MODERN NONLINEAR OPTICS

Part 1

Second Edition

ADVANCES IN CHEMICAL PHYSICS
VOLUME 119

Edited by

Myron W. Evans

Series Editors

I. PRIGOGINE

Center for Studies in Statistical Mechanics and Complex Systems
The University of Texas
Austin, Texas
and
International Solvay Institutes
Université Libre de Bruxelles
Brussels, Belgium

and

STUART A. RICE

Department of Chemistry
and
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INTRODUCTION

Few of us can any longer keep up with the flood of scientific literature, even in specialized subfields. Any attempt to do more and be broadly educated with respect to a large domain of science has the appearance of tilting at windmills. Yet the synthesis of ideas drawn from different subjects into new, powerful, general concepts is as valuable as ever, and the desire to remain educated persists in all scientists. This series, *Advances in Chemical Physics*, is devoted to helping the reader obtain general information about a wide variety of topics in chemical physics, a field that we interpret very broadly. Our intent is to have experts present comprehensive analyses of subjects of interest and to encourage the expression of individual points of view. We hope that this approach to the presentation of an overview of a subject will both stimulate new research and serve as a personalized learning text for beginners in a field.

I. PRIGOGINE
STUART A. RICE

PREFACE

This volume, produced in three parts, is the Second Edition of Volume 85 of the series, *Modern Nonlinear Optics*, edited by M. W. Evans and S. Kielich. Volume 119 is largely a dialogue between two schools of thought, one school concerned with quantum optics and Abelian electrodynamics, the other with the emerging subject of non-Abelian electrodynamics and unified field theory. In one of the review articles in the third part of this volume, the Royal Swedish Academy endorses the complete works of Jean-Pierre Vigier, works that represent a view of quantum mechanics opposite that proposed by the Copenhagen School. The formal structure of quantum mechanics is derived as a linear approximation for a generally covariant field theory of inertia by Sachs, as reviewed in his article. This also opposes the Copenhagen interpretation. Another review provides reproducible and repeatable empirical evidence to show that the Heisenberg uncertainty principle can be violated. Several of the reviews in Part 1 contain developments in conventional, or Abelian, quantum optics, with applications.

In Part 2, the articles are concerned largely with electrodynamical theories distinct from the Maxwell–Heaviside theory, the predominant paradigm at this stage in the development of science. Other review articles develop electrodynamics from a topological basis, and other articles develop conventional or U(1) electrodynamics in the fields of antenna theory and holography. There are also articles on the possibility of extracting electromagnetic energy from Riemannian spacetime, on superluminal effects in electrodynamics, and on unified field theory based on an SU(2) sector for electrodynamics rather than a U(1) sector, which is based on the Maxwell–Heaviside theory. Several effects that cannot be explained by the Maxwell–Heaviside theory are developed using various proposals for a higher-symmetry electrodynamical theory. The volume is therefore typical of the second stage of a paradigm shift, where the prevailing paradigm has been challenged and various new theories are being proposed. In this case the prevailing paradigm is the great Maxwell–Heaviside theory and its quantization. Both schools of thought are represented approximately to the same extent in the three parts of Volume 119.

As usual in the *Advances in Chemical Physics* series, a wide spectrum of opinion is represented so that a consensus will eventually emerge. The prevailing paradigm (Maxwell–Heaviside theory) is ably developed by several groups in the field of quantum optics, antenna theory, holography, and so on, but the paradigm is also challenged in several ways: for example, using general relativity, using O(3) electrodynamics, using superluminal effects, using an extended electrodynamics based on a vacuum current, using the fact that longitudinal waves may appear in vacuo on the U(1) level, using a reproducible and repeatable device, known as the *motionless electromagnetic generator*, which extracts electromagnetic energy from Riemannian spacetime, and in several other ways. There is also a review on new energy sources. Unlike Volume 85, Volume 119 is almost exclusively dedicated to electrodynamics, and many thousands of papers are reviewed by both schools of thought. Much of the evidence for challenging the prevailing paradigm is based on empirical data, data that are reproducible and repeatable and cannot be explained by the Maxwell–Heaviside theory. Perhaps the simplest, and therefore the most powerful, challenge to the prevailing paradigm is that it cannot explain interferometric and simple optical effects. A non-Abelian theory with a Yang–Mills structure is proposed in Part 2 to explain these effects. This theory is known as O(3) *electrodynamics* and stems from proposals made in the first edition, Volume 85.

As Editor I am particularly indebted to Alain Beaulieu for meticulous logistical support and to the Fellows and Emeriti of the Alpha Foundation's Institute for Advanced Studies for extensive discussion. Dr. David Hamilton at the U.S. Department of Energy is thanked for a Website reserved for some of this material in preprint form.

Finally, I would like to dedicate the volume to my wife, Dr. Laura J. Evans.

