Time ordering effects in the generation of entangled photons using nonlinear optical processes

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We study the effects of time ordering in photon generation processes such as spontaneous parametric down-conversion (SPDC) and four wave mixing (SFWM). The results presented here are used to construct an intuitive picture that allows us to predict when time ordering effects significantly modify the joint spectral amplitude (JSA) of the photons generated in SPDC and SFWM. These effects become important only when the photons being generated lie with the pump beam that travels through the non-linear material for a significant amount of time. Thus sources of spectrally separable photons are ideal candidates for the observation of modifications of the JSA due to time ordering.

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Introduction — Particles exhibiting strong, non-classical correlations play a central role in the implementation of quantum-enhanced information processing tasks. Nonlinear photonic materials provide some of the most versatile sources of these particles. The interactions that give rise to the quantum correlated photons are typically rather weak, but by using a strong classical pump field they can be enhanced by orders of magnitude. Thus the theoretical treatment of photon generation usually involves a time dependent Hamiltonian in the interaction picture. Because this Hamiltonian generally does not commute with itself at different times, the time evolution operator in these types of problems is not just a simple exponential involving the time integral of the Hamiltonian, and so-called “time ordering” corrections arise in the description of the quantum state evolution [1, 2, 5].

These time ordering effects can be ignored if the pump pulse is weak enough; to first order in perturbation theory the interaction Hamiltonian appears only once, and the time ordering effects start to become an important issue that must be addressed. One strategy to deal with them is to use the Dyson series [1, 2] for the evolution operator. As discussed elsewhere [11], this method has the drawback that the resulting approximate evolution is not unitary if the series is truncated at any finite order. Perhaps more importantly, the method does not preserve the photon statistics that the exact solution of the problem is known to have, and thus leads to unphysical predictions. An approach to achieving the correct photon statistics was proposed by Christ et al.[3], involving a numerical method that seeks solutions of the Heisenberg equations of motion satisfying the “right” photon statistics.

In this letter, we introduce an analytical model that describes the effects of time ordering in several nonlinear quantum optical processes. The model automatically captures the photon statistics and unitary evolution that characterize the exact solution of the problem. With this model we provide a simple explanation of when time ordering effects become important: As long as the generated photons do not significantly copropagate (i.e., reside in the same spatial region at the same time) with the pump photons, time ordering effects are irrelevant for the description of the generated quantum state.

Model — We consider the following interaction picture Hamiltonian, which can be used to model both spontaneous parametric down-conversion (SPDC) and spontaneous four-wave mixing (SFWM) in a one dimensional geometry, within the undepleted pump and rotating wave approximations:

\[ \hat{H}_I(t) = -\hbar \epsilon \int d\omega_p d\omega_i d\omega_b e^{i\Delta t \Phi(\omega_i, \omega_b, \omega_p)} \alpha(\omega_p) \hat{a}^{\dagger}(\omega_i) \hat{b}^{\dagger}(\omega_b) + \text{h.c.} \]  

Here \( a^{\dagger}(\omega) \) and \( b^{\dagger}(\omega) \) are creation operators for two different modes (perhaps waveguide modes), and \( \Phi \) is the phase matching function, which we take to be a Gaussian function,

\[ \Phi(\omega_i, \omega_b, \omega_p) = \exp \left( -(s_a \delta \omega_i + s_b \delta \omega_b - s_p \delta \omega_p)^2 \right). \]  

When we take \( s_i = \sqrt{3} L/(2v_i) > 0 \), where \( v_i \) is the group velocity at frequency \( \omega_i \), this provides a good approximation to the usual sinc function resulting from a uniform nonlinearity over a region of length \( L \) under the neglect of dispersion effects; the choice of \( \gamma = 0.193 \) guarantees that the full width at half maximum of the sinc and Gaussian functions agree. Here \( \omega_i = \omega_i - \omega_i \); in SPDC the central frequencies \( \bar{\omega}_i \) are constrained by energy conservation, \( \bar{\omega}_i + \bar{\omega}_b = \bar{\omega}_p \); , and momentum conservation, \( k_a(\bar{\omega}_a) + k_b(\bar{\omega}_b) = k_p(\bar{\omega}_p) \), while for SFWM \( \omega_p \) is twice the frequency of the pump and momentum conservation reads \( k_a(\bar{\omega}_a) + k_b(\bar{\omega}_b) = 2k_p(\bar{\omega}_p) / 2 \) [11]. The parameter \( \epsilon \) is a dimensionless constant that characterizes the strength of the interaction[11]; it depends on physical properties such as the effective non-linearity of the
medium and the area of the interaction region. We also assume that the classical pump can be described by a Gaussian function,

\[ \alpha(\omega_p) = \frac{\tau}{\sqrt{\pi}} \exp(-\tau^2\delta\omega_p^2), \]  

with \( \tau \) characterizing the temporal duration of the pump pulse. Finally \( \Delta = \omega_a + \omega_b - \omega_p \) and in Eq. (1) we adopt the convention (used throughout the paper) that whenever the limits of an integral are not specified they range from \(-\infty\) to \(\infty\).

Using the Magnus expansion, it was recently shown [11] that the unitary evolution operator generated by the Hamiltonian (1) can be factored as \( \hat{U} = \hat{U}_a\hat{U}_b \) with \( \hat{U}_a \), a pure two-mode squeezing term (generated by terms that are extensions of \( \hat{a}^\dagger \hat{b}^\dagger + \text{h.c.} \)) and \( \hat{U}_b \), a pure single mode frequency conversion term (generated by terms that are extensions of \( \hat{a}^\dagger \hat{a} + \hat{b}^\dagger \hat{b} \)),

\[ \hat{U} = e^{-2\pi \eta a,b} \sum_n (J_n - iK_n) \hat{a}^\dagger (\omega_n) \hat{b}^\dagger (\omega_n) + \text{h.c.}, \]  

with \( n \) odd and \( m \) even. Each of these indices labels the order of the expansion in \( \varepsilon \); e.g., \( n = 1 \) is a term \( \propto \varepsilon \) and \( m = 2 \) is a term \( \propto \varepsilon^2 \). If a spontaneous process is considered, where the unitary operator \( \hat{U} \) is applied to the vacuum, it is readily seen that the second exponential in (4) does not modify the initial state; \( \hat{U}_b |\text{vac}\rangle = |\text{vac}\rangle \), and hence the output state \( |\psi_{\text{out}}\rangle = \hat{U} |\text{vac}\rangle = \hat{U}_a |\text{vac}\rangle \) is generated solely by the squeezing part \( \hat{U}_a \). With these observations it is easy to identify \( J = \sum_n (J_n - iK_n) \) as the Joint Spectral Amplitude (JSA) of the photons generated in modes \( a \) and \( b \) in a spontaneous process. The first term of the Magnus expansion for the Hamiltonian (1) is obtained by integrating the Hamiltonian from \(-\infty\) to \(\infty\); \( K_1 = 0 \), and the result for \( J_1 \) can be written in the following simple form:

\[ J_1(\delta\omega_a, \delta\omega_b) = -\frac{\varepsilon \tau}{\sqrt{\pi}} \exp(-uNu^T), \]  

\[ N = \begin{pmatrix} \mu_a^2 & \mu_a \mu_b \\ \mu_a \mu_b & \mu_b^2 \end{pmatrix}, \quad u = (\delta\omega_a, \delta\omega_b), \]  

\[ \eta_a,b = s_p - s_{a,b}, \quad \mu^2 = \tau^2 + \eta_a \eta_b, \quad \mu_{a,b}^2 = \tau^2 + \eta_{a,b}^2. \]  

