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Nonlinear quantum effects in optics

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Abstract. Nonlinear optical effects where quantum mechanical predictions are at variance with the results of classical calculations are studied. Certain initial conditions, those of unstable equilibrium or those that lead to unstable equilibrium, classically, give rise to an aperiodicity in the intensity of the emitted radiation. One example of such a system is second harmonic generation, which is studied in detail because of the potential amenability of the aperiodicity to experimental observation. Numerical and analytic predictions for the evolution of the intensity and photon statistics of the second harmonic light are presented. The photon statistics of the second harmonic light are predicted to undergo a sharp transition following the first maximum of the intensity.

1. Introduction

Since the advent of the laser, which is itself a nonlinear device, nonlinear optical effects have been the subject of considerable research. Such effects arise, for example, in the emission of radiation from atoms where cooperative spontaneous emission, or superradiance has been a particularly popular topic. Similar effects occur in nonlinear optics, for example parametric amplification, frequency conversion and second harmonic generation (SHG).

These phenomena have been analysed theoretically by a variety of techniques. Perhaps the most popular approaches have been classical or semiclassical analyses owing to the inherent difficulty in solving nonlinear quantum problems by other than perturbative methods. In predicting the evolution of the intensity of the emitted radiation classical methods have for the most part enjoyed a considerable degree of success. The extremely large number of photons typically present in laser experiments validates a classical treatment of the electromagnetic field in a large number of phenomena. Such treatments, however, break down in situations where classically the initial conditions are those of unstable equilibrium. This occurs for example for a system of N atoms initially all excited with no photons present. Classically this system does not radiate. Quantum mechanically the atoms decay by spontaneous emission. Classical theorists overcame this deficiency by considering a very small displacement from the position of unstable equilibrium to simulate the onset of the radiation by spontaneous emission.

Recently, evidence has been presented that for systems which classically are initially in a state of unstable equilibrium the quantum effects play a greater role than merely initiating the emission of quanta. Numerical analyses (Abate and Haken 1964, Walls and Barakat 1970) for the intensity of radiation emitted by N excited atoms have shown an aperiodicity not previously suspected. Arguments linking this aperiodicity to the quantum mechanical uncertainty principle have been advanced (Senitzky 1970, 1971).

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Experimental observation of this aperiodicity however, presents problems. The difficulty lies in creating initial conditions with all the atoms excited and no photons present. In this paper we wish to investigate further examples where anomalous quantum effects leading to aperiodicity are prevalent and which are perhaps more easily accessible to experimental observation.

In the next section we introduce a phenomenological Hamiltonian which has relevance to a number of nonlinear optical phenomena. In § 3 a detailed quantum analysis is made of second harmonic generation, which is found to display anomalous quantum effects. Preliminary results of such calculations have been reported in an earlier letter (Walls and Tindle 1971). Our interest in SHG is enhanced by the possibility that the anomalous effects predicted are amenable to experimental observation. In § 4 results of analytic and numerical calculations on the photon statistics of the second harmonic (SH) light are presented. In the final section we draw some general conclusions.

2. General considerations in nonlinear optics

Consider the following Hamiltonian describing the interaction of three coupled boson field modes:

$$H = \hbar\omega_1 a_1^\dagger a_1 + \hbar\omega_2 a_2^\dagger a_2 + \hbar\omega_3 a_3^\dagger a_3 + \hbar\kappa(a_1 a_3 a_2^\dagger + a_1^\dagger a_3^\dagger a_2) \tag{2.1}$$

where the a_j are boson annihilation operators and κ is the coupling constant. This Hamiltonian may describe for example, the process of parametric amplification where a pump photon with frequency ω_2 and a signal photon with frequency ω_1 interact to produce an idler photon at the difference frequency $\omega_3 = \omega_2 - \omega_1$. Alternatively it may describe frequency up conversion where a pump photon of frequency ω_3 and a signal photon with frequency ω_1 interact to produce an idler photon at the sum frequency $\omega_2 = \omega_1 + \omega_3$. We note that this is an idealized model owing to the restriction to monochromatic modes and the complete absence of loss terms.

The Hamiltonian describing the interaction of N two level atoms with a single mode of the radiation field may be written (neglecting nonresonant terms) as

$$H = \hbar\omega_3 a_3^\dagger a_3 + \hbar\omega_3 \sum_{j=1}^{N} \sigma_z^j + \hbar\kappa \sum_{j=1}^{N} (a_3 \sigma_j^+ + a_3^\dagger \sigma_j^-) \tag{2.2}$$

where a_3 is a boson annihilation operator; σ_j^+, σ_j^- and σ_z^j are the raising, lowering and inversion operators for the jth atom.