Myron W. Evans

Ithaca, New York

CONTENTS

QUANTUM NOISE IN NONLINEAR OPTICAL PHENOMENA

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CONTENTS

I. INTRODUCTION

More than a century has passed since Planck discovered that it is possible to explain properties of the blackbody radiation by introducing discrete packets of energy, which we now call *photons*. The idea of discrete or quantized nature of energy had deep consequences and resulted in development of quantum mechanics. The quantum theory of optical fields is called *quantum optics*. The construction of lasers in the 1960s gave impulse to rapid development of nonlinear optics with a broad variety of nonlinear optical phenomena that have been experimentally observed and described theoretically and now are the subject of textbooks [1,2]. In early theoretical descriptions of nonlinear optical phenomena, the quantum nature of optical fields has been ignored on the grounds that laser fields are so strong, that is, the number of photons associated with them are so huge, that the quantum properties assigned to individual photons have no chances to manifest themselves. However, it turned out pretty soon that quantum noise associated with the vacuum fluctuations can have important consequences for the course of nonlinear phenomena. Moreover, it appeared that the quantum noise itself can change essentially when the quantum field is subject to the nonlinear transformation that is the essence of any nonlinear process. The quantum states with reduced quantum noise for a particular physical quantity can be prepared in various nonlinear processes. Such states have no classical counterparts; that is, the results of some physical measurements cannot be explained without explicit recall to the quantum character of the field. The methods of theoretical description of quantum noise are the subject of Gardiner's book [3]. This chapter is not intended as a presentation of general methods that can be found in the book; rather, we want to compare the results obtained with a few chosen methods for the two, probably most important, nonlinear processes: second-harmonic generation and downconversion with quantum pump.

Why have we chosen the second-harmonic generation and the downconversion to illustrate consequences of field quantization, or a role of quantum noise, in nonlinear optical processes? The two processes are at the same time similar and different. Both of them are described by the same interaction Hamiltonian, so in a sense they are similar and one can say that they show different faces of the same process. However, they are also different, and the difference between them consists in the different initial conditions. This difference appears to be very important, at least at early stages of the evolution, and the properties of the fields produced in the two processes are quite different. With these two best-known and practically very important examples of nonlinear optical processes, we would like to discuss several nonclassical effects and present the most common theoretical approaches used to describe quantum effects. The chapter is not intended to be a complete review of the results concerning the two processes that have been collected for years. We rather want to introduce the reader who is not an expert in quantum optics into this fascinating field by presenting not only the results but also how they can be obtained with presently available computer software. The results are largely illustrated graphically for easier comparisons. In Section II we introduce basic definitions and the most important formulas required for later discussion. Section III is devoted to presentation of results for second-harmonic generation, and Section IV results for downconversion. In the Appendixes A and B we have added examples of computer programs that illustrate usage of really existing software and were actually used in our calculations. We draw special attention to symbolic calculations and numerical methods, which can now be implemented even on small computers.

II. BASIC DEFINITIONS

In classical optics, a one mode electromagnetic field of frequency ω, with the propagation vector $\mathbf{k}$ and linear polarization, can be represented as a plane wave

$$E(\mathbf{r},t) = 2E_0 \cos(\mathbf{k}\cdot\mathbf{r} - \omega t + \varphi) \tag{1}$$

where E_0 is the amplitude and φ is the phase of the field. Assuming the linear polarization of the field, we have omitted the unit polarization vector to simplify the notation. Classically, both the amplitude E_0 and the phase φ can be well-defined quantities, with zero noise. Of course, the two quantities can be considered as classical random variables with nonzero variances; thus, they can be noisy in a classical sense, but there is no relation between the two variances and, in principle, either of them can be rendered zero giving the noiseless classical field. Apart from a constant factor, the squared real amplitude, E_0^2, is the intensity of the field. In classical electrodynamics there is no real need to use complex numbers to describe the field. However, it is convenient to work with exponentials rather than cosine and sine functions and the field (1) is usually written in the form

$$E(\mathbf{r},t) = E^{(+)}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + E^{(-)}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \tag{2}$$

with the complex amplitudes $E^{\pm} = E_0 e^{\pm i\varphi}$. The modulus squared of such an amplitude is the intensity of the field, and the argument is the phase. Both intensity and the phase can be measured simultaneously with arbitrary accuracy.

In quantum optics the situation is dramatically different. The electromagnetic field E becomes a quantum quantity; that is, it becomes an operator acting in a Hilbert space of field states, the complex amplitudes $E^{\pm}$ become the annihilation and creation operators of the electromagnetic field mode, and we have

$$\hat{E} = \sqrt{\frac{\hbar\omega}{2\varepsilon_0 V}}\left[\hat{a}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + \hat{a}^{+}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\right] \tag{3}$$

with the bosonic commutation rules

$$[\hat{a}, \hat{a}^{+}] = 1 \tag{4}$$

for the annihilation ($\hat{a}$) and creation ($\hat{a}^{+}$) operators of the field mode, where ε_0 is the electric permittivity of free space and V is the quantization volume. Because of laws of quantum mechanics, optical fields exhibit an inherent quantum indeterminacy that cannot be removed for principal reasons no matter how smart we are. The quantity

$$\mathscr{E}_0 = \sqrt{\frac{\hbar\omega}{2\varepsilon_0 V}} \tag{5}$$

appearing in (3) is a measure of the quantum optical noise for a single mode of the field. This noise is present even if the field is in the vacuum state, and for this reason it is usually referred to as the *vacuum fluctuations of the field* [4]. Quantum noise associated with the vacuum fluctuations, which appears because of noncommuting character of the annihilation and creation operators expressed by (4), is ubiquitous and cannot be eliminated, but we can to some extent control this noise by 'squeezing' it in one quantum variable at the expense of "expanding" it in another variable. This noise, no matter how small it is in comparison to macroscopic fields, can have very important macroscopic consequences changing the character of the evolution of the macroscopic fields. We are going to address such questions in this chapter.

