Effects of $\chi^{(3)}$ nonlinearities in traveling-wave second-harmonic generation

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We investigate the effects of self-phase and cross-phase $\chi^{(3)}$ nonlinearities, in the process of traveling-wave second-harmonic generation. We derive a semiclassical analytical solution for the field intensities, comparing this with the numerically obtained fully quantum solutions. We also investigate the effects of the cross-phase modulation on the quantum statistical properties of the fields. We find that, as the $\chi^{(3)}$ components increase, there are qualitative changes to both the field intensities and the quantum statistics.

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I. INTRODUCTION

It has long been known that nonlinear parametric processes such as second-harmonic generation (SHG), optical parametric oscillation (OPO), and amplification (OPA) can produce nonclassical states of the electromagnetic field [1]. Much theoretical and experimental work has been done on these cases, in both of which electromagnetic fields at differing frequencies are coupled by a second-order, $\chi^{(2)}$ nonlinearity. As all real nonlinear materials are expected to have an effective $\chi^{(3)}$ component, it is of interest to calculate the effects of this component in these nonlinear optical processes.

There have been a number of theoretical analyses of systems in which both $\chi^{(2)}$ and higher-order nonlinearities are present, although few of these are for frequency upconversion processes and most make a number of approximations, such as classical, undepleted pumping [2–4]. We have previously performed an analysis of SHG with added $\chi^{(3)}$ nonlinearities in both the traveling-wave and intracavity cases, comparing and contrasting the fully quantum solutions with those found by the common process of linearization [5]. In this previous work we gave an analytical semiclassical solution for the field intensities in the process of traveling-wave SHG with self-phase modulation, finding that this was closely similar to the fully quantum solutions, as opposed to the case with pure SHG, where the semiclassical and quantum solutions are markedly different [6]. In this present paper we give an analytical solution for the intensities with both self-phase and cross-phase modulation present, comparing this to the full quantum solutions, obtained using the positive-P representation [7].

II. ANALYTICAL SOLUTION

We consider a nonlinear $\chi^{(2)}$ and $\chi^{(3)}$ crystal, in which a pump field at frequency ω produces an harmonic field at frequency 2ω. We consider here only the case of perfect phase-matching between the two fields, with both fields considered as plane waves. In the traveling-wave regime we can write an interaction Hamiltonian, with the trivial ω dependence of the fields removed, as

$$\mathcal{H}=\frac{i\hbar\kappa}{2}[\hat{a}^{\dagger 2}\hat{b}-\hat{a}^2\hat{b}^\dagger]+\hbar\chi[\hat{a}^{\dagger 2}\hat{a}^2+\hat{b}^{\dagger 2}\hat{b}^2]+2\hbar\xi\hat{a}^\dagger\hat{a}\hat{b}^\dagger\hat{b}, \tag{1}$$

where $\hat{a}$ and $\hat{b}$ are the annihilation operators for photons at frequencies ω and 2ω, respectively, at position z inside the nonlinear crystal, κ represents the effective strength of the nonlinear interaction between the two modes, χ represents the effective strength of the self-phase modulation $\chi^{(3)}$ nonlinearity, and ξ represents the strength of the cross-phase modulation $\chi^{(3)}$ nonlinearity. We consider here the case where the Kerr-type interaction has equal effective strengths for each mode. The cross-phase modulation strength will depend on such things as the mode overlap and can typically vary up to the maximum of the self-interaction strength.