If time ordering effects were ignored, \( K \) would vanish, and \( J_1 \) would be the only contribution to \( J \); \( J_1 \) would thus completely describe the properties of the state, and could be identified with the joint spectral amplitude of the generated photons. A measure of entanglement is the Schmidt number [8], which for the state characterized by \( J_1 \) is [9, 10]

\[ S = \mu_a \mu_b/\sqrt{\mu_a^2 \mu_b^2 - \mu^4}. \]  

In the limit that the matrix \( N \) is diagonal, \( \mu \to 0 \) and the down converted photons are uncorrelated; here \( S = 1 \). If in this limit \( N \) were also proportional to the identity matrix, then the photons would not only be uncorrelated but would share the same spectral characteristics; for this to happen \( \eta_a = -\eta_b \). In the opposite limit, when

\[ \tau \ll |\eta_{a,b}|, \]  

\( N \) becomes rank deficient, the photons generated in the modes \( a \) and \( b \) are strongly correlated, and \( S \to \infty \). A simple way to understand the physics in the limit (7) is to rewrite the inequality as

\[ v_p \tau/|v_{a,b} - v_p| \ll L/|v_{a,b}|. \]

That is, in this limit the time the down converted photons spend with the pump photons is much less than the time they spend in the nonlinear structure. The down converted photons once created thus escape the nonlinear structure without spending appreciable time in the same region as the pump photons, and if the pump intensity is high enough to generate a number of pairs of photons then the stream of pairs is spread over a long time; equivalently, their frequencies are tightly constrained.

**Time ordering effects in spontaneous photon generation** — For the simple Gaussian model considered in this letter, the real part of the first non vanishing correction to the JSA due to time ordering is given by

\[ J_3(\omega_a, \omega_b) = W(\omega_a, \omega_b) - Z(\omega_a, \omega_b)V(\omega_a, \omega_b), \]  

\[ W(\omega_a, \omega_b) = \frac{2\pi^{3/2} \tau^{3/2}}{3R^2} \exp(-uQu^T/R^2), \]  

\[ Z(\omega_a, \omega_b) = 4\pi\tau^{3/2} \exp(-uNu^T/3), \]  

\[ V(\omega_a, \omega_b) = \int_0^\infty dp \int_0^\infty dq \exp(-q(p, q)M(p, q)^T) \times \]  

\[ \cos(4\tau \eta_{a,b}(\delta\omega_a q + \delta\omega_b p)/\sqrt{3}), \]  

with

\[ Q = \begin{pmatrix} M^4 & \mu_a^2 M^2 & \mu_b^2 M^2 \\ \mu_a^2 M^2 & M^2 & \mu_b^2 \\ \mu_b^2 M^2 & \mu_b^2 & M^2 \end{pmatrix}, \quad M = \begin{pmatrix} 2\mu_a^2 & 2\mu_b^2 \\ 2\mu_a^2 & 2\mu_b^2 \end{pmatrix}, \]  

\[ R^4 = 4\mu_a^2 \mu_b^2 - \mu^4 = 4\eta_{a,b}^2 \tau^{2} + 3\mu^4, \]  

\[ M^4 = 4\mu_a^2 \mu_b^2 - 3\mu^4, \quad \eta_{a,b} = -\eta_{b,a} = \eta_a - \eta_b. \]

The imaginary part of the correction \( K_3 \) is given by

\[ K_3(\omega_a, \omega_b) = -\varepsilon^3 \pi^{3/2} \tau^3 \exp(-uQu^T/R^4) \times \]  

\[ \left( \text{erfi}(y_{a,b}) + \text{erfi}(y_{b,a}) \right), \]  

\[ y_{a,b} = \frac{2}{3} \tau \eta_{a,b} (2\delta\omega_a \mu_a^2 - \delta\omega_b \mu_b^2) \]  

\[ \sqrt{R^2 \mu_a^2 \mu_b^2}, \]  

and \( \text{erfi}(z) = \text{erf}(iz)/i \), with \( \text{erf} \) being the standard error function. Details of the calculation are given in the Supplementary Material. A useful feature of using Gaussian functions to model both the phase-matching function and the pump pulse is that estimates of the relative sizes of the corrections due to time-ordering in SPDC and
SFWM can be easily extracted; this also holds for the time-ordering corrections to frequency conversion. We demonstrate this all below, introducing figures of merit which characterize the size of the time-ordering corrections relative to the uncorrected prediction that would follow from the first order Magnus calculation. Some of the mathematical details are relegated to the Supplementary Material.

To estimate the effects of time ordering on SPDC and SFWM we introduce the following figure of merit,

\[ r = \frac{\max_{\omega_a, \omega_b} |J_3(\omega_a, \omega_b)|}{\max_{\omega_a, \omega_b} |J_1(\omega_a, \omega_b)|} = \frac{\sqrt{\pi}}{\varepsilon \tau} \max_{\omega_a, \omega_b} |J_3(\omega_a, \omega_b)|. \]

In the Supplementary Material we show that

\[ |J_3(\omega_a, \omega_b)| \leq \frac{2\pi^3/2\varepsilon^3}{R^2}, \quad (15) \]

and so we identify the bound

\[ r \leq 2\pi^2 \varepsilon^2 \left( \frac{\tau^2}{R^2} \right). \quad (16) \]

There are two interesting limits of the important ratio \( \tau^2/R^2 \) that appears in (16). We find

\[ \tau \ll |\eta_{a,b}| \Rightarrow \frac{\tau^2}{R^2} \sim \frac{1}{\sqrt{3}} \frac{\tau}{|\eta_a| |\eta_b|} \ll 1, \quad (17) \]

\[ \mu = 0 \Rightarrow \frac{\tau^2}{R^2} = \frac{\tau}{2|\eta_{ab}|}. \]

From this we can see that the importance of the time ordering corrections depends strongly on the entanglement of the photons in modes \( a \) and \( b \), and we can also understand this scaling physically. As discussed in the previous section, if the generated photons are highly entangled with a large Schmidt number, the time they overlap the pump field is short. We would then expect on physical grounds that time ordering corrections should be small, and indeed we see from the first of (17) and (16) that a very large interaction strength \( \varepsilon \) would be necessary for the time ordering corrections to come into play. In the reverse limit, the second of (17), the group velocities of the generated photons satisfy \( \eta_a \eta_b = -\tau^2 \) (\( \mu = 0 \)).

