Pump depletion in optical parametric amplification

Wanli Xing and T. C. Ralph
Centre for Quantum Computation and Communication Technology, School of Mathematics and Physics, University of Queensland, St Lucia, Queensland 4072, Australia

(Received 3 January 2022; accepted 26 January 2023; published 14 February 2023)

We derive analytic solutions for Heisenberg evolution under the trilinear parametric Hamiltonian which are correct to the second order in the interaction strength but are valid for all pump amplitudes. The solutions allow pump depletion effects to be incorporated in the description of optical parametric amplification in experimentally relevant scenarios and the resulting phenomena to be rigorously described.

DOI: 10.1103/PhysRevA.107.023712

I. INTRODUCTION

Optical parametric amplification [1] is the workhorse of quantum optics, being a source for single photons in the weak amplification regime [2,3] and a source of squeezed states in the strong amplification regime [4,5]. A simple description of this interaction for the nondegenerate (two-mode squeezing) case is given by the unitary [6]

$$U = \exp\{-i\chi(a^\dagger b^\dagger \alpha + ab\alpha^*)\}, \quad (1)$$

where a and b are annihilation operators in the rotating frame describing the squeezed modes, χ is the interaction strength, and α is the amplitude of the coherent pump field. Both χ and α are dimensionless. Applying this unitary to the vacuum produces the well-known two-mode squeezed state

$$|\lambda\rangle_{ab} = U|0\rangle = \sqrt{1-\lambda^2}\sum \lambda^n |n\rangle_a |n\rangle_b, \quad (2)$$

where $\lambda = \tanh \chi\alpha$. Alternatively, the Heisenberg evolution of the annihilation operators is given by

$$a_0 = U^\dagger a U = a \cosh \chi\alpha - ib^\dagger \sinh \chi\alpha,$$
$$b_0 = U^\dagger b U = b \cosh \chi\alpha - ia^\dagger \sinh \chi\alpha. \quad (3)$$

Being quadratic in the operators, the squeezing unitary is Gaussian, i.e., mapping Gaussian states to Gaussian states, and so the first and second moments of the Heisenberg operators and their Hermitian conjugates are sufficient to completely characterize squeezed states [7].

Sophisticated models based on this interaction can be built that successfully describe a large range of devices and protocols in quantum optics [8,9], quantum communication [10], quantum computing [11], and quantum metrology [12,13]. Yet at a fundamental level this interaction is *unphysical* as it is not energy conserving. This is because the pump laser is treated as a reservoir that is unaffected, i.e., undepleted, by the interaction. Under typical experimental conditions this is a good approximation, as the efficiency of the interaction is very low; however, efficiencies are improving all the time and experiments are moving into the regime where depletion effects cannot be neglected [14,15]. Whilst full numerical solutions have been known for many years [16,17] and have been used in theoretical studies [18–20] in conjunction with short time perturbative approaches, these rapidly become intractable when treating realistic systems where the pump power is large. Specifically, numerical simulation stops being practical when $\alpha \gtrsim 30$.

In this work we derive Heisenberg equations of motion which include the lowest-order nontrivial corrections to the standard equations due to pump depletion in a consistent manner that allows for large pump powers. Although nonlinear in the mode operators, our equations are straightforward to work with and allow an exploration of the physics that arises and a description of the most accessible experimental signatures of pump depletion. We note that some work on pump depletion has been carried out with respect to the microwave domain by the superconducting circuit community (e.g., Ref. [21]); however, the physics can be somewhat different there, and here we focus on the optical domain.

II. HEISENBERG EVOLUTION BY THE TRILINEAR HAMILTONIAN

A better approximation for the unitary describing the parametric amplification process is given by

$$U = \exp\{-i\chi(a^\dagger b^\dagger c + abc^\dagger)\}. \quad (4)$$

This unitary is obtained using the rotating wave approximation (RWA) [as in Eq. (1)], which is not strictly justified due to the large detuning of the pump from the nearest excited state. However, at optical frequencies, the only effect of including non-RWA terms is a change to the effective value of the interaction strength χ (as shown explicitly for the related case of the Raman laser [22,23]). Given that χ is introduced as a parameter, the RWA is justified. Notice that the approximation that leads us back from Eq. (4) to the quadratic form in Eq. (1) is the replacement $c \to \langle c\rangle = \alpha$. We want to know the full form of operators a, b, c in the Heisenberg picture, however we no longer obtain the simple closed-form linear equations of Eq. (3). Nevertheless, they can be evaluated to any desired order using the Baker-Campbell-Hausdorff formula [24],

$$a_o = e^G a e^{-G} = a + [G, a] + \tfrac{1}{2!}[G, [G, a]] + \tfrac{1}{3!}[G, [G, [G, a]]] + \cdots \tag{5}$$

The Heisenberg operators get very complicated as we include terms of higher and higher orders. The evolved operators up to order χ^8 are given explicitly in Appendix A. The first few terms of this expansion have been studied since the 1970s [25–27]. However, we find that a brute force expansion in orders of χ is not the most tractable approach in situations of experimental interest. This is because, for typical experimental parameters, not all terms with the same power of χ contribute equally when we calculate expectation values. To see this, we can write the pump operator as $c = \alpha + \delta c$, where the expectation value of c, $\langle c\rangle = \alpha$, is the coherent amplitude of the pump, which we assume to be real here; δc is an operator representing the noise/quantum part of the pump; and $\langle \delta c\rangle = 0$ by definition. If we carry out this expansion, then, for example, the first few terms of a_o become

$$a_o = a - i\chi\alpha b^\dagger - i\chi b^\dagger \delta c + \frac{\chi^2}{2!}(-ab^\dagger b + a\alpha^2 + \alpha a\delta c^\dagger + \alpha a\delta c + a\delta c^\dagger \delta c) + \cdots \tag{6}$$

α can be much larger than 1, whereas χ is a small number much less than 1. We assume $\alpha\chi$ is of order unity. In this case we see that terms like $-\frac{\chi^2}{2!}ab^\dagger b$, which is of the order χ^2, will contribute much less than terms like $\frac{\chi^2}{2!}\alpha^2 a$, which is of the order $(\alpha\chi)^2 \sim 1$. The result is that we need to consider both α and χ when doing the expansion, and the size of a term is determined by the difference in powers between α and χ. In the end we wish to derive consistent Heisenberg operator equations which can be used to evaluate first-, second-, and third-order expectation values that are accurate to the second order in χ and to all orders in $\chi\alpha$.

Hence, we perform the $c = \alpha + \delta c$ expansion and only keep terms of the forms $\alpha^n\chi^n$ and $\alpha^{n-1}\chi^n$, and ignore any other terms (there are also no terms where the power of α is higher than the power of χ). Then the operator for the signal looks like

$$\begin{aligned} a_o &= a\left(1 + \frac{\alpha^2\chi^2}{2!} + \frac{\alpha^4\chi^4}{4!} + \frac{\alpha^6\chi^6}{6!} + \frac{\alpha^8\chi^8}{8!} + \cdots\right) \\ &+ a(\delta c + \delta c^\dagger)\left(\frac{\alpha\chi^2}{2!} + \frac{2\alpha^3\chi^4}{4!} + \frac{3\alpha^5\chi^6}{6!} + \frac{4\alpha^7\chi^8}{8!} + \cdots\right) \\ &- ib^\dagger\left(\alpha\chi + \frac{\alpha^3\chi^3}{3!} + \frac{\alpha^5\chi^5}{5!} + \frac{\alpha^7\chi^7}{7!} + \cdots\right) \\ &- ib^\dagger\delta c\left(\chi + \frac{2\alpha^2\chi^3}{3!} + \frac{3\alpha^4\chi^5}{5!} + \frac{4\alpha^6\chi^7}{7!} + \cdots\right) \\ &- ib^\dagger\delta c^\dagger\left(\frac{\alpha^2\chi^3}{3!} + \frac{2\alpha^4\chi^5}{5!} + \frac{3\alpha^6\chi^7}{7!} + \cdots\right), \end{aligned} \tag{7}$$

where we have collected terms with the same operators, leading to several expansions in $\alpha\chi$. We see a clear pattern from each of the expansions. Assuming the pattern persists (checked to order χ^{15}), we can write the terms in each bracket as infinite sums, which are found to have closed-form expressions. The coefficients of each operator are

$$\begin{aligned} a &: \sum_{n=0}^{\infty}\frac{\alpha^{2n}\chi^{2n}}{(2n)!} = \cosh\alpha\chi, \\ a(\delta c + \delta c^\dagger) &: \sum_{n=1}^{\infty}\frac{\alpha^{2n-1}\chi^{2n}n}{(2n)!} = \frac{\chi}{2}\sinh\alpha\chi, \\ -ib^\dagger &: \sum_{n=0}^{\infty}\frac{\alpha^{2n+1}\chi^{2n+1}}{(2n+1)!} = \sinh\alpha\chi, \\ -b^\dagger\delta c &: \sum_{n=0}^{\infty}\frac{\alpha^{2n}\chi^{2n+1}(n+1)}{(2n+1)!} \\ &= \frac{i\chi}{2}\cosh\alpha\chi + \frac{i}{2\alpha}\sinh\alpha\chi, \\ -b^\dagger\delta c^\dagger &: \sum_{n=1}^{\infty}\frac{\alpha^{2n}\chi^{2n+1}n}{(2n+1)!} = \frac{i\chi}{2}\cosh\alpha\chi - \frac{i}{2\alpha}\sinh\alpha\chi. \end{aligned} \tag{8}$$