The operator equations of motion for the system are found as

$$\frac{d\hat{a}}{dz}=\kappa\hat{a}^\dagger\hat{b}-2i\chi\hat{a}^\dagger\hat{a}^2-2i\xi\hat{a}\hat{b}^\dagger\hat{b},$$

$$\frac{d\hat{b}}{dz}=-\frac{\kappa}{2}\hat{a}^2-2i\chi\hat{b}^\dagger\hat{b}^2-2i\xi\hat{a}^\dagger\hat{a}\hat{b}, \tag{2}$$

for which no analytical solution is known. The first level of approximation often used in solving operator equations is linearization, or assuming that the operators can be directly replaced by complex numbers to give the mean values of the fields. In the case of traveling-wave pure SHG, this method has been shown to have limited validity [6], but in the present case the analytical solution for the photon number follows more closely the full quantum solutions when the cross-phase modulation term, ξ, is set to zero [5]. Following a similar procedure, we make the substitutions $\hat{a}\rightarrow\alpha=\langle\hat{a}\rangle$ and $\hat{b}\rightarrow\beta=\langle\hat{b}\rangle$, giving the following semiclassical equations:

$$\frac{d\alpha}{dz}=-2i\chi|\alpha|^2\alpha-2i\xi|\beta|^2\alpha+\kappa\alpha^*\beta,$$

$$\frac{d\beta}{dz} = -2i\chi|\beta|^2\beta - 2i\xi|\alpha|^2\beta - \frac{\kappa}{2}\alpha^2. \tag{3}$$

Note that we have not bothered with the normal method of calculating fluctuations around the classical solutions, as experience with the pure SHG system has shown the results to be highly inaccurate after a short-interaction length and we would expect this to be the case here also.

Defining $a(z) = |\alpha(z)|^2$ and $b(z) = |\beta(z)|^2$ (note that these are real numbers, not the operators used above), we find that Eq. (3) can be written as

$$\frac{da}{dz} = kv,$$

$$\frac{db}{dz} = -\frac{k}{2}v, \tag{4}$$

where

$$v = \alpha^{*2}\beta + \alpha^2\beta^*. \tag{5}$$

From Eq. (4) and the principle of conservation of energy, it follows that $c_0[=a(z)+2b(z)]$ is a constant of the propagation. In pure SHG, where $|\beta(0)|^2 = 0$, we have $c_0 = |\alpha(0)|^2$. If we now introduce the variable

$$w(z) = i(\alpha^{*2}\beta - \alpha^2\beta^*), \tag{6}$$

we can write Eq. (3) in the form

$$\frac{dv}{dz} = ka(4b-a) + 2[(2\chi-\xi)a + (2\xi-\chi)b]w,$$

$$\frac{dw}{dz} = 2[(\chi-2\xi)b + (\xi-2\chi)a]v. \tag{7}$$

Using the fact that $a = c_0 - 2b$ and introducing a new variable

$$x(z) = 2(\chi-2\xi)b + 2(\xi-2\chi)a = (10\chi-8\xi)b + 2c_0(\xi-2\chi), \tag{8}$$

we can combine Eqs. (4) and (7) in the form

$$\frac{dx}{dz} = -gv,$$

$$\frac{dw}{dz} = xv,$$

$$\frac{dv}{dz} = \alpha_0 - \alpha_1 x - \alpha_2 x^2 - xw, \tag{9}$$

where

$$g = \frac{k\sigma}{2},$$

$$\alpha_0 = -kc_0^2 + \frac{8kc_0\nu}{\sigma} - \frac{12k\nu^2}{\sigma^2},$$

$$\alpha_1 = \frac{24k\nu}{\sigma^2} - \frac{8kc_0}{\sigma},$$

and

$$\alpha_2 = \frac{12k}{\sigma^2}, \tag{10}$$

in which

$$\sigma = 10\chi - 8\xi,$$

and

$$\nu = 2c_0(2\chi-\xi). \tag{11}$$

Using the first and second equations of Eq. (10), we can now define another constant of the motion

$$\frac{1}{2}x^2(z) + gw(z) = c_1, \tag{12}$$

where $c_1 = 2(2\chi-\xi)^2|\alpha(0)|^4$. We can now utilize Eqs. (10) and (12) to find an equation of motion for the variable $x(z)$

$$\frac{d^2}{dz^2}x = -\alpha_0 g + (c_1 + \alpha_1 g)x + \alpha_2 g x^2 - \frac{1}{2}x^3. \tag{13}$$

