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Quantum theory of nonlinear optical phenomena

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Abstract. A treatment of nonlinear optical phenomena beginning with a microscopically correct Hamiltonian containing the Bose operators of the light field and the Fermi operators for the optically active electrons in the medium is presented. Using techniques recently developed, a nonperturbative solution to the Schrödinger equation is possible. No linearization or semiclassical approximations are introduced. Two photon emission and absorption including both the degenerate and nondegenerate cases are considered. Parametric amplification and frequency conversion are investigated as examples of multiphoton processes. Atomic cooperation is shown to enhance the above phenomena. A formal treatment of a model illustrating super and subradiance is included.

1. Introduction

Nonlinear optical effects arise as a result of the nonlinear response of a medium to intense light fields. A successful method of analysis developed by Bloembergen (1965) (Armstrong *et al.* 1962) expands the polarization in powers of the electric field, the proportionality constant being a nonlinear susceptibility tensor for the medium. Treating the electromagnetic field classically one obtains a set of coupled differential equations for the amplitudes of the interacting waves. The nonlinear susceptibility of the medium is taken into account only phenomenologically as a coupling constant. Quantum mechanical calculations of the tensor susceptibility have been made independently (Armstrong *et al.* 1962, Bloembergen 1965, Butcher 1965). A deficiency of the classical treatment of the electromagnetic field is the omission of the contribution of spontaneous emission to the stimulated processes. The first quantum mechanical model for nonlinear optical effects was proposed by Louisell *et al.* (1961). They set up a phenomenological Hamiltonian for the interacting quantized field modes in a parametric amplifier. The nonlinear susceptibility tensor of the medium was included as a coupling constant. The pump field was treated as a constant classical variable in order to linearize the Heisenberg equations of motion for the mode operators. Recently methods have been developed capable of solving the nonlinear quantum problem for special cases (Walls and Barakat 1970).

A microscopically correct Hamiltonian would contain the Bose operators of the light modes and the Fermi operators of the optically active electrons in the medium. Such an approach was adopted by Graham and Haken (1968) in studying the optical parametric oscillator. They derived quantum mechanical Langevin equations for the field and electron operators which contain dissipation and fluctuation terms. The electron operators were then eliminated by an iteration procedure which leaves a set of coupled nonlinear field equations. These were solved by linearization below threshold and by quantum mechanical quasilinearization above threshold. A similar Hamiltonian was employed by Shen (1967) in describing multiphoton emission and absorption. Shen derived an equation of motion for the density operator of the total system, then traced out over the atomic variables leaving an equation of motion for the density operator of the field alone.

In this treatise we shall present a general analysis of nonlinear optical phenomena starting from such a microscopically correct Hamiltonian. Recently techniques have been developed which enable a nonperturbative solution to the Schrödinger equation with the Hamiltonian containing both field and electron operators to be found. This technique was developed to analyse the interaction of a single mode of the electromagnetic field with N two level atoms (Jaynes and Cummings 1963, Tavis and Cummings 1968, Mallory 1969, Walls and Barakat 1970, Scharf 1970). It has since found applications in studies of the Raman effect (Walls 1970, 1971a) and double resonance phenomena (Walls 1971b). No linearization or semiclassical approximations are introduced. Relaxation effects are however neglected in our analysis.

In § 2 we present a general Hamiltonian formulation of the interaction between a quantized electromagnetic field and an ensemble of N f -level atoms. The general method of solution for this type of problem is outlined in § 3. The following sections are concerned with specific examples of phenomena described by the general Hamiltonian.

In § 4 and § 5 we consider two photon emission and absorption. Here we distinguish between the degenerate case involving only two atomic levels and the nondegenerate case which involves three atomic levels. The absorption and emission processes are shown to be enhanced by the presence of atomic cooperation.

In § 6 and § 7 parametric amplification and frequency conversion are studied as examples of multiphoton processes involving the absorption and subsequent re-emission of photons with different frequencies (Winter 1969). Such processes benefit greatly from atomic cooperation since both the absorption and subsequent re-emission processes are enhanced. The growth of the emitted photons is shown to be proportional to the fourth power of the number of cooperating atoms.

The concept of cooperation in spontaneous emission processes (superradiance) was first introduced by Dicke (1954) who presented a perturbative quantum treatment. In § 8 as an illustrative model we consider two 2-level atoms interacting with a single mode of the radiation field with one quantum of energy present. An exact solution is found for the time dependent wavefunction of this system. The solution is shown to contain the cases of super and subradiance.

Another important nonlinear phenomenon, namely the Raman effect is described by the general Hamiltonian. However, since this effect has been treated extensively in two previous publications Walls (1970), Walls (1971a) it has been omitted here.

