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Quantum-noise-induced macroscopic revivals in second-harmonic generation

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We investigate the behavior of the fundamental and second-harmonic fields in phase-matched traveling plane-wave second-harmonic generation, using the full-operator equations of motion. We find that, after a certain interaction length, both the macroscopic and quantum-statistical properties of the harmonic and fundamental fields are qualitatively different from those found in previous analyses. The mean fields do not vary in a monotonic way, but oscillate with the propagation length, leading to an unexpected periodic revival of the fundamental field, triggered by the quantum fluctuations always present in the mode. Accordingly, the amplitude noise of the fundamental, previously predicted to be perfectly squeezed for long interaction lengths, actually reaches a very small minimum for a definite length, then increases again.

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Second-harmonic generation (SHG) has been studied in great detail since the first years of nonlinear optics, and is often taken as the simplest example of a nonlinear optical process. In the simple traveling plane-wave configuration, the solution for the generated fields as a function of the propagation length z is well known [1,2]. It predicts that, when one starts from a nonzero fundamental field and no second harmonic, one gets a total and irreversible transfer to the second harmonic mode when z grows to infinity. SHG is also of great importance for the generation of nonclassical states of light, either in an intracavity configuration [3–8] or in the pure propagation case [9–13]. In this second case, the amount of squeezing present in the fields has been calculated using the standard linearized fluctuation analysis. In the case of perfect phase matching, this analysis [9] predicts that the fundamental field evolves into a perfectly amplitude squeezed vacuum, whereas the second-harmonic field undergoes a 50% amplitude squeezing, and strong quantum correlations develop between the two modes [14]. However, it is clear that the prediction that a perfectly squeezed vacuum is generated is in contradiction to the assumption that the quantum fluctuations of the different fields are much smaller than mean fields, which is at the basis of the linearization technique. Moreover, there is reason to doubt that the situation predicted by the classical model, where the second-harmonic field is large and the fundamental is zero, can remain stable when z tends to infinity. It is well known that, starting with these values of the mean fields, one finds a growing of the fundamental mode by parametric splitting of the second harmonic triggered by parametric fluorescence.

In this article, using more accurate approaches that do not rely on linearization, we show that the behavior both of the mean fields and of the quantum fluctuations is qualitatively different from the previous results for long interaction lengths. In particular, we find that the mean fields do not have a monotonic variation, but oscillate with z. We thus predict a very spectacular macroscopic revival of the fundamental field that is induced by the quantum noise present in the interacting modes.

Let us call $\hat{a}$ and $\hat{b}$ the annihilation operators for the fundamental and harmonic mode at point z, and κ the effective strength of the nonlinear interaction between the light modes in the nonlinear crystal. The exact propagation equations for these operators are [9,11]

$$\frac{d\hat{a}}{dz}=\kappa\hat{a}^{\dagger}\hat{b},\quad \frac{db}{dz}=-\frac{\kappa}{2}\hat{a}^{2}. \tag{1}$$

No exact analytical solutions are known for these operator equations. However, it is possible to find numerical solutions by stochastic simulations in the phase-space representations of quantum optics [15], either exactly in the positive-P [16], or approximately in the Wigner representation [17,18].

When one replaces the operators $\hat{a}$ and $\hat{b}$ by the c numbers α and β, one retrieves the well-known classical propagation equations of nonlinear optics, which can be solved exactly. In the pure SHG case with $\beta(z=0)=0$ and $\alpha(z=0)\in\mathbb{R}$, one finds

$$\alpha(\zeta)=\alpha(0)\mathrm{sech}(\zeta),\quad \beta(\zeta)=-\frac{\alpha(0)}{\sqrt{2}}\tanh(\zeta), \tag{2}$$

where $\zeta=z[\kappa|\alpha(0)|/\sqrt{2}]$. Setting

$$\hat{a}(\zeta)=\alpha(\zeta)+\delta\hat{a}(\zeta),\quad \hat{b}(\zeta)=\beta(\zeta)+\delta\hat{b}(\zeta), \tag{3}$$

and assuming that the fluctuations are small, one can linearize the operator equations (1) around the classical solutions, which leads to simple analytical expressions for the quadrature variances and the correlation functions of the two fields [9]. However, the linearization procedure is not valid when ζ

is large, because the phase quadrature fluctuations of the fundamental field are found to diverge exponentially, whereas the mean field decreases. In this model the mean field and the rms fluctuations are equal when $\zeta=\zeta_0=\frac{1}{4}\ln[32|\alpha(0)|^2]$, with linearization only being valid where $\zeta\ll\zeta_0$.