It is clear that Eq. (13) can be written in the form

$$\frac{d^2}{dz^2}x = -\frac{\partial}{\partial x}U(x), \tag{14}$$

where the pseudopotential $U(x)$ has the form

$$U(x) = -\frac{1}{2}(a_1 x + a_2 x^2 + a_3 x^3 + a_4 x^4). \tag{15}$$

In the above,

$$a_1 = -2\alpha_0 g,$$

$$a_2 = c_1 + \alpha_1 g,$$

$$a_3 = \frac{2}{3}\alpha_2 g,$$

and

$$a_4 = -\frac{1}{4}. \tag{16}$$

It is now evident that, by treating a total pseudoenergy as a constant of the motion, we can write

$$\frac{1}{2}\left(\frac{dx}{dz}\right)^2+U(x)=E, \tag{17}$$

which leads to the first-order differential equation for $x(z)$

$$\frac{dx}{dz}=\pm\sqrt{a_0+a_1x+a_2x^2+a_3x^3+a_4x^4}, \tag{18}$$

where $a_0=2E$. The formal solution of Eq. (18) is

$$z=\pm\int_{x(0)}^{x(z)}\frac{dx}{\sqrt{a_0+a_1x+a_2x^2+a_3x^3+a_4x^4}}, \tag{19}$$

where $x(0)=-2(2\chi-\xi)|\alpha(0)|^2$.

We find that there are three cases where Eq. (19) has periodic solutions. Defining

$$f(x)=\sum_{k=0}^{4}a_kx^k=-\lambda^2\prod_{k=1}^{4}(x-x_k), \tag{20}$$

where $a_4=-\lambda^2$, with $\lambda=1/2$, we can now examine the roots of the polynomial $f(x)=0$.

In the first-two cases, there are four real roots: $x_1>x_2>x_3>x_4$ and the solution can be written as

$$x(z)=M+\frac{N}{D+\mathrm{sn}^2(\Omega z+\phi,k)}, \tag{21}$$

where sn is the Jacobi sine amplitude of modulus k [8] and

$$\Omega=\frac{\lambda}{2}\sqrt{(x_1-x_3)(x_2-x_4)},$$

$$k=\sqrt{\frac{(x_1-x_2)(x_3-x_4)}{(x_2-x_4)(x_1-x_3)}}, \tag{22}$$

and the constant ϕ is determined from the initial condition by

$$\phi=\mathrm{sn}^{-1}\left(\sqrt{\frac{N-D(x(0)-M)}{(x(0)-M)}},k\right). \tag{23}$$

The function $x(z)$ is periodic, with the period given by

$$T=\frac{2}{\Omega}\int_0^1\frac{dt}{\sqrt{(1-t^2)(1-k^2t^2)}}$$

$$=\frac{2}{\Omega}K(k), \tag{24}$$

where $K(k)$ is the full elliptic integral. It is clear from the definition that the period of $x(z)$ is the same as that of $|\alpha(z)|^2$.

We find that there are two separate cases for the solution given by Eq. (21). The first of these cases, which is that encountered for the parameters we have used in this investigation is where

(i) $x_3\geqslant x\geqslant x_4$

In this case $M=x_1$, $N=-(x_1-x_4)(x_1-x_3)/(x_3-x_4)$, and $D=(x_1-x_3)/(x_3-x_4)$.

(ii) $x_1\geqslant x\geqslant x_2$.

In this case, $M=x_4$, $N=(x_1-x_4)(x_2-x_4)/(x_1-x_2)$, and $D=(x_2-x_4)/(x_1-x_2)$. These two-cases correspond to motion of a pseudoparticle in the two different branches of a quartic pseudpopotential.

