Spontaneous generation of phase waves and solitons in stimulated Raman scattering: Quantum-mechanical models of stimulated Raman scattering

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We use the dressed-state representation to generate the full quantum-mechanical treatment of stimulated Raman scattering (SRS). Equations of motion and propagation are obtained from the Heisenberg equations of motion in the plane-wave limit and under conditions such that anti-Stokes and higher-order Stokes processes can be neglected. Equations for a stochastic model, based upon antinormal ordering and a quantum-classical correspondence are developed from them, suitable for numerical ensemble calculations for the study of amplified quantum fluctuations and spontaneously generated phase waves and solitons. Several alternative approaches, which have more general validity but present greater difficulty in numerical implementation, are also developed by introduction of quasiprobability functions and their associated Fokker-Planck equations from which corresponding stochastic equations of motion and propagation are derived. The Q representation, whose moments correspond to expectation values of products of antinormally ordered fields, and normally ordered atomic operators yield stochastic equations identical to the equations for the stochastic model developed directly from the Heisenberg equations for the same initial and boundary conditions, giving strong credence to the stochastic model. The Wigner representation, whose moments correspond to expectation values of symmetrically ordered operators, leads to a set of equations that are valid where the stochastic model fails, namely, where conditions are such that quantum fluctuations and nonlinearity are simultaneously important. The stochastic model, which is shown to be, by far, the simplest to implement in numerical calculations, is shown also to be valid for the conditions of SRS.

I. INTRODUCTION

One usually thinks of a Raman-active molecule as providing a mechanism by which incident pump energy is transferred to Stokes energy. As with any charged oscillator, however, the absorption and emission of radiation is conditioned by the phase relationship that exists among the polarizability of the material medium and the modes of the radiation field. Should this relationship be disturbed, it is possible for the molecule to instead become an absorber of Stokes energy and an emitter of pump energy. This condition would then persist until a dephasing process (here a collision with another molecule) restored the condition of pump loss and Stokes gain. Hence the relative phase of the pump, Stokes, and molecular oscillations would exhibit a disturbance—a *phase wave*—corresponding to a local peak in the pump pulse and a trough in the Stokes pulse.

As we have shown,[1] such features can be generated spontaneously during stimulated Raman scattering (SRS) by large, abrupt phase shifts that arise in the Stokes vacuum during quantum initiation. Moreover, the nonlinear dynamics of SRS is such that these features are preserved well after the Stokes pulse has built to a macroscopic scale. Thus it is that macroscopic peaks of temporal width approximately equal to the mean collision time are sporadically observed in an otherwise depleted pump pulse. Because of the relative stability of these solitary waves, we speak of them as *spontaneously generated Raman solitons.*

Amplified quantum-mechanical temporal and spatial fluctuations in nonlinear light-matter interactions have been a subject of intense interest for many years. Interest in superfluorescence (SF), the phenomenon that can occur when a collection of atoms or molecules is prepared in an initial state of complete inversion and then allowed to undergo relaxation by collective, spontaneous decay,[2] has stemmed mainly from the ability to observe and analyze large-scale fluctuations of quantum origin. The fluctuations originate in the quantum-initiation regime and are subsequently amplified in the SF pulse buildup. Both experimental[3] and theoretical[2] efforts in this area have been extensive. A related area of current interest has been the statistical characterization of the SF to amplified spontaneous emission (ASE) transition region.[4]

A phenomenon closely related to SF, but which has received somewhat less attention, is swept-gain superfluorescence[5,6] (SGSF). Unlike SF, SGSF is a spatially asymptotic condition that occurs when the inversion of the medium is prepared from the ground state by a constant, sharp-excitation π pulse traveling at the velocity of light in the medium. If the gain-to-loss ratio is greater than unity, a solitary pulse is generated at a large propagation distance, which follows the pump pulse with peak delay and temporal width less than the dephasing time of the medium. These solitary pulses are direct analogs of SF pulses,[7] but are distinct from them in that SF pulses evolve in a medium which is initially inverted. They are, however, generated from quantum noise in the manner that SF pulses are generated. Unlike the solitons of SRS to be discussed in this paper, SGSF solitary pulses do not originate from phase waves.

It was first noted by Raymer *et al.*[8] that the process of quantum initiation in SRS is identical to that for SF, and pulse-energy fluctuations stemming from this process have been measured and analyzed by Walmsley *et al.*[9] The formal equivalence of the three phenomena, SF, SGSF, and SRS, in the quantum initiation and linear region of pulse buildup, as well as their distinct macroscopic and statistical characteristics within the respective nonlinear regimes of subsequent pulse amplification, has been discussed recently by Bowden and Englund.[10]

The importance of phase, as well as amplitude, fluctuations in SF quantum initiation was first pointed out and discussed by Hopf,[11] who demonstrated that quantum phase fluctuations are the direct cause of rarely occurring phase waves, which are generated by macroscopic, local, abrupt shifts of the phase of the SF pulse; the effects on SF pulse distortion, statistical variation, and delay time statistics were analyzed in detail.[11] Due to the equivalence of SF, SGSF, and SRS in the quantum initiation and linear region of pulse buildup, the phase-wave phenomenon is intrinsic to each. The mechanisms of phase-wave generation are fully operative in the statistical treatments and analyses of SF pulse generation and evolution;[2] however, the experimental data were never analyzed to specifically identify the effects of phase waves. Furthermore, the effects were clearly within the experimental uncertainties, and thus phase waves in SF have borne no clear signature.

The effects of phase waves in SGSF were thoroughly analyzed by Hopf and Overman[12] using the semiclassical Maxwell-Bloch formalism[13] in the plane-wave limit, in which quantum initiation formally proceeds from radiation reaction associated with the decay of the state of complete inversion prepared by the traveling excitation. They predict strong, episodic, spatial variations in the total pulse energy due to phase waves. Their results indicate that the process is pulse degrading, and precludes the existence of an asymptotic steady state in SGSF generated from quantum noise. Although the signature of phase waves (episodic pulse breakup in detected pulses from shot to shot) is quite evident in SGSF as predicted by Hopf and Overman,[12] no experiments, to our knowledge, have been carried out to explicitly analyze this particular manifestation of amplified quantum fluctuations. These processes are also expected to be operative in the SF-to-ASE transition region,[10] and phase waves are expected to significantly affect the statistical variation of pulse width, height, and delay time as well as pulse breakup. A complete analysis of these effects as well as the full treatment of effects of amplified quantum fluctuations in this regime using a fully quantum-mechanical approach is currently in progress,[14] and will be the subject of a future publication.[15]

It was shown recently by Englund and Bowden,[1,9] using a fully quantum-mechanical development, that Raman solitons can be generated from quantum noise and depend entirely upon the amplification of quantum phase fluctuations in a pump-field-induced, spontaneously generated Stokes field. Raman solitons appear as partial or nearly complete repletions in the pump field depletion zone, of temporal width comparable to the collisional dephasing time in the Raman medium, and are accompanied by corresponding dark solitons in the Stokes field. They established a one-to-one correspondence between the occurrence of a Raman soliton and a phase wave generated from quantum phase fluctuations. The stochastic properties of spontaneous soliton generation in terms of system parameters and phase wave statistics has been further discussed by Englund and Bowden,[16–18] as well as the fundamental relationship among amplified quantum fluctuations in SF, SGSF, and spontaneous Raman soliton generation[10,19,20] (SRSG). The distinguishing feature of SRS lies in how the phase waves affect the nonlinear regime to induce solitons.

Spontaneously induced Raman solitons are a unique, robust, macroscopic manifestation of quantum phase fluctuations which are the direct result of the effect of phase waves in the amplified nonlinear regime of SRS. The study of the stochastic properties of these solitons and their generation is therefore useful for analyzing effects of amplified quantum fluctuations, and in this case, specifically quantum phase fluctuations. Quite recently, spontaneously generated Raman solitons have been observed in hydrogen by Carlsten and MacPherson,[21] and stochastic properties have been studied experimentally.[22–24] Preliminary comparison has shown strong qualitative and encouraging quantitative agreement between their results and our predictions.[24] It is anticipated that these results are the vanguard of vigorous activity in this relatively new and important area of investigation.

Although soliton solutions to the classical equations of SRS have been known for more than a decade,[25] it has been only recently that solitons have been observed in SRS experiments, where Drühl, Wenzel, and Carlsten[26] used CO_2 pumped parahydrogen with a Stokes "seed" and observed occasional solitary pulses as nearly complete repletions in the depletion zone of the pump pulse. The frequency of occurrence of the solitary pulses appeared to be stochastic, suggesting the origin as noise generated. Solitary pulses were produced deterministically in subsequent experiments[27] by inducing a π phase change in the Stokes "seed" prior to its entry into the Raman gain cell.

The experimental results were simulated numerically[27] using the classical SRS equations[25] with initial and boundary conditions of the experiments. Previous theoretical treatments[25] had dealt mainly with existence and characterization of solitons in SRS, but had left indefinite the initial and boundary conditions that cause them. The experimental and theoretical analysis of Drühl, Carlsten, and Wenzel demonstrated sufficient conditions for the generation of Raman solitons deterministically. Later, Ackerhalt and Milonni[28] discussed a variety of induced soliton effects, including second Stokes and anti-Stokes generation and the use of two-pump Raman scattering to study solitons in four-wave mixing. Recently, Bowden and Englund have related Raman solitons to possible utilization in spectroscopy.[29]

Some rather straightforward considerations can illustrate the effect of an independent local phase change in the Stokes field amplitude A_S relative to the pump field amplitude A_L in SRS. If ground-state depletion is neglected, the usual set of classical SRS nonlinear coupled equations for A_S, A_L, and the polarization R are[25,26]

$$\partial_\zeta A_L = K_{LS} R A_S \ , \tag{1}$$

$$\partial_\zeta A_S = -K_{LS}^* R^* A_L \ , \tag{2}$$

$$\partial_\tau R = -\gamma R - N K_{LS}^* A_S^* A_L \ , \tag{3}$$

where N is the number of molecules, K_{LS} is the complex coupling constant, ζ is the coordinate in the direction of propagation, τ is retarded time, $\tau = t - \zeta/c$, and γ is the collisional dephasing rate. (The equations used in Refs. 1 and 16 may be obtained by making the substitutions $R \to -R^*$ and $K_{LS} \to K_{LS}^*$.) For the condition of strong coherence decay, R may be adiabatically eliminated. Then Eq. (3) results in the relation

$$R(\zeta,\tau) = -\frac{N K_{LS}^*}{\gamma} A_S^*(\zeta,\tau) A_L(\zeta,\tau) \ , \tag{4}$$

which fixes the relative phase of the complex amplitudes, and Eqs. (1) and (2) yield

$$\partial_\zeta |A_L|^2 = -\frac{N|K_{LS}|^2}{\gamma} |A_S|^2 |A_L|^2 \tag{5}$$

and

$$\partial_\zeta |A_S|^2 = \frac{N|K_{LS}|^2}{\gamma} |A_L|^2 |A_S|^2 \ . \tag{6}$$

Clearly, pump loss and Stokes gain are insensitive to phase changes in this adiabatic limit.

To analyze the effects of phase fluctuations, it is convenient to impose the following transformation:

$$P = A_L A_S^*, \quad Q = |A_L|^2 - |A_S|^2 \ . \tag{7}$$

When Eqs. (7) are used in Eqs. (1)–(3), the result is

$$\partial_\zeta P = -K_{LS} R Q \ , \tag{8}$$

$$\partial_\zeta Q = 2 K_{LS} R P^* + \text{c.c.} \ , \tag{9}$$

$$\partial_\tau R = -\gamma R - N K_{LS}^* P \ . \tag{10}$$

Note that the form of Eqs. (8) and (9) is identical to that for the resonant optical Bloch equations in the absence of relaxation and dephasing,[13] coupled to the linearized Maxwell equation with linear loss, Eq. (10), but with the roles of spacelike and timelike variables interchanged. Here, P plays the analogous role of polarization and Q plays the role of inversion, whereas R plays the role of the Maxwell field. These equations can be transformed to a sine-Gordon equation which is well known to have soliton solutions in the asymptotic regime.[5] The transformation (7) applied to Eqs. (1)–(3) has been used in connection with the introduction of the Bäcklund transformation for the wave-wave scattering problem to obtain one- and two-soliton solutions.[30] Of course, the constant of propagation $I = |A_L|^2 + |A_S|^2$ is preserved in the transformation (7) and in Eqs. (8)–(10).

Effects of relative phase change become apparent if the variables in Eqs. (8)–(10) are written in terms of amplitude and phase,

$$P = |P| e^{i\phi_P}, \quad R = |R| e^{i\phi_R}, \quad K_{LS} = |K_{LS}| e^{i\phi_K} \ . \tag{11}$$

Then, in terms of the relative phase, Eq. (9) takes the form

$$\partial_\zeta Q = -4 |K_{LS}|^2 |P| |R| \cos\phi \ , \tag{12}$$

where $\phi \equiv \pi + \phi_K + \phi_R - \phi_P$ is the relative phase. For the conditions that led to (5) and (6), $\phi = 0$, and Eq. (12) again expresses the condition of pump loss and Stokes gain. However, if ϕ is a stochastic variable, or if a change in ϕ is arbitrarily induced of sufficient magnitude, the sign can change on the right-hand side (r.h.s.) of Eq. (12), thus causing a reverse of the role of gain-to-loss between pump and Stokes intensities:

$$0 \le |\phi| \le \pi/2 \ , \tag{13a}$$

$$\pi/2 < |\phi| < \pi \ , \tag{13b}$$

where (13a) represents pump loss and Stokes gain, whereas (13b) represents Stokes loss and pump gain.

Now, one can suppose that if a phase flip in the range $\pi/2 < |\phi| < \pi$ occurs within the dephasing time γ^{-1} (which results in a temporal and spatial instability in the system), the response within the coherence time γ^{-1} will be a transition, temporally and spatially, to the most stable condition. Since soliton solutions of Eqs. (8)–(10) are well known,[25,30] it is expected that a solitary pulse will result, i.e., repletion of the pump intensity in its depletion zone with a corresponding dip in the Stokes intensity. This is precisely what is observed in numerical experiments on the classical Eqs. (1)–(3), with a sharp, induced phase flip in the Stokes amplitude.[27] The results of Ref. 27 fit the corresponding experimental data very well, where the phase flip was induced deterministically by passing the Stokes "seed" through a Pockel's cell between crossed polarizers and applying a sudden switch in the voltage across the cell.[27]

Such pulses were generated stochastically from quantum phase fluctuations in numerical experiments of quantum SRS by Englund and Bowden[1,9] and recently experimentally observed.[21–24] (We shall refer to these solitary pulses as "solitons" in keeping with the use of this term in the literature.) The purpose of this report is to lay a foundation for modeling this phenomenon; results will be described in a companion paper.

