

Effects of time ordering in quantum nonlinear optics

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We study time-ordering corrections to the description of spontaneous parametric down-conversion (SPDC), four-wave mixing (SFWM), and frequency conversion using the Magnus expansion. Analytic approximations to the evolution operator that are unitary are obtained. They are Gaussian preserving, and allow us to understand order-by-order the effects of time ordering. We show that the corrections due to time ordering vanish exactly if the phase-matching function is sufficiently broad. The calculation of the effects of time ordering on the joint spectral amplitude of the photons generated in SPDC and SFWM are reduced to quadrature.

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I. INTRODUCTION

Spontaneous parametric down-conversion (SPDC), spontaneous four-wave mixing (SFWM), and frequency conversion (FC) are three of the most common nonlinear processes used in quantum optics. In the first two a nonlinear medium is used to convert photons from a pump laser into pairs of quantum-correlated photons [1]. In the third, the frequency of a photon is increased or decreased after it interacts with a strong pump field inside a nonlinear medium. Pair generation is often modeled in a straightforward manner using the first order of a Dyson series, or the first term in a Taylor-like series that results if consequences of time-ordering are ignored [2]. But when the nonlinear interaction becomes sufficiently strong a more involved theoretical treatment is required. In particular, the Taylor-like series is suspect beyond a few orders because the interaction Hamiltonian does not commute with itself at different times.

In previous work [3,4] the Dyson series has been used to investigate parametric down-conversion, and it was shown that when the initial state of the down-converted modes is vacuum (SPDC), the Taylor-like and Dyson series give identical results up to second order. In a subsequent study, Christ *et al.* [5] used the Heisenberg picture to investigate time-ordering issues in both SPDC and FC. They noticed that when the pump is approximated as undepleted and treated classically, the Hamiltonian is quadratic in the bosonic operators of the down-converted fields, and thus the equations of motion of the operators are necessarily *linear*. This implies that the outgoing fields are related to the incoming fields by a linear Bogoliubov transformation [6]. They used this observation to build an ansatz solution, and develop a numerical algorithm that permits the calculation of the linear transformation relating output to input. Another consequence of the linear Bogoliubov relation between output and input is that the state of the down-converted photons in SPDC cannot be more complicated than a two-mode squeezed vacuum. This is at odds with a result of Branczyk *et al.* [4], who found that the third-order correction to the SPDC state contains a six photon energy correlated state. As we shall show, this discrepancy is related to the fact that the Dyson series does not retain the Gaussian-preserving nature of the

quadratic Hamiltonian, or equivalently the linear Bogoliubov nature of the transformation relating input and output fields.

In this paper we employ the Magnus expansion (ME), which is able to deal properly with time ordering at each level, and thus is unitary to all orders. Yet, unlike the Dyson series, it respects the Gaussian-preserving nature of the process for the class of Hamiltonians that are quadratic in bosonic operators, giving rise to a linear Bogoliubov transformation to whatever order it is taken. Thus we feel it should be the preferred strategy for the description of photon generation and conversion in nonlinear quantum optics.

In Sec. II we introduce the ME and mention some of its important properties, deriving a useful simplification of its terms when dealing with a broad class of Hamiltonians used in quantum optics; we also show why the ME automatically respects the Gaussian nature of usual nonlinear quantum optical processes. In Sec. III we apply the Magnus series generated by the ME to SPDC and SFWM Hamiltonians. While the ME has been used in the past in quantum nonlinear optics (see, e.g., Yang *et al.* [7] and references cited therein), usually only the first term in the expansion has been kept. We calculate the second and third order terms, and also introduce diagrams akin to Feynman diagrams that allow for a simple tabulation of the higher-order processes in the ME. Using these diagrams we easily demonstrate why it is necessary to go to third order in the expansion to see a correction to the SPDC and SFWM states. In Sec. IV we extend these ideas to FC processes. We show that the expansion formulas derived for FC are completely analogous to those derived for SPDC and SFWM. In Sec. V we show how the ME can be taken to a form more amenable to direct calculation and develop a perturbation series for the joint spectral amplitude (JSA) of the down-converted photons in SPDC and SFWM. In Sec. VI we show that for perfect phase matching the second and third order Magnus terms vanish. Finally, conclusions and remarks are presented in Sec. VII.

II. PROPERTIES OF THE MAGNUS EXPANSION

In quantum mechanics it is often necessary to solve the time-dependent Schrödinger equation for the time evolution operator $\mathcal{U}(t, t_0)$,

$$i\hbar \frac{d}{dt} \mathcal{U}(t, t_0) = H_I(t) \mathcal{U}(t, t_0), \quad (1)$$

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where $H_I(t)$ is the interaction picture Hamiltonian. One common way to approximate the solution of Eq. (1) is to use the Dyson series in which the evolution operator is expanded as a power series:

$$\begin{aligned} \mathcal{U}(t_1, t_0) &= \mathbb{I} + T_1 + T_2 + \dots \\ &= \mathbb{I} + (-i) \int_{t_0}^{t_1} dt' \frac{H_I(t')}{\hbar} \\ &\quad + (-i)^2 \int_{t_0}^{t_1} dt' \int_{t_0}^{t'} dt'' \frac{H_I(t')H_I(t'')}{\hbar^2} + \dots \end{aligned} \quad (2)$$

Often after time ordering, some of the terms appearing, depicted as Feynman diagrams, can be grouped into classes and summed. Here instead we use the Magnus expansion [8,9] in which a series expansion is developed as the argument of an exponential,

$$\mathcal{U}(t_1, t_0) = \exp[\Omega_1(t_1, t_0) + \Omega_2(t_1, t_0) + \Omega_3(t_1, t_0) + \dots], \quad (3)$$

where unless otherwise indicated we take $t_0 \rightarrow -\infty$ and $t_1 \rightarrow \infty$. In the following we calculate the terms in such a series for a rather broad class of Hamiltonians relevant to nonlinear quantum optics. We write the time dependence of the interaction Hamiltonian as

$$H_I(t) = \hbar \int d\omega [h(\omega)e^{i\Delta t} + h^\dagger(\omega)e^{-i\Delta t}], \quad (4)$$

where we use the shorthand notation

$$\begin{aligned} h(\omega) &= h(\omega_1, \omega_2, \dots, \omega_n), \\ d\omega &= d\omega_1 d\omega_2 \dots d\omega_n, \\ \Delta &= \Delta(\omega_1, \omega_2, \dots, \omega_n). \end{aligned} \quad (5)$$

For the moment we leave the integration limits of the frequencies ω unspecified; in the next sections we will specify them for particular cases.

For the first-order Magnus term one simply finds

$$\Omega_1 = -\frac{i}{\hbar} \int dt H_I(t) \quad (6)$$

$$= -2\pi i \int d\omega [h(\omega) + \text{H.c.}] \delta(\Delta). \quad (7)$$

In the last equation we have adopted the convention that whenever the lower (or upper) limit of an integral over time is not specified it is meant to be understood as $-\infty$ (or $+\infty$). The term (6), exponentiated, corresponds to the sum of the Taylor-like series for \mathcal{U} that could be constructed by time-ordering the series (2) and ignoring the fact that the Hamiltonians $H_I(t)$ at different times t need not commute. In fact, if $[H_I(t), H_I(t')] = 0$ for all t and t' , then $\exp \Omega_1$ constitutes the *exact* solution of the problem, since all the higher-order Magnus terms vanish. Hence we refer to terms in the Magnus expansion beyond Ω_1 as “time-ordering corrections.”

In general no simplifications can be made to the higher-order Magnus terms if no further assumptions are made about $H_I(t)$. We make the following:

$$[h(\omega), h(\omega')] = [h^\dagger(\omega), h^\dagger(\omega')] = 0, \quad (8)$$

which is indeed valid for the Hamiltonians usually employed to describe both type I and II parametric down-conversion, four-wave mixing, and frequency conversion, either if the pump

is treated classically or if it is treated quantum mechanically. Using property (8) and the fact that

$$\begin{aligned} \int dt \int dt' e^{i\Delta t - i\Delta' t'} \\ = 2\pi^2 \delta(\Delta) \delta(\Delta') + 2\pi i \delta(\Delta - \Delta') \frac{\mathcal{P}}{\Delta}, \end{aligned} \quad (9)$$

with \mathcal{P} indicating a principal value, the second-order Magnus term, generally given by

$$\Omega_2 = \frac{(-i)^2}{2\hbar^2} \int dt \int dt' [H_I(t), H_I(t')], \quad (10)$$

can be simplified to

$$\Omega_2 = -2\pi i \int d\omega d\omega' [h(\omega), h^\dagger(\omega')] \frac{\delta(\Delta - \Delta')}{\Delta}. \quad (11)$$

In the last equation we have used the principal value integral symbol \int . This is necessary because the result (9) is only valid under a principal value integration sign. Only the second term of (9) contributes to the second-order term. This is to be expected since the first term is just

$$2\pi^2 \delta(\Delta) \delta(\Delta') = \frac{1}{2} \int dt \int dt' e^{i\Delta t - i\Delta' t'}, \quad (12)$$

which is the expression that would be obtained from (9) ignoring the time ordering; note the difference in limits between Eq. (12) and Eq. (9).