To have a significant time ordering correction, we have from the second of (17) that the group velocities of the downconverted photons must be similar. Note that if the velocities were equal \( J_3 \) would vanish identically; see the Supplementary Material. It can also be shown that the maximum of \( \tau^2/R^2 \) under the constraint of having \( \mu = 0 \) is obtained for

\[ \eta_a = -\eta_b = \pm \tau \Rightarrow \frac{v_p \tau}{|v_{a,b} - v_p|} = \frac{\sqrt{\pi}}{2} \frac{L}{v_{a,b}}. \quad (18) \]

then \( \tau^2/R^2 \) equals 1/4. Eq. (18) simply tells us that the time the downconverted photons spend with the pump is comparable with the time they spend in the crystal. Note that the entanglement in the downconverted state as predicted by using \( J_1 \) for \( J \) and the condition (18) is very small, but because of the continued presence of photons being generated with the pump pulse, time-ordering corrections would be physically expected to come into play at a smaller interaction strength \( \varepsilon \); this is confirmed by using (18) in (16). Furthermore, besides being simply larger, time ordering corrections can be expected to be of greater significance here, since they can qualitatively modify the very small entanglement that the approximation of \( J \) by \( J_1 \) would imply.

When appropriate values of \( \varepsilon \) are used in (16), we see that our results are consistent with what was found by Christ et al. [3], in which simulations showed that extremely high pump intensities are necessary to obtain observable time ordering effects.

Time ordering effects in stimulated photon generation — We now turn to stimulated processes, in which the unitary operator \( U_{\text{out}} \) in (4) characterizing single mode frequency conversion becomes important. The first contribution to the frequency conversion unitary is given by

\[ G_2^a(\omega_a, \omega_a') = \varepsilon e^{\frac{2\pi i \tau^2}{2 |\mu_b|}} e^{-x^2 - x'^2} \text{erfi}(x_+), \quad (19) \]

\[ x_+ = \frac{\tau \eta_{ba} (\delta \omega_a + \delta \omega_b)}{\sqrt{2} |\mu_b|}, \quad x_- = \frac{(\omega_a - \omega_a') \mu a}{\sqrt{2}}. \]

An analogous formula can be found for \( G_2^b(\omega_b, \omega_b') \) by simply switching \( a \leftrightarrow b \) in the above formula. We consider the scenario of a process seeded by a coherent state in mode \( a \), described by the state

\[ |f_a\rangle = \exp \left( i \int d\omega_a (f_a(\omega_a) a^\dagger(\omega_a) + \text{h.c.}) \right) |\text{vac}\rangle. \]

Using (4) we see that the lowest (second) order effect associated with time ordering is [11]

\[ |\psi_{\text{out}}\rangle = U_{\text{out}} U_{\text{in}} |f_a\rangle = U_{\text{in}} \times \exp \left( i \int d\omega_a (f_a(\omega_a) a^\dagger(\omega_a) + \delta f_a(\omega_a) a(\omega_a) + \text{h.c.}) |\text{vac}\rangle, \quad (20) \]

\[ \delta f_a(\omega_a) = -2\pi i \int d\omega_a' f_a(\omega_a') G_2(\omega_a, \omega_a'). \quad (21) \]

The last equation can be interpreted as follows: the first (\( \propto \varepsilon \)) of the interaction is that the non-linear medium acts as a two mode squeezer, generating pairs of photons in modes \( a \) and \( b \). The second order effect (\( \propto \varepsilon^2 \)), which appears solely due to time ordering, describes the “dressing” of the seed photons by the pump. That is, the frequency amplitudes of the photons of polarization \( a \) are no longer described by \( f_a(\omega_a) \) but rather by \( f_a(\omega_a) + \delta f_a(\omega_a) \). Finally, note that if instead the seed were prepared in a single photon state \( |\text{1}(f_a)\rangle = \int d\omega_a f_a(\omega_a) a^\dagger(\omega_a) |\text{vac}\rangle \), the effect of time ordering would be to dress it according to \( |\text{1}(f_a + \delta f_a)\rangle \).

To quantify the effects of time ordering we estimate the correction given by the second term of (20) relative
to the first term. If we take \(f_a(\omega_a) = \nu \exp(-\tau_a^2 \Delta \omega_a^2)\), in the limit \(\tau_a \ll \tau, |\eta_a|\),

\[
\int d\omega_a f_a(\omega_a) G_2(\omega_a, \omega_a') = \varepsilon^2 \nu \sqrt{2\pi} \exp(-q^2) \text{erfi}(q),
\]

\[q = \sqrt{2} \omega_a \eta_a \tau \mu_a / N^2, \quad N^4 = \mu^4 + 2\tau^2 \eta_ab. \tag{22}\]

Note that the maximum value the function \(\exp(-q^2) \text{erfi}(q)\) attains is \(\xi \approx 0.610503\) and that the function is identically zero if \(q = 0\) (which only occurs for the physical situation \(\eta_a = \eta_b\)). As before, we can compare the relative strengths of the amplitudes of the coherent state with and without including time ordering effects to obtain the following figure of merit:

\[
\rho = \frac{\max_{\omega_a} |\delta f_a(\omega_a)|}{\max_{\omega_a} f_a(\omega_a)} = 2\pi^3 \xi (\varepsilon^2 \tau^2 / N^2). \tag{23}\]

This quantity measures how much the pump is dressed by the time-ordering effects of the stimulated process. It has a very simple interpretation: To have important time ordering effects the seed photons, and the \(a\) and \(b\) photons created by the nonlinear process, must spend a significant amount of time traveling with the pump pulse. Note in particular that if the photons of type \(a\) move significantly faster than those of type \(b\), for example, then \(|\eta_{ab}|\) would make the bound (23) go to zero; if on the other hand the \(a\) photons travel much faster than those of the pump, then \(\eta_a \gg \tau\) and \(\eta_b \sim \tau \Rightarrow \mu^2 = \tau^2 + \eta_a \eta_b \gg \tau^2\) and then again \(\rho \to 0\).