These terms give us expressions that are valid up to order χ. Now, we are interested in the second-order moments which will be to order χ^2, hence to be safe we should expand to order χ^2 in a similar way to what we have done when expanding to order χ. This full expansion is presented in Appendix B. However, the vast majority of the second-order terms derived in the Appendix do not contribute to the expectation values at $O(\chi^2)$. We find that for the purpose of calculating expectation values, we may take

$$\begin{aligned} a_o = a&\left[\cosh\chi' + (\delta c + \delta c^\dagger)\frac{\chi}{2}\sinh\chi'\right] \\ &- ib^\dagger\left[\sinh\chi' + \frac{\chi}{2}\cosh\chi'(\delta c + \delta c^\dagger) \right. \\ &\left. + \frac{\chi}{2\chi'}\sinh\chi'(\delta c - \delta c^\dagger)\right] + \chi^2(iAb^\dagger + Ba), \end{aligned} \tag{9}$$

$$\begin{aligned} b_o = b&\left[\cosh\chi' + (\delta c + \delta c^\dagger)\frac{\chi}{2}\sinh\chi'\right] \\ &- ia^\dagger\left[\sinh\chi' + \frac{\chi}{2}\cosh\chi'(\delta c + \delta c^\dagger) \right. \\ &\left. + \frac{\chi}{2\chi'}\sinh\chi'(\delta c - \delta c^\dagger)\right] + \chi^2(Bb + iAa^\dagger), \end{aligned} \tag{10}$$

$$\begin{aligned} c_o = \alpha_o + \delta c_o &= \alpha - \frac{\chi}{2\chi'}\sinh^2\chi' + \chi^3 C \\ &+ \delta c - (a^\dagger a + b^\dagger b)\frac{\chi}{2\chi'}\sinh^2\chi' \\ &- ia^\dagger b^\dagger\frac{\chi}{2}\left(1 - \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right) \\ &- iab\frac{\chi}{2}\left(1 + \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right) + \chi^2 D\delta c^\dagger, \end{aligned} \tag{11}$$

where

$$A = \frac{-5\chi' \cosh\chi' + 2\sinh\chi' - \chi'^2 \sinh\chi' + \sinh 3\chi'}{8\chi'^2},$$

$$B = -\frac{-\cosh\chi' - \chi'^2\cosh\chi' + \cosh 3\chi' - 3\chi'\sinh\chi'}{8\chi'^2},$$

$$C = \frac{-3 - 4\chi'^2 + (2 - 4\chi'^2)\cosh 2\chi' + \cosh 4\chi' - 2\chi'\sinh 2\chi'}{32\chi'^3},$$

$$D = -\frac{1 - \cosh 2\chi' + \chi'\sinh 2\chi'}{4\chi'^2},$$

and $\chi' = \alpha\chi$. The third-order term in c_o can lead to a second-order term in the expectation values due to cross-multiplication with α [see Eq. (12)]. This is the only third-order term in our model that contributes to correlations at second order (or third order). These are "effective operators" in the sense that they give the correct results for all normally ordered second-order moments, i.e., $\langle a_o^\dagger a_o\rangle$, $\langle a_o b_o\rangle$, $\langle \delta c_o^\dagger \delta c_o\rangle$, $\langle \delta c_o \delta c_o\rangle$, and α_o^2, and therefore all variances calculated from these operators are correct. Terms that arise in any calculation that are not normally ordered must first be reordered using the standard Boson commutator relations, e.g., $[a_o, a_o^\dagger] = 1$, before proceeding with the calculation.

Equations (9)–(11) are the main results of this paper. They provide a tractable and physically intuitive way to investigate the lowest-order corrections to the behavior of two-mode squeezing when pump depletion becomes significant.

III. EXPECTATION VALUES

We are now in a position to investigate the physics of the pump-depleted squeezer. Let us first consider the photon number in the pump and the squeezed modes. Assuming the pump is initially in a coherent state and the squeezed modes are initially in vacuum states, the photon number in the pump after the interaction is given by

$$\begin{aligned}\langle c_o^\dagger c_o\rangle &= \alpha_o^2 + \langle \delta c_o^\dagger \delta c_o\rangle = \alpha^2 - \sinh^2\chi' + \left(\frac{\chi}{2\chi'}\right)^2 \sinh^4\chi' \\ &\quad + 2\chi^2\chi' C + \frac{\chi^2}{4}\left(1 - \frac{2}{\chi'}\sinh\chi'\cosh\chi' \right. \\ &\quad \left. + \frac{1}{\chi'^2}\sinh^2\chi'\cosh^2\chi'\right). \end{aligned} \tag{12}$$

As expected, the pump is now depleted by the interaction with $\langle c_o^\dagger c_o\rangle < \alpha^2$. In addition, there is now a coherent contribution to the photon number, α_o^2, and an incoherent contribution, $\langle \delta c_o^\dagger \delta c_o\rangle$. The photon numbers in the squeezed modes are given by

$$\begin{aligned}\langle a_o^\dagger a_o\rangle = \langle b_o^\dagger b_o\rangle &= \sinh^2\chi' - 2\chi^2\sinh\chi' A \\ &\quad + \frac{\chi^2}{4}\left(\cosh^2\chi' - \frac{2}{\chi'}\sinh\chi'\cosh\chi' \right. \\ &\quad \left. + \frac{1}{\chi'^2}\sinh^2\chi'\right). \end{aligned} \tag{13}$$

The photon numbers in the squeezed modes are also lower than predicted by the undepleted pump model. It is straightforward to confirm that energy conservation now holds as

$$\langle c_o^\dagger c_o\rangle + \tfrac{1}{2}(\langle a_o^\dagger a_o\rangle + \langle b_o^\dagger b_o\rangle) = \alpha^2, \tag{14}$$

where we have taken into account that the energy of the squeezed mode photons is half that of the pump photons.

The other nonzero expectation values up to the third order can also be calculated and give

$$\begin{aligned}\langle a_o b_o\rangle &= -\frac{i}{2}\sinh 2\chi' + \frac{i\chi^2}{16\chi'^2}[-4\chi' - 6\chi'\cosh 2\chi' \\ &\quad + (1 - 4\chi'^2)\sinh 2\chi' + 2\sinh 4\chi'], \\ \langle \delta c_o \delta c_o\rangle &= \frac{1}{32}\left[-8\chi^2\left(1 + \frac{1}{\chi'}\sinh 2\chi'\right) \right. \\ &\quad \left. + \frac{\chi^2}{\chi'^2}(8\cosh 2\chi' + \cosh 4\chi' - 9)\right], \\ \langle a_o b_o \delta c_o\rangle &= \frac{i\chi}{2\chi'}\sinh\chi'\cosh^3\chi' - \frac{i\chi}{2}\cosh^2\chi', \\ \langle a_o b_o \delta c_o^\dagger\rangle &= \frac{i\chi}{2\chi'}\sinh^3\chi'\cosh\chi' - \frac{i\chi}{2}\sinh^2\chi'. \end{aligned} \tag{15}$$

From these we can calculate other interesting observables such as the quadrature variances of the output pump beam. The amplitude variance is given by

$$\begin{aligned} V_{xc} &= \langle(\delta c_o + \delta c_o^\dagger)^2\rangle = 2\langle \delta c_o^\dagger \delta c_o\rangle + \langle \delta c_o \delta c_o\rangle + \langle \delta c_o^\dagger \delta c_o^\dagger\rangle + 1 \\ &= 1 - \frac{\chi^2}{\chi'}\sinh 2\chi' + \frac{\chi^2}{8\chi'^2}(-5 + 4\cosh 2\chi' + \cosh 4\chi'), \end{aligned} \tag{16}$$

whilst the phase variance is given by

$$\begin{aligned} V_{pc} &= -\langle(\delta c_o - \delta c_o^\dagger)^2\rangle \\ &= 2\langle \delta c_o^\dagger \delta c_o\rangle - \langle \delta c_o \delta c_o\rangle - \langle \delta c_o^\dagger \delta c_o^\dagger\rangle + 1 \\ &= 1 - \chi^2\left(\frac{\sinh^2\chi'}{\chi'^2} - 1\right). \end{aligned} \tag{17}$$

We notice that the output pump has $V_p < 1 < V_x$, indicating it has become phase squeezed through the interaction. Because $\langle a_o a_o\rangle = \langle b_o b_o\rangle = 0$, the quadrature variances of the signal and idler are isotropic and given by $V_{xj} = V_{pj} = 1 + 2\langle j_o^\dagger j_o\rangle$, where $j = \{a, b\}$. Given the phase convention we have adopted, the correlations between the signal and idler exist between orthogonal quadratures. Hence, the difference (V_{xp}^-) and sum (V_{xp}^+) squeezing between the signal and idler are given by

$$\begin{aligned} V_{xp}^\pm &= \frac{\langle[a_o + a_o^\dagger \pm i(b_o^\dagger - b_o)]^2\rangle}{2} = 2\langle a_o^\dagger a_o\rangle \mp 2i\langle a_o b_o\rangle + 1 \\ &= \frac{1}{16}(\cosh\chi' \mp \sinh\chi') \times \left\{-\frac{3\chi^2}{\chi'^2}\cosh 3\chi' \right. \\ &\quad + \left[\frac{3\chi^2}{\chi'^2} \mp \frac{20\chi^2}{\chi'} + 8(2 + \chi^2)\right]\cosh\chi' \\ &\quad + 2\left[\pm\frac{5\chi^2}{\chi'^2}(1 + \cosh 2\chi') \right. \\ &\quad \left.\left. + \frac{2\chi^2}{\chi'} \mp 4(2 + \chi^2)\right]\sinh\chi'\right\}. \end{aligned} \tag{18}$$

We find $V_{xp}^+ < 1 < V_{xp}^-$, indicating entanglement between the signal and idler beams as expected.

