IOPscience iopscience.iop.org

Home Search Collections Journals About Contact us My IOPscience

Quantum theory of nonlinear optical phenomena

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1979 J. Phys. A: Math. Gen. 12 2233

(http://iopscience.iop.org/0305-4470/12/11/031)

View the table of contents for this issue, or go to the journal homepage for more

Download details:
IP Address: 129.252.86.83
The article was downloaded on 30/05/2010 at 19:14

Please note that terms and conditions apply.

J. Phys. A: Math. Gen., Vol. 12, No. 11, 1979. Printed in Great Britain

Quantum theory of nonlinear optical phenomena

Miguel Orszag

Department of Physics, Ryerson Polytechnical Institute, Toronto, Ontario, Canada

Received 16 August 1978, in final form 8 May 1979

Abstract. The equivalence between the Hamiltonians representing nonlinear optical phenomena and N two-level atoms interacting via the electromagnetic field is exploited to find a solution to a parametric amplifier, a frequency converter and to the problems of Brillouin and Raman scattering. The n-photon probability and average number of photons as a function of time are obtained for all these cases. The results of the parametric amplifier can be readily applied to the Brillouin and Raman effects. In particular the results obtained for spontaneous fluorescence are applicable to the Stokes line of the stimulated Raman effect. Other nonlinear effects are discussed in the context of the present theory.

1. Introduction

This paper deals with the basic phenomena of the coupling between three light waves at three different frequencies, the coupling taking place in a nonlinear crystal. A number of nonlinear crystals are currently known, such as ADP, KDP, ADA, CDA, $LiNbO_3$ and many others, each one having a unique frequency range for efficient second-harmonic generation or parametric amplification.

The first observation of nonlinear optical effects was made by Franken *et al* (1961) when they obtained optical second-harmonic generation using a ruby laser. In the same year Kaiser and Garret (1961) observed a nonlinear absorption due to a two-photon process, also using a ruby laser.

In the early and middle nineteen sixties nonlinear optical effects were rapidly being discovered.

The theoretical treatment of nonlinear optical phenomena was developed by Armstrong *et al*(1962) assuming that the fields behave classically.

There have been several authors who approached the parametric amplification and frequency conversion problem, using quantum mechanical models (Louisell *et al* 1961, Louisell 1964, Mollow and Glauber 1967, Graham and Haken 1968a, b, Tucker and Walls 1969). In these models the parametric approximation is used, where the pump is assumed to behave classically with constant amplitude. The depletion of the pump field is therefore neglected. We shall see that these approximations are valid only for short times when the scaled time $\tau \ll (2R)^{-1/2}$. These assumptions are avoided in the present work.

The model for parametric amplification and frequency conversion is presented in § 2 and a formal analogy is established between the models for the atomic system coupled with electromagnetic radiation and the parametric amplifier.

Section 3 is devoted to the nonlinear spontaneous fluorescence process and stimulated parametric amplification. In the case of spontaneous fluorescence the solution

 © 1979 The Institute of Physics

found in paper I (Orszag 1979a) can be used directly, provided the formal correspondence between the parameters of the atomic system and the nonlinear process is made. A solution for the time-dependent average number of idler photons is found.

In the case of stimulated parametric amplification a mathematical procedure is developed to find the n-photon probability distribution and the time-dependent average number of idler photons. The technique used here is similar to the one used in paper I.

Section 4 deals with frequency converters and § 5 with other nonlinear optical processes, such as stimulated Raman scattering, the inverse Raman effect, Brillouin scattering and super-radiance.

2. The model

We consider three electromagnetic fields coupled through a nonlinear dielectric; the three fields are labelled by 1, 2 and 3 such that

$$\omega_1 = \omega_2 + \omega_3, \tag{2.1}$$

which is the energy conservation condition, and also

$$K_1 = K_2 + K_3, \tag{2.2}$$

corresponding to the conservation of momentum or phase matching condition.

If a_1, a_2, a_3 denote the annihilation operators of the three fields, the well known boson commutation rules may be written

$$[a_i, a_j^+] = \delta_{ij}, \qquad i, j = 1, 2, 3. \tag{2.3}$$

In the case of parametric amplification, a pump or laser field at frequency ω_1 combines with a signal field at frequency ω_2 to generate an idler field at the difference frequency $(\omega_1 - \omega_2)$. (The laser frequency is ω_1 and the signal frequency ω_2.) The process is described graphically in figure 1 (Yariv 1975, Yariv and Pearson 1969). The transition in question can be described quantum mechanically as follows:

$$|n_1, n_2, n_3\rangle \rightarrow |n_1 - 1, n_2 + 1, n_3 + 1\rangle,$$

where a laser photon is annihilated and a photon is created both in the signal and idler fields.

However, in frequency conversion the pump field at frequency ω_3 combines with the signal frequency at frequency ω_2 to generate the idler field at the sum frequency $(\omega_2 + \omega_3)$. This process is displayed graphically in figure 2. The corresponding quantum

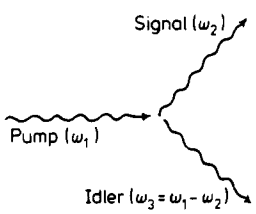

Figure 1. The parametric amplifier; a pump or laser field at frequency ω_1 combines with a signal field at frequency ω_2 producing an idler field at frequency $\omega_3 = \omega_1 - \omega_2$. The requirement of conservation of momentum is $K_1 = K_2 + K_3$.