2. Hamiltonian formulation of the nonlinear interaction

The electric field operator for the free field may be expanded in terms of normal modes as (Glauber 1963)

$$E(\mathbf{r}, t) = i \sum_k (\frac{1}{2}\hbar\omega_k)^{1/2} (a_k(t)u_k(\mathbf{r}) - a_k^\dagger(t)u_k^*(\mathbf{r})) \quad (2.1)$$

where $a_k(t)$ and $a_k^\dagger(t)$ are the boson annihilation and creation operators for the k th mode. The effects of the linear polarizability in a nondissipative isotropic medium may be accounted for by including a dielectric constant $\epsilon = 1 + 4\pi\chi$ where χ is the linear susceptibility of the medium (Shen 1967). The mode functions are then taken to satisfy the wave equation

$$\left(\nabla^2 + \frac{\omega_k^2 \epsilon_k}{c^2}\right) u_k(\mathbf{r}) = 0 \quad (2.2)$$

with the orthonormality condition

$$\int (\epsilon_k \epsilon_l)^{1/2} u_k^*(\mathbf{r}) u_l(\mathbf{r}) d^3r = \delta_{kl}. \quad (2.3)$$

The total Hamiltonian describing the interaction of the electromagnetic field with a nonlinear medium may be expressed in the following form:

$$H = H_0 + H_1 \quad (2.4)$$

where H_0 the Hamiltonian for the noninteracting system is

$$H_0 = \sum \hbar \omega_k a_k^\dagger a_k + H_{\text{medium}}. \quad (2.5)$$

We consider the medium to consist of an ensemble of N f -level atoms with free Hamiltonian

$$H_{\text{medium}} = \hbar \sum_{i=1}^N \sum_{\alpha=1}^f \Omega_{\alpha i} c_{\alpha i}^\dagger c_{\alpha i} \quad (2.6)$$

where the $c_{\alpha i}$ are the fermion annihilation operators for the α level of the i th atom.

A general interaction Hamiltonian describing a $m+n$ photon process may be written as

$$H_1 = \hbar \sum_{i=1}^N \sum_{\alpha, \beta} \epsilon_{\alpha, \beta}^{(m+n)} c_{\beta i}^\dagger c_{\alpha i} \prod_{j=1}^m E_j^{(-)}(\mathbf{r}_i) \prod_{k=1}^n E_k^{(+)}(\mathbf{r}_i) + \text{adjoint} \quad (2.7)$$

where $\epsilon_{\alpha, \beta}^{(m+n)}$ is the matrix element for a $m+n$ photon transition, consisting of m emissions and n absorptions and an atomic transition from state α to state β . The term $E_k^{(+)}$ represents a single mode of the electromagnetic field. (This is appropriate for cavity modes.)

$$E_k^{(-)}(\mathbf{r}_i) = \{E_k^{(+)}(\mathbf{r}_i)\}^\dagger = i(\frac{1}{2}\hbar\omega_k)^{1/2} u_k(\mathbf{r}_i) a_k. \quad (2.8)$$

Substituting equation (2.8) for $E_k^{(-)}$ and $E_k^{(+)}$ into equation (2.7) and assuming an even distribution of atoms throughout the interaction volume V the interaction Hamiltonian becomes

$$H_1 \simeq \sum_{\alpha, \beta} \mu_{\alpha, \beta}^{(m+n)} \sum_{i=1}^N c_{\beta i}^\dagger c_{\alpha i} \prod_{j=1}^m a_j \prod_{k=1}^n a_k^\dagger + \text{adjoint} \quad (2.9)$$

where

$$\mu_{\alpha, \beta}^{(m+n)} = \epsilon_{\alpha, \beta}^{(m+n)} \prod_{j=1}^{m+n} i(\frac{1}{2}\hbar\omega_j)^{1/2} \int_V d^3r \prod_{j=1}^m u_j(\mathbf{r}) \prod_{k=1}^n u_k^*(\mathbf{r}). \quad (2.10)$$

The coupling constants $\mu_{\alpha, \beta}^{m+n}$ will be taken to be real for convenience. This can always be accomplished by appropriate choice of the arbitrary phases of the mode functions.

3. Method of solution

The Heisenberg equations of motion resulting from a Hamiltonian of the form described in § 2 are nonlinear operator equations. One may however circumvent this intractable problem in the following manner. If the interaction described by equation (2.9) is perfectly energy conserving the following commutation relations may be shown to hold:

$$[H_0^0, H] = [H_1, H] = [H_0^0, H_1] = 0 \quad (3.1)$$

where H_0^0 denotes the perfectly energy conserving free Hamiltonian. Thus H_0^0 and H_1 are constants of the motion. Highly nonenergy conserving terms such as $c_2^\dagger c_1 a^\dagger$ ($E_2 > E_1$) are necessarily excluded from the Hamiltonian (rotating wave approximation). Such terms merely give rise to small frequency shifts. However, an energy mismatch in a basically energy conserving term such as $c_2^\dagger c_1 a$ ($E_2 > E_1$) may be taken into account in the following fashion. We may write the total Hamiltonian in the form

$$H = H_0^0 + H_1' \quad (3.2)$$

where

$$H_1' = H_1 + H_0 - H_0^0. \quad (3.3)$$

Then provided equation (3.1) holds, it follows that

$$[H_0^0, H] = [H_1', H] = [H_0^0, H_1'] = 0. \quad (3.4)$$

Thus H_0^0 and H_1' are constants of the motion. Since H_0^0 and H_1' commute a representation exists in which the total Hamiltonian H is diagonal (Messiah 1961). This diagonal representation is the solution to the Schrödinger equation.