We now show that it is possible to obtain a more accurate, but still approximate, propagation equation for the mean fields that does not rely on linearization. Letting $\hat{N}_1=\hat{a}^\dagger\hat{a}$ and $\hat{N}_2=\hat{b}^\dagger\hat{b}$, and using $[\hat{a}(z),\hat{a}^\dagger(z')]=[\hat{b}(z),\hat{b}^\dagger(z')]=\hat{1}\,\delta(z-z')$, we find exact propagation equations for these two operators,

$$\frac{d\hat{N}_1}{dz}=-2\frac{dN_2}{dz}=-\hat{S}, \tag{4}$$

where $\hat{S}=\kappa[\hat{a}^2\hat{b}^\dagger+\hat{a}^{\dagger\,2}\hat{b}]$. Equation (4) implies that $\hat{N}_1+2\hat{N}_2$ is constant in the propagation, as required by energy conservation. From Eq. (1), one finds that the evolution of the operator $\hat{S}$ is given by

$$\frac{d\hat{S}}{dz}=\kappa^2[\hat{N}_1^2-4\hat{N}_1\hat{N}_2-\hat{N}_1-2\hat{N}_2]. \tag{5}$$

The energy invariant requires that $\hat{N}_2(z)=\frac{1}{2}[\hat{N}_0-\hat{N}_1(z)]$, where $\hat{N}_0=\hat{N}_1(0)+2\hat{N}_2(0)$. We can therefore write an exact quantum propagation equation which only involves the operators $\hat{N}_1(z)$ and $\hat{N}_0$,

$$\frac{d^2\hat{N}_1}{dz^2}=-\kappa^2[3\hat{N}_1^2-2\hat{N}_0\hat{N}_1-\hat{N}_0]. \tag{6}$$

This second-order equation cannot be solved alone, because it depends on the operators $\hat{N}_1^2(z)$ and $\hat{N}_0\hat{N}_1(z)$, which obey other propagation equations that one can also derive from Eq. (4), giving an infinite hierarchy of propagation equations. In order to get an approximate solution, one must stop this hierarchy at a given level. The first level of approximation is to neglect all correlations and write

$$\langle\hat{N}_1^2(z)\rangle=\langle\hat{N}_1(z)\rangle^2,\quad \langle\hat{N}_1(z)\hat{N}_0\rangle=\langle\hat{N}_1(z)\rangle\langle\hat{N}_0\rangle. \tag{7}$$

The operator equation (6) is then transformed into an ordinary differential equation for the mean photon number $[N_1(z)=\langle\hat{N}_1(z)\rangle]$, depending on the initial photon number N_0. It is easy to show that there is a quantity conserved in the propagation

$$\frac{d}{dz}\left[\frac{1}{2}\left(\frac{dN_1}{dz}\right)^2+\kappa^2(N_1^3-N_0N_1^2-N_0N_1)\right]=0. \tag{8}$$

This quantity can be considered as the total mechanical energy for a pseudoparticle of position N_1, with $\frac{1}{2}(dN_1/dz)^2$ as the kinetic energy and $\kappa^2(N_1^3-N_0N_1^2-N_0N_1)$ as the potential energy, which is shown in Fig. (1a). The pseudoparticle will oscillate without damping in this potential well, which means that N_1 will exhibit full periodic revivals of the fundamental intensity. We can now understand why $N_1(z)$ cannot vanish, as, starting from $N_1(z=0)=N_0$ with zero kinetic energy (because $dN_1/dz=0$ when the second-harmonic field is zero), the total pseudoenergy has a negative value, so that the second turning point of the periodic motion is reached at a nonzero value of N_1. The minimum value for N_1 is found to be equal to $\sqrt{N_0}$.

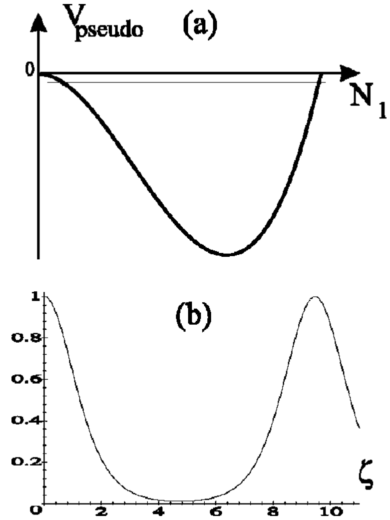

FIG. 1. (a) The effective semiclassical potential in which N_1 *moves* with a slightly negative total energy. (b) Analytical solution for the proportion of photons in the fundamental mode as a function of normalized propagation length ζ, with $N_0=10^6$.

