

Conservation laws and nonclassical states in nonlinear optical systems

Mark Hillery*

*Institute for Modern Optics, Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico 87131
and Max-Planck Institut für Quantenoptik, D-8046 Garching bei München, West Germany*

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It is shown that the states produced from the vacuum by subharmonic- and harmonic-generation processes are nonclassical for all times. This is a consequence of the Manley-Rowe relations. The stability of the nonclassical nature of these states under small perturbations (such as noise) is also discussed.

I. INTRODUCTION

It is possible in many circumstances to describe the electromagnetic field classically. There do exist, however, quantum states of the field for which no classical analog exists. That is, the set of correlation functions which characterizes such a state cannot be reproduced by a classical field. Such nonclassical states are of interest because they provide instances in which the quantum-mechanical nature of the field manifests itself and, hence, demonstrate the necessity of the quantum-mechanical description.

Nonclassical states can often be generated by nonlinear optical processes. For example, squeezed states, which are nonclassical, can be generated by a degenerate parametric amplifier¹⁻³ and in harmonic generation processes.^{4,5} Both of these processes also produce states whose photon statistics are subpoissonian, another class of nonclassical states.⁶⁻¹³ It is also possible to produce nonclassical states by means of four-wave mixing.¹⁴

These processes are generally described by model Hamiltonians. Despite the apparent simplicity of these Hamiltonians the resulting equations of motion have not been solved exactly. The results quoted above are obtained from either lowest order time-dependent perturbation theory or from the consideration of the steady state of the system in an oscillator configuration. Recently progress has been made in going beyond simple perturbation theory in the time-dependent problem, though these calculations are rather involved.^{15,16} The difficulty in obtaining solutions leads one to ask whether there are any properties of these systems which allow one to make statements about the nature of the states produced without having to solve the equations of motion.

The answer to this question is, in fact, yes, and the required properties are nothing other than the quantum-mechanical versions of the Manley-Rowe relations. These are simply conservation laws, and they constrain the dynamics of these systems to such an extent that it is often possible to predict that the states which are produced will be nonclassical for all times. This allows one to make statements about a time domain which is inaccessible to perturbation theory.

In this paper the production of nonclassical states in harmonic- and subharmonic-generation processes will be

discussed. Subharmonic processes will be considered in Sec. II and harmonic processes in Sec. III. In both cases it will be shown that if the signal mode (the harmonic or subharmonic) is initially in the vacuum state then the state at later times is either the vacuum state or nonclassical.

II. SUBHARMONIC PROCESSES

A. A criterion for finding nonclassical states

Before proceeding to a discussion of the states produced by subharmonic-generation processes it is useful to first discuss a result which allows one to identify nonclassical states. In order to do this it is first necessary to define what a classical state is more precisely. A state is classical if (a) its P representation has no singularities worse than a δ function and (b) its P representation is positive definite.¹⁷ A state which does not satisfy these conditions is nonclassical.

The criterion which we want to use to identify nonclassical states is as follows. Let ρ be the density matrix (or reduced density matrix) describing a mode of the radiation field. Consider a set of number states of this mode $S = \{ |n_\mu\rangle \}$ and the operator which projects onto this set

$$Q = \sum_{\mu} |n_\mu\rangle \langle n_\mu| . \tag{2.1}$$

The set S can be either finite or infinite. If the expectation value of Q in the state ρ is zero, i.e.,

$$\langle Q \rangle = \text{Tr}(\rho Q) = 0 , \tag{2.2}$$

then either ρ is the vacuum state or ρ is nonclassical.

This is relatively straightforward to show. Let us assume that ρ is classical, so that its P representation, $P(\alpha)$, satisfies conditions (a) and (b). Equation (2.2) can then be expressed as

$$\langle Q \rangle = \sum_{\mu} \int d^2\alpha P(\alpha) e^{-|\alpha|^2} \frac{|\alpha|^{2n_\mu}}{(n_\mu)!} = 0 , \tag{2.3}$$

where, by definition, $\int d^2\alpha P(\alpha) = 1$. The only way in which this equation can be satisfied if $P(\alpha) \geq 0$ is if S does not contain the vacuum state and $P(\alpha) = \delta^{(2)}(\alpha)$, i.e., only if $\rho = |0\rangle \langle 0|$. Therefore, if ρ is classical and satis-

fies Eq. (2.2) it must be the vacuum state. Any other state which satisfies Eq. (2.2) must be nonclassical. Note that if S contains the vacuum state and Eq. (2.2) is satisfied then we have the stronger result that ρ must be nonclassical.

