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Time Ordering in Spontaneous Parametric Down-conversion

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Abstract. We consider the multi-mode parametric down-conversion Hamiltonian that governs the evolution of the fields inside a nonlinear crystal. If the Hamiltonian does not commute with itself at all times—as is the case for the multimode down-conversion Hamiltonian—then the expansion of the evolution operator must take the form of the time-ordered Dyson series, as opposed to the simplified Taylor series. By expanding the evolution operator to third order, the conditions under which the Taylor series is a valid approximation are revealed. In addition, some new and interesting behaviour is predicted.

Spontaneous parametric down-conversion (SPDC) is a nonlinear optical process in which a photon from a pump laser, incident on a nonlinear birefringent crystal, converts into two single photons under conservation of energy and momentum. Photon sources based on this phenomenon are an ubiquitous tool for quantum computation [1], quantum communication [2] and quantum metrology [3, 4]. They are also becoming increasingly important in more specialised applications such as quantum imaging [5], quantum lithography [6] or optical coherence tomography [7].

To study the spectral properties of the down-conversion process, one needs to examine the evolution within the crystal. The time dependent Hamiltonian, which governs this evolution, does not commute with itself at all times. This implies that the expansion of the evolution operator, and therefore the output state, should take the form of the time-ordered *Dyson* series.

The spectral properties of the down-converted state have been extensively analysed to first order in the output state, see e.g. [8]. To first order, a simple Taylor expansion gives the same results as the Dyson series—when only one pair is created, time ordering is not relevant. In this paper, however, we consider the output state to higher orders, truncating the output state beyond the 6-photon term.

While the Dyson series gives the correct expansion, it would be considerably easier for future calculations to use the Taylor series. We therefore aim to identify if there are any conditions under which the Taylor series is a good approximation.

The down-converted state is given by

$$|\psi\rangle = \mathscr{T}e^{-\frac{i}{\hbar}\int_{-\infty}^{\infty}dt'\hat{H}(t')}|\psi(t_0)\rangle\,, \tag{1}$$

where $\mathscr{T}$ is the time-ordering operator and the Hamiltonian is [8]

$$\hat{H}(t) = AL \iiint d\omega_i d\omega_s d\omega_p f(\omega_i, \omega_s, \omega_p) e^{i\Delta\omega_1 t} \hat{a}_i^\dagger(\omega_i) \hat{a}_s^\dagger(\omega_s) + \text{H.c.}, \qquad (2)$$

where the integration is over the positive or negative frequency parts of the fields and

$$f(\omega_i, \omega_s, \omega_p) = \alpha(\omega_p)\Phi(\Delta k(\omega_i, \omega_s, \omega_p)), \qquad (3)$$

is the joint spectral amplitude where $\alpha(\omega_p)$ is the pump function and $\Phi(\Delta k(\omega_i, \omega_s, \omega_p)) = \text{sinc}(\Delta k(\omega_i, \omega_s, \omega_p)L/2)$ is the phase-matching function. $\Delta k(\omega_i, \omega_s, \omega_p) = k_i(\omega_i) + k_s(\omega_s) - k_p(\omega_p)$ is the phase mismatch and $\Delta\omega = \omega_i + \omega_s - \omega_p$.

The Dyson series expansions give the down-converted state to third order

$$|\Psi\rangle \approx \frac{1}{\sqrt{\mathscr{N}}} \left(|\Psi^{(0)}\rangle + |\Psi^{(1)}\rangle + |\Psi^{(2)}\rangle + |\Psi^{(3)}\rangle \right), \qquad (4)$$

where

$$|\Psi^{(0)}\rangle = |0\rangle, \qquad (5)$$

$$|\Psi^{(1)}\rangle = \frac{1}{i\hbar} \int_{-\infty}^{\infty} dt_1 \hat{H}(t_1)|0\rangle, \qquad (6)$$

$$|\Psi^{(2)}\rangle = \left(\frac{1}{i\hbar}\right)^2 \int_{-\infty}^{\infty} dt_1 \hat{H}(t_1) \int_{-\infty}^{t_1} dt_2 \hat{H}(t_2)|0\rangle, \qquad (7)$$

$$|\Psi^{(3)}\rangle = \left(\frac{1}{i\hbar}\right)^3 \int_{-\infty}^{\infty} dt_1 \hat{H}(t_1) \int_{-\infty}^{t_1} dt_2 \hat{H}(t_2) \int_{-\infty}^{t_2} dt_3 \hat{H}(t_3)|0\rangle. \qquad (8)$$

$|\Psi^{(n)}\rangle$ represents the nth order expansion of $|\Psi\rangle$. $\mathscr{N}$ is a normalisation constant which ensures that the probabilities sum to unity.