It is possible to find an exact solution of Eq. (6) using the approximation (7), in terms of Jacobi elliptic functions, as in the general classical solution of three-wave mixing found in [1]. Letting $f(\zeta)=N_1(z)/N_0$ with ζ as in Eq. (2), Eq. (6) becomes

$$\frac{d^2f}{d\zeta^2}=-6f^2+4f+2\epsilon, \tag{9}$$

where $\epsilon=1/N_0$. If ϵ is taken equal to zero, one obtains the classical limit given in Eq. (2). When $\epsilon\neq0$, the solution of Eq. (9) is

$$f(\zeta)=(1-\sqrt{\epsilon})\,\mathrm{cn}^2(\sqrt{1+\sqrt{\epsilon}}\zeta)+\sqrt{\epsilon} \tag{10}$$

where cn is the Jacobi cosine-amplitude [19] of modulus $k=\sqrt{1-\epsilon}/(1+\sqrt{\epsilon})$. This solution is displayed in Fig. (1b). One observes the expected periodic behavior of the fundamental, with full revivals, for any nonzero value of ϵ. The inclusion of ϵ, arising from the commutators of the field operators, means that we have included some quantum fluctuations, at least in the initial conditions. Though ϵ may be almost vanishingly small, it has a huge macroscopic effect on the system dynamics. It is apparent that the quantum noise, which is always present, causes oscillations between the regimes of up- and down-conversion, with the period of the oscillations becoming infinite as ϵ vanishes.

The period ζ_R of the revival has a simple expression when $N_0\gg1$,

$$\zeta_R = \ln(8\sqrt{N_0}), \tag{11}$$

which is roughly twice the validity length of the linearized solution.

The approximation made in Eqs. (7) consists in neglecting, at first order, the intensity noise of the fundamental field and the intensity correlations between the field at the considered position and the field at its starting point. This obviously becomes less valid for large values of ζ, when the fundamental field decreases and the quantum correlations develop. It is possible to go further than this approximation by correcting Eq. (7) with the intensity noise and correlations calculated by the linearized technique, valid until half of the period. We have numerically calculated the mean field using this second-order approximation with $N_0 = 10^6$, and found again a total revival of the fundamental field, shifted to a slightly larger ζ value.

To solve exactly the long-range behavior of the mean fields in SHG, and also their quantum fluctuations, we use numerical stochastic integration. Using the method of operator correspondences [20], and proceeding via the master and Fokker-Planck equations, we find the system of equations in the positive-*P* representation,

$$\begin{aligned}
\frac{d\alpha}{dz} &= \kappa\alpha^\dagger\beta + \sqrt{\kappa\beta}\,\eta_1(z),\\
\frac{d\alpha^\dagger}{dz} &= \kappa\alpha\beta^\dagger + \sqrt{\kappa\beta^\dagger}\,\eta_2(z),\\
\frac{d\beta}{dz} &= -\frac{\kappa}{2}\alpha^2,\\
\frac{d\beta^\dagger}{dz} &= -\frac{\kappa}{2}\alpha^{\dagger 2},
\end{aligned} \tag{12}$$

where α and $\alpha^\dagger$, as well as β and $\beta^\dagger$, are independent *c*-number variables that are not complex conjugate except in the average over a large number of trajectories. The noise sources are real and δ correlated : $\overline{\eta_i(z)\eta_j(z')} = \delta_{ij}\delta(z-z')$.

The differential equation found for the Wigner quasiprobability distribution has third-order derivatives, which means that there is no Fokker-Planck equation in this representation. As there is no simple way to deal with third-order derivatives in a stochastic differential equation [21], we find an approximate equation by truncating these third-order terms. This leaves us with the same equations found by linearization, but with one very important difference: the initial conditions for each stochastic trajectory are found from the Wigner distribution for a coherent state. The advantage of the Wigner distribution is that the numerical simulations are generally more stable than with the positive-*P* representation, but we must remember that the truncation means that higher-order nonlinear effects are partially neglected. The advantage of the positive-*P* representation is that, where the integration converges, it gives exact solutions for the full operator equations.