In contrast to the undepleted case, neither the output pump or signal and idler are in minimum uncertainty states. That is, we find $V_{xc}V_{pc} > 1$ and $V_{xp}^+V_{xp}^- > 1$. Given the overall unitarity of the interaction, this indicates that either non-Gaussianity or entanglement, or both, is emerging between the pump and the signal and idler. Indeed, it is both. There is no correlation between the pump or signal or idler in the second-order moments, as would be required if Gaussian entanglement was emerging. Instead, we find a correlation between the pump and signal and idler in the third-order moments, indicating non-Gaussian entanglement. In particular, we can consider the quadrature correlations $V_{abc} = \langle X_{ax}X_{bp}\delta X_{cx}\rangle$ and show

$$\begin{aligned} V_{abc} &= \langle i(a_o + a_o^\dagger)(b_o^\dagger - b_o)(\delta c_o + \delta c_o^\dagger)\rangle \\ &= -2i(\langle a_o b_o \delta c_o\rangle + \langle a_o b_o \delta c_o^\dagger\rangle) \\ &= \frac{\chi}{2}\left(\frac{1}{\chi'}\cosh\chi'\sinh\chi' - 1\right)(\cosh^2\chi' + \sinh^2\chi'). \end{aligned} \tag{19}$$

The fact that this moment is nonzero (whilst all related first-order moments are zero) indicates a non-Gaussian quantum correlation, i.e., entanglement.

IV. STRONG PUMP REGIME

A parameter regime which is expected to be relevant for experimental tests of these effects is the strong pump regime. That is, we take α sufficiently large that $\chi' > 1$, such that the positive power exponentials in our sinh and cosh terms dominate the negative power exponentials, whilst still insisting χ is sufficiently small that our second-order expansion remains valid. We note that although this regime is inaccessible to numerical approaches, it is easily explored with our analytical expressions. By neglecting the negative exponentials in our cosh and sinh terms and keeping only the largest of the positive exponentials, we can significantly simplify our expectation values. The average photon numbers of the pump, signal, and idler become

$$\langle c_o^\dagger c_o\rangle = \alpha^2 - \frac{e^{2\chi'}}{4} + \frac{\chi^2}{16\chi'^2}\ e^{4\chi'} \tag{20}$$

and

$$\langle a_o^\dagger a_o\rangle = \langle b_o^\dagger b_o\rangle = \frac{e^{2\chi'}}{4} - \frac{\chi^2}{16\chi'^2}\ e^{4\chi'}. \tag{21}$$

The pump amplitude quadrature variance becomes

$$V_{xc} = 1 + \frac{\chi^2}{16\chi'^2}\ e^{4\chi'}, \tag{22}$$

whilst the phase quadrature remains at the quantum noise level, $V_{pc} = 1$, given this approximation. The sum squeezing between the signal and idler is given by

$$V_{xp}^+ = e^{-2\chi'} + \frac{\chi^2}{16\chi'^2}\ e^{2\chi'}, \tag{23}$$

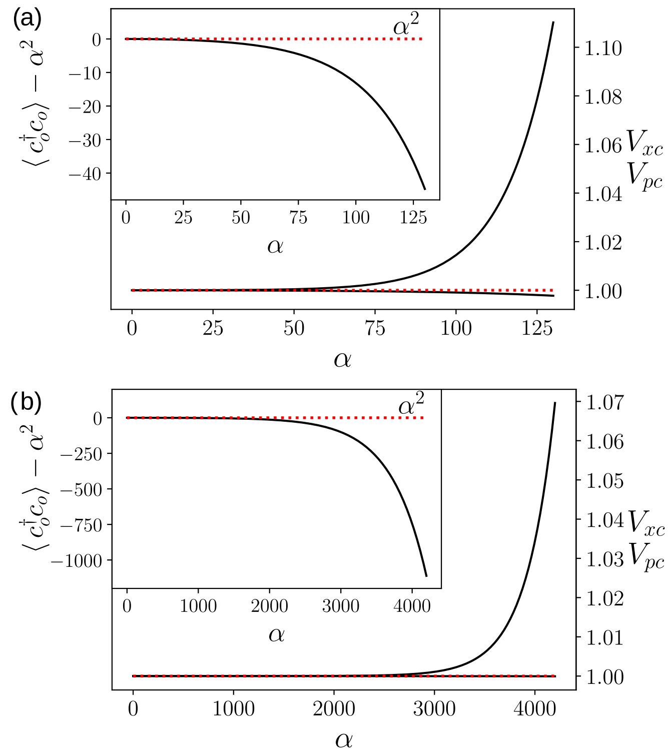

FIG. 1. Amplitude and phase variances of the output pump as a function of input pump amplitude α: (a) the amplitude variance is the upper trace and the phase variance is the lower trace. The red dashed line is the quantum noise limit. Here $\chi = 0.02$. A small amount of squeezing is seen for these parameters, which are plotted using Eqs. (16) and (17). (b) the amplitude variance is shown as the upper trace and the phase variance is the lower trace. Here $\chi = 0.001$. The stronger pump powers in this regime mean that now $V_{pc} = 1$ and V_{xc} is given by Eq. (22). The insets show the depletion of the pump for the same parameters as the main figures.

whilst the difference squeezing is given by

$$V_{xp}^- = e^{2\chi'} - \frac{\chi^2}{4\chi'^2}\ e^{4\chi'}. \tag{24}$$

Notice that this leads to the uncertainty product $V_{xp}^-V_{xp}^+ = 1 + \frac{\chi^2}{16\chi'^2}\ e^{4\chi'}$, indicating the departure from a pure Gaussian entangled state. Note that to be confident that higher-order terms $[> O(\chi^2)]$ can be neglected, we require that the entire correction term remains small, meaning (for fixed χ) χ' cannot be too large in these expressions. In Fig. 1 we show the effect of the interaction on the pump variance for both quadratures for two different values of χ. We also show the corresponding decrease in the pump photon number. In Appendix C we present additional plots highlighting effects on the pump and the signal and idler squeezing.

Perhaps surprisingly, the strongest effect is seen in the third-order correlations. The quadrature correlation between the phase quadrature of the idler and the amplitude quadratures of the signal and pump becomes

$$V_{abc} = \frac{\chi}{16\chi'}\ e^{4\chi'} - \frac{\chi}{4}e^{2\chi'}. \tag{25}$$

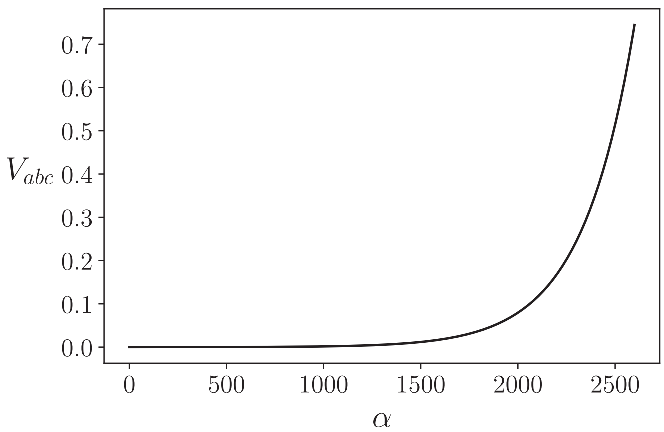

FIG. 2. Third-order correlation V_{abc} [Eq. (19)] as a function of input pump amplitude α. Here $\chi = 0.001$. Significant effects are seen at relatively low pump amplitude.

In the absence of pump depletion this term would be zero. Its nonzero value indicates tripartite entanglement between the pump, signal, and idler, and non-Gaussian statistics. As the lowest-order correction to this moment is linear in χ, it should be the first quantum effect to become observable as we enter the pump depletion regime at high pump powers. In Fig. 2 we illustrate this effect by plotting the third-order correlation [Eq. (19)] against pump amplitude. A significant deviation from the Gaussian case of zero correlation is seen for relatively low pump amplitude. We remind the reader that the equations in this section are only valid when $\chi' > 1$, so it is only in the region where $\alpha > 1000$ that Eq. (25) becomes a good approximation to the plot in Fig. 2.

V. CONCLUSION

We have derived nonlinear Heisenberg equations describing the evolution of quantum fields through the trilinear Hamiltonian, which models parametric amplification with pump depletion. Unlike previous treatments, we perform our perturbative expansion in such a way as to allow the strong pump regime to be explored. We expect our results to be immediately useful in describing and motivating squeezing experiments in the strong pump regime. Being Heisenberg picture equations, they provide good intuition about the physics and can be easily adapted to account for imperfections such as loss and excess noise and/or to model different states of the input fields. We also expect our solutions to stimulate investigations into novel quantum protocols and technologies which may be enabled by the non-Gaussian correlations [28] that emerge as we push further into the depleted pump regime of squeezing.

Note added. Recently, we became aware of a related but distinct approach to pump depletion in single-mode squeezing following a Schrödinger picture approach [29].

ACKNOWLEDGMENT

This research was supported by the Australian Research Council (ARC) under the Centre of Excellence for Quantum Computation and Communication Technology (No. CE170100012).