The electric field operator (3) can be rewritten in the form

$$\hat{E} = \mathscr{E}_0\left[\hat{Q}\cos(\mathbf{k}\cdot\mathbf{r} - \omega t) + \hat{P}\sin(\mathbf{k}\cdot\mathbf{r} - \omega t)\right] \tag{6}$$

where we have introduced two Hermitian quadrature operators, $\hat{Q}$ and $\hat{P}$, defined as

$$\hat{Q} = \hat{a} + \hat{a}^+ , \qquad \hat{P} = -i(\hat{a} - \hat{a}^+) \tag{7}$$

which satisfy the commutation relation

$$[\hat{Q}, \hat{P}] = 2i \tag{8}$$

The two quadrature operators thus obey the Heisenberg uncertainty relation

$$\langle(\Delta\hat{Q})^2\rangle\langle(\Delta\hat{P})^2\rangle \;\geq\; 1 \tag{9}$$

where we have introduced the quadrature noise operators

$$\Delta\hat{Q} = \hat{Q} - \langle\hat{Q}\rangle , \qquad \Delta\hat{P} = \hat{P} - \langle\hat{P}\rangle \tag{10}$$

For the vacuum state or a coherent state, which are the minimum uncertainty states, the inequality (9) becomes equality and, moreover, the two variances are equal

$$\langle(\Delta\hat{Q})^2\rangle = \langle(\Delta\hat{P})^2 = 1 \tag{11}$$

The Heisenberg uncertainty relation (9) imposes basic restrictions on the accuracy of the simultaneous measurement of the two quadrature components of the optical field. In the vacuum state the noise is isotropic and the two components have the same level of quantum noise. However, quantum states can be produced in which the isotropy of quantum fluctuations is broken—the uncertainty of one quadrature component, say, $\hat{Q}$, can be reduced at the expense of expanding the uncertainty of the conjugate component, $\hat{P}$. Such states are called *squeezed states* [5,6]. They may or may not be the minimum uncertainty states. Thus, for squeezed states

$$\langle(\Delta\hat{Q})^2\rangle < 1 \quad \text{or} \quad \langle(\Delta\hat{P})^2\rangle < 1 \tag{12}$$

Squeezing is a unique quantum property that cannot be explained when the field is treated as a classical quantity—field quantization is crucial for explaining this effect.

Another nonclassical effect is referred to as *sub-Poissonian photon statistics* (see, e.g., Refs. 7 and 8 and papers cited therein). It is well known that in a coherent state defined as an infinite superposition of the number states

$$|\alpha\rangle = \exp\left(-\frac{|\alpha|^2}{2}\right)\sum_{n=0}^{\infty}\frac{\alpha^n}{\sqrt{n!}}\,|n\rangle \tag{13}$$

the photon number distribution is Poissonian

$$p(n) = |\langle n|\alpha\rangle|^2 = \exp(-|\alpha|^2)\frac{|\alpha|^{2n}}{n!} = \exp(-\langle\hat{n}\rangle)\frac{\langle\hat{n}\rangle^n}{n!} \tag{14}$$

which means

$$\langle(\Delta\hat{n})^2\rangle = \langle\hat{n}^2\rangle - \langle\hat{n}\rangle^2 = \langle\hat{n}\rangle \tag{15}$$

If the variance of the number of photons is smaller than its mean value, the field is said to exhibit the sub-Poissonian photon statistics. This effect is related to the second-order intensity correlation function

$$G^{(2)}(\tau) = \langle{:}\hat{n}(t)\hat{n}(t+\tau){:}\rangle = \langle\hat{a}^+(t)\hat{a}^+(t+\tau)\hat{a}(t+\tau)\hat{a}(t)\rangle \tag{16}$$

where :: indicate the normal order of the operators. This function describes the probability of counting a photon at t and another one at $t+\tau$. For stationary fields, this function does not depend on t but solely on τ. The normalized second-order correlation function, or second-order degree of coherence, is defined as

$$g^{(2)}(\tau) = \frac{G^{(2)}(\tau)}{\langle \hat{n} \rangle^2} \tag{17}$$

If $g^{(2)}(\tau) < g^{(2)}(0)$, the probability of detecting the second photon decreases with the time delay τ, indicating *bunching* of photons. On the other hand, if $g^{(2)}(\tau) > g^{(2)}(0)$, we have the effect of *antibunching* of photons. Photon antibunching is another signature of quantum character of the field. For $\tau = 0$, we have

$$g^{(2)}(0) = \frac{\langle \hat{a}^+ \hat{a}^+ \hat{a}\, \hat{a} \rangle}{\langle \hat{a}^+ \hat{a} \rangle^2} = \frac{\langle \hat{n}(\hat{n}-1) \rangle}{\langle \hat{n} \rangle^2} = 1 + \frac{\langle (\Delta\hat{n})^2 \rangle - \langle \hat{n} \rangle}{\langle \hat{n} \rangle^2} \tag{18}$$

which gives the relation between the photon statistics and the second-order correlation function. Another convenient parameter describing the deviation of the photon statistics from the Poissonian photon number distribution is the Mandel q parameter defined as [9]

$$q = \frac{\langle (\Delta\hat{n})^2 \rangle}{\langle \hat{n} \rangle} - 1 = \langle \hat{n} \rangle (g^{(2)}(0) - 1) \tag{19}$$

Negative values of this parameter indicate sub-Poissonian photon statistics, namely, nonclassical character of the field. One obvious example of the nonclassical field is a field in a number state $|n\rangle$ for which the photon number variance is zero, and we have $g^{(2)}(0) = 1 - 1/n$ and $q = -1$. For coherent states, $g^{(2)}(0) = 1$ and $q = 0$. In this context, coherent states draw a somewhat arbitrary line between the quantum states that have "classical analogs" and the states that do not have them. The coherent states belong to the former category, while the states for which $g^{(2)}(0) < 1$ or $q < 0$ belong to the latter category. This distinction is better understood when the Glauber–Sudarshan quasidistribution function $P(\alpha)$ is used to describe the field.