**Time ordering effects in photon conversion** — For frequency conversion (FC), a strong pump pulse is used to mediate the conversion of photons of form, for example, a lower frequency to photons of a higher frequency. The Hamiltonian governing such processes for a one-dimensional geometry is

\[
\hat{H}_f(t) = -\hbar c \int d\omega_p d\omega_a d\omega_b e^{i \Delta t} \hat{\Phi}(\omega_a, \omega_b, \omega_p) \tag{24}
\]

\[
\Delta \omega_b = \omega_b - \omega_a - \omega_p, \quad \text{with} \quad \hat{\Phi}(\omega_a, \omega_b, \omega_p) = \exp \left( - (s_p \delta \omega_b - s_p \delta \omega_p - s_a \delta \omega_a)^2 \right), \tag{25}
\]

and the pump function as given in equation (3). Note that the Hamiltonian (24) can be obtained from equation (1) by changing \(a(\omega_a) \leftrightarrow a^\dagger(\omega_a)\) and switching the sign of all the scalars associated with \(a\. Using the Magnus expansion it can be shown that the time evolution operator \(\hat{U}\) for frequency conversion can be factorized in terms of a two mode frequency conversion term \(\hat{U}_{\text{fc}}\) (generated by terms that are extensions of \(\hat{a}^\dagger + \text{h.c.}\)) preceded by a pure single mode frequency conversion term \(\hat{U}_{\text{fc}}\) (generated by terms that are extensions of \(a^\dagger a + b^\dagger b\))

\[
\hat{U} = e^{-2\pi i \int d\omega_a d\omega_b \sum_a \hat{J}_a(\omega_a, \omega_b) \hat{a}(\omega_a) \hat{b}(\omega_b) + \text{h.c.}} \times e^{-2\pi i \sum_{c\neq a, b} \int d\omega_c d\omega_c' \sum_m \hat{G}_m(\omega_c, \omega_c') \hat{c}(\omega_c) \hat{c}(\omega_c')} \tag{26}
\]

For the Gaussian functions assumed here for the phase-matching and the pump pulse, we have \(\hat{J}_1(\delta \omega_a, \delta \omega_b) = J_1(\delta \omega_a, \delta \omega_b)\) and \(\hat{G}_2(\omega_c, \omega_c') = G_2(\omega_c, \omega_c')\). Were the sum in the first exponential in (26) truncated at the first term, and the second exponential ignored, we would obtain the sum of the Taylor series associated with the Hamiltonian (24). If the Hamiltonian commuted with itself at different times, this would be the exact solution of the problem. Assuming time ordering effects are irrelevant, the unitary \(\hat{U}_{\text{fc}}\) can be written, via the Schmidt decomposition, as a multimode beam-splitter that connects modes \(\hat{A}_n\) and \(\hat{B}_n\)

\[
\hat{U}_{\text{fc}} = e^{-i \sum_{n=0}^\infty j_n(\hat{A}_n \hat{B}_n^\dagger + \text{h.c.})} = \prod_{n=0}^\infty e^{-ig_j(\hat{A}_n \hat{B}_n^\dagger + \text{h.c.})},
\]

\[
\hat{A}_j = \int d\omega_a \frac{H_j(\mu_a \delta \omega_a / \sqrt{2S/2} e^{-\mu_a^2 \delta \omega_a^2 / S} \hat{a}(\omega_a),
\]

\[
\hat{B}_j = \int d\omega_b \frac{H_j(\mu_b \delta \omega_b / \sqrt{2S/2} e^{-\mu_b^2 \delta \omega_b^2 / S} \hat{b}(\omega_b),
\]

\[
g_j = \theta_0 \sqrt{1 + s^2 s^3}, \quad \theta_0 = 2\pi \varepsilon / \sqrt{2\mu_a \mu_b}. \tag{27}\]

where the \(H_j(x)\) are Hermite polynomials of degree \(j\), the quantity \(s\) is implicitly defined via \(S = (1 + s^2)/(1 - s^2)\), and \(0 \leq s \leq 1\); note that the operators \(A_j, B_j\) satisfy the usual bosonic commutation relations \([A_j, A_k^\dagger] = [B_j, B_k^\dagger] = \delta_{jk}\), with all other commutators vanishing. To convert a coherent state (or a single photon of type \(a\) and frequency centered at \(\omega_a\) to one of type \(b\) centered at frequency \(\omega_b\) one chooses an \(n\) – usually \(n = 0\) is the simplest choice, since it corresponds to a unimodal Gaussian profile – then prepares the state \([A_n] = e^{i \alpha(\hat{A}_n + \hat{A}_n^\dagger)} [\text{vac}] |\{1A_n\}_n = \hat{A}_n^\dagger [\text{vac}]\), and tunes the pump and nonlinear medium in such a way that \(g_n = \pi / 2\). This will guarantee that the output state is \([B_n] = e^{i \alpha(\hat{B}_n + \hat{B}_n^\dagger)} [\text{vac}] = \hat{U}_{\text{fc}}[g_n = \pi/2 |A_n\} |\{1B_n\}_n = \hat{B}_n^\dagger [\text{vac}] = \hat{U}_{\text{fc}}[g_n = \pi/2 |1A_n\}\). As it is clear from Eq. (26), time ordering effects will lead to two modifications of this scenario. The first is that the conversion amplitude \(\hat{J}(\omega_a, \omega_b)\) is no longer \(J_1(\omega_a, \omega_b)\), but involves corrections \(J_n(\omega_a, \omega_b)\). The second and more important is that the unitary connecting input and output is given by (26), which contains not only a two mode frequency conversion unitary but also a single frequency conversion term \(\hat{U}_{\text{fc}}\). To lowest order, this correction is encoded in the functions \(G_2(\omega_c, \omega_c') = \hat{G}_2(\omega_c, \omega_c')\) in (26). Because of this equality, the action of the second order Magnus correction on a coherent or single photon state is identical to (20), and we can obtain a bound on the time ordering effects which is identical to (23) and has the same interpretation: Time ordering effects are unimportant if the pump and frequency converted photons do not significantly copropagate.
FIG. 1. First and third order Magnus terms for the parameters from Table I. Note that the function $J_1$ as been normalized by $\varepsilon$ and $J_3, K_3$ by $\varepsilon^3 \tau$.

| Mode label | Central frequency | Central wavelength | Index of Refraction | Group velocity | Polarization |
|------------|-------------------|--------------------|---------------------|----------------|--------------|
| $p$        | 2.0000 PHz        | 0.9418 $\mu$m      | 2.1554              | $c/2.20054$    | $e$          |
| $a$        | 1.2707 PHz        | 1.4824 $\mu$m      | 2.2111              | $c/2.26276$    | $o$          |
| $b$        | 0.7293 PHz        | 2.5827 $\mu$m      | 2.10269             | $c/2.17833$    | $e$          |

TABLE I. Parameters for PPLN to observe significant time ordering effects. To obtain zeroth order phase-matching a periodicity of $\Lambda = 1/|n_e(\lambda_p)/\lambda_p - n_o(\lambda_a)/\lambda_a - n_e(\lambda_b)/\lambda_b| = 58.25 \mu$m is required. $n_{o/e}$ are the indices of refraction of the two different polarizations.