Now let us look at the third-order Magnus term, which in general

$$\begin{aligned} \Omega_3 &= \frac{(-i)^3}{6\hbar^3} \int dt \int dt' \int dt'' [H_I(t), [H_I(t'), H_I(t'')]] \\ &\quad + [[H_I(t), H_I(t')], H_I(t'')]. \end{aligned} \quad (13)$$

Here the relevant time integrals take the form

$$\begin{aligned} \int dt \int dt' \int dt'' e^{i\mu t + i\nu t' + i\xi t''} \\ = 2\pi^3 \delta(\mu) \delta(\nu) \delta(\xi) \\ - 2\pi^2 i \delta(\nu + \xi) \delta(\mu + \nu + \xi) \frac{\mathcal{P}}{\xi} \\ + 2\pi^2 i \delta(\mu + \nu) \delta(\mu + \nu + \xi) \frac{\mathcal{P}}{\mu} \\ - 2\pi \delta(\mu + \nu + \xi) \frac{\mathcal{P}}{\mu + \nu} \frac{\mathcal{P}}{\mu}. \end{aligned} \quad (14)$$

Because of the condition (8) the only term that contributes to Ω_3 in (14) is the last one, and then upon relabeling one obtains

$$\begin{aligned} \Omega_3 &= -2\pi i \int d\omega d\omega' d\omega'' ([h(\omega), [h(\omega'), h^\dagger(\omega'')]] + \text{H.c.}) \\ &\quad \times \frac{\delta(\Delta + \Delta' - \Delta'')}{3} \left(\frac{1}{\Delta \Delta'} + \frac{1}{\Delta \Delta''} \right). \end{aligned} \quad (15)$$

We emphasize that the simple form obtained for Ω_3 critically depends on assumption (8). If this did not hold, then the second and third terms in (14) would also contribute, leading to a much more complicated expression. However, even if (8) were not to hold the first term of (14) would never contribute to Ω_3 ,

which is related to the fact that it is proportional to terms that would arise if time order were ignored, and their effects have all been included in Ω_1 .

Finally, let us note that the Magnus expansion is more robust in preserving qualitative features of the exact solution for \mathcal{U} than the Dyson expansion, particularly for problems in nonlinear quantum optics. Generally, of course, the Magnus expansion truncated at any level will always give rise to a unitary evolution operator if the Hamiltonian is Hermitian, as is assumed when writing (4), while a truncated Dyson expansion will not. More specifically, the Magnus expansion truncated at any level will always give rise, in the Heisenberg picture, to a linear Bogoliubov transformation of the bosonic operators if the Hamiltonian is quadratic in them. Since such quadratic bosonic Hamiltonians (QBH) often appear in nonlinear quantum optics, and since for such a Hamiltonian the exact solution for the Heisenberg picture evolution of the bosonic operators indeed takes the form of a linear Bogoliubov transformation [6], this is a very desirable feature of the Magnus approximation. Here is the proof that any Magnus approximation displays this feature: From the general commutator identity

$$\begin{aligned} [AB, CD] &= [A, C]BD + A[B, C]D \\ &\quad + C[A, D]B + CA[B, D], \end{aligned} \quad (16)$$

we see that if A, B, C, D are all bosonic operators then the commutators on the right-hand side are all c numbers, and in fact if they do not vanish they are delta functions, either Kronecker or Dirac. Since all the terms Ω_1, Ω_2 , etc., in the Magnus series involve commutators, it is clear that for a QBH all the terms Ω_i will be quadratic functions of the bosonic operators, and the M th-order approximation to the Magnus expansion

$$\begin{aligned} \mathcal{U}_M(t_f, t_0) &= \exp[\Omega_1(t_f, t_0) + \Omega_2(t_f, t_0) + \dots + \Omega_M(t_f, t_0)] \\ &= \exp[\mathcal{O}_M(t_f, t_0)], \end{aligned} \quad (17)$$

where we reinstate general initial and final times, t_0 and t_f respectively, is characterized by an operator $\mathcal{O}(t_f, t_0)$ that is anti-Hermitian and a quadratic function of the boson operators. Now in the Heisenberg picture and initial operator $A(t_0)$ evolves to a later operator $A(t_f)$ according to

$$A(t_f) = \mathcal{U}^\dagger(t_f, t_0)A(t_0)\mathcal{U}(t_f, t_0). \quad (18)$$

Using the approximation given by (17) and the Baker-Campbell-Hausdorff (BCH) formula

$$e^X Y e^{-X} = Y + [X, Y] + \frac{1}{2!}[X, [X, Y]] + \dots, \quad (19)$$

we find

$$\begin{aligned} A(t_f) &= A(t_0) + [A(t_0), \mathcal{O}_M(t_f, t_0)] \\ &\quad + \frac{1}{2!}[[A(t_0), \mathcal{O}_M(t_f, t_0)], \mathcal{O}_M(t_f, t_0)] + \dots \end{aligned} \quad (20)$$

Using the general commutator identity

$$[A, BC] = [A, B]C + B[A, C], \quad (21)$$

we see that if $A(t_0)$ is a bosonic operator the commutators on the right-hand side of (20) will all be bosonic operators, and thus the Heisenberg bosonic operators at t_f are indeed linear functions of the Heisenberg bosonic operators at t_0 . This completes the proof. In the Schrödinger picture, the result implies that any Magnus approximation for evolution under a QBH will share the property of the exact solution that the general Gaussian nature of an initial state will be preserved; that is, the exact evolution constitutes a unitary Gaussian channel, and any Magnus approximation for the evolution constitutes a unitary Gaussian channel as well [10].

III. TIME ORDERING IN SPONTANEOUS PARAMETRIC DOWN-CONVERSION AND FOUR-WAVE MIXING

In type II SPDC an incoming photon at frequency ω_p is transformed into two photons of different polarizations at frequencies ω_a and ω_b which are roughly half the frequency of the incoming photon. Within the usual rotating wave and undepleted pump approximations, the interaction Hamiltonian governing this process in an effectively one-dimensional structure with all relevant modes propagating in the same direction, such as a channel or ridge waveguide, is (see Appendix B for a derivation)

$$\begin{aligned} H_I(t) &= -\hbar\varepsilon \int d\omega_a d\omega_b d\omega_p e^{i\Delta t} \Phi(\omega_a, \omega_b, \omega_p) \\ &\quad \times \alpha(\omega_p) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}, \end{aligned} \quad (22)$$

$$\Delta = \omega_a + \omega_b - \omega_p, \quad (23)$$

where $a(\omega_a)$ and $b(\omega_b)$ are photon destruction operators for modes associated with the wave vectors $k_a(\omega_a)$ and $k_b(\omega_b)$, constructed to satisfy the commutation relations

$$[a(\omega), a^\dagger(\omega')] = [b(\omega), b^\dagger(\omega')] = \delta(\omega - \omega'), \quad (24)$$

and all other commutators between destruction and creation operators vanishing; $\alpha(\omega_p)$ is a classical function proportional to the electric field amplitude of the undepleted pump mode with wave vector $k(\omega_p)$. The assumption (24) is well justified because for type II SPDC the down-converted photons appear in orthogonal polarizations, and their frequency support is far away from the frequency support of the pump mode characterized by $\alpha(\omega_p)$. Because of this the limits of integration for the frequencies of the modes will range from 0 to ∞ . Nevertheless, since the bandwidth of the photons is much smaller than their central frequencies these integrals can be safely extended in their lower limit to $-\infty$.

The phase-matching function Φ incorporates the spatial dependence of the modes and the nonlinearity profile. It takes the form

$$\Phi(\omega_a, \omega_b, \omega_p) = \text{sinc}\left(\frac{\Delta k L}{2}\right), \quad (25)$$

$$\Delta k = k_a(\omega_a) + k_b(\omega_b) - k_p(\omega_p), \quad (26)$$

when the nonlinearity is assumed to be uniform in a region of space from $-L/2$ to $L/2$, and in Eq. (22) ε is a dimensionless constant incorporating parameters such as the strength of the

nonlinearity profile and the effective cross sectional area of the crystal. There will be corrections to the integrand of (22) depending in a benign way on the frequencies [2], and for simplicity we neglect them here.

In SFWM a pair of photons from a pump mode at frequency ω_p are converted into a pair of photons of different frequencies $\omega_p \pm \Delta\omega_p$. The description of this process is inherently more complicated than that of SPDC, but we show in Appendix C that if the pump field is treated classically and undepleted, then the Hamiltonian governing the process is formally identical to (22) if the effects of group velocity dispersion on the propagation of the pump pulse can be neglected. In that limit we find

$$H_I(t) = -\hbar\varepsilon \int d\omega_a d\omega_b d\omega_+ e^{i\tilde{\Delta}t} \tilde{\Phi}(\omega_a, \omega_b, \omega_+) \times \tilde{\alpha}(\omega_+) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}, \quad (27)$$

where here $\tilde{\Delta} = \omega_a + \omega_b - \omega_+$, and

$$\tilde{\Phi}(\omega_a, \omega_b, \omega_+) = \text{sinc}\left(\frac{\Delta k L}{2}\right), \quad (28)$$

$$\Delta k = k_a(\omega_a) + k_b(\omega_b) - 2k_p\left(\frac{\omega_+}{2}\right), \quad (29)$$

and now $\tilde{\alpha}(\omega_+)$ is proportional to the square of the amplitude of the electric field of the pump. Again we have assumed that the generated photons at ω_a and ω_b are characterized by distinct polarizations and/or frequency ranges—the latter, for example, might arise due to phase-matching constraints [11]—and thus the only nonvanishing commutation relations are given by (24).

While the approach introduced in this paper can be applied to more complicated scenarios for SFWM, we restrict ourselves here to situations where (27) is applicable; then the analysis of SPDC and SFWM is essentially the same. The interaction Hamiltonian $H_I(t)$ takes the form (4) with

$$h(\omega_a, \omega_b, \omega_p) = F(\omega_a, \omega_b, \omega_p) a^\dagger(\omega_a) b^\dagger(\omega_b), \quad (30)$$

$$F(\omega_a, \omega_b, \omega_p) = -\varepsilon\alpha(\omega_p)\Phi(\omega_a, \omega_b, \omega_p), \quad (31)$$

for SPDC; for SFWM we simply replace ω_p by ω_+ , $\alpha(\omega_p)$ by $\tilde{\alpha}(\omega_+)$, and $\Phi(\omega_a, \omega_b, \omega_c)$ by $\tilde{\Phi}(\omega_a, \omega_b, \omega_+)$. So although in the calculations below we explicitly use the notation for SPDC, the generalization to SFWM is trivial.