It is convenient to introduce the collective operators

$$\begin{aligned} J_+ &= \sum_{j=1}^{N} \sigma_j^+ \\ J_- &= \sum_{j=1}^{N} \sigma_j^- \\ J_z &= \sum_{j=1}^{N} \sigma_z^j. \end{aligned} \tag{2.3}$$

The phenomena described by the Hamiltonians (2.1) and (2.2) may be formally related using the Schwinger representation for angular momentum (Schwinger 1965)

$$\begin{aligned} J_+ &= a_1 a_2^\dagger \\ J_- &= a_1^\dagger a_2 \\ J_z &= \tfrac{1}{2}(a_2^\dagger a_2 - a_1^\dagger a_1). \end{aligned} \tag{2.4}$$

The operators a_1 and a_2 obey boson commutation relations, however the eigenvalues of $a_1^\dagger a_1$ and $a_2^\dagger a_2$ span the reduced integer spectrum from 0 to N_c, where N_c is the number of atoms acting in cooperation. With the transformation (2.4) the Hamiltonians (2.1) and (2.2) are formally identical. The eigenvalues n_1 and n_2 of $a_1^\dagger a_1$ and $a_2^\dagger a_2$ represent the occupation numbers of the lower and upper atomic levels with $n_1 + n_2 = N$, provided $N_c = N$.

The numerical (Abate and Haken 1964, Walls and Barakat 1970, Walls 1970a) and analytic solutions (Bonifacio and Preparata 1970) found in the literature reveal that the properties of the emitted radiation vary considerably according to the initial conditions. The initial conditions $n_1 = n_3 = 0$, correspond to spontaneous emission from N excited atoms, or to the spontaneous parametric decay of a pump photon into a signal and idler photon. Classically, of course, this initial condition corresponds to an unstable equilibrium and no radiation takes place.

A number of solutions of the Hamiltonians (equations (2.1), (2.2)) for such initial conditions have already been given. For a macroscopically large number of atoms an approximate analysis given by Bonifacio and Preparata (1970) yields an elliptic function behaviour for the intensity of the emitted photons. Numerical calculations (Abate and Haken 1964, Walls and Barakat 1970) show an aperiodic behaviour at variance with the elliptic function predictions. The variance of the photon distribution of the emitted light has been shown (Walls and Barakat 1970, Bonifacio and Preparata (1970) to increase as the mean number of photons squared, characteristic of a Bose–Einstein distribution. This feature is reflected in the probability distribution of the atomic energy. Thus as has been pointed out by Walls and Barakat (1970) it is not possible to form a superradiant state (characterized by $n_1 = n_2 = N/2$, (Dicke 1954)) from an initial state with all atoms excited. This is in agreement with the analytic results of Bonifacio and Gronchi (1971) who show that the variance of the atomic energy increases as N^2 as the system approaches a state in which $N/2$ atoms are in the ground level. This however disagrees with the results of Eberly and Rehler (1970), and Argarwal (1970) who found that the variance of the atomic energy under the same conditions is proportional to N.

For initial conditions corresponding to a superradiant state classical and quantum mechanical predictions are in agreement. The system radiates a train of pulses whose shape closely resembles the square of a sine function. The statistics of the emitted photons have been shown to a good approximation to be poissonian (Walls and Barakat 1970, Bonifacio and Preparata 1970) characteristic of the distribution of photons in a coherent state (Glauber 1963a, 1963b).

The fundamental difference between the two sets of initial conditions may be understood in the following manner. Radiation from a system of atoms all excited occurs by spontaneous emission. Spontaneous emission may be considered as the amplification of the vacuum fluctuations, an essentially chaotic process. The presence of atoms in the ground state gives rise to a macroscopic transverse dipole which causes the atoms to begin radiating in a coherent or 'classical' manner. A comparison with the case of three interacting electromagnetic field modes (equation (2.1)) reveals that the presence of atoms in the ground state corresponds to an input signal giving rise to stimulated emission.

