Quantum-theoretical treatments of three-photon processes

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We perform and compare different analyses of triply degenerate four-wave mixing in the regime where three fields of the same frequency interact via a nonlinear medium with a field at three times the frequency. As the generalized Fokker-Planck equation (GFPE) for the positive-P function of this system contains third-order derivatives, there is no mapping onto genuine stochastic differential equations. Using techniques of quantum field theory, we are able to write stochastic difference equations that we may integrate numerically. We compare the results of this method with those obtained by the use of approximations based on semiclassical equations, and on truncation of the GFPE leading to stochastic differential equations. In the region where the difference equations converge, the stochastic methods agree for the field intensities, but give different predictions for the quantum statistics.

I. INTRODUCTION

The theoretical study of the interaction of electromagnetic waves via a nonlinear medium has a long history, going back at least to the groundbreaking paper of Armstrong *et al.* [1], wherein a classical treatment was performed for the processes of second- and third-harmonic generation, degenerate and nondegenerate down-conversion, and four-wave mixing. Experimentally, second-harmonic generation (SHG) and parametric down-conversion are well-known sources of quantum states of the electromagnetic field. Third-harmonic generation (THG), wherein input fields at frequency ω produce output fields at frequency 3ω is a process that has been observed experimentally in a number of different situations. An early experiment [2] produced both third- and fifth-harmonic light at the interface of glass and liquids and it was suggested that odd-multipole generation may be a widespread phenomenon. THG has been observed in a number of other situations, for example, in the interaction of laser light with a nematic liquid-crystal cell [3], and in the interaction of pulsed light from an Nd:YAG (yttrium aluminum garnet) laser with organic vapors [4] and with polyimide films [5].

As all the processes mentioned above are highly nonlinear, a full quantum-theoretical treatment is often difficult without resorting to the phase-space representations of quantum optics, generally the positive-P [6] or Wigner representations [7]. In the usual approach, the system Hamiltonian is mapped onto a Fokker-Planck equation for the particular pseudoprobability distribution being used, which may either be solved directly or further mapped onto a set of stochastic differential equations [8,9]. As the usual methods only allow for the mapping of genuine Fokker-Planck equations, that is, equations with derivatives of no higher than second order (this is the content of Pawula's theorem [10]), onto stochastic differential equations, the systems that can be investigated using this approach are limited. There is at least one other known method for the derivation of stochastic differential equations for interacting bosonic systems [11]. Although this method gave enhanced stability in the numerics over the normal positive P, it was assumed at the beginning of the deri-

vation that the time development could be modeled by Itô stochastic differential equations, so that it is subject to the limitations of Pawula's theorem when it comes to the representation of third-order noises.

The present situation of triply degenerate four-wave mixing results in generalized Fokker-Planck equations of third or higher order and hence Langevin equations may not be derived. A common procedure in these cases is to use a Wigner equation truncated at second order, which is equivalent to the semiclassical theory of stochastic electrodynamics [12]. This procedure necessarily discards the deeper quantum aspects of the problem and gives answers at odds with quantum mechanics for several systems [13,14]. There are also situations where even a P-representation Fokker-Planck equation must also be written in generalized form [15–18], and more are likely to be investigated in the future. The present problem is one of these situations.

Using the techniques of quantum field theory, we have previously developed methods to represent this class of problems using stochastic difference equations [18–20]. These equations may be numerically integrated using computers, so that the fact that stochastic differential equations cannot be defined for these systems is not a problem. As the problem of triply degenerate four-wave mixing has a Hamiltonian cubic in creation and annihilation operators, it results in a generalized Fokker-Planck equation with third-order derivatives for the positive-P pseudoprobability distribution. We note here that a related problem, namely, that of intracavity third-harmonic generation, has previously been dealt with [15,16]. In Ref. [15], the quantum properties of the fields are analyzed in a linearized approximation, Ref. [16] looks at macroscopic quantum interference. In both cases the steady states, which are natural for a resonator, rather than transients, which are natural in the traveling-wave case, are investigated. It should also be noted that there are certain technical problems with Refs. [15,16]. It appears the authors were unaware that the stochastic equations used could not, in fact, be considered Langevin equations in the formal sense, and that these should be considered stochastic difference equations. Furthermore, the third-order noise contribution to

these was not presented constructively. In this paper we will also investigate the related process of spontaneous triplet-photon production. This process may be thought of as deeply quantum as, without the third-order noises, it may not proceed. Intracavity three-photon down-conversion has previously been investigated using the density matrix to calculate the Wigner function [21] and a quantum Monte Carlo method to simulate homodyne detection of the output fields [22].

We compare the results obtained using our generalized positive-P approach with those obtained using different approximations. Based on approximately factorizing certain quantum averages, we can find analytic solutions for the mean intensities in terms of Jacobi elliptic functions, although this approach can tell us nothing about the quantum properties of the fields, and, worse than this, has been shown to be inaccurate after a certain interaction length in other parametric processes [23,24]. Two other approximations used require truncation of the differential equations for the Wigner and positive-P pseudoprobability functions at second order, resulting in Fokker-Planck equations that may then be mapped onto stochastic differential equations.