With the notation introduced in the paragraphs above, the first-order Magnus term is

$$\Omega_1 = -2\pi i \int d\omega_a d\omega_b [J_1(\omega_a, \omega_b) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}], \quad (32)$$

with

$$J_1(\omega_a, \omega_b) = F(\omega_a, \omega_b, \omega_a + \omega_b). \quad (33)$$

This process can be represented by panel (a) in Fig. 1 in which a photon of frequency of ω_p (or two photons of frequency ω_p for SFWM) is converted to two photons of frequency ω_a and ω_b ; we associate it with the term $J_1(\omega_a, \omega_b) a^\dagger(\omega_a) b^\dagger(\omega_b)$. To have a Hermitian Hamiltonian the reverse process needs to

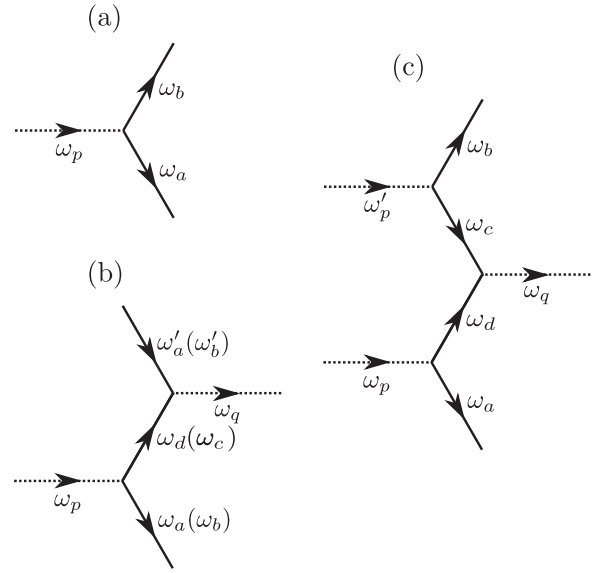


FIG. 1. Diagrams representing the first, second, and third order Magnus terms for SPDC. Dashed lines are used to represent pump photons; full lines are used to represent lower-energy down-converted ones. Panel (a) depicts a photon of frequency ω_p being converted to two photons of frequencies ω_a and ω_b . Panel (b) depicts the second-order Magnus term in which one of the photons from a down-conversion event is, with the help of a low-energy photon previously present, up-converted to a pump photon. Finally, panel (c) depicts the third-order Magnus term in which a pair of photons from two previous down-conversion events is converted to a pump photon.

be possible and this is associated with $\bar{J}_1(\omega_a, \omega_b) a(\omega_a) b(\omega_b)$ where \bar{x} is the complex conjugate of x .

We note that with the approximation of keeping only the first term in the Magnus expansion, as well as the assumption that its effect is small, we have

$$|\psi(\infty)\rangle = \mathcal{U} |\psi(-\infty)\rangle \approx e^{\Omega_1} |\psi(-\infty)\rangle \approx (\mathbb{I} + \Omega_1) |\psi(-\infty)\rangle = (\mathbb{I} + T_1) |\psi(-\infty)\rangle, \quad (34)$$

where T_1 is the first term of the Dyson series (2), this is, as shown in the last equation, identical to the first-order Magnus term Ω_1 . For the second-order Magnus term we need the following:

$$[h(\omega), h^\dagger(\omega')] = -\delta(\omega'_b - \omega_b) F(\omega_a, \omega_b, \omega_p) \times \bar{F}(\omega'_a, \omega'_b, \omega'_p) a^\dagger(\omega_a) a(\omega'_a) - \delta(\omega'_a - \omega_a) F(\omega_a, \omega_b, \omega_p) \times \bar{F}(\omega'_a, \omega'_b, \omega'_p) b^\dagger(\omega_b) b(\omega'_b). \quad (35)$$

After some relabeling the second-order Magnus term is

$$\Omega_2 = -2\pi i \left[\int d\omega_a d\omega'_a G_2^a(\omega_a, \omega'_a) a^\dagger(\omega_a) a(\omega'_a) + \int d\omega_b d\omega'_b G_2^b(\omega_b, \omega'_b) b^\dagger(\omega_b) b(\omega'_b) \right], \quad (36)$$

with

$$G_2^a(\omega_a, \omega'_a) = \int d\omega_d \int \frac{d\omega_p}{\omega_p} F(\omega_a, \omega_d, \omega_a + \omega_p + \omega_d) \times \bar{F}(\omega'_a, \omega_d, \omega'_a + \omega_p + \omega_d), \quad (37)$$

$$G_2^b(\omega_b, \omega'_b) = \int d\omega_c \int \frac{d\omega_p}{\omega_p} F(\omega_c, \omega_b, \omega_b + \omega_c + \omega_p) \times \bar{F}(\omega_c, \omega'_b, \omega'_b + \omega_p + \omega_c). \quad (38)$$

The last expression shows that the second-order term corresponds to a frequency conversion process in which a photon

of frequency ω'_a (ω'_b) is converted to one of frequency ω_a (ω_b) with probability amplitude G_2^a (G_2^b). If one recognizes that F is associated with down-conversion and \bar{F} with up-conversion, then Eq. (37) [Eq. (38)] can be read as follows: first a photon at the pump energy (or two photons for the case of SFWM) is down-converted to two photons of energies ω_a (ω_b) and ω_d (ω_c). Later the photon of energy ω_d (ω_c) interacts with a photon of energy ω'_a (ω'_b) to be up-converted to another photon of energy close to the pump energy (or a pair of photons for SFWM), which effectively gives a frequency conversion process from ω'_a (ω'_b) to ω_a (ω_b). This is sketched in panel (b) in Fig. 1.

For the third-order Magnus term we need to calculate the following:

$$[h(\omega), [h(\omega'), h^\dagger(\omega'')]] = F(\omega_a, \omega_b, \omega_p) F(\omega'_a, \omega'_b, \omega'_p) \bar{F}(\omega''_a, \omega''_b, \omega''_p) [\delta(\omega''_a - \omega'_a) \delta(\omega''_b - \omega_b) b^\dagger(\omega'_b) a^\dagger(\omega_a) + \delta(\omega''_a - \omega_a) \delta(\omega''_b - \omega'_b) b^\dagger(\omega_b) a^\dagger(\omega'_a)]. \quad (39)$$

Using (15), after some rearranging of the dummy integration variables we obtain

$$\Omega_3 = -2\pi i \int d\omega_a d\omega_b a^\dagger(\omega_a) b^\dagger(\omega_b) \int d\omega_c d\omega_d d\omega_p d\omega_q \frac{\bar{F}(\omega_d, \omega_c, -\omega_a - \omega_b + \omega_p + \omega_q)}{3} F(\omega_a, \omega_c, \omega_p) F(\omega_d, \omega_b, \omega_q) \times \left[\frac{2}{(\omega_a + \omega_c - \omega_p)(\omega_b + \omega_d - \omega_q)} + \frac{1}{(\omega_a + \omega_c - \omega_p)(\omega_a + \omega_b + \omega_c + \omega_d - \omega_p - \omega_q)} + \frac{1}{(\omega_b + \omega_d - \omega_q)(\omega_a + \omega_b + \omega_c + \omega_d - \omega_p - \omega_q)} \right]. \quad (40)$$

Switching $\omega_p \rightarrow \omega_p + \omega_a + \omega_c$ and $\omega_q \rightarrow \omega_q + \omega_b + \omega_d$ the innermost integral becomes

$$\int d\omega_c d\omega_d \int d\omega_p d\omega_q \frac{\bar{F}(\omega_d, \omega_c, \omega_c + \omega_d + \omega_p + \omega_q)}{3} F(\omega_a, \omega_c, \omega_a + \omega_c + \omega_p) F(\omega_d, \omega_b, \omega_b + \omega_d + \omega_q) \times \left[\frac{2}{\omega_p \omega_q} + \frac{1}{\omega_p + \omega_q} \left(\frac{1}{\omega_p} + \frac{1}{\omega_q} \right) \right]. \quad (41)$$

At this point we use the identity [12]

$$\frac{\mathcal{P}}{\omega_p + \omega_q} \left(\frac{\mathcal{P}}{\omega_p} + \frac{\mathcal{P}}{\omega_q} \right) = \frac{\mathcal{P}}{\omega_p} \frac{\mathcal{P}}{\omega_q} + \pi^2 \delta(\omega_p) \delta(\omega_q), \quad (42)$$

which is valid under the assumption that the test function over which the principal values are being calculated is well behaved. We can then write

$$\Omega_3 = -2\pi i \int d\omega_a d\omega_b [J_3(\omega_a, \omega_b) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}], \quad (43)$$

with

$$J_3(\omega_a, \omega_b) = \int d\omega_c d\omega_d \left[\frac{\pi^2}{3} \bar{F}(\omega_c, \omega_d, \omega_c + \omega_d) F(\omega_a, \omega_d, \omega_a + \omega_d) F(\omega_c, \omega_b, \omega_b + \omega_c) + \int \frac{d\omega_p}{\omega_p} \frac{d\omega_q}{\omega_q} \bar{F}(\omega_c, \omega_d, \omega_c + \omega_d + \omega_p + \omega_q) F(\omega_a, \omega_d, \omega_a + \omega_d + \omega_p) F(\omega_c, \omega_b, \omega_b + \omega_c + \omega_q) \right]. \quad (44)$$

As before, a diagram representing the third-order processes can be drawn. This corresponds to two down-conversion processes in which one of the converted photons from each process is later used for up-conversion, as is seen in panel (c) of Fig. 1.

Note that the treatment for type I SPDC follows easily from what was done above. One needs only to replace b by a to characterize the fact that both down-converted photons are generated with the same polarization and in the same frequency

range; the quantity F in (30) will be symmetric in the first two arguments, $F(x, y, z) = F(y, x, z)$, and the expression for ε will differ by a factor of 2 (see Appendix B).

Finally, let us mention that we have developed a numerical library [13] that uses the CUBATURE software package [14] to efficiently calculate the different Magnus terms for SPDC and SFWM within the approximations introduced in this paper.