The same basic principles occurring in the emission of radiation from atoms arise in certain nonlinear optical phenomena. Parametric frequency up conversion is equivalent to exciting N atoms with a pulse of radiation. Numerical solutions for the intensity of the up converted radiation were given by Walls and Barakat (1970). The intensity of up converted photons follows closely a sine squared behaviour when the number of pump photons greatly exceeds the number of signal photons. The statistics of the emitted photons was shown to a good approximation to be poissonian. From this we may conclude that frequency up conversion under these conditions is essentially a coherent process. Atomic excitation by a $\pi/2$ pulse corresponds to the case where the number of photons in the exciting pulse is equal to half the total number of atoms ($n_3 = n_1/2$). As Walls and Barakat (1970) have pointed out this is a suitable method to form a superradiant state owing to the coherent nature of the process.

However as the number of pump and signal photons become comparable ($n_3 \simeq n_1$) one observes a deviation from the sinusoidal behaviour. This deviation becomes most apparent for the degenerate case ($n_3 = n_1$) where a marked aperiodicity occurs. We observe that this degenerate case is an initial condition which classically would lead to a position of unstable equilibrium. A similar effect occurs in the excitation of an ensemble of atoms with a π pulse. Once excited the atomic system must decay by spontaneous emission resulting in the anomalous quantum effects discussed previously.

These examples correspond to the second solution of the double root found by Senitzky (1971) in seeking solutions which yield results significantly different from classical solutions. Initial conditions corresponding to unstable equilibrium classically have been intensively studied since they represent the initial conditions for the onset of idealized maser and laser action. However it has passed unnoticed that initial conditions which classically lead to unstable equilibrium occur in the most widely studied phenomena in nonlinear optics; second harmonic generation. In view of this we consider the process of SHG in some detail below.

3. Second harmonic generation

SHG is the degenerate case of frequency up conversion where the annihilation operators for the pump and signal modes are identical. It follows from equation (2.1) that SHG may be described by the following Hamiltonian:

$$H = \hbar\omega a_1^\dagger a_1 + \hbar 2\omega a_2^\dagger a_2 + \hbar\kappa(a_1 a_1 a_2^\dagger + a_1^\dagger a_1^\dagger a_2). \tag{3.1}$$

A classical analysis of SHG (Armstrong *et al* 1962) for perfect phase matching predicts that the intensity of SH photons from the vacuum will grow as

$$\bar{n}_2(\tau) = \frac{n_1}{2}\tanh^2(\sqrt{n_1}\,\tau) \tag{3.2}$$

where n_1 is the initial number of fundamental photons and $\tau = \kappa t$. This predicts the number of SH photons to grow to a maximum equal to $n_1/2$ and remain at this maximum for all times. That this is a position of unstable equilibrium may be verified by introducing a small displacement from the equilibrium solution into the classical equations of motion. In this way one simulates the effects of spontaneous emission by which the SH field may recreate the fundamental field.

We now outline a quantum mechanical approach to the problem. We consider an initial state $|n_1, 0\rangle$ with n_1 fundamental photons and no SH photons present. The probability amplitude to have n_2 SH photons present at time t is

$$a_{n_2}(t) = \langle n_1 - 2n_2, n_2| \exp(-\mathrm{i}Ht/\hbar)|n_1, 0\rangle. \tag{3.3}$$

On differentiating this expression with respect to time we arrive at the differential equation

$$\frac{\mathrm{i}\mathrm{d}a_{n_2}(\tau)}{\mathrm{d}\tau} = \{(n_1 - 2n_2)(n_1 - 2n_2 - 1)(n_2 + 1)\}^{1/2} a_{n_2+1}(\tau) + \{(n_1 - 2n_2 + 1)(n_1 - 2n_2 + 2)n_2\}^{1/2} a_{n_2-1}(\tau) \tag{3.4}$$

where $\tau = \kappa\tau$. For a macroscopically large number of fundamental photons an approximate solution to this equation may be derived using the method developed by Bonifacio and Preparata (1970). This solution predicts the intensity of SH photons to oscillate as an elliptic function (Walls 1970b)

$$\bar{n}_2(\tau) = \frac{n_1}{2}(1 - cn^2(\sqrt{n_1}\tau, k)) \tag{3.5}$$

where $k = n_1/(n_1+1)$ is the elliptic parameter of the jacobian elliptic function *cn*. The period of the oscillations is $\ln n_1/\sqrt{n_1}$.