One proceeds by choosing as a basis set the eigenstates of the free Hamiltonian H_0^0 . We may write these as a column vector

$$\Psi = \begin{pmatrix} \psi_1 \\ \vdots \\ \psi_j \\ \vdots \\ \psi_N \end{pmatrix} \quad (3.5)$$

where N is the dimension of the Hilbert space of the system studied.

The Schrödinger equation for the modified interaction Hamiltonian H' assumes the form

$$H_1' \Psi = \hbar A \Psi \quad (3.6)$$

where A is the matrix representation of H with dimension $N \times N$. The diagonalization of the matrix A yields the eigenvalues λ and eigenstates Φ of the system

$$\Phi = U \Psi \quad (3.7)$$

where U is the unitary matrix that diagonalizes A . The unitary property follows from the Hermitian character of the Hamiltonian. The probability amplitude of the system being in a particular state at time t may be calculated exactly from the known eigenstates and eigenvalues.

Alternatively, one may calculate the probability amplitudes directly by expanding the time dependent wavefunction of the system as a linear combination of the eigenstates of H_0^0 . The validity of this expansion as a basis set of states for the whole system rests on the commutative property (equation (3.4)). The wavefunction $\Psi(t)$ of the system at time t may be written

$$\Psi(t) = \sum_{j=1}^N c_j(t) \psi_j \quad (3.8)$$

where the $c_j(t)$ are the probability amplitudes of the system being in state j at time t .

The time dependent Schrödinger equation is

$$\frac{i \hbar d\Psi(t)}{dt} = H\Psi(t). \tag{3.9}$$

The $c_j(t)$ are then obtained as solutions of a coupled set of differential equations. Though in this treatise we shall use the former approach both methods are equivalent.

The above method provides a powerful tool for analysing certain problems involving the interaction of boson and fermion fields. It has its best applicability for systems where the dimension of the Hilbert space is small since an analytic solution is then feasible. Thus it is ideally suited for systems involving two or three fermion levels.

4. Two photon emission and absorption—degenerate case

Two photon transitions may be divided into two categories, degenerate and nondegenerate (Oka and Shimizu 1970). In the degenerate case shown in figure 1 each atomic level is a state of mixed parity. These transitions may only occur in media which lack inversion symmetry.

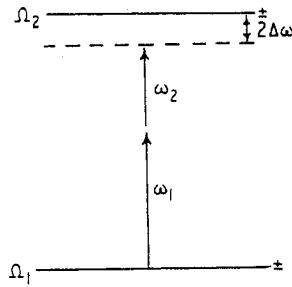


Figure 1. Double photon transition in the degenerate case.

The Hamiltonian for the two photon transition shown in figure 1 is taken to be

$$H_0 = \hbar\Omega_1 \sum_{j=1}^N c_{1j}^\dagger c_{1j} + \hbar\Omega_2 \sum_{j=1}^N c_{2j}^\dagger c_{2j} + \hbar\omega_1 a_1^\dagger a_1 + \hbar\omega_2 a_2^\dagger a_2 \tag{4.1}$$

$$H_1 = \hbar\mu^{(2)} \sum_{i=1}^N (a_1 a_2 c_{1i} c_{2i}^\dagger + a_1^\dagger a_2^\dagger c_{1i}^\dagger c_{2i}). \tag{4.2}$$

We allow for a frequency mismatch between the photons and the atomic energy levels

$$\omega_1 + \omega_2 = \Omega_2 - \Omega_1 - 2\Delta\omega. \tag{4.3}$$

4.1. *Incoherent atomic coupling*

We shall first consider the atoms in the medium to act independently, that is, with ‘cooperation number’ $N_c = 1$ (Dicke 1954, Arecchi and Courtens 1970). Thus we may restrict our attention to the interaction of the light field with a single atom. We choose as a basis set the states $|j, n_1, n_2\rangle$, where $|j\rangle$ are the stationary states of the atom and $|n_k\rangle$ are the number states for the field modes with frequencies ω_k . A

complete set of basis states assuming the relaxation time of the upper level is infinite is

$$\Psi^\dagger = (|1, n_1, n_2\rangle, |2, n_1 - 1, n_2 - 1\rangle). \quad (4.4)$$