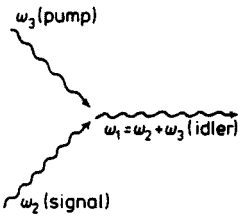

Figure 2. The frequency up-converter; a pump field (ω_3) combines with a signal field (ω_2) to generate an idler field at the sum frequency ($\omega_1=\omega_2+\omega_3$). Conservation of momentum is required.

transition is

$$|n_1, n_2, n_3\rangle \rightarrow |n_1+1, n_2-1, n_3-1\rangle.$$

The basic Hamiltonian describing both the frequency converter and the parametric amplifier is (Walls and Barakat 1970, Louisell 1964)

$$H=\hbar\omega_1 a_1^+ a_1+\hbar\omega_2 a_2^+ a_2+\hbar\omega_3 a_3^+ a_3+\hbar K(a_1 a_2^+ a_3^+ + a_1^+ a_2 a_3). \tag{2.4}$$

In the case of parametric amplification a_1 represents the pump mode, a_2 the signal mode and a_3 the idler. However, if the Hamiltonian (2.4) describes a frequency converter, a_1 is the idler, a_2 the signal mode and a_3 the pump. In a given experimental configuration one of the processes will be eliminated because of the phase matching condition.

It has been shown that the Hamiltonian (2.4) can also describe a system of N two-level atoms interacting via the radiation field (Bonifacio and Preparata 1970, Bonifacio and Masserini 1968). The Hamiltonian of the N two-level system is

$$H=\hbar\omega a^+ a+\hbar\omega R_3+\hbar K(aR^+ + a^+ R^-) \tag{2.5}$$

where R_3, R^+ and R^- are the z component, raising and lowering angular momentum operators respectively and a, a^+ are the annihilation and creation operators of the field. The connection between the Hamiltonians (2.4) and (2.5) can be readily established by defining

$$R^+=a_2 a_1^+, \qquad R^-=a_2^+ a_1. \tag{2.6}$$

Notice that here we are presenting a formal equivalence between the Hamiltonians of the parametric amplifier and frequency converter (equation (2.4)) and the Hamiltonian corresponding to an N two-level system in the rotating-wave approximation (equation (2.5)). Since we shall later use the results obtained in the restricted rotating-wave approximation, an error is introduced and the present analysis is only approximate. A critical study of this approximation is presented in the final section.

From the well known rules of commutation for R^+ and R^-, R_3 must be defined as

$$R_3=\tfrac{1}{2}(a_1^+ a_1-a_2^+ a_2), \tag{2.7}$$

and if we interpret $a^+ a$ as $a_3^+ a_3$ the two Hamiltonians are identical and any solution for the two-level system can be applied directly to the frequency converter and parametric amplifier (Walls and Barakat 1970). The operators $a_1^+ a_1$ and $a_2^+ a_2$ are interpreted as the effective populations of the upper and lower levels of the atoms respectively.

Consistently with equations (2.6) and (2.7) we can write

$$R = \tfrac{1}{2}(a_1^+ a_1 + a_2^+ a_2). \tag{2.8}$$

Using as working states for our Hilbert space the product state of Dicke's and $|n\rangle$ states,

$$|m\rangle|n\rangle, \tag{2.9}$$

and since the operators $N = a^+ a + R_3$ and R^2 are conserved, we will rely on two good quantum numbers R and $M = n + m$, which can be written as

$$\begin{aligned} M &= n_3(\tau) + \tfrac{1}{2}(n_1(\tau) - n_2(\tau)) = n_3 + \tfrac{1}{2}(n_1 - n_2), \\ R &= \tfrac{1}{2}(n_1(\tau) + n_2(\tau)) = \tfrac{1}{2}(n_1 + n_2), \end{aligned} \tag{2.10}$$

where M and $R(R+1)$ are the eigenvalues of N and R^2 respectively and n_1, n_2 and n_3 are the initial number of photons of the three fields.

3. The parametric amplifier

3.1. Spontaneous emission

This is the case where there are initially n_1 pump photons and no signal photons ($n_2 = 0$). We will also assume that the initial number of idler photons is also zero ($n_3 = 0$). From equations (2.10) it is clear that

$$M = R = \tfrac{1}{2}n_1, \tag{3.1}$$

and

$$\begin{aligned} n_1(\tau) &= n_1 - n_3(\tau), \\ n_2(\tau) &= n_3(\tau). \end{aligned} \tag{3.2}$$

In terms of the two-level atom the case of the parametric amplifier for spontaneous emission corresponds to the atomic system initially prepared in the state of complete inversion (for the two-level system, it corresponds to spontaneous emission, since $n(0) = n_3 = 0$). This problem was solved exactly by the author (paper I), valid for all times (Orszag 1979a). The solution for the n-photon probability is

$$\begin{aligned} |p(n,\tau)|^2 = {} & \left[\prod_{i=1}^{n} (i)(2R - i + 1)\right](n!)^{-3} 2^{-4R} \\ & \times \left(\sum_{s_1=0}^{n} \sum_{s_2=0}^{2R-n} \binom{n}{s_1}\binom{2R-n}{s_2}(-1)^{n-s_1}[2\tau(s_1+s_2-R)]^n \right. \\ & \left. \times \exp[-2\tau^2(s_1+s_2-R)^2]\right)^2 \Big/ \sum_{n=0}^{2R} |p(n,\tau)|^2. \end{aligned} \tag{3.3}$$

Equation (3.3) represents the n-photon emission probability as a function of the scaled time $\tau = Kt$.