The other type of periodic solution arises when we find two real roots, x_1 and x_2, with $x_1>x_2$, and two complex roots for $f(x)$. Writing

$$f(x)=-\lambda^2(x-x_1)(x-x_2)(x^2-2\mu x+\nu), \tag{25}$$

the solution has the form, for $x_1\geqslant x\geqslant x_2$

$$x(z)=M_0+\frac{N_0}{D_0-\mathrm{cn}(\Omega_0z+\phi_0,k_0)}, \tag{26}$$

where cn signifies the Jacobi cosine amplitude. Defining

$$y_1=\sqrt{x_1^2-2\mu x_1+\nu}\quad\text{and}\quad y_2=\sqrt{x_2^2-2\mu x_2+\nu}, \tag{27}$$

we have

$$M_0=\frac{y_1x_2-y_2x_1}{y_1-y_2},$$

$$N_0=\frac{2y_1y_2(x_1-x_2)}{(y_1-y_2)^2},$$

$$D_0=\frac{y_1+y_2}{y_1-y_2},$$

$$\Omega_0=\lambda\sqrt{y_1y_2},$$

$$k_0=\sqrt{\frac{y_1y_2-x_1x_2+\mu(x_1+x_2)-\nu}{2y_1y_2}},$$

$$\phi_0=\mathrm{cn}^{-1}\left(\frac{D_0(x(0)-M_0)-N_0}{x(0)-M_0},k_0\right). \tag{28}$$

In this case the period of $x(z)$ has the form

$$T_0=\frac{4}{\Omega_0}\int_0^1\frac{dt}{\sqrt{(1-t^2)(1-k_0^2t^2)}}=\frac{4}{\Omega_0}K(k_0). \tag{29}$$

III. SEMICLASSICAL SOLUTIONS

Solving the semiclassical equations (3) for the field amplitudes numerically using a fourth- and fifth-order Runge-Kutta method also shows that the mean-field intensities undergo periodic revivals, as shown in Fig. 1. The horizontal axis is a normalized interaction distance, $\rho=\kappa z|\alpha(0)|/\sqrt{2}$. Note that there is no visible difference in the solutions for $\chi^{(3)}=10^{-7}$ whether we ignore the effects of the cross-phase modulation or set it to its maximum value, $\xi=\chi=\chi^{(3)}$, however when the Kerr nonlinearity is increased by an order of

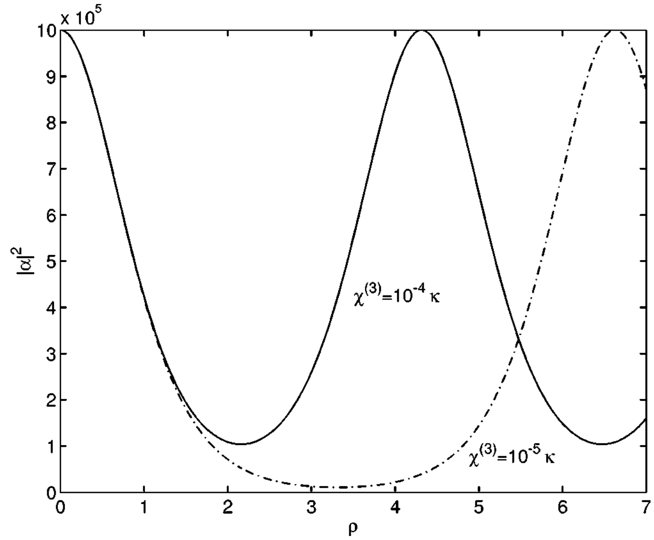

FIG. 1. The semiclassically calculated intensities of the fundamental as functions of the normalized interaction distance, ρ, for $|\alpha(0)|^2=10^6$, $\kappa=0.01$, and values of $\chi=\xi=10^{-7}$ and 10^{-6}. Note that all quantities plotted in the figures are dimensionless.

magnitude, the two solutions do become slightly different. When the $\chi^{(3)}$ component is increased even further, to 10^{-5}, as seen in Fig. 2, self-phase modulation makes a marked difference to the solutions, changing both the period and amplitude of the oscillations. Although the Kerr nonlinearity used in this example is rather high for nonlinear optical crystals, this result suggests that the oscillations between atomic and molecular condensates predicted in photoassociation of Bose-Einstein condensates [9] should be sensitive to the actual atom-atom, atom-molecule, and molecule-molecule scattering lengths, as these are typically huge compared to the nonlinearities found in optical systems.