The interaction matrix A (equation (3.6)) in this representation is

$$A = \begin{pmatrix} -\Delta\omega & g_{ab} \\ g_{ab} & \Delta\omega \end{pmatrix} \quad (4.5)$$

where

$$g_{ab} = (n_1 n_2)^{1/2} \mu^{(2)}. \quad (4.6)$$

The eigenvalues of the system are $\pm G_{ab}$ where

$$G_{ab} = (\Delta\omega^2 + g_{ab}^2)^{1/2}. \quad (4.7)$$

From the eigenstates and eigenvalues we may calculate exactly the probability for a two photon transition to take place. If the atom is initially in the ground state the probability for a two photon absorption is

$$P_{ab}^i(t) = \frac{g_{ab}^2}{G_{ab}^2} \sin^2 G_{ab} t. \quad (4.8)$$

When the atom is initially in the excited state it can be shown that the probability for a two photon emission is

$$P_e^i(t) = \frac{g_e^2}{G_e^2} \sin^2 G_e t \quad (4.9)$$

where

$$g_e = \{(n_1 + 1)(n_2 + 1)\}^{1/2} \mu^{(2)} \quad (4.10)$$

and

$$G_e = (\Delta\omega^2 + g_e^2)^{1/2}.$$

For an ensemble of N atoms with N_1 initially in the ground state and N_2 initially excited the net two photon absorption rate is

$$P(t) = N_1 P_{ab}^i(t) - N_2 P_e^i(t). \quad (4.11)$$

For short times and $\Delta\omega = 0$ this agrees with the result of first order perturbation theory

$$P(t) = \{N_1 n_1 n_2 - N_2 (n_1 + 1)(n_2 + 1)\} \mu^{(2)2} t^2. \quad (4.12)$$

4.2. Coherent atomic coupling

We now consider the atoms to act collectively with cooperation number $N_c = N$. In treating atomic coherence effects we find it convenient to make the following transformations on the atomic operators

$$\begin{aligned} \sum_{j=1}^N c_{1j} c_{2j}^\dagger &= \sum_{j=1}^N \sigma_j^+ = S^+ \\ \sum_{j=1}^N c_{1j}^\dagger c_{2j} &= \sum_{j=1}^N \sigma_j^- = S^- \\ \frac{1}{2} \left(\sum_{j=1}^N c_{2j}^\dagger c_{2j} - \sum_{j=1}^N c_{1j}^\dagger c_{1j} \right) &= \sum_{j=1}^N \sigma_z^j = S_z. \end{aligned} \quad (4.13)$$

Here σ_j^+ , σ_j^- are the raising and lowering operators for the individual atoms. S^+ and S^- are the collective raising and lowering operators for the N atoms. S_z is proportional to the total atomic energy operator with eigenvalues running from $N/2$ to $-N/2$.

Atomic coherence phenomena can perhaps be illustrated with greater lucidity by introducing the Schwinger representation for angular momentum (Schwinger 1965)

$$\begin{aligned} S^+ &= a_1 a_2^\dagger \\ S^- &= a_1^\dagger a_2 \\ S_z &= \frac{1}{2}(a_2^\dagger a_2 - a_1^\dagger a_1) \end{aligned} \quad (4.14)$$

where a_1 and a_2 obey boson commutation relations. However, they are not true boson operators since the eigenvalues of $a_1^\dagger a_1$ and $a_2^\dagger a_2$ only span the integer spectrum from 0 to N_c . For a system of N atoms with $N_c = N$ the eigenvalues of $a_2^\dagger a_2$ and $a_1^\dagger a_1$ are the actual occupation numbers of the upper and lower levels.

We consider an initial state with N_1 atoms in the ground state and N_2 excited where $N_1 + N_2 = N_c = N$. For a single two photon absorption process we take as a basis set

$$\Psi_c^\dagger = (|N_1, N_2, n_1, n_2\rangle, |N_1 - 1, N_2 + 1, n_1 - 1, n_2 - 1\rangle). \quad (4.15)$$

Here $|N_j\rangle$ refers to the occupation number of the atomic levels and $|n_j\rangle$ refers to the number states of the field. The two states in equation (4.15) do not form a complete set of basis states for a single two photon absorption. Other states contribute to processes corresponding to a real absorption of a pair of photons accompanied by virtual absorption and re-emission of photon pairs. However, neglecting these states is analogous to the ladder approximation to the Bethe-Salpeter equation (Salpeter and Bethe 1951). The validity of this approximation in the present context has been discussed by Walls (1971c). By taking the two states of equation (4.15) as a basis set we still include arbitrarily high powers of H_1 . Hence this approximation is much superior to a perturbation calculation.