A time-dependent average number of photons is simply

$$n(\tau) = n_3(\tau) = \sum_{n=0}^{2R} n|p(n,\tau)|^2. \tag{3.4}$$

An APL program was written to calculate the n-photon probability and the average number of photons. Some of the numerical results are shown in figures 3, 4 and 5. Figure 3 shows $n(\tau)$ (or $n_3(\tau)$) when the parameters are

$$M = R = \tfrac{3}{2}, \qquad n_1 = 3, \qquad n_2 = 0.$$

For long times

$$n_3(\tau) \to n_{\max} = 3, \tag{3.5}$$

corresponding to a complete depletion of the pump. Normally this limit is not achieved experimentally, since it requires a long interaction time, or equivalently, a very large nonlinear medium. Figures 4 and 5 show a similar behaviour. Figure 4 has parameters

$$M = R = \tfrac{9}{2}, \qquad n_1 = 9, \qquad n_2 = 0,$$

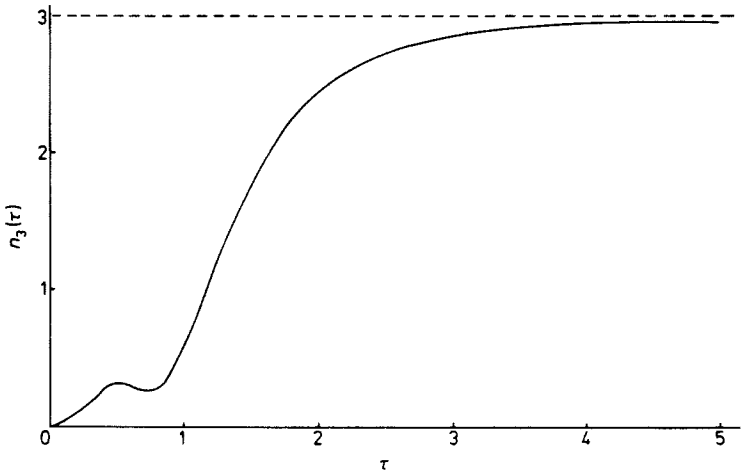

Figure 3. Number of idler photons versus time τ for spontaneous fluorescence. The parameters are $R = M = 1{\cdot}5$, $n\,(0) = 0$ or $n_1 = 3$ (pump), $n_2 = 0$ (signal), $n_3 = 0$ (idler).

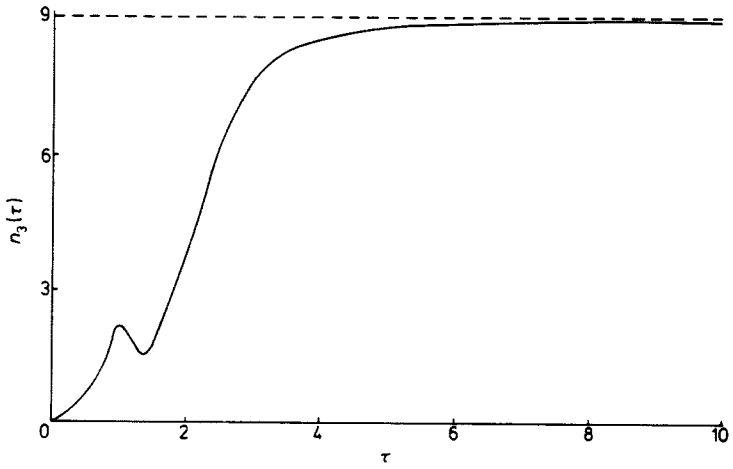

Figure 4. As figure 3 for $R = M = 4{\cdot}5$ and $n(0) = 0$ or $n_1 = 9$ (pump), $n_2 = 0$ (signal), $n_3 = 0$ (idler).