Substituting the Hamiltonian in Equation (2) into Equation (6), the first order term becomes

$$|\Psi^{(1)}\rangle = \mathscr{A} \iint d\omega_i d\omega_s f(\omega_i, \omega_s, \omega_i + \omega_s) \hat{a}_i^\dagger(\omega_i) \hat{a}_s^\dagger(\omega_s)|0\rangle, \qquad (9)$$

where the time integral was evaluated using the Fourier transform of a constant, $\int_{-\infty}^{\infty} e^{ixt} dt = 2\pi\delta(x)$. $|\Psi^{(1)}\rangle$ is a state consisting of two photons of frequencies ω_i and ω_s, whose joint spectral profile is given by $f(\omega_i, \omega_s, \omega_i + \omega_s)$, defined in Equation (3). This is identical to the solution for the Taylor series. Similarly for the second order term, we substitute the Hamiltonian in Equation (2) into Equation (7) to give

$$|\Psi^{(2)}\rangle = \frac{\mathscr{A}^2}{2} \left(G_0 + \iiiint d\omega_i d\omega_s d\omega_i' d\omega_s' G_2(\omega_i, \omega_s, \omega_i', \omega_s') \right.$$
$$\left. \times \hat{a}_i^\dagger(\omega_i) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i') \hat{a}_s^\dagger(\omega_s') \right) |0\rangle. \qquad (10)$$

To evaluate the integrals over time, we use the Fourier transform of a Heaviside step function $\int_{-\infty}^{t_1} e^{ixt} dt = \int_{-\infty}^{\infty} \Theta(t_1 - t) e^{ixt} dt = \pi\delta(x) - ie^{-it_1 x}/x$, to give $G_2 = F_2 - ig_2$.

$F_2 = f(\omega_i, \omega_s, \omega_i + \omega_s) f(\omega_i', \omega_s', \omega_i' + \omega_s')$ corresponds to the total two-photon amplitude, had the Taylor series been used, while $g_2 = \frac{1}{\pi} \int \frac{d\omega_p}{\Delta\omega} f(\omega_i, \omega_s, \omega_p) f(\omega_i', \omega_s', \omega_i' + \omega_s' + \Delta\omega)$ can be interpreted as the amplitude for the creation of a four-photon entangled state, where $\Delta\omega_2 = \omega_i' + \omega_s' - \omega_p'$. The frequencies of these photons are constrained by a combined energy conservation condition $\omega_i + \omega_s + \omega_i' + \omega_s' = \omega_p + \omega_p'$. However, we find that due to destructive interference inside the crystal, this term goes to zero. Therefore, the amplitude for the four-photon state simplifies to $G_2(\omega_i, \omega_s, \omega_i', \omega_s') = F_2(\omega_i, \omega_s, \omega_i', \omega_s')$. The amplitude for the correction to the vacuum state also simplifies to $G_0 = \iint d\omega_i d\omega_s |f(\omega_i, \omega_s, \omega_i + \omega_s)|^2$. *The Dyson and Taylor series give the same results to second order.* For the third order term, we follow a similar process, substituting the Hamiltonian in Equation (2) into Equation (8) to give

$$|\Psi^{(3)}\rangle = \frac{\mathscr{A}^3}{3!}\left(\iint d\omega_i d\omega_s G_1(\omega_i, \omega_s)\hat{a}_i^\dagger(\omega_i)\hat{a}_s^\dagger(\omega_s) + \iiiiii d\omega_i d\omega_s d\omega_i' d\omega_s' d\omega_i'' d\omega_s'' \right.$$
$$\left. \times G_3(\omega_i, \omega_s, \omega_i', \omega_s', \omega_i'', \omega_s'')\hat{a}_i^\dagger(\omega_i)\hat{a}_s^\dagger(\omega_s)\hat{a}_i^\dagger(\omega_i')\hat{a}_s^\dagger(\omega_s')\hat{a}_i^\dagger(\omega_i'')\hat{a}_s^\dagger(\omega_s'') \right)|0\rangle.$$

(11)