The coherent states (13) can be used as a basis to describe states of the field. In such a basis for a state of the field described by the density matrix ρ, we can introduce the quasidistribution function $P(\alpha)$ in the following way:

$$\rho = \int d^2\alpha\, P(\alpha) |\alpha\rangle\langle\alpha| \tag{20}$$

where $d^2\alpha = d\,\mathrm{Re}(\alpha) d\,\mathrm{Im}(\alpha)$. In terms of $P(\alpha)$, the expectation value of the normally ordered products (creation operators to the left and annihilation operators to the right) has the form

$$\langle (\hat{a}^+)^m \hat{a}^n \rangle = \mathrm{Tr}\,[\rho(\hat{a}^+)^m \hat{a}^n] = \int d^2\alpha\, P(\alpha)(\alpha^*)^m \alpha^n \tag{21}$$

For a coherent state $|\alpha_0\rangle$, $\rho = |\alpha_0\rangle\langle\alpha_0|$, and the quasiprobability distribution $P(\alpha) = \delta^{(2)}(\alpha - \alpha_0)$ giving $\langle (a^+)^m a^n \rangle = (\alpha^*)^m \alpha^n \rangle$. When $P(\alpha)$ is a well-behaved, positive definite function, it can be considered as a probability distribution function of a classical stochastic process, and the field with such a P function is said to have "classical analog." However, the P function can be highly singular or can take negative values, in which case it does not satisfy requirements for the probability distribution, and the field states with such a P function are referred to as *nonclassical states*.

From the definition (13) of coherent state it is easy to derive the completeness relation

$$\frac{1}{\pi}\int d^2\alpha\, |\alpha\rangle\langle\alpha| = 1 \tag{22}$$

and find that the coherent states do not form an orthonormal set

$$|\langle\alpha|\beta\rangle|^2 = \exp(-|\alpha - \beta|^2) \tag{23}$$

and only for $|\alpha - \beta|^2 \gg 1$ they are approximately orthogonal. In fact, coherent states form an overcomplete set of states.

To see the nonclassical character of squeezed states better, let us express the variance $\langle (\Delta\hat{Q})^2 \rangle$ in terms of the P function

$$\begin{aligned}
\langle (\Delta\hat{Q})^2 \rangle &= \langle (\hat{a} + \hat{a}^+)^2 \rangle - \langle (\hat{a} + \hat{a}^+) \rangle^2 \\
&= \langle \hat{a}^2 + \hat{a}^{+2} + 2\hat{a}^+\hat{a} + 1 \rangle - \langle \hat{a} + \hat{a}^+ \rangle^2 \\
&= 1 + \int d^2\alpha\, P(\alpha)[(\alpha + \alpha^*)^2 - \langle \alpha + \alpha^* \rangle^2]
\end{aligned} \tag{24}$$

which shows that $\langle (\Delta\hat{Q})^2 \rangle < 1$ is possible only if $P(\alpha)$ is not a positive definite function. The unity on the right-hand side of (24) comes from applying the commutation relation (4) to put the formula into its normal form, and it is thus a manifestation of the quantum character of the field ("shot noise").

Similarly, for the photon number variance, we get

$$\begin{aligned}
\langle (\Delta\hat{n})^2 \rangle &= \langle \hat{n} \rangle + \langle \hat{a}^{+2}\hat{a}^2 \rangle - \langle \hat{a}^+\hat{a} \rangle^2 \\
&= \langle \hat{n} \rangle + \int d^2\alpha\, P(\alpha)[|\alpha|^2 - \langle |\alpha|^2 \rangle]^2
\end{aligned} \tag{25}$$

Again, $\langle(\Delta\hat{n})^2\rangle < \langle\hat{n}\rangle$ only if $P(\alpha)$ is not positive definite, and thus sub-Poissonian photon statistics is a nonclassical feature.

In view of (24), one can write

$$\langle(\Delta\hat{Q})^2\rangle = 1 + \langle:(\Delta\hat{Q})^2:\rangle\,, \qquad \langle(\Delta\hat{P})^2\rangle = 1 + \langle:(\Delta\hat{P})^2:\rangle \tag{26}$$

where : : indicate the normal form of the operator. Using the normal form of the quadrature component variances squeezing can be conveniently defined by the condition

$$\langle:(\Delta\hat{Q})^2:\rangle < 0 \quad \text{or} \quad \langle:(\Delta\hat{P})^2:\rangle < 0 \tag{27}$$

Therefore, whenever the normal form of the quadrature variance is negative, this component of the field is squeezed or, in other words, the quantum noise in this component is reduced below the vacuum level. For classical fields, there is no unity coming from the boson commutation relation, and the normal form of the quadrature component represents true variance of the classical stochastic variable, which must be positive.

