SQUEEZING BY MULTIPLE HIGHER-HARMONIC GENERATION

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It is shown that by generating higher harmonic in a cascade of thin nonlinear plates and filtering out the generated harmonic behind each plate, considerable squeezing in the fundamental mode occurs. The multiple higher-harmonic generation processes are aimed to simulate ideal multi-photon absorbers without undesirable lower-order absorption and emission processes.

1. INTRODUCTION

The 1970's witnessed a considerable number of publications concerning the search for photon antibunching – a direct manifestation of the quantum nature of light. Moreover, some highly sophisticated methods of its enhancement [1–4] have been proposed. One of the methods mentioned above consists in multiple second-harmonic generation (M.S.H.G.) in thin plates when filtering out the generated second harmonic radiation behind each plate [3–5].

Squeezing which has recently become the subject of intense study is another facet of the quantum nature of light. Some schemes for its enhancement have been proposed as well [6, 7]. Squeezing has first been observed by Slusher et al. [8] in four-wave mixing.

Certain nonlinear optical phenomena produce fields displaying photon antibunching but not squeezing, and vice versa. However, there exist phenomena producing jointly both these unique quantum effects. To the class of such phenomena belong processes of multi-photon absorption (M.P.A.) [9–11] and higher-harmonic generation (H.G.) [12–16].

It is our aim to show that multiple higher-harmonic generation (M.H.H.G.) in thin nonlinear plates also exhibits considerable squeezing in the fundamental mode.

2. FUNDAMENTAL REMARKS

In the electric dipole approximation, at perfect phase matching, the process of k-th harmonic generation in a nondissipative medium is described by the following interaction Hamiltonian [12]

$$H = \hbar \varkappa_k a_k^+ a_f^k + \text{h.c.}\,, \tag{1}$$

where $\varkappa_k$ is the coupling constant, the subscripts k and f denote the k-th harmonic and fundamental mode, respectively, and h.c. represents the Hermitian conjugate. a^+ and a are the photon creation and annihilation operators, respectively.

The time evolution of photon annihilation operators a_f and a_k has been solved as yet in the short-time approximation only, and hence photon antibunching [12, 14] and squeezing [12, 15, 16] have been predicted solely at the beginning of the process. On the other hand, numerical calculations [17, 18] show that photon antibunching is absent in the long-time approximation due to the spontaneous reemision of photons between the harmonic and fundamental mode. Hence, we cannot search for a decrease of quantum fluctuations by using longer and longer media as in the case of M.P.A., which is a one-mode process. However, when H.H.G. takes place in a cascade of thin plates and the harmonic is filtered out behind each plate, the process runs in a way similar to M.P.A. [5]. Then, moreover, the short-time approximation provides an almost realistic description of generation in each approximately thin plate. We assume that the thickness L of all the plates is identical. Thus the time of nonlinear interaction in each plate is also the same, and $\tau = L/v$, where v is the velocity of light in the medium, equal for both beams at phase matching.

The boundary conditions for m-th plate are

$$a_{f,m}(0) = a_{f,m-1}(\tau)\,, \quad n_{k,m}(0) = 0\,, \tag{2}$$

where $a_{f,m-1}$ is the annihilation operator of the fundamental mode emerging from the preceding $(m-1)$-th plate and $n_{k,m}$ is the number of harmonic photons.

The electromagnetic field is in a squeezed state if one of the variances of the quadrature components

$$Q = a + a^+\,, \quad P = -\mathrm{i}(a - a^+) \tag{3}$$

is less than unity:

$$\langle(\Delta Q)^2\rangle < 1 \quad \text{or} \quad \langle(\Delta P)^2\rangle < 1\,. \tag{4}$$

3. RESULTS

The short-time approximation leads to the following solution for the slowly varying part of the annihilation operator a_f in the one-step H.H.G. process [12]

$$a_f(\tau) = a_f(0) - \mathrm{i}k\varkappa_k(a_f^+)^{k-1}(0)\,a_k(0)\,\tau + \tfrac{1}{2}k\varkappa_k^2\,. \cdot\left\{\sum_{s=1}^{k-1} s!\binom{k-1}{s}\binom{k}{s}(a_f^+)^{k-1-s}(0)\,a_f^{k-s}(0)\,a_k^+(0)\,a_k(0) - (a_f^+)^{k-1}(0)\,a_f^k(0)\right\}\tau^2. \tag{5}$$