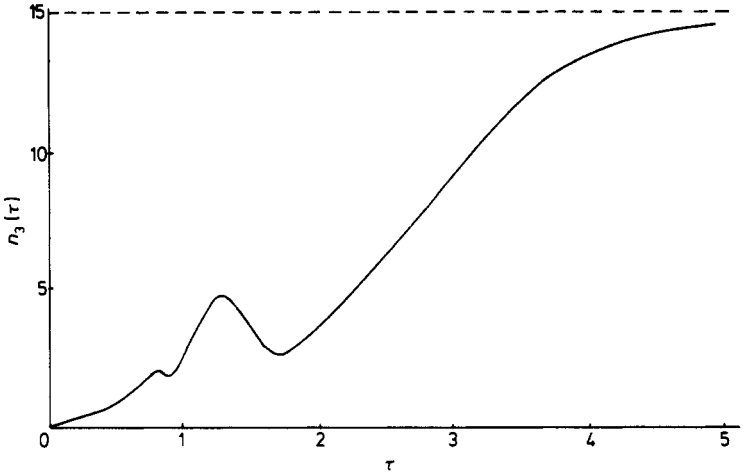

Figure 5. As figure 3 for $R = M = 7{\cdot}5$, $n(0) = 0$ or $n_1 = 15$ (pump), $n_2 = 0$ (signal), $n_3 = 0$ (idler).

and figure 5

$$M = R = \tfrac{15}{2}, \qquad n_1 = 15, \qquad n_2 = 0.$$

Finally, the spontaneous emission case, also termed spontaneous parametric fluorescence, is a typical quantum mechanical effect which cannot be described in semiclassical terms. Its two-level atomic counterpart corresponds, as mentioned above, to initial full inversion. The radiation emitted from the atoms is of a purely quantum nature.

3.2. Stimulated emission

In this case both $n_1 \neq 0$ and $n_2 \neq 0$. The special case we are going to solve is:

$$n_1 = n_2; \qquad n_3 = 0, \tag{3.6}$$

or

$$M = 0; \qquad R = n_1. \tag{3.7}$$

Using equations (2.10), it is easy to verify that

$$n_1(\tau) = n_1 - n_3(\tau), \qquad n_2(\tau) = n_1 + n_3(\tau). \tag{3.8}$$

In the two-level system language, the case $M = 0$, $R = n_1$ corresponds to a system of two-level atoms initially prepared in the super-radiant state (spontaneous emission, since $n_3 = 0$).

The system, therefore, is expected to behave semiclassically and the quantum features or corrections should have little effect. As in paper I, we will use Dicke's model in the restricted rotating-wave approximation and $p(n, \tau)$ can be written as

$$p(n, \tau) = \langle n|\langle M - n| \exp[-\mathrm{i}\tau(a + a^+)(R^+ + R^-)]|M\rangle|0\rangle \exp[-\mathrm{i}\omega_1 M\tau] \tag{3.9}$$

where

$$\tau = Kt, \qquad \omega_1 = \omega/K.$$

Using the unscrambling theorems of Arecchi *et al* (1972) we can write

$$e^{X_+R^+}\, e^{(\ln X_Z)R_3}\, e^{X_-R^-} = \begin{pmatrix} X_Z^{1/2} + \dfrac{X_+X_-}{X_Z^{1/2}} & \dfrac{X_+}{X_Z^{1/2}} \\ \dfrac{X_-}{X_Z^{1/2}} & X_Z^{-1/2} \end{pmatrix} \tag{3.10}$$

and

$$e^{\omega_+R^+ + \omega_-R^- + \omega_Z R_3} = \begin{pmatrix} \cosh k + \frac{1}{2}\omega_Z \sinh k & \omega_+ \dfrac{\sinh k}{k} \\ \omega_- \dfrac{\sinh k}{k} & \cosh k - \frac{1}{2}\omega_Z \sinh k \end{pmatrix} \tag{3.11}$$

where for our specific purpose

$$\begin{aligned} &\omega_+ = \omega_- = -\mathrm{i}\tau(a + a^+), \\ &\omega_Z = 0, \qquad k = \mathrm{i}\tau(a + a^+). \end{aligned} \tag{3.12}$$

Identifying each term of the matrices (3.10) and (3.11) and using (3.12), we obtain

$$\begin{aligned} &X_+ = X_- = -\mathrm{i}\tan\tau(a + a^+), \\ &X_Z = \cos^{-2}\tau(a + a^+), \end{aligned} \tag{3.13}$$

and $p(n, \tau)$ can be written as

$$p(n, \tau) = \exp(-\mathrm{i}\omega_1\tau M)\langle n|\langle M - n|\exp(X_+R^+)\exp[(\ln X_Z)R_3]\exp[X_-R^-]|M\rangle|O\rangle. \tag{3.14}$$

Using the following properties of the angular momentum operators,

$$(R^-)^q|M\rangle = \prod_{s=1}^{q}[(R + M + 1 - s)(R - M + s)]^{\frac{1}{2}}|M - q\rangle, \tag{3.15}$$

$$(R^-)^q|M\rangle \equiv \prod_{s=1}^{q}(f_s)|M - q\rangle, \tag{3.16}$$

and

$$\begin{aligned} (R^+)^r|M - q\rangle &= \prod_{t=1}^{r}[(R - M + q + 1 - t)(R + M - q + t)]^{1/2}|M - q + r\rangle \\ &\equiv \prod_{t=1}^{r}(f_t)|M - q + r\rangle, \end{aligned} \tag{3.17}$$

$p(n, \tau)$ can be written as

$$\begin{aligned} p(n, \tau) = \exp(-\mathrm{i}\tau M\omega_1)\langle n|\langle M - n|\sum_{r=0}^{\infty}\sum_{q=0}^{\infty}\frac{(X_+)^r}{r!}(X_Z)^{M-q}\frac{(X_-)^q}{q!} \\ \times \prod_{s=1}^{q}(f_s)\prod_{t=1}^{r}(f_t)|M - q + r\rangle|0\rangle. \end{aligned} \tag{3.18}$$