II. HAMILTONIAN AND SEMICLASSICAL SOLUTIONS

The generic system that we consider is one of four-wave mixing in a nonlinear medium, where three of the waves have frequency ω and the fourth has frequency 3ω. In our analysis we will make the approximation that only two modes are important, which necessarily ignores such effects as group-velocity dispersion, but will serve to demonstrate our method in the most simple way possible. Traveling-wave third-harmonic generation and degenerate three-photon down-conversion can then be described by the interaction Hamiltonian

$$\mathcal{H}(z) = \frac{i\hbar\kappa}{3}[\hat{a}^{\dagger 3}\hat{b} - \hat{a}^3\hat{b}^\dagger], \qquad (1)$$

where $\hat{a}(z)$ and $\hat{b}(z)$ are the annihilation operators for quanta at frequencies ω and 3ω, respectively, at position z inside the nonlinear medium, and κ represents the effective couping between the modes.

From the above Hamiltonian we can immediately derive the Heisenberg operators of motion for the operators

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

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

$$\frac{d\hat{b}}{dz} = -\frac{\kappa}{3}\hat{a}^3,$$

$$\frac{d\hat{b}^\dagger}{dz} = -\frac{\kappa}{3}\hat{a}^{\dagger 3}, \qquad (2)$$

which, being nonlinear operator equations, have no obvious analytical solution. In the case of three-photon down-conversion, as long as we are interested in the regime where the number of photons in the high-frequency field is almost unchanged, we may use the undepleted pump approximation, as is common for the optical parametric amplifier [25,26]. This involves treating the product of κ and $\hat{b}$ as a complex number and leads to the following equations of motion:

$$\frac{d\hat{a}}{dz} = \chi \hat{a}^{\dagger 2},$$

$$\frac{d\hat{a}^\dagger}{dz} = \chi \hat{a}^2, \qquad (3)$$

where $\chi = \kappa \langle \hat{b}(0) \rangle$.

Naively replacing the operator products in Eq. (3) with c numbers leads to the prediction that spontaneous three-photon down-conversion will not occur. However, by setting $\hat{N} = \hat{a}^\dagger \hat{a}$, we can find a second-order differential equation for $\hat{N}(z)$,

$$\frac{d^2\hat{N}}{dz^2} = \chi^2(6\hat{N}^2 + 6\hat{N} + 3). \qquad (4)$$

If we now assume that all operator products factorize and exchange operators for c numbers, we immediately see that, even starting from the vacuum, the process may proceed. However, what we also see is that the photon number, beginning from zero, will increase without limit. In fact, it has been shown that it will become infinite in a finite time [27]. We will, therefore, allow for pump depletion and find solutions for the photon number following the same methods.

When we consider the system with depletion of the pump field, we can find solutions for the intensities in both third-harmonic generation and three-photon down-conversion, although these are of no help if we wish to calculate the quantum statistics of the fields. These solutions, in terms of Jacobi elliptic functions, are fully periodic and are given in the Appendix. To investigate the quantum properties of the fields and the extent of the semiclassically predicted revivals, we will revert to numerical stochastic integration using phase-space techniques, comparing the results of a full representation of the problem using a generalized positive-P representation with those obtained by truncation of the equations in the positive-P and Wigner representations.

III. THE STOCHASTIC APPROACH

A. Truncated positive-P equations

Following the usual procedures [9], we may map the system Hamiltonian of Eq. (1) onto a generalized Fokker-Planck equation for the positive-P distribution

$$\frac{\partial P}{\partial z} = \left\{ -\left[\frac{\partial}{\partial \alpha}(\kappa \alpha^{*2}\beta) + \frac{\partial}{\partial \alpha^*}(\kappa \alpha^2 \beta^*)\frac{\partial}{\partial \beta}\left(-\frac{\kappa}{3}\alpha^3 \right) \right. \right.$$

$$\left. + \frac{\partial}{\partial \beta^*}\left(-\frac{\kappa}{3}\alpha^{*3} \right) \right] + \frac{1}{2}\left[\frac{\partial^2}{\partial \alpha^2}(2\kappa \alpha^* \beta) \right.$$

$$\left. + \frac{\partial^2}{\partial \alpha^{*2}}(2\kappa \alpha \beta^*) \right]$$

$$\left. -\frac{1}{6}\left[\frac{\partial^3}{\partial \alpha^3}(2\kappa \beta) + \frac{\partial^3}{\partial \alpha^{*3}}(2\kappa \beta^*) \right] \right\}$$

$$\times P(\alpha, \alpha^*, \beta, \beta^*, z). \tag{5}$$

In the above equation, there is a correspondence between the c-number variables $[\alpha, \alpha^*, \beta, \beta^*]$ and the operators $[\hat{a}, \hat{a}^\dagger, \hat{b}, \hat{b}^\dagger]$ of the interaction Hamiltonian. Equation (5) is not a genuine Fokker-Planck equation as it contains derivatives of higher than second order. There are two features that are immediately noticeable. The first is that the coefficients of the third-order derivatives are not obviously small and, in fact, may be larger than those of the second-order terms under some circumstances. The second point is that the process of spontaneous triple down-conversion depends on the presence of the third-order terms. However, by making the same type of approximation that is often used in the Wigner representation, we may truncate Eq. (5) at second order and find the following set of stochastic differential equations in Itô calculus:

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

$$\frac{d\alpha^\dagger}{dz} = \kappa \alpha^2 \beta^\dagger + \sqrt{2\kappa \alpha \beta^\dagger}\,\eta_2(z),$$

$$\frac{d\beta}{dz} = -\frac{\kappa}{3}\alpha^3,$$

$$\frac{d\beta^\dagger}{dz} = -\frac{\kappa}{3}\alpha^{\dagger 3}, \tag{6}$$

where the two noise terms have the properties that $\overline{\eta_i(z)\eta_j(z')} = \delta_{ij}\delta(z-z')$. As always with the positive P approach, the pairs of field variables (α and $\alpha^\dagger$, for example) are not complex conjugate except in the mean of a large number of integrated trajectories. This set of equations can now be integrated numerically and means taken over a large number of trajectories to calculate any desired normally ordered operator product.