The Glauber–Sudarshan P representation of the field state is associated with the normal order of the field operators and is not the only c-number representation of the quantum state. Another quasidistribution that is associated with antinormal order of the operators is the Q representation, or the Husimi function, defined as

$$Q(\alpha) = \frac{1}{\pi}\langle\alpha|\rho|\alpha\rangle \tag{28}$$

and in terms of this function the expectation value of the antinormally ordered product of the field operators is calculated according to the formula

$$\langle\hat{a}^m(\hat{a}^+)^n\rangle = \frac{1}{\pi}\int d^2\alpha\,\langle\alpha|\rho|\alpha\rangle\alpha^m(\alpha^*)^n \tag{29}$$

It is clear from (28) that $Q(\alpha)$ is always positive, since ρ is a positive definite operator. For a coherent state $|\alpha_0\rangle$, $Q(\alpha) = (1/\pi)\exp(-|\alpha-\alpha_0|^2)$ is a Gaussian in the phase space $\{\mathrm{Re}\,\alpha, \mathrm{Im}\,\alpha\}$ which is centered at α_0. The section of this function, which is a circle, represents isotropic noise in the coherent state (the same as for the vacuum). The anisotropy introduced by squeezed states means a deformation of the circle into an ellipse or another shape.

Generally, according to Cahill and Glauber [10], one can introduce the s-parametrized quasidistribution function $\mathscr{W}^{(s)}(\alpha)$ defined as

$$\mathscr{W}^{(s)}(\alpha) = \frac{1}{\pi}\mathrm{Tr}\{\rho\,\hat{T}^{(s)}(\alpha)\} \tag{30}$$

where the operator $\hat{T}^{(s)}(\alpha)$ is given by

$$\hat{T}^{(s)}(\alpha) = \frac{1}{\pi}\int d^2\xi \, \exp(\alpha\xi^* - \alpha^*\xi)\hat{D}^{(s)}(\xi) \tag{31}$$

and

$$\hat{D}^{(s)}(\xi) = \exp\left(\frac{s\xi^2}{2}\right)\hat{D}(\xi) \tag{32}$$

where $\hat{D}(\xi)$ is the displacement operator and ρ is the density matrix of the field. The operator $\hat{T}^{(s)}(\alpha)$ can be rewritten in the form

$$\hat{T}^{(s)}(\alpha) = \frac{2}{1-s}\sum_{n=0}^{\infty}\hat{D}(\alpha)|n\rangle\left(\frac{s+1}{s-1}\right)^n\langle n|\hat{D}^+(\alpha) \tag{33}$$

which gives explicitly its s dependence. So, the s-parametrized quasidistribution function $\mathcal{W}^{(s)}(\alpha)$ has the following form in the number-state basis

$$\mathcal{W}^{(s)}(\alpha) = \frac{1}{\pi}\sum_{m,n}\rho_{mn}\langle n|\hat{T}^{(s)}(\alpha)|m\rangle \tag{34}$$

where the matrix elements of the operator (31) are given by

$$\begin{aligned}\langle n|\hat{T}^{(s)}(\alpha)|m\rangle = \sqrt{\frac{n!}{m!}}\left(\frac{2}{1-s}\right)^{m-n+1}\left(\frac{s+1}{s-1}\right)^n e^{-i(m-n)\theta}|\alpha|^{m-n} \\ \times \exp\left(-\frac{2|\alpha|^2}{1-s}\right)L_n^{m-n}\left(\frac{4|\alpha|^2}{1-s^2}\right)\end{aligned} \tag{35}$$

in terms of the associate Laguerre polynomials $L_n^{m-n}(x)$. In this equation we have also separated explicitly the phase of the complex number α by writing

$$\alpha = |\alpha|e^{i\theta} \tag{36}$$

The phase θ is the quantity representing the field phase.

With the quasiprobability distributions $\mathcal{W}^{(s)}(\alpha)$, the expectation values of the s-ordered products of the creation and annihilation operators can be obtained by proper integrations in the complex α plane. In particular, for $s = 1, 0, -1$, the s-ordered products are normal, symmetric, and antinormal ordered products of the creation and annihilation operators, and the corresponding distributions are the Glauber–Sudarshan P function, Wigner function, and Husimi Q function. By virtue of the relation inverse to (34), the field density matrix can be retrieved from the quasiprobability function

$$\rho = \int d^2\alpha\, \hat{T}^{(-s)}(\alpha)\mathscr{W}^{(s)}(\alpha) \tag{37}$$

Polar decomposition of the field amplitude, as in (36), which is trivial for classical fields becomes far from being trivial for quantum fields because of the problems with proper definition of the Hermitian phase operator. It was quite natural to associate the photon number operator with the intensity of the field and somehow construct the phase operator conjugate to the number operator. The latter task, however, turned out not to be easy. Pegg and Barnett [11–13] introduced the Hermitian phase formalism, which is based on the observation that in a finite-dimensional state space, the states with well-defined phase exist [14]. Thus, they restrict the state space to a finite $(\sigma + 1)$-dimensional Hilbert space $\mathrm{H}^{(\sigma)}$ spanned by the number states $|0\rangle, |1\rangle, \ldots, |\sigma\rangle$. In this space they define a complete orthonormal set of phase states by

$$|\theta_m\rangle = \frac{1}{\sqrt{\sigma+1}}\sum_{n}^{\sigma} \exp(in\theta_m)|n\rangle\,, \qquad m = 0, 1, \ldots, \sigma \tag{38}$$

where the values of θ_m are given by

$$\theta_m = \theta_0 + \frac{2\pi m}{\sigma+1} \tag{39}$$

The value of θ_0 is arbitrary and defines a particular basis set of $(\sigma + 1)$ mutually orthogonal phase states. The number state $|n\rangle$ can be expanded in terms of the $|\theta_m\rangle$ phase-state basis as

$$|n\rangle = \sum_{m=0}^{\sigma} |\theta_m\rangle\langle\theta_m|n\rangle = \frac{1}{\sqrt{\sigma+1}}\sum_{m=0}^{\sigma} \exp(-in\theta_m)|\theta_m\rangle \tag{40}$$