Since x_+, x_- and x_z are functions only of the field operators, the summation over q

contains only one term, the one satisfying $q = n + r$. Therefore, $p(n, \tau)$ can be expressed as

$$p(n_1\tau) = \exp(-\mathrm{i}\omega_1 M\tau) \prod_{r=0}^{\infty} \left[\prod_{s=1}^{n+r} (f_s) \prod_{t=1}^{r} (f_t) \Big/ r!(n+r)! \right] A_{n0}, \tag{3.19}$$

where

$$A_{n0} \equiv \langle n|[\tan \tau(a+a^+)]^{2r+n}[\cos \tau(a+a^+)]^{2(n+r-M)}|0\rangle(-\mathrm{i})^{2r+n}. \tag{3.20}$$

To derive equation (3.19), $q = n + r$ and equation (3.18) were used. The calculation of the n-photon probability amplitude has been reduced to the computation of A_{no}:

$$A_{n0} = \langle n|[\sin \tau(a+a^+)]^{2r+n}[\cos \tau(a+a^+)]^{n-2M}|0\rangle(-\mathrm{i})^{2r+n}. \tag{3.21}$$

Expanding the sine and cosine functions in terms of exponentials, and then expanding the binomials, we obtain

$$A_{n0} = \frac{(-1)^n}{2^{2(r+n-M)}} \langle n| \sum_{s_1=0}^{2r+n} \sum_{s_2=0}^{n-2M} \binom{2r+n}{s_1} \binom{n-2M}{s_2} \exp[\alpha(a+a^+)](-1)^{2r-s_1}|0\rangle, \tag{3.22}$$

where

$$\alpha \equiv 2\mathrm{i}\tau(s_1 + s_2 + M - r - n).$$

According to the Baker–Campbell–Hausdorff (BCH) formula (Wilcox 1967) we write

$$\exp[\alpha(a+a^+)] = \exp[\alpha a^+]\exp[\alpha a]\exp[\alpha^2/2], \tag{3.23}$$

and a straightforward calculation leads to

$$\langle n|\exp[\alpha(a+a^+)]|0\rangle = \exp[\alpha^2/2](\alpha^n/(n!)^{1/2}). \tag{3.24}$$

Substituting equation (3.24) into equation (3.22) and A_{n0} in the n-photon probability amplitude (equation 3.20), we obtain

$$\begin{aligned} |p(n,\tau)|^2 = [4^{2M-n}/n!]\Bigg(& \sum_{r=0}^{r\,\mathrm{max}} \left[\prod_s (f_s) \prod_t (f_t) \Big/ 2^r r!(n+r)! \right] \\ & \times \sum_{s_1=0}^{2r+n} \sum_{s_2=0}^{n-2M} \binom{2r+n}{s_1} \binom{n-2M}{s_2} (-1)^{s_1} \tau^n (s_1+s_2+M-r-n)^n \\ & \times \exp[-2\tau^2(s_1+s_2+M-r-n)^2] \Bigg)^2 \Big/ \sum_{n=0}^{M+R} |p(n,\tau)|^2. \end{aligned} \tag{3.25}$$

Notice that in equation (3.25) we have replaced the ∞ for $r_{\max}$ in the upper limit of the summation over r. From the equation (3.16), we can see that $q_{\max} = M + R$, therefore $r_{\max} = M + R - n$. Since we expanded the cosine function inution (3.21), it was assumed that $n \geqslant 2M$. This inequalipy valid for all times only if $M \leqslant 0$. Our particular interest is $M = 0$, for which the n-photon probability becomes

$$\begin{aligned} |p(n,\tau)|^2 = \left(\frac{1}{n!16^n}\right) \Bigg\{ & \left[\sum_{r=0}^{R-n} \left(\prod_{s=1}^{n+r} (f_s) \right) \left(\prod_{t=1}^{r} f_t \right) \Big/ r!(n+r)!2^{2r} \right] \\ & \times \sum_{s_1=0}^{2r+n} \sum_{s_2=0}^{n} \binom{2r+n}{s_1} \binom{n}{s_2} (-1)^{s_1} \\ & \times (s_1+s_2-r-n)^n \tau^n \exp[-2\tau^2(s_1+s_2-r-n)^2] \Bigg\}^2 \Big/ \sum_{n=0}^{R} |p(n,\tau)|^2. \end{aligned} \tag{3.26}$$

The average number of photons, which is also the number of idler photons at a frequency $(\omega_1 - \omega_2)$, is

$$n(\tau) = n_3(\tau) = \sum_{n=0}^{R} n|p(n, \tau)|^2. \tag{3.27}$$

The stimulated emission case can be applied to the following physical situations:

(i) Parametric amplification, where a pump field at frequency ω_1 and with initial number of photons n_1 interacts with a signal field at frequency ω_2 with initial number of photons n_2 and an idler field at frequency $\omega_1 - \omega_2$ with initial number of photons zero.

