Quantum-classical correspondences for atomic operators: A doubled atomic-space approach

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An operational method is presented that can avoid some of the difficulties associated with numerically simulating the Langevin operator equations for a dissipative quantum optical system. The approach is based on a set of quantum-classical correspondence rules that relate atomic operators in the Heisenberg picture to their corresponding double-dimensioned classical atomic variables. With this approach, the Langevin operator equations can be transformed into a doubled set of classical stochastic differential equations (SDE's). The relationship to the quantum regression theory up to the two-time correlation function and similarities to the positive P representation are discussed. As an illustrative example, we show that both the resonance fluorescence spectrum (Mollow spectrum) as well as the absorption spectrum for a two-level atom interacting with a near-resonance field can be simulated from the doubled set of classical SDE's without explicit reference to the quantum regression theorem.

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I. INTRODUCTION

One of the most important differences between quantum and classical descriptions of a physical system is due to the noncommuting (or commuting) nature of system variables [1–4]. Quantum mechanically, system variables are operators that in general do not commute with each other, while in classical physics, system variables are pure c numbers that commute with each other. This quantum character of noncommuting (consequently nonassociative) quantities has caused significant difficulties in the numerical solution of a problem, even though the quantum-mechanical formulation in the Heisenberg pictures results in exactly the same form of the equations for system variables as the classical system [4].

For a Hamiltonian system the above mentioned problem can sometimes be considered as purely conceptual, since in the Schrödinger representation, wave functions of the system are described by classical amplitudes (which are commuting). In fact, the difficulties in commuting are still hidden in the basis set itself since it possesses a much larger dimension than the degrees of freedom a classical description would require. This is where most of the action of semiclassical or classical studies of quantum-mechanical systems takes place [5].

However, there is another class of problems which are non-Hamiltonian, or open in the sense that dissipation of the system due to coupling with a reservoir (other trivial degrees of freedom) exists [1–4]. The study of the open systems utilizes standard quantum-statistical methods. Two formulations for the equations of motion of the system operators are frequently used. (i) In the Heisenberg picture, a set of Langevin operator equations can be derived for system operators. The form of these operator equations is virtually the same as the equations for the corresponding classical variables (now including noncommuting noise), which has the advantage of being conceptually simple. But the degrees of freedom of the reservoir are also inherent in Langevin operator equations. They show up in damping coefficients as well as in generically multiplicative noise terms. Noncommuting noise terms relate to the damping coefficient through the quantum fluctuation-dissipation theorem. Technically, no generic and reliable computational methods exist for solving Langevin operator equations because of the noncommuting nature. (ii) In the Schrödinger picture, a master equation for the density matrix operator can be derived by tracing away the degrees of freedom associated with the reservoir. This approach often proves to be more fruitful, particularly when numerical computations is required. The influence of the reservoir shows up only through a superoperator made up of all damping coefficients in the reduced Hilbert space. Explicit reference to the degrees of freedom of the reservoir is therefore impossible, even though their influence on the system variables is correctly accounted for with the help of the quantum regression theorem (QRT) [1–4].

Since we usually understand classical systems much better, it is quite often helpful to investigate quantum-classical correspondences. Through analogy, we hope to understand peculiar quantum behaviors from their classical counterparts [5–8]. The most commonly used techniques are based on various classical phase-space approaches, which by itself is an enormous subject field

[7–9]. In this paper, we present a set of quantum-classical correspondences for atomic operators describing general dissipative systems. The resulting phase space is made up of a dual set, which we call left and right space. This method, termed a doubled atomic-space approach by us, has as a resulting feature, a doubled set of variables for the atomic degrees of freedom. It is similar to the familar positive P classical phase-space technique [10–12]. The underlying philosophy is also very similar to the work on classical stochastic equations by Martin *et al.* [13]. However, unlike the positive-P method, we do not need to use the large-N approximation which is based upon the central limit theorem argument for the Gaussian statistics. (When considering propagation problems, we do assume that the noise from different coarse-grained volumes are uncorrelated so that Gaussian statistics should be valid.) Furthermore, we show that (again unlike the positive-P representation) both normal and antinormal ordered second-order two-time correlation functions can be calculated from the same doubled set of classical stochastic differential equations (SDE's). As an example, we demonstrate that both the resonance fluorescence spectrum (Mollow spectrum) and the absorption spectrum can be simulated from these doubled set of SDE's using classical statistical averages for products without explicit reference to the QRT.

The paper is organized as follows. In Sec. II we formulate and briefly review the Langevin operator equation approach for a model system composed of a multilevel atom interacting with a classical near-resonant field as well as a continuum spectrum vacuum electromagnetic field reservoir responsible for spontaneous emission. We use conventional quantum optics notation to illustrate the key points. In Sec. III we illustrate and prove the doubled space approach that we have developed for the quantum-classical correspondences. Connections with other relevant work, particularly the positive-P representation method, are discussed. In Sec. IV we demonstrate the technique by applying it to the calculation of the resonance fluorescence spectrum as well as absorption spectrum. We conclude in Sec. V. This work was developed during the course of attempting to formulate a quantum theory of the propagation of nonclassical light in a near-resonance medium. We will concentrate here on the result related to the atomic degrees of freedom. The extension of this approach to the propagating field is more difficult and will be briefly discussed in Sec. V.

II. FORMULATION AND LANGEVIN EQUATIONS

The Hamiltonian of the model can be expressed as

$$H_{\rm tot} = H_A + H_B + H_{\rm int}, \tag{1}$$

where H_A is the atomic (system) Hamiltonian and H_B is the free reservoir Hamiltonian. $H_{\rm int}$ is the interaction Hamiltonian, which includes both the coherent interaction of the atom with a given external field and the incoherent interaction of the atom with the vacuum fields.

The atomic Hamiltonian

$$H_A = \sum_{\mu} \hbar\omega_\mu \hat{\sigma}_{\mu\mu} \tag{2}$$

satisfies

$$H_A|\mu\rangle = \hbar\omega_\mu|\mu\rangle, \tag{3}$$

with $\hbar\omega_\mu$ and $|\mu\rangle$ being the eigenvalue and the eigenvector, respectively, of the electronic part of the atomic Hamiltonian. It has the familiar eigenrepresentation form with the projection operator

$$\hat{\sigma}_{\mu\nu}(t=0) = |\mu\rangle\langle\nu|. \tag{4}$$

They satisfy the following Lie algebra:

$$[\hat{\sigma}_{\mu\nu}(t), \hat{\sigma}_{\mu'\nu'}(t)] = \delta_{\nu\mu'}\hat{\sigma}_{\mu\nu'}(t) - \delta_{\nu'\mu}\hat{\sigma}_{\mu'\nu}(t). \tag{5}$$

The reservoir Hamiltonian H_B and the dipole interaction Hamiltonian $H_{\rm int}$ will not be explicitly written [2–4,7] since we will not give the details of the derivation. In the Heisenberg picture, Langevin operator equations can be derived for the atom projector operators $\hat{\sigma}_{\mu\nu}$ by eliminating the equations of motion for the reservoir degrees of freedom using the standard Bohn-Markov approximation [3,4]. For our system, we obtain the general form

$$\frac{d\vec{\hat{\sigma}}}{dt} = \mathcal{R}\vec{\hat{\sigma}} + \mathcal{P}\vec{\hat{\sigma}} + \vec{\hat{F}} \tag{6}$$

under the general constraint that

$$\sum_{\mu} \hat{\sigma}_{\mu\mu} = \hat{1}, \tag{7}$$

where we have defined the vectors

$$\vec{\hat{\sigma}} = (\hat{\sigma}_{\mu\nu}), \quad \vec{\hat{F}} = (\hat{F}_{\mu\nu}). \tag{8}$$

The quantum noise $\vec{\hat{F}}$ is in general noncommuting and is of vital importance is maintaining the quantum consistency requirement Eq. (5).