B. Truncated Wigner equations

In an exact treatment, it is a matter of choice which stochastic representation to use. This is no longer the case when approximations are invoked. We will consider, as an example, a truncated Wigner representation for the system in

question. Again following the usual methods [9], a partial differential equation is found for the Wigner distribution of the system

$$\frac{\partial W}{\partial z} = \frac{\kappa}{3}\left\{ -\left[\frac{\partial}{\partial \alpha}(3\alpha^{*2}\beta) + \frac{\partial}{\partial \alpha^*}(3\alpha^2 \beta^*) - \frac{\partial}{\partial \beta}(\alpha^3) \right. \right.$$

$$\left. -\frac{\partial}{\partial \beta^*}(\alpha^{*3}) \right] - \frac{1}{6}\left[\frac{\partial^3}{\partial \alpha^3}\left(\frac{3}{2}\beta \right) + \frac{\partial^3}{\partial \alpha^{*3}}\left(\frac{3}{2}\beta^* \right) \right.$$

$$\left. + \frac{\partial^3}{\partial \alpha^2 \partial \beta^*}\left(\frac{9}{2}\alpha^* \right) \right.$$

$$\left. \left. + \frac{\partial^3}{\partial \alpha^{*2}\partial \beta}\left(\frac{9}{2}\alpha \right) \right] \right\} W(\alpha, \alpha^*, \beta, \beta^*, z). \tag{7}$$

As this equation also has derivatives of third order, it also cannot be mapped onto stochastic differential equations. However, it is possible to drop the third-order terms to find two coupled deterministic equations with no noise terms, in an approximation equal to stochastic electrodynamics. In this case the noise comes from the initial conditions for the two modes, as we integrate the equations

$$\frac{d\alpha}{dz} = \kappa \alpha^* \beta,$$

$$\frac{d\beta}{dz} = -\frac{\kappa}{3}\alpha^3, \tag{8}$$

a large number of times with the initial conditions chosen appropriately from the Wigner pseudoprobability distribution. This can be thought of as equivalent to adding vacuum fluctuations equal to half a photon to each mode.

C. Stochastic difference equations

As the above approaches use a truncation of the full equations and hence will necessarily result in the loss of some information, we wish to map the full interaction Hamiltonian onto some system that can be solved. Following the methods developed in Ref. [19], and used in Refs. [18,20], we may develop stochastic difference equations, which, although they have no continuous limit as stochastic differential equations, may be simulated numerically.

Generally speaking, c-number techniques of quantum stochastics are *mappings* of quantum problems onto c-number stochastic problems, such that quantum expectation values of interest equal certain classical averages. A well-known example of such quantum-classical mapping is the positive-P representation [6], in which time-normal quantum averages of the Heisenberg field operators are mapped onto corresponding stochastic averages. For a two-mode system,

$$\langle T_- \hat{a}^\dagger(z) \cdots \hat{b}^\dagger(z') \cdots T_+ \hat{a}(z'') \cdots \hat{b}(z''') \cdots \rangle$$

$$= \overline{\alpha^\dagger(z) \cdots \beta^\dagger(z') \cdots \alpha^\dagger(z'') \cdots \beta(z'') \cdots}. \tag{9}$$

(In our case, z is a position variable; we, however, prefer to use the conventional term *time-normal average* for the left-hand side of this equation.) The upper bar in Eq. (9) denotes an averaging over the statistics of the four c-number fields $\alpha(z), \beta(z), \alpha^\dagger(z), \beta^\dagger(z)$. These statistics are characterized via stochastic differential equations for the c-number fields. There are rare exceptions (such as linear systems coupled to heat baths) where $\alpha^\dagger(z) = \alpha^*(z)$, $\beta^\dagger(z) = \beta^*(z)$ may be maintained, but in general having a genuine positive probability in Eq. (9) requires doubling of the phase space.

The positive-P mapping is (at least in principle) exact, yet it is only applicable to problems where interactions are no more than quadratic in creation or annihilation operators. A much wider class of arbitrary polynomial interaction Hamiltonians may be covered if we allow the mapping (9) to hold only approximately, by considering classical stochastic processes in discretised time or, as in the present case, space. We thus assume that z is a discrete variable changing in finite steps of Δz. The classical fields obey the generic system of stochastic *difference* equations, [with $\Delta \alpha(z) = \alpha(z + \Delta z) - \alpha(z)$ and likewise for the rest of the variables]

$$\Delta \alpha(z) = \sigma_1(z), \quad \Delta \alpha^\dagger(z) = \sigma_1^\dagger(z),$$

$$\Delta \beta(z) = \sigma_2(z), \quad \Delta \beta^\dagger(z) = \sigma_2^\dagger(z). \quad (10)$$

The techniques of Ref. [19] then allow one to derive the random sources (i.e., the σ's) directly from the interaction Hamiltonian.

The derivation of the sources may be divided into two major steps. First, stochastic cumulants of the σ's are obtained via quantum-field-theoretical techniques. In the standard phase-space techniques [9] this corresponds to the derivation of the drift terms and the noise matrix. The latter are nothing but the first- and second-order cumulants of the sources; whereas failure of the phase-space techniques manifests itself as the presence of higher-order cumulants. Secondly, Hubbard-Stratonovich transformations (HST) [28] are used to find explicit expressions for the sources (i.e., to restore the sources from their cumulants). This step corresponds to factorization of the noise matrix in the standard techniques. We shall see that using the HST virtually trivializes this factorization, and hence can also be of much assistance in the usual phase-space approaches.