The interaction matrix A (equation (3.6)) in the representation equation (4.15) is

$$A = \begin{pmatrix} \Delta\omega & g_c \\ g_c & -\Delta\omega \end{pmatrix} \quad (4.16)$$

where

$$g_c = \mu^{(2)} \{n_1 n_2 (N_2 + 1) N_1\}^{1/2}.$$

The eigenvalues of the system are $\pm G_c$ where

$$G_c = \{\Delta\omega^2 + n_1 n_2 N_1 (N_2 + 1) \mu^{(2)2}\}^{1/2}. \quad (4.17)$$

The probability for a two photon absorption process to take place is calculated to be

$$P_c(t) = \frac{g_c^2}{G_c^2} \sin^2 G_c t. \quad (4.18)$$

For short times and $\Delta\omega = 0$ this becomes

$$P_c(t) = n_1 n_2 N_1 (N_2 + 1) \mu^{(2)2} t^2. \quad (4.19)$$

The maximum absorption rate occurs for an equal population of upper and lower atomic levels $N_1 = N_2 = N/2$. That is, a 'superabsorbing atomic state'. The

absorption rate for a superabsorbing atomic state is proportional to the number of atoms squared as opposed to simply the number of atoms when all the atoms are in the ground state. It would be possible to reach such a superabsorbing state starting from all atoms in the ground state provided the duration of the incident light pulse is less than or comparable to the lifetime of the excited levels. A similar result to the above may be derived for two photon emission.

5. Two photon emission and absorption—nondegenerate case

We now consider the nondegenerate case where each level has a definite parity. Three atomic levels are then required to satisfy conservation of angular momentum in a two photon process (figure 2).

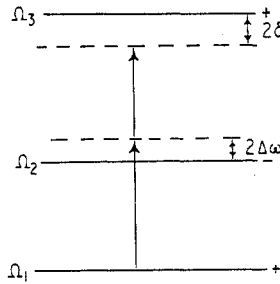


Figure 2. Double photon transition in the nondegenerate case.

This process may be regarded as an example of double resonance phenomena (Walls 1971b). The Hamiltonian for the two photon transition shown in figure 2 is

$$\begin{aligned}
 H_0 = & \hbar\Omega_1 \sum_{j=1}^N c_{1j}^\dagger c_{1j} + \hbar\Omega_2 \sum_{j=1}^N c_{2j}^\dagger c_{2j} + \hbar\Omega_3 \sum_{j=1}^N c_{3j}^\dagger c_{3j} \\
 & + \hbar\omega_1 a_1^\dagger a_1 + \hbar\omega_2 a_2^\dagger a_2
 \end{aligned} \tag{5.1}$$

$$\begin{aligned}
 H_1 = & \hbar\mu_{12}^{(1)} \sum_{j=1}^N (c_{1j} c_{2j}^\dagger a_1 + c_{1j}^\dagger c_{2j} a_1^\dagger) \\
 & + \hbar\mu_{23}^{(1)} \sum_{j=1}^N (c_{2j} c_{3j}^\dagger a_2 + c_{2j}^\dagger c_{3j} a_2^\dagger).
 \end{aligned} \tag{5.2}$$

We set $\delta = 0$, that is $\omega_1 - \omega_2 = \Omega_3 - \Omega_1$ and allow for an energy mismatch $2\Delta\omega$ with the intermediate level

$$2\Delta\omega = \omega_1 - (\Omega_2 - \Omega_1) = \Omega_3 - \Omega_2 - \omega_2. \tag{5.3}$$

A finite value for δ could have been chosen but then a solution for the eigenvalues in closed form is not possible without approximation (Walls 1971b).

We shall consider the case of incoherent atomic coupling, that is, $N_c = 1$, thus we need only consider the interaction with a single atom. A complete set of basis states describing a two photon absorption process is

$$\Psi^\dagger = (|1, n_1, n_2\rangle, |2, n_1 - 1, n_2\rangle, |3, n_1 - 1, n_2 - 1\rangle). \tag{5.4}$$

The interaction matrix A (equation (3.6)) in this representation is

$$A = \begin{pmatrix} -\Delta\omega & g_1 & 0 \\ g_1 & \Delta\omega & g_2 \\ 0 & g_2 & \Delta\omega \end{pmatrix} \quad (5.5)$$

where

$$\begin{aligned} g_1 &= \mu_{12}^{(1)} n_1^{1/2} \\ g_2 &= \mu_{23}^{(1)} n_2^{1/2}. \end{aligned} \quad (5.6)$$

The eigenvalues are $\Delta\omega, \pm G_{12}$ where

$$G_{12} = (g_1^2 + g_2^2 + \Delta\omega^2)^{1/2}. \quad (5.7)$$

For N_1 atoms all initially in the ground state the two photon absorption rate is given by

$$P(t) = \frac{N_1 g_1^2 g_2^2}{G_{12}^4} \left| \exp(i\Delta\omega t) \left(\cos G_{12}t + \frac{i\Delta\omega}{G_{12}} \sin G_{12}t \right) - 1 \right|^2. \quad (5.8)$$

