PHYSICAL REVIEW A VOLUME 24, NUMBER 2 AUGUST 1981

Quasiprobability methods for nonlinear chemical and optical systems

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(Received 23 December 1980)

A review of quasiprobability methods for transforming chemical and quantum-optical master equations into Fokker-Planck equations is presented. For cases where conventional representations lead to Fokker-Planck equations with non-positive-definite diffusion coefficients; e.g., sub-Poissonian statistics, a generalization of the representation involving an extension to the complex plane enables analytic results to be obtained for certain nonlinear chemical and optical processes. Alternatively, a different integration measure may be chosen which ensures a positive distribution and Fokker-Planck equation with positive-semidefinite diffusion coefficients. This enables stochastic differential equations to be defined. These methods are applied to two-photon absorption and dispersive bistability in quantum optics where nonclassical photon statistics arise and to two models of nonlinear chemical reactions where sub-Poissonian statistics occur.

I. INTRODUCTION

In the rather different realms of chemical reaction theory and quantum optics, the methods of quasiprobabilities have provided technical tools of great power, which at the same time give insights into the physics of the processes under investigation. The first quasiprobability method was that introduced by Wigner[1] in a quantum-mechanical context. In quantum optics, however, the P representation introduced by Glauber[2,3] and Sudarshan[4] provided many practical applications of quasiprobabilities. The development of quantum-mechanical master equations by Van Hove,[5] Swanson,[6] Zwanzig,[7] Prigogine and Resibois,[8,9] in forms shown to be on the whole equivalent by Zwanzig[10] was fruitfully combined with the Glauber P representation to give a Fokker-Planck equation for the laser by Weidlich, Risken, and Haken[11,12] and Lax and Louisell.[13] A treatment of the damped harmonic oscillator using Glauber's P function was presented almost simultaneously by Louisell and Marburger.[14] Excellent treatises on the application of quantum-mechanical master equations to quantum optics have been written by Lax,[15] Haken,[16] Louisell,[17] Graham,[18] Haake,[19] Agarwal,[20] and Whitney and Scully.[21]

It was realized long ago[15-19] that the Fokker-Planck equations which arose from these treatments did not always have positive-definite diffusion coefficients, and thus the meaning and existence of their solutions was subject to doubt. In many cases the argument was made that the terms leading to the non-positive-definite form were small and could be omitted.

In recent work by the authors and co-workers, quasiprobability methods were developed for treating birth-death stochastic master equations, which had been introduced into chemistry by McQuarrie[22] and extensively developed by many authors.[23-28] Further, Malek-Mansour, Brenig, and Horsthemke[29] have applied these kinds of master equations to the kinetic theory of gases. The Poisson representation methods[30-33] which were introduced to transform chemical master equations into Fokker-Planck equations facilitated the analytic solution of these problems.[34] However, the Fokker-Planck equations did not always have positive-definite diffusion coefficients. These non-positive-definite diffusion coefficients could be handled by extending the domain of integration into the complex plane. This was initially done without rigorous justification, but correct results ensued.

The similarities between the quantum-optical and the chemical Fokker-Planck equations led to an extension of the complex-plane techniques to situations occurring in quantum-optical situations, which had hitherto been avoided, because of the non-positive semidefiniteness of the diffusion coefficient. A number of theoretical cases were successfully treated this way, including two-photon absorption,[35] sub- and second-harmonic generation,[36,37] and dispersive optical bistability.[38] In many of these examples exact steady-state solutions for the quasiprobability distributions may be obtained.

More recently it has been possible to provide a rigorous justification for these procedures which were being used without a formal definition. It is the purpose of the present paper to review and compare the results achievable in various cases in the light of this recent work, and to demonstrate the great similarity in techniques which can be employed in quantum optics and in chemical reaction models.

II. MASTER EQUATIONS

A. Quantum optics

Master equations provide a powerful tool for dealing with dissipation in quantum systems. Consider a quantum system described by an operator O undergoing a reversible interaction described by a Hamiltonian H_{rev} and interacting weakly with a large thermal reservoir described by an operator Γ. The Hamiltonian for the interaction of the quantum system and the reservoir is

$$\begin{aligned} H &= H_{\text{rev}} + H_{\text{irrev}} , \\ H_{\text{irrev}} &= (O\Gamma^\dagger + O^\dagger \Gamma). \end{aligned} \tag{2.1}$$

The quantum theory of dissipation has been developed by a number of authors.[15-17] An equation of motion for the density operator of the system alone may be obtained in the Markoff and Born approximations in the following form[15-17]:

$$\begin{aligned} \frac{\partial\rho}{\partial t} = \frac{1}{i\hbar}[\rho, H_{\text{rev}}] &+ \lambda([O\rho, O^\dagger] + [O, \rho O^\dagger]) \\ &+ \mu([O, O^\dagger\rho] + [\rho O, O^\dagger]), \end{aligned} \tag{2.2}$$

where λ and μ are constants proportional to expectation values of the reservoir operators. Consider, for example, a coherently driven harmonic oscillator damped by interaction with a thermal reservoir. The interaction Hamiltonian for this system is

$$H_I = i\hbar(\epsilon a^\dagger - \epsilon^* a) + a\Gamma^\dagger + a^\dagger\Gamma, \tag{2.3}$$

where a, $a^\dagger$ are the annhilation and creation operators for the harmonic oscillator and ϵ is the amplitude of the field driving the harmonic oscillator. The master equation for the density operator of the harmonic oscillator in the interaction picture is[15-17]

$$\begin{aligned} \frac{\partial\rho}{\partial t} = [\epsilon a^\dagger - \epsilon^* a, \rho] &+ \frac{\gamma}{2}(2a^\dagger\rho a - \rho a^\dagger a - a^\dagger a\rho) \\ &+ \gamma\bar{n}(a^\dagger\rho a + a\rho a^\dagger - a^\dagger a\rho - \rho a a^\dagger), \end{aligned} \tag{2.4}$$

where γ is the damping rate and $\bar{n} = 1/(e^{\hbar\omega/kT} - 1)$ is the mean excitation number in the reservoir. The above operator master equation is best solved by transforming to a convenient c-number form. One possible representation is to take matrix elements in the number or Fock states $|n\rangle$. For the above master equation with no driving field ($\epsilon = 0$), this yields a diagonal master equation for ρ_{nn} similar in form to those encountered in chemical reactions described in Sec. II B. However, with the driving term included the master equation is much more complicated, involving an infinite set of coupled equations for the matrix elements $\rho_{nn'}$.

An alternative route is to use a representation for ρ in terms of quasiprobability distributions. This allows the quantum-mechanical master equation to be converted to a c-number equation of Fokker-Planck form. The various possible representations of ρ are discussed in Sec. II B.

B. Chemical systems (Refs. 22-27)

The stochastic master equation approach to chemically reacting systems is a phenomenological approach, in which a time-development equation is derived for $p(x,t)$, the probability of there being x molecules of chemical substance x in the system at time t. One assumes that reactions occur because of collisions, and the probability per unit time of a reaction proceeding, in a system of sufficiently small volume, is given by the number of appropriate collisions per unit time multiplied by an intrinsic reaction probability. For example, for the dimerization reaction

$$Y \rightleftarrows 2X, \tag{2.5}$$

the probability of collisions between two X molecules is proportional to $\frac{1}{2}x(x-1)$, the number of possible pairs of X molecules. The reaction rate from left to right would be simply proportional to the number y of Y molecules. Thus we write the transition probabilities per unit time as

$$\begin{aligned} &\text{(a)}\quad Y \rightarrow 2X\text{:}\quad t^+(x,y) = k_1 y, \\ &\text{(b)}\quad 2X \rightarrow Y\text{:}\quad t^-(x,y) = k_2 x(x-1). \end{aligned} \tag{2.6}$$

A simple Markovian picture is now introduced, whereby the probability $P(x,y,t+\Delta t)$ is given by summing over all possible transitions from states whose probabilities were given by $p(x,y,t)$. This leads to the *chemical master equation*

$$\begin{aligned} \frac{\partial p(x,y,t)}{\partial t} = &\, k_2(x+2)(x+1)p(x+2, y-1, t) \\ &+ k_1(y+1)p(x-2, y+1, t) \\ &- [k_2 x(x-1) + k_1 y]\, p(x,y,t), \end{aligned} \tag{2.7}$$

where the terms on the right are interpreted as flow of probability from the states $(x+2, y-1)$ and $(x-2, y+1)$ into the state (x,y) from which is subtracted the flow of probability from the state (x,y) to these other two states.

