

Photon number divergence in the quantum theory of n -photon down conversion

Mark Hillery

*Department of Physics and Astronomy, Hunter College of the City University of New York,
695 Park Avenue, New York, New York 10021*

(Received 12 February 1990)

The process of n -photon down conversion is considered both with a quantized pump and in the parametric approximation. In the parametric approximation it is formally found that the photon number diverges at a finite time for $n > 2$. This divergence limits the times for which the parametric approximation is useful. The divergence does not occur if the pump is quantized. Furthermore, it is shown that the Hamiltonian with a quantized pump is essentially self-adjoint. This means that the questions that have been raised about the existence of the time-development operator in the parametric approximation do not arise when the pump is quantized.

I. INTRODUCTION

A two-photon down converter, otherwise known as a degenerate parametric amplifier, can be used to produce squeezed states.¹ In this device a pump field at frequency 2ω is converted into one at ω inside a nonlinear crystal. If the mode at frequency ω is initially in a coherent state, then it will be squeezed at later times by the action of the device. The oscillator version of the two-photon down converter has been used to produce squeezed states in the laboratory.² Because the quantum noise properties of squeezed states have proven to be interesting and useful it has been natural to inquire into the corresponding properties of the states which are produced by an n -photon down-conversion process. Here, controversy has arisen.^{3,4} The problem is whether the time-development transformation for an n -photon down converter is a meaningful object.

Before discussing this dispute let us examine the Hamiltonians which can be used to describe n -photon down conversion. If both the pump and signal modes are quantized then the Hamiltonian in the rotating-wave approximation is

$$H_n = \omega a^\dagger a + n\omega b^\dagger b + \kappa_n [a^n b^\dagger + (a^\dagger)^n b], \quad (1.1)$$

where a is the annihilation operator for the signal mode, b is the annihilation operator for the pump mode, and κ_n is a coupling constant which is proportional to the n th-order nonlinear polarizability coefficient of the crystal. If the pump mode is in a highly excited coherent state then one can treat it as a classical field. This is the parametric approximation and with it the Hamiltonian becomes

$$H_{pn} = \omega a^\dagger a + \kappa_n \beta [a^n e^{in\omega t} + (a^\dagger)^n e^{-in\omega t}], \quad (1.2)$$

where β is the amplitude of the pump field and, for simplicity, it has been chosen to be real. In the interaction picture this Hamiltonian is

$$H_{Ipn} = \kappa_n \beta [a^n + (a^\dagger)^n]. \quad (1.3)$$

The existence of the time-development transformation

for the n -photon down-conversion process was first considered by Fisher, Nieto, and Sandberg.³ They examined the time-development transformation generated by the Hamiltonian in Eq. (1.3). Formally this operator is given by the power series

$$U_{In}(t) = \exp(-itH_{Ipn}) = \sum_{k=0}^{\infty} (-itH_{Ipn})^k / k!. \quad (1.4)$$

In Ref. 3 this power series was used to find a series for the expectation value $\langle 0|U_{In}(t)|0\rangle$ by taking the vacuum expectation value of both sides of the above equation. It was found that this series does not converge for $n > 2$. This shows that the vacuum is not an analytic vector of i times the Hamiltonian for $n > 2$, i.e., the power series considered as a function of t does not converge for any value of $t > 0$. From this they concluded that it is not possible to define states which result from applying $U_{In}(t)$, for $n > 2$, to the vacuum.

This was disputed by Braunstein and McLachlan.⁴ They pointed out that the fact that the vacuum is not an analytic vector of iH_{Ipn} does not imply that a unitary time evolution operator does not exist. By using Padé approximants they were able to obtain numerically the Q functions for the states $U_{I3}(t)|0\rangle$ and $U_{I4}(t)|0\rangle$ for a limited range of time.

In this paper a number of aspects of the problem of the existence of a time-development operator for n -photon down conversion will be considered. In Sec. II the basic mathematical issues will be discussed. The theory in the parametric approximation will be examined in Sec. III. There it is shown that if we assume that everything is well defined, then for n equal to 3 or 4 the photon number becomes infinite in a finite time. This result presumably holds for higher values of n as well. The situation with a quantized pump is considered in Sec. IV. It is demonstrated that in this case the photon number is always finite and that the Hamiltonian is essentially self-adjoint. This implies the existence of a unitary time-development operator. Finally, the conclusions which can be drawn from these results are summarized.