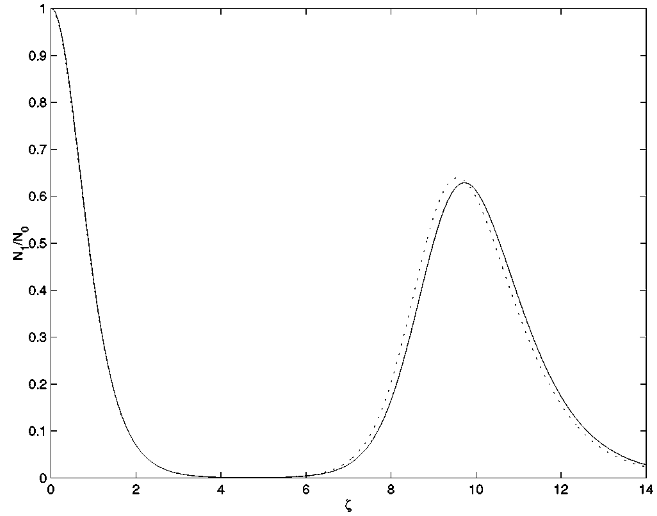

FIG. 2. Proportion of photons in the fundamental mode as a function of normalized propagation length ζ, given by numerical simulations using the positive-*P* representation (dotted line), and the Wigner representation (full line), with $N_0 = 10^6$.

The behavior of the mean fields, obtained by averaging 10 000 computed trajectories, is shown in Fig. 2, for values of $N_0 = 10^6$. The two methods give results that are in good agreement. One still finds an oscillatory behavior in the photon number, although the first revival is no longer total. The minimum value for N_1 found here is $\approx 30\%$ less than the value predicted by expression (9). For comparison, we have also plotted the semiclassical solution, which gives a revival for almost the same value of ζ, but is obviously not accurate for very long interaction lengths.

Figure 3 gives the computed variance of the amplitude quadrature $X = a + a^\dagger$. We observe that it reaches a nonzero minimum, then suddenly increases drastically to give a large

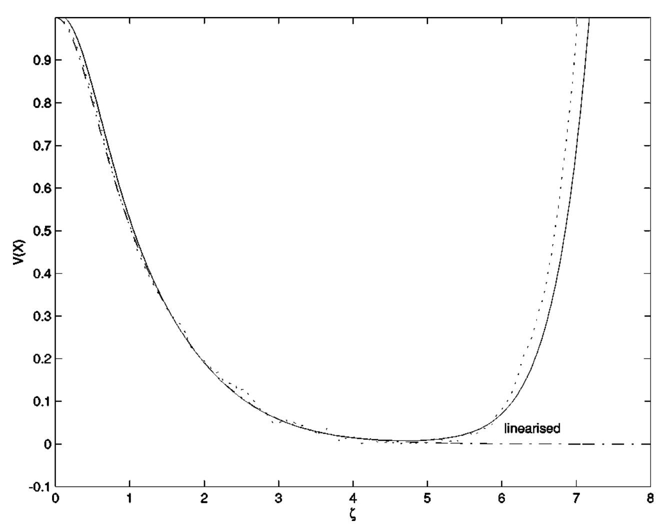

FIG. 3. Variance of the amplitude quadrature of the fundamental as a function of normalized propagation length ζ, calculated using the positive-*P* representation (dotted line), and the Wigner representation (full line), with $N_0 = 10^6$. The linearized solution is shown for comparison.

degree of excess noise when the revival begins. This is analogous to, but more drastic than a result previously found with the optical parametric oscillator [22], where increasing fluctuations in the phase quadrature fed back into the squeezed amplitude quadrature, and the quantum noise suppression had a maximum at a particular pumping value. The $Y=-i(a-a^{\dagger})$ quadrature always exhibits a noise larger than the standard quantum limit for as far as we have run our simulations. It is apparent that the solution for the mean fields found from Eq. (6) becomes inaccurate at just the point where the quantum noise increases.

In conclusion, by solving the quantum propagation equations, we have shown that the mean fields depart strongly from the classical solution of SHG for long interaction lengths, and also that the linearization procedure previously used to determine the quantum fluctuations breaks down relatively quickly. This is a distinct signature of the effect of quantum noise on the macroscopic behavior, with the revivals found not being possible without the inclusion of the small $(1/N_0)$ term in the propagation equation. We have also found that the analytical solution (10) has a restricted region of validity. The full quantum evolution of the mean fields diverges from it when the quantum noise increases drastically. In particular, the full periodic revivals of the fundamental field are no longer present.

Note that the propagation equation that we have obtained, and its analytical solution, can also be used in other physical situations. For example, this equation also describes parametric down-conversion, when one starts from the second-harmonic field without any fundamental wave, because the initial quantum noise of spontaneous parametric fluorescence is directly built in. This is not the case for the classical equations of nonlinear optics, which will not describe down-conversion without the artificial addition of fluctuations in the fundamental.

In order to predict actual experimental data for such macroscopic changes induced by very small quantum effects in SHG, one first needs to extend our analysis to Gaussian beams instead of simple plane waves. We expect that the main conclusions drawn in this paper will remain valid as long as the revival length is small compared to the Rayleigh length of the light beams. Such a situation can be encountered using very powerful laser systems.

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