Revista Brasileira de Física, Volume 13, nº 1, 1983

Dynamics of Third Harmonic Generation

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Recebido em 23 de novembro de 1981

Abstract The build-up of the third harmonic and the depletion of the pump mode are investigated describing both the fields quantum mechanically. The Heisenberg equations of motion describing the dynamics of the pump and of the third harmonic mode are solved in the short-time approximation. It is found that the pump mode shows antibunching effect due to the nonlinear interaction between the modes.

1. INTRODUCTION

Third harmonic generation has been studied both theoretically as well as experimentally by several workers[1-11] in the field of nonlinear optics. The third harmonic is being used to produce ultraviolet radiation not available from existing lasers by nonlinear optical mixing. The initial build-up of the third harmonic has been studied theoretically, in general, using the parametric approximation i.e. considering the pump depletion to be negligible. It is a reasonable approximation so long as the conversion efficiency is quite low, but recent experiments have shown increased improvement in the conversion efficiency[11]. Under these circumstances, it is necessary to include pump depletion while studying the harmonic generation. The present communication aims at investigating the dynamics of the third harmonic generation including pump depletion and treating both the pump and the harmonic fields quantum mechanically. The model Hamiltonian of the system is written along the line of Shen[12] and Walls[13]. The Heisenberg equations of motion describing the time evolution of the pump and the harmonic mode are solved in the short-time approximation[14]. Our analysis shows that as a result of nonlinear interaction, the pump photons manifest antibunching, if the pump mode is in the coherent state and the harmonic mode in the vacuum state initially. The photons generated in the harmonic mode are uncorrelated for the quoted initial conditions.

2. MODEL HAMILTONIAN

In the present work, we model our Hamiltonian along the lines of Shen[12] and Walls[13]. It is the simplest and adequate to describe the third harmonic generation. Such a Hamiltonian is

$$H = H_0 + H_1 \tag{1}$$

$$H_0 = \hbar\omega_a a^{+}(t)a(t) + \hbar\omega_b b^{+}(t)b(t) \tag{2}$$

$$H_1 = \hbar k\left[a^3(t)b^{+}(t) + a^{+3}(t)b(t)\right] \tag{3}$$

where H_0 is the free Hamiltonian of the pumpand of the third harmonic field. The operators $a^{+}(t)$, $a(t)$ and $b^{+}(t)$, $b(t)$ are the creation and destruction operators of the pump and of the third harmonic modes respectively. The H_1 represents interaction between the two modes. The expression for the coupling constant k depends also on the integral

$$\int e^{i(\vec{k}_a-\vec{k}_b)\vec{r}}\, d\vec{r}$$

where k_a and k_b are the wave vectors of the pump and of the third harmonic modes respectively. We assume that we have a phase-matched situation, i.e. $k_a(\omega) = k_b(3\omega)$. The creation and destruction operators of both the fields satisfy the following commutation relations:

$$[a,a^{+}] = [b,b^{+}] = 1 \tag{4}$$

$$[a, b^{+}] = [a^{+}, b^{+}] = [a, b] = 0 \tag{5}$$

3. EQUATIONS OF MOTION

The Heisenberg equation of motion for an operator $O(t)$ is given

$$i\hbar \frac{d}{dt} = [O, H] \tag{6}$$

Using equations (1) and (6), we obtain the following coupled equations

$$i\frac{da(t)}{dt} = \omega_a\, a(t) + 3k\, a^{+2}(t)b(t) \tag{7}$$

$$i\frac{db(t)}{dt} = \omega_b\, b(t) + ka^3(t) \tag{8}$$

for the operators $a(t)$ and $b(t)$ respectively. If we denote the number operators $a^+(t)\, a(t)$ and $b^+(t)\, b(t)$ by $N_a(t)$ and $N_b(t)$, we readily find that

$$\frac{d}{dt}\left[N_a(t) + 3N_b(t)\right] = 0 \tag{9}$$

Thus $N_a(t) + 3N_b(t)$ is a constant of motion. It implies the conservation of the photon number in the system. The equations (7) and (8) are coupled nonlinear equations for the two modes. In order to solve them, we recourse to the short-time approximation used by Agarwal and Mehta[14] to study the dynamics of the parametric processes. Using their tecnique, we obtain

$$a(t) = \left[a-3ikta^{+2}b+3/2k^2t^2\{6(a^+a+1)b^+b-a^{+2}a^2\}a\right]e^{-i\omega_a t}, \tag{10}$$

$$b(t) = \left[b-ikta^3-3/2k^2t^2(3a^{+2}a^2+6a^+a+2)b\right]e^{-i\omega_b t}, \tag{11}$$

as solution of equations (7) and (8) respectively correct up to second order in t. In eqs. (10) and (11) a and b are the initial time values of the operators.

4. CHARACTERISTIC FUNCTION

To study the build-up of the third harmonic and depletion of the pump field as a function of time, we employ the normally ordered characteristic function defined by the relation[15-16]

$$\chi(\eta,t) \equiv \mathrm{Tr}(\rho(0)e^{\eta c^+(t)}\, e^{-\eta^* c(t)}) \tag{12}$$

where $\rho(0)$ is the initial density operator for the boson fields involved. The operator $c(t)$ corresponds to either the pump or the harmonic field. Here in the present study, it is assumed that the pump mode is in a coherent state and the harmonic mode in the vacuum state initially, so the density operator is given as

$$\rho(0) = |\alpha_0, 0\rangle \langle\alpha_0, 0| \tag{13}$$

at the time $t = 0$. The state $|\alpha_0\rangle$ is the normalized eigenstate of the destruction operator a with complex eigenvalue α_0. The characteristic function $\chi(\eta, t)$ contains all the statistical information regarding the temporal behaviour of the mode involved.