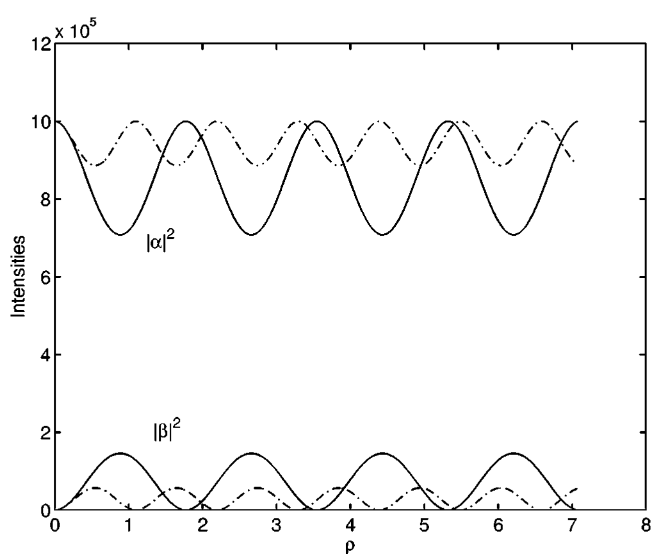

FIG. 2. The semiclassically calculated intensities of the fundamental and harmonic as functions of the normalized interaction distance, ρ, for $|\alpha(0)|^2=10^6$, $\kappa=0.01$, and $\chi=10^{-5}$. The full lines are for $\xi=\chi$, while the dash-dotted lines are without self-phase modulation.

The fact that the numerical solutions for the field amplitudes show oscillations is interesting in itself when compared with the semiclassical solution for the field amplitudes in traveling-wave SHG, which does not exhibit any periodicity [10], although writing equations for the field intensities leads to a periodic solution in terms of Jacobi elliptic functions [6]. The essential difference is that the semiclassical solutions for the field amplitudes in pure SHG stay real when we have phase matching, unlike the solutions with added $\chi^{(3)}$ component, which causes the phase of the amplitude variables to rotate. The real-valued solution for $\alpha(z)$ in pure SHG can go to zero, after which $\beta(z)$ cannot change, as the equation for β then becomes $d\beta/dz=0$. This can be further understood because in the pure-SHG case, β becomes negative and real, while α remains positive and real. This means that $d\alpha/dz \leqslant 0$ and $d\beta/dz \leqslant 0$, but with the phase rotation due to the χ^3 interaction, $d\alpha/dz$ can periodically become positive, hence the revivals in the fundamental. Quantum mechanically, there are always fluctuations present in the amplitudes in either case, which also prevents $\alpha(z)$ from reaching zero.

However, as has been shown previously [6,11,12], neither the analytical nor the numerical solutions of the classical equations allow us to reliably calculate any of the quantum statistics of the two fields after a certain interaction length. To do this we turn to one of the phase-space representations of quantum optics.

IV. QUANTUM PROPERTIES

Although the inclusion of what we would expect to be the maximum value of the cross-phase term makes no visible difference to the mean fields for small values of the Kerr nonlinearity [5], it is still of interest to investigate what effect it may have on the quantum statistics of the two fields. We can also investigate whether the quantum solutions for the mean-fields diverge from the semiclassical solutions as the Kerr nonlinearity is increased.

Using the usual methods [13], this system can be mapped exactly onto stochastic partial differential positive-*P* equations (note that we are using Itô calculus), via the master and Fokker-Planck equations. We find that, unlike the case with no cross-phase modulation, the positive-*P* Fokker-Planck equation for the system no longer has a diagonal diffusion matrix, which means that no simple and obvious factorization resulting in the stochastic differential equations suggests itself. However, the factorization we have chosen (which is by no means unique), leads to the following system of stochastic equations:

$$\frac{d\alpha}{dz}=\kappa\alpha^{\dagger}\beta-2i\chi\alpha^2\alpha^{\dagger}-2i\xi\alpha\beta^{\dagger}\beta+\sqrt{\frac{-2i}{\chi}}\xi\alpha\eta_1(z)+\sqrt{\kappa\beta-2i\alpha^2(\chi-\xi^2/\chi)}\,\eta_3(z),$$

$$\frac{d\alpha^{\dagger}}{dz}=\kappa\alpha\beta^{\dagger}+2i\chi\alpha^{\dagger 2}\alpha+2i\xi\alpha^{\dagger}\beta^{\dagger}\beta+\sqrt{\frac{2i}{\chi}}\xi\alpha^{\dagger}\eta_2(z)+\sqrt{\kappa\beta^{\dagger}+2i\alpha^{\dagger 2}(\chi-\xi^2/\chi)}\,\eta_4(z),$$

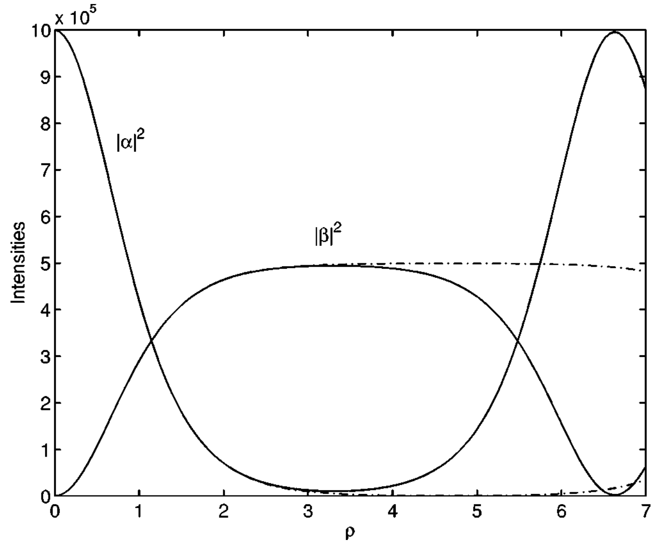

FIG. 3. The intensities of the fundamental and harmonic as functions of the normalized interaction distance, ρ, for $|\alpha(0)|^2=10^6$, $\kappa=0.01$, and $\chi=\xi=10^{-7}$, calculated using the positive-P representation. The dash-dotted lines are for $\chi^{(3)}=0$, the case of pure second-harmonic generation.

$$\frac{d\beta}{dz}=-\frac{\kappa}{2}\alpha^2-2i\chi\beta^2\beta^\dagger-2i\xi\alpha^\dagger\alpha\beta+\sqrt{-2i\chi\beta^2}\,\eta_1(z),$$

$$\frac{d\beta^\dagger}{dz}=-\frac{\kappa}{2}\alpha^{\dagger 2}+2i\chi\beta^{\dagger 2}\beta+2i\xi\alpha^\dagger\alpha\beta^\dagger+\sqrt{2i\chi\beta^{\dagger 2}}\,\eta_2(z), \tag{30}$$

where the noise terms $\eta_j(z)$ are real and Gaussian such that

$$\overline{\eta_j(z)\,\eta_k(z')}=\delta_{jk}\delta(z-z'). \tag{31}$$

Due to the independence of these noise terms, the variables α and $\alpha^\dagger$ [also $(\beta,\,\beta^\dagger)$] are not complex conjugate except in the mean of a large number of stochastic trajectories.