Although the rigorous derivation of the cumulants is rather involved [19], in practice it boils down to a very simple recipe. One starts by writing the interaction Hamiltonian in normally ordered form,

$$\mathcal{H} = :h(\hat{a}^\dagger, \hat{b}^\dagger, \hat{a}, \hat{b}):, \quad (11)$$

where $h(a^\dagger, b^\dagger, a, b)$ is a c-number function of four c-number arguments. For the present interaction,

$$h(a^\dagger, b^\dagger, a, b) = \frac{i\hbar\kappa}{3}(a^{\dagger 3}b - a^3 b^\dagger), \quad (12)$$

the normal ordering in Eq. (11) is in fact redundant. The properties of the σ sources are given by the formulas (dropping the z dependence for notational simplicity)

$$\overline{\exp\left[\sum_z (\xi_1^\dagger \sigma_1 + \xi_2^\dagger \sigma_2 + \xi_1 \sigma_1^\dagger + \xi_2 \sigma_2^\dagger)\right]}$$

$$= \exp\sum_z \overline{s_{\mathrm{int}}(\xi_1, \xi_2, \xi_1^\dagger, \xi_2^\dagger, \alpha, \beta, \alpha^\dagger, \beta^\dagger)}, \quad (13)$$

$$s_{\mathrm{int}}(\xi_1, \xi_2, \xi_1^\dagger, \xi_2^\dagger, \alpha, \beta, \alpha^\dagger, \beta^\dagger)$$
$$= -i\hbar^{-1}\Delta z h(\alpha^\dagger + \xi_1^\dagger, \beta^\dagger + \xi_2^\dagger, \alpha, \beta) + \mathrm{conj}. \quad (14)$$

Here, the upper bar denotes averaging over the statistics of the sources, the ξ's are four arbitrary c-number functions, and "conj" acts as a formal Hermitian conjugation, interchanging quantities with and without dagger, $\alpha(z) \leftrightarrow \alpha^\dagger(z)$, and likewise for the rest, and complex conjugating other c numbers. For the Hamiltonian (1), we have

$$s_{\mathrm{int}} = \Delta z\left[\xi_1^\dagger \kappa \alpha^{\dagger 2}\beta + \xi_1^{\dagger 2}\kappa \alpha^\dagger \beta + \xi_1^{\dagger 3}\frac{\kappa}{3}\beta + \xi_2^\dagger\left(-\frac{\kappa}{3}\alpha^3\right)\right]$$
$$+ \mathrm{conj}. \quad (15)$$

This completes the first step of the derivation, which results in the characteristic functional of the averages of the sources, Eq. (13), expressed in terms of the cumulants, Eq. (15).

The deterministic terms in the resulting equations of motion are immediately obvious, being those to first order in the ξ's in Eq. (15) [and exactly the same as in Eqs. (6), as they must be]. The noise contributions to the σ's are readily found using real and complex Hubbard-Stratonovich transformations, respectively,

$$\overline{\exp\frac{x^2}{2}} = \overline{\exp x\chi}\left[\frac{x^2}{2} \xrightarrow{x} x\chi\right], \quad (16)$$

$$\overline{\exp(xy)} = \overline{\exp(x\eta + y\eta^*)}[xy \Rightarrow x\eta + y\eta^*]. \quad (17)$$

Here, x and y are arbitrary quantities, and χ and η, respectively, are the real and complex standardized Gaussian random variables, with the properties $\overline{\chi} = \overline{\eta} = \overline{\eta^2} = 0$, $\overline{\chi^2} = \overline{|\eta|^2} = 1$. The formulas in square brackets introduce a convenient short hand for the real and complex HSTs. In this short hand, applying a real HST to the quadratic term in Eq. (15) yields

$$\Delta z \xi_1^{\dagger 2}\kappa \alpha^\dagger \beta \xrightarrow{\chi_1} \xi_1^\dagger \chi_1 \sqrt{2\kappa \alpha^\dagger \beta \Delta z}. \quad (18)$$

Comparing this to Eq. (13), the second-order contribution to σ_1 is found to be

$$\sigma_1^{(2)} = \chi_1 \sqrt{2\kappa \alpha^\dagger \beta \Delta z}. \quad (19)$$

Divided by Δz, this is exactly the η_1 term in Eqs. (6) (more precisely speaking, this is what the η_1 term becomes with discretization). The corresponding contribution to $\sigma_1^\dagger$ is found by formally conjugating Eq. (19)

$$\sigma_1^{\dagger(2)} = \chi_1^{\dagger}\sqrt{2\kappa\alpha\beta^{\dagger}\Delta z}. \tag{20}$$

The noise sources χ_1 and $\chi_1^{\dagger}$ are uncorrelated and the sources introduced at different z are also uncorrelated. More subtle choices may be possible (cf. Ref. [29]), but we will not consider them here. Note that by construction the $\chi_1, \chi_1^{\dagger}$ pair at z must be uncorrelated with the fields at the same z. This is equivalent to saying that Eqs. (10) imply (a discretized variety of) Itô calculus. This conjecture is actually an oversimplification: with the α's and β's being solutions of the stochastic Eqs. (10), the HSTs at different positions are not independent. The reason why this conjecture nevertheless leads to the right answer, is that, due to the causal and Markovian nature of the classical process, it is only the HST at the most recent position that is important for the correct unravelling of the process. For a detailed argument, see Refs. [19,30].