Experimental proposal — Although in many applications [6, 12] a simple perturbative treatment of the nonlinear processes described here is appropriate, there are at least two experiments [7, 13] in which this approximation breaks down and a non-perturbative treatment is needed. In both experiments a periodically poled Lithium Niobate (PPLN) crystal was used to upconvert a photon from the telecommunication wavelength to the visible with near unit efficiency. The fact that the single photon is converted with nearly unit efficiency implies that one of the $g_j$ in (27) is roughly equal to $\pi/2$; that is, the nonlinear medium acts as a frequency beam splitter with reflectivity approaching unity. This immediately implies that $\varepsilon \sim 1$. One could question whether or not in those experiments time ordering effects are important. It turns out that they are not, simply because non-degenerate Type-0 phase matching was used. Since there is only one polarization available and the group velocity curve as a function of frequency is a monotonic function, it is impossible to have pump and down converted photons travelling together for a significant amount of time, and $\tau^2/R^2 \ll 1$. Nevertheless, it is possible to make PPLN birefringent by adding small amounts of Magnesium Oxide (MgO). In the study of Gayer et al. [4], Sellmeier equations were determined for the ordinary and extraordinary polarizations of PPLN doped with MgO in the wavelength window 0.5 to 4 nm. Although in the experiments mentioned so far a PPLN crystal was used for FC, they could also be used for SPDC. In Table I we show parameters for which it is possible to have pump and down converted photons travelling together. Using the parameters described in Table I it would be possible to set up an experiment in which, for the first time, a JSA that is nonlinear in the pump electric field could be observed. Using the parameters from Table I, with $\chi^2 = 10 \text{ pm/V}$, a crystal length of $L = 4 \text{ cm}$, a pulse duration $\tau = 1 \text{ ps}$, and a peak electric field of 6 MV/m, we obtain $\varepsilon = 0.30$ and $\tau \leq 2\pi^2 \varepsilon^2 \tau^2 / R^2 = 0.46$.

As it is seen in Fig. 1 time ordering effects (encoded in $J_3$ and $K_3$) are comparable to the first order Magnus term $J_1$. These modifications could open an avenue for the generation of squeezed states with very interesting JSAs, and in any case these effects will become important as soon as very squeezed states are generated.

Conclusion — In this letter we have presented a theory that allows us to understand the effects of time ordering in nonlinear quantum optics processes. In the last three sections we investigated the effects of time ordering for spontaneous and stimulated photon generation, and frequency conversion. In each case a figure of merit quantifying the effects of time ordering was introduced, and it was shown that these effects are only sizable whenever the photons in the down-converted/frequency converted modes copropagate with the pump photons. This agrees with the simple physical intuition that the only way the effect of the Hamiltonian at some posterior time $\hat{H}(t_f)$ can be influenced by the former version of itself $\hat{H}(t_i)$, under the assumption of an undepleted pump, is if the fields associated with the pump photons, any seed photons, and the generated photons spend a significant amount of time in the same regions of space. Under these conditions the time ordering corrections will also have the
most significant qualitative effect, since they will modify
the Schmidt number of the quantum correlated photons
from the uncorrected value of close to unity.

We have used Gaussian functions to approximately de-
scribe both the pump pulse and the phase-matching func-
tions that enter the description of the photon generation
or conversion. This led to the possibility of constructing
analytic expressions for the figures of merit. Nonethe-
less, the general approach of the Magnus expansion de-
scribed earlier [11] could be applied to the use of arbitrary
functions to model the pump pulse and phase matching.
A general result of our investigations here, that strong
pump pulses are required for any deviation from the first
order Magnus term to be relevant, can be expected to
hold more generally. There is thus a wide range of
problems where an elementary first order perturbative
approach would fail, but a calculation using the first or-
der order Magnus term would be sufficient. Yet working at
pump intensities high enough that time ordering correc-
tions appear should lead to new strategies for producing
novel quantum states.

Supplementary Material for “Time ordering effects in the generation
of entangled photons using nonlinear
optical processes”

In this Supplementary Material we calculate the third order Magnus term \( \Omega_3 \) [11] and associated JSA contribution \( J_3 \)
for the Hamiltonian considered in the main text. Note that the second order correction \( \Omega_2 \) (and associated functions \( G_a^2, G_b^2 \)) and the imaginary part of the third order correction \( K_3 \) can be calculated directly from the results in [11].
For the calculation of \( J_3 \) we will not use the results presented in [11]. The reason is that integrals of the form \( \int \frac{dx}{x} \int \frac{dy}{y} f(x, y) \), which are the ones necessary to calculate the third order correction, are hard to bound and cannot be computed analytically for the situation when \( f \) is a Gaussian function. To circumvent this difficulty we assume that the Hamiltonian can be written as a function of the frequencies of the down-converted photons and time \( (\omega_a, \omega_b, t) \) but without any explicit dependence of the pump frequency \( (\omega_p) \) which will be assumed to have been integrated out. This can be done analytically for Gaussian pump profiles and phase-matching functions, as we shall show shortly. Because the time dependence of the Hamiltonian will no longer be harmonic but Gaussian, the time order integrals will
not involve any principal value integrals but rather incomplete \( (i.e. \text{not extending over the whole real line}) \) Gaussian
integrals.