IV. TIME ORDERING IN FREQUENCY CONVERSION

Frequency conversion (FC) is another nonlinear process in which three fields interact. In this section we explicitly consider FC employing a χ_2 nonlinearity, but the extension to treat FC by a χ_3 nonlinearity follows in the same way the description of photon generation by SFWM followed from the description of photon generation by SPDC. For a χ_2 nonlinearity, FC occurs when a pump photon with frequency ω_p fuses with a photon with frequency ω_a to create a higher-energy photon with frequency $\omega_b = \omega_a + \omega_p$. The Hamiltonian governing this process is derived in a completely analogous manner to the way (22) is derived in Appendix B. The Hamiltonian governing the process is

$$H_I(t) = -\hbar\varepsilon \int d\omega_a d\omega_b d\omega_p e^{i\Delta t} \Phi(\omega_a, \omega_b, \omega_p) \times \alpha(\omega_p) a(\omega_a) b^\dagger(\omega_b) + \text{H.c.}, \quad (45)$$

where all the variables have the same meaning as in Eq. (22) except that now the phase-matching function for uniform nonlinearity in the region $-L/2$ to $L/2$ is given by

$$\Phi(\omega_a, \omega_b, \omega_p) = \text{sinc}\left(\frac{\Delta k L}{2}\right), \quad (46)$$

$$\Delta k = k_b(\omega_b) - k_a(\omega_a) - k_p(\omega_p), \quad (47)$$

and

$$\Delta = \omega_b - \omega_a - \omega_p. \quad (48)$$

The interaction Hamiltonian (45) can be rewritten as

$$H_I(t) = \int d\omega_a d\omega_b d\omega_p [e^{i\Delta t} h(\omega_a, \omega_b, \omega_p) + \text{H.c.}], \quad (49)$$

with

$$h(\omega_a, \omega_b, \omega_p) = a(\omega_a) b^\dagger(\omega_b) F(\omega_a, \omega_b, \omega_p), \quad (50)$$

$$F(\omega_a, \omega_b, \omega_p) = -\varepsilon \alpha(\omega_p) \Phi(\omega_a, \omega_b, \omega_p). \quad (51)$$

In this case the first Magnus term is

$$\Omega_1 = -2\pi i \int d\omega_a d\omega_b [J_1(\omega_a, \omega_b) a(\omega_a) b^\dagger(\omega_b) + \text{H.c.}], \quad (52)$$

with

$$J_1(\omega_a, \omega_b) = F(\omega_a, \omega_b, \omega_b - \omega_a). \quad (53)$$

For the higher-order terms we find

$$\Omega_2 = -2\pi i \left[\int d\omega_a d\omega'_a G_2^a(\omega_a, \omega'_a) a^\dagger(\omega_a) a(\omega'_a) + \int d\omega_b d\omega'_b G_2^b(\omega_b, \omega'_b) b^\dagger(\omega_b) b(\omega'_b) \right], \quad (54)$$

with

$$G_2^a(\omega_a, \omega'_a) = - \int d\omega_q \int \frac{d\omega_p}{\omega_p} F(\omega'_a, \omega_q, \omega_q - \omega'_a + \omega_p) \times \bar{F}(\omega_a, \omega_q, \omega_q - \omega_a + \omega_p), \quad (55)$$

$$G_2^b(\omega_b, \omega'_b) = \int d\omega_q \int \frac{d\omega_p}{\omega_p} F(\omega_q, \omega_b, \omega_b - \omega_q + \omega_p) \times \bar{F}(\omega_q, \omega'_b, \omega'_b + \omega_p - \omega_q), \quad (56)$$

and

$$\Omega_3 = -2\pi i \int d\omega_a d\omega_b [J_3(\omega_a, \omega_b) a(\omega_a) b^\dagger(\omega_b) + \text{H.c.}], \quad (57)$$

with

$$J_3(\omega_a, \omega_b) = \int d\omega_c d\omega_d \left[\frac{-\pi^2}{3} \bar{F}(\omega_c, \omega_d, \omega_d - \omega_c) \times F(\omega_a, \omega_d, \omega_d - \omega_a) F(\omega_c, \omega_b, \omega_b - \omega_c) + \int \frac{d\omega_p}{\omega_p} \frac{d\omega_q}{\omega_q} \bar{F}(\omega_c, \omega_d, \omega_d - \omega_c + \omega_p - \omega_q) \times F(\omega_a, \omega_d, \omega_d - \omega_a - \omega_q) \times F(\omega_c, \omega_b, \omega_b - \omega_c + \omega_p) \right]. \quad (58)$$

In the last equation it has been assumed again that the function F is well behaved.

Note the very elegant connection the Magnus series provides between FC and SPDC. To obtain the Hamiltonian of FC from that of SPDC it suffices to make the substitutions $a(\omega_a) \rightarrow a^\dagger(\omega_a)$, $a^\dagger(\omega_a) \rightarrow a(\omega_a)$, and $\omega_a \rightarrow -\omega_a$. The corresponding Magnus terms of FC are obtained by performing exactly the same substitutions in the SPDC terms, and rearranging the sign of the variables in the principal value integrals over ω_q and ω_p .

V. DISENTANGLING THE MAGNUS EXPANSION

The calculations for photon generation used the fact that the commutator of two two-mode squeezing Hamiltonians gives two single-mode frequency conversion terms,

$$[a^\dagger(\omega_p) b^\dagger(\omega_q) + \text{H.c.}, a^\dagger(\omega_r) b^\dagger(\omega_s) + \text{H.c.}] = \delta(\omega_q - \omega_s) a^\dagger(\omega_r) a(\omega_p) + \delta(\omega_p - \omega_r) b^\dagger(\omega_s) b(\omega_q) - \text{H.c.}, \quad (59)$$

and the fact that the commutator of a single-mode frequency conversion term with a two-mode squeezing term is again a two-mode squeezing term,

$$[a^\dagger(\omega_s) a(\omega_r) + \text{H.c.}, a^\dagger(\omega_p) b^\dagger(\omega_q) + \text{H.c.}] = \delta(\omega_s - \omega_p) b^\dagger(\omega_q) a^\dagger(\omega_r) + \delta(\omega_r - \omega_p) b^\dagger(\omega_q) a^\dagger(\omega_s) - \text{H.c.} \quad (60)$$

Likewise for FC they used the fact that the commutator of two two-mode frequency conversion terms gives single-mode frequency conversion terms,

$$[a(\omega_p) b^\dagger(\omega_q) + \text{H.c.}, a(\omega_r) b^\dagger(\omega_s) + \text{H.c.}] = \delta(\omega_q - \omega_s) a^\dagger(\omega_p) a(\omega_r) + \delta(\omega_p - \omega_r) b^\dagger(\omega_q) b(\omega_s) - \text{H.c.}, \quad (61)$$

and that the commutator of a two-mode frequency conversion term and a single-mode frequency conversion term gives again a two-mode frequency conversion term,

$$\begin{aligned} & [a^\dagger(\omega_s)a(\omega_r) + \text{H.c.}, a^\dagger(\omega_p)b(\omega_q) + \text{H.c.}] \\ &= \delta(\omega_s - \omega_p)a^\dagger(\omega_r)b(\omega_q) \\ &+ \delta(\omega_r - \omega_p)a^\dagger(\omega_s)b(\omega_q) - \text{H.c.} \end{aligned} \quad (62)$$

These results can be used to inductively show that the unitary that connects states at $t_0 \rightarrow -\infty$ and $t \rightarrow \infty$ has to be of the form

$$\begin{aligned} \mathcal{U}(t, t_0) = \exp \left[-2\pi i \int d\omega_1 d\omega_2 \{ \tilde{G}^a(\omega_1, \omega_2) a^\dagger(\omega_1) a(\omega_2) \right. \\ \left. + \tilde{G}^b(\omega_1, \omega_2) b^\dagger(\omega_1) b(\omega_2) \right. \\ \left. + (\tilde{J}(\omega_1, \omega_2) a^\dagger(\omega_1) b^\dagger(\omega_2) + \text{H.c.}) \right], \end{aligned} \quad (63)$$

with

$$\tilde{J} = \sum_i J_{2i+1}, \quad \tilde{G}^a = \sum_i G_{2i}^a, \quad \tilde{G}^b = \sum_i G_{2i}^b, \quad (64)$$

for SPDC and SFWM. For FC one only need to change

$$a^\dagger(\omega_1)b^\dagger(\omega_2) \Rightarrow a^\dagger(\omega_1)b(\omega_2), \quad (65)$$

in Eq. (63). The result (63) confirms in detail the proof given at the end of Sec. II. Equation (63) shows the great utility of the Magnus expansion. It provides us with the general form of the solution. One only needs to calculate the higher corrections to the coefficients that appear in front of the squeezing and frequency conversion terms to get an increasingly better approximation to the solution.

Diagrams similar to those presented in Fig. 1 can be drawn for the higher-order terms. We emphasize that they are used here just as a convenient way of representing the terms in the Magnus expansion; they are *not* Feynman diagrams, of course, and we have not provided any rule to actually do calculations using them. Yet they can help in understanding the physics. In particular, given that the amplitudes they represent will always be multiplied by only pairs of bosonic operators, no matter what the order of the diagram there can be only two free legs at the low down-converted frequencies, which correspond precisely to the frequencies of the bosonic operators that they multiply. The diagrams in Fig. 1 are precisely the three simplest diagrams satisfying this selection rule.