From Eqs. (38) and (40) we see that a system in a number state is equally likely to be found in any state $|\theta_m\rangle$, and a system in a phase state is equally likely to be found in any number state $|n\rangle$.

The Pegg–Barnett Hermitian phase operator is defined as

$$\hat{\Phi}_\theta = \sum_{m=0}^{\sigma} \theta_m |\theta_m\rangle\langle\theta_m| \tag{41}$$

Of course, the phase states (38) are eigenstates of the phase operator (40) with the eigenvalues θ_m restricted to lie within a phase window between θ_0 and $\theta_0 + 2\pi\sigma/(\sigma + 1)$. The Pegg–Barnett prescription is to evaluate any observable of interest in the finite basis (38), and only after that to take the limit $\sigma \to \infty$.

Since the phase states (38) are orthonormal, $\langle\theta_m|\theta_{m'}\rangle = \delta_{mm'}$, the kth power of the Pegg–Barnett phase operator (41) can be written as

$$\hat{\Phi}_\theta^k = \sum_{m=0}^{\sigma} \theta_m^k |\theta_m\rangle\langle\theta_m| \tag{42}$$

Substituting Eqs. (38) and (39) into Eq. (41) and performing summation over m yields explicitly the phase operator in the Fock basis:

$$\hat{\Phi}_\theta = \theta_0 + \frac{\sigma\pi}{\sigma + 1} + \frac{2\pi}{\sigma + 1} \sum_{n \neq n'} \frac{\exp[i(n - n')\theta_0]|n\rangle\langle n'|}{\exp[i(n - n')2\pi/(\sigma + 1)] - 1} \tag{43}$$

It is readily apparent that the Hermitian phase operator $\hat{\Phi}_\theta$ has well-defined matrix elements in the number-state basis and does not suffer from the problems as those the original Dirac phase operator suffered. Indeed, using the Pegg–Barnett phase operator (43) one can readily calculate the phase-number commutator [13]

$$\left[\hat{\Phi}_\theta, \hat{n}\right] = -\frac{2\pi}{\sigma + 1} \sum_{n \neq n'} \frac{(n - n')\exp[i(n - n')\theta_0]}{\exp[i(n - n')2\pi/(\sigma + 1)] - 1} |n\rangle\langle n'| \tag{44}$$

This equation looks very different from the famous Dirac postulate for the phase-number commutator.

The Pegg–Barnett Hermitian phase formalism allows for direct calculations of quantum phase properties of optical fields. As the Hermitian phase operator is defined, one can calculate the expectation value and variance of this operator for a given state $|f\rangle$. Moreover, the Pegg–Barnett phase formalism allows for the introduction of the continuous phase probability distribution, which is a representation of the quantum state of the field and describes the phase properties of the field in a very spectacular fashion. For so-called physical states, that is, states of finite energy, the Pegg–Barnett formalism simplifies considerably. In the limit as $\sigma \to \infty$ one can introduce the continuous phase distribution

$$P(\theta) = \lim_{\sigma \to \infty} \frac{\sigma + 1}{2\pi} |\langle\theta_m|f\rangle|^2 \tag{45}$$

where $(\sigma + 1)/2\pi$ is the density of states and the discrete variable θ_m is replaced by a continuous phase variable θ. In the number-state basis the

Pegg–Barnett phase distribution takes the form [15]

$$P(\theta) = \frac{1}{2\pi}\left\{1 + 2\mathrm{Re}\sum_{m>n}\rho_{mn}\exp\left[-i(m-n)\theta\right]\right\} \tag{46}$$

where $\rho_{mn} = \langle m|\rho|n\rangle$ are the density matrix elements in the number-state basis. The phase distribution (46) is 2π-periodic, and for all states with the density matrix diagonal in the number-state basis, the phase distribution is uniform over the 2π-wide phase window. Knowing the phase distribution makes the calculation of the phase operator expectation values quite simple; it is simply the calculation of all integrals over the continuous phase variable θ. For example,

$$\langle f|\hat{\Phi}_\theta^k|f\rangle = \int_{\theta_0}^{\theta_0+2\pi} d\theta\,\theta^k P(\theta) \tag{47}$$

When the phase window is chosen in such a way that the phase distribution is symmetrized with respect to the initial phase of the partial phase state, the phase variance is given by the formula

$$\langle(\Delta\hat{\Phi}_\theta)^2\rangle = \int_{-\pi}^{\pi} d\theta\,\theta^2 P(\theta) \tag{48}$$

For a partial phase state with the decomposition

$$|f\rangle = \sum_n b_n e^{in\varphi}|n\rangle \tag{49}$$

the phase variance has the form

$$\langle(\Delta\hat{\Phi}_\theta)^2\rangle = \frac{\pi^2}{3} + 4\sum_{n>k} b_n b_k \frac{(-1)^{n-k}}{(n-k)^2} \tag{50}$$

The value $\pi^2/3$ is the variance for the uniformly distributed phase, as in the case of a single-number state.