We remind the reader that the Hamiltonian considered in the main text is given by Eq. (1) and that the phase
matching and pump profile functions are defined by Eq. (2) and (3) respectively. We also note that the factor \( \Delta \)
appearing in Eq. (1) can be written as

\[
\Delta = \omega_a + \omega_b - \omega_p = \delta \omega_a + \delta \omega_b - \delta \omega_p.
\]

Given the simplicity of the pump and phase matching functions we can perform analytically the integral over \( \omega_p \)

\[
F(\omega_a, \omega_b, t) = (-\varepsilon) \int d\omega_p e^{i\Delta t} \alpha(\omega_p) \Phi(\omega_a, \omega_b, \omega_p) =
\]

\[
(-\varepsilon) \tau \sqrt{\frac{1}{s_a^2 + \tau^2}} \exp \left( \frac{(-2s_a \delta \omega_a + s_b \delta \omega_b) + it}{4 (s_a^2 + \tau^2)} \right) + it (\delta \omega_a + \delta \omega_b) - (s_a \delta \omega_a + s_b \delta \omega_b)^2 \right). \tag{29}
\]

After the integration the Hamiltonian is now

\[
H_I(t) = \hbar \int d\omega_a d\omega_b F(\omega_a, \omega_b, t) a^\dagger(\omega_a) b^\dagger(\omega_b) + \text{h.c.} \tag{30}
\]
For the third order Magnus term we get

\[
\Omega_3 = \left(\frac{-i}{6\hbar^3}\right) \int dt \int dt' \int dt'' \{[H_I(t), [H_I(t'), H_I(t'')]] + [[H_I(t), H_I(t')], H_I(t'')]\}
\]

\[= \frac{i}{6} \int dt \int dt' \int dt'' \int d\omega_d d\omega_e d\omega_{\omega_c} d\omega_{\omega_c'} d\omega_{\omega_c''} d\omega_{\omega_c'} d\omega_{\omega_c''} d\omega_c
\]

\[\times \left( \tilde{\delta} (\omega''_e - \omega_e) \delta (\omega''_c - \omega_c) F (\omega_e, \omega_e, t) F (\omega'_e, \omega'_e, t') F (\omega''_e, \omega''_e, t'')^* a^\dagger (\omega_e) b (\omega_e) - 2\tilde{\delta} (\omega''_e - \omega_e) \delta (\omega''_o - \omega_o) F (\omega_e, \omega_e, t) F (\omega''_o, \omega''_o, t'') F (\omega'_o, \omega'_o, t')^* a^\dagger (\omega_o) b (\omega_e) + \delta (\omega''_o - \omega_o) \delta (\omega''_o - \omega_o) F (\omega_e, \omega_e, t) F (\omega''_o, \omega''_o, t'') F (\omega'_o, \omega'_o, t')^* a^\dagger (\omega_o) b (\omega_o) - 2\tilde{\delta} (\omega'_c - \omega'_c) \delta (\omega''_o - \omega_o) F (\omega_e, \omega_e, t) F (\omega''_o, \omega''_o, t'') F (\omega'_o, \omega'_o, t')^* a^\dagger (\omega_o) b (\omega_o) + \delta (\omega'_e - \omega'_e) \delta (\omega''_o - \omega_o) F (\omega_e, \omega_e, t) F (\omega''_o, \omega''_o, t'') F (\omega'_o, \omega'_o, t')^* a^\dagger (\omega_o) b (\omega_o) - \text{h.c.}\right)
\]

In the last equation we use \(\tilde{\delta}\) for the Dirac distribution. To simplify this expression we perform the following changes of variables in each of the six terms in the last equation

\[
\begin{align*}
\omega_e &\rightarrow \omega_d, \omega''_e \rightarrow \omega_d, \omega'_e \rightarrow \omega_c, \omega''_c \rightarrow \omega_c, \omega'_c \rightarrow \omega_b, \omega_o \rightarrow \omega_a, \\
\omega_e &\rightarrow \omega_d, \omega'_e \rightarrow \omega_c, \omega''_e \rightarrow \omega_e, \omega'_e \rightarrow \omega_b, \omega_o \rightarrow \omega_a, \\
\omega'_e &\rightarrow \omega_d, \omega''_e \rightarrow \omega_d, \omega'_e \rightarrow \omega_c, \omega''_e \rightarrow \omega_b, \omega_o \rightarrow \omega_a, \\
\omega'_e &\rightarrow \omega_d, \omega''_e \rightarrow \omega_d, \omega'_e \rightarrow \omega_c, \omega''_e \rightarrow \omega_b, \omega_o \rightarrow \omega_a, \\
\omega'_e &\rightarrow \omega_d, \omega''_e \rightarrow \omega_d, \omega'_e \rightarrow \omega_c, \omega''_e \rightarrow \omega_b, \omega_o \rightarrow \omega_a, \\
\omega'_e &\rightarrow \omega_d, \omega''_e \rightarrow \omega_d, \omega'_e \rightarrow \omega_c, \omega''_e \rightarrow \omega_b, \omega_o \rightarrow \omega_a,
\end{align*}
\]

(32)

to get

\[
\Omega_3 = \frac{i}{6} \int dt \int dt' \int dt'' \int d\omega_d d\omega_e d\omega_{\omega_c} d\omega_{\omega_c'} d\omega_{\omega_c''} d\omega_{\omega_c'} d\omega_{\omega_c''} d\omega_c
\]

\[\times \left( F (\omega_a, \omega_d, t) F (\omega_c, \omega_b, t') F (\omega_c, \omega_d, t'')^* - 2F (\omega_a, \omega_d, t) F (\omega_c, \omega_b, t'') F (\omega_c, \omega_d, t')^* + F (\omega_a, \omega_d, t) F (\omega_c, \omega_d, t') F (\omega_c, \omega_d, t'')^* + F (\omega_a, \omega_d, t) F (\omega_c, \omega_d, t')^* + F (\omega_a, \omega_d, t) F (\omega_c, \omega_d, t'')^* - 2F (\omega_a, \omega_d, t') F (\omega_c, \omega_b, t) F (\omega_c, \omega_d, t')^* + F (\omega_a, \omega_d, t') F (\omega_c, \omega_b, t')^* + F (\omega_a, \omega_d, t') F (\omega_c, \omega_d, t')^* - \text{h.c.}\right)
\]

We can now perform the change of variables \(t = q + 2r + s, t' = q - r + s, t'' = q - r - 2s\) that transforms the integral according to \(\int dt \int dt' \int dt'' = 9 \int dq \int dq' \int dq'' \int ds\), and also use the fact that \(F (\omega_a, \omega_b, -t) = F (\omega_a, \omega_b, t)^*\), where \(x^*\) denotes the complex conjugate of \(x\), to obtain

\[
\Omega_3 = \frac{3i}{2} \int dq \int dq' \int dq'' \int ds \int d\omega_d d\omega_e d\omega_{\omega_c} d\omega_{\omega_c'} d\omega_{\omega_c''} d\omega_{\omega_c'} d\omega_{\omega_c''} d\omega_c
\]

\[\times \left( F (\omega_a, \omega_d, q + 2r + s) F (\omega_c, \omega_b, q - r + s) F (\omega_c, \omega_d, -q + 2r + s) - 2F (\omega_a, \omega_d, q + 2r + s) F (\omega_c, \omega_b, q - r - 2s) F (\omega_c, \omega_d, -q + r + s) + F (\omega_a, \omega_d, q - r + s) F (\omega_c, \omega_b, q + 2r + s) F (\omega_c, \omega_d, -q + r - 2s) + F (\omega_a, \omega_d, q - r + s) F (\omega_c, \omega_b, q + 2r + s) F (\omega_c, \omega_d, -q - 2r - s) - 2F (\omega_a, \omega_d, q - r - 2s) F (\omega_c, \omega_b, q + 2r + s) F (\omega_c, \omega_d, -q + r - s) + F (\omega_a, \omega_d, q - r - 2s) F (\omega_c, \omega_b, q + 2r + s) F (\omega_c, \omega_d, -q - 2r - s) - \text{h.c.}\right)
\]