The rate for two photon emission may be derived in an analogous fashion. For perfect resonance and short times the above result agrees with perturbation theory calculations

$$P(t) = N_1 n_1 n_2 (\mu^{(1)} \mu^{(2)})^2 t^4. \quad (5.9)$$

The initial absorption rate grows as t^4 as opposed to the t^2 growth for the degenerate case. This t^4 growth reflects the two step nature of the process. However, the dipole matrix elements $\mu^{(1)}$ are many orders of magnitude larger than the matrix elements $\mu^{(2)}$ for the two photon transition. The relative magnitudes of these matrix elements are compared in the papers of Gold (1969), and Oka and Shimizu (1970).

6. Frequency up-conversion and second harmonic generation

The process of frequency up-conversion may be considered as a two step process consisting of a two photon absorption followed by a single photon emission at the sum frequency as illustrated in figure 3 (Winter 1969).

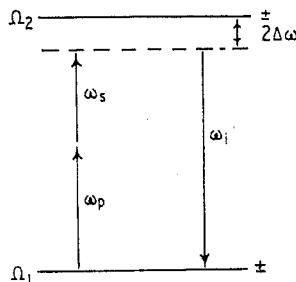


Figure 3. Frequency up-conversion as a multiphoton process.

The frequency up-conversion process must take place in a crystal lacking inversion symmetry. We consider three modes of the electromagnetic field, the pump, signal and idler modes with frequencies ω_p, ω_s and ω_i respectively. We allow for a

frequency mismatch with the atomic levels

$$\omega_i = \omega_p + \omega_s = \Omega_2 - \Omega_1 - 2\Delta\omega. \quad (6.1)$$

The Hamiltonian for the frequency up-conversion process depicted in figure 3 is

$$H_0 = \hbar\omega_p a_p^\dagger a_p + \hbar\omega_s a_s^\dagger a_s + \hbar\omega_i a_i^\dagger a_i + \hbar\Omega_1 \sum_{j=1}^N c_{1j}^\dagger c_{1j} + \hbar\Omega_2 \sum_{j=1}^N c_{2j}^\dagger c_{2j} \quad (6.2)$$

$$H_1 = \hbar \sum_{j=1}^N \mu^{(2)} (a_p a_i c_{1j} c_{2j}^\dagger + a_p^\dagger a_i^\dagger c_{1j}^\dagger c_{2j}) + \hbar \sum_{j=1}^N \mu^{(1)} (a_s^\dagger c_{1j}^\dagger c_{2j} + a_s c_{1j} c_{2j}^\dagger). \quad (6.3)$$

6.1. Incoherent atomic coupling

We first consider the atoms to act independently and choose the following set of basis states for a one photon up-conversion process

$$\Psi^\dagger = (|1, n_p, n_s, n_i\rangle, |2, n_p - 1, n_s - 1, n_i\rangle, |1, n_p - 1, n_s - 1, n_i + 1\rangle) \quad (6.4)$$

where the states $|j\rangle$ are the stationary states of the atom and the states $|n_k\rangle$ are the number states of the field modes with frequency ω_k . In principle an n photon up-conversion process may be treated, however, an analytic solution is then mathematically complex. The choice of the three states in equation (6.4) to represent a basis set for a one photon up-conversion process is again analogous to the ladder approximation to the Bethe-Salpeter equation (Walls 1971c).

The interaction matrix A (equation (3.6)) in the representation equation (6.4) is

$$A = \begin{pmatrix} -\Delta\omega & g_{ps} & 0 \\ g_{ps} & \Delta\omega & g_i \\ 0 & g_i & -\Delta\omega \end{pmatrix} \quad (6.5)$$

where

$$\begin{aligned} g_{ps} &= (n_p n_s)^{1/2} \mu^{(2)} \\ g_i &= (n_i + 1)^{1/2} \mu^{(1)}. \end{aligned} \quad (6.6)$$

The eigenvalues found by diagonalizing the matrix A are $\Delta\omega, \pm G_f$ where

$$G_f = (\Delta\omega^2 + g_{ps}^2 + g_i^2)^{1/2}. \quad (6.7)$$

The corresponding eigenstates are given by equation (3.7) with Ψ defined by equation (6.4) and where U is the 3×3 matrix

$$U = \begin{pmatrix} -\frac{g_i}{g} & 0 & \frac{g_{ps}}{g} \\ \frac{g_{ps}}{\alpha} & \frac{D}{\alpha} & \frac{g_i}{\alpha} \\ -\frac{g_{ps}}{\beta} & \frac{S}{\beta} & -\frac{g_i}{\beta} \end{pmatrix} \quad (6.8)$$

where

$$\begin{aligned} g &= (g_{ps}^2 + g_1^2)^{1/2} \\ D &= G_f - \Delta\omega & \alpha^2 &= D^2 + g^2 \\ S &= G_f + \Delta\omega & \beta^2 &= S^2 + g^2. \end{aligned}$$

We note here the close similarity between the above eigenstates and eigenvalues and those for Raman scattering from a three level molecule (Walls 1971a). In fact the exact analogy to the frequency up-conversion process is the generation of anti-Stokes radiation. The generation of Stokes radiation corresponds to the parametric amplification process considered in § 7. While in frequency up-conversion two photons combine to create a photon at the sum frequency, in anti-Stokes generation a photon plus a molecular excitation combine to create a photon at the sum frequency.