The matrices $\mathcal{R}$ (superoperator) and $\mathcal{P}$ describe respectively the rate (dissipative) part due to the interaction of the system with the reservoir and with the coherent classical field. Above, the stochastic differential equation is in the traditional Stratonovich sense [8,15]. If no additional assumption about the noise is made, even classically, Eq. (6) is very complicated due to the multiplicative nature of the noise terms. In the Markovian limit valid for this model, the Ito SDE form takes the same form as Eq. (6) [4].

Direct numerical simulation of the operator equation Eq. (6) is in general impossible because of the nonassociative nature of the noncommuting operator. Many studies have been directed toward this problem. Within the Heisenberg picture, one of particular importance is the so-called QRT, which is based on the Markovian assumption of the quantum noise and has been shown to allow for the calculation of multitime averages of products of operators [3,4] through quantum averages of Eq. (6) or the density matrix equation. However, there are distinct

advantages of using the Langevin operator equations directly for some problems [14]. It is desirable to overcome the numerical simulation problem with the Langevin operator equations approach. The adjoint equation approach developed by Gardiner [4] has proven to be very useful in this direction. In Sec. III we will present our doubled space operational method developed for this purpose. It may help to study the nonclassical light propagation in an active medium.

The density matrix elements

$$\rho_{\mu\nu} = \langle\langle \hat{\sigma}_{\nu\mu} \rangle\rangle \tag{9}$$

obey the equation

$$\frac{d\langle\langle \vec{\hat{\sigma}} \rangle\rangle}{dt} = \mathcal{R}\langle\langle \vec{\hat{\sigma}} \rangle\rangle + \langle\langle \mathcal{P}\vec{\hat{\sigma}} \rangle\rangle, \tag{10}$$

which can be obtained by averaging Eq. (6) over the reservoir. $\mathcal{P}$ is contained within the average as it may contain the reservoir coordinates. The quantum-mechanical average notation has the following interpretation:

$$\langle\langle \cdots \rangle\rangle = \mathrm{Tr}_{\mathrm{system+reservoir}}\{\cdots \hat{W}\} \tag{11}$$

with $\hat{W}$ the statistical operator for the whole system.

This gives us the first condition on the noise operators

$$\langle\langle \hat{F}_{\mu\nu} \rangle\rangle = 0. \tag{12}$$

The Markovian approximation amounts to the assumption

$$\langle\langle \hat{F}_{\mu\nu}(t)\hat{F}_{\mu'\nu'}(t') \rangle\rangle = 2D_{\mu\nu;\mu'\nu'}\delta(t-t'), \tag{13}$$

with diffusion coefficient $D_{\mu\nu;\mu'\nu'}$ and $\delta(t-t')$ to be understood to correspond to a time scale short compared with the time scale of the coherent driving $||\mathcal{P}||$ or the dissipation $||\mathcal{R}||$.

Due to the noncommuting nature of the noise, we do *not* require that

$$D_{\mu\nu;\mu'\nu'} \neq D_{\mu'\nu';\mu\nu}, \tag{14}$$

though the Hermitian conjugation requires that

$$D_{\mu\nu;\mu'\nu'} = (D_{\nu'\mu';\nu\mu})^*. \tag{15}$$

The diffusion coefficient $D_{\mu\nu;\mu'\nu'}$ can be calculated according to the generalized time-dependent Einstein relations (from the fluctuation-dissipation theorem) [2]. In particular, for any two projection operators $\hat{o}_L$ and $\hat{o}_R$ from $\vec{\hat{\sigma}}$, we have

$$\begin{aligned}\langle\langle 2D_{L;R}(t) \rangle\rangle &= \frac{d}{dt}\langle\langle \hat{o}_L(t)\hat{o}_R(t) \rangle\rangle - \left\langle\left\langle \left(\frac{d}{dt}\hat{o}_L(t)\right)\hat{o}_R(t) \right\rangle\right\rangle \\ &\quad - \left\langle\left\langle \hat{o}_L(t)\frac{d}{dt}\hat{o}_R(t) \right\rangle\right\rangle, \\ &= \frac{d}{dt}\langle\langle \hat{o}_L(t)\hat{o}_R(t) \rangle\rangle - \langle\langle (\mathcal{R}\vec{\hat{o}} + \mathcal{P}\vec{\hat{o}})_L\hat{o}_R(t) \rangle\rangle \\ &\quad - \langle\langle \hat{o}_L(t)(\mathcal{R}\vec{\hat{o}} + \mathcal{P}\vec{\hat{o}})_R \rangle\rangle. \end{aligned} \tag{16}$$

The evaluation is straightforward, as all the products of operators are evaluated at the same time and therefore can be contracted with the aid of the equal-time commutation relations Eq. (5).

For this generic model problem of an N-level atom interacting with a laser, a systematic method has been developed [6,7]. It was shown that, in general, if the noise can be written in the form

$$\vec{\hat{F}} = \mathcal{L}(t)\vec{\hat{\sigma}} + \vec{N}(t), \tag{17}$$

then the quantum-mechanical consistency requirement Eq. (5) gives

$$\begin{aligned} 2D_{\mu\nu;\mu'\nu'} = \sum_{\alpha\beta}(&\delta_{\nu\mu'}\mathcal{R}_{\mu\nu';\alpha\beta} - \delta_{\beta\nu'}\mathcal{R}_{\mu\nu;\alpha\mu'} \\ &- \delta_{\alpha\mu}\mathcal{R}_{\mu'\nu';\nu\beta})\langle\langle \hat{\sigma}_{\alpha\beta} \rangle\rangle. \end{aligned} \tag{18}$$

Since, in general,

$$\langle\langle \cdots \hat{o}_L(t')\hat{o}_R(t'')\cdots \rangle\rangle \neq \cdots \langle\langle \hat{o}_L(t') \rangle\rangle\langle\langle \hat{o}_R(t'') \rangle\rangle \cdots, \tag{19}$$

direct calculation of the operator products from the density matrix equation Eq. (10) is impossible. The QRT was developed to circumvent this problem. QRT states that the quantum fluctuations regress (i.e., evolve) with the same dynamics as those governing the evolution of averaged quantities, namely, the dynamics of the density matrix equations. Therefore the equations for the averages of the operator products obey the same homogeneous equations as the equations for the averages of the operators [density matrix equation Eq. (10)] [3]. The application of QRT to the calculation of second-order correlation functions for the case of a two-level atom will be presented in Sec. IV.