APPENDIX A: FULL MODE EXPANSIONS TO χ^8

The exact unitary describing the parametric amplification process is given by

$$U = \exp\{-i\chi(a^\dagger b^\dagger c + abc^\dagger)\}.$$

Notice that the approximation that leads us back to the quadratic form in Eq. (1) of the main text is the replacement $c \to \langle c\rangle = \alpha$. We want to know the full forms of operators a, b, c in the Heisenberg picture, which we denote a_o, b_o, c_o. We no longer obtain the simple closed-form linear equations of Eq. (3), nevertheless these Heisenberg operators can be evaluated to any desired order using the Baker-Campbell-Hausdorff formula. For example, the signal mode is

$$a_o = e^G a e^{-G} = a + [G, a] + \frac{1}{2!}[G, [G, a]] + \frac{1}{3!}[G, [G, [G, a]]] + \cdots$$

In this case $G = i\chi(a^\dagger b^\dagger c + abc^\dagger)$. For reference, the Heisenberg operator for the signal mode a_o to order χ^8 is

$$\begin{aligned}
a_o = {} & a - i\chi b^\dagger c + \frac{\chi^2}{2!}(-ab^\dagger b + ac^\dagger c) + \frac{i\chi^3}{3!}(2a^\dagger ab^\dagger c - b^\dagger c^\dagger c^2 + b^\dagger c + b^{\dagger 2}bc - 2a^2bc^\dagger)\\
& + \frac{\chi^4}{4!}(4a^\dagger b^{\dagger 2}c^2 + 4a^\dagger a^2 b^\dagger b - 4a^\dagger a^2 c^\dagger c + ab^\dagger b - 10ab^\dagger bc^\dagger c + ab^{\dagger 2}b^2 - 7ac^\dagger c + ac^{\dagger 2}c^2)\\
& + \frac{i\chi^5}{5!}(28a^\dagger ab^\dagger c^\dagger c^2 - 16a^\dagger ab^\dagger c - 28a^\dagger ab^{\dagger 2}bc + 8a^\dagger a^3 bc^\dagger - 8a^{\dagger 2}a^2 b^\dagger c + 25b^\dagger c^\dagger c^2 - b^\dagger c^{\dagger 2}c^3 - b^\dagger c\\
& + 14b^{\dagger 2}bc^\dagger c^2 - 3b^{\dagger 2}bc - b^{\dagger 3}b^2c + 16a^2b^\dagger b^2c^\dagger + 12a^2bc^\dagger - 16a^2bc^{\dagger 2}c)\\
& + \frac{\chi^6}{6!}(44a^\dagger b^{\dagger 2}c^\dagger c^3 - 72a^\dagger b^{\dagger 2}c^2 - 44a^\dagger b^{\dagger 3}bc^2 - 28a^\dagger a^2 b^\dagger b + 216a^\dagger a^2 b^\dagger bc^\dagger c - 44a^\dagger a^2 b^{\dagger 2}b^2\\
& + 68a^\dagger a^2 c^\dagger c - 44a^\dagger a^2 c^{\dagger 2}c^2 - 72a^{\dagger 2}ab^{\dagger 2}c^2 - 16a^{\dagger 2}a^3 b^\dagger b + 16a^{\dagger 2}a^3 c^\dagger c - ab^\dagger b + 216ab^\dagger bc^\dagger c\\
& - 91ab^\dagger bc^{\dagger 2}c^2 - 3ab^{\dagger 2}b^2 + 91ab^{\dagger 2}b^2c^\dagger c - ab^{\dagger 3}b^3 + 41ac^\dagger c - 85ac^{\dagger 2}c^2 + ac^{\dagger 3}c^3 - 40a^3b^2c^{\dagger 2})
\end{aligned}$$

$$
\begin{aligned}
&+\frac{i\chi^7}{7!}(-1386a^\dagger ab^\dagger c^\dagger c^2+270a^\dagger ab^\dagger c^{\dagger 2}c^3+98a^\dagger ab^\dagger c-1204a^\dagger ab^{\dagger 2}bc^\dagger c^2+598a^\dagger ab^{\dagger 2}bc\\
&+270a^\dagger ab^{\dagger 3}b^2c-416a^\dagger a^3b^\dagger b^2c^\dagger-128a^\dagger a^3bc^\dagger+416a^\dagger a^3bc^{\dagger 2}c+160a^{\dagger 2}b^{\dagger 3}c^3\\
&-496a^{\dagger 2}a^2b^\dagger c^\dagger c^2+144a^{\dagger 2}a^2b^\dagger c+496a^{\dagger 2}a^2b^{\dagger 2}bc-32a^{\dagger 2}a^4bc^\dagger+32a^{\dagger 3}a^3b^\dagger c\\
&-401b^\dagger c^\dagger c^2+264b^\dagger c^{\dagger 2}c^3-b^\dagger c^{\dagger 3}c^4+b^\dagger c-602b^{\dagger 2}bc^\dagger c^2+135b^{\dagger 2}bc^{\dagger 2}c^3+7b^{\dagger 2}bc\\
&-135b^{\dagger 3}b^2c^\dagger c^2+6b^{\dagger 3}b^2c+b^{\dagger 4}b^3c-338a^2b^\dagger b^2c^\dagger+700a^2b^\dagger b^2c^{\dagger 2}c-138a^2b^{\dagger 2}b^3c^\dagger\\
&-70a^2bc^\dagger+910a^2bc^{\dagger 2}c-138a^2bc^{\dagger 3}c^2)\\
&+\frac{\chi^8}{8!}(-4680a^\dagger b^{\dagger 2}c^\dagger c^3+408a^\dagger b^{\dagger 2}c^{\dagger 2}c^4+1104a^\dagger b^{\dagger 2}c^2-2064a^\dagger b^{\dagger 3}bc^\dagger c^3+1752a^\dagger b^{\dagger 3}bc^2\\
&+408a^\dagger b^{\dagger 4}b^2c^2+168a^\dagger a^2b^\dagger b-11\,616a^\dagger a^2b^\dagger bc^\dagger c+7272a^\dagger a^2b^\dagger bc^{\dagger 2}c^2+936a^\dagger a^2b^{\dagger 2}b^2\\
&-7272a^\dagger a^2b^{\dagger 2}b^2c^\dagger c+408a^\dagger a^2b^{\dagger 3}b^3-840a^\dagger a^2c^\dagger c+4536a^\dagger a^2c^{\dagger 2}c^2-408a^\dagger a^2c^{\dagger 3}c^3\\
&+896a^\dagger a^4b^2c^{\dagger 2}-3216a^{\dagger 2}ab^{\dagger 2}c^\dagger c^3+3264a^{\dagger 2}ab^{\dagger 2}c^2+3216a^{\dagger 2}ab^{\dagger 3}bc^2+272a^{\dagger 2}a^3b^\dagger b\\
&-3872a^{\dagger 2}a^3b^\dagger bc^\dagger c+912a^{\dagger 2}a^3b^{\dagger 2}b^2-496a^{\dagger 2}a^3c^\dagger c+912a^{\dagger 2}a^3c^{\dagger 2}c^2+1088a^{\dagger 3}a^2b^{\dagger 2}c^2\\
&+64a^{\dagger 3}a^4b^\dagger b-64a^{\dagger 3}a^4c^\dagger c+ab^\dagger b-3602ab^\dagger bc^\dagger c+10\,410ab^\dagger bc^{\dagger 2}c^2-820ab^\dagger bc^{\dagger 3}c^3\\
&+7ab^{\dagger 2}b^2-4134ab^{\dagger 2}b^2c^\dagger c+3414ab^{\dagger 2}b^2c^{\dagger 2}c^2+6ab^{\dagger 3}b^3-820ab^{\dagger 3}b^3c^\dagger c+ab^{\dagger 4}b^4\\
&-239ac^\dagger c+3607ac^{\dagger 2}c^2-810ac^{\dagger 3}c^3+ac^{\dagger 4}c^4+1392a^3b^\dagger b^3c^{\dagger 2}+1792a^3b^2c^{\dagger 2}-1392a^3b^2c^{\dagger 3}c)+\cdots
\end{aligned}
$$

And the time-evolved operator for the idler mode b_o can be found by swapping a and b in the above formula. The formula for the pump mode is