B. Subharmonic generation

Subharmonic generation is a two-mode process in which a pump mode at frequency $n\omega$ produces a signal at ω through the action of a nonlinear medium. It is described by the model Hamiltonian

$$H_n = n\omega b^\dagger b + \omega a^\dagger a + \kappa_n [a^n b^\dagger + (a^\dagger)^n b], \quad (2.4)$$

where b and b^\dagger are the annihilation and creation operators for the pump mode, and a and a^\dagger are the operators for the signal mode. The initial states which we will consider are of the form

$$\rho(0) = |0\rangle_a \langle 0| \otimes \rho_b, \quad (2.5)$$

i.e., the signal mode in the vacuum state and the pump mode in an arbitrary state.

What will now be shown is that $\rho(t)$ describes a state which contains only numbers of signal photons which are a multiple of n . This result is plausible because each pump photon produces n signal photons and we started in a state with no signal photons present. It then follows immediately that the state of the signal mode is either nonclassical or the vacuum. This is because the reduced density matrix for the signal mode

$$\rho_a(t) = \text{Tr}_b[\rho(t)] \quad (2.6)$$

satisfies

$$\text{Tr}_a(\rho_a |n+1\rangle_a \langle n+1|) = 0, \quad (2.7)$$

where $|n+1\rangle_a$ is the state with $n+1$ photons in the a mode. We can then apply the result from Sec. II A.

In order to show formally that one obtains a signal mode state in which only states whose number of photons is a multiple of n are present we first note that the operator

$$M_n = nb^\dagger b + a^\dagger a \quad (2.8)$$

commutes with the Hamiltonian

$$[M_n, H_n] = 0. \quad (2.9)$$

Define the subspaces of the total Hilbert space of the system \mathcal{H} ,

$$\mathcal{H}_m = \{ |\Psi\rangle \in \mathcal{H} \mid M_n |\Psi\rangle = m |\Psi\rangle \}, \quad (2.10)$$

and let P_m be the projection onto \mathcal{H}_m . The operators P_m form a resolution of the identity, i.e.,

$$I = I_a \otimes I_b = \sum_{m=0}^{\infty} P_m. \quad (2.11)$$

The density matrix at $t=0$ is given by Eq. (2.5) and one can see that

$$P_m \rho(0) = 0 \quad (2.12)$$

unless $m=ln$ for some integer l . From Eq. (2.9) it follows that

$$P_m \rho(t) = 0 \quad (2.13)$$

unless $m=ln$.

Consider now the operator

$$K_{n_a} = |n_a\rangle_a \langle n_a| \otimes I_b.$$

This operator has the following two properties:

$$\text{Tr}[K_{n_a} \rho(t)] = \text{Tr}_a[|n_a\rangle_a \langle n_a| \rho_a(t)] \quad (2.14)$$

and

$$K_{n_a} P_m = 0 \quad (2.15)$$

unless $m=n_a+ln$ for some integer l . We now have that

$$\begin{aligned} \text{Tr}[K_{n_a} \rho(t)] &= \sum_{m=0}^{\infty} \text{Tr}[K_{n_a} P_m \rho(t)] \\ &= \sum_{l=0}^{\infty} \text{Tr}[K_{n_a} P_{ln} \rho(t)]. \end{aligned} \quad (2.16)$$

As can be seen from Eq. (2.15) each term in the sum in Eq. (2.16) will be zero unless n_a is a multiple of n . Therefore,

$$\text{Tr}_a[|n_a\rangle_a \langle n_a| \rho_a(t)] = 0 \quad (2.17)$$

unless n_a is a multiple of n .