While the form (3) of the unitary operator given by the Magnus expansion identifies the physics of the evolution, particularly as illustrated in the example of (63), it cannot be directly used to calculate the evolution of an initial state, since the terms associated with the functions J_{2i+1} do not commute with the terms associated with the functions G_{2i}^a and G_{2i}^b . In this section we will provide a simple strategy to disentangle the Magnus expansion. We will be able to refactorize the unitary (63) as a product of two unitaries. For the SPDC and SFWM Hamiltonians (22), (27) these unitaries will correspond to a pure squeezing Hamiltonian and pure single-mode frequency

conversion:

$$\mathcal{U} = \mathcal{U}_{\text{sq}} \mathcal{U}_{\text{fc}}, \quad (66)$$

$$\mathcal{U}_{\text{sq}} = e^{-2\pi i \int d\omega_a d\omega_b J(\omega_a, \omega_b) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}}, \quad (67)$$

$$\begin{aligned} \mathcal{U}_{\text{fc}} = e^{-2\pi i \int d\omega_a d\omega'_a G(\omega_a, \omega'_a) a^\dagger(\omega_a) a(\omega'_a)} \\ \times e^{-2\pi i \int d\omega_b d\omega'_b G(\omega_b, \omega'_b) b^\dagger(\omega_b) b(\omega'_b)}. \end{aligned} \quad (68)$$

The case of FC is completely analogous and one only need to follow prescription (65). The factorization (66) has two important features. The first one is that if time ordering is irrelevant then $J = J_1$ as given in Eq. (33) and $G^a = G^b = 0$. The second is that when acted on the vacuum the SPDC or SFWM state is simply

$$\begin{aligned} |\psi\rangle = \mathcal{U} |\text{vac}\rangle = \mathcal{U}_{\text{sq}} \mathcal{U}_{\text{fc}} |\text{vac}\rangle = \mathcal{U}_{\text{sq}} |\text{vac}\rangle \\ = e^{-2\pi i \int d\omega_a d\omega_b J(\omega_a, \omega_b) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}} |\text{vac}\rangle; \end{aligned} \quad (69)$$

that is J is proportional to the joint spectral amplitude (JSA) of the down-converted photons.

To obtain the factorization (66) we will use the BCH formula (19) and the Zassenhaus formula [15],

$$e^{X+Y} = e^X e^Y e^{-\frac{1}{2}[X,Y]} e^{\frac{1}{6}(2[Y,[X,Y]] + [X,[X,Y]])} \dots \quad (70)$$

The lowest-order factorization that can be obtained by just going to second order in the Magnus expansion is to write

$$\mathcal{U} = e^{\Omega_1 + \Omega_2} \approx e^{\Omega_1} e^{\Omega_2} = \mathcal{U}_{\text{sq}} \mathcal{U}_{\text{fc}}. \quad (71)$$

When applied to vacuum one simply obtains

$$|\psi_2\rangle = \mathcal{U} |\text{vac}\rangle = e^{\Omega_1} |\text{vac}\rangle; \quad (72)$$

that is, up to second order the state is not modified by time ordering (since $\Omega_2 |\text{vac}\rangle = 0$), reminiscent of what happens when the Dyson series is used for this same problem [4]. Nevertheless, in the Magnus expansion the meaning of the null contribution of the second order in SPDC and SFWM is transparent: this second-order term requires at least one down-converted photon to be present and hence it will vanish when applied to the vacuum.

Thus it is clear that to see time-ordering corrections to the expression for the state of the generated photons it is necessary to go to at least to third order. In this case one finds

$$\begin{aligned} e^{\Omega_1 + \Omega_2 + \Omega_3} &\approx e^{\Omega_1 + \Omega_3} e^{\Omega_2} e^{-\frac{1}{2}[\Omega_1 + \Omega_3, \Omega_2]} \\ &\approx e^{\Omega_1 + \Omega_3} e^{\Omega_2} e^{-\frac{1}{2}[\Omega_1, \Omega_2]} \\ &= e^{\Omega_1 + \Omega_3} e^{\Omega_2} e^{-\frac{1}{2}[\Omega_1, \Omega_2]} e^{-\Omega_2} e^{\Omega_2} \\ &\approx e^{\Omega_1 + \Omega_3} e^{-\frac{1}{2}e^{\Omega_2}[\Omega_1, \Omega_2]e^{-\Omega_2}} e^{\Omega_2} \\ &\approx e^{\Omega_1 + \Omega_3} e^{-\frac{1}{2}([\Omega_1, \Omega_2] + [\Omega_2, [\Omega_1, \Omega_2]])} e^{\Omega_2} \\ &\approx e^{\Omega_1 + \Omega_3} e^{-\frac{1}{2}[\Omega_1, \Omega_2]} e^{\Omega_2} \\ &\approx e^{\Omega_1 + \Omega_3 - \frac{1}{2}[\Omega_1, \Omega_2]} e^{\Omega_2}. \end{aligned} \quad (73)$$

Here we ignore any fourth-order term, resolve the identity as $\mathbb{I} = e^{-\Omega_2} e^{\Omega_2}$, and use the BCH and Zassenhaus identities and the relation $e^W e^V e^{-W} \equiv e^{e^W V e^{-W}}$. From the last derivation is seen that to calculate the first nonvanishing correction to the JSA we not only need Ω_3 but also $[\Omega_1, \Omega_2]$. This extra third-order correction is easy to understand: The diagram

corresponding to Ω_2 needs an incoming photon, which can only be provided by a previous down-conversion process event included in Ω_1 . This process will be of course of third order since it includes two down-conversion events and one up-conversion event. With Ω_1 a two-mode squeezing operator and Ω_2 containing two single-mode frequency converters, their commutator is another two-mode squeezing operator:

$$\frac{[\Omega_1, \Omega_2]}{2} = -2\pi i^2 \int d\omega_a d\omega_b \times [K_3(\omega_a, \omega_b) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.}], \quad (74)$$

with

$$K_3(\omega_a, \omega_b) = \pi \int d\omega [G_2^a(\omega_a, \omega) J_1(\omega, \omega_b) + J_1(\omega_a, \omega) G_2^b(\omega_b, \omega)]. \quad (75)$$

Equation (73) provides a compact formula accounting for the effects of time ordering, and leads to an identification of the form of the joint spectral amplitude, which to this level of approximation is proportional to

$$J(\omega_a, \omega_b) = J_1(\omega_a, \omega_b) + [J_3(\omega_a, \omega_b) - iK_3(\omega_a, \omega_b)], \quad (76)$$

where the first contribution $J_1(\omega_a, \omega_b)$ neglects effects of time ordering, while the additional terms take corrections into account to all orders lower than the fifth (this last fact we shall prove shortly). This illustrates that as the intensity of the pump field increases and the corrections become larger not only are more squeezed photons generated in the same modes, but their spectral properties change. In particular, if the functions characterizing the pump pulse and the phase-matching functions are real, then as the pump intensity is increased the phase structure of the joint spectral amplitude evolves from trivial to nontrivial.

From the general properties of the Magnus expansion, i.e., the fact that the n th-order Magnus terms is made of $n - 1$ commutators, we know that all even Magnus terms will be pairs of single-mode frequency conversion generators, whereas all odd terms will be two-mode squeezing operators. This fact can be used to show that the commutator of two Magnus terms $[\Omega_i, \Omega_j]$ (which is an $i + j$ order term) will be a frequency conversion term if $i + j$ is even and a squeezing term if $i + j$ is odd. With these two facts and after some algebra it can be shown that the extension of (73) to fifth order is

$$\mathcal{U} = \exp(\Omega_1 + \Omega_2 + \Omega_3 + \Omega_4 + \Omega_5) = \mathcal{U}_{\text{sq}} \mathcal{U}_{\text{fc}}, \quad (77)$$

where the squeezing and frequency conversion unitaries \mathcal{U}_{sq} , \mathcal{U}_{fc} are

$$\mathcal{U}_{\text{sq}} = \exp(X), \quad (78)$$

$$X = \Omega_1 + (\Omega_3 - \frac{1}{2}[\Omega_1, \Omega_2]) + (\Omega_5 - \frac{1}{2}[\Omega_1, \Omega_4] - \frac{1}{2}[\Omega_3, \Omega_2] + \frac{5}{6}[\Omega_2, [\Omega_2, \Omega_1]] + \frac{1}{2}[\Omega_1, [\Omega_1, \Omega_3]]), \quad (79)$$

$$\mathcal{U}_{\text{fc}} = \exp(Y), \quad (80)$$

$$Y = \Omega_2 + (\Omega_4 + \frac{1}{2}[\Omega_1, [\Omega_1, \Omega_2]]). \quad (81)$$

We now consider difference frequency generation, where besides an injected pump pulse that is treated classically we also seed the modes at ω_a and/or ω_b , with injected pulses described by coherent states. Here the output state can be written as

$$|\psi_3\rangle = e^{\Omega_1 + \Omega_3 - \frac{1}{2}[\Omega_1, \Omega_2]} e^{\Omega_2} \times e^{i \sum_{c=a,b} [\int d\omega_c f_c(\omega_c) c^\dagger(\omega_c) + \text{H.c.}]} |\text{vac}\rangle = (e^{\Omega_1 + \Omega_3 - \frac{1}{2}[\Omega_1, \Omega_2]}) \times (e^{\Omega_2} e^{i \sum_{c=a,b} [\int d\omega_c f_c(\omega_c) c^\dagger(\omega_c) + \text{H.c.}]} e^{-\Omega_2}) |\text{vac}\rangle. \quad (82)$$

Keeping only terms up to third order, we find

$$e^{\Omega_2} e^{i \sum_{c=a,b} [\int d\omega_c f_c(\omega_c) c^\dagger(\omega_c) + \text{H.c.}]} e^{-\Omega_2} = e^{i \sum_{c=a,b} [\int d\omega_c f_c(\omega_c) e^{\Omega_2} c^\dagger(\omega_c) e^{-\Omega_2} + \text{H.c.}]} = e^{i \sum_{c=a,b} (\int d\omega_c f_c(\omega_c) [c^\dagger(\omega_c) + [\Omega_2, c^\dagger(\omega_c)]] + \text{H.c.})}. \quad (83)$$

The commutator inside the exponential can be easily calculated to be

$$[\Omega_2, a^\dagger(\omega'_a)] = -2\pi i \int d\omega_a a^\dagger(\omega_a) G_2^a(\omega_a, \omega'_a), \quad (84)$$

$$[\Omega_2, b^\dagger(\omega'_b)] = -2\pi i \int d\omega_b b^\dagger(\omega_b) G_2^b(\omega_b, \omega'_b). \quad (85)$$

This shows that the first effect due to time-ordering corrections in stimulated processes is to modify the shape of the injected seed pulses.