For N atoms all initially in the ground state and acting independently the rate of frequency up-conversion calculated from the above eigenfunctions and eigenvalues is

$$P(t) = \frac{Ng_{ps}^2g_1^2}{G_f^4} \left| \exp(i\Delta\omega t) \left(\cos G_f t + \frac{i\Delta\omega}{G_f} \sin G_f t \right) - 1 \right|^2. \quad (6.9)$$

For short times and $\Delta\omega = 0$ this reduces to

$$P(t) = Nn_p n_s (n_1 + 1) (\mu^{(1)} \mu^{(2)})^2 t^4. \quad (6.10)$$

The t^4 dependence again reflects the two step process.

6.2. Atomic coherence

In a medium where there is some degree of atomic cooperation the frequency up-conversion process is enhanced in a similar way to two photon absorption. The calculations may be performed in an analogous fashion to that described in § 4.2. We shall just present the results here. For an ensemble of N atoms with cooperation number $N_c = N$ with N_1 atoms in the ground state and N_2 excited the rate of frequency up-conversion is (for short times and $\Delta\omega = 0$)

$$P(t) = n_p n_s (n_1 + 1) N_1^2 (N_2 + 1)^2 (\mu^{(1)} \mu^{(2)})^2 t^4. \quad (6.11)$$

The maximum rate of up-conversion occurs for $N_1 = N_2 = N/2$. The rate of up-conversion is then proportional to the number of atoms to the fourth power since both the absorption of two quanta and the subsequent emission of a single quantum are enhanced by the atomic cooperation. That is, a superabsorbing state followed by a superradiant state.

6.3. Second harmonic generation

A special case of frequency up-conversion is the much studied process of second harmonic generation. This occurs when $\omega_p = \omega_s = \omega_i/2$. The results for second harmonic generation are identical to the above results for frequency up-conversion with the substitutions $n_p = n_s + 1 = n_1$, where n_1 is the number of photons in the fundamental field.

7. Parametric amplification

The process of parametric amplification corresponds to the time reversed process of frequency up-conversion. A pump photon with frequency ω_p is absorbed followed

by the emission of a signal and idler photon with frequencies ω_s and ω_i respectively (see figure 4).

Thus the illumination of the medium with a strong pump field results in the amplification of the signal field at frequency ω_s .

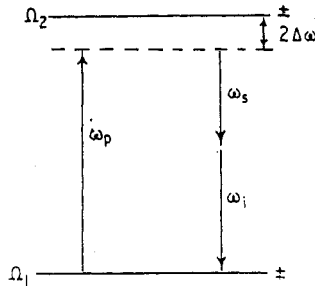


Figure 4. Parametric amplification as a multiphoton process.

The free Hamiltonian is identical to equation (5.2). The interaction Hamiltonian is

$$\begin{aligned}
 H_1 = \hbar\mu^{(1)} \sum_{j=1}^N (a_p c_{1j} c_{2j}^\dagger + a_p^\dagger c_{1j}^\dagger c_{2j}) \\
 + \hbar\mu^{(2)} \sum_{j=1}^N (c_{1j}^\dagger c_{2j} a_1^\dagger a_s^\dagger + c_{1j}^\dagger c_{2j} a_1 a_s). \quad (7.1)
 \end{aligned}$$

This system may be analysed in an exactly analogous fashion to that described in the preceding section. For N atoms acting independently, and all initially in the ground state the signal field is amplified at a rate given by

$$P(t) = \frac{N g_{si}^2 g_p^2}{G_a^4} \left| \exp(i \Delta\omega t) \left(\cos G_a t + \frac{i \Delta\omega \sin G_a t}{G_a} \right) - 1 \right|^2$$

where

$$\begin{aligned}
 g_p &= n^{1/2} \mu^{(2)} \\
 g_{si} &= \{(n_s + 1)(n_i + 1)\}^{1/2} \mu^{(1)} \\
 G_a &= (g_p^2 + g_{si}^2 + \Delta\omega^2)^{1/2}. \quad (7.3)
 \end{aligned}$$

For short times and $\Delta\omega = 0$ this reduces to

$$P(t) = N n_p (n_s + 1)(n_i + 1) (\mu^{(1)} \mu^{(2)})^2 t^4. \quad (7.4)$$

For an initial state with signal and idler modes in the vacuum the amplification process begins by spontaneous emission. The amplification process is enhanced by atomic cooperation in a like fashion to frequency conversion.

