^2} = \frac{B + N_3 + A\sin^2(2\theta_3) + 1}{N_3^2} \ll 1, \tag{110e}$$

$$\frac{\langle \hat{N}_3^3 \rangle - N_3^3}{N_3^3} = \frac{3\{B + A\sin^2(2\theta_3)\}}{N_3^2} + \frac{B}{N_3^2} \ll 1, \tag{110f}$$

since $B \geqslant N_3(t) \geqslant (B-2A) \gg 1$, and $B > 7A$. Hence the conditions (52) are satisfied. Here we remark that if the initial field is chaotic, then the conditions (52) cannot be satisfied (for example

$$\frac{\langle \hat{N}_3^2 \rangle - N_3^2}{N_3^2} = \frac{B^2 + B + A\sin^2(2\theta_3)}{N_3^2} > 1).$$

5.2 *Stimulated two-photon emission by fully excited atoms*

In this case, the modes 1 and 2 are in the number states $|N_{10}\rangle$ and $|0\rangle$ respectively and the mode 3 is assumed to be in a coherent state $|z\rangle$ so that

$$N_{10} = A, \quad N_{20} = 0, \quad N_{30} = |z|^2 = B - 2A, \tag{111}$$

and

$$(\mathrm{d}N_3/\mathrm{d}t)_{t=0} = 0. \tag{112}$$

Therefore, from (59) and (61),

$$f(N_3) = N_3^3 - (B-1)N_3^2 - (B+2A-1)N_3 - 2A(B-2A+1) - B. \tag{113}$$

To determine its real root α, we note that

$$f(B) = -2B(2B-2A+1) < 0, \tag{114}$$

$$f(B+1) = (B-2A)^2 + 3(B-2A) + 2A + 3 > 0, \tag{115}$$

so that

$$B < \alpha < B+1. \tag{116}$$

Since α is the maximum value of $N_3(t)$ (see (71)), it cannot exceed B (see (50)). Therefore we put

$$\alpha = B. \tag{117}$$

The small error in the value of α is due to the approximations (52). To determine the complex roots β, β^*, we note that from (69), (113) and (117),

$$\left.\begin{aligned} \beta + \beta^* &= 2\,\mathrm{Re}(\beta) \approx -1 \\ \beta\beta^* &= |\beta|^2 \approx \frac{2A(B-2A)}{B} \end{aligned}\right\}. \tag{118}$$

Therefore, from (74),

$$\left.\begin{aligned} &P \approx B, \quad Q \approx B - 2A \\ &\tau \approx 2\{B(B-2A)\}^{1/2} gt, \quad k^2 \ll 1. \end{aligned}\right. \tag{119}$$

The mode population $N_3(t)$ is then obtained from (73) (using (105)),

$$N_3(t) = \frac{B(B-2A)}{(B-A)+A\cos\tau}. \tag{120}$$

The time-evolution of the modes 1 and 2 is given by (88) with $\theta_3(t)$ given by (see (89) and (120))

$$\tan\theta_3(t) = \left\{\frac{B-2A}{B}\right\}^{\frac{1}{2}} \tan(\tau/2). \tag{121}$$

In particular, the mode populations are

$$\left.\begin{aligned} N_1(t) &= A\cos^2\theta_3(t) \\ N_2(t) &= A\sin^2\theta_3(t) \end{aligned}\right\}. \tag{122}$$

It is seen that the results (120) and (122) are consistent with (50). Also, using (109), we note that

$$\frac{\langle \hat{B}\hat{N}_3\rangle - BN_3}{BN_3} = \frac{N_{30}}{BN_3} \ll 1 \tag{123a}$$

$$\frac{\langle \hat{A}\hat{B}\hat{N}_3\rangle - ABN_3}{ABN_3} = \frac{N_{30}}{BN_3} \ll 1 \tag{123b}$$

$$\frac{\langle \hat{B}^2\hat{N}_3\rangle - B^2N_3}{B^2N_3} = \frac{N_{30}(2B+N_3+1)}{B^2N_3} \ll 1, \tag{123c}$$

$$\frac{\langle \hat{N}_3^2\rangle - N_3^2}{N_3^2} = \frac{N_{30}+A\sin^2(2\theta_3)}{N_3^2} \ll 1, \tag{123d}$$