(ii) Since photons and phonons obey the same commutation rules, the parametric process solved in this section can also describe the interaction of a pump photon (ω_1), a signal photon (ω_2) and an idler phonon $(\omega_1 - \omega_2)$. This process describes Brillouin scattering if the phonon is a pressure wave (acoustic branch) or Raman scattering if the phonon corresponds to internal molecular rotation or vibration (optical branch).

4. The frequency converter

As outlined in the Introduction, the frequency converter can be dealt with using the same Hamiltonian as for the parametric amplifier, provided field 2 is interpreted as the pump, field 3 as the signal and field 1 as the idler.

Assuming n_1 initial photons at the sum frequency $(n_1 \neq 0)$, we can write

$$M = n_3 + \tfrac{1}{2}(n_1 - n_2), \qquad R = \tfrac{1}{2}(n_1 + n_2), \tag{4.1}$$

which in terms of N two-level atoms, become

$$M \neq R, \qquad n(0) = n_3 \neq 0, \tag{4.2}$$

which corresponds to the general case solved in paper I. However, the accuracy of the solution is only acceptable if we are dealing with a reasonably large number of atoms, or more precisely, $M + R - n(0) \geqslant 3$. In the nonlinear optical language, this inequality can be written as $n_1 \geqslant 3$. If the latter condition is satisfied, then the general solution found in paper I (Orszag 1979a) can be readily used.

5. Other nonlinear processes

Besides the above nonlinear optical phenomena a number of effects can be dealt with by the present theory. Every one of these second-order nonlinear effects can be characterised by a set of constants n_1, n_2, n_3 or equivalently, in terms of the two-level system notation, by the set M, R, $n(0)$.

When dealing with photons mixing at various frequencies, as in the parametric amplification and frequency conversion cases, the role of the nonlinear material is a passive one in the sense that the crystal only acts as a suitable material where the interaction takes place.

There are other nonlinear phenomena in which the medium imposes characteristic frequencies when excited by a strong electromagnetic field. These characteristic frequencies are due to mechanical vibrations or rotations of molecules; the effect of this mechanical motion is modulation of the light wave. This is Raman scattering.

If the modulation is due to pressure waves or density changes in the medium (typically in a liquid like benzene), again interaction between a photon and a phonon takes place, producing shifts in the frequency of the outgoing light wave. This is Brillouin scattering.

We chose to study in this section the following effects: *1.* stimulated Raman scattering, *2.* inverse Raman effect, *3.* Brillouin effect and *4.* super-radiance. In all these effects the medium plays an active role. In the first three, the operator a_2 represents a phonon ratheruh a photon. In the fourth (super-radiance) the interaction takes place between the atoms and the radiation field.

5.1. Stimulated Raman scattering

The mixing of mechanical and electromagnetic vibrations produces upward and downward shifts in the frequency of the outgoing radiation (anti-Stokes and Stokes processes respectively).

(i) *Stokes Process*

This process is described in figure 6. Since we wish to describe stimulated Raman scattering, where the electromagnetic field excites a phonon, this case corresponds to the parameters $n_2 = n_3 = 0$, $n_1 \neq 0$ or $M = R = \frac{1}{2}n_1$, $n(0) = 0$. This case is formally identical to the case of spontaneous emission from a parametric amplifier, the only difference being that a_2 is the annihilation operator of a phonon instead of a photon. Therefore the results of § 3.1 can be applied here.

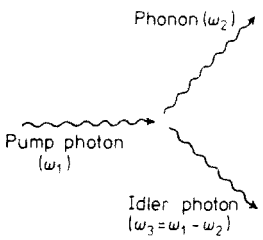

Figure 6. Stokes process in Raman effect. This process is similar to the parametric amplifier except that here the signal field is a phonon (molecular rotation or vibration) rather than a photon.

The anti-Stokes Raman process is similar to second harmonic generation. These two cases cannot be described accurately here. We shall shortly publish a solution for these two nonlinear effects.

5.2. Inverse Raman effect

The inverse Raman Stokes and anti-Stokes processes are shown in figures 7 and 8 respectively.

Let us assume that two light sources interact in a Raman active medium. If a pulsed laser and a CW laser are used, the medium displays a negative absorption or enhanced emission at the Stokes frequency $\omega_3 = \omega_1 - \omega_2$ (figure 7) and a reduced transmission at the anti-Stokes frequency $\omega_1 = \omega_2 + \omega_3$ (figure 8), having chosen a transparent medium at these frequencies (Baldwin 1974, Yariv 1975).

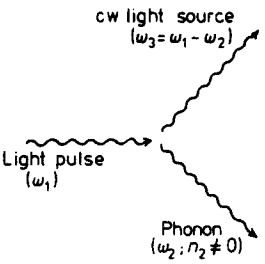

Figure 7. Inverse Raman effect, Stokes process. The CW light source at the Stokes frequency $\omega_3 = \omega_1 - \omega_2$ displays an enhanced emission when passing through a Raman medium, the pump being a pulsed laser at frequency ω_1. This is due to the presence of phonons (ω_2) in the material.