An essential characteristic of the master equations of this type, in which the transition probabilities are given by simple combinational forms like those in Eq. (2.6), is the Poissonian nature of the stationary solutions in chemical equilibrium. For, in this case, *detailed balance* is satisfied, which means that in *any reaction*, the flow of probability from one state to another is exactly canceled by the flow in the reverse direction. This means that the Poisson distribution

$$p(x)=\frac{e^{-\alpha}\alpha^x}{x!} \tag{2.8}$$

plays a central role in the study of such systems, and gives rise to the Poisson representation[30-33] explained in Sec. III.

III. QUASIPROBABILITY REPRESENTATION

A. Quasiprobability methods in quantum optics

The quantum statistics of a single mode of the electromagnetic field (as well as other quantum problems involving bosons) is equivalent to that of a harmonic oscillator with annihilation and creation operators $(a, a^\dagger)$. All physical observables are obtained from the multinomial moments and correlations of $(a, a^\dagger)$ which for most cases in quantum optics are in normally ordered form. These in turn are determined using the quantum-density operator ρ, and it is often simplest to represent ρ using a distribution function over a c-number phase space. It is usual to expand ρ with the aid of the coherent states, defined as eigenstates of the annihilation operator

$$a|\alpha\rangle=\alpha|\alpha\rangle. \tag{3.1}$$

The Glauber-Sudarshan P representation is an expansion in diagonal coherent-state projection operators[2-4]

$$\rho=\int d^2\alpha\,|\alpha\rangle\langle\alpha|P(\alpha,\alpha^*). \tag{3.2}$$

This representation provides an easy recipe for calculating the average of the normally ordered operator products

$$\langle a^{\dagger n}a^{n'}\rangle=\int (\alpha^\dagger)^n\alpha^{n'}P(\alpha)d^2\alpha. \tag{3.3}$$

Despite the formal similarity to averaging with a classical probability distribution, $P(\alpha)$ is not a true probability distribution but belongs to a class of quasiprobability distributions. While $P(\alpha)$ exists for thermal light fields (a Gaussian distribution) and coherent laser fields (a δ-function distribution) for fields with nonclassical photon statistics, $P(\alpha)$ does not exist as a well-behaved positive function (although Klauder and Sudarshan[39] have shown that it does exist in terms of distributions with singularities). Such nonclassical fields have been observed in experiments on atomic fluorescence by Kimble *et al.*[40] and Leuchs *et al.*[41] following the predictions of Carmichael and Walls[42] Alternative quasiprobability distributions which avoid some of the problems of the P representation exist. The Wigner function which was the first quasiprobability method may be obtained from the P representation by the following integral:

$$W(\alpha)=\frac{2}{\pi}\int P(\beta)e^{-2|\beta-\alpha|^2}d^2\beta. \tag{3.4}$$

The Wigner function always exists as a nonsingular function but may assume negative values. The Wigner distribution simplifies averaging symmetrically ordered operator products but is less convenient for averaging the usual normally ordered operator products arising in quantum optics.

An alternative representation which is always positive is the Q representation or diagonal matrix elements of the density operator in terms of the coherent states

$$Q(\alpha,\alpha^*)=\langle\alpha|\rho|\alpha\rangle. \tag{3.5}$$

Though this representation is positive it has the disadvantage that not every positive Q function corresponds to a positive-semidefinite Hermitian density operator. The consequences of this are discussed in Sec. V. In addition, evaluating moments is only simple in the Q representation for antinormally ordered operator products.

Glauber generalized the diagonal P representation to an off-diagonal projection on the coherent states defined by[2]

$$\begin{aligned}R(\alpha^*,\beta)&=\langle\alpha|\rho|\beta\rangle\exp(\tfrac{1}{2}|\alpha|^2+\tfrac{1}{2}|\beta|^2)\,,\\ \rho&=\frac{1}{\pi^2}\int|\alpha\rangle R(\alpha^*,\beta)\langle\beta|\exp[-\tfrac{1}{2}(|\alpha|^2+|\beta|^2)]\,d^2\alpha\,d^2\beta.\end{aligned} \tag{3.6}$$

While the representation is analytic in α^*, β (and therefore nonsingular), it is also, by definition, nonpositive and has a normalization that includes a Gaussian weight factor. For this reason, it cannot have a Fokker-Planck equation or any direct interpretation as a quasiprobability. Nevertheless, the existence of this representation does demonstrate that a calculation of normally ordered observables for any ρ is possible with a nonsingular representation. In order to treat problems in nonlinear quantum optics where nonclassical photon statistics arise, a class of generalized P representations were introduced by Drummond and Gardiner[43] by expanding in nondiagonal coherent-state projection operators. The representation is defined as follows:

$$\rho = \int_{\mathfrak{D}} \Lambda(\alpha, \alpha^\dagger) P(\alpha, \alpha^\dagger) d\mu(\alpha, \alpha^\dagger), \tag{3.7}$$

where

$$\Lambda(\alpha, \alpha^\dagger) = \frac{|\alpha\rangle\langle(\alpha^\dagger)^*|}{\langle(\alpha^\dagger)^*|\alpha\rangle},$$

and $d\mu(\alpha, \alpha^\dagger)$ is the integration measure which may be chosen to define different classes of possible representations, and $\mathfrak{D}$ is the domain of integration. The projection operator $\Lambda(\alpha, \alpha^\dagger)$ is analytic in $(\alpha, \alpha^\dagger)$.

Useful choices of the integration measure are

(1) Glauber-Sudarshan P representation

$$d\mu(\alpha, \alpha^\dagger) = \delta^2(\alpha^* - \alpha^\dagger) d^2\alpha\, d^2\alpha^\dagger. \tag{3.8}$$

This measure corresponds to the diagonal Glauber-Sudarshan P representation defined in Eq. (3.2).

(2) Complex P representation

$$d\mu(\alpha, \alpha^\dagger) = d\alpha\, d\alpha^\dagger. \tag{3.9}$$

Here $(\alpha, \alpha^\dagger)$ are treated as complex variables which are to be integrated on individual contours C, C'. The existence of this representation under certain circumstances has been proved by Drummond and Gardiner.[43] In particular, this representation exists for an operator expanded in a finite basis of number states. This is a characteristic situation where nonclassical photon statistics (photon antibunching) may arise, and where the diagonal Glauber-Sudarshan P representation would be singular. This representation has been called the complex P representation since complex values of $P(\alpha, \alpha^\dagger)$ may occur. The representation gives rise to a $P(\alpha, \alpha^\dagger)$ which can be shown to satisfy a Fokker-Planck equation obtained by replacing (α, α^*) with $(\alpha, \alpha^\dagger)$ in the usual Glauber-Sudarshan type of Fokker-Planck equation.

Under certain circumstances, exact solutions to Fokker-Planck equations occur which cannot be normalized as Glauber-Sudarshan diagonal P functions. These can be handled with the present representation by choosing C, C' (paths of integration) in the complex phase space of $(\alpha, \alpha^\dagger)$.

(3) Positive P representation

$$d\mu(\alpha, \alpha^\dagger) = d^2\alpha\, d^2\alpha^\dagger. \tag{3.10}$$

This representation allows $(\alpha, \alpha^\dagger)$ to vary independently over the whole complex phase. It has been proved by Drummond and Gardiner[43] that $P(\alpha, \alpha^\dagger)$ always exists for a physical density operator, and can always be chosen positive, in which case we call it the positive P representation. This means that $P(\alpha, \alpha^\dagger)$ has all the properties of a genuine probability. It was also shown[43] that provided any Fokker-Planck equation exists for time development in the Glauber-Sudarshan representation, a corresponding Fokker-Planck equation exists with a positive-semidefinite diffusion coefficient for the positive P representation. This enables stochastic differential equations to be derived.