$$\frac{\langle \hat{B}\hat{N}_3^2\rangle - BN_3^2}{BN_3^2} = \frac{N_{30}(B+2N_3+1)+AB\sin^2(2\theta_3)}{BN_3^2} \ll 1, \tag{123e}$$

$$\frac{\langle \hat{N}_3^3\rangle - N_3^3}{N_3^3} = \frac{1}{N_3^3}\{3N_3(N_{30}+A\sin^2(2\theta_3))+N_{30}\} \ll 1, \tag{123f}$$

which shows that the conditions (52) for the mode 3 to be "strong", are satisfied. We finally note that (see (123))

$$\Delta N_3^2 = \langle \hat{N}_3^2\rangle - N_3^2 = N_{30}+A\sin^2(2\theta_3),$$

so that

$$\Delta N_3^2 - N_3 = 2A\sin^2\theta_3(t)\{2\cos^2\theta_3(t)-1\}. \tag{124}$$

It shows that

$$\Delta N_3^2 - N_3 < 0, \tag{125}$$

if

$$\cos^2\theta_3(t) < \tfrac{1}{2} \quad \text{i.e. if } N_1(t) < \frac{A}{2}. \tag{126}$$

Then the degree of second order coherence (for zero time delay)

$$g^{(2)} = \frac{\langle \hat{a}_3^\dagger \hat{a}_3^\dagger \hat{a}_3 \hat{a}_3\rangle}{\langle \hat{a}_3^\dagger \hat{a}_3\rangle^2} < 1. \tag{127}$$

Hence the field generated by stimulated two-photon emission from fully excited atoms will show photon-antibunching (Sunil Kumar *et al* 1981), when at least half of the atoms are deexcited. We remark again that if the initial field is chaotic, then the conditions (52) cannot be satisfied.

6. Comparison with other treatments

We shall now compare our method with the treatments of other authors. All these treatments (Bloembergen and Levenson 1976; Letokhov and Chebotaev 1977; Narducci *et al* 1977; Grynberg *et al* 1980; Loudon 1973, pp. 298–307) invariably use the perturbation theory in which the change in the unperturbed initial state of the system is assumed to be small. The perturbation theory is valid for off-resonant transitions or for weak fields (so that the damping in energy units is larger than the magnitude of the interaction Hamiltonian) (Bloembergen and Levenson 1976, p. 321). Hence such treatments are not applicable to resonant two-photon transitions by strong fields. Further, in these treatments, atomic coherence is not taken into account, which may be important in dense media. Also, except in the studies by Loudon (1973), the treatment is semiclassical so that one cannot determine the change in the statistical properties of the field due to interaction. The fully quantum-mechanical perturbation treatment of (Loudon 1973) is not applicable to the experiments (Krasinski 1978) in which the incident field is strongly absorbed. On the other hand, our treatment is applicable to the resonant two-photon transitions by fields of arbitrary strength. It takes into account the atomic coherence. It can give the time-evolution of the field statistics. We finally mention that Simaan and Loudon (1975) have considered the quantum statistics of two-photon absorption using single beam and double beams of light. However they have used the rate equation approach which is not justified in quantum statistical treatments. McNeil and Walls (1974) have given a master equation treatment of the problem. However their results are exponential in time which is unphysical, for long time.

7. Conclusions

In this paper, we have given the theory of coherent, two-photon resonant interaction of a monochromatic field with an atomic system. We have first derived the Hamiltonian describing such an interaction from the usual dipole Hamiltonian, by using a time-independent canonical transformation. It is seen that the equations of motion can be solved if the field is "strong" i.e. if the conditions (52) are satisfied. The explicit solutions are then obtained for this case. To illustrate the method, we have considered two specific examples: (i) two-photon absorption by atoms in ground state, and (ii) stimulated two-photon emission by fully excited atoms, assuming a coherent field in both cases. We have found that in the second case, the field shows photon-antibunching when at least half of the atoms have emitted. Finally, we have compared our approach with other treatments. The other treatments, based on the perturbation theory, do not take into account the atomic coherence and generally do not give information about the change in field statistics. These shortcomings have been overcome in our treatment. We have also mentioned that since the same Hamiltonian (44) also describes degenerate four-wave mixing processes satisfying the condition (46), our results are applicable to such processes as well.

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