$$
\begin{aligned}
c_o=\;&c-i\chi ab+\frac{\chi^2}{2}(-a^\dagger ac-b^\dagger bc-c)+\frac{i\chi^3}{3!}(2a^\dagger b^\dagger c^2+a^\dagger a^2b+ab^\dagger b^2+ab-2abc^\dagger c)\\
&+\frac{\chi^4}{4!}(10a^\dagger ab^\dagger bc-4a^\dagger ac^\dagger c^2+3a^\dagger ac+a^{\dagger 2}a^2c-4b^\dagger bc^\dagger c^2+3b^\dagger bc+b^{\dagger 2}b^2c-4c^\dagger c^2-4a^2b^2c^\dagger+c)\\
&+\frac{i\chi^5}{5!}(8a^\dagger b^\dagger c^\dagger c^3-20a^\dagger b^\dagger c^2-16a^\dagger b^{\dagger 2}bc^2-14a^\dagger a^2b^\dagger b^2-3a^\dagger a^2b+28a^\dagger a^2bc^\dagger c-16a^{\dagger 2}ab^\dagger c^2\\
&-a^{\dagger 2}a^3b-3ab^\dagger b^2+28ab^\dagger b^2c^\dagger c-ab^{\dagger 2}b^3-ab+40abc^\dagger c-8abc^{\dagger 2}c^2)\\
&+\frac{\chi^6}{6!}(216a^\dagger ab^\dagger bc^\dagger c^2-148a^\dagger ab^\dagger bc-91a^\dagger ab^{\dagger 2}b^2c+148a^\dagger ac^\dagger c^2-16a^\dagger ac^{\dagger 2}c^3-7a^\dagger ac+44a^\dagger a^3b^2c^\dagger\\
&-40a^{\dagger 2}b^{\dagger 2}c^3-91a^{\dagger 2}a^2b^\dagger bc+44a^{\dagger 2}a^2c^\dagger c^2-6a^{\dagger 2}a^2c-a^{\dagger 3}a^3c+148b^\dagger bc^\dagger c^2-16b^\dagger bc^{\dagger 2}c^3-7b^\dagger bc\\
&+44b^{\dagger 2}b^2c^\dagger c^2-6b^{\dagger 2}b^2c-b^{\dagger 3}b^3c+60c^\dagger c^2-16c^{\dagger 2}c^3+44a^2b^\dagger b^3c^\dagger+60a^2b^2c^\dagger-72a^2b^2c^{\dagger 2}c-c)\\
&+\frac{i\chi^7}{7!}(-704a^\dagger b^\dagger c^\dagger c^3+32a^\dagger b^\dagger c^{\dagger 2}c^4+222a^\dagger b^\dagger c^2-416a^\dagger b^{\dagger 2}bc^\dagger c^3+490a^\dagger b^{\dagger 2}bc^2+138a^\dagger b^{\dagger 3}b^2c^2\\
&+208a^\dagger a^2b^\dagger b^2-1204a^\dagger a^2b^\dagger b^2c^\dagger c+135a^\dagger a^2b^{\dagger 2}b^3+7a^\dagger a^2b-1022a^\dagger a^2bc^\dagger c+496a^\dagger a^2bc^{\dagger 2}c^2\\
&-416a^{\dagger 2}ab^\dagger c^\dagger c^3+490a^{\dagger 2}ab^\dagger c^2+700a^{\dagger 2}ab^{\dagger 2}bc^2+135a^{\dagger 2}a^3b^\dagger b^2+6a^{\dagger 2}a^3b-270a^{\dagger 2}a^3bc^\dagger c\\
&+138a^{\dagger 3}a^2b^\dagger c^2+a^{\dagger 3}a^4b+7ab^\dagger b^2-1022ab^\dagger b^2c^\dagger c+496ab^\dagger b^2c^{\dagger 2}c^2+6ab^{\dagger 2}b^3-270ab^{\dagger 2}b^3c^\dagger c\\
&+ab^{\dagger 3}b^4+ab-522abc^\dagger c+848abc^{\dagger 2}c^2-32abc^{\dagger 3}c^3+160a^3b^3c^{\dagger 2})\\
&+\frac{\chi^8}{8!}(-17\,472a^\dagger ab^\dagger bc^\dagger c^2+3872a^\dagger ab^\dagger bc^{\dagger 2}c^3+1826a^\dagger ab^\dagger bc-7272a^\dagger ab^{\dagger 2}b^2c^\dagger c^2+3246a^\dagger ab^{\dagger 2}b^2c\\
&+820a^\dagger ab^{\dagger 3}b^3c-3768a^\dagger ac^\dagger c^2+3376a^\dagger ac^{\dagger 2}c^3-64a^\dagger ac^{\dagger 3}c^4+15a^\dagger ac-2064a^\dagger a^3b^\dagger b^3c^\dagger\\
&-1512a^\dagger a^3b^2c^\dagger+3216a^\dagger a^3b^2c^{\dagger 2}c-896a^{\dagger 2}b^{\dagger 2}c^\dagger c^4+2384a^{\dagger 2}b^{\dagger 2}c^3+1392a^{\dagger 2}b^{\dagger 3}bc^3\\
&-7272a^{\dagger 2}a^2b^\dagger bc^\dagger c^2+3246a^{\dagger 2}a^2b^\dagger bc+3414a^{\dagger 2}a^2b^{\dagger 2}b^2c-2736a^{\dagger 2}a^2c^\dagger c^2+912a^{\dagger 2}a^2c^{\dagger 2}c^3\\
&+25a^{\dagger 2}a^2c-408a^{\dagger 2}a^4b^2c^\dagger+1392a^{\dagger 3}ab^{\dagger 2}c^3+820a^{\dagger 3}a^3b^\dagger bc-408a^{\dagger 3}a^3c^\dagger c^2+10a^{\dagger 3}a^3c\\
&+a^{\dagger 4}a^4c-3768b^\dagger bc^\dagger c^2+3376b^\dagger bc^{\dagger 2}c^3-64b^\dagger bc^{\dagger 3}c^4+15b^\dagger bc-2736b^{\dagger 2}b^2c^\dagger c^2+912b^{\dagger 2}b^2c^{\dagger 2}c^3
\end{aligned}
$$

$$+ 25b^{\dagger 2}b^2c - 408b^{\dagger 3}b^3c^\dagger c^2 + 10b^{\dagger 3}b^3c + b^{\dagger 4}b^4c - 744c^\dagger c^2 + 1552c^{\dagger 2}c^3 - 64c^{\dagger 3}c^4 - 1512a^2b^\dagger b^3c^\dagger$$
$$+ 3216a^2b^\dagger b^3c^{\dagger 2}c - 408a^2b^{\dagger 2}b^4c^\dagger - 744a^2b^2c^\dagger + 6384a^2b^2c^{\dagger 2}c - 1088a^2b^2c^{\dagger 3}c^2 + c) + \cdots$$

APPENDIX B: FULL OUTPUT OPERATORS TO $O(\chi^2)$

We perform a $c = \alpha + \delta c$ expansion, and retain only the terms of the form $\alpha^n\chi^n$ (assumed to be of order 1) and $\alpha^{n-1}\chi^n$ ([of $O(\chi)$]. For example, with the pump mode c_o, only a small number of terms could potentially contribute at $O(\chi)$:

$$c_o = c - i\chi ab + \frac{\chi^2}{2}(-c - a^\dagger ac - b^\dagger bc) + \frac{i\chi^3}{3!}(2a^\dagger b^\dagger c^2 - 2abc^\dagger c) + \frac{\chi^4}{4!}(-4c^\dagger c^2 - 4a^\dagger ac^\dagger c^2 - 4b^\dagger bc^\dagger c^2)$$
$$+ \frac{i\chi^5}{5!}(8a^\dagger b^\dagger c^\dagger c^3 - 8abc^{\dagger 2}c^2) + \frac{\chi^6}{6!}(-16c^{\dagger 2}c^3 - 16a^\dagger ac^{\dagger 2}c^3 - 16b^\dagger bc^{\dagger 2}c^3)$$
$$+ \frac{i\chi^7}{7!}(32a^\dagger b^\dagger c^{\dagger 2}c^4 - 32abc^{\dagger 3}c^3) + \frac{\chi^8}{8!}(-64c^{\dagger 3}c^4 - 64a^\dagger ac^{\dagger 3}c^4 - 64b^\dagger bc^{\dagger 3}c^4) + \cdots$$

Now substitute $c = \alpha + \delta c$ (and therefore $c^\dagger = \alpha + \delta c^\dagger$), keeping only $O(\chi)$ terms, we have

$$c_o = \alpha + \delta c - i\chi ab + \frac{\chi^2}{2!}(-\alpha - \alpha a^\dagger a - \alpha b^\dagger b) + \frac{i\chi^3}{3!}(2\alpha^2 a^\dagger b^\dagger - 2\alpha^2 ab) + \frac{\chi^4}{4!}(-4\alpha^3 - 4\alpha^3 a^\dagger a - 4\alpha^3 b^\dagger b)$$
$$+ \frac{i\chi^5}{5!}(8\alpha^4 a^\dagger b^\dagger - 8\alpha^4 ab) + \frac{\chi^6}{6!}(-16\alpha^5 - 16\alpha^5 a^\dagger a - 16\alpha^5 b^\dagger b)$$
$$+ \frac{i\chi^7}{7!}(32\alpha^6 a^\dagger b^\dagger - 32\alpha^6 ab) + \frac{\chi^8}{8!}(-64\alpha^7 - 64\alpha^7 a^\dagger a - 64\alpha^7 b^\dagger b) + \cdots$$

Group the terms according to the operators they are multiplied to:

$$\Rightarrow c_o = \alpha - \left(\frac{\alpha\chi^2}{2!} + \frac{4\alpha^3\chi^4}{4!} + \frac{16\alpha^5\chi^6}{6!} + \frac{64\alpha^7\chi^8}{8!} + \cdots\right) + \delta c - (a^\dagger a + b^\dagger b)\left(\frac{\alpha\chi^2}{2!} + \frac{4\alpha^3\chi^4}{4!} + \frac{16\alpha^5\chi^6}{6!} + \frac{64\alpha^7\chi^8}{8!} + \cdots\right)$$
$$+ ia^\dagger b^\dagger\left(\frac{2\alpha^2\chi^3}{3!} + \frac{8\alpha^4\chi^5}{5!} + \frac{32\alpha^6\chi^7}{7!} + \cdots\right) - iab\left(\chi + \frac{2\alpha^2\chi^3}{3!} + \frac{8\alpha^4\chi^5}{5!} + \frac{32\alpha^6\chi^7}{7!} + \cdots\right).$$

Note that just as we decomposed $c = \alpha + \delta c$, we can decompose the Heisenberg operator into an amplitude part and a noise part, $c_o = \alpha_o + \delta c_o$, where both parts are time dependent. We see from above that the first line of c_o consists only of pure numbers and no operators, and is therefore the amplitude α_o; anything on the second line and below are the noise part δc_o.

There are clear patterns to the first few terms of each of the infinite series above. Assuming the patterns persist indefinitely (checked to order $\alpha^{14}\chi^{15}$), we may express each infinite series as a sum, and use MATHEMATICA to find the closed forms of these series:

$$\frac{\alpha\chi^2}{2!} + \frac{4\alpha^3\chi^4}{4!} + \frac{16\alpha^5\chi^6}{6!} + \cdots = \sum_{n=1}^{\infty}\frac{\alpha^{2n-1}\chi^{2n} \times 4^{n-1}}{(2n)!} = \frac{\chi}{2\chi'}\sinh^2\chi',$$
$$\frac{2\alpha^2\chi^3}{3!} + \frac{8\alpha^4\chi^5}{5!} + \frac{32\alpha^6\chi^7}{7!} + \cdots = \sum_{n=1}^{\infty}\frac{\alpha^{2n}\chi^{2n+1} \times 2 \times 4^{n-1}}{(2n+1)!} = -\frac{\chi}{2} + \frac{\chi}{2\chi'}\sinh\chi'\cosh\chi',$$
$$\chi + \frac{2\alpha^2\chi^3}{3!} + \frac{8\alpha^4\chi^5}{5!} + \frac{32\alpha^6\chi^7}{7!} + \cdots = \frac{\chi}{2} + \frac{\chi}{2\chi'}\sinh\chi'\cosh\chi',$$

where we again defined $\chi' \equiv \alpha\chi$. So c_o is

$$c_o = \alpha_o + \delta c_o = \alpha - \frac{\chi}{2\chi'}\sinh^2\chi' + \delta c - (a^\dagger a + b^\dagger b)\frac{\chi}{2\chi'}\sinh^2\chi'$$
$$- ia^\dagger b^\dagger\frac{\chi}{2}\left(1 - \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right) - iab\frac{\chi}{2}\left(1 + \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right),$$

where we see that

$$\alpha_o = \alpha - \frac{\chi}{2\chi'}\sinh^2\chi',$$

$$\text{and}\quad \delta c_o = \delta c - (a^\dagger a + b^\dagger b)\frac{\chi}{2\chi'}\sinh^2\chi' - ia^\dagger b^\dagger\frac{\chi}{2}\left(1 - \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right) - iab\frac{\chi}{2}\left(1 + \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right).$$