Though this result includes the recreation of the fundamental by spontaneous emission it appears to be an approximation to the exact result. Equation (3.4) has been solved numerically and the resulting behaviour for $\bar{n}_2(\tau)$, the mean number of SH photons as a function of time, has been plotted in figure 1 for the initial conditions $n_1 = 200$, 199; $n_2 = 0$. One observes aperiodic behaviour at variance with the elliptic function

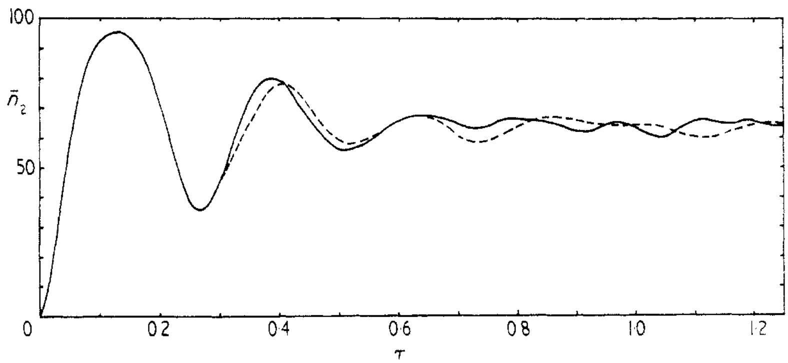

Figure 1. Mean number $\bar{n}_2$ of SH photons as a function of normalized time τ for initial conditions $n_1 = 200$, $n_2 = 0$ (full curve); $n_1 = 199$, $n_2 = 0$ (broken curve).

prediction. This aperiodicity may be expected as a consequence of the initial conditions inherent in SHG which lead classically to a state of unstable equilibrium from which the SH photons may only decay by spontaneous emission. Strictly speaking the position of unstable equilibrium is only attained classically for an initial even number of fundamental photons. However we see from figure 1 that the aperiodicity is also present for an initial odd number of fundamental photons. This distinction does not arise in actual experiments with lasers where the total number of photons is ill defined and is replaced by a probability distribution about a mean number. In any event spontaneous emission back into the fundamental mode always prevents the full conversion of the fundamental to SH. Thus of 200 fundamental photons only 190 are converted to yield a maximum of 95 SH photons.

Mathematically the aperiodicity arises in the following manner. The mean number of SH photons may be written (Walls and Barakat 1970) in the form

$$\bar{n}_2(\tau) = \sum_{\lambda\lambda'} a_{\lambda\lambda'} \exp\{i(\lambda-\lambda')\tau\} \tag{3.6}$$

where λ, λ' are the eigenvalues of the system and the $a_{\lambda\lambda'}$ are coefficients related to the eigenfunctions. If the eigenvalues were spaced equidistantly a periodic behaviour of $\bar{n}_2(\tau)$ would necessarily follow. Various calculations of the eigenvalues for systems which classically are initially in unstable equilibrium (Tavis and Cummings 1968, Mallory 1969, Walls and Barakat 1970, Scharf 1970) yield a nonequidistant spacing. In figure 2 we display the eigenvalues for SHG for the initial conditions $n_1 = 200, n_2 = 0$. It is seen that the eigenvalues are not spaced equidistantly, hence an aperiodicity in the evolution of the intensity results.

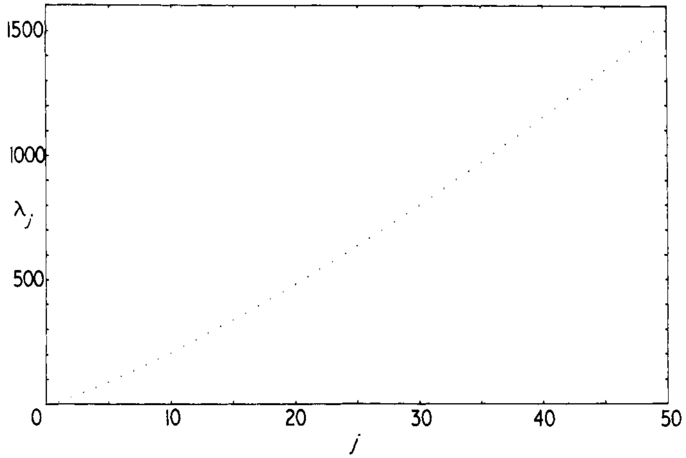

Figure 2. Eigenvalues (λ_j) for initial conditions $n_1 = 200$, $n_2 = 0$. Negative eigenvalues not shown since $\lambda_{-j} = -\lambda_j$.