It should be noted that the production of nonclassical states from a subharmonic-generation process does not depend upon the initial state of the pump. This is in contrast to squeezing which requires a pump with a well-defined phase. Any phase uncertainty degrades the squeezing. This uncertainty does not, however, affect the nonclassical nature of the state produced.

Let us conclude this subsection by showing how one can use the nonclassical states produced by subharmonic-generation processes to produce other types of nonclassical states. For simplicity we consider the case of second subharmonic generation ($n=2$).

Consider the following Gedanken experiment. Suppose that we have a cavity containing radiation produced from the signal-mode vacuum by a second subharmonic-generation process. This state, ρ_a , will contain only even numbers of photons (even photon state) and is nonclassical. It is possible to use this state to produce another state, ρ'_a , which will contain only odd numbers of photons (odd photon state). This state will also be nonclassical. The state ρ'_a can be generated by injecting a two-level atom, which is resonant with the signal mode, in its ground state into the cavity and allowing it to interact with the radiation for some time T . The atom is then removed from the cavity and measured. If it is found to be in its upper state then the state of the radiation field inside the cavity will contain only states with odd numbers of photons. This assertion is proved in Appendix A.

C. Stability of nonclassical nature

The nonclassical states which we have been considering are generated by somewhat idealized models of certain

nonlinear optical processes. Let us now consider the effects of making the systems less ideal. What happens, for example, if the system is connected to a reservoir? One can show that coupling the pump mode to a reservoir does not change the fact that the only states which become populated are those containing a number of signal-mode photons which is a multiple of n . It is, therefore, useful to consider the effects of noise and dissipation in the signal mode upon the nonclassical nature of the states which are produced. One would expect that the resulting state would contain both even and odd numbers of photons though more of one than the other. Is this state still nonclassical? In this subsection it will be shown that under certain conditions this is indeed the case.

Consider for simplicity the case of second subharmonic generation. Define the projection operators

$$\begin{aligned} Q_e &= \sum_{l=0}^{\infty} |2l\rangle_a \langle 2l|, \\ Q'_e &= Q_e - |0\rangle_a \langle 0|, \\ Q_0 &= \sum_{l=0}^{\infty} |2l+1\rangle_a \langle 2l+1|. \end{aligned} \quad (2.18)$$

We now prove two propositions.

(1) If a state has the property that the expectation value of Q_e in that state is less than $\frac{1}{2}$, i.e., $\langle Q_e \rangle < \frac{1}{2}$, then the state is nonclassical.

(2) If a state has the property that $\langle Q'_e \rangle \geq \langle Q_0 \rangle$, then the state is nonclassical.

Both of these statements are relatively straightforward to prove. Let us start with statement (1). If a state is classical, then

$$\begin{aligned} \langle Q_0 \rangle &= \int d^2\alpha P(\alpha) e^{-|\alpha|^2} \sum_{l=0}^{\infty} \frac{|\alpha|^{2(2l+1)}}{(2l+1)!} \\ &= \int d^2\alpha P(\alpha) e^{-|\alpha|^2} \sinh |\alpha|^2 \\ &\leq \sup_{x \geq 0} (e^{-x} \sinh x) = \frac{1}{2}, \end{aligned} \quad (2.19)$$

where we have made use of the fact that for a classical state, $P(\alpha)$ contains no worse than δ function singularities. Therefore, a state is nonclassical if $\langle Q_0 \rangle > \frac{1}{2}$ and because

$$\langle Q_0 \rangle + \langle Q_e \rangle = 1 \quad (2.20)$$

we have that a state is nonclassical if $\langle Q_e \rangle < \frac{1}{2}$. This proves proposition (1). In order to prove proposition (2) we note that for a classical state

$$\begin{aligned} \langle Q_0 \rangle - \langle Q'_e \rangle &= \int d^2\alpha P(\alpha) e^{-|\alpha|^2} \\ &\quad \times [\sinh |\alpha|^2 - (\cosh |\alpha|^2 - 1)] \\ &\geq 0. \end{aligned} \quad (2.21)$$

A state is then nonclassical if $\langle Q'_e \rangle \geq \langle Q_0 \rangle$.