The above representations may be used to obtain time-development equations in a c-number representation from the quantum-operator equations in Sec. II. This may be achieved using simple operator correspondence rules. These are as follows[17]:

$$\begin{aligned} a\Lambda(\vec{\alpha}) &= \alpha\Lambda(\vec{\alpha}), \\ a^\dagger\Lambda(\vec{\alpha}) &= (\partial_\alpha + \alpha^\dagger)\Lambda(\vec{\alpha}), \\ \Lambda(\vec{\alpha})a &= (\alpha + \partial_{\alpha^\dagger})\Lambda(\vec{\alpha}), \\ \Lambda(\vec{\alpha})a^\dagger &= \alpha^\dagger\Lambda(\vec{\alpha}). \end{aligned} \tag{3.11}$$

The implementation of these rules to obtain Fokker-Planck equations from the operator master equations introduced in Sec. III are described in the following section. Explicit examples are given in Sec. V.

B. Quasiprobability methods in chemical physics

Similar quasiprobability techniques have been recently developed for application to the stochastic models of chemical reactions as described in Sec. II B. The technique (developed in fact before that of generalized P representations) consists simply of expanding the probability distribution over the number of molecules in Poisson distributions, thus

$$p(x) = \int d\mu(\alpha) f(\alpha) \frac{e^{-\alpha}\alpha^x}{x!}. \tag{3.12}$$

Here $\mu(\alpha)$ is a measure, which we shall shortly specify in a manner similar to that for the generalized P representation.

Comparing with the P representations, we can see that this expansion is analogous to the P representation of the density matrix ρ which is diagonal in a number state basis. For, in this case,

$$\langle x|\rho|x\rangle = \int d\mu(\alpha, \alpha^\dagger) P(\alpha, \alpha^\dagger) \frac{e^{-\alpha\alpha^\dagger}(\alpha\alpha^\dagger)^x}{x!} \tag{3.13}$$

and one can see immediately from Eq. (3.13), setting $p(x) = \langle x|\rho|x\rangle$, that if

$$d\mu(\beta) f(\beta) = \int d\mu(\alpha, \alpha^\dagger) \delta_\mu(\alpha\alpha^\dagger - \beta) P(\alpha, \alpha^\dagger), \tag{3.14}$$

the Poisson representation arises as a special case of the P representation. Here $\delta_\mu(\alpha-\beta)$ is the Dirac δ function defined with respect to the measure $\mu(\alpha)$, i.e., it satisfies

$$\int d\mu(\alpha)\phi(\alpha)\delta_\mu(\alpha-\beta)=\phi(\beta). \tag{3.15}$$

One sees that the correspondence between the Poisson variable α and the P-representation variables $(\alpha,\alpha^\dagger)$ is the slightly deceptive replacement

$$\alpha\,(\text{Poisson}) \longrightarrow \alpha^\dagger\alpha\,(P \text{ representation}), \tag{3.16}$$

which reflects the fact that the number operator in a quantum system is given by

$$N=a^\dagger a. \tag{3.17}$$

Of course, historically, this is not how the Poisson representation was viewed, and the relationship between the two representations turns out to be useful only because the existence theorems for the generalized P representations can be used to establish analogous existence theorems for the Poisson representation. There is a profound physical difference between the coherent states $|\alpha\rangle$ of the radiation field and the chemical Poissonian states. The quantum coherent states which arise as the quantum definition of a completely coherent field (e.g., a laser, or a radio field), are used to describe a situation which is far from thermodynamic equilibrium. The thermodynamic equilibrium situation is the blackbody radiator, which is completely differently described. The chemical Poissonian states are, in contrast, the states which naturally arise in chemically reacting dilute systems in thermodynamic equilibrium, and the occurrence of such a distribution has an origin which is completely different from that of the coherent states. The connection between the P representations and the Poisson representations is thus mathematical rather than physical.

1. Choice of measures

The most useful measures $\mu(\beta)$ are completely analogous to those used for the P representations, namely,

(a) *Real Poisson Representation*:

$d\mu(\alpha)=d\alpha$ (α on the real line).

(b) *Complex Poisson Representation*:

$d\mu(\alpha)=d\alpha$ (α a complex number, which ranges over a contour in the complex plane).

(c) *Positive Poisson Representations*:

$d\mu(\alpha)=d^2\alpha$ (α moves over the whole complex plane).

Since any probability distribution $p(x)$ can be used to construct a diagonal quantum-density matrix by

$$\rho_P=\sum_x |x\rangle\langle x|\,p(x), \tag{3.18}$$

where $|x\rangle$ are number states, it is easy to adapt the existence theorems on the P representations to give corresponding theorems for the various Poisson representations. In particular, one can see immediately that, using the relation given in Eq. (3.15), there always exists a positive Poisson representation with a non-negative $f(\alpha)$.

2. Fokker-Planck equations

Fokker-Planck equations can easily be derived for the Poisson representation quasiprobability $f(\alpha)$ by integrating by parts after substituting in the chemical master equation. The results are equivalent to the following rules. For an n-component reacting system involving 5 different reactions,

$$\sum_{i=1}^{n} N_i^P X_i \underset{k_P^B}{\overset{k_P^F}{\rightleftharpoons}} \sum_{i=1}^{n} M_i^P X_i,\quad P=1,2,\ldots s, \tag{3.19}$$

we define a multivariate quasiprobability $f(\alpha_1,\alpha_2,\ldots,t)$ in which α_i corresponds to x_i.

Define the currents

$$J_P(\vec{\alpha})=\left(\prod_{i=1}^{n} k_P^F\alpha_i^{N_i^P}-\prod_{i=1}^{n} k_P^B\alpha_i^{M_i^P}\right) \tag{3.20}$$

and the quantities

$$A_i^P=M_i^P-N_i^P, \tag{3.21}$$

$$B_{ij}(\vec{J}(\vec{\alpha}))=\sum_P J_P(\vec{\alpha})(M_i^P M_j^P-N_i^P N_j^P-\delta_{ij}A_i^P). \tag{3.22}$$

Then we have shown[31] that $f(\vec{\alpha},t)$ obeys the Fokker-Planck equation

$$\frac{\partial f(\vec{\alpha},t)}{\partial t}=-\sum_{i=1}^{n}\frac{\partial}{\partial\alpha_i}\left(\sum_P A_i^P J_P(\vec{\alpha})f(\vec{\alpha},t)\right)+\frac{1}{2}\sum_{i,j=1}^{n}\frac{\partial^2}{\partial\alpha_i\partial\alpha_j}[B_{ij}(\vec{J}(\alpha))f(\vec{\alpha},t)], \tag{3.23}$$

provided only that the reactions are no more than bimolecular, which specifically means that

$$\sum_{i=1}^{n} N_i^P \leqslant 2,\qquad \sum_{i=1}^{n} M_i^P \leqslant 2. \tag{3.24}$$

It becomes evident on inspection that the diffusion matrix $B_{ij}(\vec{J}(\vec{\alpha}))$ does not necessarily satisfy the positivity requirement for a Fokker-Planck equation, namely that the diffusion matrix $B_{ij}(\vec{J}(\vec{\alpha}))$

is not positive semidefinite, even when $\vec{\alpha}$ is real. We shall indicate in the next section how to deal with this problem by using the positive and complex Poisson representation.

IV. FOKKER-PLANCK EQUATION

Using the operator correspondences appropriate to the diagonal (Glauber-Sudarshan) P representation, an extensive formalism has been developed for converting quantum-mechanical master equations into Fokker-Planck equations for the corresponding quasiprobability $P(\alpha,\alpha^*)$, and this may be extended to give similar Fokker-Planck equations for generalized P representations by using the operator correspondences listed in Eq. (3.11). However, in exactly the same way as mentioned at the end of the previous section, the Fokker-Planck equations do not have positive-semidefinite diffusion matrices and interpretation difficulties arise. We will show how these difficulties may be overcome. The explanation will be given for the P representations, the adaptation to the Poisson representation being obvious.