Note that although the discussion in this section was phrased in terms of the SPDC and SFWM Hamiltonians all its conclusions are directly applicable to the case of FC.

VI. TIME-ORDERING CORRECTIONS IN BROADLY PHASE-MATCHED PROCESSES

Here we consider time-ordering corrections in the special limit that the phase-matching function is independent of its variables,

$$\Phi(\omega_a, \omega_b, \omega_p) \propto 1, \quad (86)$$

in Eqs. (25) and (46). This implies that the function F in (30) and (51) is a function only of its last argument, $F(x, y, z) = F(z)$. We show below that if this holds the second and third order Magnus terms vanish. We conjecture that in this limit *all* Magnus terms Ω_i beyond Ω_1 vanish for PDC and FC; For FC this has been shown to hold [16].

To proceed, we look at $G_2^a(\omega_a, \omega'_a)$ as a typical term. For PDC, in the limit (86) it becomes

$$G_2^a(\omega_a, \omega'_a) = \int d\omega_q \int \frac{d\omega_p}{\omega_p} F(\omega_a, \omega_q, \omega_a + \omega_p + \omega_q) \times \bar{F}(\omega'_a, \omega_q, \omega'_a + \omega_p + \omega_q) = \int d\omega_q \int \frac{d\omega_p}{\omega_p} F(\omega_a + \omega_p + \omega_q) \times \bar{F}(\omega'_a + \omega_p + \omega_q). \quad (87)$$

In the last equation we are free to move the origin of the integral in ω_q by ω_p to get

$$G_2^a = \int d\omega_q F(\omega_a + \omega_q) \bar{F}(\omega'_a + \omega_q) \int \frac{d\omega_p}{\omega_p}; \quad (88)$$

using the identity

$$\int \frac{dx}{x+y} = 0, \quad (89)$$

the innermost integral is identically zero. The same kind of argument shows that G_2^b vanishes. Finally, to show that the third-order term is also zero we proceed along similar lines. Using expression (41), with $F(x, y, z) = F(z)$ we obtain

$$\begin{aligned} & \int d\omega_c d\omega_d \int d\omega_p d\omega_q \frac{\bar{F}(\omega_c + \omega_d + \omega_p + \omega_q)}{3} \\ & \times F(\omega_a + \omega_c + \omega_p) F(\omega_b + \omega_d + \omega_q) \\ & \times \left[\frac{2}{\omega_p \omega_q} + \frac{1}{\omega_p + \omega_q} \left(\frac{1}{\omega_p} + \frac{1}{\omega_q} \right) \right], \quad (90) \end{aligned}$$

and as before we shift the integration axes, in this case, $\omega_c \rightarrow \omega_c - \omega_q$, $\omega_d \rightarrow \omega_d - \omega_p$, to get

$$\begin{aligned} & \int d\omega_c d\omega_d \bar{F}(\omega_c + \omega_d) F(\omega_a + \omega_d) F(\omega_b + \omega_c) \\ & \times \int d\omega_p d\omega_q \left[\frac{2}{\omega_p \omega_q} + \frac{1}{\omega_p + \omega_q} \left(\frac{1}{\omega_p} + \frac{1}{\omega_q} \right) \right] \\ & = 0. \quad (91) \end{aligned}$$

VII. CONCLUSIONS

In this paper we have used the Magnus expansion to construct solutions to the time-dependent Schrödinger equation describing three nonlinear processes governed by Hamiltonians that do not commute at different times: spontaneous parametric down-conversion, spontaneous four-wave mixing, and frequency conversion. In the derivation of the Magnus terms for these three processes we found a rather simple form for the second and third order Magnus terms for a broad class of Hamiltonians that satisfy Eq. (8). The Magnus expansion solution that we found for the time evolution operator was shown to be Gaussian preserving; equivalently, when the unitary operator is used to transform input and output bosonic operators the transformation is Bogoliubov linear. With the expansion we developed, it is easy to see why the second-order effect due to the time ordering in SPDC vanishes: Essentially the second-order process needs to be stimulated by a previous photon. We also showed how to calculate corrections to the joint spectral amplitude of the photons in SPDC and SFWM, and how even when the phase-matching function and pump functions are real the JSA acquires a nontrivial phase structure due to time ordering; this structure can be investigated via tomographic methods [17]. We also explored how the effect of time ordering acts in stimulated PDC; in particular, we looked at the case when the lower-energy photons are prepared in coherent states and showed that the first correction due to time ordering is to modify their spectral structure. Finally, the Magnus expansion also allowed us to argue that time ordering is irrelevant if the phase-matching function is infinitely broad.

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VIII. APPENDICES

In these appendices we construct estimates for the form of the terms appearing in the interaction Hamiltonians we use in the text. If waveguides are employed in any actual experiment, a full description would need to take into account the form of the mode fields; if bulk crystals are employed instead, a full description would require the details of the focusing of the pump and radiation pattern of the generated photons. Since our purpose here is not to give a precise calculation for any particular system, but rather to identify the characteristic size and form of the terms needed in our calculations, we simply approximate the relevant field at each frequency in the linear regime as if it consisted of an area A “cut out” of a linearly polarized plane wave propagating in a direction perpendicular to the plane of the area; this can be taken as a “toy model” of a waveguide mode. In Appendix A we relate the energy in a pulse propagating in such a toy model of a waveguide to the field amplitude. With further approximations relating to the relevant tensor components of the nonlinear susceptibilities, in Appendix B we then construct the interaction Hamiltonian for a χ_2 process, and in Appendix C we give the corresponding calculation for a χ_3 process.

APPENDIX A: ENERGY IN AN ELECTROMAGNETIC PULSE

A solution to Maxwell’s equations in a medium with index of refraction $n(\omega)$ is given by

$$\mathbf{E}(z, t) = (\hat{\mathbf{x}} E_0 e^{i(kz - \omega t)}) + \text{c.c.}, \quad (A1)$$

$$\mathbf{B}(z, t) = (\hat{\mathbf{y}} B_0 e^{i(kz - \omega t)}) + \text{c.c.}, \quad (A2)$$

with $E_0/B_0 = c/n(\omega)$ and $k = k(\omega)$. To construct pulses we take superpositions,

$$\begin{aligned} \mathbf{E}(z, t) &= \hat{\mathbf{x}} \int_0^\infty d\omega E_0(\omega) e^{i(kz - \omega t)} + \text{c.c.}, \\ \mathbf{B}(z, t) &= \hat{\mathbf{y}} \int_0^\infty d\omega B_0(\omega) e^{i(kz - \omega t)} + \text{c.c.} \quad (A3) \end{aligned}$$

In what follows we will assume that the amplitude functions $E(\omega)$ and $B(\omega)$ are very narrowly peaked around some central frequency ν , and thus over the relevant integration range to good approximation we can write $E(\omega) = E_0 f(\omega)$ and $B(\omega) = B_0 f(\omega)$, where $E_0/B_0 = c/n(\nu)$, taking $f(\omega)$ to vanish for $\omega < 0$. With this in mind, in a nonmagnetic medium the Poynting vector is

$$\mathbf{S}(z, t) = \frac{\mathbf{E}(z, t) \times \mathbf{B}(z, t)}{\mu_0} \quad (A4)$$

$$\begin{aligned} &= \hat{\mathbf{z}} \frac{E_0 B_0}{\mu_0} \int_0^\infty d\omega d\omega' \\ & \times [f(\omega) f(\omega') e^{i[k(\omega) + k(\omega')]z - (\omega + \omega')t} \\ & + f(\omega) \bar{f}(\omega') e^{i[k(\omega) - k(\omega')]z - (\omega - \omega')t}] + \text{c.c.}, \quad (A5) \end{aligned}$$

where we use an overbar to indicate complex conjugation. Writing a directed element of area $d\mathbf{A}$ as $\hat{\mathbf{z}}dA$, from Poynting's theorem the energy carried in the pulse over an area A is then given by

$$U_{em} = \int_{-\infty}^{\infty} dt \int_A d\mathbf{A} \cdot \mathbf{S}(z, t) \quad (\text{A6})$$

$$= \frac{2\pi A E_0 B_0}{\mu_0} \int_0^{\infty} d\omega [f(\omega)f(-\omega)e^{i(k(\omega)+k(-\omega))z} + |f(\omega)|^2 + \text{c.c.}] \quad (\text{A7})$$

The first term is identically zero since $f(\omega) = 0$ if $\omega < 0$; if we then, for example, approximate $f(\omega)$ for $\omega > 0$ by

$$f(\omega) = \frac{1}{\sqrt{2\pi}\sigma} \zeta(\omega - \nu), \quad (\text{A8})$$

$$\zeta(\omega) = \exp[-\omega^2/(2\sigma^2)],$$

and take

$$k(\omega) = k(\nu) + (\omega - \nu)/v_g, \quad (\text{A9})$$

with v_g the group velocity at frequency ν , we have

$$\mathbf{E}(z, t) = 2\hat{\mathbf{x}}E_0 e^{-\frac{1}{2}\sigma^2(t-z/v_0)^2} \cos[k(\nu)z - \nu t], \quad (\text{A10})$$

$$\mathbf{B}(z, t) = 2\hat{\mathbf{y}}B_0 e^{-\frac{1}{2}\sigma^2(t-z/v_0)^2} \cos[k(\nu)z - \nu t], \quad (\text{A11})$$

where we have assumed that $\nu \gg \sigma$ and thus one can safely extend the lower integration limit in (A3) from 0 to $-\infty$. Averaged over the period $2\pi/\nu$, the Poynting vector following from (A4) has a standard deviation $\sqrt{2}\sigma$, leading to a full width at half maximum (FWHM) of the intensity of the pulse given by $\text{FWHM} = 2\sqrt{2\ln 2}(\sqrt{2}\sigma)$, or $\sigma = \text{FWHM}/4\sqrt{\ln 2} \approx 0.3 \text{FWHM}$. From Eqs. (A6) and (A7), under the same approximations leading to Eqs. (A10) and (A11), we have

$$U_{em} = 2A\epsilon_0 n(\nu) c E_0^2 \frac{\sqrt{\pi}}{\sigma}. \quad (\text{A12})$$

This is a useful result that allows us to relate the peak amplitude of the electric field E_0 to the net energy in the pulse U_{em} :

$$E_0 = \sqrt{\frac{(U_{em}/\sqrt{\pi})\sigma}{2\epsilon_0 n(\nu) c A}}. \quad (\text{A13})$$

Note that even if group dispersion were included in (A8),

$$k(\omega) = k(\nu) + (\omega - \nu)/v_g + \frac{1}{2}\kappa_\nu(\omega - \nu)^2, \quad (\text{A14})$$

with $\kappa_\nu = (d^2k/d\omega^2)_{\omega=\nu}$, we would still obtain the same relation between E_0 and U_{em} .