Let us examine the consequences of all of this. Suppose we tried to generate an odd photon state through some process which was similar to that in our Gedanken experi-

ment only this time when the signal mode was coupled to a reservoir. The result would not be a pure odd photon state because the noise and damping would have caused some of the even photon number states to become populated. On the other hand, we would expect that if the noise were small and we were considering times short compared to the damping time these even-number states would be populated far less than the odd-number states, i.e., $\langle Q_0 \rangle \gg \langle Q_e \rangle$ or $\langle Q_e \rangle < \frac{1}{2}$ so that the resulting state would still be nonclassical. Similarly if we were trying to produce an even photon state from the vacuum by means of a subharmonic-generation process in the presence of noise we would expect to find that some of the odd photon number states had become populated. Again, for weak noise and times short compared to the damping time we would expect that the population of the odd-number states would be smaller than that of the even-number states with occupation numbers greater than zero (these states become populated in time $1/\kappa_2$ while the noise and damping are assumed small enough so that the odd-number states become populated much more slowly). Thus, $\langle Q'_e \rangle \geq \langle Q_0 \rangle$ and the state which is produced is still nonclassical. Our two propositions, then, indicate that even in the presence of noise subharmonic-generation processes should be capable of producing nonclassical states.

III. HARMONIC-GENERATION PROCESSES

Harmonic generation is also a two-mode process but in this case n pump photons combine to form one n th harmonic photon. This process is also described by the Hamiltonian in Eq. (2.4) but now the a mode is the pump and the b mode is the harmonic. The initial state of the system is now taken to be

$$\rho(0) = |\alpha\rangle_a \langle \alpha| \otimes |0\rangle_b \langle 0|, \quad (3.1)$$

where $|\alpha\rangle_a$ is a coherent state for the a mode. It will be shown, as before, that for later times the harmonic is either in a nonclassical state or in the vacuum state.

The initial state in Eq. (3.1) is considerably less general than that in Eq. (2.5). It is not necessary to be quite so restrictive. The results in this section will also be true if the a mode is in a classical state with the property that its P representation is identically zero for $|\alpha| > R$ for some R . It should be possible to generalize this result still further.

The essential idea behind the proof is that it takes n pump photons to produce one harmonic photon. The number distribution of the harmonic will, therefore, tend to be more concentrated around zero than that of the pump. It will, in fact, become sufficiently concentrated for the resulting state to become nonclassical.

For simplicity let us consider the case $n=2$ though the result holds for arbitrary n . In order to proceed we again note that $[M_2, H_2] = 0$ so that the total Hilbert space \mathcal{H} is the direct sum of the subspaces \mathcal{H}_m which are invariant under the action of the time development transformation. Define the operator Q_{n_b} as

$$Q_{n_b} = \left[\sum_{n \geq n_b} |n\rangle_b \langle n| \right] \otimes I_a. \quad (3.2)$$

Because $P_m(|\psi\rangle_a \otimes |n\rangle_b) \neq 0$ only if $m \geq 2n$, we have

$$\langle Q_{n_b} \rangle = \sum_{m \geq 2n_b} \langle P_m Q_{n_b} \rangle. \quad (3.3)$$

One can use the fact that $[P_m, Q_{n_b}] = 0$ to show

$$\langle P_m Q_{n_b} \rangle \leq \langle P_m \rangle. \quad (3.4)$$

For a pure state Ψ this follows from

$$\|Q_{n_b} P_m \Psi\|^2 \leq \|P_m \Psi\|^2 \quad (3.5)$$

because

$$\|P_m \Psi\|^2 = \langle \Psi | P_m^2 | \Psi \rangle = \langle \Psi | P_m | \Psi \rangle$$

and

$$\|Q_{n_b} P_m \Psi\|^2 = \langle \Psi | P_m Q_{n_b} | \Psi \rangle. \quad (3.6)$$