Using the above solution and the boundary conditions (2) for calculating the variances (4) after the m-th step of k-th harmonic generation at coherent input radiation, we get

$$\langle(\Delta Q)^2_{f,m}\rangle = 1 - mk(k-1)\,\varkappa_k^2|\alpha_f|^{2(k-1)}\,\tau^2\cos(2\varphi)\,, \tag{6a}$$

$$\langle(\Delta P)^2_{f,m}\rangle = 1 + mk(k-1)\,\varkappa_k^2|\alpha_f|^{2(k-1)}\,\tau^2\cos(2\varphi)\,, \tag{6b}$$

where $|\alpha_f|$ is the amplitude and φ the phase of the input fundamental radiation: $\alpha_f = |\alpha_f| \exp(i\varphi)$.

The deviation from unity is now m times greater than that in [12]. For $m = 1$ and $k = 2$ Mandel's result [15] is recovered. It is easily seen that, depending on φ, either $Q_{f,m}$ or $P_{f,m}$ is squeezed according to the conditions (4). Squeezing grows with increasing number m of k-th harmonic passages.

It is obvious from the forms of the variances (6) that their product is less than unity. This might suggest a violation of the uncertainty principle for Q and P. The deviation from unity is proportional to τ^4. In equation (5) we have omitted higher order terms up to τ^4 which ought to be taken into account if one wishes to obtain the correct form of the uncertainty principle in this order of approximation, so the principle is in fact not violated.

4. DISCUSSION

It can be shown by a procedure similar to that for M.S.H.G. that the multiple k-th harmonic generation acts in the same way as k-photon absorption and, thus, any theoretical model describing the one process can be used for the other. Consequently, the exact solution of M.P.A. obtained in [11] is assumed to suit the description of M.H.H.G. as well. However, the rigorous solution in [11] is beset with serious problems in the evaluation of the resulting formulas for greater numbers of input photons, say $\langle n_f(0)\rangle \gtrapprox 20$. From the practical point of view, it is the approximate solution for strong ($\langle n_{f,m}\rangle \gg 1$) interacting radiation that is very important. Such a solution has recently been found for two-photon absorption (T.P.A.) by Bandilla [19].

Using the results of [19], we obtain the following formulas for the evolution of strong field squeezing in M.S.H.G. ($k = 2$)

$$\text{(7a)} \qquad \langle(\Delta Q)_f^2\rangle = 1 + \frac{\xi^2(3 + 2\xi)}{6(1 + \xi)^3} - \frac{\xi}{1 + \xi}\left[1 - \frac{\xi(3 + 2\xi)}{6(1 + \xi)^2}\right]\cos(2\varphi),$$

$$\text{(7b)} \qquad \langle(\Delta P)_f^2\rangle = 1 + \frac{\xi^2(3 + 2\xi)}{6(1 + \xi)^3} + \frac{\xi}{1 + \xi}\left[1 - \frac{\xi(3 + 2\xi)}{6(1 + \xi)^2}\right]\cos(2\varphi),$$

where $\xi = 2m\varkappa_2^2|\alpha_f|^2\,\tau^2$. The last formulas describe the evolution of squeezing up to the quasi-steady state [5, 9–11], which represents the theoretical limit that can be reached in real experiments. Thus, the ultimate value of squeezing in M.S.H.G. is $\frac{2}{3}$, and approximately the same values are assumed to occur for extreme squeezing in M.H.H.G. for $k > 2$ [11].

The approximate formulas for M.H.H.G. ($k > 2$), similar to equations (7), may result from the same procedure as that used for T.P.A. in [19], similarly as done for photon antibunching in M.P.A. in [20].

The method of multiple k-th harmonic generation seems to be very advantageous for the simulation of k-photon absorption because it enables us to avoid the undesirable lower-order absorptions as well as emission processes. On the other hand, unfortunately, the higher-order k-th harmonic generations ($k > 3$) can be accompanied by undesirable phase mismatched lower frequency generations with non-negligible efficiencies. Therefore the only multiple second- and third-harmonic generations are expected to be suitable for simulation of two- and three-photon absorptions, respectively.

As for the experimental demonstration of the squeezing phenomena predicted above, they can further be enhanced considerably in destructive interference when measured by means of increased photon antibunching [1, 2, 19].

Note that, alternatively, M.H.H.G. in an optical resonator can be considered [3–5].

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