II. MATHEMATICAL BACKGROUND

In order to generate a unitary time-development transformation the Hamiltonian of a quantum-mechanical system must be self-adjoint. What complicates matters is that most Hamiltonians, including those considered here, are unbounded operators. This means that the operator cannot act on all vectors in the Hilbert space but only on a particular subset which is called the domain of the operator. Both the operator and its domain must be considered when determining whether it is self-adjoint.

In order to illustrate some of these considerations we can examine the number operator $N = a^\dagger a$. The fact that N has eigenstates with arbitrarily large eigenvalues implies that it is unbounded. A necessary condition for a vector $|\psi\rangle$ to be in the domain of N is that $N|\psi\rangle$ be a normalizable vector, i.e., that $N|\psi\rangle$ be in the Hilbert space. For example, the vector

$$|\phi\rangle = \sum_{n=1}^{\infty} (1/n) |n\rangle \quad (2.1)$$

is not in the domain of N because although $|\phi\rangle$ has a finite norm $N|\phi\rangle$ does not, i.e.,

$$\|N|\phi\rangle\|^2 = \sum_{n=1}^{\infty} \langle n|n\rangle = \infty. \quad (2.2)$$

The largest choice of domain for N would be the set of all vectors such that $\|N|\phi\rangle\| < \infty$. It is also possible to define N on domains which are subsets of this one.

Before defining what is meant by self-adjoint we first need to define the adjoint of an operator.⁵ Let T be defined on a dense domain $D(T)$ which is a subset of a Hilbert space \mathcal{H} . Let the domain of T^\dagger , $D(T^\dagger)$, be the set of $\phi \in \mathcal{H}$ for which there is an $\eta \in \mathcal{H}$ with

$$\langle T\psi|\phi\rangle = \langle \psi|\eta\rangle, \quad (2.3)$$

for all $\psi \in D(T)$. For each $\phi \in D(T^\dagger)$ we define $T^\dagger\phi = \eta$, and T^\dagger is the adjoint of T . Note that this definition gives both the action of T^\dagger and its domain.

An operator T is symmetric if $T \subset T^\dagger$, i.e., if $D(T) \subset D(T^\dagger)$ and $T\phi = T^\dagger\phi$ for $\phi \in D(T)$. It is self-adjoint if it is symmetric and $D(T) = D(T^\dagger)$. It is the issues involving the domains which can make proving the self-adjointness of an operator difficult.

Note that for a symmetric operator the adjoint is an extension of the original operator; if T is symmetric, then T^\dagger has the same action as T on $D(T)$ and has a domain which is at least as big. If T^\dagger is self-adjoint, then T is said to be essentially self-adjoint. An essentially self-adjoint operator has a unique self-adjoint extension. In particular, if a Hamiltonian is essentially self-adjoint then its self-adjoint extension can be used to generate a unitary time evolution operator, and the dynamics is well defined. What we would like to do is to show that the Hamiltonians mentioned in the Introduction are essentially self-adjoint. This will be done in Sec. IV for H_n .

One way of proving that an operator is essentially self-adjoint is Nelson's theorem.⁶ This theorem makes use of the concept of analytic vectors. A vector ϕ is an analytic vector of an operator T if ϕ is in the domain of T^n for all

$n \geq 1$, and if there is a $t > 0$ such that

$$\sum_{n=0}^{\infty} t^n \|T^n \phi\| / n! < \infty. \quad (2.4)$$

In order to state the theorem we also need to know what a total set of vectors is. A set of vectors S is total in a Hilbert space \mathcal{H} if the set of all finite linear combinations of elements of S is dense in \mathcal{H} . Nelson's theorem states that if T is a symmetric operator and $D(T)$ contains a total set of analytic vectors, then T is essentially self-adjoint. This theorem provides a connection between analytic vectors and self-adjointness. As Braunstein and McLachlan pointed out, showing that the vacuum is not an analytic vector of iH_{Ipn} does not imply that H_{Ipn} is not self-adjoint, so that the self-adjointness of this operator is an open question.⁴ On the other hand, Nelson's theorem can be used to settle this issue for H_n .