Higher-order moments are in general more complicated and all of them are required in order to solve for $\langle\vec{\sigma}\rangle$. However, depending on the observables to be evaluated, we may not want higher moments or can make a Gaussian assumption in some cases. For example, if we require only the calculation of the mean (density matrix) and second-order correlation functions of the above atomic operators

$$\rho_{\nu\mu} = \langle\langle \hat{\sigma}_{\mu\nu} \rangle\rangle, \quad \varphi_{\mu\nu;\mu'\nu'} = \langle\langle \hat{\sigma}_{\mu\nu}(t)\hat{\sigma}_{\mu'\nu'}(t') \rangle\rangle, \tag{20}$$

then the first-order and second-order correlation functions of the noise would be sufficient. Based on these observations, we develop, in the following section, an operational scheme that allows us to convert directly any Langevin operator equations of the form Eq. (6) into classical calculable equations represented by c-number quantities.

III. DOUBLED ATOMIC-SPACE APPROACH

We note that the difficulties of numerically simulating operator equation Eq. (6) are due to the noncommuting nature of the operators involved. Almost all the previously developed phase-space techniques choose certain ordering of operators to resolve the problem [4]. The

price to pay is that all the observables have to be converted into normal and time ordered form before the connection with the classical averages can be made.

For some problems, such as studying light propagation, we realize that actually for the observables of interest (i.e. intensity, spectrum, etc.) we only need to find the solutions for the averages and for the correlation functions. Hence only first- and second-order moments are required. We can formally solve the operator equation Eq. (6) with a Green's function approach. Simple analysis shows that in order to correctly generate first- and second-order moments, we have to generate correctly through classical methods only the first- and second-order moments of the noise $\hat{F}_{\mu\nu}$. The noncommuting nature can be handled by a doubling of the atomic space, when the classical stochastic equation is introduced. This is very similar to the approach taken with the positive-P representation [10–12] to avoid nonpositive-definite diffusion coefficients.

Key points are illustrated in the following. When the first-order moments of the atomic operator equations (density matrix equations) are evaluated, the white noise has zero mean. Therefore, any noise with zero mean can be used to within first order. When the stochastic average is performed, we get the correct results for the first-order moments of the atomic variables. However, no advantage is shown here because the equation is deterministic. When we also require that the second-order moments (the correlation functions) be obtained from the same stochastic equations using the stochastic quantity to represent the noise, we can get the correct results to second order. This is accomplished by making the following formal connections. We introduce

$$\langle\langle \hat{\sigma}_{\mu\nu} \longrightarrow (\sigma_{\mu\nu}|, \quad \hat{\sigma}_{\mu\nu}\rangle\rangle \longrightarrow |\sigma_{\mu\nu}). \tag{21}$$

We emphasize that the representation above is purely symbolic. $(\cdots|$ and $|\cdots)$ should be understood as representing different stochastic quantities in complex space (rather than the somewhat similar Dirac bra-ket notation for wave functions in the Hilbert space), such that

$$\begin{aligned} \langle\langle \hat{\sigma}_{\mu\nu}(t)\rangle\rangle &\Longrightarrow \langle(\sigma_{\mu\nu}(t)|\rangle_{\rm av} \\ &\Longrightarrow \langle|\sigma_{\mu\nu}(t))\rangle_{\rm av}, \\ \langle\langle \hat{\sigma}_{\mu\nu}(t)\hat{\sigma}_{\mu'\nu'}(t')\rangle\rangle &\Longrightarrow \langle(\sigma_{\mu\nu}(t)||\sigma_{\mu'\nu'}(t'))\rangle_{\rm av}, \end{aligned} \tag{22}$$

where $\langle\cdots\rangle_{\rm av}$ denotes a classical stochastic average over the noise statistics. Of course, in doing this we have abandoned the usual requirement that

$$|\sigma_{\mu\nu}) \neq [(\sigma_{\mu\nu}|]^*, \quad |\sigma_{\mu\mu}) \neq \text{real}, \quad (\sigma_{\nu\nu}| \neq \text{real}, \tag{23}$$

as in the positive-P representation. Correspondingly, we have represented the quantum noise by a left $(\vec{f}|$ and right $|\vec{f})$ stochastic noise and have also abandoned

$$|f_{\mu\nu}) \neq [(f_{\nu\mu}|]^*. \tag{24}$$

The noise $(\vec{f}|$ and $|\vec{f})$ satisfy

$$\begin{aligned} \langle|f_{\mu\nu})\rangle_{\rm av} &= \langle(f_{\mu\nu}|\rangle_{\rm av} = 0, \\ \langle(f_{\mu\nu}(t)||f_{\mu'\nu'}(t'))\rangle_{\rm av} &= 2D_{\mu\nu;\mu'\nu'}\delta(t-t'), \end{aligned} \tag{25}$$

while the rest of the first- and second-order moments can be of any form. In particular, they can be chosen to be of the associative type, i.e.,

$$\begin{aligned} \langle(f_{\mu\nu}|(f_{\mu'\nu'}|\rangle_{\rm av} &= \langle(f_{\mu\nu}|^2\rangle_{\rm av}, \\ \langle|f_{\mu\nu})|f_{\mu'\nu'})\rangle_{\rm av} &= \langle|f_{\mu\nu})^2\rangle_{\rm av}, \\ \langle|f_{\mu\nu}(t))(f_{\mu'\nu'}(t')|\rangle_{\rm av} &= \langle(f_{\mu'\nu'}(t')||f_{\mu\nu}(t))\rangle_{\rm av} \\ &= 2D_{\mu'\nu';\mu\nu}\delta(t-t'). \end{aligned} \tag{26}$$

We have found generic forms for both left and right spaced noise that satisfy the above conditions for any Langevin operator equations of the form of Eq. (6) [14].