8. Superradiance

We present here an exact solution to a model problem illustrating the phenomena of superradiance. We consider two 2-level atoms denoted by α and β interacting with a single mode of the radiation field with frequency ν . The two atoms are assumed to lie within a 'cooperation length' (Dicke 1954, Arecchi and Courtens 1970) of each

other. Effects related to the spatial separation of the two atoms involve the interaction with many field modes. This problem has been considered by Stephen (1964) and Arecchi and Courtens (1970).

The Hamiltonian describing the interaction is

$$H_0 = \hbar\nu a^\dagger a + \hbar\omega c_{2\alpha}^\dagger c_{2\alpha} + \hbar\omega c_{2\beta}^\dagger c_{2\beta} \quad (8.1)$$

$$H_1 = \hbar\mu^{(1)}(ac_{2\alpha}^\dagger c_{1\alpha} + a^\dagger c_{2\alpha} c_{1\alpha}^\dagger) + \hbar\mu^{(1)}(ac_{2\beta}^\dagger c_{1\beta} + a^\dagger c_{2\beta} c_{1\beta}^\dagger) \quad (8.2)$$

where we allow for a frequency mismatch between the field and the atomic energy levels $2\delta = \omega - \nu$.

We choose as a basis set the states $|j_\alpha, j_\beta, n\rangle$, where $|j\rangle$ are the stationary states of the atoms and $|n\rangle$ are the number states of the field. For the case where only one quantum of energy is present a complete set of basis states is given by

$$\Psi^\dagger = (|2_\alpha, 1_\beta, 0\rangle, |1_\alpha, 2_\beta, 0\rangle, |1_\alpha, 1_\beta, 1\rangle). \quad (8.3)$$

The interaction matrix A (equation (3.6)) in this representation is

$$A = \begin{pmatrix} \delta & 0 & \mu^{(1)} \\ 0 & \delta & \mu^{(1)} \\ \mu^{(1)} & \mu^{(1)} & -\delta \end{pmatrix}. \quad (8.4)$$

The eigenvalues of the system are $\delta, \pm G$ where

$$G = (2\mu^{(1)2} + \delta^2)^{1/2}. \quad (8.5)$$

The wavefunction of the system at time t may be described by a linear combination of the basis states

$$\Psi(t) = a_{21}(t)|2_\alpha, 1_\beta, 0\rangle + a_{12}(t)|1_\alpha, 2_\beta, 0\rangle + a_{11}(t)|1_\alpha, 1_\beta, 1\rangle. \quad (8.6)$$

The probability amplitudes $a_{ij}(t)$ calculated from the eigenstates and eigenvalues of A are

$$\begin{pmatrix} a_{21}(t) \\ a_{12}(t) \\ a_{11}(t) \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1+\Gamma & \Gamma-1 & -\sqrt{2}\Lambda^* \\ \Gamma-1 & 1+\Gamma & -\sqrt{2}\Lambda^* \\ \sqrt{2}\Lambda & \sqrt{2}\Lambda & 2\Gamma \end{pmatrix} \begin{pmatrix} a_{21}(0) \\ a_{12}(0) \\ a_{11}(0) \end{pmatrix} \quad (8.7)$$

where

$$\Gamma = \exp(-i\delta t) \left(\cos Gt + \frac{i\delta}{G} \sin Gt \right)$$

$$\Lambda = \frac{i\sqrt{2}\mu^{(1)}}{G} \exp(i\delta t) \sin Gt.$$

If initially there is an atomic excitation ($a_{11}(0) = 0$) the probability amplitude for a photon emission at time t is

$$a_{11}(t) = \frac{i\mu^{(1)}}{G} \exp(i\delta t) \sin Gt \{a_{21}(0) + a_{12}(0)\}. \quad (8.8)$$

Thus for an initial state with wavefunction

$$\Psi(0) = \frac{1}{\sqrt{2}} (|2_\alpha, 1_\beta, 0\rangle - |1_\alpha, 2_\beta, 0\rangle) \quad (8.9)$$

the probability of emitting a photon is zero. This antisymmetrical state is known as a subradiant state.

If the initial state has the symmetrical wavefunction

$$\Psi(0) = \frac{1}{\sqrt{2}}(|2_{\alpha}, 1_{\beta}, 0\rangle + |1_{\alpha}, 2_{\beta}, 0\rangle) \quad (8.10)$$

the probability for a photon emission is

$$P_{11}(t) = |a_{11}(t)|^2 = \frac{2\mu^{(1)^2}}{G^2} \sin^2 Gt. \quad (8.11)$$

This is twice the probability of emission from an isolated excited atom. Thus the symmetrical state is known as a superradiant state.

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