The cubic term in s_{int}, $\Delta z \xi_1^{\dagger 3}\kappa\beta/3$, is also simply taken care of, by applying a complex HST followed by a real one,

$$\Delta z \xi_1^{\dagger 3}\frac{\kappa\beta}{3} \overset{\eta}{\Rightarrow} \Delta z \xi_1^{\dagger 2}\frac{p}{2}\,\eta + \xi_1^{\dagger}q\,\eta^*, \tag{21}$$

$$\overset{\chi_2}{\rightarrow} \xi_1^{\dagger}(\chi_2\sqrt{p\,\eta\Delta z} + q\,\eta^*). \tag{22}$$

After accounting for the conjugated cubic term, we finally obtain the set of coupled stochastic difference equations

$$\Delta\alpha = \kappa\alpha^{\dagger 2}\beta\Delta z + \chi_1\sqrt{2\kappa\alpha^{\dagger}\beta\Delta z} + \chi_2\sqrt{p\,\eta\Delta z} + q\,\eta^*,$$

$$\Delta\alpha^{\dagger} = \kappa\alpha^2\beta^{\dagger}\Delta z + \chi_1^{\dagger}\sqrt{2\kappa\alpha\beta^{\dagger}\Delta z} + \chi_2^{\dagger}\sqrt{p^{\dagger}\,\eta^{\dagger}\Delta z} + q^{\dagger}\,\eta^{\dagger *},$$

$$\Delta\beta = -\frac{\kappa}{3}\alpha^3\Delta z,$$

$$\Delta\beta^{\dagger} = -\frac{\kappa}{3}\alpha^{\dagger 3}\Delta z, \tag{23}$$

with the p's and q's constrained by the conditions

$$pq = \frac{2\kappa\beta}{3}, \quad p^{\dagger}q^{\dagger} = \frac{2\kappa\beta^{\dagger}}{3} \tag{24}$$

due to our choice of complex transformations, but otherwise arbitrary. Equations (23) contain four real ($\chi_1, \chi_2, \chi_1^{\dagger}, \chi_2^{\dagger}$) and two complex ($\eta, \eta^{\dagger}$) Gaussian noises. These are now $\delta_{zz'}$ (Kronecker) correlated, unlike η_1, η_2 in Eqs. (6) which are $\delta(z-z')$ (Dirac) correlated. We note here that Eq. (23) without the third-order noises has a natural continuous limit identical to the positive-P equations obtained above via the usual methods, but that the derivation is much shorter. Indeed, leaving bookeeping aside, the actual derivation consists of calculating s_{int} using Eq. (14), and then processing it as per Eqs. (18), (21), and (22); all this takes no more than three lines. For situations with noises of less than third order, this method of finding the equations is almost trivial as compared to proceeding via the master and Fokker-Planck equations.

As in the normal positive-P method, which has a freedom in the choice of noise terms constrained only by the factorization of the diffusion matrix of the Fokker-Planck equation, there are other possible choices for the noises in our difference equations, following on from the factorization of Eq. (21). An obvious degree of freedom is in the choice of the p's and q's, which may be used to reduce the sampling noise in the stochastic integration of the difference equations. In the numerical integrations we performed, we simply set

$$p = q = \sqrt{\frac{2\kappa\beta}{3}}, \quad p^{\dagger} = q^{\dagger} = \sqrt{\frac{2\kappa\beta^{\dagger}}{3}}. \tag{25}$$

Using the said freedom in order to get better behavior of the numerical integration remains a subject for further investigation.

IV. RESULTS AND COMPARISON OF METHODS

In the case of third-harmonic generation, numerical integration of the classical equations of motion for the field amplitudes shows a complete and irreversible conversion to the high-frequency mode after some interaction length, as in traveling-wave second-harmonic generation. In the case of spontaneous down-conversion, this approach predicts that the system will remain in the initial state, regardless of the interaction length. The next approximation which we used, that of beginning with Heisenberg equations of motion for the field intensities and progressing to classical second-order differential equations, predicts fully periodic behavior in both cases. As it has previously been seen that both these methods give misleading results in the case of pure SHG and spontaneous down-conversion, and in any case do not allow for the calculation of the statistical properties of the fields, we have resorted to stochastic integration in the phase space representations.