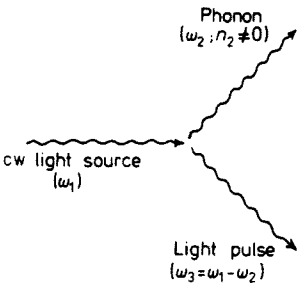

Figure 8. Inverse Raman effect, anti-Stokes process. The light source shows an increased absorption at the anti-Stokes frequency (ω_1) in a normally optical transparent Raman active medium. Phonons are present and the pulsed laser is at the frequency $\omega_3 = \omega_1 - \omega_2$.

The parameters characterising these two effects are $n_1 \neq 0$, $n_2 \neq 0$, $n_3 \neq 0$ or

$$R = \tfrac{1}{2}(n_1 + n_2), \qquad M = n_3 + \tfrac{1}{2}(n_1 - n_2), \qquad n(0) = n_3, \tag{5.1}$$

which means that in the two-level system language we are dealing with the most general case $M \neq R$, $n(0) \neq 0$. This case has been solved by the author in paper I (Orszag 1979a) for limited times.

5.3. Brillouin scattering

This differs from Raman scattering only in that we are dealing with phonons of the acoustic rather than the optical branch. A sound wave travels through the nonlinear material producing frequency shifts in the optical fields, these shifts being much smaller than Raman shifts. Although the coupling in Brillouin scagwng is with pressure waves rather than with the internal motions of molecules, the theoretical formalism is the same and the results found for Raman scattering are also applicable to Brillouin scattering.

5.4. Super-radiance

Dicke (1954) originally proposed that a collection of atoms can correlate themselves when suitably excited by an electromagnetic pulse (at optical frequency) and, using time dependent perturbation theory, found that they can emit a gigantic pulse proportional to the square of the number of atoms involved in the process.

Considerable effort has been devoted to the understanding of this effect since, although it is well known to the electrical engineer, the collective response of the atoms was surprising.

Experiments have been performed to prove the existence of optical super-radiance (Compaan and Abello 1971, Shoemaker and Brewer 1972, Skribanowitz *et al* 1973) but the issue has not yet been settled (see also Shen 1976). From the point of view of our formalism, the atomic system must be prepared initially in the super-radiant state, which corresponds to $M = 0$. The solution found in § 3.2 corresponds exactly to this choice of parameters ($R, M = 0, n(0) = 0$).

6. Discussion

We shall now discuss some of the present results and, in particular, compare them with previous work. For the case of spontaneous fluorescence, and assuming an intense pump that behaves classically, one obtains (Yariv 1975)

$$n_3(t) = \sinh^2(st/2) \tag{6.1}$$

where s in our notation is

$$s = 2K(n_1)^{0\cdot5}. \tag{6.2}$$

Obviously $n_3(t)$ cannot grow indefinitely, and equation (6.1) is valid for short times.

In table 1 a comparison between the results derived in § 3 and the approximate formula (6.1) is presented for times $\tau = 0\cdot1$ and $\tau = 0\cdot2$.

The table shows that the expression of equation (6.1) becomes worse as we increase either τ or R. Its accuracy is good if

$$(2R)^{0\cdot5}\tau \ll 1. \tag{6.3}$$

The general validity of the present theory will now be discussed.

Table 1. A numerical comparison between the present theory for spontaneous fluorescence and the results assuming that the pump behaves classically, for τ values 0·1 and 0·2. Agreement between the two theories is good provided $\tau(2R)^{1/2} \ll 1$. In the case $\tau(2R)^{1/2} > 1$, the semiclassical theory predicts an $n_3(\tau)$ which grows exponentially with time.

τ	R	$n_3(t) = \sinh^2[\tau(2R)^{1/2}]$	Results of § 3.1 for $n_3(\tau)$
0·1	0·5	0·0100	0·0099
	1·0	0·0197	0·0196
	1·5	0·0299	0·0291
	2·0	0·0405	0·0384
	4·5	0·09273	0·08121
	5·0	0·1024	0·08903
0·2	0·5	0·0405	0·0384
	1·0	0·0823	0·0739
	1·5	0·1239	0·1059
	2·0	0·1687	0·1339
	4·5	0·4053	0·2157
	5·0	0·4570	0·2232

From paper I (Orszag 1979a) we define the following operators:

$$\hat{\theta}_{\text{Exact}} = \exp\{(-\mathrm{i}\omega t)[\hat{N} + \alpha(a + a^{+})(R^{+} + R^{-})]\}$$
$$\hat{\theta}_{\text{RRWA}} = [\exp(-\mathrm{i}\omega t\hat{N})]\{\exp[(-\mathrm{i}\omega t)\alpha(a + a^{+})(R^{+} + R^{-})]\}$$
$$\hat{\theta}_{\text{RWA}} = [\exp(-\mathrm{i}\omega t\hat{N})][\exp(-\mathrm{i}\omega t)(aR^{+} + a^{+}R^{-})] \tag{6.4}$$

where $\alpha = K/\omega$.