The quantum-mechanical model is presented in Sec. II in terms of the dressed-state representation. The Heisenberg equations of motion are derived and presented in Sec. III and a stochastic model, useful for numerical calculations for the dynamical evolution of the system, is derived from them. In Sec. IV we present several alternative approaches by introducing quasiprobability functions and obtaining their Fokker-Planck equations. We obtain the associated stochastic differential equations and compare the various approaches. A summary of the results and concluding remarks are presented in the final section. Results calculated from the stochastic model presented in Sec. III will be given and analyzed in a future paper.[31]

II. MODEL

Consider first a system of N identical, stationary, noninteracting molecules each possessing an energy spectrum $\hbar\omega_i$ $(i=1,2,\ldots,\infty)$ corresponding to the eigenstates $|i\rangle$. Introducing the projection operators $\sigma_n^{ij}\equiv|i\rangle\langle j|$, where the subscript n distinguishes individual molecules, we may write the Hamiltonian of the system as

$$H_A=\hbar\sum_{n=1}^{N}\sum_{i=1}^{\infty}\omega_i\sigma_n^{ii} \,. \tag{14}$$

We quantize the free, plane-wave, electromagnetic field within a volume V of length L in the direction of propagation z. Normal modes of wave number $k_\lambda=2\pi\lambda/L$ $(\lambda=1,2,\ldots,\infty)$ and frequency $\omega_\lambda=k_\lambda c$ we assume to be polarized in the direction specified by the unit vector $\hat{\epsilon}_\lambda$. (Since angular momentum conservation enters in a trivial way into the problem treated here, we do not label photon helicities, just as we have not included angular momentum quantum numbers in our description of the molecules.) In terms of the "electric field per photon,"

$$\mathbf{e}_\lambda\equiv\left[\frac{2\pi\hbar\omega_\lambda}{V}\right]^{1/2}\hat{\epsilon}_\lambda \,, \tag{15}$$

we may then expand the electric-field operator in terms of creation $(a_\lambda^\dagger)$ and annihilation (a_λ) operators, operating on the Fock space $|\{n_\lambda\}\rangle$, as

$$\mathbf{E}(z,t)=\mathbf{E}^{(+)}(z,t)+\mathbf{E}^{(-)}(z,t) \,, \tag{16}$$

where

$$\mathbf{E}^{(+)}(z,t)=i\sum_\lambda \mathbf{e}_\lambda a_\lambda(t)e^{ik_\lambda z} \tag{17}$$

and $\mathbf{E}^{(-)}\equiv[\mathbf{E}^{(+)}(z,t)]^\dagger$. The free field then possesses the Hamiltonian

$$H_F=\hbar\sum_{\lambda=1}^{\infty}\omega_\lambda a_\lambda^\dagger a_\lambda \,. \tag{18}$$

We now allow the molecules and field to interact, at sites z_n, in the electric dipole approximation. Introducing the molecular dipole operator $\mathbf{d}$ and assuming identical matrix elements $\mathbf{d}_{ij}\equiv\langle i|\mathbf{d}|j\rangle$ for individual molecules, we obtain the interaction potential

$$H_{AF}=-\mathbf{d}\cdot\mathbf{E} = -\hbar\sum_{\lambda=1}^{\infty}\sum_{n=1}^{N}\sum_{i=1}^{\infty}\sum_{j=1}^{\infty} g_{ij}^{\lambda n}\sigma_n^{ij}a_\lambda+\text{H.c.} \,, \tag{19}$$

where

$$g_{ij}^{\lambda n}\equiv\frac{i}{\hbar}(\mathbf{d}_{ij}\cdot\mathbf{e}_\lambda)e^{ik_\lambda z_n} \,. \tag{20}$$

The Hamiltonian of the coupled system is thus

$$H=H_A+H_F+H_{AF} \,. \tag{21}$$

We identify the states $|1\rangle$ and $|2\rangle$ with the pumped Raman transition, with the associated frequency $\omega\equiv\omega_2-\omega_1$. Accordingly, we set $\mathbf{d}_{12}=0$. In addition, we assume that these states possess no permanent electric dipole moment (as in a symmetric diatomic molecule) and take $\mathbf{d}_{11}=\mathbf{d}_{22}=0$.

The free-molecule–free-field eigenstates $|\{i\}\{n_\lambda\}\rangle$ clearly do not constitute the most appropriate basis for describing the evolution generated by (21), which couples the pumped transition to the other levels. Because the coupling is weak, however, it is reasonable to construct perturbatively a unitary transformation of H to a dressed-state Hamiltonian H' for which $|\{i\}\{n_\lambda\}\rangle$ are once again eigenstates.[32] Doing so, and retaining only terms driven by the pump, we obtain (see Appendix A)

$$\begin{aligned}H'=\tfrac{1}{2}\hbar\omega\sum_{n=1}^{N}\sigma_n^z+\hbar\sum_{\lambda=1}^{\infty}\omega_\lambda a_\lambda^\dagger a_\lambda &-\tfrac{1}{2}\hbar\sum_{n=1}^{N}\sum_{\lambda=1}^{\infty}\sum_{\mu=1}^{\infty}e^{-i(k_\lambda-k_\mu)z_n}\kappa_{\lambda\mu}^z\sigma_n^z a_\lambda^\dagger a_\mu \\ &-\hbar\sum_{n=1}^{N}\sum_{\lambda=1}^{\infty}\sum_{\mu=1}^{\infty}(e^{-i(k_\lambda-k_\mu)z_n}\kappa_{\lambda\mu}\sigma_n^- a_\lambda^\dagger a_\mu+\text{H.c.}) \,,\end{aligned} \tag{22}$$

where $\sigma_n^+\equiv\sigma_n^{21}$, $\sigma_n^-\equiv\sigma_n^{12}$, $\sigma_n^z\equiv\sigma_n^{22}-\sigma_n^{11}$, and where the constants $\kappa_{\lambda\mu}$ and $\kappa_{\lambda\mu}^z$ are, respectively, the Raman and self-energy transition rates

$$\kappa_{\lambda\mu}\equiv\frac{1}{\hbar^2}\delta_{\omega_\lambda,\omega_\mu+\omega}\sum_{i=3}^{\infty}\left[\frac{(\mathbf{e}_\lambda^*\cdot\mathbf{d}_{1i})(\mathbf{d}_{i2}\cdot\mathbf{e}_\mu)}{\omega_{i1}-\omega_\lambda}+\frac{(\mathbf{e}_\mu\cdot\mathbf{d}_{1i})(\mathbf{d}_{i2}\cdot\mathbf{e}_\lambda^*)}{\omega_{i2}+\omega_\lambda}\right] \tag{23}$$

and

$$\kappa_{\lambda\mu}^z\equiv\frac{1}{\hbar^2}\delta_{\omega_\lambda,\omega_\mu}\sum_{i=3}^{\infty}\left[\left[\frac{(\mathbf{e}_\lambda^*\cdot\mathbf{d}_{2i})(\mathbf{d}_{i2}\cdot\mathbf{e}_\mu)}{\omega_{i2}-\omega_\lambda}+\frac{(\mathbf{e}_\mu\cdot\mathbf{d}_{2i})(\mathbf{d}_{i2}\cdot\mathbf{e}_\lambda^*)}{\omega_{i2}+\omega_\lambda}\right]-\left[\frac{(\mathbf{e}_\lambda^*\cdot\mathbf{d}_{1i})(\mathbf{d}_{i1}\cdot\mathbf{e}_\mu)}{\omega_{i1}-\omega_\lambda}+\frac{(\mathbf{e}_\mu\cdot\mathbf{d}_{1i})(\mathbf{d}_{i1}\cdot\mathbf{e}_\lambda^*)}{\omega_{i1}+\omega_\lambda}\right]\right] \,. \tag{24}$$

(The summations over i exclude values for which the denominators vanish.)

The Hamiltonian (22) admits both Stokes and anti-Stokes transitions. We neglect the latter. This is valid if only a small fraction of molecules occupy level 2 (negligible ground-state depletion), or if the transition rate $\kappa_{\lambda\mu}$ is considerably larger for the Stokes transition. We assume that the pump and Stokes wave numbers are grouped closely about average values k_L and k_S, respectively, with (assuming negligible dispersion) $\omega_L \equiv k_L c$ and $\omega_S \equiv k_S c$, and have polarizations $\hat{\epsilon}_L$ and $\hat{\epsilon}_S$. Adopting a convention in which pump (Stokes) modes are labeled by $\lambda, \lambda', \ldots$ $(\mu, \mu', \ldots)$, we approximate $\kappa_{\lambda\mu} \approx K_{LS} e_\lambda e_\mu$, $\kappa^z_{\lambda\lambda'} \approx K_{LL} e_\lambda e_{\lambda'}$, and $\kappa^z_{\mu\mu'} \approx K_{SS} e_\mu e_{\mu'}$, where

$$K_{LS} \equiv \frac{1}{\hbar^2} \sum_{i=3}^{\infty} \left[\frac{(\hat{\epsilon}_L^* \cdot \mathbf{d}_{1i})(\mathbf{d}_{i2} \cdot \hat{\epsilon}_S)}{\omega_{i1} - \omega_L} + \frac{(\hat{\epsilon}_S \cdot \mathbf{d}_{1i})(\mathbf{d}_{i2} \cdot \hat{\epsilon}_L^*)}{\omega_{i2} + \omega_L} \right], \tag{25}$$

$$K_{LL} \equiv \frac{1}{\hbar^2} \sum_{i=3}^{\infty} \left[\left[\frac{|\mathbf{d}_{i2} \cdot \hat{\epsilon}_L|^2}{\omega_{i2} - \omega_L} + \frac{|\mathbf{d}_{2i} \cdot \hat{\epsilon}_L|^2}{\omega_{i2} + \omega_L} \right] - \left[\frac{|\mathbf{d}_{i1} \cdot \hat{\epsilon}_L|^2}{\omega_{i1} - \omega_L} + \frac{|\mathbf{d}_{1i} \cdot \hat{\epsilon}_L|^2}{\omega_{i1} + \omega_L} \right] \right], \tag{26}$$

and

$$K_{SS} \equiv \frac{1}{\hbar^2} \sum_{i=3}^{\infty} \left[\left[\frac{|\mathbf{d}_{i2} \cdot \hat{\epsilon}_S|^2}{\omega_{i2} - \omega_S} + \frac{|\mathbf{d}_{2i} \cdot \hat{\epsilon}_S|^2}{\omega_{i2} + \omega_S} \right] - \left[\frac{|\mathbf{d}_{i1} \cdot \hat{\epsilon}_S|^2}{\omega_{i1} - \omega_S} + \frac{|\mathbf{d}_{1i} \cdot \hat{\epsilon}_S|^2}{\omega_{i1} + \omega_S} \right] \right]. \tag{27}$$

We replace the Stokes operators $a_\mu \to b_\mu$ for clarity and finally obtain

$$H' = \tfrac{1}{2}\hbar\omega \sum_{n=1}^{N} \sigma_n^z + \hbar \sum_{\lambda} \omega_\lambda a_\lambda^\dagger a_\lambda + \hbar \sum_{\mu} \omega_\mu b_\mu^\dagger b_\mu - \tfrac{1}{2}\hbar K_{LL} \sum_{n=1}^{N} \sum_{\lambda} \sum_{\lambda'} e^{-i(k_\lambda - k_{\lambda'})z_n} e_\lambda e_{\lambda'} a_\lambda^\dagger a_{\lambda'} \sigma_n^z$$
$$- \tfrac{1}{2}\hbar K_{SS} \sum_{n=1}^{N} \sum_{\mu} \sum_{\mu'} e^{-i(k_\mu - k_{\mu'})z_n} e_\mu e_{\mu'} b_\mu^\dagger b_{\mu'} \sigma_n^z - \hbar \left[K_{LS} \sum_{n=1}^{N} \sum_{\lambda} \sum_{\mu} e^{-i(k_\lambda - k_\mu)z_n} e_\lambda e_\mu a_\lambda^\dagger b_\mu \sigma_n^- + \text{H.c.} \right]. \tag{28}$$

One element remains to be included in the model. As mentioned in the Introduction, incoherent dephasing plays an essential role in soliton generation. In the experiments of Carlsten *et al.*[21–24,26–27] this is provided by molecular collisions characterized by the rate γ. We adopt the standard phenomenological model, in which each molecule is weakly coupled to an infinite reservoir through elastic processes. This adds a non-Hamiltonian term

$$L(\rho) \equiv \frac{\gamma}{4} \sum_{n=1}^{N} ([\sigma_n^z \rho, \sigma_n^z] + [\sigma_n^z, \rho \sigma_n^z]), \tag{29}$$

resulting in the Liouville equation

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H', \rho] + L(\rho) \tag{30}$$

for the density operator ρ. Equation (30) is the starting point for Sec. IV. In Sec. III we obtain the equivalent result by using the fluctuation-dissipation theorem.[33]

III. HEISENBERG REPRESENTATION

In this section we use the dressed-state Hamiltonian (28) to formulate dynamical equations for SRS, and in the process, add phenomenological terms describing collisional dephasing. Our goal is to use the linear solutions to these equations to justify the use of a stochastic model whose mean values correspond to the quantum-mechanical expectation values of a particular ordering of operators. We base our argument on the assumptions that quantum sources of noise are relevant to soliton generation only during initiation, and that, in this regime, the dynamical equations may be linearized about initial (boundary) values. We will examine the validity of this procedure more closely in Sec. IV, where stochastic differential equations are derived from Fokker-Planck equations within the Schrödinger representation.

A. Equations and initial, boundary conditions

We now obtain Heisenberg equations from the Hamiltonian H', adding fluctuation $[g_n^-(t)]$ and dissipation $[-\gamma\sigma_n^-(t)]$ terms to $\dot{\sigma}_n^-(t)$ in order to model dephasing processes. This results in the following:

$$\dot{a}_\lambda = -i\omega_\lambda a_\lambda + \tfrac{1}{2} i K_{LL} \sum_{n=1}^{N} \sum_{\lambda'} e^{i(k_{\lambda'} - k_\lambda)z_n} e_\lambda e_{\lambda'} \sigma_n^z a_{\lambda'} + i K_{LS} \sum_{n=1}^{N} \sum_{\mu} e^{i(k_\mu - k_\lambda)z_n} e_\lambda e_\mu \sigma_n^- b_\mu, \tag{31}$$

$$\dot{b}_\mu = -i\omega_\mu b_\mu + \tfrac{1}{2} i K_{SS} \sum_{n=1}^{N} \sum_{\mu'} e^{i(k_{\mu'} - k_\mu)z_n} e_\mu e_{\mu'} \sigma_n^z b_{\mu'} + i K_{LS}^* \sum_{n=1}^{N} \sum_{\lambda} e^{i(k_\lambda - k_\mu)z_n} e_\lambda e_\mu \sigma_n^+ a_\lambda, \tag{32}$$

$$\dot{\sigma}_n^- = -(\gamma + i\omega)\sigma_n^- + i K_{LL} \sum_{\lambda} \sum_{\lambda'} e^{i(k_{\lambda'} - k_\lambda)z_n} e_\lambda e_{\lambda'} a_\lambda^\dagger a_{\lambda'} \sigma_n^- + i K_{SS} \sum_{\mu} \sum_{\mu'} e^{i(k_{\mu'} - k_\mu)z_n} e_\mu e_{\mu'} b_\mu^\dagger b_{\mu'} \sigma_n^-$$
$$- i K_{LS}^* \sum_{\lambda} \sum_{\mu} e^{i(k_\lambda - k_\mu)z_n} e_\lambda e_\mu b_\mu^\dagger a_\lambda \sigma_n^z + g_n^-, \tag{33}$$

and

$$\dot{\sigma}_n^z = -2iK_{LS} \sum_\lambda \sum_\mu e^{i(k_\mu - k_\lambda)z_n} e_\lambda e_\mu a_\lambda^\dagger b_\mu \sigma_n^- + \text{H.c.} \tag{34}$$