Thus, as far as the first two moments of the atomic operators are concerned, the Langevin operator equation Eq. (6) can be cast into the doubled space form

$$\begin{aligned} \frac{d(\vec{\sigma}|}{dt} &= \mathcal{R}(\vec{\sigma}| + \mathcal{P}(\vec{\sigma}| + (\vec{f}|, \\ \frac{d|\vec{\sigma})}{dt} &= \mathcal{R}|\vec{\sigma}) + \mathcal{P}|\vec{\sigma}) + |\vec{f}), \\ \sum_{\mu}|\sigma_{\mu\mu}) &= \sum_{\nu}(\sigma_{\mu\mu}| = 1. \end{aligned} \tag{27}$$

The left and right space noise terms are given by the associative forms

$$\begin{aligned} (f_{\mu\nu}| &= \sum_{\mu_1\nu_1}\xi_{\mu\nu;\mu_1\nu_1}\sqrt{2D_{\mu\nu;\mu_1\nu_1}}, \\ |f_{\mu'\nu'}) &= \sum_{\mu_2\nu_2}\xi_{\mu_2\nu_2;\mu'\nu'}\sqrt{2D_{\mu_2\nu_2,\mu'\nu'}}. \end{aligned} \tag{28}$$

The (complex) diffusion coefficients $D_{\mu\nu;\mu'\nu'}$ are the same as those given in Eq. (18), except now with the classical average $\langle\cdots\rangle_{\rm av}$ in place of the quantum average $\langle\langle\cdots\rangle\rangle$. $\xi_{\mu\nu;\mu'\nu'}$ are independent real Wiener processes such that

$$\begin{aligned} \langle\xi_{\mu\nu;\mu'\nu'}\rangle_{\rm av} &= 0, \\ \langle\xi_{\mu\nu;\mu'\nu'}(t)\xi_{\mu_3\nu_3;\mu_4\nu_4}(t')\rangle_{\rm av} &= \delta_{\mu\mu_3}\delta_{\nu\nu_3}\delta_{\mu'\mu_4}\delta_{\nu'\nu_4}\delta(t-t'). \end{aligned} \tag{29}$$

Therefore, we have doubled our atomic space (real diagonal elements now turn into complex ones in addition to a doubling), such that now the atomic operator equations have been transformed into two sets of stochastic equations. The difficulties of the nonassociative nature is avoided as the diffusion coefficients $D_{\mu\nu;\mu'\nu'}$ and $D_{\mu'\nu';\mu\nu}$ describe the diffusion of completely different noise pairs and they do not have to be equal. However, they are still not completely classical. The left and right space noise terms given by Eq. (28) were constructed to reproduce Eq. (18). In terms of real and imaginary parts of the doubled space variable $(\vec{\sigma}|$ and $|\vec{\sigma})$, the doubled space diffusion matrix $D_{\mu\nu,\mu'\nu'}$ can still have negative elements [14] because intrinsically they represent the diffusion of nonclassical quantum stochastic processes, while a positive-P approach will result in a double-dimensioned diffusion matrix that is semipositive definite [10].

When a correlation function is calculated, we can simply draw from the two sets of trajectories of the stochastic

equations and the classical stochastic average will be correctly given. The QRT need not be stated and applied separately. The proof is sketched in the following. The formal solution can be written as

$$\begin{aligned}(\vec{\sigma}(t)| &= \exp[(\mathcal{R}+\mathcal{P})(t-t_0)](\vec{\sigma}(t_0)| \\ &\quad + \int_{t_0}^{t} dt_1 \exp[(\mathcal{R}+\mathcal{P})(t-t_1)](\vec{f}(t_1)|, \\ |\vec{\sigma}(t')) &= \exp[(\mathcal{R}+\mathcal{P})(t'-t_0)]|\vec{\sigma}(t_0)) \\ &\quad + \int_{t_0}^{t'} dt_2 \exp[(\mathcal{R}+\mathcal{P})(t'-t_2)]|\vec{f}(t_2)).\end{aligned} \tag{30}$$

Any other constant inhomogeneous terms on the right-hand side of Eq. (6) can be considered in a similar fashion as the noise terms. Since the classical noise has zero mean, it is obvious that the first-order moment (density matrix) equation is

$$\begin{aligned}\vec{\sigma}(t) &= \langle\langle \vec{\tilde{\sigma}}(t) \rangle\rangle \\ &= \left\langle \exp[(\mathcal{R}+\mathcal{P})(t-t_0)](\vec{\sigma}(t_0)| \right\rangle_{\text{av}} \\ &= \left\langle \exp[(\mathcal{R}+\mathcal{P})(t-t_0)]|\vec{\sigma}(t_0)) \right\rangle_{\text{av}} \\ &= \exp[(\mathcal{R}+\mathcal{P})(t-t_0)]\vec{\sigma}(t_0).\end{aligned} \tag{31}$$

The steady-state solution of the density matrix is

$$\vec{\sigma}^s = \lim_{t\to\infty} \vec{\sigma}(t) = \lim_{t\to\infty} \exp[(\mathcal{R}+\mathcal{P})(t-t_0)]\vec{\sigma}(t_0). \tag{32}$$

For the second-order moments, we obtain

$$\begin{aligned}\varphi_{\mu\nu;\mu'\nu'} &= \langle\langle \hat{\sigma}_{\mu\nu}(t)\hat{\sigma}_{\mu'\nu'}(t') \rangle\rangle \\ &= \vec{\sigma}^s_{\mu\nu}\vec{\sigma}^s_{\mu'\nu'} + d\varphi_{\mu\nu;\mu'\nu'},\end{aligned} \tag{33}$$

where

$$\begin{aligned}d\varphi_{\mu\nu;\mu'\nu'} &= \langle\langle (\hat{\sigma}_{\mu\nu} - \langle\langle \hat{\sigma}_{\mu\nu} \rangle\rangle)(\hat{\sigma}_{\mu'\nu'} - \langle\langle \hat{\sigma}_{\mu'\nu'} \rangle\rangle) \rangle\rangle \\ &\Longrightarrow \left(\left\langle \int_{t_0}^{t} dt_1 \exp[(\mathcal{R}+\mathcal{P})(t-t_1)](\vec{f}(t_1)| \int_{t_0}^{t'} dt_2 \exp[(\mathcal{R}+\mathcal{P})(t'-t_2)]|\vec{f}(t_2)) \right\rangle_{\text{av}} \right)_{\mu\nu;\mu'\nu'} \\ &= \int_{t_0}^{t} dt_1 \int_{t_0}^{t'} dt_2 \sum_{\mu_1\nu_1,\mu_2\nu_2} \left\{ \exp[(\mathcal{R}+\mathcal{P})(t-t_1)] \right\}_{\mu\nu;\mu_1\nu_1} \\ &\quad \times 2D_{\mu_1\nu_1;\mu_2\nu_2}\delta(t_1-t_2) \left\{ \exp[(\mathcal{R}+\mathcal{P})(t'-t_2)] \right\}_{\mu'\nu';\mu_2\nu_2}.\end{aligned} \tag{34}$$

Since all the diffusion coefficients and the noise-free density matrix equations are correctly reproduced, the above correlation function should be the correct one. In particular, this doubled space approach can reproduce all the second-order correlation functions, including those for the resonance fluorescence spectrum and the absorption spectrum. Note that we did not explicitly use the QRT. The Markovian nature of the noise, which is more fundamental, is built into the stochastic noise. It is worth noting that the doubled space approach developed here in fact reproduces diffusion coefficients at all times t even though we emphasized the physical picture of a stationary limit during the above proof. Therefore it is equally valid in time-dependent cases.