APPENDIX B: THE INTERACTION HAMILTONIAN FOR A χ_2 PROCESS

We will be using the notation and conventions introduced by Yang *et al.* [7]. The nonlinear Hamiltonian in the Schrödinger picture (Eq. (13) of [7]) is written as

$$H_I = - \int_k dk_a dk_b dk_c S_I(k_c, k_a, k_b) c(k_c) a^\dagger(k_a) b^\dagger(k_b) + \text{H.c.}, \quad (\text{B1})$$

where $[c(k), c^\dagger(k')] = [b(k), b^\dagger(k')] = [a(k), a^\dagger(k')] = \delta(k - k')$ and any other commutator between operators equals zero. We assume that the wave number ranges and/or the polarizations of the pump photons [destruction operator $c(k_c)$] and the down-converted photons [destruction operators $a(k_a)$ and $b(k_b)$] are distinct. Letting the frequency associated with k_l be ω_l , where $l = a, b$, or c , the coupling coefficient $S_I(k_c, k_a, k_b)$ is given by

$$S_I(k_c, k_a, k_b) = \frac{2}{\epsilon_0} \sqrt{\frac{\hbar\omega_c \hbar\omega_a \hbar\omega_b}{(4\pi)^3}} \frac{\chi_2}{\epsilon_0 n_c^2(\omega_c) n_a^2(\omega_a) n_b^2(\omega_b)} \times \int_A dx dy d_c(x, y) \bar{d}_a(x, y) \bar{d}_b(x, y) \times \int_{-L/2}^{L/2} dz e^{i(k_c - k_a - k_b)z}, \quad (\text{B2})$$

where $n_l(\omega_l)$ is effective index of the indicated mode type. Here we have simplified the corresponding expression of Yang *et al.* [7] by neglecting the tensor nature of χ_2 and the vector nature of the $d_l(x, y)$, which characterize the mode profiles of the displacement field; in our expression (B2) for $S_I(k_c, k_a, k_b)$ the parameter χ_2 can be taken to characterize the strength of the nonlinearity, which we assume to be present only from $-L/2$ to $L/2$, and in the spirit of our approximations in the previous appendix we take the $d_l(x, y)$ to be uniform over an area A . An extra factor of 2 appears in (B2) compared with the corresponding Eq. (13) of Yang *et al.* [7] because here we assume that the creation operators $a^\dagger(\omega_a)$ and $b^\dagger(\omega_b)$ involve different ranges of frequency and/or polarization. Since the actual vector $\mathbf{d}_l(x, y)$ at ω_l are normalized according to (Eq. (8) of [7])

$$\int dx dy \frac{\mathbf{d}_l(x, y) \cdot \bar{\mathbf{d}}_l(x, y)}{\epsilon_0 n_l^2(\omega_l)} \frac{v_p(l)}{v_g(l)} = 1,$$

where $v_p(l) = c/n_l(\omega_l)$ and $v_g(l) = d\omega_l/dk_l$ are the phase and group velocities, this leads to

$$d_l = \sqrt{\frac{v_g(l) n_l^2(\omega_l) \epsilon_0}{v_p(l) A}}, \quad (\text{B3})$$

and using this in (B2) we have

$$S_I(k_c, k_a, k_b) = 2L \sqrt{\frac{\hbar\omega_c \hbar\omega_a \hbar\omega_b}{(4\pi)^3}} \chi_2 \times \sqrt{\frac{v_g(a)v_g(b)v_g(c)}{\epsilon_0 A v_p(a)v_p(b)v_p(c) n_a^2 n_b^2 n_c^2}} \times \text{sinc}\left[\frac{L}{2}(k_c - k_a - k_b)\right]. \quad (\text{B4})$$

For fields propagating in a single direction it is conventional to work with frequency rather than wave number labels; hence we take, for example, $c(k_c) \rightarrow \sqrt{d\omega_c/dk_c} c(\omega_c)$, such that $[c(\omega_a), c^\dagger(\omega'_a)] = \delta(\omega_a - \omega'_a)$ (see, e.g., Eq. (50) in [7]). Then changing to frequency variables in (B2)

we have

$$\begin{aligned}
H_I(t) &= -2 \int_0^\infty d\omega_a d\omega_b d\omega_c L \chi_2 c(\omega_c) a^\dagger(\omega_a) b^\dagger(\omega_b) e^{i\Delta t} \\
&\times \sqrt{\frac{\hbar\omega_c \hbar\omega_a \hbar\omega_b}{(4\pi)^3 \epsilon_0 A c^3 n_a(\omega_a) n_b(\omega_b) n_c(\omega_c)}} \\
&\times \text{sinc}\left(\frac{L}{2}[k(\omega_c) - k(\omega_a) - k_b(\omega_b)]\right) + \text{H.c.},
\end{aligned} \tag{B5}$$

where we have also moved to the interaction picture defined by linear evolution, $H_I \rightarrow H_I(t)$, with $c(\omega_c) a^\dagger(\omega_a) b^\dagger(\omega_b)$ picking up the phase factor $\exp(i\Delta t)$, where $\Delta = \omega_a + \omega_b - \omega_c$. In the same interaction picture the electric field operator can be related to the displacement field operator in the usual way; using the expression for the displacement field operator (Eq. (4) of [7]) and our expression (B3) for the mode profile function we find

$$\begin{aligned}
\mathbf{E}(\mathbf{r}, t) &= \sum_l \int_0^\infty \frac{d\omega_l}{n_l} \sqrt{\frac{\hbar\omega_l}{4\pi\epsilon_0 v_p(l)A}} q_l(\omega_l) \\
&\times e^{ik(\omega_l)z - i\omega_l t} + \text{H.c.},
\end{aligned}$$

where as l is summed over a, b , and c , $q_l(\omega_l)$ ranges over the destruction operators $a(\omega_a)$, $b(\omega_b)$, and $c(\omega_c)$. Taking the expectation value of the above operator with respect to a coherent state for the pump field (and vacuum for the down-converted photon frequencies) turns $c(\omega_c)$ into a c number $\beta(\omega)$, where we drop the subscript c . Now we can compare this with a classical pulse with energy U_{em} centered at ν_l and frequency variance σ^2 (A13):

$$\begin{aligned}
\mathbf{E}(\mathbf{r}, t) &= E_0 \int_0^\infty d\omega e^{i[k(\omega)z - \omega t]} \frac{1}{\sqrt{2\pi\sigma}} e^{-(\omega - \nu_l)^2/2\sigma^2} \\
&= \int_0^\infty d\omega \sqrt{\frac{(U_{em}/\sqrt{\pi})\sigma}{2\epsilon_0 n(\omega) c A}} e^{i[k(\omega)z - \omega t]} \\
&\times \frac{1}{\sqrt{2\pi\sigma}} e^{-(\omega - \nu_l)^2/2\sigma^2},
\end{aligned} \tag{B6}$$

and identify

$$\beta(\omega) = \sqrt{\frac{U_{em}/\sqrt{\pi}}{\hbar\omega\sigma}} e^{-(\omega - \nu_l)^2/2\sigma^2}.$$

It is convenient to replace σ by with the inverse standard deviation of the time-dependent pump intensity, $\tau = 1/\sqrt{2}\sigma = 2\sqrt{2\ln 2}$ FWHM, and write

$$\beta(\omega) = \sqrt{\frac{U_{em}/\sqrt{\pi/2}}{\hbar\tau\omega}} \tau e^{-\tau^2(\omega - \nu_l)^2}. \tag{B7}$$

With this in hand we can return to the nonlinear Hamiltonian (B5) and write

$$\begin{aligned}
H_I(t) &= -\hbar\varepsilon \int_0^\infty d\omega_a d\omega_b d\omega_c \frac{\tau}{\sqrt{\pi}} e^{-\tau^2(\omega_c - \nu_c)^2} \\
&\times \text{sinc}\left(\frac{L}{2}[k(\omega_c) - k_a(\omega_a) - k_b(\omega_b)]\right) \\
&\times e^{i\Delta t} a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{H.c.},
\end{aligned} \tag{B8}$$

for an SPDC process with a classically described undepleted pump mode c , where we introduced the dimensionless perturbation parameter ε ,

$$\varepsilon = 2L\chi_2 \sqrt{\frac{\sqrt{2}U_{em}\pi v_b v_a}{\sqrt{\pi}(4\pi)^3 \epsilon_0 A c^3 n_a(\nu_a) n_b(\nu_b) n_c(\nu_c) \tau}},$$

and in benign frequency factors we have evaluated ω_c at ν_c and ω_a and ω_b at the frequencies ν_a and ν_b , where the phase-matching condition is assumed to be satisfied. Defining

$$\alpha(\omega_c) = \frac{\tau}{\sqrt{\pi}} e^{-\tau^2(\omega_c - \nu_c)^2},$$

which has units of time, and changing the notation for the pump frequency from ω_c to ω_p , (B8) reduces to (23), with $\Phi(\omega_a, \omega_b, \omega_p)$ given by (26).