III. PHOTON NUMBER IN THE PARAMETRIC APPROXIMATION

In this section we shall examine the fourth-order down-conversion process from a formal point of view, i.e., domain questions will be ignored. We shall find that in the parametric approximation the photon number becomes infinite in a finite period of time. This result is unphysical and results from the fact that pump depletion is neglected in the parametric approximation. The parametric approximation is clearly not valid beyond the time at which the photon number becomes infinite. In fact, it is only a good approximation for times at which pump depletion and quantum fluctuations of the pump are not important.^{7,8} It is expected that these times are much smaller than the time at which the photon number diverges.

The Hamiltonian for the four-photon down-conversion process in the parametric approximation is

$$H_{p4} = \omega a^\dagger a + \kappa_4 \beta [e^{4i\omega t} a^4 + e^{-4i\omega t} (a^\dagger)^4]. \quad (3.1)$$

This Hamiltonian can be used to calculate the time derivative of the photon number operator $N = a^\dagger a$

$$\begin{aligned} \frac{dN}{dt} &= -i[N, H_{p4}] \\ &= -4i\mu [A^\dagger(t)^4 - A(t)^4], \end{aligned} \quad (3.2)$$

where $\mu = \kappa_4 \beta$ and $A(t) = a(t)e^{i\omega t}$. The second derivative of N can also be calculated and is

$$\frac{d^2 N}{dt^2} = 64\mu^2 (2N^3 + 3N^2 + 7N + 3). \quad (3.3)$$

Let us consider the vacuum expectation values of these expressions. First, note that for $t > 0$

$$\frac{d\langle N(t) \rangle}{dt} = \left[\frac{d\langle N(t) \rangle}{dt} \right]_{t=0} + \int_0^t dt_1 \frac{d^2 \langle N(t_1) \rangle}{dt^2}, \quad (3.4)$$

where the angular brackets denote the vacuum expectation value. From Eq. (3.2) we see that the first term of the right-hand side of Eq. (3.4) is zero. It is also true that the integrand in the second term is positive. This follows

from Eq. (3.3) and the fact that N and its powers are positive operators. We can, therefore, conclude that

$$\frac{d\langle N(t) \rangle}{dt} \geq 0, \quad (3.5)$$

for $t \geq 0$. The mean photon number is an increasing function of time.

It is now useful to derive some inequalities involving expectation values of powers of the number operator. A direct application of the Schwarz inequality gives

$$\langle N(t)^2 \rangle \geq \langle N(t) \rangle^2, \quad \langle N(t)^4 \rangle \geq \langle N(t)^2 \rangle^2 \geq \langle N(t) \rangle^4. \quad (3.6)$$

Because $N(t)$ is a positive operator it has a well-defined square root. Making use of this fact and again applying the Schwarz inequality we find

$$\langle N(t)^2 \rangle^2 = \langle N(t)^{1/2} N(t)^{3/2} \rangle^2 \leq \langle N(t) \rangle \langle N(t)^3 \rangle. \quad (3.7)$$

Combining this relation with the second inequality in Eq. (3.6) gives

$$\langle N(t)^3 \rangle \geq \langle N(t) \rangle^3. \quad (3.8)$$

It is now possible to derive an inequality for $d\langle N(t) \rangle/dt$. Equation (3.3) and the results of the preceding paragraph imply that

$$\frac{d^2\langle N \rangle}{dt^2} \geq 64\mu^2(2\langle N \rangle^3 + 3\langle N \rangle^2 + 7\langle N \rangle + 3). \quad (3.9)$$