A quasiperiod for the oscillations may be derived by considering the eigenvalue problem utilizing the method developed by Scharf (1970). For large n_1 it may be shown that the eigenvalues are spaced approximately equidistantly yielding a quasiperiod equal to $\ln n_1/\sqrt{n_1}$. This is in agreement with the period extracted from the elliptic function solution (equation (3.5)). However it is apparent from figure 1 that this quasiperiod only has any real meaning for the first two oscillations.

It may be surmised that the aperiodicity arises as a consequence of the small photon number involved in these numerical calculations and that as the photon number becomes macroscopically large a periodic, classical type behaviour will ensue. However, solutions of equation (3.4) for $10 < n_1 \leqslant 200$ show no change in the general shape of the $\bar{n}_2(\tau)/\tau$ curve. Further, Senitzky (1970, 1971) in studying the N atom problem has argued that the aperiodicity is a consequence of the quantum mechanical uncertainty principle and will persist even for macroscopically large quantum numbers. Senitzky bases his arguments on the observation that for initial conditions near those of unstable equilibrium, the period is very sensitive to slight differences in the initial conditions. In taking expectation values one averages over the quantum fluctuations; that is, an ensemble average is taken over a range of periods thereby resulting in aperiodicity.

To date, however, no evidence of this aperiodicity has been detected. The experimental difficulties involved in preparing an excited ensemble of atoms with no photons present are considerable. The initial presence of photons or unexcited atoms results in a considerably more periodic behaviour (Walls and Barakat 1970). SHG, however offers much better hope of observing the aperiodicity since the initial condition of no SH photons present is exactly what one gets with a pulsed laser. For this reason we feel that SHG is the best example to illustrate anomalous nonlinear quantum effects.

The effect of having some SH photons initially present has also been examined. In figure 3 we have plotted $\bar{n}_2(\tau)/\tau$ for the initial conditions $n_1 = 50$ and $n_2 = 0, 1, 10$. It is seen that while the presence of a small number of SH photons initially increases the periodicity, this effect is not enhanced by having a larger initial number of SH photons present.

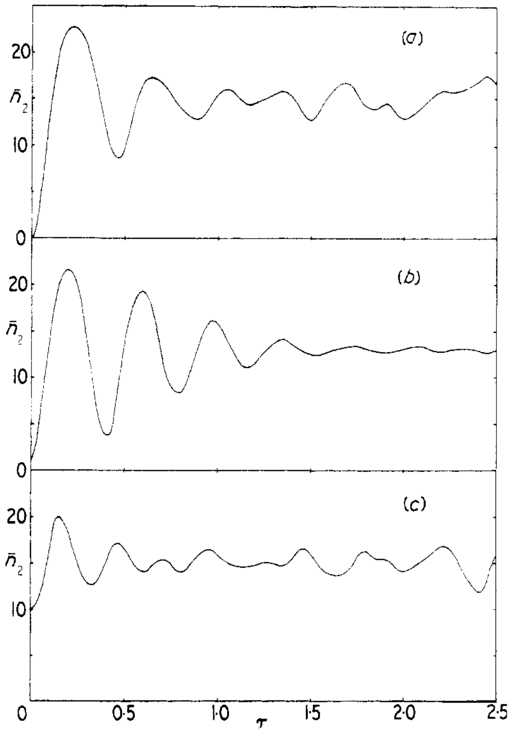

Figure 3. Mean number of SH photons as a function of τ for initial conditions (*a*) $n_1 = 50$, $n_2 = 0$; (*b*) $n_1 = 50$, $n_2 = 1$; (*c*) $n_1 = 50$, $n_2 = 10$.

The preceding work has all been done in the number state representation for the fundamental and SH modes since the computations are simpler in this representation. However a number state does not provide a good representation for a laser beam.

It has been shown by Glauber (1963a, 1963b) that a laser beam may be represented to a good approximation by a coherent state. To approximately simulate a coherent state we consider an input beam with a poissonian distribution of photons (this does not exactly simulate a coherent state since all phase information is lost).

In figure 4 we have plotted $\bar{n}_2(\tau)/\tau$ for an initial poissonian input of photons with mean number $\bar{n}_1 = 50$. Upon comparison with figure 3(*a*) ($n_1 = 50$) we note that the aperiodicity persists while a smoothing of the random fluctuations is apparent.