IV. TWO-LEVEL ATOM

In this section, we illustrate this method for the case of a simple two-level system pumped by a near-resonance field. Using the standard spin operator notation, the Langevin operator equations take the form

$$\begin{aligned}\frac{d\hat{S}_+}{dt} &= -\left(\frac{\gamma}{2} + i\delta_p\right)\hat{S}_+ - i\Omega_p\hat{S}_z + \hat{F}_+, \\ \frac{d\hat{S}_z}{dt} &= -\gamma\left(\frac{1}{2} + \hat{S}_z\right) + \frac{1}{2}i(\Omega_p\hat{S}_- - \Omega_p^*\hat{S}_+) + \hat{F}_z, \\ \frac{d\hat{S}_-}{dt} &= -\left(\frac{\gamma}{2} - i\delta_p\right)\hat{S}_- + i\Omega_p^*\hat{S}_z + \hat{F}_-,\end{aligned} \tag{35}$$

where γ is the spontaneous emission rate, Ω_p is the Rabi frequency of the pump field, and

$$\delta_p = \omega_p - \omega_{21} \tag{36}$$

is the detuning. The additional inhomogeneous term $-\frac{\gamma}{2}$ is responsible for the nonzero steady-state values. Since only radiative damping reservoir is included, the following relations can be shown:

$$\begin{aligned}\langle\langle \hat{F}_\mu \rangle\rangle &= 0, \qquad \mu \in (+,-,z) \\ \langle\langle \hat{F}_\mu(t)\hat{F}_\nu(t') \rangle\rangle &= 2D_{\mu\nu}\delta(t-t'), \quad \mu,\nu \in (+,-,z)\end{aligned} \tag{37}$$

with all nonzero diffusion coefficients listed below [3]:

$$\begin{aligned}2D_{-+} &= \gamma, \\ 2D_{-z} &= \gamma\langle\langle \hat{S}_- \rangle\rangle, \\ 2D_{z+} &= \gamma\langle\langle \hat{S}_+ \rangle\rangle, \\ 2D_{zz} &= \gamma\langle\langle \hat{S}_z + \tfrac{1}{2} \rangle\rangle.\end{aligned} \tag{38}$$

With our left and right space technique, we have the following doubled space classical SDE:

$$\frac{d(S_+|}{dt} = -\left(\frac{\gamma}{2} + i\delta_p\right)(S_+| - i\Omega_p(S_z| + (F_+|,$$
$$\frac{d(S_z|}{dt} = -\gamma\left(\frac{1}{2} + (S_z|\right) + \frac{1}{2}i[\Omega_p(S_-| - \Omega_p^*(S_+|] + (F_z|, \tag{39}$$
$$\frac{d(S_-|}{dt} = -\left(\frac{\gamma}{2} - i\delta_p\right)(S_-| + i\Omega_p^*(S_z| + (F_-|,$$

$$\frac{d|S_+)}{dt} = -\left(\frac{\gamma}{2} + i\delta_p\right)|S_+) - i\Omega_p|S_z) + |F_+),$$
$$\frac{d|S_z)}{dt} = -\gamma\left(\frac{1}{2} + |S_z)\right) + \frac{1}{2}i[\Omega_p|S_-) - \Omega_p^*|S_+)] + |F_z), \tag{40}$$
$$\frac{d|S_-)}{dt} = -\left(\frac{\gamma}{2} - i\delta_p\right)|S_-) + i\Omega_p^*|S_z) + |F_-),$$

and the associative noise ($(F_\mu|$ and $|F_\nu)$) can be made to satisfy the diffusion coefficients relations Eq. (25) according to Eq. (28). However, since the decomposition of the noise terms is by no means unique, for this specific model involving a two-level atom, we found a better choice (with fewer independent ξ's) for the doubled space noise terms as given below

$$\begin{aligned}
(F_+| &= 0,\\
(F_z| &= \sqrt{\gamma}\langle\hat{S}_+\rangle_{\rm av}\xi_1\\
&\quad + \sqrt{\gamma}\sqrt{\langle\hat{S}_z + \tfrac{1}{2}\rangle_{\rm av} - \langle\hat{S}_+\rangle_{\rm av}\langle\hat{S}_-\rangle_{\rm av}}\,\xi_2, \qquad (41)\\
(F_-| &= \sqrt{\gamma}\xi_1,
\end{aligned}$$

$$\begin{aligned}
|F_+) &= \sqrt{\gamma}\xi_1,\\
|F_z) &= \sqrt{\gamma}\langle\hat{S}_-\rangle_{\rm av}\xi_1\\
&\quad + \sqrt{\gamma}\sqrt{\langle\hat{S}_z + \tfrac{1}{2}\rangle_{\rm av} - \langle\hat{S}_+\rangle_{\rm av}\langle\hat{S}_-\rangle_{\rm av}}\,\xi_2, \qquad (42)\\
|F_-) &= 0,
\end{aligned}$$

where ξ_j are independent real Wiener processes such that

$$\langle\xi_j\rangle_{\rm av} = 0, \quad \langle\xi_j(t)\xi_{j'}(t')\rangle_{\rm av} = \delta_{jj'}\delta(t - t'). \tag{43}$$

Of course, direct physical interpretation should never go beyond the fact that they only reproduce up to second-order moments of all atomic projection operators Eq. (4) when classical stochastic averages are taken.

We have numerically solved the above two SDE's. Equations (39) and (40) are first written down in the standard Ito SDE form with Ito noise increments. We used a central difference algorithm developed in Refs. [12,14], which proved to be stable and accurate. The Ito noise terms $dW_j = \xi_j dt$ obey Gaussian statistics. They were generated using the Box-Muller method from a uniformly distributed random numbers of a pseudorandom number generator. Consistent with the analytical proof, all the second-order correlations functions were correctly generated through pure classical stochastic averages defined in Eq. (22). In the illustration given below, we will show calculations leading to the incoherent spectrum only, as the nonzero steady-state means is trivial. Basically we take the inhomogeneous term $-\frac{\gamma}{2}$ out of the equation for $\hat{S}_z$.