Because of Eq. (3.5) we can multiply both sides of the above inequality by $d\langle N \rangle/dt$ without changing its sense, yielding

$$\begin{aligned} \frac{d^2\langle N \rangle}{dt^2} \frac{d\langle N \rangle}{dt} &\geq 64\mu^2(2\langle N \rangle^3 + 3\langle N \rangle^2 + 7\langle N \rangle + 3) \frac{d\langle N \rangle}{dt} \\ \text{or} \end{aligned}$$

$$d \left[\left(\frac{d\langle N \rangle}{dt} \right)^2 \right] / dt \geq \frac{dV(\langle N \rangle)}{dt}, \quad (3.10)$$

where

$$V(\langle N \rangle) = 64\mu^2(\langle N \rangle^4 + 2\langle N \rangle^3 + 7\langle N \rangle^2 + 6\langle N \rangle). \quad (3.11)$$

Integrating both sides gives

$$\begin{aligned} \left[\left(\frac{d\langle N \rangle}{dt} \right)^2 \right]_t - \left[\left(\frac{d\langle N \rangle}{dt} \right)^2 \right]_0 &\geq V(\langle N(t) \rangle) - V(\langle N(0) \rangle). \quad (3.12) \end{aligned}$$

Because the system is in the vacuum state at $t=0$ we have that $\langle N(0) \rangle = 0$. We have already noted that $d\langle N(t) \rangle/dt$ is zero at $t=0$ so that Eq. (3.12) can be expressed as

$$\frac{d\langle N(t) \rangle}{dt} \geq [V(\langle N(t) \rangle)]^{1/2}. \quad (3.13)$$

It is possible to obtain a simpler result by noting that each of the terms in $V(\langle N \rangle)$ is positive. Dropping all but the $\langle N \rangle^4$ term gives

$$\frac{d\langle N \rangle}{dt} \geq 8\mu\langle N \rangle^2, \quad (3.14)$$

which can be expressed as

$$\frac{-d(1/\langle N \rangle)}{dt} \geq 8\mu. \quad (3.15)$$

Integrating both sides from t_1 to t_2 yields

$$1/\langle N(t_1) \rangle - 1/\langle N(t_2) \rangle \geq 8\mu(t_2 - t_1). \quad (3.16)$$

We now return to Eq. (3.9) and drop all but the last term on the right-hand side

$$\frac{d^2\langle N \rangle}{dt^2} \geq 192\mu^2. \quad (3.17)$$

Integrating twice and making use of the fact that $\langle N(t) \rangle$ and its first derivative vanish at $t=0$ gives

$$\langle N(t) \rangle \geq 96(\mu t)^2. \quad (3.18)$$

If we now set $t=t_1$ and invert both sides of the above inequality we obtain

$$1/\langle N(t_1) \rangle \leq 1/[96(\mu t_1)^2]. \quad (3.19)$$

Finally, this result can be combined with Eq. (3.16) to give

$$\langle N(t_2) \rangle \geq \{[1/96(\mu t_1)^2] - 8\mu(t_2 - t_1)\}^{-1}. \quad (3.20)$$

This should hold for all $t_1 < t_2$. Note that if

$$t_2 = t_1 + [1/(768\mu^3 t_1^2)], \quad (3.21)$$

then the right-hand side of Eq. (3.20) diverges.

Let us see what this inequality for $\langle N(t_2) \rangle$ implies. If $t_2 < t_d$, where $t_d = (3/8\mu)(1/6)^{1/3}$, then Eq. (3.21) has no solution, and for all $t_1 < t_2$ the right-hand side of Eq. (3.20) is finite. If $t_2 = t_d$, then when $t_1 = \frac{2}{3}t_d$ Eq. (3.21) is satisfied and $\langle N(t_2) \rangle$ must be infinite. Therefore, the latest time at which $\langle N(t) \rangle$ can diverge is t_d .

This divergence clearly places limits on the times for which the Hamiltonian in Eq. (3.1) is useful. In the case of two-photon down conversion, which has been extensively studied in connection with squeezing, the photon number also diverges. In that case, however, the divergence occurs only in the limit that t goes to infinity, and therefore does not present a problem. The four-photon process is clearly more divergent, and the Hamiltonian in Eq. (3.1) will provide a valid description only for times which are much less than t_d .