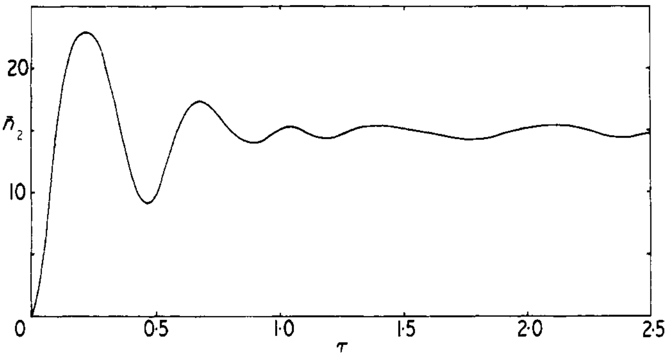

Figure 4. Mean number of SH photons as a function of τ for an initial poissonian distribution of fundamental photons with $\bar{n}_1 = 50$, and $n_2 = 0$.

4. Photon statistics of the SH light

As previously mentioned, the statistics of the photons emitted in the radiation process are strongly dependent on the initial conditions of the system. The photon statistics contain information relating to the degree of coherence inherent in the process and we shall now investigate them in some detail.

We consider first the photon statistics produced from an initial number state of fundamental photons $|n_1\rangle$ with no SH photons present. For short times such that the only coefficients $a_{n_2}(\tau)$ appreciably different from zero are the ones for which $n_2 \ll n_1$ we may replace equation (3.4) by the approximate equation

$$i\dot{a}_{n_2}(\tau) = n_1\{n_2^{1/2}a_{n_2-1}(\tau)+(n_2+1)^{1/2}a_{n_2+1}(\tau)\}. \tag{4.1}$$

This equation may be readily solved yielding $Pn_2(\tau)$ the probability of having n_2 SH photons at time τ

$$Pn_2(\tau) = |a_{n_2}(\tau)|^2 = \exp(-\bar{n}_2(\tau))\frac{\bar{n}_2(\tau)^{n_2}}{n_2!} \tag{4.2}$$

where $\bar{n}_2(\tau) = n_1^2\tau^2$. This solution represents a good approximation to the exact solution for times $\tau \ll \tau_{\max_1} \simeq \frac{1}{2}\ln n_1/n_1^{1/2}$ so that $\bar{n}_2 \ll n_1$. Thus the probability distribution of the SH photons has been shown to be poissonian for times $\tau \ll \frac{1}{2}\ln n_1/n_1^{1/2}$.

The time evolution of the SH photon distribution for $n_1 = 200$ is indicated in figure 5. As the mean number of SH photons grows up to the first maximum (see figure 1) the SH photons show a smooth distribution with a variance which is approximately

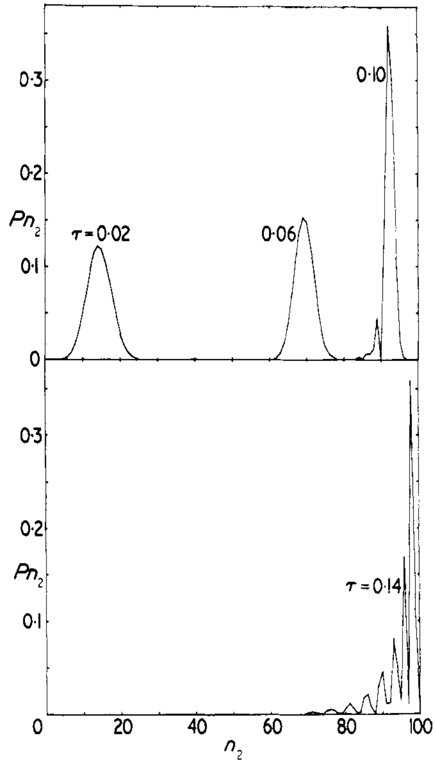

Figure 5. Evolution of the probability distribution of SH photons for initial conditions $n_1 = 200, n_2 = 0$. The distributions are discrete the points having been joined for clarity.

equal to $\bar{n}_2(\tau)$, characteristic of a Poisson distribution. This is in agreement with the analytic result derived above. As the peak is reached spontaneous emission becomes significant. This is a chaotic process and the smoothness of the distribution is lost. Beyond the first maximum the distribution becomes spread out and more chaotic as time progresses. For later times the distribution also shows a distinction between odd and even numbers of SH photons. This hereto unexplained effect was also observed in the emission of photons from an ensemble of excited atoms (Walls and Barakat 1970).