The three phase-space methods we used, the truncated Wigner, a positive-P truncated at second order, and a generalized positive P that allows the modeling of higher-order noises, give the same results for the mean-field intensities, at least in the region where the generalized positive-P integration converged. This method is less stable than even the normal positive-P approach, and allowed for integration over approximately 60% of the range shown in Fig. 1. As the fundamental intensity began to revive, some trajectories underwent huge and sudden excursions, something also seen in Ref. [18]. Tuning the noise terms helped a little, but it was not possible to proceed far enough to see the form of the partial revival shown in Fig. 1. For all three methods we used initial conditions of $|\alpha(0)|^2 = 10^6$ in a coherent state, $|\beta(0)|^2 = 0$ and $\kappa = 10^{-5}$ for the results presented in the figures. These parameters were chosen purely for computational convenience, so that it was possible to see the main features of the behavior without the time required for the numerical integration becoming impractically long. What is immediately obvious from the figure is that the revival in the intensity of the fundamental is less marked than in traveling-wave SHG, possibly due to the fact that a three-photon process
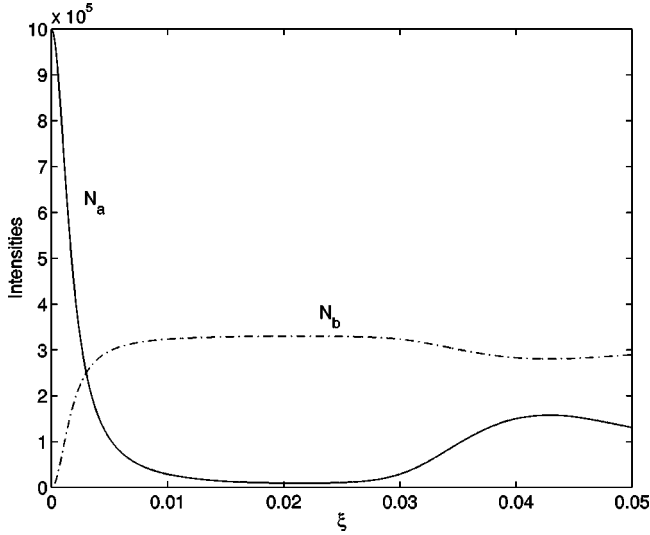
FIG. 1. The development of the intensities of the two fields as they traverse the nonlinear medium. The horizontal axis is a scaled interaction distance $\xi = \kappa |\alpha(0)| z$ and the quantities plotted in this and subsequent graphs are dimensionless. This plot is the result of 1.85×10^5 stochastic trajectories in the truncated positive-P representation. Results found by the other stochastic methods were indistinguishable until the generalized positive-P representation failed. (In this realization at $\xi \approx 0.032$.)

places stronger requirements on the coherence properties of the fields.

In the process of spontaneous three-photon down-conversion, both the positive-P and truncated Wigner representations predict that no mean field will appear at the fundamental, starting with only the third-harmonic mode occupied. While the Wigner result showed a lot of noise in the integration, the mean field averaged out to zero, indicating that this process cannot be explained as a result of vacuum fluctuations and hence cannot be adequately described using stochastic electrodynamics. The generalized positive P began to predict spontaneous down-conversion, but was extremely unstable and quickly diverged, so was not really a viable option for the investigation of this process.

In the case of THG, we found differences between the representations when we began to investigate the statistics of the fields. We calculated the variances in the X (defined as $\hat{a} + \hat{a}^\dagger$) quadratures of the two fields, as well as the Fano factors, i.e.,

$$F(N_{a,b}) = \frac{V(N_{a,b})}{\langle N_{a,b} \rangle}, \qquad (26)$$

where in both cases a coherent state exhibits a value of one. A value of less than one signifies decreased fluctuations from the coherent state (or vacuum) value. As our input state $[\alpha(0)]$ is real, it is the X quadrature that exhibits the maximum of noise suppression. Interestingly enough, the truncated positive-P and truncated Wigner methods gave almost indistinguishable results for these quantities, both being overoptimistic with regard to the maximum amount of squeezing available, as can be seen from Figs. 2 and 3.
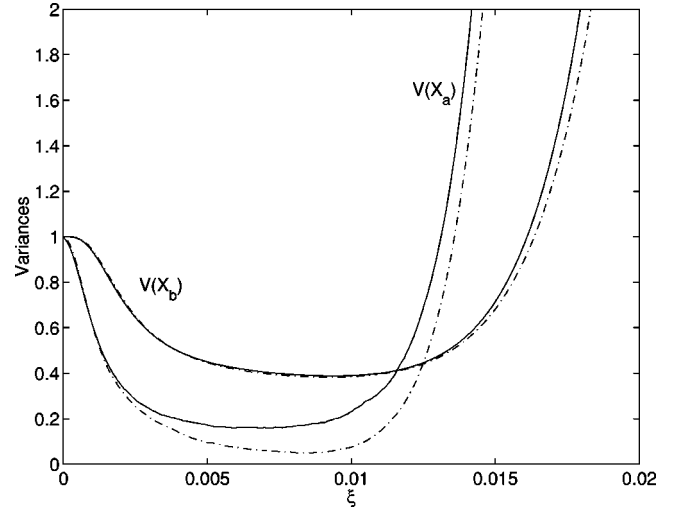

FIG. 2. The development of the X quadrature variances of the two fields as they traverse the nonlinear medium. The solid lines were calculated using the generalized positive-P representation and are the result of 4.6×10^5 trajectories. The dash-dotted lines were calculated using the truncated positive-P representation, with 1.85×10^5 trajectories.

Where the prediction was for excess noise above the vacuum level, all three methods showed good agreement. The actual form of the squeezing in both quadrature and intensity is very reminiscent of traveling-wave SHG [23], with excess noise being seen as the fundamental begins to revive. This is a result of the partially spontaneous nature of the down-conversion process necessary for this revival, and results in fields that exhibit almost thermal statistics.

We also integrated the equations for third-harmonic generation numerically in the three representations, for parameters ranging from $\kappa = 10^{-5} \rightarrow 10^{-7}$ and $\alpha(0) = 10^2 \rightarrow 10^3$.
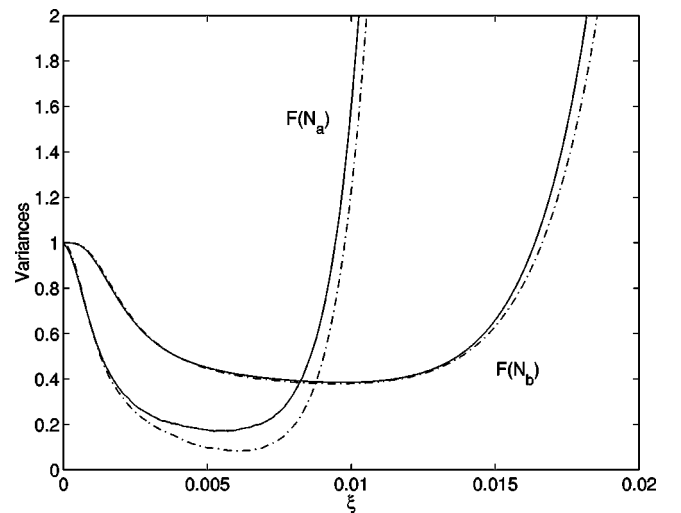