The inequality (3.5) itself is a result of the fact that $\|Q_{n_b}\| = 1$. The extension to the case of a density matrix is straightforward. Substituting inequality (3.4) into Eq. (3.3) yields

$$\langle Q_{n_b} \rangle \leq \sum_{m \geq 2n_b} \langle P_m \rangle. \quad (3.7)$$

Let us now suppose that the reduced density matrix for the b mode is classical. We then have that

$$\langle Q_{n_b} \rangle = \sum_{n \geq n_b} \int d^2\beta P(\beta) \frac{|\beta|^{2n}}{n!} e^{-|\beta|^2}, \quad (3.8)$$

where $P(\beta) \geq 0$. It is also possible to find $\langle P_m \rangle$ because this quantity does not change with time. Therefore,

$$\begin{aligned} \langle P_m \rangle &= \text{Tr}[\rho(t) P_m] = \text{Tr}[\rho(0) P_m] \\ &= e^{-|\alpha|^2} \frac{|\alpha|^{2m}}{m!}. \end{aligned} \quad (3.9)$$

Substituting these expressions into inequality (3.7) we obtain

$$\begin{aligned} \int d^2\beta P(\beta) \sum_{n \geq n_b} e^{-|\beta|^2} \frac{|\beta|^{2n}}{n!} \\ \leq \sum_{m \geq 2n_b} e^{-|\alpha|^2} \frac{|\alpha|^{2m}}{m!}. \end{aligned} \quad (3.10)$$

The object now is to show that if $P(\beta)$ is non-negative and satisfies inequality (3.10) for all n_b then it must be the case that $P(\beta) = \delta^{(2)}(\beta)$.

In order to prove this let us first note that if the function $A_n(x, y)$ is defined as

$$A_n(x, y) = \left[\sum_{l \geq 2n} \frac{y^l}{l!} \right] / \left[\sum_{j \geq n} \frac{x^j}{j!} \right], \quad (3.11)$$

then for any value of $x > 0$ we have that

$$\lim_{n \rightarrow \infty} A_n(x, y) = 0. \quad (3.12)$$

This is proved in Appendix B. Assume now that for some $R > 0$ that

$$\int_{|\beta| \geq R} d^2\beta P(\beta) = \Gamma \geq 0. \quad (3.13)$$

Because the function

$$F_n(x) = \sum_{j \geq n} \frac{x^j}{j!} e^{-x} \quad (3.14)$$

is monotonically increasing for $x \geq 0$ (as is easily demonstrated by taking its derivative), we have

$$\begin{aligned} F_{n_b}(R^2) \Gamma &\leq \int_{|\beta| \geq R} d^2\beta P(\beta) F_{n_b}(|\beta|^2) \\ &\leq \int d^2\beta P(\beta) F_{n_b}(|\beta|^2) \end{aligned} \quad (3.15)$$

so that

$$\Gamma \leq e^{(R^2 - |\alpha|^2)} A_{n_b}(R^2, |\alpha|^2). \quad (3.16)$$

Because of Eq. (3.12) we see that $\Gamma = 0$. Therefore, for all $|\beta| > 0$ we have that $P(\beta) = 0$ and because $P(\beta)$ cannot be more singular than a delta function it must also be true that $P(\beta) = \delta^{(2)}(\beta)$. Thus the harmonic is either in a nonclassical state or it is in the vacuum state.

IV. CONCLUSION

What has been shown is that both harmonic- and subharmonic-generation processes produce nonclassical states from the vacuum. That is, at any time t , the harmonic or subharmonic is either in a nonclassical state or the vacuum state. This result follows directly from the conservation laws which these systems obey and does not depend upon any approximations. We thus see that nonclassical states occur commonly as the output of nonlinear devices.