From numerical integration of these equations we find that the intensities of the two fields are not noticeably changed from the semiclassical solutions, whether $\xi=0$ or is equal to χ. This can be seen in Fig. 3, where we show the quantum solutions for the field intensities with $\chi=\xi=10^{-7}$, with the solutions for pure SHG given for purposes of comparison. The fact that inclusion of the quantum features does not invalidate the semiclassical predictions can be explained by the fact that it is the phase rotation that has the dominant effect on the dynamics and this is well described by the semiclassical equations. This type of effect is also apparent in the superchemistry of Bose-Einstein condensates, where as long as the coupling lasers are not too strong, the process of molecular photoassociation is well described by the semiclassical equations [9,14]. The solutions with Kerr nonlinearity present do not noticeably change here whether we include the cross-phase modulation or not. When we examine the variance of the $X_a(=\hat{a}+\hat{a}^\dagger)$ quadrature for the same parameters, as shown in Fig. 4, we see that there is slightly less squeezing available when the cross-phase term is introduced, although whether this difference would be significant in practice is difficult to judge. Both results with Kerr nonlinearity experience excess noise well before the variance for pure SHG begins to increase. The peak value of the variance, where the fields exhibit almost thermal statistics due to the semispontaneous nature of the downconversion process, is about 10% greater with the full value of ξ included. This peak value is so large that it cannot be shown in Fig. 4 while still leaving the amount of noise reduction visible.

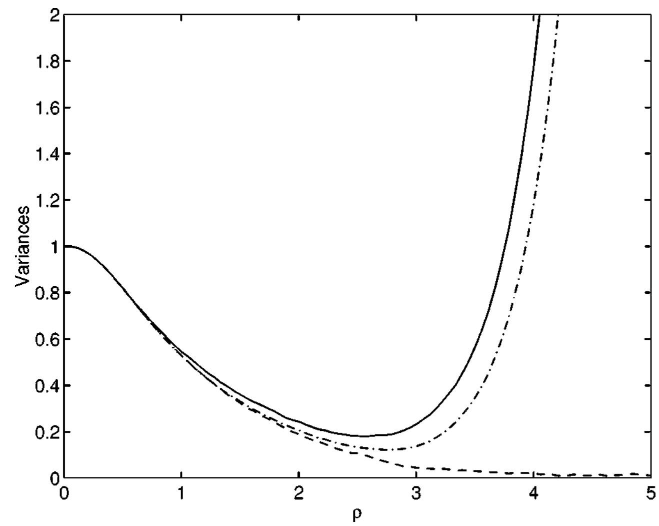

FIG. 4. The X_a quadrature variances, calculated using 10^5 stochastic trajectories, for $|\alpha(0)|^2=10^6$ and $\kappa=0.01$. The solid line is for $\chi=\xi=10^{-7}$, the dash-dotted line is for $\chi=10^{-7}$ and $\xi=0$, while the variance for pure SHG is shown by the dashed line.

When we increase the Kerr nonlinearity to 10^{-6}, the quantum solutions, shown in Fig. 5 are still indistinguishable from the semiclassical solutions, but, as can be seen, the

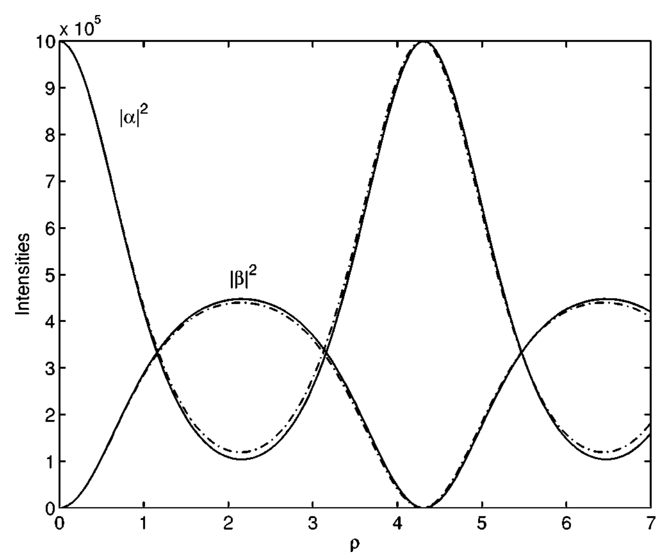

FIG. 5. The intensities of the fundamental and harmonic as functions of the normalized interaction distance, ρ, for $|\alpha(0)|^2=10^6$, $\kappa=0.01$, and $\chi=10^{-6}$, calculated using the positive-P representation. The solid line is for $\xi=10^{-6}$, while the dash-dotted line is for $\xi=0$.