A. Positive P representations

In the situations where a quantum-mechanical master equation yields a Fokker-Planck equation, one normally finds, as an intermediate step in the derivation, an equation for the time development of the master equation of the form [where $(\alpha,\alpha^\dagger)=\vec{\alpha}\equiv(\alpha^{(1)},\alpha^{(2)})$]

$$\partial_t\rho=\iint\Lambda(\vec{\alpha})\partial_t P(\vec{\alpha})d^2\alpha\, d^2\alpha^\dagger$$
$$=\iint P(\vec{\alpha})[A^\mu(\vec{\alpha})\partial_\mu+\tfrac{1}{2}D^{\mu\nu}(\vec{\alpha})\partial_\mu\partial_\nu]\Lambda(\vec{\alpha})d^2\alpha\, d^2\alpha^\dagger . \tag{4.1}$$

Here, and in the following, repeated indices are summed over. Before integrating by parts, we carry out some transformations. Firstly, the symmetric diffusion matrix $\overleftrightarrow{\mathrm{D}}(\vec{\alpha})$ can always be factorized into the form

$$\overleftrightarrow{\mathrm{D}}(\vec{\alpha})=\overleftrightarrow{\mathrm{B}}(\vec{\alpha})\overleftrightarrow{\mathrm{B}}^T(\vec{\alpha}). \tag{4.2}$$

We now separate into real and imaginary parts,

$$\vec{\mathrm{A}}(\vec{\alpha})=\vec{\mathrm{A}}_x(\vec{\alpha})+i\vec{\mathrm{A}}_y(\vec{\alpha}), \tag{4.3}$$

$$\overleftrightarrow{\mathrm{B}}(\vec{\alpha})=\overleftrightarrow{\mathrm{B}}_x(\vec{\alpha})+i\overleftrightarrow{\mathrm{B}}_y(\vec{\alpha}), \tag{4.4}$$

$$\vec{\alpha}=\vec{\alpha}_x+i\vec{\alpha}_y, \tag{4.5}$$

and use the analyticity of $\Lambda(\vec{\alpha})$ to write

$$\partial_\mu\Lambda(\vec{\alpha})=\partial^x_\mu\Lambda(\vec{\alpha})=-i\partial^y_\mu\Lambda(\vec{\alpha}). \tag{4.6}$$

We then find that from (4.1) we may write

$$\iint\Lambda(\vec{\alpha})\partial_t P(\vec{\alpha})d^2\alpha\, d^2\alpha^\dagger=\iint P(\vec{\alpha})[A^\mu_x(\vec{\alpha})\partial^x_\mu+A^\mu_y\partial^y_\mu+\tfrac{1}{2}(B^{\mu\sigma}_xB^{\nu\sigma}_x\partial^x_\mu\partial^x_\nu+B^{\mu\sigma}_yB^{\nu\sigma}_y\partial^y_\mu\partial^y_\nu+2B^{\mu\sigma}_xB^{\nu\sigma}_y\partial^x_\mu\partial^y_\nu)]\Lambda(\vec{\alpha})d^2\alpha\, d^2\alpha^\dagger . \tag{4.7}$$

We now integrate by parts, discarding surface terms, to derive the Fokker-Planck equation

$$\partial_t P(\vec{\alpha})=\{-\partial^x_\mu A^\mu_x(\vec{\alpha})-\partial^y_\mu A^\mu_y(\vec{\alpha})+\tfrac{1}{2}[\partial^x_\mu\partial^x_\nu B^{\mu\sigma}_x(\vec{\alpha})B^{\nu\sigma}_x(\vec{\alpha})+\partial^y_\mu\partial^y_\nu B^{\mu\sigma}_y(\vec{\alpha})B^{\nu\sigma}_y(\vec{\alpha})+2\partial^x_\mu\partial^y_\nu B^{\mu\sigma}_x(\vec{\alpha})B^{\nu\sigma}_y(\vec{\alpha})]\}P(\vec{\alpha}). \tag{4.8}$$

The Fokker-Planck equation is not unique, since the $\Lambda(\vec{\alpha})$ are not linearly independent—however, the original master equation is a consequence of it, and this is all that is physically required.

However, the Fokker-Planck equation (4.8) now possesses a positive-semidefinite diffusion matrix in a four-dimensional space whose vectors are

$$(\vec{\alpha}^{(1)}_x,\vec{\alpha}^{(2)}_x,\vec{\alpha}^{(1)}_y,\vec{\alpha}^{(2)}_y)\equiv(\vec{\alpha}_x,\vec{\alpha}^{\dagger}_x,\vec{\alpha}_y,\vec{\alpha}^{\dagger}_y). \tag{4.9}$$

We find the drift vector is

$$\vec{\mathfrak{A}}(\alpha)\equiv(\vec{\mathrm{A}}^{(1)}_x(\vec{\alpha}),\vec{\mathrm{A}}^{(2)}_x(\vec{\alpha}),\vec{\mathrm{A}}^{(1)}_y(\vec{\alpha}),\vec{\mathrm{A}}^{(2)}_y(\vec{\alpha})) \tag{4.10}$$

and the diffusion matrix is

$$\overleftrightarrow{\mathfrak{D}}(\vec{\alpha})\equiv\begin{bmatrix}\overleftrightarrow{\mathrm{B}}_x\cdot\overleftrightarrow{\mathrm{B}}^T_x & \overleftrightarrow{\mathrm{B}}_x\cdot\overleftrightarrow{\mathrm{B}}^T_y\\ \overleftrightarrow{\mathrm{B}}_y\cdot\overleftrightarrow{\mathrm{B}}^T_x & \overleftrightarrow{\mathrm{B}}_y\cdot\overleftrightarrow{\mathrm{B}}^T_y\end{bmatrix}(\vec{\alpha})\equiv\overleftrightarrow{\mathfrak{B}}(\vec{\alpha})\overleftrightarrow{\mathfrak{B}}^T(\vec{\alpha}), \tag{4.11}$$

where

$$\overleftrightarrow{\mathfrak{B}}(\alpha)\equiv\begin{bmatrix}\overleftrightarrow{\mathrm{B}}_x & 0\\ \overleftrightarrow{\mathrm{B}}_y & 0\end{bmatrix}(\vec{\alpha}) \tag{4.12}$$

and the matrix is thus explicitly positive semidefinite. We have thus obtained an explicitly probabilistic Fokker-Planck equation for the positive $P(\alpha,\alpha^\dagger)$, so that in terms of this quasiprobability, the quantum stochastic process yields a classical stochastic process.

One may now use the well-known correspondence between Fokker-Planck and stochastic differential equations to write the following Itô stochastic differential equation (for an exposition of this relationship, see Refs. 44 and 45):

$$\frac{\partial}{\partial t}\begin{pmatrix}\vec{\alpha}_x\\ \vec{\alpha}_y\end{pmatrix}=\begin{pmatrix}\vec{\mathrm{A}}_x(\vec{\alpha})\\ \vec{\mathrm{A}}_y(\vec{\alpha})\end{pmatrix}+\begin{pmatrix}\overleftrightarrow{\mathrm{B}}_x(\vec{\alpha})\cdot\vec{\xi}(\vec{\mathrm{t}})\\ \overleftrightarrow{\mathrm{B}}_y(\vec{\alpha})\cdot\vec{\xi}(\vec{\mathrm{t}})\end{pmatrix} \tag{4.13}$$

or, recombining real and imaginary parts,

$$\frac{\partial\vec{\alpha}}{\partial t}=\vec{\mathrm{A}}(\vec{\alpha})+\vec{\mathrm{B}}(\alpha)\cdot\vec{\xi}(t)\,. \qquad (4.14)$$

Apart from the substitution $\alpha^*\rightarrow\alpha^\dagger$, Eq. (4.14) is just the stochastic differential equation which would be obtained by using the Glauber-Sudarshan representation, and naively converting the Fokker-Planck equation with a non-positive-definite diffusion matrix into an Itô stochastic differential equation.

In our derivation, the two formal variables (α,α^*) have been replaced by variables in the complex plane $(\alpha,\alpha^\dagger)$ that are allowed to fluctuate independently. The positive P representation as defined here thus appears as a mathematical justification of this procedure.

B. Positive-Poisson representation

A similar technique can be used to develop positive Fokker-Planck equations from the positive Poisson representation, using equations of the form (3.23). The validity of the result depends on the analyticity of the Poisson function $e^{-\alpha}\alpha^x/x!$ in the same way as the previous result depended on the analyticity of $\Lambda(\alpha)$. We find that $\vec{\alpha}$ in this case become complex fluctuating variables, and the resulting stochastic differential equation for this complex $\vec{\alpha}$ is

$$\frac{d\vec{\alpha}}{dt}=\sum_P\vec{\mathrm{A}}^P J_P(\vec{\alpha})+\overleftrightarrow{\mathrm{C}}(\vec{\mathrm{J}}(\vec{\alpha}))\cdot\vec{\xi}(t)\,, \qquad (4.15)$$

where

$$\overleftrightarrow{\mathrm{C}}(\vec{\mathrm{J}}(\vec{\alpha}))\overleftrightarrow{\mathrm{C}}^T(\vec{\mathrm{J}}(\vec{\alpha}))=\overleftrightarrow{\mathrm{B}}(\vec{\mathrm{J}}(\alpha))\,. \qquad (4.16)$$

We develop specific examples of these equations in Sec. VI.