We have also shown in paper I that in the case of the emission of one photon, we have the following relations:

$$\langle 1|\langle M-1|\hat{\theta}_{\text{Exact}}|M\rangle|0\rangle\, \mathrm{e}^{\mathrm{i}\omega tM} = \alpha C_1 + \alpha^3 C_{\text{Exact}} + \ldots$$
$$\langle 1|\langle M-1|\hat{\theta}_{\text{RRWA}}|\mathrm{M}\rangle|0\rangle\, \mathrm{e}^{\mathrm{i}\omega tM} = \alpha C_1 + \alpha^3 C_{RRWA} + \ldots$$
$$\langle 1|\langle M-1|\hat{\theta}_{\text{RWA}}|\mathrm{M}\rangle|0\rangle\, \mathrm{e}^{\mathrm{i}\omega tM} = \alpha C_1 + \alpha^3 C_{RWA} + \ldots, \tag{6.5}$$

where

$$C_1 = (-\mathrm{i}\omega t)[(R+M)(R-M+1)]^{1/2},$$
$$C_{\text{Exact}} = [\mathrm{i}(\omega t)^3/6]\langle M-1|\langle 1|(a+a^{+})^3(R^{+}+R^{-})^3|M\rangle|0\rangle,$$
$$C_{\text{RRWA}} = [\mathrm{i}(\omega t)^3/6]\langle M-1|\langle 1|(a+a^{+})^3(R^{+}+R^{-})^3|M\rangle|0\rangle,$$
$$C_{\text{RWA}} = [\mathrm{i}(\omega t)^3/6]\langle M-1|\langle 1|(aR^{+}+a^{+}R^{-})^3|M\rangle|0\rangle. \tag{6.6}$$

The equations (6.5) and (6.6) clearly indicate that the rotating-wave approximation model introduces an error with respect to the exact case of the order of $(\alpha)^3$, while the restricted rotating-wave approximation is exact in this order. In this work, the formal analogy between the Hamiltonians (2.4) and (2.5) is exploited. However, the results using the RRWA are used, thus introducing an error of the order of $(\alpha)^3$. This is clear from equation (6.7) if we simply compare C_{RRWA} and C_{RWA}. As we can see from the above discussion, this theory is only approximate, the accuracy of the approximation being comparable with the accuracy of the standard rotating-wave approximation when solving the problem of N atoms interacting via the electromagnetic field.

Finally, regarding the loss of unitarity of the time-evolution operator, we refer the reader to the discussion in paper I. The error is due to the use of the BCH identity when dealing with bound $|n\rangle$ states (bound spectrum) and this error is of the order of $(\alpha)^{M+R-n(0)}$ and it can be neglected in all the cases considered here. However, the cases of second-harmonic generation and the anti-Stokes Raman process correspond exactly to $M+R-n(0)=0$ and the present theory cannot be used. These two cases will be dealt with in a separate publication.

Acknowledgment

I should like to thank Mr Jaime Menendez for writing the several APL computer programs used in this work.

References

Arecchi F T, Courtens E, Gilmore R and Thomas H 1972 *Phys. Rev.* **A6** 2211
Armstrong J A, Bloembergen N, Ducuing J and Pershan P S 1962 *Phys. Rev.* **127** 1918

Baldwin G C 1974 *An Introduction to Nonlinear Optics* (New York: Plenum)
Bonifacio R and Masserini G 1968 *Phys. Lett.* **28A** 359
Bonifacio R and Preparata G 1970 *Phys. Rev.* **1** 446
Compaan A and Abello I D 1971 *Phys. Rev. Lett.* **27** 23
Dicke R H 1954 *Phys. Rev.* **93** 99
Franken P A, Hill A E, Peters C W and Weinreich G 1961 *Phys. Rev. Lett.* **7** 118
Graham R and Haken H 1968a *Z. Phys.* **210** 276
—— 1968b *Z. Phys.* **210** 319
Jaynes E T and Cummings F W 1963 *Proc. I.E.E.E.* **51** 89
Kaiser W and Garret C G B 1961 *Phys. Rev. Lett.* **7** 229
Louisell W H 1964 *Radiation and Noise in Quantum Electronics* (New York: McGraw–Hill)
Louisell W H, Yariv A and Siegman A E 1961 *Phys. Rev.* **124** 1646
Mollow B R and Glauber R J 1967 *Phys. Rev.* **160** 1077
Orszag M 1979a *J. Phys. A: Math. Gen.* **12** 2205
—— M 1979b *J. Phys. A: Math. Gen.* **12** 2225
Shen Y R 1975 *Raman Scattering in Solids* ed. M Cardona (Berlin: Springer)
—— 1976 *Rev. Mod. Phys.* **48** 1
Skribanowitz N, Herman I D, MacGillivray J C and Feld M S 1973 *Phys. Rev. Lett.* **30** 309
Shoemaker R L and Brewer R G 1972 *Phys. Rev. Lett.* **28** 1430
Tucker J and Walls D F 1969 *Ann. Phys.* **52** 1
Walls D F and Barakat R 1970 *Phys. Rev.* **1** 446
Wilcox R M 1967 *J. Math. Phys.* **8** 962
Yariv A 1975 *Quantum Electronics* (New York: Wiley)
Yariv A and Pearson J E 1969 *Progress in Quantum Electronics* vol 1 (Oxford: Pergamon)