Finally, let us mention that the techniques employed in this section can be applied to H_{p3} as well. In this case one also finds that the photon number diverges in a finite time.

IV. QUANTIZED PUMP

Let us now examine the four-photon down-conversion process when the pump mode is quantized. This means

that we need to consider the Hamiltonian

$$H_4 = \omega a^\dagger a + 4\omega b^\dagger b + \kappa_4 [a^4 b^\dagger + (a^\dagger)^4 b] . \quad (4.1)$$

We want to use Nelson's theorem to show that this Hamiltonian is essentially self-adjoint. In order to do so it is necessary to find a domain on which H_4 is symmetric which contains a total set of analytic vectors.

Consider the set of states $S = \{|n_a, n_b\rangle | n_a = 0, 1, 2, \dots \text{ and } n_b = 0, 1, 2, \dots\}$ where $|n_a, n_b\rangle$ is a state with n_a photons in the a mode and n_b photons in the b mode. This set is total, i.e., the set of all finite linear combinations of vectors of this form is dense in the two-mode Hilbert space. Let us choose the domain of H_4 , $D(H_4)$, to be all finite linear combinations of elements of S . The Hamiltonian H_4 is symmetric on this domain. If it can be shown that each of the vectors $|n_a, n_b\rangle$ is analytic, then the conditions of Nelson's theorem will have been satisfied.

The Hamiltonian H_4 has a conservation law because the operator

$$M = 4b^\dagger b + a^\dagger a \quad (4.2)$$

commutes with H_4 . This allows us to break up the full Hilbert space into invariant subspaces \mathcal{H}_m . The space \mathcal{H}_m is the subspace on which M has the eigenvalue m and consists of all linear combinations of the vectors $\{|m, 0\rangle, |m-4, 1\rangle, \dots, |r, (m-r)/4\rangle\}$ where $r=0, 1, 2$, or 3 and is the remainder when m is divided by 4. If $\psi \in \mathcal{H}_m$, then the fact that $[M, H_4] = 0$ implies that any power of H_4 acting on ψ is also in \mathcal{H}_m .

If $\psi \in \mathcal{H}_m$ let us find a bound for $\|H_4 \psi\|$ in terms of $\|\psi\|$. If $\psi \in \mathcal{H}_m$, then it can be expressed as

$$|\psi\rangle = \sum_{k=0}^{[m/4]} c_k |m-4k, k\rangle , \quad (4.3)$$

where $[m/4]$ is the greatest integer less than or equal to $m/4$. We then have that

$$H_4 \psi = m\omega \psi + \psi_1 + \psi_2 , \quad (4.4)$$

where

$$\begin{aligned} \psi_1 &= \kappa_4 \sum_{k=0}^{[m/4]-1} c_k \sqrt{k+1} [(m-4k)!/(m-4k-4)!]^{1/2} \\ &\quad \times |m-4k-4, k+1\rangle , \\ \psi_2 &= \kappa_4 \sum_{k=1}^{[m/4]} c_k \sqrt{k} [(m-4k+4)!/(m-4k)!]^{1/2} \\ &\quad \times |m-4k+4, k-1\rangle . \end{aligned} \quad (4.5)$$

For $0 \leq k \leq [m/4]-1$ we have

$$\sqrt{k+1} [(m-4k)!/(m-4k-4)!]^{1/2} \leq m^{5/2}/2 , \quad (4.6)$$

and for $0 \leq k \leq m/4$

$$\sqrt{k} [(m-4k+4)!/(m-4k)!]^{1/2} \leq m^{5/2}/2 . \quad (4.7)$$

These imply that

$$\|\psi_1\| \leq \kappa_4 (m^{5/2}/2) \|\psi\| , \quad \|\psi_2\| \leq \kappa_4 (m^{5/2}/2) \|\psi\| , \quad (4.8)$$

which in turn gives us that

$$\|H_4 \psi\| \leq (m\omega + \kappa_4 m^{5/2}) \|\psi\| . \quad (4.9)$$

Because $H_4^n \psi$ is in \mathcal{H}_m if $\psi \in \mathcal{H}_m$ we can apply Eq. (4.9) repeatedly to give