C. Complex P representation

The procedure here is very similar to that used above: the master equation is reduced to the form

$$\begin{aligned}\partial_t\rho&=\int_C\int_{C'}\Lambda(\vec{\alpha})\partial_t P(\vec{\alpha})\,d\alpha\,d\alpha^\dagger\\&=\int\int P(\vec{\alpha})[A^\mu(\vec{\alpha})\partial_\mu+\tfrac{1}{2}D^{\mu\nu}(\vec{\alpha})\partial_\mu\partial_\nu]\Lambda(\vec{\alpha})d\alpha\,d\alpha^\dagger\,,\end{aligned} \qquad (4.17)$$

where now $(\alpha,\alpha^\dagger)\equiv(\alpha^{(1)},\alpha^{(2)})$ are independent complex variables integrated over contours C, C' in their respective complex planes. We now integrate by parts in the complex plane, and obtain an *analytic* Fokker-Planck equation

$$\partial_t P(\vec{\alpha})=[-\partial_\mu A^\mu(\alpha)+\tfrac{1}{2}\partial_\mu\partial_\nu D^{\mu\nu}(\vec{\alpha})]P(\vec{\alpha})\,. \qquad (4.18)$$

That is, since this equation possesses coefficients, which are in all practical cases analytic functions of $(\alpha,\alpha^\dagger)$, it possesses solutions which are also analytic, provided the initial condition is analytic.

This kind of equation is often useful in comparing stationary properties, since we often find that the coefficients satisfy potential conditions[18,46]

$$\partial_\mu v_\nu=\partial_\nu v_\mu\,, \qquad (4.19)$$

where

$$v_\rho(\vec{\alpha})=[\overleftrightarrow{\mathrm{D}}(\vec{\alpha})]^{-1}_{\rho\nu}[2A_\nu(\vec{\alpha})-\partial_\sigma D_{\nu\sigma}(\vec{\alpha})]\,. \qquad (4.20)$$

When these potential conditions are valid, the stationary solution $P_s(\vec{\alpha})$ can be written as

$$\begin{aligned}P(\vec{\alpha})&=\exp\left(-\int^{\vec{\alpha}}d\alpha'_\mu v_\mu(\vec{\alpha}')\right)\\&=\exp[-\Phi(\vec{\alpha})]\,,\end{aligned} \qquad (4.21)$$

where $\Phi(\alpha)$ is referred to as the potential function. The complex P representation is not interpretable as a probability but, nevertheless, the existence of some exact solutions, where the corresponding positive P representation does not possess such solutions, can be of great practical utility.

D. Complex Poisson representation

We can similarly interpret the Fokker-Planck equation for chemical reactions (3.23) as analytic Fokker-Planck equations in the complex variables $\vec{\alpha}$, and again, in certain situations, potential solutions are possible. This is demonstrated in examples in Sec. VI.

V. EXAMPLES IN QUANTUM OPTICS

A. Two-photon absorption and dispersive optical bistability

We consider a coherently driven interferometer filled with an intracavity nonlinear medium. We shall treat cases in which

(a) the medium is a two-photon absorber,
(b) the medium has a nonlinear polarizability.

A Hamiltonian which describes both situations, by an appropriate choice of parameter values, is

$$\begin{aligned}H&=\sum_{j=1}^{5}H_j\,,\\H_1&=\hbar\omega_c a^\dagger a\,,\\H_2&=i\hbar(\epsilon e^{-i\omega_L t}a^\dagger-\epsilon^* e^{i\omega_L t}a)\,,\\H_3&=a\Gamma_c+a^\dagger\Gamma_c\,,\\H_4&=\hbar\chi'' a^{\dagger 2}a^2\,,\\H_5&=a^{\dagger 2}\Gamma_2+a^2\Gamma_2^\dagger\,.\end{aligned} \qquad (5.1)$$

For two-photon absorption, H_3 and H_4 vanish, while for dispersive optical bistability H_5 vanishes. These two cases have been treated in Refs. 35 and 38. Here H_1 describes the cavity mode a with frequency ω_c. H_2 describes the coupling with the coherent driving field with amplitude ϵ and frequency ω_L. H_3 describes the coupling to the cavity reservoirs $\Gamma_c, \Gamma_c^\dagger$. H_4 describes a nonlinear dispersive medium with nonlinear susceptability χ''. H_5 describes an intracavity two-photon absorber with reservoir operators $\Gamma_2, \Gamma_2^\dagger$.

In a reference system rotating at a frequency ω_L, the master equation for the density operator of the cavity field mode is obtained using standard techniques[15-17]:

$$
\begin{aligned}
&\frac{\partial \rho}{\partial t} = \sum_{j=1}^{5} \mathcal{L}_j(\rho)\,, \\
&\mathcal{L}_1(\rho) = -i\,\Delta\omega[a^\dagger a, \rho]\,, \\
&\mathcal{L}_2(\rho) = [\epsilon a^\dagger - \epsilon^* a, \rho]\,, \\
&\mathcal{L}_3(\rho) = \kappa'(2a\rho a^\dagger - \rho a^\dagger a - a^\dagger a \rho) + 2n_{\text{th}}([a,\rho]a^\dagger)\,, \\
&\mathcal{L}_4(\rho) = -i\,\chi''[a^{\dagger 2}a^2, \rho]\,, \\
&\mathcal{L}_5(\rho) = \chi'(2a^2\rho a^{\dagger 2} - \rho a^{\dagger 2}a^2 - a^{\dagger 2}a^2\rho)\,,
\end{aligned}
\tag{5.2}
$$

where $\Delta\omega = \omega_L - \omega_c$, κ' is the cavity relaxation, and χ' is the two-photon absorption rate, n_{th} is the thermal occupation number due to Gaussian fluctuations in the thermal reservoir for the cavity modes. This master equation may be transformed to the following Fokker-Planck equation via the generalized P representation[43]

$$
\begin{aligned}
\frac{\partial}{\partial t} P(\vec{\alpha}) = \bigg(&\frac{\partial}{\partial \alpha}[(\kappa + i\Delta\omega)\alpha + 2\chi\alpha^2\alpha^\dagger - \epsilon] \\
&+ \frac{\partial}{\partial \alpha^\dagger}[(\kappa - i\Delta\omega)\alpha^\dagger + 2\chi^*\alpha^{\dagger 2}\alpha - \epsilon^*] \\
&-\chi\frac{\partial^2}{\partial\alpha^2}\alpha^2 - \chi^*\frac{\partial^2}{\partial\alpha^{\dagger 2}}\alpha^{\dagger 2} + \Gamma_1\frac{\partial^2}{\partial\alpha\,\partial\alpha^\dagger}\bigg)P(\vec{\alpha})\,,
\end{aligned}
\tag{5.3}
$$

where $\chi = \chi' + i\chi''$, and $\Gamma_1 = 2\kappa' n_{\text{th}}$. This equation satisfies the potential conditions[18,46] and has the following steady-state solution:

$$
P(\alpha, \alpha^\dagger) = \alpha^{(c-2)}\alpha^{\dagger(c^*-2)}\exp\left[\frac{\epsilon}{\chi}\left(\frac{1}{\alpha} + \frac{1}{\alpha^\dagger}\right) + 2\alpha\alpha^\dagger\right]\,,
\tag{5.4}
$$

where $c = (\kappa + i\Delta\omega)/\chi$ and we have chosen the phase of the driving field so that ϵ/χ is real. The steady-state solution obtained above [Eq. (5.4)] with the generalized P representation has a completely different character from the Landau-Ginsberg type of distribution associated with thermal fluctuations. It can be seen immediately that the use of the Glauber-Sudarshan integration domain with $\alpha^\dagger = \alpha^*$ is not possible, since the distribution $P(\alpha, \alpha^*)$ would diverge for $|\alpha|^2 \to \infty$. Thus no steady-state Glauber-Sudarshan P function exists (except as a highly singular distribution). However, it is possible to evaluate steady-state moments usings the complex P representation, where the domain is chosen to have independent contours on the α and $\alpha^\dagger$ planes. If the variables are changed to

$$
\begin{aligned}
\bar{\alpha} &= \frac{1}{\alpha}\,, \\
\bar{\alpha}^\dagger &= \frac{1}{\alpha^\dagger}\,,
\end{aligned}
\tag{5.5}
$$

the expectation value of an arbitrary normal ordered moment of the field operator is given by

$$
\begin{aligned}
\langle (a^\dagger)^m a^n \rangle &\propto \int\int_C \bar{\alpha}^{-(n-c)}\,\bar{\alpha}^{\dagger(-m-c^*)}\exp\left(\frac{\epsilon(\bar{\alpha}+\bar{\alpha}^\dagger)}{\chi} + \frac{2}{\bar{\alpha}\bar{\alpha}^\dagger}\right) d\bar{\alpha}\,d\bar{\alpha}^\dagger \\
&\propto \int\int_C \sum_{j=0}^{\infty}\left(\frac{2^j}{j!}\right)\bar{\alpha}^{(-j-n-c)}\bar{\alpha}^{\dagger(-j-m-c^*)}\exp\left(\frac{\epsilon}{\chi}(\bar{\alpha}+\bar{\alpha}^\dagger)\right) d\bar{\alpha}\,d\bar{\alpha}^\dagger\,.
\end{aligned}
\tag{5.6}
$$