A. Resonance fluorescence spectrum

Define

$$\delta\hat{S}_\mu = \hat{S}_\mu - \langle\!\langle\hat{S}_\mu\rangle\!\rangle. \tag{44}$$

The resonance fluorescence spectrum is related to the Fourier transform of the normal ordered dipole operator correlation function $g_+^n(\tau)$, where

$$g_\mu^n(\tau) = \lim_{t\to\infty}\langle\!\langle\delta\hat{S}_\mu(t+\tau)\delta\hat{S}_-(t)\rangle\!\rangle. \tag{45}$$

QRT tells us that for $\tau > 0$, equations of motion for $g_\mu^n(\tau)$ take the same form as equations of motion for $\langle\!\langle\delta\hat{S}_\mu\rangle\!\rangle$. The initial conditions are obtained using the Lie algebra Eq. (5) to be

$$\begin{aligned}
g_+^n(0) &= \langle\!\langle\tfrac{1}{2} + \hat{S}_z\rangle\!\rangle - \langle\!\langle\hat{S}_+\rangle\!\rangle\langle\!\langle\hat{S}_-\rangle\!\rangle,\\
g_z^n(0) &= -\langle\!\langle\tfrac{1}{2} + \hat{S}_z\rangle\!\rangle\langle\!\langle\hat{S}_-\rangle\!\rangle, \qquad (46)\\
g_-^n(0) &= -\langle\!\langle\hat{S}_-\rangle\!\rangle^2.
\end{aligned}$$

The additional symmetry relation

$$g_+^n(-\tau) = [g_+^n(\tau)]^* \tag{47}$$

is used to obtain values for negative τ. We have plotted a typical calculation for $\Omega_p = 4\gamma$ and $\delta_p = 3\gamma$ in Fig. 1.

The resonance fluorescence spectrum (Mollow spectrum) is given by [16]

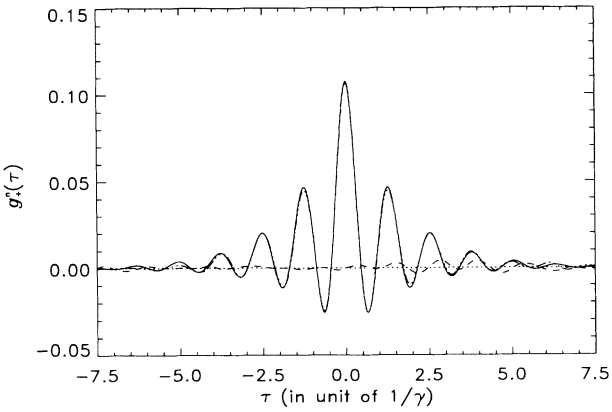

FIG. 1. Normal ordered autocorrelation functions for the dipole operator. Solid and dotted lines represent the real and the imaginary parts of the (exact) QRT solutions, respectively. Dash–triple-dotted and dashed lines denote the corresponding results from doubled space SDE simulations with 4000 sampling averages with $\Omega_p = 4\gamma$ and $\delta_p = 3\gamma$.

$$S^n(\omega-\omega_p)=\int_{-\infty}^{\infty}e^{i(\omega-\omega_p)\tau}g_+^n(\tau)d\tau. \tag{48}$$

This is shown in Fig. 2.

The classical ensemble averages Eq. (22) have been approximated by a time series average using the ergodic argument. To obtain statistical independence, different sampling windows centered around t_w separated by at least a few spontaneous emission times were taken and

$$g_+^n(\pm\tau)=\langle(\delta S_+(t_w\pm\tau)||\delta S_-(t_w))\rangle_{\rm av}. \tag{49}$$

We compute here the averages for both positive and negative τ from the same set of classical SDE's. Comparison with the quantum calculation is shown in Fig. 1.

With the solutions of the full doubled space SDE, we can calculate the resonance fluorescence spectrum by using the definition of Eq. (48). Within this scheme, the stationarity of the correlation functions are assumed. We can also calculate the spectrum utilizing both trajectory sequences from left and right spaced quantities. Then, we have

$$\begin{aligned}S^n(\omega-\omega_p)\Longrightarrow&\langle(\delta S_+(\omega-\omega_p)||\delta S_-(\omega_p-\omega))\rangle_{\rm av}\\=&\int_{-\infty}^{\infty}e^{i(\omega-\omega_p)\tau}d\tau\int_{-\infty}^{\infty}e^{-i(\omega-\omega_p)\tau'}d\tau'\\&\times\langle(\delta S_+(\tau)||\delta S_-(\tau'))\rangle_{\rm av},\end{aligned} \tag{50}$$

which is related to the definition Eq. (48) through Wiener-Khintchine theorem for a stationary process [2]. [Note that the limits of time integration ($\pm\infty$) are replaced by finite values corresponding to the width of the sampling window in numerical samplings.] It turns out that this scheme of calculation gives better agreement with quantum results, presumably because more random numbers are being sampled, as the whole time-dependent trajectory $|\delta S_-(\tau'))$ has been utilized rather than only at t_w (one point) according to Eq. (49). Comparisons are made in Fig. 2.

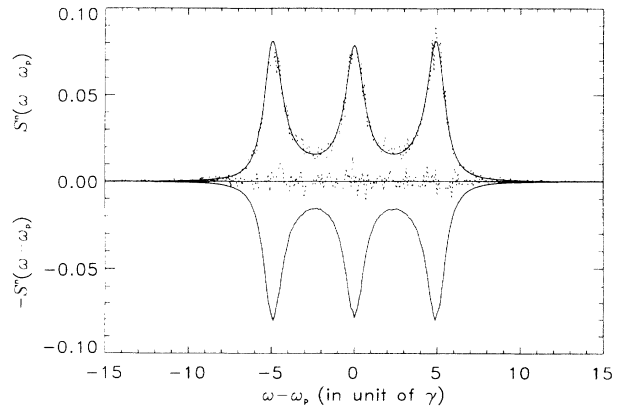

FIG. 2. Resonance fluorescence spectrum (in arbitrary units). Solid lines represent both real and imaginary parts of the spectrum as obtained from the QRT solution curve (with imaginary part being zero). Dotted lines represent the corresponding results from the doubled space SDE solution. The inverted solid spectrum line is calculated from Eq. (50) with negligible imaginary parts and statistical errors undisplayed.

B. Absorption spectrum

The weak probe absorption spectrum is related to [17]

$$S^a(\omega-\omega_p)=\int_{-\infty}^{\infty}e^{i(\omega-\omega_p)\tau}[g_-^a(\tau)-g_+^n(\tau)]d\tau, \tag{51}$$

where antinormal ordered correlation functions

$$g_\mu^a(\tau)=\lim_{t\to\infty}\langle\langle\delta\hat{S}_\mu(t+\tau)\delta\hat{S}_+(t)\rangle\rangle \tag{52}$$

again can be calculated using QRT for $\tau\geq 0$. They obey the same equations of motion as for $\langle\langle\delta\hat{S}_\mu\rangle\rangle$. This time, the initial conditions are

$$\begin{aligned}g_+^a(0)&=-\langle\langle\hat{S}_-\rangle\rangle^2,\\g_z^a(0)&=\langle\langle\tfrac{1}{2}-\hat{S}_z\rangle\rangle\langle\langle\hat{S}_+\rangle\rangle,\\g_-^a(0)&=\langle\langle\tfrac{1}{2}-\hat{S}_z\rangle\rangle-\langle\langle\hat{S}_-\rangle\rangle\langle\langle\hat{S}_+\rangle\rangle.\end{aligned} \tag{53}$$

The correlation function $g_-^a(\tau)$ is compared with classical calculations in Fig. 3, while the spectrum $S^a(\omega-\omega_p)$ is compared in Fig. 4. The classical calculations were obtained in exactly the same fashion as outlined in Eqs. (49) and (50).