5. TIME EVOLUTION OF THIRD HARMONIC

The normally ordered characteristic function for the third harmonic mode is

$$\chi_b(\eta, t) = \mathrm{Tr}\{\rho(0) e^{\eta b^+(t)} e^{-\eta^* b(t)}\} \tag{14}$$

Using equation (11) and expression for $\rho(0)$ from Eq. (13), the characteristic function $\chi_b(\eta, t)$ is evaluated, retaining terms up to second order in kt. It turns out that

$$\chi_b(\eta, t) = e^{\eta L^+ - \eta^* L} \tag{15}$$

where

$$L = -ikt\, \alpha_0^3\, e^{-i\omega_b t}$$

The average number of third harmonic photons present at time t can be obtained from the following relation[15]

$$N_b(t) = \langle b^+(t) b(t)\rangle = \left.\frac{\partial^2 \chi_b(\eta, t)}{\partial(-\eta^*)\partial\eta}\right|_{\eta=\eta^*=0} \tag{16}$$

From Eqs. (15) and (16), one obtains

$$N_b(t) = k^2 t^2 |\alpha_0|^6 = k^2 t^2\, N_a(0)^3 \tag{17}$$

where $|\alpha_0|^2 = N_a(0)$ is the average number of photons initially present in the pump mode. It is clear from Eq. (17) that the number of third harmonic photons produced is proportional to the cube of the number of pump photons initially present or in other words it is proportional tothecube of the initial pump intensity-which is a well-known result. In the short--time approximation $N_b(t)$ is proportional to the square of the interaction time t.

The variance of intensity[17] for the third harmonic is defined by

$$<(\Delta N)^2> \equiv <b^+(t)^2 b^2(t)> - (<b^+(t)b(t)>)^2 \tag{18}$$

which in terrns of the characteristic function can be written as

$$<(\Delta N)^2> = \left.\frac{\partial^4 \chi_b(\eta,t)}{\partial^2(-\eta^*)\partial^2\eta}\right|_{\eta=\eta^*=0} - \left(\left.\frac{\partial^2 \chi_b(\eta,t)}{\partial(-\eta^*)\partial\eta}\right|_{\eta=\eta^*=0}\right)^2 \tag{19}$$

In the short-time approximation, when terms only up to t^2 are being retained, the value of $<(\Delta N)^2>$ is zero. It implies[17] that the photons in the third harmonic mode are uncorrelated if the pump mode is inacoherent state and the harmonic mode is in the vacuum state initially.

6. PUMP MODE DEPLETION

To investigate the depletion and variance of the pump field as a function of time, we use the normally ordered characteristic function for the pump mode which is given by

$$\chi_p(\xi,t) = \mathrm{Tr}\ \{\rho(0)\ e^{\xi a^+(t)}\ e^{-\xi^* a(t)}\} \tag{20}$$

as we already mentioned in Eq. (12). On substituting the expressions for $a^+(t)$ and $a(t)$ from Eq. (10) on the right hand side of Eq. (20) and then expanding it in powers of kt up to second order, after tracing the resultant expression for $\chi_p(\xi t)$ is

$$\chi_p(\xi,t) = e^{\xi A^+(t) - \xi^* A(t) + \xi^2/2\ B^+(t) + \xi^2/2^* B(t)} \tag{21}$$

where

$$A(t) = \alpha_0(1-3/2k^2t^2|\alpha_0|^4)e^{-i\omega_a t} \tag{22}$$

and

$$B(t) = -3k^2t^2|\alpha_0|^2\alpha_0^2 \; e^{-2i\omega_a t} \tag{23}$$

Once $\chi_p(\xi,t)$ is known, one can easily calculate relevant quantities from it. The average number of photons present in the pump mode at time t is

$$\langle a^+(t)a(t)\rangle = \left.\frac{\partial^2\chi_p(\xi,t)}{\partial(-\xi^*)\partial\xi}\right|_{\xi=\xi^*=0} = N_a(0)-3k^2t^2N_a(0)^3 \tag{24}$$

The Eq. (24) shows that the depletion of the pump mode is proportional to the cube of the number of the pump photons initially present as expected from Eq. (9).

The variance of intensity of the pump mode is evaluated in the same manner as done in the previous section for the third harmonic mode and it is found as

$$\langle(\Delta N)^2\rangle = \langle a^+(t^2)a^2(t)\rangle - (\langle a^+(t)a(t)\rangle)^2 = -6k^2t^2N_a^3(0) \tag{25}$$

which comes out to be a negative quantity. Using the criteria of reference[17], we find that, in third harmonic generation, the pump photons show antibunching if initially the pump mode is in the coherent state and the third harmonic mode in the vacuum state.

In a recent semiclassical investigation[11], the third harmonic generation has been studied including the effect of pump depletion and of saturation of two-photon absorption. The antibunching of laser photons which we have predicted above cannot come out of their theory. Antibunching is a quantum effect, only quantized fields couldaccount for it.

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RESUMO

Investigamos o crescimento do terceiro harmônico e a atenuação de modo excitador descrevendo quanticamente os dois campos. As equações de movimento que descrevem a dinâmica dos modos excitor e terceiro harmônico são resolvidas na aproximação de tempos curtos. Verifica-se que o modo excitador apresenta efeitos de antibunching devido à interação não linear entre os modos.