For a fully self-consistent model capable of calculating nontrivial expectation values, we need to include the $O(\chi^2)$ terms in the output modes a_o, b_o, c_o as well. That is, after the $c = \alpha + \delta c$ expansion, on top of the $\alpha^n\chi^n$ and $\alpha^{n-1}\chi^n$ terms, we now also retain terms of the form $\alpha^{n-2}\chi^n$. This introduces many additional infinite series. For illustrative purposes, let us focus on a couple of them. In c_o, consider terms proportional to $a^\dagger a c^{\dagger n} c^m$, the terms that could contribute are

$$c_o = \cdots - \frac{\chi^2}{2}a^\dagger ac - \frac{\chi^4}{4!}4a^\dagger ac^\dagger c^2 - \frac{\chi^6}{6!}16a^\dagger ac^{\dagger 2}c^3 - \frac{\chi^8}{8!}64a^\dagger ac^{\dagger 3}c^4 + \cdots,$$

now performing the $c = \alpha + \delta c$ expansion, we have

$$\begin{aligned} c_o &= \cdots - a^\dagger a\left[\frac{\chi^2}{2}(\alpha+\delta c) + \frac{\chi^4}{4!}4(\alpha+\delta c^\dagger)(\alpha+\delta c)^2 + \frac{\chi^6}{6!}16(\alpha+\delta c^\dagger)^2(\alpha+\delta c)^3 + \frac{\chi^8}{8!}64(\alpha+\delta c^\dagger)^3(\alpha+\delta c)^4 + \cdots\right] \\ &= \cdots - a^\dagger a\left(\frac{\chi^2}{2}\alpha + \frac{4\chi^4}{4!}\alpha^3 + \frac{16\chi^6}{6!}\alpha^5 + \frac{64\chi^8}{8!}\alpha^7 + \cdots\right) - a^\dagger a\left(\frac{\chi^2}{2}\delta c + \frac{8\chi^4}{4!}\alpha^2\delta c + \frac{48\chi^6}{6!}\alpha^4\delta c + \frac{256\chi^8}{8!}\alpha^6\delta c + \cdots\right) \\ &\quad - a^\dagger a\left(\frac{4\chi^4}{4!}\alpha^2\delta c^\dagger + \frac{32\chi^6}{6!}\alpha^4\delta c^\dagger + \frac{192\chi^8}{8!}\alpha^6\delta c^\dagger + \cdots\right), \end{aligned}$$

giving three infinite series. The first one we've already seen; it is of $O(\chi)$ and equals to $-a^\dagger a\frac{\chi}{2\chi'}\sinh^2\chi'$. The last two series are new, and can be put into closed-form expressions as

$$\begin{aligned} c_o &= \cdots - a^\dagger a\delta c\,\chi^2\sum_{n=0}^{\infty}2^{2n-1}(2n+2)\times\frac{\chi'^{2n}}{(2n+2)!} - a^\dagger a\delta c^\dagger\,\chi^2\sum_{n=1}^{\infty}2^{2n}n\frac{\chi'^{2n}}{(2n+2)!} \\ &= \cdots - a^\dagger a\delta c\,\chi^2\frac{1-\cosh 2\chi' + 2\sinh^2\chi' + \chi'\sinh 2\chi'}{4\chi'^2} - a^\dagger a\delta c^\dagger\,\chi^2\frac{1-\cosh 2\chi' + \chi'\sinh 2\chi'}{4\chi'^2}. \end{aligned}$$

With some work, all second-order terms can be grouped into series which can then be expressed as closed-form expressions like the ones above.

We now simply list the final results. We find that the signal mode a_o to order χ^2 is

$$\begin{aligned} a_o &= a\left[\cosh\chi' + (\delta c+\delta c^\dagger)\frac{\chi}{2}\sinh\chi'\right] - ib^\dagger\left[\sinh\chi' + \frac{\chi}{2}\cosh\chi'(\delta c+\delta c^\dagger) + \frac{\chi}{2\chi'}\sinh\chi'(\delta c-\delta c^\dagger)\right] \\ &\quad + \chi^2(A_a a + A_{b^\dagger}b^\dagger + A_{a^2b}a^2b + A_{ab^\dagger b}ab^\dagger b + A_{a^\dagger ab^\dagger}a^\dagger ab^\dagger + A_{a^\dagger b^{\dagger 2}}a^\dagger b^{\dagger 2} + A_{a^\dagger a^2}a^\dagger a^2 + A_{a\delta c^2}a\delta c^2 \\ &\quad + A_{a\delta c^{\dagger 2}}a\delta c^{\dagger 2} + A_{a\delta c^\dagger\delta c}a\delta c^\dagger\delta c + A_{b^{\dagger 2}b}b^{\dagger 2}b + A_{b^\dagger\delta c^2}b^\dagger\delta c^2 + A_{b^\dagger\delta c^\dagger\delta c}b^\dagger\delta c^\dagger\delta c + A_{b^\dagger\delta c^{\dagger 2}}b^\dagger\delta c^{\dagger 2}), \end{aligned} \tag{B1}$$

where

$$\begin{aligned} A_a &= -\sum_{n=2}^{\infty}\left(\frac{9^n-1}{8} - \frac{n(n+1)}{2}\right)\times\frac{\chi'^{2n-2}}{(2n)!} = -\frac{-\cosh\chi' - \chi'^2\cosh\chi' + \cosh 3\chi' - 3\chi'\sinh\chi'}{8\chi'^2}, \\ A_{b^\dagger} &= i\sum_{n=1}^{\infty}Y(n)\frac{\chi'^{2n-1}}{(2n+1)!} = i\times\frac{-5\chi'\cosh\chi' + 2\sinh\chi' - \chi'^2\sinh\chi' + \sinh 3\chi'}{8\chi'^2}, \\ A_{a^2b} &= -i\sum_{n=1}^{\infty}[a(n)+n]\times\frac{\chi'^{2n-1}}{(2n+1)!} = -i\times\frac{4\chi'\cosh\chi' - 7\sinh\chi' + \sinh 3\chi'}{16\chi'^2}, \\ A_{ab^\dagger b} &= -\sum_{n=1}^{\infty}\frac{9^n-1}{8}\times\frac{\chi'^{2n-2}}{(2n)!} = \frac{\cosh\chi' - \cosh 3\chi'}{8\chi'^2}, \\ A_{a^\dagger ab^\dagger} &= i\sum_{n=1}^{\infty}\left[Y(n) + \frac{n(n+1)}{2}\right]\times\frac{\chi'^{2n-1}}{(2n+1)!} = i\times\frac{-4\chi'\cosh\chi' + \sinh\chi' + \sinh 3\chi'}{8\chi'^2}, \\ A_{a^\dagger b^{\dagger 2}} &= -A_{a^\dagger a^2} = \sum_{n=2}^{\infty}\left(\frac{9^n-1}{16} - \frac{n}{2}\right)\times\frac{\chi'^{2n-2}}{(2n)!} = \frac{-\cosh\chi' + \cosh 3\chi' - 4\chi'\sinh\chi'}{16\chi'^2}, \\ A_{a\delta c^2} &= A_{a\delta c^{\dagger 2}} = \sum_{n=1}^{\infty}\frac{n(n+1)}{2}\times\frac{\chi'^{2n}}{(2n+2)!} = \frac{\chi'\cosh\chi' - \sinh\chi'}{8\chi'}, \end{aligned}$$

$$A_{a\delta c^\dagger\delta c} = \sum_{n=0}^{\infty}(n+1)^2\frac{\chi'^{2n}}{(2n+2)!} = \frac{\chi'\cosh\chi' + \sinh\chi'}{4\chi'},$$

$$A_{b^{\dagger 2}b} = i\sum_{n=1}^{\infty}a(n)\times\frac{\chi'^{2n-1}}{(2n+1)!} = i\times\frac{-4\chi'\cosh\chi' + \sinh\chi' + \sinh 3\chi'}{16\chi'^2},$$

$$A_{b^\dagger\delta c^2} = \frac{1}{2}A_{b^\dagger\delta c^\dagger\delta c} = -i\sum_{n=1}^{\infty}\frac{n(n+1)}{2}\times\frac{\chi'^{2n-1}}{(2n+1)!} = -i\times\frac{\chi'\cosh\chi' - \sinh\chi' + \chi'^2\sinh\chi'}{8\chi'2},$$

$$A_{b^\dagger\delta c^{\dagger 2}} = -i\sum_{n=1}^{\infty}\left(\frac{n(n+1)}{2}\right)\times\frac{\chi'^{2n+1}}{(2n+3)!} = -i\times\frac{-3\chi'\cosh\chi' + 3\sinh\chi' + \chi'^2\sinh\chi'}{8\chi'^2}.$$