We introduce the slowly varying electric-field operators

$$A_L^{(+)}(z,t) \equiv \left[i \sum_\lambda e_\lambda a_\lambda(t) e^{ik_\lambda z} \right] e^{-i(k_L z - \omega_L t)} , \tag{35}$$

$$A_S^{(+)}(z,t) \equiv \left[i \sum_\mu e_\mu b_\mu(t) e^{ik_\mu z} \right] e^{-i(k_S z - \omega_S t)} , \tag{36}$$

and their respective conjugates, $A_L^{(-)}(z,t)$ and $A_S^{(-)}(z,t)$, and similarly define the molecular operators

$$R^{(+)}(z_n,t) \equiv i\sigma_n^-(t) \exp\{-i[(k_L - k_S)z_n - (\omega_L - \omega_S)t]\} , \tag{37}$$

$$G^{(+)}(z_n,t) \equiv ig_n^-(t) \exp\{-i[(k_L - k_S)z_n - (\omega_L - \omega_S)t]\} , \tag{38}$$

and

$$R^{(3)}(z_n,t) \equiv \sigma_n^z(t) , \tag{39}$$

with

$$R^{(-)}(z_n,t) \equiv [R^{(+)}(z_n,t)]^\dagger$$

and

$$G^{(-)}(z_n,t) \equiv [G^{(+)}(z_n,t)]^\dagger .$$

By setting $\omega = (k_L - k_S)c$, as is appropriate for spontaneous initiation, we show in Appendix B that these evolve, in the continuum limit, according to

$$\left[\partial_z + \frac{1}{c}\partial_t \right] A_L^{(+)}(z,t) = (Ne_L^2/c)[\tfrac{1}{2}iK_{LL} R^{(3)}(z,t) A_L^{(+)}(z,t) + K_{LS} R^{(+)}(z,t) A_S^{(+)}(z,t)] , \tag{40}$$

$$\left[\partial_z + \frac{1}{c}\partial_t \right] A_S^{(+)}(z,t) = (Ne_S^2/c)[\tfrac{1}{2}iK_{SS} R^{(3)}(z,t) A_S^{(+)}(z,t) - K_{LS}^* R^{(-)}(z,t) A_L^{(+)}(z,t)] , \tag{41}$$

$$\partial_t R^{(+)}(z,t) = -\gamma R^{(+)}(z,t) + G^{(+)}(z,t) + i[K_{LL} A_L^{(-)}(z,t) A_L^{(+)}(z,t) + K_{SS} A_S^{(-)}(z,t) A_S^{(+)}(z,t)] R^{(+)}(z,t) + K_{LS}^* A_L^{(+)}(z,t) A_S^{(-)}(z,t) R^{(3)}(z,t) , \tag{42}$$

and

$$\partial_t R^{(3)}(z,t) = -2K_{LS} A_L^{(-)}(z,t) A_S^{(+)}(z,t) R^{(+)}(z,t) + \text{H.c.} \tag{43}$$

with reservoir averages

$$\langle G^{(+)}(z,t) \rangle_R = \langle G^{(+)}(z,t) G^{(+)}(z',t') \rangle_R = 0 , \tag{44}$$

$$\langle G^{(+)}(z,t) G^{(-)}(z',t') \rangle_R = \gamma(L/N)[1 - R^{(3)}(z,t)] \delta(z - z') \delta(t - t') , \tag{45}$$

and

$$\langle G^{(-)}(z,t) G^{(+)}(z',t') \rangle_R = \gamma(L/N)[1 + R^{(3)}(z,t)] \delta(z - z') \delta(t - t') . \tag{46}$$

Higher-order correlation functions obey the Gaussian decomposition rule, e.g.,

$$\langle G^{(+)}(z_1,t_1) G^{(-)}(z_2,t_2) G^{(+)}(z_3,t_3) G^{(-)}(z_4,t_4) \rangle_R = \langle G^{(+)}(z_1,t_1) G^{(-)}(z_2,t_2) \rangle_R \langle G^{(+)}(z_3,t_3) G^{(-)}(z_4,t_4) \rangle_R .$$

Finally, we introduce a transformation to retarded-time coordinates

$$\tau \equiv t - z/c, \quad \zeta \equiv z \tag{47}$$

and scale parameters and variables through the following substitutions:

$$\zeta \to L\zeta, \quad \tau \to (L/c)\tau, \quad \gamma \to (c/L)\gamma ,$$
$$A_L^{(+)} \to e_L A_L^{(+)}, \quad A_S^{(+)} \to e_S A_S^{(+)} ,$$
$$R^{(+)} \to R^{(+)}/N, \quad G^{(+)} \to G^{(+)}/N, \quad R^{(3)} \to R^{(3)}/N , \tag{48}$$

$$K_{LS} \to (c/Le_L e_S) K_{LS}, \quad K_{LL} \to (c/Le_L^2) K_{LL} ,$$
$$K_{SS} \to (c/Le_S^2) K_{SS} .$$

Then,

$$\partial_\zeta A_L^{(+)}(\zeta,\tau) = \tfrac{1}{2}iK_{LL} R^{(3)}(\zeta,\tau) A_L^{(+)}(\zeta,\tau) + K_{LS} R^{(+)}(\zeta,\tau) A_S^{(+)}(\zeta,\tau) , \tag{49}$$

$$\partial_\zeta A_S^{(+)}(\zeta,\tau) = \tfrac{1}{2}iK_{SS} R^{(3)}(\zeta,\tau) A_S^{(+)}(\zeta,\tau) - K_{LS}^* R^{(-)}(\zeta,\tau) A_L^{(+)}(\zeta,\tau) , \tag{50}$$

$$\partial_\tau R^{(+)}(\zeta,\tau) = -\gamma R^{(+)}(\zeta,\tau) + G^{(+)}(\zeta,\tau) + i[K_{LL} A_L^{(-)}(\zeta,\tau) A_L^{(+)}(\zeta,\tau) + K_{SS} A_S^{(-)}(\zeta,\tau) A_S^{(+)}(\zeta,\tau)] R^{(+)}(\zeta,\tau) + K_{LS}^* A_L^{(+)}(\zeta,\tau) A_S^{(-)}(\zeta,\tau) R^{(3)}(\zeta,\tau) , \tag{51}$$

and

$$\partial_\tau R^{(3)}(\zeta,\tau) = -2K_{LS} A_L^{(-)}(\zeta,\tau) \times A_S^{(+)}(\zeta,\tau) R^{(+)}(\zeta,\tau) + \text{H.c.} \tag{52}$$

Thus, both the length of the medium and its transit time are now unity, while the molecular operators, like those for the field, are extensive, with

$$\langle G^{(+)}(\zeta,\tau)\rangle_R=\langle G^{(+)}(\zeta,\tau)G^{(+)}(\zeta',\tau')\rangle_R=0\ , \quad (53)$$

$$\langle G^{(+)}(\zeta,\tau)G^{(-)}(\zeta',\tau')\rangle_R =\gamma[N-R^{(3)}(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau')\ , \quad (54)$$

and

$$\langle G^{(-)}(\zeta,\tau)G^{(+)}(\zeta',\tau')\rangle_R =\gamma[N+R^{(3)}(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau')\ . \quad (55)$$

Equations (49) and (50) furthermore imply that

$$A_L^{(-)}(\zeta,\tau)A_L^{(+)}(\zeta,\tau)+A_S^{(-)}(\zeta,\tau)A_S^{(+)}(\zeta,\tau)=I(\tau)\ , \quad (56)$$

a function of τ only, a statement of the conservation of photon number; in fact, with the scaling (48), $I(\tau)$ is the number of photons within the medium for given τ.

It is of course necessary to specify boundary and initial conditions for the solutions of (49)–(52). These are determined by

$$A_{L0}^{(+)}(\tau)\equiv A_L^{(+)}(\tau,0)=i\sum_\lambda (e_\lambda/e_L)a_\lambda(0)e^{-i(\omega_\lambda-\omega_L)\tau} \quad (57)$$

and

$$A_{S0}^{(+)}(\tau)\equiv A_S^{(+)}(\tau,0)=i\sum_\mu (e_\mu/e_S)b_\mu(0)e^{-i(\omega_\mu-\omega_S)\tau}\ , \quad (58)$$

the continuum limit of

$$R_0^{(+)}(\zeta_n)\equiv R^{(+)}(\zeta_n,0)=iN\sigma_n^-(0) \quad (59)$$

and

$$R_0^{(3)}(\zeta_n)\equiv R^{(3)}(\zeta_n,0)=N\sigma_n^z(0)\ , \quad (60)$$

and the state of the system, which is invariant in the Heisenberg representation. We assume that the pump is initially in a classical state

$$A_{L0}^{(-)}(\tau)=A_{L0}(\tau)\equiv i\sum_\lambda (e_\lambda/e_L)\alpha_\lambda e^{-i(\omega_\lambda-\omega_L)\tau}\ , \quad (61)$$

where the α_λ are c numbers, and that the Stokes is in the vacuum state, so that (see Appendix B)

$$\langle A_{S0}^{(+)}(\tau)\rangle=\langle A_{S0}^{(+)}(\tau)A_{S0}^{(+)}(\tau')\rangle=0\ , \quad (62)$$

$$\langle A_{S0}^{(+)}(\tau)A_{S0}^{(-)}(\tau')\rangle=\delta(\tau-\tau')\ , \quad (63)$$

and

$$\langle A_{S0}^{(-)}(\tau)A_{S0}^{(+)}(\tau')\rangle=0\ . \quad (64)$$

Assuming also that the molecules initially all occupy the state $|1\rangle$, we find that

$$\langle R_0^{(+)}(\zeta)\rangle=\langle R_0^{(+)}(\zeta)R_0^{(+)}(\zeta')\rangle=0\ , \quad (65)$$

$$\langle R_0^{(+)}(\zeta)R_0^{(-)}(\zeta')\rangle=N\delta(\zeta-\zeta')\ , \quad (66)$$

and

$$\langle R_0^{(-)}(\zeta)R_0^{(+)}(\zeta')\rangle=0\ , \quad (67)$$

with

$$R_0^{(3)}(\zeta)=-N\ , \quad (68)$$

a c number. The higher-order correlation functions of both $A_{S0}^{(+)}(\tau)$ and $R_0^{(+)}(\zeta)$ obey the Gaussian decomposition rule.

B. Stochastic models

The task of solving the operator Eqs. (49)–(52) appears formidable. In Sec. IV we shall present c-number stochastic differential equations whose statistics are related directly to expectation values of the operators satisfying (49)–(52); the only approximation will be the Fokker-Planck (diffusion) approximation. Here we adopt a simpler approach, one more restricted in its valid application, but one which is sufficient to describe ongoing experiments. The basis for our approach is the assumption that quantum fluctuations become negligible in Eqs. (49)–(52) before their nonlinearity becomes important, i.e., the system becomes macroscopic before it becomes nonlinear. This allows us to substitute for these equations a c-number, stochastic model tailored in such a way that (i) when linearized about initial values, it gives averages in agreement with the results of the linearized operator equations, and (ii) it reproduces the semiclassical evolution in the nonlinear regime.

To accomplish this, we start out by linearizing Eqs. (49)–(52) about the values (61), (62), (65), and (68). The model then reduces to two equations:

$$\partial_\zeta A_S^{(+)}(\zeta,\tau)=-\tfrac{1}{2}iK_{SS}NA_S^{(+)}(\zeta,\tau) -K_{LS}^*A_{L0}(\tau)R^{(-)}(\zeta,\tau) \quad (69)$$

and

$$\partial_\tau R^{(+)}(\zeta,\tau)=-\gamma R^{(+)}(\zeta,\tau)+G^{(+)}(\zeta,\tau) +iK_{LL}I_{L0}(\tau)R^{(+)}(\zeta,\tau) -K_{LS}^*NA_{L0}(\tau)A_S^{(-)}(\zeta,\tau)\ , \quad (70)$$

with $I_{L0}(\tau)\equiv|A_{L0}(\tau)|^2$,

$$\langle G^{(+)}(\zeta,\tau)G^{(-)}(\zeta',\tau')\rangle_R=2\gamma N\delta(\zeta-\zeta')\delta(\tau-\tau')\ , \quad (71)$$

and

$$\langle G^{(-)}(\zeta,\tau)G^{(+)}(\zeta',\tau')\rangle_R=0\ . \quad (72)$$

Solutions are found in a straightforward manner through a Laplace transform with respect to ζ; the results are

$$A_S^{(+)}(\zeta,\tau)=\exp(-\tfrac{1}{2}iK_{SS}N\zeta)A_{S0}^{(+)}(\tau)$$
$$+|K_{LS}|^2NA_{L0}(\tau)\int_0^\tau d\tau'\exp\{-[\gamma\tau'+\tfrac{1}{2}iK_{SS}N\zeta+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}$$
$$\times\sqrt{\zeta/g(\tau,\tau')\tau'}I_1[2\sqrt{g(\tau,\tau')\zeta\tau'}]A_{L0}^*(\tau-\tau')A_{S0}^{(+)}(\tau-\tau')$$
$$-K_{LS}A_{L0}(\tau)\int_0^\zeta d\zeta'\exp\{-[\gamma\tau+\tfrac{1}{2}iK_{SS}N\zeta'+iK_{LL}\bar{I}_{L0}(\tau,\tau)\tau]\}I_0[2\sqrt{g(\tau,\tau)\zeta'\tau}]R_0^{(-)}(\zeta-\zeta')$$
$$-K_{LS}A_{L0}(\tau)\int_0^\zeta d\zeta'\int_0^\tau d\tau'\exp\{-[\gamma\tau'+\tfrac{1}{2}iK_{SS}N\zeta'+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}$$
$$\times I_0[2\sqrt{g(\tau,\tau')\zeta'\tau'}]G^{(-)}(\zeta-\zeta',\tau-\tau') \quad (73)$$

and

$$R^{(-)}(\zeta,\tau)=\exp\{-[\gamma\tau+iK_{LL}\bar{I}_{L0}(\tau,\tau)\tau]\}R_0^{(-)}(\zeta)$$
$$+\int_0^\tau d\tau'\exp\{-[\gamma\tau'+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}G^{(-)}(\zeta,\tau-\tau')$$
$$-K_{LS}N\int_0^\tau d\tau'\exp\{-[\gamma\tau'+\tfrac{1}{2}iK_{SS}N\zeta+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}$$
$$\times I_0[2\sqrt{g(\tau,\tau')\zeta\tau'}]A_{L0}^*(\tau-\tau')A_{S0}^{(+)}(\tau-\tau')$$
$$+\int_0^\zeta d\zeta'\exp\{-[\gamma\tau+\tfrac{1}{2}iK_{SS}N\zeta'+iK_{LL}\bar{I}_{L0}(\tau,\tau)\tau]\}$$
$$\times\sqrt{g(\tau,\tau)\tau/\zeta'}I_1[2\sqrt{g(\tau,\tau)\zeta'\tau}]R_0^{(-)}(\zeta-\zeta')$$
$$+\int_0^\zeta d\zeta'\int_0^\tau d\tau'\exp\{-[\gamma\tau'+\tfrac{1}{2}iK_{SS}N\zeta'+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}$$
$$\times\sqrt{g(\tau,\tau')\tau'/\zeta'}I_1[2\sqrt{g(\tau,\tau')\zeta'\tau'}]G^{(-)}(\zeta-\zeta',\tau-\tau')\,, \quad (74)$$

where I_0 and I_1 are modified Bessel functions,

$$\bar{I}_{L0}(\tau,\tau')\equiv\frac{1}{\tau'}\int_{\tau-\tau'}^{\tau}d\tau''I_{L0}(\tau'') \quad (75)$$

is the average incident pump intensity from $\tau-\tau'$ to τ, and

$$g(\tau,\tau')\equiv|K_{LS}|^2N\bar{I}_{L0}(\tau,\tau')\,. \quad (76)$$

Notice that $A_S^{(+)}$ (and $R^{(-)}$) is an integral transform of the operators $A_{S0}^{(+)}$, $R_0^{(-)}$, and $G^{(-)}$ but not their conjugates. This is important to the present approach, for the following reason. We would like to replace the quantum noise sources $A_{S0}^{(+)}$, etc., with c-number stochastic sources. The correlation functions of the latter will obviously be independent of ordering, unlike the situation in Eqs. (63) and (64), (66) and (67), and (71) and (72). If, however, we restrict our attention to a particular ordering prescription for $A_S^{(+)}$ and $A_S^{(-)}$, we encounter in computations only the same ordering of the quantum sources; this makes it possible to identify the correlations of the stochastic sources with that particular ordering. Averages obtained from the stochastic model will thus be identified also with that ordering.