These integrals correspond to the definition of the gamma function. Hence it is appropriate to define each path of integration to be a Hankel path from $-\infty$ on the real axis around the origin in an anticlockwise direction and back to $-\infty$. The infinite series is the defining series for the generalized Gauss hypergeometric function ${}_0F_2$. Including the normalizing factor, the final result is

$$
\langle (a^\dagger)^m a^n \rangle = \left(\frac{\epsilon}{\chi}\right)^{m+n} \frac{\Gamma(c)\Gamma(c^*)\,{}_0F_2(m+c^*, n+c, |2\epsilon/\chi|)}{\Gamma(m+c^*)\Gamma(n+c)\,{}_0F_2(c^*, c\,|2\epsilon/\chi|)}\,.
\tag{5.7}
$$

The mean photon number $\langle a^\dagger a \rangle$ and the second-order correlation function $g^{(2)}(0) = \langle a^\dagger a^\dagger a a \rangle / \langle a^\dagger a \rangle^2$ are shown for dispersive bistability and two-photon absorption in Figs. 1 and 2, respectively. The semiclassical result for the photon number is shown for comparison on the same graphs. In both systems a $g^{(2)}(0) < 1$ characteristic of photon antibunching, a nonclassical feature of the electromagnetic field is displayed. These results require the use of the generalized P representation and cannot be obtained using the Glauber-Sudarshan P representation.

B. Comment on the use of the Q representation

By comparison, the use of the other representations (corresponding to different operator order-

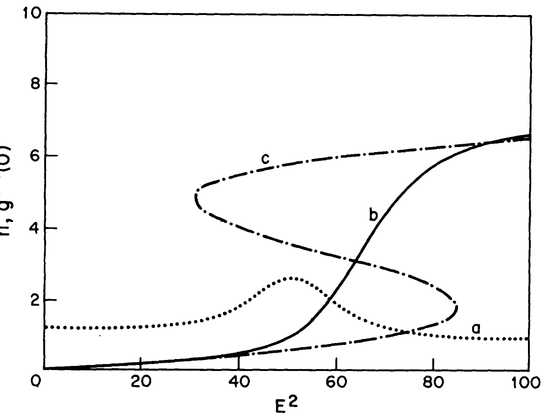

FIG. 1. Dispersive bistability, correlation, and photon number versus input power ($\kappa = 2.5 - i10$, $\chi' = 0$, $\chi'' = 1.0$, $\Gamma = 0$). (a) $g^{(2)}(0)$-exact, (b) $\bar{n}$-exact, (c) $\bar{n}$-semiclassical.

ings) is far from simple. If the Q representation is utilized, one would obtain the following equation:

$$\frac{\partial Q}{\partial t}(\alpha, {}^*\alpha) = \left\{ \frac{\partial}{\partial\alpha}(-\epsilon + \bar{\kappa}\alpha + 2\chi\alpha^2\alpha^*) + \frac{\partial^2}{\partial\alpha^2}\chi\alpha^2 + \kappa' \frac{\partial^2}{\partial\alpha\,\partial\alpha^*} + \chi' \frac{\partial^2}{\partial\alpha\,\partial\alpha^*}\left[4\left(\frac{\partial}{\partial\alpha}\alpha + \alpha\alpha^* - 1\right)\right] + \chi' \frac{\partial^4}{\partial^2\alpha\,\partial^2\alpha^*} + \text{c.c.} \right\} Q(\alpha, \alpha^*), \tag{5.8}$$

where $\bar{\kappa} = \kappa - 4\chi$.

In the case of dispersive bistability ($\chi' = 0$), the equation is nonpositive definite and does not satisfy detailed balance. Even worse, with two-photon absorption ($\chi' \neq 0$) these are fourth-order derivatives present.

This presents the following paradox—it is known that the solution of a time-evolution equation for an arbitrary positive initial condition is only positive if the evolution equation is of order two or less, or of infinite order.[47,48]

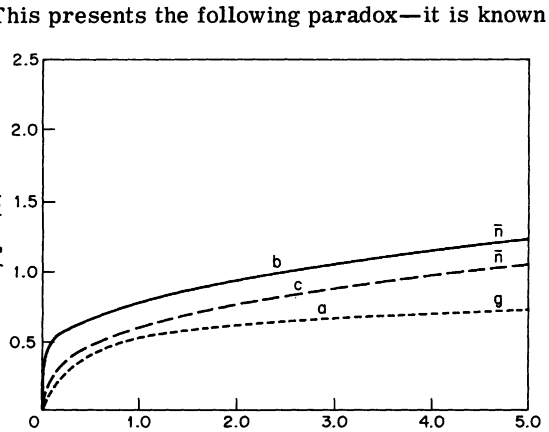

FIG. 2. Nonlinear absorber, correlation, and photon number versus input power ($\kappa' = 0.1$, $\chi' = 1.0$, $\chi'' = 0$, $\Gamma = 0$). (a) $g^{(2)}(0)$-exact, (b) $\bar{n}$-exact, (c) $\bar{n}$-semiclassical.

Therefore it is clear that there are solutions of Eq. (5.8) which are not positive, since fourth-order terms occur. This must be reconciled with the fact that Q is by construction positive. The reconcilation comes with the realization that a Q function of the form $\delta^2(\alpha - \alpha_0)$ is physically impossible (as ρ would not have the required positivity) and therefore it is not possible to choose an arbitrary positive initial condition. That is, only those initial conditions which have a certain minimum spread in α are permissible. Since Q is by construction explicitly positive,[49] the subset of solutions corresponding to such permissible initial conditions must be positive in spite of the fact that the equation is of fourth order.

C. Stochastic differential equations

Further results may be obtained by using the positive P representation to derive stochastic differential equations from the Fokker-Planck equation. The exact stochastic differential equations corresponding to the Fokker-Planck Eq. (5.3) in the Ito calculus are

$$\frac{d}{dt}\begin{pmatrix} \alpha \\ \alpha^\dagger \end{pmatrix} = \begin{pmatrix} \epsilon - \kappa\alpha - 2\chi\alpha^2\alpha^\dagger \\ \epsilon^* - \kappa^*\alpha^\dagger - 2\chi^*\alpha^{\dagger 2}\alpha \end{pmatrix} + \begin{pmatrix} -2\chi\alpha^2 & \Gamma_1 \\ \Gamma_1 & -2\chi^*\alpha^{\dagger 2} \end{pmatrix}^{1/2} \begin{pmatrix} \xi_1(t) \\ \xi_2(t) \end{pmatrix}. \tag{5.9}$$