We found that, as expected, all second- (as well as first-) order moments were correctly reproduced through the doubled space approach given above. As an additional comment, we note that the specific form of doubled space noise we chose in Eqs. (41) and (42) possesses additional symmetry, namely, we note

$$|F_+)=[(F_-|]^*,\quad |F_z)=[(F_z|]^*,\quad |F_-)=[(F_+|]^*. \tag{54}$$

Under the substitution

$$|S_+)\Longrightarrow[(S_-|]^*,\quad |S_z)\Longrightarrow[(S_z|]^*,\quad |S_-)\Longrightarrow[(S_+|]^*, \tag{55}$$

equations for the right spaced quantities Eq. (40) are exactly the complex conjugate of Eq. (39). What this implies is that for this particular choice of classical noise

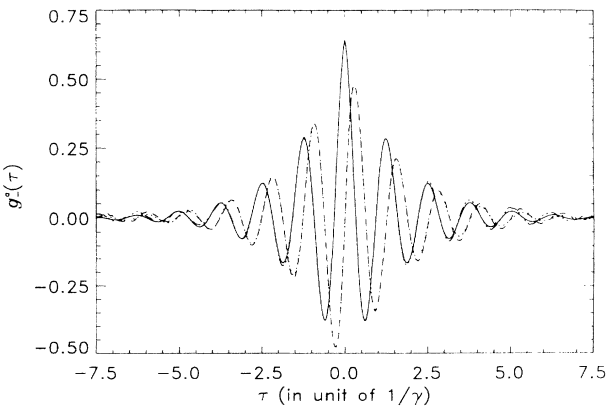

FIG. 3. Antinormal ordered autocorrelation functions for the dipole operator with same notation of lines and parameters as in Fig. 1. With 4000 averages, it is hard to tell the differences between the results from the QRT and classical doubled space SDE simulations.

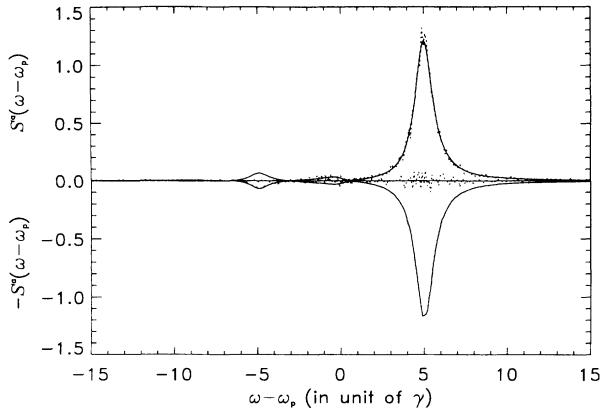

FIG. 4. Same as Fig. 2, except for the absorption spectrum.

as given by Eqs. (41) and (42), only solving one set of the doubled set of SDE's would provide all the necessary information. [Note this is not true for the generic noise terms of Eq. (28). The choice of different noise representations is not unique. Equations (41) and (42) are a very special case that is not of the form given by Eq. (28), even though they also reproduce all diffusion coefficients. Of course, using Eq. (28) gives exactly the same results, even though they involve more random variables.] In fact, this is what we did for the calculations illustrated in Figs. 1–4. Basically, we have

$$\langle(\delta S_+(t)||\delta S_-(t'))\rangle_{\rm av} = \left\langle(\delta S_+(t)||[(\delta S_+(t')||]^*\right\rangle_{\rm av},$$
$$\langle(\delta S_-(t)||\delta S_+(t'))\rangle_{\rm av} = \left\langle(\delta S_-(t)||[(\delta S_-(t')||]^*\right\rangle_{\rm av}. \quad (56)$$

V. CONCLUSIONS

In summary, we have developed quantum-classical correspondences for atomic operators describing general dissipative systems. This method, termed a doubled atomic-space approach has, as a resulting feature, a doubled set of variables for the atomic degrees of freedom, similar to the familar positive-P classical phase-space technique. However, unlike the positive-P method (in the large-N limit with a Gaussian approximation), both normal and antinormal ordered second-order two-time correlation functions can be calculated from the same set of classical stochastic differential equations. We have shown that both the resonance fluorescence spectrum (Mollow spectrum) as well as the absorption spectrum can be simulated from the same set of classical SDE's without explicit reference to the QRT.

Langevin operator equations rather than the traditional method of going from a master equation have been used. The doubled space equations have classical noise terms that can be obtained from the diffusion coefficients of the noncommutating Langevin noise terms. For a generic atomic system with coherent state driving fields, we have derived generic noise terms for the quantum-classical correspondence. Furthermore, the mathematical work involved is significantly simpler than what is usually required for the derivation of the correspondence rules with the classical phase-space technique for atoms.

Different from the traditional phase-space techniques, this doubled space approach works as long as the diffusion coefficients for the noise operators in the Langevin operator equations can be found. We obtained the classical SDE's without the use of a large-N approximation (Gaussian approximation). Therefore the method works for a single atom, as well as a collection of many atoms. While in the traditional phase-space technique, SDE's can only be obtained through a large-N expansion and by neglecting terms of order $1/N$ or higher. Effectively, this means SDE's from traditional phase-space techniques (with the help of Fokker-Planck equations) only work if the stochastic motion is of much smaller amplitude than classical (or semiclassical) mean motions. This is the reason why in the traditional phase-space SDE's, diffusion coefficients are of order $1/N$. But with our approach, this restriction is lifted.

Another point that is worth mentioning is that simulation of SDE's obtained with traditional phase-space techniques has always been very difficult because of the extreme singularities associated with some characteristic functions, even though their noise terms are $1/\sqrt{N}$ smaller than the mean motion. However, with our doubled space approach, which is based on a purely operational observation, we found it to be numerically stable and it converged much faster despite the fact that the noise being considered here is of the same order as the mean motion. We illustrate in Fig. 5 convergence properties of the classical stochastic averaged quantities with increasing number of samplings. Throughout the simulation for this work, we did not experience any divergent trajectories. This is in sharp contrast to the positive-P method [12]. We believe one of the reasons for this is that the atom spectrum calculations presented here rely on simulating linear SDE's Eqs. (39) and (40) rather than traditional nonlinear SDE problems encounted in the positive-P method. In fact, we can prove that, with the central difference algorithm implementation, no divergent trajectories would ever occur from Eqs. (39) and (40), as successive multipliers have norms all less than unity due to damping terms (negative real parts for all three eigenvalues associated with matrix $\mathcal{R}+\mathcal{P}$) [14].