$$\|H_4^n \psi\| \leq (m\omega + \kappa_4 m^{5/2})^n \|\psi\| . \quad (4.10)$$

Finally, this result can be applied to the exponential series to give for $\psi \in \mathcal{H}_m$

$$\begin{aligned} \sum_{n=0}^{\infty} \|H_4^n \psi\| t^n / n! &\leq \sum_{n=0}^{\infty} [(m\omega + \kappa_4 m^{5/2})^n t^n / n!] \|\psi\| \\ &\leq \exp[(m\omega + \kappa_4 m^{5/2})t] \|\psi\| . \end{aligned} \quad (4.11)$$

This implies that ψ is an analytic vector of H_4 . As each of the vectors $|n_a, n_b\rangle$ is in one of the subspaces \mathcal{H}_m (m for this state will be $n_a + 4n_b$) each is also an analytic vector of H_4 . Nelson's theorem now allows us to conclude that H_4 defined on the domain $D(H_4)$ is essentially self-adjoint.

The conservation law which H_4 obeys also implies that if the initial photon numbers in the a and b modes are finite, then they will be finite for all times. In fact, we have that if $\psi(t)$ is the wave function of the system at time t , then

$$\begin{aligned} \langle \psi(0) | M | \psi(0) \rangle &\geq \langle \psi(t) | a^\dagger a | \psi(t) \rangle , \\ \frac{1}{4} \langle \psi(0) | M | \psi(0) \rangle &\geq \langle \psi(t) | b^\dagger b | \psi(t) \rangle . \end{aligned} \quad (4.12)$$

Therefore the divergence problem which plagues this Hamiltonian in the parametric approximation is not present when the pump is quantized.

In this section we have proved the essential self-adjointness of H_4 defined on the domain consisting of all finite linear combinations of vectors of the form $|n_a, n_b\rangle$. The method of proof which was employed can be easily generalized to show that H_n defined on the same domain is also essentially self-adjoint. Therefore the quantum theory of n -photon down conversion with a quantized pump in the rotating-wave approximation is well defined.

V. CONCLUSION

We have discussed a number of issues in the quantum theory of n -photon down conversion. Both the case in which the pump was treated classically and that in which it was quantized were considered.

If the pump is classical (parametric approximation) it was formally shown that the photon number diverges at a finite time in the cases $n=3$ and 4. Because the theory is expected to become more singular as n increases, the suspicion is that this behavior holds for all $n > 2$. This divergence places limits on the use of the parametric approximation. This approximation will provide an accurate description of these processes only for times which

are much less than the divergence times.

If the pump is quantized the divergence of the photon number does not occur. Furthermore, it is possible to show explicitly that the Hamiltonian is essentially self-adjoint. This means that a unitary time-development operator exists in this case. The theory with a quantized pump is, therefore, well defined for all times.

ACKNOWLEDGMENTS

This research was supported by the National Science Foundation under Grant No. PHY 8802683 and by a grant under the Professional Staff Congress-City University of New York (PSC-CUNY) Research Award Program.

¹D. Stoler, Phys. Rev. D **1**, 3217 (1970).

²L. Wu, H. J. Kimble, J. L. Hall, and H. Wu, Phys. Rev. Lett. **57**, 2520 (1986).

³R. A. Fisher, M. M. Nieto, and V. D. Sandberg, Phys. Rev. D **29**, 1107 (1984).

⁴S. L. Braunstein and R. I. McLachlan, Phys. Rev. A **35**, 1659 (1987).

⁵M. Reed and B. Simon, *Functional Analysis*, Vol. 1 of *Methods*

of Modern Mathematical Physics (Academic, New York, 1972), p. 255.

⁶M. Reed and B. Simon, *Fourier Analysis and Self Adjointness*, Vol. 2 of *Methods of Modern Mathematical Physics* (Academic, New York, 1972), p. 200.

⁷M. Hillery and M. S. Zubairy, Phys. Rev. A **29**, 1275 (1984).

⁸D. D. Crouch and S. L. Braunstein, Phys. Rev. A **38**, 4696 (1988).