(34)

Note the following: First, \(r\) and \(s\) can always be swapped since they have the same integration ranges and second the change \(q \rightarrow -q\) can always be performed since \(\int_{-\infty}^{\infty} dq f(q) = \int_{-\infty}^{\infty} dq f(-q)\). Because of this it is easily seen that in the last equation the sixth term is the complex conjugate of the first, the fifth is the complex conjugate of the second
and the fourth is the complex conjugate of the third. So we can more compactly write
\[ \Omega_3 = -2\pi i \int d\omega_d d\omega_b a^\dagger(\omega_a)b^\dagger(\omega_b)J_3(\omega_a, \omega_b) - \text{h.c.} \] (35)
\[ = -2\pi i \int d\omega_d d\omega_b a^\dagger(\omega_a)b^\dagger(\omega_b) \left( -\frac{3}{4\pi} \int dq dq^\prime dq^\prime d\omega_d(L_1 - 2L_2 + L_3 + \text{c.c.}) \right) - \text{h.c.} \] (36)
\[ L_1 = \int_0^\infty dr \int_0^\infty ds F(\omega_a, \omega_d, q + 2r + s) F(\omega_c, \omega_b, q - r + s) F(\omega_c, \omega_d, -q + r + 2s) \] (37)
\[ L_2 = \int_0^\infty dr \int_0^\infty ds F(\omega_a, \omega_d, q + 2r + s) F(\omega_c, \omega_b, q - r - s) F(\omega_c, \omega_d, -q + r - s) \] (38)
\[ L_3 = \int_0^\infty dr \int_0^\infty ds F(\omega_a, \omega_d, q - r + s) F(\omega_c, \omega_b, q + 2r + s) F(\omega_c, \omega_d, -q + r + 2s). \] (39)

To get a simpler expression we can perform the following changes of variables: For \( L_1 \) in the last equation we put \( r \to r' + s', s \to -s' \) to obtain:
\[ L_1 = \int_{-\infty}^{0} ds' \int_{-\infty}^{\infty} dr' F(\omega_a, \omega_d, q + 2r' + s') F(\omega_c, \omega_b, q - r' - 2s') F(\omega_c, \omega_d, -q + r' - s'). \] (40)

Likewise for \( L_3 \) we change \( r \to -r' - s' \) and \( s \to r' \) to obtain:
\[ L_3 = \int_0^\infty dr' \int_{-\infty}^{-r'} ds' F(\omega_a, \omega_d, q + 2r' + s') F(\omega_c, \omega_b, q - r' - 2s') F(\omega_c, \omega_d, -q + r' - s'). \] (41)

Note that now the integrands of \( L_1, L_2 \) and \( L_3 \) are identical and that \( \int_{-\infty}^{0} ds' \int_{-\infty}^{\infty} dr' + \int_0^\infty dr' \int_{-\infty}^{-r'} ds' = \int_0^\infty dr' \int_{-\infty}^{0} ds' \) so we can write more simply
\[ L_1 - 2L_2 + L_3 = \left( \int_0^\infty dr \int_{-\infty}^{0} ds - 2 \int_0^\infty dr \int_{-\infty}^{\infty} ds \right) \times F(\omega_a, \omega_d, q + 2r + s) F(\omega_c, \omega_b, q - r - 2s) F(\omega_c, \omega_d, -q + r + s), \] (42)
we can now write \( J_3 \) as
\[ J_3(\omega_a, \omega_b) = \frac{3}{4\pi} \int dq dq^\prime dq^\prime d\omega_d \left( \int_0^\infty dr' \int_{-\infty}^{0} ds - 2 \int_0^\infty dr' \int_{-\infty}^{\infty} ds \right) \times F(\omega_a, \omega_d, q + 2r + s) F(\omega_c, \omega_b, q - r - 2s) F(\omega_c, \omega_d, -q + r + s). \] (43)

Up to this point we have not used the fact that \( F \) is Gaussian; now we will use it: The product of three \( F \)'s is a Gaussian function in \( q, r, s, \omega_a, \omega_b, \omega_c, \omega_d \) and hence the integrals over \( q, \omega_c, \omega_d \) are complete (i.e. extending over the whole real line) Gaussian integrals and thus can be done analytically, leaving us with a Gaussian function in \( r, s, \omega_a, \omega_b \). Doing these integrals we obtain
\[ \int dq dq^\prime dq^\prime d\omega_d F(\omega_a, \omega_d, q + 2r + s) F(\omega_c, \omega_b, q - r - 2s) F(\omega_c, \omega_d, -q + r + s) = -\frac{2\pi^{3/2} \tau^3}{3\sigma_a \sigma_b} e^{-\frac{uN}{3\gamma} (f(r, s) + \text{c.c.})}, \] (44)
with
\[ f(r, s) = \exp(x^T A x + iv^T x), \quad A = -\frac{3}{4\tau^2} \begin{pmatrix} \frac{\mu^2}{\sigma_a^2} & \frac{\mu^2}{\sigma_b^2} & \frac{\mu^2}{\sigma_c^2} \\ \frac{\mu^2}{\sigma_a^2} & \frac{\mu^2}{\sigma_b^2} & \frac{\mu^2}{\sigma_c^2} \\ \frac{\mu^2}{\sigma_a^2} & \frac{\mu^2}{\sigma_b^2} & \frac{\mu^2}{\sigma_c^2} \end{pmatrix}, \quad v^T = 2\eta \begin{pmatrix} \delta \omega_a \\ \delta \omega_b \\ \delta \omega_c \end{pmatrix}. \] (45)
and \( \mathbf{N} \) and \( \mathbf{u} \) being defined in Eq. (6) of the main text. With this result we can write
\[ J_3(\omega_a, \omega_b) = \sqrt{\frac{\pi \tau^3}{2\sigma_a \sigma_b}} \exp\left(-\frac{uN}{3\gamma} T/3\right) I \] (45)
\[ I = \left( \int_0^\infty dr' \int_{-\infty}^{0} ds - 2 \int_0^\infty dr' \int_{-\infty}^{\infty} ds \right) f(r, s) + \text{c.c.} \] (46)
Now we will add and subtract the quantity \( \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \text{c.c.} \) to \( I \) to get

\[
I = \left( \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \text{c.c.} \right) - \left( 3 \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \text{c.c.} \right) = I_1 - I_2. \tag{47}
\]