On integrating the quasiprobability distribution $\mathcal{W}^{(s)}(\alpha)$, given by (34), over the "radial" variable $|\alpha|$, we get a "phase distribution" associated with this quasiprobability distribution. The s-parametrized phase distribution is thus given by

$$P^{(s)}(\theta) = \int_0^\infty d|\alpha|\,\mathcal{W}^{(s)}(\alpha)|\alpha| \tag{51}$$

which, after performing of the integrations, gives the formula similar to the Pegg–Barnett phase distribution

$$P^{(s)}(\theta) = \frac{1}{2\pi}\left\{1 + 2\mathrm{Re}\sum_{m>n} \rho_{mn}\, e^{-i(m-n)\theta} G^{(s)}(m,n)\right\} \tag{52}$$

The difference between the Pegg–Barnett phase distribution (46) and the distribution (52) lies in the coefficients $G^{(s)}(m,n)$, which are given by [16]

$$G^{(s)}(m,n) = \left(\frac{2}{1-s}\right)^{(m+n)/2} \sum_{l=0}^{\min(m,n)} (-1)^l \left(\frac{1+s}{2}\right)^l \times \sqrt{\binom{n}{l}\binom{m}{l}}\, \frac{\Gamma\left(\frac{m+n}{2} - l + 1\right)}{\sqrt{(m-l)!(n-l)!}} \tag{53}$$

The phase distributions obtained by integration of the quasidistribution functions are different for different s, and all of them are different from the Pegg–Barnett phase distribution. The Pegg–Barnett phase distribution is always positive while the distribution associated with the Wigner distribution ($s = 0$) may take negative values. The distribution associated with the Husimi Q function is much broader than the Pegg–Barnett distribution, indicating that some phase information on the particular quantum state has been lost. Quantum phase fluctuations as fluctuations associated with the operator conjugate to the photon-number operator are important for complete picture of the quantum noise of the optical fields (for more details, see, e.g., Refs. 16 and 17).

III. SECOND-HARMONIC GENERATION

Second-harmonic generation, which was observed in the early days of lasers [18] is probably the best known nonlinear optical process. Because of its simplicity and variety of practical applications, it is a starting point for presentation of nonlinear optical processes in the textbooks on nonlinear optics [1,2]. Classically, the second-harmonic generation means the appearance of the field at frequency 2ω (second harmonic) when the optical field of frequency ω (fundamental mode) propagates through a nonlinear crystal. In the quantum picture of the process, we deal with a nonlinear process in which two photons of the fundamental mode are annihilated and one photon of the second harmonic is created. The classical treatment of the problem allows for closed-form solutions with the possibility of energy being transferred completely into the second-harmonic mode. For quantum fields, the closed-form analytical solution of the problem has not been found unless some approximations are made. The early numerical solutions [19,20] showed that quantum fluctuations of the field prevent the complete transfer of energy into the second harmonic and the solutions become oscillatory. Later studies showed that the quantum states of the field generated in the process have a number of unique quantum features such as photon antibunching [21] and squeezing [9,22] for both fundamental and second harmonic modes (for a review and literature, see Ref. 23). Nikitin and Masalov [24] discussed the properties of the quantum state of the fundamental mode by calculating numerically the quasiprobability distribution function $Q(\alpha)$. They suggested that the quantum state of the fundamental mode evolves, in the course of the second-harmonic generation, into a superposition of two macroscopically distinguishable states, similar to the superpositions obtained for the anharmonic oscillator model [25–28] or a Kerr medium [29,30]. Bajer and Lisoněk [31] and Bajer and Peřina [32] have applied a symbolic computation approach to calculate Taylor series expansion terms to find evolution of nonlinear quantum systems. A quasiclassical analysis of the second harmonic generation has been done by Alvarez-Estrada et al. [33]. Phase properties of fields in harmonics generation have been studied by Gantsog et al. [34] and Drobný and Jex [35]. Bajer et al. [36] and Bajer et al. [37] have discussed the sub-Poissonian behavior in the second- and third-harmonic generation. More recently, Olsen et al. [38,38] have investigated quantum-noise-induced macroscopic revivals in second-harmonic generation and criteria for the quantum nondemolition measurement in this process.

Quantum description of the second harmonic generation, in the absence of dissipation, can start with the following model Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_I \tag{54}$$

where

$$\hat{H}_0 = \hbar\omega\hat{a}^+\hat{a} + 2\hbar\omega\hat{b}^+\hat{b}\,, \qquad \hat{H}_I = \hbar\kappa(\hat{a}^2\hat{b}^+ + \hat{a}^{+2}\hat{b}) \tag{55}$$

and $\hat{a}$ ($\hat{a}^+$), $\hat{b}$ ($\hat{b}^+$) are the annihilation (creation) operators of the fundamental mode of frequency ω and the second harmonic mode at frequency 2ω, respectively. The coupling constant κ, which is real, describes the coupling between the two modes. Since $\hat{H}_0$ and $\hat{H}_I$ commute, there are two constants of motion: $\hat{H}_0$ and $\hat{H}_I$, $\hat{H}_0$ determines the total energy stored in both modes, which is conserved by the interaction $\hat{H}_I$. The free evolution associated with the Hamiltonian $\hat{H}_0$ leads to $\hat{a}(t) = \hat{a}(0)\exp(-i\omega t)$ and $\hat{b}(t) = \hat{b}(0)\exp(-i2\omega t)$. This trivial exponential evolution can always be factored out and the important part of the evolution described by the interaction Hamiltonian $\hat{H}_I$, for the slowly varying operators in the Heisenberg picture, is given by a set of equations