To clarify this, let us consider the following c-number stochastic model:

$$\partial_\zeta A_L(\zeta,\tau)=\tfrac{1}{2}iK_{LL}R_3(\zeta,\tau)A_L(\zeta,\tau)$$
$$+K_{LS}R(\zeta,\tau)A_S(\zeta,\tau)\,, \quad (77)$$

$$\partial_\zeta A_S(\zeta,\tau)=\tfrac{1}{2}iK_{SS}R_3(\zeta,\tau)A_S(\zeta,\tau)$$
$$-K_{LS}^*R^*(\zeta,\tau)A_L(\zeta,\tau)\,, \quad (78)$$

$$\partial_\tau R(\zeta,\tau)=-\gamma R(\zeta,\tau)+i[K_{LL}I_L(\zeta,\tau)$$
$$+K_{SS}I_S(\zeta,\tau)]R(\zeta,\tau)$$
$$+K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R_3(\zeta,\tau)\,, \quad (79)$$

$$\partial_\tau R_3(\zeta,\tau)=-2K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau)+\text{c.c.}\,, \quad (80)$$

with $I_L(\zeta,\tau)\equiv|A_L(\zeta,\tau)|^2$, $I_S(\zeta,\tau)\equiv|A_S(\zeta,\tau)|^2$, and boundary conditions

$$A_L(0,\tau)\equiv A_{L0}(\tau)\,, \quad (81)$$

$$A_S(0,\tau)\equiv A_{S0}(\tau)\,, \quad (82)$$

$$R(\zeta,0)=0\,, \quad (83)$$

$$R_3(\zeta,0)=-N\,, \quad (84)$$

such that $A_{S0}(\tau)$ is a complex Gaussian process with

$$\langle A_{S0}(\tau)\rangle=\langle A_{S0}(\tau)A_{S0}(\tau')\rangle=0 \quad (85)$$

and

$$\langle A_{S0}^*(\tau)A_{S0}(\tau')\rangle=\delta(\tau-\tau')\,. \quad (86)$$

The corresponding linearized system

$$\partial_\zeta A_S(\zeta,\tau)=-\tfrac{1}{2}iK_{SS}NA_S(\zeta,\tau)$$
$$-K_{LS}^*A_{L0}(\tau)R^*(\zeta,\tau)\,, \quad (87)$$

$$\partial_\tau R(\zeta,\tau)=-\gamma R(\zeta,\tau)+iK_{LL}I_{L0}(\tau)R(\zeta,\tau)$$
$$-K_{LS}^*NA_{L0}(\tau)A_S^*(\zeta,\tau) \quad (88)$$

has solutions

$$A_S(\zeta,\tau)=\exp(-\tfrac{1}{2}iK_{SS}N\zeta)A_{S0}(\tau)+|K_{LS}|^2NA_{L0}(\tau)\int_0^\tau d\tau'\exp\{-[\gamma\tau'+\tfrac{1}{2}iK_{SS}N\zeta+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}\times\sqrt{\zeta/g(\tau,\tau')\tau'}\,I_1[2\sqrt{g(\tau,\tau')\zeta\tau'}]A^*_{L0}(\tau-\tau')A_{S0}(\tau-\tau')\ , \tag{89}$$

$$R^*(\zeta,\tau)=-K_{LS}N\int_0^\tau d\tau'\exp\{-[\gamma\tau'+\tfrac{1}{2}iK_{SS}N\zeta+iK_{LL}\bar{I}_{L0}(\tau,\tau')\tau']\}I_0[2\sqrt{g(\tau,\tau')\zeta t'}]A^*_{L0}(\tau-\tau')A_{S0}(\tau-\tau')\ . \tag{90}$$

It is now easy to show using (73) and (89) that

$$\langle|A_S(\zeta,\tau)|^2\rangle=\langle A_S^{(+)}(\zeta,\tau)A_S^{(-)}(\zeta,\tau)\rangle=\delta(0)+g(\tau,0)\left[\zeta+\int_0^\tau d\tau' e^{-2\gamma\tau'}I_1^2[2\sqrt{g(\tau,\tau')\zeta\tau'}]\left[\frac{g(\tau-\tau',0)\zeta}{g(\tau,\tau')\tau'}\right]\right]\ , \tag{91}$$

i.e., the stochastic model yields the antinormally ordered intensity. This agreement is clearly a result of the fact that the quantum calculation involves only the correlation functions $\langle A_{S0}^{(+)}(\tau)A_{S0}^{(-)}(\tau')\rangle$, $\langle R_0^{(-)}(\zeta)R_0^{(+)}(\zeta')\rangle$, and $\langle G^{(-)}(\zeta,\tau)G^{(+)}(\zeta',\tau')\rangle$. It was therefore possible to adopt stochastic sources $A_{S0}(\tau)$, satisfying (85) and (86), and $R_0(\zeta)=G(\zeta,\tau)=0$ with no ambiguity in regard to ordering.

We must therefore have in mind a particular ordering convention when setting up a correspondence between quantum and stochastic models. In the context of the problem under discussion, it is clearly irrelevant just which ordering convention is adopted, since we are ultimately interested in macroscopic features—the solitons. On the other hand, there is some computational advantage in adopting the antinormal-ordering correspondence used to formulate the model of Eqs. (77)–(86). It should be clear that, if we choose normal ordering instead, we need nonzero stochastic sources $G(\zeta,\tau)$ and $R_0(\zeta)$ instead of $A_{S0}(\tau)$. The former must then be generated over the whole range of (ζ,τ), and must be introduced at each step of the integration of $R(\zeta,\tau)$. In contrast, with antinormal ordering, one simply adopts a noisy boundary value $A_{S0}(\tau)$ for $A_S(\zeta,\tau)$ and propagates this deterministically via Eqs. (77)–(80). Following propagation, one may obtain the normally ordered intensity by subtracting the vacuum intensity (which is of course finite in numerical computations):

$$\langle A_S^{(-)}A_S^{(+)}\rangle=\langle A_S^{(+)}A_S^{(-)}\rangle-\langle[A_S^{(+)},A_S^{(-)}]\rangle=\langle|A_S|^2\rangle-\langle[A_{S0}^{(+)},A_{S0}^{(-)}]\rangle=\langle|A_S|^2\rangle-\langle|A_{S0}|^2\rangle\ . \tag{92}$$

For these reasons, we have used the model (77)–(86) for our reported results.[1,10,16–20,29]

Let us close this section with the observation that the self-energy terms in (77)–(80) can cause computational difficulty. Although these are slowly varying relative to the optical-frequency carriers, they may still undergo a great many oscillations during the course of pump depletion. Because we are interested in situations in which the pump depletes with relatively little ground-state depletion [i.e., $N\gg I_{L0}(\tau)$], the oscillations in (77) and (78) are the more serious, and it is advisable to remove them by a transformation of variables. One possibility (there is an infinite number) is

$$B_L(\zeta,\tau)\equiv A_L(\zeta,\tau)\exp\left[-iK_{LL}\left[\tfrac{1}{2}\int_0^\zeta d\zeta' R_3(\zeta',\tau)-\int_0^\tau d\tau' I_{L0}(\tau')\right]\right]\ , \tag{93}$$

$$B_S(\zeta,\tau)\equiv A_S(\zeta,\tau)\exp\left[-iK_{SS}\left[\tfrac{1}{2}\int_0^\zeta d\zeta' R_3(\zeta',\tau)+\int_0^\tau d\tau' I_{S0}(\tau')\right]\right]\ , \tag{94}$$

and

$$Q(\zeta,\tau)\equiv R(\zeta,\tau)\exp\left[-\tfrac{1}{2}i(K_{LL}-K_{SS})\int_0^\zeta d\zeta' R_3(\zeta',\tau)+i\int_0^\tau d\tau'[K_{LL}I_{L0}(\tau')+K_{SS}I_{S0}(\tau)]\right]\ , \tag{95}$$

which, as one can show using the identity $\frac{1}{2}\partial_\tau R_3=-\partial_\zeta I_L=\partial_\zeta I_S$, yields from (77)–(80),

$$\partial_\zeta B_L(\zeta,\tau)=K_{LS}Q(\zeta,\tau)B_S(\zeta,\tau)\ , \tag{96}$$

$$\partial_\zeta B_S(\zeta,\tau)=-K^*_{LS}Q^*(\zeta,\tau)B_L(\zeta,\tau)\ , \tag{97}$$

$$\partial_\tau Q(\zeta,\tau)=-\gamma Q(\zeta,\tau)+2i[K_{LL}I_L(\zeta,\tau)+K_{SS}I_S(\zeta,\tau)]Q(\zeta,\tau)+K^*_{LS}B_L(\zeta,\tau)B^*_S(\zeta,\tau)R_3(\zeta,\tau)\ , \tag{98}$$

and

$$\partial_\tau R_3(\zeta,\tau)=-2K_{LS}B^*_L(\zeta,\tau)B_S(\zeta,\tau)Q(\zeta,\tau)+\text{c.c.} \tag{99}$$

One must of course similarly transform $A_{L0}(\tau)$ and $A_{S0}(\tau)$.

IV. SCHRÖDINGER REPRESENTATION

In the preceding section we developed a model, based on an antinormal-ordering correspondence for averages of field operators, that is applicable in situations where nonlinearity and quantum noise are not simultaneously important. Here we derive similar models by introducing quasiprobability functions, finding Fokker-Planck equations describing their evolution, and finally associating them with stochastic differential equations. The resulting models are then compared with Eqs. (77)–(86).

A. Fokker-Planck equations

We begin by recasting the Hamiltonian (28) in terms of local operators. We define pump and Stokes operators

$$a_l \equiv \frac{1}{\sqrt{N}} \sum_{\lambda=[-N/2]}^{[N/2]} a_\lambda e^{ik_\lambda z_l} \tag{100}$$

and

$$b_m \equiv \frac{1}{\sqrt{N}} \sum_{\mu=[-N/2]}^{[N/2]} b_\mu e^{ik_\mu z_m} \tag{101}$$

(brackets in the summation limits indicate the "greatest integer" operation) by discretizing phase space according to

$$z_n \equiv \frac{(n-1)L}{N} \tag{102}$$

and

$$k_\lambda \equiv k_L + \frac{2\pi\lambda}{L}, \quad k_\mu \equiv k_S + \frac{2\pi\mu}{L}, \tag{103}$$

where z_n is a molecular site ($n=1,2,\ldots,N$). Then a_l, b_m, and their conjugates obey Bose-Einstein commutation relations:

$$[a_l, a_{l'}^\dagger] = \delta_{ll'}, \quad [b_m, b_{m'}^\dagger] = \delta_{mm'} . \tag{104}$$

Defining

$$\omega_{ll'} \equiv \frac{1}{N} \sum_{\lambda=[-N/2]}^{[N/2]} \omega_\lambda e^{ik_\lambda(z_l - z_{l'})}, \tag{105}$$

$$e_{ln} \equiv \frac{1}{\sqrt{N}} \sum_{\lambda=[-N/2]}^{[N/2]} e_\lambda e^{ik_\lambda(z_l - z_n)}, \tag{106}$$

and analogous expressions for $\omega_{mm'}$ and e_{mn}, we obtain from (28)

$$\begin{aligned} H' = {} & \tfrac{1}{2}\hbar\omega \sum_n \sigma_n^z + \hbar \sum_l \sum_{l'} \omega_{ll'} a_l^\dagger a_{l'} + \hbar \sum_m \sum_{m'} \omega_{mm'} b_m^\dagger b_{m'} \\ & - \tfrac{1}{2}\hbar K_{LL} \sum_l \sum_{l'} \sum_n e_{nl}^* e_{nl'} a_l^\dagger a_{l'} \sigma_n^z \\ & - \frac{1}{2}\hbar K_{SS} \sum_m \sum_{m'} \sum_n e_{nm}^* e_{nm'} b_m^\dagger b_{m'} \sigma_n^z \\ & - \hbar \left[K_{LS} \sum_l \sum_m \sum_n e_{nl}^* e_{nm} a_l^\dagger b_m \sigma_n^- + \text{H.c.} \right], \end{aligned} \tag{107}$$

with all summations ranging from 1 to N.

We saw in Sec. III that the quantum sources of field and molecular fluctuations enter into the linear evolution with opposite orderings. For example, the expectation values $\langle A_S^{(+)} A_S^{(-)} \rangle$ and $\langle R^{(-)} R^{(+)} \rangle$, computed from (73) and (74), depend only on the orderings $\langle A_{S0}^{(+)} A_{S0}^{(-)} \rangle$, $\langle R_0^{(-)} R_0^{(+)} \rangle$, and $\langle G^{(-)} G^{(+)} \rangle$ (arguments omitted). Hence, the model (77)–(86), while associated with antinormally ordered field averages, clearly yields normally ordered averages of $R^{(+)}$. A quasiprobability function Q having similar properties can be derived from the characteristic function

$$\begin{aligned} C_Q(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t) \equiv \text{tr}[& e^{i\mathbf{u}\cdot\mathbf{a}} e^{i\mathbf{u}^*\cdot\mathbf{a}^\dagger} e^{i\mathbf{v}\cdot\mathbf{b}} e^{i\mathbf{v}^*\cdot\mathbf{b}^\dagger} \\ & \times e^{i\mathbf{r}^*\cdot\sigma^+} e^{i\mathbf{z}\cdot\sigma^z} e^{i\mathbf{r}\cdot\sigma^-} \rho(t)] , \end{aligned} \tag{108}$$

written here in terms of the N-dimensional vectors $\mathbf{a} \equiv (a_1, a_2, \ldots, a_N)$, $\mathbf{u} \equiv (u_1, u_2, \ldots, u_N)$, etc., where $\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z}$ are c numbers. Performing the inverse transformation to a "phase space" of c numbers, we define

$$\begin{aligned} Q(\alpha,\beta,\rho,\zeta,t) \equiv \frac{1}{(2\pi)^{7N}} \int d^{2N}u \int d^{2N}v \int d^{2N}r \int d^{N}z \exp[& -i(\mathbf{u}\cdot\boldsymbol{\alpha} + \mathbf{u}^*\cdot\boldsymbol{\alpha}^* + \mathbf{v}\cdot\boldsymbol{\beta} + \mathbf{v}^*\cdot\boldsymbol{\beta}^* + \mathbf{r}\cdot\boldsymbol{\rho} \\ & + \mathbf{r}^*\cdot\boldsymbol{\rho}^* + \mathbf{z}\cdot\boldsymbol{\zeta})] C_Q(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t) , \end{aligned} \tag{109}$$

in which

$$d^{2N}u \equiv \prod_{l=1}^{N} [\text{Re}(du_l)\text{Im}(du_l)] ,$$

etc. It follows from this definition that, for example,

$$\langle |\alpha_l|^2 \rangle = \langle a_l a_l^\dagger \rangle \tag{110}$$

while

$$\langle |\rho_n|^2 \rangle = \langle \sigma_n^+ \sigma_n^- \rangle . \tag{111}$$

For reasons that will become apparent below, we also introduce the Wigner function W, whose moments equal the expectation values of symmetrically ordered operators, e.g.,

$$\langle |\alpha_l|^2 \rangle = \tfrac{1}{2}(\langle a_l^\dagger a_l \rangle + \langle a_l a_l^\dagger \rangle) , \tag{112}$$

$$\langle |\rho_n|^2 \rangle = \tfrac{1}{2}(\langle \sigma_n^+ \sigma_n^- \rangle + \langle \sigma_n^- \sigma_n^+ \rangle) . \tag{113}$$