APPENDIX C: THE INTERACTION HAMILTONIAN FOR A χ_3 PROCESS

For a third-order process, the straightforward extension of the nonlinear Hamiltonian (B1) [7] in the Schrödinger picture is given by

$$H_I = -\frac{1}{4\epsilon_0} \int_V d\mathbf{r} \Gamma_3^{ijkl} D^i(\mathbf{r}) D^j(\mathbf{r}) D^k(\mathbf{r}) D^l(\mathbf{r}), \tag{C1}$$

where indices refer to Cartesian components and are to be summed over when repeated; Γ_3^{ijkl} can be written in terms of the more usual third-order optical susceptibility χ_3^{ijkl} and the linear indices at the appropriate frequency components involved [18]; as in the appendix above we use the symbol χ_3 (without tensor indices) to indicate the size of the component(s) that are important when the relevant mode fields are used in the different $D^i(\mathbf{r})$. For SFWM we require destruction operators for the pump fields ($c(\omega_c)$ and $c(\omega'_c)$) and creation operators for the photons generated described by $a(\omega_a)$ and $b(\omega_b)$, and in analogy with the result of Appendix B we find

$$\begin{aligned}
H_I(t) &= - \int_{-L/2}^{L/2} dz \int_0^\infty d\omega_c d\omega'_c d\omega_b d\omega_a \\
&\times \frac{3\chi_3}{\epsilon_0^3 n_c^2(\omega_c) n_c^2(\omega'_c) n_b^2(\omega_b) n_a^2(\omega_a)} \\
&\times \sqrt{\frac{\hbar\omega_c \hbar\omega'_c \hbar\omega_a \hbar\omega_b}{4\pi \ 4\pi \ 4\pi \ 4\pi} A} \frac{1}{\sqrt{v_g(a)v_g(b)v_g(c)v_g(c')}} \\
&\times \sqrt{\frac{v_g(c')n_c^2(\omega'_c)\epsilon_0 v_g(c)n_c^2(\omega_c)\epsilon_0}{v_p(c')A v_p(c)A}} \\
&\times \sqrt{\frac{v_g(a)n_a^2(\omega_a)\epsilon_0 v_g(b)n_b^2(\omega_b)\epsilon_0}{v_p(a)A v_p(b)A}} \\
&\times a^\dagger(\omega_a) b^\dagger(\omega_b) c(\omega_c) c(\omega'_c) e^{-i(\omega_c + \omega'_c - \omega_a - \omega_b)t} \\
&\times e^{i[k_c(\omega_c) + k_c(\omega'_c) - k_a(\omega_a) - k_b(\omega_b)]z} + \text{H.c.}
\end{aligned} \tag{C2}$$

The factor of 3 in the fraction containing χ_3 is $(4!/2!)/4$, with $(4!/2!)$ the number of permutations involving the ordering of the creating and annihilation operators; note that this is twice the corresponding factor appearing in [18] because here we assume that the creation operators $a^\dagger(\omega_a)$ and $b^\dagger(\omega_b)$

involve different ranges of frequency and/or polarization. The refractive index factors in the denominator, and the appearance of ϵ_0^3 , arise because of the conversion from Γ^{ijkl} to χ_3^{ijkl} and the prefactor of ϵ_0^{-1} in (C1); compare with the corresponding terms in (B2). The terms of the form $\sqrt{\hbar\omega_l/4\pi}$ arise from the expansion of the displacement field in terms of raising and lowering operators, and again find their correspondence in terms in (B2). The factor of A comes from integrating over the area in our “toy model” for our waveguide and its modes; the factor $[v_g(a)v_g(b)v_g(c)v_g(c')]^{-1/2}$ arises from the change of variables from wave numbers to frequencies; and the next terms are the expressions for the mode profiles (B3). Supposing that all the frequency prefactors vary slowly relative to the harmonic dependence in space and time due to the phase factors, in those prefactors we set ω_c and ω'_c at the center frequency ν_c of the pump, and evaluate ω_a and ω_b at the frequencies ν_a and ν_b where the phase-matching is optimum; (C2) then reduces to

$$H_I(t) = -\frac{3\chi_3\hbar^2}{\epsilon_0 A(4\pi)^2 c^2} \sqrt{\left(\frac{\nu_c}{n_c(\nu_c)}\right)^2 \frac{\nu_a}{n_a(\nu_a)} \frac{\nu_b}{n_b(\nu_b)}} \times \int_{-L/2}^{L/2} dz \int_0^\infty d\omega_c d\omega'_c d\omega_b d\omega_a \times a^\dagger(\omega_a) b^\dagger(\omega_b) c(\omega_c) c(\omega'_c) e^{-i(\omega_c + \omega'_c - \omega_a - \omega_b)t} \times e^{i[k_c(\omega_c) + k_c(\omega'_c) - k_a(\omega_a) - k_b(\omega_b)]z} + \text{H.c.} \quad (\text{C3})$$

As in Appendix B we replace the c operators by classical functions (B7) and assume that $\sigma \ll \nu_c$, which allows us to extend the range of the ω_c and ω'_c integrals to $-\infty$ to ∞ , and we find

$$H_I(t) = -\frac{3\chi_3\hbar^2}{\epsilon_0 A(4\pi)^2 c^2} \sqrt{\left(\frac{\nu_c}{n_c(\nu_c)}\right)^2 \frac{\nu_a}{n_a(\nu_a)} \frac{\nu_b}{n_b(\nu_b)}} \times \frac{U_{em}/\sqrt{\pi/2}}{\hbar\nu_c\sigma} \int_{-L/2}^{L/2} dz \int d\omega_c d\omega'_c d\omega_b d\omega_a \times a^\dagger(\omega_a) b^\dagger(\omega_b) e^{-(\omega_c - \nu_c)^2/2\sigma^2} e^{-(\omega'_c - \nu_c)^2/2\sigma^2} \times e^{i[k_c(\omega_c) + k_c(\omega'_c) - k_a(\omega_a) - k_b(\omega_b)]z} e^{-i(\omega_c + \omega'_c - \omega_a - \omega_b)t} + \text{H.c.}$$

To simplify this further we look at the integrals over ω_c and ω'_c ,

$$I = \int d\omega_c d\omega'_c e^{i[k_c(\omega_c) + k_c(\omega'_c)]z - i(\omega_c + \omega'_c)t} \times e^{-(\omega_c - \nu_c)^2/2\sigma^2} e^{-(\omega'_c - \nu_c)^2/2\sigma^2}. \quad (\text{C4})$$

Expanding $k_c(\omega_c)$ and $k_c(\omega'_c)$ as was done earlier (A14), and introducing $\omega_\pm = \omega_c \pm \omega'_c$, we find that (C4) equals

$$I = \int d\omega_+ e^{ik_+(\omega_+)z} e^{-i\omega_+ t} e^{-(\omega_+ - 2\nu_c)^2/4\sigma^2} \times \int \frac{d\omega_-}{2} e^{-\frac{\omega_-^2}{4}(\frac{1}{\sigma^2} - i\kappa_c z)}, \quad (\text{C5})$$

where

$$k_+(\omega_+) = 2k_c(\nu_c) + (\omega_+ - 2\nu_c)/\nu_c + \frac{\kappa_c}{4}(\omega_+ - 2\nu_c)^2. \quad (\text{C6})$$

The innermost integral is

$$\int \frac{d\omega_-}{2} e^{-\frac{\omega_-^2}{4}(\frac{1}{\sigma^2} - i\kappa_c z)} = \sqrt{\pi}\sigma g(z), \quad (\text{C7})$$

where

$$g(z) \equiv \frac{1}{\sqrt{1 - i\kappa_c z \sigma^2}}.$$

We can then finally write the Hamiltonian as

$$H_I(t) = -\hbar\epsilon \int d\omega_+ \int d\omega_b \int d\omega_a \tilde{\Phi}(\Delta k) \times e^{i\tilde{\Delta}t} a^\dagger(\omega_a) b^\dagger(\omega_b) \tilde{\alpha}(\omega_+) + \text{H.c.}, \quad (\text{C8})$$

where

$$\tilde{\alpha}(\omega_+) \equiv \frac{\tilde{\tau} e^{-\tilde{\tau}^2(\omega_+ - 2\nu_c)^2}}{\sqrt{\pi}}$$

(with units of time) characterizes the bandwidth of the pump, and we have introduced $\tilde{\tau} = 1/(2\sigma)$ and $\tilde{\Delta} \equiv \omega_a + \omega_b - \omega_+$ (note the difference between these quantities and the corresponding $\tilde{\alpha}$, τ , and Δ of Appendix B); the dimensionless perturbation parameter

$$\epsilon = \frac{6\sqrt{2\pi}\chi_3 U_{em} L \sigma}{\epsilon_0 A(4\pi)^2 n_c c^2} \sqrt{\frac{\nu_a \nu_b}{n_a n_b}}$$

characterizes the strength of the nonlinear interaction, and the modified phase matching function is

$$\tilde{\Phi}(\Delta k) \equiv \int_{-L/2}^{L/2} \frac{dz}{L} e^{i[k_a(\omega_a) + k_b(\omega_b) - k_+(\omega_+)]z} g(z). \quad (\text{C9})$$

If the effect of group velocity dispersion on the pump pulse is negligible over the length L of the nonlinear region, for all relevant z we have $|\kappa_c z \sigma^2| \ll 1$, we can set $g(z) \rightarrow 1$ and $k_+(\omega_+) = 2k_c(\omega_+/2)$ [cf. (C6)] and (C9) reduces to (22). Finally, note that we can rewrite expression (C8) in a slightly more suggestive manner,

$$H_I(t) = -\hbar\epsilon \int d\omega_+ \int d\omega_b \int d\omega_a \tilde{\Phi}(\Delta k) \times e^{i\tilde{\Delta}t} a^\dagger(\omega_a) b^\dagger(\omega_b) \frac{\zeta(\omega_+/2 - \nu_c)^2}{2\sigma} + \text{H.c.}, \quad (\text{C10})$$

where $\zeta(\omega)$ is proportional to the electric field amplitude as defined in (A8), explicitly displaying the dependence of $H_I(t)$ on the square of that amplitude. Of course, the details of the argument given here rely on the assumed Gaussian nature of the pump. But we can expect that the general form (C8) will survive even for more general pump pulses, as long as the effect of group velocity dispersion on the pump pulse is negligible.

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