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APPENDIX A

Here it will be shown that if an atom absorbs a photon from a state which contains only even numbers of photons then the result is a state with only odd numbers of photons. For simplicity let us consider the case of a pure state so that at $t=0$ the state of the system is

$$\Psi(0) = |g\rangle \otimes |\psi_e\rangle, \quad (A1)$$

where $|g\rangle$ is the ground state of the atom ($|e\rangle$ is the excited state) and $|\psi_e\rangle$ is a state of the field which is a superposition of states containing even numbers of photons. The system is described by the Hamiltonian

$$H = \frac{1}{2} \omega (\sigma_3 + I) + \omega a^\dagger a + \lambda \sigma_1 (a^\dagger + a), \quad (A2)$$

where ω is the energy level spacing (and mode frequency), and Pauli matrices are being used to describe the atom. Define the unitary operator

$$U_0 = \sigma_3 e^{i n a^\dagger a} \quad (A3)$$

and note that

$$U_0 a^\dagger U_0^{-1} = -a^\dagger, \quad U_0 \sigma_1 U_0^{-1} = -\sigma_1, \quad (\text{A4})$$

$$U_0 a U_0^{-1} = -a.$$

This implies that

$$U_0 e^{-iHt} U_0^{-1} = e^{-iHt}. \quad (\text{A5})$$

Therefore,

$$U_0 \Psi(t) = U_0 e^{-iHt} U_0^{-1} U_0 \Psi(0) = -\Psi(t) \quad (\text{A6})$$

because $\sigma_3 |g\rangle = -|g\rangle$ and $e^{i\pi a^\dagger a} |\psi_e\rangle = |\psi_e\rangle$. It is also possible to express $\Psi(t)$ as

$$\Psi(t) = |g\rangle \otimes |\psi_1\rangle + |e\rangle \otimes |\psi_2\rangle, \quad (\text{A7})$$

where $|\psi_1\rangle$ and $|\psi_2\rangle$ are field states. If U_0 is applied to Eq. (A7) and use is made of Eq. (A6) we find

$$e^{i\pi a^\dagger a} |\psi_1\rangle = |\psi_1\rangle, \quad (\text{A8})$$

$$e^{i\pi a^\dagger a} |\psi_2\rangle = -|\psi_2\rangle,$$

so that $|\psi_1\rangle$ is a superposition of states with an even number of photons and $|\psi_2\rangle$ is a superposition of states with an odd number of photons. Therefore, if the atom is measured and found in the excited state the field contains only odd numbers of photons.

APPENDIX B

In this appendix it is shown that

$$\lim_{n \rightarrow \infty} A_n(x, y) = 0. \quad (\text{B1})$$

We first note that for $2n+1 > y$,

$$\begin{aligned} \sum_{m \geq 2n} \frac{y^m}{m!} &= \frac{y^{2n}}{(2n)!} \sum_{m=2n}^{\infty} \frac{y^{m-2n}}{m(m-1) \cdots (2n+1)} \\ &\leq \frac{y^{2n}}{(2n)!} \sum_{m=0}^{\infty} \left[\frac{y}{2n+1} \right]^m \\ &\leq \frac{y^{2n}}{(2n)!} \left[\frac{2n+1}{(2n+1)-y} \right] \end{aligned} \quad (\text{B2})$$

and

$$\sum_{m \geq n} \frac{x^m}{m!} \geq \frac{x^n}{n!}. \quad (\text{B3})$$

Therefore, we have that

$$A_n(x, y) \leq \left[\frac{y^2}{x} \right]^n \frac{n!}{(2n)!} \left[\frac{2n+1}{(2n+1)-y} \right]. \quad (\text{B4})$$

It is also the case that

$$\frac{n!}{(2n)!} \leq \left[\frac{e}{4} \right]^n \frac{(n+1)^{n+1}}{n^{2n}} \quad (\text{B5})$$

so that

$$A_n(x, y) \leq \left[\frac{e}{4} \frac{(n+1)^{(1+1/n)} y^2}{n^2 x} \right]^n \left[\frac{2n+1}{(2n+1)-y} \right]. \quad (\text{B6})$$

As $n \rightarrow \infty$ the first group in parentheses goes to 0 and the second group in parentheses goes to 1. Equation (B1) then follows.

*Address after February 1, 1985: Department of Physics and Astronomy, Hunter College of the City University of New York, 695 Park Avenue, New York, N.Y. 10021.

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