It is defined as

$$W(\boldsymbol{\alpha},\boldsymbol{\beta},\boldsymbol{\rho},\boldsymbol{\zeta},t)\equiv\frac{1}{(2\pi)^{7N}}\int d^{2N}u\int d^{2N}v\int d^{2N}r\int d^{N}z\,\exp[-i(\mathbf{u}\cdot\boldsymbol{\alpha}+\mathbf{u}^*\cdot\boldsymbol{\alpha}^*+\mathbf{v}\cdot\boldsymbol{\beta}+\mathbf{v}^*\cdot\boldsymbol{\beta}^*+\mathbf{r}\cdot\boldsymbol{\rho}+\mathbf{r}^*\cdot\boldsymbol{\rho}^*+\mathbf{z}\cdot\boldsymbol{\zeta})]C_W(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t) \tag{114}$$

using the characteristic function

$$C_W(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t)\equiv\mathrm{tr}\{\exp[i(\mathbf{u}^*\cdot\mathbf{a}^\dagger+\mathbf{u}\cdot\mathbf{a}+\mathbf{v}^*\cdot\mathbf{b}^\dagger+\mathbf{v}\cdot\mathbf{b}+\mathbf{r}^*\cdot\boldsymbol{\sigma}^++\mathbf{r}\cdot\boldsymbol{\sigma}^-+\mathbf{z}\cdot\boldsymbol{\sigma}^z)]\rho(t)\}\ . \tag{115}$$

Applying these definitions to the Liouville equation (30), we obtain kinetic equations for Q and W that contain derivatives of all orders. (If ground-state depletion is ignored, with ζ assumed constant, then the equation for Q truncates at third order.) The Fokker-Planck or diffusion approximation may be justified for large N using the familiar technique of a system-size expansion. Accordingly, we retain only second-derivative terms and obtain the following:

$$\begin{aligned}\frac{\partial Q}{\partial t}=\Bigg\{\Bigg[&i\omega\sum_n\frac{\partial}{\partial\rho_n}\rho_n+i\sum_{l,l'}\omega_{ll'}\frac{\partial}{\partial\alpha_l}\alpha_{l'}+i\sum_{m,m'}\omega_{mm'}\frac{\partial}{\partial\beta_m}\beta_{m'}\\
&+\tfrac{1}{2}iK_{LL}\sum_{l,l',n}e_{nl}^*e_{nl'}\left[-\frac{\partial}{\partial\alpha_l}\alpha_{l'}\zeta_n+2\frac{\partial}{\partial\rho_n^*}\alpha_l^*\alpha_{l'}\rho_n^*+2\frac{\partial^2}{\partial\alpha_l\partial\rho_n^*}\alpha_{l'}\rho_n^*\right]\\
&+\tfrac{1}{2}iK_{SS}\sum_{m,m',n}e_{nm}^*e_{nm'}\left[-\frac{\partial}{\partial\beta_m}\beta_{m'}\zeta_n+2\frac{\partial}{\partial\rho_n^*}\beta_m^*\beta_{m'}\rho_n^*+2\frac{\partial^2}{\partial\beta_m\partial\rho_n^*}\beta_{m'}\rho_n^*\right]\\
&+iK_{LS}\sum_{l,m,n}e_{nl}^*e_{nm}\Bigg[-\frac{\partial}{\partial\alpha_l}\beta_m\rho_n+\frac{\partial}{\partial\beta_m^*}\alpha_l^*\rho_n-\frac{\partial}{\partial\rho_n^*}\alpha_l^*\beta_m\zeta_n+2\frac{\partial}{\partial\zeta_n}\alpha_l^*\beta_m\rho_n\\
&\qquad-\frac{\partial^2}{\partial\alpha_l\partial\rho_n^*}\beta_m\zeta_n+2\frac{\partial^2}{\partial\alpha_l\partial\zeta_n}\beta_m\rho_n+\frac{\partial^2}{\partial\rho_n^{*2}}\alpha_l^*\beta_m\rho_n^*-2\frac{\partial^2}{\partial\zeta_n^2}\alpha_l^*\beta_m\rho_n\Bigg]\\
&+\gamma\sum_n\left[\frac{\partial}{\partial\rho_n}\rho_n+\frac{1}{2}\frac{\partial^2}{\partial\rho_n\partial\rho_n^*}(1+\zeta_n)\right]\Bigg]+\text{c.c.}\Bigg\}Q\end{aligned} \tag{116}$$

and

$$\begin{aligned}\frac{\partial W}{\partial t}=\Bigg\{\Bigg[&i\omega\sum_n\frac{\partial}{\partial\rho_n}\rho_n+i\sum_{l,l'}\omega_{ll'}\frac{\partial}{\partial\alpha_l}\alpha_{l'}+i\sum_{m,m'}\omega_{mm'}\frac{\partial}{\partial\beta_m}\beta_{m'}+\tfrac{1}{2}iK_{LL}\sum_{l,l',n}e_{nl}^*e_{nl'}\left[-\frac{\partial}{\partial\alpha_l}\alpha_{l'}\zeta_n+2\frac{\partial}{\partial\rho_n^*}\alpha_l^*\alpha_{l'}\rho_n^*\right]\\
&+\tfrac{1}{2}iK_{SS}\sum_{m,m',n}e_{nm}^*e_{nm'}\left[-\frac{\partial}{\partial\beta_m}\beta_{m'}\zeta_n+2\frac{\partial}{\partial\rho_n^*}\beta_m^*\beta_{m'}\rho_n^*\right]\\
&+iK_{LS}\sum_{l,m,n}e_{nl}^*e_{nm}\left[-\frac{\partial}{\partial\alpha_l}\beta_m\rho_n+\frac{\partial}{\partial\beta_m^*}\alpha_l^*\rho_n-\frac{\partial}{\partial\rho_n^*}\alpha_l^*\beta_m\zeta_n+2\frac{\partial}{\partial\zeta_n}\alpha_l^*\beta_m\rho_n\right]\\
&+\gamma\sum_n\left[\frac{\partial}{\partial\rho_n}\rho_n+\frac{1}{2}\frac{\partial^2}{\partial\rho_n\partial\rho_n^*}\right]\Bigg]+\text{c.c.}\Bigg\}W\ .\end{aligned} \tag{117}$$

The initial values of Q and W, which for brevity we do not write down, can be obtained from the initial density operator by applying the definitions[33] (108) and (109) and (114) and (115).

B. Stochastic differential equations

Let us attempt to associate the results (116) and (117) with stochastic differential equations. The rules of Ito's calculus yield for the former the system

$$\dot\alpha_l(t)=-i\sum_{l'}\omega_{ll'}\alpha_{l'}(t)+\tfrac{1}{2}iK_{LL}\sum_{l',n}e_{nl}^*e_{nl'}\alpha_{l'}(t)\zeta_n(t)+iK_{LS}\sum_{m,n}e_{nl}^*e_{nm}\beta_m(t)\rho_n(t)+\Gamma_{\alpha_l}(t)\ , \tag{118}$$

$$\dot\beta_m(t)=-i\sum_{m'}\omega_{mm'}\beta_{m'}(t)+\tfrac{1}{2}iK_{SS}\sum_{m',n}e_{nm}^*e_{nm'}\beta_{m'}(t)\zeta_n(t)+iK_{LS}^*\sum_{l,n}e_{nl}e_{nm}^*\alpha_l(t)\rho_n^*(t)+\Gamma_{\beta_m}(t)\ , \tag{119}$$

$$\begin{aligned}\dot\rho_n(t)=&-(\gamma+i\omega)\rho_n(t)\\
&+i\left[K_{LL}\sum_{l,l'}e_{nl}e_{nl'}^*\alpha_l(t)\alpha_{l'}^*(t)+K_{SS}\sum_{m,m'}e_{nm}e_{nm'}^*\beta_m(t)\beta_{m'}^*(t)\right]\rho_n\\
&-iK_{LS}^*\sum_{l,m}e_{nl}e_{nm}^*\alpha_l(t)\beta_m^*(t)\zeta_n(t)+\Gamma_{\rho_n}(t)\ ,\end{aligned} \tag{120}$$

and

$$\dot{\zeta}_n(t)=\left[-2iK_{LS}\sum_{l,m}e_{nl}^*e_{nm}\alpha_l^*(t)\beta_m(t)\rho_n(t)+\text{c.c.}\right]+\Gamma_{\zeta_n}(t)\ , \tag{121}$$

where the noise sources $\Gamma_{\alpha_l}(t)$, $\Gamma_{\beta_m}(t)$, etc., are Gaussian with zero mean and

$$\langle\Gamma_{\alpha_l}(t)\Gamma_{\rho_n^*}(t')\rangle=\left[iK_{LL}\sum_{l'}e_{nl}^*e_{nl'}\alpha_{l'}(t)\rho_n^*(t)-iK_{LS}\sum_m e_{nl}^*e_{nm}\beta_m(t)\zeta_n(t)\right]\times\delta(t-t')\ , \tag{122}$$

$$\langle\Gamma_{\alpha_l}(t)\Gamma_{\zeta_n}(t')\rangle=2iK_{LS}\sum_m e_{nl}^*e_{nm}\beta_m(t)\rho_n(t)\delta(t-t')\ , \tag{123}$$

$$\langle\Gamma_{\beta_m}(t)\Gamma_{\rho_n^*}(t')\rangle=iK_{SS}\sum_{m'}e_{nm}^*e_{nm'}\beta_{m'}(t)\rho_n^*(t)\delta(t-t')\ , \tag{124}$$

$$\langle\Gamma_{\rho_n}(t)\Gamma_{\rho_{n'}}(t')\rangle=-2iK_{LS}^*\sum_{l,m}e_{nl}e_{nm}^*\alpha_l(t)\beta_m^*(t)\times\rho_n(t)\delta_{nn'}\delta(t-t')\ , \tag{125}$$

$$\langle\Gamma_{\rho_n}(t)\Gamma_{\rho_{n'}^*}(t')\rangle=\gamma\sum_n[1+\zeta_n(t)]\delta_{nn'}\delta(t-t')\ , \tag{126}$$

and

$$\langle\Gamma_{\zeta_n}(t)\Gamma_{\zeta_{n'}}(t')\rangle=-4iK_{LS}\sum_{l,m}e_{nl}^*e_{nm}\alpha_l^*(t)\beta_m(t)\rho_n(t)\times\delta_{nn'}\delta(t-t')+\text{c.c.} \tag{127}$$

All other second-order correlation functions, other than conjugates of the above, must vanish.

To facilitate a comparison with the stochastic model, we introduce the definitions, analogous to (35)–(39),

$$A_L(z,t)\equiv\left[i\sum_\lambda e_\lambda\alpha_\lambda(t)e^{ik_\lambda z}\right]e^{-i(k_Lz-\omega_Lt)}\ , \tag{128}$$

$$A_S(z,t)\equiv\left[i\sum_\mu e_\mu\beta_\mu(t)e^{ik_\mu z}\right]e^{-i(k_Sz-\omega_St)}\ , \tag{129}$$

$$R(z_n,t)\equiv i\rho_n(t)\exp\{-i[(k_L-k_S)z_n-(\omega_L-\omega_S)t]\}\ , \tag{130}$$

$$R_3(z_n,t)\equiv\zeta_n(t)\ , \tag{131}$$

and

$$F_L(z,t)\equiv\left[\frac{i}{\sqrt{N}}\sum_l\Gamma_{\alpha_l}(t)\sum_\lambda e_\lambda e^{ik_\lambda(z-z_l)}\right]e^{-i(k_Lz-\omega_Lt)}\ , \tag{132}$$

$$F_S(z,t)\equiv\left[\frac{i}{\sqrt{N}}\sum_m\Gamma_{\beta_m}(t)\sum_\mu e_\mu e^{ik_\mu(z-z_m)}\right]\times e^{-i(k_Sz-\omega_St)}\ , \tag{133}$$

$$F_R(z_n,t)\equiv i\Gamma_{\rho_n}(t)\exp\{-i[(k_L-k_S)z_n-(\omega_L-\omega_S)t]\}\ , \tag{134}$$

$$F_3(z_n,t)\equiv\Gamma_{\zeta_n}(t)\ . \tag{135}$$

Similarly, we transform to retarded-time coordinates and adopt the scaling (48), supplemented by

$$F_L\to(e_L/L)F_L,\quad F_S\to(e_S/L)F_S\ ,\quad F_R\to(c/LN)F_R,\quad F_3\to(c/LN)F_3 \tag{136}$$

and obtain in the continuum limit

$$\partial_\zeta A_L(\zeta,\tau)=\tfrac{1}{2}iK_{LL}R_3(\zeta,\tau)A_L(\zeta,\tau)+K_{LS}R(\zeta,\tau)A_S(\zeta,\tau)+F_L(\zeta,\tau)\ , \tag{137}$$

$$\partial_\zeta A_S(\zeta,\tau)=\tfrac{1}{2}iK_{SS}R_3(\zeta,\tau)A_S(\zeta,\tau)-K_{LS}^*R^*(\zeta,\tau)A_L(\zeta,\tau)+F_S(\zeta,\tau)\ , \tag{138}$$

$$\partial_\tau R(\zeta,\tau)=-\gamma R(\zeta,\tau)+i[K_{LL}I_L(\zeta,\tau)+K_{SS}I_S(\zeta,\tau)]R(\zeta,\tau)+K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R_3(\zeta,\tau)+F_R(\zeta,\tau)\ , \tag{139}$$

$$\partial_\tau R_3(\zeta,\tau)=[-2K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau)+\text{c.c.}]+F_3(\zeta,\tau)\ , \tag{140}$$

with

$$\langle F_L(\zeta,\tau)F_R^*(\zeta',\tau')\rangle=[iK_{LL}A_L(\zeta,\tau)R^*(\zeta,\tau)-K_{LS}A_S(\zeta,\tau)R_3(\zeta,\tau)]\times\delta(\zeta-\zeta')\delta(\tau-\tau')\ , \tag{141}$$

$$\langle F_L(\zeta,\tau)F_3(\zeta',\tau')\rangle=2K_{LS}A_S(\zeta,\tau)R(\zeta,\tau)\delta(\zeta-\zeta')\times\delta(\tau-\tau')\ , \tag{142}$$

$$\langle F_S(\zeta,\tau)F_R^*(\zeta',\tau')\rangle=iK_{SS}A_S(\zeta,\tau)R^*(\zeta,\tau)\delta(\zeta-\zeta')\times\delta(\tau-\tau')\ , \tag{143}$$

$$\langle F_R(\zeta,\tau)F_R(\zeta',\tau')\rangle=2K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R(\zeta,\tau)\times\delta(\zeta-\zeta')\delta(\tau-\tau')\ , \tag{144}$$

$$\langle F_R(\zeta,\tau)F_R^*(\zeta',\tau')\rangle=\gamma[N+R_3(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau')\ , \tag{145}$$

and

$$\langle F_3(\zeta,\tau)F_3(\zeta',\tau')\rangle=[-4K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau)+\text{c.c.}]\delta(\zeta-\zeta')\delta(\tau-\tau')\ . \tag{146}$$

Adopting the state vector of Sec. III B, with a classical pump $A_{L0}(\tau)$, an unpolarized, unexcited medium, and vacuum-state Stokes, we apply the correspondences (110) and (111) to find precisely the initial and boundary conditions (81)–(86).

Let us examine the resulting model (137)–(146). Notice first that, when linearized about initial values, all Langevin sources disappear, since the correlation functions (141)–(146) vanish. Thus, quantum initiation is effected by the Stokes vacuum alone, as in the stochastic model of Sec. III. Moreover, we see by comparing (137)–(140) with (77)–(80) that the difference between the two systems lies solely in the Langevin sources of the former. Here, we have further evidence of the validity of the model (77)–(80) for situations in which quantum fluctuations and nonlinearity are not of simultaneous importance.

For other situations, one might adopt the model (137)–(146). As written, however, these equations are problematic. In particular, we cannot in general have (143) hold while $\langle F_S(\zeta,\tau)F_S^*(\zeta',\tau')\rangle=0$. A way out of this dilemma is achieved by adopting a representation in which β_m and β_m^*, and hence $A_S(\zeta,\tau)$ and $A_S^*(\zeta,\tau)$, are not complex conjugates, but independent complex variables. (Such an approach is associated with the positive-P representation[35,36] when the normal-ordering correspondence is used.) Because these quantities are coupled to the other variables $A_L(\zeta,\tau)$, $A_L^*(\zeta,\tau)$, etc., it then becomes necessary to likewise consider these as independent rather than conjugate. The effect is then to double the "phase space" over which averages are to be performed and the number of equations that must be solved.