The first term corresponds to the creation of two photons while the second term corresponds to the creation of six photons. The six-photon amplitude is $G_3 = \frac{3}{2}(F_3 + ig_{3a} + ig_{3b} + h_3)$ where $F_3(\omega_i, \omega_s, \omega_i', \omega_s', \omega_i'', \omega_s'') = f(\omega_i, \omega_s, \omega_i + \omega_s)f(\omega_i', \omega_s', \omega_i' + \omega_s')f(\omega_i'', \omega_s'', \omega_i'' + \omega_s'')$ corresponds to the total six-photon amplitude, had the Taylor series been used, while $g_{3a} = -\frac{1}{\pi}f(\omega_i'', \omega_s'', \omega_i'' + \omega_s'') \int \frac{d\omega_p''}{\Delta\omega_2} f(\omega_i, \omega_s, \omega_i + \omega_s + \Delta\omega_2)f(\omega_i', \omega_s', \omega_p')$ and $g_{3b} = \frac{1}{\pi}f(\omega_i, \omega_s, \omega_i + \omega_s) \int \frac{d\omega_p'}{\Delta\omega_2} f(\omega_i', \omega_s', \omega_p')f(\omega_i'', \omega_s'', \omega_i'' + \omega_s'' + \Delta\omega_2)$ can be interpreted as the amplitude for the creation of a four-photon entangled state along with an independent photon-pair. The frequencies of these photons are constrained by a combined energy conservation condition $\omega_i + \omega_s + \omega_i' + \omega_s' = \omega_p + \omega_p'$ as well as an independent condition $\omega_i'' + \omega_s'' = \omega_p''$ [1]. The last term in G_3 is $h_3 = \frac{1}{\pi^2} \int \frac{d\omega_p d\omega_p''}{\Delta\omega_1 \Delta\omega_3} f(\omega_i, \omega_s, \omega_p)f(\omega_i'', \omega_s'', \omega_p'')f(\omega_i', \omega_s', \omega_i' + \omega_s' + \Delta\omega_1 + \Delta\omega_3)$ and can be interpreted as the amplitude for the creation of a six-photon entangled state. The frequencies of these photons are constrained by a combined energy conservation condition $\omega_i + \omega_s + \omega_i' + \omega_s' + \omega_i'' + \omega_s'' = \omega_p + \omega_p' + \omega_p''$.

We find that due to destructive interference inside the crystal, g_{3a} and g_{3b} cancel each other. Therefore, the amplitude for the six-photon state simplifies to $G_3 = \frac{3}{2}\big(F_3(\omega_i, \omega_s, \omega_i', \omega_s', \omega_i'', \omega_s'') + h_3(\omega_i, \omega_s, \omega_i', \omega_s', \omega_i'', \omega_s'')\big)$. The amplitude for the correction to the two-photon state also simplifies to $G_1(\omega_i, \omega_s) = \frac{3}{2}\big(F_1(\omega_i, \omega_s) - h_1(\omega_i, \omega_s)\big)$ where $F_1 = \iint d\omega_i' d\omega_s' f^*(\omega_i', \omega_s', \omega_i' + \omega_s')\big(f(\omega_i, \omega_s, \omega_i + \omega_s)f(\omega_i', \omega_s', \omega_i' + \omega_s') + \sum_{\substack{[j,k]= \\ \mathscr{P}[\omega_i, \omega_i']}} \sum_{\substack{[m,n]= \\ \mathscr{P}[\omega_s, \omega_s']}} f(j, m, j+m)f(k, n, k+n)\big)$ corresponds to the two-photon correc-

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[1] This is the case for the state described by g_{3a}. For the state described by g_{3b}, the energy conservation conditions are $\omega_i + \omega_s + \omega_i'' + \omega_s'' = \omega_p + \omega_p''$ and $\omega_i' + \omega_s' = \omega_p'$.

tion[2] , had the Taylor series been used, while

$$
\begin{aligned}
h_1 = {} & \frac{1}{\pi^2} \iint d\omega_i' d\omega_s' \iint d\omega_p d\omega_p'' \big(f^*(\omega_i', \omega_s', \omega_p) \\
& \times \sum_{\substack{[j,k]= \\ \mathscr{P}[\omega_i,\omega_i']}} \sum_{\substack{[m,n]= \\ \mathscr{P}[\omega_s,\omega_s']}} \frac{f(k,n,\omega_p'')f(j,m,j+m+\omega_i'+\omega_s'-\omega_p-k-n+\omega_p'')}{(\omega_i'+\omega_s'-\omega_p)(k+n-\omega_p'')} \\
& - \frac{f(\omega_i,\omega_s,\omega_p)f(\omega_i',\omega_s',\omega_p'')}{\Delta\omega(\omega_i'+\omega_s'-\omega_p'')} f^*(\omega_i',\omega_s',2\omega_i'+2\omega_s'-\omega_p''-\Delta\omega)\big).
\end{aligned}
\tag{12}
$$

DISCUSSION

Expanding to first order, we see no difference between the Dyson and Taylor series. At second order, there appears to be an additional phenomenon for creating four-photon states in the Dyson series, however, these events are not observable due to destructive interference of the fields within the crystal. Only at third order, do we see a real difference between the two series: the joint spectral amplitude predicted by the Dyson series consists of a component that is equal to the amplitude predicted by the Taylor series— i.e. the creation of three independent pairs of photons—and an additional component corresponding to the creation of six-photon entangled state whose energies sum nontrivially to the energy of three pump photons.

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[2] We define $\sum_{[i,j]=\mathscr{P}[a,b]} f(i,j) = f(a,b) + f(b,a)$.