Notice that the term in parenthesis, \( I_1 \), can be evaluated since:

\[
I_1 = \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \text{c.c.} = \int_0^\infty dr \int_0^\infty ds \: f(r, s) \tag{48}
\]

\[
= \frac{4\pi s_0 s_b \tau^2}{3R^2} \exp \left( -\frac{4\pi^2 s_0^2}{3R^4} uWu^T \right), \quad W = \begin{pmatrix} 2\mu_a^2 - \mu^2 & -\mu^2 \\ -\mu^2 & 2\mu_b^2 \end{pmatrix}. \tag{49}
\]

As for \( I_2 \) we can proceed as follows,

\[
I_2 = 3 \int_0^\infty dr \int_0^\infty ds \: f(r, s) + \text{c.c.} = 6 \int_0^\infty dr \int_0^\infty ds \: \exp \left( x^T Ax \right) \cos \left( \sqrt{3} x \right). \tag{50}
\]

But now we can let \( s \to 2s_0 \tau q/\sqrt{3} \) and \( r \to s_0 \tau p/\sqrt{3} \) to get:

\[
I_2 = 8s_0 s_\tau^2 \int_0^\infty dq \int_0^\infty dp \: \exp \left( -(p, q) M(p, q)^T \right) \cos \left( 4\pi \eta_{ab} (\delta \omega_a q + \delta \omega_b p)/\sqrt{3} \right). \tag{51}
\]

Now we can put all these results together to obtain

\[
J_3 = \frac{\sqrt{\pi} \varepsilon^3}{2s_0 s_b} \exp \left( -\frac{uNu}{3} \right) \left\{ \frac{4\pi s_0 s_b \tau^2}{3R^2} \exp \left( -\frac{4\pi^2 s_0^2}{3R^4} uWu^T \right) -8s_0 s_b \tau^2 \int_0^\infty dq \int_0^\infty dp \: \exp \left( -(p, q) M(p, q)^T \right) \cos \left( 4\pi \eta_{ab} (\delta \omega_a q + \delta \omega_b p)/\sqrt{3} \right) \right\}. \tag{52}
\]

Simplifying and noting that \( N/3 + W = Q/R^4 \) we obtain

\[
J_3 = \frac{2\pi^3/\tau^3 \varepsilon^3}{3R^2} \exp \left( -\frac{uQu^T}{R^4} \right)
-4\sqrt{\pi} \tau^3 \varepsilon^3 \exp \left( -\frac{uNu}{3} / 3 \right) \int_0^\infty dq \int_0^\infty dp \: \exp \left( -(p, q) M(p, q)^T \right) \cos \left( 4\pi \eta_{ab} (\delta \omega_a q + \delta \omega_b p)/\sqrt{3} \right). \tag{53}
\]

This last equation can be rewritten as Eqs. (8-11) using the definitions of Eq. (12) in the main text. We now obtain a bound for \( J_3 \). First note that \( |J_3| < |W| + |VZ| = W + |V|Z \); the only term that is hard to bound from the last inequality is \( |V| \), and to this end we note the following chain of inequalities:

\[
\exp \left( -(p, q) M(p, q)^T \right) \cos \left( 4\pi \eta_{ab} (\delta \omega_a q + \delta \omega_b p)/\sqrt{3} \right) \leq \exp \left( -(p, q) M(p, q)^T \right) \tag{54}
\]

\[
\int_0^\infty dp \int_0^\infty dq \: \exp \left( -(p, q) M(p, q)^T \right) \cos \left( 4\pi \eta_{ab} (\delta \omega_a q + \delta \omega_b p)/\sqrt{3} \right) \leq \int_0^\infty dp \int_0^\infty dq \: \exp \left( -(p, q) M(p, q)^T \right). \tag{55}
\]

Now note that for any function \( g(p, q) \), \( \int_0^\infty dp \int_0^\infty dq \: g(p, q) \leq \int_0^\infty dp \int_0^\infty dq \: |g(p, q)| \) and thus we can finally write

\[
|V(\omega_a, \omega_b)| = \int_0^\infty dp \int_0^\infty dq \: \exp \left( -(p, q) M(p, q)^T \right) \cos \left( 4\pi \eta_{ab} (\delta \omega_a q + \delta \omega_b p)/\sqrt{3} \right) \leq \int_0^\infty dp \int_0^\infty dq \: \exp \left( -(p, q) M(p, q)^T \right) = V(\tilde{\omega}_a, \tilde{\omega}_b). \tag{56}
\]

The right hand side of the last inequality can be easily evaluated

\[
V(\tilde{\omega}_a, \tilde{\omega}_b) = \begin{cases} \frac{\pi + \tan^{-1}(R^2/\mu^2)}{\tan^{-1}(R^2/\mu^2)}, & \text{if } \mu^2 < 0 \\ \frac{\tan^{-1}(R^2/\mu^2)}{2\mu^2}, & \text{if } \mu^2 > 0. \end{cases} \tag{56}
\]

Note from Eq. (12) of the main text that \( R^4 \geq 3\mu^4 \) this immediately implies that \( V(\tilde{\omega}_a, \tilde{\omega}_b) < \pi/(3R^2) \) and the general bound

\[
|J_3(\tilde{\omega}_a, \tilde{\omega}_b)| < \frac{2\pi^3/\tau^3 \varepsilon^3}{3R^2} \left( \exp \left( -\frac{uQu^T}{R^4} \right) + 2 \exp \left( -\frac{uNu}{3} \right) \right) \leq \frac{2\pi^3/\tau^3 \varepsilon^3}{R^2}, \tag{57}
\]

\[
\text{for } \mu^2 < 0.
\]
which is Eq. (15) The last bound is obtained by taking \( u = 0 \). Note that \( \max_{\eta_a, \eta_b} \frac{\tau^2}{R^2} = 1/\sqrt{3} \) is achieved for \( \eta_a = \eta_b = 0 \). Nevertheless note that for this parameter values (\( \eta_{ab} = \eta_a - \eta_b = 0 \)) \( J_3 \) is identically zero. To show this note that if \( \eta_{ab} = 0 \) then \( R^4 = 3\mu^4, \ M^4 = \mu^4, \ \mu^2 > 0, \ Q/R^4 = N/3 \) and

\[
V(\omega_a, \omega_b) = V(\bar{\omega}_a, \bar{\omega}_b) = \frac{\pi}{6R^2}.
\]

(58)