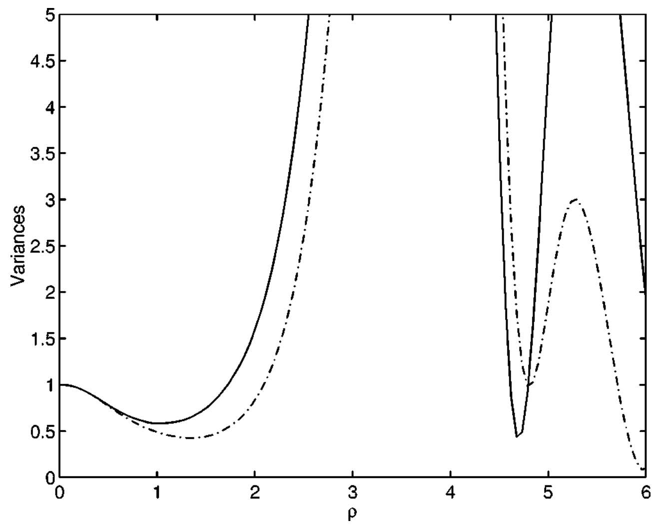

FIG. 6. The X_a quadrature variances, calculated using 10^5 stochastic trajectories, for $|\alpha(0)|^2=10^6$, $\kappa=0.01$, and $\chi=10^{-6}$. The solid line is for $\xi=10^{-6}$ and the dash-dotted line is for $\xi=0$.

addition of cross-phase modulation does make a perceptible difference to the mean intensities. For this value of nonlinearity there is significantly less quadrature squeezing present with cross-phase modulation, as shown in Fig. 6. The maximum value of the squeezing is now found at $\rho\approx 6$, well beyond the experimentally achievable parameter regimes. All the results of stochastic integration shown have sampling errors of typically less than 1%.

The fact that the addition of self-phase modulation decreases the degree of squeezing available can be explained by the fact that χ^2 and χ^3 processes introduce different types of dynamical phase matching and hence interfere with each other. As quadrature squeezing is phase sensitive, it naturally decreases. The further degree to which the cross-phase modulation decreases the squeezing can be understood as a shearing, rotation, and deformation of the contours of the Wigner function, a property of χ^3 processes [15]. When we look at Eq. (3), we can see that when $\xi=\chi$, both fields have the maximum value of phase-modulation nonlinearity, i.e., $2i\chi(|\alpha|^2+|\beta|^2)$, so that these contours will deform at their maximum rate. This naturally leads to an increase in the quadrature variances.

V. CONCLUSION

We have analyzed traveling-wave second-harmonic generation in the case where the nonlinear crystal has added $\chi^{(3)}$ nonlinearities, finding a semiclassical analytical solution for the generalized case where both self-phase and cross-phase modulation are present. We find marked differences between the dynamical behavior of the fields with and without the $\chi^{(3)}$ components. As the nonlinearity is increased, both the period and amplitude of the oscillations between the fundamental and harmonic are changed. This feature is also of relevance to proposals for coherent molecular photoassociation of Bose-Einstein condensates (BECs), where the self-interaction and cross-interaction terms are typically much larger than in optical systems. Although BECs are not single-mode systems, the effects we find here should be present at least in a qualitative sense. As far as the quantum statistics of the fields are concerned, we find that less squeezing is achievable in the $\chi^{(3)}$ case, with the addition of cross-phase modulation worsening the squeezing as the modulation strength increases.

As all materials have some $\chi^{(3)}$ component, and the ratios of $\chi^{(3)}/\chi^{(2)}$ that we have used are typical of nonlinear media, from optical crystals to BEC, it is of interest to know what the signatures of this component are. We have found several signatures that should be accessible to experimental observation and have shown that if it is either a maximum of noise supression or large amplitude oscillations that is sought, materials should have as small a $\chi^{(3)}$ component as possible.

ACKNOWLEDGMENTS

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