And for the pump mode,

$$\begin{aligned} c_o = {}& \alpha - \frac{\chi}{2\chi'}\sinh^2\chi' + \chi^3 C_\alpha + \delta c - (a^\dagger a + b^\dagger b)\frac{\chi}{2\chi'}\sinh^2\chi' - ia^\dagger b^\dagger\frac{\chi}{2}\left(1 - \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right) \\ & - iab\frac{\chi}{2}\left(1 + \frac{1}{\chi'}\sinh\chi'\cosh\chi'\right) + \chi^2(C_{\delta c}\delta c + C_{a^\dagger a\delta c}a^\dagger a\delta c + C_{b^\dagger b\delta c}b^\dagger b\delta c + C_{ab\delta c}ab\delta c + C_{ab\delta c^\dagger}ab\delta c^\dagger \\ & + C_{a^\dagger b^\dagger\delta c}a^\dagger b^\dagger\delta c + C_{\delta c^\dagger}\delta c^\dagger + C_{a^\dagger a\delta c^\dagger}a^\dagger a\delta c^\dagger + C_{b^\dagger b\delta c^\dagger}b^\dagger b\delta c^\dagger + C_{a^\dagger b^\dagger\delta c^\dagger}a^\dagger b^\dagger\delta c^\dagger), \end{aligned} \tag{B2}$$

where

$$C_\alpha = \sum_{n=1}^{\infty}Z(n)\times\frac{\chi'^{2n-1}}{(2n+2)!} = \frac{-3 - 4\chi'^2 + (2 - 4\chi'^2)\cosh 2\chi' + \cosh 4\chi' - 2\chi'\sinh 2\chi'}{32\chi'^3}$$

$$C_{\delta c} = C_{a^\dagger a\delta c} = C_{b^\dagger b\delta c} = -\sum_{n=0}^{\infty}2^{2n-1}(2n+2)\times\frac{\chi'^{2n}}{(2n+2)!} = -\frac{1 - \cosh 2\chi' + 2\sinh^2\chi' + \chi'\sinh 2\chi'}{4\chi'^2},$$

$$C_{ab\delta c} = C_{ab\delta c^\dagger} = -i\sum_{n=1}^{\infty}2^{2n-2}(2n)\times\frac{\chi'^{2n-1}}{(2n+1)!} = -i\frac{2\chi'\cosh 2\chi' - \sinh 2\chi'}{8\chi'^2},$$

$$C_{a^\dagger b^\dagger\delta c} = i\sum_{n=2}^{\infty}\frac{2^{2n-2}n}{2}\times\frac{\chi'^{2n-3}}{(2n-1)!} = i\frac{-4\chi' + 2\chi'\cosh 2\chi' + \sinh 2\chi'}{8\chi'^2},$$

$$C_{\delta c^\dagger} = C_{a^\dagger a\delta c^\dagger} = C_{b^\dagger b\delta c^\dagger} = -\sum_{n=1}^{\infty}2^{2n}n\frac{\chi'^{2n}}{(2n+2)!} = -\frac{1 - \cosh 2\chi' + \chi'\sinh 2\chi'}{4\chi'^2},$$

$$C_{a^\dagger b^\dagger\delta c^\dagger} = i\sum_{n=1}^{\infty}2^{2n}(2n)\times\frac{\chi'^{2n+1}}{(2n+3)!} = i\frac{4\chi' + 2\chi'\cosh 2\chi' - 3\sinh 2\chi'}{8\chi'^2}.$$

In the above, the expressions $a(n)$, $X(n)$, $Y(n)$, and $Z(n)$ are given by

$$a(n) = \frac{3^{2n+1} - 8n - 3}{16},$$

$$X(n) = 54a(n-1) + 25n - 18 - \frac{n(n-1)}{2},$$

$$Y(n) = 18a(n-1) + 7n - 6 - \frac{n(n-1)}{2},$$

$$\text{and}\quad Z(n) = (2n+1)!\sum_{k=0}^{n}\frac{1}{(2k)!}\frac{1}{[2(n-k)+1]!}[X(k-1) + Y(n-k) - k(n-k)].$$

All n and k are integers. The first few numbers in each sequence are listed in Table I.

One can check that the operators are physical in the sense that the commutation relations are satisfied to $O(\chi^2)$, namely,

$$[a_o, a_o^\dagger] = [b_o, b_o^\dagger] = [c_o, c_o^\dagger] = 1 + O(\chi^3),$$

$$\text{all other commutation relations} = 0 + O(\chi^3).$$

The vast majority of the terms in the output modes do not contribute to the expectation values at $O(\chi^2)$. Specifically, it turns out the only second-order term in a_o that contributes to $\langle a_o^\dagger a_o\rangle$ is $\chi^2 A_{b^\dagger}b^\dagger$; the only second-order terms that contribute to $\langle a_o b_o\rangle$ are $\chi^2(A_{b^\dagger}b^\dagger + A_a a)$. Similarly, the only second-order term in c_o that contributes to $\langle\delta c_o\delta c_o\rangle$ is $\chi^2 C_{\delta c^\dagger}\delta c^\dagger$, and the only second-order or above term that contributes to $\langle c_o^\dagger c_o\rangle$

TABLE I. Table showing the first few numbers in the series $a(n)$, $X(n)$, $Y(n)$, $Z(n)$.

n	$a(n)$	$X(n)$	$Y(n)$	$Z(n)$
−1	1/3	0	2/3	—
0	0	0	0	0
1	1	7	1	1
2	14	85	25	60
3	135	810	264	1552
4	1228	7366	2446	29 632
5	11 069	66 409	22 123	506 112
6	99 642	597 843	199 263	8 289 280

is $\chi^3 C_\alpha$. All other terms either annihilate $\langle 0|$ or $|0\rangle$ to give 0 contributions, or they only contribute to $O(\chi^3)$ terms. So for the purpose of calculating expectation values, we may simply take

$$\begin{aligned} a_o =& a\bigg[\cosh\chi' + (\delta c+\delta c^\dagger)\frac{\chi}{2}\sinh\chi'\bigg] \\ &- ib^\dagger\bigg[\sinh\chi' + \frac{\chi}{2}\cosh\chi'(\delta c+\delta c^\dagger) \\ &+ \frac{\chi}{2\chi'}\sinh\chi'(\delta c-\delta c^\dagger)\bigg] + \chi^2(A_a a + A_{b^\dagger} b^\dagger), \end{aligned} \tag{B3}$$

$$\begin{aligned} b_o =& b\bigg[\cosh\chi' + (\delta c+\delta c^\dagger)\frac{\chi}{2}\sinh\chi'\bigg] \\ &- ia^\dagger\bigg[\sinh\chi' + \frac{\chi}{2}\cosh\chi'(\delta c+\delta c^\dagger) \\ &+ \frac{\chi}{2\chi'}\sinh\chi'(\delta c-\delta c^\dagger)\bigg] \\ &+ \chi^2(A_a b + A_{b^\dagger} a^\dagger), \end{aligned} \tag{B4}$$

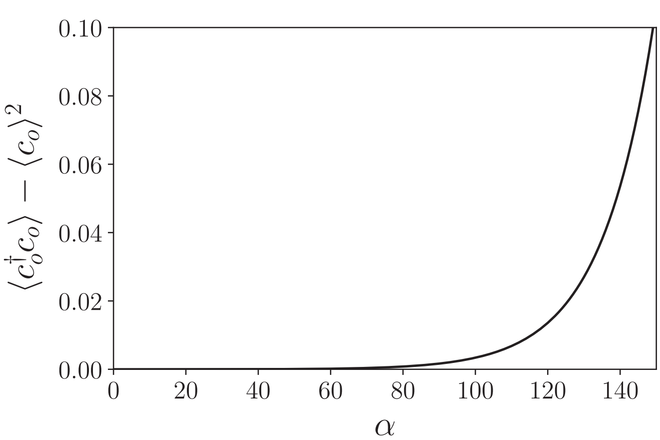

FIG. 3. The incoherent component of the pump photon number when $\chi = 0.02$.

$$\begin{aligned} c_o =& \alpha_o + \delta c_o \\ =& \alpha - \frac{\chi}{2\chi'}\sinh^2\chi' + \chi^3 C_\alpha \\ &+ \delta c - (a^\dagger a + b^\dagger b)\frac{\chi}{2\chi'}\sinh^2\chi' \\ &- ia^\dagger b^\dagger\frac{\chi}{2}\Big(1 - \frac{1}{\chi'}\sinh\chi'\cosh\chi'\Big) \\ &- iab\frac{\chi}{2}\Big(1 + \frac{1}{\chi'}\sinh\chi'\cosh\chi'\Big) \\ &+ \chi^2 C_{\delta c^\dagger}\delta c^\dagger. \end{aligned} \tag{B5}$$

These are effective operators in the sense that they give the correct results for $\langle a_o^\dagger a_o\rangle$, $\langle a_o b_o\rangle$, $\langle \delta c_o^\dagger \delta c_o\rangle$, $\langle \delta c_o \delta c_o\rangle$, and α_o^2, and therefore all variances calculated from these operators are correct. As such, one can also check that energy is conserved,

$$\alpha^2 = \langle c_o^\dagger c_o\rangle + \langle a_o^\dagger a_o\rangle = \alpha_o^2 + \langle \delta c_o^\dagger \delta c_o\rangle + \langle a_o^\dagger a_o\rangle.$$

Although not obvious, it turns out the three effective operators above also give the correct formulae for $\langle ab\delta c\rangle$ and $\langle ab\delta c^\dagger\rangle$ up to $O(\chi^2)$.

The only drawback of using the effective operators is that the commutation relations given by these operator are only correct to $O(\chi)$, not the desired $O(\chi^2)$. Therefore, we should not use them to calculate nonnormally ordered operator products, for example, $\langle a_o a_o^\dagger\rangle$ or $\langle \delta c_o \delta c_o^\dagger\rangle$. Instead, we should first normal order them such that, for example, $\langle a_o a_o^\dagger\rangle \to \langle 1 + a_o^\dagger a_o\rangle$ and $\langle \delta c_o \delta c_o^\dagger\rangle \to \langle 1 + \delta c_o^\dagger \delta c_o\rangle$, before evaluating them.

The bottom line is, we may use Eqs. (B3)–(B5) to calculate any second- or third-order correlations to $O(\chi^2)$, provided the correlations are normal ordered. If we want our theory to be fully self-consistent without reordering and capable of predicting any correlations to $O(\chi^2)$, we should use the full Eqs. (B1) and (B2).