We have also solved the same problem considered here with the traditional positive-P method [14]. The SDE's obtained have forms exactly the same as Eqs. (39) and (40), except that the noise terms have correlations different from those in Eqs. (41) and (42) [14]. Numerically, we found that for the parameters considered, solutions from the positive-P SDE's converge at least a factor of 10 slower than the doubled space SDE's developed in this work. This is illustrated in Fig. 6, which shows the results of the positive-P SDE solutions (with $N = 100$) under the same conditions as used for Fig. 5, but with 10 times more samplings. We have also solved the positive-P SDE's with noise terms corresponding to $N = 1$ (exactly the same as the doubled space work devel-

oped here). With an additional factor of 10 times more samplings, we still could not obtain convergent results comparable to those displayed in Fig. 5.

As remarked earlier, this work was developed during the course of unified theoretical work on the propagation of nonclassical light in a near-resonance medium. Under a slowly varying amplitude approximation, the usual Langevin operator coupled Maxwell-Bloch equations can be also extended to doubled space classical SDE's by including the following equations for fields (both left and right spaced quantities as well):

$$\left[\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\pm i\frac{1}{2k_p}\nabla_\perp^2\right](\Omega_p^\pm(\vec{r},t)| = i\frac{3\pi}{k_p^2}\gamma\mathcal{N}(\vec{r})(\vec{S}_\pm(\vec{r},t)|, \tag{57}$$

$$\left[\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\pm i\frac{1}{2k_p}\nabla_\perp^2\right]|\Omega_p^\pm(\vec{r},t)) = i\frac{3\pi}{k_p^2}\gamma\mathcal{N}(\vec{r})|\vec{S}_\pm(\vec{r},t)),$$

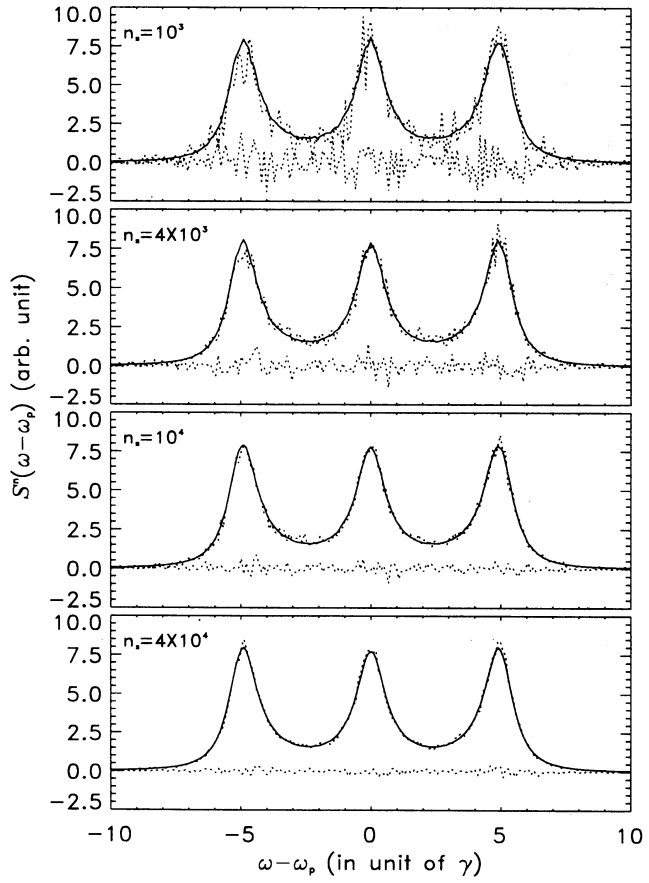

FIG. 5. Resonance fluorescence spectrum with different samplings n_s as given in the figure. Solid lines represent the results calculated according to Eq. (50), which has negligible error for $n_s = 4 \times 10^4$. Dotted lines represent both real and imaginary parts of results calculated from Fourier transform of the autocorrelations according to Eq. (48).

where $\mathcal{N}(\vec{r})$ is the density of the two-level atomic medium and $k_p = \omega_p c$ is the wave vector of the propagating near-resonance wave. Notice that

$$(\Omega_p^\pm(\vec{r},t)| \neq [|\Omega_p^\mp(\vec{r},t))]^*, \tag{58}$$

although the averages are equal just as in the case of the atomic variables. Correspondingly, in the left spaced atom SDE Eq. (39), we have to make the substitutions

$$\Omega_p \longrightarrow (\Omega_p^+(\vec{r},t)|, \quad \Omega_p^* \longrightarrow (\Omega_p^-(\vec{r},t)|, \tag{59}$$

with similar substitutions for right spaced fields in Eqs. (40). This extension to the field reproduces the correlation functions correctly also only up to the second order, while the propagation formulation in principle requires all orders of the field correlations. Note, however, that the quantities on the right-hand sides of Eqs. (57) are coarse-grained atom dipole variables. When there are a large number of atoms within the slowly varying amplitude volume, the central limit theorem would indicate that Gaussian statistics should be an excellent approximation. We intend to investigate further the propagational in a future work [18].

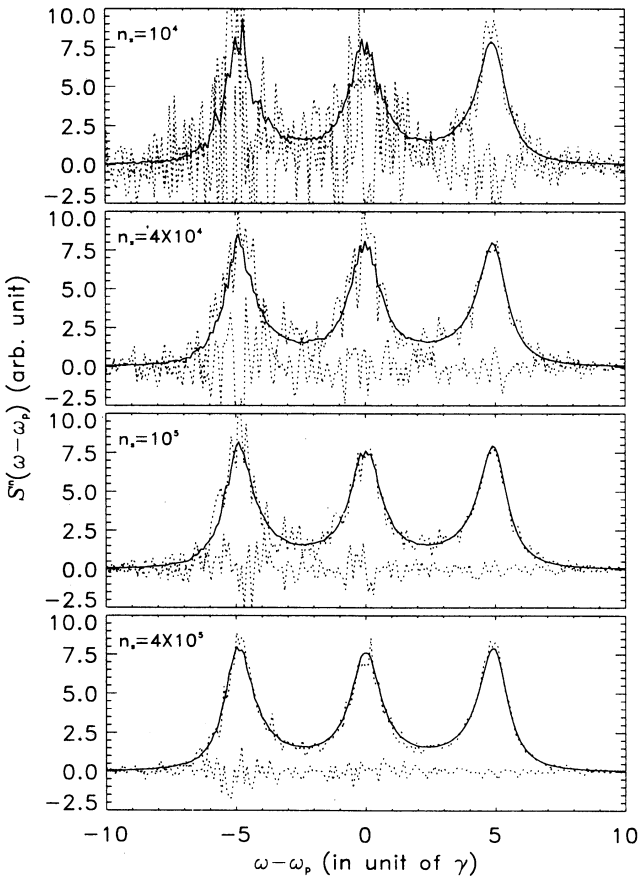

FIG. 6. Same as Fig. 5, but calculated from positive P SDE (for N=100) with 10 times more samplings.

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