$$\frac{d}{dt}\hat{a}(t) = \frac{1}{i\hbar}[\hat{a}, \hat{H}_I] = -2i\kappa\,\hat{a}^{+}(t)\hat{b}(t)$$
$$\frac{d}{dt}\hat{b}(t) = \frac{1}{i\hbar}[\hat{b}, \hat{H}_I] = -i\kappa\,\hat{a}^{2}(t) \qquad (56)$$

where for notational convenience we use the same notation for the slowly varying operators as for the original operators — it is always clear from the context which operators are considered. In deriving the equations of motion (56), it is assumed that the operators associated with different modes commute, while for the same mode they obey the bosonic commutation rules (4).

Usually, the second-harmonic generation is considered as a propagation problem, not a cavity field problem, and the evolution variable is rather the path z the two beams traveled in the nonlinear medium. In the simplest, discrete two mode description of the process the transition from the cavity to the propagation problem is done by the replacement $t = -z/v$, where v denotes the velocity of the beams in the medium (we assume perfect matching conditions). We will use here time as the evolution variable, but it is understood that it can be equally well the propagation time in the propagation problem. So, we basically consider an idealized, one-pass problem. In fact, in the cavity situation the classical field pumping the cavity as well as the cavity damping must be added into the simple model to make it more realistic. Quantum theory of such a model has been developed by Drummond et al. [39,40]. Another interesting possibility is to study the second harmonic generation from the point of view of the chaotic behavior [41]. Such effects,however, will not be the subject of our concern here.

A. Classical Fields

Before we start with quantum description, let us recollect the classical solutions which will be used later in the method of classical trajectories to study some quantum properties of the fields. Equations (56) are valid also for classical fields after replacing the field operators $\hat{a}$ and $\hat{b}$ by the c-number field amplitudes α and β, which are generally complex numbers. They can be derived from the Maxwell equations in the slowly varying amplitude approximation [1] and have the form.

$$\frac{d}{dt}\alpha(t) = -2i\kappa\alpha^{*}(t)\beta(t)$$
$$\frac{d}{dt}\beta(t) = -i\kappa\alpha^{2}(t) \qquad (57)$$

For classical fields the closed-form analytical solutions to equations (57) are known. Assuming that initially there is no second-harmonic field ($\beta(0) = 0$), and the fundamental field amplitude is real and equal to $\alpha(0) = \alpha_0$ the solutions for the classical amplitudes of the second harmonic and fundamental modes are given by [1]

$$\begin{aligned}\alpha(t) &= \alpha_0 \operatorname{sech}(\sqrt{2}\,\alpha_0 \kappa t)\\ \beta(t) &= \frac{\alpha_0}{\sqrt{2}} \tanh(\sqrt{2}\,\alpha_0 \kappa t)\end{aligned} \tag{58}$$

The solutions (58) are monotonic and eventually all the energy present initially in the fundamental mode is transferred to the second-harmonic mode.

In a general case, when both modes initially have nonzero amplitudes, $\alpha_0 \neq 0$ and $\beta_0 \neq 0$, introducing $\alpha = |\alpha|e^{i\phi_a}$ and $\beta = |\beta|e^{i\phi_b}$, we obtain the following set of equations:

$$\begin{aligned}\frac{d}{dt}|\alpha| &= -2\kappa|\alpha||\beta|\sin\vartheta\\ \frac{d}{dt}|\beta| &= \kappa|\alpha|^2\sin\vartheta\\ \frac{d}{dt}\vartheta &= \kappa\left(\frac{|\alpha|^2}{|\beta|} - 4|\beta|\right)\cos\vartheta\\ \frac{d}{dt}\phi_a &= -2\kappa|\beta|\cos\vartheta\\ \frac{d}{dt}\phi_b &= -\kappa\frac{|\alpha|^2}{|\beta|}\cos\vartheta\end{aligned} \tag{59}$$

where $\vartheta = 2\phi_a - \phi_b$. The system (59) has two integrals of motion

$$C_0 = |\alpha|^2 + 2|\beta|^2, \qquad C_I = |\alpha|^2|\beta|\cos\vartheta \tag{60}$$

which are classical equivalents of the quantum constants of motion $\hat{H}_0$ and $\hat{H}_I$ ($C_0 = \langle\hat{H}_0\rangle$, $C_I = \langle\hat{H}_I\rangle$). Depending on the values of the constants of motion C_0 and C_I, the dynamics of the system (59) can be classified into several categories [42,43]:

1. *Phase-stable* motion, $C_I = 0$, in which the phases of each mode are preserved and the modes move radially in the phase space. The phase difference ϑ is also preserved, which appears for $\cos\vartheta = 0$ and $\vartheta = \pm\pi/2$. The solutions (58) belong to this category.
2. *Phase-changing* motion, $C_I \neq 0$, in which the dynamics of each mode involves both radial and phase motion. In this case both modes must be initially excited and their phase difference cannot be equal to $\pm\pi/2$.