One finds similar complications arising in all representations in which normal (antinormal)-ordering correspondences are adopted (see Appendix C). As one may already infer by comparing the Fokker-Planck equations [(116) and (117)], the Wigner representation affords a significantly simpler model. Indeed, there is only one Langevin source; in the notation of Eqs. (118)–(121), $\Gamma_{\alpha_l}(t)=\Gamma_{\beta_m}(t)=\Gamma_{\zeta_n}(t)=0$ while

$$\langle \Gamma_{\rho_n}(t)\rangle=\langle \Gamma_{\rho_n}(t)\Gamma_{\rho_{n'}}(t')\rangle=0 \tag{147a}$$

and

$$\langle \Gamma_{\rho_n}(t)\Gamma_{\rho_{n'}^*}(t')\rangle=\gamma\delta_{nn'}\delta(t-t') . \tag{147b}$$

Hence, applying the definitions (128)–(131) and (134), assuming the continuum limit, and scaling variables and parameters as above, we obtain in the Wigner representations the model

$$\partial_\zeta A_L(\zeta,\tau)=\tfrac{1}{2}iK_{LL}R_3(\zeta,\tau)A_L(\zeta,\tau)+K_{LS}R(\zeta,\tau)A_S(\zeta,\tau) , \tag{148}$$

$$\partial_\zeta A_S(\zeta,\tau)=\tfrac{1}{2}iK_{SS}R_3(\zeta,\tau)A_S(\zeta,\tau)-K_{LS}^*R^*(\zeta,\tau)A_L(\zeta,\tau) , \tag{149}$$

$$\partial_\tau R(\zeta,\tau)=-\gamma R(\zeta,\tau)+i[K_{LL}I_L(\zeta,\tau)+K_{SS}I_S(\zeta,\tau)]R(\zeta,\tau)+K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R_3(\zeta,\tau)+F_R(\zeta,\tau) , \tag{150}$$

and

$$\partial_\tau R_3(\zeta,\tau)=-2K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau)+\text{c.c.} \tag{151}$$

with $F_R(\zeta,\tau)$ a zero-mean complex Gaussian process for which

$$\langle F_R(\zeta,\tau)F_R^*(\zeta',\tau')\rangle=\gamma N\delta(\zeta-\zeta')\delta(\tau-\tau') . \tag{152}$$

Because of the symmetric-ordering correspondence (112) and (113), the state vector of Sec. III B now implies the initial and boundary values

$$A_L(0,\tau)=A_{L0}(\tau) , \tag{153}$$

$$A_S(0,\tau)=A_{S0}(\tau) , \tag{154}$$

$$R(\zeta,0)=R_0(\zeta) , \tag{155}$$

$$R_3(\zeta,0)=-N , \tag{156}$$

where the quantum-noise sources $A_{S0}(\tau)$ and $R_0(\zeta)$ are Gaussian with

$$\langle A_{S0}(\tau)\rangle=\langle R_0(\zeta)\rangle=\langle A_{S0}(\tau)A_{S0}(\tau')\rangle=\langle R_0(\zeta)R_0(\zeta')\rangle=0 \tag{157}$$

and

$$\langle A_{S0}^*(\tau)A_{S0}(\tau')\rangle=\tfrac{1}{2}\delta(\tau-\tau') , \tag{158}$$

$$\langle R_0^*(\zeta)R_0(\zeta')\rangle=\tfrac{1}{2}N\delta(\zeta-\zeta') . \tag{159}$$

Examining (148)–(152), we see that the Wigner representation yields a system of stochastic differential equations that, but for the single, additive noise source $F_R(\zeta,\tau)$, would be identical to the plane-wave semiclassical model of SRS. Quantum noise due to collisional dephasing enters through this source, while vacuum fluctuations and radiation reaction enter through noisy initial and boundary conditions on $A_S(\zeta,\tau)$ and $R(\zeta,\tau)$ via Eqs. (158) and (159). While this model is a bit more elaborate than the stochastic model constructed in Sec. III B, it can be expected to maintain its validity in cases for which the latter is expected to fail—when nonlinearity arises while the system is still being influenced by quantum noise. In such cases, this model is clearly superior to those based upon normal or antinormal orderings, at least from a computational point of view.

V. CONCLUSIONS

We have used the dressed-state representation to present a quantum-mechanical model suitable for disussing the quantum-mechanical aspects of stimulated Raman scattering. The Heisenberg equations of motion were derived and presented in Sec. III, and the equations for the stochastic model, based upon an antinormal-ordering correspondence for averages of field operations, and a quantum-classical correspondence, were developed from them [Eqs. (77)–(86)]. This model is particularly useful for numerical calculations for the dynamical evolution of the system in cases where quantum fluctuations and nonlinear evolution are not simultaneously important. We use this model in a future paper,[31] to generate ensembles to characterize the statistics of amplified, macroscopic realizations of quantum fluctuations, particularly in the form of effects due to phase waves and spontaneously generated solitons.

In addition to the stochastic model developed in Sec. III, we have presented several alternative approaches in Sec. IV by introducing quasiprobability functions, Eqs. (109) and (114), and their associated Fokker-Planck equations, Eqs. (116) and (117), respectively, from which we obtained the corresponding dynamical stochastic equations of motion and propagation, which are presented in the continuum limit by Eqs. (137)–(146), and Eqs. (148)–(152). The Q representation, whose moments are equivalent to expectation values of products of antinormally ordered field operators and normally ordered atomic operators, yields stochastic differential equations which are identical to the equations for the stochastic model developed in Sec. III, for the same initial and boundary conditions and the condition that quantum fluctuations and nonlinearity are not simultaneously important. This result gives further credence to the validity of the model developed in Sec. III. For more general cases, one may be required to adopt a representation in which the Stokes amplitudes, $A_S(\zeta,\tau)$ and $A_S^*(\zeta,\tau)$ are not complex conjugates, which automatically requires the same condition for the other variables of the system, leading to the positive-P representation,[35,36] when the normal-ordering correspondence is used.

The Wigner representation, where moments correspond to expectation values of symmetrically ordered operators, was also developed in Sec. IV and leads to a considerably simpler set of equations as compared to the Q representation, there being only one Langevin source. The state vector of Sec. III implies boundary conditions, for this case, which determine the quantum noise sources commensurate with the symmetric-ordering correspondence. For the general case, the Wigner representation is valid where the stochastic model fails, and is evidently the least complicated to implement numerically with respect to other alternatives.

For the stochastic characterization of SRS, the stochastic model developed in Sec. III is valid and is by far superior to the alternatives with regard to ease of numerical implementation. We therefore use this method to generate numerical algorithms to characterize the macroscopic stochastic effects of quantum initiation, particularly in regard to spontaneous phase-wave and soliton generation. The results will be presented and discussed in relation to reported experimental results in a future paper.[31]

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APPENDIX A: DRESSED-STATE HAMILTONIAN

We follow Coulter's procedure,[32] using a unitary operator e^{iK} to determine, to second order in the interaction energy, the dressed-state Hamiltonian $H' \equiv e^{iK} H e^{-iK}$ for H given by (21). The first- and second-order contributions to K we call, respectively, K_1 and K_2. As shown by Coulter, these Hermitian operators generate energy terms H_1 and H_2 satisfying

$$H_1 = i[K_1, H_0] + H_{AF} \tag{A1}$$

and

$$H_2 = i[K_2, H_0] - \tfrac{1}{2}[K_1, [K_1, H_0]] + i[K_1, H_{AF}] \tag{A2}$$

under the assumption that

$$[H_0, H_1] = [H_0, H_2] = 0 \ . \tag{A3}$$

where $H_0 \equiv H_A + H_F$ is the free-molecule–free-field Hamiltonian.

An operator H_1 satisfying (A3) can be constructed from the terms in H_{AF} that commute with H_0. Accordingly,

$$H_1 = -\hbar \sum_n \sum_\lambda \sum_i \sum_j \delta_{\omega_\lambda, \omega_{ij}} g_{ij}^{\lambda n} \sigma_n^{ij} a_\lambda + \text{H.c.} \ , \tag{A4}$$

where $\omega_{ij} \equiv \omega_i - \omega_j$. (Note that the Kronecker delta ensures the rotating-wave approximation through energy conservation for the dressed states.) Then

$$K_1 = -i \sum_n \sum_\lambda \sum_i \sum_j \frac{\bar{\delta}_{\omega_\lambda, \omega_{ij}}}{\omega_\lambda - \omega_{ij}} g_{ij}^{\lambda n} \sigma_n^{ij} a_\lambda + \text{H.c.} \ , \tag{A5}$$

where $\bar{\delta}_{ij} \equiv 1 - \delta_{ij}$ is the complement to the Kronecker delta, satisfies (A1).

To satisfy (A3), we construct H_2 from those terms in the combination

$$-\tfrac{1}{2}[K_1, [K_1, H_0]] + i[K_1, H_{AF}] \tag{A6}$$

that commute with H_0. This procedure yields the general result

$$H_2 = -\tfrac{1}{2}\hbar \sum_n \sum_\lambda \sum_\mu \sum_i \sum_j \sum_k \left[\delta_{\omega_\lambda + \omega_\mu, \omega_{jk}} \left(\frac{\bar{\delta}_{\omega_\lambda, \omega_{ik}}}{\omega_{ik} - \omega_\lambda} g_{ji}^{\mu n} g_{ik}^{\lambda n} \sigma_n^{jk} a_\mu a_\lambda + \frac{\bar{\delta}_{\omega_\lambda, \omega_{ji}}}{\omega_{ij} + \omega_\lambda} g_{ji}^{\lambda n} g_{ik}^{\mu n} \sigma_n^{jk} a_\lambda a_\mu \right) + \delta_{\omega_\lambda - \omega_\mu, \omega_{kj}} \left(\frac{\bar{\delta}_{\omega_\lambda, \omega_{ki}}}{\omega_{ik} + \omega_\lambda} g_{ji}^{\mu n} g_{ki}^{\lambda n *} \sigma_n^{jk} a_\mu a_\lambda^\dagger + \frac{\bar{\delta}_{\omega_\lambda, \omega_{ij}}}{\omega_{ij} - \omega_\lambda} g_{ij}^{\lambda n *} g_{ik}^{\mu n} \sigma_n^{jk} a_\lambda^\dagger a_\mu \right) \right] + \text{H.c.} \tag{A7}$$

(Note again the explicit energy conservation, as well as the exclusion of the resonances $\omega_\lambda = \omega_{ik}$, etc.) The generator K_2, which we do not write, can now be derived from (A2).

We now delete from $H' = H_0 + H_1 + H_2$ those terms not involving the Raman-connected states $|1\rangle$ and $|2\rangle$. Thus we ignore H_1 entirely. The operator H_2 retains terms of three distinct types, reflecting self-energy, Raman, and direct two-photon contributions. The latter transitions are not pumped, and we therefore delete them also. Finally, we introduce the operators $\sigma_n^+ \equiv \sigma_n^{21}$, $\sigma_n^- \equiv \sigma_n^{12}$, and $\sigma_n^z \equiv \sigma_n^{22} - \sigma_n^{11}$ and normally order H_2 with respect to the photon operators. Assuming that the frequency shifts incurred by this process are negligible, we obtain the result (22).

APPENDIX B: HEISENBERG EQUATIONS

Formal integration of the Heisenberg equations (31) and (32), followed by the application of definitions (35)–(39), yields

$$A_L^{(+)}(z,t)=i\sum_\lambda e_\lambda a_\lambda(0)\exp\{i[(k_\lambda-k_L)z-(\omega_\lambda-\omega_L)t]\}$$
$$+\tfrac{1}{2}iK_{LL}\sum_{n=1}^{N}\sum_\lambda\int_0^t dt' e_\lambda^2 R^{(3)}(z_n,t')A_L^{(+)}(z_n,t')\exp\{i[(k_\lambda-k_L)(z-z_n)-(\omega_\lambda-\omega_L)(t-t')]\}$$
$$+K_{LS}\sum_{n=1}^{N}\sum_\lambda\int_0^t dt' e_\lambda^2 R^{(+)}(z_n,t')A_S^{(+)}(z_n,t')\exp\{i[(k_\lambda-k_L)(z-z_n)-(\omega_\lambda-\omega_L)(t-t')]\} \quad \text{(B1)}$$

and

$$A_S^{(+)}(z,t)=i\sum_\mu e_\mu b_\mu(0)\exp\{i[(k_\mu-k_S)z-(\omega_\mu-\omega_S)t]\}$$
$$+\tfrac{1}{2}iK_{SS}\sum_{n=1}^{N}\sum_\mu\int_0^t dt' e_\mu^2 R^{(3)}(z_n,t')A_S^{(+)}(z_n,t')\exp\{i[(k_\mu-k_S)(z-z_n)-(\omega_\mu-\omega_S)(t-t')]\}$$
$$-K_{LS}^*\sum_{n=1}^{N}\sum_\mu\int_0^t dt' e_\mu^2 R^{(-)}(z_n,t')A_L^{(+)}(z_n,t')\exp\{i[(k_\mu-k_S)(z-z_n)-(\omega_\mu-\omega_S)(t-t')]\} . \quad \text{(B2)}$$

The free-field contributions are clearly present in the first term on the right-hand side of each of these expressions. Applying the operator $\partial_z+(1/c)\partial_t$, we obtain

$$\left[\partial_z+\frac{1}{c}\partial_t\right]A_L^{(+)}(z,t)=\tfrac{1}{2}i(K_{LL}/c)\sum_{n=1}^{N}R^{(3)}(z_n,t)A_L^{(+)}(z_n,t)\sum_\lambda e_\lambda^2 e^{i(k_\lambda-k_L)(z-z_n)}$$
$$+(K_{LS}/c)\sum_{n=1}^{N}R^{(+)}(z_n,t)A_S^{(+)}(z_n,t)\sum_\lambda e_\lambda^2 e^{i(k_\lambda-k_L)(z-z_n)} \quad \text{(B3)}$$

and

$$\left[\partial_z+\frac{1}{c}\partial_t\right]A_S^{(+)}(z,t)=\tfrac{1}{2}i(K_{SS}/c)\sum_{n=1}^{N}R^{(3)}(z_n,t)A_S^{(+)}(z_n,t)\sum_\mu e_\mu^2 e^{i(k_\mu-k_S)(z-z_n)}$$
$$-(K_{LS}^*/c)\sum_{n=1}^{N}R^{(-)}(z_n,t)A_L^{(+)}(z_n,t)\sum_\mu e_\mu^2 e^{i(k_\mu-k_S)(z-z_n)} . \quad \text{(B4)}$$

Adopting the continuum limit, we let

$$\sum_\lambda e_\lambda^2 e^{i(k_\lambda-k_L)(z-z_n)}\to\frac{L}{2\pi}\int dk'\left[\frac{2\pi\hbar k'c}{V}\right]e^{i(k'-k_L)(z-z_n)}$$
$$=Le_L^2[\delta(z-z_n)-ik_L^{-1}\delta'(z-z_n)] \quad \text{(B5)}$$

if the integration limits are taken to infinity. The second term in this expression, when used in integrals, will produce spatial derivatives of the slowly varying terms divided by k_L. In the spirit of the slowly-varying-envelope approximation, it is therefore reasonable to drop this term, letting

$$\sum_\lambda e_\lambda^2 e^{i(k_\lambda-k_L)(z-z_n)}\approx Le_L^2\delta(z-z_n) . \quad \text{(B6)}$$

Also taking

$$\sum_{n=1}^{N}\to(N/L)\int_0^L dz' ,$$

with $z_n\to z'$, we then obtain (40). Equation (41) is similarly derived.