Here $\epsilon_1(t)$, $\epsilon_2(t)$ are δ-correlated random Gaussian functions so that α, $\alpha^\dagger$ are complex conjugate in the mean. The stochastic differential equations provide a convenient starting point for calculating correlation functions via an asymptotic expansion. In particular, two time-correlation functions may be conveniently calculated via a linearized analysis of the stochastic differential equation. In a linearized fluctuation analysis the stochastic differential equation may be written in the form

$$\frac{d}{dt}\vec{\alpha}_1(t) = -\overleftrightarrow{\mathrm{A}}\vec{\alpha}_1(t) + [\overleftrightarrow{\mathrm{D}}(\alpha_0)]^{1/2}\vec{\xi}(t), \tag{5.10}$$

where $\overleftrightarrow{\mathrm{A}}$ is the linearized drift and $\overleftrightarrow{\mathrm{D}}$ is the diffusion array evaluated at $\vec{\alpha} = \vec{\alpha}_0$. The expression for the incoherent spectrum is then given by[32]

$$S(\omega_L + \omega) = \left(\frac{1}{2\pi}(\overleftrightarrow{\mathrm{A}} + \overleftrightarrow{\mathrm{I}}i\omega)^{-1}\overleftrightarrow{\mathrm{D}}(\overleftrightarrow{\mathrm{A}}^T - \overleftrightarrow{\mathrm{I}}i\omega)^{-1}\right)_{21}. \tag{5.11}$$

The resulting spectrum for dispersive bistability[38] on the upper branch is shown in Fig. 3. The symmetric spectrum results from pure quantum fluctuations ($\Gamma_1 = 0$) whereas the asymmetric spec-

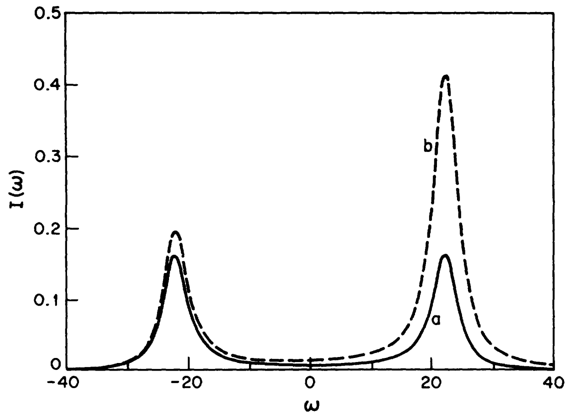

FIG. 3. Dispersive bistability spectrum, upper-branch region (arbitrary units). (a) Zero temperature and (b) finite temperature.

trum results when thermal fluctuations are also present.

VI. EXAMPLES IN CHEMICAL SYSTEMS

A. Schlögl's second-order phase transition model

Suppose we consider the rather well-studied second-order phase transition model[24, 27, 31] originated by Schlögl,[50]

$$\text{(a)}\quad A+X \underset{k_4}{\overset{k_2}{\rightleftharpoons}} 2X, \quad \text{(b)}\quad B+X \underset{k_3}{\overset{k_1}{\rightleftharpoons}} C, \tag{6.1}$$

for which it is easy to show[31] that the Fokker-Planck equation for the quasiprobability is

$$\frac{\partial f(\alpha,t)}{\partial t} = -\frac{\partial}{\partial\alpha}[\kappa_3 V + (\kappa_2-\kappa_1)\alpha - \kappa_4 V^{-1}\alpha^2]f(\alpha,t) + \frac{1}{2}\frac{\partial}{\partial\alpha}[2(\kappa_2\alpha - \kappa_4 V^{-1}\alpha^2)f(\alpha,t)]\ , \tag{6.2}$$

where

$$\kappa_3 V = k_3 C, \quad \kappa_2 = k_2 A, \quad \kappa_1 = k_1 B, \quad \kappa_4 V^{-1} = k_4, \tag{6.3}$$

A, B, C are the numbers of molecules of the corresponding chemical species, and V is the system volume. The steady-state solution to (6.2) is obviously obtained from the potential solution, since there is only one variable, and is able, and is

$$f_s(\alpha) = e^{\alpha}(\kappa_2 V - \kappa_4\alpha)^{V(\kappa_1/\kappa_4 - \kappa_3/\kappa_2)-1}\alpha^{(\kappa_3 V/\kappa_2 - 1)}\ . \tag{6.4}$$

If one uses the Poisson representation and derives (6.2) directly (using integration by parts), it is not difficult to show that Eq. (6.4) gives a steady-state solution of (6.2) provided (6.4) satisfies

$$\begin{aligned} &\text{I} \quad f_s(0) = 0\,, \\ &\text{II} \quad (\kappa_2\alpha - \kappa_4 V^{-1}\alpha^2)f_s(\alpha) = 0 \text{ when } \kappa_4\alpha = \kappa_2 V, \\ &\text{III} \quad \text{The range of } \alpha \text{ is } [0,\ \kappa_2 V/\kappa_4]\,. \end{aligned} \tag{6.5}$$

Condition I requires $\kappa_3/\kappa_2 > 1/V$ for its validity. We shall assume this in what follows. We now distinguish three situations depending on the value of $\delta = \kappa_1/\kappa_4 - \kappa_3/\kappa_2$. The quantity δ gives a measure of the direction in which the reaction system (6.1) is proceeding when a steady state exists. If $\delta > 0$, we find that, when X has its steady-state value, reaction (a) is producing X, while reaction (b) consumes X. When $\delta = 0$, both reactions balance separately —thus we have chemical equilibrium. When $\delta < 0$, reaction (a) consumes X, while reaction (b) produces X.

(I) $\delta > 0$. According to Eq. (6.5), this is the condition for $f(\alpha)$ to be valid quasiprobability on the real interval $[0, \kappa_2 V/\kappa_4]$. In this range, the diffusion coefficient $D(\alpha) = 2(\kappa_2 - \kappa_4\alpha^2/V) \geqslant 0$. The deterministic mean of α, given by

$$\alpha = \frac{V}{2\kappa_4}\{(\kappa_2 - \kappa_1) + [(\kappa_2 - \kappa_1)^2 + 4\kappa_3\kappa_4]^{1/2}\}\,, \tag{6.6}$$

lies within the interval $[0, \kappa_2 V/\kappa_4]$. We are therefore dealing with the case of a genuine Fokker-Planck equation and $f_s(\alpha)$ is a function vanishing at both ends of the interval, and peaked near the deterministic steady state.

(II) $\delta = 0$. Since both reactions now balance separately, we expect a Poissonian steady state. We note that $f_s(\alpha)$ in this case has a pole at $\kappa_2 V = \kappa_4\alpha$, and we choose the range of α to be a contour in the complex plane enclosing this pole. Since this is a closed contour, there are no boundary terms arising from partial integration, and $p_s(x)$ given by choosing this type of Poisson representation clearly satisfies the steady-state master equation. Using now the calculus of residues, we see that

$$p_s(x) = \frac{e^{-\alpha_0}\alpha_0^x}{x!}\ , \tag{6.7}$$

with $\alpha_0 = \kappa_2 V/\kappa_4$.

(III) $\delta < 0$. When $\delta < 0$ we meet some very interesting features. The steady-stage solution Eq. (6.4) now no longer satisfies the condition (6.5). However, if the range of α is chosen to be a contour C in the complex plane [Fig. 4(a)], and we employ the *complex Poisson representation*, it is not difficult to show that $p_s(x)$ constructed as

$$p_s(x) = \int_C d\alpha\, f_s(\alpha)\frac{e^{-\alpha}\alpha^x}{x!} \tag{6.8}$$

is a solution of the master equation. The deterministic steady state now occurs at a point on the real axis to the right of the singularity at $\alpha = \kappa_2 V/\kappa_4$, and asymptotic evaluations of means, moments, etc. may be obtained by choosing C to pass through

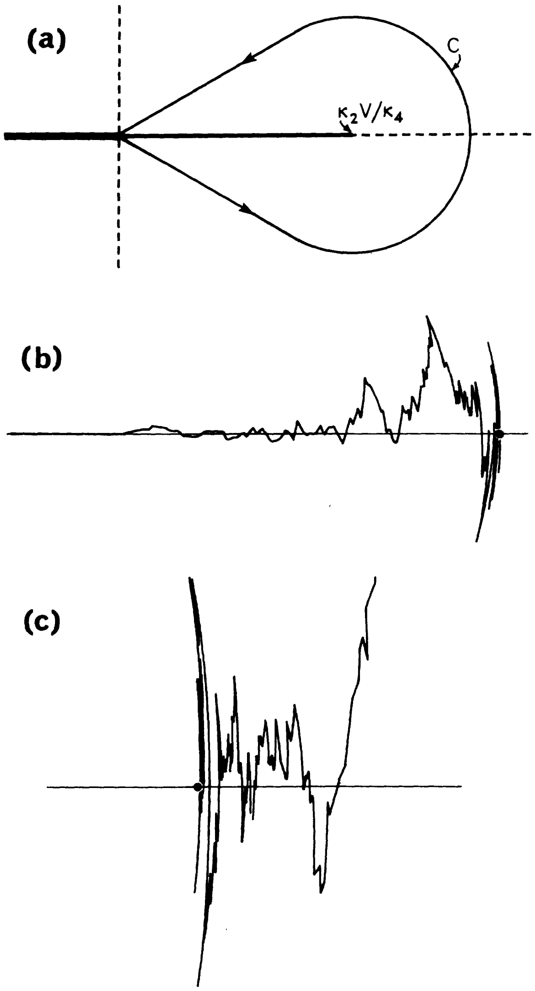

FIG. 4. (a) Curve C in the complex plane used for defining normalization and moments for the complex Poisson representation. (b) Reactions $A+X \rightleftarrows 2X$, $B+X \rightleftarrows C$. Simulated path of a point which starts initially at the left where the noise is real, and approaches a stationary point on the right, where it fluctuates with a negative variance. (c) Reactions $B\rightarrow X$, $2X\rightarrow A$. Point starts at top right, and eventually reaches equilibrium at center, fluctuating with negative variance.