FIG. 3. The development of the Fano factors of the two fields as they traverse the nonlinear medium. The solid lines were calculated using the generalized positive-P representation and the dash-dotted lines were calculated using the truncated positive-P representation, with numbers of trajectories as in Fig. 2.
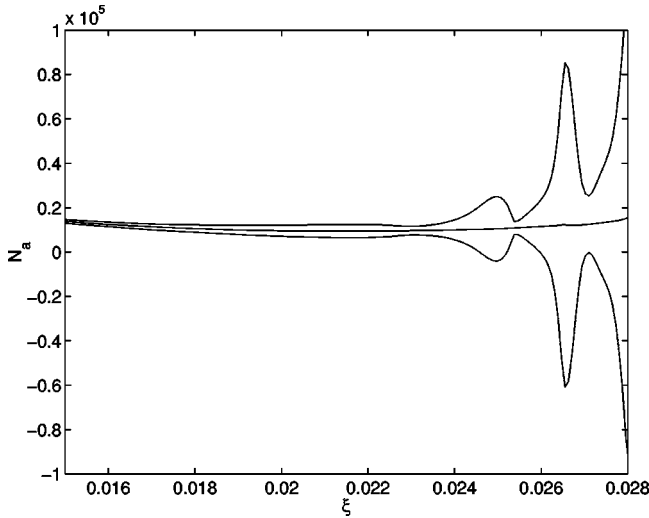
FIG. 4. The sampling errors for N_a in the generalized positive-P representation as it nears a divergence, calculated using 5×10^3 trajectories. The center line is the mean value while the outer ones represent plus or minus one standard deviation.

We found good agreement for the mean intensities for all the representations, where there was convergence of the generalized positive-P method. The generalized positive-P approach always predicted less maximum noise suppression, with the general form of the intensities and variances being the same over the range of parameters examined. We were not able to achieve convergence of the generalized positive-P method for long enough to see a revival in the fundamental. What happened is that a number of trajectories began to undergo rapid and arbitrarily large excursions before this point, causing divergence of the integration. This can be seen in the sampling errors plotted in Fig. 4, which increase dramatically near the end of the interval. This eventual failure of the integration is reminiscent of problems with the positive-P representation in highly nonlinear, undamped systems and it may be that, just as recent works have exhibited some success in attacking this problem [29,31], ways can be found to make the generalized positive-P method more convergent.

V. CONCLUSION

We have performed a fully quantum-mechanical analysis of the process of traveling-wave third-harmonic generation and compared the results obtained with those obtained using approximate, but more stable methods. The first approximation used, that of writing second-order differential equations for the field intensities, predicts periodic behavior with full revivals of the fundamental. The Wigner and positive-P equations, both truncated at second order, predict only a partial revival but agree with the full quantum predictions of the generalized positive-P method where the integration of the latter converges. They do, however, predict significantly more squeezing in the output fields than would seem to be the case.

In the process of spontaneous triplet-photon production, none of these methods proved to be useful for calculating the statistical properties of the fields. The semiclassical equations for the intensities again predict periodic behavior, but can say nothing about the statistics. This process simply will not proceed in the truncated representations. While the generalized positive-P representation begins to show spontaneous down-conversion, it rapidly falls victim to enormous sampling errors. Whether the freedom we have in writing the noise terms can be successfully used to overcome this problem is a subject for further investigation.

APPENDIX: DERIVATION OF SEMICLASSICAL SOLUTIONS

Beginning with the Heisenberg equations of motion (2) and proceeding as in Sec. II, with $\hat{N}_{a,b}$ being the number operators for the two modes, we find the nonlinear operator equation,

$$\frac{d^2\hat{N}_a}{dz^2} = \kappa^2 \left[(6\hat{N}_a^2 + 6\hat{N}_a + 4)\hat{N}_b - \frac{2}{3}\hat{N}_a^3 + 2\hat{N}_a^2 - \frac{4}{3}\hat{N}_a \right], \tag{A1}$$

which has no obvious solution. Assuming all products factorize and replacing the operator expressions by their expectation values, $(N_{a,b} = \langle \hat{N}_{a,b} \rangle)$, gives an equation in two real variables. We now have two different initial conditions, depending on which process we wish to investigate.

1. Third-harmonic generation

In pure third-harmonic generation, the initial condition is that $N_a(0) \neq 0$ and $N_b(0) = 0$. Using conservation of energy within the nonlinear material (we are not considering any other processes), we may write $N_b(z) = [A - N_a(z)]/3$, where $A = N_a(0)$. This allows us to write an equation in terms of N_a only

$$\frac{d^2N_a}{dz^2} = -\frac{\kappa^2}{3}(8N_a^3 - 6AN_a^2 - 6AN_a + 4A). \tag{A2}$$