The molecular equations (42) and (43) are the simple consequence of applying the definitions (37)–(39) to the Heisenberg equations (33) and (34), then letting $z_n\to z$. Deriving the correlation functions $G^{(+)}(z,t)$ requires in addition the use of the quantum fluctuation-dissipation theorem.[33] This implies that, under the Markov approximation, with (33) and its conjugate written as

$$\dot\sigma_n^-(t)=f_n^-(t)+g_n^-(t) \quad \text{(B7)}$$

and

$$\dot\sigma_n^+(t)=f_n^+(t)+g_n^+(t) \quad \text{(B8)}$$

that

$$\langle g_n^+(t)g_{n'}^-(t')\rangle_R=\left[\frac{d}{dt}\langle\sigma_n^+\sigma_{n'}^-\rangle_R-\langle\sigma_n^+f_{n'}^-\rangle_R-\langle f_n^+\sigma_{n'}^-\rangle_R\right]\delta(t-t')$$
$$=\gamma[1+\sigma_n^z(t)]\delta_{nn'}\delta(t-t') \qquad (B9)$$

and

$$\langle g_n^-(t)g_{n'}^+(t')\rangle_R=\left[\frac{d}{dt}\langle\sigma_n^-\sigma_{n'}^+\rangle_R-\langle\sigma_n^-f_{n'}^+\rangle_R-\langle f_n^-\sigma_{n'}^+\rangle_R\right]\delta(t-t')$$
$$=\gamma[1-\sigma_n^z(t)]\delta_{nn'}\delta(t-t') \ . \qquad (B10)$$

The definitions (38) and (39), and the replacement $\delta_{nn'}\to(L/N)\delta(z-z')$ in the continuum limit then give (45) and (46).

The properties of $A_{S0}^{(+)}(\tau)$ defined by (58) are easily derived. Assuming that the Stokes occupies the vacuum state, then

$$\langle A_{S0}^{(+)}(\tau)A_{S0}^{(-)}(\tau')\rangle=\sum_\mu\sum_{\mu'}\left[\frac{e_\mu e_{\mu'}}{e_S^2}\right]e^{-i(\omega_\mu-\omega_S)\tau}\times e^{i(\omega_{\mu'}-\omega_S)\tau'}\langle b_\mu(0)b_{\mu'}^\dagger(0)\rangle$$
$$=e_S^{-2}\sum_\mu e_\mu^2 e^{-i(\omega_\mu-\omega_S)(\tau-\tau')} \ . \qquad (B11)$$

Arguments similar to those leading to (B6), taking into account the scaling (48) that has been adopted, lead directly to (63). The Gaussian decomposition rule is found from the factorization of products such as

$$\langle b_{\mu_1}(0)b_{\mu_2}(0)b_{\mu_3}^\dagger(0)b_{\mu_4}^\dagger(0)\rangle=\langle b_{\mu_1}(0)b_{\mu_3}^\dagger(0)\rangle\langle b_{\mu_2}(0)b_{\mu_4}^\dagger(0)\rangle+\langle b_{\mu_1}(0)b_{\mu_4}^\dagger(0)\rangle\langle b_{\mu_2}(0)b_{\mu_3}^\dagger(0)\rangle \ . \qquad (B12)$$

Similarly, using the definition (59), we find

$$\langle R_0^{(+)}(\zeta_n)R_0^{(-)}(\zeta_{n'})\rangle=N^2\langle\sigma_n^-(0)\sigma_{n'}^+(0)\rangle=N^2\delta_{nn'} \ . \qquad (B13)$$

Because $\delta_{nn'}\to(1/N)\delta(\zeta-\zeta')$ in the continuum limit when scaled variables are used, Eq. (66) follows directly. In higher-order products, e.g.,

$$\langle\sigma_{n_1}^-(0)\sigma_{n_2}^-(0)\sigma_{n_3}^+(0)\sigma_{n_4}^+(0)\rangle=\delta_{n_1n_3}\delta_{n_2n_4}+\delta_{n_1n_4}\delta_{n_2n_3}-2\delta_{n_1n_2n_3n_4} \ , \qquad (B14)$$

non-Gaussian contributions, like the last term in (B14), become negligible for large N.

APPENDIX C: NORMAL AND ANTINORMAL REPRESENTATIONS OF SRS

We list here the Fokker-Planck and stochastic differential equations for plane-wave SRS. For the former we adopt the following notation: we designate the quasiperiodicity function as P or Q, depending upon whether *field* averages yield, respectively, normally (P) or antinormally (Q) ordered expectation values, and attach a subscript N (normal) or A (antinormal) to indicate the ordering correspondence of *molecular* averages. Thus, the function Q defined by Eqs. (108) and (109) would be written as Q_N. The three other possibilities are as follows:

$$C_{P_N}(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t)\equiv\mathrm{tr}[e^{i\mathbf{u}^*\cdot\mathbf{a}^\dagger}e^{i\mathbf{u}\cdot\mathbf{a}}e^{i\mathbf{v}^*\cdot\mathbf{b}^\dagger}e^{i\mathbf{v}\cdot\mathbf{b}}e^{i\mathbf{r}^*\cdot\sigma^+}e^{i\mathbf{z}\cdot\sigma^z}e^{i\mathbf{r}\cdot\sigma^-}\rho(t)] \ , \qquad (C1a)$$

$$P_N(\boldsymbol{\alpha},\boldsymbol{\beta},\boldsymbol{\rho},\boldsymbol{\zeta},t)\equiv\frac{1}{(2\pi)^{7N}}\int d^{2N}u\int d^{2N}v\int d^{2N}r\int d^Nz\exp[-i(\mathbf{u}\cdot\boldsymbol{\alpha}+\mathbf{u}^*\cdot\boldsymbol{\alpha}^*+\mathbf{v}\cdot\boldsymbol{\beta}+\mathbf{v}^*\cdot\boldsymbol{\beta}^*+\mathbf{r}\cdot\boldsymbol{\rho}+\mathbf{r}^*\cdot\boldsymbol{\rho}^*+\mathbf{z}\cdot\boldsymbol{\zeta})]C_{P_N}(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t) \ ; \qquad (C1b)$$

$$C_{P_A}(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t)\equiv\mathrm{tr}[e^{i\mathbf{u}^*\cdot\mathbf{a}^\dagger}e^{i\mathbf{u}\cdot\mathbf{a}}e^{i\mathbf{v}^*\cdot\mathbf{b}^\dagger}e^{i\mathbf{v}\cdot\mathbf{b}}e^{i\mathbf{r}\cdot\sigma^-}e^{i\mathbf{z}\cdot\sigma^z}e^{i\mathbf{r}^*\cdot\sigma^+}\rho(t)] \ . \qquad (C2a)$$

$$P_A(\boldsymbol{\alpha},\boldsymbol{\beta},\boldsymbol{\rho},\boldsymbol{\zeta},t)\equiv\frac{1}{(2\pi)^{7N}}\int d^{2N}u\int d^{2N}v\int d^{2N}r\int d^Nz\exp[-i(\mathbf{u}\cdot\boldsymbol{\alpha}+\mathbf{u}^*\cdot\boldsymbol{\alpha}^*+\mathbf{v}\cdot\boldsymbol{\beta}+\mathbf{v}^*\cdot\boldsymbol{\beta}^*+\mathbf{r}\cdot\boldsymbol{\rho}+\mathbf{r}^*\cdot\boldsymbol{\rho}^*+\mathbf{z}\cdot\boldsymbol{\zeta})]C_{P_A}(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t) \ ; \qquad (C2b)$$

$$C_{Q_A}(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t)\equiv\mathrm{tr}[e^{i\mathbf{u}\cdot\mathbf{a}}e^{i\mathbf{u}^*\cdot\mathbf{a}^\dagger}e^{i\mathbf{v}\cdot\mathbf{b}}e^{i\mathbf{v}^*\cdot\mathbf{b}^\dagger}e^{i\mathbf{r}\cdot\sigma^-}e^{i\mathbf{z}\cdot\sigma^z}e^{i\mathbf{r}^*\cdot\sigma^+}\rho(t)] \ , \qquad (C3a)$$

$$Q_A(\boldsymbol{\alpha},\boldsymbol{\beta},\boldsymbol{\rho},\boldsymbol{\zeta},t)\equiv\frac{1}{(2\pi)^{7N}}\int d^{2N}u\int d^{2N}v\int d^{2N}r\int d^Nz\exp[-i(\mathbf{u}\cdot\boldsymbol{\alpha}+\mathbf{u}^*\cdot\boldsymbol{\alpha}^*\cdot\mathbf{v}\cdot\boldsymbol{\beta}+\mathbf{v}^*\cdot\boldsymbol{\beta}^*+\mathbf{r}\cdot\boldsymbol{\rho}+\mathbf{r}^*\cdot\boldsymbol{\rho}^*+\mathbf{z}\cdot\boldsymbol{\zeta})]C_{Q_A}(\mathbf{u},\mathbf{v},\mathbf{r},\mathbf{z},t) \ . \qquad (C3b)$$

The Fokker-Planck equations associated with these functions are

$$\frac{\partial P_N}{\partial t} = \Bigg\{ \Bigg[i\omega \sum_n \frac{\partial}{\partial \rho_n}\rho_n + i\sum_{l,l'} \omega_{ll'} \frac{\partial}{\partial \alpha_l}\alpha_{l'} + i \sum_{m,m'} \omega_{mm'} \frac{\partial}{\partial \beta_m}\beta_{m'}$$

$$+ \tfrac{1}{2} iK_{LL} \sum_{l,l',n} e^*_{nl} e_{nl'} \left[-\frac{\partial}{\partial \alpha_l}\alpha_{l'}\zeta_n - 2\frac{\partial}{\partial \rho_n}\alpha^*_l \alpha_{l'}\rho_n + 2\frac{\partial^2}{\partial \alpha_l \partial \rho_n}\alpha_{l'}\rho_n \right]$$

$$+ \tfrac{1}{2} K_{SS} \sum_{m,m',n} e^*_{nm} e_{nm'} \left[-\frac{\partial}{\partial \beta_m}\beta_{m'}\zeta_n - 2\frac{\partial}{\partial \rho_n}\beta^*_m \beta_{m'}\rho_n + 2\frac{\partial^2}{\partial \beta_m \partial \rho_n}\beta_{m'}\rho_n \right]$$

$$+ iK_{LS} \sum_{l,m,n} e^*_{nl} e_{nm} \Bigg[-\frac{\partial}{\partial \alpha_l}\beta_m \rho_n + \frac{\partial}{\partial \beta^*_m}\alpha^*_l \rho_n - \frac{\partial}{\partial \rho^*_n}\alpha^*_l \beta_m \zeta_n + 2\frac{\partial}{\partial \zeta_n}\alpha^*_l \beta_m \rho_n$$

$$+ \frac{\partial^2}{\partial \beta^*_m \rho^*_n}\alpha^*_l \zeta_n - 2\frac{\partial^2}{\partial \beta^*_m \zeta_n}\alpha^*_l \rho_n + \frac{\partial^2}{\partial \rho^{*2}_n}\alpha^*_l \beta_m \rho^*_n - 2\frac{\partial^2}{\partial \zeta^2_n}\alpha^*_l \beta_m \rho_n \Bigg]$$

$$+ \gamma \sum_n \left[\frac{\partial}{\partial \rho_n}\rho_n + \frac{1}{2}\frac{\partial^2}{\partial \rho_n \partial \rho^*_n}(1+\zeta_n) \right] \Bigg] + \text{c.c.} \Bigg\} P_N \;, \tag{C4}$$

$$\frac{\partial P_A}{\partial t} = \Bigg\{ \Bigg[i\omega \sum_n \frac{\partial}{\partial \rho_n}\rho_n + i\sum_{l,l'} \omega_{ll'} \frac{\partial}{\partial \alpha_l}\alpha_{l'} + i \sum_{m,m'} \omega_{mm'} \frac{\partial}{\partial \beta_m}\beta_{m'}$$

$$+ \tfrac{1}{2} iK_{LL} \sum_{l,l',n} e^*_{nl} e_{nl'} \left[-\frac{\partial}{\partial \alpha_l}\alpha_{l'}\zeta_n + 2\frac{\partial}{\partial \rho^*_n}\alpha^*_l \alpha_{l'}\rho^*_n - 2\frac{\partial^2}{\partial \alpha_l \partial \rho^*_n}\alpha_{l'}\rho^*_n \right]$$

$$+ \tfrac{1}{2} iK_{SS} \sum_{m,m',n} e^*_{nm} e_{nm'} \left[-\frac{\partial}{\partial \beta_m}\beta_{m'}\zeta_n + 2\frac{\partial}{\partial \rho^*_n}\beta^*_m \beta_{m'}\rho^*_n - 2\frac{\partial^2}{\partial \beta_m \partial \rho^*_n}\beta_{m'}\rho^*_n \right]$$

$$+ iK_{LS} \sum_{l,m,n} e^*_{nl} e_{nm} \Bigg[-\frac{\partial}{\partial \alpha_l}\beta_m \rho_n + \frac{\partial}{\partial \beta^*_m}\alpha^*_l \rho_n - \frac{\partial}{\partial \rho^*_n}\alpha^*_l \beta_m \zeta_n + 2\frac{\partial}{\partial \zeta_n}\alpha^*_l \beta_m \rho_n$$

$$+ \frac{\partial}{\partial \alpha_l \partial \rho^*_n}\beta_m \zeta_n - 2\frac{\partial^2}{\partial \alpha_l \partial \zeta_n}\beta_m \rho_n - \frac{\partial^2}{\partial \rho^{*2}_n}\alpha^*_l \beta_m \rho^*_n + 2\frac{\partial^2}{\partial \zeta^2_n}\alpha^*_l \beta_m \rho_n \Bigg]$$

$$+ \gamma \sum_n \left[\frac{\partial}{\partial \rho_n}\rho_n + \frac{1}{2}\frac{\partial^2}{\partial \rho_n \partial \rho^*_n}(1-\zeta_n) \right] \Bigg] + \text{c.c.} \Bigg\} P_A \;, \tag{C5}$$

and

$$\frac{\partial Q_A}{\partial t} = \Bigg\{ \Bigg[i\omega \sum_n \frac{\partial}{\partial \rho_n}\rho_n + i\sum_{l,l'} \omega_{ll'} \frac{\partial}{\partial \alpha_l}\alpha_{l'} + i \sum_{m,m'} \omega_{mm'} \frac{\partial}{\partial \beta_m}\beta_{m'}$$

$$+ \tfrac{1}{2} iK_{LL} \sum_{l,l',n} e^*_{nl} e_{nl'} \left[-\frac{\partial}{\partial \alpha_l}\alpha_{l'}\zeta_n - 2\frac{\partial}{\partial \rho_n}\alpha^*_l \alpha_{l'}\rho_n - 2\frac{\partial^2}{\partial \alpha_l \partial \rho_n}\alpha_{l'}\rho_n \right]$$

$$+ \tfrac{1}{2} iK_{SS} \sum_{m,m',n} e^*_{nm} e_{nm'} \left[-\frac{\partial}{\partial \beta_m}\beta_{m'}\zeta_n - 2\frac{\partial}{\partial \rho_n}\beta^*_m \beta_{m'}\rho_n - 2\frac{\partial^2}{\partial \beta_m \partial \rho_n}\beta_{m'}\rho_n \right]$$

$$+ iK_{LS} \sum_{l,m,n} e^*_{nl} e_{nm} \Bigg[-\frac{\partial}{\partial \alpha_l}\beta_m \rho_n + \frac{\partial}{\partial \beta^*_m}\alpha^*_l \rho_n - \frac{\partial}{\partial \rho^*_n}\alpha^*_l \beta_m \zeta_n + 2\frac{\partial}{\partial \zeta_n}\alpha^*_l \beta_m \rho_n$$

$$- \frac{\partial^2}{\partial \beta^*_m \partial \rho^*_n}\alpha^*_l \zeta_n + 2\frac{\partial^2}{\partial \beta^*_m \partial \zeta_n}\alpha^*_l \rho_n - \frac{\partial^2}{\partial \rho^{*2}_n}\alpha^*_l \beta_m \rho^*_n + 2\frac{\partial^2}{\partial \zeta^2_n}\alpha^*_l \beta_m \rho_n \Bigg]$$

$$+ \gamma \sum_n \left[\frac{\partial}{\partial \rho_n}\rho_n + \frac{1}{2}\frac{\partial^2}{\partial \rho_n \partial \rho^*_n}(1-\zeta_n) \right] \Bigg] + \text{c.c.} \Bigg\} Q_A \;. \tag{C6}$$