It is of greater interest to consider as before a coherent fundamental mode since this provides a good representation for the input of a laser beam. We consider therefore an initial density operator

$$\rho(0) = |\alpha_1, 0\rangle\langle\alpha_1, 0| \tag{4.3}$$

where the fundamental mode is in a coherent state $|\alpha_1\rangle$ and the SH mode is in the vacuum.

We shall present a derivation of the density operator for the SH mode at time t. We begin with the Heisenberg equations of motion for the operators $a_1(t)$, $a_2(t)$ derived from the Hamiltonian (equation (3.1)). With the substitutions

$$\begin{aligned} a_1(t) &= A_1(t)\exp(-\mathrm{i}\omega t) \\ a_2(t) &= A_2(t)\exp(-2\mathrm{i}\omega t) \end{aligned} \tag{4.4}$$

the Heisenberg equations of motion may be written in the form

$$\begin{aligned} \dot{A}_1(t) &= 2\mathrm{i}\kappa A_1^\dagger(t)A_2(t) \\ \dot{A}_2(t) &= \mathrm{i}\kappa A_1(t)A_1(t). \end{aligned} \tag{4.5}$$

A second differentiation of $\dot{A}_2(t)$ yields

$$\ddot{A}_2(t) + 2\kappa^2(2A_1^\dagger(t)A_1(t)+1)A_2(t) = 0. \tag{4.6}$$

From the above equations it follows that:

$$\begin{aligned} A_2(0) &= a_2(0) \\ \dot{A}_2(0) &= \mathrm{i}\kappa a_1(0)a_1(0) \\ \ddot{A}_2(0) &= -2\kappa^2(2a_1^\dagger(0)a_1(0)+1)a_2(0). \end{aligned} \tag{4.7}$$

The solution for $A_2(t)$ may now be expanded in a Taylor series

$$A_2(t) = a_2(0) + \mathrm{i}\kappa t a_1(0)a_1(0) - (\kappa t)^2(2a_1^\dagger(0)a_1(0)+1)a_2(0) \tag{4.8}$$

where we have included terms up to second order in t.

The P representation for the SH mode at time t as defined by Glauber (1965) is

$$P(\alpha_2, t) = \frac{1}{\pi^2}\int \mathrm{d}^2\eta \exp(\alpha_2\eta^* - \alpha_2^*\eta)\ \mathrm{Tr}[\rho(0)\exp(\eta a_2^\dagger(t) - \eta^* a_2(t))]. \tag{4.9}$$

Substituting equations (4.3), (4.4) and (4.8) into equation (4.9) we obtain

$$\begin{aligned} P(\alpha_2, t) &= \frac{1}{\pi^2}\int \mathrm{d}^2\eta \exp[\eta^*\{\alpha_2 - \mathrm{i}\kappa t\exp(-2\mathrm{i}\omega t)\alpha_1^2\}] \\ &\qquad \times \exp[-\eta\{\alpha_2^* + \mathrm{i}\kappa t\exp(2\mathrm{i}\omega t)\alpha_1^{*2}\}] \\ &= \delta^2\{\alpha_2 - \mathrm{i}\kappa t\alpha_1^2\exp(-2\mathrm{i}\omega t)\}. \end{aligned} \tag{4.10}$$

Thus the density operator at time t for the SH field correct up to second order in t is

$$\rho_2(t) = |\alpha_2(t)\rangle\langle\alpha_2(t)| \tag{4.11}$$

where

$$\alpha_2(t) = \mathrm{i}\kappa t\alpha_1^2\exp(-2\mathrm{i}\omega t).$$

Thus to a good approximation the SH light is initially produced in a coherent state.

To simulate the input of a coherent state numerically we consider an input of photons with a Poisson distribution. The evolution of the photon distribution of the SH light with time is plotted in figure 6 for a poissonian input with mean number $\bar{n}_1 = 50$.

The character of the distribution is determined by plotting $\sigma/\sqrt{\bar{n}_2}$ against τ (figure 7) where σ is the standard deviation. A value close to unity then indicates a Poisson distribution. The statistics of the SH photons are seen to be approximately poissonian for times out to the first maximum of the intensity ($\tau_{\max_1}$). This is in agreement with the analytic result derived above. Thus SHG approximately preserves coherence at least over interaction times attainable by present experiments. This has been verified experimentally by Clark *et al* (1970) who find that the process of SHG causes an enhancement by a factor of four of the small fundamental fluctuations while still approximately preserving the shape of the input Poisson distribution.