APPENDIX C: ADDITIONAL FIGURES

In Figs. 3 and 4 we plot the difference between the coherent amplitude squared of the pump and its actual photon number for two different values of χ. This highlights the incoherent contribution to the pump photon number, $\langle \delta c_o^\dagger \delta c_o\rangle$, after the interaction. In Figs. 5 and 6 we plot the sum squeezing be-

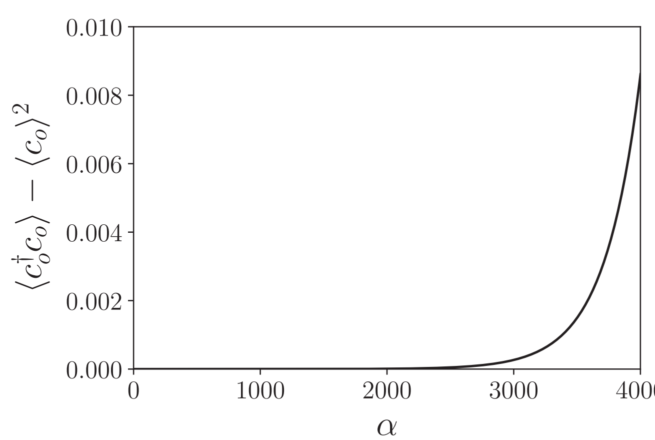

FIG. 4. The incoherent component of the pump photon number when $\chi = 0.001$.

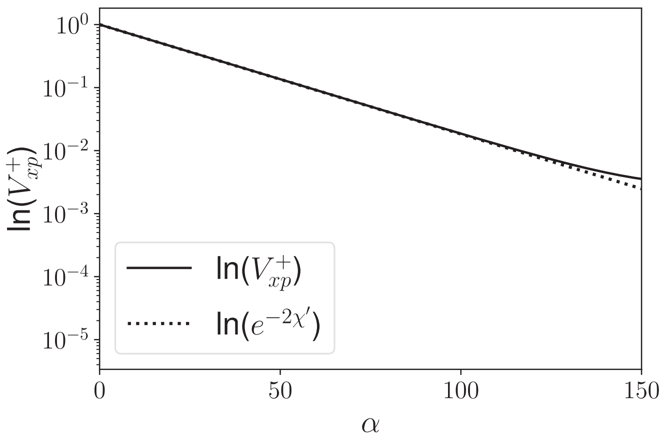

FIG. 5. The sum squeezing between the signal and idler when $\chi = 0.02$, plotted on a log scale. The squeezing in the undepleted case ($e^{-2\chi'}$) is also shown. A saturation of the squeezing is visible for high pump rates, which would limit the achievable squeezing.

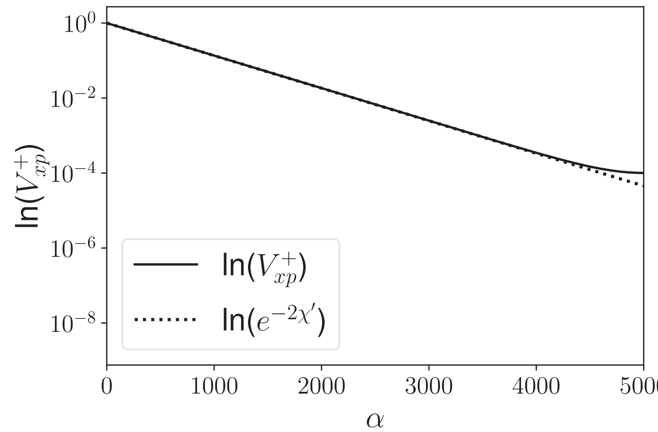

FIG. 6. The sum squeezing between the signal and idler when $\chi = 0.001$. In this high pump regime the saturation effect is weaker, only appearing at much higher squeezing levels compared to Fig. 5.

tween the signal and idler for two different values of χ. For the higher value of χ in Fig. 5 a saturation effect on the sum squeezing is evident for squeezing variances lower than about 0.01 . That is, the variance (on a log scale) stops decreasing linearly with pump amplitude at higher α. This effect only occurs for much stronger squeezing for the lower value of χ in Fig. 6.

[1] D. F. Walls and G. J. Milburn, *Quantum Optics*, 2nd ed. (Springer-Verlag, New York, 2008).

[2] C. K. Hong and L. Mandel, Experimental Realization of a Localized One-Photon State, Phys. Rev. Lett. **56**, 58 (1986).

[3] R. B. Jin, R. Shimizu, K. Wakui, Mikio Fujiwara, T. Yamashita, S. Miki, H. Terai, Z. Wang, and M. Sasaki, Pulsed Sagnac polarization-entangled photon source with a PPKTP crystal at telecom wavelength, Opt. Express **22**, 11498 (2014)

[4] L.-A. Wu, M. Xiao, and H. J. Kimble, Squeezed states of light from an optical parametric oscillator, J. Opt. Soc. Am. B **4**, 1465 (1987).

[5] T. Eberle, V. Händchen, and R. Schnabel, Stable control of 10 dB two-mode squeezed vacuum states of light, Opt. Express **21**, 11546 (2013).

[6] H.-A. Bachor and T. C. Ralph, *A Guide to Experiments in Quantum Optics*, 3rd ed. (Wiley, New York, 2019).

[7] C. Weedbrook, S. Pirandola, R. García-Patrón, N. J. Cerf, T. C. Ralph, J. H. Shapiro, and S. Lloyd, Gaussian quantum information, Rev. Mod. Phys. **84**, 621 (2012).

[8] R. Baumgartner and R. Byer, Optical parametric amplification, IEEE J. Quantum Electron. **15**, 432 (1979).

[9] V. V. Dodonov, "Nonclassical" states in quantum optics: A "squeezed" review of the first 75 years, J. Opt. B: Quantum Semiclass. Opt. **4**, R1 (2002).

[10] Z. Y. Ou, S. F. Pereira, H. J. Kimble, and K. C. Peng, Realization of the Einstein-Podolsky-Rosen Paradox for Continuous Variables, Phys. Rev. Lett. **68**, 3663 (1992).

[11] N. C. Menicucci, P. van Loock, M. Gu, C. Weedbrook, T. C. Ralph, and M. A. Nielsen, Universal Quantum Computation with Continuous-Variable Cluster States, Phys. Rev. Lett. **97**, 110501 (2006).

[12] A. A. Clerk, M. H. Devoret, S. M. Girvin, F. Marquardt, and R. J. Schoelkopf, Introduction to quantum noise, measurement, and amplification, Rev. Mod. Phys. **82**, 1155 (2010).

[13] F. Hudelist, J. Kong, C. Liu, J. Jing, Z. Ou, and W. Zhang, Quantum metrology with parametric amplifier-based photon correlation interferometers, Nat. Commun. **5**, 3049 (2014).

[14] A. Allevi, O. Jedrkiewicz, E. Brambilla, A. Gatti, J. Peřina, Jr., O. Haderka, and M. Bondani, Coherence properties of high-gain twin beams, Phys. Rev. A **90**, 063812 (2014).

[15] J. Flórez, J. S. Lundeen, and M. V. Chekhova, Pump depletion in parametric down-conversion with low pump energies, Opt. Lett. **45**, 4264 (2020).

[16] D. F. Walls and R. Barakat, Quantum-mechanical amplification and frequency conversion with a trilinear Hamiltonian, Phys. Rev. A **1**, 446 (1970).

[17] G. Drobný and I. Jex, Quantum properties of field modes in trilinear optical processes, Phys. Rev. A **46**, 499 (1992).

[18] P. D. Nation and M. P. Blencowe, The trilinear Hamiltonian: A zero-dimensional model of Hawking radiation from a quantized source, New J. Phys. **12**, 095013 (2010).

[19] S. Ding, G. Maslennikov, R. Hablützel, and D. Matsukevich, Quantum Simulation with a Trilinear Hamiltonian, Phys. Rev. Lett. **121**, 130502 (2018).

[20] R. J. Birrittella, P. M. Alsing, and C. C. Gerry, Phase effects in coherently-stimulated down-conversion with a quantized pump field, Phys. Rev. A **101**, 013813 (2020).

[21] A. Roy and M. Devoret, Quantum-limited parametric amplification with Josephson circuits in the regime of pump depletion, Phys. Rev. B **98**, 045405 (2018).

[22] P. A. Roos, S. K. Murphy, L. S. Meng, J. L. Carlsten, T. C. Ralph, A. G. White, and J. K. Brasseur, Quantum theory of the far-off-resonance continuous-wave Raman laser: Heisenberg/Langevin approach, Phys. Rev. A **68**, 013802 (2003).

[23] P. A. Roos, The diode-pumped continuous-wave Raman laser: Classical, quantum and thermo-optic fundamentals, Ph.D. thesis, Montana State University, 2002.

[24] J. J. Sakurai and J. Napolitano, *Modern Quantum Mechanics*, 2nd ed. (Cambridge University Press, Cambridge, 2017).

[25] G. P. Agrawal and C. L. Mehta, Dynamics of parametric processes with a trilinear Hamiltonian, J. Phys. A: Math. Nucl. Gen. **7**, 607 (1974).

[26] J. Peřina, Dynamics of non-linear optical processes, Czech. J. Phys. B **26**, 140 (1976).

[27] A. Bandilla, G. Drobny, and I. Jex, Nondegenerate parametric interactions and nonclassical effects, Phys. Rev. A **53**, 507 (1996).

[28] I. Straka, L. Lachman, J. Hloušek, M. Miková, M. Mičuda, M. Ježek, and R. Filip, Quantum non-Gaussian multiphoton light, npj Quantum Inf. **4**, 4 (2018).

[29] R. Yanagimoto, E. Ng, A. Yamamura, T. Onodera, L. G. Wright, M. Jankowski, M. M. Fejer, P. L. McMahon, and H. Mabuchi, Onset of non-Gaussian quantum physics in pulsed squeezing with mesoscopic fields, Optica **9**, 379 (2022).