Proceeding as in Refs. [23,32,33], we find a pseudopotential in which the photon number moves,

$$U(N_a) = \frac{2\kappa^2}{3}\left[N_a^4 - AN_a^3 - \frac{3}{2}AN_a^2 + 2AN_a + C \right], \tag{A3}$$

where C is a constant of integration. Treating the total pseudoenergy as a constant of the motion leads to a first-order differential equation for $N_a(z)$,

$$\frac{dN_a}{dz} = \pm \sqrt{2[E - U(N_a)]}$$

$$= \pm \frac{2\kappa}{\sqrt{3}} \sqrt{E_0 - N_a^4 + AN_a^3 + \frac{3}{2}AN_a^2 - 2N_a}, \quad \text{(A4)}$$

where $E_0 = -C + 6E/4\kappa^2$. The formal solution of Eq. (A4) is then written as

$$z = \pm \frac{\sqrt{3}}{2\kappa} \int_A^{N_a} \frac{dN_a}{\sqrt{E_0 - N_a^4 + AN_a^3 + \frac{3}{2}AN_a^2 - 2N_a}}. \quad \text{(A5)}$$

We find that there are three cases where Eq. (A5) has periodic solutions. Defining

$$f(x) = E_0 - x^4 + Ax^3 + \frac{3}{2}Ax^2 - 2x = -\prod_{k=0}^{4}(x - x_k), \quad \text{(A6)}$$

so that we may write

$$z = \pm \int_{x(0)}^{x(z)} \frac{dx}{\sqrt{f(x)}}, \quad \text{(A7)}$$

we examine the roots of the polynomial $f(x) = 0$. (Note that we have changed the variable to x because not all the solutions we find will be possible physical solutions for N_a).

To find which solution is appropriate in this case, we note that the pseudokinetic energy must be equal to zero at $z = 0$, so that $E = U(N_0)$. As we may add any constant value to a potential without changing the dynamics, we choose $C = 0$ in Eq. (A3). For an initial condition of $N_a(0) = 10^6$ (which we used in simulations), we find that $f(x)$ has two real roots, x_1 and x_2, with $x_1 > x_2$, and two complex roots. Writing

$$f(x) = -(x - x_1)(x - x_2)(x^2 - 2\mu x + \nu), \quad \text{(A8)}$$

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

$$x(z) = M_0 + \frac{N_0}{D_0 - \text{cn}(\Omega_0 z + \phi_0, k_0)}, \quad \text{(A9)}$$

where cn signifies the Jacobi cosine amplitude [34]. Defining

$$y_1 = \sqrt{x_1^2 - 2\mu x_1 + \nu} \quad \text{and} \quad y_2 = \sqrt{x_2^2 - 2\mu x_2 + \nu}, \quad \text{(A10)}$$

we have

$$M_0 = \frac{y_1 x_2 - y_2 x_1}{y_1 - y_2},$$

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

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

$$\Omega_0 = \sqrt{y_1 y_2},$$

$$k_0 = \sqrt{\frac{y_1 y_2 - x_1 x_2 + \mu(x_1 + x_2) - \nu}{2y_1 y_2}},$$

$$\phi_0 = \text{cn}^{-1}\left(\frac{D_0(x(0) - M_0) - N_0}{x(0) - M_0}, k_0\right). \quad \text{(A11)}$$

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

$$T_0 = \frac{4}{\Omega_0} \int_0^1 \frac{dt}{\sqrt{(1-t^2)(1 - k_0^2 t^2)}} = \frac{4}{\Omega_0} K(k_0), \quad \text{(A12)}$$

where $K(k_0)$ is the full elliptic integral. This solution has the same formal structure as those for the intensities in traveling-wave second-harmonic generation with an added $\chi^{(3)}$ nonlinearity [33], due to the quartic nature of the pseudopotential, but is different in detail.

2. Spontaneous triplet production

Mathematically, the difference between third-harmonic generation and spontaneous three-photon down-conversion lies in the initial condition. In this case $N_b(0) \neq 0$ and $N_a(0) = 0$. Beginning with Eq. (A1), we again change from operators to real numbers to find an equation for $N_a(z)$,

$$\frac{d^2 N_a}{dz^2} = -\frac{\kappa^3}{3}[8N_a^3 - 18BN_a^2 - (18B - 8)N_a - 12B]. \quad \text{(A13)}$$

We then find that the equivalent pseudopotential may be written as

$$U(N_a) = \frac{2\kappa^2}{3}\left[N_a^4 - 3BN_a^3 - \left(\frac{9}{2}B - 2\right)N_a^2 - 6BN_a\right], \quad \text{(A14)}$$

where we have set the constant of integration equal to zero. In this case we find that

$$\frac{dN_a}{dz} = \pm \frac{2\kappa}{\sqrt{3}} \sqrt{E_0 - N_a^4 + 3BN_a^3 + \left(\frac{9}{2}B - 2\right)N_a^2 + 6BN_a}, \quad \text{(A15)}$$

where $E_0 = 3E/4\kappa^2$. This equation has the formal solution

$$z = \pm \frac{\sqrt{3}}{2\kappa} \int_0^{N_a} \frac{dN_a}{\sqrt{E_0 - N_a^4 + 3BN_a^3 + (\frac{9}{2}B - 2)N_a^2 + 6BN_a}}. \quad \text{(A16)}$$

We may again find a value for E_0 by considering that all the pseudoenergy is potential at $z=0$, giving $E_0=0$. Writing the polynomial under the square root as

$$g(x) = -\prod_{k=1}^{4} (x - x_k), \qquad (A17)$$

we find that for $N_b(0) = 1/3 \times 10^6$, which is the value we used in simulations, the polynomial has two real roots, with $x_2 \leq x \leq x_1$, and two roots that are complex conjugates. The motion of the photon number in this case is also periodic and obeys the general form of Eq. (A9), but with different parameters.

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