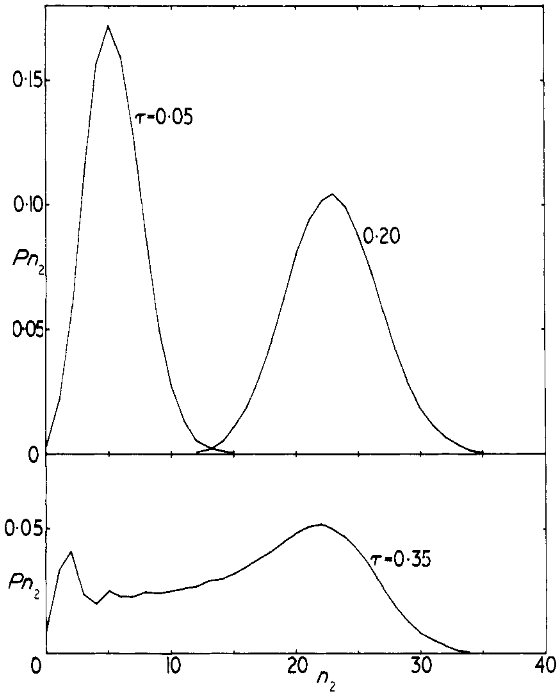

Figure 6. Evolution of the probability distribution of SH photons for an initial poissonian distribution with $\bar{n}_1 = 50$, $n_2 = 0$.

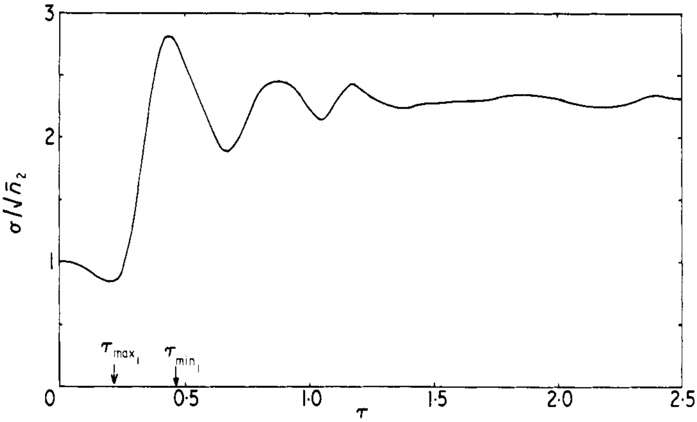

Figure 7. $\sigma/\sqrt{\bar{n}_2}$ (where σ is the standard deviation of the photon distribution of the SH light) as a function of τ for an initial poissonian input distribution with $\bar{n}_1 = 50$, $n_2 = 0$.

However as τ exceeds τ_{max_1} the statistics of the SH light undergo a sudden change. This is vividly illustrated in figure 7 where at τ_{max_1} the standard deviation σ is no longer equal to $\sqrt{\bar{n}_2}$ and the photon distributions (figure 6) which were initially poissonian in shape flatten out.

An understanding of this phenomenon may be reached by observing that at $\tau = \tau_{max_1}$ the spontaneous emission of SH photons back into the fundamental mode becomes the dominant effect. This is as previously noted a chaotic process. Thus one has a sharp transition between a coherent process, the creation of SH photons, and a chaotic process, the spontaneous decay into fundamental photons. This transition should be accessible to experimental observation providing the efficiency of existing SHG experiments is increased to reach times greater than $\tau_{\max_1}$.

5. Conclusions

A review has been made of a number of radiating systems under a variety of initial conditions. For systems which classically would be initially either in a state of unstable equilibrium, or in a state which would lead to unstable equilibrium, numerical calculations predict an aperiodicity in the intensity of the emitted radiation. An ensemble of excited atoms with no photons present initially is a well known example of the first situation. This however is a difficult state to prepare and thus experimental observation of the predicted aperiodicity would be difficult. An example of the second situation is provided by SHG. The predicted aperiodicity in SHG is more readily accessible to experimental observation since the initial conditions may be readily satisfied with the use of a pulsed laser.

The photon distribution of the SH light, which initially has a variance equal to the mean number of SH photons characteristic of a Poisson distribution, assumes a chaotic character upon reaching the first maximum of the intensity. This effect occurs as the result of the spontaneous decay of the SH back into the fundamental mode, an essentially chaotic process. This transition should be accessible to experimental observation with an efficient SHG experiment capable of attaining interaction times greater than $\tau_{\max_1}$.

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