the saddle point that occurs there. In doing so, one finds that the variance of α, defined as

$$\mathrm{var}(\alpha)=\langle\alpha^2\rangle-\langle\alpha\rangle^2\,, \tag{6.9}$$

is negative, so that

$$\mathrm{var}(x)=\langle x^2\rangle-\langle x\rangle^2=\langle\alpha^2\rangle-\langle\alpha\rangle^2+\langle\alpha\rangle<\langle\alpha\rangle\,. \tag{6.10}$$

This means that the steady state is narrower than the Poissonian. Finally, it should be noted that all three cases can be obtained from the contour C. In the case that $\delta=0$, the cut from the singularity at $\alpha=\kappa_2V/\kappa_4$ to $-\infty$ vanishes, and C may be distorted to a simple contour round the pole, while if $\delta>0$, the singularity at $\alpha=\kappa_2V/\kappa_4$ is now integrable, so the contour may be collapsed onto the cut, and the integral evaluated as a discontinuity integral over the range $[0,\kappa_2V/\kappa_4]$. (When δ is a positive integer, this argument requires modification.)

The use of the positive Poisson representation applied to this system yields the (*Itô*) stochastic differential equation

$$\frac{d\alpha}{dt}=\kappa_3V+(\kappa_2-\kappa_1)\alpha-\kappa_4V^{-1}\alpha^2+[2(\kappa_2\alpha-\kappa_4\alpha^2)]^{1/2}\xi(t)\,. \tag{6.11}$$

In the case $\delta>0$, we note that the noise term vanishes at $\alpha=0$, and $\alpha=V\kappa_2/\kappa_4$ is positive between these points, and the drift term is such as to turn α to the range $[0,V\kappa_2/\kappa_4]$ whenever it approaches the end points. Thus, for $\delta>0$, the Eq. (6.11) represents a real stochastic differential equation on the real interval $[0,V\kappa_2/\kappa_4]$.

In the case $\delta<0$, the stationary points lies outside the interval $[0,V\kappa_2/\kappa_4]$, and a point initially in this interval will move along this interval governed by Eq. (6.11) until it meets the right-hand end, where the noise vanishes, and the drift continues to drive it towards the right. On leaving the interval, the noise becomes imaginary, and the point will follow a path like that shown in Fig. 4(b) until eventually reaches the interval $[0,V\kappa_2/\kappa_4]$ again.

The case of $\delta=0$ is not very dissimilar, except that once the point reaches the right-hand end of the interval $[0,V\kappa_2/\kappa_4]$ both drift and diffusion vanish, so it remains there from then on.

B. Reaction with negative variance

Another interesting example is provided by the following reaction mechanism[23,28]:

$$B\xrightarrow{k_1}X\,,\quad 2X\xrightarrow{k_2}A \tag{6.12}$$

for which the Fokker-Planck equation is

$$\frac{\partial f(\alpha,t)}{\partial t}=\frac{-\partial}{\partial\alpha}\left((\kappa_1V-2\kappa_2V^{-1}\alpha^2)f(\alpha,t)+\frac{\partial}{\partial\alpha}[(\kappa_2V^{-1}\alpha^2)f(\alpha,t)]\right)\,, \tag{6.13}$$

where $\kappa_1V=k_1B$, $\kappa_2V^{-1}=k_2$. Note that the diffusion coefficient in the above Fokker-Planck equation is negative on all the real line.

The potential solution of (6.13) is

$$f(\alpha)=\alpha^{-2}\exp(2\alpha+aV^2/\alpha) \tag{6.14}$$

with $a=2\kappa_1/\kappa_2$ and the α integration is to be performed along a closed contour encircling the origin.

Thus, by putting $\alpha=\eta V$, we get

$$\langle x^r\rangle_f=\frac{V^r\oint dn\, e^{V(2\eta+a/\eta)}\eta^{r-2}}{\oint d\eta\, e^{V(2\eta+a/\eta)}\eta^{-2}}\,. \tag{6.15}$$

The function $(2\eta+a/\eta)$ does not have a maximum at the deterministic steady state. In fact, it has a minimum at the deterministic steady state $\eta=+(a/2)^{1/2}$. However, in the complex η plane this point is a saddle point and provides the dominant contribution to the integral.

Thus, the negative diffusion coefficient in (6.13) refelcts itself by giving rise to a saddle point at the determinate steady state, which results in the variance in X being less than $\langle X\rangle$.

From (6.15) all the steady-state moments can be calculated exactly. The results are

$$\langle X^r\rangle_f=\left[V\left(\frac{a}{2}\right)^{1/2}\right]^r\frac{I_{r-1}(2(2a)^{1/2}V)}{I_1(2(2a)^{1/2}V)}\,, \tag{6.16}$$

where $I_r(2(2a)^{1/2}V)$ are the modified Bessel functions. Using the large-argument expansion for $I_r(2(2a)^{1/2}V)$, we get

$$\langle X\rangle=V(a/2)^{1/2}+\tfrac{1}{8}+O(1/V)\,,$$

$$\langle X^2\rangle-\langle X\rangle^2=\tfrac{3}{4}V(a/2)^{1/2}-\tfrac{1}{16}+O(1/V)\,. \tag{6.17}$$

C. Stochastic differential equations

The stochastic differential equation corresponding to (6.13) is

$$\frac{d\eta}{dt}=\kappa_1-2\kappa_2\eta^2+i\epsilon(2\kappa_2)^{1/2}\eta\xi(t)\,, \tag{6.18}$$

where $\alpha=\eta V$ and $\epsilon=V^{-1/2}$. The stochastic differential equation (6.18) can be computer simulated, and a plot of motion in the complex η plane generated. Figure 4(c) illustrates the behavior. The point is seen to remain in the vicinity of $\mathrm{Re}(\alpha)=(\alpha/2)^{1/2}$, but to fluctuate mainly in the imaginary direction on either side, thus giving rise to a negative variance in α.

VI. DISCUSSION

We have shown how quasiprobability methods may be used to solve nonlinear problems in quantum optics and chemical reaction theory. For certain problems, e.g., those giving rise to nonclassical photon statistics in quantum optics of distributions narrower than the equilibrium distribution, the conventional methods fail completely. Here by introducing generalized representations which extend Fokker-Planck methods into the complex plane we are able to obtain steady-state quasiprobabilities from which expressions for the moments may be calculated. This has been demonstrated for the case of two-photon absorption and dispersive bistability in quantum optics and for models exhibiting a second-order phase transition and a negative variance in chemical reactions. Alternatively, a different integration measure may be chosen which ensures a positive $f(\alpha)$ or $P(\alpha,\beta)$ in the chemical or quantum optical case. By means of these representations we may define Fokker-Planck equations which have positive-semidefinite diffusion coefficients. This is particularly important for certain nonlinear processes where conventional representations lead to Fokker-Planck equations with non-positive-definite diffusion coefficients. Since we now have Fokker-Planck equations with positive-semidefinite diffusion, appropriate stochastic differential equations may be developed. These may be solved by asymptotic procedures or numerical simulation.

ACKNOWLEDGMENTS

D.F. Walls wishes to acknowledge the receipt of a J.I.L.A. fellowship and C. W. Gardiner wishes to thank Professor H. Haken and the Universität Stuttgart for the award of a visiting professorship.

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