The stochastic differential equations derived from these results are all of the form given by Eqs. (137)–(140), differing only in the correlation functions associated with the noise sources $F_L(\zeta,\tau)$, $F_S(\zeta,\tau)$, $F_R(\zeta,\tau)$, and $F_3(\zeta,\tau)$. The set of nonzero second-order correlation functions are, from (C4) (normal field and normal molecules),

$$\langle F_L(\zeta,\tau)F_R(\zeta',\tau')\rangle = iK_{LL}A_L(\zeta,\tau)R(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C7}$$

$$\langle F_S(\zeta,\tau)F_R(\zeta',\tau')\rangle = [iK_{SS}A_S(\zeta,\tau)R(\zeta,\tau) + K_{LS}^*A_L(\zeta,\tau)R_3(\zeta,\tau)] \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C8}$$

$$\langle F_S(\zeta,\tau)F_3(\zeta',\tau')\rangle = -2K_{LS}^*A_L(\zeta,\tau)R^*(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C9}$$

$$\langle F_R(\zeta,\tau)F_R(\zeta',\tau')\rangle = 2K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C10}$$

$$\langle F_R(\zeta,\tau)F_R^*(\zeta',\tau')\rangle = \gamma[N+R_3(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C11}$$

$$\langle F_3(\zeta,\tau)F_3(\zeta',\tau')\rangle = [-4K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau) + \text{c.c.}]\delta(\zeta-\zeta')\delta(\tau-\tau') ; \tag{C12}$$

from (C5) (normal field and antinormal molecules),

$$\langle F_L(\zeta,\tau)F_R^*(\zeta',\tau')\rangle = [-iK_{LL}A_L(\zeta,\tau)R^*(\zeta,\tau) + K_{LS}A_S(\zeta,\tau)R_3(\zeta,\tau)] \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C13}$$

$$\langle F_L(\zeta,\tau)F_3(\zeta',\tau')\rangle = -2K_{LS}A_S(\zeta,\tau)R(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C14}$$

$$\langle F_S(\zeta,\tau)F_R^*(\zeta',\tau')\rangle = -iK_{SS}A_S(\zeta,\tau)R^*(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C15}$$

$$\langle F_R(\zeta,\tau)F_R(\zeta',\tau')\rangle = -2K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau) , \tag{C16}$$

$$\langle F_R(\zeta,\tau)F_R^*(\zeta',\tau')\rangle = \gamma[N-R_3(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C17}$$

$$\langle F_3(\zeta,\tau)F_3(\zeta',\tau')\rangle = [4K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau) + \text{c.c.}]\delta(\zeta-\zeta')\delta(\tau-\tau') ; \tag{C18}$$

and from (C6) (antinormal field and antinormal molecules),

$$\langle F_L(\zeta,\tau)F_R(\zeta',\tau')\rangle = -iK_{LL}A_L(\zeta,\tau)R(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C19}$$

$$\langle F_S(\zeta,\tau)F_R(\zeta',\tau')\rangle = [-iK_{SS}A_S(\zeta,\tau)R(\zeta,\tau) - K_{LS}^*A_L(\zeta,\tau)R_3(\zeta,\tau)] \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C20}$$

$$\langle F_S(\zeta,\tau)F_3(\zeta',\tau')\rangle = 2K_{LS}^*A_L(\zeta,\tau)R^*(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C21}$$

$$\langle F_R(\zeta,\tau)F_R(\zeta',\tau')\rangle = -2K_{LS}^*A_L(\zeta,\tau)A_S^*(\zeta,\tau)R(\zeta,\tau) \times\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C22}$$

$$\langle F_R(\zeta,\tau)F_R^*(\zeta',\tau')\rangle = \gamma[N-R_3(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau') , \tag{C23}$$

$$\langle F_3(\zeta,\tau)F_3(\zeta',\tau')\rangle = [4K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R(\zeta,\tau) + \text{c.c.}]\delta(\zeta-\zeta')\delta(\tau-\tau') . \tag{C24}$$

[We emphasize that (C7)–(C24) reflect the scaling transformations (48) and (136).] Finally, the initial and boundary conditions reflecting the associated ordering prescription of each case are, respectively, for (C7)–(C12),

$$A_{S0}(\tau)=0 , \tag{C25}$$

$$R_0(\zeta)=0 ; \tag{C26}$$

for (C13)–(C18),

$$A_{S0}(\tau)=0 , \tag{C27}$$

$$\langle R_0(\zeta)R_0(\zeta')\rangle = N\delta(\zeta-\zeta') ; \tag{C28}$$

and for (C19)–(C24),

$$\langle A_{S0}(\tau)A_{S0}^*(\tau')\rangle = \delta(\tau-\tau') , \tag{C29}$$

$$\langle R_0(\zeta)R_0^*(\zeta')\rangle = N\delta(\zeta-\zeta') , \tag{C30}$$

with in each case $R_3(\zeta,0)=-N$ and $A_{L0}(\tau)$ given by the incident pump-pulse profile.

It is thus evident that quantum initiation proceeds from different sources in each case. As remarked following (146), the Stokes vacuum exclusively triggers initiation with the antinormal-field–normal-molecules prescription, with no contribution from the Langevin noise sources. On the other hand, radiation reaction and collisions are both sources of quantum initiation when the normal-field–antinormal-molecules ordering prescription is adopted, according to (C17) and (C28). The other cases are more difficult to interpret. In the case of the normal-field–normal-molecules ordering, neither the Stokes vacuum, radiation reaction, nor collision play an explicit role [cf. Eqs. (C11), (C25), and (C26)]. While (C7)–(C9) are also initially nonzero, only (C8) enters into computations of the Stokes buildup in the linear regime, so it can be identified as the source of initiation. However, $F_S(\zeta,\tau)$ and $F_R(\zeta,\tau)$ must also satisfy the conditions

$$\langle F_S(\zeta,\tau)F_S^*(\zeta',\tau')\rangle = \langle F_R(\zeta,\tau)F_R^*(\zeta',\tau')\rangle = 0 ,$$

which requires that one double the "phase-space" dimension, interpreting $A_S(\zeta,\tau)$ and $A_S^*(\zeta,\tau)$ as independent complex variables [see discussion preceding Eq. (147)]. This poses no problem in analytical calculations within the linear regime, but introduces considerable complications into numerical computations of the nonlinear dynamics as well as the interpretation of the initiation process. One can say the same regarding the antinormal-field–antinormal molecules prescription.

There is thus a considerable disparity in utility among the four normal or antinormal representations. Let us emphasize in closing that they are equivalent dynamically, to the extent that the Fokker-Planck approximation is valid. The multiplicity of equally valid interpretations of quantum initiation that may be derived from these representations is similar to what is found in studies of spontaneous emission.[37]

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[1]J. C. Englund and C. M. Bowden, Phys. Rev. Lett. **57**, 2661 (1986).

[2]R. H. Dicke, Phys. Rev. **93**, 95 (1954); R. Bonifacio and L. A. Lugiato, Phys. Rev. A **11**, 1507 (1975); *Cooperative Effects in Matter and Radiation,* edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977); R. Glauber and F. Haake, Phys. Lett. **68A**, 29 (1978); F. Haake, J. Haus, H. King, G. Schröder, and R. Glauber, Phys. Rev. Lett. **45**, 558 (1980); F. Haake, H. King, G. Schröder, J. Haus, and R. Glauber, Phys. Rev. **20**, 2047 (1979); D. Polder, M. F. H. Schuurmanns, and Q. H. F. Vrehen, Phys. Rev. A **19**, 1192 (1979); C. M. Bowden and C. C. Sung, Phys. Rev. Lett. **50**, 156 (1983); F. P. Matter, H. M. Gibbs, S. L. McCall, and M. S. Feld, *ibid.* **46**, 1123 (1981); E. A. Watson, H. M. Gibbs, F. P. Mattar, M. Cormier, Y. Claude, S. L. McCall, and M. S. Feld, Phys. Rev. A **27**, 1427 (1983); F. P. Mattar, P. R. Berman, A. W. Matos, Y. Claude, C. Gauthier, and C. M. Bowden, in *Topics in Current Physics,* edited by C. D. Cantrell (Springer-Verlag, New York, 1986), Vol. 35, p. 223; C. M. Bowden, in *Laser Physics,* Vol. 182 of *Lecture Notes in Physics,* edited by J. D. Harvey and D. F. Walls (Springer-Verlag, Berlin, 1983), p. 195.

[3]H. M. Gibbs, Q. H. F. Vrehen, and H. M. J. Hickspoors, Phys. Rev. Lett. **39**, 547 (1977); in *Laser Spectroscopy III,* edited by J. S. Hall and J. L. Carlsten (Springer-Verlag, Berlin, 1977), p. 213; N. Skribanowitz, I. P. Herman, J. C. MacGillivray, and M. S. Feld, Phys. Rev. Lett. **30**, 309 (1973); M. Gross, C. Fabre, P. Pillet, and S. Haroche, *ibid.* **36**, 1035 (1976); A. Flusberg, T. Mossberg, and S. R. Hartmann, Phys. Lett. **58A**, 373 (1976); Q. H. F. Vrehen, H. M. J. Hickspoors, and H. M. Gibbs, Phys. Rev. Lett. **38**, 764 (1977): A. T. Rosenberger and T. A. DeTemple, Phys. Rev. A **24**, 858 (1981).

[4]M. S. Malcuit, J. J. Maki, D. J. Simkin, and R. W. Boyd, Phys. Rev. Lett. **59**, 1189 (1987); J. J. Maki, M. S. Malcuit, M. G. Raymer, R. W. Boyd, and P. D. Drummond, Phys. Rev. A **40**, 5135 (1989).

[5]R. Bonifacio, F. A. Hopf, P. Meystre, and M. O. Scully, Phys. Rev. A **12**, 2568 (1975); F. A. Hopf and P. Meystre, *ibid.* **12**, 2534 (1975); F. A. Hopf, P. Meystre, and D. W. McLaughlin, *ibid.* **13**, 777 (1976); R. Bonifacio, F. A. Hopf, P. Meystre, and M. O. Scully, in *Laser Induced Fusion and x-ray Laser Studies, Physics of Quantum Electronics,* edited by S. F. Jacobs, M. O. Scully, M. Sargeant III, and C. D. Cantrell (Addison-Wesley, London, 1976), Vol. III, p. 487.

[6]J. J. Ehrlich, C. M. Bowden, D. W. Howgate, S. H. Lehnigk, A. T. Rosenberger, and T. A. DeTemple, in *Coherence and Quantum Optics IV,* edited by L. Mandel and E. Wolf (Plenum, New York, 1978), p. 923; A. T. Rosenberger, T. A. DeTemple, C. M. Bowden, and C. C. Sung, J. Opt. Soc. Am. **68**, 700 (1978); A. T. Rosenberger and T. A. DeTemple, Phys. Rev. A **24**, 858 (1981).

[7]C. M. Bowden and F. P. Mattar, in *Coherence and Quantum Optics V,* edited by L. Mandel and E. Wolf (Plenum, New York, 1984), p. 87.

[8]M. G. Raymer and J. Mostowski, Phys. Rev. A **24**, 1980 (1981); M. G. Raymer, K. Rzazewski, and J. Mostowski, Opt. Lett. **7**, 71 (1982).

[9]I. A. Walmsley, M. G. Raymer, T. Sizer II, J. N. Duling III, and J. D. Kafka, Opt. Commun. **53**, 137 (1985).

[10]C. M. Bowden and J. C. Englund, Opt. Commun. **67**, 71 (1988).

[11]F. A. Hopf, Phys. Rev. A **20**, 2064 (1979).

[12]F. A. Hopf, and E. A. Overman II, Phys. Rev. A **19**, 1180 (1979).

[13]L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[14]J. Rai, R. Inguva, and C. M. Bowden, in *Coherence and Quantum Optics VI,* edited by L. Mandel and E. Wolf (Plenum, New York, in press); J. Rai and C. M. Bowden, Optics News **15**, A-44 (1989).

[15]C. M. Bowden and J. Rai (unpublished).

[16]J. C. Englund and C. M. Bowden, SPIE Proceedings, **874**, 218 (1988).

[17]J. C. Englund and C. M. Bowden, Opt. News **14**, 224 (1988).

[18]C. M. Bowden and J. C. Englund, in *Quantum Optics V,* edited by J. C. Harvey and D. F. Walls (Springer-Verlag, New York, 1989), p. 222.

[19]C. M. Bowden and J. C. Englund (unpublished).

[20]J. C. Englund and C. M. Bowden, Opt. News **15**, A-36 (1989).

[21]D. C. MacPherson and J. L. Carlsten, in *International Quantum Electronics Conference 1987, Technical Digest Series* (Optical Society of America, Washington, D.C., 1987), Vol. 21.

[22]D. C. MacPherson, R. C. Swanson and J. L. Carlsten, Phys. Rev. Lett. **61**, 66 (1988); D. C. MacPherson and J. L. Carlsten, J. Opt. Soc. Am. B **4**, 32 (1987).

[23]D. C. MacPherson, R. C. Swanson and J. L. Carlsten, Opt. News **14**, 222 (1988).

[24]D. C. MacPherson, R. C. Swanson, and J. L. Carlsten, Phys. Rev. A **40**, 6745 (1989).

[25]F. Y. F. Chu and A. C. Scott, Phys. Rev. A **12**, 2060 (1975); N. Tan-No, F. Shirakata, K. Kaloto, and H. Inaba, *ibid.* **12**, 159 (1975); T. M. Makhviladze, M. E. Sanychev, and L. A. Shelepin, Zh. Eksp. Teor. Fiz. **69**, 499 (1975) [Sov. Phys.—JETP **42**, 255 (1976)]; D. J. Kaup, Physica **6D**, 143 (1983); H. Steudel, *ibid.* **6D**, 155 (1983); R. Meinel, Opt. Commun. **49**, 224 (1984); T. M. Makhviladze and M. E. Sanychev, Zn. Eksp. Teor. Fiz. **71**, 896 (1976) [Sov. Phys.—JETP **44**, 471 (1976)]; A. Hasegawa, Appl. Opt. **23**, 3302 (1984); Y. Gürsey, Phys. Rev. A **36**, 4792 (1987).

[26]K. J. Drühl, R. G. Wenzel and J. L. Carlsten, Phys. Rev. Lett. **51**, 1171 (1983).

[27]K. J. Drühl, J. L. Carlsten, and R. G. Wenzel, J. Stat. Phys. **39**, 615 (1985); **39**, 621 (1985).

[28]J. R. Ackerhalt and P. W. Milonni, Phys. Rev. A **33**, 3195 (1986).

[29]C. M. Bowden and J. C. Englund in *Vibrational Spectra and Structure,* edited by H. R. Durig (Elsevier, Amsterdam, 1989),

Vol. 17B, p. 107.

[30]F. Y. F. Chu, Phys. Rev. A **12**, 2065 (1975).

[31]J. C. Englund and C. M. Bowden (unpublished).

[32]C. A. Coulter, Phys. Rev. A **10**, 1946 (1974).

[33]M. Lax, Phys. Rev. **145**, 110 (1966).

[34]W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).

[35]P. D. Drummond and C. W. Gardiner, J. Phys. A **13**, 2353 (1980).

[36]C. W. Gardiner, *Handbook of Stochastic Methods* (Springer-Verlag, Berlin, 1983).

[37]J. R. Ackerhalt, P. L. Knight, and J. H. Eberly, Phys. Rev. Lett. **30**, 456 (1973); J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D **10**, 3350 (1974).